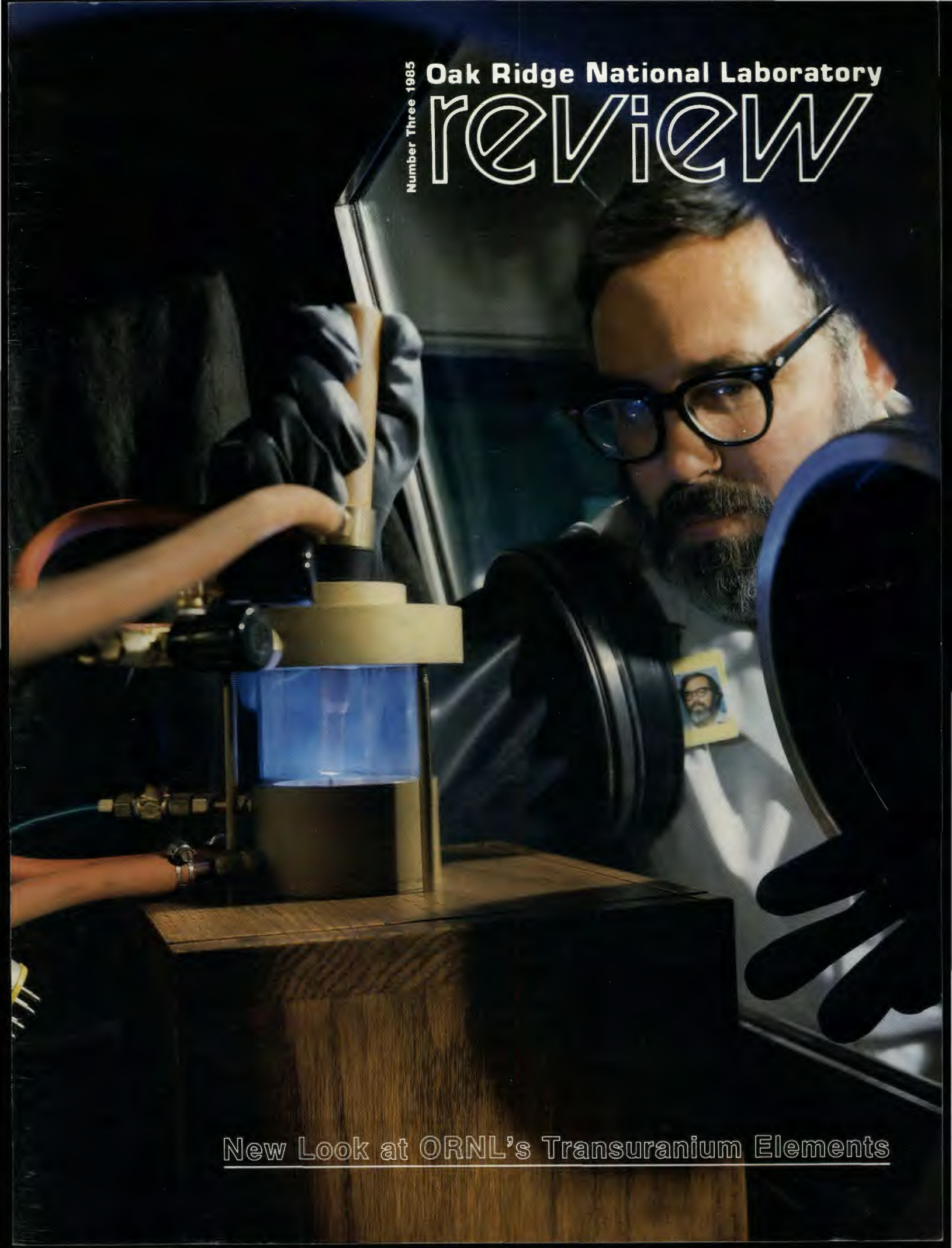
Number Three 1985

Oak Ridge National Laboratory

review



New Look at ORNL's Transuranium Elements

THE COVER: Paul Huray, professor of physics at the University of Tennessee, works in a gloved box with a plasma used to make transuranium metals and alloys. ORNL and UT scientists conduct collaborative studies on heavy elements produced at ORNL's High Flux Isotope Reactor and Transuranium Processing Plant. These man-made, radioactive elements are also vitally needed for international research. See article on page 49.

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DEPARTMENTS

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Samuel H. Liu was born in Taiyuan, China, and received his Ph.D. degree in physics from Iowa State University in 1960. He joined ORNL's Solid State Division in 1981 as a senior research staff member. Before coming to Oak Ridge, he was a research staff member at the IBM Thomas J. Watson Research Center (1960–64); associate professor (1964–67) and professor (1967–81) of physics at Iowa State University; and section chief, Solid State Theory, at the Department of Energy's Ames Laboratory in Iowa (1975–81). He was a visiting professor at Ruhr University, Federal Republic of Germany; University of Copenhagen, Denmark; University of California, Berkeley; and Free University, Berlin. In 1980 he was on a nine-member team from the United States that participated in the Indo-U.S. Conference on the Science and Technology of Rare-Earth Materials in Cochin, India. His research interests include the theory of magnetism in metals, electronic structure of metals, intermetallic compounds and metal surfaces, electrode-electrolyte interactions, mixed valence and heavy fermion materials, and fractals in condensed matter physics. He has written a chapter on the electronic structure of rare earth metals in the *Handbook on the Physics and Chemistry of Rare Earths*. He has been a fellow of the American Physical Society since 1967 and was recently elected to the Executive Committee of the Division of Condensed Matter Physics, American Physical Society, for a three-year term (1985–88). Here, Liu (left) discusses fractals with John Bates in front of the frequency response analyzers used to measure the conducting properties of an electrode-electrolyte interface.

Fractals: *Realm of Monster Curves and Irregular Solids*

By SAMUEL H. LIU

So, Nat'ralists observe, a Flea
Hath smaller Fleas that on him prey,
And these have smaller Fleas to bite'm,
And so proceed ad infinitum.
—Jonathan Swift, 1667–1745

Sometime in 1983 John Bates, a senior research staff member and leader of the Solid Electrolyte and Superionic Conductor group in ORNL's Solid State Division, showed me a paper that puzzled him. I was intrigued by the thesis, that the interface between a metal electrode and an aqueous or solid electrolyte has strange electrical properties. I was even more intrigued by the suggestion that the fractal structure of the interface might be the reason for the odd behavior.

This idea prompted me to start a research program to study the physical properties of fractals, a mathematical concept newly adopted by theoretical physicists to describe a large class of irregular objects in nature (such as the features of mountains, clouds, trees, and the rough surfaces of solid

A solid-state physicist has turned to fractals to understand the strange electrical properties of the interface between an electrode and various electrolytes. He suggests that fractals—a mathematical concept that describes a large class of irregular natural objects—could be useful in other areas of ORNL research.

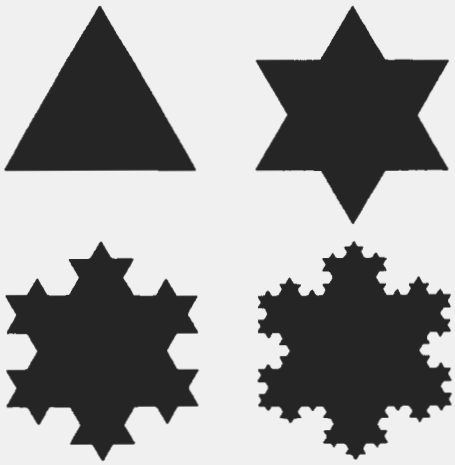


Fig. 1. The first four steps in the construction of a Koch snowflake.

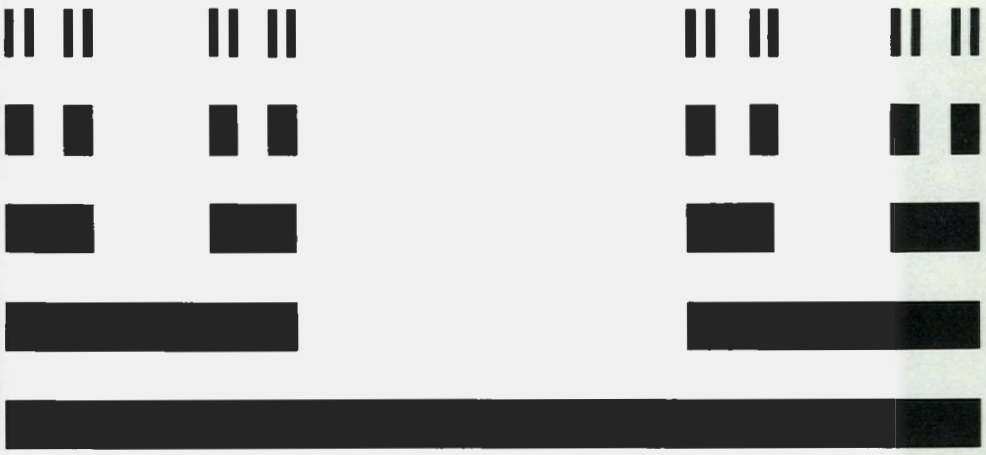


Fig. 2. The first five members of a Cantor bar.

materials, the structure of complex biological molecules, and the paths of particles circulating through the air). The problems are so challenging and fascinating that I have since devoted virtually all my effort to them. So far I have solved problems involving the electrical response of the interface, the lattice vibrations, magnetic ordering, and small-angle neutron and X-ray scattering of fractals.

My enthusiasm is shared by many colleagues at ORNL. The short course on fractals that I gave in the fall of 1984 attracted about 30 participants from many disciplines ranging from physics to biology. In the past 10 years a sudden burst of interest in fractals has occurred worldwide for reasons that I will explain.

What Is a Fractal?

A fractal is a geometrical object that is self-similar under a scale transformation. One excellent example is the flea described in the poem cited above. Jonathan Swift, the British writer, anticipated this new field of physics by more than two centuries. If the flea is magnified sufficiently, each smaller flea would look identical to the original flea, down to the full hierarchy of smaller fleas that infest it. In the same manner, the pockmarked surface of the moon, with its randomly distributed set of craters of all sizes, looks very similar whether viewed through an earthbound telescope or from the much closer vantage point of a moon-orbiting spaceship. A head of cauliflower appears to be a magnified version of its parts. Several dozen examples of fractals in nature are collected in a 1976 book and its 1983 revision by Benoit Mandelbrot, a French mathematician working in the United States. He coined the term fractal because self-similar objects usually have fractional dimensions, and he also publicized the notion of self-similarity as a fundamental symmetry property of fractals. Because scientists have become increasingly familiar with the concept of fractals and have recognized them in the objects they work with in their laboratories, the field of fractals has grown explosively.

The earliest fractals emerged about 100 years ago when a small group of mathematicians looked beyond ordinary smooth curves and invented some monster curves. A classic example is the Koch snowflake shown in Fig. 1. This curve, named after its inventor, the Swedish mathematician Helge von Koch, is constructed by the following process: Start with an equilateral triangle. Next, add one smaller equilateral triangle to the middle of each side to obtain a six-pointed star. In the third stage, add 12 still smaller triangles, one to each side of the star. When the procedure is repeated without end, the resultant shape is the Koch snowflake. The analogy between this shape and the flea envisioned by Swift is quite obvious, provided that we depict the hierarchy of these pests by equilateral triangles.

The boundary of the Koch snowflake (Koch curve) is continuous but not smooth. It makes an infinite number of zigzags between any two points on the curve. The length of the curve between any two points is infinity. Most of us have seen strange objects of this kind at some point during our mathematical training. However, we tend to think of them as aberrations, as skeletons in the closet of otherwise neat mathematics. Like monsters, they are summoned only to scare inexperienced students into thinking rigorously along the line of modern analysis.

At the dawn of the 20th century, the German mathematician Felix Hausdorff suggested a way to generalize the notion of dimension,

Fig. 3. The Cantor bar model for a rough interface.

thereby putting the monster curves in a class of their own. His idea is based on scaling—that is, measuring the same object with different units of measurement. Suppose we measure an area with a measuring square. We do this by finding out how many times the square fits into the area. A 1 m × 1 m square fits into a 2 m^2 area twice, while a 10 cm × 10 cm square fits into the same area 200 times. The ratio between the two results, 200 and 2, is 100, or 10^2. Notice that the exponent of 10, which is the ratio of the length scales 1 m to 10 cm, is the dimension of area. Similarly, we can measure line segments or volumes with unequal measuring sticks or cubes. In every case the dimension of the object measured appears in the ratio of the results as the exponent of the ratio of the length scale of the measuring units.

Applying the same consideration to the Koch curve, Hausdorff found it necessary to add the condition that one must not count any detail smaller than the unit of measurement. Physical scientists may understand the unit as the limit of resolution, namely our ability to discern fine details. Suppose at the beginning stage the Koch curve is 1 cm on a side. With a resolution of 1 cm we see the curve as a triangle (three line segments). If the resolution is improved to 1/3 cm, we begin to see 12 segments each 1/3 cm long, as in the second stage. Every time the unit of measurement is reduced by

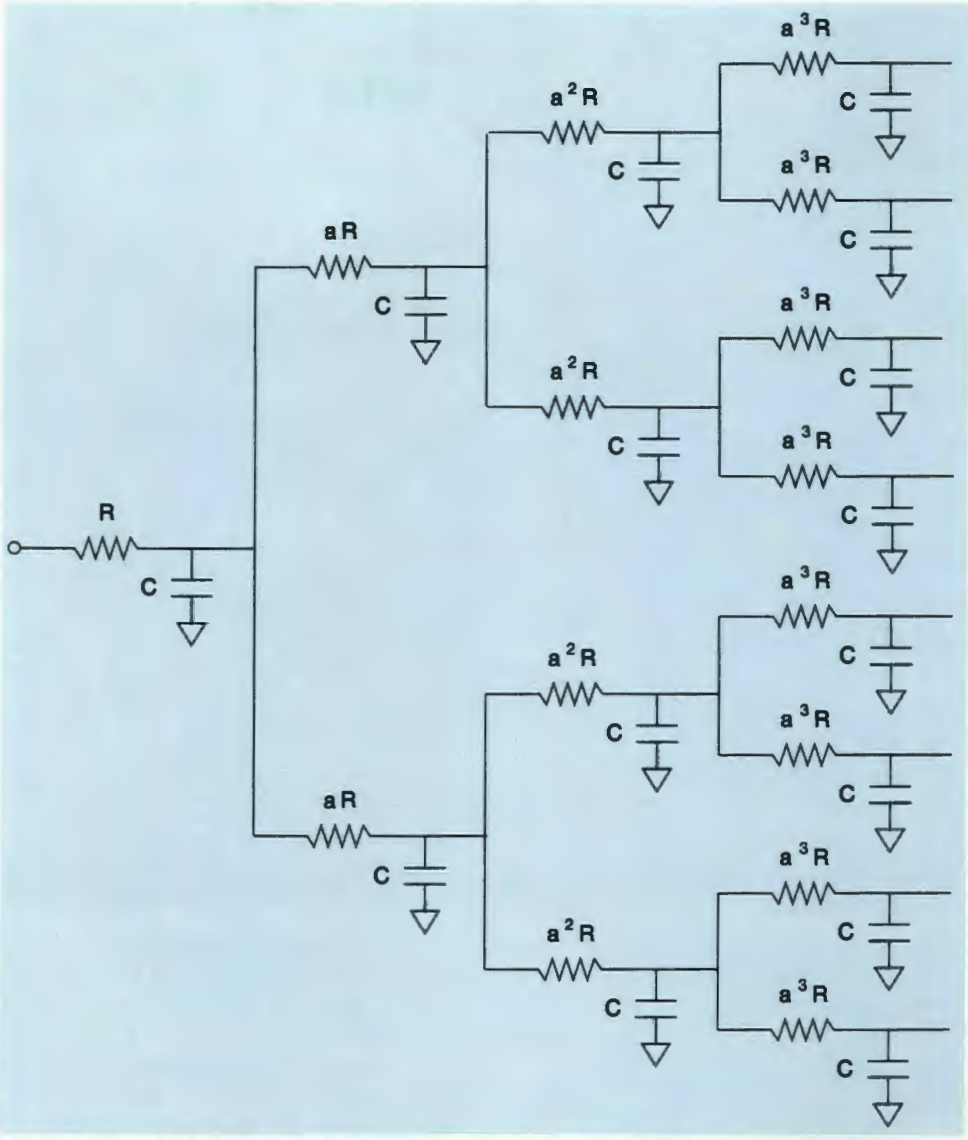


Fig. 4. The equivalent electrical network for the rough interface. Liu constructed this network to simulate the electrical properties of the Cantor bar model (turned on its side).

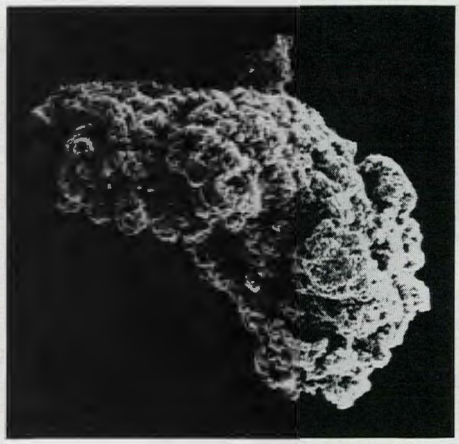
a factor of 3, the number of visible segments increases four times. Following the rule of the last paragraph, we equate 4 to 3^d to obtain the Hausdorff dimension of the Koch curve: $d = \ln 4/\ln 3 = 1.26$. Therefore, the strange properties of the snowflake stem from the fact that it is not a one-dimensional object. It belongs in the never-never land of fractional dimensions. Hence the name fractal.

Like the Koch curve, the North American coastline appears longer when measured with finer resolution. Its Hausdorff dimension is around 1.2. Typically, fractals in nature lack the regularity of the snowflake; however, they are self-similar in the statistical sense—that is, with a large enough collection, one can magnify a small portion of one member and match it closely with some other member of the collection.

The Interface Problem

The problem that aroused my interest in fractals is the following. According to theory the electrode-electrolyte interface should behave like a pure capacitor, an electronic device that stores and releases electrical charges. When an

The negative of the electron micrograph of a zinc electrode is being inspected with a laser light source. The resemblance between the image and a head of cauliflower in the foreground is striking.



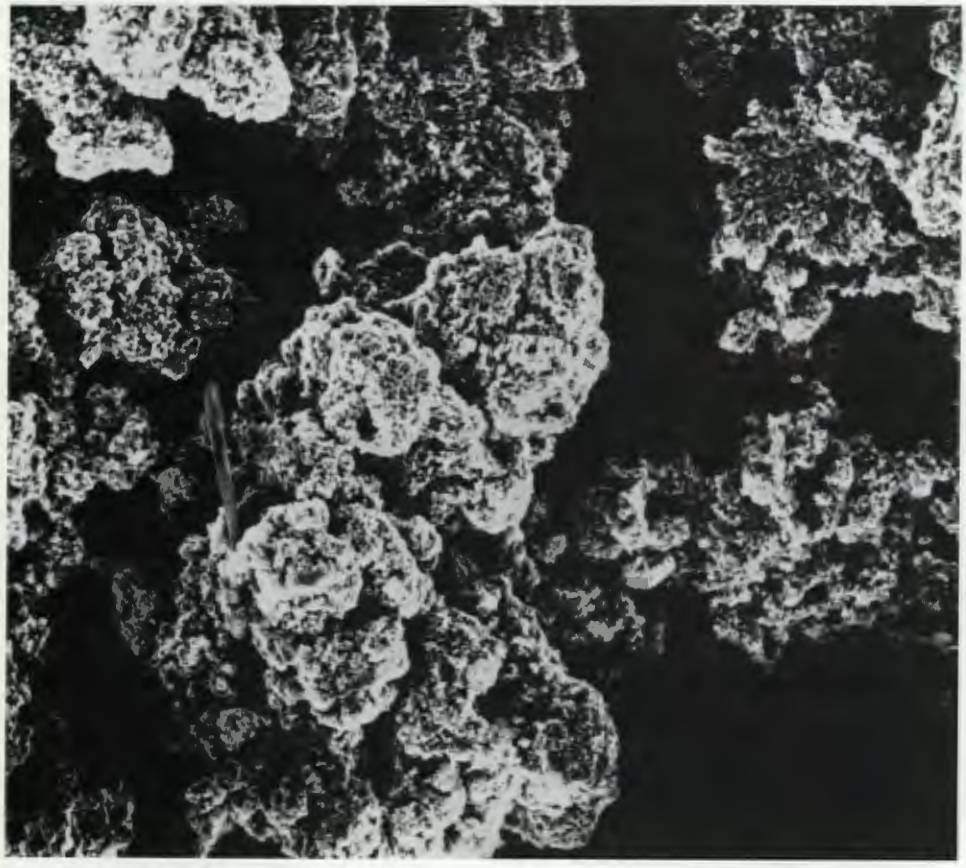
The zinc electrode used in the impedance measurement magnified 30 times. The electrode was grown in Bates's laboratory by electrodeposition. At right is the same zinc electrode magnified 100 times. The similarity between the two images is an indication of the fractal nature of the electrode surface.

alternating current (ac) passes through the interface, it is expected to encounter an impedance (resistance to the current), which is simply related to the frequency (cycles per second) of the current. What is seen in the laboratory, however, disagrees completely with the theoretical expectation. In fact, the impedance has been found to be inversely proportional to the frequency raised to a fractional power between zero and one. Such a behavior cannot be understood on the basis of the existing model of the interface.

The science of electrode-electrolyte systems has a long history, owing to its importance in electrochemical technology, storage batteries, and corrosion. The earliest mention of the unexpected electrical property of the interface appeared in the literature in 1926. Through careful experimentation many scientists concluded recently that the anomalous property is related to the roughness of the electrode surface. When the surface is made increasingly smooth, the exponent steadily approaches one. Under magnification even well-polished solid surfaces show long grooves with jagged surfaces. The fractal connection stems from this observation. In the paper Bates showed me, the authors made an attempt to prove the connection mathematically. Unfortunately their proof fails at the most important step. Could I do it right?

When theoretical physicists approach a problem, they seldom confront it head-on because nature is invariably far more complex than we can analyze with our limited mathematics. Theorists must idealize the problem into a soluble model, and they hope that the solution will allow a glimpse of nature's inner secrets. They do not attempt to paint a realistic portrait of nature. Instead, they try to capture its spirit with a caricature. Very often the making of a model is as creative as, say, writing poetry.

The path that led me to the model interface was almost as tortuous as the monster curve. No fewer than half a dozen models appear in the scientific literature, but none has the fractal property. Needless to say, they all fail to explain the observed electrical property. I had no problem thinking up several fractal models, but I could solve none of them. Finally an inspiration came to me when I was giving a lecture for my short course. I discussed a fractal called the Cantor bar (see Fig. 2) to make the point that a fractal dimension could be less than one. The bar, named in honor of the German mathematician Georg Cantor, begins with a solid bar. Next, the



bar is broken into two pieces, each of length $1/a$ of the first, with $a > 2$. In the third step the two shorter bars are broken according to the same rule. The procedure is repeated ad infinitum. The dimension of this object is $d = \ln 2/\ln a < 1$.

While sketching this fractal on the blackboard, I suddenly conjured up a mental image of the Cantor bar with all pieces linked together, as in Fig. 3. I somehow perceived a resemblance between this Rorschach inkblot and the cross section of a rough surface. A set of bars looks like a groove on the electrode. The groove carries smaller grooves to make it a fractal. Undoubtedly other scientists will question how realistic this model is, but I had my mind set on solving the model as my first try at understanding the strange electrical properties of the interface.

The next step in the model-making process is to give electrical properties to the geometric pattern. When an electric current crosses the interface from the electrolyte side (black) to the electrode side (white), it can go via two different paths, either across the interfacial capacitance or down the branches of the grooves. The electrolyte is usually the high-resistance material while the electrode, a metal, has negligible resistance by comparison. Therefore, I represent the branches of a groove as resistors in the network shown in Fig. 4. The resistances increase by a factor of a at every branch because the cross-sectional area decreases by that ratio. The capacitances represent the alternative paths across the interface. I have made another idealization at this point by assigning the same capacitance to every branch. This approximation amounts to counting only the interfacial capacitance on the two side faces of the branch and ignoring the one in the dip between branches. As the branches become thinner the interfacial area of the dip becomes very small, and the simplification does not affect the final result. The alternating electric current passes from the electrolyte into the input terminal of the network and eventually enters the common ground, which represents the electrode.

Now mathematics takes over. The input impedance of the network in Fig. 4 can be expressed as an infinite continued fraction (shown below in box).
In this equation ω is the angular frequency (2π times the frequency), j is the square root of -1, and the symbol $Z(\omega)$ indicates that the impedance, Z, is a function of the frequency. We use complex numbers in circuit theory as a convenient way to keep track of the phase shift of the current.

Finite continued fractions are subjects of elementary algebra, but infinite ones require special handling. Here the self-similarity becomes very useful. I was able to use this symmetry to derive the frequency scaling relation

$$Z(\omega) = R + \cfrac{1}{j\omega C + \cfrac{2}{aR + \cfrac{1}{j\omega C + \cfrac{2}{a^2R + \cfrac{1}{j\omega C + \ldots}}}}}$$

$$Z\left(\frac{\omega}{a}\right) = \frac{a}{2} Z(\omega) \ .$$

for $\omega RC \ll 1$. The solution of this equation is simply

$$Z(\omega) = K(j\omega)^{-\eta}$$

where K is a scale factor and $\eta = 1 - \ln 2/\ln a = 1 - d$. Thus the impedance is inversely proportional to the frequency raised to the power η, and the exponent is related to the geometry of the electrode surface.

After this in-depth analysis of one model, I felt confident enough to take a great leap of faith and apply the result to the real interface. We normally determine the dimension of a rough surface by measuring its area using different resolutions. For the model interface the surface dimension turns out to be $d_s = 2 + d$. Thus the relation between the exponent and the dimension of the surface is $\eta = 3 - d_s$. I propose that this relation holds in general. The dimension of a surface is a measure of its roughness. A smooth surface has a d_s close to 2. Consequently η is close to 1, in qualitative agreement with experiments.

The ball is now back in the court of Bates and his experimental group. They are devising ways to check out my theory quantitatively in their laboratory. They will try to measure the parameters in my model to see whether the relation between the impedance and the frequency would be observed in the correct frequency range. They must determine the dimension of the rough electrode surface independently to verify the relation between η and d_s.

Thus what seemed at the beginning to be a theorist's pipe dream has become a crucial step in the arduous course of scientific investigation. I have related one example of how an abstract concept in theoretical physics has contributed to progress in interfacial science, which includes the science of electrolysis, batteries, and corrosion.

"ORNL researchers have found fractals useful for describing the molecular structure of cellulose and graphite fibers, the adsorption of gases on solid surfaces, and the growth of dendrites in metal alloys."

The turbulence in the hot gases emitted by Mount St. Helens during the May 18, 1980, eruption gives the plume its fractal appearance.

ORNL researchers have found fractals useful for describing the molecular structure of cellulose and graphite fibers, the adsorption of gases on solid surfaces, and the growth of dendrites in metal alloys. Others might find them useful for modeling diffusion processes, such as oil or groundwater seeping through porous rock, plant species spreading in a forest, or the onset of superconductivity in a thin metallic film; for studying the tortuous path that air pollutants take in collisions with atmospheric molecules; or for describing the irregular surface of a fractured piece of metal.

Will our inquiry lead to a more efficient battery or to a way to prevent corrosion? Will fractals move into other areas of the natural sciences? We cannot predict the future any better than the inventors of monster curves could have imagined that their strange world of in-between dimensions would one day become the happy hunting ground of theoretical physicists. ornl

BOOKS

Weapons and Hope, *Freeman Dyson, Harper & Row, New York, 1984 (340 pages). Reviewed by Jack Barkenbus, senior researcher and specialist in nuclear arms issues at the Institute for Energy Analysis, Oak Ridge Associated Universities.*

Most books written on nuclear weapons today are intended to scare the reader. *Weapons and Hope* seeks instead to educate the reader—a much more difficult task—and succeeds beyond question.

This book, which recently was given the National Book Critics Circle Award for general nonfiction, conveys a sense of optimism despite its gloomy subject. The optimism is based not on complacency about the current state of affairs but on the belief that we, as fallible human beings, can find a way out of the nuclear impasse if we search hard enough.

Far from being a Pollyanna, Dyson rejects the easy but ultimately futile routes of escaping from the nuclear predicament, such as unilateral disarmament and the nuclear freeze. Instead he finds hope in new technologies that incorporate advancing computer and sensor capabilities. Dyson's thesis is that these emerging capabilities fundamentally shift the balance of forces away from offensive dominance ("brute force") and toward defensive dominance, which is based on the possession of advanced information- and battle-management capabilities.

Consequently, he envisions the eventual development of small (1-kg, or 2-lb!), highly accurate, nonnuclear, ground-based missiles or rockets that can intercept lumbering offensive missiles (a "non-nuclear David slaying the nuclear Goliath"). He is also heartened by the continued development of Precision Guided Munitions—small, nonnuclear missiles designed to strike tanks, airplanes, and ships—and believes they will serve admirably in the defense of western Europe, thereby allowing for the eventual removal of tactical nuclear weapons from the European front.

Despite his faith in technology, Dyson holds no hope for a space-based, antimissile system, as conceived by President Reagan's "Star Wars" plan. Space systems and futuristic technologies such as high-energy lasers and particle-beam generators are too elaborate and too fragile or vulnerable, Dyson writes, and he calls the plan a "technical-follies future."

The "Star Wars" emphasis on technology concerns Dyson, because he rejects the possibility that technical fixes—even when they are his own preferred technologies—can lead us out of the debilitating arms race. New technologies, he asserts must be accompanied by negotiated arms reductions and, more importantly, by a change in government policies so that each superpower opts for assured survival of its own nation, as opposed to the assured destruction of the other. Mutually assured destruction, or MAD, would then become a thing of the past.

Dyson is hopeful that this policy or security concept—one he calls "Live and Let Live"—will ultimately be adopted by both U.S. and Soviet leaders and that it can bridge the gap he sees between the perceptions of the military (the "warriors") and the general public (the "victims"). He is asking for a lot; yet the case he makes against relying on the concept of assured destruction for our security is compelling.

