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IMPACT OF NEW PROCESS TECHNOLOGY UPON CONTINUOUS TASK I AND TASK II INSTALLATIONS

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INTRODUCTION

A new process for the conversion of plutonium nitrate solutions to a dry compound suitable for reduction to massive metal has been developed at Savannah River. Plutonium trifluoride is precipitated directly from the nitrate in an aqueous medium; after filtering and drying, the trifluoride powder can be efficiently reduced to plutonium metal. Both laboratory and plant-scale applications of a batch plutonium trifluoride process have been demonstrated successfully at Savannah River and are under study at Rocky Flats. Adaptation to a continuously operated system has been initiated at HAPO with preliminary results appearing highly favorable.

When compared to the present HAPO procedure of precipitating plutonium oxalate, filtering, drying, and hydrofluorinating the cake to plutonium tetrafluoride, it is apparent that the newer process combines the present wet chemistry and dry chemistry operations into one step. Under certain conditions the newer process may cause rapid technological obsolesence of the present HAPO procedure. Since the expenditure of approximately $750,000 is currently being proposed to install continuously operated production-line equipment within the 234-5 Building for the older process (including a supernatant evaporator), consideration should be given to the possible impact of the new technology upon the continuous Task I and II installations and the desirability of enjoying the advantages of the newer process without the intermediate expenditure of funds for the older process.

OBJECTIVE

It is the purpose of this study: (1) to review the advantages and disadvantages of the plutonium trifluoride process and the processing conditions under which the procedure would be applicable at HAPO, and (2) to consider the possibility of delaying the equipment installation for the present process until such time as the new process can be thoroughly evaluated.

SUMMARY AND CONCLUSIONS

The prime advantage of the plutonium trifluoride process is the possibility of low plutonium losses and the direct disposal of the filtrate to waste. Precipitation of plutonium trifluoride, however, does not provide an appreciable separation of plutonium from cationic impurities and is therefore sensitive to feed quality. Because of this disadvantage, the process is incompatible with the present plant processing scheme at HAPO and could not be adopted immediately even if it were suitably developed. Urgent production capacity requirements prevent delaying the installation of the proposed Task I and II facilities until the required development programs for the Purex and Redox product solutions could be completed and the trifluoride process made compatible. With the large capital investment to be placed in oxalate-fluorination production and recovery equipment, the relative advantages of the trifluoride process would not economically justify replacement during the service life of the installation. Thus, the adoption of the plutonium trifluoride process at HAPO prior to 1962 would appear to offer no economic advantages.

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ADVANTAGES AND DISADVANTAGES OF TRIFLUORIDE PROCESS

The plutonium trifluoride process, as presently envisaged, has three advantages:

(1) The waste filtrate from the precipitation and filtration of plutonium trifluoride is theoretically low enough in plutonium content (less than 0.1 per cent) to permit direct disposal to a waste crib. It should be noted, however, that low losses are dependent upon an efficient filter and a proper feed stream. With present filtering techniques, batch plutonium losses from plant oxalate precipitations vary from one to ten per cent and average three to four per cent (as compared to the one per cent obtained in the laboratory) due to mechanical by-passing of the filter. While improved performance is anticipated for continuous filtration, mechanical factors may occasionally permit losses high enough that recovery would be required. Furthermore, certain impurities in feed streams may result in unpredictable filtrate losses and filtration problems.

(2) The "kill" procedure for the plutonium trifluoride filtrate is relatively simple. If filtrate recovery should be required because of filter difficulties or unusual feed conditions, the fluoride ion would be complexed with an excess of aluminum ion and the solution processed through the Recuplex facility. Recovery of the present oxalate filtrate requires a more complicated "kill" involving treatment with permanganate ion, then hydrogen peroxide, and followed by salting with aluminum nitrate for Recuplex operations. If direct disposal to waste is permissible for the trifluoride filtrate, simple neutralization should be adequate.

(3) The present use of hydrogen fluoride at high temperatures would be largely eliminated. The operating conditions required for removing residual hydrofluoric acid and drying the fluoride cake, however, are not firmly established. The temperatures involved may be high enough to offset this advantage.

The plutonium trifluoride process has serious limitations that are of particular concern to the HAPO plant processing scheme. Of prime importance is the sensitivity of the process to impurities in the feed stream. There is no comparable separation of cationic impurities, such as iron and aluminum, in the precipitation of plutonium trifluoride, as there is in the precipitation of plutonium oxalate. The feed plutonium must therefore be of exceptional purity as compared to present requirements to insure final product quality. At Savannah River this process requirement is not a major limitation, since the product solutions from the main separations plants and the recovery plant are routinely processed through batch cation exchange systems prior to their precipitation. At HAPO, however, cation exchange units are not presently employed; product concentration is achieved by evaporation in the Redox and Purex plants and by refluxing in the Recuplex facility. While elimination of evaporator corrosion would probably result in a Purex product normally compatible with the trifluoride process, process revisions would be required for the Redox and Recuplex systems.

Other factors of concern include:

(1) The need for reducing the feed plutonium to the (III) valence state. At high plutonium concentrations an efficient valence reduction may be difficult, which would offset the filtrate waste losses. Moreover, storage of plutonium(III) nitrate solutions may be undesirable due to valence instability so that it would be preferable to perform the valence reduction step just prior to the precipitation step.

(2) The corrosion problems attending the use of aqueous hydrofluoric acid and nitric acid mixtures. Special materials of construction would be required for the precipitation and filtration equipment.

(3) The possible requirement of an alcohol wash for the plutonium trifluoride cake, creating a potential handling problem.

