MOLTEN-SALT REACTOR CONCEPTS WITH REDUCED POTENTIAL FOR PROLIFERATION OF SPECIAL NUCLEAR MATERIALS

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ABSTRACT[*]

This study examines design alternatives for molten-salt breeder reactors (MSBRs) with breeding ratios near 1.0 to evaluate their nonproliferation characteristics. Only those systems are examined for which sufficient information exists to describe adequately the power plant system characteristics in terms of both practicality as a source of electricity and susceptibility to diversion of special nuclear material (SNM). In this precursory study evaluating performance and non-diversion features, various candidate systems have been examined with the following results: (1) molten-salt reactors could eliminate the transport requirements of SNM to or from the reactor for long periods of time and make the extraction of SNM from the reactor inventory difficult; (2) two candidate MSR configurations, the CeF_3 processing scheme and the scheme with no chemical processing, can be highly resistant to diversion but cannot be classed as diversion-proof; (3) two additional systems, less resistant than the two above, are the reductive-extraction process without Pa isolation and the salt distillation process; and, (4) the system based on the reference MSBR, requiring salt fluorination is significantly less resistant to diversion than a system without fluorination. Diversion-resistant MSBRs, if developed, might afford resistance to diversion of SNM comparable to solid-fueled reactors without fuel reprocessing and would require less uranium for deployment and operation.

INTRODUCTION

In considering the feasibility of producing a nuclear reactor system for export that does not admit a diversion of fissile material, molten-salt reactors (MSR) have been suggested. It is the purpose of this report to examine the available design alternatives to determine if one or more arrangements will satisfy the criteria established for non-proliferating reactor plants.

This study was confined to the examination of those systems for which a sufficient body of developed information exists to describe adequately the character of power plants, both as practical sources of electrical energy and with regard to susceptibility to diversion of special nuclear material (SNM).

A study was conducted by a small team of individuals from Oak Ridge National Laboratory (ORNL) and EBASCO Services Inc. familiar with the molten-salt breeder reactor (MSBR) developmental work that has been done; this was then reviewed by a different group from the Institute for Energy Analysis and ORNL. This approach was felt to be desirable as a precursor to any more comprehensive and definitive study, to determine quickly whether it was likely that an attractive system could be developed.

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[*] Note Added in Proof: Since this report was written, a molten-salt system was identified that may be remarkably resistant to materials diversion as well as having a conversion ratio near 1.0. This system uses a denatured fuel mixture of ^{233}U-^{238}U; processing consists only of removing fission products. These results are only tentative and require more analysis before the described properties can be confirmed.

The manner of presentation is aimed at facilitating the comparison of the various candidate conceptual arrangements, with an evaluation of performance and non-diversion features. An assessment of the developmental status and commercial attractiveness then follows in which the candidate systems are compared.

CANDIDATE SYSTEMS

General

In this section are described five members of the general family of molten-salt reactors that appear to offer some advantages from the standpoint of non-proliferation of SNM. All of the systems considered are variations of the extensively studied "reference-design" MSBR which is described in detail in another report.[1] No consideration is given to a wide variety of other conceivable MSRs (e.g., two-fluid systems) which are less well-developed, since such systems would not be expected to exhibit greatly different characteristics with respect to proliferation potential.

Although the systems to be described offer varying degrees of resistance to the potential diversion of SNM, there are some characteristics of MSRs in general that are significant in this regard. These reactors would be expected to operate principally in the Th-^{233}U fuel cycle with breeding ratios near 1.0. Hence, the fuel would be primarily ^{233}U with significant amounts of the isotopes ^{234}U, ^{235}U, and ^{236}U. (The equilibrium uranium isotopic composition for the reference-design MSBR is about 65, 23, 6, 6 atomic percent o£ these four isotopes.) The systems would contain specific inventories of fissile isotopes in the range of 1.5 to ~3 kg/MWe for 1000-MWe reactors, with somewhat higher values for smaller versions. Thus a 300 MWe system would contain 500 to 1000 kg of fissile nuclides (^{233}U + ^{235}U) at high enrichment, and these nuclides would be present as UF_4 in a mixture of fluoride salts, including fission-product fluorides. This mixture could be fluorinated (bubbling gaseous F_2 through the molten salt) in relatively simple equipment to recover UF_6 in a highly purified state. (Uranium decontamination factors of 10^9 with respect to fission products were demonstrated with this process in the molten-salt reactor experiment [MSRE].) Consequently, the MSRs described below—as well as all other MSRs— are potential sources of large quantities of high quality SNM.

Any operating MSR will, to some degree, be subject to contamination of the fuel by oxides (moisture or air intrusion), which could ultimately lead to precipitation of uranium as UO_2. Since the point where such precipitation is likely is on the order of 50 to 100 ppm oxide in the salt, any practical MSR installation must include capability for treating the salt with HF to remove oxides. This implies the existence of facilities for sparging the salt with a gas and for introducing and removing gaseous materials from within the reactor containment, either continuously or on an intermittent basis. The potential for transforming this capability into capability for adding F_2 and removing UF_6 probably is not negligible.

Although most monitoring of the chemical condition of the fuel salt in an MSR could probably be accomplished by in-line analytical techniques, it is likely that capability would have to be provided to occasionally remove samples of the fuel salt for laboratory analysis. The amount of material that could be removed from the reactor through a sampling system would be quite small (less than 5g of SNM per day), but the sampler would represent another penetration

of the system enclosure. Thus no MSR, however independent it may be of external fueling requirements, can be regarded as a totally closed system.