This is not an ordinary book about nuclear weapons, though it covers much of the standard ground, including discussion of various nuclear weapons systems and the strategic theories or concepts that have grown up around them. What distinguishes this book, apart from its tightly woven logic and wisdom, is the author's well-constructed prose and his unique approach to the topic.

In many ways it is a personal memoir. Dyson, who is now a physicist with the Institute for Advanced Studies at Princeton University, served as a technical expert for the British Royal Air Force in World War II. His uncle was killed in World War I. The author recounts wartime experiences in considerable detail, not to establish his credentials but to illustrate the follies that have accompanied humans into war and to warn against their reappearance.

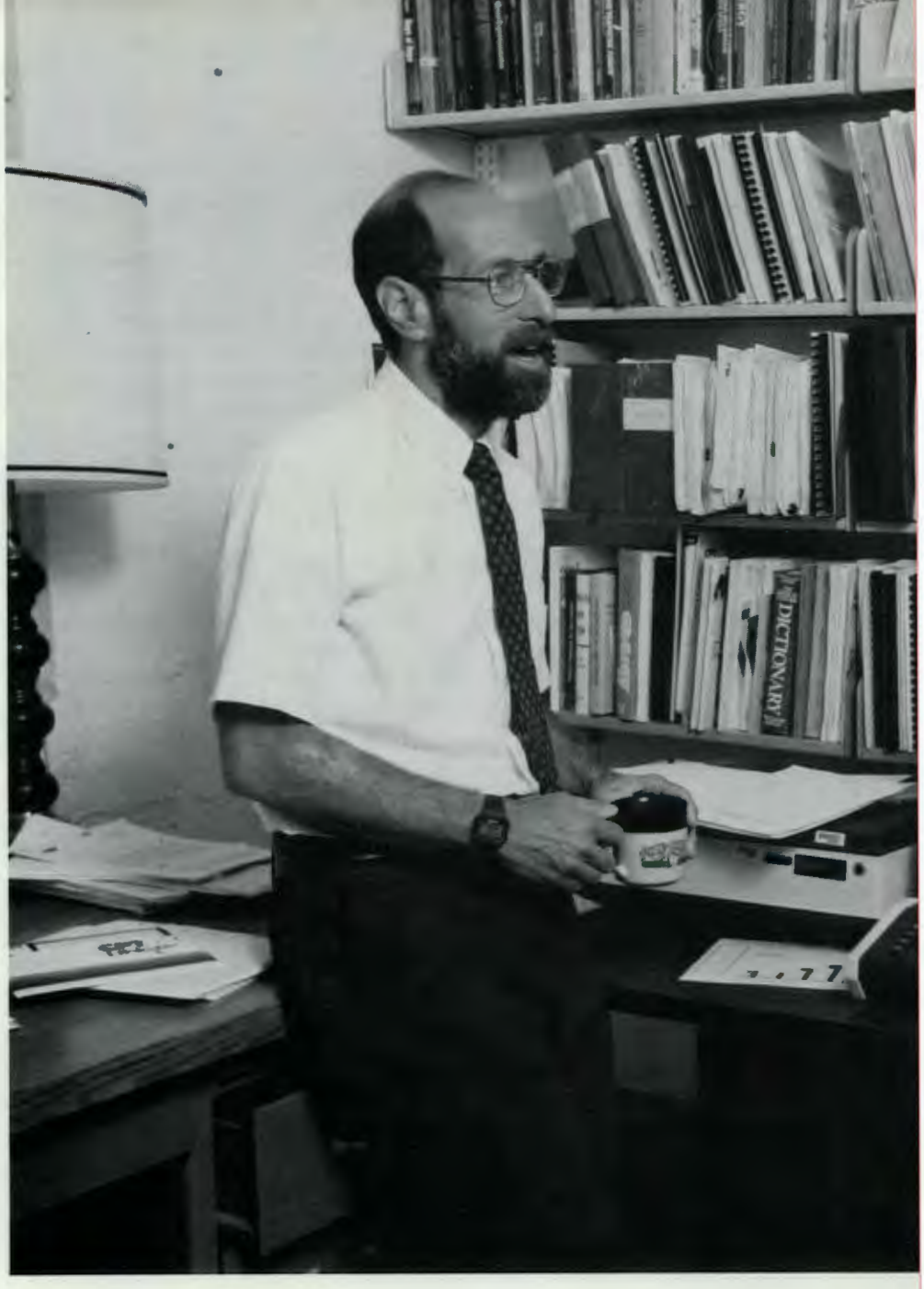
Dyson has not given us a blueprint for achieving a stable and secure peace. Much additional thought, analysis, and political consensus-building remains. He has, however, pointed us in a direction that increasingly will be seen as wise and correct.

On the last page of his book, he states, "We shall not be finished with nuclear weapons in a year or in a decade. But we may, if we are lucky, be finished with them in half a century. . ." That is the hope, and Dyson challenges us to muster the will and insight necessary for its realization. ornl

Conservation as an Energy Resource:

Electricity Savings from a Utility Program

By ERIC HIRST



A fable: On January 22, 1983, in the state of Washington, John Smith arrived home from work, sorted the mail, and opened the monthly bill from his electric utility, the Clark County Public Utility District (PUD). He was astounded to see that he owed $105, the highest electric bill his family had ever received. (It sounds low to us in East Tennessee, where electricity costs 5 cents/kWh, but 3 cents/kWh is expensive to them!)

For John, this bill touched a raw nerve. Angrily, he called the utility to complain and threatened to convert his heat source from electricity to wood.

Jean Jones, the Clark County PUD customer service representative, suggested that John let the utility conduct a free home energy audit. The audit, she

What are the energy and economic impacts of a utility program in the Pacific Northwest that offered homeowners incentives to reduce electricity use? An ORNL team has completed a study of the benefits and costs of such a program.

Eric Hirst, a mechanical engineer who has worked at ORNL since 1970, is a leading expert on energy conservation. His findings have been of interest to utilities and many policymaking groups, including the U.S. Department of Energy, the U.S. Congress, and state energy agencies. As leader of the Evaluation Group in the new Decision Systems Research Section of the Energy Division, he is responsible for designing and carrying out detailed quantitative evaluations of utility and government energy conservation programs. Recent projects involve developing methods to accurately determine the energy savings from and economics of utility-sponsored conservation programs. These methods have been used to evaluate home energy audit-and-loan programs in the Pacific Northwest, Minnesota, Connecticut, Florida, and New Jersey. In 1979 Hirst was on assignment with the Minnesota Energy Agency. For 15 months in 1974–75 he served as director of the Office of Transportation Research in the Federal Energy Administration. He holds a Ph.D. degree in mechanical engineering from Stanford University. He has published more than 200 reports and papers on the engineering, economic, and policy issues of energy use and efficiency, especially in the residential and commercial sectors. Here, Hirst (left) and Rick Goeltz of the Energy Division discuss the economics of utility conservation programs in various regions.

explained, identifies ways to cut electricity use for space and water heating through retrofit measures such as installing attic insulation, caulking and weatherstripping, and adding insulation to the water heater tank. Persons taking measures that the energy audit identifies as cost-effective, she added, are eligible for a cash rebate from the Bonneville Power Administration (BPA).

According to Jean, other Clark County PUD customers who received energy audits and rebates have had 90% of their retrofits paid for by BPA. In other words, a $2000 package of energy conservation measures for John's home would cost him only $200. In addition, the reduction in John's electricity bill should amount to about $150 a year.

John was sold on the program. He asked for an audit, which was conducted in early March. After the audit identified several retrofit measures needed in John's house, he and the utility agreed on a contractor, who installed storm windows, insulation, and weatherstripping in late April. The total cost was $1700, but BPA paid $1500 of that amount. John happily paid only $200 and felt more comfortable about opening his monthly electric bills.

John Shaw, district meterman for Pacific Power and Light Co., tests one of the monitoring units to be installed on a sample of 320 Hood River area homes. These homes will be monitored continuously to determine consumption of energy by space and water heating, both before and after weatherization measures are installed.

Installing residential insulation in a home as part of the BPA conservation program.

Conservation Issues

We made up the preceding scenario, but similar events did occur in 104,000 households throughout the BPA service area of Washington, Oregon, Idaho, and western Montana during 1982 and 1983. The families in these households enjoyed the benefits of having BPA pay 90% of the cost of their conservation retrofits. One question raised by this and other utility conservation programs throughout the United States concerns how much actual electricity savings can be attributed to them.

Private and public utilities like BPA and the Tennessee Valley Authority fund residential conservation programs out of the conviction that saving energy costs less than the construction and operation of generating plants that would be needed if energy demands were not restrained. Some economists, however, argue that the recent rise in energy prices alone reduces energy demand enough to head off the need for additional power plants. Policy issues that utilities want addressed include these: Do programs that induce residents to weatherize their homes stimulate greater reduction in electricity use than rising energy prices do? How much money does a utility and its customers save if the utility invests in residential conservation programs and defers construction of new power plants? Do rises in energy prices alone keep demand for electricity low enough to avoid the need for new power plants?

The answers to these questions depend on accurate estimates of actual program-induced electricity savings. These savings represent the primary benefit of such programs, against which the program costs must be weighed. In the case of the BPA program, $160 million were spent for 104,000 retrofits in 1982 and 1983.

What did BPA and its customers receive for their money? To determine the actual program-induced electricity savings in one program, Oak Ridge National Laboratory recently led an evaluation of BPA's Residential Weatherization Pilot Program, the forerunner to BPA's ten-year, region-wide program. The evaluation involved Rick Goeltz, Linda Berry, Bruce Tonn, Dennis White, and me in ORNL's Energy Division, as well as staff members in the Evaluation Research Corporation office in Portland, Oregon. BPA's Office of Conservation sponsored the evaluation.

The pilot program was operated by BPA from 1980 through 1982 and administered by 11 small,

This 24-MW hydroelectric power plant on the BPA system is in Portland, Oregon. The shortage of new hydro sites and the high cost of coal and nuclear power plants sparked the great interest in conservation as an energy resource in the Northwest.

public utilities throughout the Pacific Northwest. The program offered free home energy audits to identify cost-effective conservation measures. It also included zero-interest, deferred-payment loans for electrically heated single-family homes to cover the cost of the weatherization measures recommended during the audit. About 7200 homes were audited and 4100 loans, averaging $2200 each, were made. The total cost of the program (including audits, loans, postinstallation inspections, and administration) was $11 million.

BPA's interest in conservation programs stemmed from the rapid growth in electricity demand during the 1970s, the shortage of additional hydroelectric sites, and the high cost and long delays associated with construction of coal and nuclear power plants. In response to these dramatic changes in the electricity supply–demand situation, the U.S. Congress passed the Pacific Northwest Electric Power Planning and Conservation Act in 1980. The act gave BPA substantial authority to implement programs to meet future electricity demands in the region; the act also emphasized conservation as an important resource in the Northwest.

Data and Analysis

We collected electric-bill data from 311 households, 179 of which participated in the program (in mid-1981). We started the year before participation and stopped three years after. In addition, we obtained daily temperature data from National Oceanic and Atmospheric Administration weather stations within each utility service area; these data were used with the billing data to adjust household electricity use for changes in winter severity from year to year. We also obtained information from the utilities on each participant's home energy audit and postinstallation inspection (to ensure, for example, that the recommended amount of insulation had been properly and fully installed by the contractor). Finally, we had results of a telephone survey of both conservation program participants and nonparticipants in the fall of 1982. Altogether, we had complete and usable data for 179 participant households—they had been audited by and received loans from the BPA program—and 132 nonparticipants.

Using this rich data base, we conducted several analyses of interest to BPA. In addition to estimating the program-induced electricity savings, we examined the economics of the program from the perspectives of the participants, the BPA power system, and the Pacific Northwest region as a whole. We also compared the characteristics of program participants with nonparticipants to suggest marketing strategies that would increase program participation. Finally, we compared actual electricity savings with audit predictions to identify the key

factors that contribute to large electricity savings.

All these analyses involved considerable effort on our part to "clean" the raw data. Even though a utility's monthly meter readings form the basis of its income, the readings contain many errors. Meters are sometimes misread, not read at all, or broken, and determining the dates on which meters were read is sometimes impossible. Families move into and out of houses, resulting in changing patterns of energy use in certain homes. All these factors complicate development of a useful data base. Hence, we conducted various engineering and statistical analyses to examine the impact of these factors on the validity of the data and to make corrections to improve the accuracy of the data base.

The most important purpose of ORNL's evaluation was to develop accurate and credible estimates of the electricity savings that could be attributed directly to the pilot program. The analyses we conducted yield estimates of the total and net energy savings induced by BPA's pilot program. *Total* saving refers to the reduction in annual electricity use in the households receiving an audit, a loan, and retrofits as part of the program. The *net* saving is the difference between the total saving and the saving that these homes would have achieved on their own had no BPA program existed.

For example, we assumed that participating households would have reduced electricity use because of increased energy prices. To infer the no-program energy savings for participants, we used information from the nonparticipants because they, too, would cut back on electricity use in response to higher prices. As far as we know, this study is the first one to closely examine the actual (measured) energy savings that result from a conservation program over such a long time span.

	Audit + loan[a] (kWh/year)	Nonparticipants[b] (kWh/year)
Electricity use		
year 1 (preretrofit)	29,350	25,410
year 2 (postretrofit)	23,940	23,840
year 3 (postretrofit)	22,820	22,670
year 4 (postretrofit)	22,420	21,890
Total savings		
years 1–2	5,410 (17)[c]	1,570 (5)
years 1–3	6,530 (21)	2,740 (9)
years 1–4	6,930 (22)	3,520 (12)

[a]179 households.
[b]132 households.
[c]The numbers in parentheses are the percentage savings relative to year 1.

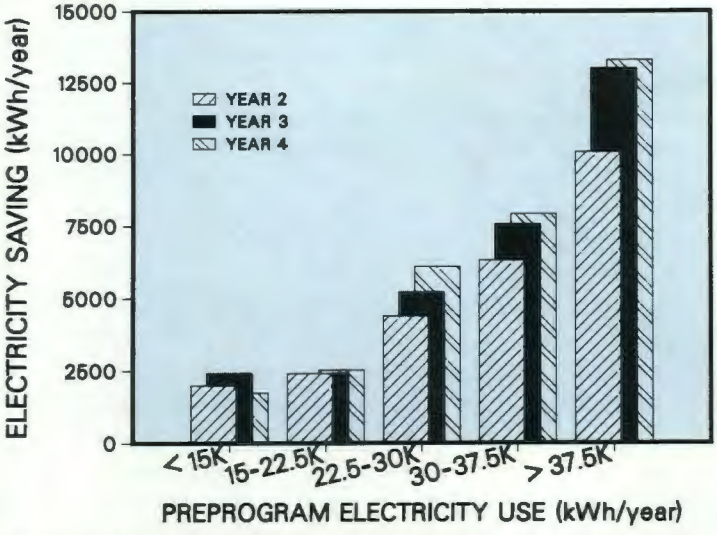
Results

Weather-adjusted electricity use the year before the BPA program began (1980–81) was about 10% higher, on average, for the audit-loan (AL) households than for the nonparticipant (NP) households. Roughly speaking, for both groups total electricity use was split equally between heating and baseload uses such as lighting and running appliances.

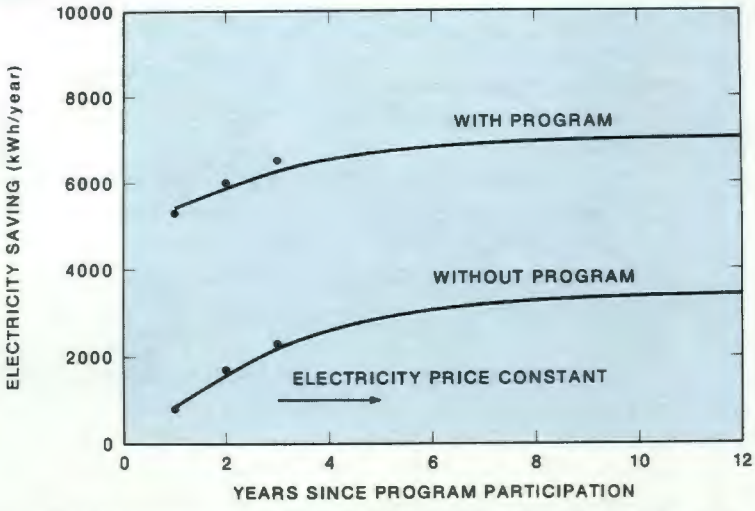
We found that reduction in electricity use from year 1 to year 2 was greater for AL households, averaging 5400 kWh per household per year compared with 1600 for NP households. However, the reduction in electricity use between years 2 and 3 was about the same for the two groups of households, roughly 1100 kWh. The reduction between years 3 and 4 was less than 1000 kWh per home for both groups of households. The overall effect was an average four-year reduction of 6900 kWh/year for the AL households and 3500 kWh/year for the NP households. In other words, the participants reduced their overall annual electricity consumption by 22% relative to that used the year before the program began; the nonparticipants used 12% less electricity.

The substantial reduction in electricity use for nonparticipants is surely due in part to the large increases in electricity prices in the Pacific Northwest during this period. Averaged across the households in the evaluation, real (excluding inflation) electricity prices increased by 72% between years 1 and 4. Other forces that may have affected household electricity use include public awareness of energy issues, knowledge of the potential for saving money through energy conservation actions, changes in household income, and overall changes in the region's economy.

Household electricity use depends on a variety of factors, including demographics (number of people in the home, their ages, household income) and structure characteristics (size and age of home, use of nonelectric fuels for heating), as well as weather conditions such as unusual heat waves. Although the analysis corrects household energy use for differences from year to year in



Increases in annual electricity saving in years 2, 3, and 4 (postprogram) as a function of preprogram electricity use for 179 audit-loan households are shown in this bar chart. Note the strong positive correlation between actual electricity saving and preprogam electricity use.



Electricity savings attributed to the Bonneville Power Administration program as a function of time are shown in this graph. This simulation assumes that the real price of electricity remains constant after 1984.

winter severity, it does not consider these other factors.

To adjust for these differences across households and thereby identify the electricity-saving effects of the BPA program more accurately, we used annual electricity consumption as the dependent variable in a statistical model. That is, for each household, values of kilowatt-hours per year for the four years are the dependent variables. We took into account factors affecting annual electricity use, including household income, number of household members, floor area, annual wood use, primary heating fuel, electricity price, and variables that distinguish between program participants and nonparticipants before and after retrofit.

Our model shows that annual electricity use increases with household income, number of household members, floor area, and use of electricity as the primary (rather than supplemental) heating fuel. Electricity consumption declines with increasing wood use and rising electricity prices.

According to the model results, the AL households used 2300 kWh/year more than the NP households during year 1 (before the program started). The reductions in postprogram electricity use for the AL households were also very significant; results show per-household savings of 4500 kWh/year in year 2, 4300 kWh/year in year 3, and 4200 kWh/year in year 4 because of participation in the BPA pilot program.

Conclusions

As more and more electric utilities operate energy conservation programs, it becomes increasingly important to determine the actual energy-saving effects that the programs have. Careful measurement of program effects is critical because utilities want to compare program benefits to the costs of the programs and of conventional power supply resources that the programs are intended to displace. (Investment in energy conservation programs may be thought of as the purchase of "conservation energy.")

For BPA's Residential Weatherization Pilot Program during 1980 through 1982, our estimates of the *net* program savings average 4500, 4300, and 4200 kWh/year per AL household in years 2, 3, and 4. The comparable *total* energy savings per AL household was 5400, 6500, and 6900 kWh/year.

During the first three years after BPA-financed retrofits were installed, program-induced electricity savings persisted, diminishing by only 10%. The portion of the total electricity saving experienced by these households that can be attributed directly to the BPA program declined from 83% (4500 kWh/5400 kWh) in year 2 to 61% (4200 kWh/6900 kWh) in year 4. This decline occurs because the primary effect of the program is in year 2 (immediately after retrofit), while rising electricity prices and other factors have an impact each year. The results suggest that, even without the BPA program, these households would ultimately have taken at least some of the energy-conserving measures financed by the BPA program. Nevertheless,

Economics of a Utility Conservation Program

The bottom line for any utility program is its overall worth. For the Bonneville Power Administration (BPA) Residential Weatherization Pilot Program, the question boils down to whether the $11-million cost of the program was justified by the value of the electricity savings.

Program benefits and costs were analyzed as part of our evaluation for BPA. Using estimates of program energy savings discussed in the main article, we computed the net present worth (NPW) of the program (benefits minus costs). These NPW figures were developed for three different perspectives: the audit-loan participants, the BPA power system, and the Pacific Northwest region as a whole. The analysis was conducted twice, once with the economic assumptions used in 1982 and once with current (early 1985) economic assumptions. The results are quite different because the marginal cost of electricity was thought to be high in 1980, when power shortages were expected; today the region enjoys a large surplus of electricity, which leads to a much lower marginal cost.

Marginal cost is an economic term that refers to the cost for the next increment. In this context, marginal cost refers to the price that BPA must pay to produce or purchase an additional kilowatt-hour of electricity. BPA's calculation of marginal costs is based on the cost of constructing a new 1000-MW coal-fired power plant.

The 1982 assumptions yield results that show large benefits from the BPA pilot program for all three perspectives. The NPW is $4400 per participant for the region, $400 for the BPA power system, and $2500 for the participants themselves. (All figures are in constant 1981 dollars.)

The 1985 assumptions, by contrast, yield NPW estimates of $1800 per participant for the region, −$1000 for BPA, and $2800 for the participants. The marginal cost assumed in the 1982 evaluation was 3.8 cents/kWh, nearly double the current value. Participant benefits are slightly higher because BPA's 1985 forecast of residential electricity prices is higher than its 1982 forecast.

BPA's estimate of marginal cost dropped dramatically, primarily because forecasts for the growth of electricity demand are much lower now than they were just a few years ago. Less need for power in the future reduces the need to construct expensive power plants; thus the marginal cost of electricity is lower.

The NPW figures represent the present worth of *lifetime* benefits. They are not *annual* benefits. The NPW approach computes program-related benefits for each year until the retrofits wear out (after about 30 years). Each year's saving in electricity bills is then discounted (at a real rate of 3%, equivalent to a nominal interest rate of about 8% nowadays) and brought back to the present. For example, a $100 saving in 10 years has a net present worth of $74 with a 3% discount rate ($46 if discounted at 8%).

Results obtained with the 1985 assumptions suggest that the program is economically attractive for the Pacific Northwest region as a whole. The negative results for the BPA system (i.e., −$1000) indicate that electricity rates for nonparticipants would be higher with the program than without it.

The apparent discrepancy between the regional and BPA results occurs because of the difference in perspective. The regional view (which includes all the people in the region) values the *total* electricity savings at the marginal cost of power, while the BPA view (which includes BPA ratepayers—both program participants and nonparticipants) values the *net* saving at the *difference* between the marginal and average costs of power (the latter reflects the revenue lost to BPA because of reduced electricity sales as a result of the conservation program). The positive results for the region suggest that the cost of electricity services—essentially the product of electricity price and consumption—is lower with the program than without it, while the negative BPA results suggest that electricity prices are higher with the program than without it. Thus the regional view, which deals with services and prices rather than with prices alone, suggests that the BPA program is indeed economically justifiable.—*E. H.*

our evidence shows that the program yields dramatic energy savings for at least a few years.

These findings demonstrate the importance of measuring the effects of conservation programs for several years after participation begins. If utilities purchase "conservation resources" as cost-effective alternatives to conventional supply resources, they must be confident that the improvements in energy efficiency will last. Our results—based on three years of postparticipation electric-bill data—show that the energy savings directly attributed to the BPA pilot program are substantial for residents and diminish slowly during the first three years. This is good news for utilities and for countless electricity consumers like John Smith. ornl

take a number

BY V. R. R. UPPULURI

Fun with Foursome

Take any four integers, say, 13, 9, 1, and 6. Look at the successive differences: $13 - 9 = 4$, $9 - 1 = 8$, $1 - 6 = -5$, and $6 - 13 = -7$. Change the negatives to positives and consider the foursome 4, 8, 5, and 7. Look at the successive differences again: $4 - 8 = -4$, $8 - 5 = 3$, $5 - 7 = -2$, and $7 - 4 = 3$. After changing the negatives to positives, we have a new foursome, 4, 3, 2, 3. In determining the successive differences, we obtain $4 - 3 = 1$, $3 - 2 = 1$, $2 - 3 = -1$, and $3 - 4 = -1$. By changing the negatives to positives, we get 1, 1, 1, 1. By repeating the above process, we obtain the foursome 0, 0, 0, and 0.

Using any four integers, the above process always ends up with zeros. However, given any three integers, this process does *not* necessarily lead to zeros. For instance, consider the triplet 1, 5, and 2. After one step we get $1 - 5 = -4$, $5 - 2 = 3$, and $2 - 1 = 1$; the results 4, 3, and 1 lead to 1, 2, and 3, and their successive differences yield 1, 1, and 2. Continuing in this vein we obtain 1, 1, and 2; then 0, 1, and 1; then 1, 0, and 1; then 1, 1, and 0; and then 0, 1, and 1. After that we are caught in the cycle of 0, 1, and 1.

Similarly, given any set of five integers, this process does not necessarily lead to zeros. It can be proven that, given any set of n integers, the above process results in zeros if and only if n is a power of 2.

Divisibility by 9

It has been commonly observed that the sum of the digits of small numbers divisible by 9 (e.g., 18, 27, 36, 45, 54, 63, 72, 81, 108) equals 9. It is less commonly known that an integer like 46656 is divisible by 9 if and only if its sum of digits is divisible by 9, which is so in this case ($4 + 6 + 6 + 5 + 6 = 27$).

Take the sixth power of any natural number (positive integers such as 1, 2, 3, 4, . . .) and denote it by n^6. It can be shown that *either* n^6 is divisible by 9 *or* $n^6 - 1$ is divisible by 9. For instance, 6^6 (or 46656) is divisible by 9. However, 8^6 (or 262144) is not divisible by 9, but $8^6 - 1$ (or 262143) is divisible by 9.

Biotechnology at ORNL

When microorganisms are used to churn out a desired product or break down a harmful substance into innocuous chemicals, biotechnology is at work. This ancient technology was used as early as 3000 B.C. to produce beer. It has been employed for centuries to make bread, wine, and cheese. In recent years, biological systems have been harnessed to produce fuels and chemicals and to clean up the environment by removing pollutants from waste streams. Users of this technology often call it bioprocessing.

In the past decade, the ancient technology has undergone a revolution. Thanks to new genetic engineering techniques, scientists have transplanted foreign genes into bacteria, yeast, and other microorganisms, thus inducing them to produce large quantities of human insulin, interferon, growth hormone, and an artificial sweetener.

In addition, new hybrid-cell techniques have been used to produce monoclonal antibodies, which can be made by fusing endlessly growing cancer cells with antibody-producing cells from mice. New fused-cell populations are grown as clones from single hybrid cells. Because monoclonal antibodies that bind to specific tumor cells can be identified, they show promise for detecting cancer or delivering toxic agents to destroy specific cancer cells, much like a magic bullet. Monoclonal antibodies have been approved for diagnosing cancer. Other potential uses include diagnostic tests for some animal diseases and the frightening human disease of acquired immune deficiency syndrome (AIDS), as well as detoxification of individuals acutely exposed to toxins. Other developments in the new biotechnology have resulted in innovations in plant genetics, which are expected to add $5 billion per year to the value of major food and energy crops in the next decade.

Oak Ridge National Laboratory has been involved in developing and using these technologies over the years. Under the direction of Chuck Scott, researchers in the Chemical Technology Division have developed bioreactors for removing nitrates and phenols from industrial wastewaters, treating municipal wastewater, and removing toxic metals such as lead and cadmium from waste streams. (See article on page 19.) Their continuous annular chromatograph shows promise for commercially separating proteins and producing purified drugs and enzymes (constituents of living cells that catalyze chemical reactions).

In addition, Jonathan Woodward of the same division is working on ways to increase the efficiency of using enzymes to convert cellulose in wood and other plant materials to fermentable sugars, which could be a source of fuels and chemicals. He is conducting fundamental studies on the mechanism by which the enzyme cellulase is adsorbed onto insoluble cellulose. New knowledge in this area could lead to the development of a method that would allow the enzyme to be used repeatedly, thereby reducing considerably the costs of the cellulose-to-sugar process.

Another example of biotechnology employs organisms, such as algae, to produce the clean fuel hydrogen. Led by Eli

ORNL is a leading laboratory in bioprocessing and is becoming more involved in other areas of biotechnology. The work ranges from optimizing bioreactors to produce desired fuels and chemicals and remove pollutants from liquids to using protein engineering to find ways of improving food and energy crop yields.

Plantlets of willow (in flask) and sweetgum (in test tube at right) were cloned from parent plants under the technical guidance of ORNL. Cloning refers to methods of generating new plants that are identical to parent plants. The plants selected for cloning usually grow fast, even in poor soils, and are tolerant to diseases and pests. The willow was grown at the University of Minnesota, and the sweetgum was cloned using a tissue culture technique developed at the University of Georgia.

Greenbaum of the Chemical Technology Division, the ORNL scientists have pioneered studies in using light and algae (or nonliving systems) to photosynthetically split water into oxygen and hydrogen, which can also be used as a chemical feedstock.

A Chemical Technology Division group led by Carl Burtis has been developing innovative bioanalytical technology. One technique will detect body chemicals that signal a human physiological response to exposure to cancer-causing substances. Such carcinogens include polycyclic aromatic hydrocarbons, which are discharged in the production of synthetic fuels. When the synthetic fuels industry matures, this technique may be used to monitor workers and determine whether they are being exposed to potentially hazardous levels of carcinogens.

The Environmental Sciences Division, under the leadership of Jack Ranney and Janet Cushman, has been guiding subcontracted work on woody and herbaceous crop genetics. The genetic research focuses on testing and selecting species for producing biomass for energy. Crops that grow fast, are tolerant to pests and diseases, and adapt to a variety of soils and climatic conditions are being identified. Several institutions are developing new hybrids for testing as biomass crops.

Tissue culture is a biotechnology being used to propagate, or reproduce, selected species and hybrids for testing in the field. Plantlets produced using tissue culture are clones—that is, they are genetically identical to the parent plant. Tissue culture enables the production of large numbers of plants identified as superior; such plants can be grown in biomass plantations. Other research, just recently initiated, is using tissue culture technology to screen species for desirable characteristics. This innovative approach to species screening should enable researchers to identify superior species quickly.

