

MASTER

MOLTEN-SALT BREEDER REACTOR FUEL PROCESSING\*

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## MOLTEN-SALT BREEDER REACTOR FUEL PROCESSING

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The Oak Ridge National Laboratory has as one of its goals the development of a molten-salt breeder reactor that will be based on the thorium fuel cycle and will operate with a thermal neutron spectrum. A reliable and economic method for processing the fuel from this reactor is necessary, and work on fuel processing constitutes one of the important areas of research and development in the molten salt reactor program. In this paper, the following questions will be considered briefly: What are the design features for the molten-salt breeder reactor? What chemical processing is required for its operation? What are the best processing methods currently available and how can these be combined into an integrated flowsheet? What is the status of development work related to the reference flowsheet? What alternative processing methods are there that might ultimately become more attractive than the present reference system? And finally, what are the possible materials of construction for processing plants and what problems are involved in building plants from these materials?

### Design Features of the Single-Fluid Breeder Reactor

A single-fluid molten-salt breeder reactor<sup>1</sup> is illustrated in simple form in Fig. 1. The fuel salt is a mixture of the fluorides of beryllium, lithium-7, thorium, and uranium and has the composition and properties shown in Table 1. The fluorides of lithium-7 and beryllium have low neutron absorption cross-sections and are used to obtain a composition having a low

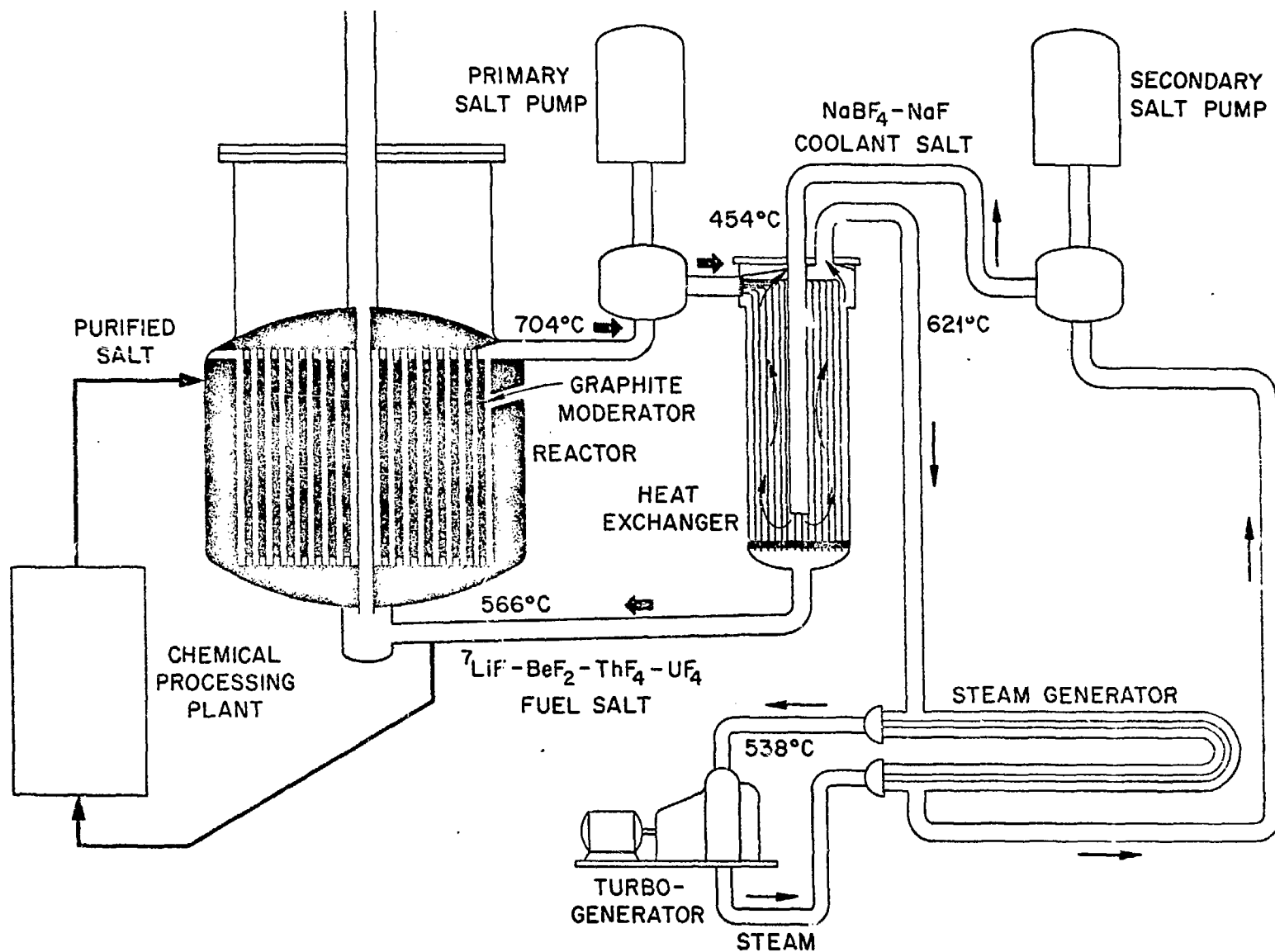


Fig. 1. Single-Fluid, Two-Region Molten Salt Breeder Reactor.

Table 1. Properties of Fuel and Coolant Salts for a Molten-Salt Breeder Reactor

	Fuel Salt		Coolant	
Composition, mole %	${}^7\text{LiF}^a$	71.7	$\text{NaBF}_4$	92
	$\text{BeF}_2$	16	$\text{NaF}$	8
	$\text{ThF}_4$	12		
	$\text{UF}_4$	0.3		
Liquidus Temperature ( $^{\circ}\text{C}$ )	500		385	
Specific Heat ( $\text{cal/g}\cdot^{\circ}\text{C}$ )	0.32		0.36	
Density ( $\text{g/cm}^3$ )	(566 $^{\circ}\text{C}$ )	3.38	(454 $^{\circ}\text{C}$ )	1.93
	(704 $^{\circ}\text{C}$ )	3.28	(621 $^{\circ}\text{C}$ )	1.81
Thermal Conductivity ( $\text{W/cm}\cdot^{\circ}\text{C}$ )	(566 $^{\circ}\text{C}$ )	0.012	(454 $^{\circ}\text{C}$ )	0.0045
	(704 $^{\circ}\text{C}$ )	0.012	(621 $^{\circ}\text{C}$ )	0.0040
Viscosity (cP)	(566 $^{\circ}\text{C}$ )	14.3	(454 $^{\circ}\text{C}$ )	1.91
	(704 $^{\circ}\text{C}$ )	7.2	(621 $^{\circ}\text{C}$ )	1.08
Vapor Pressure (torr)	(566 $^{\circ}\text{C}$ )	< 0.1	(454 $^{\circ}\text{C}$ )	7.6
	(704 $^{\circ}\text{C}$ )	< 0.1	(621 $^{\circ}\text{C}$ )	250

<sup>a</sup>Isotopically separated lithium having a composition of 99.99%  ${}^7\text{Li}$  can be obtained from the USAEC (subject to export regulations) as lithium hydroxide monohydrate. Sufficient quantities for large-scale use in reactors are expected to be available.<sup>26</sup>

liquidus temperature and other physical properties<sup>2,3</sup> that are desirable for reactor use. As noted in the table, the salt has a high specific heat and a very low vapor pressure.

The fuel salt is circulated through the reactor core which contains graphite; the salt does not wet graphite and will not penetrate it if the material has sufficiently small pore sizes. The central region of the reactor has a salt volume fraction of 13%; this region is surrounded by annular regions having salt volume fractions of 37 and 100%. A graphite reflector, which contains essentially no salt, is located adjacent to the reactor vessel. By varying the moderator-to-fuel ratio in the manner described, a well moderated core and an under-moderated fertile blanket where neutron captures in thorium predominate<sup>4</sup> is obtained.

The heat produced by fission is transferred from the fuel salt to a coolant salt in an intermediate heat exchanger. The coolant salt, a eutectic mixture of sodium fluoborate and sodium fluoride, has a low liquidus temperature (385°C), is inexpensive, and has the physical properties indicated in Table 1. The coolant salt is circulated through a steam generator where supercritical steam is produced for generation of electricity. All metal surfaces in the reactor that contact salt are made from Hastelloy N, a nickel base alloy specially developed for use in molten salt reactors.