The MSR systems considered in this study would all be expected to have lifetime average breeding ratios very close to 1.0. However, in practice the breeding or conversion ratio would vary somewhat with time. This would be pronounced if the reactor were started up with ^{235}U rather than with ^{233}U. Furthermore, the reactor operator would require enough flexibility in his fissile inventory and potentially available reactivity to operate the plant under foreseeable transient conditions. Not only would he need to go through a reasonable plant startup transient, but he would expect to be able to follow normal electrical load demand variations and also be able to meet load demand when all or parts of the processing plant may not be operating. He probably also should be able to operate the processing plant with the reactor shut down. Consequently, to break even on the average, there would be periods when the instantaneous conversion ratio would exceed 1.0 and/or when the fissile inventory would exceed the minimum for criticality. Thus, one could not depend on the reactor shutting down promptly if any fissile material were diverted, if one expected to have a plant which a utility would find acceptable from a reliability standpoint. This implies that plant operability, alone, would be inadequate as a monitor for the full system inventory of SNM in an MSR. The amount of SNM that could be diverted from any given reactor system without rendering it inoperable is, of course, dependent on the quantity of excess material that must be present to support the projected normal operating cycle. This excess inventory may be significantly smaller in some MSR concepts than in solid-fueled systems for comparable duty.

If an MSR were started up with ^{235}U, the initial conversion ratio would of necessity be < 1.0 and an excess fissile inventory (or feed) would be required (perhaps along with burnable poison). Subsequently, as ^{233}U built up, the conversion ratio could exceed 1.0 and then eventually decline as fission-product poisons grew in. In the event that the reactor were started with ^{233}U, or an equilibrium isotopic mixture, the clean initial conversion ratio would exceed 1.0 initially (after achievement of equilibrium Pa loading), and approach 1.0 at steady state.

We believe that the intent of most of the design specifications would be met if the fissile inventory were secured within the biological shield with access carefully monitored. For this purpose, it might prove desirable to maximize breeding gain in the early stages of reactor operation in preference to feeding a larger amount of fissile material from an external source, as would otherwise be required to meet the desired reactor lifetime.

The MSBR reference design is for a high-performance, high-power-density system with a low specific inventory of fissile material. The long-term operation of this system would require, among other things, periodic (every 4 years or so) replacement of a portion of the graphite moderator in the reactor. These operations would require a high level of system maintenance capability (and, by inference, ability to modify the plant) along with the requirement for routine opening of reactor shield. In considering systems for export, it was concluded that the core graphite should not require replacement over the life of the plant. This criterion dictates that the candidate systems be based on the low-power-density version of the MSBR, which has been studied as a breeder and as a high-gain converter.[2,3,4] This reactor system would be well suited to a total power level corresponding to a few hundred megawatts (electric), and it could be limited to a net breeding ratio of essentially 1.0. All the systems described below are variations on this reactor concept. Five candidate systems are described below as variations on this basic reactor concept. They are

1) a break-even MSBR with the reference fuel reprocessing system,
2) a system with processing by reductive extraction only, and no Pa isolation,
3) an MSBR with processing to remove only rare-earth fission products by exchange with CeF_3,
4) an MSBR with rare-earth fission-product removal by vacuum distillation, and
5) an upgraded molten-salt converter reactor with no on-site chemical processing.

In all of the candidate systems, it is presumed that gas stripping[*] would be employed to remove noble-gas fission products and some volatile semi-noble metals (but not halogens).

Break-Even MSBR with Reference Fuel Processing

In this concept, all the essential features of the reference-design single-fluid MSBR would be retained but modified to enhance the non-proliferation characteristics. Decreasing the breeding ratio to a nominal value of 1.0 would be accomplished by reducing the fertile and fissile inventories to about 70 percent of the calculated values for higher performance systems (see Ref. 2). Thus, a 300 MWe plant would have a specific fissile inventory of about 2 kg/MWe.

The processing scheme for the reference MSBR (Figure 1) comprises the following steps performed on a side stream from the reactor system.

a. Most of the uranium (>95 percent) and volatile fission products are stripped in a continuous fluorinator. The UF_6 so removed is decontaminated and promptly returned to the fuel salt and reduced to UF_4.

b. Protactinium is removed from the uranium-depleted fuel-salt stream by reductive extraction into bismuth containing dissolved metallic ^7Li as the reductant. The Pa and residual U in the bismuth stream are oxidized with HF and transferred to a captive salt volume where the isolated ^{233}Pa is stored until it decays to ^{233}U. A continuous secondary fluorinator recovers uranium from the Pa salt as UF_6 which is reduced and returned to the primary fuel salt.

c. Uranium and protactinium-free salt from the protactinium extractor enters a second reductive extractor where it is contacted with Bi containing 0.2 percent Li to extract some o£ the rare-earth, alkaline-earth, and alkali-metal fission products. The resultant bismuth stream is treated in a metal transfer system in which the fission products are transferred to a LiCl stream from which they are again stripped by contacting with Bi-Li alloys, containing high Li concentration, in two packed columns. The isolated fission products are stored in the circulating bismuth alloy systems for partial decay and are periodically extracted and packaged for disposal by using semi-continuous processes.

After removal of the rare earths, the fuel salt is reconstituted with internally recycled uranium and returned to the reactor system after adjusting the composition by addition of BeF_2 and ThF_4, and treatment to eliminate Bi, remove noble metals and adjust the U^{4+}/U^{3+} ratio. Use of Li reductant results in a potential buildup of LiF which is compensated by discarding barren salt in conjunction with additions of BeF_2 and ThF_4.

