Molten salts and nuclear energy production

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Abstract: Molten salts (fluorides or chlorides) have been taken in consideration very soon in nuclear energy production researches. This was initially due to their advantageous physical properties: good heat transfer capacity, radiation insensitivity, high boiling point, .. and they can be used in various situations: heat transfer, core coolants with solid fuels, liquid fuel in molten salt reactor, solvents for spent nuclear solid fuel in the case of pyro- reprocessing, fusion. Molten salt reactors which are one of the six innovative concepts chosen by the Generation IV international forum may be particularly interesting in the case of waste incinerators or of the thorium cycle. As the neutron balance is very tight, the possibility to quickly extract poisoning fission products is very attractive. The most important questions addressed to demonstrate the scientific feasibility of Molten Salt Reactor will be reviewed.

Keywords: Molten salts, nuclear reactors, heat transfer, thorium cycle

1. Introduction

The main characteristic of nuclear energy production is the large energy release by nuclear reaction (fission) compared to chemical energy production reactions (at least a factor 10^7). So the choice of a coolant able to transfer under irradiation large amount of heat is an important concern and the choice is further reduced when taking into account the nuclear properties. The objective of the paper is to discuss the possibilities offered by molten salts in the nuclear energy production. First we discuss the properties of the molten salts based on fluorides or chlorides which make them an attractive choice. We will then discuss the elementary composition of the salt taking into account the nuclear properties (neutron capture and diffusion), the desirable chemical and physical properties and the treatment it must undergo. Molten salts are particularly interesting because they may be used either for the whole heat transfer between two places or as both heat and fuel carriers in reactors. Very soon after the beginning of the nuclear era, molten salt reactors were studied especially for the thorium cycle which presents very interesting features. These will also be briefly discussed in the paper.

2. Why molten salts in nuclear energy production?

A nuclear reactor core is a place where a large amount of heat is produced and has to be carried out quickly in an environment characterized by high neutrons fluxes and a high radiation level. There are few possible fluids able to effectively bear these constraints and to remove the heat and transfer it to the first heat exchanger. Amongst them, the molten salts were considered very early for the following reasons:

- The capability to heat transfer is very good compared to the gases and the liquid metals [Forsberg, 2005]
- The high boiling point which allows them to function at high temperature at near atmospheric pressure
- The great insensitivity to radiations which prevents from gas creation and changes in chemical properties.

Another interesting feature is that each fluorinated actinide is more or less soluble and may be mixed with chosen salts according to the requested properties. This idea leads to the molten salt reactor where the fuel is part of the salt moving in and out of the core. Two experiments with molten salt reactors were successfully conducted in Oak Ridge: the ARE (Aircraft Reactor Experiment) was running for approximately one hundred hours around 1955 at a thermal power of 2.5 MWth. Ten years later the MSRE (Molten Salt Reactor Experiment) started [Rosenthal, 1970] an 8MWth reactor fuelled at the beginning with ^{235}U and at the end with ^{233}U, ran for five years without any problem As the power was low, there was no need to perform one- line salt reprocessing. This preparatory work led the ORNL team to present a power reactor project, the Molten Salt Breeder Reactor [Bettis 1970] fuelled with thorium and ^{233}U as fissile material. As the maximum breeding ratio was the main objective, a very constrained salt reprocessing was required where the whole salt ($48m^3$) has to be reprocessed in 10 days

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with efficiencies depending the element (e.g. 100% for Pa). Besides the demonstration of the validity of the concept, the experiments have given very important informations about the corrosion of structural materials and graphite and lead to the choice of Hastelloy- N for the tank and the pipes. In the molten salt reactor, the question of the fuel conditioning, of its cladding and of the resistance to high temperature, irradiation and corrosion are avoided. The salt composition may be controlled and adjusted each time, there is no need of reactivity reserve inside the reactor and very high burn- up is possible.