#### Chemical Processing Requirements

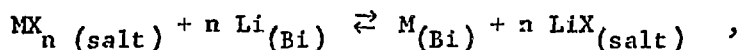
Operation of a molten-salt breeder reactor requires the isolation of <sup>233</sup>Pa from the region of high neutron flux during its decay to <sup>233</sup>U and the removal of fission products (principally the rare earths) in order to hold neutron absorption in these materials to an acceptably low level. The removal of protactinium and the fission products is effected in a

chemical processing plant which is an integral part of the reactor system and will be operated continuously. Processes involving the selective chemical reduction of materials from the fuel salt into liquid bismuth appear to be the most promising processing methods currently available and the development of these processes has been the subject of most of the work on fuel processing. We have noted previously<sup>5</sup> that the isolation of protactinium is straightforward since its extraction behavior is significantly different from that of uranium, thorium, and lithium. However, until recently, the removal of rare earths was difficult since the rare earths and thorium extract in almost the same manner from molten fluoride mixtures. Smith and Ferris<sup>6</sup> noted that the rare earths distribute selectively into molten lithium chloride from bismuth solutions containing thorium; this observation allowed McNeese<sup>7</sup> to devise a significantly improved rare earth removal system.

#### Distribution of Metals Between Molten Salts and Bismuth

Bismuth is a low-melting (271°C) metal that is essentially immiscible with molten halide mixtures consisting of fluorides, chlorides, and bromides. The vapor pressure of bismuth in the temperature range of interest (500 to 700°C) is negligible, and the solubilities of lithium, thorium, uranium, protactinium, and most of the fission products are adequate for processing applications.

Under the conditions of interest, reductive extraction reactions between materials in salt and metal phases can be represented by the following reaction:



in which the metal halide  $MX_n$  in the salt reacts with lithium from the bismuth phase to produce M in the bismuth phase and the respective lithium halide in the salt phase. The valence of M in the salt is  $+n$  and X represents fluorine, chlorine, or bromine. It has been found<sup>8</sup> that at a constant temperature the distribution coefficient D for metal M depends on the lithium concentration in the metal phase (mole fraction),  $X_{Li}$ , as follows:

$$\log D = n \log X_{Li} + \log K_m^*$$

The quantity  $K_m^*$  is dependent only on temperature and the distribution coefficient is defined by the relation:

$$D = \frac{\text{mole fraction of M in metal phase}}{\text{mole fraction of } MX_n \text{ in salt phase}}$$

The ease with which one component can be separated from another is indicated by the ratio of the respective distribution coefficients, that is, the separation factor. As the separation factor approaches unity, separation of the components becomes increasingly difficult. On the other hand, the greater the deviation from unity, the easier the separation.

Distribution data obtained by Ferris, Mailen, and Smith<sup>8</sup> for a number of materials between fuel salt (72-16-12 mole % LiF-BeF<sub>2</sub>-ThF<sub>4</sub>) and bismuth at 640°C are summarized in Fig. 2. The lines for the various elements have slopes that correspond to the indicated oxidation states. Under the expected process conditions, the Pa-Th separation factor is about 1200 which indicates that protactinium as well as uranium and zirconium can be easily extracted from a salt stream containing ThF<sub>4</sub>. However, the rare earth-thorium separation factors are close to unity (1.2 to 3.5), indicating that removal of the rare earths from a salt containing thorium fluoride will be

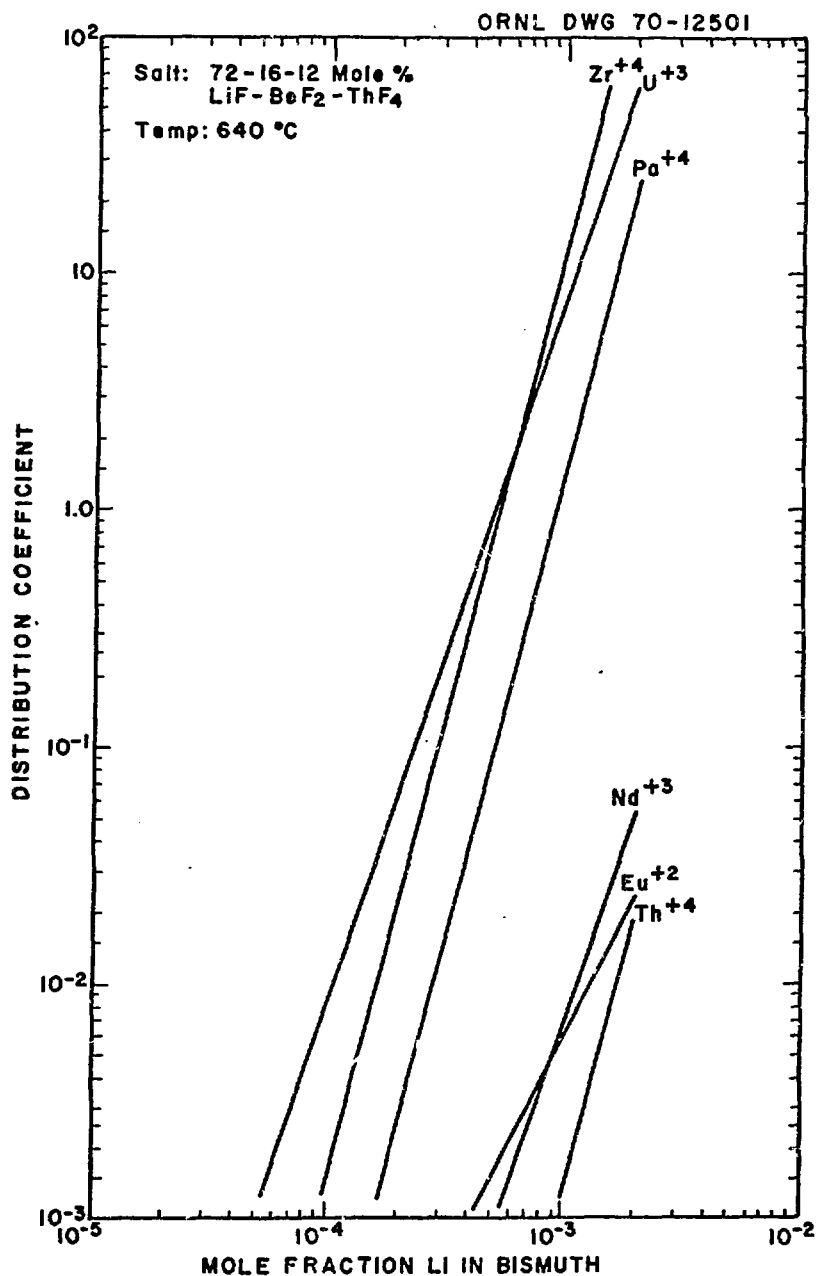


Fig. 2. Distribution Data Between Fuel Salt and Bismuth.

difficult. The previous rare-earth removal system, which was based on these low separation factors, required a large number of stages, a high metal-to-salt flow ratio, and a large electrolytic cell for providing thorium and rare earth reflux at the ends of the extraction cascade.<sup>9</sup>

We have found, however, that with LiCl or LiBr, much more favorable thorium rare-earth separation factors are obtained.<sup>10</sup> Distribution data for LiCl<sup>11,12</sup> at 640°C are shown in Fig. 3. The data fall roughly into three groups. The divalent rare earth and alkaline earth elements distribute most readily to the LiCl with thorium-rare earth separation factors of about  $10^8$ . The trivalent rare earths form the second group, and the thorium-rare earth separation factors are about  $10^4$ . Tetravalent materials, such as thorium and protactinium, distribute only slightly to the LiCl. Studies on the temperature dependence of the distribution data show essentially no effect for the divalent elements, a minor effect for the trivalent elements, and a somewhat greater effect for the tetravalent elements. The distribution coefficient for thorium is decreased sharply by the addition of fluoride to the LiCl although the distribution coefficients for the rare earths are affected by only a minor amount. Thus, contamination of the LiCl with fluoride will not affect the removal of the rare earths but will cause a sharp increase in the thorium discard rate. Data with LiBr<sup>12</sup> are similar to those with LiCl and the distribution behavior with LiCl-LiBr mixtures would likely not differ appreciably from the data with the pure materials.

