

MASTER

THE SALT CYCLE PROCESS

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ABSTRACT

The Salt Cycle Process is a nuclear fuel processing approach designed to be applied in a compact facility located at the reactor site. Irradiated uranium dioxide fuels would be processed through a brief sequence of steps and partially purified uranium dioxide or uranium dioxide-plutonium dioxide powders recovered, suitable for refabrication into fuel elements.

The major steps of the process are the dissolution of uranium oxides in molten NaCl-KCl eutectic by chlorination to form soluble uranyl chloride and the reduction of uranyl chloride to uranium dioxide, which separates as a solid. The preferred method of reduction is the electrolytic method, which yields uranium dioxide as a conveniently handled, adherent deposit on the cathode.

Means are described for separation of plutonium from uranium and for co-deposition of the two. Also included are discussions of the probable nature of the uranium and plutonium species present in the molten salt, side reactions in which uranyl chloride may participate, and the influence of certain variables on the properties of the uranium dioxide produced.

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## THE SALT CYCLE PROCESS

by

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The AEC-sponsored Plutonium Recycle Program seeks to establish proven and economic technology for utilizing plutonium as an alternate for or to supplement uranium-235 as a fuel enrichment material in power reactors. This program is therefore somewhat unique among current nuclear power development programs in that the goal is not a specific reactor type but rather a plutonium fuel cycle technology which will be appropriate to a wide variety of thermal, heterogeneous reactors.

Research and development on fuel reprocessing approaches appropriate to this program has been pursued in the Hanford Laboratories Operation for several years. The Salt Cycle Process is the most recent development to emerge from this program. Because of the broad range of reactor and fuel types to which recycle of plutonium may have applicability, it is somewhat unlikely that a single fuel reprocessing approach will apply economically to all plutonium recycle applications. However, we believe that the Salt Cycle Process will be of interest to a large majority of such cases for the near future and for this reason have elected to present this summary at this time despite the fact that this work is only now progressing past the preliminary stage.

The Salt Cycle Process is what is generally termed a close-coupled process, i.e., it is designed to be applied in a plant located at the reactor site. In what we foresee as a typical application the feed to the process will consist of spent uranium dioxide fuel encased in one of the conventional cladding alloys. The objective is to separate the fuel material from the cladding, dissolve the fuel material, readjust the plutonium to uranium ratio to yield the desired reactivity in the material to be recycled, and recover oxide products suitable for remote refabrication into fuel elements. It may be noted that nothing has been said about separation from fission products. It goes without saying that a high degree of separation from fission products would be welcome. However, it must be recognized that the degree of fission product decontamination to be scught

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in the Salt Cycle Process (as in any close-coupled process reconciled to remote fuel element fabrication), must be established by economic considerations. Complete recycle of non-volatile fission products contained in fuel irradiated to 30,000 MWD/T would require that the enrichment in the recycled fuel be increased by perhaps 0.4 to 0.5 percent to over-ride the fission product poisons. Against this penalty must be balanced the cost of any additional operations included in the chemical process to improve fission product decontamination. We are perhaps optimistic in our hope that conditions can be found within the framework of the basic Salt Cycle Process which will yield sufficient decontamination from the rare earths, which constitute the most serious fission product poisons. In any event we are in the enviable position at the moment of having only very meager data on fission product behavior and are, therefore, free to speculate, unhampered by facts.

The first step in the Salt Cycle Process entails separation of the oxide fuel material from its cladding. We hope to do this by a mechanical method since this will make the remainder of the process relatively invulnerable to changes in the cladding alloy.

We plan to cut fuel rods into short sections, thereby exposing the oxide fuel, and then oxidize the  $UO_2$  to  $U_3O_8$  by treatment with air or oxygen at a controlled temperature. The gross change in crystal lattice which is accomplished by this reaction serves to convert  $UO_2$  ceramic shapes into a finely-divided  $U_3O_8$  powder which can be separated from the cladding remnants by mechanical sieving. Many of you will recognize this clad removal process as the one proposed and studied by L. A. Hanson (NAA-SR-3591, "Removal of Irradiated  $UO_2$  Fuel from the Cladding by Controlled Oxidation") at Atomics International. We have verified the workability of this process in small-scale work with both unirradiated and irradiated material and are currently employing it in hot cell experiments on a 200 gram scale.

