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DECONTAMINATION OF REFRACTORY FUEL ELEMENTS - PART II

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INTRODUCTION

The possibility exists that nuclear reactions may be operated with fuels composed of refractory materials. Smith[1] in part I of this series of papers has outlined a decontamination process based on chloride volatility. It appears that another possible method which should be considered would include a single cycle extraction step as a decontamination method.

PROPOSED PROCESS

The complete flow sheet for this process is given in Fig. 1. The first step would be dissolution of the refractory material in acid to produce a feed compatible with tributyl phosphate extraction system. Research would be required on the dissolution step. Possibly mixed hydrofluoric and nitric acids could be utilized. This step would no doubt depend on the physical form of the material and its past history such as firing temperature. Highly irradiated refractories may suffer enough radiation damage to render them reactive to strong acids.

The feed adjustment step would simply be a hold tank where the concentration of the feed would be adjusted to the proper level. If fluoride ion were used in the dissolution step, aluminum nitrate would be added both as a complexing agent for the fluoride ion and also to provide salting strength for the extraction step.

Technology on extraction by TBP is well advanced as both the Purex[2] and Thorex[3] processes have been operated successfully. Thus, the main requirement for this step would be picking a suitable size column and flow sheet to perform the extraction. The same also applies to the stripping column where the uranium and/or thorium is re-extracted into dilute nitric acid yielding a solution of uranyl nitrate or thorium nitrate.

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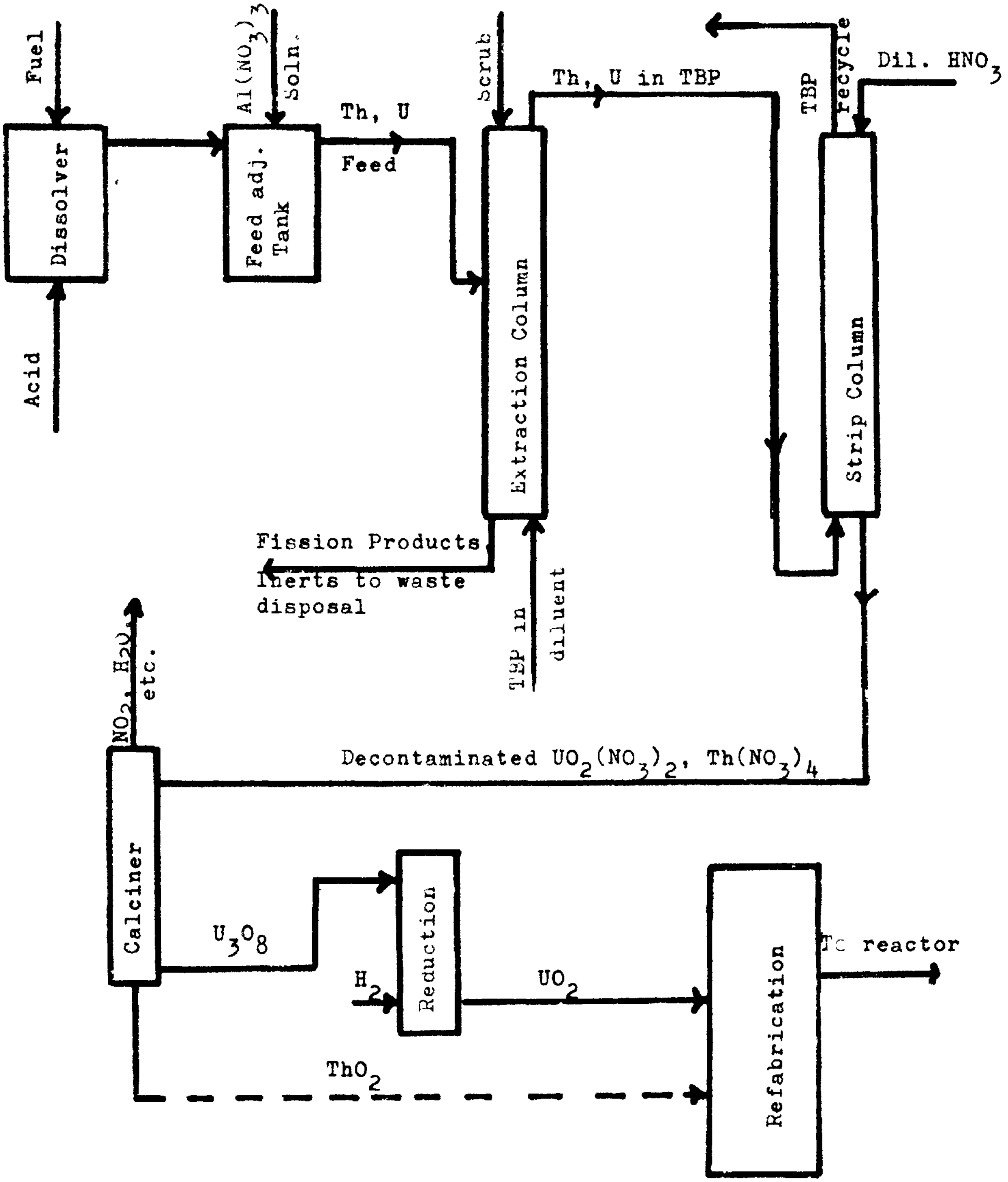

