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TECHNICAL DIVISION

Section IV, Engineering Materials  
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AIR CONTAMINATION AND ATTENDANT METAL LOSSES  
DURING PRODUCTION OF URANIUM-ALUMINUM ALLOYS

BY

G. M. ADAMSON

AEC RESEARCH AND DEVELOPMENT REPORT

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TECHNICAL DIVISION

Section IV, Engineering Materials

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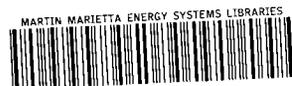
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I. ABSTRACT

An investigation of the foundry air contamination and resulting uranium losses during the production and casting of U-Al alloys is reported. Tests were made using both enriched and natural uranium. Data show that although actual uranium losses are low, if they are not properly controlled, they can cause serious air contamination. A simple but adequate ventilating system is shown.

II. INTRODUCTION

The method of producing U-Al alloys at ORNL consists of the reduction of  $U_3O_8$  by molten aluminum in the presence of cryolite. The pig produced in this operation is then remelted without a flux and cast into an ingot. In this process the uranium can be accounted for as follows:

- (1) alloy ingot
- (2) slag, dross and crucible losses
- (3) air losses.

This report is an investigation of the magnitude and effects of item three. These losses are of interest both from the standpoint of air contamination and uranium accountability. The effects of both of these factors are magnified when enriched material is handled. A suitable exhaust and collection system would accommodate both of them.

During early work on the U-Al alloys Health Physics made several air tests and found no activity. When a large batch of  $U^{235}$  was reduced and cast it was thought advisable to obtain a record of air contamination and uranium air losses. The first sample was found to be well below tolerance; however, a check sample and several taken subsequently gave values considerably above tolerance level.

In light of these results an investigation was made to determine: (1) safe working conditions for the reduction of  $U^{235}$  oxide to U-Al alloy and (2) uranium losses. Since a limited amount of  $U^{235}$  is available for such tests natural uranium was substituted.

### III. PROCEDURE AND EQUIPMENT

In these tests the  $U_3O_8$  was mixed with cryolite in a weight ratio of 1:4 before being charged. The mix was added to molten aluminum either by being poured from a bottle into the crucible or by being added while held in aluminum foil cans. In either case the mix floated on the aluminum. There was no difference in ingot uranium recovery between the two methods. The time required for the charging and melting was between fifteen and twenty-five minutes depending upon the technique used by the operator. After melting, the mix was held at temperature ( $1150^{\circ}C$ ) for thirty minutes with frequent stirring. During this interval the alloy was covered with a six to eight inch layer of molten slag.

Remelting is a simple operation consisting of the following steps: (1) melt the pig at  $825^{\circ}C$ ; (2) hold at temperature for ten minutes; (3) dross to remove oxide and other light impurities; (4) pour into an ingot mold. With natural uranium all tests were made on heats producing 1500 grams of 25% uranium alloy. The conditions for the enriched heats varied and are listed individually.

Figure I shows the hood and its relationship to the coil and crucible. The photograph was made after the investigation was completed and shows the sides and top that were not part of the hood

