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ABSTRACT

Verbatim transcriptions of various seminars on ultra high flux research reactors, originally issued as separate ORNL memoranda, have been revised slightly and here issued as a unit. A brief summary of these seminars is included as an introduction to the series and to point out the more important conclusions.

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TABLE OF CONTENTS

	<u>Page</u>
Summary - J. A. Lane .....	3
The Production of Transuranic Elements - A. Chetham-Strode .....	10
The Production of Isotopes with High Fluxes - A. F. Rupp .....	25
Neutron Diffraction - H. A. Levy .....	38
Neutron Diffraction with Powder Samples - E. O. Wollan .....	53
Velocity Selector Experiments - J. A. Harvey .....	64
Solid State Physics - D. S. Billington .....	80
Physics of Flux-Trap and Other High Flux Reactors - W. K. Ergen .....	98
Design Considerations of Aqueous Flux-Trap Reactors - J. A. Lane .....	116
Laboratory Program - A. M. Weinberg .....	137

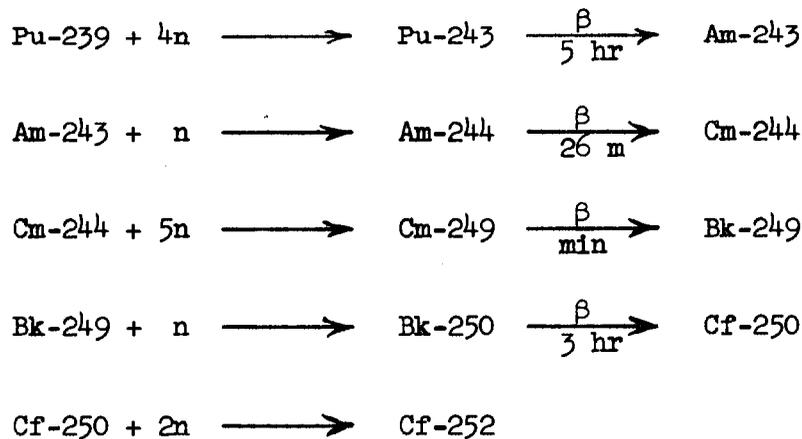
SUMMARY OF THE SEMINARS ON ULTRA HIGH FLUX RESEARCH REACTORS

Introduction

At a meeting on December 6, 1957, attended by various members of the staff of the Oak Ridge National Laboratory, the possible need for neutron fluxes an order of magnitude higher than presently available, for isotope production, improved research in neutron diffraction, and solid state physics, was discussed. As a result of this meeting it was decided to hold a series of informal seminars to discuss the uses to which an ultra high flux research reactor might be put and to review the problems associated with the design and construction of such a reactor. It was further decided to record and transcribe a verbatim account of the talks presented and the accompanying discussions. These transcriptions have been issued as the following memoranda: Seminar No. 1, "Isotope Production," A. Chetham-Strode and A. F. Rupp, ORNL CF-58-1-98; Seminar No. 2, "Neutron Diffraction," H. A. Levy and E. O. Wollan, ORNL CF-58-2-99; Seminar No. 3, "Velocity Selector Experiments," J. A. Harvey and "Solid State Physics," D. S. Billington, ORNL CF-58-2-132; Seminar No. 4, "Solid State Physics," D. S. Billington (continued), ORNL CF-58-6-49; Seminar No. 5, "Physics of Flux-Trap and Other Ultra High Flux Reactors," W. K. Ergen, ORNL CF-58-6-50; Seminar No. 6, "Design Problems of Aqueous Flux-Trap Reactors," J. A. Lane, ORNL CF-58-6-51; Seminar No. 7, "The Laboratory Program on Ultra High Flux Reactors," A. M. Weinberg, ORNL CF-58-6-89. The information presented in each of these seminars is summarized as follows:

The Production of Transuranic Elements - A. Chetham-Strode

The production of transuranic elements through successive neutron captures in a reactor proceeds according to the following reactions:



Thus, it is seen that thirteen successive neutron captures are required to produce Cf-252 from Pu-239 or ten neutron captures starting with Pu-242. Because of this, the initial production rate of Cf-252 from plutonium goes up as the 10th to 13th power of the flux. Although this initial rate drops due to burn-up of lighter element materials and by the approach to equilibrium of some of the high cross-section intermediates, however, after a year's irradiation the flux dependence is still somewhere between the second and sixth order. For example, the continuous irradiation of 6 grams of Pu-242 (produced from 100 grams of Pu-239) at  $3 \times 10^{14}$  flux, yields after one year about 0.01 micrograms of Cf-252. At  $5 \times 10^{15}$  flux, on the other hand, the production of Cf-252 would be a factor of about  $3 \times 10^5$  greater. In this case, the yield of Cf-252 from 6 grams of Pu-242 would be 3 milligrams. This is many thousand times the total amount of californium available in the United States to date. If it is desired to produce these heavy elements in a reactor, therefore, thermal neutron fluxes in the range of 3 to  $5 \times 10^{15}$  are essential.

A second important use of ultra high fluxes is the production of special heavy element isotopes such as Pu-244 by bypassing short-lived  $\beta$  decay elements. At  $5 \times 10^{15}$  flux, about 2% of the Pu-242 can be transformed into Pu-244, whereas at lower fluxes only negligible amounts are produced due to the short half-life of Pu-243. The isotope Pu-244 could be used to great advantage for certain nuclear research work because of its long alpha half-life and 0 spin.

Ultra high neutron fluxes can also be used to vary the isotopic ratios of certain transuranic elements by burn-out of high cross-section isotopes. The curium isotopes, which normally contain up to 95% Cm-244, 4% Cm-246, and 1% Cm-245, are an example of this. By irradiating such a mixture at ultra high fluxes, it is possible to obtain curium which is largely Cm-248 and which contains little or no Cm-244.

One of the major aims of the field of transuranic elements is the production of weighable quantities of elements 101 and above. Unfortunately, a high flux reactor is not suitable for the production of such new elements. Even starting with Cf-252 it takes seven successive captures before reaching the first fermium beta emitter, Fm-259. To get to Fm-259 it is necessary to pass through fermium isotopes which have a very short spontaneous fission half-life (i.e., Fm-256, 3 hours; Fm-258,  $\sim$  minutes). So it is not possible to get through these intermediate steps. Though starting with einsteinium isotopes such as E-253 and E-254 avoids the spontaneous fission problem, one encounters the problem of short half-lives for beta decay. Most plans for producing new heavy elements, therefore, consider using reactor-produced californium as target material for heavy ion bombardment.

#### The Production of Isotopes with High Fluxes - A. F. Rupp

The primary advantages of ultra high neutron fluxes for isotope production are: (a) the ability to obtain higher specific activities of

short-life isotopes, (b) the speeding up in the production of long-lived materials, (c) the burning out of undesired atoms in an isotopic mixture, (d) the conservation of high cost target materials, (e) the production of carrier-free isotopes, and (f) the reduction in cost of isotopes. Examples of these advantages of high fluxes for isotope production are as follows:

1. The production of high specific activity, short-lived isotopes, such as 12.8 hour Cu-64, 14.2 hour Ga-72, 12.6 hour I-130, 12.5 hour K-42, and 15 hour Na-24, may be improved by up to a factor of 10 by irradiating the parent atoms at  $10^{15}$  flux instead of  $10^{14}$  flux. However, in some cases, such as Eu-152 which is produced from a very high cross section target material Eu-151 (7000 barns), exposure at  $10^{15}$  flux produces no higher activity per gram starting material than  $10^{14}$  flux. This is due to burn-out of the parent atoms. The actual specific activity of the product is, however, higher. In other cases when the thermal neutron cross section of the product atom is high, such as Au-198 (26,000 barns), increasing the flux does not give a corresponding increase in the activity of the product due to the more rapid burning out of product.

2. The production of long-lived isotopes such as 7.2 year Ba-133 and 5.3 year Co-60 may be speeded up by irradiating at higher fluxes. The time to reach the maximum activity of Ba-133, for example, is decreased from 19.8 years at  $10^{14}$  flux to 6.6 years at  $10^{15}$  flux. Also, for Co-60 the time for maximum activity is decreased from 7.8 years to 1.6 years at these two flux levels. However, an ultra high flux reactor may not be suitable for producing large quantities of Co-60 since in such a case a large volume for irradiation and a large number of excess neutrons are required.

3. High fluxes can also be used to burn out undesired atoms in an isotopic mixture. For example, at  $10^{15}$  flux, 99% of Co-59 could be burned out of Co-60 in about 5 years. Similarly, Eu-152 and Eu-154 could be removed from fission product Eu-155.

4. Many isotopes are prepared by irradiating isotopically-enriched target material, such as Cr-50 to make Cr-51. At higher fluxes, less target material would be needed - reducing the cost of product.

5. High fluxes can also be used to advantage in producing carrier-free isotopes. For example, Ca-45, now made by an n,p reaction with Sc-45 followed by chemical separation, could be made from Ca-44 at  $10^{15}$  flux at considerably lower cost.

#### Neutron Diffraction - H. A. Levy

The use of higher fluxes for neutron diffraction experiments would permit measurements to be made more rapidly, more easily, and with greater accuracy. At present it is possible to analyze a crystal with 20 atoms, such as n-acetyl glycine, by neutron diffraction techniques using beams from the X-10 graphite pile. Because of the problem of resolution of the

beam, the intensity of individual reflections and the time for collecting data, the desired neutron flux is proportional to the cube of the number of atoms in the unit cell being investigated. To analyze a crystal with 200 atoms, such as Vitamin B<sub>12</sub>, which is the best that can be done with X-ray diffraction, would require, therefore, a flux of about  $3 \times 10^{14}$  at the interior of the beam hole. To analyze a molecule with  $\sim 2000$  atoms per unit cell, such as that of a simple protein, would require at least  $3 \times 10^{17}$  n/cm<sup>2</sup>(sec). Actually, because of the possible need for discriminating against a higher fast-neutron component or a reduction in sample size, the flux necessary for protein analysis could easily be as much as  $3 \times 10^{19}$ . In general, for neutron diffraction work, the ratio of slow to fast neutrons and slow neutrons to gamma rays is equally as important as higher fluxes per se.

#### Neutron Diffraction with Powder Samples - E. O. Wollan

In order to make neutron powder photographs of a quality and resolution comparable to those obtainable with X-rays, a beam of about  $10^9$  monoenergetic (0.06 ev) n/cm<sup>2</sup>(sec) would be required. This corresponds to a total thermal neutron flux of  $3 \times 10^{15}$ . With the  $10^5$  n/cm<sup>2</sup>(sec) beam from the X-10 pile, the resolution is 20-100 times poorer and the time to obtain a pattern about 20 times longer than with  $10^{10}$  photons/cm<sup>2</sup>(sec) at 1 ft. Although the sample size can be increased, which tends to offset some of these effects, a factor of 10,000 is still needed to do work comparable to that with X-rays.

#### Velocity Selector Experiments - J. A. Harvey

With regard to cross section measurements using a velocity selector, a factor of 100 in flux would improve the resolution of the measurements a factor of three. Fluxes in the range of  $10^{14}$  are adequate for measuring cross sections at neutron energies up to 100 ev with a resolution of 0.3 ev. At  $5 \times 10^{15}$  flux, the resolution would be 0.06 ev at 100 volts, 0.4 ev at 400 volts, and 50 ev at 10,000 volts. Above 10 kv, better measurements can be made using a Van de Graaff generator. Although an ultra high flux reactor would permit neutron cross section measurements to be made more accurately and at higher neutron energies, equal gains at lower cost might be made by improving the performance of the detecting equipment.

#### Solid State Physics - D. S. Billington

In order to safely specify materials to be used in a  $10^{14}$  flux reactor, a knowledge of what happens at  $10^{15}$  flux or above is desirable. One example of this is the creep rate of metals. However, fast neutron irradiations are required for such work, rather than thermal neutrons. The effects of radiation on the mechanical properties of reactor structural materials do not show up until the integrated fast flux dose reaches  $10^{21}$  nvt. To actually be safe, the effect of doses of  $10^{22}$  should be determined. At  $1 \times 10^{14}$  fast flux, this point is reached in about 3 years; at  $1 \times 10^{15}$ , in 4 months; and at  $5 \times 10^{15}$ , in about 3 weeks. The difference between

4 months and 3 weeks, however, is not sufficient to justify a  $5 \times 10^{15}$  reactor compared to a  $1 \times 10^{15}$  reactor. Fast fluxes of this magnitude are already obtainable in the ETR.

In addition to reducing the time for making radiation damage studies, a higher fast-flux reactor would open up the possibility of doing radiation damage studies with fast-neutron beams, rather than inside the reactor. This would cut down on the amount of gamma heating and permit insertion and withdrawal of samples without shutting down the reactor.

High fast fluxes could be also used to better investigate the effect of radiation on mechanical properties such as brittle fracture and Young's modulus of metals, to study the effect of radiation on the properties of ceramics, to study solid state reactions such as the phase reversion of alloys, and to carry out neutron transmission and diffraction experiments with metals. The effect of thermal neutron transmutation on the properties of solids could be studied at higher fluxes. For most of the experiments described above, fluxes of  $10^{15}$  or less are acceptable.

#### Physics of Flux Trap and Other Ultra High Flux Reactors - W. K. Ergen

A preliminary comparison of reactors designed to provide high fast flux irradiations indicates that molten salt reactors are particularly well-suited for such an application. This is because the fast flux at the surface of a fuel region comes mainly from neutrons which are generated within one slowing-down length of the surface and molten salt reactors have a large slowing-down length. Also in such reactors, the power density or number of watts per cc is large, so that the production of fission neutrons in the fuel region concerned is large.

To maximize the thermal neutron flux, on the other hand, it is necessary to reduce the macroscopic fission cross section or, in other words, reduce the concentration of fissionable material per unit volume. However, even in an infinite reactor the absorption of neutrons by moderator, structure, impurities, etc. sets a limit below which the macroscopic fission cross section cannot go. Below this value it is not possible to maintain a critical system. On this basis it can be shown that the maximum thermal flux obtainable is equal to  $4 \times 10^{10} \div$  the parasitic neutron absorption cross section. Assuming that the power density cannot go above 100 watts per cc and that the only parasitic absorptions are those due to moderator, thermal neutron fluxes up to  $6 \times 10^{16}$  are obtainable with  $D_2O$ ,  $10^{16}$  with carbon, 3 to  $5 \times 10^{15}$  with beryllium or beryllium oxide, and  $2 \times 10^{14}$  with  $H_2O$ . If the total power is limited to 100 Mw rather than the power density to 100 watts per cc, however, the maximum thermal fluxes will be in the range of 1.7 to  $3.5 \times 10^{15}$  for all moderators considered with beryllium oxide, beryllium,  $D_2O$ ,  $H_2O$ , and carbon in order of preference.

An investigation of spherical flux-trap reactors consisting of thin shells of fuel in an infinite moderator indicates that for each moderator

there exists a shell radius at which the thermal flux at the center of the sphere per Mw of power is a maximum. This radius for maximum flux is approximately equal to  $2\sqrt{\tau}$  where  $\sqrt{\tau}$  is the slowing-down length in the moderator. Estimated values of  $\phi_{\max}/\text{Mw}$  for beryllium, beryllium oxide,  $\text{D}_2\text{O}$ , and carbon were  $7.2 \times 10^{13}$ ,  $6.9 \times 10^{13}$ ,  $3.8 \times 10^{13}$ , and  $3.2 \times 10^{13}$ , respectively.  $\text{H}_2\text{O}$  was not included since a thin shell of enriched fuel cannot be made critical in  $\text{H}_2\text{O}$  at any radius. A spherical shell reactor moderated by  $\text{H}_2\text{O}$  and  $\text{D}_2\text{O}$  either mixed or in separate regions can, however, be made critical and such a system may give higher values of  $\phi_{\max}/\text{Mw}$  than the single moderator systems considered.

#### Design Considerations of Aqueous Flux-Trap Reactors - J. A. Lane

In order to achieve thermal neutron fluxes significantly above  $10^{15}$  n/cm<sup>2</sup>(sec) in a heterogeneous reactor, higher power densities must be achieved than in existing reactors. This requires thinner fuel elements, higher coolant velocities, and higher operating temperatures. Calculations indicate a limit of about  $3 \times 10^8$  Btu/hr (cu ft) or 3100 watts per cc for fuel elements obtainable with present technology and with a practical heat removal system. On this basis, for a 90-cm-long core with a 60-cm-diameter island, a 30-cm-thick fuel annulus would be required to remove 800 Mw of heat. The unperturbed center flux in such a reactor moderated, cooled, and reflected with  $\text{D}_2\text{O}$  and loaded to give a 5-day fuel cycle would be 7 to  $9 \times 10^{15}$  n/cm<sup>2</sup>(sec). Increasing the power level above 800 Mw would have no effect on the center flux since a larger fuel annulus and greater fuel loading would be required, resulting in a reduced flux peaking. The only way to increase the thermal flux in the center would be to use a different moderator in the island or increase the power density in the fuel region. It does not seem possible, therefore, to achieve a flux of  $10^{16}$  in a  $\text{D}_2\text{O}$  moderated flux-trap reactor under the assumed conditions of heat removal. Calculations for mixed systems (i.e.,  $\text{H}_2\text{O}$  in island,  $\text{D}_2\text{O}$  in core and reflector) now underway will determine whether  $10^{16}$  fluxes are obtainable in such reactors.

#### Laboratory Program - A. M. Weinberg

The following conclusions can be drawn from the preceding seminars on ultra high flux reactors:

1. The best justification for building an expensive ultra high flux reactor is in connection with the production of transuranic elements and other isotopes.
2. Thermal and fast fluxes available from the best research reactors built to date (MITR, ETR, ORR) are, in general, adequate for cross-section measurements, neutron diffraction work, solid state physics, and other nuclear research programs.
3. Further consideration should be given to high flux reactors with poor moderators as a means of providing high epithermal fluxes.

4. A number of smaller research reactors (ORR type) with specialized facilities for individual nuclear research or isotope production programs might contribute more to the over-all program than a single ultra high flux multipurpose machine.

5. A number of reactor designs such as aqueous homogeneous, heterogeneous flux-trap, and molten salt reactors seem capable of achieving thermal neutron fluxes in the range of  $5 \times 10^{15}$  to  $10^{16}$  n/cm<sup>2</sup>(sec).

## INTRODUCTION

The seminar was opened with a consideration of persons other than those present who might be interested in attending subsequent meetings. J. A. Lane asked that Division Directors submit names of those who should receive the schedule of future seminars. Following this, Mr. Chetham-Strode was introduced as the first speaker.

## TRANSURANIC ISOTOPES\* - A. Chetham-Strode

I think it is particularly important in the discussion of high flux reactors to include a session on the transuranium isotopes. Indeed, the high flux reactor has been of very large interest to the transuranium program over the years and does occupy a very important place in its future serving in a sense as a mine, mill, and research laboratory in this field. It is probably not necessary to dwell in any detail on the reasons for our continued interest in the heavy elements, or the Commission's support of such activities. The research fields of fission, spontaneous fission, alpha decay, collective aspects of nuclear motion, and other of these topics, have become rather intimately associated with the field of heavy elements. Of more immediate practical importance, the ultimate plans for plutonium burners running under the economically advantageous conditions of high burn-up make it necessary that we acquire a more thorough knowledge of the technology, metallurgy, and general behavior of many of these heavy elements. The transuranium elements do, however, differ from the lighter elements in a rather unique way. That is, the first order of business in heavy element research must be to make these elements, to provide the materials on which the research is going to be carried out. In this line the early workers realized quite clearly that, although the cyclotron would probably skim off the cream and get the glory for new element production and identification, the ultimate program of production had to be tied to high flux piles. The reasons for this are pretty clear - one must start with large quantities of material and must have high fluxes for a long period of time. These are the conditions one can satisfy in a pile. There is another reason for the importance of pile-produced nuclides, a more fundamental one, in that they correspond in a sense in the heavy element region to the stable isotopes in the light element region. In general, the nuclides that one makes by prolonged irradiation of uranium or plutonium say, are those which are of long half-life and amenable to both isolation in quantity and to further work on them. One may cite as an example of the advantages of a long half-life the case of Cm 242. Cm 242 is a 162 day alpha emitter. People doing chemistry on this material find that when they centrifuge it in the bottom of a cone it boils itself back up in solution by its own alpha activity. So for chemical investigations, as well as for physical investigations, the pile-produced isotopes are the important ones. Finally, an analogy can be drawn between the production in reactors and the production of the lighter elements by the element build up process that took place presumably in the early stages of our universe and resulted, we suppose, at one time in rather large quantities of transuranium elements.

Weinberg: It's still going on people say, isn't that right?

\* This is a transcribed record of Mr. Chetham-Strode's talk and the discussion.

Chetham-Strods: Well, there's pretty good evidence for the presence of Cf 254 in  $10^{12}$  ton lots in supernova, or something like this.

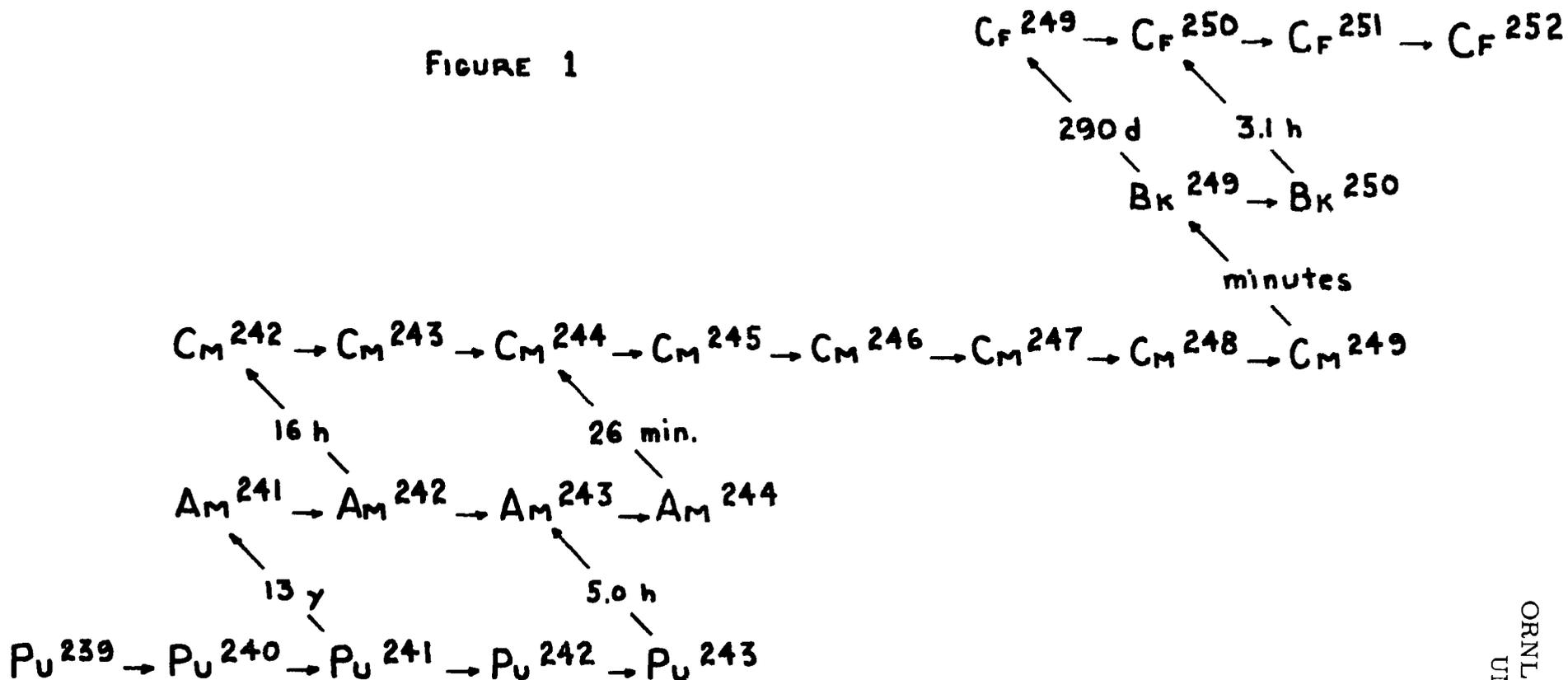
For practical reasons, the production of heavy elements and heavy isotopes in piles today has been extremely tiny. The amounts, for example, of berkelium, californium which have been isolated are orders of magnitude less than the amounts that have been separated by the Stable Isotopes Division in our own Laboratory. To give you an idea of where the situation stands now, we might detail a few of these quantities available. In the case of Pu 242, for example, the total known production in the United States, Britain, and we have no information on Russia of course, is in the range of 10-100 milligrams - something of this sort; Am 243, Cm 244, 10 milligrams - something on this order. Finally, in the case of some of the heavy curium isotopes, 246 for example, you have microgram quantities; and the elements 97 and 98, Bk 249 - and let's just lump the californium isotopes together - here quantities are considerably under a microgram.

[Editor's Note: It was pointed out in the subsequent discussion that the quantity of Pu 242 mentioned referred to separated material, whereas the other numbers were for totals produced in unseparated form in all known reactors (excluding the Russian reactors). It can be shown that the production of Pu 242 is of the order of a few hundred milligrams per day at the present time.]

To illustrate these rates of growth, let's just roughly sketch the rate of production of something like Cf 252 from plutonium at  $3 \times 10^{14}$  flux, being somewhere close to a maximum flux available now at long periods of time. In terms of atom per atom or approximately grams per gram, that is how many grams of californium we get per gram of plutonium we put in, we first begin to get  $10^{-10}$  grams of californium in the neighborhood of about a year and a half. This is continuous irradiation at  $3 \times 10^{14}$ . Before one is up to a microgram, you have gone in the neighborhood of six years at continuous irradiation. So this is an extremely steep rate of rise and this is why unless one deliberately goes to long exposures of plutonium the production of some of these heavier things is small. Well, notwithstanding the small amounts that are available and have been worked with, there has been a rather impressive collection of nuclear and chemical information obtained. It is nevertheless true to say that the field of heavy element research is very much in the production phase of its history and that its limitations now are principally those of quantities of isotopes available to work with. It is for this reason that the high flux reactor really is of basic importance to the future of this sort of work. Just exactly why this should be true is tied up of course with the production paths and the kinetics of pile production of these heavy isotopes.\* Thanks to some fifteen years of technology in production, we are no longer forced to start with U 238, but we can consider instead starting with Pu 239; and through successive captures with a very long neutron irradiation, the chain proceeds in a straightforward fashion to Pu 240, Pu 241, and Pu 242. At this point due to the relatively small cross sections for capturing Pu 242, most schemes envision stopping, reprocessing the Pu 242 and reinserting it in the reactor. The reasons for this are many. One obtains from an initial 100 grams of Pu 239 about

\* See Figure 1.

FIGURE 1



REACTION PATHS IN THE PRODUCTION OF Cf FROM Pu<sup>239</sup>

6 grams of Pu 242 at maximum - something like that; so Item A, one has only 6% of the original mass to handle; Item B, the heat production of course is down by several orders of magnitude. As the first approximation most of the fissionable things have been burned out certainly in quantities. Finally, by reprocessing at this or a close point, one can get away from the 90 some grams of fission product poisoning that have accumulated in the initial stage. From Pu 242 then, whether it is a separate irradiation, a repackaged capsule, or whether it is a continuation of the original process, the reaction path goes through a short-lived, five hour, beta emitter to Am 243. Again another capture and another short beta emission to Cm 244.

Blizard: How do you separate the 242 from the rest of the plutonium?

Chetham-Strode: You exploit the cross section differences. It turns out that one can burn out essentially 99% of the Pu 239, with similar high quantities of 240 and 241 burned out ....

Lane: The separation is from fission products, not plutonium.

Chetham-Strode: Well, from plutonium too. The irradiation burns it out and runs to the point where you have essentially pure Pu 242.

Blizard: So you have six grams of Pu 242 and a lot of fission products.

Chetham-Strode: Right.

Weinberg: What is the capture cross section of 242?

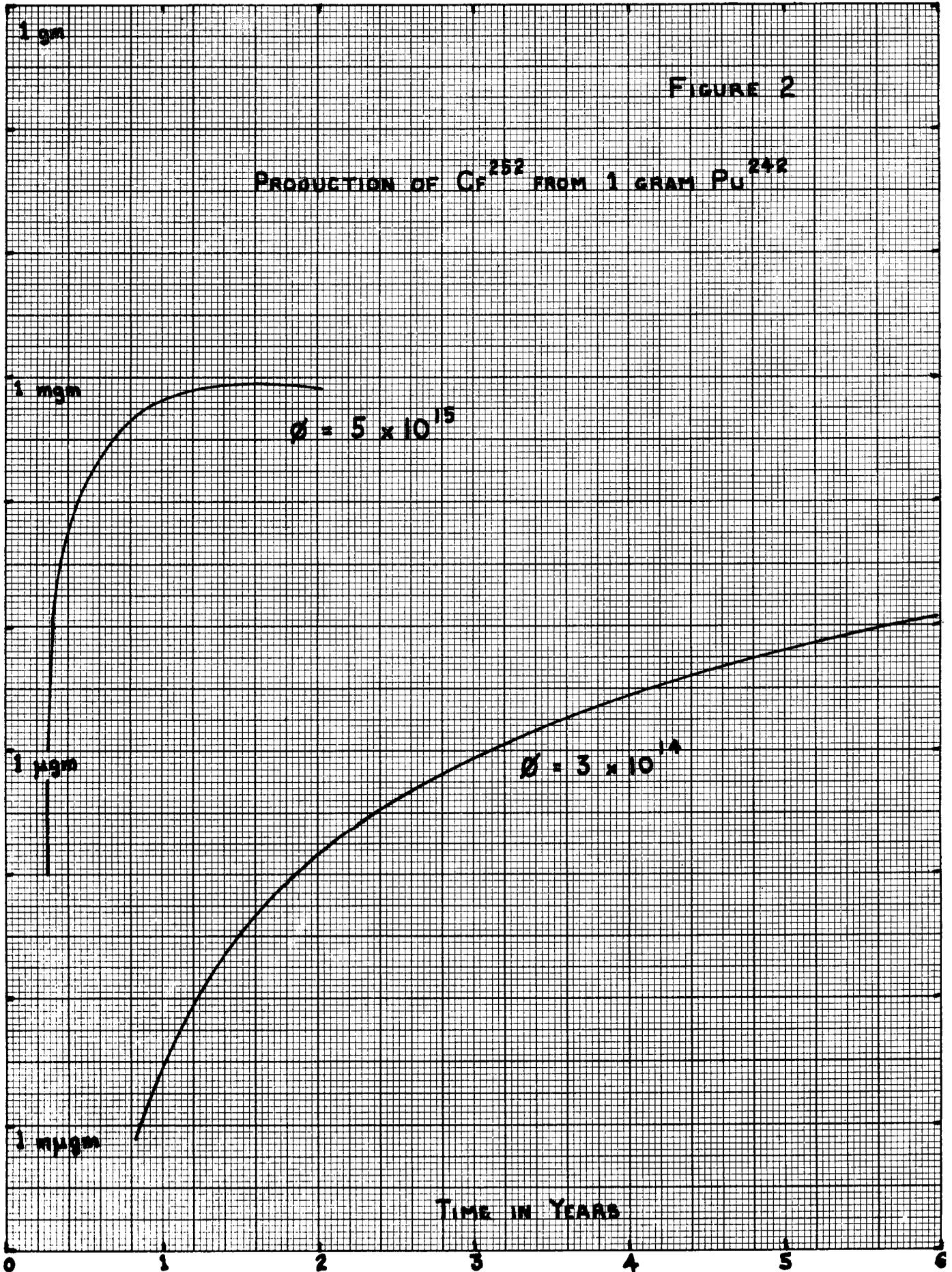
Chetham-Strode: Oh, it's 20 barns or something like that. It is small enough in comparison with the cross sections of the intermediate plutonium isotopes that this turns out to be a very favorable method of producing Pu 242.

Then follows a series of some four successive capture reactions through the chain of curium isotopes to the next beta emitting step, Cm 249, then to Bk 249, which has a very short half-life of the order of minutes. At Bk 249 there is the first case of important branching in this chain; namely, the decay to the next isotope, Cf 249 which proceeds by way of a 290 day beta emission. Bk 249 has a capture cross section of almost 1000 barns and a very sizeable fraction of the reactions go through Bk 250, which is again a short beta emitter, and to Cf 250. Two more captures then gets you up to the next logical stopping point, Cf 252. This is an alpha emitter with about a 2.2 year half-life. When one goes much beyond this, with fluxes that are available now you run into the problem of short half-lives, and Cf 252 logically then makes a good parameter to discuss the kinetics of this reaction. Taking the case of Cf 252, one goes through 13 captures in order to get there. So in the first approximation then, the initial production of Cf 252 at a given time, say a month or so, goes up as the 13th power of the flux in the reactor you are dealing with. Of course, after the irradiation has proceeded to some extent, this high rate of flux

dependence is reduced both by burn-up of lighter element materials and by the approach to equilibrium of some of the high cross section intermediates so that the actual flux dependence boils down after say a year or two to somewhere between second and sixth order; nevertheless, an extremely high dependence on total flux. It follows then that if one talks about going from present fluxes, say  $4\frac{1}{2} \times 10^{15}$  maximum in the MTR to ten times this much, you are talking about increase in the yields, although not a decrease in the time required to get to a given yield, of factors considerably in excess of that. For a given time if one takes, for example, one year, and one examines the products at the end of this, then you find that the amount you have produced at two fluxes differing by a factor of ten is up considerably as shown in Figure 2. This shows the amount of Cf 252, here again taken as a convenient parameter, produced as a function of time at two fluxes from one gram of Pu 242. Here one sees that at two years' irradiation period, the two fluxes produce amounts of californium differing by roughly four orders of magnitude. It is, of course, also true that if one produces say milligram amounts of Cf 252, one produces much larger amounts of some of the intermediate things. Berkelium stays in terms of absolute amount produced fairly close to the californium. It differs in general by less than an order of magnitude. Here in a typical case in which we started in with 100 grams of plutonium, we could expect in the high flux pile, at the end of two years, in the neighborhood of one to ten milligrams of californium - berkelium, an order of magnitude less - curium and americium, and here we are talking about some of the heavier curiums such as 246 and 248 in particular, both in the half gram scale - something of this sort. It is also interesting to point out that in order to get the same yield of californium in present piles, one finds out that you have to leave the samples in for in the neighborhood of 15 to 18 years.

Well, these arguments are really arguments based on a concept of how long it takes you to get to a given point. There is another area of interest in the high flux piles, which is not subject to this limitation, and this is the case in which one is interested in producing certain special heavy element isotopes by means of bypassing some of the short beta decay steps. The step at Pu 242 is a good example of this. Again, there is a five hour beta decay involved (Pu 243). The next step is a very long-lived Pu 244, with a half-life somewhere in the neighborhood of  $10^8$  years, I believe. It turns out that at  $5 \times 10^{15}$  flux (sort of an intermediate region in the range set for this discussion) about 2% of the total Pu 242 which disappears will essentially bypass the five hour decay step and go to the Pu 244. There are, as far as production implications are concerned, no great changes at these fluxes in this overall scheme and no large increases in yield. One should point out, however, that in fluxes an order of magnitude higher,  $10^{17}$  for example, one would begin to lose through the formation of the product, Bk<sup>251</sup>, which according to the rather sketchy systematics of neutron capture magnitudes and capture to fission ratios is expected to be rather highly fissionable.  $10^{15}$  to  $10^{16}$ , however, is a good solid range in which you have essentially maximized production of californium, here taken as an example.

There are in the very heavy element regions some other examples of the bypass of moderately short-lived decays, to give, in one case, the 70 day Cf 254,



which is the one the astrophysicists have been interested in lately. This is a nuclide which is of particular interest because it decays predominantly by spontaneous fission and might very well provide the most convenient isotope for spontaneous fission neutron sources and for spontaneous fission sources in general. Similarly, there is a one-year isomer of element 99, E 254, which is, as far as is known, the longest lived isotope of this element. These are other examples of the bypass of the short-lived decay steps.

One finally should mention the possibilities for varying isotope ratios over wide ranges by burn-out. The case of the curiums is a good example of this. The normal material which is produced at present with somewhere between two and four and one half years of radiation history in the MTR is made up about 95% of Cm 244, about 4% of Cm 246, and about 1% of Cm 245, with essentially negligible quantities of the heavier materials. It is possible by taking material of this composition and putting it into a  $2 \times 10^{15}$  flux, to essentially invert this ratio and come out with material in which the last of the beta stable curium, Cm 248, is the preponderant species, Cm 246 is next in line, and Cm 244 could be essentially burned out completely. Similar arguments apply to the californium isotopes, of which there are three or four beta stable species.

The last thing that I would like to mention is something that the  $10^{15}$  to  $10^{16}$  flux reactor probably will not do, and this is make new elements. The reasons for this are pretty straightforward. One should normally think that, given milligrams of californium, you could put them in a flux of this sort and proceed by analogous heating to go through the process of successive beta decay, successive neutron captures and beta decays, and eventually work up as high as one wanted to go. Two things intervene to prevent this. One is that, according to the well-developed systematics of decay energies, in the case of the fermium isotopes, element 100, one must go through quite a series of them out to Fm 259 before you get the first beta decay. After Fm 256 none of these isotopes are known. Figure 3 contains a section of the nuclide chart for this region. So the first thing is, one has to go through quite a number of captures before you reach the next beta emitters, starting out here with Cf 252, for example. The second and more significant reason is that this is also the region in which spontaneous fission begins to come in with a vengeance. In the case of Fm 254, for example, the spontaneous fission half-life (partial half-life for spontaneous fission, that is the half-life it would show if it decayed entirely by this mode) is about six tenths of a year. Fm 256 has a half-life in the order of three hours, and decays almost exclusively by spontaneous fission. If one wants to make a guess based on correlation of the known properties of spontaneous fissioning isotopes, one would guess that Fm 258 has a half-life for spontaneous fission of the order of minutes. This is a situation in which you begin to lose material too fast simply because the decays are too fast. It is also not possible to go through the intermediate steps, such as the einsteinium isotopes. We might consider E 253 and 254 which have a one year isomer. We might consider using this as a starting material and going up the odd mass elements, thus avoiding the spontaneous fission complication. What cuts you off, of course, is that as you get farther from stability, beta

FIGURE 3

				(M <sub>v</sub> <sup>259</sup> ) α	
F <sub>M</sub> <sup>254</sup> 3.4 h α SF	F <sub>M</sub> <sup>255</sup> 20 h α (SF) ε 100	F <sub>M</sub> <sup>256</sup> 3.1 h SF	(F <sub>M</sub> <sup>257</sup> ) α	(F <sub>M</sub> <sup>258</sup> ) SF	(F <sub>M</sub> <sup>259</sup> ) β <sup>-</sup>
E <sup>253</sup> 20 d α SF ~200	E <sup>254</sup> 37 h β <sup>-</sup> k ε 15	E <sup>255</sup> 30 d β <sup>-</sup> ~40	E <sup>256</sup> Short β <sup>-</sup>	(E <sup>257</sup> ) β <sup>-</sup>	
C <sub>F</sub> <sup>252</sup> 2.2 y α SF ~30	C <sub>F</sub> <sup>253</sup> 18 d β <sup>-</sup>	C <sub>F</sub> <sup>254</sup> 70 d SF ε 2			

PILE - PRODUCED ISOTOPES OF CF, E, AND FM

decay energies get larger and half-lives get shorter, and one winds up in the same fix. It is entirely possible that one could, starting with milligram quantities of californium, make detectable amounts, perhaps even enough for tracer experiments in such a pile of elements 101, 102, for example. It is, however, very unlikely that this method will be a means of producing weighable quantities of these heaviest elements.

I have emphasized throughout, the aspects relating to production for reasons which should be evident; namely because of the rather unique character of the heavy element field, they are all synthetic elements and to a very large extent production and research are equated under these conditions. It is also true that production of a new isotope, or in this case a new element, is also the absolutely necessary prerequisite for studying its properties. It seems then that the best way to describe the situation with respect to the heavy element field is in terms of the tremendous production capabilities of this high flux reactor. It is also pretty clear that if in the next ten or fifteen years we hope to have multi-milligram amounts of berkelium and californium, this is the program we are going to have to follow.

#### DISCUSSION

Lane: How much more is it worth in terms of increased rate of production of higher isotopes to have a  $10^{16}$  flux reactor, compared to say  $5 \times 10^{15}$ ?

Chetham-Strode: My feeling is, probably not very much. We are talking about now going from essentially Pu 242, which will in all probability have to be made in conventional reactors in any case.

Lane: Such as in long burn-up civilian power reactor fuel elements?

Chetham-Strode: Yes. You don't want to put whole fuel elements full of plutonium in the middle of a flux trap reactor, so one will start with some of the higher burn-up material. Now in two years we have reached essentially saturation at  $5 \times 10^{15}$  for the Cf 254. Also when we talk about this region in here we are talking about a very low flux dependence, so it means that going to  $10^{16}$  might get the same amount in one year, or something like that.

Lane: Well, wouldn't the saturation point be higher?

