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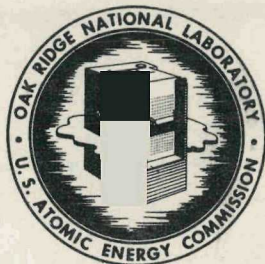
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FROM: R. E. Blanco  
D. E. Ferguson

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DATE MAR 13 1957  
For The Atomic Energy Commission  
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## SUMMARY

### Homogeneous Reactor Fuel Processing

Tracer experiments with ruthenium in simulated homogeneous reactor fuel solution showed that at 250°C ruthenium is soluble to the extent of 8 to 30 µg/kg H<sub>2</sub>O and is not absorbed on silver in the vapor phase above such solutions.

### Homogeneous Reactor Fuel and Blanket Studies

Hindered settling studies on samples of slurry from the 200A-5 run indicated that the addition of sulfate markedly affected the properties of the slurry at 300°C. At 1000 ppm the settling rate was > 4 cm/sec with a settled concentration of 1400 g Th/kg H<sub>2</sub>O and at 1800 ppm sulfate the settling rate was 0.3 cm/sec and settled concentration 600 g Th/kg H<sub>2</sub>O.

In LITR experiment number 24, a total of 178 hr of out-of-pile stirring, and 50 hr in the reactor have proceeded without incident. The relative viscosity has remained constant at 5 cstk over this period.

### Uranium-233 Isolation

When U-233 was eluted from 1 liter of Dowex-50 resin with acidified TBP-Amsco, 96.3% of the uranium was eluted at a concentration of 76 g/l. This uranium contained  $1.9 \times 10^3$  γ c/m/mg, which is a factor of 50 less than uranium eluted from the same resin with acetate-citrate.

### Mass-233 Studies

Carrier precipitation of Pa-233 from pilot plant AP solution was > 99.8% complete with both thorium hydroxide and thorium iodate.

Solvent extraction of the acetate-citrate uranium product gave decontamination for ruthenium of  $10^3$ , gross  $\gamma$ -40 and an AW loss of 0.2%. Thorex used solvent was decontaminated by contact with activated charcoal, alumina, or soda-lime.

#### Fused Salt Dissolution

Different specimens of zirconium and Zircaloy-2 exhibit different dissolving rates in some cases, but the differences are small compared to experimental error. ~~SUR~~ specimens appear to dissolve slowly in  $\text{NaF} \cdot \text{ZrF}_4$  when exposed to dried nitrogen gas, in the absence of HF gas.

There is only a small dependence of dissolution rate on temperature.

#### Excer

Simulated ore leach has been run on the 2-in. contactor with a consumption of 2-1/2 lbs of sodium chloride per pound of uranium and a maximum product concentration of 98 g U/l.

#### Ion Exchange Studies

##### TBP Treatment

Waste TBP may be decontaminated by ion exchange by a factor of 20.

#### Heterogeneous Reactor Fuel Reprocessing

##### Chloride Removal from Dissolver Solutions

About 95% of the chloride in stainless steel dissolver solutions was removed and recovered as HCl in a continuous stripping column when the feed was  $< 0.6 \text{ M}$  in chloride.

##### Dissolution of Zirconium

Zirconium metal can be dissolved rapidly in a 900% excess of boiling 15 M sulfuric acid. Elemental sulfur is formed during a part of the

reaction.

Uranium silicon and uranium molybdenum alloys have low dissolution rates in sulfuric acid but are rapidly dissolved in nitric acid.

#### Hydrochlorination

About 38% of the  $\beta$  and 45% of the  $\gamma$  fission activity sublimed with the  $ZrCl_4$  when a section of irradiated STR fuel element was hydrochlorinated.

#### Feed Materials

##### Hermex

The rate of dissolution and solubility of uranium in 3 M sodium amalgam are about one-half that in mercury but the stability of the resulting amalgam to oxygen is greater.

##### Metallex

A rough correlation between the free voidage of sintered thorium metal and its oxide content is indicated.

