SAFE ²³⁵U ENRICHMENTS FOR THE PUREX FACILITY

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The safe processing of uranium enrichments greater than 1.0 weight percent U-235 in the Purex facility required a study to determine what enrichments could safely be processed. This document describes the rationale used in determining these safe enrichments.

SUMMARY

The control of criticality safety outside of geometrically favorable equipment in the Purex facility is based upon the following:

1. Where precipitation is not probable (requiring three or more independent operational errors), concentration control is used and advantage is taken of the nitrogen poisoning in uranyl nitrate solutions with or without plutonium.
2. Where precipitation of uranium solutions is possible (requiring less than three independent operational errors), the following maximum uranium density of the given precipitate is used to determine the enrichment limitation:

<u>Precipitate</u>	<u>Maximum Plant U Concentration g U/l</u>	<u>Maximum Safe Enrichment Wt% U-235</u>
Uranium Sulphate	970	1.78
Polyuranates	905	1.87

3. For precipitated uranium-plutonium mixtures the Pu/U ratio of the precipitate is considered to be twice that of the original solution and the limiting equivalent enrichment is based upon that ratio.
4. Safe equipment mass limits are based upon a safe uniformly distributed slab of precipitate at a maximum uranium density of 905 g U/l (if sulfate precipitation cannot be excluded, 970 g U/l must be used).

The minimum critical enrichment of uranyl nitrate has been determined by experiment to be 2.104 wt% with a standard deviation of 0.01 wt% resulting in a minimum of 2.07 wt% at the 99 percent confidence level. Since the calculational method used agreed very closely with this and other experiments, a calculated k_{00} of 0.98 was considered to give an adequate safety margin for specifying the maximum safe enrichments of precipitates.

DISCUSSION

Uranium solutions in the Purex plant are normally in the uranyl nitrate form even when the uranium is in the organic phase (TBP solvent). Physical Constants Testing Reactor (PCTR) experiments (reference 1) have confirmed the calculations (reference 2) showing uranyl nitrate enriched to less than 2.07 wt% in U-235 is safe to handle in any concentration as long as the nitrogen to uranium atomic ratio is ≥ 2 . Since process solution vessel temperatures are limited to less than 188°C, the only way of separating the nitrogen from uranium is by precipitation.

C. A. Colvin and others (reference 3) have experimented with reagents used in the Purex process that could accidentally be added to UNH solutions to cause precipitation. In the experiments, the maximum uranium precipitate densities were obtained by centrifugation. The maximum uranium concentrations obtained were 970 grams of uranium per liter as a sulphate precipitate and the next highest of 905 grams of plutonium per liter as a polyuranate.

In an accidental precipitation, the resulting solids will settle by gravity to the bottom of the containing vessel and will have a uranium concentration less than the experimental maximum densities. Therefore, the use of these maximum densities for calculating uranium enrichments that are safe to process in Purex is considered to be conservative. Figure I (reference 2) is a plot of k_{∞} versus uranium concentration for a $\text{UO}_3\text{-H}_2\text{O}$ system calculated by the GAMTEC two-group code (reference 5). Compared to PCTR experiments for $\text{U}(3.04)\text{O}_3$ in polyethylene, the $\text{UO}_3\text{-H}_2\text{O}$ calculations for k_{∞} appear to be conservative in the well moderated region, yielding a $k_{\infty}=1$ at a uranium concentration of 510 grams uranium per liter versus 540 grams uranium per liter for the UO_3 -polyethylene experiment. A calculated point of the UO_3 -polyethylene experiment at an $\text{H/U}=43.85$ yielded a k_{∞} of 1.030 versus an experimental k_{∞} of $0.996 \pm .006$. Additional conservatism may be claimed for the poisoning effect of the nitrate solution that would exist in the settled precipitate as shown in Figure II. Because of this conservatism and the accuracy of the GAMTEC k_{∞} calculation of $\text{U}(2)\text{NH}$ experiments (reference 1), a k_{∞} of 0.98 was considered to give an adequate safety margin. Thus, polyuranate precipitates (905 g U/l) at $k_{\infty}=0.98$ are safe up to an enrichment of 1.87 wt% U-235 and sulphate precipitates (970 g U/l) up to 1.78 wt% U-235 as shown in Figure II.