Chetham-Strode: No, because ultimately the product of flux-time comes into it, and these higher order reactions are all linear with respect to the flux-time product. So what one is saying is if you take twice the flux you get a curve that looks similar except for the time scale. We can reproduce these curves simply by changing the time scale.

Wollan: Is it worth very much to cut these times down so much? I notice here that in six years you can come within a factor of 10 to 20 or something like that of what you can get very soon in the high flux reactor. Six years is a short length of time.

Chetham-Strode: That's true.

Weinberg: Well, there is something that really puzzles me, because isn't it true that by and large work in heavy element chemistry now is limited more by the number of workers in the field rather than by the amount of material that is available, or am I wrong on that?

Chetham-Strode: Well, I don't think so. One of the things that contributes to essentially the slow pace of progress is that the techniques one has to use get to be very complicated as the amount of available material decreases. One has to, for example, think in terms of doing microchemical manipulations; it also means that there are whole ranges of investigations in chemistry and indeed in physics which are essentially excluded.

Weinberg: The thing that puzzles me a little bit is this - if you have say 100 times the flux that you now have, would you do different things than you are now doing or would you do the same things better and quicker?

Chetham-Strode: I think you would do both.

Weinberg: Could you give two examples, one where you would be doing the same thing better, and another example where you would be doing the same thing faster?

Chetham-Strode: Let's take first doing the same thing better, and the first thing that comes to mind is miscellaneous investigations of decay schemes of various nuclear properties which is pretty much standard for nuclear people these days. How well you can do this job depends upon what kind of high resolution instruments you can bring to bear on the subject. This in turn means how much of the given material in terms of activity you can produce. So, in general, producing more of the stuff faster gives you the opportunity of doing this sort of job better. One can also cite, of course, the production of a whole range of isotopes I haven't mentioned, the short-lived beta emitters, one and two steps out from these longer ones. These are things that you make in proportionately greater yield in a high flux reactor. In the case of chemistry, let us cite one very simple piece of information, and fundamental information - solubility measurements. This is something that one is hard-pressed to do on a microgram scale. It is possible, but one worries about interpretation of the results even if you do do it. This is the sort of thing one likes to do on a somewhat larger scale.

Weinberg: Well yes. On the other hand, one can argue on the question of inorganic chemistry of these materials. Why is the inorganic chemistry say of fermium, intrinsically more interesting than the inorganic chemistry say of europium which is one of the rarest of the elements?

Boyd: How can you assume they are going to be the same, even analogous?

Weinberg: No, it isn't that, but isn't it true that if you look below 92 on the periodic chart there are lots of real rare things whose inorganic chemistry is not known? Protactinium is a good example.

Boyd: This is nuclear chemistry, and we are interested in both.

Chetham-Strode: Even on a fundamental basis I think one can argue that one of the most interesting aspects of the chemistry of these heavy elements is comparison between the actinide and lanthanide series. It is one of the few ways we can get to bring more light to bear on the nature of these inner transitional series of elements.

Weinberg: Well, I can see that. I simply raised the question is there some intrinsic reason why the chemistry of these things is more interesting say than the chemistry of things below 92 for which we have not made any dent on the chemistry, and protactinium was suggested as an example where we've made little dent. I suppose there are other things also. We know more about technetium than we do hafnium for example.

Boyd: Let's review my argument. My argument is that you don't really know too much about the periodic table, and you don't know too much about the Aufbau principle, and you don't know that these 6D, 7S, 8P electrons, etc., are going to go into these shells quite in the way we thought they did. This is always true. The really fundamental thing about chemistry is knowing how these electrons go into their orbits. This is what you are going to find out.

Weinberg: Not so much by measuring the solubility.

Boyd: Sure, the chemical properties do reflect this.

Weinberg: Yes, but there are more direct ways of finding out whether it is 4F, 5F, or 6D.

Boyd: There is still a lot of controversy on this.

Weinberg: I ask this very general question, why is heavy element chemistry basically in a different situation say than astronomy? What bearing really does it have on things? They both are expensive, they both require a great big instrument, and basically, aside from the heavy element chemists in one case or the astronomers in the other case, who cares? Let me say I'm raising these arguments a little bit facetiously, sure, and yet these will have to be the issues that one kind of makes one's peace with when one asks for the \$100 million.

Boyd: I'd like to answer that argument. If all of our predecessors in science had asked themselves, "who cares," I don't think we would have had uranium fission today; I don't think we would have the electromagnetic theory; I don't think we would have all the great things we have.

Weinberg: Well, but there is a difference. Electromagnetic theory was worked out by one scientist. If you read about how Hertz begged, borrowed, and stole stuff, and didn't require the German Government to make an appropriation of \$100 million to back his first Hertzian oscillators, you see that there is a difference.

Boyd: I think that is the great fallacy today that you ask, "who cares."

Weinberg: Don't misunderstand, I think it is real good. The only real basic issue that I think is going to keep coming up over and over again throughout our discussions is why are these issues in science the ones that we really want to pursue.

Taylor: We are scientists.

Chetham-Strode: I wonder if that isn't the point. I wonder if it is not a case of "why should we do this," and I think perhaps the answer is that this is just exactly the sort of thing that the AEC and its facilities can do.

Blizard: There are lots of things that they can do, and I am sure they would do it if you could do it for nothing. They would also do it for \$10 or \$1000, but the question is whether we want to build an extremely difficult reactor in order to do these things.

Weinberg: There is the question of the rates at which you want to do it. Now, Glenn Seaborg has a very valid point because he argues as follows - he says that if you argue on the basis of the usefulness or even the advance in human knowledge, then one certainly can argue that advances in this area are intrinsically as interesting to as large a group of people, say as our advances in hyperon physics. He is right! If you are willing to spend \$100 million for Panofsky's electron accelerator, then we should be willing to spend \$100 million for this gadget.

Blizard: Well, what qualitatively is different about the chemistry of the very heavy elements as opposed to the chemistry of elements up to 92, for example. When you talk about hyperons this is quite different from nucleons and electrons.

Boyd: Well, you're right at the end of the actinide series. Okay, the next element, is it going to be say like protactinium, or is it going to be like something else, such as hafnium?

Blizard: Is there a theory of electron construction which will be straightened out by the answer to this if we get the next isotope?

Taylor: We just heard that we don't get to the next one.

Chetham-Strode: Let me make myself clear on this. The way one goes up to the next element and the ones following this is tied up in a very intimate way with a high flux reactor in making lots of californium, for example. The way one does this is take and use this material as target material for heavy ion bombardment.

Lane: I see, but you can't do it in a reactor?

Chetham-Strode: Not at these fluxes.

Boyd: Let me put this real dramatically. The reason why these are important is that these are the nuclear sputniks. You are going to put one of these on

the face of the earth, and by doing this you really show your technology. If you orbit one of these elements, that shows you have achieved a certain position.

Wollan: Do you think the Russians will ever orbit one of these sputniks?

Boyd: All right, but if the Russians should suddenly announce element 102, and really prove it to us, this would be a shock to many of us as severe as the sputnik. It means they had to have a certain apparatus behind them to do it. That is a demonstrative argument.

Rupp: Isn't the chemistry on this a means to an end? Aren't you trying to get purer transuranics here for other things to be studied, not just the chemistry?

Chetham-Strode: That's right.

Rupp: It is for physicists to work on, and in order to do this you have to understand the chemistry to make the separation.

Chetham-Strode: At the risk of being thrown out of the chemists' union, I think the physical implications are at least as great, if not greater, and in his case one can make a really good case of unique character of very heavy things if on no other grounds than that of spontaneous fission.

Rupp: Sure, if the physicists are really interested in these things, the chemists separate them.

Boyd: You are at the end of the actinide series, and something new may happen.

Wollan: I don't understand yet. Can you get to the same place in a reasonable length of time, say at the MTR, as against the high flux reactor? I see that you have a very high rate of build-up here, what about if I wait fifteen years, can I come to the same technical situation fifteen years hence, against a few years?

Chetham-Strode: Yes, provided one restricts oneself to small sample arguments. What you come into is a case of reactors paced to do the initial burn-outs, reactors paced to do the intermediate steps, and the final steps.

Weinberg: Well, suppose you built three MTRs just for that purpose?

Chetham-Strode: Then you are presumably at the same point as far as ultimate production goes, you have lost yourself maybe ten years.

Weinberg: Well, in that sense there is a qualitative difference between this and either astronomy or hyperon physics, because here you can exchange time for money.

Chetham-Strode: Well, provided one restricts oneself to considering only the production of a given isotope then you lose all the rest of these things, such

as the possibility of, within reasonable periods of time, being able to make Pu 244, for example. This is something which would be quite easy with this reactor.

Wollan: You still have to pit time against economy. I think your original argument was good, especially if you consider the fact that sometime later we will get this knowledge, and fifteen years is a pretty short time in a research program, and after all you won't have this reactor in less than five years probably.

Lane: But I don't know whether they are doing any irradiating now to get these things in present reactors, are they?

Chetham-Strode: No, not now.

Weinberg: Well, that of course is an administrative faux pas of tremendous magnitude. The Laboratory is going to present tomorrow in Washington a proposal for starting this program. It is clearly an error of large proportion that at the time the MTR started they didn't start cooking these things.

Chetham-Strode: Not in large enough quantities.

Boyd: Can I ask one scientific kind of question? Since you pointed out that you can't get beyond Cf 254, E 254 has an isomer and a long half-life, now why doesn't E 256 also have an isomer, and that will give you the bridge then to get up to these new elements?

Chetham-Strode: You don't go quite far enough; the thing that gets you first of all is the even mass, even Z elements with short spontaneous fission half-lives. Even supposing that there is another long isomer E 256 - and there might be - supposing there is, this still doesn't get you past the real hooker here which is the Fm 258 in this chain. Now then, suppose just for the sake of argument we do get past that, and we made the element 101. Well, by analogy with most of the rest of the elements we expected to have one stable isotope, and probably the next one is a beta emitter, and this gets us to element 102 pretty quickly. 102 is again an even one, and if one makes the usual sort of guesses and extrapolations from alpha decay and this sort of thing, you are back in the same fix - you run through perhaps four or six stable isotopes before you get to the next decay. Then the spontaneous fission competition is even worse, and in view of the fact that the Swedes, British, and Argonne people already claim 102, things look pretty black for 103 for example. No, I think it is quite possible that one can make tracer quantities of these materials (even mendelevium) and they will be of considerable interest in themselves because they will be isotopes that are not available in cyclotron programs, for example, which by nature gives neutron deficient products.

Snell: You think that 102 is just about as far as you can get by heavy ion build-up, that neutron emission and fission will keep you from going further?

Chetham-Strode: No, No I don't. I think the thing that is going to set the limit is basically the half-lives involved, and if one were going to make a

guess you would say that with sufficiently sophisticated techniques, the half-life limit would be set roughly by the resolving time of electronic equipment - somewhere in the neighborhood of  $10^8$  seconds. If one makes any extrapolation of the known spontaneous fission half-lives and their mass dependence, you come to the conclusion that you are going to run out of steam, perhaps at element 108, maybe 110, 112, somewhere in this region.

Snell: This seems to leave a nice little margin to work on.

Chetham-Strode: The heavy ion people aren't out of business yet.

Weinberg: I would like to raise once more this point that Lane raised; namely  $10^{15}$  is there anyway in which one can get a handle on how useful say  $2 \times 10^{15}$  is against  $8 \times 10^{15}$  as far as your business is concerned?

Chetham-Strode: I think this is very largely an arbitrary decision. It is involved with how long you are willing to wait to get to a given level of a given product.

Wollan: You have to set down a time schedule and take a look at it.

Chetham-Strode: On the other hand, one has to realize that to feed into a reactor of this sort one gets involved with perhaps a year, maybe a year and one half of burn-out processing time on starting materials, and it clearly doesn't make sense to run at such a high flux that your time from there on is small compared to this. I suspect if one optimizes, it might come out by some sort of reasoning that you would shoot for something about equal times. That is one of the reasons why this  $5 \times 10^{15}$  figure which gives you two years to achieve saturation .....

Lane: Yes, but how long would it take to get reasonably close to that number at  $3 \times 10^{15}$  flux? Since each  $10^{15}$  unit requires removing 100 megawatts of heat, and 100 megawatts costs say \$10 million, it is desirable to find the minimum acceptable flux.

Chetham-Strode: I think the decision probably isn't going to be made with regard to the production of a given isotope. I think it is rather going to be made with regard to other factors which were mentioned, such as the beta decay steps that you are trying to get around.

Rupp: Well, it's bound up in the relation between the cross section of the target atom that you are bombarding, the product atom of some particular couple that you are looking at, and the decay of both of those, and this governs it and you come right up to a maximum on the thing. You shouldn't go any further than that. I'll point out most of the things that I've looked at qualitatively, and it actually doesn't seem like much more than  $10^{15}$  is useful for ordinary isotope work.

Boyd: Why should it be any different from ordinary than this?

Rupp: Well, I shouldn't have implied that it is. I'm just saying that is what I looked at.

Snell: Just one more comment on the heavy ion bombardment. This looks as if it would work nicely with the heavy ion cyclotron. The two perhaps should be put together.

Weinberg: I might say that Seaborg is trying to get a heavy ion cyclotron now.

Chetham-Strode: It is quite possible, and should also be realized, that by the time a heavy ion cyclotron comes to our Laboratory, there is not going to be a lot of very easy gravy left to pick up. Berkeley people, of course, have their linear heavy ion accelerator.

Weinberg: Yes, but the currents you can get would be much higher in the cyclotron.

Chetham-Strode: That's true. This in general is apt to mean that allowing for the delay in time and the normal increase of cyclotron currents in time, one does a lot of things much better.

Taylor: What is this argument about resolving time, limiting ultimately the last step? It seems to me that you have got to aim your accelerator down the snout of the reactor.

Chetham-Strode: I don't follow.

Taylor: You said that in deciding how far you could go toward short half-lives you would be limited by electronics perhaps along with kind of instruments, rather than by chemistry. You certainly don't want to do two chemical steps on short-lived things, so you will have to make all your transformations in one place then.

Chetham-Strode: Well, one normally thinks about this in terms of, for example, bombarding californium with first carbons, then oxygens, then neons, etc., so that you use heavier and heavier bombarding particles. So really this is sort of a two stage operation.

Boyd: Ultimately all new element discoveries will be based on physical rather than chemical evidence.

Lane: If there are no other questions, perhaps we can hear from Mr. Rupp on "Other Isotopes."

#### OTHER ISOTOPES - A. F. Rupp

One of the things I noted in discussing regular radioisotope production, contrasted with rather specialized types, is that we are dealing with a lot of target atoms in radioisotope production; and we have, of course, a wide range of cross sections we have to deal with and isotopic composition of the elements, of course, differing. All of these factors have to be taken into account in considering what is the best way to produce a certain radioisotope.

Since the reactor production of radioisotopes is primarily concerned with the reaction between neutrons and a great variety of target atoms, the concentration of the reactant common to all of the production reactions - the neutrons - is of the greatest importance in producing high specific activity radioisotopes. The isotopic activation cross-sections of the various target atoms are, of course, fixed values, and in only a comparatively few cases can one gain a factor of 10 by concentrating the target atoms by isotopic separation. Since in neutron-gamma reactions (which are by far the most common) the product atom is the same chemical species as the target atom and therefore cannot be separated chemically, the best way, and almost the only way, to increase the concentration of the product atoms is to increase the neutron flux. One other possible way to greatly increase the specific activity of nuclides produced only by  $(n, \gamma)$  reaction is to isotopically separate the radioactive product atoms from the target atoms in machines such as our electromagnetic separators. However, little is known in this field, and in any case, the method would not be applicable to fairly short-lived radioisotopes because of the time required to carry out such a complicated operation.

It therefore seems clear that if we are interested in producing all the possible radioisotopes in as nearly "carrier-free" form as possible, an ultra high flux reactor is required. But one would be justified in asking, "Why do we need pure, carrier-free radioisotopes?" Unfortunately, the best answer is the one that sounds the weakest: We intuitively know that radioisotopes will become more versatile and useful as their isotopic and chemical purity increases. There are many cases that could be cited to support this view, but I believe that the recitation of numerous specific cases is unnecessary. It is significant that in dealing with radioisotope customers, particularly those engaged in research work, we have seldom, if ever, encountered one who did not want higher specific activity material. In biological research, the ideal situation is one in which adequate tracer is added without disturbing the delicate chemical balance of living organisms. The same holds true in many studies of chemical equilibria. Physicists want radioactive sources of high intensity, but with as little substance as possible. Radiographers want high intensity point sources to improve definition. If we simply want to gain more knowledge about the radioactive nuclides themselves - their radiations, cross-sections, magnetic moments, spin, etc. - it is highly desirable to have the radioisotopes chemically and isotopically pure.

In general all radioisotope production would be benefited by having a higher flux, obviously because you could do things faster by having this higher flux. One of the things to mention would be gaining very high saturation values with short-lived material, where you essentially have to go to equilibrium where the atoms are decaying at the same rate they are being produced, and of course if you are producing the higher rates this value becomes higher in proportion to the flux. Some of these isotopes are listed in Table I. The first one is  $\text{Cu } 64$ , which has a half-life of 12.8 hours and a cross section of about 4.1 barns. It might be of interest to note at this point that the theoretical specific activity of isotopes like this are all pretty high, being around  $10^8$  curies per gram. Now with  $10^{14}$  flux reactors we will get only about 73 curies per gram, and at  $10^{15}$  we get about 263 curies per gram, based on

Table I - Examples of Short-Lived Isotopes

Nuclide	Cu-64	Ga-72	I-130
T-1/2	12.82 hours	14.2 hours	12.6 hours
Target Atom Abundance	Cu-63 (69.1%)	Ga-71 (39.8%)	I-129**
Isotopic $O_a$ , Barns	4.1	4.9	11
Theoretical Specific Activity	$3.83 \times 10^6$ c/g	$3.04 \times 10^6$ c/g	$1.87 \times 10^6$ c/g
* Activity at Maximum, $10^{14}$ , c/g	72	45	73
Activity at Maximum, $10^{15}$ , c/g	725	456	727
Time for Maximum, $10^{15}$	6.4 days	6.76 days	5.48 days
Nuclide	K-42	Na-24	Au-198
T-1/2	12.47 hours	15 hours	2.7 days
Target Atom Abundance	K-41 (6.91%)	Na-23 (100%)	Au-197 (100%)
Isotopic $O_a$ , Barns	1.17	0.53	98
Theoretical Specific Activity	$6.01 \times 10^6$ c/g	$8.69 \times 10^6$ c/g	$2.44 \times 10^5$ c/g
* Activity at Maximum, $10^{14}$ , c/g	3.4	37	167
Activity at Maximum, $10^{15}$ , c/g	34	374	815
Time for Maximum, $10^{15}$	7.09 days	9 days	2.28 days

\* Not true specific activity, but amount of radioisotope per gram of starting material.

\*\* I-129 is very long-lived radioactive nuclide obtained by U-f.

grams of material that you originally put into the reactor. This is not the true specific activity since it is based on the target that you put in. The time for maximum activity in this case is around 6.3 days.

Taylor: You could have gotten 263 curies per gram by irradiating for a little longer, couldn't you?

Rupp: No. Now let's select something with a different cross section. Take Na 24 which has a half-life of 15 hours and a cross section of only about 0.53 barns. In this case for  $10^{14}$  flux you get about 37 curies per gram, and for  $10^{15}$  flux essentially ten times as much or about 374 curies per gram. In comparison to these isotopes, with Au 198, with a 2.7 day half-life and a cross-section of 98 barns but also with a large cross-section for the radioactive product atom, we get about 167 curies per gram at  $10^{14}$  flux and only 5 times the activity at 10 times the flux. In this case the reduction in activity is due mainly to burn-up of product atom, Au 198, which has a 26,000 barn cross-section. Also in the case of the europium isotopes, where Eu 151, the parent atom has a cross-section of 7000 barns, the specific activity per gram starting material is the same at  $10^{15}$  flux as at  $10^{14}$  flux due to the burn-up of the parent. In other words you don't gain with such isotopes by going to higher fluxes. The thing I want to point out here is that this fact is not necessarily bad since the actual specific activity of a high cross-section isotope is considerably higher than that calculated on the basis of original target material (if the radioactive product atom doesn't have too high a cross-section). Although these figures might be fairly good on sodium, for example, for the specific activity that we actually find when we take it out of the reactor, the value given for gold would be way off. In other words, the specific activity would be very much higher than this (if the product atom isn't burned).

Question: You mean you don't put pure gold in the reactor?

Rupp: You put in pure gold, but you burn up the target material since it has a very high cross section.

Weinberg: Don't you burn it to a gold isotope that isn't radioactive? Isn't that the point?

Rupp: No. You burn it to radioactive gold, which decays to mercury. (Also, the radioactive gold is burned up because of its high cross-section.)

Weinberg: But then the mercury captures and goes to gold.

Rupp: No, it goes to another mercury, so what you can do then is separate them chemically and have a very high specific activity.

All I wanted to point out in this one case is that you can get a very much higher saturation value by burning out target atoms. To summarize the situation for short-lived isotopes since you can get very much higher saturation values it appears that an ultra high flux reactor for many of these things would be very good. You can actually get, in most cases, a factor of 10 or so higher specific activity, unless you run into materials with high cross sections.

The second item to be mentioned is the speeding up production of long-lived materials shown in Table II where irradiation times are too long in ordinary

TABLE II

Examples of Long-Lived Isotopes

Nuclide	Ba <sup>133</sup>	Tl <sup>204</sup>	Ni <sup>63</sup>	Co <sup>60</sup>
$T_{\frac{1}{2}}$	7.2 years	3 years	80 years	5.3 years
Target	Ba <sup>132</sup> (0.097%)	Tl <sup>203</sup> (29.5%)	Ni <sup>62</sup> (3.66%)	Co <sup>59</sup> (100%)
Isotopic $\sigma_a$ , Barns	7	11	15	34
Time for Max., $10^{14}$	19.8 years	9.6 years	44 years	7.8 years
Time for Max., $10^{15}$	6.6 years	3.38 years	18.5 years	1.6 years

reactors. This would be somewhat the same argument that has been made for the transuranics. I might point out some of the differences in time that you get here for the maximum production of materials that are fairly long-lived, of the order of a few years. Now in the case of Ba 133, which has a half-life of 7.2 years and a cross section of 7 barns, the time at  $10^{14}$  flux for maximum production would be 19.8 years and at  $10^{15}$  maximum would occur at 3.38 years. For Ni 63, which has a half-life of 80 years and a cross section of 15 barns, the maximum time would be about 44 years to get to a point where there is no use of keeping it in there any longer. This will occur in 18-1/2 years at  $10^{15}$ . Co 60, about which I will speak a little more later, has a half-life of 5.3 years and a cross section of 34 barns. We have a maximum at  $10^{14}$  or 7.8 years. This occurs in only 1.6 years at  $10^{15}$  flux. Now, this again is tied up with the burn-out of the target which usually isn't of much interest to us in the low flux reactors that we have been using, but becomes a very important factor in a high flux reactor. I might note that for the production of some of the very long-lived things, such as C 14 which is produced by (n,p) reaction, and 105 year Cl 36, and things like very large quantities of Co 60, an ultra high flux reactor would not be required. It would be a great waste to even consider using an ultra high flux reactor for that kind of production.

Boyd: You are not saving any time, but you would be producing much more in a given time.

Rupp: Yes, but you can do it a lot cheaper. You can use neutrons that are tremendously cheaper in other reactors and get essentially the same results since it is a question of total neutrons, rather than neutron flux, so you would never consider doing that in a reactor of this kind.

Boyd: You are saying for a radioisotope that doesn't decay appreciably you should use a low flux reactor?

Rupp: That's right. An ultra high flux reactor is not of any use for long-lived materials, and it is of limited use for even moderate-lived materials.

Boyd: What do you do now? Do you load that beryllium nitride in the outside of a reactor when you want to produce it?

Rupp: Yes. It can be produced in any large reactor.

The third item I would like to discuss is one that has been mentioned before and that is the burning out of undesired atoms that are isotopic with radioisotope products. In some ways this is the most interesting thing about having a high flux reactor. An example of this would be Co 60, which achieves a maximum production per unit weight of starting target material in 1.6 years in the  $10^{15}$  flux. The theoretical specific activity of Co 60 is 1140 curies per gram. The maximum activity per unit weight of starting target at  $10^{15}$  flux is 692 curies per gram; and, since the target is partially burned-out, the actual specific activity is higher than this. At  $10^{15}$  flux, it would require about 5 years to burn 99% of the original target, so it can be seen that a fairly close approach to the theoretical specific activity can be made in 2-3 years' irradiation. You can keep burning out the remainder of the Co 59 as you continue the irradiation and approach, of course, the limiting value of specific activity. So, in a case of this kind you see it would be very

practical in a  $10^{15}$  flux reactor to get in 3 years a sample, that is a thin sample, that would have 90% or better of the theoretical specific activity of Co 60, and samples of this kind would be of very great interest in producing highly concentrated sources to be used for a lot of radiographic work or research work. So you actually can burn out the remaining atoms, and that I think is one of the more interesting things about the high flux reactor. To take a case in which this effect is very pronounced is 13-year Eu 152, which has a companion of 16-year Eu 154. Europiums are notable, all of them, that they have terrific cross sections, in the order of thousands of barns. It is interesting to note that in a  $10^{15}$  flux reactor the maximum production occurs in only 14 days. In other words, the burn-up of the radioactive atoms is so great there is just not much use of leaving it in longer than 14 days. This indicates that you can use this to burn Eu 152, 154 out of fission product Eu 155, for example. We have a problem of fission product Eu 155, which has a soft gamma ray, the type that is very hard to obtain, but it is contaminated with secondary-produced Eu 152, 154 which occurs because of neutron capture of inactive fission products produced in a fission chain. There are other instances when you could use the reactor to burn impurities or undesired nuclides out of preparations, and if you use fluxes much below  $10^{15}$  this simply takes too long. But at  $10^{15}$ , in general, it seems that things like this are practical.

Wollan: Don't you have to pit the cost of this against the manufacture of radioactive materials at lower fluxes and then sending them through a calutron?

Rupp: That's right.

Cole: In regard to Co 60, you stated that this material might be better produced in lower flux reactors, but then you did point out where there were certain advantages for irradiating cobalt. Were you thinking of this latter aspect in terms of perhaps a small total flux?

Rupp: I should have made that clear. In the case of producing very large amounts, that is, thousands and thousands of curies of Co 60 such as we produce now in Savannah River and Hanford, it would not be practical to do this in a reactor of ultra high flux, or the ORR or MTR for that matter. It is wasteful because the neutrons are so very expensive. But for producing specialized sources of all kinds you could certainly use a reactor of this kind to produce the very high specific activities which would be difficult to get any other way. In the case of cobalt, I think I could almost say this by inspection and from experience that it would be very much cheaper to make the ultra high specific activity cobalt this way than in the calutron because all we have to do is make a thin film of cobalt, put it in there, and just leave it for a year or so.

Wollan: Are you amortizing \$100 million on that statement?

Rupp: No, it depends on what kind of ultra high flux reactor we build, of course.

Lane: Are you suggesting an ultra high flux of  $10^{15}$  as distinct, I suppose, from the super high flux of  $5 \times 10^{15}$ ?

Rupp: No, I was just using the term ultra high flux reactor to distinguish it from any high flux reactor that we have at the present time.

Lane: Well, you made the statement that you weren't interested too much in fluxes over  $10^{15}$ .

Rupp: That's right.

Weinberg: Why was that now?

Rupp: Well, if you take a number of cases and go through them, you find that at  $10^{15}$  flux, it is possible to get reasonable burn-out or a high specific activity in times of the order of a year or so. You don't find many cases where you really need more flux than that to accomplish reasonable burn-out or to produce reasonable activities. Indeed, in some cases if you don't want to separate the material afterward, and you're interested in both production and high specific activity, you don't want too high a flux. As a matter of fact, there is a good case for irradiating these targets successively in two or three different fluxes.

Lane: Well, take the case of 99% burn-out in cobalt - 5 years. That's a little too long isn't it?

Rupp: That is a little bit too long, but if it were five times as much it wouldn't even be considered.

Winters: When you are talking there, you're talking about actual delivered flux though?

Rupp: Yes, delivered flux in the reactor.

Lane: Well, but the point is that these samples aren't very big massive samples, they are not going to depress the flux tremendously are they?

Rupp: No, really the type that we are thinking about here are very thin films. You wouldn't want to use a reactor of this kind for just ...

Weinberg: Does this mean that Lane's idea of getting a very high localized flux would be very useful for you?

Rupp: Yes, it would.

Weinberg: The same type of reactor would also be useful for Chetham-Strode's work although he spoke of somewhat larger samples.

Winters: But he wouldn't put it into the reactor in the high flux region until after he got it boiled down to 6 grams.

Rupp: You have a heating problem there though.

Winters: For your first stage it doesn't pay to have a real high flux reactor.

Boyd: Would this nuclear incinerator burn some of the waste products?

Rupp: Yes, one thing I wanted to mention is that we don't really know very much about the cross section of some of these things, and we do want to put things like Sr 90 and the like into the ORR and try to find out a little bit more about their properties. I mentioned one case where it would be useful with fission products in burning out europium from Eu 155, and I am sure there are other cases like that.

Boyd: Like operation bootstraps, you burn out the strontium, but you are producing fission products of course in order to get neutrons. Do you make anything this way or not?

Rupp: Oh, you cannot burn up Sr 90 as far as we know and get anywhere on it as far as quantities are concerned. The only way you could do that is if you had a thermonuclear reactor.

Now, another item to be mentioned deals with secondary reactions which go up with the square of the flux. Another item would be in conserving isotopically-enriched target material. Some of these target materials are really quite expensive. We like to have things like Ca 47, which is a gamma emitting calcium isotope, which would be useful if we had much of it. It is now made by a (pp,n) reaction on Ca 48 - a very expensive business. In fact, we just don't have any way of making any decent amounts of Ca 47. In a case like this, you could use the Ca 46 (n, $\gamma$ ) reaction, but the natural abundance of Ca 46 is 0.003%, so that we have a very, very small amount of target material, and it is extremely expensive and we want to conserve the material. There are other cases where we need, and do use, enriched material, such as Cr 51 by using enriched Cr 50, I 130 by using fission product I 129, Fe 55 and Fe 59 using enriched Fe 54 and Fe 58, and Sr 85 using enriched Sr 84. Again Sr 85 is a strontium isotope that has a gamma ray, so both of these are very important because the other isotopes, like calcium, are soft beta emitters, and are hard to follow through in biological systems and the same way with strontium. There is a lot of work waiting to be done with both calcium and strontium gamma emitters. If high fluxes were available, we could use a hundredth or maybe even a thousandth as much target material and get the same production as at lower fluxes, which would be very good of course. I am sure this would be economical unless the reactor were really terrifically expensive. Many radioisotopes are now made carrier-free by an expensive (n,p) reaction, plus chemical separation processes, and they could be produced by (n, $\gamma$ ) reaction with an acceptable specific activity. Now in this case I might cite Ca 45, which is made by (n,p) with Sc 45. We are not able to get a very high yield; the target material is expensive; and the chemical separation is pretty complicated. Right now, using an MTR flux and Ca 44 isotopically separated, we can get preparations that are within a factor of 10 in specific activity compared to the Ca 45 that is made from scandium. This is because that even if you make a very pure separation carrier-free, unless you baby it along every step of the way you are going to get casual calcium contamination in it. So with another factor of 10 in flux we

could prove even to the people who do research with teeth (they are the most particular on what kind of calcium they get) that it would just be nonsense to try to make it from scandium because using Ca <sup>44</sup> in an ultra high flux reactor you could make very, very good Ca <sup>45</sup>. The same thing applies to P 32 which is made from sulfur. P 32 is used in large quantity, and is widely used in medicine. Right now we feel that with a 10<sup>17</sup> reactor, we could make it directly by irradiating phosphorus to more than 100 curies per gram, and that is quite acceptable for most medical uses. That would bring down the cost of P 32 tremendously. Also, in the past, we have used 1 ft. long cans filled with sulfur for making one curie in the graphite reactor, and we are prepared to do it in the ORR with some small tubes with about 100 grams. So even with the (n,p) reaction you can go way down with the target material, and this cuts down the processing expense a whole lot. The same thing would apply to S 35 which is made from chlorine, and any other (n,p) product, that you could use the (n,γ) reaction and get the same thing. You could do it a lot cheaper and cut the cost of the material.

Well, that's about all the specific cases I wanted to bring up. However, I might mention one thing here. In talking about an ultra high flux reactor I feel that there are several kinds of reactors within that classification that should be studied. One is a reactor that is designed for radioisotope work exclusively, and I feel like a reactor of this kind would be very, very much cheaper than one that is designed to do all kinds of work. Anybody who has worked with reactors, built reactors, designed reactors, and knows where a lot of the cost goes, knows that it goes into the appurtenances of a reactor designed to do many different jobs. So I think that if it turns out that a reactor is not feasible for general research, that is from the standpoint of cost, I would like to say here and now that we shouldn't throw the idea of a high flux reactor out. It should then be considered for radioisotope work or for transuranic work or things of that type, and it might cost only one-tenth as much as the other type reactor in my opinion.

Lane: Well, you can reverse that argument and say that if you are building the reactor for basic research, then you get it free for isotope production.

Rupp: Of course, this argument has always been used in the past, and this has never (in dollars) been true. I don't accept that argument at all.

Cole: I have come more and more to the conclusion that when you build reactors you should build them for a specific job.

Rupp: Absolutely, and that's the reason I feel that if it looks like you can't afford to build a general research reactor, I'd like to see us go ahead and study a radioisotope reactor for once.

Lane: It seems to me that it is going to be very difficult to put a dollar value on high flux neutrons for basic research. I think we are going to have to muster all of these arguments - the research, as well as the radioisotopes.

Rupp: Well, mind you, I'm not urging against that point, and I think the first time around that's alright. But if it turns out that having a general research

reactor of such high flux is out of the question, let's go back and look at it for a cheaper reactor for radioisotope work.

Lane: Well, would you be able to put a dollar value on how much more it would be worth for these high specific activity isotopes and high saturation isotopes? How much, based on your present business, would be the dollar sale of these isotopes per year on the material that you could make with high fluxes that you can't make now? Any idea?

Rupp: No, not right at the present time.

Lane: Could you derive that sort of number?

Rupp: I would have to put a lot of real crystal-balling in that, and if somebody pinned you down you would have a hard time bringing up enough supporting arguments.

Lane: We may have to stick our necks out on that sort of basis to get such a reactor.

Rupp: Well, let me put it this way. I think that we can come closer to putting a dollar value on it than anybody else can. I feel that there has not been one radioisotope reactor built in the entire world that I know of - there have been I don't know how many reactors built for power, for neutrons - there has never in the whole world been a reactor built for radioisotopes.

Comment: Now is no time to start! (Laughter)

Rupp: No, on the contrary, I think it's time to build one for radioisotope work because this is one end of the nuclear energy business that has really proved itself, and there has never been a reactor designed and built for radioisotope work.

Lane: Yes there has, in Buffalo.

Rupp: Oh no, that's not a radioisotope reactor, that's a joke. You mean this cobalt reactor?

Lane: They are making as much from the cobalt as the steam!

Rupp: No, they may not build it until there is a guaranteed market for the cobalt.

Lane: Well, everybody that wants to build a pure radioisotope reactor can't justify the market, so it is never built.

Cole: I think that your requirements are not essentially different from those for the transuranic elements.

Boyd: I do not think there is any merit in trying to put a dollar value on higher isotopes since you get yourself tied up ....

Rupp: Well, for example, on dollar value we can only talk about sales value. The actual value of something that we don't have now, that is something of very high specific activity, can't be determined. There is just no way of doing it.

Cole: You can evaluate it in terms of your own production, and how much of this money is tied up in the reactor end of it and how much is tied up in the products that come out of the reactor.

Rupp: Now if the reactor were designed with hydraulic tubes, and if it were designed specifically for radioisotope production where you could put loops in it and do continuous C 14 work, then you could prove it on a dollars and cents basis. I'm not saying the flux would be  $10^{15}$ , however; it might still be a  $10^{14}$  reactor.

Boyd: How many requests do you now get for high specific activity isotopes over and beyond what you can produce now?

Rupp: We get some requests for certain high specific activity isotopes, but I can't give a quantitative answer on this.

Boyd: What useful new radioisotopes can you get by multiple capture?

Rupp: We don't have enough information on this; there hasn't been enough work. I feel that there must be a lot of them, but I can't pinpoint them.

Lane: Haven't they ever irradiated material in the MTR to get multiple capture?

Rupp: There hasn't been any real production of these, at least not that I know of.

Lane: Any other questions?

Snell: I would like to bring up another aspect of the matter that may be confusing to some people, I don't know. All of the isotopes produced in reactors comprise only one-half of the isotopic chart. What about using a cyclotron for producing isotopes in the lower half of the chart? Aren't there some attractive ones there that are useful?

Rupp: There are. We haven't done as much thinking along that line as we have the reactor-produced isotopes, but there definitely would be many of them that would be very useful. Your point is here whether it would be better to have a big cyclotron than have a big reactor?

Snell: No, not exactly. I didn't want to make it that sharp.

Lane: I don't understand what you mean by the lower half of the isotopic chart.

Rupp: The reference is to neutron deficient isotopes, specifically positron emitters, etc. These are isotopes we just don't have now at all. The field is wide open.

Snell: It is a really different point.

Fowler: Well, it's not completely different, however, since there is a matter of ultimate high flux that one can achieve with a machine. I think one can marshal arguments to the effect that the really ultimate in high flux is better achieved with a machine since the amount of heat which must be dissipated per neutron added is less at 900-Mev protons than it is for neutrons produced with fission. So if the limitation is the power you have to dissipate to get the high flux, the ultimate limit would be obtained with protons.

Lane: But so far that doesn't appear to be the limitation, at least from what we've heard today.

Fowler: Well, we haven't talked about ultimate high flux.

Rupp: I certainly haven't studied that too much, but I know that there are a lot of nuclides that would really be interesting if you could get them.

Snell: What I was really wondering - can one really develop a case for the 900-Mev cyclotron on the isotope production side. However, it would require horribly expensive running time, I believe.

Rupp: I know that most other cyclotron-produced radioisotopes have been very, very expensive. Well, usually there is a factor of 100 difference in unit cost.

Taylor: Do these procure any customers?

Rupp: Yes, most of these things are needed by research groups, groups that don't have a lot of money unless they have government contracts. I used to think that perhaps money didn't make any difference in buying such things as radioisotopes, and very often it makes up a very small amount of the total cost of the research project; but even so, for some reason or other which I don't understand, the price still seems to make a difference. A factor of 100 would make a real noticeable difference. Of course, there are some things that unless you get a reasonable amount of radioactivity you can't do good work anyway. That has been one of the limitations in cyclotron production - not being able to get enough to work with.

The seminar was adjourned by Mr. Lane.

## INTRODUCTION

The seminar was opened by J. A. Lane with Mr. Levy being introduced as the first speaker.

## NEUTRON DIFFRACTION - H. A. Levy

I am going to have a few remarks on what we could do in the field of neutron diffraction with a new high flux reactor. When Dr. Weinberg brought up the subject, he suggested that we might aim our efforts toward a study of protein crystals, and with this in mind I have used that rather long-range goal as something against which to measure the utility of a neutron beam that we might get from a high flux reactor. We will find out rather shortly that it is a long range goal, that it is not definitely in the realm of the reactor that is being talked about now. We have a second speaker coming, on neutron diffraction, and I hope that we will not overlap too much in our remarks and that between us we can manage to cover the field.

Now, in asking what we could do with a high flux reactor in the field of neutron diffraction, there are several aspects of the problem that occur to me. First of all, and most simply, we can say that we could make the kind of measurements we are making now faster and better, and more easily. That is, a problem on which we would now spend about two months taking data could certainly be done by whatever factor increase in flux we have in that much shorter time. I don't think this constitutes an excuse for a new reactor, however; we should look further. We could make the measurements we are making now more accurately, we would think, if we had higher fluxes available. We could count for longer times and get a smaller statistical error in the measurements. Now this doesn't necessarily follow that the measurements are thereby better; they wouldn't be any better if our background corrections rise at a certain rate as the flux rises. I am going to have a little more to say later about whether a reactor is a better one for neutron diffraction simply because it has a higher flux. We are really interested though in extending problems to things that we can't do now, and in this direction, the first thing that might occur to us is that we are very limited at the present time when we can't get large single crystals.

[Weinberg: Before you go on to the question of extending your investigations to things that you can't do now, could you pursue a little bit further the question of doing present things faster. As far as I can gather you are abundantly busy right now dealing with the data that you get at the present rate; now what would you do if the data came in ten times faster?

Levy: You might say that we would have to try to keep ten times as many people working. I don't think this is really the whole story though, but it is part of it. Actually, we have a group of four dealing primarily with the interpretation of neutron diffraction data, and we don't quite keep our present spectrometer busy, although we come very close to it. We could obviously then accomodate a larger group on problems of the same type as we are doing now, but we would certainly be more inclined to proceed to problems that we can't do now.