(4) The relatively less efficient reduction yield obtained with plutonium trifluoride as compared to plutonium tetrafluoride, and the greater quantity of iodine booster required in the reduction "bomb".

Weighing the advantages and disadvantages of the plutonium trifluoride process, it is apparent that the process is incompatible with the plant processing schemes presently employed at HAPO. Moreover, if the manyfold advantages of a continuous vs. a batch process are realized by the oxalate-fluorination process rather than by trifluoride process, the further advantages of the trifluoride process would probably not be great enough to warrant a later conversion of the installed equipment to the newer system.

This latter conclusion would most likely be true even if the present studies of the separations plants' product handling systems resulted in pure enough solutions that the trifluoride process was compatible. The expected throughput of the proposed Task I and II installations should provide more than enough capacity and operating reliability for presently anticipated production requirements during the service life of the installation. Continuous evaporation of the oxalate filtrate will minimize any effect the recovery operation would have upon Recuplex capacity, while elimination of filtrate recovery by the trifluoride process would not permit shutdown of the Recuplex facility or reduce operating expenses appreciably. Other forseeable operating cost differentials would not provide a sufficiently rapid payout (viz: less than five years) to warrant newer facilities. Under these conditions, obsolescence by a new technology within the service life of the proposed facility would probably require elimination of the use of fluorine or some similarly drastic step. The trifluoride process, however, would be quite similar to the present process in terms of equipment requirements (except for materials of construction). Thus, once the continuous Task I and II equipment for the oxalate-fluorination process is installed, the use of the plutonium trifluoride process in Z-Plant prior to 1962 would not appear economically advantageous.

From these considerations, it can be concluded that the conditions under which the plutonium trifluoride process could be advantageously employed at HAPO are a compatible product solutions from the main separations plants and the need for new wet chemistry facilities.

DELAYING CONSTRUCTION OF TASKS I AND II

The most immediate means by which the favorable conditions for employing the plutonium trifluoride process could be met would be to delay installation of the continuous Task I and II equipment for the oxalate-fluorination process until present studies on Purex tail-end processes and the trifluoride process are completed. Such a delay is considered in terms of development program and production commitment schedules presently envisaged for the next two and one-half years.

Based upon present estimates of prototype development programs and plant installation requirements, the production application of the plutonium trifluoride process at HAPO would not be feasible prior to January, 1959. The prime factor is the tail-end treatment studies being undertaken for the Purex plant. Results of these studies will lead to the installation in Purex of either an ion exchange system or an evaporator that will be relatively free of corrosion effects: either should provide an iron-free product solution suitable for feed to a trifluoride reactor. Installation of a new evaporator in the Redox plant, however, would have to be accompanied by a flowsheet revision or an ion exchange process that would eliminate the aluminum contamination from the third cycle scrub stream. The Recuplex facility, on the other hand, would definitely require an ion exchange system to remove aluminum. The tail-end package program in Purex will probably be completed by July, 1957. The technology of adapting the trifluoride process to a continuously operated system should be well developed by July, 1957.* Assuming a one and one-half year construction period for both installations, the process would be available for production by January, 1959. Since both development programs include the operation of plant-scale prototype equipment, little difficulty other than the normal "bugs" should be experienced upon start-up and relatively firm production commitment could be made shortly thereafter. If, more optimistically, the prototype ion exchange unit now planned for Purex should prove to be suitable for production operation, final compatibility of the separations plants and the trifluoride process would still have to await equipment installations in Redox and therefore could not be firmly anticipated much before January, 1959.

Plutonium production requirements, however, prevent such a long delay in the installation of Tasks I and II. In fact, a request has recently been made by the Manufacturing Department that the target date for completion of the Task I and II installation be revised from October, 1957 to March, 1957.**

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* H. H. Hopkins, Jr., Personal Communication, June 7, 1956.

** W. N. Mobley by J. G. Attanas, "Improved Task I and II Project (B-2222) Z-Plant", HW-43765, June 15, 1956

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The prime justification for the early installation of the continuous equipment is the present lack of production capacity to handle peak loads. Thus, although the estimated production requirement for FY 1958 could probably be processed through the present facilities on an average basis, the instantaneous processing capacity required because of variances in the separations plants' output exceeds present capacity limits. Means of reducing the peak loads would involve immediate capital expenditures or high operating costs. The installation of lag storage facilities for the Redox and Purex product solutions could presumably provide adequate capacity, but would involve new storage cans and vault areas. A second alternative would be to ship directly to Rocky Flats any plutonium output from the separations plants that exceeded the Z-Plant storage and processing capacity. This might necessitate continued operation of the 231 Building, although initial results of a product test for the direct concentration of Purex product solutions to 400 g/l appear very promising. Off-site shipment of nitrate, however, would upset plans of the AEC and Rocky Flats for the shipment of only metal product from HAPO. Any large cost of temporarily employing such schemes to reduce peak loads probably would not be recovered through the savings permitted by installation of the plutonium trifluoride process.

Secondary factors affecting delay of construction are: (1) the economic loss involved in not realizing the operating and maintenance cost advantages of a continuous system; and (2) the risk inherent in closely scheduling critical development programs for production start-up.

Delaying the construction of the continuous Task I and II equipment for the oxalate-fluorination process therefore does not appear attractive. Unless subsequent revisions in the plant's production objectives should require installation of a new wet chemistry facility - in the Purex plant, for example - the adoption of the plutonium trifluoride process at HAPO would offer no large-scale economic advantages prior to 1962.

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