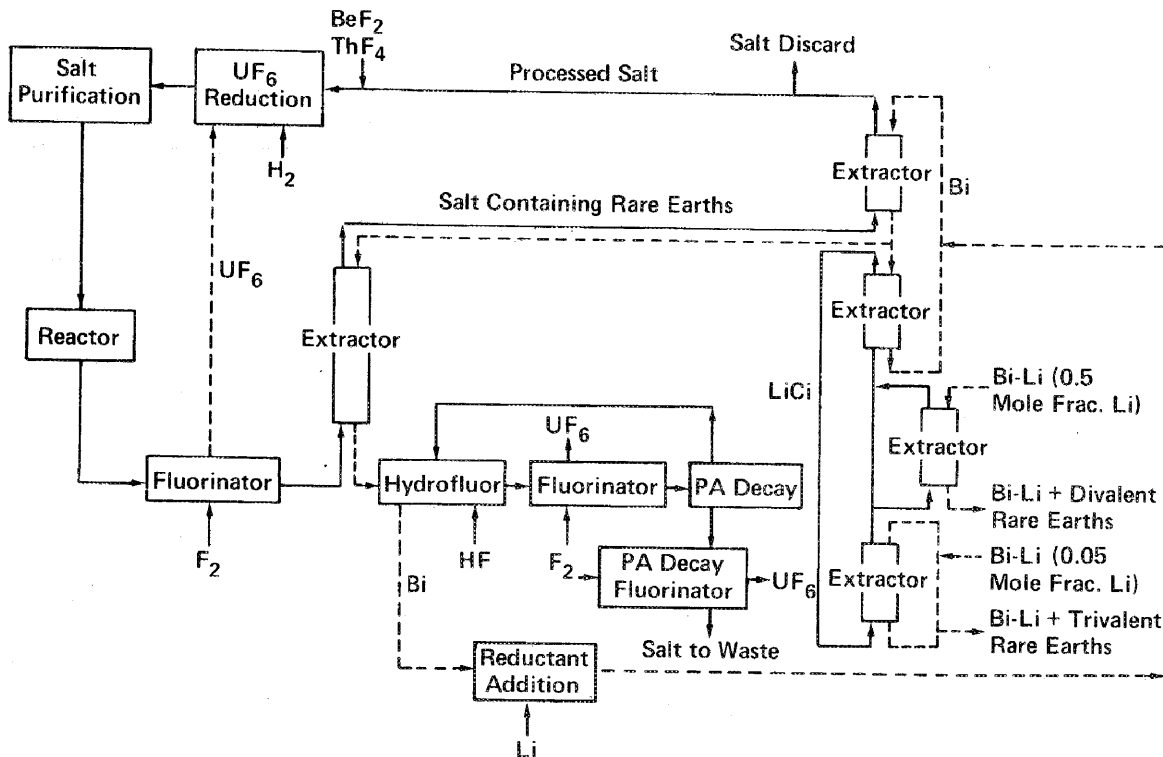[*] This gas-stripping system would not be adaptable for removal of SNM.

Figure 1: Conceptual flow sheet for processing a single-fluid MSBR by fluorination-reductive extraction and the metal transfer process

The reference MSBR plant also has systems for removal of gaseous fission products and noble metals and means for extracting bred uranium and recovering trace amounts of uranium from waste streams. Since the export system with reduced breeding ratio would not produce excess fissile material, no equipment would be provided for removal of uranium from the system enclosure.

Since this approach would permit complete processing on site, it would have the best overall nuclear performance of the five candidate systems considered. As indicated above, the nominal MSBR performance could easily be downgraded to achieve a breeding ratio of 1.0 while, at the same time, reducing the fissile specific inventory to ~2 kg/MWe. In addition, the Pa holdup and decay loop would provide the means for adjusting the fissile concentration in the fuel salt as required during the life of the plant.

This system has a number of features that would make it relatively susceptible in comparison to other MSRs to the diversion of SNM. In addition to the features common to all MSRs, the system would have at least two built-in fluorinators which could produce UF_6 in a highly purified form. Extraction of the UF_6 would require some modification of the system which would be quite difficult in view of the high levels of nuclear radiation throughout the system. However, such modifications would be substantially easier than if there were no fluorinators in the system. The reference processing system also includes final fluorination of salt that is to be discarded, to recover any residual uranium. While such residues would normally be small, the processing system could, in principle, be operated in such a way as to leave more uranium for recovery from the salt that is discarded, especially in the discard from the Pa-

isolation system. Retention of the "discarded" salt within the system enclosure would reduce, but would not totally eliminate, this avenue for diversion of SNM by the operator of the plant.

Reductive Extraction Without Protactinium Isolation

In this concept the reference MSBR processing scheme would be modified by eliminating all fluorination steps. (Figure 2) Both uranium and protactinium would be removed from the process stream by reductive extraction into Bi-Li alloy. The rare-earth removal system would be basically identical with that for the reference MSBR. Instead of using an isolated Pa salt, the uranium and protactinium would be transferred directly into the barren salt leaving the rare-earth extractor, using HF as the chemical oxidant. A captive storage volume would be provided in the fuel-salt reconstitution loop to permit transient storage of uranium in excess of current reactor system needs, along with a fraction of the Pa inventory. The breeding ratio would be permitted to exceed 1.0 early in the reactor operating life with any excess uranium stored in the captive storage volume and subsequently returned to the reactor system to compensate for declining reactivity later in plant life as non-saturating poisons were built up. The same volume could be charged with extra fuel at startup of the plant to provide for requirements during the initial transient. The plant would have no built-in capability for removing fissile material from the biological shield and, in principle, would approach asymptotically a self-sustaining capability with a breeding ratio of essentially 1.0.

The elimination of the primary fluorinator for removal of uranium would lead to a relatively large requirement for reductant lithium with a corresponding tendency to build up the LiF concentration in the fuel salt. This would be counteracted by additions of BeF_2 and ThF_4 and a purge of barren fuel salt to maintain the desired fuel-salt composition. With a postulated fuel processing cycle time of 50 days (vs. 10 days for the reference design), the salt purge rate would be about 10 percent of the reactor inventory per year. In principle this salt, which would contain only trace concentrations of SNM, could be stored within the system containment for the life of the plant.

Elimination of the Pa-isolation step is made practical by the low power density of the proposed reactor system. The breeding-ratio penalty would be only about 0.004 greater than that in the reference (high power density) system with Pa isolation on a 10-day cycle. The salt discard rate of 10 percent per year is comparable to the rate proposed for the reference-design MSBR, so the buildup of fission-product poisons that remain in the salt would be comparable to the reference system.