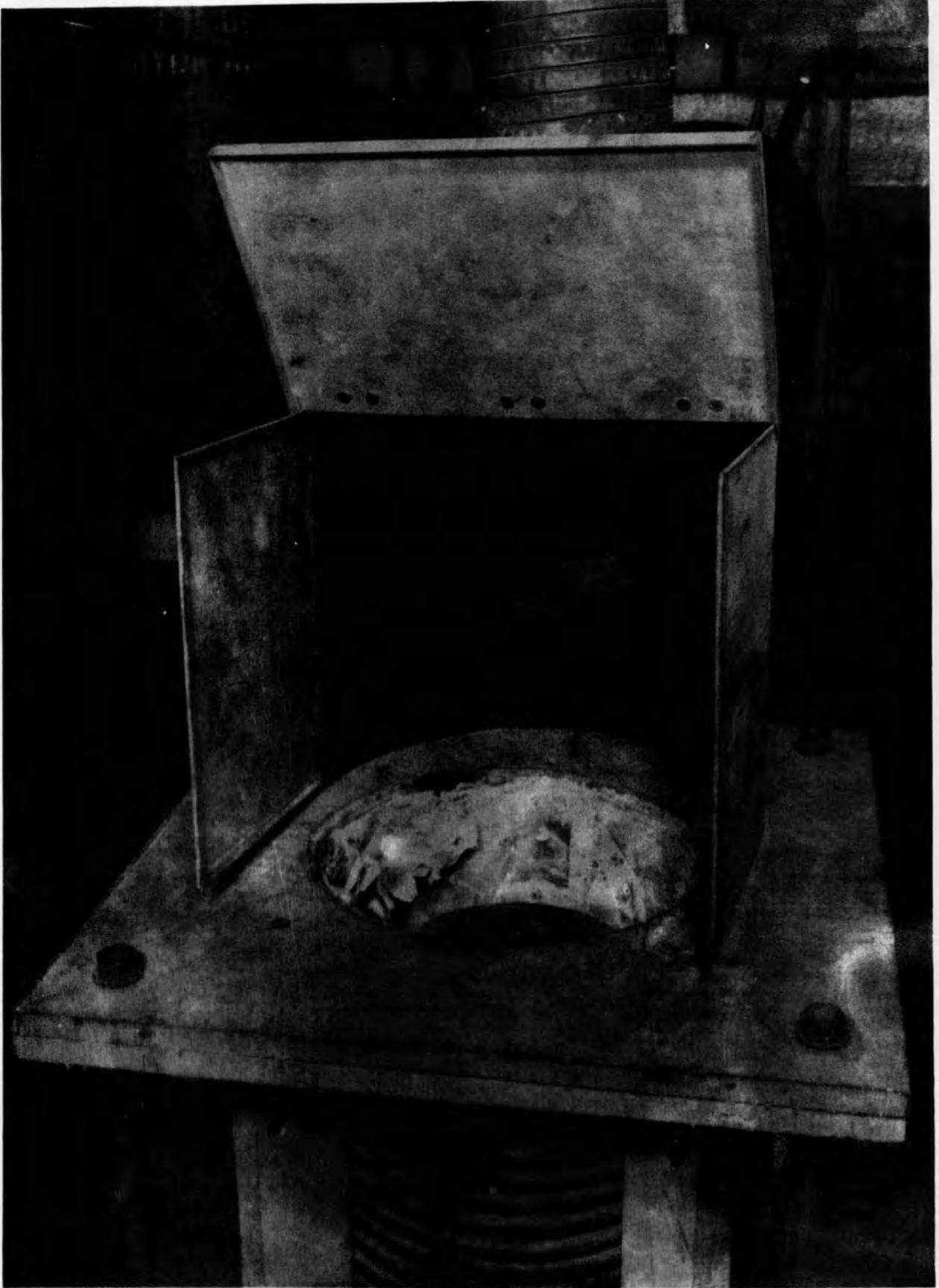


FIG. 1  
FURNACE HOOD

during these tests. The face of the hood is nine inches across and eight inches high. It is connected to the fan by means of a four inch flexible pipe. The blower is a Trane air blower model B-5852 and is powered by a one-half horsepower, 1725 RPM motor with a 1:2 pulley ratio. The capacity of this system is 166 c.f.m. In addition a small window fan was in operation continually.

All air sampling was by means of Precipitrons. The sample tubes were located directly above the crucible and/or approximately five feet out into the room. The length of sampling time and the exact location of the sample tube are given in the Tables. After a twenty-four decay period, the samples were counted by the Health Physics Division.

