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REDUCTION OF BETA ACTIVITY FROM DEPLETED DERBIES, INGOTS AND CRUCIBLES

By

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ABSTRACT

The reduction of beta radiation on uranium ingot and crucible surfaces was demonstrated in the production casting operation by adding a mixture of slag liner material (MgF_2) and calcium fluoride to the remelt charge. The beta emitters (Th_{234} and Pa_{234}) are largely discharged with the fluorides into drums during a remote crucible burnout operation; thereby, reducing operator exposure to beta radiation.

A production test showed that very low beta radiation from uranium flat castings can be achieved by using derbies recently prepared by reduction.

Plant tests with fluoride addition indicate that pickling of derbies may not be necessary for casting uranium flats from a plasma sprayed (ZrO_2) crucible. Also, ingots produced with fluoride additions had less pipe as compared to standard production technique.

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References

[1] D. J. Allard et al., Beta Dosimetry Experiences at a Depleted Uranium Metal Fabrication Facility, Nuclear Metals, Inc., Concord, Massachusetts (February 1983).

[2] A. Travesi, F. de la Cruz, and R. F. Cellini, The Composition of the Solids in the Smoke Released in the Calciothermic Process for the Manufacture of Uranium, Vol 4, Peaceful Uses of Atomic Energy, 1958 (pp 93-100).

INTRODUCTION

This report covers the most recent work that has been done with the aim of reducing the levels of radiation from newly cast uranium ingot surfaces and also from the interiors of the graphite crucibles in which the ingot charges are melted. The problem is of considerable importance because the radiation levels now encountered in the casting area are sufficiently severe to require the rotation of personnel at five different work stations where tasks bring them into proximity with the ingots and crucibles. Under present conditions these personnel are limited to only fifteen operating shifts per quarter year at the stations in question. Moreover, they are not allowed to work more than five shifts per month before they must be transferred to tasks with much lower exposures. The exposure problem has recently become more burdensome because operations have expanded to three-shift coverage and a seven-day work week. These restrictions affect a large percentage of the total casting plant work force and necessitate a large amount of administrative control.

This report will be devoted to what appears to be the most promising method explored to date for reducing the radiation emitted from the cylindrical ingots and their melting crucibles. The method involves the addition of a few pounds of a mixture of magnesium and calcium fluorides to each casting charge. The data have been gathered primarily on the casting of 38 ingots of the Mark 31B type (10-inch). A small body of data have been obtained on the effects of adding the fluorides to "flat" casting charges. Another very promising method for reducing radiation from flats is also discussed briefly.

NATURE OF THE RADIATION ASSOCIATED WITH THE CASTING PROCESS

A phenomenon associated with our castings of uranium is that the surfaces of the ingots, after stripping from the molds, exhibit rather high radiation levels which are many times greater than that emitted from a cut face of the same ingot. This radiation contains alpha particle, beta particle, and gamma components, but it is the strong beta component that is of greatest concern. This beta radiation penetrates several feet of air and constitutes a skin dose hazard. We now recognize that the radioactivity stems from the decay daughters of U_{238} as outlined below:

$U_{238} \rightarrow Th_{234} + \alpha$, Half-life, $T_{\frac{1}{2}} = 4.68 \times 10^{9}$ years

$Th_{234} \rightarrow Pa_{234} + \bar{\beta}$ $T_{\frac{1}{2}} = 24.1$ days

$Pa_{234} \rightarrow U_{234} + \bar{\beta}$ $T_{\frac{1}{2}}$ (98.84%) $= 1.17$ minutes

$U_{234} \rightarrow Th_{230} + \alpha$ $T_{\frac{1}{2}} = 2.445 \times 10^{5}$ years

In the partial decay scheme shown above, it is the decay of the Pa_{234} that yields very energetic beta particles (electrons) of 2.28 MEV maximum energy and which contributes dose at depths greater than 7 mg/cm^2 (i.e., typical skin thickness above dividing cells at risk). Reference [1].