Thus, rare-earth removal on a 50-day cycle would lead to overall neutronic performance only slightly less favorable than that for a plant using the reference processing scheme. There appears to be adequate margin in the nominal breeding ratio to accommodate this loss without loss of the self-sustaining character of the reactor. However, this system would require a modestly higher specific fissile inventory.

Removal of the fluorinators from the processing flowsheet would significantly reduce the potential for diversion of SNM from the reactor system. (Providing and installing fluorination capability is presumed to be much more difficult than modifying existing capability or operating procedures.) Thus, this approach would retain most of the attractive fuel-cycle features of the system with the reference processing scheme while enhancing its non-proliferation characteristics.
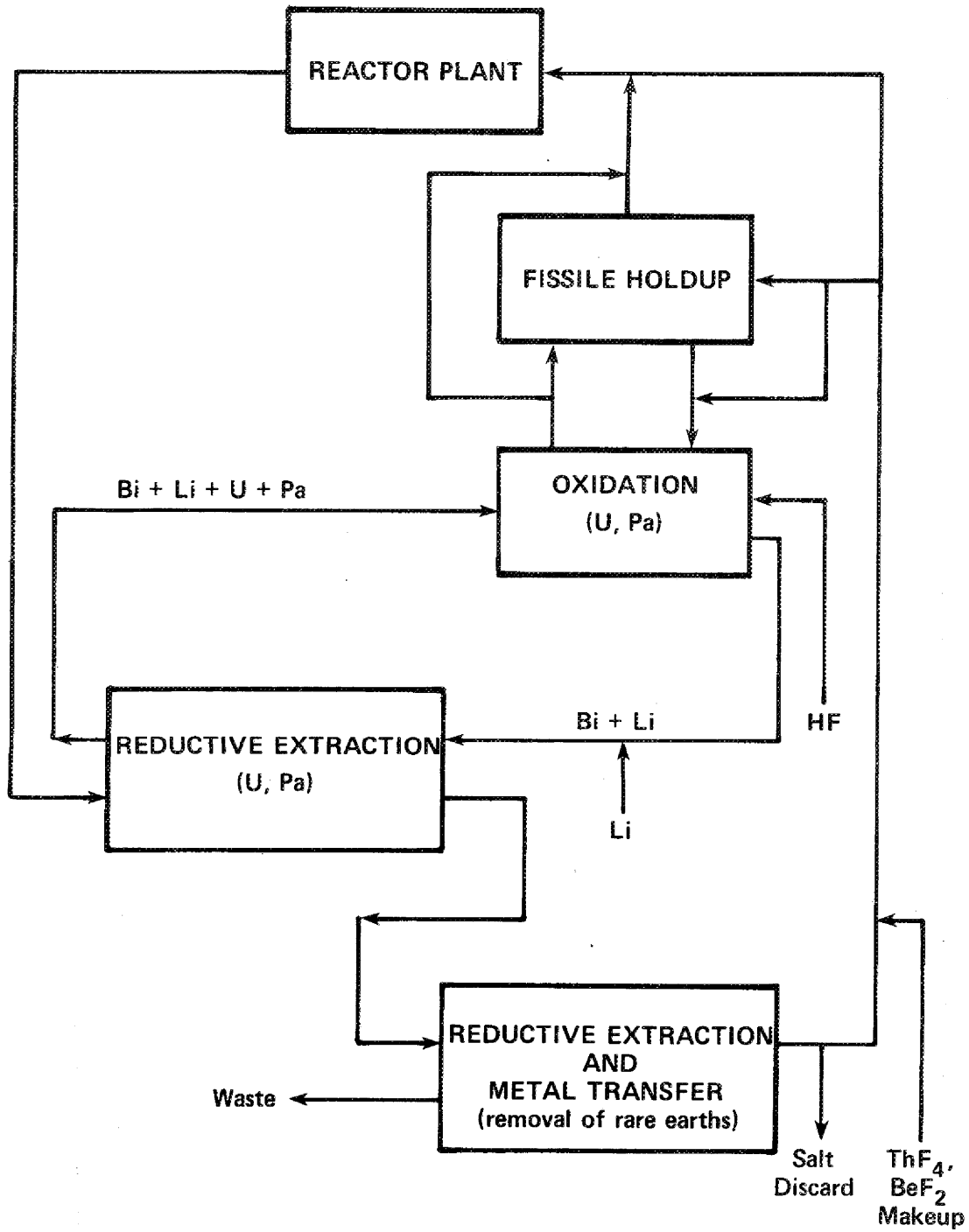
Figure 2: Conceptual flow sheet for rare earth removal without fluorination and Pa isolation.

MSBR with Only Rare-Earth Removal by Exchange with CeF$_3$

In this concept, chemical reprocessing, other than xenon removal with its associated off-gas handling and the processing (presumably involving treatment with HF) for control of oxide contamination, would be limited to removal of rare-earth fission products by exchange with beds of CeF$_3$.

The rare-earth trifluorides are all very similar, both chemically and crystallographically. They are sparingly soluble in the molten-fluoride mixtures of interest as reactor fuel, and from such fluorides they crystallize as the simple trifluorides upon cooling of saturated solutions. When more than one lanthanide trifluoride is dissolved and the melt cooled below the saturation temperature, a solid solution of the rare-earth fluorides results. The ratio of the rare earths in the solid trifluoride is essentially identical to the ratio of the rare earths in the molten fluoride from which the solid solution crystallized.

Accordingly, if a portion of the reference fuel were removed from the reactor, treated with CeF$_3$ equivalent to twice the quantity that would dissolve at 500°C (that is with 41 grams of CeF$_3$ per kg of fuel), heated to a temperature such that this quantity dissolved, homogenized, and then slowly cooled to 500°C to recrystallize the excess CeF$_3$ (20.5 grams/kg), the recovered crystalline CeF$_3$ would contain one-half of the rare earths contained in the fuel. Laboratory experiments of this sort have shown that no LiF, BeF$_2$, ThF$_4$, or UF$_4$ would be included in the crystals. The fuel would, however, contain 20.5 grams of CeF$_3$ per kg and would have to be returned to the reactor in this condition. Substantial removal of the rare-earth poisons would have been accomplished, but the poisoning by dissolved CeF$_3$ would be appreciable.