Weinberg: So rather than do things that you now can do, you would do other things.

Levy: We would spend a smaller part of our effort on the kind of things we can do now.

Weinberg: But you haven't solved all the problems that can be done by a long shot at the lower flux.

Levy: That is certainly true.

Weinberg: What you are finally saying again is that the field, to be thoroughly exploited, really requires ten times as many people. In fact, there is no limit to the number of people that you can effectively use.

Levy: There is really no limit, but at the same time we can foresee the time at which there will be many more groups working in this field.

MacPherson: You are engaging in an analysis of rather complex crystal structures?

Levy: We are engaging now in the analysis of rather simple crystal structures, and doing a fairly thorough job, not only in placing the positions of atoms, but also in describing their thermal motions. This we can do now at a fairly slow rate with our present spectrometer, but there are coming into existence quite a number of new neutron diffraction groups with neutron fluxes available to them in the order of magnitude of our graphite reactor up to ten times that much. I think we can foresee a more rapid advance in the field of analysis of simple crystal structures by neutron diffraction.

Blizard: How much of an improvement will you get by going to the ORR?

Levy: The question here is really what constitutes an improvement. We will certainly have a higher flux; we will have higher beam intensities; and we will be able to get higher resolution. I am going to go into more detail on what I mean by these terms a little bit later. As we get into the design problem on the ORR, it becomes increasingly clear that it is not entirely a blessing though, because we are going to deal with where we have perhaps 100 times the slow flux, we are going to have 1000 times the fast flux. We are going to have to do something about that.

Blizard: This is what I was thinking. Is it true that one of the advantages of going to a higher flux machine is that you can afford then to throw away some of the flux if you preferentially throw away fast flux?

Levy: We don't have to do it at the graphite reactor; it comes naturally in a very favorable ratio. But at the ORR, it is going to be necessary to sacrifice some of the potential increase in order to make use of what we have.

Blizard: That's what I'm saying, only if you went to a still higher flux reactor you would presumably be able to get a better ratio.

Levy: Then I would say this higher flux reactor, if it is aimed toward neutron diffraction, ought to be built with the flux ratio in mind.

Weinberg: It should be a heavy water reactor?

Levy: Well, it should be maybe a graphite reactor. At least, it should have a region of high ratio of thermal to fast flux; our beams should look at moderator rather than fuel.]

I was about to mention the kind of things that we might take up that we can't accomplish now, and the first thing I am simply going to mention and pass over because Dr. Wollan is going to talk more about it. At the present time we can do rather little in the way of crystal structure analysis with powder samples. If single crystals are not available we are confined to very simple crystal structures indeed to be able to analyze complete powder data. On the other hand, there are many, many problems where one can't hope to get single crystal specimens, and the extension of powder data techniques certainly then requires a high resolution instrument, which in turn calls for a high flux source. Along the same lines there are very many problems that could be attacked if we had small single crystals available at the same order of magnitude as is used in X-ray crystal analysis. These range from a factor of 100 to 1000 times smaller in volume than what we try to use now, and a flux increase of the same order of magnitude would be called for to make use of samples of this size.

Now I would like to take up what I think are the two important topics in this discussion. One is the analysis of more complex crystal structures and what this calls for, using the protein problem as a yardstick against which to measure the effort. Then second is a discussion of what we might call inelastic neutron diffraction, which is really a triple scattering experiment in which not only is the angular distribution of the scattered neutron measured, but also its energy distribution and what scientific interest, particularly chemical interest, might lie in this field. Well, now first of all the question of analysis of complex structures. The general measure of complexity that I am going to use is the number of atoms which is in the asymmetric unit of the structure, the unit cell. At the present time our most ambitious crystal, n-acetyl glycine, contains about 20 atoms, and I think that we consider this to be pretty much on the limit of feasibility with our present equipment. We are now dealing with a flux at the interior of our beam hole of  $3 \times 10^{11}$ , which is not quite what we might get if we were at a more favorable position.

[Weinberg: Where is the  $3 \times 10^{11}$  flux?

Levy: At the center of the reactor opposite our beam hole.

Weinberg: Oh, but that's not really quite relevant to the issue here, is it? Wouldn't it be much better to speak of the flux you can get at your sample?

Levy: I want to extrapolate from our present experience, and I think this is the only relevant number.

Weinberg: Well, then we are going to have to go from that to what the beam intensity is in the ORR, and the factor by which the beam is degraded from center to edge.

Levy: We have no experience yet in the ORR .....

Weinberg: No, but we have wonderful calculators though! They can calculate this to a "fare-thee-well".

Levy: I wish I knew just how to do it, but for the sake of extrapolating our present experience, I would like to start with this number.

Weinberg: I would just like to say ... - Well, let's see how it comes out; there may be a factor of 10 floating around there.

Levy: There may be. I would certainly like to get the reaction of you people on it.

Weinberg: I think the ratio of beam flux to central flux will be much higher in the ORR than in the graphite reactor because you are so much closer to the core.

Levy: This is the number that the operations people tell us is flux in the moderator outside our beam hole. We know what it will do.

Weinberg: But it's 15 feet from your beam hole to where your sample is.

Levy: I'm not sure that is a relevant factor here.

Weinberg: Well, there is then the question of how much it is collimated, so you have to look at really what is the angle.]

Now, at the other end of the scale, a crystal of a fairly simple protein will have about 2000 atoms in it, about 100 times as complex as on the scale which seems to us to be relevant. In between there are all gradations of problems some of which are quite interesting, and one must think fairly hard though about whether that type of problem is going to justify a new reactor or not. I think that we could argue that the protein problem is sufficiently important perhaps so that if we thought we had a good chance of success in solving this problem, it would be a strong argument for the reactor. I am not making this as a flat statement, but as a suggestion. In between we have to think a little harder. The geometric mean structure with about 200 atoms would be something like Vitamin B<sub>12</sub>, which is the most complicated structure that is approaching a complete analysis by X-ray diffraction. Now, if we make the following argument that the resolution of our present instrument is just adequate .....

[Weinberg: I don't understand. For the Vitamin B<sub>12</sub>, the number 200 includes the hydrogens, as well as everything else?

Levy: It's about 100 if you don't include the hydrogens though. The protein would be a 1000 atom problem without hydrogens.

Weinberg: If it were with X-rays, it would be a 1000 atom problem.]

If we argue that our present instrument has just adequate resolution for the 20 atom problem, and we ask for one that has just adequate resolution for the 2000 atom one; and if further we say that our individual measurements should be just as precise from the point of view of counting statistics as they are now; and if we go even a step further and ask that we be able to collect enough data for the difficult problem in the same time as we now use to collect enough data for the easier one; we are going to come out with a desired flux proportional to  $n^3$ , the number of atoms cubed - that is if we make those three requirements, although that's asking a lot. Now this isn't quite fair; we can argue that it is overestimated in some ways, but it is also underestimated in others, and I would like to go into more detail here. Now the three factors that I mentioned are (1) the resolution problem, (2) the intensity of the individual reflections, and (3) the time for collecting data. What we require in the way of resolution is that our beam be defined sufficiently closely geometrically and in wave length spread so that we can pick out one reflection free of its adjacent ones. This, of course, is something that is not a constant from experiment to experiment, but we will have to argue a little bit roughly. At the present time the inner reflections where resolution is naturally greater can be easily resolved one from another; with the outer ones we are starting to get into trouble especially at high scattering angles. I think that by and large we can say that we have about the limiting resolution which would enable us to solve this problem. Now in defining the beam geometrically, what is required is that the angular divergence of the neutron beam coming into the sample go down as approximately the linear dimensions of the unit cell. The reflections will occur closer together in Bragg angle as the linear dimensions increase, and they will occur closer together in crystal rotation angle the same way. So then we can say that we have two geometric dimensions which have to be squeezed down. We have to get our same intensity into a smaller and smaller cone as we go to bigger unit cells. This gives us a factor of  $a$ , the linear unit cell dimension for each of these. Then because the crystal disperses the neutron's wave lengths, and we have to separate closer and closer reflections on wave length scale also, we have to put in another factor; and this also goes as the linear dimension of the crystal. But that is just a volume factor, and the number of atoms for similar crystals will be approximately as the volume of the unit cell, so this amounts to a factor of  $n$  - a factor proportional to  $n$ , the number of atoms in the unit cell.

The next factor, the intensity per reflection, is well known - has been well known for some years. The average intensity that one can get from a crystal over the whole collection of data is proportional to the sum of the squares of the scattering factors of the atoms in the unit cell. The intensity from one unit cell has to be multiplied by the square of the number of unit cells in a crystal. This is an interference problem. The intensity of a given crystal reflector is proportional to the square of the number of unit cells per unit volume, times the square of the structure factor per unit cell. If

we combine these two things and ask for the average intensity of a crystal of a fixed volume as the unit cell increases in size, but the kind of atoms present doesn't change (in going from an amino acid to a protein, this would be about true); if we don't change the size of the specimen, but simply make the crystal more complex, we are dealing with the same number of atoms in fewer unit cells. We come out with the average intensity proportional to  $1/n$ , so that if we require the same total counting rate as the crystal becomes more complex, we have another power of  $n$ .

Then finally the third factor, the time for collecting the data. As we increase the number of atoms, we are increasing the number of parameters at the same rate. It takes three parameters per atom to locate an atom in space. If we also describe the thermal motion completely it takes six more, making a total of nine. Of course, here it is a question of what you are after. If we are satisfied with determining positions only, as we might very well be in a complex problem, it is three per atom. However, it is still proportional to the number of atoms however you do it. So the number of answers that we desire goes up proportional with the number of atoms. The number of reflections available to the experimental method, as it turns out, goes up in exactly the same proportion. Since we are collecting all available data at this point, that is another factor of the number of atoms. Now here I think we can argue that perhaps that is not really necessary, but it depends again on what you are after. If we are going to make a real contribution to solving the structure of proteins which as yet are unknown in detail, it is very likely necessary that all data that it is possible to collect be collected. On the other hand, if the structure is known by and large, and we are asked only to refine and to determine more accurately parameters that are already partially known, I think we could get by with fewer.

[Weinberg: Well, you seem to leave out the very basic question of whether it makes sense to even think of looking at proteins with neutrons at all before the thing has been looked at thoroughly with X-rays.

Levy: That is a pertinent question, and I do have some remarks on that. I think it's questionable, let me say; and I have some reasons, and I do plan to make some remarks.

Weinberg: Before you leave this question, do you have any actual practical cases in which you compare say a real simple one with your  $n$ -acetyl glycine, and is it in fact true that it goes like  $n^3$  in actual practice, or is that a foolish question?

Levy: No, we don't have any experience, and I don't think that we have the range of complexity that would justify making the comparison. After all our tools are more than adequate for more simple things.]

Well, one might say that we could knock off a half a power of  $n$  - instead of  $n^3$ , perhaps  $n^{2-1/2}$  power. But on the other hand, we are now dealing

with very large crystals. A crystal of 2 millimeters in diameter and 5 millimeters tall is quite large compared to what crystallographers usually deal with, and if we were to attack the protein problem we would most likely find it quite impossible to obtain crystals of interesting materials in this size. So a reduction in the sample size of a factor of 10 or perhaps a factor of 100 is in order, and this only pushes up the problem of getting enough flux.

[MacPherson: Don't you have an instrumentation difficulty in getting the extremely sharp angular resolutions that you are talking about here?

Levy: No, I don't think that would be a real difficulty. The angular requirements are not as high as one might think. I think a total spread of something like a quarter of a degree, which is perhaps larger than one has in the X-ray case, could be made to do the job. This isn't hard to achieve, and as for fine wave length spread, this is achieved fairly easily by using a perfect crystal monochromator. These would be available.]

Then if one takes an  $n^3$  relationship, the flux goes up to  $3 \times 10^{17}$  for the protein problem and somewhere in between for other things which are interesting but don't have quite the glamour of the protein problem. Also, the flux could be pushed up another factor of 10 or 100 to account for the reduction of sample size. Another thing that could push the flux requirement further might be the necessity for discriminating further against a fast neutron component that might come from the reactor, and this might be done by a second monochromatizing crystal. This would add another factor of 10 to the neutron flux.

[Lane: You're talking now about  $10^{18}$ ?

Levy: I could make it  $10^{19}$ , but I hope it wouldn't have to go that high. I would say that  $3 \times 10^{17}$  is the minimum though that would make it worthwhile to try to work in this field.

Ergen: It appears that since you are throwing away so many neutrons, before one builds a \$100 million reactor it might be worthwhile to consider if one could use the neutrons more efficiently, or am I wrong on this?

Levy: There are some possibilities, but it doesn't seem to me they offer an order of magnitude improvement. It is simply necessary to have a very fine probe to examine a complex structure, and the neutrons must be put into a very finely collimated beam. If one wants to make use of more neutrons by enlarging the dimensions of the beam, the probe is no longer fine enough to examine a complex structure.

Ergen: Is it completely out of question to make the neutrons which you reject somehow come back and be selected again?

Boyd: The question of a neutron lens, perhaps?

Levy: Well, I don't know whether invoking the second law of thermodynamics is really appropriate here, but if it amounts to getting a parallel beam of greater brilliance than your source, you can't do it.

Weinberg: You need a little Maxwell demon.]

I said I wanted to say a little bit about whether it makes sense to study proteins. Proteins are materials which at best are rather poorly crystalline, and one has to keep in mind then that you can't describe a protein structure unless one is discovered that is highly crystalline, and you can't expect to describe it in the same detail that you can describe a simple structure. Disorders enter the problem; the atoms perhaps do not have definite positions that you could describe by definite parameters. They have greater thermal motion because of the loose binding that occurs from place to place in the structure. Finally, I think one can ask whether a structure whose repeat unit is so enormously long really remembers from one cell to the next exactly how it should be constructed. This all reflects itself in the X-ray data in a dearth of individual reflections. Where a good crystalline material would give a total number of reflections proportional to the number of atoms in the unit cell, proteins do not do that. The data die off in smaller scattering angles, and with a smaller number of data you do not have the opportunity to describe the structure in such detail. I think the reason for it is that the structure doesn't have the detail to describe.

[Weinberg: More like a liquid?

Levy: Part way, let's say, between a good crystal and a liquid, but only a short distance.

Weinberg: Closer to a crystal then?

Levy: Yes, closer to a crystal than a liquid.]

Well, then there is also the question of timing. The groups who are working with X-ray diffraction on protein structure are rather optimistic that they will be able to do what we call "solving the structure," that is, giving the main aspects of the atomic positions in not more than a few years. This kind of a project would surely have to wait that long or longer, and it may be that by the time a suitable reactor is available, the initial stages, the solving of the first protein structure, would no longer be available to us. At the time, it is extremely safe to say there will always be interesting problems, and if the method is going to make a contribution at all it could do so just as well at a later time than at an earlier one.

[Weinberg: You hadn't thought of using neutrons to determine the backbone of this structure had you?

Levy: I really think that is foolish, but it is a possibility.

Weinberg: Oh, but you haven't done that with anything else?

Levy: We haven't done that with anything else.

Weinberg: Why start with proteins?

Levy: Well, because there is the possibility of making a contribution, perhaps not with proteins but with other things. The solving of a structure is getting to be less and less of a problem, I think, as time goes on, but in the past it has always been the thing that took a stroke of genius in a difficult case. You had to get an idea and then show that it was right. As I say, it is getting to be less and less of a problem because more powerful methods are being invented. If it should turn out that the X-ray method has failed, then the idea of having two sets of scattering data from different radiations, with different scattering powers of the atoms, is certainly a possible way of pushing the thing further.

Weinberg: But that has never yet been .....

Levy: Never yet been done. There have been one or two structures that have not been determined by X-rays that have been determined by neutrons, but it isn't because they couldn't be determined by X-rays. It isn't because people tried and failed.]

That's all I have to say about more complex structures.

The other field I want to take up is that of inelastic scattering of neutrons which has been given its start by Brockhouse and his associates at Chalk River.

[Weinberg: Excuse me. Before you go on to the other subject - actually the thing that I had in mind was this. There are in many of these structures, insofar as I understand it, rather strategic hydrogen ions, the strategically placed ones which somehow are responsible for say either the polymerization or binding of adjacent structures. Now if you say that what you really want to do is to find out everything possible about just that particular hydrogen atom, does that greatly simplify the matter or reduce the amount of detail that you need, or is that in general not the case?

Levy: I think in general it would not be the case, but you could say this - if you knew everything else about the structure except the locations of the hydrogen atoms, then there is a possibility that by collecting a smaller amount of data, assigning approximate positions to everything else that you didn't know, then you might tie down one hydrogen atom.

Weinberg: Well, on the n-acetyl glycine you knew where the N's and the O's were from the X-rays, and all you were trying to do was get the H's?

Levy: Yes.

Weinberg: How much simpler was that? It doesn't go proportional to the number of H's, does it?

Levy: We probably could have verified the positions of all the H's, having guessed where they ought to be, but with a partial collection of data.

Weinberg: A tenth as much?

Levy: I think not. Half as much perhaps. Actually we were more ambitious; we wanted to describe both the positions and the thermal motions of all the H's accurately, and this required us to refine the positions of the other atoms further than it had been done.

Weinberg: So your impression is that even to find strategically placed hydrogen ions, whose general location you know in advance, still doesn't grossly simplify the matter.

Levy: Not grossly, I think.

Weinberg: You don't get a factor even of 10 in flux, you don't think, or 100.

Levy: Certainly not. I wouldn't want to count on it.]

Several writers have shown and pointed out recently that if one is able to measure not only the scattering angles of neutrons and neutron diffraction, but also their energy change, one can open up a new field of interest in the behavior of the scattering material. This would not be confined to crystals, but would apply also to liquids and other kinds of things. What one obtains is information about the dynamics, as well as the statics of the scattering system. As I said, Brockhouse at Chalk River has made a start in the experimental exploitation of this field, and his experience in what he can do with the flux available to him at NRX is a guide to what might be done if we had higher flux. Brockhouse has studied the scattering of neutrons from single crystals of aluminum and of vanadium. The experimental measurement is to count for, in his case, 7-1/2 minutes per point. A point is described by a given orientation of a scattering crystal and a given setting of the analyzing monochromator so that he has four quantities which need adjusting as he moves from one point to another. A complete examination of the aluminum single crystal must have required thousands of individual measurements. At least six months were spent in collecting the data, and then more time in analyzing it. He states in his report that the energy resolution was surely inadequate to obtain as much information as he would like, even to give a first description to the dynamics of the aluminum crystal; and I think the same could be said for the angular resolution, although not quite as urgently. I think that NRX would have a central flux of about  $5 \times 10^{13}$  and if you ask that the problem be simplified even just as to time of collecting data, then a considerable increase in flux, all that one could give it, would surely be useful.

[Wollan: I have some patterns that are Brockhouse's if anybody wants to look at them. They give some idea of the resolutions you get with some of Brockhouse's work.]

Now, I would like to say just a few words about what one could do of chemical interest. Perhaps Dr. Wollan will have more to say about crystal dynamics, which is a little separated from chemistry.

[MacPherson: I am not exactly clear on what he learns about the aluminum crystals.

Levy: What he is able to do is describe the direction, polarization, and intensity of the phonons in the aluminum crystal. He studies in detail how the vibrations are taking place, in what direction with respect to the crystallographic axis, and whether the vibration is longitudinal or transverse.

Weinberg: How does he get that? --- But that is beside the point.

Levy: I'm afraid I can't tell you offhand. It can be deduced from the measurements, however.]

In general, Van Hove has shown that what one can do with data of this sort is derive a function which is closely related to the radial distribution function that one can get by X-ray or neutron diffraction; that is, the function  $\rho(r, t)$ , the radial distribution function which describes the probability of finding two atoms or two scattering units separated by a distance "r". If one is examining a liquid or an amorphous material this is usually the end result of the structural investigation. Now Van Hove showed that this can be generalized so that it is a function of the vector distance and time if you have three dimensional data from a crystal or scalar distance and time from the amorphous material or a liquid. As I see it, we could answer some very interesting questions. If we were able to derive a series of radial distribution functions corresponding to different times of correlation as shown in Figure 1, in which the top curve represents the distribution  $t = 0$  which is the probability of finding a pair of atoms at this distance "r" apart at the same time. The middle curve represents  $t = t_1$  .....

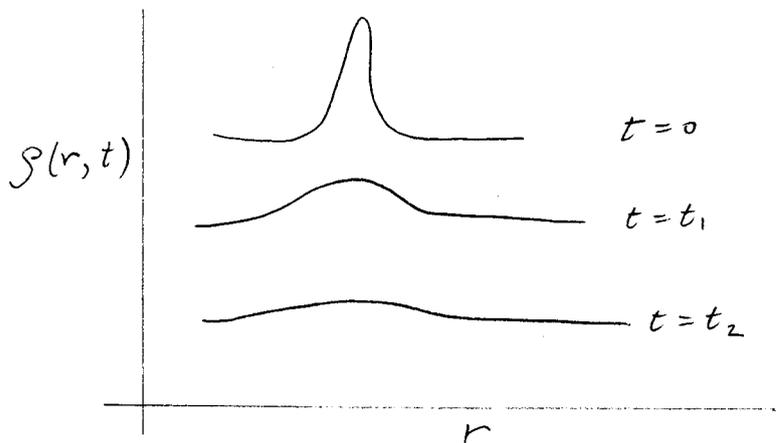


Figure 1

Radial Distribution Functions

[Weinberg: You measure the time from what instant now?

Levy: This is the probability of finding a pair of atoms a distance "r" apart. When one atom is at point "p", the second atom is at "p<sub>1</sub>", a distance "r" apart at  $t = t_1$ .

Weinberg: That's arbitrary - that "t<sub>1</sub>"? When you start looking is arbitrary?

Levy: When you start looking is arbitrary. Instead of a correlation at the instant .....

Weinberg: Is  $\rho$  a periodic function of "t"?

Levy: No, it wouldn't be periodic. As "t" gets large, the correlation of an atom at point "p" with an atom at point "q" a long time later would be zero. It would have forgotten any information it possessed as to where the first atom was.

Weinberg: It is evident I don't understand this very well.

Levy: If you have a molecule over times that are comparable to laboratory times, the function wouldn't change. If you have a crystal where this is true in three dimensions, then you get Bragg reflections which are elastic. There is no energy change on scattering.

Weinberg: The thing I don't understand is how you choose the zero of time.

Levy: The zero of time means an instantaneous correlation. A time not zero means a delay. We are dealing with a delta time.]

If we have a molecule, then the curves shown in Figure 1 would not change over a period of let's say microseconds anyway - at least many thermal vibrations. If we have a molecule which has transient existence, we could see its decay, and we can say something then about how long molecules stay together. In a problem such as the structure of molten salts, we now can learn by X-ray diffraction the static correlation. We can say what is the probability that two atoms are a certain distance apart at a given instant. But it is very interesting to ask whether this is a permanent configuration or a transient one, and that could only be answered with this kind of an analysis.

[Holmes: I would like to say with reference to this thing you were mentioning before, isn't this just a matter of a neutron scattering from the phonons in the crystal and an interaction with a number of atoms at one time?

Levy: The neutron gives up or takes up energy from the crystal by exciting or deexciting a phonon, that is, a vibrational mode.

MacPherson: Is this time function done with X-rays too?

Levy: No. With X-rays, the only inelastic scattering is the Compton effect, and this is the only one that is important. But this is much too crude a degradation, and the energy change is much too high. The reason it is possible with neutrons and not with X-rays is connected with the mass of the neutron. When a certain momentum change takes place with a neutron, an appreciable energy change accompanies it; whereas with X-rays the energy change is completely immeasurable.

Weinberg: Would you like to then answer the question, "Do we need a  $10^{16}$  reactor for neutron diffraction?"

Levy: I was afraid that question was coming. I would say that if we have a  $10^{16}$  reactor we can make good use of it in the various ways that I described, by doing our present jobs better and faster, by doing more complex jobs, and by going into this field of inelastic scattering. I don't think I am in a position to say whether the field by itself justifies a new reactor. All I can say is we could make good use of it if we had it.

Ergen: Going back to my previous question. Do you think that a factor of 10 could be gained by designing the reactor so it would be most appropriate to your work, rather than just by brute force going up a factor of 10 in power or in neutron flux?

Levy: Oh, you have reminded me of some things that I forgot to say. I would say that we could not do any better than the present graphite reactor in that respect. We have a very good geometrical situation and a very good ratio of slow to fast neutrons.

Blizard: Supposing we ran up the power of the graphite reactor, would this please you?

Levy: Very much.

Weinberg: That means that the enriched graphite reactor would have an even better ratio of slows to fast.

Levy: Yes it would be a very good tool for neutron diffraction. Unfortunately, it might be a better one than the ORR, but we don't know yet.

Wollan: You know the Brookhaven people were very disappointed with their enriched loading. They haven't gained more than about a factor of 2 in the neutron experiments.

Levy: Is that right? Well, that's another point.]

There are two things we would ask of a reactor as a neutron source. We would ask that the slow to fast flux, and I include gammas with this, be as high as possible.

[Charpie: We will have to take a vote on whether we want slow or fast flux! (Laughter)

Levy: Well, there should be a region at which the beams could be aimed in which the fast flux is relatively low.

Blizard: We need a reactor with one side slow, the other fast.

Levy: The other is that the ratio of the current to flux be high, and I don't really know just what this calls for.

Weinberg: This means that the  $k$  should be high. What you want is a heavy water enriched reactor like CP-5. That's what you want.

Levy: I should think that this would mean that if the flux implies that a neutron is scattered many times through a point before it eventually disappears, then that flux is not a good measure of what we would get out of the beam because our neutrons in the beam could come out only once.

MacPherson: Are people doing neutron diffraction at Hanford or Savannah River?

Levy: At Hanford there is a spectrometer that is used in measuring the energy dependence of cross sections. I don't know of any crystallographic work, and I don't know of any at Savannah River.

MacPherson: The question is how far will people travel to do such experiments.

Levy: Well, I don't know of any crystallographers who have gone out to Arco either.

Weinberg: I guess the MTR doesn't have a spectrometer, does it?

Levy: There is one for cross sections.

Weinberg: But not a crystal spectrometer for structural work?

Levy: No.

Weinberg: I suggest that your view that there will be many groups throughout the world may actually not quite be correct.

Levy: Well, they are taking steps for one at Michigan, North Carolina, Stockholm, Saclay, Penn State - I'm sure they are interested and are coming along.

Weinberg: On the question of the usefulness of the ORR, do you gain or lose, or has anybody really looked at the question of whether you gain or lose, by say putting a chunk of beryllium in the beam hole? They must have done that at the LITR.

Wollan: I don't know about the LITR .....

Harvey: We have a chunk of three inches of beryllium in the beam hole.

Weinberg: Does it help, or does it hurt?

Harvey: The fellows before us put it in, and they left it in. Because it is a big problem to get it out, and since we are moving, we didn't think it worth our effort to take it out to see what happens.

Blizard: It has to be a slab, rather than a chunk. If it is just a chunk it is all surely going to scatter out. If you have a big slab, then you would have essentially a bad geometry experiment, and you have a very different situation.

Levy: There is one suggestion .....

Weinberg: Well, I thought in any case the piece of beryllium has more effect on the fast neutrons than on the slow neutrons.

Blizard: It has in effect. If you have a hole coming through the shield, and put a piece of beryllium in this hole, the beryllium will actually cut down the total beam very strongly.

Weinberg: Sure, but won't it cut down the fast more than it cuts down the slow?

Levy: No.

Harvey: If it is a good geometry, then it would scatter everything else as much as it scatters high energy neutrons, then the cross section for low energy is 6 barns and high energy is only  $1-1/2$ , so you lose with good geometry.

Weinberg: Yes, that's right. You're right about that.

Levy: There is a way you could make it work. It is very difficult, but it has been suggested; namely, to make this beryllium a single crystal so that for Bragg reflecting neutrons it is transparent.

Ergen: Is this so impossible? I thought that if you stretched beryllium in one direction it does become essentially transparent.

Harvey: Beryllium is usually just sintered, and you couldn't get much effect.

Levy: When you look into the possibility of getting a beryllium crystal for a monochromator it becomes very expensive.

Billington: I am not sure that is so, because there has not been much effort in that direction. It may be possible with suitable casting techniques.

Levy: We should look into the situation.

Weinberg: Well, you should get a tremendous factor if you did that. Isn't this right?

Levy: You could get a normal relaxation as far as fast neutrons are concerned, and the slow neutrons would essentially go on through. It might be necessary to cool this crystal too, because of inelastic scattering that I have talked about in other connections.

Weinberg: The gamma rays would be bad.

Levy: The gamma rays would be bad.

Ergen: But it may not remain a single crystal after exposure.

MacPherson: It would be an extremely well collimated beam that comes through a single crystal. The part that comes through is attenuated by many factors.

Levy: No, at that stage it would not be well collimated. We ask what could happen to a slow neutron as it goes through a single crystal. Well, it would be Bragg reflected, but that requires a critical orientation which we would avoid, or it could be scattered by a phonon, and if the crystal is hard and the temperature low if necessary this would not happen. Otherwise it would have to go right on through.

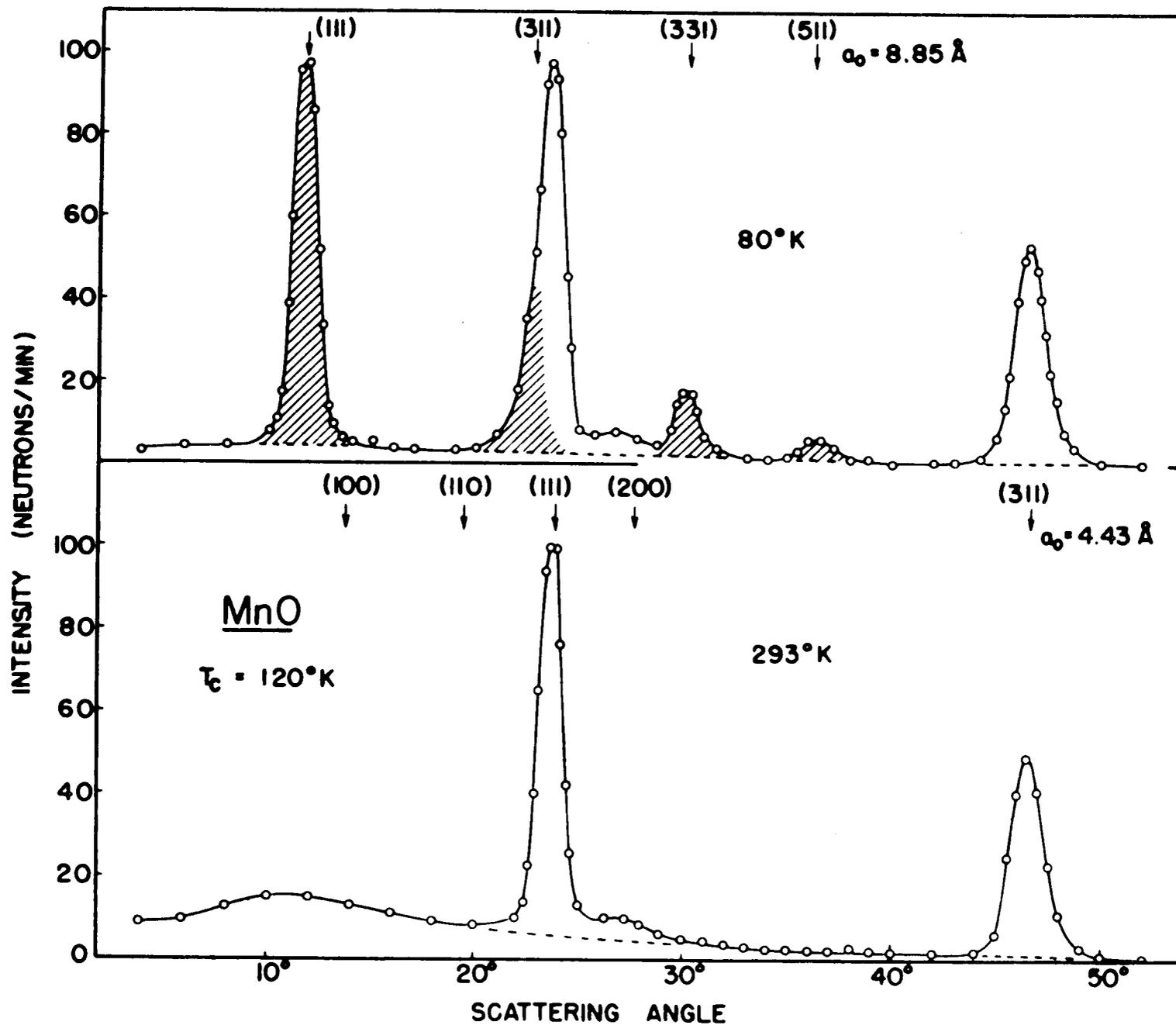
Lane: I suggest that we move on, the time is getting away from us.

Weinberg: I wonder if perhaps we could plan to make this session two sessions so that we can have enough time to hear everything that Harvey has to say?

Lane: That's agreeable with me if it is agreeable with Harvey, since we have only one scheduled talk for the next seminar. We will next hear from Dr. Wollan.

#### NEUTRON DIFFRACTION - E. O. Wollan

I would like to consider the powder diffraction technique and see what improvement by factors of 10, 100, or 1000 can mean for the kind of work that we are doing now. To do this, I would like to make some comparisons between the X-ray technique and the present neutron technique. I have here a modern X-ray diffractometer pattern (Fig. 1) (not shown) from a filtered Cu target X-ray tube which gives an idea of the quality of results that are obtainable by this technique, and I will refer to this pattern again a little later. For comparison, I show also a typical set of neutron powder patterns (Fig. 2). The lower pattern in this figure represents nuclear reflections only, and the top one includes magnetic reflections. A comparison of these patterns in terms of the resolving power of the



instruments will give some idea of the relative situations for X-rays and neutrons. Another neutron pattern that may come into the discussion is shown here (Figure 3). I would now like to show a table (Table I) which gives a calculated comparison between the X-ray and neutron diffraction techniques. For this comparison we consider the neutron situation in the graphite reactor and the X-rays from an X-ray tube with a copper target. Neutrons are shown in the first column and X-rays in the second. For the sake of definiteness, I do want to indicate that we consider here an X-ray tube used with filters, which then gives both the copper  $K\alpha_1$  and  $K\alpha_2$  lines in the monochromatic beam. This gives us a measure of the resolution of the system which we will later consider. In the neutron case, in our spectrometer the monochromatic flux at the sample is of the order of  $10^5$ . In the case of an X-ray tube, the calculated equivalent flux is of the order of  $10^{10}$  photons per square centimeter per second. I might mention here also that in an X-ray diffractometer where a counter is used, the efficiency of detection is close to one, and this is true also for our neutron counter so the useful fluxes in the two cases are directly comparable.

[Weinberg: Is that  $10^5$  what you actually now get?

Wollan: Yes, it is what we now get at the sample on the center of the spectrometer of monoenergetic radiation. I'll talk about the quality or monochromaticity later.

Weinberg: That means that the people at Chalk River right now are better than you in this respect by a factor of  $10^5$ ?

Wollan: 10 or 20, yes - in that range.

Weinberg: And they are the highest in the world?

Wollan: As far as I know they are - that's right.]

Now returning to Table I, let us consider the angular divergence for both cases. With a diffractometer you use a focusing technique for which one has a fair angular divergence of about one degree. In the case of neutrons we also use some focusing, which blurs out at large angles. The angular divergence in our incident neutron beam is at present also about one degree.

Now, let us consider the wave length spread in the incident radiations, or in other words the ratio of  $d\lambda/\lambda$ . In our work, as shown in Table I, the value of  $d\lambda/\lambda$  for the width at half maximum is something like 1 to 20. Now in the X-ray case the individual lines cannot be resolved, but the  $K\alpha_1$  and  $K\alpha_2$  lines can, and their  $d\lambda/\lambda$  value for this case is about 1 to 400. So at the moment everything is obviously in favor of X-rays, and one wonders how one can do neutron diffraction at all. But, of course, in the case of neutrons the sample size is much greater. Ten grams is a fairly representative sample. Some workers at Brookhaven, for example, use up to a couple of hundred grams. In the X-ray case the sample is more like a hundredth of a gram.

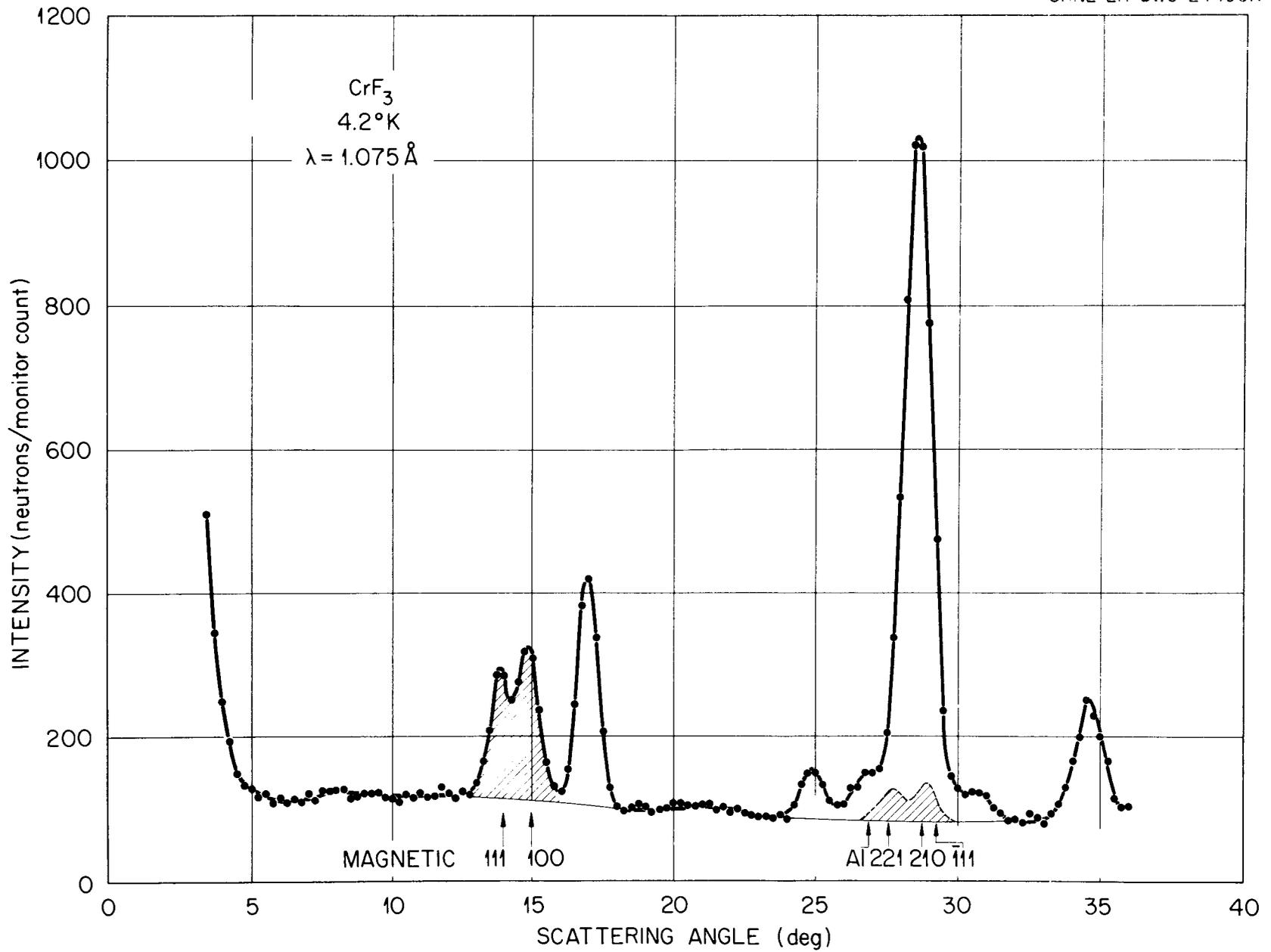


Fig. 3. Another Neutron Pattern Which Strikingly Demonstrates the Need for Better Resolution and Higher Intensities.

TABLE I  
COMPARISON OF NEUTRON AND X-RAY POWDER METHODS

	Neutrons Graphite Reactor	X-Rays (35 kv, 25 ma Cu Target)
Monoenergetic Flux for Angular Divergence in Incident Plane	$\sim 10^5$ neutrons/cm <sup>2</sup> sec  1°	$\sim 10^{10}$ photons/cm <sup>2</sup> sec (at 1 ft.)  1°
$\frac{d\lambda}{\lambda}$	$\sim \frac{1}{20}$	$\sim \frac{1}{400}$ (Cu K $\alpha_1$ - Cu K $\alpha_2$ )
Powder Samples	$\sim 10$ gms	$\sim 0.01$ gm
Time to Obtain Pattern	$\sim 20$ hrs	$\sim 1$ hr
Detecting Efficiency for Counting	$\sim \frac{1}{(B^{10}F_3)}$	$\sim \frac{1}{(Xe)}$
Detecting efficiency for photographic recording.	This can probably be made as much as 100 times better for neutrons than for X-rays because of the larger energy associated with B <sup>10</sup> (n, $\alpha$ ) $\sim$ 2.7 Mev vs. the $\sim$ 10 kev per Cu K $\alpha$ photon. Small samples must be used in both cases.	