The feed to the dissolving step thus comprises  $U_3O_8$  (with some unreacted  $UO_2$ ),  $PuO_2$ , and fission products. The reaction medium which we have used in almost all our work to date is an equimolar mixture of NaCl and KCl which melts at about 670 C. The uranium oxides can be dissolved into this medium in several ways. Certain of the dissolving reactions which we have briefly investigated are shown in the first two slides.

As indicated on the first slide, simply suspending a uranium oxide powder in NaCl-KCl and sparging with chlorine converts the oxide to species soluble in molten NaCl-KCl. The "relative rates" shown are simple ratios of the fraction of the powder sample dissolved in five minutes when (-100 +325) mesh fractions of the powders were treated in identical fashion with chlorine at 700 C. In the case of  $UO_3$  almost 80 percent of the sample was dissolved in five minutes whereas with  $UO_2$  only four percent was dissolved in five minutes and only 13 percent in 30 minutes. Quite apparently, roasting  $UO_2$  to  $U_3O_8$  prior to the dissolving step will greatly facilitate dissolution with chlorine.

Plutonium dioxide dissolves more slowly than uranium dioxide on chlorination in NaCl-KCl, as shown by the fact that when a solid solution of  $\text{PuO}_2$  in  $\text{UO}_2$  was subjected to chlorination under NaCl-KCl, uranium entered the molten salt more rapidly than did plutonium.

On the second slide are shown equations representing the reactions which are thought to occur when uranium and plutonium oxides are dissolved with hydrogen chloride. In general, rates of dissolution with hydrogen chloride are greater than with chlorine.

The formation of soluble uranyl compounds on chlorination of any of the uranium oxides in NaCl-KCl is indicated by the fact that no uranium species other than uranyl are detected in aqueous solutions prepared by dissolving samples of the crystallized salt phase. The situation when uranium oxides are dissolved into molten NaCl-KCl by treatment with hydrogen chloride is more complicated. With  $\text{UO}_3$  apparently only uranyl species are formed. The spectra of molten salt solutions prepared by dissolving  $\text{UO}_2$  with hydrogen chloride indicate that some of the uranium is present in solution as uranium(IV) and some in a form other than uranium(IV) or (VI). On dissolving  $\text{U}_3\text{O}_8$  with hydrogen chloride, spectra of the molten salt solutions indicate the presence of little if any uranium(IV), a species other than uranium(IV) or (VI), and perhaps uranium(VI). Although no uranium(IV) was observed in this molten salt solution, uranium(IV) was observed in aqueous solutions obtained on dissolution of samples of the crystallized melt. Because of the known disproportionation of uranium(V) in aqueous solution, it is inferred that a uranium(V) species is present in the molten salt.

The soluble plutonium species formed by dissolving  $\text{PuO}_2$  in NaCl-KCl by chlorination or hydrochlorination appears to be plutonium(III). Dissolution of  $\text{PuO}_2$  by treatment with hydrogen chloride and dissolution of  $\text{PuCl}_3$  in molten NaCl-KCl produced salt solutions which when crystallized and dissolved in dilute hydrochloric acid showed no evidence for any plutonium species other than plutonium(III). Similarly, prolonged chlorination of a melt prepared by dissolving  $\text{PuCl}_3$  in molten NaCl-KCl showed no evidence for species other than plutonium(III) when the melt was allowed to freeze and then dissolved in dilute hydrochloric acid.

The third step in the Salt Cycle Process as we envision it will be to readjust the ratio of plutonium to uranium to the degree necessary to establish the desired reactivity in the fuel to be recycled. The equations shown on the next slide detail some of the reactions which could in theory be used to effect separation of plutonium and uranium.