Methanol and petroleum ether from which dissolved oxygen has been removed have less oxidative effect upon thorium amalgam than water with or without dissolved oxygen.

##### Mice

An apparatus for creating temperatures of 20-40,000<sup>o</sup>K for use in high temperature U and Pu chemistry studies has been assembled and tested.

##### Fluorox

The specific surface area of  $UF_4$  appears to be constant over the temperature range 25-600<sup>o</sup>C. Above 600<sup>o</sup>C the specific surface area

decreases as sintering begins.

Flowing oxygen at 200°C for 5 hours failed to remove carbon from UF<sub>4</sub>-C pellets.

#### Dissolution Studies

The mercury-catalyzed dissolution of U-Al alloy in nitric acid is well known to be very complex. Additional work has been done to elucidate the mechanisms of dissolution.

A small vial of Nd<sub>2</sub>O<sub>3</sub> was shown to be attracted with a force of very roughly 1/40 that of gravity when suspended near a 10 kg permanent magnet.

#### Volatility Studies

The settling behavior and solubility of NiF<sub>2</sub> was observed to be about the same in the molten NaF-ZrF<sub>4</sub>-UF<sub>4</sub> (50-46-4 mole %) system as in uranium-free salt. Passage of gaseous HF through molten NaF-ZrF<sub>4</sub>-UF<sub>4</sub> containing 2 wt % Ni resulted in NiF<sub>2</sub> concentrating at the top.

## 1.0 HOMOGENEOUS REACTOR FUEL PROCESSING (R. A. McNees)

### 1.1 Fission Product Chemistry (S. Peterson)

Repetition in triplicate of various iodine experiments has clearly indicated that some variable in the experimental work has not been under control. Efforts are being made to determine the nature of that variable and bring it under control.

From 0.02  $\underline{m}$   $\text{UO}_2\text{SO}_4$ , 0.005  $\underline{m}$   $\text{H}_2\text{SO}_4$  solutions ruthenium precipitates almost completely at 250°C. Ruthenium remaining in solution ranged from 8 to 30  $\mu\text{g}/\text{kg H}_2\text{O}$  when the original ruthenium was varied from  $3.6 \times 10^{-5} \underline{m}$  to  $1.55 \times 10^{-4} \underline{m}$ . The original solutions were also  $2.8 \times 10^{-5} \underline{m}$  in  $\text{HIO}_3$ . These experiments were conducted in pyrex ampoules containing silver in the vapor phase. Less than 0.02% of the ruthenium appeared on the silver.

## 2.0 HOMOGENEOUS REACTOR FUEL AND BLANKET STUDIES (J. P. McBride)

### 2.1 Laboratory Studies (V. D. Allred)

Indication of the changes that can be introduced into a slurry by temperature changes and sulfate addition has been shown by high temperature hindered settling studies on the slurry now being circulated as run 200A-5 at Y-12. Typical data at 300°C showed settling rates of greater than 4 cm/sec and a settled bed concentration of 1400 g Th/kg  $\text{H}_2\text{O}$ , and 0.3 cm/sec and settled concentration of 600 g Th/kg  $\text{H}_2\text{O}$  for this oxide with 1000 ppm and 1800 ppm of sulfate respectively. A systematic study of the effect of additive concentration with temperature upon the settling characteristics of slurries has been initiated.



## 2.2 Irradiation Studies (A. R. Jones, N. A. Krohn)

LITR experiment number 24 was inserted on 5/16/56. Oxygen pressure of 250 lb was added. Stirring had proceeded without incident out-of-pile for about 178 hr. During this period the viscosity remained constant at 5 cstk (last week's report of 9 cstk was erroneous). The windings show no deterioration.

The bomb has now stirred well for 50 hr in-pile (25 hr at full power). No viscosity change is evident. No gas buildup has been noticed.

