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DECONTAMINATION OF REFRACTORY FUEL ELEMENTS - PART II

By

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The possibility exists that nuclear reactions may be operated with fuels composed of refractory materials. Smith 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 and Thorox 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.
Fig. 1 Reprocessing Flow Sheet
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\textsuperscript{233} or
U\textsubscript{233}O\textsubscript{2}(NO\textsubscript{3})\textsubscript{2} solution would be added in the calcining step.

If only a thorium solution is being dealt with, i.e., the bred U\textsuperscript{233} is
separated in a partition column during the extraction cycle, then the ThO\textsubscript{2}
product from the calciner would go directly to the refabrication step.

If a uranium fuel is being dealt with, the U\textsubscript{3}O\textsubscript{8} product from the
calciner would go through a hydrogen reduction step to produce UO\textsubscript{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 thorium 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, 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.

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

3. Wischow and Mansfield, ORNL 1944, June 1956

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