As indicated in the first series of equations the soluble uranyl species in molten NaCl-KCl can be reduced to insoluble  $UO_2$  with a wide variety of reductants. The reactions are essentially quantitative and the solubility of uranium dioxide sufficiently low that essentially quantitative precipitation of uranium should be possible. The work of M. C. Lambert at Hanford has established an upper limit of four milligrams of uranium per kilogram of salt for the solubility of  $UO_2$ . Actually, the true solubility of  $UO_2$  is probably considerably lower since the salt-soluble uranium detected in this experiment was entirely uranium(VI). ( $UO_3$  and  $U_3O_8$  have substantially larger solubilities - at least 66 g U/Kg salt for  $UO_3$  and 13 g/Kg salt for  $U_3O_8$ .) Since most of the reductants listed would not be capable of reducing plutonium(III), it should be possible to effect separation of uranium and plutonium by selectively reducing uranyl and precipitating  $UO_2$ , leaving plutonium in solution. We have verified that this is possible. For example, simply sparging a melt containing uranium(VI) and plutonium with ammonia results in precipitation of  $UO_2$ . However, we have not as yet been able to avoid carrying considerable amounts of the plutonium with the  $UO_2$  precipitate.

As indicated by these equations, we have found that simply sparging a plutonium-bearing salt solution with air results in essentially quantitative precipitation of plutonium as  $PuO_2$ . No reaction occurs with uranyl ion whereas if uranium(IV) is present in the solution it is simply converted to soluble uranyl species. This then represents a second approach whereby plutonium and uranium may be separated - in this case by selective precipitation of  $PuO_2$  while leaving uranium in solution. Again we have demonstrated that this is a potentially feasible approach. However, once again we have not been able to recover more than about 95 percent of the plutonium in this way. From the fact that the plutonium precipitate is very finely divided and settles poorly and the fact that plutonium recovery can be improved by providing a scavenger precipitate to carry the plutonium (for example, by precipitating a fraction of the uranium with ammonia), we suspect that this difficulty simply reflects our inability to make a clean separation of the very finely divided  $PuO_2$  from the salt melt.

Our preferred method for effecting separation of uranium and plutonium depends upon selective electrolytic deposition of uranium dioxide. As I indicated on the preceding slide, simple direct current electrolysis of a molten NaCl-KCl system containing the soluble uranyl species will result in deposition of uranium dioxide as an adherent deposit on a metallic or graphite cathode. The problem in separating uranium from plutonium is avoiding simultaneous deposition of a plutonium species (e.g.,  $PuC_2$ ) along with the uranium. If our inference as to the stable plutonium species in the melt is correct then this should be simple enough, the only requirement being that the potential be held below the discharge potential for plutonium(III).

This indeed proves to be the case. At a concentration of ten weight percent uranium in the melt, uranyl ion can be reduced to uranium dioxide at cell potentials as low as 0.5 volt and if the deposition is made under anhydrous and oxygen-free conditions plutonium can be maintained in solution in the salt phase. In a typical experiment employing a salt solution containing uranium and plutonium in a 300 to 1 ratio, 40 percent of the uranium was deposited as  $UO_2$  and this deposit contained only 0.3 percent of the plutonium. To effect this separation it suffices merely to dry the melt thoroughly with a dry chlorine and/or hydrogen chloride sparge prior to the deposition and to maintain a dry chlorine atmosphere over the cell during the deposition. A useful technique which was employed in the experiment I cited entails adding graphite powder to the melt during the chlorine sparge. This results in conversion of a portion of the uranyl ion in the melt to soluble uranium(IV) which then serves as a rather efficient oxygen getter during the electrolysis.

We have not attempted quantitative removal of uranium from plutonium by this approach. However, this should not be necessary in a plutonium recycle application. Our desire is simply to increase the plutonium to uranium ratio to the point where the remaining partially depleted uranium together with the plutonium will constitute a workable fuel. Thus, removal of 10 to 20 percent of the uranium might well suffice and it is rather unlikely that removal of more than 40 percent of the uranium would ever be necessary.