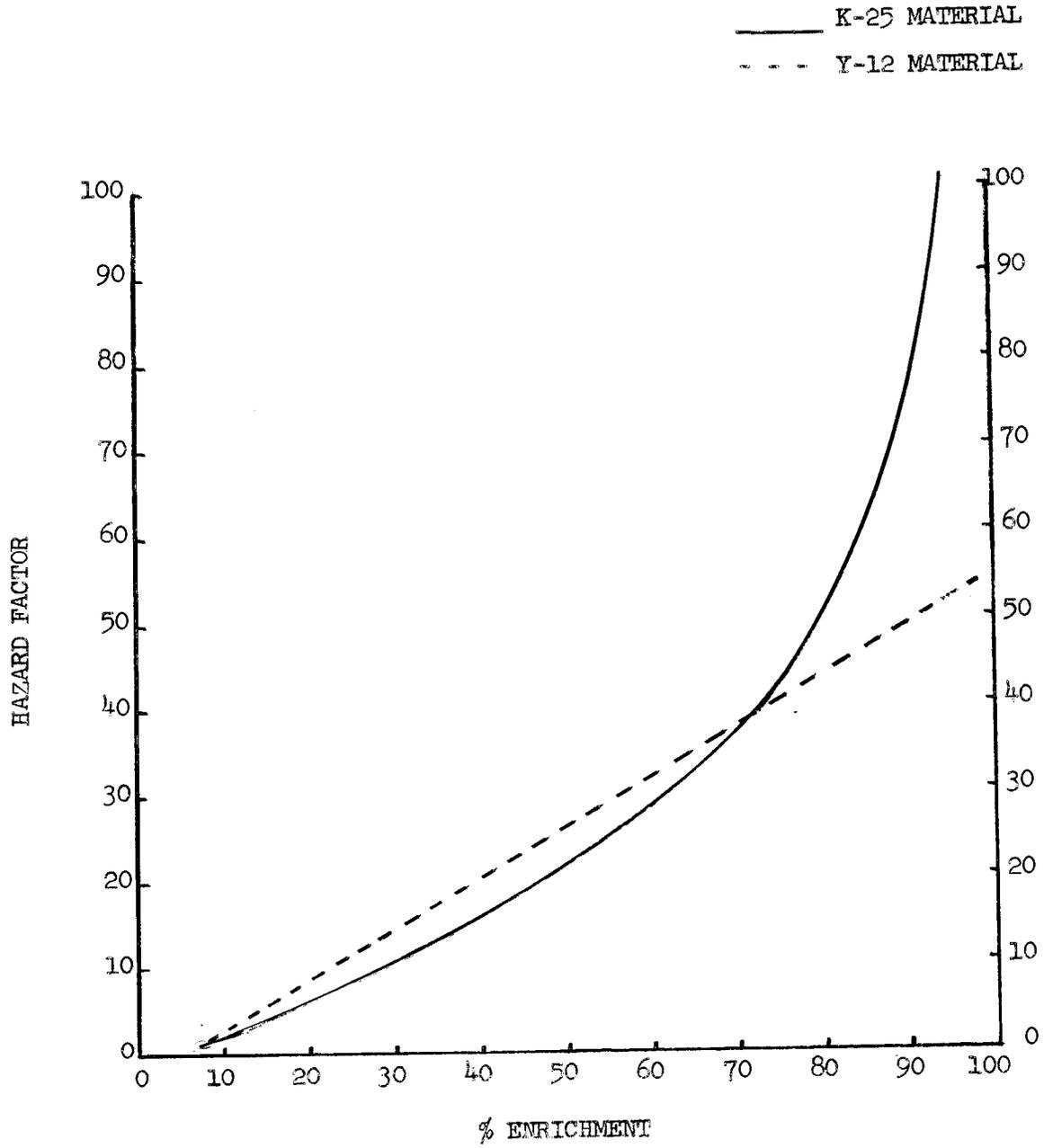
Both types of  $U_3O_8$  were high purity material in the form of fine dry powder and were obtained from the Atomic Energy Commission or the Manhattan District. No information on particle size is available for either material.

#### IV. DATA AND DISCUSSION

##### A. Air Contamination.

This section of this report has been divided into two parts: (a) tests during natural heats and (b) tests during enriched heats. The data for the first section are plotted in Table I and for the second in Table II. For both of these sections the continuous exposure tolerance level, above which a mask is required, is taken as  $3.1 \times 10^{-11} \mu/\text{cc}^*$  for long-life alpha.<sup>(1)</sup> It should be pointed out

\*  $\mu/\text{cc}$  - Microcuries/cubic centimeter of air tested.



