

The Molten Salt Reactor Adventure

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Abstract - A personal history of the development of molten salt reactors in the United States is presented. The initial goal was an aircraft propulsion reactor, and a molten fluoride-fueled Aircraft Reactor Experiment was operated at Oak Ridge National Laboratory in 1954. In 1956, the objective shifted to civilian nuclear power, and reactor concepts were developed using a circulating UF₄- ThF₄ fuel, graphite moderator, and Hastelloy N pressure boundary. The program culminated in the successful operation of the Molten Salt Reactor Experiment in 1965 to 1969. By then the Atomic Energy Commission's goals had shifted to breeder development; the molten salt program supported on-site reprocessing development and study of various reactor arrangements that had potential to breed. Some commercial and foreign interest contributed to the program which, however, was terminated by the government in 1976. The current status of the technology and prospects for revived interest are summarized.

L'aventure "réacteur à sels fondus"

Resumé - Le mémoire traite de l'histoire du développement de réacteurs sels fondus aux Etats Unis du point de vue de l'expérience personnelle de l'auteur. Le but initial fut un reaaeur de propulsions d'avion, et une expe'rience denommee Aircraft Reactor Experiment fut entreprise dans l'Oak Ridge National Laboratory en 1954 avec le fluorure fondu corn me combustible. En 1956 le but original fut abandonné en faveur de la puissance nucléaire civile, et des conceptions de reacteur furent mises au point basees sur le combustible UF₄-ThF circulant, le graphite comme modérateur et Hastelloy N cornme enceinte sous pression. Le programme atteignit son point culminant avec le succes caracterisant l'experience Molten Salt Reactor Experiment poursuivie de 1965 jusqu'à 1969. Les buts visés par l'Atomic Energy Commission s'e'taient ensuite orientés vers le développement du surgenerateur rapide et le programme de reacteurs a sels fondus fut con tin ue' pour appuyer le retraitement sur le terrain de la station nucléaire ainsi que l'e'tude de diffe'rentes configurations en pile offrant un potentiel de surré'ne'ration. Il y avait un certain nombre de partenaires industriels et étrangers contribuant au programme qui, cependant, fut arre'te' par le gouvernement en 1976. Le me'moire donne un aper de l'état actuel de la technologie ainsi que des perspectives pour un renouvellement de l'int'e'ret.

Das Abenteuer "Salzschmelzenreaktor"

Zusammenfassung - Die Geschichte der Entwicklung des Salzschnmelzenreaktors in den Vereinigten Staaten wird aus der persönlichen Sicht des Verfassers dargestellt. Ursprüngliches Ziel war ein Antriebsreaktor für Flugzeuge, und im Jahre 1954 wurde im Oak Ridge National Laboratory ein Aircraft Reactor Experiment mit einer Fluoridschnmelze als Brennstoff durchgeführt. Im Jahre 1956 änderte sich die Zielrichtung zugunsten des zivilen Sektors der Kernenergie, und es wurden Konzepte für Reaktoren mit umlaufendem UF₄-Th₄F-Brennstoff, Graphitmoderator und Hastelloy-N-Druck einschluß entwickelt. Das Programm gipfelte in der erfolgreichen Durchführung des Molten Salt Reactor Experiment in den Jahren 1965 bis 1969. Zu diesem Zeitpunkt hatte sich die Atomic Energy Commission für die Brüterentwicklung entschieden, und das Kernschnmelzenprogramm wurde ein Unterstützungsprogramm bei der Entwicklung der Wiederaufarbeitung auf Kraftwerksgeländen und bei der Untersuchung verschiedener Reaktorarrordnungen, die für den Brutvorgang in Frage kamen. Das Programm fand die Unterstützung einiger interessierter Industriefirmen und ausländischer Partner, wurde aber 1976 von der Regierung eingestellt. Der derzeitige Stand der Technik und Aussichten auf ein Wiederaufleben des Interesses werden im vorliegenden Bericht zusammenfassend dargestellt.

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This story of the development of the molten salt reactor (MSR) is told the way I remember it. For me, it started in 1956 after I had decided that I would like to become more directly involved in the development of nuclear power. To do this I needed a new job, and to get such a job I needed a few references. Thus I wrote to Alvin Weinberg to see whether he would put in a good word for me.

