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SAFE ACTINIDE DISPOSITION IN MOLTEN SALT REACTORS[a]

Uri Gat
Oak Ridge National Laboratory
P. O. Box 2009
Oak Ridge, Tennessee 37831-8087
(423) 574-0560

ABSTRACT

Safe molten salt reactors (MSR) can readily accommodate the burning of all fissile actinides. Only minor compromises associated with plutonium are required. The MSRs can dispose safely of actinides and long lived isotopes to result in safer and simpler waste. Disposing of actinides in MSRs does increase the source term of a safety optimized MSR. It is concluded that the burning and transmutation of actinides in MSRs can be done in a safe manner. Development is needed for the processing to handle and separate the actinides. Calculations are needed to establish the neutron economy and the fuel management.

I. MOLTEN SALT REACTORS SAFETY

Molten Salt Reactors (MSR) are endowed with many safety attributes and great flexibility.[1,2] When MSRs are optimized for safety they can be designed such that no credible severe accident is identified for them. The extremely safe MSR concept was dubbed the U.S.R.[3] Safe MSRs are predicated on frequent on-line processing to continuously remove fission products and retain criticality of the reactor by temperature control alone, and by utilizing "external cooling" that is circulating the fuel itself from the core to a primary heat exchanger external to the core. Both of these features are unique to fluid fuel reactors (FFR), which includes the MSR. Frequent processing of the fuel essentially eliminates the severe accidents of excess reactivity or criticality excursion, and the overheating, or melt-down, that may be associated with a loss of coolant accident (LOCA). The external cooling and the temperature control of the reactor result in a safety-optimized core that is not subject to compromises that may require special safety precautions that are subject to failure.

The molten salts proposed for use in the MSRs are chemically inert and operate at high temperature at low pressure. The salts are compatible with available selected structural materials. All of these properties enhance the safety of MSRs and protect them from accidents.

Molten Salt Reactors are extremely flexible. The same design can handle a variety of fissile materials and additives in the fuel. The flexibility and safety of MSRs was well demonstrated in the operation of the Molten Salt Reactor Experiment (MSRE).[4] The MSRE operated initially on a U-235. The U-235 was later removed and replaced with U-233 with no change in design. Six hundred grams of plutonium were added to the MSRE, during its operation, in addition to the plutonium that was bred from the uranium in its initial operation with U-235. Thus, the MSRE operated on the three major fissile materials with no safety incidents.

The disposition of actinides in an MSR may, at times, require some compromise from a safety optimized MSR, as will be discussed later.

The safety of an MSR is, in most cases, passive, inherent and non-tamperable (PINT). This renders the safety

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optimized MSR mostly immune against severe incidents with respect to sabotage and terrorism.

It is assumed here that the MSRs discussed also generate useful power.

II. NON PROLIFERATION

Non proliferation is not a safety issue per se, nevertheless it is closely related to safety and thus will be addressed to some extent here. Non proliferation attributes of MSRs are detailed in previous publications.[5,6] As a rule, the fuel handling in MSRs protects the fissile material against proliferation and diversion. Fissile material, when required to be transported, can be shipped in arbitrary small quantities that can be well protected, and the diversion of one shipment alone is not sufficient to constitute a threat. Further, the fissile material can be readily diluted, denatured or spiked at the outset, usually by simple chemical means. The fact that the material transfers in the reactor are done in fluid form at elevated temperatures requires sophisticated and elaborate handling facilities that are difficult to conceal or hide. While the fuel is in the reactor complex, it is mixed with highly radioactive materials that necessitate remote handling. Unless the MSR's processing is designed for breeding, there are no facilities to remove fissile material from the reactor. State-of-the-art technology would require major modifications to facilitate separation of "clean" fissile material. Removal of uranium by the uranium volatility process can be readily counteracted by either blending the uranium to low enrichment or by allowing some U-232 to be bred into the fuel. The U-232 acts as a strong diversion and proliferation deterrent because of the strong gamma emission of its daughter products. The penetrating 2.6 MeV gamma of Tl-208 allows for ready detection and exposes any handler to excessive radiation.