A feature of the radiation problem encountered in the casting process is that the radioactive daughter product Th_{234} largely separates from the bulk of the molten metal and becomes highly concentrated in a thin layer on the surface of the solidified ingot. It is this strong separation of the Th_{234} out of uniform solution in the bulk of the metal that gives rise to the high beta field in the air surrounding the ingot. If the minute amount of Th_{234} had remained uniformly dissolved throughout the volume of the ingot, the radiation problem would not exist. In such a case the Th_{234} decays to Pa_{234} and this in turn ejects high energy beta particle electrons within the ingot, the fast moving beta electrons will be stopped by a very thin thickness of the dense uranium. The rapid slowing down of the beta electrons will produce "braking" or bremsstrahlung x-rays which in turn will be largely absorbed before reaching the surface of the ingot.

The separated and concentrated Th_{234} at the ingot surface constitutes what is called an "unsupported" daughter product activity. (It is "unsupported" because the ratio of daughter Th_{234} to parent U_{238} atoms in the thin surface layer is many times greater than that corresponding to secular equilibrium between parent and daughter.) The specific activity of the surface radiation is greatest immediately after casting at the time that the ingot is stripped from the mold and sawed to remove the top crop. The surface radiation will diminish with time as controlled by the half-life decay time of 24.1 days for Th_{234}. Conversely, the cut faces of the ingot will be lowest in specific activity immediately after casting because the ratio of Th_{234} to U_{238} atoms within the metal is now less than that corresponding to secular equilibrium. The radiation from a cut face will rise with time in accordance with a 24.1 day half life as Th_{234} grows back in to reach the concentration corresponding to secular equilibrium. In about five months the radiation from the ingot surface should die down until it equals that from the cut face.

Perhaps the most curious and least well understood feature of the radiation from the ingots is its variation over the ingot surface. Invariably, the highest readings ($\beta + \gamma$) are found at the flat bottoms of the cylindrical ingots where the metal solidifies most rapidly against the mold cup. The readings generally taper off from the bottom to top of the cylindrical surface although this variation may be quite irregular. Radiation readings from the tops of the uncropped ingots tend to be higher than those from the uppermost side surface, but are also quite variable.

TECHNICAL CONSIDERATIONS LEADING TO THE TESTING OF MgF_2 - CaF_2 ADDITIONS IN CASTING MELTS

When uranium is purified by chemical separation methods such as precipitation or solvent extraction, virtually all of the products of radioactive decay that are present will be stripped away from the uranium. An exception will be the U_{234} isotope which appears in the decay chain of U_{238}. However, if the uranium has also been through the gaseous diffusion process (cascades), the U_{234} will also have been largely removed along with the U_{235} isotope. As time passes following the purification, the concentrations of daughter products Th_{234} and Pa_{234} will grow back into the uranium metal or uranium compound. Secular equilibrium will be essentially reestablished for these two isotopes in about five months. The equilibrium will be reestablished as determined by the half-life decay time of the Th_{234} which is 24.1 days. After six half-lives or about 4.7 months, the Th_{234} (and Pa_{234}) will have "grown in" to reach 63/64 times their maximum concentrations finally reached at secular equilibrium. At secular equilibrium, the numbers of U_{238}, Th_{234}, and Pa_{234} atoms undergoing decay per unit time will have become equal. Since the half-lives of Th_{234} and Pa_{234} are very short compared to the half-life of U_{238}, the actual concentrations of the two daughter products in the uranium or uranium compound will be very small. The atomic ratios of the U_{238}, Th_{234} and Pa_{234} will be proportional to the ratios of their half-lives. We can calculate, for example, that 1400 pounds of uranium (the weight of uranium in a Mark 31B ingot) should contain 9.2 micrograms of Th_{234} and 310 micro-micrograms of Pa_{234} at secular equilibrium. Because the half-life decay time of the uranium isotope U_{234} is very long, secular equilibrium between this isotope and U_{238} will not be reestablished for several hundred thousand years.

Depleted UF_4 which has been in storage for several years will thus always contain highly radioactive Th_{234} and Pa_{234} in secular equilibrium, and these elements will be uniformly distributed throughout the UF_4 in solid solution.