The potential held by LiCl for selective extraction of the rare earths from MSBR fuel salt is best illustrated by considering the equilibrium concentrations of rare earths, thorium, and lithium in fuel salt, bismuth containing reductant, and LiCl as shown in Table 2. The concentrations of the rare earths and alkaline earths in the fluoride salt correspond to a 25-day

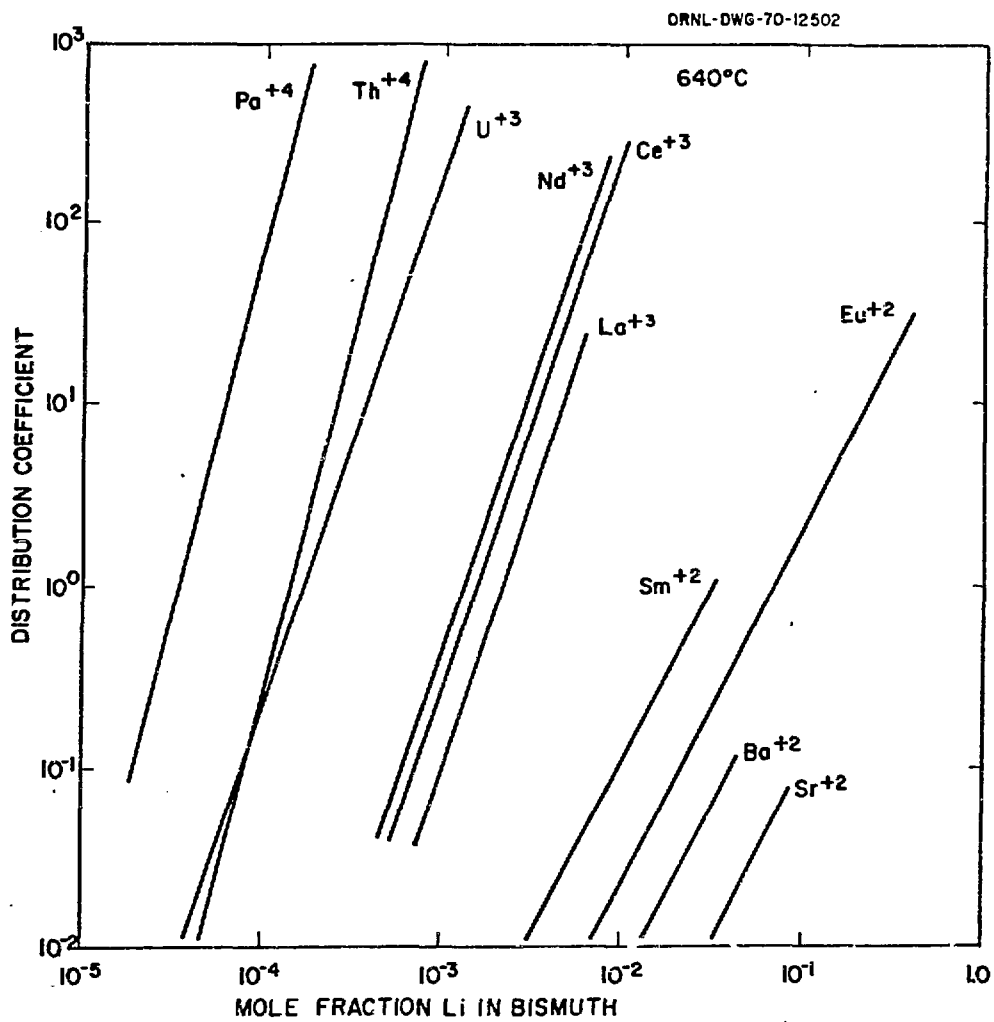


Fig. 3. Distribution Data Between LiCl and Bismuth.

Table 2. Equilibrium Concentrations<sup>a</sup> in Fuel Carrier Salt, Bismuth, and LiCl at 640°C

Element	Fuel Carrier Salt <sup>c</sup>	Bismuth	LiCl
Li	0.72	0.00201	
Be	0.16		
Th	0.12	0.0025	3.31 x 10 <sup>-8</sup>
Sr	7.44 x 10 <sup>-6</sup>	0.664 x 10 <sup>-6</sup>	0.0155
Zr	33.8 x 10 <sup>-6</sup>	0.00802	0.236 x 10 <sup>-6</sup>
Ba	2.83 x 10 <sup>-6</sup>	0.253 x 10 <sup>-6</sup>	0.00123
La	5.46 x 10 <sup>-6</sup>	0.266 x 10 <sup>-6</sup>	0.375 x 10 <sup>-6</sup>
Ce	19.3 x 10 <sup>-6</sup>	1.38 x 10 <sup>-6</sup>	0.636 x 10 <sup>-6</sup>
Nd	12.1 x 10 <sup>-6</sup>	0.680 x 10 <sup>-6</sup>	0.219 x 10 <sup>-6</sup>
Pm	1.26 x 10 <sup>-6</sup>	0.0439 x 10 <sup>-6</sup>	0.0424 x 10 <sup>-6</sup>
Sm	1.34 x 10 <sup>-6</sup>	0.0622 x 10 <sup>-6</sup>	0.000019
Eu	1.55 x 10 <sup>-6</sup>	0.00359 x 10 <sup>-6</sup>	4.39 x 10 <sup>-6</sup>
Y <sup>b</sup>	4.98 x 10 <sup>-6</sup>	0.281 x 10 <sup>-6</sup>	0.090 x 10 <sup>-6</sup>
Pr <sup>b</sup>	4.69 x 10 <sup>-6</sup>	0.264 x 10 <sup>-6</sup>	0.0849 x 10 <sup>-6</sup>

<sup>a</sup>All concentrations are in mole fractions.

<sup>b</sup>Concentrations for Y and Pr were calculated using Nd distribution data.

<sup>c</sup>Concentrations of the fission products in the fuel carrier salt are based on an assumed processing cycle time of 10 days and a removal efficiency of 40%, which results in a 25-day removal time.

removal time for these materials in the reference MSBR. The thorium concentration in the bismuth is 90% of the thorium solubility at 640°C. As can be seen, the rare earth and alkaline earth elements are present in the LiCl at low concentrations and are associated with a negligible amount of thorium. A rare earth removal system, based on this effect, will be practical only if a suitable means is available for removing the rare earth and alkaline earth elements from the LiCl. The distribution coefficients for these elements are strongly affected by the concentration of lithium in the bismuth phase, and the best method for removing these materials from the LiCl appears to be extraction into bismuth containing lithium at a concentration of 0.05 to 0.50 mole fraction. Sufficient data have been obtained with lithium concentrations in the bismuth as high as 0.38 mole fraction to show that no deviation occurs from the relations established initially with much lower lithium concentrations.

#### Protactinium Removal System

The reference protactinium removal system<sup>13</sup> shown in Fig. 4 is based on fluorination for uranium removal and reductive extraction for protactinium isolation. Fuel salt containing 0.33 mole %  $UF_4$  and approximately 0.0035 mole %  $PaF_4$  is withdrawn from the reactor. About 99% of the uranium is removed from the salt by fluorination in order to avoid the use of large quantities of reductant in the subsequent protactinium removal step. The salt stream is fed countercurrent to a bismuth stream containing lithium and thorium where the remaining uranium and the protactinium transfer to the metal stream. These materials are transferred from the bismuth to a captive secondary salt by hydrofluorinating the bismuth stream leaving the extraction column in the presence of the secondary salt. The secondary salt which flows

