

MOLTEN-SALT REACTORS—HISTORY, STATUS, AND POTENTIAL

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REACTORS

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Molten-salt breeder reactors (MSBR's) are being developed by the Oak Ridge National Laboratory for generating low-cost power while extending the nation's resources of fissionable fuel. The fluid fuel in these reactors, consisting of UF_4 and ThF_4 dissolved in fluorides of beryllium and lithium, is circulated through a reactor core moderated by graphite. Technology developments over the past 20 years have culminated in the successful operation of the 8-MW(th) Molten-Salt Reactor Experiment (MSRE), and have indicated that operation with a molten fuel is practical, that the salt is stable under reactor conditions, and that corrosion is very low. Processing of the MSRE fuel has demonstrated the MSR processing associated with high-performance converters. New fuel processing methods under development should permit MSR's to operate as economical breeders. These features, combined with high thermal efficiency (44%) and low primary system pressure, give MSR converters and breeders potentially favorable economic, fuel utilization, and safety characteristics. Further, these reactors can be initially fueled with ^{233}U , ^{235}U , or plutonium. The construction cost of an MSBR power plant is estimated to be about the same as that of light-water reactors. This could lend to power costs ~ 0.5 to 1.0 mill/kWh less than those for light-water reactors. Achievement of economic molten-salt breeder reactors requires the construction and operation of several reactors of increasing size and their associated processing plants.

THE HISTORY OF MOLTEN-SALT REACTORS

Investigation of molten-salt reactors started in the late 1940's as part of the United States' pro-

gram to develop a nuclear powered airplane. A liquid fuel appeared to offer several advantages, so experiments to establish the feasibility of molten-salt fuels were begun in 1947 on "the initiative of V. P. Calkins, Kermit Anderson, and E. S. Bettis. At the enthusiastic urging of Bettis and on the recommendation of W. R. Grimes, R. C. Briant adopted molten fluoride salts in 1950 as the main line effort of the Oak Ridge National Laboratory's Aircraft Nuclear Propulsion¹ program." The fluorides appeared particularly appropriate because they have high solubility for uranium, are among the most stable of chemical compounds, have very low vapor pressure even at red heat, have reasonably good heat transfer properties, are not damaged by radiation, do not react violently with air or water, and are inert to some common structural metals.

A small reactor, the Aircraft Reactor Experiment, was built at Oak Ridge to investigate the use of molten fluoride fuels for aircraft propulsion reactors and particularly to study the nuclear stability of the circulating fuel system. The ARE fuel salt was a mixture of NaF, ZrF_4 , and UF_4 , the moderator was BeO, and all the piping was Inconel. In 1954 the ARE was operated successfully for 9 days at steady-state outlet temperatures ranging up to $1580^\circ F$ and at powers up to 2.5 MW (th). No mechanical or chemical problems were encountered, and the reactor was found to be stable and self-regulating.²

That molten-salt reactors might be attractive for civilian power applications was recognized from the beginning of the ANP program, and in 1956 H. G. MacPherson formed a group to study the technical characteristics, nuclear performance, and economics of molten-salt converters and breeders.³ After considering a number of concepts over a period of several years, MacPherson and his associates concluded that graphite-moderated thermal reactors operating on a

thorium fuel cycle would be the best molten-salt systems for producing economic power. The thorium fuel cycle with recycle of ^{233}U was found to give better performance in a molten-salt thermal reactor than a uranium fuel cycle in which ^{238}U is the fertile material and plutonium is produced and recycled. Homogeneous reactors in which the entire core is liquid salt were rejected because the limited moderation by the salt constituents did not appear to make as good a thermal reactor as one moderated by graphite, and intermediate spectrum reactors did not appear to have high enough breeding ratios to compensate for their higher inventory of fuel. Studies of fast spectrum molten-salt reactors^{4,5} indicated that good breeding ratios could be obtained, but very high power densities would be required to avoid excessive fissile inventories. Adequate power densities appeared difficult to achieve without going to novel and untested heat removal methods.

