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**THE SOURCE TERM AND WASTE OPTIMIZATION  
OF MOLTEN SALT REACTORS WITH PROCESSING**

Uri Gat  
Oak Ridge National Laboratory\*

H. L. Dodds  
University of Tennessee

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# THE SOURCE TERM AND WASTE OPTIMIZATION OF MOLTEN SALT REACTORS WITH PROCESSING

Uri Gat  
Oak Ridge National Laboratory  
P.O. Box 2008, Building 7917  
Oak Ridge, TN 37831-6399  
(615) 574-0560

H. L. Dodds  
University of Tennessee  
Nuclear Engineering Department  
Knoxville, TN 37996  
(615) 974-2525

## ABSTRACT

The source term of a molten salt reactor (MSR) with fuel processing is reduced by the ratio of processing time to refueling time as compared to solid fuel reactors. The reduction, which can be one to two orders of magnitude, is due to removal of the long-lived fission products. The waste from MSRs can be optimized with respect to its chemical composition, concentration, mixture, shape, and size. The actinides and long-lived isotopes can be separated out and returned to the reactor for transmutation. These features make MSRs more acceptable and simpler in operation and handling.

## INTRODUCTION

Molten salt reactors are fluid fuel reactors. One of the advantages of fluid fuel reactors over solid fuel reactors is the ability to do on-line fueling and fuel processing.<sup>1</sup> Specifically, the MSRs that are fueled with uranium fluoride have the fluoride volatility process available to them, which enables simple, proven, full processing.<sup>2</sup>

Molten salt reactors can be designed with or without on line processing, or with various degrees of processing. This paper deals only with MSRs that have complete on-line processing. For fluid fuel reactors with processing, some of the common nuclear reactor terminology has no meaning. Except for some start-up period, the reactors are operated at an equilibrium steady state. The fuel concentration and content do not vary with time. Fissioned fuel is replenished by feeding or by breeding. The term fuel burnup thus has no meaning as there is no specific amount of energy generation related to a particular identifiable original amount of fissile material. For the same reason, there is no excess reactivity to compensate for burnup or for the continuous poisoning caused by fission products accumulation in solid fuel reactors. Also, fluid fuel suffers neither fuel damage, nor is a meltdown possible for a fuel that is already molten. Furthermore, when the fluid fuel

itself is circulated out of the core to an external heat exchanger ("external cooling"), fluid fuel reactors cannot have a loss-of-coolant (LOCA) accident, they do not have a separate coolant that will leave the fuel to generate heat with no cooling.

## MOLTEN SALT PROCESSING

Processing of the fuel is performed on a side stream that is drawn off the main fuel into a processing plant. An equivalent stream is fed back into the reactor. The return stream can be made up of components of the withdrawn stream and can be augmented with (or totally comprised of) unrelated fresh components. In general, fertile material, usually thorium and fissile makeup, as necessary, will be added as fresh components. The side stream and the return stream can be periodic batch quantities or a continuous stream. For the discussion here, an average constant stream is assumed as it does not impact the results as long as the batches are frequent and small in comparison to the reactor inventory. The reactor fuel inventory is the fuel mass, or volume, in the core, the external heat exchanger and all connecting pipes. The fuel holdup in the processing facility is assumed to be small in comparison to the fuel inventory. The fuel holdup in processing may not be small if the processing rate is very large.

Processing time is the time it takes to process the equivalent of a fuel inventory. A typical processing time considered for the molten salt breeder reactor (MSBR) is ten days.<sup>3</sup>

A much simplified schematic molten salt processing can be viewed as comprised of three stages (Fig. 1). In the first stage, the noble gases, other gases, and volatiles are removed. This purging of gases and volatiles can be done by sparging. This first stage is also the one that is done for all MSR's lest there be gas accumulation that may result in bubbles, pressure buildup, and instabilities. The second stage is the fluoride volatility process. In this stage, the uranium tetrafluoride is fluorinated with fluorine to uranium hexafluoride, which is very volatile and is quantitatively removed. The uranium hexafluoride is then reduced with hydrogen to the tetrafluoride which is returned to the reactor. When the molten salt reactor experiment (MSRE) switched from uranium-235 to uranium-233, the entire inventory was processed in three days.<sup>4</sup> The fissile free salt is passed on to the third stage. The salt may contain protactinium, a precursor of uranium-233, and other fissile isotopes, or their precursors. It is assumed here that all of

these are removed or that their concentration in the remaining salt is negligible.