Thus we have chosen to focus our major development effort for the near future on selective electrodeposition of uranium dioxide as a means of effecting the reportioning of plutonium and uranium which will be necessary in a plutonium recycle operation.

The next step in the Salt Cycle Process entails recovery of the fuel material to be recycled to the reactor. In view of the present unsettled status of the plutonium recycle concept, we are again faced with an arbitrary decision. Conceivably, recycle of plutonium could be accomplished in either of two ways.

So-called plutonium "spike" elements consisting of plutonium, in admixture with inert material only, could be inserted in a reactor as integral fuel elements to supplement a  $UO_2$  fuel loading or mixed  $UO_2$ - $PuO_2$  fuel elements could be employed as the entire reactor loading. From the standpoint of chemical processing we favor the second alternative where mixed  $UO_2$ - $PuO_2$  fuel is received at the processing plant and mixed  $UO_2$ - $PuO_2$  fuel material is likewise the product.

The precipitation reactions which I summarized on an earlier slide could presumably be utilized to effect recovery of separated  $UO_2$  and  $PuO_2$  appropriate for a spike loading. For a uniform reactor loading in which all fuel elements consist of a solid solution of  $PuO_2$  in  $UO_2$  we favor an electrolytic deposition of a mixed plutonium-uranium deposit as a means of recovering the products to be recycled. For a reactor designed to handle a uniform mixed oxide fuel loading such an approach has obvious advantages. The products are recovered in a single step as a blended material thereby obviating any necessity for powder blending operation. Likewise, an electrolytic deposition enables the oxide product to be recovered as an adherent deposit on the cathode.

Thus molten salt filtration is avoided - the necessary phase separation is made simply by withdrawing the cathode from the molten salt solution.

To allow co-deposition of the plutonium with the  $UO_2$  it is only required to introduce oxygen into the atmosphere over the cell. We do not have sufficient information at present to permit a definitive conclusion as to the mechanism by which plutonium is incorporated in the  $UO_2$  deposit. The process has the appearance of a genuine co-deposition process in the sense that the concentrations of uranium and plutonium in the melt decrease in similar fashion (and under certain conditions at identical relative rates) throughout the period of the electrolysis. Without detailing the results of several attempts to elucidate this process, all of which fail to yield entirely conclusive results because of known complicating side reactions, let me simply describe a plausible mechanism which is consistent with all our experience with this system to date.

In this mechanism it is assumed that the presence of oxygen at the top of the melt results in precipitation of  $PuO_2$  as a finely-divided suspension in the melt. However, during the electrolysis chlorine is continually evolved from the anode and reaction of this chlorine with the precipitated  $PuO_2$  causes it to redissolve. If this were all that occurred then a steady state would be established such that a constant fraction of the plutonium would remain in solution and the remainder would be present as solid, suspended  $PuO_2$ . If, however, we also presume that  $PuO_2$  precipitated in the vicinity of the cathode can be assimilated into the  $UO_2$  deposit being formed on the cathode and thereby be protected from redissolution then this provides a means, and the only means, for removing  $PuO_2$  from the system. Continued electrolysis could then result in near-quantitative removal of plutonium from the melt, as we have observed. In short, then, the plutonium is precipitated as  $PuO_2$  by reaction with oxygen and the precipitated  $PuO_2$  is forced to localize in the  $UO_2$  deposit.

Quite obviously we have as yet done little toward elucidating this process and can only stipulate that we have arrived at conditions which do permit plutonium as some as yet undefined species to be incorporated into the  $UO_2$  deposit in a reproducible manner.

A process whereby the plutonium to uranium ratio is increased by first selectively depositing the excess uranium and then co-depositing the remaining uranium and essentially all the plutonium thus appears quite feasible.

Since the aim in the Salt Cycle Process is to recover the product oxides in a form suitable for refabrication into ceramic fuel elements with a minimum number of intervening steps, the nature of the  $UO_2$  product formed is of considerable concern. It would be ideal to obtain a deposit which, after simply rinsing and drying, had chemical and physical properties necessary to assure efficient compaction into fuel elements and proper behavior during subsequent irradiation.