A partial processing method, which could prove attractive but which has not yet been demonstrated, would draw from the reactor a small sidestream of fuel (saturated at 500°C with CeF$_3$), cool the stream to 500°C, and pass it through a column packed with crystalline CeF$_3$ and maintained at 500°C. The fission-product trifluorides (primarily yttrium and the rare earths)[*] would equilibrate to form solid solutions with the CeF$_3$ and a portion would be retained on the column. The salt emerging from the column for return to the reactor would, of course, be saturated (at 500°C) with CeF$_3$. It appears that the concentrations of high cross-section rare earths could be kept to sufficiently low values in the fuel by proper choice of column design and processing rate. If rates of equilibration were such that the crystalline CeF$_3$ could be used with relatively high efficiency, the requirements for this compound would appear to be a few tens of tons per year at most.

If the process could perform as described above, the fuel would require little auxiliary processing. To avoid loss of uranium to the CeF$_3$ it would be necessary to ensure that the uranium all be present as UF$_4$; it would also be necessary to ensure that europium be present as EuF$_3$[**]. Such oxidation could be accomplished by use of HF and might be done in combination with the oxide removal process. In addition, since some UF$_3$ in the fuel is desirable or even essential, slight reduction of the fuel after passage through the column would be required. A small addition of lithium or (probably more conveniently) of beryllium would suffice.

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[*] Plutonium, if any, in the fuel will exist as PuF$_3$ and will also be retained on the column. It may be economical to recover this when the CeF$_3$ is ultimately returned with the spent fuel from the reactor facility.

[**] UF$_3$ and EuF$_3$ will be partially removed by the column; UF$_4$ and EuF$_2$ will not.

It should be emphasized that no other useful fission product separation can be expected from this system[†] and that no other removal (as of ThF_4) can be accomplished by it. Some fuel mixtures differing from the reference fuel—but probably still in the useful range—can apparently have a lower (perhaps a twofold lower) CeF_3 solubility. Should the system appear to be of interest, this possibility should be examined. It should also be noted that no genuinely "insoluble" rare-earth exchanger is known or is likely to be found; CeF_3 is likely to be the best material available for the purpose.

Much research and development would remain before (or even if) such a process could be realized. Long-term stability of such beds in the slowly flowing fuel could well be a problem. Recrystallization of the CeF_3 will likely occur. It may be that large crystals would form and limit exchange only to near-surface layers. Heat loadings on the bed would become relatively large, and close temperature control such as would be required would probably be difficult. Though the major fuel components appear not to load on the bed—and although no useful removal of other than rare-earth fission products could be expected—it may be that other fission products would adversely affect the separation.

It is not now possible to estimate the development cost of such a system. Much would remain to be done and it is certain that many years of sequential development would be required before the system could be realized in practice. It is possible, moreover, that a proper choice of engineering experiments might, in a relatively short time, show that the notion has no really attractive embodiment.

If this processing approach were developed and coupled to a low-power-density MSR, a reasonably attractive overall fuel cycle would result that might allow reactor operation for as much as 30 years without fuel addition. With the poisoning due to rare-earth fission products stabilized at a low level, it would be necessary to compensate only for the near-linear buildup of non-saturating and slowly saturating fission products. This likely could be accomplished by tuning the system to have a breeding ratio very slightly greater than 1.0—a reasonable prospect considering the excess breeding gain available in the reference system. It still would be necessary to provide some excess fuel in the initial loading and a burnable poison—^6Li may be a reasonable candidate—tailored to fit the initial negative reactivity transient due to buildup of the steady-state Pa inventory (and buildup of ^{233}U if the system were started on ^{235}U). Such a system would have a specific fissile inventory near that of the nominal breeding systems, that is, ~3 kg/MWe in the 300 MWe range.

The system, as described, would contain no provisions for removal of fissionable materials or of other constituents, besides noble gases, rare-earth trifluorides and some volatile fission products plus oxide ion, from the fuel. Accordingly, it would be necessary to install additional equipment if removal of fissionable material from the system were to be accomplished. (Such installation should be obvious to even a casual inspection but it could, of course, be done.) Addition, for example, of equipment for purification and collection of UF_6 at the outlet of the oxygen removal system and substitution of fluorine for HF as the treatment gas would suffice.[*] Extensive corrosion of the oxide removal system would result, but—given that removal was limited to a few tens of kg of SNM—the reactor would probably be operable, albeit with some impairment in performance, for a considerable period.

[†] It may be that the column may have filtering action on suspended insolubles such as fission product Mo, Ru, etc., and some limited exchange to I^- for F^- may occur. Such actions may be mildly beneficial to reactor operation.
[*] That this is not an inherently safe thing to do would hardly be guaranteed to deter an organization sufficiently interested.

MSBR with Rare-Earth Removal by Vacuum Distillation

In this concept, chemical reprocessing, again other than xenon removal and its associated off-gas handling and the processing for control of contaminant oxide,[**] would be limited to removal of fission products that are retained in the residue from vacuum distillation of the major fuel constituents. Fission products that should be effectively removed in this process appear to be limited to the rare-earths and the alkaline-earths (Sr and Ba) whose fluorides are relatively nonvolatile. The small fraction of noble metals (Mo, Ru, etc.) that would accompany the fuel in the metallic state should also remain in the distillation residues.

