

# INCINERATION OF TRANSURANIC WASTE IN A MOLTEN SALT REACTOR BASED ON THE THORIUM-URANIUM-233 FUEL CYCLE

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## 1. Introduction

The Molten Salt Reactor (MSR) is suitable for the removal of actinides, accumulated in light water reactors, of dismantled nuclear weapons, and for the efficient and economical energy production. It has a simpler design as a reactor with solid fuel. The neutron economy is much better, because mainly non-fissile materials with very low neutron absorption cross section are present and no artificial absorbers in the coolant and control rods are necessary. MSR-fuel can consist of all three major fissile fuels, U 233, U 235, and plutonium. At FZR, the MSR-effort is focused to minimize the stock of plutonium and minor actinides by spent fuel transmutation. Stimulated by the MOST-project, first preliminary calculations on MSR were carried out.

## 2. Molten Salt Reactor

The U. S. Nuclear Energy Research Advisory Committee and the Generation IV International Forum rank the MSR among the six most promising nuclear reactor-systems [1]. In a MSR fission power will be produced in a circulating molten salt fuel mixture. MSRs are fueled with uranium or plutonium fluorides dissolved in a mixture of molten fluorides, with Li 7 and Be 9 fluorides as the primary option. Molten fluoride salts have a very low vapor pressure. This reduces stresses on the vessel and piping and hazards associated with high pressures, such as ruptures or depressurizations. Potentially, MSRs have a high availability - refueling, processing, and fission product removal can be performed on-line. High temperature, ranging from 500 to 800 °C, offers the potential for thermochemical hydrogen production.

Four fuel cycle options are under discussion: (1) Maximum conversion ratio using a Th-U-233 fuel cycle, (2) denatured Th-U-233 converter with minimum inventory of weapons grade material, (3) denatured once-through actinide burning (Pu and minor actinides) fuel cycle with minimum chemical processing, and (4) actinide burning with continuous recycling.

Two liquid fuel reactors have been designed, built, and operated by Oak Ridge National Laboratory (ORNL): the Aircraft Reactor Experiment (ARE) and the Molten Salt Reactor Experiments (MSRE). The operation of the Molten Salt Reactor Experiment (MSRE) with various fuels from 1964 to 1969 was very successful [2]. Based on these experiences the concept of the molten salt breeder reactor (MSBR) was developed [3]. The present investigations in Europe are based mainly on the papers of the latter program. The original program was finally closed in 1976. It stimulated molten salt reactor studies in many other places, for example in the Soviet Union [4]. One program was conducted at KFA Jülich with the Molten Salt Epithermal (MOSEL) Reactor in the sixties [5].

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### **3. Safety and waste**

The safety properties of the MSR are much more favorable than those of the solid fuel reactors. The fuel is critical only if the moderator - the graphite - is present. If the fuel escapes from the core, it will become subcritical. A meltdown is impossible. Because of the expansion of the fluid the fluid fuel has a negative temperature coefficient. The continuous removal of Xe 135 by Helium and of other fission gases reduces significantly the radioactivity within the core. The fission products with short half-lives can be removed by on-line processing, those with a long half-life can be retained in the salt, for example I 129, as long as they are transmuted into stable nuclides. The salts are chemically stable and nonflammable, and there are no dangerous chemical interactions between the salts and water.

In case of fluid fuel it is possible to shutdown the reactor by dumping the salt into outer containers from which the decay heat can be removed by thermal conduction and natural convection. If no Pa 233 is removed from the fuel, which decays with a half-life of 27 days into the fissile U 233, the MSR is proliferation resistant, as proposed in the Denatured Molten Salt Reactor concept [6]. It was summarized that MSRs can be designed in an extremely safe and economical manner.

The safety and the handling of nuclear waste mainly determine the acceptability of nuclear reactors. The continuous processing of the waste, the separation of short-lived nuclides and the potential transmutation of long-lived fission products and actinides minimizes the waste problem. This reduces the need for very long controlled storage time of the waste.