HAZARD FACTORS FOR ENRICHED URANIUM

FIGURE 2

that while values below this are considered acceptable they are not desirable, as any level of activity is to be avoided if at all possible. Since the number of enriched heats is limited, it is not practical to conduct an extensive air contamination study with this material. To correlate the data obtained with the natural heats to what would be expected with enriched material, Health Physics furnished data<sup>(1)</sup> to plot the curves shown in Figure II. The values are only approximations but are sufficiently accurate for this work. The hazard factors show how many times more dangerous than natural uranium a material of a given enrichment is. Since the processes at K-25 and Y-12 affect the concentration of  $U^{234}$  differently, the hazard factors will vary.

a. Natural Uranium Tests

An examination of Table I shows that the reduction of  $U_3O_8$  in an open furnace is inadvisable even with natural uranium. Adding the mix in aluminum cans results in some improvement but the counts although below tolerance are still very high. Comparing these results indicates that while some of the activity results from the pouring, most of it is carried out of the crucible by the gases and vapors rising from the bath. When the tests were repeated with the hood operating, both methods gave counts well below tolerance level.

Section C of Table I proves that most of the activity is carried from the bath before the mix has melted. It is evident that the escaping gases and vapors entrain small particles of the  $U_3O_8$  while rising through

TABLE I

AIR COUNTS FROM PROCESSING NATURAL URANIUM

Test	Sample Tube Location	Operation	Fan off			Fan on		
			Air Count	Average	Corrected Average	Corrected Average	Average	Air Count
A	1 ft. above crucible	Bottle charging	3.94	3.80	3.57	0.0	0.22	0.22
	"	" "	3.66					
B	"	Can charging	3.91	2.84	2.61	0.04	0.27	0.27
	"	" "	1.94					
	"	" "	2.94					
	"	" "	2.58					
C	"	Stirring	0.39	0.50	0.27	0.10	0.33	0.50
	"	"	0.63					0.50
	"	"						0.20
	"	"						0.14
D	"	Remelting	0.33	0.33	0.10			
E	5 ft. away 6 in. below	Entire heat	0.38	0.26	0.03	-0.16	0.07	0.03
	"	" "	0.32					0.12
	"	" "	0.07					
F	Inside exhaust funnel	Can charging				0.74	0.97	1.25
G	Inside Stack Outlet	Bottle charging				1.97	2.20	2.20
H	In main rm. of bldg.	None	0.11		0.23			
	"	"	0.25					
	"	"	0.33					

the unmelted mix. During the stirring or holding period of the reduction heats, very little activity was detected even with the hood fan off. The first two comparatively high results in this section, measured with the hood on, were caused by activity remaining in the room from heats that were charged with the hood off. Even when these results are included, the average is still very low either with or without the hood in operation.

Conditions during a remelt heat are similar to those in the stirring period of a reduction heat. In a reduction heat the bath is protected by a thick layer of slag while in the remelt heats the protection is achieved by an oxide layer; also, in neither case is there any powdered oxide present. For this reason, similar low counts would be expected and Section D confirms this. Only one remelt heat was tested.

Even when activities well above tolerance are measured near the crucible, the window fan and regular ventilation system keeps the major portion of the room well below the danger point. This is confirmed by the results tabulated in Section E. With the furnace hood off, the average value at a point five feet from the crucible is less than 10% of tolerance over a one hour period. To cut down on the number of samples being counted, the room counts were taken for an entire heat rather than for individual operations. However, these values are so low it does not appear likely that they exceeded the tolerance value even for a short interval.

Three tests were made to determine the long-life alpha background activity. From the three individual values reported in Section H, the average background was found to be  $0.23 \times 10^{-11}$   $\mu$ /cc. Two of the samples were taken in the main room shortly after runs in the foundry but the third ( $0.25 \times 10^{-11}$   $\mu$ /cc) was taken in the foundry on a Monday morning before any material was handled. Since an air current passes from the main room into the foundry, the foundry operation should not affect tests in the main room. This average value ( $0.23 \times 10^{-11}$   $\mu$ /cc) has been subtracted from the averages for each test to give the corrected averages as listed in Table I.