## 3.0 URANIUM-233 ISOLATION (R. E. Leuze)

### 3.1 Elution with Acidified TBP-Amsco (R. D. Baybarz, W. E. Tomlin)

One liter of Dowex-50 resin loaded with U-233 was obtained from the Thorex Pilot Plant. The resin was washed with 42-1/2% TBP in Amsco to displace the water. Uranium was then eluted with 42-1/2% TBP-Amsco containing 1.6 N HNO<sub>3</sub>. After a displacement volume of 400 ml, 98.6% of the uranium was eluted in about 4 column volumes. The product cut was composed of 1820 ml of organic phase and 81 ml of aqueous phase. The organic phase was 0.14 N HNO<sub>3</sub> and contained 96.3% of the uranium at 76 g/l. Gamma activity in this uranium was  $1.9 \times 10^3$  cts/m/mg. The aqueous phase was 0.45 M HNO<sub>3</sub> and contained 2.3% of the uranium at 40 g/l. Gamma activity in the uranium was  $5 \times 10^4$  cts/m/mg. Uranium eluted from some of the same resin with the acetate-citrate elutriant contained  $1.2 \times 10^5$   $\gamma$  c/m/mg or a factor of 50 more than the uranium eluted with acidified TBP-Amsco. The gamma activity in the TBP product was essentially all ruthenium.

In a small scale laboratory experiment, uranium eluted with acidified TBP-Amsco from Dowex-50 resin loaded with uranium, thorium, and aluminum contained 70% of the thorium and 3% of the aluminum.

About 45 ml of Dowex-50 resin was loaded with 8.7 g U, 0.25 g Th, and 0.2 g Al. After elution with acidified 42.5% TBP in Amsco, the eluate contained 27.6 mg U/ml, 0.56 mg Th/ml, and 0.02 mg Al/ml.

#### 4.0 VOLATILITY STUDIES (G. I. Cathers)

##### 4.1 NiF<sub>2</sub> Behavior in NaF-ZrF<sub>4</sub>-UF<sub>4</sub> System (M. R. Bennett)

The sedimentation of NiF<sub>2</sub> in the NaF-ZrF<sub>4</sub>-UF<sub>4</sub> system (50-46-4 mole %) was found to be similar to that in the NaF-ZrF<sub>4</sub> (50-50 mole %) system. At an initial Ni concentration of 2 wt % settling was incomplete at 48 hr due probably to the same viscosity effect observed previously. At 1 and 0.5% Ni, settling was faster, being fairly complete after 48 hr. In all tests the Ni concentration in the zone of complete settling varied over the range of 0.2-0.3 wt %, indicating that NiF<sub>2</sub> has approximately the same solubility in both the uranium-bearing and uranium-free systems.

Passage of gaseous HF for 6 hr through a NiF<sub>2</sub> dispersion in NaF-ZrF<sub>4</sub>-UF<sub>4</sub> at 600°C resulted in partial flotation of the NiF<sub>2</sub> to the top of the material. The Ni concentration at the top was 4.1 wt % in contrast to the initial concentration of 2.0%. This test was carried out in attempting to duplicate the sludge formation encountered in two other cases. Although the flotation effect was opposite to that being sought, the increase in the degree of heterogeneity occurring in both cases could be related. In a second test with HF being present as a blanket gas for 6 hr the NiF<sub>2</sub> settled downward at the slow rate normal in all tests where a N<sub>2</sub> blanket was used. It is possible that the flotation effect in the first test was due to the combination of agitation and the presence of HF.

5.0 MASS-233 STUDIES  
(R. H. Rainey)

5.1 Thorex U<sup>233</sup> Product (R. H. Rainey)

A tentative Thorex uranium-233 product specification, that a hard shell cutie pie shall not read over 300 mr in contact with the product container, has been set to allow continuous processing of the material in the peroxide precipitation facility at Los Alamos. Some of the recent products have not met this specification. In order to obtain additional decontamination when needed, a second solvent extraction using 15% TBP has been proposed. Since the first cycle product is eluted from the ion exchange resin as acetate-citrate, the extraction of the uranium from this material was investigated by laboratory batch countercurrent experiments. The ruthenium DF was about  $10^3$ , gross  $\gamma$ -40, and uranium loss to AW 0.2%.