III. WEAPONS FISSILE MATERIAL

The MSRs are particularly suitable for disposing of weapons fissile materials. The weapons fissile material can be completely burned in an MSR by "converting" it to U-233. This is done by adding thorium to the fuel which successively breeds U-233 while the weapons fissile materials is consumed. The critical mass is converted, from the weapons fissile, to the U-233. As desired the conversion can be done toward the "end of life"(when the reactor is destined to cease operation). When the fissile material is HEU all the safety attributes of the MSR can be retained as the HEU is not different, from the MSR aspects, from a design uranium fuel. When the weapons fissile is plutonium, a few developments and compromises are required. The developed fuel process does not include plutonium in the processing scheme. The processing, that included the protection of the fuel, can no longer rely on U-232 for protection. There is need to assure that in no place in the process plutonium is available in a pure form. This will require the retention of more of the radioactive fission products in the reactor, which means that the source term and the after heat can no longer be optimized for safety alone. Detailed calculations for the best compromise have yet to be performed.

An additional concern associated with the addition of plutonium to the salt is the lack of information regarding the thermodynamic phase diagrams, sometimes (incorrectly) referred to as the missibility of plutonium. There is a limit of the amount of plutonium that can be added, lest there be some precipitation which can result in a serious safety incident. Another concern, due to lack of data, is that excessive addition of plutonium to the melt may at some point reduce the high negative temperature coefficient of the MSR, which is the basis for some aspects of its PINT safety. When these concerns are fully evaluated, the amount of plutonium that can be disposed of in an MSR may be limited to either combining the plutonium with HEU, U-233, or less than fully enriched uranium.

An alternative MSR has been proposed for burning plutonium. This alternative foregoes the fuel processing except for separation of gases and volatiles, which do not include fissile materials; all other materials are retained with the fuel.[7,8] This version of the MSR concept retains much of the source term, though the volatile and readily dispersable part, the obnoxious part, is removed. It is also more difficult to completely burn the plutonium in this version as the complete consumption of the plutonium is predicated on building in U-233 in lieu of the plutonium and not breeding new plutonium from U-238. This mode of MSR operation retains some plutonium with the final critical mass.

IV. SPENT FUEL

Spent fuel from nuclear reactors can be burned in MSRs. The operation is expected to be simple and safe. In general, the spent fuel can be fed into the MSR with no special preparation. The spent fuel can simply be dissolved, in small quantities, in the MSR fuel. The direct feed of spent fuel puts an extra demand and burden on the fuel processing of the MSR. Since spent fuel also contains plutonium and U-238, which is converted into plutonium, there are restrictions on the quantities and rate at which spent fuel can be mixed into the MSR. The advantages of disposing of spent fuel in an MSR are that the fuel is not discarded with large amounts of usable fissile material, particularly plutonium, that can be chemically separated. There is no need for reprocessing nor for fuel refabrication.

V. PLUTONIUM AND SPENT FUEL RESTRICTIONS

The disposal and burning of plutonium and spent fuel in MSRs are restricted by two major factors. The first of these restrictions is the anticipated limited amount of plutonium that can be added to the salt melt. The molten salt must retain its fluidity in a practical temperature range. At the upper temperature, the limits are set by the integrity of the structural materials and the possibility of reaching temperatures at which some components of the salt may evaporate, requiring increased pressure to avoid boiling, or the salts may disintegrate. At the lower temperature, the restrictions are given by the melting point, which cannot be too close to the upper limit, and the possibility of selective precipitation, or freezing out, of a phase of the mixture. To some extent the thermodynamic properties of the mixture could also restrict the fraction of plutonium that can be tolerated. Most of these restrictions are poorly known and require extensive research and possibly some development to completely understand and utilize.

The second constraint is associated with the nuclear properties of the molten salt with larger proportions of plutonium. There is some concern that the large negative temperature coefficient may decline, or even disappear, with added proportions of plutonium. There is clear need to completely explore the nuclear properties of melts with increasing proportions of plutonium.

A secondary concern is the decreased safety associated with the increased amounts of plutonium in the fuel. An unknown, that requires development, is the impact on fuel processing. When uranium is the sole fissile component, it can be separated and returned to the core, quantitatively by using the fluoride volatility process. The uranium is still protected by the ever present U-232. The carrier salts, with all fission products, except for the volatiles, are then available for treatment and optimization as waste.[3,9] There is no developed processing technology when plutonium is a significant component, and the safety consequences that may result from the processing are not known.