If one were to attempt to correlate a natural uranium value to one that would be obtained with K-25 93% enriched material, it is evident from Figure 2 that multiplying by a factor of almost 100 is necessary. From this it is evident that the background alone is enough to produce values above tolerance when converted to highly enriched material. To make matters worse, the difference between the extreme background figures ( $0.22 \times 10^{-11}$   $\mu$ /cc) is also enough to produce errors larger than the tolerance value. For natural material a value below  $0.03 \times 10^{-11}$   $\mu$ /cc would be necessary before it could be converted and give a value below tolerance for highly enriched material. An examination of the individual values making up the averages in Table I shows variations much larger than this figure.

From the foregoing discussion it is evident that tests made with natural material will give only approximations of what can be expected with enriched materials. It is possible to establish the most desirable conditions but it is not possible to predict exact air contamination values. When the background values are subtracted from the averages in Table I the results obtained when the hood is in operation are at least of the proper order of magnitude for safety. A conclusive figure for air contamination with enriched material can be determined only by using the enriched material itself.

b. Enriched Uranium Tests.

The results tabulated in Table II are from tests made during the processing of K-25 enriched material of approximately 93% enrichment. These results instigated this investigation.

Of the tests made during the reduction operations (1-6) all except the first produced counts well above the tolerance level. The tests during this series were taken for thirty minute intervals rather than being timed for a single operation. As a result they include some stirring time in addition to the charging and melting periods during which the contamination reaches a maximum. At the request of the Atomic Energy Commission the hood was turned off during charging which consisted of from fifteen to twenty-five minutes of the thirty minute test. From the results presented in Table I it is now evident that this is the interval during which the air contamination reaches a maximum. For purposes of comparison

TABLE II

AIR COUNTS FROM PROCESSING ENRICHED URANIUM

Test	Sample Tube Location, inches above crucible	Melt, % U.	Heat size gm.	Fan Operation	Operation	M/cc 10 <sup>-11</sup>
1	18	18	1000	After charging	Charging by Pouring	0.2
2	18	18	1000	" "	" " "	41.8
3	18	18	1000	" "	" " "	39.8
4	18	18	1000	Entire heat - hood 6 in. to side during charging	" " "	5.2
5	12	30	800	Entire heat - hood 6 in. above crucible	Charging by cans	15.8
6	6	18	1000	After charging	Charging by Pouring	77.2
7	12	40	150	Entire heat	Entire heat	1.3
8	12	30	800	Entire heat - hood 6 in. above crucible	Stirring	1.6
9	18	4.5	2500	Entire heat	Remelting	0.7
10	"	"	"	" "	"	0.0

these heats should be considered as being made with the fan off. In test 4, the hood was left on, but lost most of its effectiveness by being moved six inches to one side of the crucible. In this operation there was only a slight draft over the crucible but a large drop in air contamination was found. Test 5 was made with the fan on, but the bottom of the hood was six inches above the crucible top. This test was taken only during charging and melting. In spite of the fact that this test did not include any stirring time and that vapor could be seen blowing into the room from the six inch space, the test gave a result much lower than the first three which were taken on similar heats.

Test 7 was on a heat much smaller in size than the others but because of this almost the entire stirring period could be included in the 30 minute interval. To compensate partially for the size difference, the sample tube was moved closer to the crucible. This was the only enriched test in which the hood was operating in the proper position and it was the only one that produced a contamination below tolerance level.

As far as safe working conditions are concerned, tests 4, 5, and 7 present a much more optimistic and representative view point than the others. While no definite conclusions can be drawn, the indications are that with the hood operating in the proper position for an entire heat, enriched material can be handled safely. Test 4 produced a value only slightly above tolerance level even with the hood only partially

effective. Tests 4 and 5 which were made with only partial hood use showed values well below those recorded for similar heats made without the hood in operation. Test 7 gave a value below tolerance.