The key to the development of successful methods of reducing the radiation associated with the casting process came with the realization that the troublesome daughter product Th_{234} (and hence also the Pa_{234}) is largely removed temporarily from uranium metal during the bomb reduction process in which the UF_4 is reduced to metal by reaction with magnesium. This phenomenon was noticed and investigated by Spanish workers several years ago in connection with the similar calciothermic reduction of UF_4 to metal. [Reference 2.] In the reduction process, a two-phase liquid-liquid system is produced in which the radioactive daughter products are largely transferred from the liquid uranium phase to the molten MgF_2 slag phase. The separation process can be viewed as quite similar in effect to a single-stage liquid-liquid solvent extraction process. With the realization that Th_{234} could be temporarily reduced in amount by contacting the molten metal with a molten salt phase, consideration was given to the possibility of applying this general scheme to the casting process. The derbies which have been prepared and stored for periods of a few months before being consumed in casting will generally contain the Th_{234} grown back in. The same will be true of the recycled solid scrap and briquetted chips which comprise a large fraction of the total charges going into the cylindrical ingots. It appeared possible that a relatively small addition of MgF_2 or a mixture of MgF_2 and CaF_2 to the melting charges in the crucibles might be quite effective in scavenging the Th_{234} from the molten metal into the molten salt phase. It was also reasoned that the mixture of the two fluorides should be more effective as a scavenging agent in the casting process than either MgF_2, or CaF_2 alone. The melting points of pure uranium, MgF_2, and CaF_2 are respectively 1133°C, 1263°C, and 1380°C. Thus, on heating a uranium metal charge with either of the pure fluorides, the fluoride phase will not melt until the uranium metal has been superheated to temperatures well above its melting point. The salt phase probably cannot act as a very efficient scavenging agent until it has melted. However, the binary system $MgF_2 - CaF_2$ contains a deep eutectic melting at about 940°C. The eutectic composition is at 52 w/o CaF_2. Any mixture of the two fluorides lying between about 16 and 75 w/o CaF_2 should begin to melt at 940°C and be completely molten at the melting point of uranium metal.

Thought has been given to possible mechanisms to explain in greater detail (1) how Th_{234} originally present as thorium metal atoms uniformly dissolved in essentially "clean" metal casting charges becomes largely extracted within a thin surface layer on cast ingots and (2) how a molten salt phase of MgF_2 - CaF_2 can act effectively to scavenge the Th_{234} into the salt phase. A possible explanation of (1) is as follows: In vacuum casting there exist low but significant partial pressures of oxygen and nitrogen in the furnace atmosphere. In general, the hot uranium (solid or molten) will react with all gas molecules of O_2 and N_2 that strike the clean metal surface. As the molecules of air are removed from the furnace atmosphere by reaction with the metal, they will be replenished by backstreaming of air through the pumps since the furnace pressure must remain essentially constant at whatever low pressure the pumps are capable of producing. Neither nitrogen nor oxygen has appreciable solubility in uranium with the result that minute inclusions of solid oxynitride must nucleate and grow within the molten metal. Since thorium forms even more stable chemical bonds with oxygen and nitrogen than does uranium, it is to be expected that a large fraction of the Th_{234} atoms will form such bonds and be incorporated into the inclusions of the uranium oxynitrides. As the inclusions nucleate and grow in size, they will develop a tendency to leave the metal phase. The inclusions are lower in density than molten uranium and will tend to liquate upwards to the surface of the melt under the influence of gravity. Probably more important are phase separating mechanisms brought about by interfacial tension forces. If the uranium melt does not strongly "wet" the inclusions, any inclusion that has reached the surface of the melt or which finds itself at the melt-crucible wall interface will tend to remain there and not re-enter the metal phase. In other words the inclusions which carry most of the Th_{234} from the melt will form "skull" material floating on the molten metal or will be trapped at the crucible wall. If the casting charges consist of essentially "clean" metal containing very little slag MgF_2 on the derbies, the skull material should consist of only a relatively small amount of the solid uranium oxynitrides (and some uranium carbide). If the small volume of skull material has extracted almost all of the Th_{234}, the specific radioactivity (mr/gram or cc) of this material should be very high. At pour some amount of the skull material will enter the mold to be trapped as a thin layer between the metal and mold surface. The rest will remain on the walls of the crucible.

