White Paper

TBD-6000 Working Group - Putzier Effect

David Allen September 2010

During the May 12th 2010 meeting of the TBD-6000 working group, NIOSH agreed to draft language describing the "Putzier Effect" and provide that language to the working group. The language is intended for incorporation into TBD-6000. The language is presented here. The references included here will be added to the reference section of TBD-6000.

The language indicates in part a review of beta to gamma ratios at several plants. The table below supports this review. It is based on dose values contained in an SC&A document titled "Review of NIOSH Issue 1 White Paper Dated December 10, 2009" (SC&A 2009). The ratios were calculated and added here. The hands to whole body ratio is not used in TBD-6000 but it is instructive to review these as well.

	Whole body dose from Table 3 (rem)	Skin dose from Table 2 (rem)	Dose to the Hands and Arms from Table 2 (rem)	Skin to Whole body ratio	Hands to Whole body ratio
TBD-6000 95th percentile	29.375	294	3250	10.01	110.64
TBD-6000 95th arithmetic mean	7.6	76	840	10.00	110.53
TBD-6000 95th median	2.08	20.8	230	10.00	110.58
Fernald maximum	12.3	52	190	4.23	15.45
Fernald 95th percentile of highest annual doses (1963)	1.769	9.59	35	5.42	19.79
MCW maximum		38.4	140		
Electro-Met maximum	3.3	17	62	5.15	18.79
Theoretical maximum	30		3000		100.00

Proposed Language for TBD-6000

When uranium metal is melted, impurities can separate from the metal matrix. Differences in densities and melting points can then cause impurities to separate from the molten uranium metal and

concentrate on the surfaces. The two processes that routinely involve molten uranium metal are the metal reduction process and the remelting process.

The metal reduction process involves mixing uranium tetrafluoride (UF_4) with magnesium chips and loading the mixture into a lined reduction vessel. The sealed vessel is heated to initiate the exothermic reaction. The reaction results in the formation of magnesium fluoride (MgF_2) and free uranium metal. The temperature inside the vessel exceeds the melting point of uranium and, due to the high specific gravity, the uranium collects in the bottom of the vessel forming a "derby". The MgF_2 is collected above the derby as a slag. After it cooled, the derby is "broken out" of the vessel. The MgF_2 slag had to be broken and chipped away to dislodge it from the derby (Chrisofano 1960).

The derby resulting from the reduction step contained impurities that made it unsuitable for reactor fuel. The metal was both purified and altered in shape in the remelt process. In this process, the derbies are melted in a vacuum furnace and molten uranium metal poured into a graphite mold (Chrisofano 1960). The vacuum casting removes volatile contaminants and allows other impurities to float to the surface concentrating impurities near the top. Impurities can also be concentrated where the molten uranium metal cools rapidly preventing (or minimizing) the time necessary for the impurities to separate. This can cause impurities to also concentrate near other surfaces of the casting. The separation can be improved by controlling the cooling of the cast uranium. If the mold is insulated near the top, a steep temperature gradient is formed causing the ingot to solidify from the bottom to the top. This allows impurities to separate and migrate to the top of the ingot without being trapped in solidifying metal. The "hot-top" that is formed is then cut off (cropped) to eliminate the impurities (Fleishman-Hillard 1967).

A third process worth mentioning is the dingot (direct ingot) process developed at Mallinckrodt. That process produced a finish ingot directly in the metal reduction step. This eliminated the remelting step by increasing the size of the metal reduction vessel and carefully controlling the temperature and ingredients. After separation, the dingot was "scalped" by machining all the surfaces (Fleishman-Hillard 1967).

Some of the impurities in the uranium include Th-234 and Pa-234m, decay products of U-238 (as well as residual magnesium, some slag, hydrogen, and others). These isotopes are beta emitters with relatively short half-lives. The process then causes a high concentration of these beta emitters in the top and other surfaces of the cast ingot. This concentration can produce higher than normal beta dose rates that then decay to a normal dose rate with a half-life of 24.1 days.

The concentrating of decay products during the reduction process does not appear to occur. When the uranium metal is first formed in the reduction process, it exists as molten droplets of uranium intermixed with MgF_2, and unreacted UF_4 and magnesium. These droplets of molten uranium settle to the bottom of the reduction vessel. The droplets are small enough that no appreciable separation can occur within the drop. Separation would be possible once the metal collects in the bottom of the vessel. There is some indication that these decay products are actually collected by the MgF_2 before the derby is formed resulting in a decrease in beta radiation levels on the derby (Briggs 1986). Even if separation were to occur in the derby, the solidified MgF_2 has to be broken or chipped away from the derby (Chrisofano 1960). This likely would remove appreciable amounts of any material concentrated in the surface layer.

The concentrated beta emitters near the surface of castings cause elevated beta radiation levels from the uranium casting with little effect on gamma dose rates. This can cause beta to gamma dose rate ratios to be significantly higher than uranium metal in equilibrium with its decay products. This elevated ratio may not be limited to facilities where recasting of uranium metal is performed. Several months are required to the beta radiation levels decrease to normal levels and castings could be shipped to other sites in that time. Film badge readings at various sites indicate those sites engaged in remelting exhibit the highest ratio. The ratio for those sites can approach 10. Therefore, a ratio of 10 is used in this document to account for this affect.

References

Briggs, G.G., T.R. Kato, and E. Schoeneff, 1986. *Process for Reducing Beta Activity in Uranium*. United States Statutory Invention registration, Reg. Number H137. October 7, 1986.

Christofano E. and W. B. Harris, 1960, *The Industrial Hygiene of Uranium Refining*. Env Health 1(5):75-96.

Fleishman-Hillard, 1967, *Fuel For the Atomic Age, Completion Report on St. Louis-Area uranium Processing Operations, 1942-1967*. September 30, 1967.

SC&A (Sanford Cohen & Associates) 2009, *Review of NIOSH Issue 1 White Paper Dated December 10, 2009*, December 2009