### **4. Project MOST**

Under the headline “Review of Molten Salt reactor Technology (MOST)” a EURATOM-project was initiated in 2001, coordinated by the French Commissariat à l’Energie Atomique. The goal of the project is to summarize the state of the art on molten salt reactor. This review analyses different scenarios in which MSRs can be used, and it focuses on the best available data and methods to define the weak points and advantages. Sixteen institutes of nine countries take part in this project with duration of two years. The project work plan is subdivided into nine work packages, and the Institute of Safety Research of the Forschungszentrum Rossendorf (FZR) is involved in the work package 2 “Reactor physics study and non-proliferation issues” and in the work package 3 “Design review on nominal operating conditions”. MOST reviews the concepts of MSRE [2], MSBR [3], and the concept Actinides Molten Salt TransmutER (AMSTER) of Electricité de France [7].

### **5. MSR model**

In co-operation of the FZR and the KIAE Moscow a code system for MSR consideration was developed linking the well-known codes MCNP-4C and ORIGEN 2.1. Our calculations of the Denatured Molten Salt Reactor [6] for a time period of three operating years showed the internal consistency between our calculations with the codes MCNP-ORIGEN and the Russian calculations with the codes MCU-ORIGEN-S. In the framework of the MOST project, preliminary studies were performed to examine the fuel cycle characteristics of a MSR of the AMSTER-type.

The studied AMSTER-model of the MSR has a cylindrical core with removable blocks of graphite, a fixed graphite reflector, and a vessel of the Ni-Mo-Cr-Fe-alloy, Hastelloy N. The removable blocks with a hexagonal shaped cross section have vertical holes in which the molten salt flows. The graphite moderates the neutrons. Usually the reactor model has two regions without graphite, the lower and the upper plenum. These parts were disregarded in the present calculations. In contrast to solid fuel reactors the fuel in liquid fuel reactors is heat carrier and coolant as well.

Our calculations were done with a salt composition of 70 % LiF (Li 7 only), 15 % BeF<sub>2</sub> and 15 % ThF<sub>4</sub> (mole percent), a graphite density of 1.68 g/cm<sup>3</sup>, a salt density of 3.328 g/cm<sup>3</sup>, a temperature of salt and graphite of 900 K and a thermal power 2250 MW. The system contains 168 metric tons of salt and 400 tons of graphite. The main geometrical and physical parameters are summarized in Table 1.

Table 1: The calculational model geometry and the main physical parameters

Radius of the MSR (cm)	411.1571
Height of active core (cm)	500
Thickness of radial reflector (cm)	50
Thickness of axial reflector (cm)	50
The side of the hexagon (cm)	13
The radius of hole in the hexagon (cm)	4
Total salt volume in primary circuit (m <sup>3</sup> )	48.4
Salt volume in active core (m <sup>3</sup> )	30.4
Thermal power of reactor (MW)	2250
Electrical power of reactor (MW)	1000

As fuel feeding strategy of the MSR burner, the inventory of Th 232 was kept constant in the core. The criticality was assured by feeding TRU-nuclides (Np, Pu, Am, and Cm).

Table 2: Composition of the feed in transuranic (TRU) nuclides in percent

Np 237	6.419
Pu 238	3.179
Pu 239	43.940
Pu 240	21.272
Pu 241	13.516
Pu 242	7.875
Am 241	0.545
Am 243	2.328
Cm 244	0.926

Under startup conditions, all fissions occur in these TRU-nuclides. Then more and more U 233 contributes to the fission process. The TRU-feeding is continued until the end of life (EOL) of the reactor with constant percentage of the TRU-nuclides, typically from spent fuel of light water reactors (LWRs) after reprocessing. The amount of feeded TRU varies and must be recalculate in each time step in order to fulfill the criticality condition.

A main reprocessing time of the MSR-salt of 300 equivalent full power days (efpd) was used with the assumption of a loss rate of  $10^{-3}$  for the transuranic nuclides (TRU). The Table 3 lists the recycling characteristics of our calculations.