To make  $\left(\frac{d\lambda}{\lambda}\right)_{\text{Neutrons}} \sim \left(\frac{d\lambda}{\lambda}\right)_{\text{X-rays}}$  would require an increase of a hundred or more in neutron flux because the angular divergence must be made smaller to effect the decrease in  $d\lambda$ .

To make neutron powder photographs comparable to those obtained with X-rays would require an increase in neutron flux of the order of 10,000.

[Weinberg: I don't understand why there is such disparity in sample size.

Wollan: Well, no matter how you dispose the sample for X-ray work, the absorption factor is so great that it still becomes a very small sample, even if you spread a little material on a very small plate as is done for the usual diffractometer work.

Weinberg: I see. For 50 kilovolt X-rays what is the attenuation likely to be?

Wollan: The absorption coefficient is very high, I do not have an exact figure. In general, when one does X-ray powder diffraction with a capillary tube, a very small capillary tube is used, frequently less than a millimeter diameter. Actually the reason that one can do neutron diffraction at all is the fact that neutrons can penetrate relatively large samples.]

One sees then that a factor of about 1000 can be gained for neutrons on the basis of sample size alone. One can also gain back something more in the time of exposures. In the case of neutrons, one takes of the order of 20 hours to get a good pattern - with X-rays it is something less than an hour.

Let us now take a look at the X-ray and neutron patterns again. In the X-ray pattern one sees that in the neighborhood of  $2\theta = 35^\circ$ , the diffraction peaks are about  $1/5$  of a degree wide at half maximum. Now if one goes up on this pattern to about  $60^\circ$  you will find that the two  $K\alpha$  lines have become resolved, and the quality of the pattern begins to become poorer even under the condition of  $d\lambda/\lambda = 1/400$ .

In the case of the neutron pattern, take this one for example (Figure 3), we are dealing here with lines that are of the order of a degree wide. The clear areas are the nuclear peaks, and the cross hatched areas are those from magnetic scattering. Let's now take a look at the difficulty we run into in a situation like this when we actually try to make an analysis of these data. We have in this pattern one pair of just resolved magnetic reflections and at larger angles a very small additional magnetic contribution. This little area of magnetic scattering was obtained in this particular pattern by taking two patterns, one at room temperature and one at  $4.2^\circ$  (which is below the magnetic ordering temperature), and subtracting the two. Of course, it is evident that you can't do a very good job in obtaining the intensity of this small magnetic contribution although with good statistics, you do reasonably well.

Now we actually learn from this pattern and almost from the first peaks alone, the magnetic structure, that is the antiferromagnetic arrangement of the spins in which half the spins point say up and half down; and in this case we obtain this information from very little data. But there is one bit of data here that we cannot easily get from such a pattern, and that has to do with the angular fall-off of the magnetic intensity. Such data gives information about the distribution of magnetic electrons which are the outermost electrons of the atom. I believe you chemists will even agree that knowing the distribution, or essentially the wave functions, as accurately as possible of these outer electrons is very important in the theory of bonding, and at the present time we do a poor job in learning anything about this subject. I can show you here the form factor data

that now exist for a manganese ion from which the distribution of magnetic electrons is obtained. You will notice that the data have been obtained only over a part of the desired range, and it becomes much more difficult to get good results over an extended range. Actually, if one is going to carry out this type of work into the large angle region, one must have good resolutions so that the diffraction lines of this type can be resolved. To do this requires resolutions of the order of that shown in the X-ray pattern. In addition to better resolution, it is evident that increased intensity is also required. Now let's take a little look at this resolution as represented in the table. In order to achieve with neutrons the equivalent in resolution to the X-ray case, one needs to improve  $d \lambda / \lambda$  by a factor of about 20, but one can't gain this by an increase of 20 in intensity because one has to get the monochromatic beam by reflection from the crystal, and one loses both from the reduction in  $d \lambda / \lambda$  and from the small angular divergence required to achieve the better resolution. I would guess that to put the two techniques on a comparable basis, an increase in flux of something like 100 would be needed.

[Weinberg: Is it as little as 100?

Wollan: Yes, but this only takes care of the resolution factor.

Weinberg: But you gained a factor of  $10^3$  in sample size.

Wollan: Yes, but I needed this  $10^3$  in the sample size even to get this little bump on the pattern, and if you compare this with the X-ray pattern you observe nice sharp clean peaks. Remember at this point I am only out to  $30^\circ$  in scattering angle. If I go out to  $2\theta$  of  $60^\circ$ , which I really need to do to evaluate the form factor, then I need more than good resolution - I also need increased intensity.]

Now another aspect of the evaluation of the distribution functions in atoms is the fact that if you use the coherent peaks, then you jump over regions of the curve. Another approach is to take the diffuse background scattering - and I did have one curve here in which the normal background scattering has been subtracted. It is evident that if one is going to measure this diffuse scattering out to large angle one must also resolve the lines so that one has space between them to measure the background scattering. On this pattern out at  $40^\circ$  one sees that there are only about 2 counts per minute in the background; so an increase in intensity of 100 would be very useful.

[Weinberg: Is it true that the same major question that was asked of Levy applies to you too, that to effectively use a much higher flux you probably will need more people? Is this right?

Wollan: I think that is right. In fact, I think it would be foolish to spend a lot of money on a reactor without having enough people to make proper use of the equipment that you are using ...

Weinberg: Are there people available who would go into neutron diffraction if we had jobs for them? How many could you hire if you had the money?

Levy: One could get three or four a year easily. The field is very attractive. But so far we haven't made a big effort to get them.

Wollan: Our situation is pretty much the same.

MacPherson: Isn't it the fact that it probably isn't so much the greater speed with which you can do things, but that you can do better things that attracts these people. The same group would have an unfair advantage over competition.

Weinberg: I have somewhat the impression though that the various groups have somehow gravitated into different areas, and that in each one of these areas there are very interesting things to be done, that the groups don't really overlap very much. Now do you think that there are enough additional areas so that new groups will be able to find things that don't overlap, or will there start being competition in some of the same areas? For example, who else does magnetic work?

Wollan: The Brookhaven group and the Chemistry Division are probably the best.

Question: Are they rather competitors of yours?

Levy: I don't know how the multiplicity of magnetic problems is, but certainly in the crystallographic field there are unlimited structures that are interesting now.

Weinberg: And that can be done uniquely with neutrons, and not with X-rays?

Comment: Of course, if you count the total number of structure groups in X-ray, there must be about 1000 throughout the world, and the total number of structure groups in neutrons is only about ten.

Levy: Not even ten. - Harwell, Oak Ridge, Argonne, Brookhaven, and possibly the Russians.

Wollan: Most of the magnetic work is done at Brookhaven and ORNL, some at Chalk River, Harwell, and Argonne.

Levy: Also the Norwegians and Swedes are getting started.

Weinberg: These questions are a little bit irrelevant to the issue of should we built a  $10^{16}$  reactor, and more to the point of should we built another reactor, whether it is  $10^{16}$  or not.

Lane: Well, I haven't really heard much of a strong case for a  $10^{16}$  reactor, except that you can do these things better if you have more people, and it opens up these new vistas. But there is some question about the protein work even so.

Levy: The protein question is beyond anything that we can achieve. One has to be satisfied short of the protein, but these problems don't quite have the glamour of the protein problem.

Swartout: I have another question. In referring to Wollan's data here you are talking about spending many millions of dollars to increase the maximum flux of

the reactor by say a factor of  $10^3$  or  $10^4$ , and he is working with a beam which is lower than in the reactor by  $10^7$ . If we were to tell them they could have \$1 million to beef up this beam some way in the graphite reactor, could they do it that way, instead of building a whole new reactor?

Wollan: If you enriched and moved in closer you could.

Weinberg: I am sure that people have not really looked in the greatest detail at research reactor design from the point of view of maximizing the beam quality.

Wollan: Well, there are certain little tricks that we have done, but we have not measured how effective they are. For example, in line with what you were saying, we originally worried about bringing a large beam out of the reactor and then reflecting it from a crystal in such a way as to produce a very narrow angle reflected beam. This is called the Fankucken method. I don't think any of us agree exactly on how much good it does, but this is an example of striving to achieve something along those lines, and there may be other tricks.

Weinberg: Still, Levy's point which you discussed about whether you really are working at the maximum distribution, I think you explained to me that you are working just about the best place now, so that you don't really gain by cooling down monochromically - in fact, you lose.

Levy: You would then want to use a longer wave length and get less data.

Holmes: I once heard some talk about putting a heavy water facility in the ORR.

Weinberg: There is heavy water around the beam holes, but there is no place where the thermal neutrons which you see have been forced to go through heavy water.

Lane: This can't be with the present design.

Weinberg: I don't know. I suppose if you tried hard enough you might be able to do it.

Lane: I don't believe so.

Charpie: It's too late. We talked about it, but it's too late.

Wollan: Can't you put heavy water in the inside of your beam hole? Is that what you have in mind?

Weinberg: Perhaps you can put cans of heavy water in the beam hole, if you line the whole beam hole.

(Subsequent discussion revolved around merits of putting  $D_2O$  in a beam hole in the ORR. It was agreed that the situation should be investigated in more detail.)

Levy: I was interested in the remark that the Brookhaven people were disappointed in increased intensity. What was the flux factor they got?

Wollan: A flux factor of about 2. They actually said it improved their situation by only about a factor of 2.

Weinberg: Isn't that because the reactor is so much smaller they have a great big reflector now, and they are too far away from the core? They would have done better if they had made an asymmetric loading of the pile.

Charpie: They should have loaded right up to the front face.

Levy: I was wondering if this indicated that when you measure the flux in the moderator you are measuring the same neutron many times, but when you take it out in the current you only measure it once.

Weinberg: Yes, you're right - sure. But I believe that one of the few things that really can be calculated if one sets one's mind to it and calculates it properly is the beam that one gets. Isn't this right, Dave, you're the expert on that.

Holmes: Yes, but of course if you're talking about the whole problem then, of course, that's a very tough problem.

Weinberg: No, I mean given the flux distribution which we know really quite well to compute what comes out of that channel. Even I could calculate that. You look skeptical!

Holmes: Well, one could make a stab at it.

Weinberg: Oh no. This is something that Mr. Neal Lansing calculated in great detail - calculated the beam. You can do it because you know the angular distribution, and you know pretty well the way the flux goes so you just calculate it. I don't get any takers, I see.

Charpie: It's been done once. It was done for this arrangement once. That's how we decided to put the  $D_2O$  around the beam holes in the ORR.

Weinberg: I thought we also did an experiment on the  $D_2O$ .

Lane: It didn't agree with the calculations, I thought.

Weinberg: Yes, but those were a little different. That was a different issue.

Lane: Wasn't it the degradation of fluxes that went along the beam hole?

Weinberg: That's right.

Holmes: It looks to me as though Swartout's point is well taken, that more should be done by the long range planning group on how to get better beams out of existing reactors. If you had  $D_2O$  around a beam hole that goes right to the active lattice, you could get much higher beams.

(A discussion on the use of the ORR followed.)

Weinberg: Well, I don't think we should try to settle that now. I must say that the general impression I get of this discussion is that we got a little bit off the question of  $10^{16}$  and quite a bit on the question basically of whether really another reactor is not needed for the Laboratory, whether it is  $10^{16}$  or not.

Lane: Well, the point is if we had a high flux reactor here, it would be another reactor .....

Weinberg: A high flux reactor always has some low flux in it.

Lane: It always has low flux, and this could be very useful for this kind of research.

Weinberg: And, in fact, if you have a really high flux reactor, then in principle you can always get that slow to fast ratio just as good as you want.

Blizard: You can afford to throw away neutrons.

Weinberg: You can get a very good thermal column in other words. Now when we built the ORR the question of a thermal column was discussed, and on the whole, people said that they didn't want the thermal column.

Lane: If one of those large engineering facilities were not needed, it is possible you could build yourself a thermal column in there. It is 5 feet in diameter.

Levy: But the thermal column isn't what we want. We want 0.06 ev neutrons in as large a beam as possible.

Lane: I don't understand then. You talked about the high ratio of fast to thermals. These aren't true thermals.

Weinberg: Fast to 0.06 ev neutrons. I bet that we will really have something nice in the ORR for neutron diffraction. How thick is that piece of beryllium there?

Charpie: Six inches. The heavy water is about the same.

Weinberg: Well, that is 15 centimeters, and the mean free path for the fast neutrons is way down.

Blizard: That means it is going to be an awful nuisance shielding all those holes.

Weinberg: That will be a problem.

Lane: Well, if there are no more questions, the seminar is adjourned.

## INTRODUCTION

After a brief discussion of future meetings, Mr. Lane introduced Mr. Harvey as the first speaker.

### VELOCITY SELECTOR EXPERIMENTS - J. A. Harvey

Before I outline the time of flight experiments that I feel would be worthwhile doing in a high flux reactor, I would like to emphasize that I am sure that in the period between now and the time the reactor is built there will be many other experiments which are a lot more valuable to do than the ones I will be talking about today. For example, when the Brookhaven, Argonne, and MTR people put in requests for fast choppers six or seven years ago, they probably just stated they were going to measure total cross sections. Out of these instruments, however, have come information fundamental to nuclear theory, such as the distribution of radiation widths, neutron and fission resonance widths, and the interference between fission resonances. I am sure this will continue to be true in the next five or ten years. Certainly if you ask anyone in the chopper business whether he could use a high flux reactor, he would emphatically say yes. Then if you asked him what he would do with it, he would say that he could improve the statistical accuracy of his measurements, improve the resolution of them, and would take the data somewhat faster, and hence accumulate somewhat more data. I feel the last point tends to be the least important one, at least here in Oak Ridge. At present, we can get about all the data we can handle right now, or we will with the ORR going, and consequently would not try to greatly increase the amount of data collected.

Now we also have a considerable amount of other equipment in addition to just a reactor for making cross section measurements, and one could ask if you could also make the same improvements by working on these other pieces of equipment and would it be cheaper? There is no question that one can make great improvements in detector design and probably a little bit more in the rotor of the chopper, but these tend to be quite expensive, too. If it were a question of increasing the flux of the reactor from  $10^{12}$  to  $10^{14}$ , then I am sure there would be no question of the advantage of the higher flux machine. But going from  $10^{14}$  to  $5 \times 10^{15}$  one has to be a little careful in justifying the higher fluxes for time of flight experiments. Consideration should be given to the possibility of improving the other pieces of equipment more cheaply.

I am sure you are all familiar with the compilation of cross sections (Report BNL-325 and its supplement). Referring to this report, it will be noted that there is a page for practically every element. A lot of them have been run with separated isotopes, and the question might be asked if one already knows enough about cross sections, and is there really anything more to be gained. Certainly if you ask this of the reactor people, you will find that they are never satisfied with the available data. They want more and better results, and as far as nuclear theory is concerned, more is needed. Let us first consider iodine as an example, shown on page 202 of

BNL-325. Figure 1 shows the total neutron cross section of iodine which we have obtained recently with the time of flight spectrometer at the LITR. In the energy range that I am principally going to be talking about there are many sharp resonances. At the lower energies, such as from 20 to 100 ev, there are many resonances, but at higher energies the number of resonances appears to decrease and even disappear. This is not true at all. What this simply means is that the resolution of the instrument is just not sufficient to see the higher energy resonances. These resonances are the order of 1/10 of an electron volt wide and are thousands of barns high. The resolution of the instrument with which the data in Figure 1 were taken was about 1.5 electron volts at 100 ev. So at about 100 ev one has a resolution some 15 times the width of the line. The situation gets worse much faster as you go up in energy.

Now to outline the principle of the time of flight technique one starts with a reactor which gives out neutrons. You make a hole into the reactor to get a beam of neutrons to come out and collimate it so it is a very narrow beam. This beam passes through what is called a "fast chopper" or a high speed rotor with a slit through it. It is only when the slit in the rotor lines up with the beam that you get a burst of neutrons through. The time of this burst can be made about a microsecond long. The neutrons which travel out along a flight path can be detected with some sort of detector at distances up to the order of 100 meters. The resolution of the equipment is given as the number of microseconds per meter. In this case, the resolution of an instrument with a 1 microsecond burst at 100 meters is 0.01 microsecond per meter. Several people talk in millimicroseconds now, and this would be 10 millimicroseconds per meter. Now the fast neutrons, of course, arrive there in very short times; the slower neutrons will take hundreds of microseconds to get there. These bursts are produced something like once every 5000 microseconds, which corresponds to about the maximum time for the slowest neutrons to reach the detector.

In order to convert into energy resolution, the following equation is used:

$$\Delta E = 0.028 E^{3/2} \left( \frac{\tau}{D} \right)$$

where,  $\tau$  = time of burst, microseconds

D = distance in meters

and  $\Delta E$  = energy resolution in electron volts.

So for the conditions outlined above, a resolution of 0.01 microseconds/meter would give an energy resolution,  $\Delta E$ , of about 0.3 ev. An instrument such as this would be a better instrument than any available at the present time.

[Lane: Pardon me. Is this instrument that you are describing a hypothetical one, or one that is actually achievable?

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NOTE: All questions and answers are enclosed in brackets [ ].

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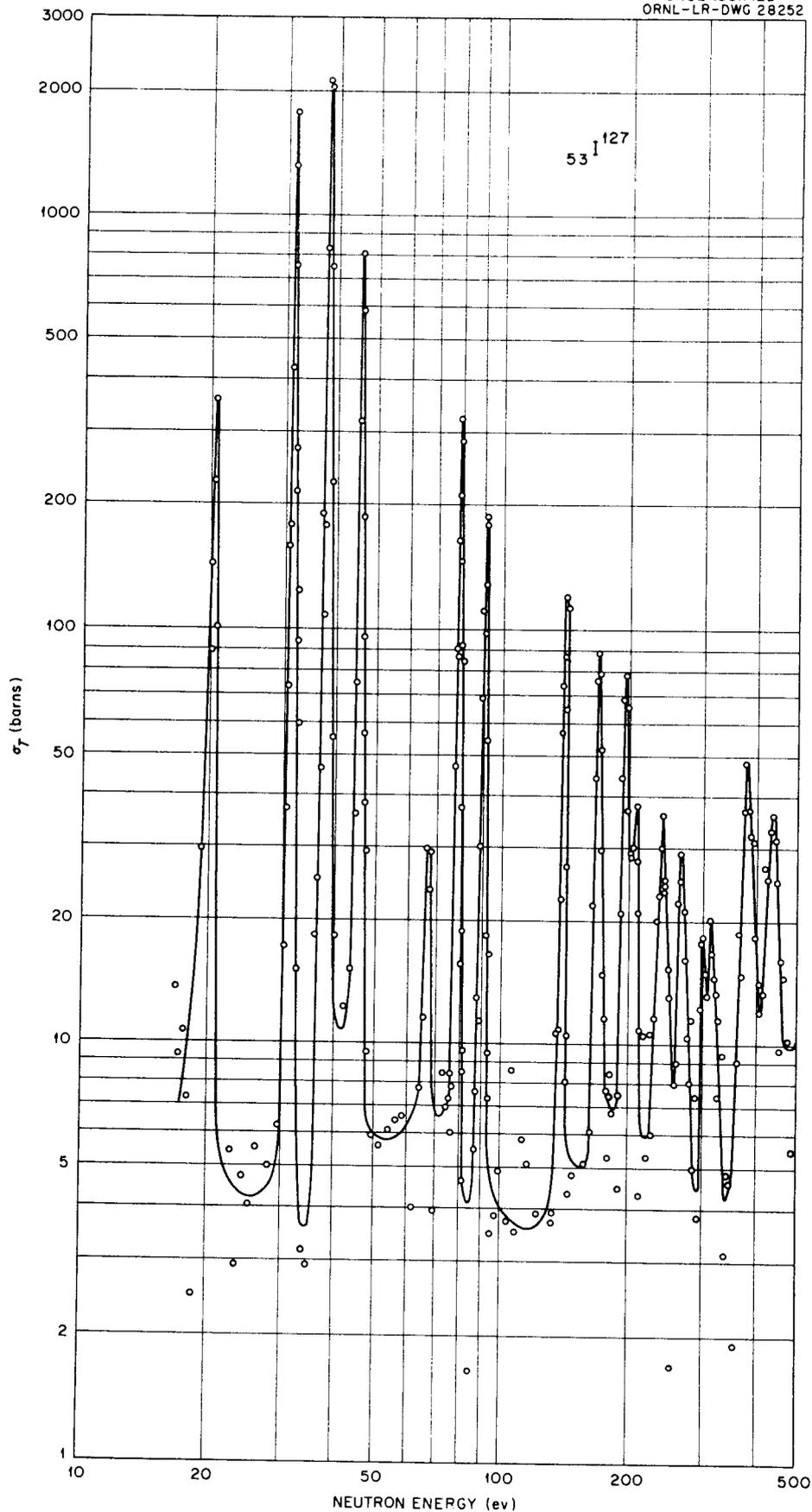


Fig. 1.

Harvey: It's achievable, but it's not available right now. It will be with the ORR, but I haven't exactly selected those parameters which we will use at the ORR. It is a hypothetical one to show what one will be able to do soon.

Lane: Is this because you are starting with  $10^{14}$  flux instead of the  $10^{12}$  you had in the graphite reactor?

Harvey: Yes, and it also includes some improvement of the associated equipment.]

Now to get a suitable counting rate for a piece of equipment like this one needs a reasonably high flux reactor, of the order of  $10^{14}$  neutrons/cm<sup>2</sup>/sec. This will give a counting rate of the order of 2500 counts per channel per day for a reasonable size detector. To detect neutrons arriving at different times, one needs a multichannel analyzer with some 1000 channels, and one records the number of counts as a function of time of flight which then of course corresponds to energy. Now, the counting rate is inversely proportional to the resolution to the 4th power, so if one wants to improve the resolution a factor of 2, other things being equal, the counting rate per channel decreases by a factor of 16. So one can use up flux very easily this way. With a factor of 100 higher beam intensity one would not use the extra intensity to get more data, but one would use it to improve the resolution. Large gains in flux are necessary in order to improve the resolution appreciably.

[Ergen: If you get more resolution, you need more data to draw a full curve.

Harvey: Well, the 4th power rule includes the fact that you take smaller channel widths and hence one needs an analyzer with more channels.

Snell: Well, this means that improving a factor of 100 in flux only means about 2 in resolution.

Harvey: It would mean a factor of 3 in resolution. The important thing is that the higher energy range can be covered with better resolution. With a factor of 100 in flux, one could cover twice the energy range with the same degree of accuracy. One would also be able to improve the data at lower energies.

Lane: Is this factor of 2 in range, or in resolution in a range?

Harvey: If the resolution with this hypothetical instrument at 100 ev is 0.3 ev, and if these resonances are 20 volts apart and are 1/10 of an ev wide, then a small resonance a few tenths of an ev away from another resonance would be missed. If one compared this to a spectrometer with 3 times the resolution, the same situation would prevail at 200 ev; the resolution would be 0.3 ev and a resonance a few tenths of an ev away would also be missed. One would be in the same boat at 200 ev as we are here now at 100 ev. On this basis, a factor of 2 increase in flux does not mean very much.]

Now, the energy range which I feel one would tend to work in five or ten years from now would probably be in the 100 volt range or higher. Because of this, thought should be given for our type of work to the suggestion that one build an intermediate type reactor in the center and have it well moderated around the outside, and try and increase the resonance flux in the center region by a large factor.

[Lane: You mean the higher energy flux?

Harvey: Yes, the higher energy flux - by not using light water, but using heavy water.

Weinberg: You might do lots better than that by using say magnesium or something like that.

Harvey: Right.

Lane: You need a neutron filter then.

Harvey: What you want is to moderate neutrons slowly so they live a long time in the several hundred volt energy range and stand a good chance of leaking out before they get further knocked down in energy.

Weinberg: Really what you are saying is that for your purposes perhaps a special type reactor, an intermediate energy reactor moderated by something with atomic weight 100, say a zirconium reactor, might be what you really want.

Harvey: I have never seen any calculations on such a reactor.

Weinberg: Well, I know that the French raised this issue, and have been talking about something of this sort.]

I feel that five years from now most of the region below 100 volts will be milked pretty dry. You can't have everything, so you say it's more important to study the high energy region.

[Weinberg: You can't have everything higher in energy though, because if you consider a zirconium-moderated reactor I would think you probably would be power limited. If you would get a factor of say 100 in your slowing down power, you would have to give up a factor of 100 in power per unit volume. How about it, Jim?

Lane: Well, can't you combine the two types though by using a special intermediate section in a thermal reactor in which you have a more poorly moderated area? That was suggested last time, I believe.

Harvey: But you probably need quite a large size volume which is poorly moderated.

Weinberg: This might be difficult because in an intermediate region the fuel density is very much higher than elsewhere.

Lane: You'll just soak up all the neutrons.

Weinberg: Yes. I don't know, it's something that perhaps deserves some very careful thought.

Blizard: Yes, but if you surround a bucket of zirconium with uranium and have a pretty good reactor outside of this, you should get a pretty high intermediate flux from the zirconium. If you drill a hole into the bucket of zirconium and look at that - into the middle of it - it might be a fine source for your experiments.

Lane: What you need is a resonance neutron column, like a thermal column with a converter at the outside so the neutrons enter as fast and are not quite slowed down completely.

Blizard: You really surround this thing with a converter so it becomes a three dimensional thermal column, instead of a one dimensional column.

Weinberg: It's something that should be looked at certainly.

Ergen: Maybe you can play some tricks with materials that have peak resonances at particular energies.

Harvey: Titanium has a very big resonance at 16 kilovolts where it goes up to hundreds of barns, and then it comes way down to about 2 barns I think. If you could slow the neutrons down fast till they get down to the kilovolt region, then you could let them live a long time.

Lane: Titanium you say?

Harvey: Titanium.

Taylor: What kind of a resonance?

Harvey: A scattering resonance.

Taylor: What width?

Harvey: Oh, 4 kilovolts about.

Ergen: Another thing. If you talk about special reactors you might consider a reactor which oscillates and just gives neutron pulses as you need them.

Harvey: Right.] This is the other thing I was going to mention. This other point is that we only take a microsecond pulse out of the reactor every 5,000 microseconds or perhaps 10,000. If you could build a reactor which, instead of operating at a constant 100 megawatt power level, goes up to the high power for as short as 100 microseconds, and then shuts off for 10,000 microseconds, you could gain a factor of 100 in flux for the same amount of heat removed.

Weinberg: What you are talking about is a factor of 100 in duty cycle, so we would say a 100 megawatt reactor could be improved 100 times which would be 10 million kilowatts.

Harvey: That is the same total heat, yes.

Weinberg: It is a little risky, though. (Laughter) Not quite crazy though, because the rocket engine people talk about such power levels. In fact, they talk about running at 10 million kilowatts for three minutes.

Ergen: The emphasis should be on "talk."

Weinberg: Joe Fowler, of course, is one of the inventors of a dragon of this sort, but I don't think he talked about quite this magnitude of thing.

Fowler: No, I was dealing with something else. But there has been talk of this magnitude of dragon, and the way the talk went it seemed safe enough. Fred Reines worked on this. What you do is put the critical material on fly wheels and turn them in opposite directions at high speed. You make your safety gadget so it is a period of the whole revolution of the fly wheel. If something happens and the power goes too high, it pulls out the safety control for the next meshing of the gears, so to speak.

Blizard: Sort of like Russian Roulette!

Weinberg: That's certainly an interesting matter that perhaps should be resurrected and looked at some more.

Question: Can't you do better with two fly wheels with slightly different gear ratios?

Fowler: Sure, that is what you do. You make them with say 100 and 101 teeth so that they come together once every 100 revolutions. People actually came to the conclusion that it was not particularly dangerous, and that was a long time ago when things seemed more dangerous than they really were.

Weinberg: Did Reines have cooling for his machine?

Fowler: I don't think cooling was provided.

Weinberg: He really wasn't talking in this power level range though.

Fowler: No, he wasn't talking in this power range.

Winters: You can scale the whole thing down and use air cooling though.

Weinberg: Well, what you are saying is that for this kind of business you can get the equivalent of 100 megawatts with 1 megawatt. It certainly is a worthwhile idea.

Harvey: Well, there is a statement around that the Russians are building something with about a 15 microsecond pulse, but I gather they are just going to pulse the whole reactor. They are not going to have a chopper to sharpen it up. They are talking about a kilometer or a thousand meter flight path at about 15 microseconds.]

If either one of these two features, a pulsed reactor or an intermediate flux reactor, could gain a factor of 10 or 100, it would mean a lot. Now, if we ignore these and ask what kind of experiments could we do with a  $5 \times 10^{15}$  flux reactor now, the most obvious one, of course, is a total cross section measurement. As I say we would improve the resolution, and if we pay that much more rent per beam hole, we could certainly justify putting more money into our detector and our time analyzer, so that we would end up with something like a 400 meter flight path, and we would use an 8,000 to 16,000 channel analyzer, and we could get a resolution of the order of 0.002 microseconds per meter. So with a factor of 50 in flux and a factor of about 4 in efficiency of detectors, this would approximately cancel the factor of  $4^4$ . Then with this instrument, with 0.002 resolution, at 100 volts we would have an energy resolution of the order of 0.06 ev.

Now, I forgot to mention one thing. The natural width of resonances is 0.1 ev, but there is always the thermal motion of the atoms in the material and this motion increases the observed width. This is called the Doppler width, which is of the order of 0.2 ev at 100 volts, and it goes up as the square root of the energy. Thus an energy resolution of about 0.06 ev would be much better than the Doppler width.

[Fowler: But you could always cool a sample?

Harvey: Well, the Doppler width goes only as the square root of the absolute temperature when it is greater than the Debye temperature. At low temperatures there is always the zero point energy, and you replace the temperature with the Debye temperature. Hence you can only gain by about a factor of 2, unless you could get real tricky and make your atoms vibrate at  $90^\circ$  to the direction of the neutron beam.

Weinberg: How would you do that?

Harvey: I don't know.

Taylor: Don't they do it with a molecular beam source with optical spectroscopy?

Weinberg: Yes, but they're not vibrating in that case, they're just moving.

Harvey: Most of the neutron measurements are made with solid samples.

Blizard: Do you really need very high density? If you are talking about a meter-long path, you ought to have lots of room for the sample.

Harvey: Yes, but we would need large quantities of material then.

Blizard: You've got lots of room.

Harvey: Yes, but the further back you go, the larger the diameter of the sample, and you still need so many grams per square centimeter, and if you increase the area you would need pounds of material.]

I have calculated a few other resolutions. At 400 volts I have 0.4 ev written down here; and at 10,000 volts it is 50 ev. Now this is the only way, using a reactor, to get neutrons less than 10 kilovolts in order to do cross section work. Above 10 kilovolts you can use Van de Graaff machines and produce monoenergetic neutrons. At present, Van de Graaff machines in this range can do maybe 300 electron volt resolution. Probably by the time this instrument is built the high voltage people will have improved their machines to get down to maybe 50 volts or so. The crossover is still somewhere around the 5-10 kilovolt region.

[Weinberg: The 300 ev is based on which technique? Is that based on Bill Good's technique?

Harvey: No.

Fowler: He is running now something around 7 or 10  $\mu\text{sec}/\text{m}$  maybe.

Harvey: I would say more like 10  $\mu\text{sec}/\text{m}$ . He would get around 250 ev resolution at 10 kev.

Fowler: But, he can certainly get down to 2 millimicroseconds.]

Now I would like to discuss the question of using the reactor instead of a pulsed machine like a cyclotron. The one example I was going to bring up is the Columbia cyclotron where they use 380-Mev protons and blast them into a tungsten target and produce lots of boiled-off neutrons, moderate them down, and then allow them to travel a certain flight path. Here you use the pulsed proton beam. Now they do much better to almost an order of magnitude as far as the pulse is concerned. They get more like a 0.1 microsecond pulse or better. The Columbia machine works about 35 meters, and it probably could go to 100 meters and still get fair counting rates; in which case they would end up with a little bit better resolution than what we would do with a reactor. But there has been this argument for the last ten years that the pulsed machines always look better on paper than the choppers. In the past ten years, however, the choppers have turned out five times more data than the pulsed machines. I think in the future it will probably be true that the choppers will do very well, even if the pulsed machines get better resolutions.

[Weinberg: Why is that really true? Is that because of the people involved?

Harvey: No, I think the technology gets tougher. We are talking about microseconds here. We only need half-microsecond channels or quarter-

microsecond channels which you can push with standard techniques. For the accelerators the time analyzer should have 0.05 microsecond channels and detectors must be in the millimicrosecond range. Choppers can use  $\text{BF}_3$  counters with  $\sim 1/2$  microsecond collection time. However, choppers require long flight paths to get high resolutions which is expensive. A reactor always has the advantage that it just sits and runs. The machines are down a lot of the time, and they are used for other things. The equipment that we use here at ORNL operates  $\sim 90\%$  of the time. It runs automatically, and once a day the sample is changed. Data are accumulated all the time. With the machine you may be on one week, and then off for a few months. A chopper looks good by comparison.

Weinberg: Well, one of the reasons one brings up this point is, suppose we do build a 900 million volt machine, would you prefer to use it? It would have a great deal better intensity than they have in the beam. What is the peak proton beam intensity in the Columbia machine?

Harvey: Havens quotes  $10^{18}$  as a peak neutron rate in the beam.

Weinberg: Protons?

Harvey: No, a peak neutron rate with 3 or 4 neutrons per proton.

Weinberg: Thus they have a proton beam of around  $3 \times 10^{17}$ , which corresponds to 0.03 amperes.

Fowler: They have a system of essentially storing their beam.

Harvey: By deflecting bunches upwards at different radii, you gain sort of an order of magnitude.

Fowler: But as Welton points out, this is possible with our cyclotron, too, by feeding it with more current continuously so that you use the same grid. You store the beam in some sort of ring and then put it onto a target all at one time; in other words, store it for a hundred microseconds then put it on a target, and per microsecond you get 100 times the average beam. This is also possible with the 900-Mev cyclotron we are talking about for ORNL, at least in principle it's possible; so one can increase the beam by this same factor.

Harvey: The 900-Mev machine has several advantages over the Columbia machine. There is a factor of 5 in the number of neutrons per proton because of the higher proton energy. There is about a factor of 10 in the instantaneous current that you could store if you went to the trouble of removing the beam and putting it in a storage ring, and still another factor of 2 if you optimize the geometry for producing moderated neutrons. With the Columbia machine the proton beam is swept up into a tungsten target and neutrons are moderated in a nylon block. The solid angle for getting neutrons into the nylon block is fairly small, and then you have

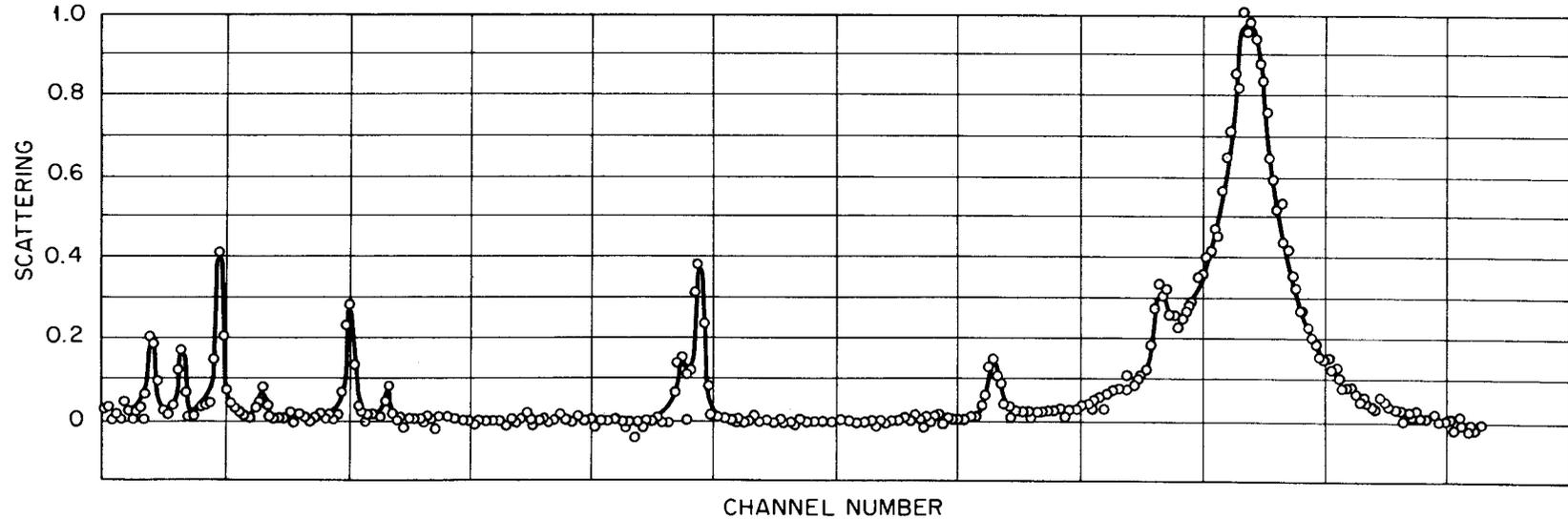
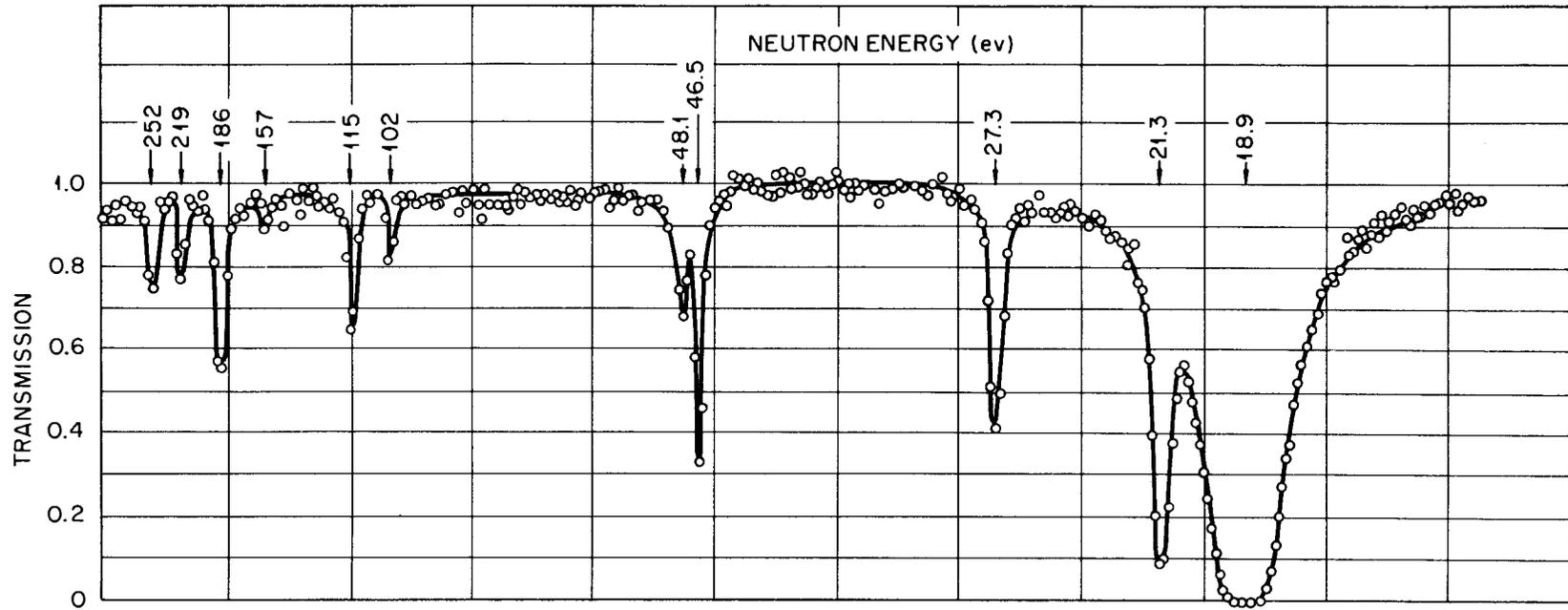
the other solid angle for getting them to the detector. With the Oak Ridge machine, one could probably better the intensity by a factor of 100 or a factor of 3 better in resolution, hence a resolution less than 1 milli-microsecond per meter. However, if you could only get the machine about 10% of the time you are at a disadvantage relative to choppers.]

Well, with the increased resolution one could certainly go up to higher energies in these total cross section measurements and get more and better data. Now, you say, "If you are just doubling or tripling the amount of data, there will not be anything new, and don't you already know what is going on up to 100 ev?" There is the point that at higher energies higher values of the angular momentum,  $\ell$ , occur and most of the work that has been done up to the present is just for  $\ell = 0$  neutrons. When we get in the higher energy region, 100 ev and up, then you can start getting the  $\ell = 1$  values, and the strengths of these resonances go up as  $E^{3/2}$ , whereas  $\ell = 0$  resonances vary as the  $\sqrt{E}$  on the average and so these  $\ell = 1$  resonances will start to come in more pronounced at the higher energies.