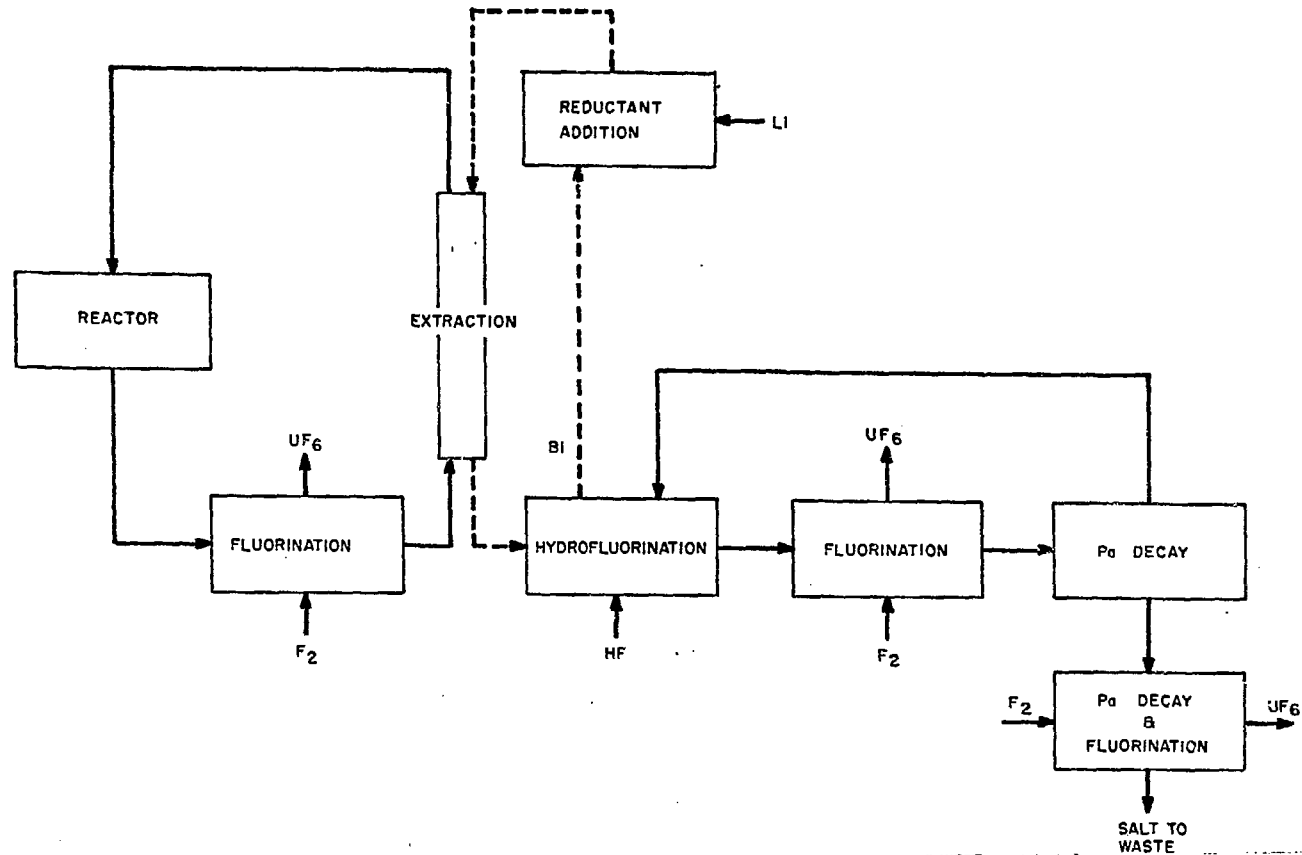


Fig. 4. Flowsheet for Isolation of Protactinium from a Single-Fluid MSBR by Fluorination-Reductive Extraction.

through the hydrofluorinator also circulates through a fluorinator where about 90% of the uranium is removed and through a tank that contains most of the protactinium. Lithium is added to the bismuth leaving the hydrofluorinator and the resulting stream is returned to the top of the extraction column. The salt leaving the extraction column is essentially free of uranium and protactinium but contains the rare earths at essentially the reactor concentration. This stream is fed to the rare earth removal system.

#### Rare Earth Removal Process

A simplified flowsheet for the rare earth removal system<sup>14</sup> is shown in Fig. 5. Fuel salt, which is free of uranium and protactinium but contains the rare earths, is countercurrently contacted with bismuth containing reductant in order to extract a significant fraction of the rare earths into the bismuth. The bismuth stream which contains the rare earths and thorium is then countercurrently contacted with lithium chloride. Because of highly favorable distribution coefficients, a significant fraction of the rare earths transfer to the LiCl along with a negligible amount of thorium. The final steps of the process consist of extracting the rare earths from the LiCl by contact with bismuth having lithium concentrations of 5 and 50 at. %.

This process has a number of very desirable characteristics. Of primary importance is the fact that there is no net consumption of reductant in the two upper contactors. The process is not sensitive to minor variations in operating conditions. Essentially no materials other than the rare earth and alkaline earth elements are removed from or added to the fuel salt; the major change consists of replacing the extracted rare earths with an equivalent amount of lithium as LiF. The amount of LiF added to the fuel salt in this manner during 30 years of operation would be less than 10% of the LiF inventory in the reactor.

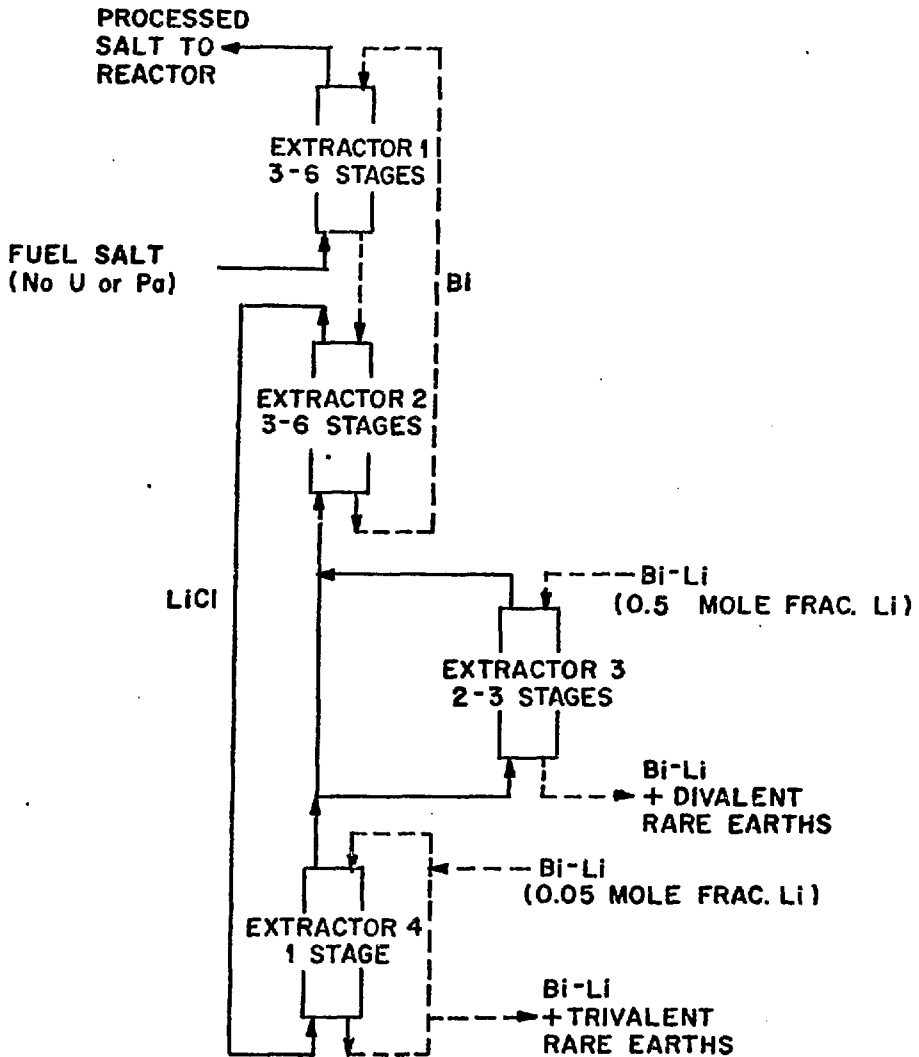


Fig. 5. Metal Transfer Process for Removal of Rare Earths from Single-Fluid MSBR Fuel Salt.

## Conceptual Processing Flowsheet

The reference processing flowsheet<sup>13</sup> is shown in Fig. 6. Fuel salt is withdrawn from the reactor on a 10-day cycle; for a 1000-MW(e) reactor, this represents a flow rate of 0.88 gpm. The fluorinator, where about 99% of the uranium is removed, has an active diameter of 5 inches and a height of about 15 feet. The protactinium extraction column is 3 in. in diameter and is packed with 3/8-in. Raschig rings. The column is equivalent to 5 equilibrium stages and has a height of about 15 ft. The bismuth flow rate through the column is 0.13 gpm and the inlet thorium concentration in the stream is 90% of the thorium solubility at the operating temperature of 640°C. The protactinium decay tank has a volume of 160 ft<sup>3</sup>. The uranium inventory in the tank is less than 0.2% of that in the reactor. Fluorides of lithium, thorium, zirconium, and nickel accumulate in the tank at a total rate of about 0.1 ft<sup>3</sup>/day. These materials are removed by periodic withdrawal of salt to a final protactinium decay and fluorination operation.

The bismuth flow rate through the two upper contactors in the rare-earth removal system is 12.5 gpm and the LiCl flow rate is 33 gpm. The extraction columns are 7 to 13 in. in diameter and are packed with 1/2-in. Raschig rings. The two upper columns are each equivalent to three equilibrium stages. The trivalent and divalent rare earths are removed in separate contactors in order to minimize the amount of lithium required. Only 2% of the LiCl or 0.66 gpm is fed to the two-stage divalent rare-earth removal contactor where it is contacted with a 2.2 liter/day bismuth stream containing 50 at. % lithium. The trivalent stripper where the LiCl is contacted with bismuth containing 5 at. % lithium is equivalent to one equilibrium stage.