In the Biology Division Howard Adler has found that membrane particles derived from bacteria that flourish in the human intestine can remove oxygen from solutions and thus hasten the growth of bacteria that live in oxygen-free environments; use of this technique can speed up diagnosis of diseases caused by such anaerobic bacteria. The technique is also expected to facilitate further study of anaerobic bacteria that produce fuels and chemicals.

Biochemists and molecular biologists led by Fred Hartman of the Biology Division have been involved in another facet of biotechnology called protein engineering, which involves changing the structure of the product of a gene, such as an enzyme or some other protein, by genetic engineering techniques. Their goal is to alter an important plant enzyme so that it no longer uses atmospheric oxygen to break down carbohydrates, which it simultaneously helps to synthesize from atmospheric carbon dioxide. If successful, the group could find a way of increasing the growth and yield of food and energy crops. (See article on page 22.)

Other work in the Biology Division uses approaches that could be applied to problems in biotechnology:

- A group of researchers led by Steve Kennel has studied the mechanisms by which monoclonal antibodies cause regression of cancerous tumors in mice. In a new project, the group is investigating the ability of monoclonal antibodies to detoxify animals treated with acute doses of a potent environmental toxin, dioxin.
- Frank Kenney and Kai-Lin Lee have used genetic engineering techniques to clone several genes from rats to identify the mechanisms by which the genes become activated (express themselves by producing proteins)

as the cells of the rat liver differentiate, or specialize, into a fully functional organ. The information gained may help explain why such genes are often not expressed in the cancerous liver.

• Wen Yang has used genetic engineering techniques to study the role of genes in causing cancer. Investigating the role played by mobile gene elements in the rearrangement of genes (a phenomenon that has been shown to be associated with cancer in mice and humans), Yang and his associates have cloned various classes of these elements (including those responsible for producing leukemia viruses) from chromosomal deoxyribonucleic acid (DNA) of the mouse. They then spliced and exchanged parts of the elements between two selected DNA clones and put the recombinants back into cells in culture by a technique called DNA transfection. Their goal is to determine which part or parts give mobile gene elements the ability to move, insert themselves among other genes, and cause rearrangement of genes at the site of insertion. Genes expected to be related to cancer can be tested by in vitro manipulation and by being "shuttled" back and forth between bacteria and mammalian cells. The important parts of the gene can then be analyzed by DNA sequencing techniques to decipher the exact genetic code information (base sequences) contained in it. In experiments on one strain of mouse in which myeloid leukemia was induced by irradiation, Yang and his colleagues detected that a particular set of mobile gene elements related to the leukemia virus had spread itself and established colonies in other locations of mouse chromosomes. This finding is one example of a gene rearrangement event.

• Dorothy Skinner is leading a study of the influence of unusual DNA sequences on the mutability of DNA. Her group's experiments deal primarily with very complex repeated DNAs that have unusual sequences at irregular intervals. Such segments are highly sensitive to some enzymes, indicating that the secondary structure of the DNA is perturbed at these sites. In the many copies of the repeated DNA, the ORNL researchers have found that major changes in primary structure occur at these sites. These sites are therefore "hot spots" for genetic mutations. Similar DNA sequences are found close to important human genes and are thought to regulate their functioning. Skinner's future research will include cloning genes active in growth and development.

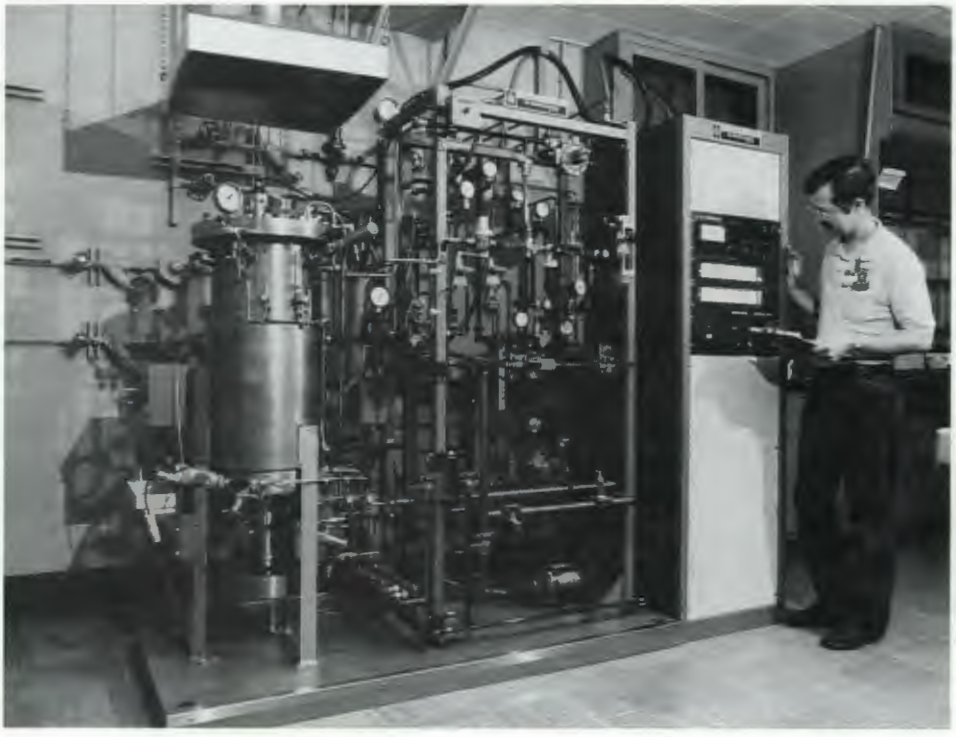
In the Analytical Chemistry Division, Leon Klatt has developed a remote analytical instrumentation concept, which could be useful for gene manipulation experiments done inside biological containments to prevent the escape of disease-causing viruses to the environment. In the Instrumentation and Controls Division, Jack Davidson has developed a television-based technique that is faster and more sensitive than X-ray film for imaging patterns of cellular protein production. The technique could be used to screen for clones of genetically engineered microorganisms.

ORNL is a leading laboratory for bioprocessing research and has made important contributions in other areas of biotechnology. This expertise has attracted national attention. For his novel approaches to the characterization of catalytic sites of enzymes (prerequisite information for designing rational experiments in enzyme engineering), Hartman was honored in 1979 by the American Chemical Society with the prestigious Charles Pfizer Award in Enzyme Chemistry; Adler and Weldon Crow received a 1984 I•R 100 award from *Research & Development* magazine for their work on the oxygen-consuming membranes; and other Biology Division researchers in biotechnology have received national honors. Scott has won three I•R 100 awards in the area of bioprocessing, and he received DOE's prestigious E. O. Lawrence Award in 1980, largely for his bioprocessing work. And DOE has approved the establishment of a national biotechnology user facility at ORNL called the Bioprocessing Research Facility User Resource; it is managed by Terry Donaldson, coordinator of the Laboratory's Biotechnology Program. It appears that ORNL is headed for the forefront of one of today's most exciting technologies.

Bioprocess Research and Development

By CHARLES D. SCOTT and TERRENCE L. DONALDSON

Microorganisms can carry out a wide variety of chemical transformations. They can combine carbon dioxide and water to produce a broad spectrum of carbohydrates, proteins, and lipids—the essence of life. They can also mediate the reverse process—degradation of organic chemicals to carbon dioxide and water. Recent advances in



Doug Lee is studying the anaerobic digestion characteristics of cellulosic materials using this 75-L digester. Anaerobic digestion is a promising technology for volume reduction of radioactive cellulosic and animal wastes. Results from this bioreactor will be used to define process operating parameters and to project performance for a full-scale digester. Bioreactors are an important part of ORNL's Bioprocessing Research Facility User Resource.

The Oak Ridge Bioprocessing Research Facility User Resource

Because of its work in developing bioreactors and various processes that use microorganisms to produce fuels and chemicals from biomass and remove pollutants from wastewaters, ORNL has become a center of expertise in bioprocessing research. As a result, in 1984 the U.S. Department of Energy established at ORNL the Oak Ridge Bioprocessing Research Facility User Resource. This user facility is open to scientists and engineers from universities, industry, and national laboratories.

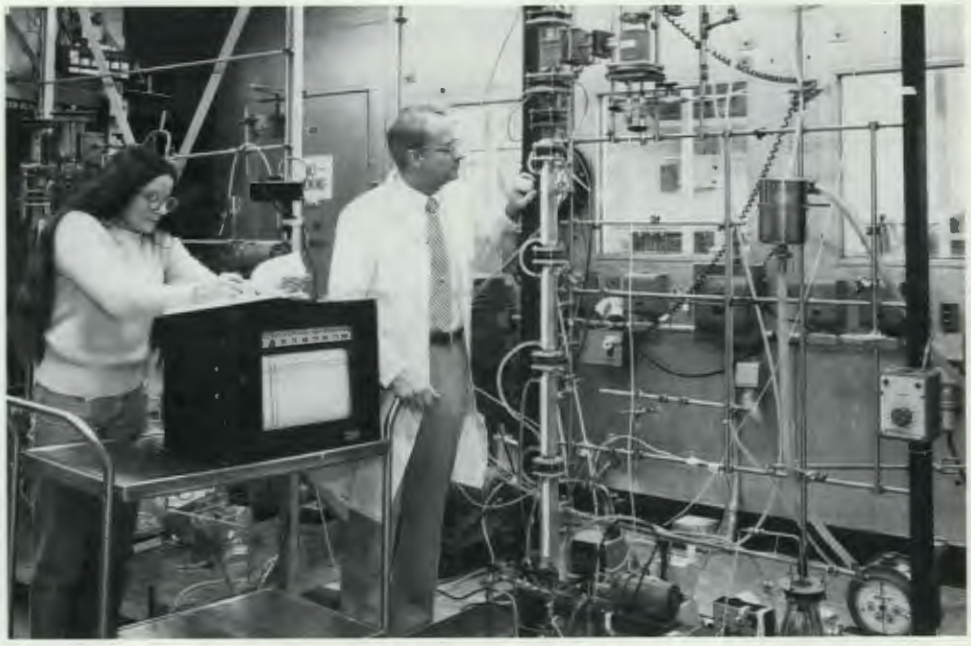
The Bioprocessing Research Facility User Resource includes laboratories for investigation of advanced bioprocessing concepts, employing stirred-tank and columnar bioreactors and a fermentation pilot plant for large-scale fermentation experiments. Using the facility's advanced systems and other state-of-the-art equipment, researchers can engage in

- pretreating and fractionating chemical feedstocks
- selecting and improving microbial cultures
- manipulating genes to produce desired proteins
- designing advanced bioreactors
- developing advanced analytical concepts
- determining the feasibility of using a bioprocess and scaling it up
- monitoring and controlling experimental bioprocesses
- conducting biochemical separations.

Researchers using the new facility will have access to a wide variety of analytical equipment, including gas chromatographs, spectrophotometers, carbon and nitrogen analyzers, centrifuges, environmentally controlled growth chambers, autoclaves, refrigerators, freeze dryers, and microscopes. Also available to researchers are ORNL laboratories equipped for mammalian tissue culture and molecular genetics, containment facilities for recombinant DNA studies, and a large facility for handling research animals. These resources are located primarily in the Chemical Technology Division at the X-10 site and in ORNL's Biology Division at the Y-12 Plant.

According to Terry Donaldson, coordinator of the Bioprocessing Research Facility User Resource, experiments that could be done at the new facility include production of gram quantities of a particular enzyme using microorganisms modified by recombinant DNA techniques and scale-up tests of a new method for immobilizing microorganisms to make biocatalyst particles.

Chuck Scott is a research fellow in ORNL's Chemical Technology Division. In 1983 he was named a Corporate Research Fellow by Union Carbide Corporation (ORNL's operating contractor at the time), and in 1980 he received the E. O. Lawrence Award from the U.S. Department of Energy for outstanding contributions to separation science and technology, bioanalytical instrumentation, and nuclear energy. During the 1970s he received four IR 100 awards for such developments as bioanalytical instrumentation (1971), a portable analytical system (1977), the continuous annular chromatograph (1978), and the tapered fluidized-bed bioreactor (1979). Scott is the organizer of two annual symposium series—one on "Automated Analyses in the Clinical Laboratory" and the other on "Biotechnology for Fuels and Chemicals." A native of Chaffee, Missouri, Scott obtained three degrees in chemical engineering from the University of Missouri and the University of Tennessee (UT), earning his Ph.D. degree from UT in 1966. He has worked at ORNL since 1957, serving in the Chemical Technology Division as a section chief of the Experimental Engineering Section (1974–76) and as associate director (1976–83). Since 1967 he has lectured on chemical engineering at UT in Knoxville. Here, Scott and Susan Arnold obtain high rates of ethanol production in a laboratory-scale fluidized-bed bioreactor using immobilized organisms.



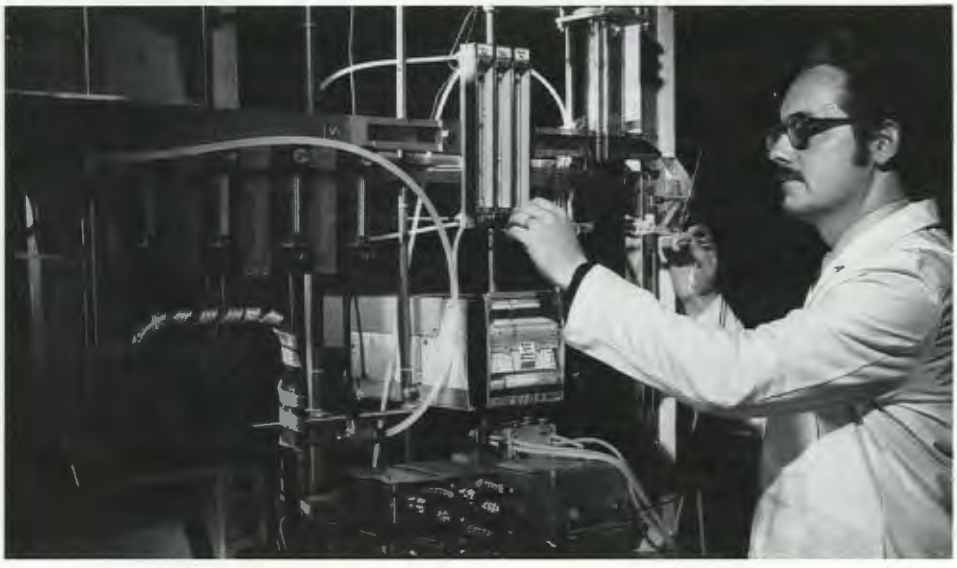
molecular biology have enabled scientists to alter the cell machinery of some microorganisms to create a new capability that they normally lack. For example, *Escherichia coli* bacteria (a common species found, for example, in the human intestine) can be altered by inserting a gene that causes each bacterium to produce a desired chemical, such as insulin for diabetics or human growth hormone for diminutive children. This new ability—and the potential it offers—is largely responsible for the current interest in biotechnology.

To do useful things with microorganisms, it is necessary to develop and operate a "process." When the process uses microorganisms or microbially derived components such as enzymes, it is often referred to as a bioprocess. Production of alcoholic beverages and biological treatment of wastewater (including residential septic tanks) are examples of classical, conventional bioprocesses. At Oak Ridge National Laboratory we are looking for new bioprocesses to make or break down certain chemicals and for ways to improve conventional bioprocesses to increase yields, efficiencies, flexibility, and stability, particularly for producing or using energy cleanly and efficiently.

Characteristics of Bioprocesses

Bioprocesses are different from other technologies that can produce similar chemical transformations:

- Bioprocesses are usually highly selective. Selectivity is an advantage if a high yield of a particular product, such as ethanol (a fuel), is desired; it can be a disadvantage if the goal is to consume a heterogeneous feedstock such as municipal solid waste.
- Because bioprocesses operate at mild temperature, pressure, and pH and are generally not corrosive, they can be energy-efficient. However, their reaction rates are usually slower than rates for nonbiological processes that operate under more severe conditions. Residence times and equipment sizes are larger, although costs may be lower because of the relative simplicity and energy efficiency of the process. Bioprocesses can be "scaled down" for relatively small



Terry Donaldson is coordinator of ORNL's Biotechnology Program and the new Oak Ridge Bioprocessing Research Facility User Resource. In this capacity he reports to ORNL's Associate Director for Biomedical and Environmental Sciences. He is also group leader for Bioprocess Development in the Chemical Technology Division. The group designs bioreactors and conducts research on developing processes for producing fuels and chemicals from biomass and for treating wastes. Before joining the ORNL staff in 1980, Donaldson was associate professor of chemical engineering at the University of Rochester in Rochester, New York. A native of Emlenton, Pennsylvania, he received his Ph.D. degree from the University of Pennsylvania. He teaches in the chemical engineering program at UT and advises graduate students in thesis research. Here, he studies the rate at which microbes in a fluidized-bed bioreactor degrade pollutants in wastewater from coal gasification.

applications. Usually little economy of scale can be gained in large installations.

- Bioprocesses usually require an aqueous environment. This requirement is an obvious advantage if water is already present; it is a problem if the feedstock or product is inherently nonaqueous (ethanol, for example) because of the expense of adding water and removing it later.

At ORNL we are working on bioprocesses for production of useful fuels and chemicals and for degradation of wastes. Our past accomplishments have included developing packed-bed bioreactor systems for anaerobic treatment of municipal wastewater (ANFLOW) and a bioprocess for removing nitrate from industrial wastewaters, and using biosorption to remove heavy or toxic metals, such as uranium, lead, and cadmium, from waste streams.

A new private company called ANFLOW, Inc., plans to commercialize the ANFLOW technology for treating municipal wastewater. The company is receiving support from the Tennessee Innovation Center and the Office of Technology Applications of Martin Marietta Energy Systems, Inc. It is planning a joint venture with the Tennessee Valley Authority to demonstrate the ANFLOW technology. The biodenitrification process developed at ORNL has been implemented at Goodyear Atomic Company's gaseous diffusion plant at Portsmouth, Ohio, and will be used in a system now being built at the Feed Materials Production Center in Fernald, Ohio.

Current Activities

Our current research and development work in the Chemical Technology Division ranges from investigating conceptual bioprocesses at the bench to operating small, pilot-scale facilities to demonstrate technical feasibility. We are developing a fixed-film, fluidized-bed bioreactor for degradation of organic chemicals in wastewater from coal gasification processes. This bioreactor contains thin films of microorganisms attached to the surface of <1-mm-diameter coal particles. The bioprocess shows potential for reducing capital and operating costs to perhaps 50% of the costs of a conventional biotreatment process such as activated sludge.

Radioactive wastes can also be treated biologically. At ORNL Doug Lee and Terry Donaldson have converted contaminated cellulosic materials, such as paper and cloth, to water-soluble forms by anaerobic digestion. The water-soluble chemicals are in turn converted biologically to methane and carbon dioxide, while the radioactive species remain with the relatively small volume of sludge. A mixed culture of organisms obtained from a municipal sewage sludge digester is used. ORNL has been operating a 75-L digester for over a year to obtain information on reaction rates and stable operating conditions. A design for bioprocessing the cellulosic wastes generated at the Oak Ridge plants of the U.S. Department of Energy has been prepared.

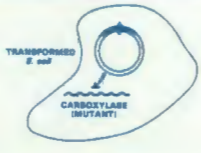
Chuck Scott, Brian Davison, and Jim Thompson are developing advanced bioreactors for efficient fermentation of sugar substrates to ethanol, a valuable chemical feedstock and gasoline extender–octane booster. These bioreactors use immobilized *Zymomonas mobilis* microorganisms to maintain high cell concentrations without washout. Rates of production of ethanol have been spectacular, exceeding rates in conventional batch fermentations by orders of magnitude. If these rates could be achieved in commercial-scale bioreactors, the effect on process economics would be substantial.

A key requirement for successful advanced bioprocesses is a highly active, stable biocatalyst. Scott has been developing a technique for immobilizing microorganisms and cell fractions such as enzymes. Immobilized catalysts tend to have favorable stability properties and can be reused easily. He has produced small gel beads from carrageenan and encapsulated microorganisms within the beads. (See *News Notes* item on p. 37 for more details.)

New work by Jerry Strandberg and Scott is also under way on microbial processes for liquefaction of coal, a relatively new application of bioprocessing. They are investigating which fuels and chemicals can be made from coal using microbes. The mild environmental conditions and the potential product selectivity in bioprocessing may lead to attractive bioprocesses for coal conversion that could be more economical and less hazardous to the environment than thermochemical liquefaction processes.

Since beer-making began 5000 years ago, bioprocessing has broadened to envelop many different techniques to degrade wastes and produce chemicals of choice. ORNL is playing an important role in developing improved bioprocesses to increase energy supplies and reduce environmental hazards.

Protein Engineering



By FRED C. HARTMAN

During the brief 13-year period since its inception, genetic engineering has revolutionized, glamorized, and commercialized biology. Some 200 companies are engaged in genetic engineering despite a paucity of products introduced into the marketplace. Research in these companies is supported primarily through venture capital and public stock offerings, so obviously the financial community anticipates future profits. Likewise, the scientific community anticipates dramatically increased use of genetic engineering to solve problems in medicine, agriculture, and industry.

A major component of genetic engineering is "recombinant DNA technology," techniques that enable scientists to manipulate genes in the laboratory like chemical reagents and to move genes from one organism to another. A gene is a segment of deoxyribonucleic acid

(DNA) with a specific sequence of bases that specifies the structure of a particular protein. In most cases, a bacterium is the recipient of the transferred gene, and usually the rationale for the transfer is to make large amounts of scarce proteins of industrial or therapeutic value. An example is insulin for humans.

Insulin, a protein hormone made in the pancreas, regulates the body's use of glucose. One form of diabetes, a life-threatening disease afflicting millions, results from the inability of the pancreas to synthesize enough insulin. In most cases, the ailment can be controlled by daily injections of insulin extracted from the pancreases of pigs or cows. Unfortunately, the supply of pancreases is limited, the purification of insulin is difficult, and the yields of insulin obtained are low. Furthermore, some individuals develop intolerances to animal insulins because they are somewhat different in structure from human insulin.

Because of advances in genetic engineering, human insulin—which causes fewer adverse reactions—has been available in the marketplace since September 1982. This commercial development was based on the independent work of W. J. Rutter and H. M. Goodman of the University of California at San Francisco and of Walter Gilbert and colleagues at Harvard University. They inserted the human gene for insulin into the genetic material of live bacteria and found that a genetically altered bacterial strain produces insulin. (Rutter is a member of the ORNL Advisory Board and past member of the Biology Division Advisory Committee.) Insulin-producing bacteria can be grown quickly, simply, inexpensively, and in unlimited amounts using a solution of basic nutrients, so the process of making human insulin was easily commercialized. Today the pharmaceutical firm Eli Lilly and Company produces ample human insulin for diabetics.

Many additional scarce proteins have been cloned by genetic engineering techniques. Some of these proteins may be used clinically to control high blood pressure, to dissolve blood clots in stroke victims, to prevent dwarfism, and to combat viruses. In fact, a protein named interferon has shown limited promise in treating some cancers and herpes virus infections.

Eventually it may be possible to transfer genes into higher organisms such as green plants and even humans. One obvious goal is to endow more agricultural plants with the ability to use atmospheric nitrogen directly to minimize the need for expensive nitrogen fertilizers. Another possible application is to correct hereditary disorders in humans caused by defects in single genes; a special virus could ferry copies of the needed "good" gene into the cells of victims. Such "gene therapy" could be used for victims of such genetic diseases as muscular dystrophy, cystic fibrosis, hemophilia, several types of arthritis, and sickle-cell anemia (discussed later in this article).

The feasibility of transferring genes into lower mammals has already been demonstrated. Richard Palmiter and his colleagues at the University of Washington in Seattle succeeded in microinjecting the rat gene for growth hormone into fertilized mouse eggs, some of which developed into mice twice the size of their untreated littermates.

Transfer from one organism to another is not the only way that genes can be manipulated in the laboratory. Genes also can be structurally altered to instruct cells to manufacture new proteins with different properties. Such work on

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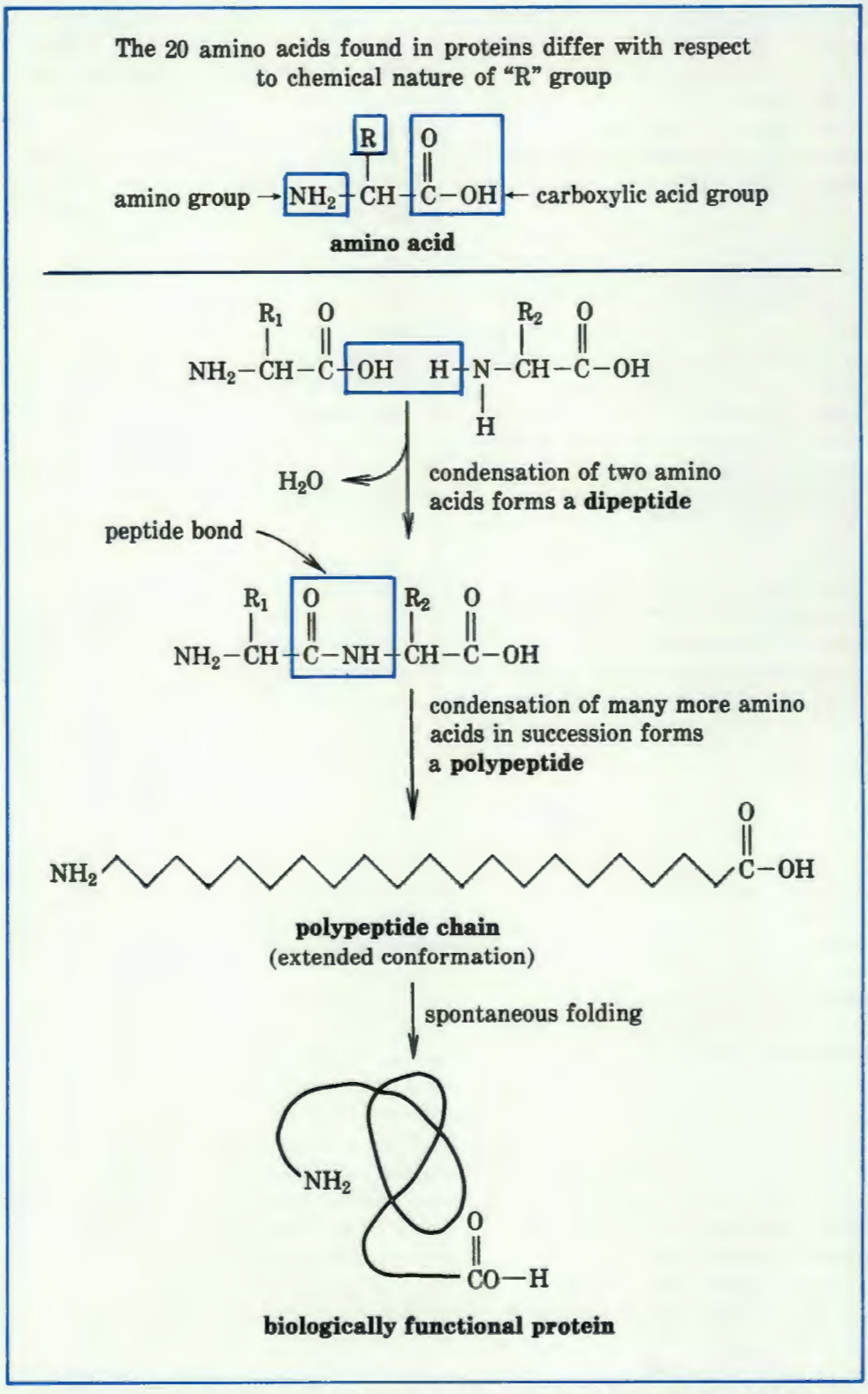
"protein engineering" is under way in ORNL's Biology Division. The program was initiated in April 1984 with partial support provided by the ORNL Director's R&D Fund.

Although the methods are reasonably well established, they involve many disciplines; hence success in this new field requires extensive collaborations. Currently, the ORNL team effort includes Bob Foote, an organic chemist who is able to synthesize pieces of genes; Frank Larimer, Richard Machanoff, and Richard Mural, molecular geneticists with expertise in cloning; Sankar Mitra and Salil Niyogi, biochemists who study mechanisms by which DNA is duplicated and translated to make proteins; Bob Fujimura, a biochemist proficient in determining base sequences of DNA; and Tom Soper and me, protein chemists who isolate and characterize the newly designed proteins.

A Protein Primer

Living organisms contain three major classes of polymers, which biochemists call "macromolecules": (1) polysaccharides, polymers of sugars, which store nutrients; (2) nucleic acids like DNA, polymers of nucleotides, which serve as informational molecules or "blueprints" for life processes including reproduction; and (3) proteins, polymers of amino acids, which are exceedingly diverse in function and participate in most life processes.

Proteins are required for sensory perceptions such as touch, vision, taste, smell, and hearing; for all voluntary body movement (muscle is virtually all protein); for involuntary acts such as breathing, the pumping of the heart to circulate blood, and maintaining



Representation of the structures of amino acids, peptides, polypeptides, and biologically functional "globular" proteins.

body temperature; and for bodily functions such as digestion and excretion of wastes. As transport agents, proteins carry materials from one part of the body to another. For example, the protein hemoglobin adsorbs oxygen from air inhaled into the lungs and delivers it to tissues. Hemoglobin then carries carbon dioxide (CO_2), a toxic byproduct of metabolism, from the tissues to the lungs, where it is exhaled. As antibodies, proteins fight disease and infection. As hormones, proteins regulate metabolism.

Not surprisingly, given their scope of function, proteins constitute 50% of the dry weight of cells. Among all types of proteins, enzymes represent the largest class.

Enzymes are nature's catalysts—that is, they increase the rates of all chemical reactions that occur within living cells. Indeed, very few biochemical reactions can proceed in the absence of enzymes under the gentle conditions of temperature and acidity (pH) required for cell viability. The lowly unicellular bacterium *Escherichia coli*, which inhabits the human intestine, contains about 3000 different proteins, 2000 of which are enzymes. Why so many? Generally one enzyme can catalyze only one (or one type) of chemical reaction. Thanks to enzymes, the myriad and complicated chemical reactions that are required to sustain life proceed efficiently and continuously; syntheses that the organic chemist could not duplicate in a lifetime, even without restrictions on reaction conditions, are accomplished within cells in a matter of seconds.