Little is known as yet about the behavior of electrolytically produced  $UO_2$  in the various methods of compaction into fuel elements. In one test, cold-swaging of electrolytic  $UO_2$  resulted in a product of identical density (65 percent of theoretical) to that obtained on cold-swaging a commercial arc-fused  $UO_2$ . A program is currently in progress to determine the compaction properties of electrolytic  $UO_2$  samples prepared under various conditions. The methods of compaction of most interest here are swaging and vibratory compaction as it is felt that these two methods could be most easily adapted to operation by remote means.

The most readily apparent result of changing the conditions under which the electrolytic  $UO_2$  is formed is the physical appearance of the cathode deposit. In slide IV are presented examples of the wide variability one can obtain. On the one hand is seen a smooth, dense deposit while on the other is seen a dendritic, voluminous deposit. The variable which is apparently of prime importance in determining whether a smooth or a dendritic deposit is obtained is the extent to which the deposit is exposed to chlorine. In the assured absence of chlorine from the cathode region, very dendritic deposits are obtained. The absence of chlorine from the cathode region was assured by surrounding the anode with a sleeve closed at the bottom with a quartz frit on which was placed  $U_3O_8$  powder to react with chlorine before it could pass through the frit into the cathode chamber. As the conditions are altered to allow more chlorine to react with the deposit, smoother deposits are obtained. Under various conditions we have obtained deposits having nearly any degree of smoothness between the extremes shown in this slide.

Another property of electrolytic  $UO_2$  which can be quite sensitive to the extent to which chlorine is present in the region of the cathode is the oxygen to uranium ratio of the product. The oxygen to uranium ratios given in this paper were calculated from the relative amounts of uranium(IV) and uranium(VI) found by coulometric titration of aqueous solutions prepared by dissolution of  $UO_2$  samples, assuming three oxygens per uranium(VI) and two per uranium(IV). In smooth, dense deposits as shown on the preceding slide, oxygen to uranium ratios in the desired range of 2.00 to 2.01 have invariably been obtained. In very dendritic deposits prepared in the absence of chlorine in the region of the cathode, oxygen to uranium ratios as high as 2.25 have been obtained. Under certain conditions of electrolysis, however, dendritic deposits having oxygen to uranium ratios of 2.01 have been obtained.

In the dendritic-type deposits the oxygen to uranium ratios become lower as the temperature during the electrolysis is lowered and as the access of chlorine to the cathode region is increased. Of course, excessive access of chlorine leads to the smooth type of deposit. The combined effects of temperature and degree of access of chlorine on the oxygen to uranium ratio appear to be due to the fact that thermal decomposition of  $UO_2Cl_2$  in  $KCl-NaCl$  results in the formation of an oxide having an oxygen to uranium ratio very near to that of  $U_3O_8$ , as shown by R. E. Ewing at Hanford. It is felt that the  $U_3O_8$  formed by thermal decomposition of the  $UO_2Cl_2$  is incorporated in the  $UO_2$  deposit unless there is sufficient chlorine present to either prevent its formation or dissolve it once it is formed. The presence of  $U_3O_8$  in the  $UO_2$  deposit would, of course, increase the oxygen to uranium ratio of the deposit. The rate of decomposition to  $U_3O_8$  is lower at lower temperatures so, other conditions being equal, less  $U_3O_8$  can be incorporated in a  $UO_2$  deposit formed at a lower temperature.