[Weinberg: Do  $\ell = 1$  resonances occur as low as 100 ev?

Harvey: Well, there is a little resonance at 10 ev in U-238 which is thought to be a "p" resonance. The barrier penetrability for this p-wave resonance is something like 20,000. Thus on the average a "p" resonance at 10 ev is about 20,000 times smaller than an "s" resonance. But you can always have a very strong "p" resonance and a very weak "s" resonance, and they appear the same strength. But as you go up in energy the "p" resonances get stronger so much faster that very likely in the 100 to 200 ev range, even though "p" resonances are still 1,000 times smaller than "s" resonances, one can pick up several of them and learn something about "p" wave resonances. Thus for total cross sections, not only do we just accumulate more data to get better statistics on resonances, but we learn something new about "p" wave resonances.]

To conclude, I can quickly list some of the other things that one would do. One could make scattering cross section measurements. Resonances in heavy nuclei have radiation widths  $\sim 0.1$  ev, and the scattering widths  $\sim 0.01$  ev. What one would like to do for nuclei which have a spin, such as Ag-107 which has spin  $1/2$ , is to find the way neutrons interact. The neutron can interact with its spin parallel or anti-parallel to that of Ag-107, and one of the best ways of getting this information is to measure the scattering. This is because the scattering cross section is proportional to the neutron width, whereas in the total cross section there is a statistical weight factor whose value depends on the direction of the neutron spin relative to that of the target nucleus. By making total and scattering measurements, you can solve for this factor and tell how many resonances are of one spin state and how many are of the other. We have started doing some of these measurements at the LITR. Figure 2 illustrates that it is pretty difficult even with the best example that we could select. This is a curve showing the scattering from tungsten in the energy range from about 15 to 300 ev. There are resonances in W-183 at 27, 46, 48 and



Transmission and Scattering of W.

102 ev. You can see that we have quite a high background here, and we are looking for resonances on top of this background. Since W-183 has a spin of  $1/2$  the statistical weight factor must be  $1/4$  or  $3/4$ . By comparing this scattering curve to the total cross section data one can determine the statistical weight factor of the resonances. For this run, the small resonance at 48 ev is about 2 times stronger in this scattering run than in the total measurements, and hence the spin of the compound state is 0 for this resonance and 1 for the other 3 resonances.

[Fowler: Is that scattering data?

Harvey: These are scattering data on tungsten.]

We can do the easy ones now at the LITR. When we move to the ORR, we will be able to do some of the slightly more difficult ones, but to do the whole works would certainly require higher intensity. Now we can reduce the background some, but we would gain more, of course, by increasing the neutron intensity and keeping the background constant.

In addition to these scattering measurements one could measure capture cross sections. Probably the biggest problem here is getting a good gamma ray detector. When you capture a neutron, you capture it up at 6 Mev excitation, and the compound nucleus loses this excitation either by the emission of a single gamma ray, or in general it decays with several cascading gamma rays. If you assume that the sensitivity of the gamma ray detector does not vary with incident neutron energy, then you just measure the counting rate as a function of neutron energy, and this is proportional to neutron capture cross section. But it would be far better to have an energy sensitive gamma ray detector whose efficiency is linear with energy. Then it wouldn't matter whether you got one high energy gamma ray with a certain efficiency or two lower energy ones of half the energy and half the efficiency. But I would say for capture measurements it is the detector that needs more development right now, rather than a higher flux reactor.

For fission measurements, of course, you can always argue that you can use more flux to improve their accuracy and to do various experiments with fissionable nuclei.

There are two other things that I would like to mention. One is to measure the gamma ray spectra coming down from these different resonances. There has been a little work done with a sodium iodide crystal, however since the energy resolution is  $\sim 8\%$ , the work at present is limited to the gamma rays quite low down near the ground state. There are just too many levels to do anything at higher excitation. You might ask whether you could use a Compton spectrometer or a pair spectrometer. I think in intensity one is still many orders of magnitude too low to do anything like the work that has been done with thermal neutrons.

I probably haven't answered the question of whether it is worth having the high flux reactor for time of flight spectrometry. In research I think

you would say it is worth having the high flux reactor, it is worth having greatly improved detectors, it is worth having everything. All you need is the money!

[Snell: Could you tell us about how efficient present day detectors are?

Harvey: The bank of  $\text{BF}_3$  counters that we plan to use at the ORR, which we know will work and with which we will be able to get a resolution of 0.008 microseconds per meter with counting rates of something like 2500 counts per channel per day, covering an area something like 10" x 32", will only be 3% efficient at 100 ev and only 1% at 1000 ev. Thus there is the choice of building a 50 times higher flux reactor or improving the efficiency of the detectors a factor of 50. Some of these detectors like the liquid scintillators, such as trimethyl borate, that Bollinger at Argonne National Laboratory uses are of the order of 50% efficient. However, it is very hard to build a large detector. Bollinger has built what is now considered quite a large one - 3" x 16". This is something like a factor of 6 smaller than the area we plan to cover with  $\text{BF}_3$  counters, but he does have 50 times the efficiency. However, there are lots of headaches that go with such detectors. The boron in the liquid scintillator captures the neutrons to give off a 2.7 Mev alpha, but in the liquid scintillator this only corresponds to something like a 30 kev gamma ray pulse. If you look at the scintillator with a single photomultiplier off to the side, you don't even see this 30 kev pulse above the tube noise. It is only when you put adjacent tubes in coincidence that you see a little peak sticking up above noise. The electronic instrumentation needed for this detector is quite complicated.

[Blizard: Why don't you use a solid scintillator?

Harvey: You mean like  $\text{Li}^6\text{I}$ ?

Blizard: Oh,  $\text{Li}^6\text{I}$  or  $\text{Li}^6\text{F}$ , for example, which is very fast.

Harvey: These are not cheap.

Blizard: At least they are cheaper than reactors.

Harvey:  $\text{Li}^6\text{I}$  is not cheap. To cover an area something like 50 square inches with  $\text{Li}^6\text{I}$ , 1 cm thick would cost about \$15,000. We would want five times this, or about \$75,000 worth of detectors.

Blizard: Is it really that expensive?

Harvey: With liquid scintillators it is the electronics that costs the money, you need fast and slow coincidence circuits, summing circuits, etc., and it gets complicated. A liquid scintillation detector to cover about 50 square inches costs about \$20,000 for detector equipment.

Maienschein: Well,  $\text{Li}^6$  has a very large Q-value so the gamma rays are no longer a problem.

Harvey: Right.

Blizard: It seems to me you could use a very thin LiI crystal.

Harvey: You need thick crystals in order to get efficiency; you need something like a cm thick crystal to get 9% efficiency at a kilovolt, and then it is quite gamma sensitive. This is the advantage of  $\text{BF}_3$  counters, you can have a fair amount of gamma ray background without having any trouble.

Blizard: Well, the same thing applies with the lithium.

Harvey: Not with  $\text{Li}^6\text{I}$ . If you want to get high efficiency, you have to have something like an inch thick.  $\text{LiF}$  would be better.

Blizard:  $\text{LiF}$  is now available. I thought they could now make it scintillate. Also the germanium crystals might work in a neutron device.

Harvey: We need big detectors. We talk about beams which are about 300 square inches. I do agree that the detector part of our program looks like the weakest part in the whole program. I wonder how much we would pay for a beam hole at a reactor with  $5 \times 10^{15}$ . I think the charge will be about \$6,000 per month or something like that for a beam hole at the ORR.

Blizard: Well, Dick Murray will be up on all these lithium things, certainly better than I am. I suppose you talked to him.

Harvey: Yes. We tried one out at the LITR with our old chopper, but it was hopeless because we had too much gamma ray background, and we haven't tried one yet with our new rotor, but we do plan to do this.

Weinberg: What is the situation with respect to the very fast neutron background in these small reactors? How much of a bother are they?

Harvey: You can get rid of it if you have enough space between you and the next fellow at the reactor by increasing the diameter of the rotor. We have an 18" rotor, and it has an attenuation for pile neutrons with our  $\text{BF}_3$  counters of  $2 \times 10^6$ . Thus we have a ratio with the rotor open to close of  $2$  million.

Weinberg: How does that compare with say the MTR chopper?

Harvey: We're quite a bit better, but I don't actually know their number. We plan to run with a duty cycle of something like 1 part in 5,000, and hence with a leakage of only 1 part in 2 million we get a background of 1 part in 400. So the fission neutron leakage is down pretty low. The background that we get is mainly room background or close to it. If the reactor had a much higher fast flux then we would just increase the diameter of the rotor to decrease the leakage through the rotor when closed.

Weinberg: Well, from your point of view the fact that the small reactors have lots of very fast neutrons is not really bad.

Harvey: No. We feel we can handle that adequately.

Weinberg: In contrast to say Henri Levy here who finds that a problem.

Harvey: It's probably the gamma rays that are more important to us even with  $\text{BF}_3$  counters. At time zero when the rotor is open we get neutrons and gammas through, and they give us quite a large peak at zero flight time. When we put a little piece of lead in the beam they disappear, thus proving we have more trouble with gamma rays than with fast neutrons.

Ergen: How about the data processing? If you had the ORACLE available all the time, could you hook it into your machinery so you took only the data which you really are using?

Harvey: Yes, we do this to some extent now. At the LITR we have a 256 channel analyzer, and we read that out every day. For these total cross section measurements we take an open beam run, and then a sample run. Whenever there is a resonance in the total cross section curve we get a transmission dip. Then we can read out these two runs on punched tape and feed them into the ORACLE. It will go through the tapes and list the number of counts we had on the open beam and on the sample run, subtract background, compute transmission, compute cross sections, convert time of flight to energy, and plot the transmission out on the curve plotter to show the resonances. We also have it compute the accumulated transmission, so if we want to know the strength of a particular resonance, we take the differences between two accumulated transmissions. But there is still a fair amount of work to be done. We run one of these runs a day, and it takes about a day for one girl and one of us to analyze and get the parameters out of these resonances. This is why I say that if we had 100 times the flux we wouldn't turn out 100 times the data. We just couldn't handle them. We would try and improve the quality of the data.

Ergen: Couldn't you somehow take a rough set and go over it once with not too many points, then make the ORACLE tell you where you want to take the next point?

Harvey: I think you are better off having lots of channels available right from the start, and just run the whole thing, and what isn't useful you just ignore.

Lane: If there are no more questions, we will hear next from Mr. Billington on solid state research.

SOLID STATE PHYSICS - D. S. Billington

I would like to make some general remarks about the design of the reactor. I would like to discuss this today, and then discuss experiments next time. My first remarks have to do with the problem of building high flux reactors, which is the thing that we are really worried about, and this has to do with the radiation damage involved in building a high flux reactor. Our group is always in the position of giving advice on what the radiation damage is going to be at fluxes greater than we are able ever to measure until a reactor of that flux is built. Now this is a bad merry-go-round to be on. Someday we are going to run into trouble. It may not be at  $10^{15}$  or  $10^{16}$ , but if we build one at  $10^{16}$ , then we are going to want to build one at  $10^{17}$ , and I don't know where the practical limit is as far as high flux is concerned. Well, the answer to this obviously is that if you want to really make high fast flux radiation damage studies you probably should do this on some sort of nuclear machine where you can get beams. We looked into the possibility of making use of A 48 out at Livermore, the old MTA setup. Unfortunately, they wanted \$6 million to fix this up and \$1-1/2 million a year to run it. It didn't seem practical offhand. There is, of course, one way around this business of the high flux, and this is simply that you do have some choice of materials. Our present high flux reactors operate very well because the materials have been selected carefully and because they operate at relatively low temperature. However, as the power level keeps going up and up, it will probably be possible to operate only at higher and higher temperatures. When you get into this situation then you worry about the problem of acceleration of reaction rates due to the vacancies induced by the radiation. You might very well get into a bad situation with something like creep, for example. Where at  $10^{15}$  or  $10^{14}$  flux you can hardly detect this, at  $10^{16}$  it might become catastrophic because creep rates which are increased a factor of 10 are catastrophic. You may be able to stand a little bit of creep in 10 years, but if you cut this down by a factor of 10 you have got the same amount in one year, and this will limit the life of the reactor considerably. This is just one of the problems involved in building high flux reactors.

Well, the other problem: I would like to comment just generally in relationship to the other people who have talked. They have usually put the emphasis on beam facilities, where the interest is in the thermal flux beam coming out of the reactors, so one is not concerned about what is going on inside the reactor. What we would like to see in a reactor is usually in opposition to what, say, the isotope people would like to have, or the neutron diffraction people would like to have. In general, we want high fast flux. Ideally, what we would like to have is essentially a hollow sphere or a cube. We want lots of places to play around in, and we want lots of fast flux. The higher the power goes in reactors, the more it becomes condensed. This just puts us in more and more trouble from the standpoint of experimental space. The other thing is the gradients in the flux become much more serious and the gamma heating will go up, and these things all affect the materials we are looking at very severely. Now it

is true that when you are looking at reactor materials you can say the gamma heating is part of the problem so you go ahead and study all of these conditions. But if you want to separate the heating effects from damage effects, then you have really got to study your sample as a function of temperature. So, what we would like to have in a reactor is not usually what other people would like to have in a reactor.

Lane: This problem of gradients is somewhat more a function of size of the sample that you are trying to test, isn't it?

Billington: Yes.

Lane: And so is heating.

Billington: Well, partly, yes.

Ergen: It depends on the power density and the size, doesn't it?

Billington: Well, this is a major problem in the MTR right now - to control the gamma heating. Sometimes you can take advantage of this, of course, and let the gamma heating heat the sample, especially if you want to make an irradiation at a high temperature. Wilson does this now in irradiating his impact specimens as a function of temperature.

Weinberg: The practical question is, is it not true nevertheless, in spite of the worse gamma heating at the MTR or LIIR, that the fact that you have more fast neutrons means that they are more effective instruments for doing irradiation damage than, say, the CP-5? Is this a fair statement, or is this not a correct statement?

Billington: Well, inside the fuel element of the CP-5 is a nice place to get good fast flux. Every place else is not too good from our standpoint.

Weinberg: Well, from your standpoint would you rather have a CP-5 or, let's say, an NRU, or would you rather have an MTR?

Billington: I'm not sure that we really like any of them.

Weinberg: Do you want to take on the job of building your own type?

Billington: Well, what we would like is what Harvey was talking about, essentially a poorly moderated reactor - something of this sort.

Weinberg: No, that doesn't help you very much because you need neutrons that are at least 100 kilovolts in energy, and an intermediate reactor is not much good for you. It is essentially a fast reactor you are talking about, and then you get into lots of other problems.

Wollan: Maybe you need a continuously operating Godiva type reactor with better space in which to work.

Weinberg: Well, you are again talking about nuclear machinery other than reactors perhaps.

Billington: Well, yes.

Weinberg: As a historical matter or a practical matter, do you have any information on the extent to which CP-5 has been used for radiation damage work?

Billington: I believe they are using the fast flux for such work.

Weinberg: At one time Argonne people would send samples down to the LITR.

Billington: That was before they made their fuel elements hollow inside.

Weinberg: I see. Now they get higher fluxes in the middle of those fuel elements than we do in the LITR?

Billington: I think it's roughly the same.

Lane: In these flux trap type reactors you get as much of a peak in fast flux in the fuel annulus as you do for the thermal flux in the center region and in the reflector. So somehow having your fast flux in one region and your thermal flux in another is a sort of compromise that might be very useful in your work. It is even better than the MTR in that respect or the CP-5.

Billington: If you just leave out the moderated core you have lots of space.

Lane: But not for thermals. Since you have to use both thermals and fast, you build a reactor which provides both.

Billington: We have talked about this in the Division on several occasions, and Holmes suggested the hollow sphere reactor which is lined with U-235.

Weinberg: Well, you recall one of the original designs for the MTR was essentially a hollow flux trap. Nobody saw how to make it, though.

Billington: Yes, it isn't clear how to do it even now.

Well, that's about all on the two remarks I wanted to make, first the problem of building a high flux reactor, and second that the people who are talking about the use of the reactor don't agree on how the reactor should look.

Weinberg: I would like to raise this point. You remember when we first started talking about the ORR, it looked quite different from what it now

looks. If we had built it that way we probably would have had it running now for two or three years. Which way do you think we are better off - having the present design and not running until the next month or so, or having the old design which you didn't like so much, but having it three years earlier?

Billington: This is a rough question. But I still say that, measuring creep under irradiation, it's something that you simply cannot do satisfactorily in the MTR or any other existing reactor. We will be able to do it here on the flat face of the ORR, which was one of the extra facilities provided.

Weinberg: You really feel that accessibility to the core of the ORR really makes it unique?

Billington: Yes. There are two unique aspects. One is the accessibility to the lattice itself, and the other is to the flat face. These things are really unique in this reactor at this level, and they are bound to be big advantages.

Weinberg: Do you think we would do well to just build a second ORR just like the present one? The Argonne people are proposing the same thing with a second CP-5, isn't that right?

Billington: I would think that all experimental groups represented here would agree that they would like to have the reactor operate the way they want; that is, they would like to have the thing go up and down or run at a certain power level to suit them. How many reactors you can justify on this basis I don't know.

Ergen: It still seems possible that with a relatively small reactor built for a specific purpose you could satisfy one requirement rather cheaply, and then this person could have his reactor for himself. It might conceivably be a better approach than one large reactor.

Weinberg: This is somewhat similar to what we did with the swimming pool.

Ergen: Yes.

Weinberg: It's no longer quite one person using it himself, though.

Lane: Since the experiments on solid state physics will be discussed at the next seminar, we can now adjourn this meeting.

HIGH FLUX RESEARCH REACTOR SEMINAR

March 7, 1958

INTRODUCTION

Lane: I would like to announce that the next seminar will be on April 11. Today Doug Billington will talk about research in solid states in an ultra high flux reactor, continuing from where he left off.

SOLID STATE PHYSICS, PART II - D. S. Billington

I mentioned in the last seminar some of our feelings about reactors in general, and today I will suggest some experiments that we might be interested in doing if we had a higher flux reactor. However, I will have to caution you that we have to specify how this reactor would operate in order for us to say whether we would really like it at these fluxes. In principle, a reactor that would operate at  $5 \times 10^{15}$  would be a very good thing as far as radiation damage effects in reactor structural materials, fuel elements, and things of this sort are concerned, for the simple reason that such a flux level is probably a factor of ten or more higher than one can expect to encounter in reactors that are being contemplated for the next few years, so that it does give you the chance to really run life-time tests on materials for these reactors, which is a very important consideration.

With respect to the effect of radiation on materials, an especially important consideration is damage by fast neutron flux. In this connection one mainly looks at the mechanical properties of reactor structural materials or metals. One of the things that would be interesting to find out is at what flux level do hardness, strength and ductility and things of this sort saturate. We have some feeling now that at fluxes of the order of  $10^{21}$  or higher many properties do reach saturation. But on the other hand, it would be nice to be able to go past this number by a factor of ten or more to make certain that we are really getting saturation and that there is no other phenomenon at these higher levels which we don't even know about.

Ergen: Just one thing, is  $10^{21}$  the integrated flux, nvt?

Billington: Yes, that is correct - nvt.

Well, we can get an integrated flux of  $10^{21}$  out at the MTR. However, it takes several months to get the samples back and forth. It sometimes runs into almost a year's time to do a complete irradiation at the MTR. A more readily available high flux reactor would be very valuable to us to study the effect of very high integrated flux exposures on the properties of reactor materials. So this general area, I think, is a field in which we are interested.

Another specific property I would like to talk a little bit about is the effect of radiation on the so-called "creep" in metals and alloys.

This has some interesting historical ramifications. In 1944, Andrade did a creep experiment on cadmium single crystals bombarded with alpha particles. He found that the creep rate of cadmium single crystals increased a factor of 5 or more after bombarding with an alpha source of  $10^6$  particles per second, which made everyone very much concerned about the whole business of creep. Shortly thereafter a number of in-pile creep experiments were done. In general a negative effect was found; the reactor radiation decreased rather than increased the creep rate. However, it has recently occurred to us that possibly if both the temperature and the flux are high enough one might expect an acceleration of creep rate. The basis for the argument is something like this: At a given temperature you have a certain equilibrium number of vacancies present in metals or alloys. By bombardment you introduce an excess number of these vacancies. If the relative fraction of these excess vacancies is high enough one might become quite concerned. A second factor is now becoming pretty well established; namely, that a secondary or steady state creep takes place by a diffusion of vacancies to edge dislocations. This allows dislocation climb and slip to take place. So there must be some relationship between the vacancies that are thermally present and the creep process. If you get an excess number from irradiation they might contribute to an increase in rate of creep, although this is only suppositional and has not yet been verified by experiments. The activation energy of the creep and for self-diffusion are essentially the same, so by inference there must be a close relationship between vacancies and the creep rate.

One thing that got us to worrying specifically about this was a letter to the editor in the Journal of Applied Physics by G. Schoek, of Westinghouse, who insisted that even at a flux of  $10^{13}$  there should be an increase in creep rate. After analyzing his equation we have come to the conclusion it is off by several orders of magnitude, by neglecting recombination and sources of additional vacancies from straining the crystal. However, by neglecting these factors he showed that at half the absolute melting point the thermal- and radiation-produced vacancies approach each other in number.

Lane: Do you suspect his results?

Billington: No, this is just a calculation I am referring to, not an experiment. He is trying to derive what the effect of radiation would be on the creep process as a function of flux.

Lane: But you suspect his calculation is wrong?

Billington: Oh yes.

Guth: You didn't do any experiments did you?

Billington: Well, we have done some experiments at a flux of  $10^{13}$  and have seen in general negative effects. However, Wilson has shown an indication towards getting a slight increase at elevated temperatures.

Question: With the same metals?

Billington: No, this is with Inconel, nickel, and stainless steel.

If we assume that one gets about  $10^{-9}$  vacancies per second by irradiation with respect to an equilibrium number of  $10^{-6}$ , then the ratio is 1 in 1000. The ratio here is not very low so that the effect of the additional vacancies should be low. But you see if this were at  $10^{13}$  then by the time you get to  $10^{16}$  the ratio would approach 1, other things being equal. So you say maybe it would be very well to have real high fluxes to test this point because as you get these higher fluxes you are liable to go higher and higher in operating temperature anyway, so it may become an important reactor problem. The creep that is of interest is the secondary or steady state creep, the creep that shows a linear extension with time for long periods of time. When a structure is used under stress, this stage of creep determines the useful lifetime of the structure. In engineering structures, the rate of extension is of the order of 1 part in  $10^6$  per second or less, e.g., 1% creep in  $10^4$  hours =  $3 \times 10^{-7}$  second. But Wilson is doing a lot of his general engineering type studies on all types of metals and alloys and observes that you get a general decrease in ductility. So the question arises: Could you possibly get a decrease during creep test too so that you might just shorten its life arbitrarily? We should be doing more creep tests than we are doing now, irrespective of flux, for two reasons: One is to study the increase in creep rate, and the other is to study the loss of ductility in creep tests.

Another property along this same line is brittle fracture, which is of practical importance. Here again you argue from a reactor standpoint, the life of the reactor to be as long as possible at as high a flux as possible. Yet radiation induced brittleness is a function of the amount of radiation. The irradiation pushes the temperature up at which a metal becomes brittle, as well as reduces the impact strength. These properties are representative of the important properties of interest both basically and technically. However, in passing, I would like to mention one other class of properties, the Young's Modulus and elastic constants. We don't have any good tests on the theory or the effect of radiation on elastic constants because we have not been able to produce sufficient defects to see an effect. We would really like to bombard at high fluxes in a short enough time so at least we could do more than one experiment during a lifetime. Here again it would be of practical, as well as basic, importance.

Probably the person in the Solid State Division who really needs the high flux reactor the most is Mark Wittels. The type of work he is doing involves bombarding various oxides, silicates, quartz, and diamond. He has to make extremely long exposures. For the MTR, a typical exposure is six months or so. Here one might gain a factor of 15 since this becomes one to two weeks in the new high flux reactor. So he will be in a position to survey a lot of materials and develop them in a short time. The reason I make a little bit of an issue of this is because there are so many potential ceramics that could be used in reactor development, as well as those that are of basic interest because of their structure; and until the theory of solid state is in much better shape than it is, we have no option for many purposes other than to irradiate them to see what happens.

Blizard: Will the results of one set of irradiations tell what the effect will be in other cases?

Billington: Well yes. Sometimes it can be a function of a lot of things. As you irradiate a certain type of structure, then this will tell you something about other structures or other materials of the same structure.

Blizard: I am wondering what would be the advantage of being able to do these things, especially if you put an awful lot of samples in the ORR and just waited a year for the results?

Billington: In order to learn from the first experiment what to study in the second experiment.

Blizard: If you have to do them in sequence then I can see it would take a lot of time.

Billington: In many cases for engineering purposes, true, you would irradiate a lot of them and determine, say, the effect on thermal conductivity and pick out those least affected; but if you were really trying to understand what is happening, then one experiment would probably suggest another. Then the other important variable, of course, is doing all these experiments as a function of temperature. Because it makes a big difference whether a sample is irradiated at helium temperature or irradiated near the melting point.

MacPherson: In this type of experiment is the examination time decreased as the number of samples irradiated increases?

Billington: No. It would be a lot more. Well, it does bring up another question, as a matter of fact, and that is, pretty soon you start getting heavy exposures and in a short time your samples are mighty radioactive. Now we often operate at a distance, in unshielded laboratories. Pretty soon we are not going to be able to do this, so we are going to need more shielded facilities. Even right now we can foresee the need for shielded X-ray diffraction equipment for ORR experiments.

Wollan: Are all your experiments of the form that require large nvt rather than nv?

Billington: This is the other general point. We have never quite cleared this point up. If we had a year's operating experience in the ORR we might be able to answer that question. Mark Wittels thinks in connection with some of his studies on  $ZrO_2$ , diamond also, that flux is very important and not the total dose. He starts seeing changes in diamond at the MTR much quicker because the flux is higher.

MacPherson: Higher, you mean at this time?

Billington: Well, in this particular case he is getting more damage.

He thinks they were done at the same temperature although he didn't say how. Our feeling about the radiation annealing is that it should take place faster at a higher flux and not the other way around.

Question: I was wondering is a longer irradiation at the same nvt, does this mean flux is more important than total dose or is temperature more important?

Wollan: It is mainly a matter of the back reaction in combination with the flux and temperature.

Billington: We think this is a problem, but we have no way of documenting it at the moment. In many of the materials including a lot of metals and alloys, as far as we can tell it just looks like total dose is most important except as you go back to the creep experiment. It has to be flux dependent.

I suppose that's about all on the mechanical property aspect of the problem. Many other properties are of equal importance.

I would like to mention one other category, what we would call solid state reactions, because it points up two things that are different in radiation damage experiments. In one case we do experiments at a low temperature; then the damage you see is a direct function of the incoming particle knocking things out of the way. But if you raise the temperature then the defects start moving around, and then they initiate various solid state reactions and you get a secondary effect. If you irradiate the sample at an elevated temperature at the same time, then the reaction rates may be considerably enhanced. It's a little bit like the creep problem. You begin to worry about reaction rate and at the low fluxes as we look at things now, we see an effect beginning to be initiated. The question is, what is going to happen if the flux goes up by a factor of 100? Something is going to happen for sure. But at the moment, we are not able to predict it. We haven't had a good chance to study flux dependency between  $10^{12}$  and  $10^{14}$ . It is a little hard for us to argue uniquely that we have got to have  $5 \times 10^{15}$ . Maybe a year from now after we have worked in the ORR on creep and the various phenomena like precipitation hardening and things of this sort we will be able to say very clearly that we should go higher or that we don't need to go higher to illustrate the point.

MacPherson: Due to gamma heating don't you introduce a temperature control problem when you start working at higher fluxes?

Billington: Yes. This is where we might amplify our original statement, because when we say high flux we want high fast flux with little or no gamma heating. Unfortunately, these things kind of run along together. From a practical standpoint a lot of these extraneous features are most important as far as radiation damage is concerned. If you can't cool it, it's going to melt.

MacPherson: With respect to flux dependence properties, I can see that an order of magnitude increased effect may be readily perceived; however, for

small effects an accurate knowledge of the flux is necessary, otherwise you may have a very large error in your estimated nvt.

Billington: This is true. Let's say we know the flux a lot better in X-10 than at the MTR. We never know for sure what we are getting out there. It changes from day to day. On the other hand, we don't get very high fast flux out at the MTR. We get high thermal, and this is very useful for fuel element studies but not for fast neutron damage because they won't let us get in the active lattice like we will be able to in the ORR.

MacPherson: Why is that?

Billington: Well, they won't let us in there with a dynamic experiment. Some exposures have been made, true. Well, I'm not sure that we have ever been in the active lattice - we have been out at the L-47 piece, the place where we do most of our irradiations.

Lane: Have you thought of using a converter out there to get a high fast flux?

Billington: Well, not at the MTR. No!

Incidentally, Oscar Sisman told me just the other day that in the L-47 piece at the MTR the thermal flux is  $7 \times 10^{14}$  now. They have raised the power to 40 Mw.

Guth: Is this the highest flux in the MTR?

Billington: I don't know. I couldn't find this out, but this is essentially an increase of a factor of two.

Answer from someone in the group: Yes, it must be.

Billington: Yes. All I am saying is that it is in the L-47 piece.

Lane: They must be running above 40 megawatts out there. It is only about 5 at 40, about 5 or 6.

MacPherson: When did this increase occur? Do you know?

Billington: Within the last two or three weeks.

Ergen: One question with respect to the effect on diamond. If you made artificial diamonds, in theory, at reasonable temperature, the diamond would be the stable configuration if the pressure is very high. At reasonable temperatures the rate of transformation is so small we can't wait to make diamonds. Then we have to go to very high temperatures and the pressures get fantastically high. Now with neutron irradiation, if the pressure is high enough, can a stable configuration be reached faster even at room temperature?

Stoughton: Are you talking about making them from graphite?

Ergen: Yes, making them out of graphite.

Billington: Our observations, of course, are the other way around. You destroy the diamond structure under irradiation at low temperatures.

Ergen: Yes, but you don't make them under pressure. You destroy them under conditions where diamond is unstable.

Billington: We worried about putting a number of things under pressure and irradiating them at temperature. So far we have not done an experiment because there are several other things we would like to try. This would be an interesting experiment with quartz.

Crawford: Do you use quartz to get the pressure?

Billington: We know we get between 10 and 15% expansion in quartz under irradiation. We can use this to generate the pressure.

Schweinler: A short estimate indicates that the pressure would have to be about  $10^4$  atmospheres at room temperature, using my recollection of the difference in energy of diamond and graphite. Now, is that a feasible pressure for pile irradiations?

Comment: That isn't very high.

Billington: No, I would think that this isn't impossible.

Question: Even in a small volume?

Blizard: If we can make diamonds, to hell with atomic energy! (Laughter)

Billington: I think the diamond trust is still in control of the situation.

Snell: You say you have not used a converter in the MTR. Are you planning to use one in the ORR?

Billington: No, we are hoping to do experiments adjacent to the active lattice and would not need a converter.

Snell: Likewise, a converter won't be necessary in a  $10^{15}$  reactor.

Billington: Well, will they build us a flat sided tank?

Snell: A converter would have problems in itself.

Billington: Any  $5 \times 10^{15}$  reactor would have to be liquid fuel because the burn-up rate would be so high.

Lane: No, not necessarily. But that will be covered at the next meeting. Recent studies by Internuclear indicate that  $10^{16}$  fluxes can be achieved at ~100 Mw in a solid fuel reactor and with a reasonable fuel cycle. You try to keep the power level as small as possible to reduce furn burn-up. However, the reactor design studies at ORNL did not indicate as advantageous a situa-

tion. The Internuclear report on this design conducted in connection with the Advanced Engineering Test Reactor has not yet been issued.

Blizard: But these reactors are thermal reactors!

Lane: We also get a high fast flux in the annular core due to the high power density.

Ergen: Is there a report available covering this information.

Lane: One will be issued soon by the Internuclear Company. This is in connection with the Advanced Engineering Test Reactor studies for the A.E.C.

Billington: What kind of moderator is used?

Lane: The design calls for a  $D_2O$  reflector and  $H_2O$  as the coolant and internal moderator.

Blizard: How do they get such high fluxes?

Lane: I think it is essentially because they are keeping the size of the island small to get high flux but this limits the size of the experiments.

Ergen: Well, in light water, if the thing would go critical would a flux be possible like that?

Lane: Well, see your studies took on a light water reflector, they had a heavy water reflector which changes it completely and makes it go critical. By keeping the power level down they keep the fuel requirements down and get a better enhancement of the flux. Well, go ahead Doug, we'll go into that next time.

Billington: Well, just one other point I would like to make. I think our strongest argument for a very high flux reactor is perhaps we would be in a position to do more beam hole experiments - in other words, to bring out a high intensity beam of fast neutrons. Then, this solves a lot of our experimental problems.

Blizard: This again points up the fact that maybe a higher reactor would give a strong beam rather than a plain old high flux reactor.

Weinberg: Is not an accelerator better for a beam?

Billington: Is it cheaper?

Weinberg: Well that's a good question - depends on what you are talking about.

Billington: I must confess I don't know at this point. But anyway, the whole point in this case is the ease of experimental manipulation. If you can move the experiment in and out of the beam, research is much easier. I think you can get a lot of very important answers by such a technique. We would like to see a high fast neutron flux reactor built for this purpose.

Weinberg: You are talking now about a reactor that is essentially unmoderated.

Blizard: It is probably moderated in most directions but not around the diffractor.

Weinberg: Well, if they're moderated you don't have as many fast neutrons.

Blizard: Yes, but you put a converter in the region where you don't do experiments.

Lane: A flux trap reactor is essentially a thermal device.

Fowler: But you can always convert to fast neutrons.

Weinberg: Have people really looked at the question of a poorly moderated reactor.

Ergen: How about the fluoride reactors?

Weinberg: That, perhaps, is the argument in favor of a fast reactor because you can also get to  $10^{15}$  from the MTR, but the power and flux rate is a little higher than it is in a fast reactor.

Wollan: You speak about going to a beam then the fall off in intensity is much larger than the factor you gain by upping the power.

Billington: Yes, I think you would be able to gain so much more by doing a better job on designing the experiment.

Wollan: Wouldn't such a facility require shielding?

Billington: Oh, yes! This would be a very elaborate facility.

Fowler: Well, it would almost certainly have to utilize the converter idea.

Holmes: Well, what I thought I had in mind was if you want thermal beams, you would put  $D_2O$  along the channel.

Blizard: Perhaps using nothing would be better.

Weinberg: I was thinking of zirconium or something like that.

Wollan: One other advantage to a beam; it gives a chance to do something that we can't do now very effectively, and that is to determine the energy spectrum of the neutron.

MacPherson: Do all your beam experiments require a high fast flux?

Billington: No, not all of them. We are really in a kind of contradictory situation. For lots of experiments we ask for lower and lower fast flux compared to thermal.

In experiments that Thompson and Holmes are doing, such as dislocation pinning, they are down to  $10^8$  and  $10^9$ . These are very basic experiments in trying to understand interaction between defects.

Along another avenue we face the problem of gamma heating. We want to do experiments at very low temperatures, and we want as high a flux as possible. These things seem to be somewhat incompatible.

Holmes: Well, could I ask a question? You are saying that a high fast flux reactor is difficult in some way to design. I have often wondered just what the real disadvantage is. Why can't one build a hollow sphere type reactor in which the fuel is on the inside of a spherical shell, some relatively heavy substance just on the outside, and outside of this a moderator? In this type of reactor, the fast flux would be directly equal to the number of fission neutrons produced per square centimeter of the wall.

Blizard: Yes, but in order to improve the flux inside the sphere, you want to have fissions contributing from as far away from the surface as possible. In order to increase the flux in that core you want to make the mean free path for fast neutrons in the material surrounding the core as small as possible. It is just the product of the mean free path and the power per unit volume that determines the flux. This is one reason, of course, why one must find a coolant and fuel which do not moderate. Now we might find something that is high enough in atomic weight, as Dr. Weinberg points out, so that it doesn't moderate to very low energies, but only down to the inelastic scattering cross section.

Boyd: Well, the Clementine reactor was a mercury cooled reactor which was a very nice fast reactor.

Weinberg: That was really a different ball park though.

Boyd: Yes, but it certainly has many advantages you just talked about.

Holmes: Yes, but this reactor I am thinking about wouldn't necessarily be a fast reactor but would be a thermal reactor. Those wanting thermal neutrons could work on the outside and those wanting fast neutrons, on the inside.

Blizard: I think maybe combinations like this would be very good. If you could have the thermal neutrons come in from one side of the sphere and have the fission take place on the other side.

Lane: You are just talking about a shell reactor with a hole in the middle.

Holmes: Right. If you have enough space inside the shell, such a reactor is ideal for the solid state business because you can shield out gammas. The nice part about a shell reactor is the geometry. No matter how big you make it, the flux is still the same for the same number of fissions per square centimeter on the wall. Thus, you can solve the cooling problem.

Billington: Well, to continue, we would like to have a higher fast flux to look at the initial damage rate of materials and its flux dependency to find out if there is a saturation effect. We would also like to look at that effect on the whole category of solid state reactions. This means a combination of radiation and temperature increases. I might point out that we are primarily just trying to look at the interaction of various types of defects, some of which we introduce by irradiation. We are being led to the conclusion that we should look at material that does not have any defects in it to start with, something like metallic whiskers. These presumably represent theoretical metal.

Weinberg: Hasn't any radiation damage work been done on them?

Billington: Well, we hope to start some.

Weinberg: Nobody else has done any?

Billington: No.

Weinberg: They're so thin that you could use charged particles to great advantage.

Billington: Anyway, we don't introduce dislocations by bombardment. What we really get with metallic structures is an interaction of defects. By studying alloying effects in a theoretical metal like a whisker and carrying out transmutation studies at very high fluxes, you really get to look at alloying the way it really is instead of getting complications like interactions with dislocations.

Lane: Where do you get these metallic whiskers?

Billington: Well, they were originally discovered at Bell Laboratory. They were noticing short circuits in some of their telephone lines because they had cadmium plated fittings and the little metallic whiskers that grew out from the cadmium short circuited the instruments. Somebody got curious about these and got to looking at the whiskers and realized that they had strained them several percent. In normal metal, the elastic strain is very, very small - a couple of thousandths of an inch or so. When whiskers do finally flow, they just kind of "kink". If you heat them back up again, they pop right back in shape. The strength of these materials agrees with theoretical calculations. You gain a factor of ten essentially in strength compared to normal metals. My old theme song is that we should spend lots of money working on whiskers. This is where the future of materials research lies.

Guth: Who has done the experiments to show that the strength of whiskers reaches the theoretical limit?

Billington: The people at Bell Laboratories and General Electric Company. The trouble is that they are only several microns in diameter and at the best, only a half an inch long.

Guth: Isn't there a variation in properties of these whiskers?

Billington: Yes, there is a wide variation in the character of these whiskers. They have also found out that the larger they get, the weaker they become, presumably because of the presence of more dislocations.

The other point that I want to make is that we are interested in high thermal fluxes as well as high fast flux, partly because of interest in use of converters; also we'd like to reinforce the arguments of Wollan and Levy for higher flux for use in neutron diffraction. We'd be interested in looking at structures like beta brass. Copper and Zinc are indistinguishable by x-ray diffraction techniques. At present flux levels, we can't get enough intensity to give us the resolution we need. In the ORR we might be in good shape but we don't know yet. Here again it seems straightforward that the higher the flux, the better the resolution that can be obtained.

There is another type of experiment that is of some interest to us. At Brookhaven they have started some neutron transmission experiments. In this case, one filters out all but the longer wavelength neutrons, then you can look at the scattering by defects directly, then this becomes an absolute method for counting defects. Their flux is a little bit low; they can't quite look at the wavelength dependence. We think that at  $10^{15}$  this type of experiment would be in good shape.

One can also make a good argument for high thermal flux reactors in regard to fuel element evaluation. For reactors that are to be operated at  $10^{14}$  flux, then in a  $10^{15}$  reactor good lifetime studies of the fuel elements can be made.

A good study of "phase reversion" could be made also. By phase reversion, I mean that when certain alloys such as Uranium - 12% Molybdenum are irradiated in the room temperature equilibrium phase, the radiation changes the structure to that structure that can be obtained thermally only above approximately  $700^{\circ}\text{C}$ . This also happens in Uranium-Niobium alloys. The situation is similar to Mark Wittels' findings in  $\text{ZrO}_2$ .

The Westinghouse people have observed with these alloys that occasionally the high pressure-high temperature water corrosion resistance of these alloys fails shortly after removal from the radiation field. This may be correlated with the radiation induced high temperature phase going back to the equilibrium room temperature state. When out of the radiation field, i.e., the higher temperature phase is corrosion resistant, the low temperature phase is not. They also observe anomalous volume increases under irradiation. Often times greater changes thus can be attributed to the fission gases.

These alloys could be readily studied using a real high flux reactor because of the shorter irradiation times required to do a comprehensive study.

Weinberg: What material is this?