5.2 Thorex Solvent Recovery (R. G. Mansfield)

The use of activated charcoal, alumina, and soda-lime both in slurry and in packed columns for cleanup of pilot plant solvent have given decontamination factors from 10 to  $10^3$ .

5.3 Protactinium Separation (A. B. Meservey)

Two methods for the concentration of protactinium from Thorex A-column waste (AP) are being evaluated on daily samples from the hot waste itself, containing about  $10^8$  gamma counts/ml/min from Pa. The methods feature carrier precipitation of the Pa on either thorium hydroxide (from AP solution containing excess NaOH) or thorium iodate (from the addition of iodic acid to AP solution). In the caustic method, over 99% of the fission products and 99.8% of the Pa appear in the precipitate. The iodate precipitate carries half to two-thirds of the fission products and over 99.9% of the Pa. With both methods, the

thorium normally present in the AP solution (0.1 to 1 g/liter) was sufficient for the scavenging operation.

#### 6.0 FUSED SALT DISSOLUTION (R. G. Wymer)

A survey of about 30 ~~STR~~ dissolutions made in  $\text{NaF}\cdot\text{ZrF}_4$  at  $600^\circ\text{C}$  using the HF lift pump dissolver indicates that there is a considerably smaller dependence of dissolution rate on HF flow rate than was found when HF was introduced into the melts through a straight nickel pipe. The rate changes from about  $4 \text{ mg/min-cm}^2$  at 25 mg HF/min to about  $5 \text{ mg/min-cm}^2$  at 90 mg HF/min. The smaller dependence may probably be attributed to the much greater agitation obtained using the pump, even at the lower flow rates. It is well known that the rates of diffusion-controlled reactions are enhanced by agitation up to a point, beyond which increased agitation is not very beneficial. In any case, the particular geometry is important, and scaling to other sizes is difficult.

The efficiency of utilization of HF gas has a strong dependence on HF flow rate, varying from about 60% at 25 mg HF/min through the HF lift pump dissolver down to about 20% at 90 mg HF/min.

Dissolution rates are not very reproducible, and factor-of-two differences are common on duplicate experiments where the only difference is that a new piece of STR element is used. This suggests that perhaps metal property differences are very important. Experiments with Zircaloy-2, obtained from R. A. McNees, and crystal bar zirconium, from C. E. Schilling, show that the dissolution rates are within the scatter of the STR data for the crystal bar specimens, but that the Zircaloy-2 specimens are probably more reactive than the STR specimens by a factor of 1.5 to 2.

Experiments have been performed at both 700 and 800°C for a range of HF flow rates, but the data scatter is such that a meaningful determination of the energy of activation for the dissolution is not possible. It is certain, however, that the energy is small, in accord with the requirements of diffusion controlled reaction mechanisms.

The results of three experiments performed at 600°C using a melt which had a NaF:ZrF<sub>4</sub> mole ratio of 65:35 indicate that the dissolution rate at constant HF flow rate may be somewhat higher than with the usual 1:1 mole ratio, though not much.

A surprising observation is that STR specimens will dissolve at the rate of 1 mg/min-cm<sup>2</sup> in a 1:1 melt at 600°C when dried nitrogen gas is bubbled through the melt. A melt was degassed by passing dried nitrogen through it for 2 hr. The nitrogen had been dried by passing it successively through Drierite, concentrated sulfuric acid, and copper turnings heated to ~ 550°C. After degassing the salt, the STR specimens were lowered into the melt from the space above the melt, where they had been held in the lift pump dissolver, and the nitrogen was passed over them in the same manner as though HF were being used.