The last three heats which were made during either stirring or remelting operations confirm the previous conclusion that most of the activity is carried out before the powdered mix has melted. Test 8 was made on the same heat and immediately following test 5, but the air activity had fallen by a factor of ten. Both of the tests made during remelting gave very low values. These values are not comparable to the others since the hood was in use during the entire operation.

B. Material Losses.

From the preceding sections of this discussion it is apparent that some uranium is being carried out of the crucible. When enriched material is handled, the magnitude of these losses is quite important.

Since no corresponding air volume or velocity figures could be determined, the air counts discussed so far cannot be converted into actual uranium losses. The easiest place to determine both of these sets of figures was in the exhaust stack. The velocity in the stack was measured by Health Physics and reported<sup>(2)</sup> as being 166 cubic feet per minute. Owing to the differences in draft between the hood and the sample tube, difficulty was encountered in obtaining representative samples. At the inlet the stack draft would be opposing the sample tube suction; therefore, the sample would be smaller than usual, and low results would be obtained. At the top of the stack the effect

would be reversed but to a smaller extent. The results of the samples taken at these points are tabulated in Sections F and G of Table I.

For purposes of calculation the corrected value obtained at the top of the stack ( $2.0 \times 10^{-11}$   $\mu$ /cc) was taken as being correct. It may be a little high owing to the stack draft increasing the sample size but this is partially compensated by including some of the stirring period with its lower losses. The samples taken at the top are slightly higher and would therefore give the maximum loss. To convert the air counts into actual uranium losses a value of  $1.47 \times 10^6$  grams of uranium per curie<sup>(3)</sup> was used. The following is the calculation for determining the uranium losses during a fifteen minute charging and melting period:

$$2.0 \times 10^{-11} \mu/\text{cc} \times 1.47 \times 10^3 \text{ mg}/\mu \times 166 \text{ c.f.m.} \times 28340 \text{ cc/c.f.} \\ \times 15 \text{ min.} = 2.1 \text{ milligrams of uranium.}$$

No samples were taken from the hood system during stirring. Section B and C of Table I show that the air count with the fan off, drops from 2.84  $\mu$ /cc during charging to 0.50  $\mu$ /cc during stirring. The ratio between these figures ( $\frac{0.50}{2.84}$ ) should be an indication of the relative amounts being lost during the two periods. A simple calculation then shows that the approximate loss during the thirty minute stirring period should be as follows:

$$2.1 \times \frac{0.50}{2.84} \times \frac{30 \text{ min.}}{15 \text{ min.}} = 0.7 \text{ milligrams of uranium.}$$

For an entire reduction heat producing 1500 grams of 25% alloy, the uranium carried out into the air would be the sum of the above two figures or about three milligrams. This figure cannot be taken as an exact value but it is at least of the proper order of magnitude and appears to be on the high side. The loss during a remelt heat would be about the same as during stirring or about one milligram.

V. CONCLUSIONS.

From this work it appears that both natural and enriched material may be reduced to form uranium aluminum alloy without exceeding tolerance levels. This can be accomplished using only very simple hood facilities. It is possible that counts slightly above tolerance may be found during the charging and melting period but this time could be held to a maximum of fifteen minutes. It would not be necessary for the operator to be near the furnace for this entire period. With only two reduction heats per day even if this brief high period exists it would not be serious. The air contamination will be reduced further by extending the sides of the hood to eliminate cross drafts as shown in Figure 1. During the stirring of reduction heats and remelting of the alloy, no difficulty from air contamination should be found even with highly enriched material.

Although difficulty was encountered with air contamination, the actual uranium losses are very low. The loss during a reduction heat being approximately three milligrams and during a remelt heat one milligram. A filter is being installed in the exhaust system to collect these small amounts.

VI. ACKNOWLEDGEMENTS

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C. F. Cutcher - furnace operation

F. Kerze - valuable aid in the preparation of this report.

VII. REFERENCES

(1) Memo from H. J. Burnett to J. C. Hart dated July 15, 1948.

ORNL Central Files Number 48-7-196.

(2) Memo from W. H. Baumann to G. M. Adamson dated July 30, 1948.

(3) Verbally from R. L. Clark.