The thermal decomposition of  $UO_2Cl_2$  to  $U_3O_8$  is one of the side reactions of considerable importance in the Salt Cycle system. Another side reaction which does occur when graphite electrodes are employed is the formation of  $UCl_4$  as shown on slide V. On this slide are also shown reactions involving  $UCl_4$  which we feel may be of considerable importance in connection with factors previously discussed. We know very little about the importance of these reactions since in all our electrolysis runs to date at least one of the electrodes has been graphite, thus allowing the formation of  $UCl_4$  to occur and making it impossible to separate the effects of chlorine and  $UCl_4$  on certain portions of the process. Thus, some reactions which have been attributed in previous discussion to chlorine may in reality involve  $UCl_4$  to a greater extent than chlorine. Examples of such possibilities shown on this slide are the dissolution of  $U_3O_8$  formed by thermal decomposition, the partial dissolution of  $UO_2$  deposits to form smooth as opposed to dendritic deposits, and the maintaining of plutonium as  $PuCl_3$  during the deposition of only  $UO_2$  from a melt containing both uranium and plutonium.

Perhaps surprisingly, the chemical purity of the  $UO_2$  powders does not vary greatly with the nature (smooth or dendritic) of the deposit. In  $UO_2$  obtained by deposition on graphite cathodes, carbon contamination of the order of 1000 ppm has been measured in the  $UO_2$  powder after removal from the cathode. Fortunately, platinum cathodes are usable in this melt and provide a means of bypassing this problem. Excluding carbon, the major impurities are the melt constituents sodium, potassium, and chloride, which are present in the 100 ppm range in apparently stoichiometric ratio. The densities of the electrolytic  $UO_2$  powders, as measured by helium displacement, have been in range 10.6 to 10.8. The specific surface of nearly all our preparations has been below the minimum value of  $0.01 \text{ m}^2/\text{gram}$  which can be measured in our BET apparatus. As an indication of the manner in which electrolytic  $UO_2$  crystals can build on one another and the size of the crystals, a photograph of the powder from a dendritic deposit is shown in slide VI. The maximum length of any of the crystals shown here is about 1 mm.

I mentioned earlier that we have very little information on fission product behavior in this system. However, we have performed a couple of small-scale co-deposition experiments with  $UO_2$  irradiated to about 1.24 atom percent burn up (of U-235) and cooled about ten months.

As expected, decontamination factors for the fission products varied rather widely. The rare earths, which we are most desirous of removing, proved the most difficult to remove, the decontamination factor being about two. At the other extreme the highly electropositive fission products such as cesium, barium and strontium were removed quite efficiently, the decontamination factors being of the order of 100. We have not yet studied the behavior of the fission products under electrolysis conditions designed to deposit  $UO_2$  but not  $PuO_2$ .

This summarizes the bulk of the background information we have so far acquired on this processing approach. By way of conclusion, I would like to show you one of several possible flowsheets for applying this process in close-coupled processing of spent  $UO_2$ - $PuO_2$  fuel from a uniformly loaded reactor recycling plutonium. Such a conceptual flowsheet is shown on slide VII.

The fuel is sectioned to expose the core material and the core material is separated from the clad material by oxidation to form  $U_3O_8$  powder and sieving. The  $U_3O_8$  powder, containing the plutonium and fission products, is suspended in molten NaCl-KCl and dissolved by chlorine or mixed chlorine-hydrogen chloride to form  $UO_2Cl_2$  and  $PuCl_3$ . The fraction of the  $UO_2$  to be removed to adjust the plutonium to uranium ratio is electrolytically deposited free of plutonium by maintaining a dry chlorine atmosphere over the cell during the deposition. This cathode is then withdrawn, air is introduced to form  $PuO_2$  (and oxidize any  $UCl_4$  to  $UO_2Cl_2$ ) and  $UO_2$  and  $PuO_2$  are co-deposited on a fresh cathode. The deposit is removed from the melt on the cathode and after cooling is mechanically removed from the cathode, washed, dried, and refabricated into fuel elements to be recycled to the reactor.