7.0 EXCER  
(I. R. Higgins)

Conductivity instruments gave excellent continuous control of the 2-in. continuous contactor using the 5 M NaCl-water elution flowsheet. Better control of the flow rates permitted a reduction in the amount of NaCl required for conversion from the sulfate to chloride forms. Consequently, the need for recycling NaCl, and the attendant removal of sulfate from the NaCl recycle stream by precipitating CaSO<sub>4</sub> with CaCl<sub>2</sub>, was eliminated. In 3 ft of column a 100% excess of NaCl over theoretical was required to reduce the sulfate ten-fold. This is a consumption of 2-1/2

lbs of NaCl per pound of uranium or less than the cost of CaCl<sub>2</sub> proposed previously. The Na<sub>2</sub>SO<sub>4</sub>·NaCl waste containing from 3 to 15% of the uranium is recycled to the ore feed slurry. The chloride concentration is < 0.1 M in the feed and is not sufficiently high to interfere with loading. The uranium recovery was > 99% on 3 ft of Permutit SK regular at a loading of 50 g of U/l of resin. Product concentrations varied depending on how much is allowed to build up in the column, but for one hour a product solution containing 98.1 g/l of U as UO<sub>2</sub>Cl<sub>2</sub> was obtained.

### 8.0 ION EXCHANGE STUDIES (I. R. Higgins)

#### 8.1 TBP Clean-Up (W. E. Shockley)

TBP from Thorex, after standard solvent treatment, contains considerable activity, 10<sup>4</sup> gross  $\gamma$  and 10<sup>3</sup> gross  $\beta$ . A demineralization ion exchange type purification was tried because ion exchange is an efficient method of removing trace contaminants from process streams.

Contaminated Thorex TBP was fed first to a H-form cation column, then to a OH-form anion column at a rate of 20 ml/min/cm<sup>2</sup> using 50 volumes of feed to one volume of resin.

	Decontamination Factors	
	<u>gross <math>\gamma</math></u>	<u>gross <math>\beta</math></u>
Cation	5.2	3.5
Anion	3.2	5.8
Total	17.0	20.0

### 9.0 HETEROGENEOUS REACTOR FUEL REPROCESSING (J. E. Savolainen)

#### 9.1 Chloride Removal from Dissolver Solutions (M. L. Hyman)

A series of scouting runs for continuously stripping Cl<sup>-</sup> from

stainless steel dissolver solution ( $\text{HCl-HNO}_3$ ) has been made. The apparatus consists of a vacuum jacketed distillation column packed with 1/8-in. glass helices. Cold dissolver solution is introduced as feed at the top of the column. A reboiler supplies  $\sim 15.2 \text{ N HNO}_3$  vapor to the column and also receives the stripped bottoms ( $\text{Cl}^-$  free dissolver solution). Overhead is taken off and condensed as the distillate with almost no reflux returned to the column. The results for various runs are given in Table 1.

Decomposition and subsequent loss of  $\text{Cl}^-$  in the feed is almost eliminated if the  $\text{Cl}^-$  concentration in the feed is  $\sim 0.6 \text{ M}$  or less. The distillate/feed ratio, which is a measure of the dilution effect in the top part of the column, undoubtedly also affects  $\text{Cl}^-$  decomposition. Other runs, not reported in Table 1, suggest that distillate/feed ratios less than 2, and also reflux ratios in the range 3 to 8, may further the decomposition of  $\text{Cl}^-$ .

When using  $15.2 \text{ M HNO}_3$  (azeotrope) in the reboiler, and a distillate/feed ratio of 2, the acidity in the upper part of the column is  $\sim 7.6 \text{ N}$ . Apparently at this acidity the ferric ion-chloride complex reported by J. E. Savolainen is broken without oxidation of the  $\text{Cl}^-$ , provided that the actual  $\text{Cl}^-$  concentration in the column is  $\sim 0.3 \text{ M}$ .

A more systematic study of the many parameters in this system is under way. Variables to be investigated include feed conditions, distillate/feed ratio, reflux ratio, reboiler stripping vapor composition, distillate composition, and the geometry of the contiguous stripping column itself.