My acquaintance with Alvin started in 1946-1947 when I was a student at Oak Ridge's "Clinch College of Nuclear Knowledge," as the training school under Eugene Wigner and Fred Seitz came to be called. Alvin, who was then assistant director of the Physics Division, was one of my teachers and I was tremendously impressed with him. I went back to a Union Carbide research laboratory in Cleveland, Ohio, at the end of my year at Oak Ridge, and in late 1947, when it was decided that Union Carbide was to take over the operation of Oak Ridge National Laboratory (ORNL), I was called to New York to brief a few vice-presidents as to who the good guys and the bad guys were. All I could think to tell them was to get on the good side of Alvin Weinberg. I told them that he was the natural leader of the place and that their success would depend on him. I have never told Alvin of this incident, and I am sure my advice was not necessary for the Union Carbide officials to recognize Alvin's worth.

When I wrote Alvin for a reference, he responded by asking me to come to Oak Ridge to head up a program to investigate power reactors fueled with molten salts. Although I did

not know much about molten salts at the time, the job sounded exciting and I gladly accepted.

Molten salt reactors were first proposed by Ed Bettis and Ray Briant of ORNL during the post-World War II attempt to design a nuclear-powered aircraft. The attraction of molten fluoride salts for that program was the great stability of the salts, both to high temperatures and to radiation. An active development program aimed at such an aircraft reactor was carried out from about 1950 to 1956. The Aircraft Reactor Experiment, a small reactor using a circulating molten fuel salt, operated for several days in 1943 [sic 1953] and reached a peak temperature of 1620° F. In 1956 interest in the airplane began to fall off, and Alvin Weinberg wished to see whether the molten fluoride fuel technology that had been developed for the aircraft could be adapted to civilian power reactors. Part of his interest stemmed from the fact that all of the other materials and coolants being suggested for reactors had been anticipated by the reactor design group at the Metallurgical Laboratory in Chicago during World War II. This was new.

In starting the new civilian program, we had a tremendous head start from the capable staff and the research under way for the aircraft program. Our civilian program was in reality a continuation of the old military program with a few changes in emphasis.

It is my impression that the Division of Reactor Development (DRD) of the U.S. Atomic Energy Commission (AEC) never showed much enthusiasm for the Molten Salt Reactor Program. At the start, in 1956, Alvin had to use all his powers of persuasion with Kenneth Davis, who was then director of the DRD, to provide us with an initial \$2 million per year to start the program. A couple of years later the DRD realized that they were supporting too many fledgling reactor concepts. While each concept could be researched at the exploratory level with only modest funding, to carry the development of any concept to fruition would require tremendously larger expenditures. With funds limited, only a few reactor concepts could survive.

One of the methods employed by the AEC to eliminate reactor concepts was to establish task forces of outside experts to evaluate the reactor concepts and, especially, to point out their weaknesses. After a couple of other reactor concepts had been eliminated by this process, the AEC formed the Fluid Fuels Reactor Task Force to evaluate and compare three different fluid fuel reactors: the aqueous homogeneous, the liquid bismuth, and the MSR. The task force met in Washington for about two months early in 1959. I was there to represent the molten salt system, Beecher Briggs of ORNL represented the aqueous homogeneous, and Frank Miles from Brookhaven National Laboratory represented the bismuth-graphite reactor. Task force members came from other AEC laboratories, from electric utilities, from architect engineering firms, and from the AEC itself. The first sentence of the Summary of the Task Force Report (TID-8505) was, "The Molten Salt Reactor has the highest probability of achieving technical feasibility."¹

This conclusion arose from the fact that the molten fluoride salts (a) have a wide range of solubility of uranium and thorium, (b) are stable thermodynamically and do not undergo radiolytic decomposition, (c) have a very low vapor pressure at operating temperatures, and (d) do not attack the nickel-based alloy used in the circulating salt system.