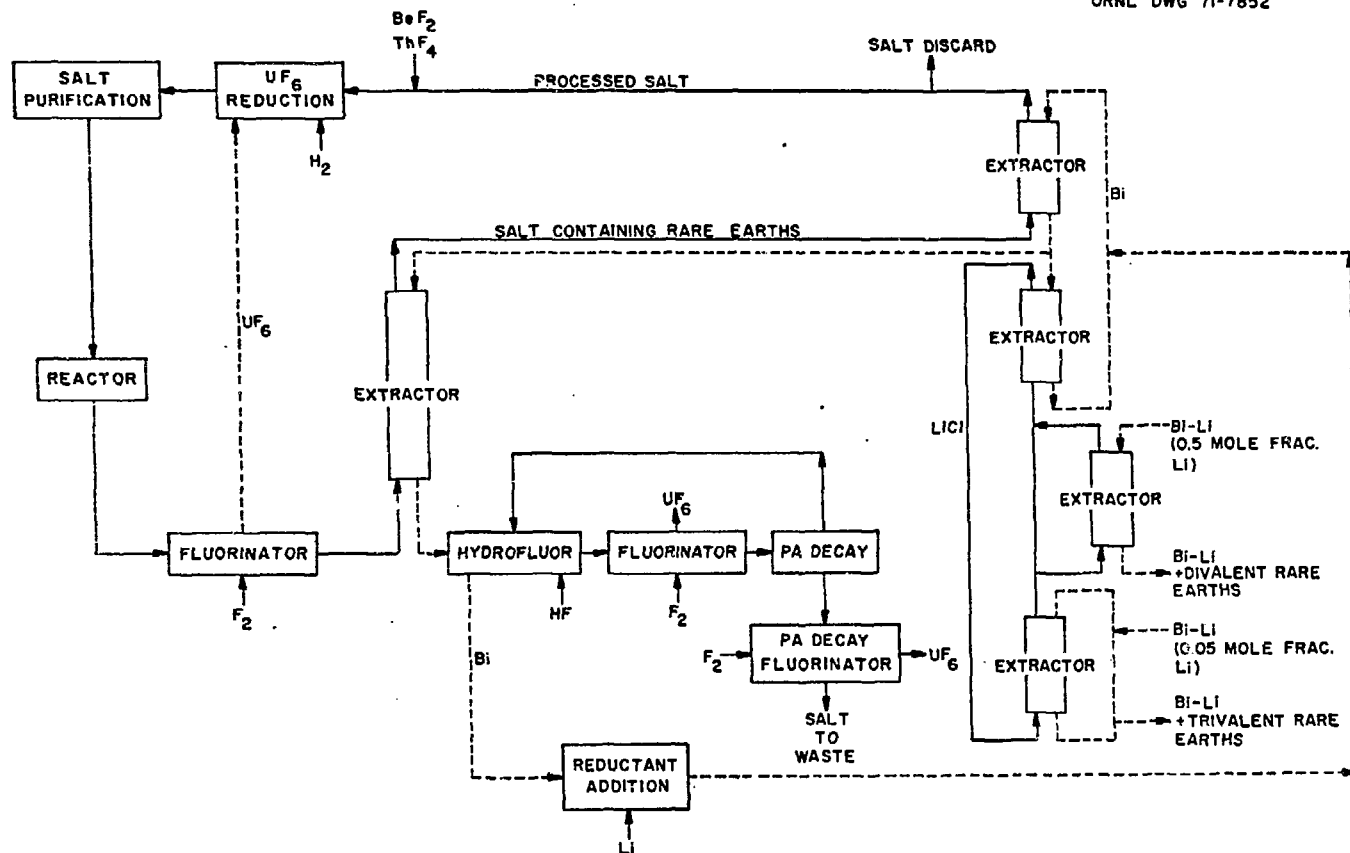


Fig. 6. Conceptual Flowsheet for Processing a Single-Fluid MSBR by Fluorination-Reductive Extraction and the Metal Transfer Process.

The bismuth stream containing the reductant necessary for the isolation of protactinium is actually fed to the recirculating bismuth stream in the rare-earth removal system. An equivalent amount of bismuth is withdrawn from the stream and is fed to the protactinium isolation column. This allows for more nearly complete extraction of the protactinium and provides a means for removing materials which might otherwise accumulate in the recirculating stream.

The remaining steps in the flowsheet consist of combining the processed salt with uranium and purifying the resulting fuel salt. The uranium addition is accomplished by absorbing the  $UF_6$  into fuel salt containing  $UF_4$  which results in the formation of soluble, nonvolatile  $UF_5$ . The  $UF_5$  is then reduced to  $UF_4$  by contact with hydrogen.

The protactinium removal time obtained with the flowsheet is 10 days and the rare earth removal times range from 17 to 51 days with the rare earths of most importance being removed on 27- to 30-day cycles. Bell and McNeese have made calculations<sup>13,14</sup> which indicate that the flowsheet is relatively insensitive to minor variations in operating conditions such as changes in temperature, flow rates, reductant concentrations, etc. It was noted earlier that the thorium-rare earth separation factor decreases sharply as the concentration of fluoride in the LiCl is increased; contamination of the LiCl would result from entrainment of fuel salt by the bismuth stream leaving the upper contactor. The effect is largely an increase in the rate at which thorium is removed with the rare earths. The thorium removal rate increases from about 0.4 mole/day with no fluoride in the LiCl to about 280 mole/day when the LiCl contains the equivalent of 5 mole % LiF. It appears that the fluoride concentration in the LiCl can economically be as high as 2 mole % which corresponds to a thorium discard rate of 7.7 mole/day. Discard of

thorium at this rate would add only about 0.0013 mill/kWhr to the power cost. The effect of fluoride in the LiCl on the removal of rare earths is negligible. In fact, the rare earth removal efficiency increases slightly as the fluoride concentration in the LiCl increases.

#### Combination of Discard Streams from the Processing System

Bell and McNeese<sup>13</sup> have observed that it is possible to combine all waste streams from the protactinium and rare-earth removal systems shown in Fig. 6 for uranium recovery prior to disposal as shown in Fig. 7. In this operation, waste salt from the protactinium decay tank would be combined with the fuel carrier salt discard stream. The lithium-bismuth stream from the trivalent rare-earth stripper would be hydrofluorinated in the presence of the resulting salt, and the combined stream would be held for protactinium decay. The protactinium concentration in the combined stream would be only 500 ppm initially, and the specific heat generation rate would be acceptably low. The salt in the waste holdup tank would be fluorinated before discard to recover uranium. The composition of the discarded salt would be 74.7-13.5-9.5-0.8 mole % LiF-ThF<sub>4</sub>-BeF<sub>2</sub>-ZrF<sub>4</sub>, 1.2 mole % trivalent rare earth fluorides, and 0.3 mole % divalent rare earth fluorides. The salt temperature would have to be maintained at about 600°C so that the trivalent rare earth fluorides would not precipitate. This processing scheme would require that salt be discarded at the rate of 60 ft<sup>3</sup> every 220 days.

#### Status of Development Work

Several important objectives for MSBR processing have been achieved recently. The most important of these is an engineering experiment made by Youngblood and McNeese which demonstrates all aspects of the metal transfer