## 9.2 Hydrochlorination (J. E. Savolainen)

A section of ~~SFR~~ fuel element was irradiated for one week in the X-10 graphite reactor and cooled for three weeks. The specimen was then hydrochlorinated at about  $330^\circ\text{C}$ . The residue, the  $\text{ZrCl}_4$  sublimate, and

Table 1

Run No.	Feed Conditions			Distillate		Pot	Distillate/ Feed Ratio	Unrecovered Cl <sup>-</sup> , % <sup>**</sup>
	<u>N</u> H <sup>+</sup>	<u>M</u> Cl <sup>-</sup>	Metal* g/l	<u>N</u> H <sup>+</sup>	<u>M</u> Cl <sup>-</sup>	Cl <sup>-</sup> , ppm		
63	4.4	0.32	58	11.1	0.148	<20	2	6
67	0.60	0.61	18	8.5	0.292	55	2	4
68	0.30	0.30	9	8.1	0.146	<5	2	3
64	1.2	1.2	35	8.6	0.26	<15	2	56
65	1.2	1.2	35	4.0	0.46	35	1.1	58
66	2.4	2.4	70	9.4	0.45	210	2	63

\*Fe, Ni, and Cr from stainless steel.

\*\*"Unrecovered Cl<sup>-</sup>" is that not accounted for by a material balance on the column and which was lost as an off-gas.



the solution from the water trap used to remove the unreacted HCl from the gas stream were analyzed. The distribution of the  $\beta$  and  $\gamma$  activity was as follows:

	<u>% of <math>\beta</math></u>	<u>% of <math>\gamma</math></u>
Hydrochlorinator residue	60.8	53.3
ZrCl <sub>4</sub> sublimate	38.1	45.3
HCl water trap	1.1	1.3

9.3 Dissolution of Uranium and Uranium Alloys in HNO<sub>3</sub> or H<sub>2</sub>SO<sub>4</sub> Solutions (A. H. Kibbey)

The dissolution rates of 0.5 and 1% Si and also 10-12% Mo-uranium alloys and uranium metal in boiling 6 M sulfuric acid were determined. The dissolution rates of the silicon alloys were also determined in boiling 8 M HNO<sub>3</sub>. Dissolution times of 3 to ~10 min were used in making the determinations. Samples of the 0.49 and 1.16% Si-uranium alloys were dissolved to completion in boiling 8 M HNO<sub>3</sub>. The silica residues were removed and washed by centrifugation. The residues for the respective alloys were found to contain 0.017 and 0.040% of the total uranium accounted for. These values for uranium loss are within the specifications for processing enriched material. These results together with pertinent earlier work are summarized in Table 2.

Table 2

U	Alloy Composition %		Dissolvent	Reaction time-min	Dissolution Rate mg/cm <sup>2</sup> /min
	Si	Mo			
100	---	---	6 M H <sub>2</sub> SO <sub>4</sub>	10	0.032
88-90	---	10-12	6 M H <sub>2</sub> SO <sub>4</sub>	10	0.0079 (wt gain)
99.51	0.49	---	6 M H <sub>2</sub> SO <sub>4</sub>	10.5	0.043
99.51	0.49	---	2 M HCl-5 M HNO <sub>3</sub>	8.0	7.60
99.51	0.49	---	8 M HNO <sub>3</sub>	3.0	58.15*
98.84	1.16	---	6 M H <sub>2</sub> SO <sub>4</sub>	10.25	0.064
98.84	1.16	---	8 M HNO <sub>3</sub>	3.0	76.74*

\*These rates drop off rapidly as dissolution proceeds, presumably owing to formation of an adherent layer of SiO<sub>2</sub> on the surface of the specimen.

9.4 Dissolution of Zirconium Metal in Sulfuric Acid (A. H. Kibbey)

Investigation of the dissolution of zirconium in  $H_2SO_4$  solutions done at Idaho indicated that reflux with 14 M  $H_2SO_4$ , present in 900% stoichiometric excess, is the optimum dissolution condition. At concentrations of 15 M or greater, passivation of zirconium occurs and at concentrations of 13 M or lower, the reaction rates are too slow to be practical.\* Other work on the dissolution of zirconium in  $H_2SO_4$  solutions, reported by Lustman and Kerze,\*\* indicate that the dissolution rate increases with increasing acid concentration over the range 9.17 to 15.4 M. These data are summarized in Table 3.