Early studies of vacuum distillation focused on decontamination and recovery of LiF, BeF_2, ZrF_4 and UF_4 from MSRE fuel and from similar mixtures anticipated as fuels for two-fluid molten-salt breeders. These studies showed that essentially quantitative recoveries of LiF, BeF_2, and ZrF_4 could be obtained in batch distillations; distillation until 98 percent by weight of the charge had been collected as condensate seems to have left only UF_4 (of the major fuel constitutents) in the still bottoms. Though attended by considerable engineering difficulties, continuous distillation of MSRE carrier salt (the uranium had been removed by fluorination) showed that relatively large separation factors (100 to 1000) could be obtained for rare-earth fluorides and for strontium and barium fluorides. These early studies suggested that distillation procedures could probably be devised to recover the fuel constituents, including the uranium from salts of this type.[†]

There can, however, be no doubt that introduction of large quantities of thorium into the fuel salt makes distillative recovery of the fuel values more difficult. The few studies performed (and which did not include UF_4) suggested that adequate distillation rates might require still temperatures of 1200°C.

In practice one could almost certainly accept an incomplete recovery of ThF_4 but this is not true of UF_4 or of PaF_4. Accordingly, and before vacuum distillation were accepted as a possible reprocessing scheme, it would be necessary to establish that very good recoveries of UF_4 and PaF_4 are realizable. The high vapor pressure of UF_4 at about 1200°C would suggest that it is possible, but the ability of UF_4 and its several complex compounds with LiF to form solid solutions with ThF_4 and its largely analogous LiF compounds would make this complete recovery difficult.

It might also be noted that molten fuels differing from the reference fuel—but still in a useful composition range—might be capable of fractional crystallization to remove a substantial fraction of their contained ThF_4, UF_4, and PaF_4 before distillation. This process would make the necessary high recoveries of the values easier, but at the expense of considerable complication of the process.

All in all, the vacuum distillation process would, if it could be developed, promise an effective separation from the rare-earths and strontium and barium. Plutonium, if any, would probably remain in the still bottoms. Fission product zirconium, cesium, rubidium, and iodine would certainly distill with the fuel values. Distillation may offer a way of discarding some thorium from the cycle—and such discard might be advantageous in some applications—if uranium and protactinium could be recovered sufficiently from the residual thorium. This possibility would certainly require detailed experimental confirmation.

[**] It is possible, but unlikely, that oxide contamination can be managed by retention of oxides in the still residues.

[†] Though it was early recognized that UF_4 was the most difficult of recovery, and since other means (such as fluoride volatility) were available for its recovery, relatively few such distilling studies were made.

It must be clear from the above that a large program of research and development would be required before this process could be considered truly useful for the high-concentration ThF_4 fuel mixtures. If sufficiently quantitative uranium recoveries were demonstrated from reasonable fuel mixtures, or from mixtures reachable by fractional crystallization from such fuels, then it is likely that useful fission-product separations could be accomplished.

The reactor associated with this processing scheme would have very nearly the same fuel-cycle performance characteristics as one using CeF_3 removal of rare earths. With the absence of CeF_3 and the removal of Sr and Ba, the system would have a slightly larger breeding margin and, hence, a somewhat lower specific fissile inventory for break-even operation. In addition, the ability to remove some Th (provided it were not accompanied by U and/or Pa) could add some operational flexibility.

The system, with its installed equipment like the CeF_3 exchange process above, would contain no provisions for removal of fissionable materials from the fuel. The process, if it could be satisfactorily demonstrated, offers the slight advantage (over CeF_3) that it can remove SrF_2 and BaF_2 from the salt along with the rare-earth trifluorides and it might offer a way to discard ThF_4 to prolong the useful life of the reactor. It would have a more real advantage (over CeF_3) if, as is unlikely, it could also serve as the mechanism for removal of oxide contamination.[*] In that event, installation of a fluorinator and a UF_6 purification and collection scheme (rather than modification of an existing hydrofluorinator) would be required to divert fissionable material. However, such installation would be possible, given a sufficiently determined effort. Thus even if the vacuum distillation procedure could be demonstrated to have every virtue mentioned above, its resistance to material diversion would not be very different from that of the system with CeF_3 processing.

The nearly marginal advantages, combined with the almost certain difficulties in genuinely successful demonstration of the system, suggest that vacuum distillation should not be given major consideration as a reprocessing method.

MSCR with No On-Site Fuel Processing

Molten-salt reactors without continuous fuel processing (except for fission gas removal) have been studied extensively.[5] While not intrinsically different from MSBRs except for processing, the conversion ratios for such systems are generally less than 1.0 (typically 0.85 to 0.95) and they have therefore been called molten-salt converter reactors (MSCRs). In the cases studied, a fuel charge would remain in the reactor for either 6 or 8 equivalent full-power years (efpy), where the reactor lifetime is taken to be 24 efpy, or about 30 years of operation at 0.8 plant factor. While the operating cycle may be long enough to be of interest for a non-proliferation reactor, the usual MSCR cycle requires continual addition of fissile fuel to maintain the reactor critical. To meet the criteria of the non-proliferation reactor, the MSCR would have to be modified to eliminate the need for fissile feed and to provide another means for controlling the reactivity (keff).

To achieve these objectives, the initial charge could be modified to contain all the fissile material required for the entire operating cycle. A good way to do this would be to boost the conversion ratio to near 1.0 by increasing the amount of thorium in the charge, which would also increase the amount of fissile material required. This system appears to be quite feasible, and

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[*] The process would probably leave any oxide contamination in the still bottoms, but this oxide may be combined with (and render undistillable) some uranium and protactinium.

would not be greatly different from at least one of the MSCR cases studied, in which a lifetime-averaged conversion ratio of 0.98 was obtained based on a fuel salt containing 14 mole percent thorium.

The problem of controlling reactivity with adding or removing fissile material also appears tractable. The largest change in reactivity occurs during the first few months of the cycle, as the rapidly saturating fission products build in. This change is suited to control by a burnable poison added to the fuel charge. Reactivity changes would be small during the remainder of the cycle and could be controlled by conventional shim rods in the core.

The fissile materials usually considered for the startup of molten-salt reactors are fully enriched uranium, recycled plutonium from light-water reactors, and recycled ^{233}U from molten-salt reactors. The overall performance of the system is little affected by the starting material, since (because of the high conversion ratio) after a few years the main fuel in the system becomes ^{233}U. For this reason, molten-salt reactors have been examined as burners for the plutonium generated in light-water reactors. The plutonium could be utilized without requiring the fabrication of fuel elements, and with minimum requirements for plutonium fuel transportation.