Stage three contains the carrier salt and the nonvolatile fission products. The complete processing, assumed here, implies that all three stages are utilized and that in stage three those parts of the carrier salt, free of fission products, for which it is economical, are returned to the reactor. In extreme cases, the entire stage three salt can become waste, and new salt is used for makeup.

If, in stages one and three, not all of the fission products are removed, then an effective processing time needs to be considered. The effective processing time is a modified processing time which is the time it would have taken had all the fission products been removed. Here the assumption is made that all fission products are removed in the processing and hence the effective processing time and processing time are identical. This terminology is applicable to solid fuel reactors if the new fuel supplied is equated with the returned fuel and the makeup fuel from stage two. The equivalent stages one and three are comprised of the complete removal of all fission products along with the residual fuel. For solid fuel reactors, the operation is a batch operation with a processing time that can be approximated, as the interval between refueling.

## EQUILIBRIUM OPERATION

At steady state operation and looking only at simple decay, not compound decay isotopes, the production rate of any isotope is  $F$  and the decay constant is  $\lambda$ . For a number of atoms  $N$ , the rate of change without processing is:

$$(dN/dt) = F - \lambda N \quad (1)$$

or

$$N = (F/\lambda)(1 - e^{-\lambda t}) + N_0 e^{-\lambda t} \quad (2)$$

or assuming the start-up ( $t = 0$ ) is with no fission products ( $N_0 = 0$ ):

$$N = (F/\lambda)(1 - e^{-\lambda t}) \quad (3)$$

With on-line processing that totally removes the fission product, the removal rate per unit time due to processing is  $N/P$  with  $P$  being the (effective) processing time. Equation (1) becomes:

$$\begin{aligned} (dN/dt) &= F - \lambda N - (1/P) N \\ &= F - [\lambda + (1/P)] N . \end{aligned} \quad (4)$$

Remembering that the decay constant ( $\lambda$ ) is the inverse of the mean life  $T_m$  ( $= T_{1/2}/0.693$ ), we can substitute in Eqs. (1) through (3) for what is now the effective decay constant

$$\lambda = (1/T_m) + (1/P) . \quad (5)$$

In the following qualitative discussion, the differences between  $T_m$  and  $T_{1/2}$  are ignored. Also, the removal rate due to neutron absorptions, isotopes that are daughter products, and any power transients are ignored.

Short-lived isotopes ( $\lambda t > 3$ ) will asymptotically reach their equilibrium [Eq. (3)] of  $F/\lambda$ . Long-lived isotopes ( $\lambda t \ll 1$ ) will build up approximately linearly at the rate of [Eq. (3)]  $Ft$ .

Once the production ceases ( $F = 0$ ), say due to an accident, the processing also stops ( $1/P = 0$ ). The removal rate, which becomes the activity  $A$  is:

$$A = (N_i/T_m) e^{-t/T_m} , \quad (6)$$

with  $N_i$  the number of atoms at time of the accident from Eq. (3).

## THE SOURCE TERM

The source term is that amount of radioactivity available to an accident for dispersal and for impact on the environment. This paper deals with qualitative comparisons and therefore will not consider the type and energies of the radiation.

The activity that has been removed in processing is separate from the reactor and therefore is not available to a reactor accident. This is particularly valid for the MSR processing considered here because the waste is separated from the fuel, and as discussed later, frequently removed from the site. This compares with storage of spent fuel from solid fuel reactors away from the core. There are of course possible accidents in the processing facility, however, these are separate from those of the reactor and have their own, different, source term. The reactor here is that part of the system that contains the fuel inventory (i.e., core, heat exchanger, and piping).