Billington: I think it's noticed in the U-Nb alloys with a little zirconium added.

Weinberg: This is something new then, because it has been thought that niobium is a poor material to add to uranium because it leaves its corrosion resistance. Uranium by itself is no good in hot water because of radiation damage and the crinkling effect and because of corrosion resistance. Alloying materials added to uranium to improve dimensional stability are usually no good from a corrosion standpoint. This was always the argument against using niobium which increased the dimensional stability but reduced its corrosion resistance. Now you are saying the reverse is true.

Billington: What I am saying is that as long as the U-Nb is in a radiation field its corrosion resistance is better.

Weinberg: Does this mean that the statement about corrosion resistance only applies to out-of-pile conditions.

Billington: Yes, that is correct.

Weinberg: Well this is quite a new invention isn't it?

Billington: Well, no one quite understands this; but there are a number of such instances. Some time after the reactor goes down, a day or so, samples become susceptible to corrosion.

MacPherson: A similar effect is noted on the oxidation of graphite. Irradiated graphite oxidizes at a lower rate than fresh graphite by steam or some other oxidizing agent outside the reactor. Yet radiation damaged graphite in a gamma field has increased oxidation.

Weinberg: Homogeneous reactor people should take note of this.

Billington: I think that is about all I have to say. Although we are not in a position to make quantitative arguments concerning the value of higher flux, we can at least make some qualitative arguments in its favor.

Snell: With regard to the relative cost of a high flux reactor and an accelerator, one should remember that a high flux reactor will not be cheap.

Schweinler: Before we leave, I would like to say a word about thermal neutron transmutations in solids. It is not clear whether this is a subject which the Physics Division people or Chemistry, as well as Solid State people, are most concerned with.

The decade before the war saw the development of knowledge of perfect crystalline solids. The decade after World War II saw the development, which is still continuing, of the knowledge of solids with one-dimensional defects, that is, dislocation lines. People with neutrons to transmute atoms have a tool for making point defects in solids. When you have a transmutation, the substance which is transmuted first of all is of adjacent Z, so it fits into the structure well. Secondly, as a complication, it is almost always an interstitial atom when first formed, because of the capture of gamma-ray and beta-

neutrino momentum recoil, so that one has the complication of getting the atoms back into their perfect crystalline sites (as imperfections of  $\Delta Z = 1$ ) before he can examine their effects on the substance. But the rewards are also fairly high, because when one has changed the ratio of electron number to atom number in the crystal he is really changing the position of the Fermi level, something which has not been studied at all thoroughly to date.

Most ionic crystals and valence crystals probably form with the Fermi level about midway between the conduction band position and valence band position, or farther down towards the gap. With electrons we produce a situation in which the Fermi level rises; that is, the electrons from beta-decay produce a situation in which the Fermi level rises to the bottom of the conduction band in most cases. This class of substances has not been studied to any appreciable extent as yet, and one needs an appreciable amount of conversion, the order of a per cent or a fraction of a per cent, to produce these substances which are at present largely unknown.

Weinberg: Why do you need an ultra-high flux reactor for that experiment?

Schweinler: Of course, one doesn't need an ultra high flux reactor. The product of  $nvt$ , the total number of neutrons crossing a square centimeter, and the absorption cross-section,  $\sigma_a$ , is what gives the fraction of atoms transmuted.

MacPherson: Can this be done easily by mixture and diffusion?

Schweinler: One always has difficulties with segregation coefficients, knowing whether the substance really goes into the mass or whether it is segregated on dislocation lines or on grain boundaries.

Blizard: Is there also the problem of getting the atoms back into their lattice points?

Schweinler: This I mentioned is one principal problem. When you start with single crystals, one hopes that these problems are minimized and that one can saturate defects like dislocation lines and then look at the "perfectly doped" crystals.

Lane: Well, if there are no more questions, the seminar is adjourned.

HIGH FLUX RESEARCH REACTOR SEMINAR

April 11, 1958

NONAQUEOUS SYSTEMS - W. K. Ergen

The title of "Nonaqueous Systems" is maybe somewhat misleading. It was selected quite a while ago and has not changed, even though the scope of the work has increased. I became interested in the problem of a high flux reactor about two years ago when the Laboratory was interested in a small scale effort directed toward an investigation of various applications of molten fluoride reactors. At that time it was thought that fluoride reactors and molten salt reactors, in general, would have certain advantages for high flux reactors, particularly if ~~a~~ high fast flux is desired. If you have a fuel region such as shown in Figure 1,

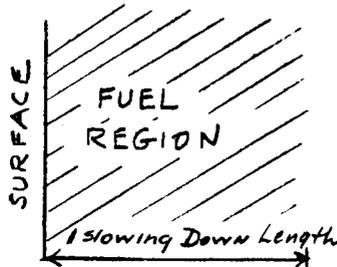


Figure 1

then the fast flux at the surface is proportional to the watts per square centimeter of this surface, or in the neighborhood of the surface itself - neighborhood being defined as the distance of an order of magnitude of the slowing down length. Fission neutrons generated at a large distance from the surface - larger than the slowing down distance - will not reach the surface as fast neutrons. So the fast flux comes from neutrons which are generated within approximately one slowing down length of the surface.

It appears that molten salt reactors have two advantages if you want a great number of watts per square centimeter as just defined. In the first place, the watts per cubic centimeter - the power density - which can be attained in molten salt reactors is rather high; and secondly the slowing down length is large, so the watts per cubic centimeter times centimeters, or in other words, watts per square centimeter, is large. Because of these two aspects, one would expect that a molten salt reactor would be suitable for high flux reactors. The results of this early work have been summarized in a memorandum written some time ago<sup>(1)</sup>. -With respect to the feasibility of such a molten salt research reactor, the group headed by Mr. MacPherson established very quickly that the work on the airplane reactor project had solved many problems of the molten salt reactors, but not those which guarantee long term operation (such as how you fix the reactor, take it apart, and

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(1) Ergen, W. K., "ANP Steering Committee Meeting of May 6, 1956", ORNL Report No. CF 56-6-4.

reassemble it remotely). Everyone finally realized that to build a research reactor fueled with molten fluorides would require a long-term effort. For this reason my work took the direction of doing some rather elementary calculations to find out what can be achieved in the way of high flux if certain limitations are imposed on either the power density or the total power available. This work is being currently reported in CF memos (2-5). There was also an interim report given at an Applied Nuclear Physics Seminar about January 22, and I will, in the course of today's seminar, briefly review what I said at that time. However, I would like mainly to emphasize what I have been doing since then. The status of the work is such that it is a very good time to report on the various items which I have considered.

One of the things that I have done since the last seminar was to consider a homogeneous reactor as contrasted to the flux trap reactor originally considered and reported in the earlier memoranda mentioned. By homogeneous, I mean a reactor that is mathematically homogeneous and can be described by one region in which the cross sections are constant. These cross sections may be space averages of varying cross sections. For instance a block of BeO, penetrated by equal and equally-spaced fuel elements would be regarded as homogeneous in this connection if the distance between the elements is small.

In such a homogeneous thermal reactor the power density  $p$  is given by

$$p = \frac{\Sigma_f \phi}{3.1 \times 10^{10}} \text{ watt/cm}^3 \dots \quad (1)$$

where  $\Sigma_f$  is the macroscopic fission cross section, in  $\text{cm}^{-1}$ , and  $\phi$  is the thermal-neutron flux, in  $\text{n/cm}^2 \text{ sec}$ . For a given power density, the thermal-neutron flux can be increased only by reduction of the macroscopic fission cross section, that is by diluting the fissionable material. If there were no absorption other than the absorption in the fissionable material, and if the reactor were infinite in extent, the macroscopic fission cross section could indeed be reduced without limit.

However, even in an infinite reactor, parasitic absorption of neutrons in the moderator, structures, impurities, etc., sets a limit below which the macroscopic fission cross section  $\Sigma_f$  cannot go. For  $\Sigma_f$  smaller than this limiting value  $\Sigma_{f\ell}$ , the reactor would not be critical.

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- (2) Ergen, W. K., "Flux Distribution in a Reactor Consisting of a Spherical Shell of Fuel in a Lifetime in an Infinite Moderator", ORNL Report No. CF-57-12-100.
  - (3) Ergen, W. K., "Fluxes Obtainable in a Flux Trap Reactor", ORNL Report No. CF-58-1-4
  - (4) Ergen, W. K., "High Thermal Neutron Fluxes from Fission Oversimplified Cases", ORNL Report No. CF-58-2-127.
  - (5) Ergen, W. K., "Flux Trap Reactor with Absorber in the Center", ORNL Report No. CF-58-3-27.
  - (6) Ergen, W. K., "Homogeneous High-Flux Reactor," ORNL Report No. 58-3-68.

If  $\Sigma_m$  is the macroscopic cross section of this parasitic absorption, then  $\Sigma_f \ell$  is given by

$$k = \frac{\nu \Sigma_f \ell}{(1+\alpha) \Sigma_f \ell + \Sigma_m} = 1 \dots \quad (2)$$

Here  $\nu$  is the number of neutrons per fission, and  $\alpha$  the ratio of capture cross section to fission cross section. From Eq. (2),  $\Sigma_f \ell$  can be computed and this limiting value can then be substituted into Eq. (1). If  $p$  is then interpreted as the maximum permissible power density, the maximum obtainable flux is given by

$$\phi_{\max} = 3.1 \times 10^{10} (\nu - 1 - \alpha) \frac{p}{\Sigma_m} \dots \quad (3)$$

With the "U.S. values" for  $\nu$  and  $\alpha$ <sup>(7)</sup> of U<sup>235</sup>,

$$\phi_{\max} = 4 \times 10^{10} \frac{p}{\Sigma_m} \dots \quad (3a)$$

Thus the maximum flux which would be obtainable even in an infinite reactor would be  $4 \times 10^{10}$  times the power density over the parasitic absorption cross section. If one takes the idealized case where the only parasitic absorption comes from the moderator itself, and of course all practical moderators do have absorption cross sections, (neither Helium 4, Carbon 14, nor Beryllium 10 are very practical) one can then compute very easily for the various moderators that are of general interest what the maximum flux would be. For this theoretical case, Table 1, column 2, lists for the five most common moderators the maximum thermal flux obtainable with a power density of 100 w/cm<sup>3</sup>. The values of  $\Sigma_m$  were computed from Table 1, Ref. 2, on the basis of  $\Sigma_m = k \Sigma_D$ .

TABLE 1  
THERMAL-NEUTRON FLUXES OBTAINABLE WITH VARIOUS MODERATORS

Moderator	Power-Density Limited	Total Power Limited	
	Maximum Flux in n/cm <sup>2</sup> sec for 100 w/cm <sup>3</sup>	Optimum R (cm)	Maximum Flux in n/cm <sup>2</sup> sec for 100 Mw
H <sub>2</sub> O	0.201 x 10 <sup>15</sup>	27.9	2.3 x 10 <sup>15</sup>
D <sub>2</sub> O	60.4 x 10 <sup>15</sup>	76.4	2.8 x 10 <sup>15</sup>
Be	3.56 x 10 <sup>15</sup>	55.3	3.3 x 10 <sup>15</sup>
BeO	5.36 x 10 <sup>15</sup>	59.6	3.5 x 10 <sup>15</sup>
C	10.2 x 10 <sup>15</sup>	90.5	1.7 x 10 <sup>15</sup>

(7) Hughes, D. J. and Schwartz, R. B., "Neutron Cross Sections," BNL-325, Supplement No. 1, p. vii, Brookhaven National Laboratory, Upton, L.I., New York.

But this applies to infinite reactors. If you have a finite reactor, Eq. (2) has to be replaced by,

$$\left[ \frac{\nu \Sigma_f}{(1 + \alpha) \Sigma_f + \Sigma_m} \right] \left[ \frac{e^{-B^2 \tau}}{1 + L^2 B^2} \right] = 1 \dots \quad (2a)$$

where B is the buckling,  $\tau$  the age, and L the diffusion length. Since  $e^{-B^2 \tau} / (1 + L^2 B^2)$  is smaller than one, the expression  $\nu \Sigma_f / [(1 + \alpha) \Sigma_f + \Sigma_m]$  and hence  $\Sigma_f$  has to be increased as compared to the value given by Eq. (2). In other words, the neutron losses by leakage necessitate an improved competitive position of the fissionable material relative to the parasitic absorbers. If  $\Sigma_f$  is increased, and more fissionable material is used, the flux obtainable for a given power density decreases below the values given by Table 1, column 2. This is, of course, very much idealized. However it is doubtful whether higher fluxes than this can be achieved for the given conditions.

Lane: That's for a homogeneous situation?

Ergen: Yes, for a homogeneous reactor.

Lane: You can do better in the non-homogeneous case.

Ergen: Yes, that is probably correct. I have not investigated systems where you either mix H<sub>2</sub>O and D<sub>2</sub>O in homogeneous mixture or else use them as proposed in the reactor you are going to describe -- have the H<sub>2</sub>O inside the flux trap and the D<sub>2</sub>O outside. So you can beat it in heterogeneous reactors, or you maybe can beat it by using combinations of moderators, of which I think the only practical one would be H<sub>2</sub>O-D<sub>2</sub>O mixture.

Let us now consider the case in which the total power P is the limiting factor rather than power density. For a spherical, bare homogeneous reactor of radius R, cm

$$P = \frac{4}{\pi} R^3 \left[ \frac{\Sigma_f \phi}{3.1 \times 10^{10}} \right] \text{ watts } \dots \quad (4)$$

$\Sigma_f$  is calculated from Eq. 2a, which can be transformed into

$$\Sigma_f = \Sigma_m \left[ \frac{1 + x^2 / K^2}{\nu e^{-x^2} - (1 + \alpha)} \right]$$

$$x = \pi \sqrt{2/R}, \quad K^2 = \tau \Sigma_m P$$

If you now substitute the  $\Sigma_f$  in Eq. (4), you find that for a certain radius there is an optimum. The reason for this is obvious. If the reactor is too large then even for a moderate power density you would get a very high power or conversely if you are power limited the power density would have to be very low which results in a low flux. If the reactor is very small and if it goes critical at all, then you have to have a very large uranium concentration again resulting in a low flux. In between there is an optimum. The radii for these optimum reactors are shown in Table 1, with the flux attainable at 100 Mw.

It is interesting to note that the wide variation in  $\phi$  max which you observe if you are power-density limited does not exist if you are limited instead by the total power. One can, of course, easily conceive of reactors with much higher fluxes but these require a higher total power. Numbers are for fluxes at the center, not the average fluxes. These rather simple calculations, are worthwhile to give an idea of the actual limitations. You also may note that the various moderators do not vary widely in performance. Carbon which is a very practical moderator does not give quite as high flux as the rest of the moderators.

Turning now to another subject, I should like to briefly review what has been done on the flux trap reactor, frequent mention of which has already been made in these seminars. As you know, Mr. Lane will discuss the flux trap reactor in some detail next time. This type of reactor consists essentially of a shell which contains the fissionable material and this is surrounded by a reflector which you usually make as thick as you possibly can so that it is practically infinite. On the inside you also have a moderator. This region is called the flux trap, sometimes also referred to as the island or the internal thermal column. This reactor differs from the reactors studied by George Safanov of Rand, inasmuch as these involved a cavity in the center of very low density material which is advantageous for certain purposes. In our reactor, part of the neutrons which are borne as fission neutrons in the shell, diffuse into and slow down (according to the age equation) inside the internal thermal column or flux trap. Thus there are some thermal neutrons borne some distance from the shell that have to diffuse to the shell and in doing so set up a flux gradient. This means that the flux is higher in the center than at the edge. The flux gradient at equilibrium is just high enough so that the neutrons which are borne are carried away by the neutron current sustained by the flux gradient. This would be the case if there would be absolutely no absorption. If there is absorption in the moderator, then some of the neutrons get eliminated by the absorption and the flux in the center would be less. Consequently, a flux trap reactor would use as low an absorption cross section moderator as possible.

Lane: I don't quite understand that because  $H_2O$ , which doesn't have a low absorption cross section, has the highest peaking in the center.

Weinberg: It is the slowing down power that determines the peaking.

Ergen: Water doesn't go critical though in this arrangement.

Lane: I see. You're just talking about the same moderator on both sides.

Ergen: I have only investigated the case of the same moderator on both sides. If you have different moderators inside and outside or if you mix light and heavy water then you may get something that is conceivably somewhat better than the figures which I have so far.

Lane: But I think Weinberg's point is still true -- it's not just absorption of the moderator. It is something like the ratio of slowing down power to absorption.

Ergen: This is right.

Weinberg: I think it is actually the ratio of square root of  $\tau$  to the mean free path against capture or something like that.

Ergen: Yes, that is right,  $\sqrt{\tau}$  times  $\ell$ , the reciprocal of the diffusion length.

Weinberg: Well, I don't think that's too relevant for us.

Ergen: Well, the physical reason why this is so may be relevant because if you have a very large  $\tau$  then the reactor gets very big and the power density gets low for a given power. Or conversely, you need lots of power for a given power density. This is where the water is advantageous. However, by itself, it doesn't go critical. On the other hand,  $D_2O$  does better than going critical and it is obvious there is some mixture which will give the optimum.

Well, the first thing that I did in making these calculations was to idealize the situation by collapsing the shell into an infinitely thin region, which means that the dimensions are small compared to the slowing down length and the diffusion length of the moderator. However, I assumed the shell was black to thermal neutrons and therefore is thick compared to the diffusion length in the shell itself. This is actually not a bad assumption, as will turn out later. If you take this idealized picture you can follow the history of a neutron which is borne in the shell, slows down in the moderator and comes back to the shell from either the inside or outside and you can compute the probability of this neutron returning to the shell rather than being absorbed or disappearing into infinity. This probability, of course, is connected with the multiplication factor,  $\eta f$  which you need to make the reactor critical and you find that  $\eta f$  is a certain function of  $K$  and  $\rho$ , where  $K = \sqrt{\tau} \ell$  and the radius,  $\rho$  is the actual radius in centimeters divided by the square root of  $\tau$ . Without repeating the mathematics, which are covered by a memorandum, the flux at the center,  $\phi_c$  can now be calculated as well as values of  $\eta f$ . The center flux can be represented by the equation  $\phi_c = F_2(K, \rho) / D\sqrt{\tau}$ , where  $F_2$  is a function of  $K$  and  $\rho$ . Now, the fact that this diffusion constant  $D$  and the square root of  $\tau$  enter in this manner means that the flux is lower if you decrease the density. This is especially of some importance in the case of either aqueous or  $D_2O$  reactors because with hot water the flux will go down. In the case of graphite it is also important and here an idealized density of 2.0 g/cc was used to represent maximum conditions.

The results are reported in CF 58-1-4 and summarized in Table 2.

TABLE 2

Idealized Flux-Trap Reactor with Radius Adjusted  
to Give Maximum Central Flux

<u>Moderator</u>	<u>Density g/cm<sup>3</sup></u>	<u>Multiplication Factor</u>	<u><math>\rho</math> (see text)</u>	<u>Radius cm</u>	<u><math>\phi_c</math> at 100 Mw</u>
D <sub>2</sub> O	1.1	1.474	2.08	23.3	3.8 x 10 <sup>15</sup>
Be	1.85	1.944	1.95	19.2	7.2 x 10 <sup>15</sup>
BeO	3.0	1.833	1.99	20.4	6.9 x 10 <sup>15</sup>
C	2.0	1.780	2.00	30.5	3.1 x 10 <sup>15</sup>

If one reverses the figures in the last column of Table 2 to calculate the power required to get  $10^{16}$  flux, values are 262, 138, 145, and 315 megawatts, respectively, for the four moderators considered.

Weinberg: To what total power does the  $3.8 \times 10^{15}$  correspond?

Ergen: This is for 100 Mw.

Weinberg: I thought for the  $D_2O$  in the previous case it was  $60 \times 10^{15}$ .

Ergen: No, that was for the limitation of 100 watts per cc.

Weinberg: Yes, of course. I see. But if one only gets  $3.8 \times 10^{15}$  per 100 Mw in  $D_2O$ , the flux trap isn't such a great advantage after all.

Lane: But it does turn out you can do better than this by using a combination of light and heavy water, better than anyone of these single moderator combinations.

Ergen: Yes, this light and heavy water case I was not able to finish. This is, I think, something that still has to be looked into. Of course, your homogeneous reactor gets better too.

Lane: A study by Internuclear indicated that at about 70 Mw they could get fluxes about  $5 \times 10^{15}$ .

Weinberg: With a flux trap?

Lane: Light water inside and heavy water out, which is about twice as good as the  $D_2O$  case considered here.

Ergen: This sounds perfectly reasonable.

Lane: Could you revise your calculations to handle mixed moderators.

Ergen: Yes, I can do that.

Lane: Inside and out?

Ergen: Yes. This can be done very easily.

Lane: Maybe it would be nice to do the one case of light water inside and heavy water out to make a comparison with these results you have just presented.

Ergen: I will do this, but it has not been done yet.

Wollan: How do you cool a solid moderator such as uses beryllium or beryllium oxide and still keep it homogeneous. I don't quite understand this.

Ergen: Well, these are academic reactors.

Lane: What he means to say is that it's rough.

Ergen: Well, actually these reactors are reasonably large; if you could visualize a reactor with tubes containing a non-neutron absorptive coolant....

Lane: Helium would not disturb the neutron flux.

Ergen: No, helium would not disturb the flux, but it would disturb the density. A more practical case would be a reactor which has a beryllium moderator and has tubes of fluorides going through it. This would be an engineering embodiment of this concept.

Wollan: But you would have to divide it into a lot of parts so you would have lots of tubes, so it seems to me that by putting something through you have already mixed the moderator, haven't you.

Ergen: Yes; however, the amount of fluoride in there would not be very large compared to the amount of other moderators there.

Wollan: Even with a homogeneous mixture of uranium and beryllium?

Lane: There's no uranium in the beryllium.

Ergen: One of the ways this idealized thing could be approached practically would be to have a beryllium matrix and have coolant tubes of fluoride. Now as long as the total amount of fluoride is not very large compared to the amount of beryllium oxide, then your approach is idealized.

I should mention that these maxima all correspond to a certain required value of  $\eta f$ , the multiplication constant and though the beryllium gives the highest flux, it must be said that with Be,  $\eta f$  comes dangerously close to the maximum that one can hope to obtain with  $U^{235}$  (i.e., 2.06). This, of course, would be the  $\eta$  for  $U^{235}$  itself with the thermal utilization,  $f = 1.0$ . Whereas with  $D_2O$ , with lots of multiplication to spare, you could mix it with light water to decrease the  $\tau$  and obtain a somewhat better effect.

Well, this was a very much idealized case and furthermore it applies to the case where you have neutrons at the center of the reactor but do not extract them in any appreciable amount. This might be of interest, for example, for exposing a very small amount of einsteinium but not enough to depress the flux.

In many experiments, however, the flux is depressed by the sample. To find the effect of this sample, one can represent it by its effective radius. In other words, you assume that the sample is a black sphere which has a certain radius. The radius of this sphere should extend to a point where the flux goes to zero. With this assumption, solve the diffusion equation and compute the number of neutrons which the sphere would absorb and compare it with the number of neutrons it would absorb if the flux depression were absent. If you do this, you find that the flux depression is proportional to  $D$ , the

diffusion constant. Consequently, the factor of D in the equation for central flux, which has been mentioned previously, is just cancelled. You can then compare the moderators on the basis of the center flux multiplied by the diffusion constant. This is a relative unit which will be of interest in the case where you do have a large absorber in the center. This gives in arbitrary units a value of 4.4 for D<sub>2</sub>O, 4.5 for Be and BeO, and 3.1 for carbon. Except for carbon, all the moderators are about the same and the large differences between BeO and D<sub>2</sub>O disappear. So the relative advantages of Be and BeO disappear if you use a flux trap reactor with a relatively heavy neutron absorbing sample.

Now these calculations were reported last January, and a question was asked at the end of the seminar concerning the effect of abandoning these idealized conditions which the calculations have assumed. To answer this we did some three region - two group calculations on the ORACLE. The results will be soon published in a memorandum<sup>(8)</sup>. To summarize our findings I can show what happened in the case of beryllium. First we took a reactor which looked very much like the one just mentioned and estimated that the center flux would be  $10 \times 10^{-4}$  n/cm<sup>2</sup>sec per fission neutron in the shell for this very much idealized case. This is practically the same as was computed taking the radius equal to the radius which had given the optimum for a Be-moderated reactor. Then we gave this reactor a finite shell thickness and we filled this shell successively with nonmoderating fuel, with fluoride fuel, specifically Flinak (LiF-NaF-KF), and then with D<sub>2</sub>O. This figure of 10 was then changed to approximately 7.9, 7.2 and 6.4, respectively. The thickness was computed to get a flux of  $10^{15}$  based on a power density of 1 kw per cc and  $10^{-4}$  n/cm<sup>2</sup>sec per fission neutron. In the next calculation we made the shell ten times as thick so as to get a flux of  $10^{16}$  with the same power density. This was the way it was computed and it turned out that the thickness in the first case was 2.6 centimeters. In the second case, for the same three fuel mixtures, the center flux went down to 4.7, 3.5 and 2.3, compared to 10 in the idealized case.

Lane: What was this second case?

Ergen: The latter case is with the same fuels but the shell thickness is increased in such a way that the volume is increased by a factor of 10. In other words, one was a reactor where the power density was almost 100 watts per cc, and the total power was selected in such a way that it would give in the idealized case  $10^{15}$  and  $10^{16}$ , respectively.

So you can see that increasing the thickness without moderation does drop the flux somewhat, that it drops even further as the moderation is increased. The same pattern is followed for the  $10^{16}$  flux reactors except that the greater thickness has a stronger influence. Now this in itself didn't look so good because we did lose quite a bit of this value of 10. The flux went down almost by a factor of 4 in the extreme case with the thick shell. We then were somewhat dismayed by the fact that the fluorides showed an appreciable drop and I ascribed this first to the fact that I used Flinak which has a very large diffusion constant. If you look at it with molecular eyes, it is

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(8) Ergen, W. K., "High Flux Reactor Machine Calculations" (ORNL CF-58-4-7).

a very fluffy substance. So I tried to use a fluoride, namely sodium-zirconium fluoride, which had a smaller value of  $D$ , but the situation was not improved. The second thing which we tried was to reoptimize the radius. We took the same thickness and varied the inner radius. We found that as this radius was decreased we did reach a maximum of the center flux per fission neutron in the shell and this maximum was equal to 9. So by reoptimization one can retrieve the effect of at least a moderate amount of moderation and effect of the finite size of the shell. I should however mention that this optimization was really not quite accurate because the thickness remained constant as the radius changed which did mean that as the radius became smaller the power density went up. However, I don't believe it made much of a difference.

Lane: Did you do it for the thicker shells also.

Ergen: Yes.

Lane: Then you can extrapolate in between. What was the result of the fluoride for the thicker shell case? Was that also nine?

Ergen: It was not quite nine, but it did come out to 7.5.

Lane: Since you can vary to that limit, then certainly the  $10^{15}$  case wouldn't be changed. But the  $10^{16}$  case would come down.

Ergen: It does come down somewhat but by reoptimization you can improve the situation to a certain extent.

Wollan: Are you talking about thermal flux?

Ergen: I am always talking about thermal flux.

I should mention very briefly that one more elaborate calculation was made using a thirty-one group code to see whether the idealization of the two group method was justified and it did turn out that all the cases came back to about 10. Assuming that this multi-group calculation was fairly realistic, this indicated that the idealized case was a good approximation. This was, of course, not obvious to begin with.

To mention other calculations performed, we did use the Cure Code at K-25 to get the influence of a hole or vacuum. However, data have not yet been plotted. For everybody who wants to use the Cure Code, this was the first time it was used in a realistic problem, in fact, we very much simplified it. It did turn out that once the machine got the problem, it ran beautifully and very quickly. It would be highly desirable, however, to get a machine which could break up the grid of this two-dimensional code because the people who had to do this had a terrible time. Once the machine had this, and had it in the right way, it worked quite well.

Finally, I should say where I hope to go from here. One loose end that is still left is the question of hydrogen and deuterium as a mixture or in different regions. Apart from this, I think I am at a good stopping point now,

and it becomes a question of where do we go from here.

One item which should be considered in this seminar is the question of whether it is better to build a bigger multi-purpose reactor or a small single-purpose reactor which would be designed for a specific purpose and then would be entirely under the control of those who intend to use it.

Weinberg: What you are saying is, is it basically better to build five \$5 million reactors or one \$50 million reactor.

Ergen: I'm not saying exactly this, but I would just like to consider the question in a general way.

Weinberg: When you put it in quantitative terms, it seems to me that five ORR's are better than one large reactor such as the Canadian NRU.

Ergen: I agree.

Winters: Do you get the NRU flux in the ORR?

Weinberg: Sure, you get the NRU flux in the ORR. As a matter of fact, the NRU cost \$60 million and the maximum thermal flux is  $3 \times 10^{14}$  or just about the same as the ORR.

Lane: Well, it may be more difficult to get approval to build five separate reactors. One way to get around this is a multiple core reactor. In the proposal of the Internuclear Co. for an advanced engineering test reactor they put seven reactor cores inside the same tank of heavy water and considered whether this would cost more or less money than seven separate reactors. I believe it was less with the multiple core reactor.

Weinberg: Underlying the whole discussion that we have been having all this time, is the question of which reactor do we really want - a  $10^{15}$  or  $5 \times 10^{15}$  flux reactor, or more space - say at  $10^{14}$  flux.

Ergen: This, to some extent, depends on the experiment, and in this connection, I asked myself what would a reactor look like if it were specifically designed for transuranic isotope production compared to one specifically designed for neutron diffraction experiments, for example. With respect to the neutron diffraction experiment, I made a back-of-the-envelope calculation to see how the number of neutrons produced per second in a reactor, say like the ORR, would compare with the number which you actually count. The factor which I got was  $10^{18}$ . Now I don't swear it is  $10^{18}$ , but I would like to call your attention to the order of magnitude of the order of the order of magnitude. It certainly appears promising to work on this factor of  $10^{18}$  and maybe reduce it to  $10^{17}$ . This might be more promising than hiking up the reactor power and the reactor flux by a factor of 10.

Lane: You say you only count  $1/10^{18}$  of the neutrons you actually produce?

Ergen: Yes, of the neutrons in the ORR and in an experiment such as Wollan does.

Wollan: We thought once though on that same point, that by doing something with collimation you could improve the ratio. But you might need a special reactor designed for it.

Ergen: Yes, this is the point I would like to make. However, even were you to design a reactor specifically for neutron diffraction experiments and not for anything else, it probably would incidentally be suitable for other things too.

Wollan: If you've got a few million dollars to play with though you can invent quite a few along those lines too. However, the way it is now with just a few people considering these things, one just can't do much.

Lane: That's a good point because we've been looking at going up in power levels rather than at other ways of making improvements.

Winters: It is not a new issue.

Ergen: No, not a new issue at least to others, however, this came into my mind through the seminar. It may have been known before, but I didn't realize it before. Well, unfortunately before I can do anything about a reactor specifically designed for neutron diffraction experiments I have to learn more of neutron diffraction and this will take some time. Consequently I started out studying a reactor designed specifically for producing transuranic elements. I would like now to make some additional points on this subject which I discussed in the seminar in January. There are in a way additions and also some corrections to my early remarks.

Generally speaking, as Mr. Chetham-Strode pointed out, if you make transuranic elements, you start with Pu 239 and produce Pu 240, 241 and then you get to Pu 242. This then by neutron absorption gives Pu 243 which very quickly decays to Americium-243 and by neutron absorption you get to Americium-244 and this decays to curium. Since curium isotopes up to Cm-249 are long lived, one goes up to the curium chain by successive captures to Cm-249. You do this with all the chains of even atomic number Z and there is a short transition over the odd chains. In the case of the even Z chains one notices that with the exception of Pu 240, which has a resonance near thermal, the even-even nuclei have a small thermal cross section, whereas the ones with an odd number of neutrons have very large absorption and fission cross sections. So if you try to produce transplutonium elements in a thermal reactor, you quickly run into these even-even nuclei as sort of a bottleneck because of their low thermal cross section. You need a high flux to get through those, but at these high fluxes the fissionable or odd-neutron containing nuclei will very rapidly be burned out. So the idea which occurred to me was that maybe you should go to higher energies and build a reactor with a higher spectrum. Taking the statistically anticipated cross sections for such even-even nuclei, from Weinberg's and Wigner's book, and comparing them with the fission cross section of Pu 239 at 100 ev, the ratio is 0.7. For 1000 ev, the ratio is 0.6.

At thermal energies, the ratio is also very low.

Lane: What is this ratio?

Ergen: This is the ratio of the absorption cross section of even-even nuclei divided by the fission cross section of Pu-239. Of course, the statistical theory is not good below 100 ev, so this is why there are no numbers available. As far as the book goes, however, the optimum would be at 100 ev. In a reactor with 100 ev average neutron spectrum, the plutonium fission cross section would be very much smaller than the cross section at the thermal energies. Consequently, for a given power density, you could tolerate a very much greater flux of 100 ev neutrons than you could a flux of thermal neutrons. On the other hand, for the even-even nuclei, the higher flux would mean that they would transmute very much faster because the cross section of the even-even nuclei at 100 ev is not much different than it is at thermal energies. This is of course only true on the average. On this basis I decided that a 100 ev reactor then would have a higher flux for a given power density and the transmutation for even-even nuclei which normally would be the bottleneck would proceed much faster. However, I ran into the following snag. If you want a 100 ev reactor then you have to prevent the neutrons from slowing down to thermal, and in order to do this you have to use a reasonably large concentration of uranium. Now the slowing down power  $\xi \Sigma_s$  in a fluoride reactor which of course is what I had particularly in mind, namely a lithium-beryllium fluoride - about 1/3 beryllium fluoride - is  $0.038 \text{ cm}^{-1}$ . So if you want to stop neutrons at 100 ev then you have to use a uranium-plutonium concentration which would give you  $\Sigma_a$  at 100 ev which is approximately equal to  $\xi \Sigma_s$ . If you then compute what the flux would be with this cross section at about 100 watts per cc, then you find that you get a 100 ev flux of  $10^{14}$ . You could, of course, in a fluoride reactor, tolerate a much higher power density, but if you do this then these reactors which contain poor moderators will become very big. You may recall that the molten fluoride reference reactor, at least the one that was current at the time of the Pittsburgh Meeting, had a power density of 187 watts per cc and the total reactor power was 600 megawatts. So at 100 watts per cc, one gets into the 100 megawatt region and if you want to go very much higher in power density you get fantastically high total powers. Because of this, I now don't quite understand about some remarks which Weinberg made about a reactor which supposedly had a very poor moderator like a zirconium moderator or something like this. Doesn't this run into the same difficulty?

Weinberg: It sounds like it does, however, I am somewhat confused at this point.

Ergen: Is what I said not clear?

Weinberg: I think what you say is not clear, but maybe that's just me. I'm sure to everyone else it's clear. (Laughter) Perhaps you can repeat in a few sentences what you just covered.

Ergen: I was trying to build a reactor in such a way (and I confine myself

now to a homogeneous reactor) that most of the fissions occur at 100 ev. I do this with the intent of taking advantage of the relatively low fission cross section of plutonium compared to thermal with the purpose of reaching the high flux, the high flux to be used to speed up the transformation.

Weinberg: This is high epithermal flux.

Ergen: Yes, high epithermal flux. For a given power density, due to the low cross section, I want to get a high epithermal flux which I then want to use to speed up transformation of higher isotopes.

Weinberg: Let's not talk about why you want it.

Ergen: But to do this, it is necessary to somehow prevent the neutrons from reaching thermal because if they do then you would get a very large number of fissions due to the large thermal cross section. But if I confine myself to this homogeneous reactor, then I have to put in a large fission cross section, at least a large absorption cross section which has to be to a certain extent fissionable. In other words you have to catch the neutrons before they become thermal.

But if I make the fission cross section large enough, then I have to fix it up in such a way that at 100 ev the macroscopic absorption cross section is approximately equal to the slowing down power.

Weinberg: Isn't it the resonance integral for everything above that energy?

Ergen: Well, sure. This is true, but nevertheless the fission cross section increases as the lethargy increases.

MacPherson: Can you look at this in a little different way in that you have to have your even isotopes in sufficient concentration that they absorb an appreciable proportion of the neutrons during the slowing down process?

Ergen: Yes, this is exactly right. However, I cannot allow the even nuclei to absorb more than one-half of the available neutrons.

The point is if I do this then I have to use the plutonium and other fissionable materials in such high concentration again that my flux wouldn't be so high after all.

Weinberg: Wouldn't that depend upon what your  $\xi \Sigma_s$  is for your moderator? The smaller that is, the smaller the concentration is.

Ergen: This is right. The second point is, even if I go to fluorides which are poor moderators, I have to make the reactor so large to avoid leakage before you get down to 100 ev that the total power even at 100 watts per cc would be excessive.

Weinberg: I won't argue that point at all. I only mentioned zirconium as an idle thought. What you are saying is that you see great difficulty in making a resonance reactor that will in fact be a very high flux resonance reactor.

Ergen: This is what I am saying so far. I confine myself to essentially a homogeneous reactor and what I wanted to say and more or less conclude with is that the next thing to do on this would be to see what I can do if I make this reactor heterogeneous.

Weinberg: Does your remark hold independent of the slowing down power of the moderator? Suppose you take something that is really a poor slowing down moderator?

Ergen: The reactor gets bigger and bigger.

Cole: The reactor gets bigger, but you're making lots and lots of heavy isotopes.

Ergen: That's right. If somebody all of a sudden should want many, many kilograms of Curium  $^{245}$  then this would be justified.

Weinberg: The thing that I had in mind was really the matter of getting lots of epithermal neutrons for other purposes.

Ergen: One point is that if you would succeed to build a reactor with high epithermal flux, this would not only give you a possibility of making transuranic elements, but this same reactor presumably would be good for cross section measurements.

Weinberg: I must say it's a little puzzling that you come up with such numbers because after all the APPR reactor actually has a fair amount of epithermal fission in it - about 20%.

Lane: It doesn't try to limit the thermals, does it? That's the thing here you have to limit thermals to keep the fissionable isotopes from burning up too fast.

Ergen: You say its 20%, but what is the energy at which these fission occur?

Weinberg: About one volt I would way.

It seems to me that this question of looking into an epithermal research reactor is something that is quite worthwhile to get a definite answer on the question, and I don't think anyone has really thought enough about it to answer the issue you have just raised. What you are saying basically is that to get a high flux epithermal reactor you have to have a very big one with very high powers.

Ergen: I'm pretty sure this is qualitatively true.

Weinberg: I can't quite believe that, because isn't it true that the fast reactors after all have a high epithermal flux.

Ergen: Well, how high? I don't know.

Weinberg: Well,  $10^{15}$ . Doesn't the ETR have several times  $10^{14}$  epithermal flux? The best way to do radiation damage experiments is in a fast reactor, by the way.

Ergen: Yes, this I don't think is in contradiction to what I have said though.

Weinberg: No, because there the mean free path is small. It's not the age that counts there, it is just the distance from one collision.

Ergen: You see at their peak of around 100 kev or something like that and at  $10^{15}$  flux, fission is very much less with plutonium and also the production of transplutonium elements is small.

Weinberg: I'm not talking about making transplutonium elements, I'm just talking about having beams or radiation damage sources.

Wollan: A heavy element tamper or reflector would help too, wouldn't it?

Ergen: Yes, this helps, but you get only a relatively small factor. I am quite sure that if you sit down and really see what you can do in the way of increasing the epithermal flux, then you can get better by having tampers, or by making the reactor really heterogeneous so it cannot be described as a homogeneous reactor. One possibility would be where the thermal cross section would shield itself and the epithermal cross section would not quite shield itself. All these things help a little.

Weinberg: I just wondered, Jim, if we could persuade Bill to come back in a month or so and tell us about his deliberations on nonthermal reactors.

Ergen: A month is too short.

Lane: I think it would be quite interesting.

Weinberg: Well, as I say, this is something that people have muttered about and it would be nice if someone really looked into it seriously.

Ergen: I think I would very much like to - but later.

Wollan: On the thermal flux in this reactor, have you plotted any curves to indicate the efficiency of producing thermal flux in the hole as a function of the thermal power? As your power goes on up, you get less and less per unit of power for the same density. You have to spread out the thickness of your layer. So it would be interesting to see where the diminishing return comes on the increasing of the power.

Lane: Well, the data show that it comes between  $10^{15}$  and  $10^{16}$  fluxes.

Wollan: I can see that in a general way, but I wonder if more specific remarks can be made.

Ergen: Well, the computing machine is excellent if you ask it specific

questions and you get those very quickly. But if you have to optimize something, then it becomes somewhat more tricky. Maybe to answer your question, I should go back to getting this thing written down in formulas then differentiate to find the optimum.

Wollan: A few more points on the curve would do this actually.

Ergen: What I did first was to just compute those reactors as they came out from the original assumption, namely make the thickness such that the volume with the given power density would give you the required flux. Then I got those poor figures. Then I wanted to optimize them and in the few cases where optimization was done, I retrieved most or almost all of the loss due to the finite thickness. This was quite an extensive computation for the code used on the machine. I don't think it is economical to do this for very many points. I think you are better off to extend the analytic formulae and find the maximum right then and there. I have the method for this worked out, incidentally.