The performance of the non-proliferation system can be estimated by comparison with appropriate cases from the MSCR studies. It was assumed that the average conversion ratio for the non-proliferation system would be roughly equal to the end-of-cycle conversion ratio for a comparable MSCR system, because of the effect of the control poison required. For a reactor designed for an average conversion ratio of 0.95, the estimated fissile specific inventory is about 3 kg/MWe and the estimated lifetime (24 efpy) fissile requirement is about 1 kg/MWe. It is expected that this system could operate on an 8-efpy cycle, which would require two fuel-salt changes during the life of the reactor. Thus the fissile inventory that would be removed with the fission-product-laden salt charge at the end of a fuel cycle would be approximately equivalent to that installed with the new salt charge. Even longer cycles, such as 12 efpy requiring only one salt change per lifetime, appear possible but would have to be confirmed by further studies.

The diversion resistance of the MSCR-type system would be approximately equivalent to the previously described systems with processing, except that the reactor would have to be opened up under supervision one or more times during its lifetime for replacement of the fuel salt.

DEVELOPMENT PERSPECTIVE

Obviously the non-proliferation objectives could not be met unless an acceptable reactor system could be devised and until such a reactor system had been developed to the point where it could be deployed. In the case of the MSR it would be necessary, as a minimum, (1) to develop, design and demonstrate a reactor system with the desired charateristics, (2) to develop, design and demonstrate the associated processing plant, and (3) to produce the fuel inventory, particularly if this were to contain ^{233}U.

Two alternative development scenarios can be visualized as leading to a novel non-proliferating reactor system. If the basic reactor concept were attractive for commercial use in the United States, it conceivably would be developed first in a version suitable for domestic deployment. Subsequently, a modified version incorporating the non-proliferation features would have to be designed and demonstrated before it would be available for export. On the

other hand, if the system were deemed to be attractive solely as a non-proliferation concept, then the total development effort and any demonstration plant(s) would have to be charged against meeting the non-proliferation goal. In the case of the molten-salt reactor, development for domestic applications has been terminated. Consequently, it would be necessary to reestablish the molten-salt reactor project with the goal of developing a commercially attractive reactor system before the first scenario could be followed. The reactor and its associated processing plant would be designed as a high performance system, presumably essentially the reference MSBR. The subsequent non-proliferation version would draw on the technology demonstrated in the domestic commercial reactor project with either suitable modification or substitution of alternative features. If the domestic commercial goal were not reestablished, the second scenario would be followed. Some features of the high-performance domestic reactor might be bypassed but essential features would still have to be demonstrated. It is not clear that there would be any significant improvement in the schedule and, as noted, the total cost of development would have to be borne by the non-proliferation project.

It should also be noted that even when the non-proliferation version had been demonstrated by either scenario and had become available for deployment, there would still be a question of whether the resulting plant would be acceptable to the prospective customer as a viable commercial plant, economically or otherwise.

Development Needs

The several non-proliferation concepts differ somewhat in development status and required further development. All would use essentially the same basic reactor design. Moderate variations in thorium concentration, fuel-salt fraction, or uranium concentration which may be needed to achieve the desired breeding gain are considered to be minor with essentially no difference in development requirement. Concepts which require use of burnable poisons or other schemes to control excess reactivity and thus hold k_{eff} relatively constant over long periods of time to avoid addition or removal of fissile material may require some modest additional development effort. By far the major differences in development requirement are occasioned by differences in the proposed processing schemes. Considerable effort has been expended on developing the conceptual flow sheet and design for the reference MSBR. However, detailed design of some of the equipment to implement the concept still requires some fundamental data and, in some cases, choice of materials and successful demonstration. This is particularly true of the frozen-wall continuous fluorinators and the contactors for the reductive-extraction and the metal transfer steps. Other schemes such as the CeF_3 process and vacuum distillation have been the subject of some development effort in connection with previous molten-salt reactor concepts and, while they are not demonstrated, may not necessarily require greater development effort than the reference MSBR processing scheme.

The first concept described above would utilize the same basic processing steps as the reference MSBR. Consequently it could be expected that the successful development of a commercial MSBR would provide all the essential technology with virtually no need for additional development effort. The second concept, reductive extraction without Pa isolation, would use technology which would be demonstrated by successful development of a commercial MSBR. Furthermore, this concept would eliminate the need for a frozen-wall continuous fluorinator and thus would not require the completely successful development of a commercial

high-performance MSBR. If the commercial MSBR were bypassed, the required development might be somewhat less than for concept 1.

Concept 3, the CeF_3 removal of rare earths, appears to be an inherently simpler system than the reference scheme, with a correspondingly smaller effort required for development if it were substituted for the reference process. If it were developed in addition to the reference process, the total development effort would be greater but, because the CeF_3 process could be developed in parallel, the elapsed time to deployment of the non-proliferation system would not necessarily increase.

The vacuum distillation concept would require considerable development effort not contemplated for the reference commercial MSBR.

Complete elimination of fuel processing would, of course, require no processing development and, depending on the particular scheme chosen for controlling excess reactivity, could require only minimal overall development effort beyond that needed for demonstration of the reactor system.

Availability of Fissile Material

It is generally conceded that a self-sustaining MSR system would operate with ^{233}U as the principal fissile nuclide (albeit with equilibrium ^{235}U content). If a particular system were to be started with ^{233}U, there would be a problem in furnishing the initial inventory. It could be derived from excess bred ^{233}U from other MSBRs. However this would require the deployment of high-gain MSBRs and many reactor years of operation. Alternatively, other reactor types operating in the converter mode could produce ^{233}U. Such reactors do not currently exist in the private sector and would have to be built (and perhaps first developed). Potential converter types include heavy-water reactors, high-temperature gas reactors, and MSRs.