As a result of the task force deliberations, the other two concepts were abandoned and the molten salt system continued its precarious existence. The reactor considered by the Task Force was a converter reactor, not a breeder, and was described as follows:

The reference design molten salt reactor is an INOR-8 (now called Hastelloy N) vessel containing a graphite assembly 12.25 feet in diameter by 12.25 feet high, through which molten salt flows in vertical channels. The fuel salt is a solution composed of 0.3 mole percent UF_4 13 mole per cent ThF_4 , 16 mole percent BeF_2 , and 70.7 mole percent 7LiF . The fuel salt is heated from 1075°F to 1225°F in the core and is circulated from the reactor vessel to four primary heat exchangers by four fuel pumps. A barren coolant salt is used as the intermediate heat exchange fluid.¹

It is interesting to me that this reactor description prepared hurriedly in early 1959 differs only in minor detail from the MSR designs of the 1970s.

Until late 1959 our exploration of MSRs was not focused sharply on the breeding possibilities of the system, although we always preferred high conversion ratio designs in our studies. Starting in 1960, however, the financial support of the Molten Salt Reactor Program was dependent on its breeding possibilities, and thereafter the program was focused on the molten salt breeder reactor (MSBR). Thermal breeders have only a small breeding margin, and, to breed comfortably, fission products must be kept at low concentrations. This means that the fuel must be purified frequently. The fact that the molten salt fuel is a liquid helps in this respect, since the gaseous fission products come off continuously and the fuel can be purified from other fission products by suitable liquid extraction methods. In principle, this purification is much easier for MSRs than it is for solid fuel reactors, since the steps of dissolution and refabrication of the fuel are avoided.

Over the course of years, suitable processing methods were found so that the single fluid reactor described above could become a breeder. In 1960, however, our limited knowledge of processing chemistry forced us to consider breeder reactors in which the fuel (fissile uranium) was kept in one fluid and the fertile material (thorium) was in a separate fluid. Both fluids circulated through the reactor, but were kept separate by walls of graphite.

By the end of 1959, our engineering development program had proceeded far enough that we felt justified in proposing an MSR experiment (MSRE), but getting money and permission appeared difficult. Then one day I heard a rumor that Frank Pittman, who had succeeded Ken Davis as director of the DRD, had expressed interest in funding as many as four "quick and dirty" reactor experiments provided that each one should cost less than a million dollars. As I remember it, I wrote a proposal that night and submitted it through channels the next day. I outlined the general features of the reactor, and by analogy with another reactor system for which a cost estimate had been made. I came up with a cost

estimate of \$4.18 million. The proposal was accepted, although by the time the design had been detailed the cost estimate had doubled.

The conceptual design of the MSRE was arrived at as follows. To keep the reactor simple we intended to simulate only the fuel stream of a two-fluid breeder reactor, so that no thorium fluoride was included. We wanted the neutron spectrum to be near thermal, as it would be in a commercial reactor, and since graphite was the moderator, this dictated the minimum physical size. The moderator was in the form of a 1.37-m-diam x 1.62-m-high right circular cylinder. Had it been smaller, the neutron leakage would have caused the neutron spectrum to be more energetic than we wished. We would have liked to have a higher power density, but cost considerations limited us to ~10 MW of heat. There was also another reason for limiting the power of the reactor. The AEC accounting rules at the time allowed us to build a 10-MW reactor as an experiment, using operating funds. A higher power reactor would have required us to obtain a capital appropriation and would have limited our freedom to make changes. Actually we miscalculated the heat transfer characteristics and the reactor operated at only 8 MW.

I ceased being director of the MSR program in 1960, at which time capable engineers took over for the construction of the MSRE and the later development work. However, I maintained my strong interest in the program during my tenure as deputy director of ORNL and even after leaving in 1970.

Design of the MSRE started in the summer of 1960 and construction started 18 months later, at the beginning of 1962. The reactor went critical in June 1965, and was briefly at full power a year later. After a shakedown period, reliable operation was achieved in December 1966, when a 30-day continuous run was made at full power. While carrying out numerous experiments, the reactor was operated at full power most of the time during the next 15 months, after which the ^{235}U was removed from the fuel salt and later replaced with ^{233}U . The reactor was operated with ^{233}U as the fuel from January through May 1969. This was the first time ^{233}U had been used as a reactor fuel, and AEC Chairman Glen Seaborg and Ray Stoughton, codiscoverers of ^{233}U , were present when the reactor first went critical with ^{233}U fuel.