3. How to choose the salt?

Several points of view have to be taken into account

3.1. Nuclear point of view

As the salt spends a lot of time in a very large neutron flux, the first investigation concerns the neutron capture probability by the salt constituents. A large capture cross- section leads to a loss of neutrons which may be not acceptable for the reactor operation so the elements with the smallest cross-sections are preferred. The second consequence of the capture is the production of a new isotope which may be undesirable from the point of view of radioprotection (production of long lived isotope such as ^{36}Cl from capture on ^{35}Cl) or after decay of a new element which may be undesirable from the point of view of the radioprotection or of the chemistry. The diffusion properties of the neutrons on the components and the atomic number of the components have also an influence on the neutron spectrum in the reactor core (neutrons more or less quickly thermalized).

3.2. Chemistry point of view

Following the choice of a good coolant with acceptable viscosity, the main requirement for the salt is its stability to avoid circulation troubles due to solid precipitation. A good knowledge of the phase- diagram is required, taking into account the fission products and actinides produced by the nuclear reactions. A good on- line redox potential control is also needed to avoid corrosion. Finally the reprocessing must also be considered. Assuming that a bubbling is able to quickly remove gaseous elements and some insoluble metallic elements, a further reprocessing step is still required to remove lanthanides from the salt. The salt choice may interfere with the chemical reprocessing methods that are available to remove various components (U, Th, minor actinides) prior to lanthanides extraction and this has to be taken into account.

4. Possible applications

The multiple applications which are now foreseen in the nuclear energy domain production are the following:

1- Salt loops for high temperature heat transfer. For example, future plants designed for hydrogen production are assumed to be based on various processes which require high temperature. Some very high temperature reactors under development are designed with gas coolants which are not well suited to long distance heat transfer between the reactor and the hydrogen production plant which must be separated for safety reasons.

2- Liquid coolant in solid fuel reactors. Molten salts are some of the very few fluid coolants which are able to sustain temperatures needed for the proposed direct hydrogen production methods (T>850°C). Hence, in the United States, one of the VHTR concepts studied is the Advanced High Temperature Reactor [Forsberg 2004] (AHTR), a thermal neutron reactor with conventional fuel (^{235}U and Pu) cooled by molten fluorides. Molten salt may also be used as a coolant in a solid fuel fast reactor (LSFR) to replace liquid metals or gas without limitations on temperature or pressure. In the fusion case, it may play a role both as heat carrier and tritium source due to neutron reaction on ^{6}Li.

3- Liquid fuel reactors. The concept, which was experimented with at ORNL, has many interesting features which allowed study of its use for many different applications. The first was the MSBR project, to produce energy via the thorium cycle, which will be discussed later. Thermal or fast neutron molten salt reactors with Uranium-Plutonium or Thorium-uranium cycle are now considered in several institutions. They may allow safety questions to be overcome and high temperature and pressure problems for energy production in breeder reactors to be solved. Furthermore, they are also considered for critical or sub- critical transmutation systems as fuel fabrication difficulties are avoided.

4- Reprocessing of spent fuels by pyro- reprocessing methods. Up to now the only industrial process at work to reprocess the spent nuclear fuel is the hydrometallic liquid- liquid process. With the new fuel supports (nitride, carbide) and the high radiation level which will characterize the innovative concepts, the limits of this process may be reached quickly. As fuel reprocessing is more or less unavoidable for future nuclear energy systems, pyro- reprocessing may be the only other solution and is currently studied.