We now consider the possible effects of adding a few pounds of mixed magnesium and calcium fluorides to the casting charge. As a more specific example, assume that a total of four pounds of the fluorides containing 19 w/o CaF_2 has been added to 1400 pounds of uranium metal (derbies plus recycled scrap metal) in a Mark 31B charge. On heating the charge in the crucible, the salt mixture should begin to melt first at 940°C and be completely melted at a temperature somewhat below the melting point of the uranium. As the metal begins to melt, the molten fluorides will float on top of the liquid metal. As the metal becomes molten under the vacuum conditions, it will be agitated by bubbles of escaping hydrogen. This ebullition of the melt will help to mix the molten metal and fluorides. As the solid inclusions of uranium oxynitrides form they will be taken into the molten fluoride phase by processes involving increased surface tension wetting of the particles by the salt phase and also because the molten fluorides can dissolve the oxynitrides to some extent. At any rate, the Th_{234} should now be largely contained within the salt phase. Moreover, because the molten salts add considerably to the total volume of skull material, the specific radioactivity (or concentration of the activity) in the skull material will be lowered. At pour, some of the skull may enter the mold and be trapped at the surfaces of the freezing ingot, but we should expect the Th_{234} to be far less concentrated in a surface layer on the ingot. Also most of the molten salt skull material should remain in the drained crucible. Very little may flow out into the mold because of its relatively low density and because it may be rendered quite viscous by its content of undissolved uranium oxynitride particles.

We may also predict that the addition of the fluorides will result in less of the total radioactivity being retained on the interior crucible wall after burnout of the crucibles. The daughter product activity will now be diluted in a larger bulk of skull material. Most of this material will drop out of the inverted crucible during the burnout to leave less activity on the crucible walls. Thus a greater fraction of the total activity should be removed and captured in a drum below the burnout station where it no longer poses a radiation hazard to operating personnel. Less of the total activity reaches the station where the crucibles are further cleaned manually and, of course, even less remains in the crucibles at the charging station.

The separation of the daughter product Th_{234} which occurs in the bomb reduction step almost certainly proceeds by a mechanism similar to that discussed above for the casting operation. We believe that the essential key is that a small content of oxygen must be present as an "impurity" in the reduction process. It must be present in sufficient concentration to react with the Th_{234} to form the very stable thorium-oxygen bonds. The oxygen content is normally present as UO_2F_2 and uranium oxides in the green salt and as traces of hydrate or hydroxyl "water" in the green salt and liner material. Depleted green salt contains little UO_2F_2 or uranium oxides, but UO_3 is purposely added to the extent of 0.6 w/o to the green salt. In the reduction process we know that the oxygen content ultimately ends up as the minor phases MgO and UO_2 in the slag MgF_2. The derby interior metal contains very little of the oxygen in the form of oxygen in solution or inclusions of UO_2. In bomb reduction the Th_{234} should form ThO_2 which is absorbed into the inclusions of UO_2 that are formed simultaneously. The inclusion material is then taken into the MgF_2 slag phase as previously discussed.

Comparative measurements of the radiation from slag-free areas at the tops of freshly prepared depleted and enriched derbies have indicated that the daughter product removal is significantly greater in the enriched reductions. We theorize that the effect is due to the generally higher oxygen content of the hydrofluorinated-type green salt used in the enriched reductions. It has been proposed to study the effect further. Specifically, it has been proposed to test the effect of adding additional oxygen to the depleted reduction charges - either in the form of an increased UO_3 addition or as an addition of calcium hydroxide.

Since the daughter products are apparently very effectively removed from the metal temporarily in the reduction process, it has been proposed to take advantage of this fact by incorporating derbies into the casting charges as soon as possible after their preparation by reduction and before the daughter product Th_{234} has time to grow back into the derby metal. This scheme appears to be quite practical for the flat castings which need not incorporate a large fraction of aged metal in their casting charges. As will be discussed in a later section, the scheme has been tested in the casting of one pair of flats with eminently successful results.

DESCRIPTION OF EXPERIMENTAL WORK PERFORMED

Procedures:

Not all the work has been performed as yet. The major part of the work completed has involved the addition of MgF_2 and CaF_2 to the metal charges of Mark 31B ingots. One of these was cut up to obtain samples for a metallographic study of the inclusions. Ingots have been isolated as a special group to be followed through to final machined cores. Thus it should be possible to evaluate the test ingots as to metal quality and core yields.

The magnesium fluoride for the test consisted of the depleted liner material (MgF_2) taken from the production supply of milled product. For some tests, the slag liner was leached with nitric acid, washed, and dried before use. This was done to remove most of the uranium which is present in the milled slag as either free metal or uranium oxides. In the test work we did not detect any obvious benefits to be gained by leaching the slag liner material. In two test castings, MgF_2 was added as coarser lumps produced by breaking up massive reduction slag with a hammer.