Despite the diversity of protein function, all proteins are linear polymers of amino acids. How can polymers so similar in structure be so different in function? Largely because these linear polymers are folded in very precise ways, and the

Stringer loads a sample of the cloned CO_2-fixation enzyme into a "spinning-cup" protein sequencer.

three-dimensional structure adopted by the protein within the cellular milieu then determines its biological function.

Visualize a metal chain made from many individual links. It may be pulled taut from end to end without breaking because of the interconnecting links. Despite this resistance to rupturing, the chain may be readily coiled, twisted, and folded into innumerable shapes. Similarly, the individual chemical bonds that connect the amino acids in linear array in a protein macromolecule are quite strong but do not prevent the protein from adopting a folded structure. In the chain analogy, each link represents one molecule of an amino acid.

Twenty chemically different amino acids are found in proteins; because the protein chain (referred to as a polypeptide) is 100–1000 amino acids long, each of the twenty will be repeated many times. A biologically functional protein may consist of a single polypeptide or multiple polypeptides held together through physical interactions. Every protein has its own unique sequence—that is, the linear arrangement of amino acids along the chain. This sequence determines the folding pattern, which in turn endows the protein with properties essential for its biological role. To illustrate the number of sequence isomers possible for even short polypeptides, consider the following hypothetical example: a polypeptide 61 amino acids long with 20 different possibilities at each position can give rise to 20^{61}, or 2.3×10^{79}, different structures, a number that exceeds the estimated number of atoms in the universe!

Protein Structure and Function

The chief aim of the protein biochemist is to correlate structure with function. Studies of protein structure can be subdivided into elucidation of amino acid sequence

and determination of three-dimensional structure. The former entails chemical degradation from one end of the macromolecule to the other, whereby one amino acid at a time is released and identified; the latter entails X-ray crystallography. Although both methods are well established, determination of protein structure is very laborious and time-consuming and may require many months or even years for a single structure. One traditional approach to correlating structure, once established, with function is somewhat akin to that of an inquisitive child learning how a watch works—take it apart and put it back together. This feat cannot be done literally with proteins, but scientists can chemically alter some of the amino acids in an intact protein and observe the biological consequences.

The other approach has been to compare the amino acid sequences of proteins that come from different organisms but carry out the same function. This approach rests on the premise that certain amino acids at given locations within the polypeptide are critical to biological function and have been conserved during evolution, while amino acids not essential to function have been replaced with other amino acids. In general, only a few key amino acids out of the hundred or more that form the enzyme molecule are absolutely critical to the binding of the reactants (which enzymologists call substrates) and to the catalytic process.

Thus the biochemist has relied on what nature has provided and on chemical alteration to correlate protein structure with function. However, the advent of genetic engineering during the past decade has provided the opportunity to change the amino acid sequence of proteins in predictable fashion. Any amino acid can be replaced by any other amino acid, and segments of polypeptide chains can be added or removed. Collectively, these manipulations of protein structure by genetic engineering constitute protein engineering. The biochemist can now make proteins that do not exist in nature. Beyond its obvious application to questions about mechanisms of protein function, protein engineering offers the potential to tailor properties of proteins for specific commercial uses.

A Look at DNA

The codes for sequences of proteins are embodied in DNA, the genetic material of an organism that determines all of the characteristic structural, biochemical, physiological, and morphological features of that organism. Indeed, only the information stored in DNA and transmitted during reproduction distinguishes a bacterium from a plant or animal or even a human being. DNA is a polymeric sugar phosphate that contains four different organic bases: (1) guanine, (2) adenine, (3) cytosine, and (4) thymine. DNA is a long thread-like molecule; if stretched end to end, it would exceed more than 1 m, a distance many

Frank Larimer and Richard Mural discuss the nucleotide sequence of a gene that codes for an altered enzyme.

thousand-fold greater than the diameter of a cell. DNA can fit into a cell only because it is tightly folded.

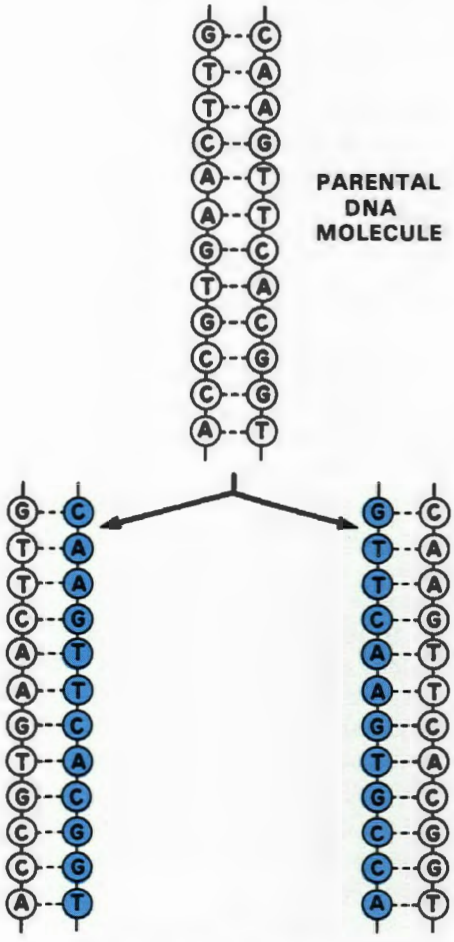
Cell division as well as reproduction of the entire organism requires precise duplication of the DNA molecule. The process is accomplished as a consequence of DNA's two polymeric strands. The strands are said to be complementary—that is, the sequence of bases on one strand of the double helix dictate one and only one sequence on the other strand.

Consider a double string of beads; the beads will be black, white, red, or green. Suppose that on one string the beads are strung randomly. However, on the opposite string, aligned in parallel to the first, a white bead will always be opposite a black one, and a green bead will always be opposite a red one. Provided that we remember this combination, the two strings of beads can be pulled apart and a double string, identical to the original one, readily reconstructed from either single string. As a consequence of physical interactions, guanine on one strand of DNA will always be opposite cytosine on the other, and adenine on one strand will be opposite thymine.

In this way, DNA is propagated from cell to cell or from parent to offspring with complete structural integrity and without change in informational content (excepting rare mutational events). To specify the amino acid sequence of a protein, it is necessary only to consider the linear sequence of bases on one strand of DNA. The sequence of three consecutive bases, referred to as a codon, prescribes a given amino acid. Hence, a strand of 1500 bases is required to direct the biosynthesis of a complete polypeptide containing 500 amino acids. A segment of DNA that corresponds to one polypeptide is called a gene. Based on the size of human DNA, as many as 100,000 different polypeptides (corresponding to an equal number of separate genes) could be synthesized within a cell.

DNA does not serve directly as a template for protein synthesis because it is located inside the cell's nucleus; the protein synthesizing machinery is outside the nucleus. Individual genes direct the synthesis of another polymer, ribonucleic acid (RNA), which

Sankar Mitra uses a liquid scintillation spectrometer to count the radioactivity of DNA labeled with tritium.



Replication (duplication) of DNA. During replication of DNA and cell division, both strands of the "parental" molecule are copied so that each of the two "first-generation" molecules is identical to the original. This fidelity in the replication process is primarily a consequence of the precise physical-chemical interactions of guanine (G) with cytosine (C) and adenine (A) with thymine (T) on opposite strands.

contains the same sequence information as the DNA template. This "messenger" RNA is shuttled out of the nucleus, after which it delivers the information originating with DNA to the protein synthesizing machinery. Before the function of messenger RNA was understood, its properties were first described in 1956 by Ken Volkin, a long-time senior staff member of the Biology Division, who recently retired. RNA polymerase, the enzyme that transcribes DNA to make messenger RNA, was codiscovered by Audrey Stevens, a senior staff member in the Biology Division.

To summarize, a linear sequence of bases in DNA, through an intermediary RNA, defines the protein's amino acid sequence. If one base in one gene is exchanged for another base, one amino acid will be exchanged for another amino acid in the protein expressed by that gene. Recent advances in gene cloning, DNA sequencing, and organic synthesis of short segments of genes have paved the way for generating the substitution of a desired amino acid for the original one at a predetermined location within the protein.

A striking example of how drastically the properties of a protein can change from only slight alterations in sequence is provided by sickle-cell hemoglobin. Sickle-cell anemia is a hereditary, usually fatal, disease prominent among black populations. All of the clinical symptoms, including anemia, chronic infections, renal failure, cardiac failure, and thrombosis, are a consequence of one amino acid substitution in the hemoglobin molecule. Like all hereditary diseases, the change reflects a mutation in DNA.

Hemoglobin consists of two pairs of nonidentical polypeptide chains, each about 150 amino acids long. The only chemical difference between normal hemoglobin and the abnormal sickle-cell hemoglobin is that the latter contains the amino acid valine instead of glutamic acid at position number 6 in one pair of polypeptide chains. This slight chemical change drastically decreases the solubility of hemoglobin within the red blood cell, leading to aggregation and precipitation. This precipitated hemoglobin is unable to transport oxygen efficiently, and hence the patient appears anemic; also, the precipitated hemoglobin distorts the shape of red blood cells (under a microscope they appear sickle-shaped instead of spherical) so that they tend to plug small blood vessels. This particular structural change illustrates the potential for changing properties of proteins through amino acid substitutions.

Bob Foote programs the synthesizer for making a particular gene segment.

ORNL's Plant Enzyme Work

Most of the ongoing efforts in protein engineering in the Biology Division are focused on an enzyme that is unique to photosynthetic

organisms. In contrast to animals, which derive energy from burning ingested food, plants trap light energy from the sun and convert it to chemical energy that is used to drive life-sustaining reactions—hence the term "photosynthesis" (synthesis from light, or photons). Many people do not fully appreciate the extent of their dependence on plants for oxygen, food, and energy. Plants produce the oxygen we breathe; before plants appeared on the earth, the atmosphere essentially lacked oxygen. Either directly or indirectly (through livestock) plants provide our nourishment. Natural gas, coal, and petroleum, which provide most of our energy for heat, light, and power, are decomposition products of plants that lived millions of years ago.

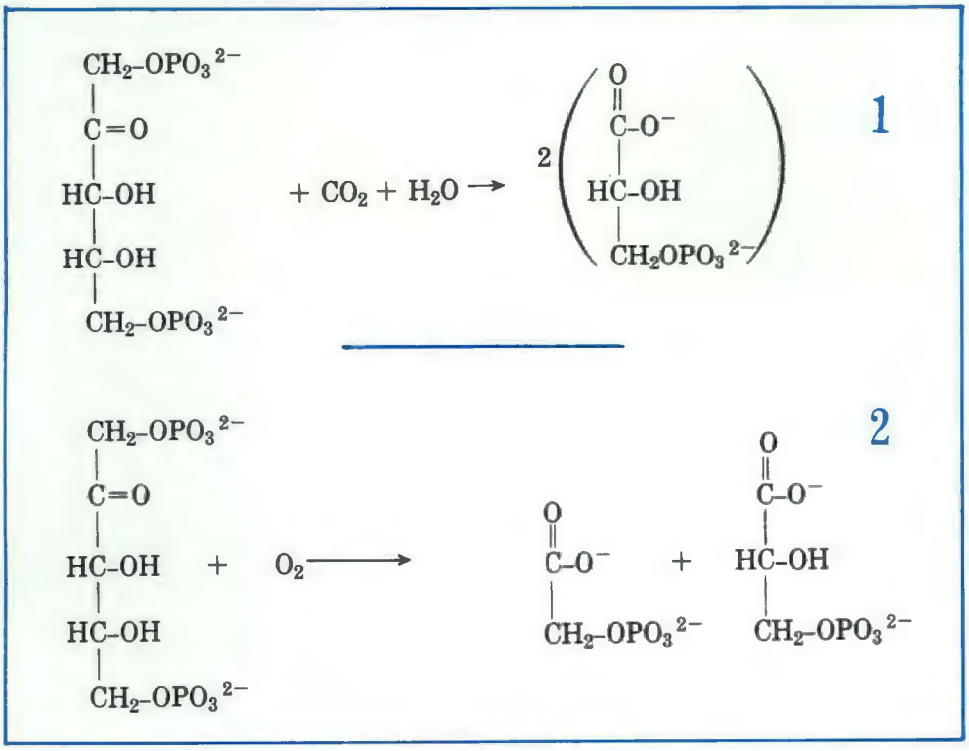
The overall chemical reaction of photosynthesis can be represented by:

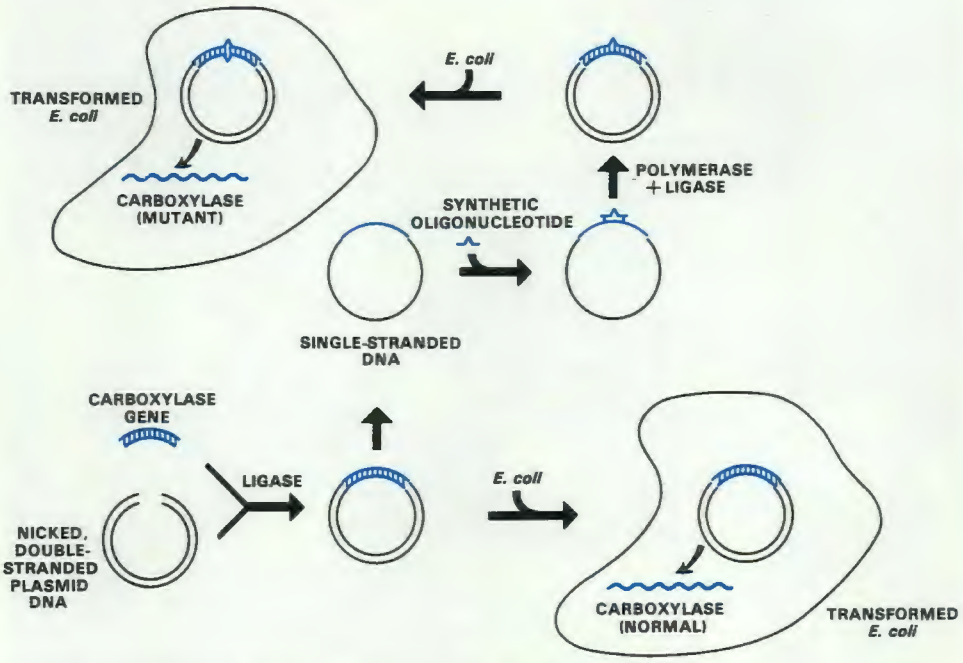
$$6CO_2 + 6H_2O \xrightarrow{light} \underset{glucose}{C_6H_{12}O_6} + 6O_2$$

Larimer analyzes recombinant DNA molecules by agarose gel electrophoresis.

whereby light energy is used to generate the basic nutrient glucose from atmospheric CO_2 and water. Glucose, a simple carbohydrate, is a precursor of complex carbohydrates (starch and cellulose), proteins, and nucleic acids, all essential constituents of living cells.

The biosynthesis of glucose from CO_2 (equation above) does not occur in one step but by a series of 15 highly coordinated reactions, each of which is catalyzed by a particular enzyme. This biosynthetic pathway was elucidated by Melvin Calvin, who subsequently was honored with a Nobel Prize. (Calvin has lectured in Oak Ridge on several occasions about the use of plant hydrocarbons as petroleum substitutes.) The initial step in this pathway is shown in the box (equation 1). In this oxidation-reduction reaction ribulosebisphosphate (a sugar phosphate) is oxidized with simultaneous reduction of atmospheric CO_2, which becomes "fixed" as 3-phosphoglycerate (another sugar phosphate). Although this reaction does not consume energy, energy is required





Transfer and chemical alteration of genes. The lower portion of the figure illustrates the insertion of the normal carboxylase gene into* E. coli *plasmid DNA, which is then placed into an* E. coli *cell to generate a transformed bacterial strain that synthesizes normal ribulosebisphosphate carboxylase. Mutants of the enzyme can be constructed by working with a single-stranded form of the plasmid DNA (center of figure). A short segment of the gene (synthetic oligonucleotide) that contains the desired mutation is synthesized and complexed with the corresponding region of the gene in the single-stranded DNA. Synthesis of the mutant gene is completed by use of appropriate enzymes (polymerase and ligase), and the new double-stranded plasmid DNA (upper right), when placed into* E. coli, *gives rise to a strain that synthesizes the "designed" mutant form of the CO_2-fixation enzyme.

by many of the ensuing 14 reactions that complete the synthesis of glucose and regenerate ribulosebisphosphate so that more CO_2 can be fixed. The energy is produced by the splitting of water to form oxygen (see equation in text on p. 29), which in nature occurs only in photosynthetic organisms.

The enzyme that catalyzes the photosynthetic reduction of atmospheric CO_2 (see equation 1 in box) is ribulosebisphosphate carboxylase, or simply the "CO_2-fixation enzyme." The importance of this enzyme cannot be exaggerated, because it brings about the only reaction on earth that produces carbohydrates from atmospheric CO_2. To put it bluntly, were it not for this plant enzyme, all life as we know it would cease. The rate of CO_2 fixation determines how rapidly plants grow and how much mass (dry weight) they accumulate.

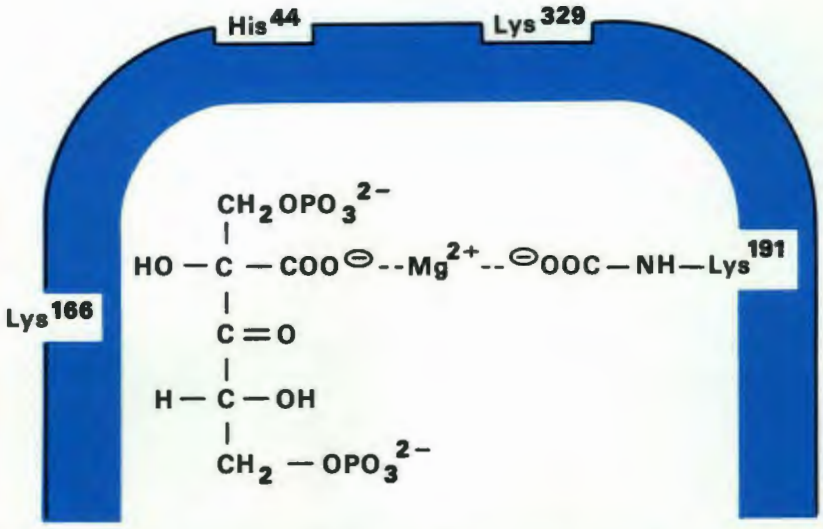
Around the turn of the century, the famous German biochemist Otto Warburg showed that many varieties of plants grow more rapidly in atmospheres with lowered levels of oxygen. This empirical observation was explained about 70 years later by N. E. Tolbert at Michigan State University (formerly a member of ORNL's Biology Division). Tolbert showed that the CO_2-fixation enzyme catalyzes a second reaction, whereby ribulosebisphosphate is oxidized by atmospheric oxygen (see equation 2 in box):

Hence, we have the uncommon situation of one enzyme catalyzing two different chemical reactions; furthermore, the two reactions operate at cross purposes. Whereas the CO_2-fixation reaction synthesizes carbohydrates, the reaction using oxygen (equation 2) lowers the concentration of ribulosebisphosphate in plant cells, thereby decreasing CO_2 fixation and inhibiting plant growth. The reason for this uncommon situation is not clear; some say it is due to an imperfection of evolution, and others believe that a not yet discovered purpose exists. What is clear is that the enzyme accomplishes a difficult task by synthesizing carbohydrates in an atmosphere that is only 0.03% carbon dioxide and 21% oxygen!

Detection of the CO_2-fixation enzyme in* E. coli *by immunoblotting. Streak cultures of* E. coli *strains were tested by treatment with antibody to the enzyme. The resulting antibody-enzyme complex can be visualized with a radioactive protein that recognizes and binds to the complex. Autoradiography (dark streak) reveals synthesis of the CO_2-fixation enzyme from a recombinant plasmid in cells that contain its respective gene, while streaks of bacteria that do not produce the enzyme are not visible.

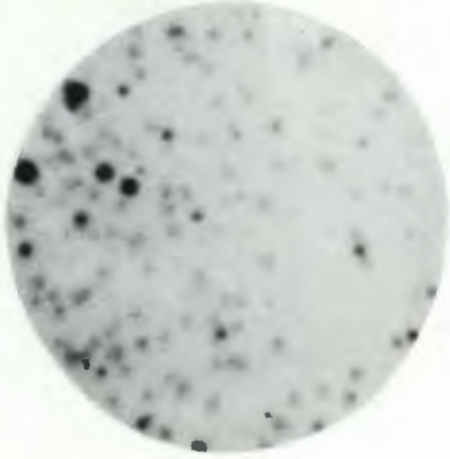
By introducing systematic structural changes into the CO_2-fixation enzyme through protein engineering, we hope to address some fundamental questions about how the enzyme works and also to ascertain the feasibility of diminishing the detrimental oxygenation activity or enhancing the desirable carboxylation activity. It is generally agreed that if the oxygenation activity could be abolished, the yield of many plants would double, with obvious beneficial impacts on food production and generation of biomass for energy.

A number of strategies are available for cloning genes and altering them to produce new gene products, but the key to all has been the discovery by Nobel laureates Hamilton Smith and Daniel Nathans at Johns Hopkins University of bacterial enzymes that act like molecular scissors to cut DNA into discrete pieces. In nature, these enzymes serve a protective role by degrading foreign DNA (hence, their designation as restriction enzymes). In the laboratory, they enable scientists to slice purified DNA into its component genes, which may then be transferred into the genome (set of chromosomes, which contain genes) of other organisms.



Schematic of the carboxylation reaction intermediate bound to the "active site" of the CO_2-fixation enzyme. Lysine at positions 166, 191, and 329 (Lys^{166}, Lys^{191}, and Lys^{329}) and histidine at position 44 (His^{44}) of the polypeptide chain are essential amino acids in close proximity to the bound substrate.

Screening for mutant CO_2-fixation enzyme genes in viruses (propagated in E. coli) by selective binding to a radioactive synthetic gene segment. The synthetic gene segment that contains the desired base change binds more tightly with the DNA of mutant viruses (dark dots) than with that of normal viruses (light dots). The viruses were fixed on a membrane.



Typically, small circular DNA from *E. coli* (called plasmid DNA), which contains just a few genes, is clipped at a single position by a restriction enzyme. By the use of other enzymes involved in DNA biosynthesis, this "linearized" DNA can be chemically combined with genes from other organisms (obtained by slicing their DNAs with restriction enzymes), converted back into circular DNA, and then reinserted into *E. coli*. This *E. coli* is referred to as a transformed strain because it contains a new gene in its DNA which will be propagated for all future cell generations. Once a gene has been "cloned" in this way, it can be isolated in pure form and its base sequence altered in vitro by a combination of chemical and biochemical manipulations. The "mutated" gene is spliced into the plasmid DNA, and the plasmid DNA is introduced into *E. coli*, resulting in the biosynthesis of a mutant protein.

Gene Transferred and Altered

We have transferred the gene for the CO_2-fixation enzyme from a photosynthetic bacterium into *E. coli* and are in the process of constructing a number of mutants. The transformed *E. coli* produces large amounts of the CO_2-fixation enzyme, which is chemically and biologically indistinguishable from that normally found in photosynthetic organisms. To simplify the synthesis of mutant forms of the gene, we transfer it into a circular viral DNA, whose two strands can be readily pulled apart. One of these strands then serves as a template for the in vitro synthesis of the altered genes.

Synthesis is initiated with a short segment of DNA, prepared in the laboratory, which contains the base change that will ultimately lead to the desired amino acid substitution. Excepting the single base change, the short synthetic fragment of DNA is perfectly

(continued on p. 33)

Biotechnology in the Soviet Union

The United States and several other countries have been on the forefront of the new biotechnology of enzyme engineering, but the Soviet Union is catching up. So said a Russian biochemist who addressed ORNL scientists at a Biology Division seminar in the fall of 1984. Enzyme engineering involves the use of genetic engineering techniques to change the structure of a specific product of a gene, such as an enzyme or other protein. According to Anatole A. Klyosov, professor and head of the Carbohydrate Research Laboratory at the A. N. Bach Institute of Biochemistry in the USSR, the Soviet Union got a late start in enzyme engineering, but Russian scientists have made up for lost time. One enzyme engineering process is being employed on an industrial scale in the USSR to make a product used for producing synthetic drugs to fight bacterial diseases.

Klyosov, an eminent biochemist and an expert on enzyme engineering, visited the Laboratory in September 1984 as part of his tour of the United States to exchange information on research efforts to convert cellulose to glucose sugar. He also visited the Massachusetts Institute of Technology and the University of California at Berkeley.

In 1981, according to Klyosov, top Soviet officials—the Presidium of the National Academy of Sciences, the Council of Ministers, and the Politburo—accorded top priority to biotechnology as a subject for research and development. At that time, however, industrial-scale production of fodder yeast had been under way for several years, reaching a billion-dollar-profit level from 1975 to 1980. Fodder yeast is grown on substrates, such as paraffins or methanol, to provide large quantities of microbial protein for animal feed. Because sugars are not used to feed the yeast, this method produces protein with a high nutritive value at low cost. In 1981 alone, production of fodder yeast through improved biotechnology totalled well over a third of the 1975–80 production. Klyosov predicts a continued escalation of fodder yeast production because of its importance in agriculture. "Producing fodder yeast, of course, is not enzyme engineering but it is biotechnology, whose goals are not that different from those of enzyme engineering," said Klyosov.

Since the 1981 decree, two agencies have been established to coordinate biotechnology in the USSR: the Interministerial Biotechnology Council and the Council on Biotechnology at the USSR Academy of Sciences. These bodies try to achieve cooperation among the ministries working on biotechnology within the Soviet Union as well as internationally.

So far the only enzyme engineering process that yields a product on an industrial scale in the USSR is the one that synthesizes 6-APA, which can be used as a building block to create an almost infinite variety of synthetic antibiotics that are superior to natural penicillin. Using natural penicillin to combat bacterial infections is undesirable for several reasons: it triggers allergic reactions, it has an undesirable administration route, and it is effective for only a limited spectrum of bacteria. Hence, the production of 6-APA to facilitate the manufacture of improved synthetic antibiotics is an important contribution to the USSR pharmaceutical industry.

The Soviet Union is also interested in producing the sugar fructose from glucose. The problem has been finding an inexpensive way to obtain enough glucose to start production. Because sugar production in the USSR has already reached maximum capacity and because importing sugar for industrial use is impractical, Soviet scientists are looking for other sources of sugar.

Biotechnology could offer a solution if an economical way can be found to convert cellulose (such as wood from forests) into glucose. Although many enzymes occur in nature to catalyze this conversion, none have been found to be suitable for industrial production because of narrow temperature and substrate concentration sensitivities. Enzyme engineering offers the possibility of altering a gene to produce an enzyme specially tailored for large-scale production of glucose from cellulose. Such an achievement would be sweet news for the Russians.—*Lydia Corrill.*

complementary to the corresponding region of the single-stranded template DNA, and hence a double-stranded complex is formed. Appropriate enzymes are used to complete the formation of circular, double-stranded DNA. Finally, the mutant gene is clipped out with restriction enzymes and shuttled, via plasmid DNA, back into *E. coli*. This transformed strain then produces the mutant CO_2-fixation enzyme.

To make meaningful choices concerning which amino acid substitutions to incorporate, a great deal of structural information about the enzyme must be available. Previous work in my laboratory has provided the complete amino acid sequence of the CO_2-fixation enzyme, whose two identical polypeptide chains are 475 amino acids long, and has pinpointed several amino acids that appear to be crucial either for binding of the substrate or for the catalytic process itself. We are evaluating the precise roles of these amino acids by systematically replacing them with other amino acids and examining the biological consequences. The figure on page 31 gives a schematic representation of that portion of the enzyme molecule where substrates bind and catalysis ensues, referred to as the "active site."

One of the crucial amino acids is lysine at position 166. Lysine is a positively charged amino acid that could be involved in the binding of negatively charged substrates; alternatively, as a general base, it could function catalytically. To distinguish whether this amino acid is involved in catalysis or binding, we have replaced lysine-166 with arginine, another positively charged amino acid that differs from lysine-166 in that it cannot function in general-base catalysis. Because the mutant protein lacks enzyme activity, we have concluded that lysine-166 functions catalytically and is not involved in substrate binding.

Gel electrophoresis is used by Mural to determine the nucleotide sequence of a mutant gene, an important step in the analysis of "engineered" proteins.

If the CO_2-fixation enzyme can be improved by alteration of gene structure, the ultimate objective would be to insert the altered gene into the chloroplasts (the subcellular compartment where CO_2 fixation occurs) of plants, thereby endowing them with the genetic information for synthesizing the superior enzyme. Presumably, these genetically altered plants would fix CO_2 more efficiently and consequently grow more rapidly. A gene transfer system for inserting recombinant genes into chloroplasts is not yet available, but several laboratories are attempting to devise one.

Protein engineering has already emerged as a powerful tool for probing many mechanistic questions about protein function that cannot be addressed by conventional techniques. However, much work remains before a critical assessment can be made concerning the potential of protein engineering in optimizing the properties of a protein for a particular application such as increasing the yield of crops for food and energy.

Strategies for Biotechnology: Taking Advantage of the Opportunity

The scientific and commercial potential of biotechnology has led to a variety of strategies for developing the potential in acceptable ways. In January 1984 the Office of Technology Assessment of the U.S. Congress published a report entitled "Commercial Biotechnology: An International Analysis," and in April 1984 the White House Office of Science and Technology Policy published a report issued by the Interagency Working Group on Biotechnology. Senator Albert Gore (D-Tenn.) has proposed legislation governing commercialization of biotechnology, and several federal agencies are competing to regulate the new biotechnology industry. Major studies have been completed by the National Academy of Sciences and the National Academy of Engineering to identify problems and opportunities in biotechnology. All of these studies conclude that a shortage of funding for research and development (R&D) and a shortage of qualified personnel threaten the continued healthy development of biotechnology in the United States and the ability of American research organizations to keep pace with the highly competitive, fast-moving groups abroad.

The National Science Foundation (NSF) is emphasizing research programs in biotechnology and encouraging participation of universities in the field. NSF has awarded funds to the Massachusetts Institute of Technology for a new Engineering Research Center in biotechnology and is supporting several other academic biotechnology centers. In addition, many major universities have established biotechnology centers to promote interdepartmental and interdisciplinary research programs; some of these centers have significant financial support from industry (particularly from companies that are members of the biotechnology centers).