Table 3: Recycling characteristics for the present calculations

Component	Removal time	Removal operation
Kr, Xe (noble gases)	50 s	Sparging with He
Zn, Ga, Ge, As, Se, Nb, Mo, Ru, Rh, Pd, Ag, Tc, Cd, In, Sn, Sb, Te	2.4 h	Plating out on surfaces and escape to off gas system
U 233, U 234, U 235, U 236, U 237, Np (tetravalent)	300 efpd	Volatilization in primary fluorinator
Br, I (halogens)	300 efpd	
Zr	300 efpd	Reductive extraction
Ni, Fe, Cr (corrosion products)	300 efpd	
Pa 233 (tetravalent)	No	
Pu, Am, Cm (trivalent TRU)	300 efpd	
Y, La, Ce, Pr, Nd, Pm, Gd, Tb, Dy, Ho, Er (trivalent lanthanides)	300 efpd	
Sm, Eu, (divalent lanthanides)	300 efpd	

## 6. Numerical results

From the calculations we get nuclide number densities of all components of the salt as a function of time. So we can calculate the inventory of all nuclides. Fig. 1 shows the inventory of the heavy nuclei and the fission products (FP) versus depletion time for a life of 80 years. The scale is logarithmic in order to present such different inventories as those of Th 232 with 79 tons and Pa 233 with 61 kg (at the EOL). As we can see, after a certain time the inventory of most of the nuclides is almost constant. The TRU-nuclides and the U 233 follow the Th 232 in the mass fraction. For the fission products and U 236 an equilibrium cannot be really achieved. At EOL an amount of 100 kg of fission products still exists. But the radiotoxic waste remains in the system in contrast to LWRs. At EOL the fraction of neutrons generated by fission is 99.5 %, those generated by (n,2n)- and (n,3n)-reactions is 0.5 %. The neutron (and energy) production by fission can be subdivided as follows: 57 % from U 233, 6 % from U 235, 12 % from Pu 239, 17 % from Pu 241, and 6 % from Cm 245. A small contribution of 1.5 % comes from the more than 15 other TRU-nuclides.

In Fig. 1 the sum of TRU-nuclides was plotted. Fig. 2 shows the transuranic inventory in more detail. The mass fraction of Pu 239 is the highest in a short period after startup only. Then the non-fissile nuclides Pu 240 and Pu 242 are the dominating TRU-nuclides, later Am 243 too. The shown TRU-distribution strongly depends on the neutron spectrum of the system. A harder spectrum than in the present AMSTER-model, attained, for example, by reducing the mass-fraction of the graphite moderator with “larger holes”, can drastically change the distribution of TRU-nuclides.

It should be mentioned that the MSR-system could also be used in an undercritical system in connection with a proton accelerator, called in an Accelerator Driven System (ADS). As mentioned above, in our critical system we feed the system continuously with TRU-nuclides and incinerate some of the radiotoxic waste of LWRs.