The initial supply of about 25 pounds of calcium fluoride was from a supply of pure precipitated material purchased several years ago. When this supply of very expensive CaF_2 ran out, a further quantity of 70 pounds of pure CaF_2 was prepared in the laboratory. The material was precipitated by slurrying technical-grade, bagged, calcium hydroxide in dilute (36% HF) obtained from the Tank Farm. The precipitated CaF_2 was filtered, washed and tray-dried in ovens. The soft cake was pulverized and passed through a 25-mesh screen before use. This preparation of CaF_2 was undertaken partly to gain experience in making the material, and also because the purchase price of the pure fluoride is very high.

The general procedure followed throughout the test program of MgF_2 - CaF_2 addition was to cast consecutive groups of three ingots, two of which contained the fluoride additions and one of which contained no fluorides. The three ingots thus all would be similar in charge makeup (number of derbies and same type of recycle scrap). The ingot cast without fluoride addition differed in no way from regular production ingots and was considered to be a "control."

Casting with the fluoride addition differed in only one detail. Roughing evacuation of the furnace was done slowly over a period of up to five minutes. This was done to make sure that the mixture of the powdery fluorides would not be partially blown out of the crucibles by the sudden expansion of air entrapped in the powders.

The radiation measurements were made on the ingots within a day after casting and usually immediately after stripping from the graphite molds.

The readings (mr/hr) are taken by I. H. and R. personnel using two Ludlum Model Geiger-Muller survey meters. (Model 3 is used in the lower ranges and Model 5 is used for higher ranges.)

During the test work, an effort was made to retain the same two crucibles for repeated use in casting those test ingots containing the added fluorides. Similarly the same crucible was repeatedly used for casting of the "control" ingots. After each use the crucibles (test and control), following burn-out, were set out and measured for radioactivity on their interior surfaces.

TEST RESULTS - 10" DIAMETER X 30" LONG INGOTS

The results of the tests for both "control" and fluoride added charges are shown in Tables 1-4.

Tables 1 and 2 are for similar charges (3 derbies plus solid scrap) while Tables 3 and 4 are for charges with 3 derbies, solid scrap and machine turnings (briquettes).

TABLE I - CONTROL INGOTS - NO FLUORIDES ADDED

CHARGE = 3 Derbies + Solid Scrap

INGOT	Readings (β + γ), mr/hr BOTTOM SURFACE	DISTANCE FROM BOTTOM (Circular Surface) 3"	12"	20"	27"	TOP SURFACE
3762	1800	1400	1400	1300	650	400
3800	1850	1000	700	1400	900	125
3827	1800	1200	800	1000	70	600
3907	1800	1500	1500	1100	75	500
3940	1600	1350	1000	800	200	550
4165	1850	350	1150	1100	450	400
4207	1500	1200	1100	900	80	150
4238	900	600	120	400	60	600
4251	1300	1050	650	-	-	600
4321	1200	775	725	800	55	250
Avg.	1560	1043	915	978	282	418

TABLE 2 - TEST INGOTS - FLUORIDES ADDED

19 w/o CaF_2/81 w/o MgF_2 added
(4 lbs/1400 lb charge)

CHARGE = 3 Derbies + Solid Scrap

	Readings (β + γ), mr/hr					
INGOT	BOTTOM SURFACE	DISTANCE FROM BOTTOM (Circular Surface)				TOP SURFACE
		3"	12"	20"	27"	
4208	1600	700	500	600	170	400
4209	1550	400	850	75	500	750
4252	1300	450	90	70	120	500
4253	1050	150	40	35	500	140
4319	1300	70	50	28	20	400
4320	1150	80	30	28	20	400
4344	700	500	200	110	80	50
Avg.	1236	336	251	135	201	377

TABLE 3 - CONTROL INGOTS - NO FLUORIDES ADDED

CHARGE = 3 Derbies, Solid Scrap, Machining Chips

	Readings ($\beta + \gamma$), mr/r					
INGOT	BOTTOM SURFACE	DISTANCE FROM BOTTOM (Circular Surface)				TOP SURFACE
		3"	12"	20"	27"	
3468	1200	600	700	-	160	1300
3718	1700	1100	750	700	500	60
3728	1750	1150	800	700	500	1500
3741	1850	1400	1200	1000	150	200
4346	1200	800	900	700	120	800
4535	1000	600	350	300	150	15*
4558	2000	950	750	350	125	250
Avg.	1529	943	779	625	244	685**