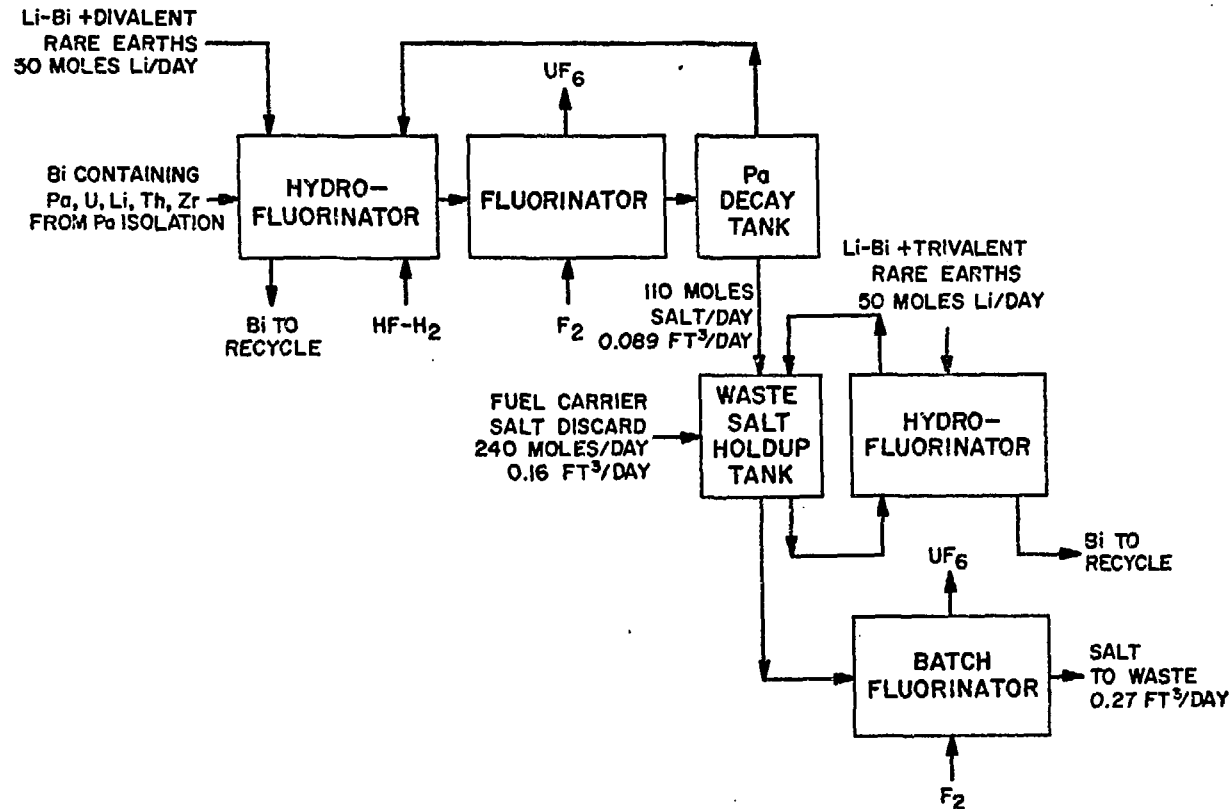


Fig. 7. Method for Combining Waste Streams from Protactinium Isolation and Rare Earth Removal Processes. Flow rates are shown for an assumed uranium removal efficiency in the primary fluorinator of 99%.

process.<sup>15</sup> The equipment consisted of a 6-in.-diam compartmented vessel in which were present about 1 liter each of MSBR fuel carrier salt, bismuth saturated with thorium, and LiCl. The fluoride salt initially contained  $^{147}\text{NdF}_3$  at the tracer level and  $\text{LaF}_3$  at a concentration of 0.04 mole fraction. During the experiment, the rare earths were selectively extracted into the LiCl along with a negligible amount of thorium. Provision was made for circulating the LiCl through a chamber containing bismuth having a lithium concentration of 38 at. % where the rare earths and thorium were removed. The distribution ratios for the rare earths remained constant during the experiment at about the expected values. About 50% of the neodymium and about 70% of the lanthanum were collected in the Li-Bi solution. The final thorium concentration in the Li-Bi solution was below 5 ppm, making the ratio of rare earths to thorium in the Li-Bi greater than  $10^5$  times the initial concentration in the fuel salt and thus demonstrating the selective removal of rare earths from a fluoride salt containing thorium. A larger metal transfer experiment<sup>16,17,18</sup> has been put into operation that uses salt and bismuth flow rates that are about 1% of the values required for processing a 1000-MW(e) MSBR, and the preliminary design has been carried out for an experiment that will use a 3-stage salt-metal contactor and flow rates that are 5 to 10% of those required for a 1000-MW(e) MSBR.<sup>19</sup>

Hannaford, Kée, and Sood have successfully operated a salt-bismuth reductive extraction facility in which uranium and zirconium were extracted from salt by countercurrent contact with bismuth containing reductant.<sup>20,21</sup> Greater than 95% of the uranium was extracted from the salt by a 0.82-in.-diam, 24-in.-long packed column. The inlet uranium concentration in the salt was about 25% of the uranium concentration in the reference MSBR. These experiments represent the first demonstration of reductive extraction of uranium

in a flowing system. Information on the rate of mass transfer of uranium and zirconium has been obtained in the system using an isotopic dilution method, and HTU values of about 4.5 ft have been obtained. Watson and McNeese<sup>22,23</sup> have developed correlations for flooding and dispersed-phase holdup in packed columns during countercurrent flow of liquids having high densities and a large difference in density such as salt and bismuth. These correlations, which have been verified by studies with molten salt and bismuth,<sup>24</sup> were developed by study of countercurrent flow of mercury and water or high density organics and water in 1- and 2-in.-diam columns packed with solid cylinders and Raschig rings varying in size from 1/8 to 1/2 in. Watson and McNeese have also obtained data on axial dispersion in the continuous phase during the countercurrent flow of high density liquids in packed columns<sup>25,26</sup> and Watson and Cochran have developed a simple relation for predicting the effects of axial dispersion on column performance.<sup>27</sup>

The successful operation of salt-metal extraction columns is dependent upon the availability of a bismuth-salt interface detector. Weeren, Nicholson, and Dodd<sup>29</sup> have recently demonstrated the successful operation of an eddy-current type interface detector. The detector consists of a ceramic form on which bifilar primary and secondary coils are wound. Contact of the coils with molten salt or bismuth is prevented by enclosing the coils in a molybdenum tube. A high-frequency alternating current is passed through the primary coil and it induces a current in the secondary coil. The induced current is dependent on the conductivities of materials adjacent to the primary and secondary coils; since the conductivities of bismuth and salt are quite different, the induced current reflects the presence or absence of bismuth. Two approaches for obtaining an output from the detector are being studied. The first is based on measuring changes in the magnitude of the

induced current; the second is based on measuring the phase shift that occurs between the voltage that is imposed on the primary coil and that which is induced in the secondary coil. Measurements in the temperature range of 500 to 700°C indicate that the detector output varies linearly with changes in bismuth level. The average sensitivity of the detector to changes in bismuth level is about 0.87 degree phase shift per inch of level change, and the average temperature coefficient is only about 0.006 in./°C. Measurements of the change in magnitude of the induced current indicate a similar response although the temperature coefficient is somewhat larger. The detector appears to be a practical and sensitive indicator of either salt-bismuth interface location or bismuth level.

We have initiated design and development work for a reductive extraction process facility<sup>28</sup> that will allow operation of the important steps for the reductive extraction process for protactinium isolation. The facility will allow countercurrent contact of salt and bismuth streams in a 2-in.-diam, 6-ft-long packed column at flow rates as high as about 25% of those required for processing a 1000-MW(e) MSBR. The facility will operate continuously and will allow measurement of mass transfer and hydrodynamic data under steady state conditions.

Progress has also been made in the development of continuous fluorinators. McNeese has previously<sup>30</sup> carried out experimental studies of fluorination of molten salt in a 1-in.-diam, 72-in.-long nickel fluorinator that allowed countercurrent contact of molten salt with fluorine. In these tests, molten salt (41-25-35 mole % NaF-LiF-ZrF<sub>4</sub>) containing UF<sub>4</sub> was countercurrently contacted with a quantity of fluorine in excess of that required for the conversion of UF<sub>4</sub> to UF<sub>6</sub>. Experiments were carried

out with temperatures ranging from 525 to 600°C, salt feed rates ranging from 5 to 30 cm<sup>3</sup>/min, fluorine feed rates ranging from 215 to 410 cm<sup>3</sup>/min, and UF<sub>4</sub> concentrations in the feed salt ranging from 0.12 to 0.35 mole %. The fraction of the uranium removed from the salt ranged from 97.5% to 99.9%.

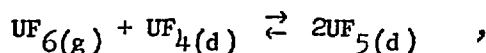
Axial dispersion in the salt phase will be important in the design of continuous fluorinators, and Watson and McNeese have measured gas holdup and axial dispersion in columns having diameters ranging from 1 to 6 in. using air and aqueous solutions. Data were obtained for viscosities of the liquid ranging from 0.9 to 12 cP, surface tensions of the liquid ranging from 27 to 72 dyne/cm, and superficial gas velocities ranging from 0.04 to 40 cm/sec. Correlations for gas holdup and axial dispersion were developed<sup>31</sup> which are believed to be applicable to the condition involving counter-current contact of molten salt and fluorine in a continuous fluorinator. The data on axial dispersion and uranium removal in the 1-in.-diam continuous fluorinator were used for estimating the performance of larger diameter continuous fluorinators.<sup>32</sup>

The combination of molten salt and fluorine results in a high corrosive environment, and it will be necessary to protect a continuous fluorinator from corrosion by maintaining a layer of frozen salt on surfaces that would otherwise contact both molten salt and fluorine in order to allow passivation of the nickel metal to occur. The feasibility of maintaining frozen salt layers in gas-salt contactors was demonstrated previously by Hannaford and McNeese<sup>33</sup> in tests in a 5-in.-diam, 8-ft-high fluorinator simulation in which molten salt (66-34 mole % LiF-ZrF<sub>4</sub>) and argon were countercurrently contacted. An internal heat source in the molten region was provided by Calrod heaters contained in a 3/4-in.-diam pipe along the center line of the

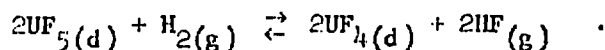
vessel. A frozen salt layer was maintained in the system with equivalent volumetric heat generation rates of 10 to 55 kW/ft<sup>3</sup>; for comparison, the heat generation rates in salt immediately after removal from the reactor and after passing through vessels having holdup times of 5 and 30 min are 56.6, 27.6, and 12.5 kW/ft<sup>3</sup>, respectively.

