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UNITED STATES ATOMIC ENERGY COMMISSION

CRITICALITY HAZARDS IN PROCESSING 1% ENRICHED URANIUM

By
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For The Atomic Energy Commission
H. F. Canale
Chief, Declassification Branch

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MEMORANDUM TO FILES

Subject: Criticality Hazards in Processing 1% Enriched Uranium

The processing of the 1% enriched uranium at Mallinckrodt Chemical must of course be done under conditions which prevent any possibility of a chain reaction occurring in the uranium. The processing procedures up to the production of ingots were outlined by Mallinckrodt representatives at a meeting in the New York AEC office, September 21, 1951. This memo analyzes the possible hazards in this processing, and specifies necessary precautions.

In eliminating criticality hazards from a mixture containing fissionable material, there are two cases. (1) If k , the material neutron reproduction factor, is less than unity, there is no criticality hazard with any amount of material. (2) If k is, or even might possibly be, greater than unity, the materials must be disposed so that neutron leakage and absorption prevents criticality. This is best done by limiting the amount of material in a single unit.

Where there is no significant amount of H_2O moderator present, the only chain reaction which need be considered is a fast neutron reaction. With low enrichment uranium, it is shown that the k is well below unity, so that there is no possible criticality problem. Where H_2O is present, it is conceivable that criticality could be approached, with the full batch of 3 tons of enriched uranium in a lattice with H_2O . In this case it is recommended that the weight of uranium in one assemblage be limited to one ton.

No H_2O Present

First consider the considerations for criticality when there is no H_2O present. It was shown experimentally (CF-1627, "Neutron Multiplication in a Mass of Uranium Metal," by Brolley et al., April 1, 1944) that k for natural metal = 0.34. For k to = 1.00, it was calculated in this report that an enrichment of 8% would be required. Calculations at H. K. Ferguson indicate that 1% enriched, unmoderated uranium would have a k 0.05 higher than natural uranium, making k = 0.39. There is no possibility, therefore, of a fast chain reaction in mass of 1% enriched metal. The reason is that at fission energies the U-238 undergoes some fission, but inelastic scattering drops most of the neutrons below the U-238 fission energy threshold. A calculation showed 0.36 fast neutrons produced by fast fission per fast neutron absorbed or inelastically scattered. (The effect of U-235 at fission energies in 1% concentration is negligible, accounting for 3% of the 0.36).

There are now 0.82 of the original fast neutrons slowed below the U-238 threshold, so that further multiplication must come from U-235. There is little elastic slowing down in the metal, so the neutrons remain at high energies. But the ratio $\sum_f(25)/\sum_a(28)$ here is only 0.05. **DECLASSIFIED**

more fast neutrons are returned, making a total of $0.36 + 0.16 = 0.52$ for k . These calculations are conservative, giving $k = 0.47$ for natural uranium, instead of the experimentally determined 0.34. (Cross sections were taken from ORNL-86, "Tables of Neutron Cross Sections for Elements Po-Cm" Haines and Way, September 9, 1948).

The addition of fluorine to form UF_4 adds a little moderation, but not enough to bring the neutrons to thermal where a chain reaction would be possible. The absorption will take place at above resonance energies, where the fission cross section of U-235 is still not appreciable in 1% enriched material, compared to the capture in U-238.

The enriched uranium comes into the Mallinicrodt plant in the form of UF_4 , and is converted to the metal. Until the metal derbies are stored in a cooler, there is no moderator present, and no possible hazard. The first time H_2O moderator might be present is in the tank where the derbies are cooled. Let us consider the possible hazard.

H2O Present

A chain reaction with low enrichment uranium is possible with H_2O as moderator if the proper lattice of uranium and water is present, and if the amount of neutron poison is limited. Then the k can be above unity, and the lattice could become critical above some mass.

A calculation was made assuming 3/4 inch diameter uranium rods in a lattice with H_2O . (This diameter has a greater chance of reacting than smaller or larger rods). The volume ratio of H_2O to uranium was varied. Thick reflectors of graphite in one case, H_2O in the other, were assumed. The resulting critical mass, in short tons, is plotted against the volume ratio in Figure 1. The minimum critical mass is 1.25 tons, with a graphite reflector which would be several feet thick. The case of the H_2O reflector, which more nearly approximates the condition in the Mallinicrodt plant, requires 2.1 tons. The volume ratios are about 3 H_2O to 1 uranium, for this worst case.

The assumptions were conservative in the direction of setting a lower limit on the possible critical mass. An enrichment of 1.05% was assumed. No poison was present other than the H_2O and uranium in the core. A spherical geometry was taken. The k was taken 0.02 higher than our present best estimate, based on lattice experiments at ORNL.

This analysis gives the worst possible criticality hazard. Let us examine the actual problem. The first time that the uranium is in a lattice with H_2O is when the uranium derbies are cooling. Here 130 lb. of metal are contained in an iron bomb weighing 275 lb. The amount of poison is so great that the k for the lattice is well below unity. There is therefore no possibility of a chain reaction.

The derbies are melted, a total of 650 lb. at a time, to form billets 5-1/8 inch diameter, 44 inches long. There is a possibility that water might surround individual billets. This would not be the same as a water lattice, however, but would come under the metallic uranium case.

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which has been shown to have k less than unity. The processing of the billets, to form $3/4$ inch rods, was not described. It is in this step that the only possibility for a chain reaction exists. Liquid water, or H_2O in wooden boxes, could provide moderation. The hazard is greatest at the smaller rod diameters. Here the amount of uranium in one stack, assemblage, or unit should be limited to less than one ton for absolute safety. Information from exponential experiments which are being set up at Brookhaven will provide more accurate estimates of safe uranium amounts and will undoubtedly result in higher permissible masses.

Summary

No danger is possible in handling any amount of nominally 1% uranium in the reduction of UF_4 to metal, cooling, or remelting. Critical amounts might conceivably be accumulated during the rolling to $3/4$ " rod, in which case an "always safe" amount will be one ton. Allowance is made in this calculation

- (a) For the possibility that the U-235 concentration may be as high as 1.05%.
- (b) For a possible ($k-1$) of a clean H_2O lattice with 1% U-235 20% above our present best estimate.

Kenneth C. Cooper

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Critical Mass
1.05% Enriched Uranium
H₂O Lattice

Figure 1