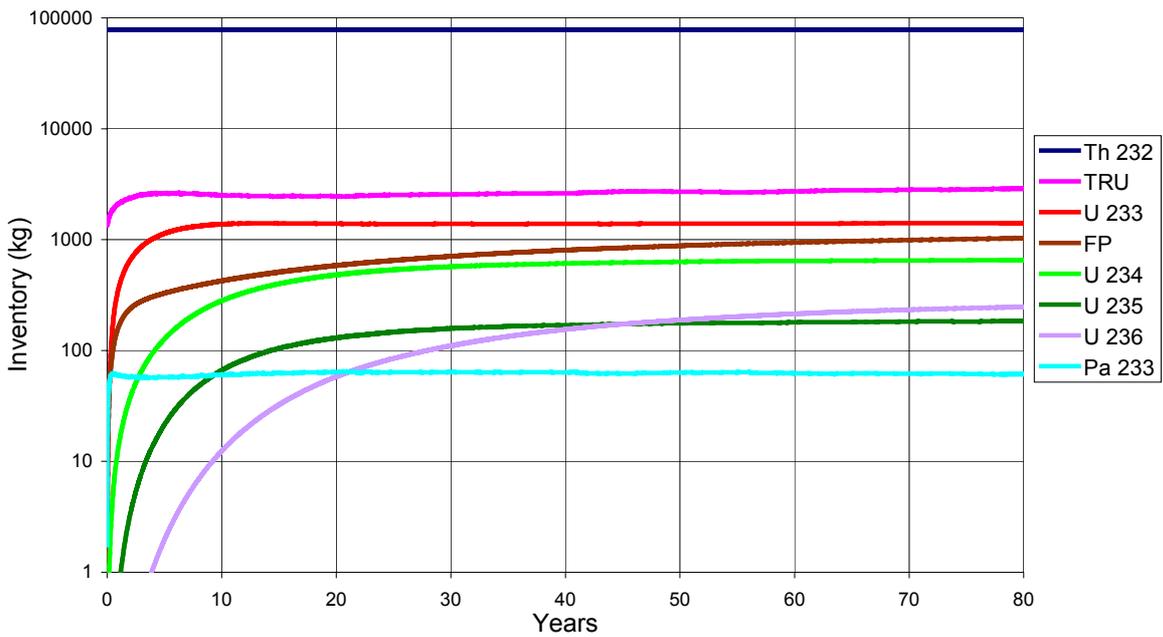


Fig. 1: Inventory of heavy nuclides and fission products (FP) versus depletion time for the AMSTER-system

Simultaneously with an energy-production of 80 GWa a surplus of almost 20 metric tons TRU-waste of the LWRs could be burned with this AMSTER-system, as shown in Fig. 3, and (almost) no new radiotoxic waste would be extracted from the system.