Operation of a continuous fluorinator with nonradioactive salt requires a means for generating heat in the molten salt that is not subject to corrosion. Hightower has studied radio frequency induction heating in fluorinator simulations using nitrate acid<sup>34</sup> and has studied autoresistance heating using 60 Hz power with molten salt (65-35 mole % LiF-BeF<sub>2</sub>) in a 6-in.-diam fluorinator simulation.<sup>35</sup> Autoresistance heating is the preferred method since it can be used over a wider range of operating conditions and since the electrical power supply is much simpler than that required for induction heating. A facility is planned in which a continuous fluorinator can be operated that has a molten zone diameter of 5 in. and a molten salt depth of 5 ft.

Studies of the absorption of UF<sub>6</sub> by MSBR fuel carrier salt containing UF<sub>4</sub> are being carried out by Bennett.<sup>37</sup> These studies have shown that absorption of UF<sub>6</sub> in fuel carrier salt containing UF<sub>4</sub> results in the formation of soluble nonvolatile UF<sub>5</sub> according to the following reaction:



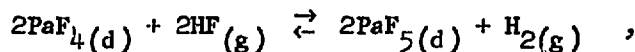
in which (g) denotes gas and (d) denotes species dissolved in the salt. The recent studies have also shown that gaseous hydrogen reacts with dissolved UF<sub>5</sub> according to the reaction:



Since both  $\text{UF}_6$  and  $\text{UF}_5$  are strong oxidants, the initial studies were conducted primarily to find a material that was inert to these species. These studies showed that, at  $600^\circ\text{C}$ , nickel, copper, and graphite were not sufficiently inert but that gold was stable both to gaseous  $\text{UF}_6$  and to salt containing up to 6 wt %  $\text{UF}_5$ . The recent studies have been conducted in gold apparatus. Results from several experiments show that  $\text{UF}_5$  dissolved in molten salt slowly disproportionates to  $\text{UF}_6$  and  $\text{UF}_4$ , and that the rate of disproportionation is second order with respect to the concentration of  $\text{UF}_5$ . The studies also indicate that the solubility of  $\text{UF}_6$  in the salt is low. Engineering experiments are being designed for further study of the absorption of  $\text{UF}_6$  into molten salt containing  $\text{UF}_4$  and the subsequent reduction of  $\text{UF}_5$  by contact of the salt with hydrogen.

#### Alternate Processing Methods

We presently know of no other rare earth removal method as attractive as the metal transfer process; however, there appear to be alternate methods for the removal of protactinium and uranium from fuel salt based on the selective precipitation of the oxides of these materials. Protactinium is present in the MSBR as  $\text{PaF}_4$ . It has been shown<sup>39</sup> that  $\text{Pa}^{4+}$  dissolved in a molten  $\text{LiF}-\text{BeF}_2-\text{ThF}_4$  solution can be oxidized to the  $5^+$  state by hydrofluorination according to the reaction:



and that the addition of oxide to molten salt containing  $\text{Pa}^{5+}$  results in the precipitation of a pure phase consisting of  $\text{Pa}_2\text{O}_5$ . The solubility product for  $\text{Pa}_2\text{O}_5$  appears to be much lower than that for the oxides of other

materials in the fuel salt; this makes possible the selective removal of protactinium from MSBR fuel salt without the prior removal of uranium. If the oxide concentration in the salt is increased to too high a level, a solid solution of uranium and thorium oxides will be precipitated; studies by Bamberger and Baes<sup>40</sup> indicate that a  $\text{UO}_2\text{-ThO}_2$  solid solution containing about 95%  $\text{UO}_2$  would be precipitated from MSBR fuel salt. It is believed that contact of the fuel salt with an  $\text{HF-H}_2\text{O-Ar}$  gas mixture of the appropriate composition would convert practically all of the protactinium to  $\text{Pa}^{5+}$  and precipitate a large fraction of the  $\text{Pa}_2\text{O}_5$  without precipitating uranium oxide. The equilibria involved in the oxidation and precipitation steps are being studied by Tallent, Ferris, and Smith<sup>41,42</sup> and Ross, Bamberger, and Baes.<sup>43,44</sup> The extent to which the protactinium concentration in MSBR fuel salt can be decreased without the precipitation of uranium oxide is not known at present<sup>42,44</sup> with sufficient accuracy to allow final evaluation of oxide precipitation processes.

Engineering studies of uranium removal by oxide precipitation have been carried out by Bell, Sood, and McNeese.<sup>45,46</sup> These experiments involved the contact of 2 liters of MSBR fuel salt with  $\text{H}_2\text{O-Ar}$  gas mixtures in a 4-in.-diam nickel precipitator. Experiments were conducted at temperatures ranging from 540 to 630°C, and the composition of the  $\text{H}_2\text{O-Ar}$  mixture was varied from 10 to 35% water. Only a slight increase in the precipitation rate with an increase in temperature was observed, and the rate of precipitation appeared to vary directly with the rate at which water was supplied to the system. The values for the water utilization were uniformly low (about 10 to 15%) and did not vary with the composition of the gas stream. Higher utilization values should be obtained by improving the contact of the gas with the salt. From 50 to 90% of the uranium was precipitated as oxide in most of the experiments. Samples of the oxide contained about

90%  $\text{UO}_2$  even though at the lower uranium concentrations in the salt, the solid in equilibrium with the salt would contain 50%  $\text{UO}_2$  or less. It is believed that under nonequilibrium precipitation conditions, such as would be present in a processing plant,  $\text{UO}_2$ - $\text{ThO}_2$  solid solutions are formed which are in equilibrium with the salt at the moment of precipitation, but which once formed do not rapidly reequilibrate. Thus, solid solutions that are formed early in the precipitation process and that contain 90 to 95%  $\text{UO}_2$  are still present during the final stages of precipitation when solid solutions are being formed that contain much less  $\text{UO}_2$ . It is believed that this effect will allow precipitation of 99% of the uranium as a solid containing 85%  $\text{UO}_2$  in a single-stage batch precipitator. Earlier calculations,<sup>47</sup> based on the assumption that the oxide and salt would remain in equilibrium throughout the precipitation process, had indicated that a 3-stage batch countercurrent precipitation system would be required to achieve removal of this fraction of the uranium without the removal of more than 1% of the thorium in the salt as  $\text{ThO}_2$ . The oxide precipitate was observed to settle rapidly and greater than 90% of the salt could be separated from the oxide by simple decantation. It is believed that the removal of uranium from MSBR fuel salt from which most of the protactinium has been removed can be accomplished easily by oxide precipitation by contacting the salt with gaseous  $\text{H}_2\text{O}$ -Ar mixtures.

Bell and McNeese have made preliminary evaluations for several conceptual flowsheets;<sup>48</sup> the most promising of these is shown in Fig. 8. Fuel salt would be withdrawn from the reactor on a 3-day cycle and about 60% of the protactinium would be removed as  $\text{Pa}_2\text{O}_5$  in order to obtain a protactinium removal time of 5 days. The  $\text{Pa}_2\text{O}_5$  precipitate would be hydrofluorinated in the presence of a captive fluoride salt phase which would be circulated