At ORNL we are working to strengthen ties with the University of Tennessee to promote interdisciplinary collaboration in biotechnology. In 1984 an ORNL-UT Life Sciences Committee was established to identify and plan research programs in areas of common interest to both institutions. This committee involves primarily the Biology Division and the Environmental Sciences Division at ORNL and the Biology Consortium (Departments of Biochemistry, Botany, Ecology, Microbiology, and Zoology) at UT. Under the auspices of the Science Alliance funded by Tennessee's Better Schools Program, a new graduate program in biotechnology started this fall. ORNL staff members will participate in the instructional program at UT, and a major laboratory course will be taught at ORNL's Biology Division. The Distinguished Scientist Program, which provides for joint appointments at UT and ORNL, is a new mechanism for attracting exceptional scientists to the ORNL-UT scientific community; an appointee with expertise in biotechnology is being sought. In addition, a joint initiative for bioprocessing R&D is being pursued by ORNL's Chemical Technology Division and UT's Department of Chemical Engineering. Oak Ridge Associated Universities (ORAU) is interested in collaborative research between ORNL staff members and ORAU member universities using ORNL's Bioprocessing Research Facility User Resource.

Multidisciplinary collaboration has been and will continue to be a major ingredient in biotechnology. ORNL has a history of successful collaboration involving many disciplines and university researchers. The importance of this collaboration is recognized, and it will be a basic element in our strategies to strengthen ORNL's role in biotechnology. ornl

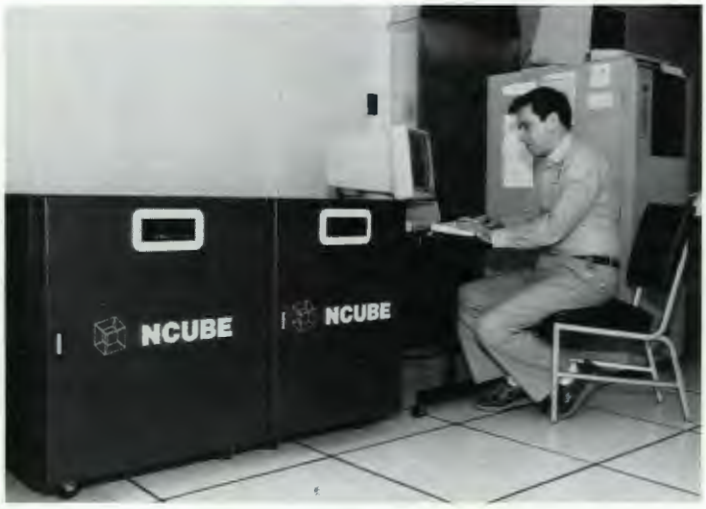
news notes

New-generation computers at ORNL

This summer ORNL acquired two powerful "hypercube" computers, designed to harness many microprocessors for cooperative problem-solving. The computers will be used for research in a variety of areas, including artificial intelligence, solid-state physics, robotics, fracture analysis, polymer studies, and environmental contamination transport. Both computers employ 64 microprocessors, linked together as the corners of cubes (hence the name "hypercube"). One of the machines is built by Intel, a leading maker of microprocessors. The other is built by a new company called NCUBE.

ORNL's Engineering Physics and Mathematics Division is in charge of both computers. The Intel machine, operated by the division's Mathematical Sciences Section, will be available to researchers throughout the Laboratory. According to Bob Ward, head of the section, the computer's capabilities will be tested first on six research projects in a variety of disciplines. The NCUBE will be used by ORNL's Center for Engineering Systems Advanced Research (CESAR) to develop and test an intelligent machine operating system, a general framework for implementing machine intelligence. CESAR will also apply the computer in a project on bionic "vision."

The key to both machines is their use of concurrent processors, loosely linked microchips that team up to solve different parts of a complex problem or calculation. Concurrent, or parallel, processing offers great promise both for complex scientific calculations and for robots and other intelligent machines, which must rapidly sense numerous images and conditions and continually modify their actions in response to what they sense. Concurrent processing may also prove important in complex military calculations, such as those likely to be involved in "Star Wars" defenses against nuclear missile attack. Both CESAR and the Mathematical Sciences Section have already received support from the Air Force for developing algorithms and operating systems for multiprocessors.



Jacob Barhen and new 64-processor supercomputer, one of two new "hypercube" computers at ORNL.

Parallel processing computers may well represent the next generation in computing, says Ward. The Intel and NCUBE machines use a new architecture based on a 1984 breakthrough at California Institute of Technology. There, two professors and five graduate students built the first successful concurrent computing ensemble, the "Cosmic Cube," for the remarkable sum of only $80,000.

Another important advance on which the hypercube computers depend is very large scale integrated (VLSI) circuitry. According to Jacob Barhen, whose Machine Intelligence and Advanced Computer Systems Group is in charge of the NCUBE machine, VLSI circuitry makes microprocessors both smaller and cheaper. Therefore, the NCUBE design is compact (smaller than an ordinary office desk), powerful (when expanded later to 1024 nodes, the roughly $2-million machine is designed to have the processing power of a dozen or more Cray supercomputers), and inexpensive (less than 5% of the cost of comparable Cray power).

Concurrent processing on a hypercube is also very efficient. That efficiency has a price, however: rather than being fed into the computer as a single job, a task must be separated into subtasks that are performed by individual nodes in a particular order. This and other points were discussed at a national conference on hypercube multiprocessors sponsored by ORNL and held in Knoxville in August.

Parallel processing moves computers a step closer to the brain, say Barhen and Ward. The machines still have a long way to go, however, before they begin to approach the power of that organ. Its concurrent processors—living neurons—number some 10 billion.

Uranium solidification program reaches milestone

A $29-million ORNL program to solidify more than 1000 kg of uranium from a liquid nitrate solution passed the 25% mark in operations this summer and is expected to be completed in early 1986.

The uranium solution, which measures 8000 L (about 1900 gal), has been stored at ORNL since the late 1960s, when it was transferred from a nuclear-fuel reprocessing plant in West Valley, New York. The plant, now closed, had experimentally reprocessed a nuclear fuel core from a Consolidated Edison reactor. Phil McGinnis, manager of the solidification program, says that two factors, the absence of a breeder reactor program and the presence in the solution of a highly radioactive uranium isotope, made reuse of the material impractical. ORNL

Technician adjusts mechanism for remote movement of uranium-oxide canisters.

was chosen to store—and, eventually, to dispose of—the uranium because of its expertise in handling highly radioactive materials.

Design efforts for the program, called the Consolidated Edison Uranium Solidification Project (CEUSP), began eight years ago. Designing, fabricating, and installing the complex process equipment required six years. A series of more than 200 "cold" tests of the process (using synthetic solutions of depleted uranium) began in 1984, and the solidification of the highly radioactive uranium solution began this April.

The remotely operated process uses evaporation to concentrate 20-L batches of the solution (down to a volume of 8 L); a high-temperature denitration step then converts the concentrate to a dry, inert oxide form. The oxide is double-sealed within cylindrical stainless-steel cans, which are about 9 cm (3.5 in.) in diameter and 61 cm (24 in.) high. A total of 380 cans will be packed, at the rate of 8 cans per week.

After being sealed, the cans are stacked by remote handling equipment inside monitored wells. The wells are within a heavily shielded cell adjacent to the processing cell.

According to McGinnis, the project has been a challenge because it involves totally remote operation and massive shielding (for protection from the highly energetic radiation), precautions against accidental criticality of the large amount of fissile uranium, highly corrosive conditions, and intricate processing and handling equipment. The processing plant, which contains a maze of piping and 1500 piping connections, was engineered to fit within existing "hot cells" measuring just 7 m square by 8 m high.

Other complicating factors include extensive reviews and analyses, personnel training requirements, and other efforts needed to ensure the safe operation of the facility.

McGinnis says that some engineers contend that the program is as complex as a "moon shot."

New nuclear-medicine agents studied in patients

Two new ORNL methods for detecting heart disease have been tried successfully in heart patients in Europe.

One is a radiolabeled form of a modified fatty acid similar to that which fuels the heart muscle; it indicates how different forms of heart disease affect the heart's metabolism of fuel. Studies of the agent's use in 19 patients have been completed at the Medical University Clinic in Vienna. Clinical trials in the United States are expected to begin soon.

The other ORNL development is a safer and more efficient system for generating a very short-lived isotope of iridium. The iridium is "milked" from a parent isotope, or "cow," of osmium (which is produced at ORNL's High Flux Isotope Reactor). The iridium is injected into the bloodstream, where gamma-camera pictures of its distribution can reveal the existence of defects such as intracardiac shunts (holes between heart chambers).

Because of its extremely short half-life (less than 5 s), the iridium agent is safe for repeated studies and for use in children. The system is now in routine use at a university clinic in Liege, Belgium, where more than 50 adult patient studies have been completed. U.S. clinical trials are expected to follow toxicity tests, which are scheduled to start this fall.

Up to 500,000 adult heart studies could be performed with the iridium-generator system each year. The system also shows promise for studies of blood flow through the brain and other organs.

technology transfer briefs:

Five more I-R 100s for ORNL

In September ORNL received five I-R 100 awards, bringing the Laboratory's total to 46 for the contest's 22-year history—the most of any DOE laboratory. The awards are given each year by *Research & Development* magazine for the year's top 100 developments having commercial potential. Since 1980, ORNL has averaged five awards per year.

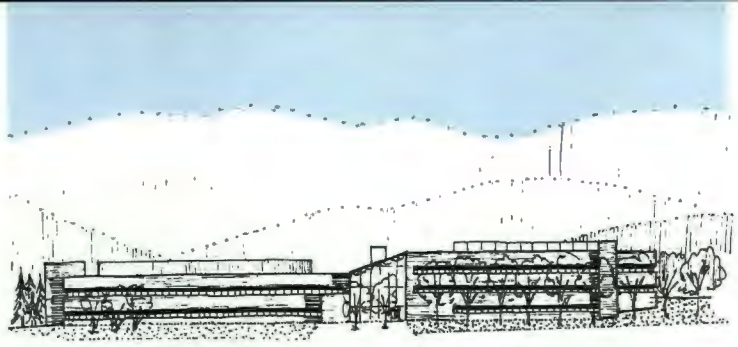
The Laboratory's latest winners:

- Metal oxide varistor
- Helium ionization detector for gas chromatography
- Radioactive waste disposal process
- Image detector for electrophoresis and chromatography
- Biaxial high-temperature fatigue extensometer

More details in the next issue of the *Review*.

ORNL inventors cash in

In June, 28 ORNL inventors, along with another 20 from other DOE facilities operated by Energy Systems, received patent awards ranging from $100 to $750. The inventors, who were recognized for patent applications from May through December of 1984, were the first beneficiaries of the company's new incentive program for inventors.



Artist's conception of multiprogram support building, part of proposed Life Sciences Complex.

The 48 inventors received a total of over $15,000; the average award was slightly more than $300. The individual award amounts were determined by a 56-member Technology Evaluation Committee comprising experts from numerous technical fields.

Technology-transfer funding for five projects

Nearly $200,000 in additional technology-transfer funding has been granted to five ORNL projects. The funding, which comes from the Energy Systems Office of Technology Applications, will help speed commercial adoption of the products and processes.

The five projects:

Fabrication of ceramic composites by chemical vapor infiltration (see the previous issue of the ORNL *Review*). Tough composites are formed by depositing a ceramic matrix around a fibrous preform. Numerous companies have requested samples of these composites for testing and evaluation. Currently, sample production is limited by the amount of time required to produce each sample (20 h) and the availability of only a single furnace. Funds will be used to modify another furnace for sample production, to purchase higher-strength, three-dimensional woven preforms for infiltration, and to buy better flow meters and controllers needed for further development of ceramic composite coatings.

Biocatalyst beads for advanced fermentation systems. Bioreactors such as fermentation systems use microorganisms as catalysts in chemical reactions. One key to more efficient bioreactors is to fix high concentrations of microorganisms into or onto a solid material, so they cannot be washed out of the reactor even at high flow rates. A particularly attractive approach for fermentation systems is to encapsulate densely packed microorganisms into a gel matrix in the form of beads. Such biocatalyst beads may be 10 times as effective as the conventional technique of suspending the micro-organisms within a culture medium in the bioreactor. Technology-transfer funding will allow production of a series of biobead samples that can be supplied to interested companies for evaluation.

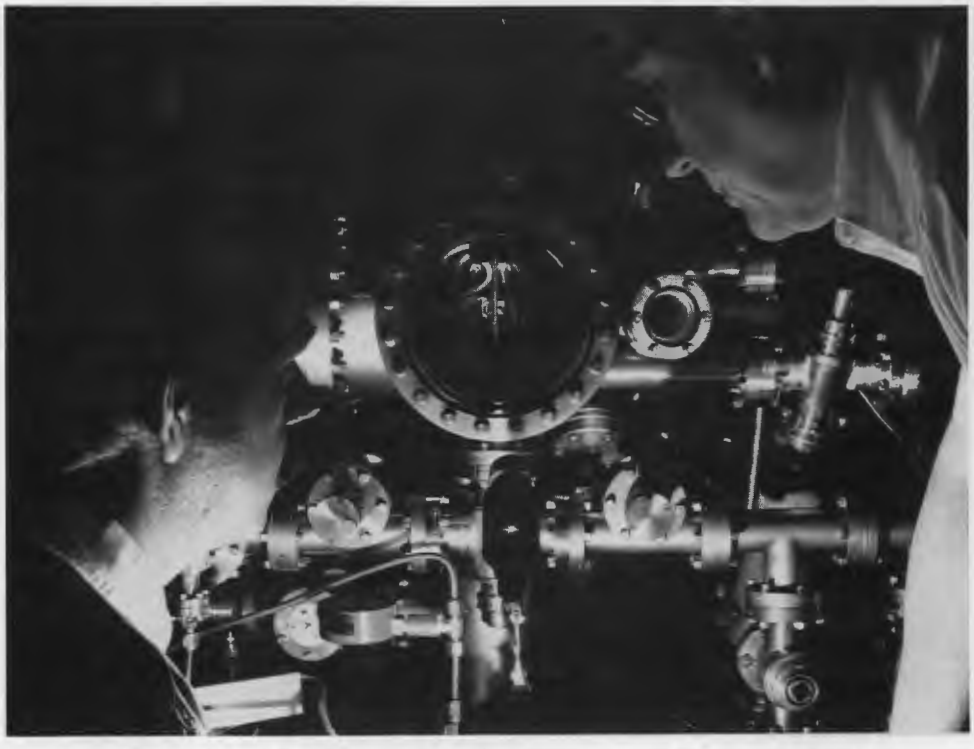
Whole-blood processing rotor. An ORNL-developed analytical device is capable of accepting a specimen of whole blood and automatically processing it into separate microliter samples of serum, which can then be individually analyzed. The device is expected to be useful for physicians' offices, hospital emergency rooms and special care units, and even bedside monitoring. The technology transfer will allow fabrication and evaluation of a modified prototype of the device.

Nickel aluminide alloy. Nickel aluminides are intermetallic alloys that possess great strength and toughness at high temperatures. They have applications in turbine and diesel engines and other high-temperature, energy-related applications. Numerous companies, including Cummins Engine Company, Combustion Engineering, Homogeneous Metals, Special Metals Company, and Ladish, have expressed strong interest in nickel aluminides. The technology-transfer funding would allow procurement and testing of additional billets of the material.

Pulsed helium ionization detection. Two ORNL researchers have developed a helium ionization detector, a highly sensitive instrument for use in gas chromatography. The instrument, which can detect gas concentrations of only a few parts per billion, has applications in industries such as semiconductor manufacturing, where the purity and concentration levels of various gases must be carefully monitored. The additional funding will allow improvements in the state-of-the-art circuitry, a complete characterization of the system, contacts with potential users, and publication of articles describing the instrument.

Construction of ORNL's High Temperature Materials Laboratory (HTML) was "inaugurated" at a May 17 ceremony. ORNL Associate Director Alex Zucker is shown here at the HTML test bent, a prototype section of the building's concrete post-and-beam structure.

Bob Clausing and others at ORNL have conducted studies to determine which cleaning techniques work best to remove the most obnoxious impurities from fusion vessel walls to prevent plasma energy losses. His experiences with fusion devices abroad suggest that adding methane to glow discharge plasmas can lead to fuel plasmas that are relatively free of energy-robbing impurities.



Lee Heatherly (left) and Bob Clausing examine a metal sample of wall material in a surface analysis station at ORNL.

A Question of Impurities:

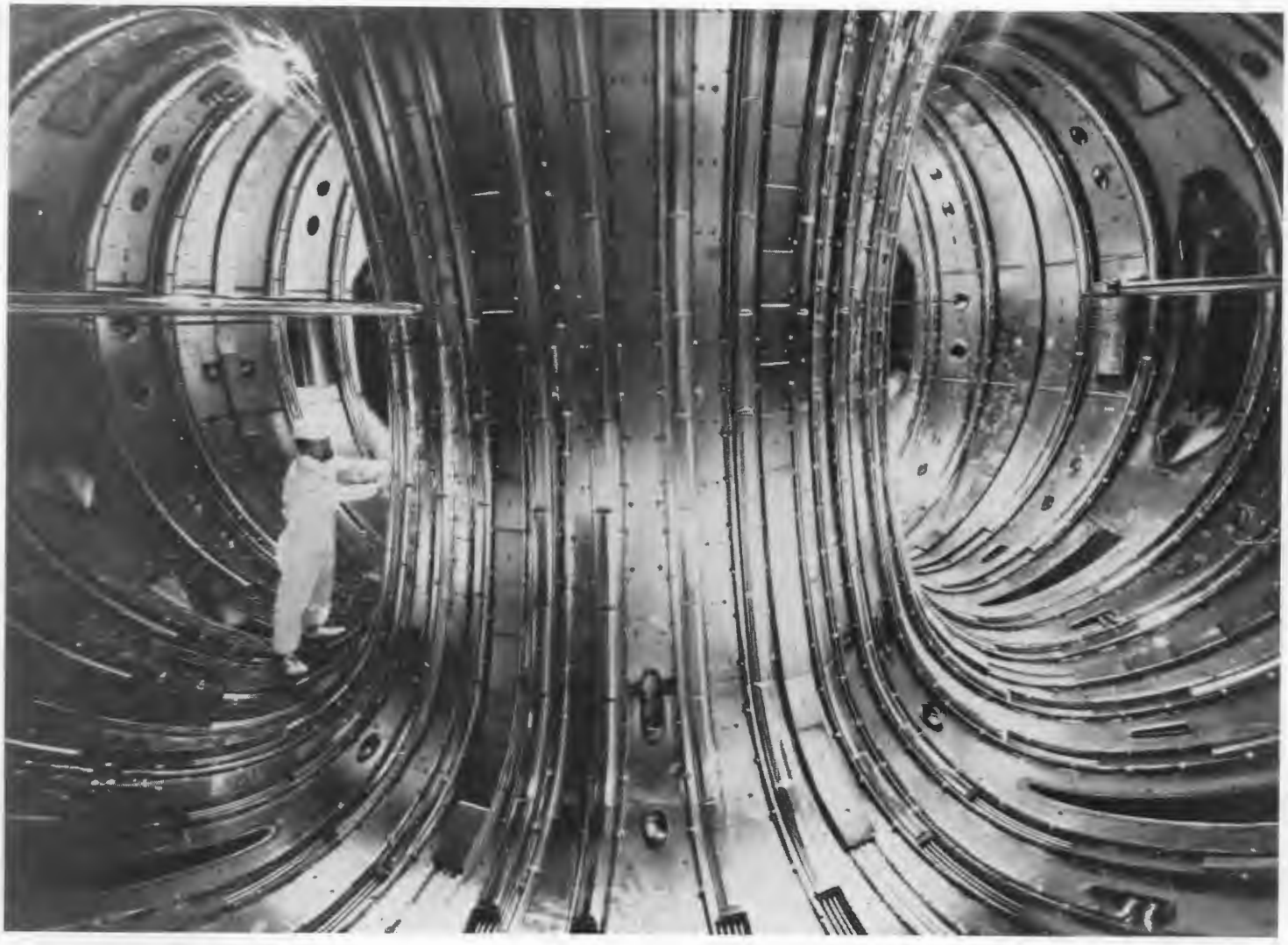
ORNL Examines a Persistent Fusion Problem

By CAROLYN KRAUSE

For fusion researchers experimenting with tokamaks, cleanliness is next to godliness. Inside these doughnut shaped devices, the curved wall nearest the fusion plasma must be as free of impurities as possible; otherwise, the impurities eventually infiltrate the plasma, robbing it of its fuel and energy. Since the early 1970s researchers in Japan, the Federal Republic of Germany (FRG), the Democratic Republic of Germany, France, Great Britain, the Soviet Union, and the United States have been developing and testing various cleaning techniques to remove impurities from tokamak "first walls." Although many cleaning formulas exist, so far no one has found the perfect Mr. Clean.

Some cleaning techniques work better than others in removing certain impurities, and some impurities are less damaging to the plasma than others. So researchers use the cleaning techniques that remove the most obnoxious impurities.

One group that has been active internationally in monitoring the progress of wall cleanup and determining which cleaning techniques remove the worst impurities is led by Bob Clausing in the Metals and Ceramics Division of Oak Ridge National Laboratory. Clausing, who has advanced degrees in metallurgical engineering and materials science, has worked with ORNL's Fusion Energy Division off and on since 1958. Clausing, Lee Heatherly, "Doc" Emerson, and Scott Halstead have traveled to tokamaks in the Soviet Union, Western Europe, and the United States with a surface analysis



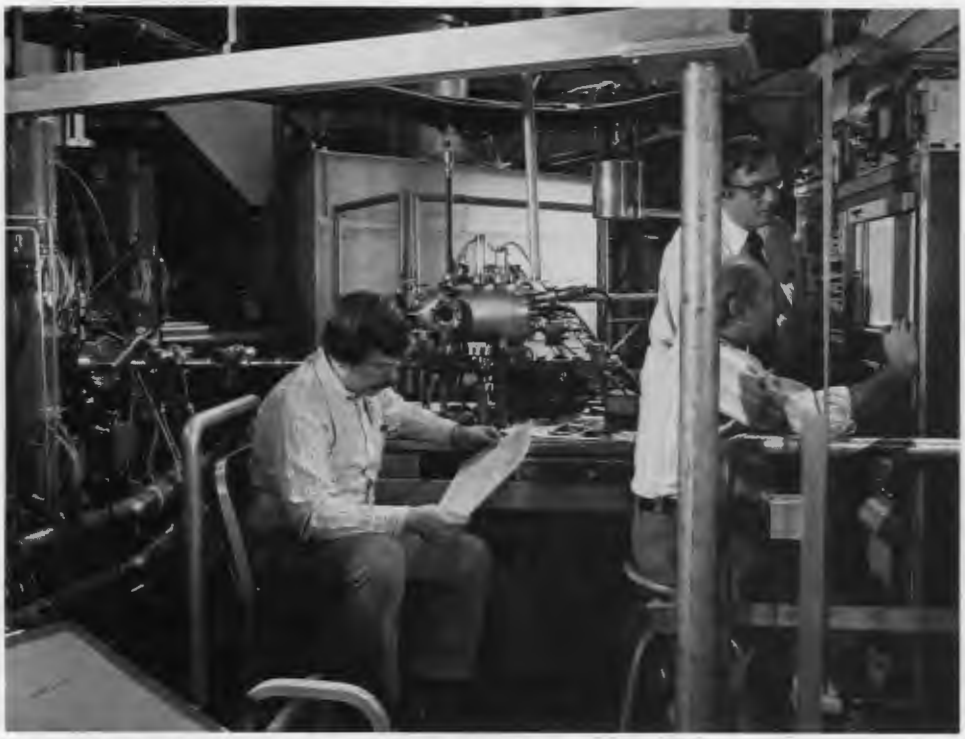
A technician inspects the plasma chamber in the Joint European Torus (JET). The vacuum vessel of this experimental fusion device has an internal projected surface area of 250 m^2 and a volume of 200 m^3. Its normal operating temperature is 300°C, but it can be baked to 500°C. At JET it was discovered that using methane to deposit thick carbon films on the vessel's inner wall allows the creation of exceptionally clean, energetic plasmas.

station built at ORNL in 1976. This group was probably the first to travel to other tokamaks to compare and analyze the values of different cleaning techniques; now other groups, including Japanese researchers, are becoming involved in such efforts.

The analytical technique used in ORNL's portable station is one of two used in 1975 to help solve an impurity problem at ORNL's original tokamak, ORMAK. At that time ORNL was one of the world's first laboratories to use glow discharge cleaning. With this technique, impurities are removed by a low-pressure gas ionized by an electrical discharge. Similar low-temperature plasmas have long been used to provide the glow in fluorescent lights and neon signs.

After the surface analysis station was built, it was used to monitor glow discharge cleaning on ORNL's Impurity Study Experiment (ISX-A) in 1977. The following year Clausing and associates took it to the T-12 tokamak in the Soviet Union and then to GA Technology's D-III tokamak in San Diego, California. In 1982 and 1983 it was used on the TEXTOR machine at Jülich, FRG, and on the Joint European Torus (JET) tokamak at the Culham Laboratory in Abingdon, England.

In 1975 Clausing and his colleagues experimented with two techniques to analyze the impurities on the surfaces of metallic samples placed in plasmas. The first technique, soft X-ray appearance-potential spectroscopy, was used inside the ORMAK tokamak to analyze the samples bombarded by electrons, ions, and energetic neutral atoms in the

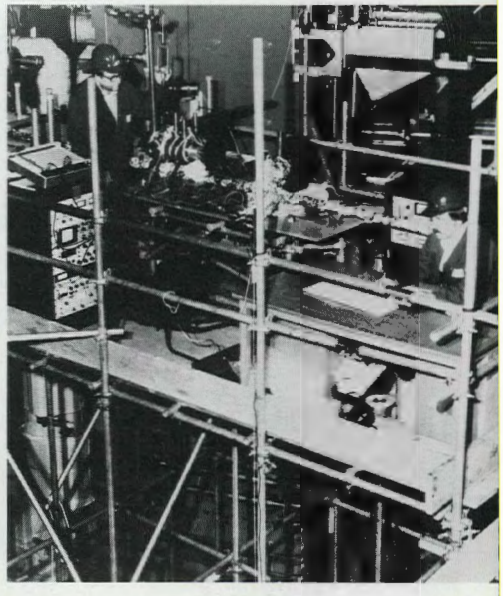


Heatherly, Doc Emerson, and Clausing with the surface analysis station on ISX-A at ORNL. Measurements of wall conditioning and other plasma edge phenomena were made on both ISX-A and ISX-B from 1978 through 1984.

plasma. The second technique, Auger electron spectroscopy, provided more information but could not be used in situ. To overcome this problem a method was developed to move samples in and out of the machine for immediate analysis by Auger spectroscopy while still under ultrahigh vacuum. This method allowed Clausing's group to develop a portable surface analysis station.

Why did the ORNL group build a portable station? Says Clausing: "Our intention was first to analyze different techniques for removing impurities from wall samples in a controlled environment in our M&C lab. Then we wanted to compare our lab results with results on complicated fusion machines such as the ORNL tokamaks at the Y-12 Plant and tokamaks in other places. So we needed a portable analysis station to characterize the usefulness of these techniques for wall cleaning and to determine what problems required more study in our lab. From our lab pilot studies we could suggest experiments and techniques that could work on fusion machines. Then with our portable station we would test how effectively modified techniques or different combinations of techniques worked as we monitored the progress of cleanup."

Auger spectroscopy was selected because of its good surface sensitivity and relatively well-developed technology—that is, its superior ability to determine the identity and quantity of elements on the very surface of the metal samples made of "wall" materials, primarily Inconel (an alloy developed by International Nickel, Inc.) and stainless steel. Says Clausing, "With this technique, we can accurately determine the composition of the first one or two atom layers on the surface. The technique is limited to the first few



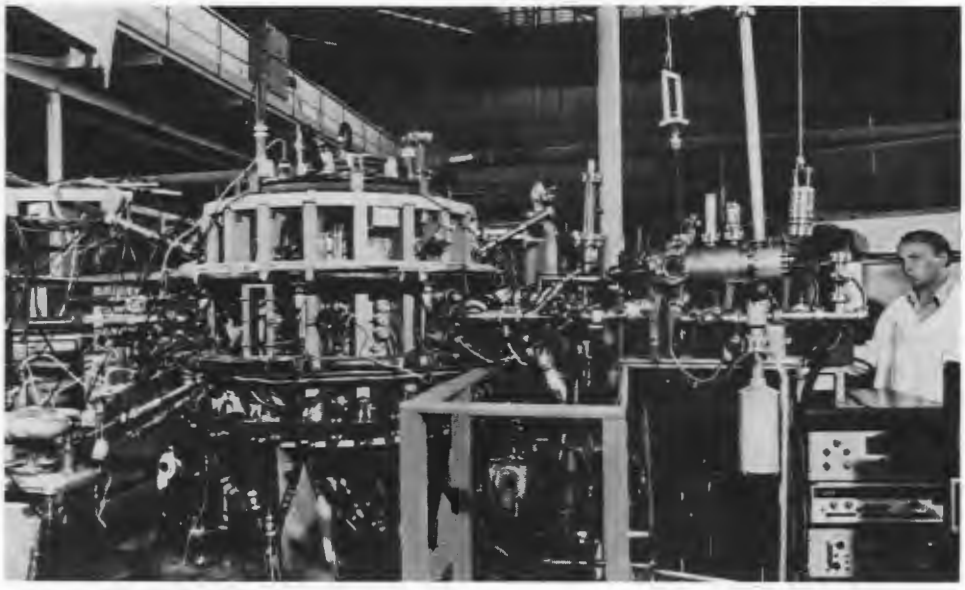
Clausing and Heatherly operate the surface analysis station on JET. The work area is about 6 m (20 ft) above the floor and at the midplane of the tokamak.

atom layers because the Auger electrons used to identify the elements present can escape with their characteristic energy only if they originate very near the surface; they lose energy as they move through the bulk of the material."