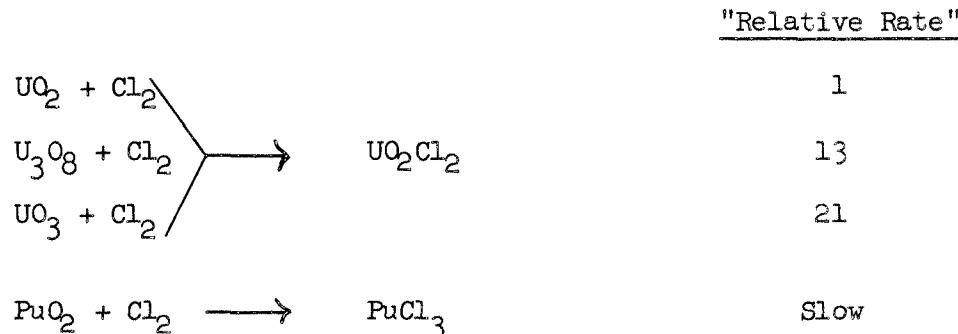
Features of this process which we consider attractive and which we hope to retain include the following.

1. Phase separation is effected by simply withdrawing the cathode and its adherent deposit from the melt. This avoids the necessity for molten salt filtration or centrifugation.
2. The desired mixed  $UO_2$ - $PuO_2$  product is formed as a "pre-blended" co-deposit. This obviates separate steps for plutonium and uranium and avoids any subsequent powder blending operations.
3. No impurities (other than fission products) are introduced into the molten NaCl-KCl salt. The salt is simply a vehicle in which the indicated electrodepositions are conducted. This makes it possible to consider re-use of the salt through multiple processing cycles. This introduces considerable flexibility in that quantitative product recovery is not mandatory in each processing cycle. Product left behind in one cycle is simply recycled to the next processing cycle.

We hope that this technique will also afford us sufficient flexibility that we will be able to effect a workable degree of separation from the rare earths.

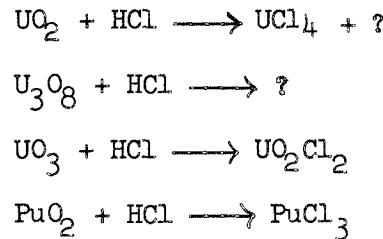
SLIDE I

DISSOLUTION OF OXIDE FUELS INTO NaCl-KCl  
WITH CHLORINE AT 700 - 800 C



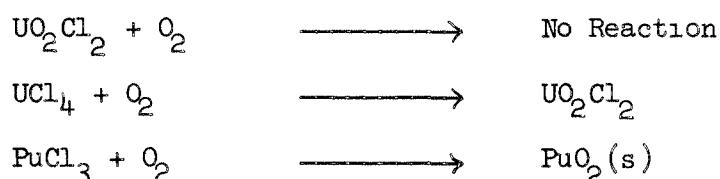
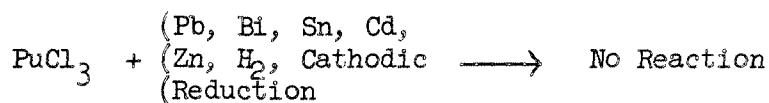
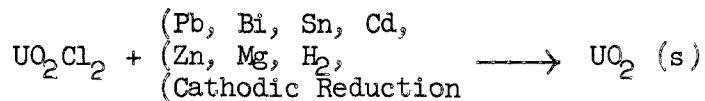
SLIDE II

DISSOLUTION OF OXIDE FUELS INTO NaCl-KCl  
WITH HYDROGEN CHLORIDE AT 700-800 C



SLIDE III

METHODS FOR SEPARATING U AND Pu IN NaCl-KCl AT 700-800 C



SLIDE IV

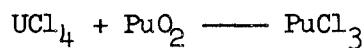
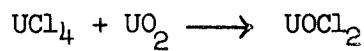
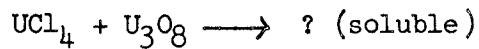
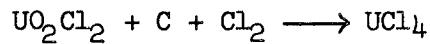
Pictures of Smooth and Dendritic  $UO_2$  Deposits

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SLIDE V

FORMATION OF  $UCl_4$  AND REACTIONS OF POSSIBLE IMPORTANCE

INVOLVING  $UCl_4$



SLIDE VI

Picture of  $UO_2$  Crystals

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SLIDE VII

SALT CYCLE PROCESS

GENERALIZED FLOWSHEET

