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By J. E. Savely 11-17-94

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Separations Section

From: Paul E. Collins, Leader PEC
234-5 Plant Assistance
Separations Technology Unit

Production of "X" Buttons In The 234-5 Building

Process Review

Since 1945 the routine production of plutonium ("A" buttons) at Atomic Energy Plants has been accomplished by heating mixtures of plutonium tetrafluoride, calcium and iodine in crucible-bomb assemblies. Yields above 97 per cent and metal of adequate purity are consistently obtained by this process.

Plutonium ("X" button) production has also been carried out routinely by including plutonium turnings with the powder mixture given above. A majority (several

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thousand) of buttons produced in the 234-5 Building have been "X" buttons made by recycling up to 1.1 units of turnings/unit of plutonium in the plutonium tetrafluoride. Benefits obtained by recycling turnings to this operation include:

1. Re-entry of turnings to the process stream only two operations prior to their production,
2. Turnings may be processed in the form in which they are produced,
3. Button sizes may be altered easily, within limits, by varying the amount of turnings used,
4. Production capacity of the equipment supplied is adequate to process the material, and
5. Button purity and product yields are not appreciably affected.

The benefits given above have been the bases for the adoption of the processes for producing plutonium and recycling turnings. Operating hazards exist in either process, however, and these are reviewed below.

Operating Hazards.

The reactions occurring during reduction are shown by the following equations:

If plutonium turnings are added they melt (some of the oxide coating may reduce) and collect with the reduction regulus.

The free energy of formation of calcium iodide is ca. $-130^{(1)}$ kilogram calories per mole. The change in free energy for reaction number 2 is ca. $-175^{(2)}$ kilogram calories/mole Pu. Therefore, the mixture of Ca, I_2 and plutonium tetrafluoride is thermodynamically unstable and a spontaneous reaction is possible. This fact, however, does not define the reaction kinetics or speeds of the reactions. Experimental work at Los Alamos⁽³⁾ established that reactions 1 and 2 do not become rapid enough to be self-sustaining below ca. 400° and 600°C respectively. The reduction mixture, however, becomes self-sustaining at ca. 325°C . The latter phenomena is confirmed by the fact that plant reduction reactions occur when the reactants are in the range of 325° to 375°C . From these data it would be concluded that handling the reduction mixture is safe at room temperature and no evidence to the contrary has been observed in the handling of several thousand reduction mixtures to date in the 234-5 Building.

When turnings are being recycled a hazard exists in Hood 9 where the crucible-bomb assemblies are being loaded. Turnings have been placed in crucibles after some of the mixture of Ca, I_2 , and PuF_4 have been added. The remainder of the

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latter mixture is then placed on the turnings and the charge is tamped with a lucite rod. The turnings spark at times when they are being handled. It is conceivable that the sparks could initiate reaction number 1 above and nucleate the reaction in the remainder of the charge. These loading procedures have been carried out several thousand times without a premature reaction occurring. However, to give an added degree of protection during this operation it is recommended that, (1) Hood 9 be equipped so that an inert atmosphere can be established during the loading operation (already accomplished), and (2) that the following crucible-loading sequence be used:

1. Place turnings in the bottom of the crucible,
2. Flush crucible turning charge with argon or helium,
3. Inspect the turnings charged for sparks or glowing fragments, and
4. If no turnings have ignited add the powder mixture and tamp or vibrate the charge to pack it.

During the firing of the reduction charge temperatures and pressures in the bomb assembly are probably as high as 1600°C and 200 lbs/sq.in.⁽⁴⁾ for a period of a few minutes. If the reactants are not confined during this time their high temperature creates a serious hazard. Two gasket failures have occurred during PuF_4 reductions in the 234-5 Building and wide spread contamination occurred both times through burned or ruptured gloves. Recently a new type of reduction bomb having two virtually independent seals has been placed in use in the R.G. Line. Prior to its adoption for use a uranium tetrafluoride reduction was carried out in similar equipment in which the inner gasket was filed to a point where its failure was assured. Only a small quantity of reactants escaped through the first gasket and the second gasket was unaffected. This bomb is undoubtedly safer to use than the one it replaced.

Summary

Although hazards exist during the preparation for and during the reduction reactions, and complete protection against these hazards cannot be assured, we believe that the safety devices and procedures which are in current use (double-sealed bombs and an inert atmosphere in Hood 9) plus the procedures recommended above for loading turnings make the reduction operation including recycled turnings a feasible process for routine use in the 234-5 Building.

Letter verbally approved by
R.B. Richards 4-8-52 P.E.C.

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- (1) Handbook values.
- (2) Data on ΔH_f and ΔS_f obtained from:-
 - a. CN-3551, Secret, "Compounds of Plutonium", pg. 45, B.B. Cunningham.
 - b. Oxidation Potentials - Latimer.
 - c. Handbook of Chemistry - Lange.
- (3) LA-13. Secret. "Preparation of Plutonium Metal By The Bomb Method", R.D.
- (4) Verbal communication from R.D. Baker, Los Alamos, New Mexico.