In the Auger technique, electrons bombard the surface atoms, creating an inner-shell vacancy in the atom. An electron from a higher energy level falls into the vacancy and transfers its extra energy to another electron, which is emitted at a characteristic energy for that particular atom. Clausing and his associates measure these energies and compare them to a catalog of Auger spectra. (Auger transitions exist for all elements except hydrogen and helium.) With this information they can identify elements on the sample surface and measure their concentrations.

Sources of Impurities

The stainless steel inner wall of a tokamak vessel can look shiny



ORNL's surface analysis station was operated in 1978 on the T-12 Tokamak at the Kurchatov Institute in Moscow. The station was used to conduct studies on the effectiveness of glow discharge cleaning using techniques developed by the Russians.

and clean, but careful analysis shows that the first few atomic layers on the surface contain a multitude of elements, including not only iron, nickel, and chromium (the main constituents of stainless steel) but also a large variety of impurities, such as oxygen, nitrogen, sulfur, potassium, calcium, sodium, chlorine, and fluorine. Carbides and hydrocarbons may also be present. Where do these impurities come from?

"If you expose clean metal to the atmosphere," says Clausing, "it will oxidize and adsorb water vapor, oxygen, nitrogen, hydrocarbons, and other airborne substances. Metals cleaned with water solutions will also pick up dissolved solids from the water, such as potassium, calcium, sodium, and chlorine. Fluorine is commonly used in cleaning and etching solutions, such as hydrofluoric acid. Sulfur is present in most metals as an impurity; it moves, or segregates, to metal surfaces at high temperatures. Hydrocarbons come not only from the air but also from working fluids and lubricants in the vacuum system. Carbides come from the metal itself and from interactions between the plasma and hydrocarbons on the metal surface."

How do impurities rob the plasma of its fuel and energy? Explains Clausing: "Any material with a high atomic number can rob the plasma of its energy and even of its hydrogen fuel by displacing hydrogen. The impurities become ionized and replace one or more hydrogen ions. For example, consider an oxygen impurity atom. *One* oxygen atom can provide up to eight electrons to the plasma and the resulting ion with eight positive charges could displace eight hydrogen ions. The reason for this displacement is that a plasma must have an equal number of positive charges, or ions, and negative charges, or electrons. If a plasma is 7% oxygen, for example, its reactivity is reduced by a factor of 4."

Besides displacement of hydrogen, another deleterious effect on the plasma is the loss of energy that occurs when the partially ionized medium or heavy impurities are excited by the high-temperature electrons and ions in the plasma. As a result of this electronic excitation and subsequent de-excitation, these impurities can quickly radiate away much of the plasma's energy. "The amount of power radiated by impurity ions increases as the fourth power of the atomic number of the impurity," says Clausing, "so it's easy to see why heavy element impurities are so damaging to the plasmas."

How do these impurities enter the plasma? Clausing says that the impurities on the wall surface may be removed by photons (radiation) from the plasma, by evaporation due to heating by contact with the high-temperature plasma, by arcing caused by the passage of an electric current through the plasma to the wall, and by sputtering—a physical process by which a particle leaving the plasma knocks an atom from the wall into the plasma.

Cleaning Techniques

Solvents and etchants can be used to remove "gross" wall impurities, but some will still remain. Like self-cleaning ovens, tokamaks can be heated to remove contaminants from their inner walls. By heating the vessel to a few hundred degrees Celsius, a process known simply as baking, tokamak operators can vaporize some wall impurities (e.g., water vapor), which are then removed as a gas or vapor (thermal outgassing) by the vacuum pumps. "Sometimes if the heating is not uniform," says Clausing, "the pumps do not completely remove the impurities, but some of them move to cooler areas of the machine where they may be less likely to interact strongly with the plasma. We'd really like to get rid of them, because if we just sweep them into

the corners or under the rug, so to speak, they will come out again and get into the plasma."

To remove the most stubborn impurities, scientists have tried tokamak shots (high-temperature hydrogen plasmas contained by magnetic fields) and glow discharge cleaning (low-temperature plasmas of hydrogen, helium, or krypton not confined by magnetic fields). The glow discharge cleaning plasmas are commonly referred to by names that describe the method of creating the plasma. The methods used to excite plasmas include

- a high-voltage (500 V) current,
- radiofrequency (RF) radiation (commonly, 13.6 MHz),
- RG power (a hybrid of high-voltage and RF excitation), and
- electron cyclotron resonance (ECR) excitation, which uses microwaves in a magnetic field.

Each experimental group tends to like the techniques it has pioneered. The Germans often use RG cleaning, and some Japanese employ ECR glow discharge techniques. Simple high-voltage glow discharges seem to be used frequently in both the Soviet Union and the United States. Low-power tokamak shots are almost universally used as a part of the wall-conditioning process.

However sophisticated these methods are, not all impurities will be removed. Says Clausing: "The most important result from our lab and machine observations is that, after initial removal of contaminants by solvents and baking and subsequent treatment by glow discharge cleaning, at least a monolayer of impurities will inevitably remain. Generally, if we try to remove the oxygen, we may end up with a layer of carbon, often in the form of carbides. If we try to remove carbon, for example, we may find oxygen or sulfur or nitrogen on the wall surface. Clean surfaces are just so attractive to impurities that they are impossible to keep impurity-free under practical conditions in today's experimental machines. If we accept the inevitability of these conditions, we have to decide how to manipulate the surface to get rid of the worst offenders and leave the impurities that we can best tolerate."

The Methane Fix

One strategy for removing the most common impurities—oxygen, carbon, and sulfur—is to run a hydrogen plasma through the tokamak vessel. The hydrogen reacts with oxygen to form water vapor, with carbon to form methane, and with sulfur to form hydrogen sulfide. These gases can be pumped out. "What works best, is removing oxygen by reacting carbon with it to make carbon monoxide, a gas that is more easily removed by pumping than is water vapor," Clausing says. "Because oxygen has a higher atomic number than carbon and therefore is more damaging to the plasma, it is more desirable to remove oxygen than carbon, anyway. This can be done by adding some methane to the hydrogen plasma. In this way, oxygen is removed, leaving either a carbon-rich surface of metallic carbides or a thin layer of carbon on the surface." This technique was demonstrated in the laboratory and subsequently verified in TEXTOR and JET.

"At JET and TEXTOR," Clausing continues, "we found that in situ deposited carbon-rich layers worked very well. They caused very little harm to the plasma during operation and protected the plasma from other impurities. When straight hydrogen plasmas were run later to remove the carbon film from JET, not only oxygen but also other impurities under the film—such as sulfur, nitrogen, chlorine, and fluorine—left the machine as volatile compounds."

Clausing tells a story about an accident at JET in 1982 that led to insights about the value of adding methane to glow discharge plasmas to improve cleaning. "When we originally put methane in JET, it was not our intention to deposit a layer of carbon but simply to provide enough methane to get rid of oxygen and other impurities on the surface, leaving a layer of metallic carbides. Because of a mistake or oversight, more methane was admitted than had been planned. As a result, the carbon deposits on some parts of the machine, including my sample, were pretty thick. But the machine worked much better than had been anticipated. In fact, the first plasmas not dominated by impurity problems were obtained.

"By contrast, in an earlier experiment in TEXTOR we put in just a little methane at a time until the oxygen content decreased and carbides increased. As soon as we saw substantial amounts of metal carbides on the surface, we stopped the methane injection. This procedure gave good results but didn't remove other impurities to the extent that thicker carbon films did in JET.

"By the happy accident at JET, we learned that by covering up metal surfaces completely with a carbon film, we can attain surface conditions that allow the creation of very good plasmas. We can later remove the carbon with glow discharge cleaning using hydrogen, making methane in the process and removing impurities at the same time with the carbon film. Because of this information, JET now routinely uses the strong methane treatment."

Knowing that the Tokamak Fusion Test Reactor (TFTR) at the Princeton Plasma Physics Laboratory has problems with

St. Basil's Cathedral in Red Square is one of the most famous landmarks in Russia. The Russian scientists who worked with Clausing showed him and his family around Moscow on their arrival and helped them settle into an apartment near the Kurchatov Institute.

impurities such as oxygen and various metals, Clausing recommends that researchers operating TFTR for energy breakeven studies consider adding methane during glow discharge cleaning.

Beryllium Limiter Experiment

In 1984 Clausing's group helped assess the effectiveness of beryllium as a limiter, a component inserted next to the plasma to protect the first wall. The limiter defines the edge of the plasma and interacts with it. Beryllium was chosen as a limiter material worth studying because it has the lowest atomic number of any metal and therefore was judged to pose less of a threat to the plasma energy and fuel. The beryllium limiter used in the experiment was designed by ORNL in collaboration with Sandia National Laboratories, which fabricated it. Its performance was compared with that of graphite coated with titanium carbide.

In a cooperative study performed with JET investigators from January through September 1984, ORNL researchers led by Phillip Edmonds and Peter Mioduszewski of the Fusion Energy Division demonstrated the value of a beryllium limiter in the concluding experiment on ORNL's ISX-B. During this experiment the beryllium limiter suffered some melting in high-power tests. However, the beryllium performed

very well as a getter—that is, the beryllium atoms evaporated from the limiter were deposited on the wall, where they reacted with the oxygen impurities to form beryllium oxide. By "gettering" the oxygen and holding it strongly as a compound, the beryllium prevented much of the oxygen from infiltrating the plasma. (Because of his background, Clausing was particularly interested in the findings on beryllium gettering; in 1960 he developed data and techniques for the large-scale use of titanium getter pumping in fusion research.)

In collaboration with the ORNL and JET researchers on the ISX-B, Clausing and his associates monitored the initial machine cleanup and analyzed the transport of beryllium and other materials at the plasma edge by inserting a probe there and collecting what appeared. On the probe were samples of stainless steel wall material, aluminum, and a cube of beryllium. The JET researchers were particularly interested in the amount of deuterium (heavy hydrogen) that had collected on the beryllium limiter and the beryllium deposits distributed around ISX-B.

"The extent to which deuterium deposits on limiter materials is important to JET investigators," says Clausing, "because deuterium behaves like its sister hydrogen isotope tritium. JET will use tritium in its future plasmas. However, because tritium is radioactive, JET researchers want to to predict how much tritium would be retained on limiter and wall materials so they can determine whether maintenance and operation of the machine or disposal of these materials later would be a serious problem. Our measurements of deuterium outgassing and deposits on the beryllium cube, together with other measurements by Ray Zuhr and Jim Roberto of ORNL's Solid State Division on the beryllium limiter and other surfaces inside ISX-B, gave JET researchers a basis from which to estimate tritium retention in JET."

New Wall Materials

In today's experimental tokamaks, first walls are made of stainless steel and Inconel. But researchers are considering other materials that not only can resist high temperatures but also have other desirable properties. For example, vanadium alloys are candidates for first walls because of their resistance to radiation damage. Carbon coatings and graphite are being considered for limiters and wall areas that receive high heat fluxes because of their resistance to heat and their relatively low atomic number. One proposal is to build a first wall of Inconel and coat it with carbon.

Clausing and his group plan to study the altered complexion of the surface impurity problem in new wall materials. "The properties of a carbon film will not be the same as those of a bulk graphite," he says. "Hydrogen recycling and retention and sputtering of surface impurities into the plasma will no doubt be different. If a deuterium-tritium plasma is run, a tritium retention problem could occur because hydrogen isotopes will become incorporated into the carbon coating. We intend to study the exchange of hydrogen isotopes between the wall and plasma to determine if tritium retention is an important problem. We will be studying carbon coatings in our laboratory here and will relay our findings to investigators at TEXTOR and JET, who are also studying and using carbon coatings."

International Cooperation

From his travels and collaboration in Western Europe, Clausing has these comments: "The European Economic Community has done a great job with JET. It was built on time and under budget. The initial shakedown and early operation of the machine were done quite professionally. It is impressive that machine components and scientists from all over Europe can be brought together effectively to achieve important results of vast interest to the entire fusion community.

"Our visit to the JET project was a mutually beneficial scientific effort in the area of plasma-wall interactions. The cleanup could not have been done as well or have been adequately documented without the resources made available from ORNL. The JET studies helped us understand the processes involved in cleanup. This type of international cooperation is an extremely good investment for all concerned and should be strongly encouraged."

Clausing concludes that his experiences are not unique; many people in the Fusion Energy Division have had similar experiences and, in fact, the whole fusion program is an excellent example of the benefits of a free exchange of personnel and information from all over the world. "Many other areas of life," he says, "could benefit from this type of open communication and international cooperation." ornl

awards and appointments

Don Ferguson has received the Glenn T. Seaborg Award, which was presented by the organizers of the 1985 Actinide Separations Workshop. He was cited for his "outstanding work in the field of actinide element separations." He also has been named Outstanding Engineer of the Year by Tennessee Technological University.

Frank Plasil has been selected as the recipient of a Senior U.S. Scientist Award by the Alexander von Humboldt Foundation of the Federal Republic of Germany.

Steven E. Lindberg has received the 1984 Scientific Achievement Award of ORNL's Environmental Sciences Division.

C. T. Liu has been appointed to the Awards Committee of the Materials Research Society.

John McGowan has been selected as the exhibit chairman for the Pressure Vessel and Piping Division Conferences for 1986, 1987, and 1988 of the American Society of Mechanical Engineers.

Tony A. Gabriel has received an award for professional excellence from the Radiation Protection and Shielding Division of the American Nuclear Society. He was honored for his contributions to the field of radiation shielding.

Bruce Peterson, Frank Southworth, Shih-Miao Chin, and **Richard Davis** have received the 1985 Citation Award from the Applied Geography Project Awards Program of the Association of American Geographers. The award cited the ORNL researchers' development of the Convoy Mobilization Automated Support System, a convoy-routing model that keeps track of 528,000 km (330,000 miles) of roads for the U.S. Army.

Bill R. Rodgers has been elected a director of the Fuels and Petrochemicals Division of the American Institute of Chemical Engineers.

Samuel H. Liu has been elected to the Executive Committee of the Division of Condensed Matter Physics of the American Physical Society.

Vivian Jacobs has been elected secretary of the national Society for Technical Communication (STC). She was also elected first vice-president of the East Tennessee Chapter of STC. **Jeanne Dole** was elected secretary of the local organization.

Billy G. Eads has been named Eminent Engineer by the Tennessee Alpha Chapter of Tau Beta Pi, the second-oldest honor society in the United States.

In the 1984–85 International Technical Publication Competition sponsored by the Society for Technical Communication, four ORNL entries received awards. They were "Use of Laser-Excited Fluorescence To Measure Mixed-Function Oxidase Activity," *Clinical Chemistry*, September 1983, by **B. Zane Egan, Norman E. Lee,** and **Carl A. Burtis**, award of distinction in scholarly and professional articles; *Synthetic Fossil Fuel Technologies* (1984), by **Kenneth Cowser** and **Vivian Jacobs**, award of excellence in the books category; *Energy Division Annual Progress Report for Period Ending September 30, 1983*, by **William Fulkerson, the Energy Division**, and **Janice M. Asher**, award of excellence in periodic activity reports; "Characterization of Hydrofracture Grouts for Radionuclide Migration," *Advances in Ceramics: Nuclear Waste Management* (1984), by **David Stinton, Earl W. McDaniel**, and **Irene Brogden**, award of achievement in scholarly and professional articles.

Tuan Vo-Dinh has received the first annual Excellence in Research Award of the Health and Safety Research Division. He is leader of the Advanced Monitoring Development Group of that division.

Martin Marietta Energy Systems, Inc., honored its employees May 24, 1985, at the first annual dinner and awards presentation program at the Hyatt Regency Hotel in Knoxville. Of the 144 awards, 75 were given to ORNL employees. An additional 24 awards were given later to publications authors from ORNL. The inventor of the year was **Brian Sales**; the scientist of the year was **Chain T. Liu**; and the author of the year was **Peter Mazur**, all ORNL employees. These three plus **Phillip Thompson** of ORNL are among the seven Energy Systems employees who received Jefferson Cups from Martin Marietta Corporation.

Eleven ORNL employees received an Operational Performance Award, which recognizes outstanding, exemplary performance in management, business, personnel, manufacturing,

Martin Marietta Energy Systems, Inc., honored its employees May 24, 1985, at the first annual dinner and awards presentation program at the Hyatt Regency Hotel in Knoxville. Of the 144 awards, 75 were given to ORNL employees. The author of the year was Peter Mazur (left) of the Biology Division; the inventor of the year was Brian Sales (center) of the Solid State Division, and the scientist of the year was Chain T. Liu (right) of the Metals and Ceramics Division.

and other similar functions. The winners were **Stephen C. Clark**, for outstanding technical contributions in resolving manufacturing complexities and fabricating prototype experimental components for a U.S.-Japanese materials experiment when all other fabrication resources failed; **Donna S. Griffith**, for outstanding leadership in achieving significant improvements in energy efficiency, saving nearly $1 million in energy costs for the ORNL Biology Division; **Edwin H. Krieg, Jr.**, for unusual cost-effectiveness in project management while maintaining full responsiveness to ORNL client requirements; **Bennie McNabb, Jr.**, for outstanding service in technical support to materials science and technology, especially in management and execution of complex engineering assignments; **Michael E. Mitchell**, for outstanding performance in formulating the model National Pollutant Discharge Elimination System (NPDES) permits for Y-12 and the permitting strategy formulation for the Oak Ridge installations; **Nancy P. Norton**, for outstanding management in increasing productivity of library services by implementing a multifunction, state-of-the-art computerized library system, resulting in an annual personnel savings of 12%; **Thomas W. Oakes**, for outstanding leadership in establishing and directing the Laboratory's environmental management program toward correction of ORNL's long-standing environmental concerns; **Phillip E. Parrett**, for outstanding leadership and achievements in recruiting professionals at ORNL, specifically, an all-time high acceptance rate of 71% for Ph.D.'s and a near record of 66% for B.S. or M.S. candidates; **W. Ronald Ragland**, for sustained outstanding performance in the development and administration of complete computerization of capital equipment, budget schedules, overhead reports, and monthly cost projections; **Robert W. Schaich**, for outstanding management of radioisotope processing and radioactive-materials packaging and shipping operations; and **G. Scott Trotter**, for outstanding performance in the Department of Energy competition for "Security Inspector of the Year," where he was awarded second runner-up.

Fourteen ORNL employees received the Inventor Award, which recognizes innovative employee contributions to the activities of Energy Systems. The winners were **Carlos E. Bamberger**, for creative application of knowledge from inorganic chemical research to inventions in nuclear fuel reprocessing, hydrogen production, and non-oxide ceramics; **Richard J. Fox**, for continuous innovation and application of novel mechanical and measurement ideas to development of new radiation- and temperature-sensing instruments, over more than 40 years; **Elias Greenbaum**, for innovative research and development in the biotechnology of microalgae and photosynthetic water-splitting for renewable fuels and chemical feedstock production; **Paul A. Haas**, for many contributions to the nuclear fuel cycle, especially development of sol-gel process technology for reactor fuel preparation from oxides of uranium and plutonium; **Joseph P. Hammond**, for sustained creativity in the fabrication of nuclear reactor fuels and development of methods for joining refractory metals and ceramics for energy applications; **George S. Hurst**, for achievement of isotopically selective detection, storage, and retrieval of small numbers of noble gas atoms, based on the technique of resonance ionization spectroscopy; **Henry Inouye**, for development of temperature- and impact-resistant precious metal alloys as cladding materials for radioisotope thermoelectric generators used to power space probes to the outer planets; **Furn F. Knapp, Jr.**, for development and testing of a new clinical radioisotope generator that allows safe and prolonged use of

iridium-191*m* to evaluate cardiovascular defects and blood flow in young patients; **Herbert A. Mook, Jr.**, for the design and development of a high-intensity, ultrasonically pulsed, time-of-flight spectrometer that greatly increases reactor-produced neutron intensity for inelastic scattering research; **Lawrence W. Owen**, for initial magnetic field and single-particle orbit calculations that established the Elmo Bumpy Square fusion plasma confinement concept as a variant of the Elmo Bumpy Torus; **Thomas C. Quinby**, for creative efforts and numerous innovations, over more than 20 years, in preparing radioactive and stable nuclides in useful forms for research, development, and engineering applications; **Brian C. Sales**, for discovery of lead-iron phosphate glass as a new, easily prepared, highly leach-resistant medium for safe and permanent disposal of commercial and defense-related high-level nuclear wastes; **James S. Wike**, for development of a process to produce yttrium-90 (free of strontium-90) as an internal radiation agent for effective treatment of liver cancer and other advanced cancer therapies; and **James M. Williams**, for developing an economic ion-implantation treatment that virtually eliminates wear and corrosion as a clinical problem for artificial hip- and knee-joint prostheses made of a titanium alloy.

ORNL employees received 58 Publication Awards, which recognize superior employee performance in the authorship of a paper, technical article, or book that represents a significant advance in the author's professional field. Five employees received two publication awards. The winners were **T. C. Awes, R. L. Ferguson, F. E. Obenshain, F. Plasil**, and **G. R. Young**, for "Energy Division in Damped Reactions"; **C. F. Barnett**, for "Particle Plasma Diagnostics"; **P. F. Becher**, for "Mechanical Reliability of Ceramic Windows in High Frequency Microwave Heating Devices. Part 2: Mechanical Behaviour of the Ceramics"; **J. M. Begovich** and **W. G. Sisson**, for "A Rotating Annular Chromatograph for Continuous Separations"; **M. V. Buchanan** and **G. Olerich**, for "Differentiation of Polycyclic Aromatic Hydrocarbons Using Electron Capture Negative Chemical Ionization"; **R. H. Busey, H. F. Holmes**, and **R. E. Mesmer**, for "The Enthalpy of Dilution of Aqueous Sodium Chloride to 673 K Using a New Heat-Flow and Liquid-Flow Microcalorimeter: Excess Thermodynamic Properties and Their Pressure Coefficients"; **B. A. Carreras, H. R. Hicks, J. A. Holmes, V. E. Lynch**, and **G. H. Neilson**, for "Zero Current High-Beta Stellarator Equilibria with Rotational Transform Profile Control"; **L. Dresner**, for "Superconductor Stability, 1983: A Review"; **W. R. Emanuel, G. G. Killough**, and **W. M. Post**, for "Modeling Terrestrial Ecosystems in the Global Carbon Cycle with Shifts in Carbon Storage Capacity Due to Land-Use Change"; **J. M. Giddings, P. J. Franco, R. M. Cushman, L. A. Hook**, and **G. R. Southworth**, for "Effects of Chronic Exposure to Coal-Derived Oil on Freshwater Ecosystems: II. Experimental Ponds"; **R. Gwin, R. R. Spencer**, and **R. W. Ingle**, for "Measurements of the Energy Dependence of Prompt Neutron Emission from ^{233}U, ^{235}U, ^{239}Pu, and ^{241}Pu for $E_n = 0.005$ to 10 eV Relative to Emission from Spontaneous Fission of ^{252}Cf"; **R. L. Huddleston**, for "An Improved Multiaxial Creep-Rupture Strength Criterion"; **R. C. Isler**, for "Impurities in Tokamaks"; **D. W. Johnson**, for "Soil-Mediated Effects of Atmospherically Deposited Sulfur and Nitrogen"; **T. D. Jones**, for "A Unifying Concept for Carcinogenic Risk Assessments: Comparison with Radiation-Induced Leukemia in Mice and Men"; **D. M. Kroeger, J. O. Scarbrough**, and **C. G. McKamey**, for "Effects of Short-Range Order on Electronic Properties of Zr-Ni Glasses as Seen from Low-Temperature Specific Heat"; **R. J. Lauf, T. B. Lindemer**, and **R. L. Pearson**, for "Out-of-Reactor Studies of Fission Product–Silicon Carbide Interactions in High-Temperature Gas-Cooled Reactor (HTGR) Fuel Particles"; **A. C. Marchok, S. F. Huang**, and **D. H. Martin**, for "Selection of Carcinogen-Altered Rat Tracheal Epithelial Cells Preexposed to 7,12-dimethyl-benz[a]anthracene by Their Loss of a Need for Pyruvate to Survive in Culture"; **P. Mazur**, for "Freezing of Living Cells: Mechanisms and Implications"; **T.-H. Peng**, for "Ocean Life Cycles and the Atmospheric CO_2 Content"; **F. Plasil, T. C. Awes, R. L. Ferguson, F. E. Obenshain**, and **G. R. Young**, for "Angular-Momentum-Dependent Fission Barriers in the Rare-Earth Region"; **S. Raman** and **C. R. Vane**, for "Implications of Heavy-Ion-Induced Satellite X-ray Emission"; **B. C. Sales** and **L. A. Boatner**, for "Lead-Iron Phosphate Glass: A Stable Storage Medium for High-Level Nuclear Waste"; **T. Vo-Dinh**, for *Room Temperature Phosphorimetry for Chemical Analysis*; and **W. B. Whitten** and **J. M. Ramsey**, for "Self-Scanning of a Dye Laser Due to Feedback from a $BaTiO_3$ Phase-Conjugate Reflector."

Two ORNL employees received a Community Service Award, which recognizes outstanding and noteworthy performance by Energy Systems employees engaged in

voluntary, uncompensated activities (social, civic, or governmental) that provide significant benefit to the community. The winners were **Lorena F. Truett**, for her outstanding work with Girl Scouts, Boy Scouts, the church, community musical activities, and the 4-H Club and **James T. White**, for 30 years of devotion and leadership in scouting activities that have helped boys and young men become better citizens of our community.

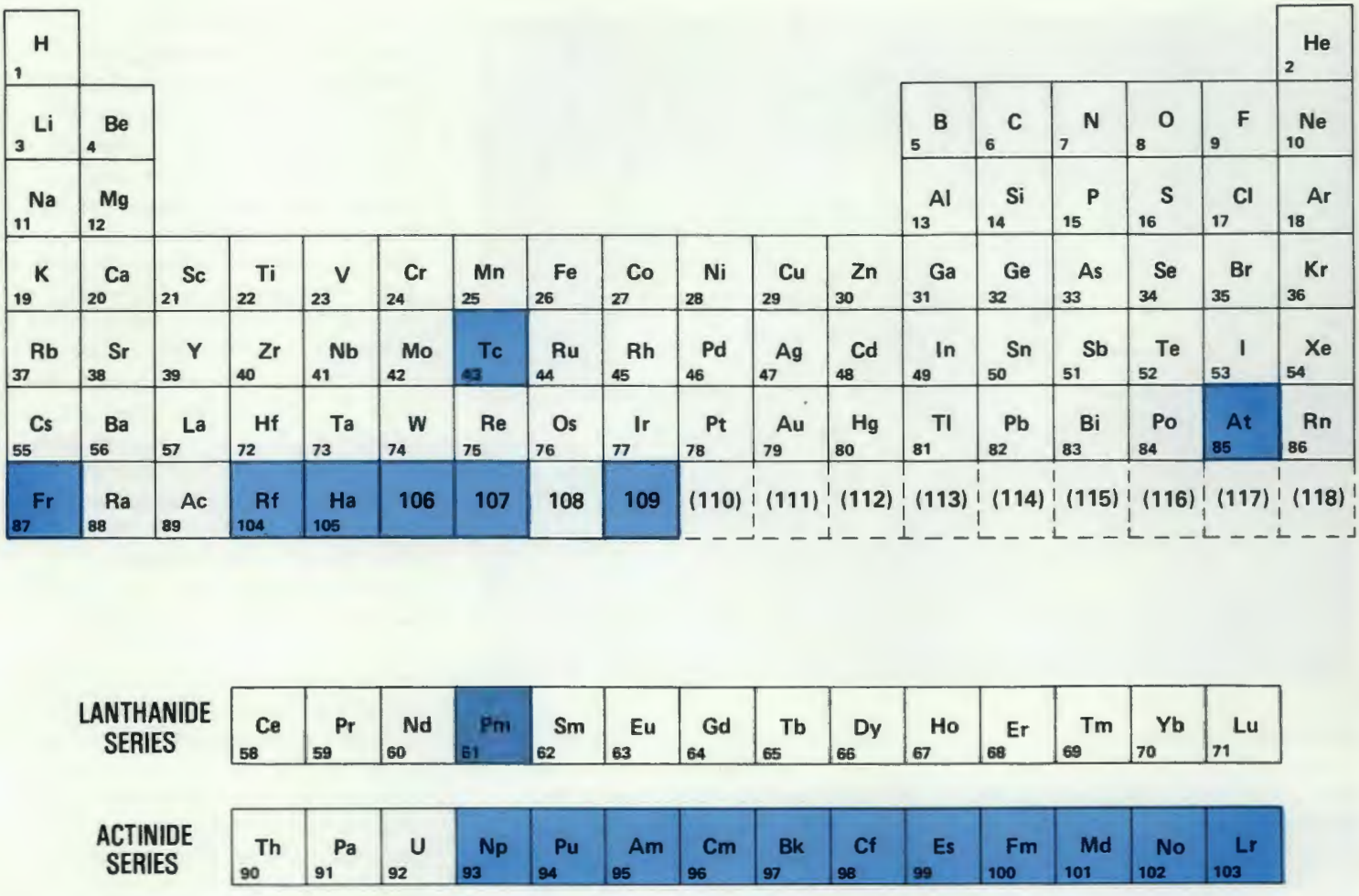
Twenty-three ORNL employees received a Technical Achievement Award, which recognizes the excellence of employee contributions of a scientific or engineering nature to the activities of Energy Systems. The winners were **William R. Busing**, for innovative use of computers to determine crystal structures by diffraction methods and for the theoretical interpretation of such structures; **William D. Cain**, for development of a method to accurately determine magnetic fields induced by complexity-deformed, three-dimensional electrical conductors, leading to the economical design of the helical coil system of the Advanced Toroidal Facility (ATF) device; **Thomas A. Carlson**, for development of techniques using synchrotron radiation to accurately determine molecular structure and for the establishment of ORNL as an international center for photoelectron spectroscopy; **Steinar J. Dale**, for exceptional contributions to research and development of electrical insulation and dielectric materials for use in electrical utilities systems; **Ward W. Engle, Jr.**, for outstanding contributions to the field of radiation shielding, including development and upgrading of the worldwide ANISN code; **David L. Greene**, for continuing contributions to understanding energy use and conservation in transportation, ranging from determining basic economic parameters of gasoline demand to energy planning for developing countries; **Frederick C. Hartman**, for establishing a genetic engineering laboratory and developing techniques to make new mutations of the natural enzyme used by photosynthetic plants to convert atmospheric CO_2 into sugars; **Bennett C. Larson**, for pioneering research using synchrotron radiation sources for time-resolved X-ray diffraction measurements and application of the results to nonequilibrium crystal growth; **Leslie W. Little**, for outstanding contributions in planning, coordinating, designing, and constructing interfaces for the mandated Y-12 treatment facilities required to eliminate outdated disposal practices; **Chain T. Liu**, for exemplary contributions in the field of ordered intermetallic alloys, resulting in development of nickel aluminides that maintain structural integrity at high temperatures; **Vickie E. Lynch**, for definitive studies of magnetic confinement fields and plasma equilibria, leading to detailed designs for several significant fusion energy experiments; **Louis K. Mansur**, for leadership in theoretical modeling and experimentation, resulting in a comprehensive understanding of the evolution of microstructure in metals and alloys during neutron irradiation; **Alfred H. Narten**, for excellence of research leading to the application of neutron and X-ray scattering for obtaining information on the atomic and molecular structure of liquids and solutions; **S. Michael Ohr**, for introduction of a new electron microscopy technique used to study crack propagation in metals and for development of a dislocation theory of fracture; **David K. Olsen**, for contributions to measurement and analysis of the resonance cross section of uranium-238, resulting in significant contributions to the development of fast breeder reactors; **Y. Martin Peng**, for significant contributions in the field of plasma engineering, leading to substantial improvements in reactor concepts such as the spherical torus; **R. Julian Preston**, for advancement in assessment of biological risk to public health, especially for definitive research on chromosomal damage caused by radiation and toxic chemicals; **Ted L. Ryan**, for demonstration of extreme flexibility and versatility in performing as principal engineer, general mechanical engineering department superintendent, and technical expert in designing the Large Coil Test Facility; **Douglas L. Selby**, for outstanding contributions leading to understanding and resolution of the pressurized-thermal-shock safety issue for nuclear reactors, resulting in savings for the nuclear industry in the millions of dollars; **Phillip B. Thompson**, for sustained contributions to the critical fusion energy experimental programs, including major contributions to the development of the Large Coil Test Facility; **Terry N. Tiegs**, for contributions to the development of silicon carbide whisker reinforced ceramic composites, resulting in substantial increases in strength and toughness at elevated temperatures; **John H. Whealton**, for contributions to the theoretical areas of plasma technology, low-divergence ion beams, atomic vapor laser isotope separation, and high-power radio-frequency plasma heating; and **John C. Whitson**, for development of computer-controlled measuring techniques to produce castings with sufficient precision to meet the required tolerances with no further machining for the ATF Project, resulting in savings of over $1 million.