Fig. 1 Reprocessing Flow Sheet

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The solution would then be fed to a calciner where the material is fired to the oxide. The bulk of the oxides of nitrogen could be absorbed in water, concentrated and returned to the beginning of the process for dissolution, feed adjustment, etc..

If the product needs reenriching the required amount of U^{233} or $U^{235}O_2(NO_3)_2$ solution would be added in the calcining step.

If only a thorium solution is being dealt with, i.e. the bred U^{233} is separated in a partition column during the extraction cycle, then the ThO_2 product from the calciner would go directly to the refabrication step.

If a uranium fuel is being dealt with, the U_3O_8 product from the calciner would go through a hydrogen reduction step to produce UO_2 and thence to the refabrication step.

The refabrication step depends on the shape, composition, whether the material is fired, etc.. Probably, some blending and hot pressing would be required if some diluent such as MgO were employed. However, whatever operations were required could be carried out remotely.

A modest research and development program would be required to prove out such a flow sheet. The dissolution step would require the major effort. The program would be dependent on what types and compositions of refractories were considered. For example, a high fired, dense thoria compact would probably be difficult to dissolve.

In the extraction step a small amount of development would be necessary to prove out the extraction flow sheet. However, disposal of the waste stream from the extraction cycle would require some development work.

Other sites have studied and are studying the calcining of uranyl nitrate solution so the development effort on this point would be adaptation of remote methods.

The refabrication step would require the development of remote handling methods for whatever processing is done in this step.

To sum up the development effort required, a major effort would be required on the dissolution and remote calcination and refabrication steps and only a minor development effort on the actual extraction cycle.

One of the main disadvantages of the aqueous reprocessing methods for metallic fuels is that the decontaminated material has to be reduced back to the metal for refabrication. Using an oxide fuel, no metal reduction step is needed and hence, the overall process is simplified.

One possible advantage of an extraction step lies in the decontamination obtained. Ordinary aqueous plants have second and third extraction cycles to reduce zirconium and ruthenium activity in the metal to a point where direct refabrication is possible. In a single cycle, decontamination factors of 10^3 or better can be expected for the rare earths, and cesium and strontium. The main fission products remaining to any degree are zirconium and ruthenium. Thus, from the nuclear standpoint, the product would be quite satisfactory.

Although the oxide product from the calciner would be sufficiently radioactive to require remote handling, this should not be a serious problem. The remote refabrication of metallic fuels is already under consideration. The remote refabrication of oxide powders should not be as difficult as that for metallic fuels.

One disadvantage of this process is the production of an aqueous waste solution which would be highly contaminated. An evaluation of the effect of increased waste volume with subsequent increased storage costs on the overall economics of the process would be required.

This process appears to be simple enough to warrant consideration as a method for reprocessing and refabrication of refractory fuels.

CONCLUSIONS AND SUMMARY

A flow sheet based on a single extraction cycle has been presented for the reprocessing of irradiated reactor fuels composed of refractory materials. The advantages and disadvantages were discussed.

It is recommended that this process be included in the consideration of methods for decontamination of such fuels.

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1. Smith, T., HAA-SM-Memo-1961, May, 1957

2. Darby and Chandler, ORNL 1519, Feb. 1954

3. Wischow and Mansfield, ORNL 1944, June 1956

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