Typical processing times for solid fuel reactors (refueling time) are 300 to 1000 days. Processing times for the MSBR were designed at 10 days.<sup>3</sup> Speculative processing times for MSRs, to enhance safety, are shortened down to one day.<sup>5</sup>

For isotopes with short half life for which  $T_m \ll P$  the effective decay constant equals the decay constant and the isotopes rapidly build up to their equilibrium level of  $F/\lambda$ . This short-lived isotopes group is of course much smaller for the MSR than for other reactors since its processing time is one to two orders of magnitude smaller than that of solid fuel reactors. This group of short-lived isotopes also decays much faster. Since most accidents have a finite time interval between the accident and the dispersal of the source term, this part of the source term is in actuality significantly diminished by the time it reaches the environment. Here again the effect is much greater for the MSR as this group includes only those isotopes with average lifetimes shorter than the processing time, which is up to a few days.

Isotopes for which the processing time is shorter than the average lifetime ( $P < T_m$ ) the effective decay constant becomes dominated by the processing time. The quantity of these isotopes is limited to the production rate times the processing time ( $FP$ ). This means that the long-lived isotopes source term is proportional to the processing time, and thus is one to two orders of magnitude lower for a MSR than for a solid fuel reactor. This group of isotopes is the important one for the source term as it includes those that if released could linger in the environment for many decades. The group includes such prominent members as strontium and cesium with half lives of decades.

Not considered here is the driving energy that actually disperses the source term. This driving energy is usually the afterheat produced by the same isotopes as the source term. As the source term is reduced, so is the energy that can drive it. (This is not applicable to a criticality accident that has a driving energy from fissioning.)

## THE WASTE

The waste is encountered in stages one and three in the MSR with full processing. The waste treatment described here is not a developed or tested process. The treatment is a suggested schematic outline that must be developed, detailed, tested, and confirmed. However molten salt chemistry in general is widely used on a large

scale in the chemical industry and is generally well known and developed.

In stage one the volatiles and gases are separated out. The nonradioactive gases can be separated and reused. The radioactive gases can be either adsorbed on adsorbing materials or collected in bottles for disposal. In either case the quantities and the configuration of the bottles or containers can be optimized. The general recommendation is to reduce accumulations on-site to a minimum and to have frequent shipments of small, low-risk, quantities to a disposal site. Shipments would be as frequent as one or more per day. The volatiles separated in stage one can either be treated together with the gases or, after altering their chemical composition, combined with the stage three materials.

Stage three materials are free of fissile material and contain the nonvolatile fission products and any actinides. The protactinium, the precursor for uranium-233, and any plutonium that may be present due to either breeding or because it was used as initial fissile material, is removed first. Any plutonium will be returned to the core. Protactinium may be held up to convert to uranium prior to return to the core.

As a next step, the actinides, if present, and some elements that contain long-lived isotopes, such as iodine-129, will be separated out and fed back into the reactor for transmutation. This step needs to be carefully analyzed. Feeding the actinides and long-term isotopes back into the reactor makes them part of the source term and affects conversion or breeding. Keeping them with the waste makes the waste a very long half-life waste that requires disposal in appropriate formations. An accommodating solution can be to dispose of the actinides and long-lived isotopes separate from the other waste. This will greatly reduce the mass and variety of material that requires very long-term disposal. This waste could be encapsulated in very secure containers in relatively small quantities. If sufficiently concentrated, they can be utilized as long-range, low-power heat sources.