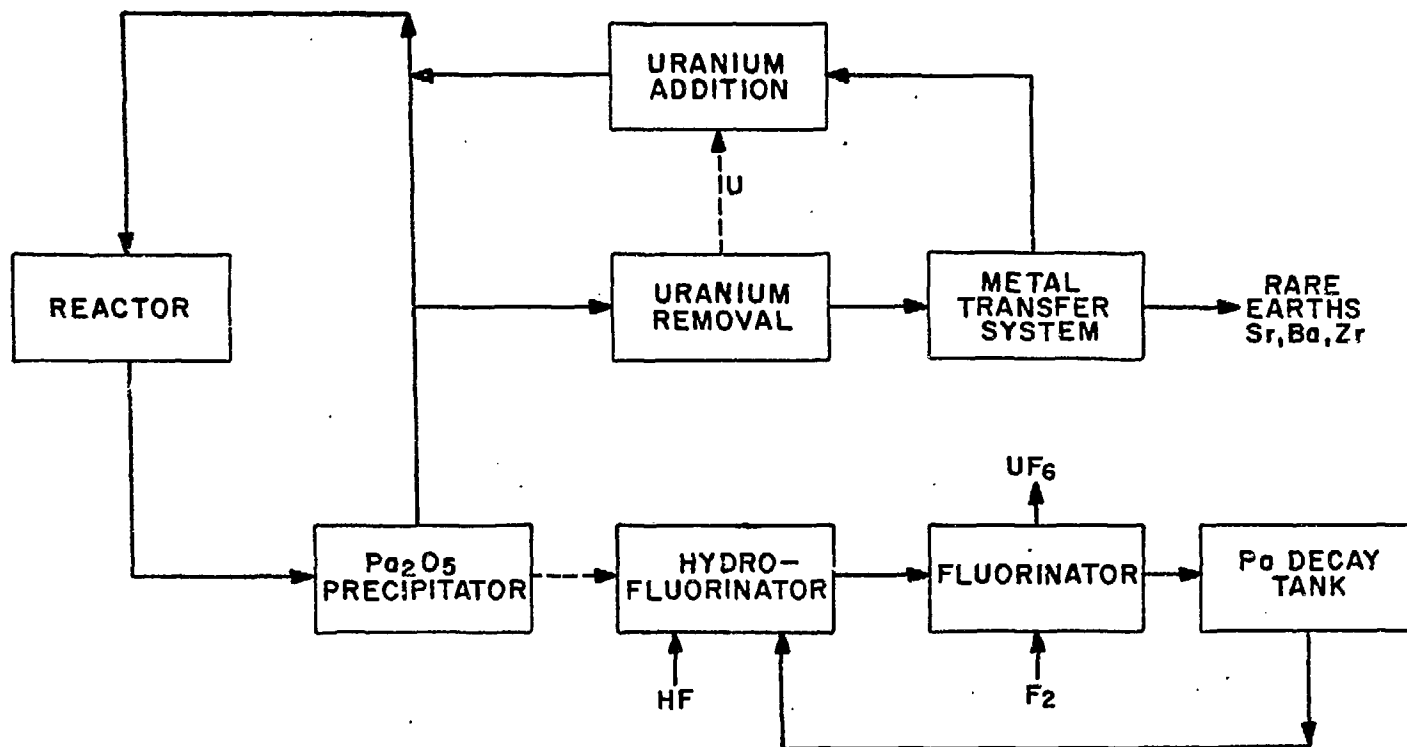


Fig. 8. Oxide Precipitation-Metal Transfer Flowsheet for Processing a Single-Fluid MSBR.

through the protactinium decay tank and through a fluorinator in order to maintain an acceptably low uranium inventory in the decay tank. Part of the salt in the decay tank would be returned to the reactor periodically to compensate for salt that is transferred to the hydrofluorinator with the  $\text{Pa}_2\text{O}_5$ . Ten percent of the salt leaving the  $\text{Pa}_2\text{O}_5$  precipitator would be processed for rare earth removal by the metal transfer process; this would result in a 30-day processing cycle and a rare earth removal time of about 50 days. Most of the uranium must be removed from the salt prior to the removal of the rare earths. This could be accomplished either by fluorination or by oxide precipitation. The separated uranium would then be recombined with the processed salt leaving the metal transfer system and would be returned to the reactor.

#### Materials of Construction for Processing Plants

The materials requirements for a processing plant based on the reference flowsheet are relatively stringent since few materials are compatible with molten bismuth. Molybdenum, tungsten, and graphite appear to be suitable materials for containing both molten salts and bismuth. Of the two refractory metals, molybdenum appears to be the better choice because of the availability of material in the form of tubing, plate, and billets. Although molybdenum is quite resistant to corrosion, the difficulty of fabricating equipment from this material has restricted its application to processing systems. Several advances in the fabrication technology of molybdenum have been made at the Oak Ridge National Laboratory as we began to build an all-molybdenum system in which bismuth and molten salt will be countercurrently contacted in a 1-in.-ID, 5-ft-high packed column that has 3.5-in.-ID upper and lower disengaging sections. Salt and bismuth will be circulated in the

system by gas-lift pumps that will elevate the streams to 3.5-in.-ID head pots for sampling, gas separation, and flow measurement. A salt-bismuth interface detector will be provided in the lower disengagement section for determining the pressure drop through the column and the holdup of bismuth in the column.

McDonald and Schaffhauser<sup>49,50,51,52</sup> have developed techniques for the production of closed-end molybdenum vessels by back extrusion which involves the flow of metal into a die and the backward flow of metal over an advancing plunger. This process has the advantages that the diameter of the part produced is as large as or larger than that of the starting metal blank, the configuration of the part can be changed by relatively simple changes in the die and mandrel design, and deformation can be accomplished below the recrystallization temperature so that a wrought structure having good mechanical properties is produced. Parts having good quality were consistently produced with this technique by the use of  $ZrO_2$ -coated plungers and dies and extrusion temperatures of 1600 to 1700°C. Eleven closed-end vessels were produced that had a 3 7/8-in.-OD, a 3.5-in.-ID, and lengths from 8 to greater than 9 in. The 5.5-ft-long molybdenum pipe for the extraction column, having an outside diameter of 1.16 in. and an inside diameter of 1 in., was produced by floating mandrel extrusion at 1600°C. Three extrusions for producing this material were performed; the second extrusion produced a pipe 11.5 ft long that was concentric to within 0.007 in. with excellent external and internal surfaces.

DiStefano,<sup>53</sup> in evaluating three sizes of commercially available molybdenum tubing (1 1/4-, 3/8-, and 1/2-in.-OD), found that the 1/2-in.-OD tubing was ductile at room temperature while the 3/8-in.-OD tubing was ductile

only at temperatures as high as 150 to 250°C, and the 1/4-in.-OD tubing was ductile only at temperatures above 300°C. It was found, however, that the 3/8-in.-OD and 1/4-in.-OD tubing could be made ductile at room temperature by the removal of 0.001 to 0.003 in. of material from the ID of the tubing by etching. It is believed that tubing that is ductile at room temperature can be produced routinely by careful control of surface contamination during tubing fabrication.

Moorehead and Housley have shown that molybdenum can be welded successfully by either the tungsten arc or the electron-beam welding technique.<sup>54,55,56,57</sup> A commercially available device for automatically tungsten-arc welding tubing under field conditions has been modified and successfully applied to the welding of molybdenum tubing. An unavoidable increase in the grain size of the molybdenum in the weld area renders the material nonductile at room temperature. However, by brazing a molybdenum band over the weld-affected zone, stresses in the weld zone can be limited to satisfactorily low values.

Cole has carried out studies for the development of braze materials for joining molybdenum that are resistant to corrosion by bismuth and molten salts.<sup>58,59,60,61</sup> An iron base alloy (Fe-15%Mo-5%Ge-4%C-1%B) has been found to have good wetting and flow properties, a moderately low brazing temperature (less than 1200°C), and adequate resistance to bismuth at 650°C.

Preliminary tests have been carried out for studying the compatibility of graphite with bismuth containing lithium. The extent of penetration of the bismuth into the graphite appears to be greater when the bismuth contains lithium than with pure bismuth. Studies are in progress for determining the nature and extent of chemical reaction between graphite and bismuth containing lithium. Experience with graphite components in systems containing

bismuth and lithium at low concentrations lead us to expect that graphite is a suitable material of construction. However, the final decision is dependent on studies now in progress.

Nickel would be a suitable material of construction for the oxide-precipitation portions of a plant based on an oxide precipitation-metal transfer flowsheet; however, the use of molybdenum and graphite would still be required in the metal-transfer portions of the plant.

### SUMMARY

The successful operation of a single-fluid molten-salt breeder reactor requires the removal of  $^{233}\text{Pa}$  and the fission products in order to hold the absorption of neutrons in these materials to an acceptable level. The reference flowsheet for effecting removal of these materials is based on fluorination for uranium removal, reductive extraction for protactinium removal, and the metal transfer process for rare-earth and alkaline-earth removal. Progress has been made in the development of continuous fluorinators which will be protected from corrosion by a frozen layer of salt. Studies have been carried out that allow improved design and evaluation of packed column salt-metal contactors. All steps of the metal transfer process for removing rare earths from MSBR fuel carrier salt have been demonstrated and experiments are in progress that use salt and bismuth flow rates that are 1% of the expected flow rates for processing a 1000-MW(e) MSBR.

An alternate processing flowsheet is being developed that is based on oxide precipitation for removal of protactinium and uranium and the metal transfer process for removal of rare earths and alkaline earths. The equilibria involved in the oxide precipitation of protactinium are not sufficiently well defined to allow final evaluation of the flowsheet. Processing

plants based on either flowsheet will require the use of graphite and molybdenum in portions of the plant that use molten bismuth, and significant progress in the fabrication technology for molybdenum has been made. These developments have significantly strengthened the technical basis for processing molten-salt breeder reactors.

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