During the remainder of 1969, the reactor was devoted to a number of experiments, including xenon stripping, fission product deposition, tritium behavior, and plutonium additions. Operation was finally terminated in December 1969 so that the available funds could be applied to other development areas.

In 1962 the AEC first took a position strongly favoring the development of breeder reactors. This position was spelled out in its 1962 "Report to the President." In that report two breeder reactors were described and discussed. One was the familiar liquid-metal-cooled fast breeder reactor (LMFBR) using the ^{238}U -Pu breeding cycle, and the other was the molten salt fueled thermal breeder using the ^{233}U -Th cycle. Somewhat surprisingly, the report devoted about 60% as much favorable space to the molten salt thermal breeder as to the fast breeder, despite the much broader countrywide participation in the fast breeder program.

The AEC's 1962 "Report to the President" was written while the MSRE was under construction, but apparently we had proceeded far enough with our development program to impress the Subcommittee on Reactors of the General Advisory Committee with the value of the molten salt system. As soon as the "Report to the President" presented its justification for the development of breeder reactors, however, the existent Liquid Metal Cooled Reactors Department of the DRD proposed an elaborate and wide-ranging program for the development of the LMFBR concept, and that program began to gain momentum. On the other hand, those of us involved in the MSR program chose to wait until the MSRE had operated successfully before trying to expand our program. This, we thought, was the prudent thing to do, but by the time we were prepared to go for a larger reactor, the momentum and money needs of the LMFBR program were massive and there was no interest in funding a competitor.

The MSRE was a very successful experiment, in that it answered many questions and posed but a few new ones. Perhaps the most important result was the conclusion that it was quite a practical reactor. It ran for long periods of time, and when maintenance was required, it was accomplished safely and without excessive delay. Also, it demonstrated the expected flexibility and ease of handling the fuel. As mentioned above, it was the first reactor in the world to operate with ^{233}U as the sole fuel, and the highly radioactive ^{233}U used would have been extremely difficult to handle if it had had to be incorporated into solid fuel elements. In preparation for the run with ^{233}U , the ^{235}U was removed from the carrier salt in 4 days by the fluoride volatility process. This process decontaminated the 218 kg of uranium of gamma radiation by a factor of 4×10^9 so that it could be handled without shielding. As an aside, this equipment used for the MSRE was sufficiently large so that it could satisfactorily handle all of the fuel processing needs for a 1000-MW molten salt converter reactor (MSCR), about which I will say more later.

Three problems requiring further development turned up during the construction and operation of the MSRE. The first was that the Hastelloy N used for the MSRE was subject to a kind of "radiation hardening," due to accumulation of helium at grain boundaries. Later, it was found that modified alloys that had fine carbide precipitates within the grains would hold the helium and restrain this migration to the grain boundaries. Nevertheless, it is still desirable to design well-blanketed reactors in which the exposure of the reactor vessel wall to fast neutron radiation is limited.

The second problem concerned the tritium produced by neutron reactions with lithium. At high temperatures the radioactive tritium, which is, of course, chemically like hydrogen, penetrates metals quite readily, and unless captured in some way, would appear in the steam generators and reach the atmosphere. After considerable development work, it was found that the intermediate salt coolant, a mixture of sodium fluoride and sodium fluoroborate, would capture the tritium and that it could be removed and isolated in the gas purge system.

The third problem came from the discovery of tiny cracks on the inside surface of the Hastelloy N piping for the MSRE. It was found that these cracks were caused by the fission product tellurium. Later work showed that this tellurium attack could be

controlled by keeping the fuel on the reducing side. This is done by adjustment of the chemistry so that about 2% of the uranium is in the form of UF_3 , as opposed to UF_4 . This can be controlled rather easily now that good analytical methods have been developed. If the UF_3 to UF_4 ratio drops too low, it can be raised by the addition of some beryllium metal, which, as it dissolves, will rob some of the fluoride ions from the uranium.

As solutions to these new problems became available, those of us familiar with the technology believed that we were ready for the next step, the construction of a reactor producing modest amounts of electrical power, or at least the development of the equipment needed for such a reactor. In any case, productive continuation of the MSR program in the United States would soon require funds an order of magnitude greater than previous expenditures.