Lane: In regard to Wollan's question, I looked at the same thing for an all heavy water flux trap reactor. If you go up enough in power, at some point you lose flux in the center as fast as you increase the power level so the flux reaches a saturation point. In that particular case, for the all heavy-water annular-type reactors, with aluminum fuel elements, it saturated at  $6 \times 10^{15}$ , and no matter what you did you couldn't get any higher.

Ergen: This brings us back to Jordan's original point that for this kind of operation the fluorides would be good because they don't saturate as fast.

Lane: Other combinations may saturate above the  $10^{16}$  point.

Wollan: You certainly wouldn't want to push right up to the full saturation point would you. You could do a lot of good down below that point.

Lane: Yes, the curve is rather steep and it flattens out quite rapidly.

Ergen: If I get a chance to come back, I may be able to cover these points.

Question: What about these remaining slides.

Ergen: I just had them in reserve.

HIGH FLUX RESEARCH REACTOR SEMINAR

April 25, 1958

Lane: Today I am going to talk about flux trap type reactors in which the fuel is located in an annular ring, or shell, surrounded on both sides by moderators. These were mentioned at the last seminar by Ergen. Studies have been carried out by the Oak Ridge National Laboratory (7-6), Argonne (8-9), Internuclear Co. (12-13), and others (10-11,14). Criticality experiments for H<sub>2</sub>O moderated, U-235 enriched, annular cylinders have been carried out at the Oak Ridge National Laboratory (4); however, data for other moderators are lacking.

The results of these studies indicate that maximum thermal neutron fluxes of the order of  $5 \times 10^{15}$  n's/cm<sup>2</sup>(sec) can be achieved at power levels of 500 Mw. For this reason I carried out a review of previous work with the objective of establishing whether an unperturbed thermal neutron flux of  $10^{16}$  neutrons/sec (cm<sup>2</sup>) can be obtained in a flux trap type reactor. In all cases, I assumed that for these ultra high flux reactors it will be necessary to use D<sub>2</sub>O through-out, as coolant, as well as internal and external reflector, although more recent studies seem to indicate that an H<sub>2</sub>O island has some advantages.

A major problem in ultra high flux reactors is that of removing the large amount of heat generated in such a reactor. Although this implies operating the reactor at as high a power density as possible (i.e., power per unit volume), for many applications (11) it is desirable to keep the power density down to minimize gamma heating problems. Fortunately, as will be shown later, the maximum achievable thermal neutron flux is a function of the specific power (power per unit weight of fissionable material) and reactor size, as well as the power density. Thus it is possible by increasing the size of the reactor core and by maximizing the specific power to get high fluxes at somewhat less than the highest power densities. The ultimate flux, however, will be achieved only at maximum power densities.

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- (1) Cheverton, R.D., "ORSORT Reactor Design and Feasibility Problem, High Flux Research Reactor", ORNL Report No. CF 56-8-206.
  - (2) Nestor, C.W., Jr., "Flux Peaking in Homogeneous Reactors", ORNL Report No. CF 57-8-84.
  - (3) "ORSORT Design Study of an Advanced Test Reactor", ORNL Report No. CF 57-8-5.
  - (4) "Physics Division Semiannual Progress Report for Period Ending March 10, 1955", ORNL Report No. ORNL-1926.
  - (5) Bolger, D'Irso, and Maak, "Feasibility Study of Superflux Reactor", ORNL Report No. EPS-X-293.
  - (6) Ergen, W.K., "Flux Distribution in a Reactor Consisting of a Spherical Shell of Fuel in Infinite Moderator", ORNL Report No. CF 57-12-100.
  - (7) Ergen, W.K., "Fluxes Obtainable in a Flux-Trap Reactor", ORNL Report No. CF 58-1-4.
  - (8) Link, L.E., "The Mighty Mouse Research Reactor Preliminary Design Study", ANL-5688.
  - (9) Kelber, C.N., "The Theoretical Physics of the Argonaut Reactor, ANL-5710.
  - (10) B & W, Lockheed Aircraft Corporation, "Proposal for an Engineering Test Reactor", June 30, 1955.
  - (11) Huffman, Conner, and Hanson, "Advanced Testing Reactors", IDO-16353.

Calculations presented in References (1 and 12) indicate that with 30 mil thick, 3 ft long plate-type fuel elements and a metal to water ratio of 0.67 (i.e., the water gap is 45 mils) a maximum power density of  $4.1 \times 10^8$  Btu/hr (cu ft) or 4250 Kw/liter can be achieved. Data for other plate thicknesses, metal to water ratios, and flow velocities are summarized in Table 1.

TABLE 1  
Maximum Power Densities vs. Fuel Plate  
Thickness, Metal to Water Ratios, and Flow Velocities

Plate Thickness, Mils	Water Gap, Mils	M/W	$\phi_{max}$ , Btu/hr (cu ft)*	
			V = 30 ft/sec	35 ft/sec
30	30	1.0	$3.1 \times 10^8$	$3.7 \times 10^8$
	45	0.67	3.5	4.1
	60	0.50	3.5	4.0
	90	0.33	2.9	3.3
45	30	1.5	2.5	3.0
	45	1.0	3.0	3.4
	60	0.75	3.0	3.4
	90	0.50	2.6	3.0
60	45	1.33	2.6	2.9
	60	1.00	2.7	3.0
	90	0.67	2.4	2.7
	120	0.50	2.0	2.3

\* Averaged over entire core

$T_s$  (max) = maximum metal surface temperature = 350° F.  
 $T_i$  = inlet water temperature = 100° F.

As seen in Table 1, the power density obtainable in these very high flux reactors is considerably above that in existing research reactors. For example, the heat removal or power density in the MTR is about 400 Kw/liter. Thus, we are talking about pushing this value up by a factor of 8-10.

Weinberg: How is this going to be possible?

Lane: With thinner fuel plates, higher rate velocities, higher surface temperatures, etc.

Winters: How long were the fuel elements?

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- (12) Elgert, Leyse and Ott, "Preliminary Investigations for an Advanced Engineering Test Reactor", AECU-3427 and Addendum (1957).
  - (13) National Science Foundation Study, Purdue University and Internuclear Co. Internuc 22, "High Flux University Research Reactor".
  - (14) Leyse, C. F., et.al., "An Advanced Engineering Test Reactor" (Internuc 23, March, 1958).

Lane: These are 3 feet long elements. One can gain something by going to shorter elements to permit increased flow velocities, but here you lose in that you don't have as long a space to irradiate samples. The longitudinal maximum to average flux would also increase and you wouldn't have as flat a flux along the length of the reactor at the midplane.

Winters: You may be able to pick up a little bit in these reactors by special techniques such as using embossed fuel plates.

Lane: Well, I looked at it from the standpoint of existing technology, and things such as you mention such as embossing fuel plates, certainly should be explored to see what kind of strength can be achieved with very thin plates by special techniques.

Wollan: Do you gain much by having your fuel not in the form of plates but rods or wires?

Lane: The particular design that was studied by the ORSORT group on the super-flux reactor did not actually involve fuel plates but used spiral elements to get more surface in a given volume. However, that design did not achieve higher power densities because the cross section for flow was lowered. Also they could not get above a flux of  $5 \times 10^{15}$  at 500 megawatts.

Ergen: Aren't you limited by the total temperature rise of the water in any case? If I am right, you have about a 200° C. temperature rise before you boil on fuel elements.

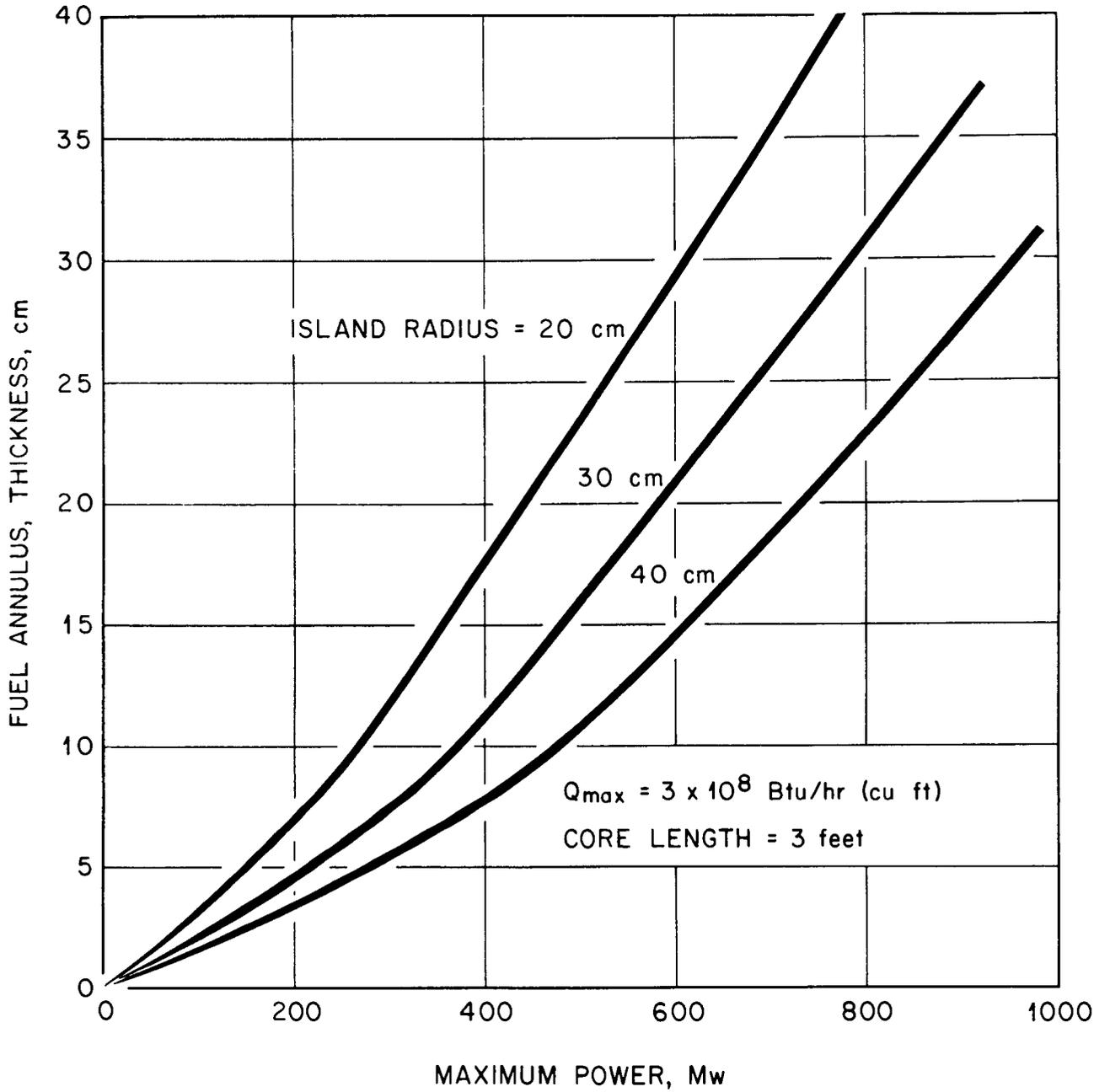
Lane: These calculations are based on no boiling at the surface of the plate.

Ergen: But you can't go much further without increasing your pressure.

Lane: That is right, you must increase the pressure to suppress boiling if you want to increase the surface temperature and also could make the plates thinner. All of these things combined might gain about 25-30%. Even with more surface however, there is still the limitation on the cross sectional area for flow of coolant in a given size annulus. The total pressure drop is also a limitation. For these calculations of high flux reactors I used a value of  $3 \times 10^8$  Btu/hr (cu ft) as the basis. This power density corresponds to 45 mil plates rather than 30 mil plates, since there is some question of the strength of 30 mil plates. On this basis, one can calculate the power extractable from a given size fuel annulus. Figure 1 shows the size of the annulus for various total powers and different island sizes. With an island radius of 20 cm, for example, the fuel annulus thickness must be about 30 cm to remove 600 megawatts. For this same annulus thickness, one can remove about 1800 Mw in a reactor with a 30 cm island and 950 Mw with a 40 cm island reactor.

This figure provides a basis for determining the thickness of the fuel annulus for any given case. Now the next step is to look at values of  $k_{eff}$  as a function of U-235 concentration, metal to water ratios, fuel annulus dimensions, and island radii. This has been calculated for D<sub>2</sub>O-moderated, annular fuel reactors by R. D. Cheverton (15). These calculations were

(15) Cheverton, R. D., "Further Calculations of a High Flux Research Reactor", (To be Published).



MAXIMUM HEAT POWER VS. FUEL ANNULUS DIMENSIONS

FIGURE 1

made with a three group - three region code on the ORACLE. In these calculations, island radii of 20-50 cm, fuel annuli of 2-30 cm and metal to water ratios of 0.5 and 0.73 were considered. The complete results will be published soon.

In Figure 2 - the value of  $k_{eff}$  for one case, namely a 30 cm island and metal to water ratio of 0.5 is shown. The original curves in which  $k_{eff}$  was plotted as a function of U-235 concentration have been replotted in this Figure as a function of the total amount of U-235 for the varying annuli thicknesses.

If one assumes that about 16% excessive reactivity will be required to make up for neutron losses due to equilibrium xenon, temperature coefficient, fission product poisoning, etc., this would correspond to a  $k_{eff}$  of 1.20. This excess reactivity would be sufficient for xenon override after shutdown of only a few minutes. At these high fluxes, the xenon builds up very rapidly after shutdown and it is almost impossible to provide enough excess reactivity to compensate for more than a few minutes. So in case of unscheduled shutdowns, the reactor must be brought back to power quickly, loaded with fresh fuel or left idle for about two days.

Winters: A few minutes, however, is not even enough time to lower the rod drive mechanisms and raise the safety rods.

Lane: But that is all the time one has so this has to be taken into consideration in the design.

Winters: This means you have to plan on reloading whenever the safety rods are completely tripped and have a set of spare fuel elements handy.

Lane: That is right. There is also the question of whether the rapid xenon build-up leads to nuclear oscillations. This has been looked at; however even for the flux levels considered, it appears that nuclear instability due to xenon is not a problem. Well, to continue, one can assume a  $k_{eff}$  of 1.20 will be necessary to make up for all losses except fuel burn-up. For a 5-day fuel cycle, about 6 grams of U-235 will be burned per Mw of heat power. In Figure 3, the amount of U-235 for two different islands is shown as a function of power level based on a  $k_{eff} = 1.20$  and a 5 day fuel cycle. In each case, the fuel annulus thickness is adjusted to permit extraction of the given power based on the heat removal conditions described previously, namely  $3 \times 10^6$  Btu/hr (cu ft). As seen in Figure 3, there is not too much difference between the two cases of a 30 and 40 cm flux trap respectively. At 500 Mw, the fuel loading would be between 7-8 kg and at 1000 Mw between 16-17 kg U-235.

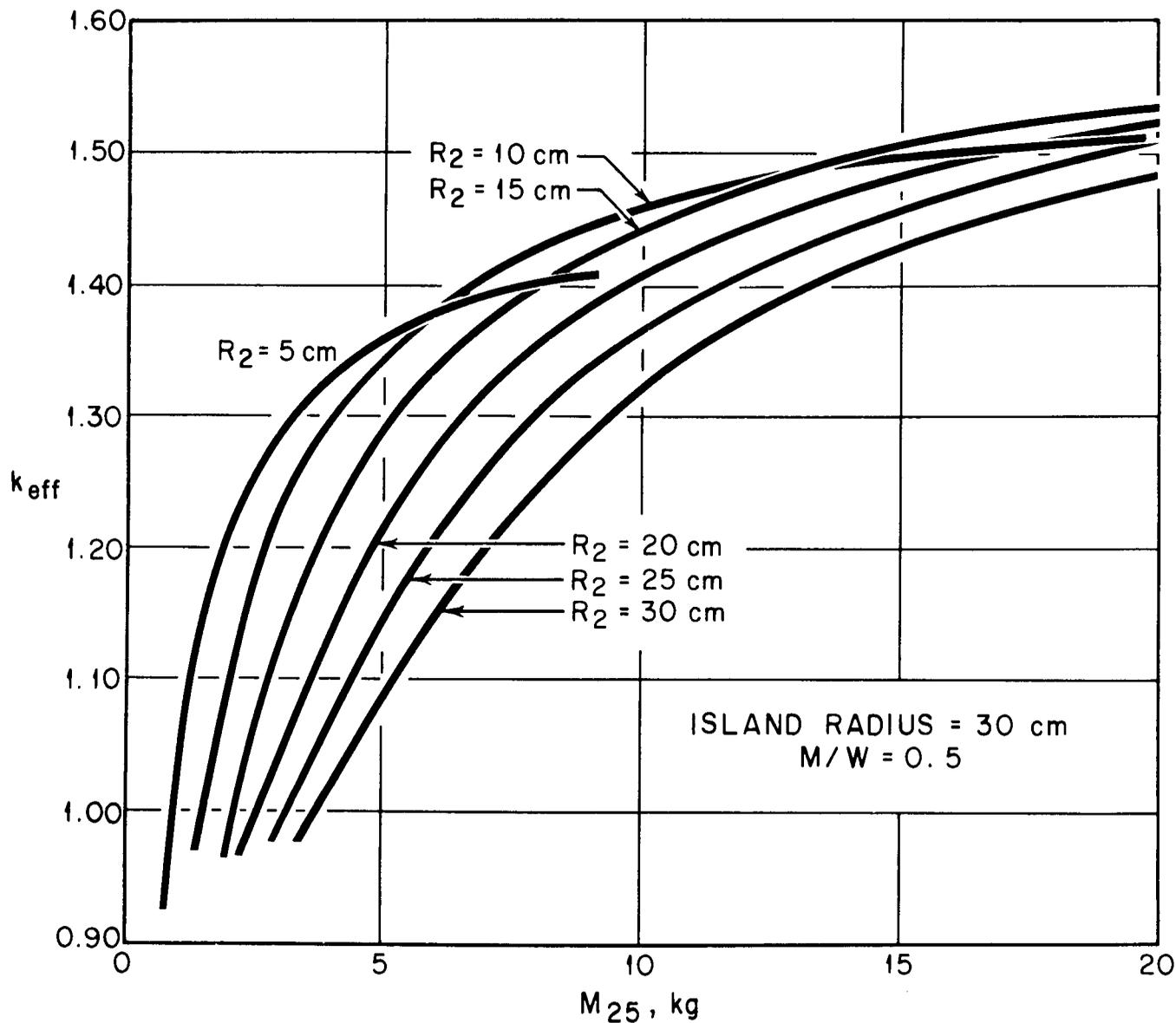
Weinberg: You say you can get 500 Mw out of about 7 kg of U-235?

Lane: Right. This is with a 5 day fuel cycle. I will discuss the effect of longer cycles shortly.

Weinberg: But this is a specific power of about 70,000 kw/kg U-235.

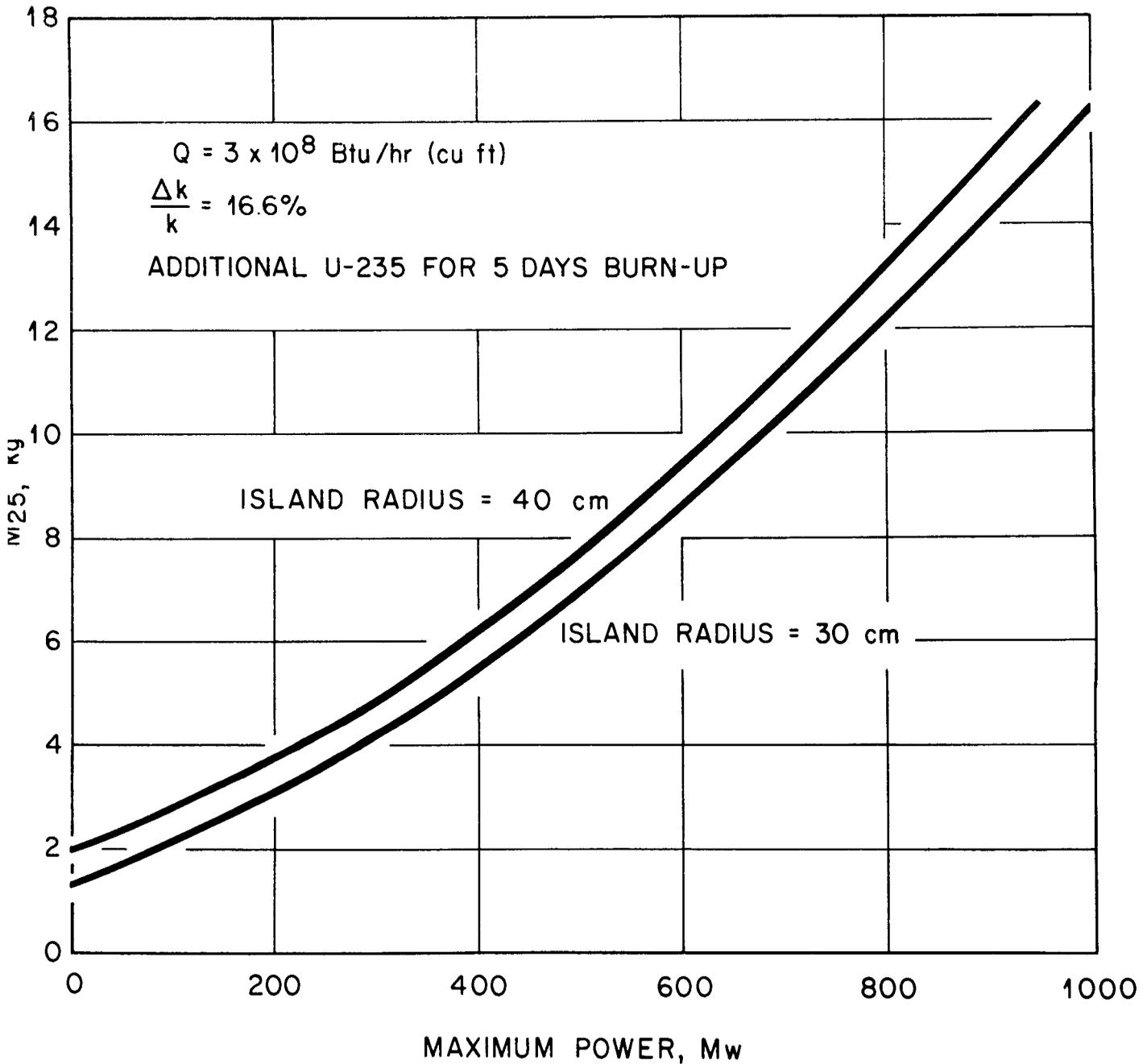
Lane: Yes, this is a very high specific power reactor.

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$k_{eff}$  VS. FUEL ANNULUS THICKNESS AND U-235 LOADING

FIGURE 2



U-235 REQUIREMENTS VS. POWER LEVEL

FIGURE 3

MacPherson: Referring to Figure 3, this is the amount necessary for burn-up only.

Lane: No, the U-235 requirement shown includes U-235 for a  $k_{eff} = 1.20$  plus that for a five day cycle.

Winters: But in going from 500 Mw to 1000 Mw, you double the amount of U-235 so you don't gain anything in flux, at least in the fuel region.

Lane: I will discuss this in a moment. Actually you are correct that you don't gain in the fuel region but the flux peaking in the island increases somewhat.

Weinberg: There is something I don't understand. This reactor already has a great deal of excess k in it. How do you take all of it up?

Lane: I will come to this also in a moment. But it would have to be done with burnable poisons or removable poisons. No one has actually devised a system for absorbing as much excess reactivity as that necessary for some of these reactors, namely a  $k_{eff}$  of 1.6 or greater.

Weinberg: But more than that, you can't use a removable poison because that reduces your effective heat transfer surface.

Lane: The removable poison I was thinking of would be a soluble material such as boric acid.

Weinberg: Oh, I see.

Lane: This would probably be the only way to get enough poison in the system. To continue, the equation for the average flux in the fuel is familiar to all of you and is simply,

$$\phi_{fuel} = \frac{3.1 \times 10^{16} P}{M_{25} \Sigma_f} \quad (1)$$

where, P is in megawatts,  $M_{25}$  in kg and  $\Sigma_f$  the macroscopic fission cross section of U-235 is in  $cm^2/kg$ .

Now by inserting  $\phi_{max}$ , the maximum flux in the island, in both sides of the equation and rearranging, one gets the following equation for the maximum flux per Mw.

$$\frac{\phi_{max}}{P} = \frac{3.1 \times 10^{16}}{M_{25} \Sigma_f} \left( \frac{\phi_{max}}{\phi_{fuel}} \right) \quad (2)$$

The expression for the ratio of the center flux to the flux in the fuel is referred to as the "flux peaking". Values for the flux peaking will vary with

the dimensions of the flux trap (island) and fuel annulus and also with the U-235 concentration or amount, and metal to water ratio. The metal to water ratio, however, has only a small effect on the flux peaking.

Referring to the comment made by Winters, increasing the U-235 loading in the reactor has two effects. In the first place it depresses the flux in the fuel region as evident from Eq. (1); but second it raises the ratio of  $\phi_{\max}/\phi_{\text{fuel}}$ . This latter effect is shown in Figure 4 in which data are plotted to show the flux peaking for varying dimensions of islands and fuel annuli as a function of the U-235 loading. The numbers in the parenthesis refer to annulus thickness and flux trap radius respectively. Although the points shown refer to the peaking for a reactor of the specified geometry loaded with the given amount of U-235, the peaking at other loadings for the same reactor can be estimated by assuming a straight line relationship with the line passing through  $\phi_{\max}/\phi_{\text{fuel}} = 1.2$  at  $M_{25} = 0$  and the point given. The validity of this assumption has not been verified for the entire range of island and fuel annuli; however, it appears to be a good approximation for the range of dimensions covered.

Thus Figure 4 permits one to determine the value of  $\phi_{\max}/\phi_{\text{fuel}}$  for different size  $D_2O$  moderated flux traps and fuel annuli with varying amounts of U-235.

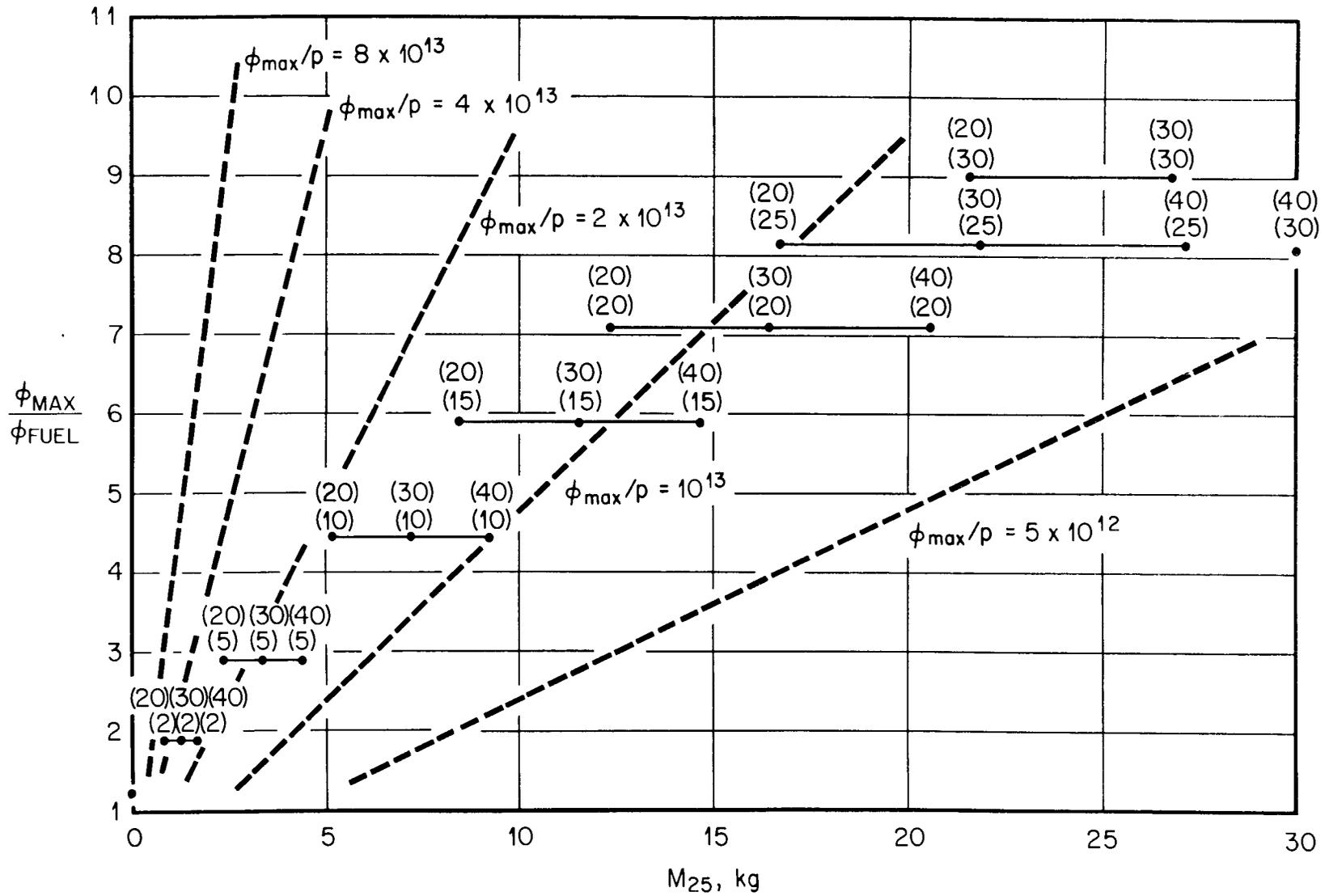
Lines of constant unit flux, that is, the maximum flux per Mw are also shown in Figure 4. Thus it is seen that for any particular reactor, increasing the amount of U-235 increases the value of  $\phi_{\max}/\phi_{\text{fuel}}$ , following the straight line relationship mentioned; however since the slope of this line is less than the slope of the lines of constant unit flux, the overall effect of increasing the U-235 loading is to reduce the value of  $\phi_{\max}/P$ . This is true for the range of conditions considered, it would not necessarily be true for all cases however.

It is also seen from Figure 4, that increasing either the thickness of the fuel annulus or the radius of the island, decreases the value of  $\phi_{\max}/P$ . Although it is not evident from Figure 4,  $\phi_{\max}/P$  actually goes through a maximum as one decreases the fuel annulus thickness or the island radius. However, the optimum occurs for small reactors, those in Figure 4 have dimensions larger than the optimum to permit high power operation necessary to get to a high flux.

It is noted from Figure 4 that points for reactors with the same outer radius of the fuel annulus (i.e., the sum of the numbers in parenthesis) all fall on a straight line passing through  $\phi_{\max}/\phi_{\text{fuel}} = 1.0$  and  $M_{25} = 0$ . This makes it possible to express the ratio  $\phi_{\max}/P$  or the center flux per Mw by a second approximation,

$$\phi_{\max}/P = 2.07 \times 10^{13} \left( C + \frac{1}{M_{25}} \right)$$

where C = constant determined by value of island radius plus annulus thickness in cm; also C is approximately  $20/R$  for  $R < 50$  cm where R is the radius of the outer edge of the fuel annulus. Thus the above equation reduces to:



-125-

FLUX PEAKING VS. FUEL ANNULUS DIMENSIONS AND U-235 LOADING

FIGURE 4

$$\phi_{\max}/P = 2.07 \times 10^{-13} (20/R + 1/M25)$$

Wollan: What are the units of P?

Lane: Megawatts. This is the maximum flux per Mw. If one multiplies through by P, this equation shows that the maximum flux itself is proportional to both the power density (power per unit volume) and the specific power (power per unit weight) of U-235. For a 3 ft long core cooled with water at 35 ft/sec, the maximum flux is given by,

$$\phi_{\max} = 2.07 \times 10^{-13} (0.893R + P_s)$$

where,  $P_s = \text{Mw heat/kilogram U-235}$

$R = \text{cm}$

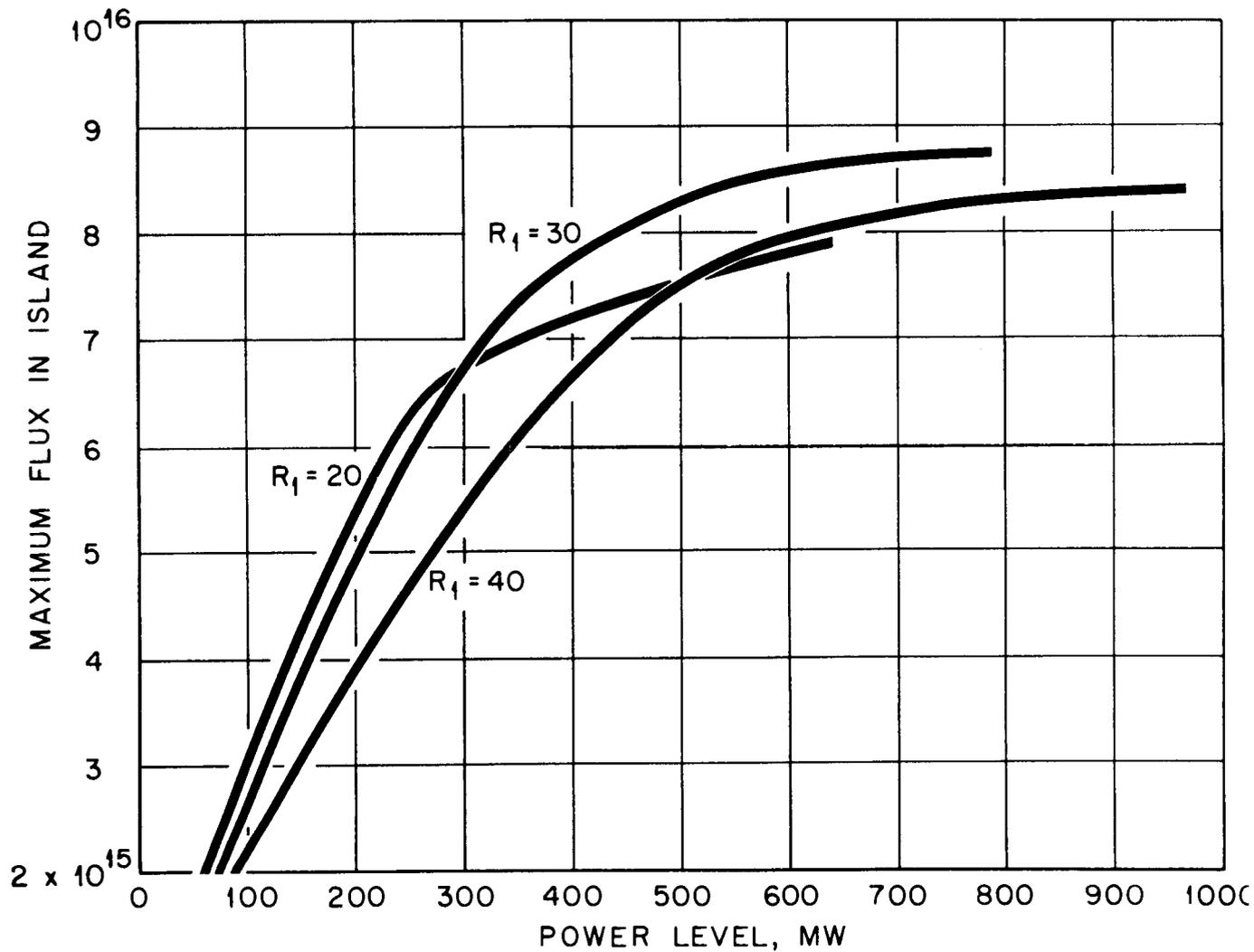
Ergen: Something must be wrong in the equation because if you run at zero specific power you still get an appreciable flux.

Lane: The equation only holds for a constant power density of  $3 \times 10^8$  Btu/hr (cu ft) which was used to find the constant 0.893. So you can't operate at zero specific power under the assumed conditions. This equation indicates that to achieve maximum flux both the size of the total reactor core plus island, as well as the specific power, must be maximized. Unfortunately, for a given power density the specific power decreases as the core size increases due to the increased loading for burnup, etc. Because of this, the maximum flux achievable in a D<sub>2</sub>O cooled and moderated solid fuel reactor is of the order of  $7-8 \times 10^{15}$  n's/cm<sup>2</sup>(sec) based on the initial loading. This is shown in Figure 5 which is a plot of maximum flux in the island as a function of power level and island radii based on a  $k_{\text{eff}} = 1.20$ . In other words, the curves represent the flux at the end of any given fuel cycle at which time the loading is just enough to compensate for experiments, etc., and  $k_{\text{eff}} = 1.20$ . It is seen here that for low power levels, higher fluxes are achieved with the smaller island, but at higher power levels there is an optimum island radius. For the conditions shown, a 30 cm island appears to give highest fluxes. The effect of increasing the fuel loading in a reactor with a 30 cm island and a 30 cm fuel annulus on  $\phi_{\max}/P$  is shown in Figure 6. Although the ratio of  $\phi_{\max}/\phi_{\text{fuel}}$  increases as the U-235 concentration increases (as indicated previously in Figure 4) the value of  $\phi_{\max}/P$  decreases. Values of  $k_{\text{eff}}$  are also shown in Figure 6 to indicate the amount of excess k that must be controlled by removable or burnable poisons. This may be difficult to do for very large U-235 additions.

Winters: You say you have to have  $k_{\text{eff}}$  equal to 1.20 at the end of the cycle. Why is that?

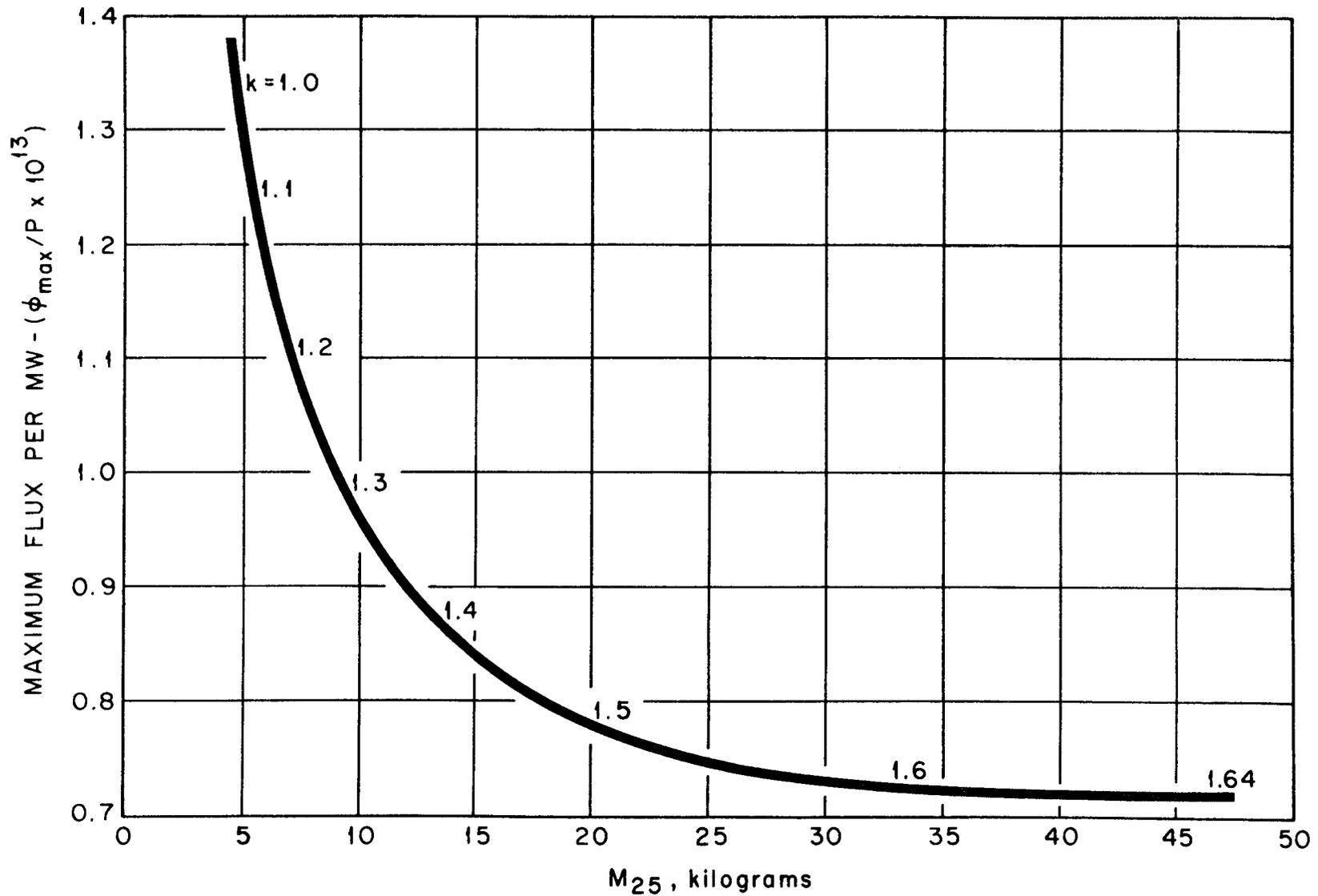
Lane: Because of temperature coefficient equilibrium xenon, fission product poisons and experiments. This amounts to 16.6% excess k to compensate for things beyond one's control.