* Cropped Face

** w/o Cropped Face

TABLE 4 - TEST INGOTS - FLUORIDES ADDED

19 w/o CaF_2/81 w/o MgF_2 added

CHARGE = 3 Derbies, Solid Scrap, Machining Chips

INGOT	Readings (β + γ), mr/hr					
	BOTTOM SURFACE	DISTANCE FROM BOTTOM (Circular Surface)				TOP SURFACE
		3"	12"	20"	27"	
4221	800	15	14	45	50	50
4222	750	75	50	130	400	50
4345	600	450	65	55	100	60
4533	1000	55	55	30	20	15*
4534	850	40	42	30	20	15*
4557	550	45	47	20	17	500
4624	400	100	50	65	25	500
Avg.	707	111	46	54	90	232**

* Cropped Surface

** w/o Cropped Surface

DISCUSSION OF TEST RESULTS ON INGOTS

Most of the radiation measurements were made with the ingot lying horizontally. Readings were taken at the top of the uncropped ingot, in most cases, at four positions along the exposed topside sides, and on the flat bottom. The readings are somewhat subjective and tend to be the values indicated by the maximum deflections of the meter needle.

Comparing the controls with the fluoride additives the following reductions in radiation were observed.

CHARGE	BOTTOM SURFACE	DISTANCE FROM BOTTOM 3"	12"	20"	27"	TOP SURFACE
3D, SS	1.3	3.1	3.6	7.2	1.4	1.1
3D, SS, brix	2.2	8.5	17	12	2.7	3

The highest levels of radiation from the Mark 31B ingots are found at the flat bottoms of the ingots where the metal solidifies against the mold cup. We still have no very cogent explanation of this phenomenon. The addition of the fluorides to the melts has not reduced the radiation from these ingot bottoms to a very satisfactory degree. At one time it was thought that some of the daughter product activity gathered in the skull material was being spewed out of the crucible during the period of the vigorous metal "boil," and that some of this material was dropping down to the bottoms of the molds where it eventually became trapped against the mold cup by the rapidly freezing metal. A pathway does exist by which spewed material can sift down into the mold. However, we have been able to close off this pathway by appropriate use of Cerafelt so that ejected material cannot enter the top of the mold. This did not reduce the radiation emitted from the bottoms of the ingots. We have found that a small amount of "dirt" does fall into the mold prior to pour, but this material is not highly radioactive. It is now clearly established that the daughter product activity that ends up on the bottom faces of the ingots enters with the metal stream at the time of pour.

There was no detectable difference in inclusion content, size or type from examinations of cut up ingots (control and test).

Chemical analyses showed no significant differences between the control and test ingots except for a slight increase in hydrogen (0.25 ppm) in the test ingots.

One noticeable effect appeared in that the ingots containing the added fluorides had no primary pipe in the top. Usually one out of five production ingots (same as controls) contain pipe. Additional work is needed here.

TEST RESULTS - CRUCIBLES

Table 5 shows the readings obtained on the inside top (2" down) and on the inside bottom of crucibles used in making fluoride additive melts (crucible S 8808 and S 8809). Similar readings were obtained on one crucible (S 8811) used in casting "control" charges.

The results show a reduction of 4.6X in radiation from the top of the crucible and a 1.13X reduction at the bottom of crucible. The overall reduction is about 2.9X.

TABLE 5 - RADIATION MEASUREMENTS PERFORMED ON CRUCIBLES USED FOR MELTING OF Mk 31B CHARGES

Radiation ($\beta + \gamma$) in mr/hr. (After Burnout and Cleaning.)