In 1972 ORNL proposed a major development program that would culminate in the construction and operation of a demonstration reactor called the Molten Salt Breeder Experiment. The program was estimated to cost a total of \$350 million over a period of 11 yr. However, those who would have had to approve such a program were already heavily committed to the LMFBR and guiding a very expensive development program that would be spending about \$400 million each year by 1975. It was asking too much of human nature to expect them to believe that a much less expensive program could be effective in developing a competing system, and the ORNL proposal was rejected. In January 1973, ORNL was directed to terminate MSR development work. For reasons I do not understand, the program was reinstated a year later, and in 1974 ORNL submitted a more elaborate proposal with suitably inflated costs calling for about \$720 million to be spent over an 11-yr period. This last proposal was also rejected, and in 1976 ORNL was again ordered to shut down the MSR program "for budgetary reasons.

The decision to cut off the funding for the MSR program was supported by an "evaluation" of the MSBR (1972) prepared internally by the Division of Reactor Development and Technology in response to a request from the Office of Science and Technology. Although this report contained no overt recommendations, the Conclusions section, after granting some attractive features of the MSR, emphasized the difficulty of solving a number of problems, including those described above. It was stated that after realistic solutions for these problems had been demonstrated, proceeding toward engineering development would require "reasonable assurances that large scale government and industrial resources can be made available on a continuing basis in light of other commitments to the commercial nuclear power program and higher priority energy developments." This was, of course, true.

This evaluation was prepared before the solutions to the tritium evolution and tellurium cracking problems were known. On a recent trip to Japan, I was asked the following question by a group of engineers who were thoroughly familiar with MSR technology: "Concerning the MSBR development program, the U.S. AEC pointed out the following major technological problems in 1972, namely the problem on the materials, the tritium and fuel salt chemical processing. And we think that most of the above problems have been settled by the splendid effort of R&D; performed by ORNL thereafter, but how was

it evaluated in the U.S.?" I do not believe that such an evaluation was made or that the Department of Energy is prepared to make one.

For the several groups of enthusiasts for molten salt technology in other countries, the reasons for the demise of the U.S. program are important. It is difficult for them to overcome the stigma of abandonment of the MSR by the country of its origin. In my opinion, these are the major factors contributing to the cessation of the program.

1. The political and technical support for the program in the United States was too thin geographically. Within the United States, only in Oak Ridge, Tennessee, was the technology really understood and appreciated.
2. The MSR program was in competition with the fast breeder program, which got an early start and had copious government development funds being spent in many parts of the United States. When the MSR development program had progressed far enough to justify a greatly expanded program leading to commercial development, the AEC could not justify the diversion of substantial funds from the LMFBR to a competing program.

It was often suggested by the DRD that evidence of industrial support for the MSR was needed to gain AEC enthusiasm. However, the evidence available for this support was ignored.

At least two privately funded technology evaluation and design studies of the MSR were made. One was carried out in 1970 by a group called the Molten Salt Breeder Reactor Associates, headed by Black and Veach and including five Midwest utilities. The other study was done by The Molten Salt Group, which was headed by Ebasco Services, Inc., and included 5 large industrial firms and 15 utilities. Both of these studies reported favorably on the promise of the system. The Molten Salt Group concluded in 1971 that the existing technology was sufficient to justify construction of a molten salt demonstration plant.

About 10 yr have elapsed since the demise of the active MSR development program at ORNL. In that interval several changes have occurred that might have some influence on how the MSR is perceived.

Foremost has been the decreased rate of expansion of electricity use in developed countries. This has sharply reduced the expected rate of building new nuclear power plants by as much as an order of magnitude. As a result, we no longer expect a crisis in the availability of uranium for at least 50 yr. Even with some renewed growth, it is unlikely that breeders will be needed before 2035 to 2050. With this delay in the need for breeders, the primary current interest should be for reactors that are economical and that have other features of merit that might encourage a revival of new reactor construction. Thus, there is no need for the MSR to emerge as a full-blown breeder, and it should now be considered on its merits as a relatively simple, low cost, converter reactor, the MSCR. The fact that the MSCR can be developed into a breeder at the suitable time by the addition of chemical processing equipment should be regarded as a positive feature. Full

engineering development of the known methods for accomplishing this, however, can be deferred for decades.