O. L. Keller is director of the Transuranium Research Laboratory (TRL), a section of ORNL's Chemistry Division. He has been associated with the transuranium program since coming to ORNL in 1960 from the Massachusetts Institute of Technology, where he received his Ph.D. degree in physical chemistry. Keller has worked in both the nuclear and inorganic chemistry aspects of the transuranium program, and much of his work between 1967 and 1974 was carried out at the Oak Ridge Isochronous Cyclotron. Using its heavy-ion beams to bombard transplutonium targets from the High Flux Isotope Reactor (HFIR) and Transuranium Processing Plant (TRU), he and his colleagues produced still heavier elements in the transfermium region for study. At that time he participated in the development of the proposal for the construction of the Holifield Heavy Ion Research Facility at ORNL. He served on the Nuclear Physics Panel of the National Academy of Sciences between 1969 and 1972. From 1974 to 1984 Keller was director of the Chemistry Division. Since returning to TRL, he has been working with a consortium of four national laboratories (Berkeley, Livermore, Los Alamos, and Oak Ridge) on the chemistry and physics of the transeinsteinium elements. His particular interest is to determine the chemical properties of elements 101–105 that will transmit information on how relativity is affecting the electronic configurations of these elements, which occur at the junction of the actinide and transactinide series. Here, Keller watches Chick Wiggins working at a gloved box in TRU performing the final purification of individual transplutonium elements (produced at HFIR) after initial separation in cell operations.

Transuranium-Element Production and Research

By O. L. KELLER

The periodic table of the elements today extends to element 109. Fifty years ago the story was very different. At that time no elements heavier than uranium had been discovered, and it appeared that chemistry and physics would be limited to the study of elements from atomic number 1 to 92. But the invention of the cyclotron, which can bombard heavy element targets with nuclear particles to produce still heavier elements, changed all that. The first of the transuranium elements, neptunium and plutonium, were discovered in 1940–41 by Glenn Seaborg, Edwin McMillan, Philip Abelson, J. W. Kennedy, and A.C. Wahl, using the Berkeley 60-inch cyclotron and uranium targets. By 1958 eight more elements had been discovered by Seaborg and his co-workers. As might be expected these new elements provided an opportunity for learning aspects of chemistry and physics that had been inaccessible before. But production rates on accelerators are very low, and the reactors back then could not produce sufficient quantities of the transplutonium isotopes. So



The periodic table as arranged by Glenn Seaborg in 1945 places the actinide series below the lanthanide series to indicate their parallel electronic properties. The elements shown in color (representing 20% of the total) have been discovered since 1935. One of the elements (promethium, element 61) was discovered at ORNL as a result of work for the Manhattan Project. Elements 104, 105, and 106 were discovered at Berkeley using californium-249 (^{249}Cf) targets from ORNL's High Flux Isotope Reactor (HFIR) and Transuranium Processing Plant (TRU). The undiscovered superheavy elements (dotted squares) are actively being sought at laboratories in the United States, the United Kingdom, the Federal Republic of Germany, and the Soviet Union.

research efforts were being seriously hampered by an inadequacy in the production capabilities.

In response to this need for larger quantities, the directors of the national laboratories of the U.S. Atomic Energy Commission (AEC) recommended at a 1958 meeting that appropriate facilities be designed and constructed at Oak Ridge to produce useful research quantities of the transplutonium elements. The AEC shortly thereafter approved this recommendation. As a result, in the mid-1960s, Oak Ridge National Laboratory's High Flux Isotope Reactor (HFIR) and Transuranium Processing Plant (TRU) were built. They have been the centerpieces of the Western world's transplutonium production program ever since. At about the same time, an important, but smaller, transplutonium production facility was built in the Soviet Union.

HFIR-TRU is the gateway to the top of the chart of the nuclides. The chief products are macroscopic quantities of isotopes of elements 96–100 (Table 1) for direct studies in nuclear and inorganic chemistry and solid-state physics. These elements also serve as targets in heavy-ion accelerators for the synthesis of the still heavier elements and their isotopes, which are used for research. However, californium-252 (^{252}Cf) is not used exclusively for research. This strong neutron source is also important in the treatment of cancer and in some industrial applications.

Most of the isotopes in the HFIR-TRU mainline production are short-lived and require continued production to support the research in this region of the periodic table. For example, einsteinium-254 (^{254}Es) has a half-life of 276 days. As the longest-lived einsteinium

Table 1. Mainline HFIR-TRU production (elements 96–100)

Isotope	Half-life (principal decay mode)	Production (amount/year)
Curium-248	3.397×10^5 years (alpha)	150 mg
Berkelium-249	320 days (beta)	50 mg
Californium-249	350.6 years (alpha)	from ^{249}Bk decay
Californium-252	2.64 years (alpha)[a]	500 mg
Einsteinium-253	20.4 days (alpha)	2 mg
Einsteinium-254	275.7 days (alpha)	4 μg
Fermium-257	100.5 days (alpha)	1 pg

[a] ^{252}Cf has a neutron output from spontaneous fission of 2.3×10^{12} neutrons/s/g.

isotope, ^{254}Es is in such demand by experimenters that ORNL is evaluating the possibility of increasing the production capacity to 40 to 60 μg per campaign from the present level of 4 μg.

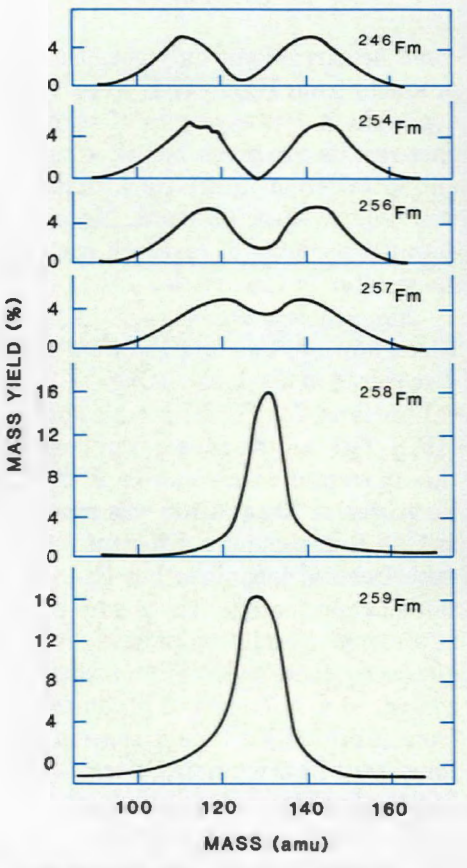
Moreover, ORNL is designing a successor to HFIR called the Center for Neutron Research (CNR). The goal of the CNR designers is to develop the world's best research reactor—one that will offer a significant improvement over the HFIR in transuranium element production, neutron scattering research, and experimentation on effects of radiation on materials. From the point of view of the transuranium research program, it is already clear that a large increase in the production of ^{254}Es for use as a target in accelerators would open up many new opportunities for understanding the chemical, atomic, and nuclear properties of the heaviest elements. In addition, increased quantities of ^{254}Es could be a springboard for discovering elements 110 through 120—the superheavy elements. Furthermore, it is expected that increased production of ^{252}Cf ($T_{1/2}$ = 2.65 years) will be needed as a new program is developed to detect structural faults in aircraft using neutron radiography.

Many advances in physics and chemistry over the last two decades could be carried out only because products from HFIR-TRU were available. A few examples of such past results are given below, with an indication of future work that will require these products. More detailed accounts of research are given later in the article.

New element discovery. Elements 104, 105, and 106 were discovered at Berkeley using californium-249 (^{249}Cf) produced at HFIR-TRU as the target. Further, the important confirmation of the identities of 104 and 105 was made at Oak Ridge using a different experimental technique but the same target isotope. The future of new element and new isotope discovery depends heavily on the continued availability of products from HFIR-TRU. ^{254}Es is especially important in this regard, because it has the highest proton and neutron numbers of any isotope with a half-life long enough for the isotope to be a target that can be used on a relatively routine basis. New element and new isotope discovery is fundamental to many advances in the transuranium field because it furnishes the raw material for discoveries in the other areas.

Symmetric spontaneous fission. In 1979 a group from Los Alamos National Laboratory (LANL) and Lawrence Livermore National Laboratory (LLNL) discovered that the nucleus of fermium-259 (^{259}Fm) fissions spontaneously principally into two equal fragments rather than into two unequal fragments, as was the case for all previously studied nuclei. That is, ^{259}Fm spontaneously fissions symmetrically rather than asymmetrically. A target of 10^9 atoms of ^{257}Fm from HFIR-TRU was used to make the ^{259}Fm isotope. The observation of symmetric spontaneous fission in ^{259}Fm opened up a whole new aspect of fission for investigation because all previously known spontaneous and low energy

For almost two decades ORNL's High Flux Isotope Reactor (HFIR) and Transuranium Processing Plant (TRU) have produced most of the transuranium elements used by researchers in the Western world. ORNL's Transuranium Research Laboratory, where research is carried out on the HFIR-TRU products, has become an international center for collaborative research. ORNL collaboration with the University of Tennessee has been particularly strong in inorganic chemistry and solid-state physics.



Mass yields for spontaneous fission of fermium (Fm) isotopes showing the symmetric character for the heavier members versus the asymmetric character for the lighter ones. Until ^{259}Fm was discovered (using a ^{257}Fm target from HFIR-TRU), all known spontaneously fissioning nuclei split asymmetrically. These new discoveries require revisions in the standard theories of fission.

Dick Hahn (left) and Dave O'Kelley ponder data relating to the very recent discovery of mendelevium-260. The discovery was made by Hahn and his collaborators at Lawrence Livermore Laboratory. This new isotope is of great interest because of its surprisingly long half-life of about 30 days.

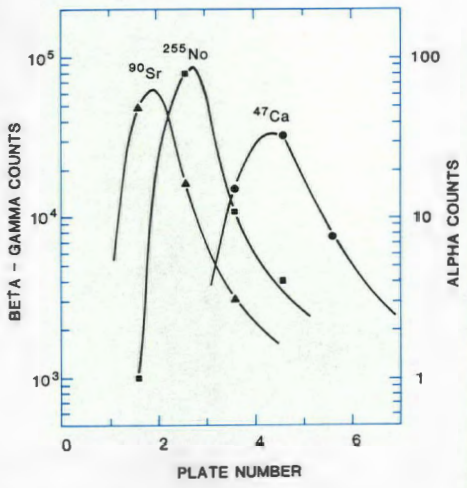
neutron-induced fission had been asymmetric.

So far three other nuclei—^{258}Fm, mendelevium-259 (^{259}Md), and rutherfordium-260 (^{260}Rf)—have been found to give symmetric mass distributions upon fissioning spontaneously. Beyond the abrupt change in the mass distribution between ^{257}Fm and ^{258}Fm, an anomalous increase of about 20% in the kinetic energy of the fission fragments also occurs. The anomalously high total release of kinetic energy is also shown by ^{259}Fm, but curiously ^{259}Md and ^{260}Rf have normal kinetic energy releases. These results are puzzling, and much more experimentation in the region of elements 100 and above is necessary before an adequate model for spontaneous fission can be developed. A prime motivation for continuing fission studies of this type is to understand the basic physics governing the limit of stability for chemical elements and, indeed, to find that limit.

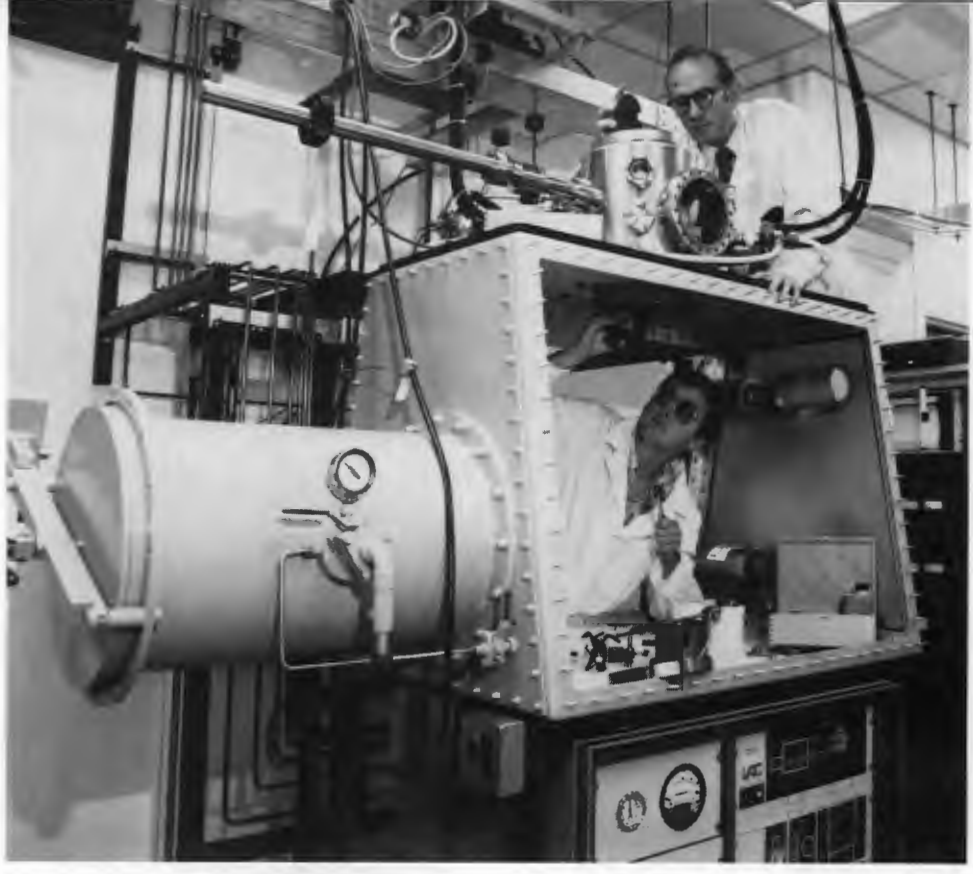
Chemistry and atomic physics. The isotopes produced at HFIR-TRU are also important for chemical studies. An early result of the discovery of the first transuranium elements was Glenn Seaborg's reformulation of the periodic table around his concept of an actinide series that would parallel the lanthanide series (see periodic table).

Developing the chemistry establishing the validity of Seaborg's form of the periodic table has been a major triumph of transuranium researchers. Much of this development depended upon the availability of macroscopic quantities of the elements curium, berkelium, californium, and einsteinium, all of which are supplied by HFIR-TRU. This dependence took two routes. The first was direct experimentation on the HFIR-TRU products themselves. The second was use of the HFIR-TRU products as targets in heavy-ion accelerators to make still heavier atoms beyond those that can be made in a reactor. These accelerator-produced elements have allowed chemical studies of mendelevium, nobelium, lawrencium, and rutherfordium to be carried out at various accelerators in the United States and in Europe. Such studies have also been done at Dubna in the Soviet Union. Because only quantities measured in atoms can be made at accelerators, the chemistry of these transfermium elements is carried out on the basis of a few atoms or just one atom at a time.

An outline of the chemical behavior of the heavy actinides and of the first member of the transactinides has been derived now from these macroscopic and microscopic chemical experiments.



The chemical behavior of nobelium (element 102) surprisingly resembles closely that of calcium and strontium in its normal divalent state, as shown by these chromatographic extraction results. The nobelium was produced using a ^{249}Cf target from HFIR-TRU. Much of the known chemistry of the transplutonium elements is based on samples produced at HFIR-TRU.



Dick Haire (top) and John Gibson assemble a mass spectrometer system for high-temperature thermodynamic and chemistry studies of transplutonium metals and compounds. Their investigations will extend to fermium (element 100), the heaviest element made in HFIR-TRU.

Two major results have been obtained. The first is that the actinide series clearly ends at lawrencium (element 103), and a new series begins at rutherfordium (element 104), as Seaborg's formulation of a 14-electron 5*f* series beginning at actinium (element 89) requires. The second result was finding out about an unexpected trend toward stability in the 2+ oxidation state that starts at californium (element 98) and progresses across the actinide series. This trend toward divalency (achieving maximum stability by contributing two electrons to participate in the bonding) culminates at nobelium (element 102), which is far more stable in the 2+ state than in 3+. This behavior is the opposite of its lanthanide homolog, ytterbium (element 70).

In fact, recent work at Berkeley and Oak Ridge has shown that nobelium in its ordinary chemistry is very similar to calcium (Ca^{2+}) and strontium (Sr^{2+}). We thus have the rather startling result that a bit of alkaline earth chemistry appears in the actinide series! Such behavior on the part of nobelium certainly serves as a spur to study the elements heavier than nobelium—namely, lawrencium, rutherfordium, and hahnium (element 105). So far in the history of the world, only one chemical experiment has been performed on lawrencium; three experiments have been done on rutherfordium; and no successful experiment has been carried out on hahnium. Future work on the chemistry of these elements will require large ^{254}Es targets.

In another important experiment researchers at Argonne National Laboratory carried out an atomic beam experiment demonstrating that fermium has a ground electronic state of $5f^{12}7s^2$ (i.e., 12 electrons in the 5*f* shell and 2 electrons in the 7*s* shell), in accordance with Seaborg's analogy with the lanthanides. This experiment was performed with ^{254}Fm ($T_{1/2}$ = 3.24 h) derived as the daughter product from a special preparation of ^{254m}Es ($T_{1/2}$ = 39.3 h) from HFIR-TRU. Fermium is the heaviest element for which an electronic configuration has been experimentally determined, but Livermore researchers have proposed a new atomic beam experiment that could determine atomic configurations through lawrencium if a 40-μg ^{254}Es target becomes available.

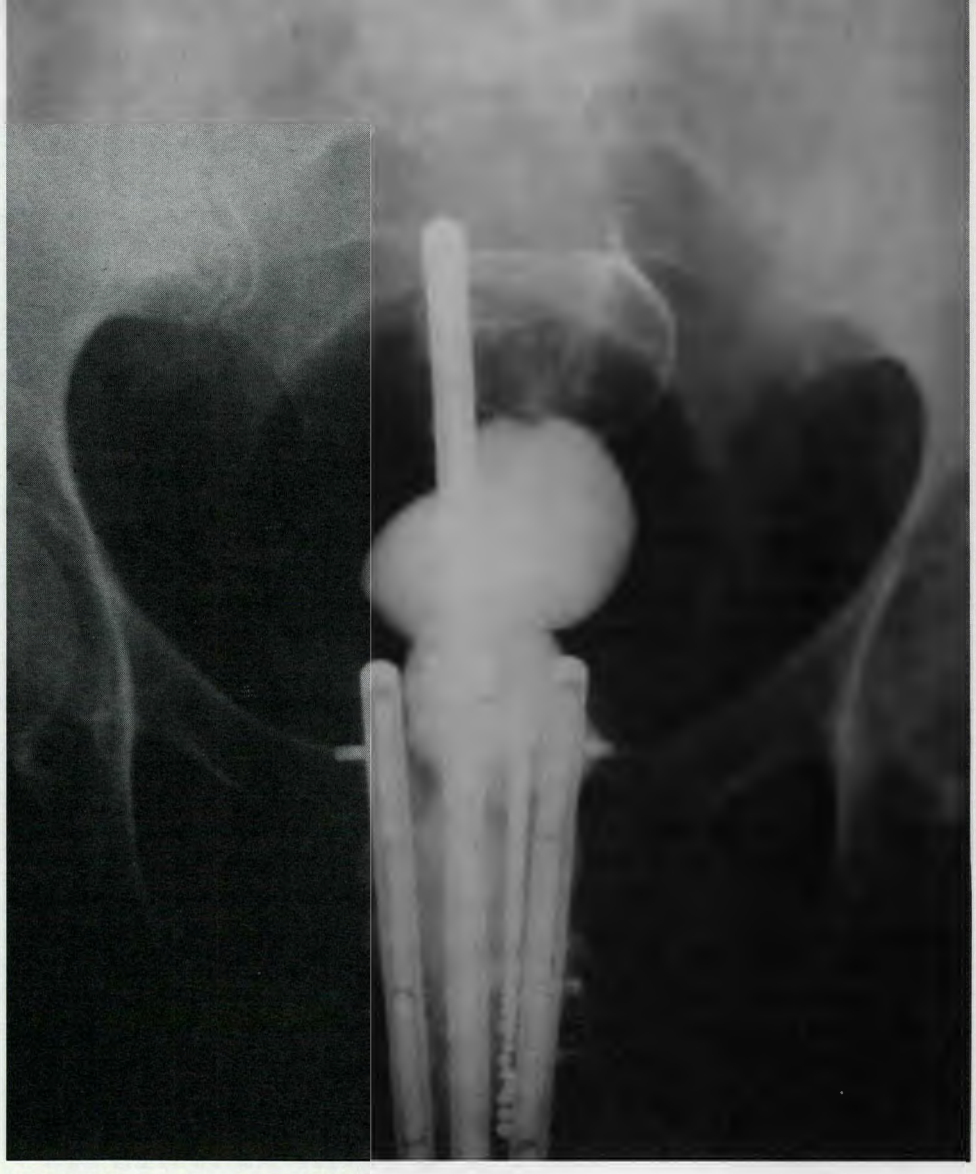
Solid-state physics. The basic problem in solid-state physics is to understand how localized atomic levels transform into the delocalized, or itinerant, extended states in a solid. In uranium and plutonium metals, the 5*f* electrons participate in the metallic bonding; these electrons are said to be itinerant, or delocalized. The actinide series presents a unique case where a class of orbitals (the

5*f* electron shell) begins as delocalized and bonding (in uranium and plutonium) and transforms into localized and nonbonding (in americium and heavier elements in the actinide series). However, in americium, curium, berkelium, and californium metals, application of high pressures can force the 5*f* electrons to participate in the bonding once again.

A quantitative understanding of these transformations will be of great interest to all of solid-state physics. As in the case of atoms and molecules, the needed theoretical development will prove formidable. It will require relativistic band calculations with electron correlation. Fortunately, theoretical work in this area is already under way in Sweden, Denmark, and the Federal Republic of Germany.

Medical applications. HFIR-TRU products are also being investigated for various applications in the medical field. The most successful is ^{252}Cf, which has been found uniquely applicable to the treatment of certain forms of advanced cancer by neutron therapy. Dr. Yosh Maruyama and co-workers at the University of Kentucky in Lexington employ ^{252}Cf neutron sources in the treatment of several forms of cancer, but mostly cancer of the uterine cervix. Of the group of patients receiving the best ^{252}Cf protocol in 1978–79, 54% have survived for five years compared with 12% for those receiving conventional therapy. These results are expected to interest more hospitals in using ^{252}Cf therapy over the next few years.

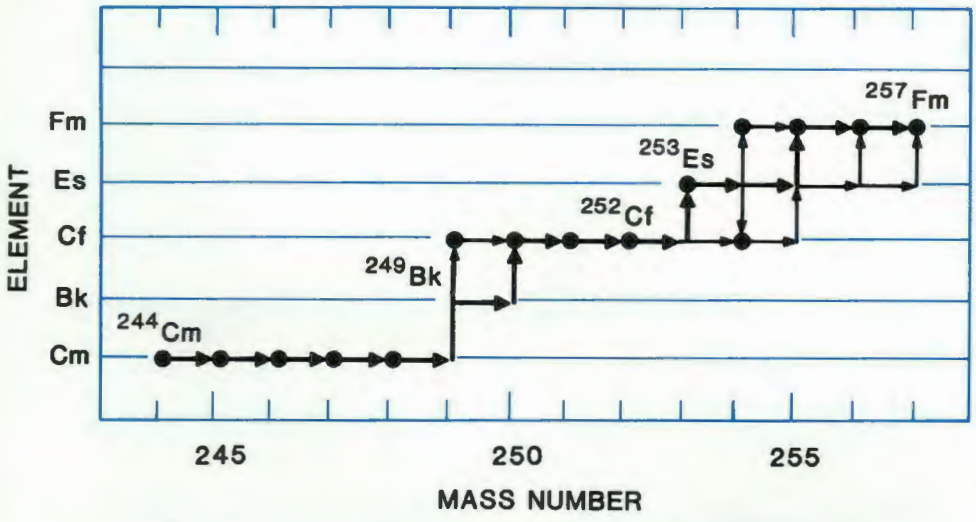
Much has been learned about chemistry, physics, and cancer therapy because of the availability of isotopes from HFIR-TRU over the last 20 years. Many new opportunities will be opened up over the next 20 years if HFIR-TRU and later CNR continue to offer a similar slate of isotopes with a special emphasis on increased ^{254}Es production.



X-ray of a female pelvis with stainless steel tubes inserted into the uterus and vaginal canal for treatment of cancer of the uterine cervix. Californium-252 sources about the width of a pencil lead and 2.5 cm (1 in.) long are inserted into the hollow tubes to irradiate the surrounding tumor with a high dose while sparing the sensitive normal tissues of the bladder and rectum. These organs are outlined here with radio-opaque dye and a plastic tube filled with lead spheres. The sources remain in the patient for about seven hours, after which the entire apparatus is removed. The best ^{252}Cf treatment protocol, used at the University of Kentucky at Lexington, has resulted in a five-year cure rate of 54% for patients having advanced cervical cancers compared with 12% for conventional therapy. ^{252}Cf is a major product of HFIR-TRU.

High Flux Isotope Reactor

HFIR produces transplutonium elements through ^{257}Fm by transmutation from curium oxide targets. The transmutation involves two processes: (1) neutron absorption, which increases the nuclear mass by 1, and (2) beta decay, which increases the atomic number (proton number) by 1. For example, curium-244 (^{244}Cm) absorbs a neutron to become ^{245}Cm. Then ^{245}Cm absorbs a neutron to become ^{246}Cm and so on up to ^{249}Cm. Neutron absorption at this point in the chain is largely replaced by beta decay because the half-life of ^{249}Cm is short enough



Path of element transmutation in HFIR, which produces transplutonium elements up through fermium-257. Neutron absorption followed by beta decay leads to transmutation of each element to a heavier one.

(64.2 min) to allow most of these nuclei to eject an electron (thus increasing the proton number) before they can capture a neutron and become ^{250}Cm. The berkelium-249 (^{249}Bk) formed by the beta decay captures a neutron and becomes ^{250}Bk, which has a half-life for beta decay of 3.2 h. The decay of ^{250}Bk leads to the formation of ^{250}Cf and so on.

The production of isotopes in HFIR stops with ^{257}Fm. The next isotope in the chain, ^{258}Fm, has a spontaneous fission half-life of only 0.38 ms. That is, the ^{258}Fm nucleus splits into two light fragments in this short time, thus eliminating the chance for neutron capture or beta decay to increase the mass or atomic number further. This ends the chain of heavy element production in the HFIR.

Operation and design. HFIR is a pressurized water reactor that uses ordinary water to cool the nuclear core and to slow down, or moderate, the neutrons, which are reflected back into the core by a beryllium reflector. The pressure vessel is located in a pool filled with light water. The fuel consists of a uranium oxide–aluminum alloy (U_3O_8-Al) containing enriched uranium-235 (^{235}U) and clad in aluminum plates. HFIR produces 100,000 kW of thermal energy. Its continuous working flux level averages 1.8×10^{15} neutrons and the unperturbed flux is 5×10^{15} neutrons $cm^{-2}\,s^{-1}$, the highest in the world.

HFIR was designed for safety and reliability. Three safety systems monitor the reactor parameters independently. If two of these systems simultaneously detect a safety shutdown condition, the reactor is shut down automatically. An extensive "on-line" system tests the response of the shutdown safety systems during operation. The reliability of and ease of human interaction with these systems have contributed greatly to HFIR's impressive operational and safety record. The amount of scheduled on-stream time since it began operating in 1966 is greater than 90%, a record unsurpassed by any other reactor in the United States and probably in the world.

Reactor core. The HFIR core consists of an annular fuel element that holds 30 target rods containing curium oxides. These targets are irradiated with a high concentration of neutrons to produce heavier isotopes. The thin fuel plates are precisely curved to prevent bending by the strong hydrodynamic forces generated as the cooling water rushes between them at a rate of about 1000 L/s (14,000 gal/min).