Two types of graphite-moderated reactors were considered by MacPherson's group—single-fluid reactors in which thorium and uranium are contained in the same salt, and two-fluid reactors in which a fertile salt containing thorium is kept separate from the fissile salt which contains uranium. The two-fluid reactor had the advantage that it would operate as a breeder; however, the single-fluid reactor appeared simpler and seemed to offer low power costs, even though the breeding ratio would be below 1.0 using the technology of that time. The fluoride volatility process,⁶ which could remove uranium from fluoride salts, had already been demonstrated in the recovery of uranium from the ARE fuel and thus was available for partial processing of salts from either type of reactor.

The results of the ORNL studies were considered by a U.S. Atomic Energy Commission task force that made a comparative evaluation of fluid-fuel reactors early in 1959. One conclusion of the task force⁷ was that the molten-salt reactor, although limited in potential breeding gain, had "the highest probability of achieving technical feasibility."

By 1960, more complete conceptual designs of molten-salt reactors had emerged. Although emphasis was placed on the two-fluid concept because of its better nuclear performance,⁸ the single-fluid reactor was also studied.⁹ ORNL concluded that either route would lead to low-power-cost reactors, and that proceeding to the breeder either directly or via the converter would achieve reactors with good fuel conservation characteristics. Since many of the features of civilian power reactors would differ from those of the ARE, and the ARE had been operated only a short period, another reactor experiment was needed to

investigate some of the technology for power reactors.

The design of the Molten-Salt Reactor Experiment was begun in 1960. A single-fluid reactor was selected that in its engineering features resembled a converter, but the fuel salt did not contain thorium and thus was similar to the fuel salt for a two-fluid breeder. The MSRE fuel salt is a mixture of uranium, lithium-7, beryllium, and zirconium fluorides. Unclad graphite serves as the moderator (the salt does not wet graphite and will not penetrate into its pores if the pore sizes are small). All other parts of the system that contact salt are made from the nickel-base alloy, INOR-8 (also called Hastelloy-N), which was specially developed in the aircraft program for use with molten fluorides. The maximum power is ~8000 kW, and the heat is rejected to the atmosphere.

Construction of the MSRE began in 1962, and the reactor was first critical in 1965. Sustained operation at full power began in December 1966. Successful completion of a six-month run in March of 1968 brought to a close the first phase of operation during which the initial objectives were achieved. The molten fluoride fuel was used for many months at temperatures > 1200°F without corrosive attack on the metal and graphite parts of the system. The reactor equipment operated reliably and the radioactive liquids and gases were contained safely. The fuel was completely stable. Xenon was removed rapidly from the salt. When necessary, radioactive equipment was repaired or replaced in reasonable time and without overexposing maintenance personnel.

The second phase of MSRE operation began in August 1968 when a small processing facility attached to the reactor was used to remove the original uranium by treating the fuel salt with fluorine gas. A charge of ^{233}U fuel was added to the same carrier salt, and on October 2 the MSRE was made critical on ^{233}U . Six days later the power was taken to 100 kW by Glenn T. Seaborg, Chairman of the U. S. Atomic Energy Commission, bringing to power the first reactor to operate on ^{233}U .

During the years when the MSRE was being built and brought into operation, most of the development work on molten-salt reactors was in support of the MSRE. However, basic chemistry studies of molten fluoride salts continued throughout this period. One discovery¹⁰ during this time was that the lithium fluoride and beryllium fluoride in a fuel salt can be separated from rare earths by vacuum distillation at temperatures near 1000°C. This was a significant discovery, since it provided an inexpensive, on-site method for recovering these valuable materials. As a

consequence, the study effort looking at future reactors focused on a two-fluid breeder in which the fuel salt would be fluorinated to recover the uranium and distilled to separate the carrier salt from fission products. The blanket salt would be processed by fluorination alone, since few fission products would be generated in the blanket if the uranium concentration were kept low. Graphite tubes would be used in the core to keep the fuel and fertile streams from mixing.