Another major change is the very high capital costs that are reported in building fast breeder reactors. The result is that the LMFBR will not be competitive with light water reactors (LWRs) until natural uranium reaches astronomically high costs; in fact, until it attains levels it may never reach in the next century. Thus, fast reactors as currently designed will not be economically competitive for perhaps two generations (of people). By the time they are competitive, a whole new crop of nuclear engineers will have to learn the technology.

Off hand one might think that the high cost of LMFBRs would open the door to other advanced reactor systems, such as the MSCR. The practical effect is negative, however, since the new information casts doubt on the projected costs of all advanced reactor systems, and this presumption is difficult to overcome without actual experience with hardware. In 1970 a careful detailed estimate was made of the capital cost of an MSR and it turned out to be within 1% of the cost of an LWR. Construction experience is needed, however, to verify such a cost estimate.

Like the LMFBR, the MSR has an intermediate heat transfer loop to isolate the steam generators from radioactive fluids. However, there are basic reasons for expecting MSCR capital costs to be lower than those for the LMFBR:

1. The fuel handling system will be much simpler.
2. The molten salts have a much higher heat capacity per unit volume than sodium, so that the physical size of pumps and piping will be smaller.
3. There is no threat of a "core disruptive accident" with the MSCR, so that safety-related equipment can be simpler.
4. The molten salts have a much lower thermal conductivity than sodium, so that sudden coolant temperature changes will provide less thermal shock to system components.
5. The coolant is more compatible with water than is sodium, so that there should be fewer problems in the design and maintenance of steam generators.

On the other hand, it should be remembered that the development of large engineering components for the MSCR is in a primitive state and that any future development program would surely reveal new difficulties of uncertain magnitude. Furthermore, the high degree of radioactivity of the MSCR's primary system will present problems of design for remote maintenance. The continuing development of robots ought to be of some help here.

Another change is the effective moratorium on fuel processing for LWRs, resulting from fears of plutonium weapons proliferation and from higher projected costs of plutonium recycling. As a result, large stockpiles of plutonium will be accumulating, stored mostly in spent fuel elements. The safe use of this plutonium is one of the reasons for the current interest in MSR in Japan. Preliminary calculations indicate the possibility of obtaining additional energy out of spent LWR fuel by dissolving it in a fluoride salt and burning it in an MSCR.

The fear of weapons proliferation has also spurred an interest in reactors that do not use highly enriched fuels and that avoid fuel reprocessing. A recent quite sophisticated study at ORNL has shown how an MSCR using thorium as the fertile material but fueled with 20% enriched uranium could be operated for 30 yr without any processing of the fuel. There would be periodic additions of the 20% enriched uranium, but no need to remove fission products other than those coming off naturally in the gas purge system. Even under these extreme conditions, the lifetime average conversion ratio is ~0.80. I was personally surprised at the favorable result of that study, which illustrates another way that the MSR technology could be used if fully developed.

A social change within the last few years is the increased sensitivity of the public to the possibility of reactor accidents. The MSRs deserve consideration on this score, especially because catastrophic accidents appear to be extremely unlikely. Although the entire primary system of an MSR is highly radioactive, there is little driving force to make the activity escape. Thus, with proper design of the containment, the public should be well protected.

For the foreseeable future, any newly proposed reactor must compete with the LWR. Because the MSCR's fuel cycle cost will be lower, it is possible that it can compete with the LWR on a cost basis. I believe the two most important considerations favoring the MSR, however, are its versatility in being able to use any fuel and its ability to be ultimately transformed into a breeder, thus solving the electric power problem for millennia.

What of the future? The details of the technology developed for the MSR at Oak Ridge are exceedingly well documented, and the knowledge, as preserved on paper, is available to any group wishing to pursue a new development effort.²⁻⁸ There continues to be a significant amount of international interest in molten salts.⁹⁻¹⁵ Sophisticated research on the chemistry and technology is going on at several Japanese universities and there is some industrial interest there. A new engineering development project has been started in Italy for which I have great hopes. Certain individuals in France, India, the USSR, and other countries are strongly interested. It is possible that one of these centers of interest will persevere and ultimately achieve commercial success. Mr. Weinberg and I would be overjoyed.

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