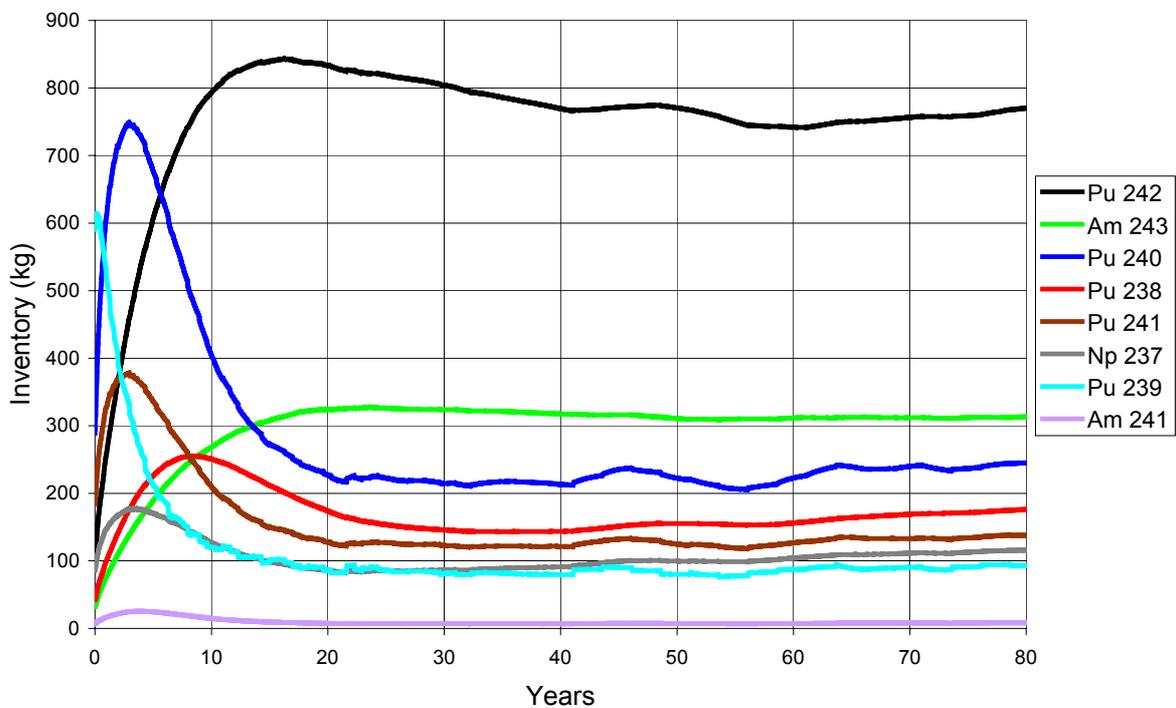


Fig. 2: Detailed transuranic (TRU) inventory versus depletion time

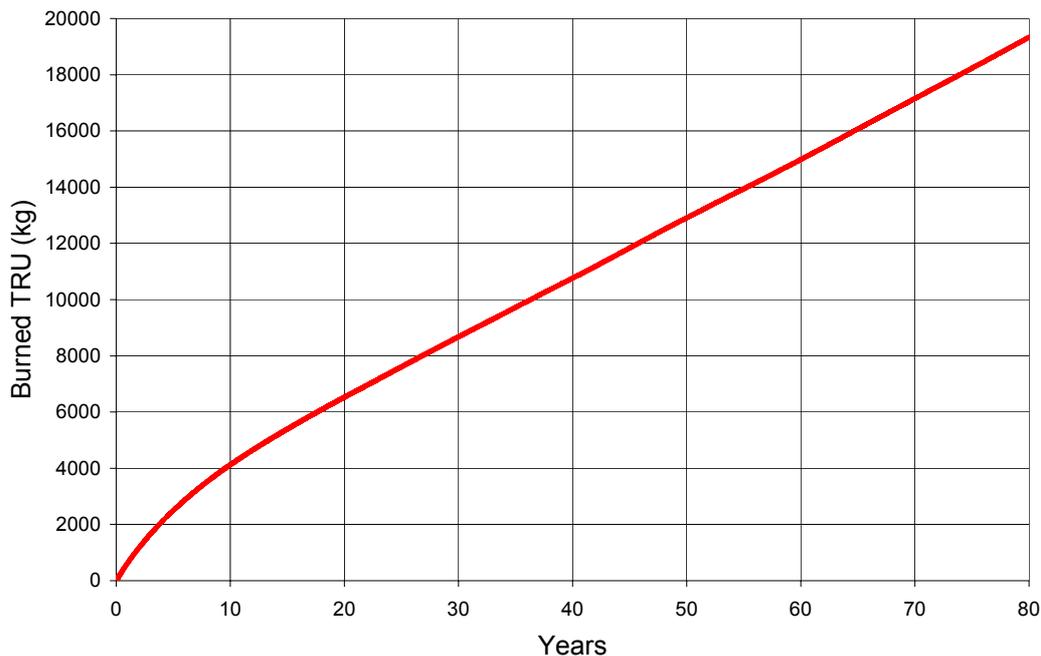


Fig. 3: The burned (“consumed”) TRU-mass in the Molten Salt Reactor of the AMSTER-type

## 7. Summary and prospects

Our results, obtained for the AMSTER-model of the burner type, which was not completely defined at the date of our calculations, demonstrate the physical feasibility of MSR. There are small differences with the results of other groups due to various approaches in the fuel policy: Somewhere the total heavy nuclei inventory was kept constant during transition to the equilibrium. In our calculations the Th 232 inventory is constant. Moreover different salt and graphite densities were used and different uranium losses in the thorium- uranium cycle, too.

The Monte-Carlo calculations with the code MCNP are very time consuming. Therefore, applications of faster deterministic codes like the well-known collision probability cell-code HELIOS and discrete ordinates transport codes as well as diffusion codes are planned. Many of these codes have never been used for calculations of reactors with fluid fuel, so the data libraries must be provided with supplemental nuclides. Since the geometry of the MSRs differs from those of LWRs, some other changes will be necessary on these deterministic codes. Moreover, fundamental research will be necessary, for example to determine kinetic and thermodynamic data, to determine the optimum process for separating fission products, including lanthanides without removal of minor actinides. Data of the solubility of minor actinides and lanthanides will be needed to design MSRs capable of burning minor actinides with minimum inventories in the reactor.

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