Analyses of these two-fluid systems showed that breeding ratios in the range of 1.07 to 1.08 could be obtained, which, along with low fuel inventories, would lead to good fuel utilization. In addition, the fuel cycle cost appeared to be quite low. Consequently, the development effort for future reactors was aimed mainly at the features of two-fluid breeders. A review of the technology associated with such reactors was published in 1967.¹¹ The major disadvantage of this two-fluid system was recognized as being that the graphite had to serve as a piping material in the core where it was exposed to very high neutron fluxes.

In late 1967, new experimental information and an advance in core design caused the molten-salt program at ORNL to change from the two-fluid breeder to a single-fluid breeder. Part of the information influencing this change concerned the behavior of graphite at higher radiation exposures than had been achieved previously, and the other part related to a development in chemical processing.

The irradiation data^{12,13} showed that the kind of graphite planned for use in an MSBR changes dimensions more rapidly than had been anticipated. This made it necessary to lower the core power density for the graphite to have an acceptable service life, and to plan on replacement of the core at fairly frequent intervals.¹⁴ Moreover, complexities in the assembly of the core seemed to require that the entire core and reactor vessel be replaced whenever a graphite element reached its radiation limit or developed a leak. Under such circumstances, many years of operation of a prototype reactor would be required to prove convincingly that the two-fluid core is practicable.

At about the time the problems associated with long graphite exposure became evident, a chemical processing development occurred¹⁵ that greatly improved the prospect for a single-fluid breeder. To obtain good breeding performance in a single-fluid reactor, ²³³Pa (27.4-day half-life) must be held up outside the core until it decays to ²³³U. The processing development that showed promise of accomplishing this was a laboratory demonstration of the chemical steps in a liquid-liquid extraction process for removing protactinium and uranium from molten fluoride salts. The tech-

nique is to exchange thorium and lithium dissolved in molten bismuth for the constituents to be removed from the salt. The process has similarities to one being developed at Argonne National Laboratory for processing fast reactor fuels and involves technology explored at Brookhaven National Laboratory for removing fission products from a liquid bismuth fuel. Additional data have confirmed the early results and have shown that the uranium can be selectively stripped from the salt into bismuth, the protactinium can be trapped in salt in a decay tank, and the uranium can be transferred back to the salt by electrolysis for return to the reactor. Calculations indicated that the extraction and electrolysis could be carried out rapidly and continuously, and that the process equipment would be relatively small.

Laboratory experiments also indicated that rare earths might be extracted from salt from which the uranium had previously been removed. Unfortunately, the chemical potentials that determine which constituents transfer to the bismuth are relatively close for thorium and the higher cross-section rare earths, and the separation is more difficult than it is for uranium and protactinium. The extraction process may still be workable, however, because with no processing the rare earths have a much smaller effect on breeding ratio than does protactinium and so need be removed only relatively slowly (50- to 80-day cycle time). Several other processes for rare earth removal are under investigation, and whether liquid metal extraction will be the most attractive is still uncertain.

The advance in core design that was important in the switch to the single-fluid breeder was the recognition that a fertile "blanket" can be achieved with a salt that contains uranium as well as thorium. The blanket is obtained by increasing the volume fraction of salt and reducing the volume fraction of graphite in the outer part of the reactor. This makes the outer region undermoderated and increases the capture of neutrons there by the thorium. With this arrangement, most of the neutrons are generated at some distance from the reactor boundary, and captures in the blanket reduce the neutron leakage to an acceptable level. H. G. MacPherson had proposed this scheme several years before, but it was only studied thoroughly after the discovery that protactinium could be removed from the salt. Optimization calculations then showed that proper selection of dimensions and volume fractions could keep the inventory of uranium in the outer region from being excessive, while producing a blanket region.

As a result of the developments cited above, design studies of single-fluid breeders were pur-