The alternative would be to use ^{235}U (or possibly Pu if deemed acceptable) as the initial fissile fuel and to generate the steady-state ^{233}U inventory in-situ. Such an approach would tend to relieve the supply requirement on the initial fuel inventory because enriched ^{235}U is available. However, the reactivity transient as ^{233}U builds in is larger for this type of startup and it remains to be shown in detail that the entire transient can be handled by an acceptable combination of burnable poisons, control rods, and in-plant storage of extra fuel.

Cost and Time for Development and Deployment

The cost and time effort for a domestic MSBR program have been estimated[*] at \$3.25 billion (1975 dollars) and 17 years. If the program objectives were concentrated upon the export reactor goal, some savings would accrue but the time scale probably could not be collapsed. Thus, taking into account both the development time and project construction time, it is hardly possible to have a reactor in operation overseas in less than 25 years.

Considering the great effort and cost involved in developing a molten-salt reactor system, it seems reasonable that export should be considered only if there is a substantial MSBR program in the United States that is justified upon its merits as a competitive power-producing system alone. Without such a program the broad-based industrial manufacturing capabilities needed could not develop. In addition, a relatively small export volume could not bear, alone, the entire development cost.

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[*] L.F.C. Reichle, EBASCO Services Inc., letter to R. W. Roberts, USERDA, March 12, 1976.

Incentives for Development

An important consideration in the evaluation of anti-proliferation measures is timing. If one accepts the view that by, say, the mid-nineties or the turn of the century many countries can have nuclear weapons if they want them strongly enough, even without civilian nuclear power, then anti-proliferation measures that could not be available for 20 to 30 years may be of questionable value; measures that can be immediately effective may lose their effectiveness in 20 to 30 years unless they bar all pathways to nuclear weapons, not just power reactors and processing plants.

It remains a valid question whether over the longer haul certain nuclear technologies or institutional arrangements may be more intrinsically resistant than others to diversion of SNM by sub-national groups, thus reducing society's vulnerability to terrorist acts. In this connection, MSRs as a class and in particular the versions described in this report appear to be inherently more diversion-resistant than the mixed-oxide fuel cycle whether used in light-water reactors or liquid-metal fast breeder reactors.

Although it is the considered opinion of the members of this study group that the MSR does not offer sufficient advantages over other reactor systems as a single, stand-alone, diversion-proof reactor to warrant its development for this single purpose, it is believed that, on the broader question of safeguards against the diversion of plutonium produced in US reactors, the MSR offers an attractive way of limiting the total buildup of plutonium. Fuel-cycle scenarios have been studied in which a high-conversion-ratio MSR, charged with Pu, would operate without fuel reprocessing to burn up the plutonium and generate ^{233}U in the process. After several years of operation, a batch reprocessing operation would recover the ^{233}U which could be used to fuel a high-performance breeder or to transform the converter to a thorium-^{233}U breeder. The end products of the Pu charged into the reactor would be essentially free of ^{239}Pu, ^{240}Pu, and ^{241}Pu, with about 12 percent of the original mass as ^{242}Pu or higher-mass capture products. The fact that this scheme would lead to predicted fuel-cycle costs which are lower than the light-water reactor fuel-recycle cost is a bonus. The risk of Pu diversion would thus be reduced by (1) limiting the total world inventory of Pu, and (2) eliminating fuel fabrication, transportation, and reprocessing of material high in ^{239}Pu.

SUMMARY AND CONCLUSIONS

The results of this study of molten-salt reactors as exportable, non-proliferating nuclear power plants may be summarized as follows:

- Molten-salt reactors could be designed to (a) eliminate requirements for traffic in SNM to or from the reactor over long periods of time (e.g., 30 years for versions with limited in-plant processing from removal of fission products or 8 to 10 years for MSCR versions with no fission-product removal except rare gases); and (b) make difficult the extraction of SNM from the reactor inventory owing to the elimination of devices and processes (e.g., fluorination of salt) for doing so and to the highly radioactive state of the salt systems and their contents.

- At least two candidate MSR configurations are highly resistant to diversion but cannot be classed as diversion-proof. These are the CeF_3 processing scheme and the concept

with no chemical processing. The use of CeF_3 to remove other rare-earth neutron absorbers with a variable processing rate can achieve a near-break-even system that is entirely enclosed with little need for access to the system. The simplicity of the reprocessing unit and its continuous operation permit the use of only short delays between extraction of the salt from the reactor and its return, with no removal of fissile material at any time. Complete elimination of the processing system would require the least interaction with the salt mixture in the reactor, and any materials that need be added would be non-fissile and could be safely inventoried for the expected fuel cycle lifetime. Based upon present data with defensible extrapolations in time, this cycle would be about 10 years. The actions that would be taken to reconstitute or replace the reactor mixture would require the same level of safeguards as in the initial commissioning of the reactor.

- Two additional systems, less resistant then the two above, are the reductive-extraction process without Pa isolation and the salt distillation process. The first of these is, perhaps, the most desirable operationally, but it may require a sizable volume of out-of-reactor salt with numerous sampling points (needed for process control) which might permit conscious diversion of SNM. Of course, the diverted material would then need to be fluorinated to extract the uranium. No fluorinator is provided in this system, and one would have to be provided elsewhere or clandestinely installed. With the reactor in operation at full power, the radiation levels would make this difficult. Even after extended shutdown the radioactivity of the system presents a considerable barrier. The salt-distillation approach offers somewhat similar barriers to diversion, but it is considered to be more questionably feasible. It cannot be excluded from consideration, however.

- Finally, the system based on the reference MSBR processing flow sheet, since it requires salt fluorination capability, is significantly less resistant to diversion of SNM than a system without fluorination.

Thus diversion-resistant MSRs, if developed, might afford resistance to diversion of SNM comparable to that of solid-fueled reactors without fuel reprocessing. They could not be available for about 25 years, but on the other hand, when available, they would require substantially less mined uranium for deployment and operation.

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