	CRUCIBLES USED WITH FLUORIDE ADDITIONS				CONTROL	
	S 8808		S 8809		S 8811	
	TOP	BOTTOM	TOP	BOTTOM	TOP	BOTTOM
	150	600	70	90	800	190
	140	400	130	170	900	600
	300*	700	200	450	700	500
	110	400	300	700	1000	800
	80	500	95	500	900	700
	250	850	150	550	900	650
	110	900	600*	700	900	800
	95	400	150	500	1200	1000
	110	650	140	500	750	800
	140	600	300	800	900	800
	90	700	120	700	500	500
	150	700	90	500	600	500
	200	750	110	600	850	750
			150	500	800	650
			190	500	700	725
			150	700	650	{600 1000
			150	700	?	?
			250	500	250	450
			250	500	Crucible cracked	
	TOP	BOTTOM	TOP	BOTTOM	TOP	BOTTOM
AVERAGE	148	627	189	537	782	660

*Note highest "top" radiation readings obtained with additions of 6 lb MgF_2 containing no CaF_2.

RADIATION REDUCTION FACTOR (Tops of Crucibles) $= \dfrac{782}{\frac{1}{2}(148 + 189)} = 4.6$

RADIATION REDUCTION FACTOR (Bottoms of Crucibles) $= \dfrac{660}{\frac{1}{2}(627 + 537)} = 1.13$

"OVERALL FACTOR" $= \dfrac{4.6 + 1.13}{2} = 2.9$

FLAT CASTINGS

The flat castings pose somewhat different and additional problems from a radiation standpoint. An additional work station requires restriction on the basis of radiation exposure. This is the saw where the flats are top cropped and sawn into halves. Also the mold cleaning and mold coating operations involve much more close contact with the contaminated molds. Here the beta radiation may be less objectionable than the dust inhalation hazard.

Normally the flats are produced from molten salt-cleaned and nitric acid-pickled derbies and the charges include little (if any) recycled scrap. The melting crucibles are flame-sprayed with zirconia to limit carbon pickup. The derbies are cleaned to minimize the amount of slag MgF_2 on them since it has been thought that even minor amounts of the slag could react with and destroy the zirconia coating and thus cause excessive carbon pickup from the graphite crucible. It may therefore appear that a scheme for lowering the radiation from flats based on adding magnesium and calcium fluoride to the melts is not feasible since it is likely to cause excessive carbon in the metal. Nevertheless, four flat castings have been made in which the charges contained 1.5 or 2.0 pounds of the mixed fluorides. The limited data from these tests has indicated that the $\beta + \gamma$ radiation was reduced by a factor of at least two. Carbon values obtained on the four flats were 5, 24, 40, and 89 ppm. The highest value did exceed the specification of 60 ppm for "Class I" flats, but the range of values is typical of that seen in regular production.

Quite obviously, more testing of the "salt addition" method in the case of flat castings is required. It is planned to increase the fluoride additions to 4.5 pounds in these 1600-pound melts in order to reduce further the radiation from these castings. It remains to be seen if the increased fluoride additions will cause appreciable carbon pickup.

In another approach to the radiation problem, one flat casting has been made in which the melting charge consisted of four derbies that had been prepared by magnesium reduction of the depleted UF_4 only two days before the casting. As stated previously the reduction process temporarily removes a large fraction of the uranium daughter products of radioactive decay, and if the derbies are consumed in the casting process without undue delay, the cast ingot surfaces should be correspondingly lowered in radioactivity.

The radiation measurements made on the pair of flats produced in the single test averaged only about 20 percent greater than the average of several readings that were made on the derbies prior to incorporation into the melting charge. The $\beta + \gamma$ radiation was reduced by a factor of at least five.

The derbies used in the test casting were not salt-cleaned and pickled as is the usual practice in regular production. Nevertheless, the carbon analysis of the flats was a very low 7 ppm.

ADDITIONAL TESTS PLANNED

This work has revealed a number of promising areas for further exploration.

1) The use of fluoride additions to eliminate or minimize ingot pipe.

2) The eliminations of the extensive cleaning now done to derbies to remove adhering MgF_2 slag. (Some derbies are cleaned by use of molten salt plus nitric acid pickling.)

3) The immediate charging of derbies into the crucible so as to minimize the Th_{234} growth.

Each of these will require additional plant tests because of possible side effects. For example, the fluoride additions may adversely affect the ability to control carbon for particular customers. This will depend on the ability of the graphite crucible coating to withstand attack from the fluorides.

Production scheduling for immediate use of derbies presents another problem.

CONCLUSIONS

A mechanism has been demonstrated which lowers the radiation emanating from the surface of castings and crucibles. This should be of considerable value in aiming toward the ALARA philosophy of minimizing employee exposure to radiation.