The discussion throughout this paper implicitly emphasizes the fluoride salts. The fluorides are the salts which have been well studied, tested and demonstrated for the MSR. There are possibilities of using other salts, chlorides for example. However, the information for nuclear applications and the associated implications are not known. A grand research and development program, with no certain outcome, is needed to study alternatives to the fluorides.

The MSRs also offer the potential for burning actinides, other than plutonium and uranium, and some long-lived isotopes, most notably I-129. The advantage of burning these isotopes is that they will be eliminated from the waste. Nuclear waste that is mostly free of long-lived isotopes (greater than thousands of years half-life) and free of fissile material can be disposed of by relatively simple and low-cost means with essentially no long-term safety concerns. It is expected that the actinides from the MSR operation can be readily transmuted in that MSR simply by leaving them in the fuel. However, there may be restrictions in dealing with larger quantities of actinides associated with, for example, disposal of spent fuel. To determine the ability and magnitude to handle actinides, it is necessary to do development in two areas: neutron economy, discussed below, and fuel processing. The fuel processing requires modification and development to retain, or return, those isotopes destined for transmutation to the reactor. The fuel processing, being a chemical process is, of course, limited to selecting elements, disregarding their isotopic composition. The inclusion of entire elements, rather than selected isotopes, may constitute a hardship on the neutron economy of the reactor.

The extreme safety of an MSR, associated with a low source term, must be compromised. The long-lived isotopes must be retained in the fuel for transmutation, but that also retains them as part of the source term.

VI. DEVELOPMENT

It was already stated that the MSR chemistry, chemical thermodynamics and thermal properties require understanding and development to accommodate expected large quantities of non-uranium fissile materials. The development of fuel processing to deal with new, hitherto not considered, components is also necessary.

Areas that must be explored and calculated are the time lines of fissile burning and the synergistics of MSRs and other reactors. A special initial issue is the already accumulated weapons fissile material and spent fuel that are awaiting disposition. The rate of burning fissiles is readily known.[5] However when actinide transmutation is included, the neutron economy of the entire system requires careful detailing and planning of rates and concentrations. When spent fuel is considered, the time to completely dispose of the plutonium may become long as it may not happen prior to conversion of the residual U-238 in the fuel. This planning is the analogue of fuel management in other reactors. The two most noticeable differences are the elimination of fuel reprocessing and (re)manufacturing and the planned absence of fissile material in the waste, which is therefore no longer spent fuel.

Once the back-log of fissile material and spent fuel are consumed, it can be anticipated that the synergism between

MSRs and other reactors will readily take care of any spent fuel. The waste will then exclude actinides and long-lived isotopes.

VII. SUMMARY

The safe MSRs that utilize fuel processing and external cooling can readily dispose of weapons-grade fissile material in a safe manner. Some compromise of the normally extreme safety of the MSRs may be required due to needed modification of the processing for plutonium and the plutonium now included in the fuel. The MSRs can also dispose of spent fuel, and they can transmute the non-fissile actinides and long-lived isotopes to render the waste safer and more readily handleable. It may require long periods of time to dispose of the back log of fissiles and fuels, in particular the complete burn of plutonium may have to wait for the conversion of any residual U-238. The final product, planned to be U-233 spiked with U-232, is considered proliferation resistant.

Development is needed and calculations are required to confirm these concepts. The fuel processing needs to be modified to handle plutonium in a safe and safeguarded manner. For actinide transmutation the selective separation of the actinides must be made possible.

While the MSRs that will burn plutonium and transmute actinides are still expected to be completely safe, some of the safety features of the extreme and ultimate safe versions must be compromised. The otherwise extremely low source term cannot be upheld as the actinides and plutonium must be retained in the fuel for burning and transmutation.

The rate and time it takes to accomplish the actinide disposition in MSRs is yet to be explored and calculated. The equivalent of fuel management in other reactors is to be planned; however, there is neither reprocessing nor re-fabrication of fuel to be considered.

The flexibility of MSRs allows them to fluctuate among various operation modes, as circumstances dictate, with no design modifications.

Molten salt reactors can safely dispose of actinides.

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