The other fission products can then be processed into the most desirable chemical combination form. The idea here is to bring them to a stable form that will not leach or otherwise escape to the environment. They can further be concentrated to any desirable volume since there are no criticality or diversion concerns to be considered. They can be mixed or diluted with other inert or stabilizing

materials. They can be fixed in some matrix material such as glass or ceramic that will assure their safe containment for their anticipated life. They can also be encapsulated in any desirable shape or size. An example of a shape that may be useful is a narrow rod. A narrow rod has good heat transfer properties that will prevent the rod from melting. The narrow rod also has relatively little activity per unit length and can be shielded within a cylindrical radiation shield of fairly small thickness. Also, a narrow rod can be disposed of in a hole in the final disposal formation with comparatively simple and lightweight remote manipulation tools.

The shipments of the waste from the reactor site should be frequent and contain a small quantity per shipment. The approximately 3 kg of fission products generated per day can be shipped in two to six daily shipments. These frequent small shipments will assure that the waste is not accumulating on the site and thus not constituting an on-site risk. The small shipments assure that the risk associated with each shipment is small, and the shipping containers do not require difficult elaborate handling.

## SUMMARY AND CONCLUSIONS

Molten salt reactors utilize fluid fuel that enables on-line fuel processing. MSR's in general are designed to remove at least the fission gases and volatiles, which reduces the source term. When all the fission products are routinely removed, the source term for MSR's, as compared to solid fuel reactors, is reduced for long-lived isotopes (those isotopes whose average life is longer than the processing time) by the ratio of the MSR processing time to the solid fuel reactor refueling time. This reduction can be one to two orders of magnitude. Also the group of isotopes included in this category is related to the relative shorter processing time rather than to the longer refueling time. The short-lived isotopes (those whose average lifetime is short compared to the processing time) build up to an equilibrium that is determined by their average lifetime. However, that group, because of their short life, also decays very fast and generally does not constitute a long-range environmental problem. For a typical MSR with a processing time of ten days or less, a few months correspond to six to ten half lives of these isotopes. The fission products removed from the reactor are no longer available to a reactor accident as a source term.



The long-lasting source term of an MSR as compared to solid fuel reactors is reduced by the ratio of processing time to refueling time. For typical reactors this reduction is one to two orders of magnitude.

The waste encountered in the fluid processing facility is nominally free of fissile fuel. Thus the waste is not subject to criticality, diversion, or proliferation concerns. The waste can be optimized for acceptance criteria. The waste can be optimized for chemical combination, dilution or concentration, elemental makeup, matrix, fixation, encapsulation, shape, size, and configuration. Transportation of the waste can be made in small frequent shipments so that each constitutes only a small hazard, and there is no accumulation on the processing site. Shaping the waste into a narrow rod form assures good heat transfer, simple shielding, and easy manipulation.

The actinides and other long-lived isotopes can be separated out from the waste and returned to the reactor for transmutation. Returning actinides to the reactor also returns them to the source term, but removes them from the long-term waste containment concern. The actinides could also be disposed as a separate waste.

The waste of an MSR can be optimized in almost any respect to make the handling and disposal simple and acceptable and to lower the risk. Further considerations and development are needed to determine optimization and acceptance criteria.

MSRs with processing can reduce the source term, optimize the waste, transmute the actinides and long-lived isotopes if desired, to make them more acceptable, safer, and alleviate many of the concerns related to nuclear reactors.

## REFERENCES

1. J. A. LANE, H. G. MACPHERSON, and F. MASLAN, Fluid Fuel Reactors, Adison-Wesley, Reading, Massachusetts (1958).
2. W. H. CARR et al., "Molten Salt Fluoride Volatility Pilot Plan: Recovery of Enriched Uranium from Aluminum-Clad Fuel Elements," ORNL-4574 (April 1971).

3. R. C. ROBERTSON (Ed.), "Conceptual Design Study of a Single-Fluid Molten-Salt Breeder Reactor," ORNL 4541 (June 1971).
4. H. G. MACPHERSON, The Molten Salt Reactor Adventure, Nucl.Sci.Eng. 90, (1995).
5. URI GAT, The Ultimate Safe (U.S.) Reactor – A Concept for the Third Millenium, 4th Int. Conf. Emerging Nuclear Energy Systems, Madrid, Spain (June 30-July 4, 1986).