Winters: I guess it depends on your definition of k.



$\phi_{max}$  IN BURNED OUT CORE ( $k_{eff} = 1.20$ )  
CYLINDRICAL D<sub>2</sub>O-D<sub>2</sub>O-D<sub>2</sub>O REACTORS

FIGURE 5



-128-

EFFECT OF FUEL LOADING ON MAXIMUM FLUX PER MW  
( $R_1 = 30$  cm,  $R_2 = 30$  cm,  $M/W = 0.5$ )

FIGURE 6

Lane: But this represents the amount of adjustable reactivity between an operating reactor and one that is shutdown. Turning now to the effect of the fuel cycle on the maximum flux, this is shown in Figure 7 for an 800 Mw reactor for a  $k_{eff} = 1.2$ . Depending where one starts, or in other words how much U-235 is added for burn-up, the initial maximum flux will be as indicated at some point on the curve. If, for example, one adds enough U-235 to permit operation on a 10 day fuel cycle the initial flux at the center of the 800 Mw reactor would be  $6.5 \times 10^{15}$ . As the U-235 burns out, the flux gradually increases until it reaches a value of  $8.8 \times 10^{15}$  at which point the cycle repeats. Although the further lowering of initial flux due to the addition of more U-235 to get a 20 or 30 day fuel cycle, for example, is not very great, the main problem would be that of compensating for the large excess k introduced in this later case.

Winters: Is this on the basis of the flux at the end of the cycle?

Lane: No, the curve represents the flux at the beginning, not end, of the cycle for any desired cycle. Starting at any point, say for a 20 day fuel cycle, the initial flux would be  $5.9 \times 10^{15}$ . At the end of 10 days it would be  $6.5 \times 10^{15}$  and at the end of 20 days it would be  $8.8 \times 10^{15}$ . So the variation over the entire fuel cycle would be about 50%.

Ergen: This doesn't take into account that the added loading might require more fuel plates does it?

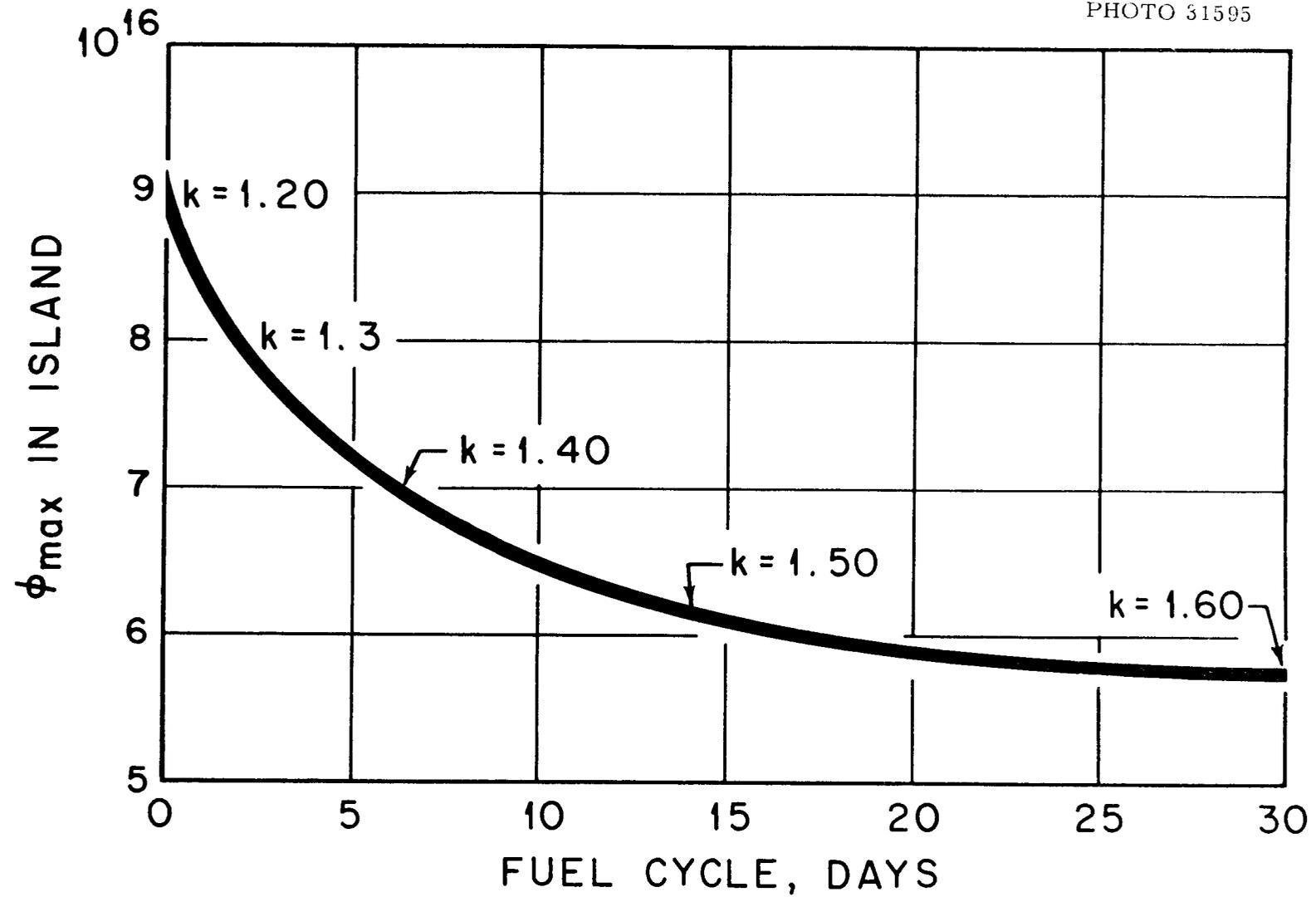
Lane: No, it was assumed that one could change the amount of uranium in the fuel plate by increasing the alloy content. But this might be a limit if too much uranium were required. However, about 5 times the uranium concentration used in MTR type fuel plates can be used before metallurgical problems arise. Also one might use uranium oxide in the fuel rather than uranium alloy.

The reactors thus far considered do not seem capable of achieving  $10^{16}$  flux, that is, the all D<sub>2</sub>O moderated and reflected reactors unless they operate at power densities higher than those assumed.

Cole: Do you have any results for reactors moderated with H<sub>2</sub>O?

Lane: Yes, I am coming to those now. The calculations described so far have dealt with D<sub>2</sub>O moderated, cooled, and reflected systems only. To investigate the effect of changing the moderating material in these regions, calculations of  $\phi_{max}/P$  were carried out by T. B. Fowler of the Reactor Experimental Engineering Division for spherical flux trap reactors<sup>(16)</sup>. The results are plotted in Figure 8 for H<sub>2</sub>O-H<sub>2</sub>O-D<sub>2</sub>O, H<sub>2</sub>O-D<sub>2</sub>O-D<sub>2</sub>O, D<sub>2</sub>O-H<sub>2</sub>O-D<sub>2</sub>O, and D<sub>2</sub>O-D<sub>2</sub>O-D<sub>2</sub>O systems as a function of island radius and fuel annulus thickness based on  $k_{eff} = 1.0$ .

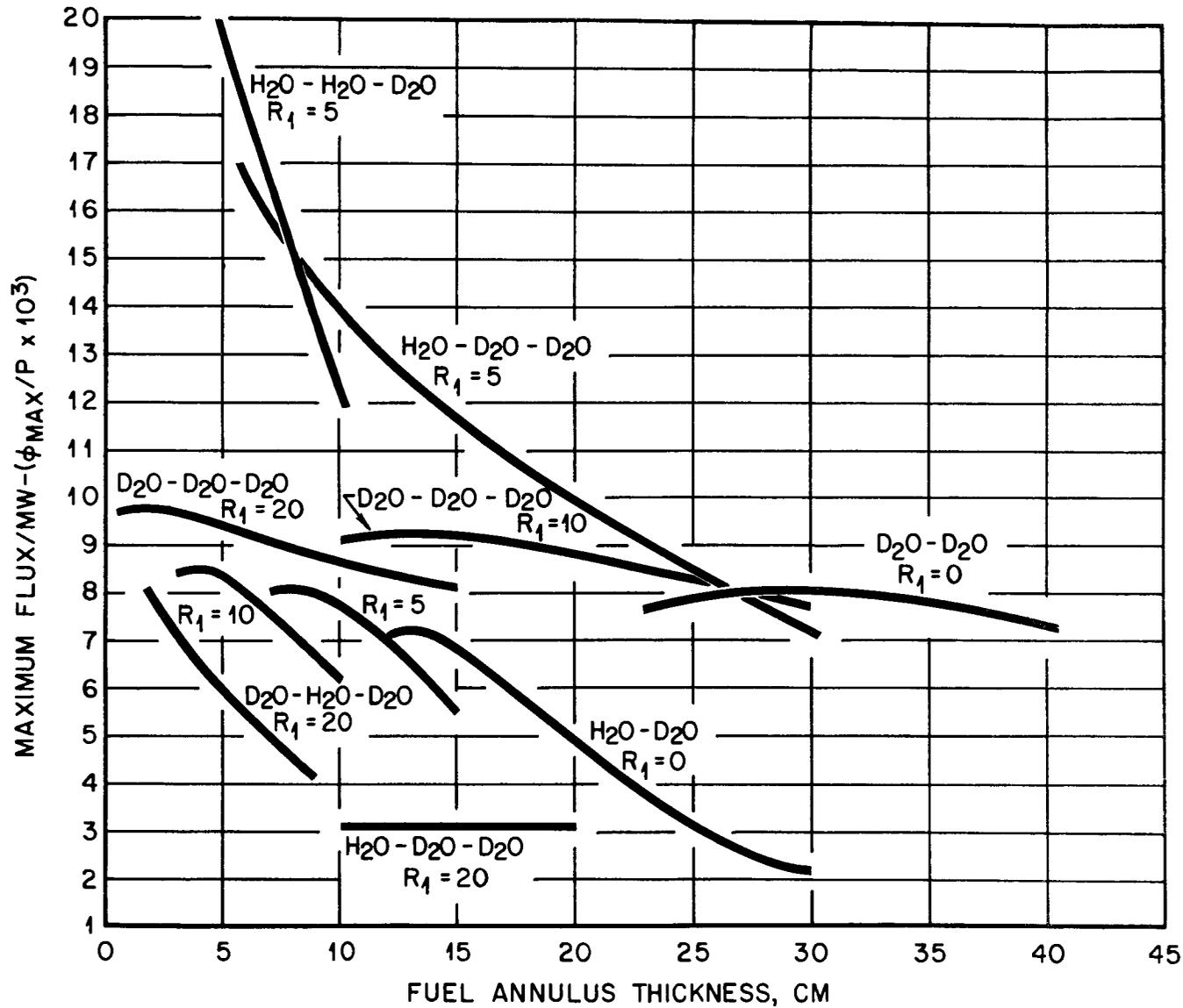
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(16) Fowler, T. B., "Maximum Thermal Flux Per Mw in Three-Region Homogeneous Reactors", ORNL Report No. CF 58-3-76.



-130-

EFFECT OF FUEL CYCLE ON MAXIMUM FLUX  
( $R_1 = 30$  cm,  $R_2 = 30$  cm,  $P = 800$  MW)

FIGURE 7



-131-

$\phi_{MAX}/P$  FOR SPHERICAL REACTORS ( $K_{eff} = 1.0$ )

FIGURE 8

As shown in Figure 8, highest values of  $\phi_{\max}/P$  are achieved for the systems which have  $H_2O$  in the flux trap. However it is noted that the radius of the  $H_2O$  island must be quite small.

Winters: That means a very small reactor doesn't it?

Lane: Right. In order to get high flux peaking, the flux trap and fuel annulus must be relatively small which implies lower power systems. For these low power reactors,  $H_2O$  gives the highest flux per megawatt; however for a fuel annulus thickness greater than 30 cm, the all  $D_2O$  moderated systems are best. As noted in Figure 8, if the radius of an  $H_2O$  island is too large (i.e., 20 cm for the  $H_2O$ - $D_2O$ - $D_2O$  case), the value of  $\phi_{\max}/P$  is reduced to about  $3 \times 10^{13}$ .

Similar calculations for cylindrical flux trap reactors have been carried out by Internuclear Company in connection with studies for an advanced engineering test reactor and a high flux university reactor. These results, given in Internuc 22 and Internuc 23, indicate that with  $H_2O$  in the flux trap values of  $\phi_{\max}/P$  up to  $10 \times 10^{13}$  may be obtained.

Stoughton: That you might expect to be the best case. Because you have a large fuel volume and  $H_2O$  is better than  $D_2O$  for slowing down fast neutrons.

Lane: Yes, but if you get the annulus too large too many neutrons are absorbed internally and the flux is depressed.

Winters: Perhaps a better case would be a mixture of  $H_2O$  and  $D_2O$  in the island.

Lane: This has not yet been considered as far as I know, but it is an interesting possibility.

Stoughton: There were some calculations on mixed  $H_2O$  and  $D_2O$  for another application; however, the mixed moderator did not appear to have any advantages. But that may not be true here.

Lane: In our last seminar, it seemed that the mixture might be better.

Ergen: I don't know, I have done no calculations on this. The way to do it would be to use experimental values for the age in  $H_2O$  and  $D_2O$  from Savannah River. The calculations could be done by hand or with the ORACLE.

Lane: Figure 8 also shows values for systems without a flux trap. An  $H_2O$  cooled core and  $D_2O$  reflector seems best for small cores, a  $D_2O$  cooled and reflected system best for large cores. Maximum values of  $\phi_{\max}/P$  of  $7-8 \times 10^{13}$  may be obtained for these reactors. Well, that is as far as we have gone on these considerations of flux trap reactors. More calculations should be carried out for the mixed systems and  $H_2O$ - $D_2O$  combinations. The Internuclear calculations for high flux reactors indicates that enough peaking can be obtained with the  $H_2O$ - $D_2O$  case so that very high power levels are not necessary to get fluxes greater than  $10^{15}$ ; at 75 Mw, fluxes of  $3-5 \times 10^{15}$  may be possible.

Weinberg: This is not very different from the METR reactors is it?

Lane: What is the METER?

Weinberg: The NDA-Belgium MTR.

Lane: Is that the reactor with the skewed loading?

Weinberg: Yes, they claim fluxes better than  $10^{15}$  at 50 Mw.

Lane: But here we are talking about a factor of three greater at that same power level.

Weinberg: Yes, I guess they have less flux peaking.

Lane: A factor of two increase in flux is hard to achieve at these high fluxes. None of the cases so far looked at, have reached the  $10^{16}$  level no matter what is done -  $8 \times 10^{15}$  is the maximum.

Weinberg: None of them really get even that because of the depression due to experiments.

Lane: That is correct. These are unperturbed fluxes.

Winters: It seems to me that the central region is so sensitive that with anything you put in the peaking advantage is lost.

Lane: No, that is not quite true. The  $H_2O$  island is not as sensitive as the  $D_2O$  island and if the objective is to make transuranic elements, the flux depression due to the experiment would be small. However, these very high-flux trap reactors are not quite as good for engineering test loops as for small-sample experiments.

Wollan: Would that also be true for a beam hole looking at this high flux?

Lane: In these systems, just as in the MTR, a flux peaking in the reflector also occurs. This peaking is about half that of the center flux trap, however it lends itself to the use of beam holes. I should think the depression of flux due to an experiment in this beam hole would be less than in the center.

Winters: This points up the comment made earlier namely that it is going to be extremely difficult to increase the flux by a factor of two, and more should be done in the direction of attempting to improve the experimental techniques.

Lane: Yes. At power levels above 200 Mw, it costs about \$10 million for each added 100 Mw, or each unit of  $10^{15}$  flux, and flux increases are costly.

Weinberg: That really raises the same old question; which is better, a number of smaller reactors or one big machine such as you have been discussing. My own personal hunch is that a greater number of smaller reactors would be more useful than one large reactor.

Winters: That still does not answer the question of whether it is better to spend lots of money increasing the flux or spend the same amount on improving the utilization of the neutrons.

Wollan: There are some experiments which can be done on a very high flux reactor that simply could not be done with a greater number of smaller ORR's; such as irradiating small samples to saturation.

Ergen: In some of these experiments, however, such as making transuranics, tricks such as using epithermal neutrons may be used. This idea has certainly not been eliminated yet and in some respects looks very good. So before building a high power reactor these things should be investigated.

Weinberg: The thing I keep coming back to is that for \$17 million, the cost of the 175 Mw ETR for example, one can build three 30 Mw reactors and these would seem to be of more general use. Actually in the ETR the flux isn't terribly high even so. Why didn't they build three smaller reactors instead of the ETR?

Lane: At 30 Mw you might be able to get very much higher fluxes than the ETR with the flux trap idea.

Weinberg: According to your formula, it should be about  $7 \times 10^{14}$  @ 30 Mw. Would a reactor of that sort be \$5 million or \$10 million?

Lane: Probably the lower figure because the flux trap idea doesn't raise the cost.

Winters: How about all the heavy water cooling - isn't that costly?

Lane: At 30 Mw one would use H<sub>2</sub>O cooling, not D<sub>2</sub>O.

Winters: That would save considerably on the cost if you can use H<sub>2</sub>O for cooling.

Ergen: Has any factor been applied to allow for the uneven power distribution in the fuel annulus?

Lane: Yes, this was taken into consideration in calculating both the power density and the  $\phi_{\max}/\phi_{\text{fuel}}$ .

Wollan: I don't remember anything having been said about the length of the core as a function of its size.

Lane: These calculations have all been made for a 3 ft core. Higher power densities may possibly be obtained with a shorter core which might give a higher flux.

Wollan: Higher flux per Mw?

Lane: Right. The unit flux might be higher but you would have less experi-

mental volume in the longitudinal direction.

Winters: Can you gain by putting fuel at the ends of the flux trap?

Lane: Yes, then the cylinder would be more like a sphere and from these calculations a factor of 1.5 might be gained.

Question: Is there any single limitation on power density or is it a combination of many things?

Lane: The power density could be increased by making fuel plates thinner or spacing them closer together.

Weinberg: Is 800,000 Btu/hr sq ft a nominal value or a maximum value of heat flux?

Lane: No, that is the maximum heat flux. The average would be less than that.

Weinberg: Can't you do better than that by allowing boiling to take place on the plates?

Lane: So far we have assumed that the surface of the fuel plates is kept below the boiling point by pressurizing the system.

Winters: If you can release that restriction you can improve the situation.

Weinberg: Well, you soon encounter the limitation of the temperature of the aluminum.

Lane: There is also the problem of getting the heat out of the reactor limited by the flow rate.

Weinberg: We always thought that even the MTR core could do better by a factor of 1.5-2.

Lane: But that is only 400 kw/liter. We are now talking about 4000. But this requires very thin plates. If you tried to circulate water faster, they might vibrate like a reed.

Wollan: Would this introduce a nuclear oscillation?

Lane: Yes, it probably would.

Weinberg: This sort of thing has already been noticed in the MTR. However, buckling of the plates was more serious.

Winters: These studies won't be complete unless this question of heat removal by nucleate boiling is considered.

Weinberg: It is generally considered that 1 million Btu/hr (sq ft) is the upper limit, however, because of burn-out.

Lane: But that is a question of flow velocity.

Winters: One can operate at 2 million Btu/hr (sq ft) with sufficiently high velocities. Data are available which show that under some conditions burn-out occurs well above a heat flux of 2 million. \*

Lane: These things must be considered in order to achieve  $10^{16}$  flux. Also more research on a laboratory scale should be done on extended surface fuel elements to find how such fuel elements behave with high flow velocities, etc.

Wollan: Can you do any better in a homogeneous system?

Lane: No, the power density would have to be much lower due to corrosion of zirconium. A large single region homogeneous reactor has been considered; however this reactor was no better than the solid fuel reactors.

Ergen: Perhaps higher power densities might be obtained with fluorides.

MacPherson: If you are approaching a limit on the  $\Delta t$  of the coolant, one can not hope to increase the power density by using a circulating fuel.

Lane: That would be a limit in any case. If there are no more questions, the seminar is adjourned. The next seminar will review the findings of the entire series.

\* Editors Note:

Gambill, W. R., and Greene, N. D., "A Preliminary Study of Boiling Burnout Heat Fluxes for Water in Vortex Flow", ORNL Report No. CF 58-4-56.

"A maximum heat flux of  $35 \times 10^6$  Btu/hr (sq ft) ( $11.1 \text{ kw/cm}^2$ ) was obtained experimentally with vortex flow. In this report reference it was made to burnout heat fluxes of  $14 \times 10^6$  Btu/hr (sq ft) for water in linear flow and  $32 \times 10^6$  Btu/hr (sq ft) for sub-cooled nucleate boiling in a continuously consumed flow channel".

THE LABORATORY PROGRAM - A. M. Weinberg

Today we have a guest, the new Director of the Division of Research of the Atomic Energy Commission, Professor Williams, formerly of the University of Minnesota. John has been at the Laboratory a number of times, but this is the first time in his new capacity. I am happy that he is able to attend this seminar on an ultra high flux research reactor because, as these matters progress, problems related to such a reactor are likely to land in his lap.

To open the discussion I should explain that we here at the Laboratory have for a number of years been thinking of the question of whether the Oak Ridge National Laboratory should embark on the development and construction of a reactor which would exceed the present highest flux reactors by a factor of at least 10. The best existing reactor, as far as thermal flux is concerned, I guess, is really the MTR. The present best flux that exists anywhere is of the order of  $5 \times 10^{14}$ . People have wondered a great deal, on and off, if one could get something like  $5 \times 10^{15}$  and possibly even  $5 \times 10^{16}$ . In considering and discussing this question it has been brought out that there are two separate ways of achieving very high fluxes, (1) by a heterogeneous reactor of special design, and (2) by a homogeneous reactor. To go back a bit, if you operate at a very high flux you encounter the problem of fuel burn-up. Operation of a reactor becomes very awkward if the fuel elements have to be changed very often. For example, MTR fuel elements are changed, roughly, every three weeks; at  $5 \times 10^{15}$  fluxes you might have to change fuel elements every two to four days - actually you might extend the cycle to every five or six days. So it was natural to look some at the possibility of a liquid fueled reactor, and we carried out a conceptual design of a huge 600-Mw homogeneous research reactor which would achieve a maximum flux of almost  $6.5 \times 10^{15}$ . The design of such a reactor has been carried out in a fair amount of detail, and, in fact, I talked to the Commission about it. It really was a monstrous device and cost of the order of \$100 million.

Well, there have been other schemes that have been proposed. Generally speaking, discussions that we have had here over the past several months have called attention again to the fact that perhaps the best way of achieving this very high flux is to go to the flux-trap geometry. Although everybody here knows what the flux-trap concept is and what it is like, perhaps John does not know.

Williams: No, I don't.

Weinberg: In a reactor such as the MTR, the fuel elements are in a central region surrounded by a reflector; in the flux-trap reactor you put the uranium in an annulus with moderator inside and a reflector on the outside. With this arrangement the thermal flux is low in the fuel region and high in the center of the annulus because the amount of uranium absorbing material is much less in the center. Estimates have been made of just what you can achieve with reactors of this general class. Jim Lane, in the last meeting,

gave an extensive discussion of what you can achieve. Bill Ergen, in a previous meeting, also discussed reactors of this design. It seems one can achieve - what would you say, Jim,  $5 \times 10^{15}$  with how many megawatts?

Lane: The best design I have heard of gets  $5 \times 10^{15}$  with only 50-100 Mw.

Williams: Well, is this over a reasonable volume?

Lane: No, the experimental volume in the center is limited. However, the peaking of the flux in the reflector is about half as much as in the core. Here one does have as large a volume as any of the enriched research reactors built so far. All in all, I would say that you could achieve  $5 \times 10^{15}$  for one experiment and  $2 \times 10^{15}$  for a number of others at power levels between 50-150 Mw.

Weinberg: Let me just summarize the following aspects of the matter. First, is it technically feasible, as far as we see it, to achieve a flux of  $5 \times 10^{15}$  with a reactor in which there are very few engineering uncertainties? I would say that we believe it probably is technically feasible to achieve  $5 \times 10^{15}$ , without going to a liquid system. With the liquid systems with fused salts, or maybe even with the aqueous fuels, perhaps you could push it up an additional factor of 2. Wouldn't you say, Bill, that  $10^{16}$  is possible?

Ergen: Yes. It is mainly a question of achieving a higher power density, as well as a better fuel processing procedure.

Weinberg: The Internuclear Company design achieved 5 kw per cc with a heterogeneous reactor, which is a good factor of 10 higher than the MTR and makes power density not a strong limitation. This is actually nothing new, but it reinforced the view that it is possible to achieve of the order of  $5 \times 10^{15}$  with reactors that are probably attainable from the engineering standpoint. If you are content with  $5 \times 10^{15}$  over a small volume, then, as Jim Lane has just implied, one can reduce the magnitude of an enterprise of this sort from something like \$100 million to something perhaps a tenth of that.

Williams: Do you pay as much for the limited volume over which you attain the high flux that you lose the purpose and general flexibility of the reactor?

Weinberg: Let me just say that that is the other question we tried to come to grips with in these seminars. As I say, first we wanted to find out if the reactor is really feasible, and now we think that it is. Second, we wanted to determine its worth. I suppose that each person listening or reading through the minutes of these seminars comes to his own conclusion on the matter and decides which things are the most important and which are not. It is my own personal opinion, and I believe that I do not yet have the whole situation completely and thoroughly understood and digested, that the main incentive to achieve  $5 \times 10^{15}$  fluxes is the work in chemistry. If one excludes such things as the most advanced reactor development, such as nuclear rockets which have to run at  $5 \times 10^{16}$  or  $10^{17}$  fluxes, and looks only at the incentive

that comes from the pursuit of basic knowledge, I am inclined to think that the chemists made the best case. In other words, the transuranic people and the radioisotope producers, from my way of looking at it, seem to have made the strongest case. Now this, I admit, is a feeling which I cannot completely document, but when you look at what the neutron diffraction people said, if I understood Henri Levy correctly, there are many really very interesting things that they can still do at the lower fluxes. Also, this is true for the work with neutron choppers carried out by Jack Harvey. Isn't that true, Jack?

Harvey: The detectors are the weakest part of our work. If you could get a gain of about 2 there you could probably utilize higher fluxes.

Weinberg: That is only part of it. The other thing is that as far as getting higher in energy is concerned, the Van de Graaff people are coming down and squeezing you out of that.

Harvey: When they come down, we go up; however, there is a dividing line and there will be one for several years.

Weinberg: Yes, and then with respect to the work by the radiation damage people, the general tone of what Doug Billington said is that as far as basic solid state physics is concerned the fluxes that you now have are probably quite enough for most purposes. Now he did point out that as far as engineering research is concerned, especially fuel element research, the higher fluxes did have advantages.

Williams: Is the volume big enough to handle that problem?

Weinberg: The fuel elements? -- Not for \$10 million. To really get  $5 \times 10^{15}$  flux in a reactor that would handle fuel elements you would have to talk about \$50 million or \$75 million.

Lane: \$15 million or \$50 million?

Weinberg: \$50 million.

Lane: No, \$15 million would be closer if you are satisfied with one loop.

Weinberg: Well, will you take a contract for this then -- oh, only one loop you say?

Lane: Yes, one loop. However, in these big engineering test reactors there is always some question as to whether you can really handle lots of complicated loops simultaneously since the control of the loop is so much tied to the control of the reactor. So the most recent thinking in connection with the Advanced Engineering Test Reactor, which was designed for loop-type testing of fuel elements, is that you build a reactor around a single loop. The flux-trap design, even though it has a rather small hole, does lend itself to such a single loop.

Ergen: It is not only the volume that counts, it is also the neutron absorption by the experiment.

Lane: Yes. You can't rob flux in all directions. Even with the single loop the flux peaking may be depressed as much as a factor of three, depending upon the amount of neutron absorption.

Weinberg: Well, Jim, since you have been running these meetings perhaps you would like to say who you think made the best case.

Lane: The original objective was to see if we need a  $10^{16}$  flux reactor. There was no strong case for that at all. If you get  $5 \times 10^{15}$  fluxes, the transuranic people are certainly going to be satisfied. None of the basic research uses, including making the lower atomic weight isotopes, required fluxes above  $5 \times 10^{15}$ . There is one case like the things Henri Levy talked about where they would rather have  $10^{17}$ , or was it  $3 \times 10^{18}$ , to do the studies on complicated molecules such as proteins, but that is out of the question. Thus there seems to be a gap where no one really wants  $10^{16}$  fluxes. As a matter of fact, I came to the conclusion that  $5 \times 10^{15}$  is somewhat on the high side of what was required, and that it was more nearly 2 to  $3 \times 10^{15}$ .

I think that an important point was made that all-purpose reactors are not very desirable. Tremendous general-purpose super reactors, which could do all things for all people, are not too good because the needs are so different - needs in terms of the ratio of fast neutrons to the thermal neutrons or needs in terms of the average energy of the neutrons. So most people felt that they would rather have more low power reactors for the same total amount of money than one real super high power reactor.

Blizard: You really mean the all-purpose reactor turns out to be a no-purpose reactor because it can't do all these things.

Lane: There were a number of discussions to that effect in the various seminars.

Ergen: There was one point which came essentially out of Levy's comments on neutron diffraction work, namely, that we use only one neutron out of  $10^{18}$ . It might be more profitable to get the  $10^{18}$  down to  $10^{17}$ , rather than to increase the reactor power by a factor of 10.

Weinberg: I want to make the general comment that it is really quite dangerous to try to settle on whether to build a reactor like this on the basis of the demands and needs that people can foresee in advance of the existence of the machine. The MTR is really the very best example of this. I remember very distinctly when we were first talking of designing the MTR in 1945, which was just when Wigner came to the Laboratory. The first question he asked was why we wanted  $10^{14}$  flux. He just wasn't very convinced at first that  $10^{14}$  would be a very good idea. In fact, if you look at what it was that first motivated the Oak Ridge National Laboratory to build the MTR,

there were two things which were listed in the justification. The first of these was that we wanted to build a homogeneous reactor which ran at  $10^{14}$  flux, but we couldn't build it at that time. Then we said we needed  $10^{14}$  flux in order to test the solutions that would be used in the homogeneous reactor. That is something which we only barely have used the MTR for, a little bit perhaps; but we still have never had a circulating aqueous fuel solution loop in the MTR.

Lane: A facility was actually built into the MTR for such a loop, but never used.

Weinberg: Yes, never used.

The second reason that we gave for building the MTR was this. We said that we would have a high flux of intermediate energy neutrons, and therefore we would be able to use both velocity selectors and crystal monochrometers to measure the variation of  $\gamma$  in fissionable isotopes. Well, I guess they finally did measure the variation of  $\gamma$  of U-233 in the MTR.

Harvey: When they first set up though they weren't even going to have a chopper there, were they?

Weinberg: A chopper was always planned because there was always a half-mile beam. The MTR has been in many ways one of the most successful, if not the most successful, pieces of equipment the AEC has ever built; but it just wasn't used for the things that one foresaw. So the fact that the case one can make in detail for these other uses is perhaps not completely convincing should not weigh too heavily in the balance of the whole thing. You probably know, John, that Glenn Seaborg is all set to start a very vigorous campaign in which he casts as his antagonists people who want to spend \$100 million for accelerators. If it is indeed the fact that, as Jim says, one can get a  $10^{15}$  reactor for only \$10 million, and not \$100 million, then the argument of its being too expensive is not really an issue.

Well, there are just a couple of other things which I wanted to mention. One of the things that did come out of the discussions was that we really have not sufficiently looked at the question of high flux reactors with poor moderators. These are reactors in which you use fairly high atomic weight moderators so that you build up the number of intermediate energy neutrons far beyond what you have in any of the present reactors.

Swartout: BNL people are doing this; we might profit from what they have done. Kouts has been doing this.

Weinberg: I didn't know this. What kind of moderators?

Swartout: I'm not sure.

Weinberg: Is he really trying to design something, or is he just looking at it?

Swartout: He is just looking at it so far.

Ergen: He is interested in getting intermediate values of  $\eta$  for U-233.

Weinberg: Well, finally there is the question of what the Laboratory ought to do about the whole matter. We have been talking about this thing for a long time, but we have not made a completely firm proposal to the Commission. One reason for this is that we are so busy with other things that we have not been able to devote full attention to this matter. Now we finally have the ORR finished and are now in a position to look at the matter with much more seriousness now than we have previously. In this regard I would suggest, Jim, that the minutes be put together and covered with a summary of what people have found in investigating each aspect of the matter. As a whole, this might prove a very valuable document for the Commission.

Lane: I wonder if other people might not like to comment on their conclusion as to the results of the seminars.

Weinberg: Well, maybe Art Snell would like to comment on his conclusions.

Snell: If I had any conclusions, I would take it all on a matter of faith that given a  $10^{15}$  reactor the experimenters would find it very useful.

Weinberg: You must remember that the MTR is just about a  $10^{15}$  reactor, Art.

Snell: I also think that since we are just getting the ORR we should take a few years getting experience with that.

Williams: What is the flux of the ORR?

Weinberg: About  $3 \times 10^{14}$ .

Snell: Perhaps there is no rush on this.

MacPherson: I would think that logically the next step, from what has been said already, would be to follow along a special purpose line and perhaps look at two or three most promising applications and see what a reactor designed for these particular applications could do. In particular, I know the radio-isotopes people would like to do this and Glenn Seaborg also has the incentive for a particular type of reactor. A third type - I think these are all different types really - is a high intermediate or high energy flux reactor suitable for solid state engineering tests.

Weinberg: I must say that appeals to me. Consider the NRU which the Canadians built and which I think by now might cost about \$60 million for the whole device. It would probably cost \$80 million to start in this country and build the same thing. Now, for \$80 million you can build about 25 ORR's, and the reason I say 25 is that when you build so many you ought to get them cheaper. (Laughter) I can't help but believe that 25 ORR's are

a better bet than 1 NRU. The NRU is a vast thing of 200 Mw - incredibly complicated.

Blizard: I wonder if a program of altering the LiTR to see how you could change a reactor of general design to become a special purpose reactor might not be a sensible way to go right now - try a variety of flux traps, etc.

Weinberg: I am not sure you want to try the flux traps there, but Jim has suggested that Dixon Callihan might get some experiments done over this next year in which you actually look some more at the flux trap concept.

Blizard: This flux trap idea is only one thing, but there are others which might be referred to as high energy flux traps for radiation damage. These may involve different moderators, for example. I wonder if real honest-to-goodness experiments along these lines to see how the design might profitably be changed for a special use wouldn't be worthwhile.

Weinberg: All the holes in both the LiTR and in the ORR have already been spoken for by now.

Blizard: How about using the Swimming Pool critical facility?

Lane: Can't these different moderators and neutron spectra be measured at zero power?

Blizard: A lot of it can be done at zero power.

Weinberg: I think the Swimming Pool would be a very good place to do it.

Blizard: The Swimming Pool is a good place, but you couldn't do cooling experiments.

Lane: That is a problem for the engineers to figure out.

Blizard: You remember why we built the MTR mock-up; we wanted to see what the heat distribution was in it. However, certainly the first step would involve critical experiments or critical arrangements.

Lane: It seems to me that the Laboratory should set up a small group to outline a series of such experiments.

Weinberg: Well, it's more basic than that, Jim. The Laboratory should possibly set up a small group whose main responsibility is  $10^{15}$  flux reactors. Experiments needed in the course of that would come up. I must say I agree with what Art Snell says, that we, after all these years, have just finished the ORR and haven't done one single experiment in it, so we would be in a poor way to seek a new reactor even before we have done much with the ORR.

Lane: Of course we started the ORR in 1950 and it's 1958 now, so it takes quite awhile before you end up with a finished machine.

Weinberg: Yes, that is true. One thing which does concern me a little bit is the question of competition between various groups. Now, I think the Commission always has to make certain that its own laboratories have the best equipment in those areas very directly relevant to the Commission's business. As you probably know, there has been talk in Texas, as well as from the NACA and various other outfits, to build the world's hottest, biggest, most expensive research reactor. Well, I suppose just out of self-defense the Commission probably has to at least look very seriously into this question.

Williams: "To take them under serious consideration" are their precise words.

Weinberg: Argonne has proposed the Mighty Mouse; I don't know how actively they are planning to proceed with it. The situation at Argonne is a little different from here because they do not have a  $3 \times 10^{14}$  reactor at Argonne. The best they have is the CP-5 which even at 5 Mw produces about  $10^{14}$  flux.

Lane: What about the EBWR?

Weinberg: I don't think that does because it has 20 kg of U-235. John, do you have any comments you would like to add to this whole discussion?

Williams: No.

Ergen: As far as this study group is concerned, which was tentatively mentioned, I would recommend that the objective should be defined for each specific purpose reactor. For example, for a reactor to produce trans-plutonium elements we should say we want one that produces them ten times as fast as we are producing them now, rather than merely to say that flux should be ten times as high. This is because it is not obvious that by just increasing the flux you get more than from changing the spectrum. The same thing applies to neutron diffraction.

Weinberg: Would anybody else like to comment on this? Henri, you were out of the room when I was guilty of making the statement that I didn't think the neutron diffraction people had proved that the thing that was holding them up was the lack of a  $10^{16}$  reactor. Do you want to disagree with that?

Levy: I didn't try to prove that. There are plenty of neutron diffraction people that would like higher fluxes, though.

Weinberg: The only people who I thought had made a strong case for  $10^{16}$  flux were the people involved in making isotopes of one kind or another.

Fowler: I would like to bring up a point which I have discussed with various people, although I really haven't done any calculations on it. The point is this -- to produce two neutrons from fission you have to dissipate 200 Mev of energy. If you were to accelerate a proton to 1 Bev and use this to produce neutrons, the dissipation of energy, not counting the energy you have to dissipate in the accelerator, is of the order of 20 Mev per neutron or perhaps a little bit larger. Thus, there would be a factor of 10, roughly, in energy that has to be dissipated. I claim that this in principle means that with protons at 1 Bev you could get 10 times the flux that you could get in a reactor - that is, if the limitation is the dissipation of heat. Anybody could set down and draw up a picture of how you would make the target in such a reactor. You would start with the Materials Testing Accelerator technology.

Weinberg: It won't be cheap though, because what you save in heat transfer equipment you more than make up in the acceleration equipment.

Fowler: No, I am talking about technical range, not the cost range. My bet would be that you could get  $10^{17}$  with such a device. I don't think the arguments apply in this range.

Weinberg: You are saying that you can't get to  $10^{17}$  with a reactor.

Fowler: Yes, I think the transuranic elements are back in again.

Weinberg: That really would be a vast undertaking. It means you would have to have maybe 100 Mw of beam energy.

Fowler: It would take, if you believe my figures, roughly the order of 10 ma. I believe that at 1 Bev that adds up to 50 Mw.

Weinberg: It really isn't what we are talking about now, though.

Fowler: Well, no it isn't, but if you want the ultimate in flux then maybe you want to look at this.

Weinberg: Well, 50 Mw of beam - I don't know. That would be pretty high.

Wollan: It would be good for other purposes.

Fowler: Yes, well that depends on the design.

Weinberg: Are there any other comments that anybody would like to make? John, is there any other comment you would like to make?

Williams: No, I have no other comments to make.

Lane: I wonder if the Research Division has \$10 million for such a reactor?

Williams: Now I'm sure I don't want to make any comment.

Swartout: It's really a little hard to ask for \$10 million because we really don't have a case for  $5 \times 10^{16}$  flux.

Lane: Certainly not in specific detail.

Swartout: I think what Art Snell said is most pertinent. We should have experience first with the ORR.

Blizard: Well, I don't know, it took eight years to get the ORR.

Weinberg: Well, you see, only four of those years were spent after the money had been appropriated.

Blizard: It is pretty pessimistic though to presume that the ORR will not do anything even in four years.

Weinberg: I think though that all John is saying is that we don't quite ask for money from the Commission today, but by no means is he recommending that we drop this matter. His contention is that we keep it alive by further study. If we didn't have the ORR just coming on the line, and if we weren't very heavily committed in many other directions, I suspect we would take a different view. I suppose the Argonne people feel a little differently than we do because they do not have a very high flux reactor.

Lane: I don't believe they are pushing the Mighty Mouse.

Williams: Partly because they are heavily committed, too. I would say that has a lot to do with it. It might be that you won't be so heavily committed.

Weinberg: That might be. We might precipitously find ourselves very unheavily committed. (Laughter) That is why the research divisions are always expected to come to our rescue to find some work for us.

Well, upon leaving, I would like to thank all of you for coming to these meetings and participating, and Jim will put them together and will provide a summary of the matter. We will continue these studies by means of a small group, members of which will be designated in the near future.

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