Table 3

Dissolution Rate of Zr in Boiling  $H_2SO_4$

<u>w/o <math>H_2SO_4</math></u>	<u>M <math>H_2SO_4</math></u>	<u>mils/yr</u>
85	15.4	Disintegration in 1 hr
82.5	14.8	570
80	14.1	270
75	12.8	30.3
70	11.5	0.56
60	9.2	0.88

A qualitative dissolution experiment was made using the optimum Idaho dissolution conditions described above. The metal used was dummy STR fuel, i.e., Zircaloy-2 clad, natural U-Zr alloy. The overall average dissolution rate determined for the sample was 38.8 mg/cm<sup>2</sup>/min. The core material showed selective attack, indicating a much faster dissolution rate than that of the Zircaloy-2 cladding. A suspension of finely divided black solid material was obtained which will be submitted for

\* IDO-14035.

\*\* "Metallurgy of Zr," National Nuclear Energy Series, Vol. VII-4, p. 690 (McGraw-Hill Publishers).

x-ray-diffraction analysis. In addition, free sulfur was obtained as a product of the reaction.

10.0 FEED MATERIALS  
(O. C. Dean)

10.1 Hermex (E. Sturch)

Studies of the dissolution of uranium in boiling Na-amalgam were carried out to determine the effect of additives to mercury on the dissolution of uranium. The rate of dissolution of U in Na-amalgam is slower than in pure mercury and the solubility is less. On the basis of qualitative results the U-Na-amalgam appears to be more stable to oxidation than the U-amalgam.

Dissolutions of a 8.2 cm<sup>2</sup> slug of uranium in 100-ml portions of ~3.5 M Na-amalgam gave the following results:

<u>Time (min)</u>	<u>U Dissolved (g)</u>	
	<u>1</u>	<u>2</u>
15	0.788	0.977
30	1.967	1.855
60	3.393	3.113
120	3.705	3.307

Each dissolution was carried out under an atmosphere of argon and using a fresh 100-ml portion of Na-amalgam. Following the dissolution, the uranium slug was washed in HNO<sub>3</sub>, water, and methanol, and dried and weighed.

On the basis of present data, the maximum solubility of uranium in ~3.5 M Na-amalgam is about 0.3% by weight. A U-Na-amalgam of this composition has been kept essentially unoxidized for 5 days in a glass-stoppered graduate with an initial argon flush.

10.2 Metallex (O. C. Dean)

There is a rough correlation between atmosphere-connected voidage in sintered thorium metal and the  $\text{ThO}_2$  content. In a series of metal samples having voidage percentages ranging from 23 to 48%, the  $\text{ThO}_2$  content was 2.07 to 3.48%, while those having voidages of 7.7 to 14.8% had  $\text{ThO}_2$  contents of 1.15 to 1.67%. These results were based on only 7 metal samples, and the correlation was not good. More evidence is needed for a firm conclusion.

The oxidative effects of 5 wash liquids for thorium quasi-amalgam have been studied. The results are presented in Table 4.

Table 4

Effect of the Oxygen Content of Washing Agents on Thorium Amalgam

<u>Wash Liquid</u>	<u>Thorium Dissolved from the Thorium Amalgam (%)</u>
Distilled water, no treatment	0.62
Argon-sparged distilled water	0.26
Distilled water, distilled from potassium pyrogallate under argon	0.42
Methanol, distilled from potassium pyrogallate under argon	0.07
Petroleum ether, sodium-treated	0.08

The 5 amalgam samples, each weighing 135 grams and containing initially 4 grams of thorium, had been previously washed in 1 N HCl and distilled water. They were agitated at the same rate under 10 ml of each of the liquids described in the above table for 40 hours in an argon atmosphere. The results show that methanol which has previously had dissolved oxygen removed by distillation from potassium pyrogallate in an inert atmosphere had the least oxidative effect on the amalgam, and that petroleum ether which had been deoxygenated by agitation with

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sodium metal was nearly as inert. Reduction of the oxygen content of the water reduced its oxidative effect, but did not eliminate it. Evidently water itself has a corrosive effect.