Reactivity calculations were made with GAMTEC II (reference 4) using a 910 g U/l polyuranate precipitate at 2.0 grams/cc overall density. These two-group k_{∞} calculations are plotted in Figure III for three enrichments. An H/U of 16 is equivalent to the maximum experimental uranium density of polyuranate precipitates. The H/U ratios less than 16 were calculated by removing water while maintaining a constant precipitate volume. These calculations show that when the precipitate is allowed to dry out, the k_{∞} and material buckling will increase slightly. The vessel vent systems and the centrifuge are the only process vessels where, after an accidental or planned precipitation, the solids might dry out unnoticed. Since the 48-inch diameter centrifuge has been calculated to be safe for enrichments up to 2.5 wt% U-235 due to the geometry of the vessel (reference 4), and since the accumulation of large amounts of uranium in the vessel vent systems (i.e., filters or silver reactors) is highly unlikely, these systems are considered safe.

If a plutonium-uranium solution is accidentally precipitated, the Pu/U ratio in the precipitate can vary widely from the Pu/U ratio of the starting solution depending upon the plutonium valence. The Pu/U ratio has been shown experimentally to be increased as much as 80 percent during polyuranate precipitation from a uranyl nitrate solution, thus increasing the effective enrichment of the precipitated uranium. Therefore, each process vessel or sump containing Pu-UNH solution must be evaluated for the possibility of accidental precipitation (precipitation of any kind is normally excluded in the Purex plant (reference 6)). If the probability of an accidental precipitation is high (less than three independent processing errors), the Pu/U ratio of the precipitate is considered to be twice the solution ratio when

calculating the safe equivalent enrichment at maximum precipitate density (reference 7).

Safe equipment mass limits are based upon a safe uniformly distributed slab of precipitate at a maximum uranium density of 905 g U/l (if sulfate precipitation cannot be excluded, 970 g U/l must be used). Any plutonium is considered to be precipitated homogeneously with an optimum amount of uranium as a uniform slab at the maximum uranium density. A safe slab is taken as eighty-five percent of a critical slab in the equipment being considered.

A curve for estimating the relative worth of plutonium in comparison to U-235 in solutions of low U-235 enrichment has been calculated using GAMTEC II (Figure IV, reference 4). The curve gives the relative worth of U-235 as compared to Pu-239 with varying Pu-240 content.

CONCLUSIONS

Thus, it is concluded that the processing at Purex of uranium enriched to 1.87 wt% in U-235 is safe, if sulfate precipitation is excluded. Should the processing of higher enrichments be required, additional administrative controls can be used to assure the low risk potential necessary.

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FIGURE I

HOMOGENEOUS URANIUM TRIOXIDE WATER MIXTURES

⊙ PCTR EXP $U(3.04)O_3$ - Polyethylene

△ GAMTEC II CALC. $U(3.04)O_3$ - Polyethylene

FIGURE II

— $\text{UO}_3 - \text{H}_2\text{O}$ ---- Polyuranate at 910 g U/l
with nitrate solution includedK&E 10 X 10 TO THE CENTIMETER 46 1510
MADE IN U.S.A.
KEUFFEL & ESSER CO.

FIGURE III

CALCULATED K_{∞} OF PRECIPITATED UNH USING 50 WT%
SODIUM HYDROXIDE

An H/U of 16 represents a moist centrifuged cake (910 gU/l). Lower H/U values were obtained by holding the uranium density constant and removing water. GAMTEC II calculations.

FIGURE IV
CALCULATED RELATIVE WORTH
OF ^{235}U /Pu
GAMTEC II

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