HFIR is a remarkable engineering achievement. The people who designed and constructed it were very successful in producing a reactor of a radically new design which operates with extreme reliability at the highest continuous neutron fluxes yet attained. Some pertinent statistics on this unique reactor are given in Table 2 on p. 56.

Transuranium Processing Plant

At ORNL's Transuranium Processing Plant, staff members (1) fabricate the target rods of curium oxide for irradiation by neutrons in HFIR, (2) chemically separate and purify the transplutonium elements produced by transmutation in these targets, and (3) package the transplutonium products and ship them to the research community and other users.

The elements to be separated in TRU from HFIR targets include transplutonium elements (actinides) such as curium, americium, berkelium, californium, einsteinium, and fermium isotopes; fission products such as the lanthanides, molybdenum, ruthenium, palladium, iodine, cesium, and barium; corrosion products such as zirconium, nickel, iron, and chromium; the target cladding, aluminum; and impurities in the aluminum such as silicon, zinc, manganese, copper, magnesium, chromium, and titanium. Because all these isotopes are highly radioactive, most chemical manipulations must be carried out in hot cells. Final purification of the transplutonium elements (other than ^{252}Cf) is carried out in gloved boxes in TRU laboratories.

The separation procedures employed in TRU capitalize on

small differences that exist in the chemical behaviors of the lanthanide and actinide elements in the 3+ state. (Lanthanide elements and most actinide elements are most stable in chemical compounds if three electrons are separated from each atom to form a 3+ ion). The small differences in chemical behavior are magnified by ion-exchange and solvent-extraction procedures that automatically put the elements through a large series of separation reactions. For example, each separation in an ion-exchange column differs only infinitesimally from the previous one; but by having millions of separation reactions occurring between the top and bottom of the column, the total effect becomes enormous. Thus the well-known principle behind the laborious series of repeated fractional crystallizations of barium salts that Madame Curie used in the discovery of radium can be applied quickly and automatically by the methods of ion exchange and solvent extraction used at TRU. It is the only plant in the world capable of handling the complex chemical procedures required to separate and purify the transplutonium elements on a scale of one gram of ^{252}Cf.

The final step at TRU is to package the various transplutonium products and ship them to the end users. This step is highly specialized, too, because of the containment and shielding requirements arising from the radioactive nature of these elements.

One secret of TRU's success over 18 years of operation is the forethought that went into the planning of the facility to ensure flexibility in equipment design and in the building itself. The design has allowed new processes and new equipment to be rapidly incorporated so that TRU can take on new tasks and operate more efficiently.

Even more important to the success of TRU has been the ability of its staff members to work closely with the research community as ideas and needs change. The TRU group's ability to meet an ever-changing horizon has been a critical driving force for progress in transuranium research.

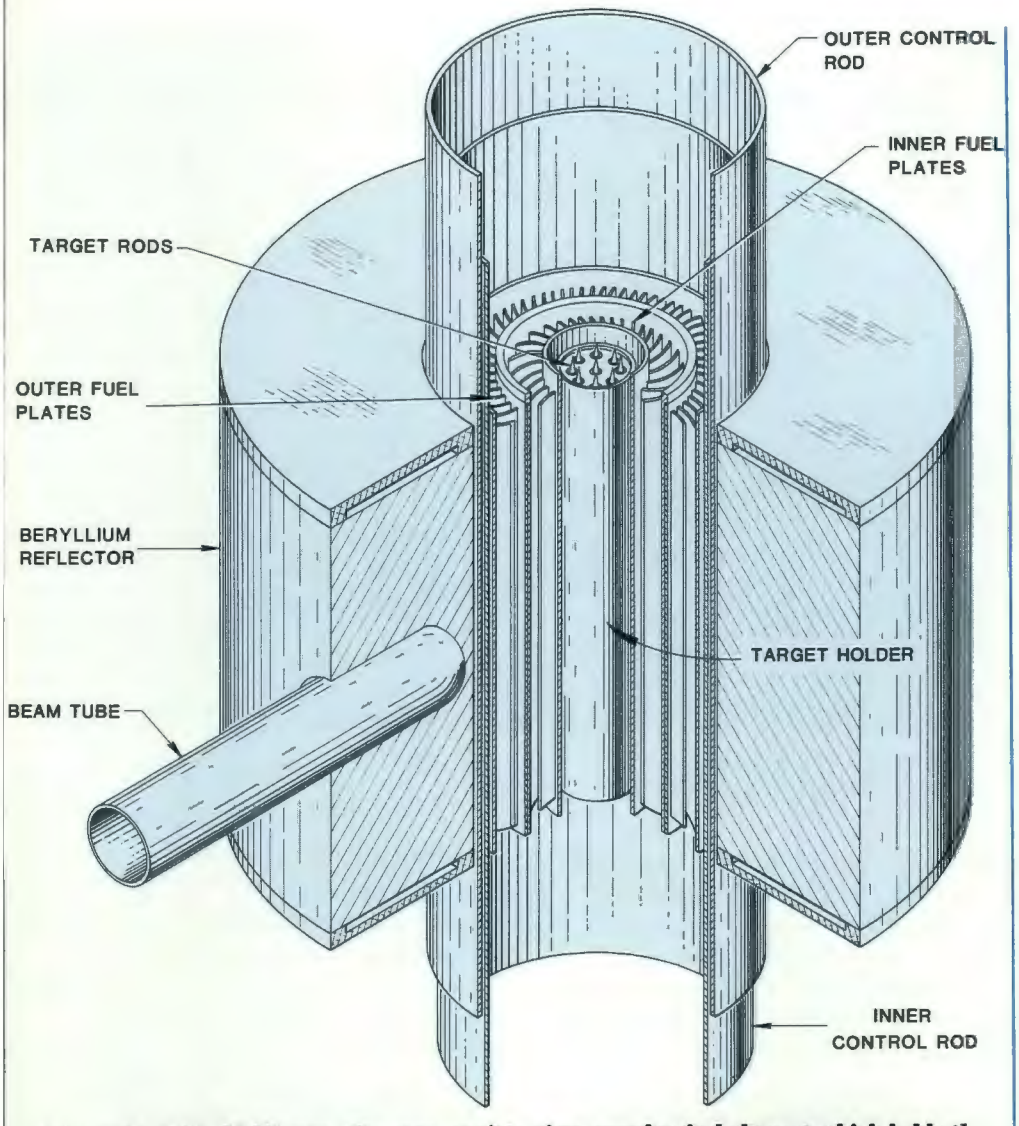
Table 2. Characteristics of the High Flux Isotope Reactor

Date of first operation	1965
Date of first operation at full power	1966
Power	100,000 kW (thermal)
Fuel	U_3O_8-Al; Al-clad, U-Al cermet
Fuel load	9.4 kg of ^{235}U
Fuel element	Two concentric annular elements each about 60 cm high; inner element: inside diameter 13 cm, outside diameter 27 cm; outer element: inside diameter 28 cm, outside diameter 45 cm. Thickness of (curved) fuel plates 1.3 mm
Reactor vessel	Carbon steel, 6.1 m high, 2.4 m diameter top located 5.2 m below the surface of the reactor pool
Control rods	Concentric cylinders; europium oxide-aluminum, tantalum-aluminum, aluminum
Moderator	Light water
Reflector	Beryllium 30 cm thick surrounded by water
Coolant	Water flow 65,000 L/min through vessel
Average power density in core	2 MW/L
Thermal neutron flux (unperturbed)	5×10^{15} neutrons/cm^2/s
Thermal neutron flux (average with targets)	1.8×10^{15} neutrons/cm^2/s
Cost	$14,718,000

Transuranium Element Research

Nuclear studies and superheavy elements. With their multiplicity of isotopes, the transuranium elements offer unique opportunities for exploring the basic physics of the nucleus—most especially that associated with exploring the limits of nuclear stability. The wide variation in spontaneous fission half-lives of the actinides implies that the balance of cohesive versus repulsive forces in these heavy nuclei must also vary widely. The nature of the proton and neutron configurations that stabilize nuclei against spontaneous fission has been of particular interest.

Spontaneous fission was discovered in uranium by Petrzhak and Flerov in the Soviet Union in 1940. The study of the mechanism of spontaneous fission was extremely difficult with isotopes of thorium, uranium, and plutonium because of their long half-lives for spontaneous fission ($T_{SF} = 10^9$ to 10^{21} years). With the discovery of the heavier transuranium elements, high specific activity sources such as ^{252}Cf ($T_{SF} = 86$ years) became available, and detailed study of the spontaneous fission process became much easier. The parameters determined included the kinetic energy distributions of the fission



Schematic of the HFIR core. The core consists of an annular fuel element which holds the 30 target rods containing curium oxides; the oxides are irradiated with the high neutron flux to produce the heavier isotopes.

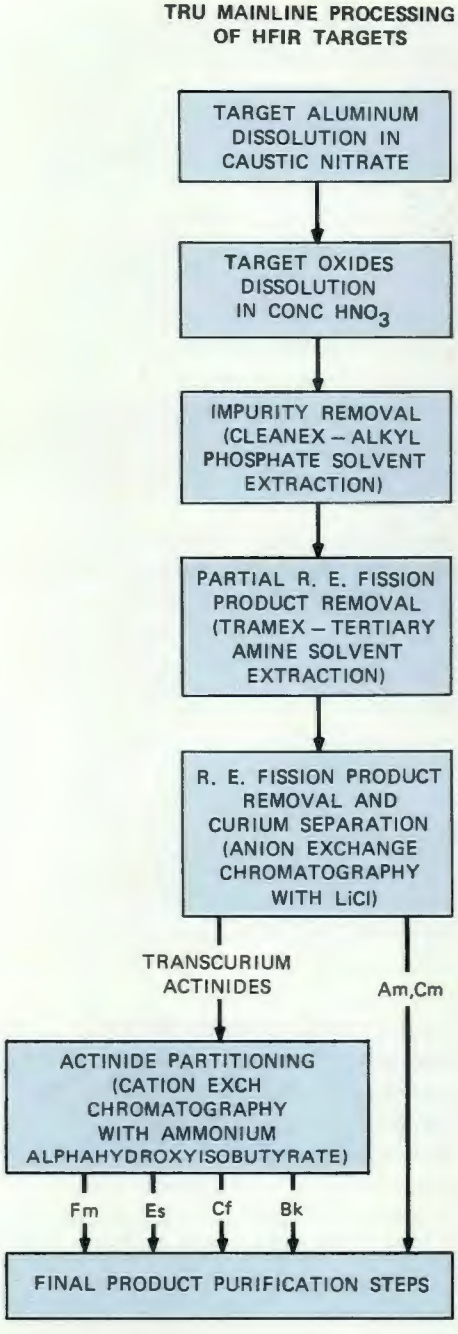
fragments as well as their mass, their nuclear charge, and the accompanying neutron and photon emission. The theoretical interpretation of such data has led to a valuable general understanding of the fission process. But more recent work has shown that spontaneous fission is a much more complicated phenomenon than previously thought.

The discovery of spontaneous fission isomers at Dubna in the Soviet Union opened a major new window on the nucleus. These isomers are nuclear matter whose shape is greatly elongated beyond that of the normal ground state. The most elegant and definitive demonstration of this distinct elongation of a nucleus made use of the fissioning isomer americium-240 ($^{240}Am^{m}$) in experiments performed at ORNL. The isomeric, elongated shape of $^{240}Am^{m}$ was compared directly to the ground state shape by spectroscopically determining the shift in electronic energy levels accompanying the change in nuclear size.

Fission isomers have motivated theorists to reach new heights of sophistication in modeling the fission process. Instead of one valley in the potential energy surface, they now see two. It is the second valley that stabilizes the elongated shape. Furthermore, the theoretical developments resulted in the idea that nuclei could be stabilized against spontaneous fission by protons and neutrons assembling into particularly favorable configurations (shells) that lower the energy of the system. Calculational codes were developed that couple the microscopic proton and neutron shell structure effects to the smoothly varying macroscopic potential energy surfaces generated in the liquid drop model. This merging of the shell model and the liquid drop model proved to be very successful and resulted in many new experimental and theoretical investigations.

Recent studies at Darmstadt, Federal Republic of Germany (FRG), and at Dubna show that nuclei in the element 107–109 region are much more stable than had been expected, although the half-lives are still quite short. Furthermore, unexpectedly, a number of the isotopes have spontaneous fission half-lives that are long enough that their predominant mode of decay is by alpha emission. So a shell stabilization against spontaneous fission of the type mentioned above has evidently come into play.

The extra stability provided by nucleonic shell effects in the 107–109 region provides a hopeful sign that a predicted island of relative nuclear stability around elements 110–120 may be discovered. Calculations indicate that these elements could be stabilized by particularly large shell effects arising from a "magic" number of 184 neutrons. If these "superheavy" elements could be synthesized, their half-lives might be in the range of years.



Outline of cell processing steps in TRU. TRU combines ion-exchange and solvent-extraction techniques to separate transplutonium (actinide) elements from lanthanide (rare-earth, or R. E.) fission products and many other elements in the HFIR-irradiated target rods. The actinide-lanthanide separation is particularly difficult because these elements are so similar chemically.

Fred Chattin at a typical cell at TRU, where highly radioactive HFIR targets are processed for removal of the transplutonium elements.

A new reaction mechanism has been discovered at Dubna that could be extremely helpful in synthesizing the superheavy elements. Researchers there observed that they could fuse certain combinations of nuclei together without imparting much excitation energy to the compound system. The low excitation energy allows more nuclei to survive the possibility of destruction by fission. This process has been given the name "cold fusion." An investigation of the cold fusion process for the asymmetric heavy target–heavy ion beam combination ^{248}Cm-^{48}Ca has been made in a German-U.S. collaboration to determine if superheavy elements can be produced in this manner. The reaction sought was

$$^{48}_{20}Ca + {}^{248}_{96}Cm \rightarrow {}^{296-x}[116]_{180-x} + xn.$$

Unfortunately, this attempt with the ^{248}Cm-^{48}Ca reaction failed to produce superheavy elements.

A target of ^{254}Es should be superior to the ^{248}Cm target, because ^{254}Es would allow a closer approach to the "magic" number of 184 neutrons required for stabilization. Also, a ^{254}Es reaction with ^{48}Ca offers another advantage: it would produce a compound nucleus with an odd number of protons (119). Nuclei with odd numbers of protons and neutrons are much more stable (less prone to spontaneous fission) than those with even numbers. The reaction would be

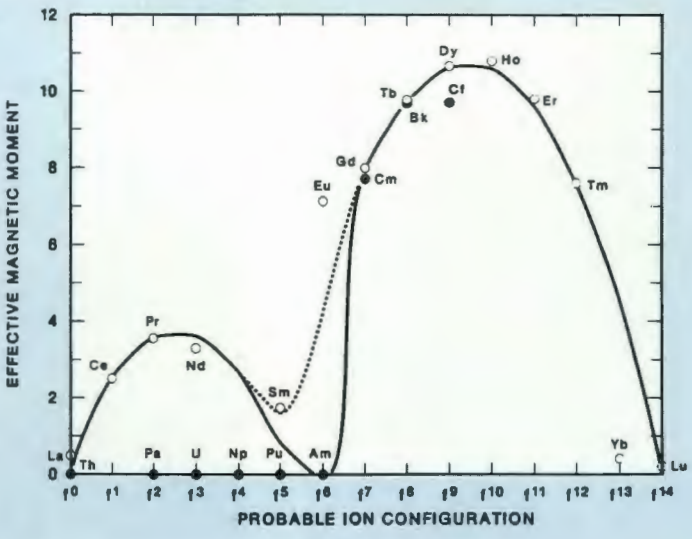
$$^{48}_{20}Ca + {}^{254}_{99}Es \rightarrow {}^{302-x}[119]_{183-x} + xn.$$

Investigations are now under way to determine the irradiation conditions in HFIR-TRU for producing a large (40-μg) target of ^{254}Es. This target would be used in heavy-ion accelerators (such as the 88-inch cyclotron at Berkeley) to produce transeinsteinium elements for investigations of their chemical, nuclear, and atomic properties. Perhaps the last, best chance to produce superheavy elements is a target of ^{255}Es ($T_{1/2}$ = 40 days). This very difficult-to-make target, in a reaction with ^{48}Ca, would give

Magnetic Moments and Electronic Behavior

A central question in solid-state physics is how electrons localized around individual atoms can become delocalized and swarm around many atoms to form metallic bonds in solids. The actinides are unique because, in the heavy part of the series, the well-defined 5*f* electrons are localized around the individual atoms in the metals, but in the light end of the series, they are delocalized into itinerant conduction electrons. The study of the physics of this transition from localization to delocalization in the actinides is of interest as a general problem in solid-state physics.

One effect of the delocalization-localization phenomenon is seen in the work of University of Tennessee researchers and co-workers at the Transuranium Research Laboratory. In individual light actinides, they have measured the magnetic moment, which reflects the strength of the magnetic field produced in the atom by its circulating electrons. The magnetic moments of light actinides such as uranium and plutonium (left) are zero. These metals cannot develop localized magnetic moments because their 5*f* electrons are moving about freely and independently in the metallic lattice and cannot assume any preferred orientation in a magnetic field. On the other hand, the lanthanides have localized 4*f* electrons. Because their electrons *can* cooperatively assume preferred orientations in a magnetic field, the lanthanide metals have finite magnetic moments.



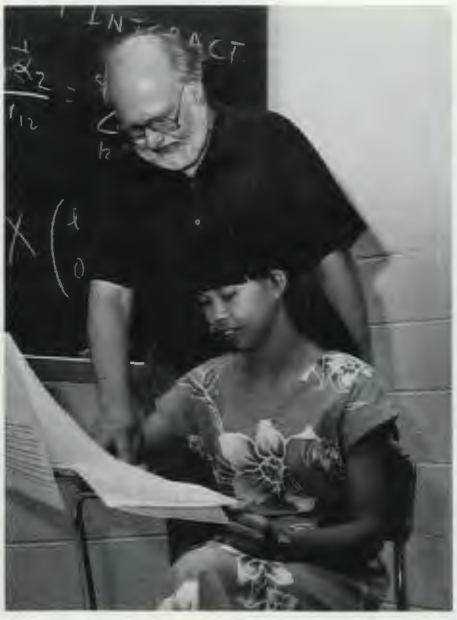
The heavy actinides curium, berkelium, and californium along the right half of the curve behave like the lanthanides because their 5*f* electrons are similarly localized and nonbonding. Thus the change in character of the magnetic moments illustrates the transition of the 5*f* electrons in the actinide series from atomic to metallic type behavior.

the 184-neutron shell in the compound nucleus thought to be needed for stability.

The discovery of relatively stable superheavy elements would open up an important new frontier of chemistry and physics comparable to that offered by the discovery of the original transuranium elements 45 years ago. The superheavy nuclei are expected to have unusual fission properties because they should be spherical rather than football-shaped like the actinides. Their chemical properties are expected to be unusual also, because their electrons will be highly relativistic (i.e., be accelerated to an appreciable fraction of the speed of light by the high nuclear charge). Chemists will then have an excellent opportunity to study changes in chemical behavior induced by relativistic effects on electronic configurations.

Chemistry and solid-state physics. In presenting the actinide concept, Seaborg predicted that the electronic configurations in the actinides would parallel those of the lanthanides. That is, just as in the lanthanides the 4*f* inner electron shell is progressively filled, so in the actinides the 5*f* shell would be progressively filled. If this were the case, the fourteen 5*f* electrons, adding from actinium (element 89), would create a half-filled shell at curium (element 96) and a filled shell at lawrencium (element 103). Such shells should have extra stability. So Seaborg reasoned that curium and lawrencium should be more lanthanide-like than the other members of the series—that is, these two members should be more stable in the oxidation state of 3+ than the others. Experimental determination of the extra chemical stability of the 3+ state of curium gave early support to the actinide concept. But just as the early members of the actinides tend toward a stability in oxidation states higher than 3+, so the heavier members tend toward an enhanced stability in the 2+ state. Against this background, the determination by Berkeley researchers that lawrencium exhibits a very stable +3 state (as expected for the last member of the actinide series) was a most important confirmation of Seaborg's formulation of the periodic table. The ^{249}Cf target used in the synthesis of the lawrencium

Joe Halperin adjusts the controls on a micromixer settler that he and Jack McDowell developed for solvent extraction chemistry of transeinsteinium elements. These elements will be synthesized on a few-atoms-at-a-time basis at the Berkeley 88-inch cyclotron using an einsteinium-254 target produced at HFIR-TRU.



Professor John Bloor and graduate student Napawan Tinpipat of the University of Tennessee discuss one of their calculations. Bloor's group is carrying out relativistic quantum mechanical calculations on atoms of the transuranium elements and their compounds.

Gerrard Payne and Jalal Mondal, postdoctoral students with Professor Joe Peterson, University of Tennessee, calibrate a microcalorimeter used to determine heats of solution of transuranium metals and compounds.

that was studied in the Berkeley experiments came from HFIR-TRU.

Furthermore, three experiments by the Berkeley, Livermore, and Dubna groups have shown that rutherfordium is definitely not a member of the actinide series. The Berkeley and Livermore experimenters used ^{248}Cm targets from HFIR-TRU. So it has been demonstrated experimentally that the actinide series ends at lawrencium. These experiments on lawrencium and rutherfordium have been difficult because only about 10 to 20 atoms of each were actually counted in the chemistry. If a 40-μg ^{254}Es target can be made at HFIR-TRU, the yield of lawrencium would be increased to about 125 atoms/min, that of rutherfordium to about 1 atom/min and that of hahnium to about 4 atoms/min. These much increased yields over those available at the time of the previous experiments would allow the exploration of a much broader range of chemistry. Of particular interest will be those chemical properties that can be linked to relativistic effects in the electronic configuratons of these elements.

As pointed out in the introduction, nobelium does not behave like its homolog ytterbium in that it is most stable in the 2+ state rather than the 3+. The difference between nobelium and ytterbium is no doubt related to differences between relativistic effects on the 5*f* and 6*d* electrons versus the 4*f* and 5*d* orbitals. Spectroscopic studies of the separation in energy (increasing differences in energy) of the electrons in the 5*f* and 6*d* shells of berkelium, californium, and einsteinium are allowing comparison of experimental results with relativistic calculations. In this way a beginning has been made for comparing the chemistry of the heavy actinides with that of the lanthanides and placing it into the framework of the Dirac theory of relativistic quantum mechanics.

The separation of energy in the 5*f* and 6*d* electron shells appears to be an important parameter for understanding the actinide series as a whole. Solid-state physics studies have focused on understanding the transition in 5*f*-electron behavior in going from plutonium to americium. Uranium and plutonium metals have 5*f* electrons that are delocalized and engage in chemical bonding. On the other hand, the 5*f* electrons in americium are found to be localized and nonbonding. But the 5*f* electrons in americium can be made to mimic those delocalized electrons in uranium by squeezing the americium metal under very high pressures! Like americium, the higher actinides also have localized 5*f* electrons. Measurements of the pressures required to make 5*f*

electrons in curium, berkelium, and californium behave in a delocalized fashion have been performed in a collaborative study by researchers from ORNL, the University of Tennessee (UT) in Knoxville, and Karlsruhe, FRG.

Transuranium Research Laboratory

The Transuranium Research Laboratory (TRL) completes the triad of transuranium facilities at Oak Ridge. It adds a research capability to the production capabilities of HFIR-TRU. In addition to having a program of its own, TRL offers scientists at American and foreign universities and research institutes an opportunity to participate in transuranium research. On the average, about half the researchers in TRL are from universities. Because of UT's proximity to ORNL, UT professors and students are especially active and have built ongoing programs of international reputation at TRL. Emphasis is in particular areas of inorganic chemistry and solid-state physics. Most of the work at TRL involves curium and the heavier elements.

To support its university and national laboratory programs, TRL has special facilities and equipment for carrying out research on very small amounts (micrograms) of the highly radioactive transuranium elements. These facilities include a Knudsen cell-mass spectrometer for high-temperature chemistry; X-ray diffraction and Raman, infrared, and ultraviolet-visible spectrometers for structure and bonding studies; inert atmosphere gloved boxes for preparing metals and compounds sensitive to air; and magnetic susceptibility apparatuses (Superconducting Quantum Interference Devices, or SQUIDS) for studies of metals and compounds. Nuclear analyzers, including an alpha particle liquid scintillation detector, are also available for detection purposes in chemical experiments. Furthermore, various microcomputers and terminals wired to mainframe computers form an excellent base for computations and analyses.

The transuranium-element researcher has at any given time several scientific areas that offer opportunities for developing new understanding of chemistry and physics. No single laboratory or country can have the equipment and background expertise needed to pursue all of these areas economically and rapidly. As a result, the transuranium area as a whole can only be developed efficiently on an international scale. Over the years UT and ORNL researchers at TRL have carried out collaborative work with scientists from the Federal Republic of Germany, the United Kingdom, France, Belgium, the Soviet Union, and a number of other countries. Depending on the circumstances, the work may be done at TRL or in the foreign country. Collaborative work is also carried out with other laboratories in the United States (especially Lawrence Berkeley, Lawrence Livermore, and Los Alamos national laboratories).

ORNL is the sole source in the Western world of heavy elements employed by the international research community. In addition, the Laboratory conducts much of its own research on the chemistry and physics of the transuranium elements in collaboration with scientists throughout the world. These researchers have opened up a number of new insights into the chemical and physical universes over the last 20 years. New prospects promise at least an equal harvest over the next 20 years. oml

Local Contributors to Transuranium Element Research

Oak Ridge National Laboratory and University of Tennessee (UT) at Knoxville researchers who contributed to investigations mentioned in the article are listed below. ORNL projects and researchers working as a team include

- Elements 104 and 105 confirmation—C. E. Bemis, P. F. Dittner, R. L. Hahn, D. C. Hensley, R. J. Silva, O. L. Keller, J. R. Tarrant, L. D. Hunt, C. D. Goodman.
- $^{240}Am^{m}$ fission isomer study—C. E. Bemis, J. R. Beene, J. P. Young, S. D. Kramer.
- Nobelium chemistry—W. J. McDowell, R. E. Meyer, R. J. Silva, G. N. Case, O. L. Keller, J. R. Tarrant, P. F. Dittner.

In the projects involving measurements of magnetic moments and effects of high pressure on metals, ORNL researcher R. G. Haire collaborated with UT researchers P. G. Huray, J. R. Peterson, S. E. Nave, and J. R. Moore.

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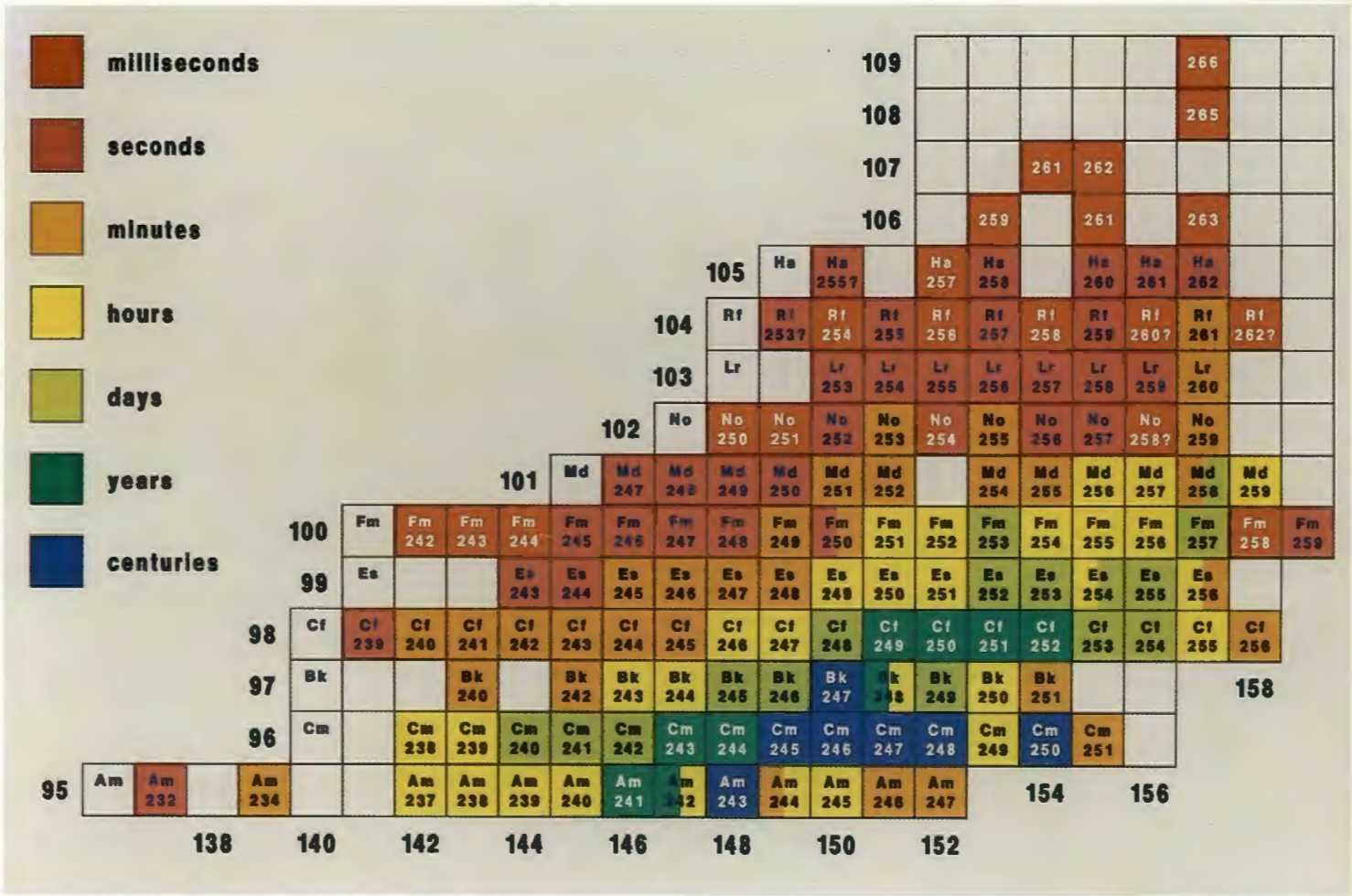

The chart of the transplutonium nuclides.