11.0 MICE  
(R. G. Wymer, L. M. Ferris)

It is desired to study the chemistries of uranium and plutonium as they would be after the metals had been exposed to temperatures in excess of one million degrees Kelvin in various media. A collection of available capacitors connected in parallel and attached to a high voltage source has made it possible to discharge  $\sim 25$  calories through fine wires in about 3 microseconds. If the heat required to just vaporize the size wire used is less than 25 calories, the excess energy goes into kinetic energy of the atomized metal. In this way, temperatures estimated at 20-40,000<sup>o</sup>K have been attained in Ni and Pt wires.

It is planned that similar uranium wire explosions will be carried out in air, water, and NaCl, and that chemical studies of the resultant uranium will be made. It is felt that results thus obtained will give insight into the chemistry which may be expected from exposures to the higher temperatures of interest.

12.0 FLUOROX  
(R. G. Wymer)

12.1 Carbon Removal from UF<sub>4</sub>-C Pellets (L. M. Ferris)

Data presented in CF memo 56-2-161 suggest the possibility of removing  $\sim 75\%$  of the residual carbon from UF<sub>4</sub> by prolonged reaction with dry O<sub>2</sub> at 200<sup>o</sup>C. However, recent experiments show that little or no carbon is removed at this temperature in reaction times up to 5 hr.

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## 12.2 Surface Area Measurements (L. M. Ferris)

A systematic investigation of the variation of the specific surface area of  $UF_4$  with temperature and time has been undertaken, since it is felt that the rate of oxidation of  $UF_4$  is at least partially controlled by the  $UF_4$  surface area available. If this proves to be the case, then knowledge of surface areas will be essential in comparing oxidation reactivities of various  $UF_4$  samples. The studies to date have been performed by heating 98%  $UF_4$  (2%  $UO_2F_2$ ) in dry helium for 1 hr at various temperatures. The apparent specific surface is essentially constant up to a temperature of  $600^\circ C$  at which point sintering begins to occur. For one type of  $UF_4$  the specific surface area remains at  $\sim 0.3$  meter<sup>2</sup>/g over the temperature range  $25-600^\circ C$  and then falls to  $\sim 0.1$  meter<sup>2</sup>/g at  $800^\circ C$ . Since this  $UF_4$  contains some  $UO_2F_2$  initially, and will be coated with increasing amounts as the oxidation proceeds, work is under way to evaluate the effect of varying amounts of  $UO_2F_2$ .

## 13.0 DISSOLUTION STUDIES (R. G. Wymer)

The mercury-catalyzed dissolution of U-Al alloy in nitric is well-known to be very complex. Additional work has been done to elucidate the mechanism of dissolution. Table 5 summarizes observations which have been made on the dissolution of extruded miniature 15% U-Al alloy slugs in boiling 8 M nitric acid both in the presence and absence of mercury.

Table 5

0.005 M Hg Used	Gas Used	Sample in Gas Pump	Pt Screen	Dissolution Rate (mg/min-cm <sup>2</sup> )
Yes	---	---	Yes	0.36
Yes	NO	---	Yes	0.38
Yes	NO	Yes	Yes	0.19
Yes	NO <sub>2</sub>	Yes	Yes	> 60
Yes	NO <sub>2</sub>	---	Yes	0.33
No	NO <sub>2</sub>	---	Yes	0.29
Yes	NO <sub>2</sub>	Yes	Yes	0.22
No	NO <sub>2</sub>	Yes	---	0.25

These results point up some of the extremely interesting features of this complicated system.

13.1 Magnetic Solids Separation (R. G. Wymer)

A small vial of Nd<sub>2</sub>O<sub>3</sub> was shown to be attracted with a force of very roughly 1/40 that of gravity when suspended near a 10 kg permanent magnet. The value of 1/40 mxg is not meaningful except in that it indicates the potentialities of using a magnetic field as a means of retaining paramagnetic as well as ferromagnetic materials. The applications of this principle are manifold and obvious, especially to the HRT.

*R E Blanco*

R. E. Blanco

*D E Ferguson*

D. E. Ferguson.

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