5. Thorium cycle and molten salts reactors

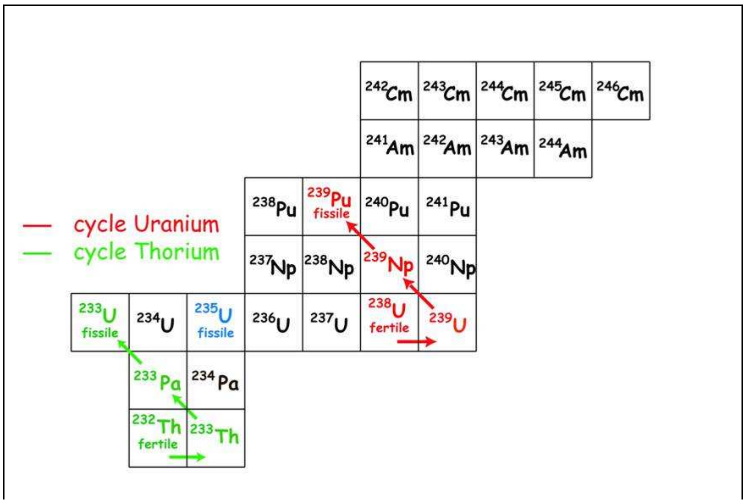

Figure 1 Actinide Chart

Nuclear production has up to now been only based on the use of ^{235}U. It is the only fissile element available in nature. Moreover figure 1 shows that there are only two other possibilities for nuclear energy production that are the fertile nuclei ^{238}U and ^{232}Th. After a neutron capture and two β- decays, the fissile nuclei ^{239}Pu and ^{233}U are produced and may be used as fuel in reactors. The only way the neutron capture can occur at a sufficient rate is to place the fertile element in a reactor with enough fissile material to start the chain reaction; the reactor will be a “breeder” if it is able to produce a number of fissile nuclei equal or greater than the number of fissile nuclei that disappear. If ν is the number of neutrons emitted by fission and α, the ratio of capture cross-section divided by the fission cross-section induced by neutrons as a function of energy, the available neutron is given by $N_d = \nu - 2(1+\alpha)$. This quantity is plotted on the figure 2 for the two fertile elements. It appears that, as the available neutron number is slightly larger than 0, breeding is possible for the whole neutron energy spectrum for thorium whereas it is only possible for neutron energy larger than a few ten keV for plutonium. This explains why, if plutonium is produced and partly burnt in the light water reactors, it is impossible to reach an interesting breeding ratio with a thermal neutron spectrum. The main advantage of the thermal spectra is that the required fissile material for starting the chain reaction is smaller (factor up to six) than for the fast neutron reactor.

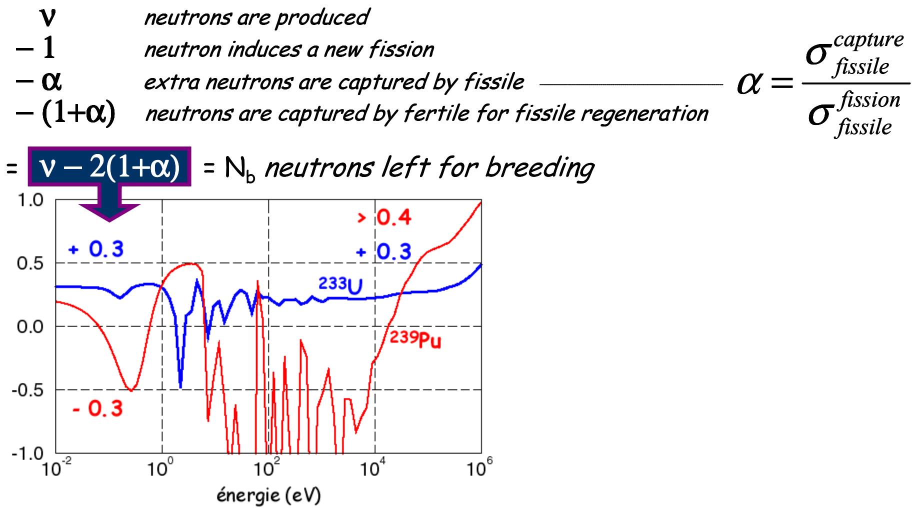

Figure 2:Available neutrons

Another interesting feature of the thorium cycle is the lower production of actinides which are the main contributors to the radiotoxicity of the spent fuel. Figure 1 shows that five successive neutron captures are necessary to reach the neptunium whereas the Plutonium is already right in the middle of the actinides. The radiotoxicity which tries to assess the risk due to the spent fuel of the various fuel cycles as a function of time is given on the figure 3 which shows clearly the advantage of the thorium cycle. As the number of available neutrons is as small as 0.3, it is very important to minimise all the potential neutron losses. As some fission products are very neutron capturing, it is very interesting to remove them as soon as possible from the reactor core and it is one of the reasons why the thorium cycle has been linked to the molten salt reactors from the start.

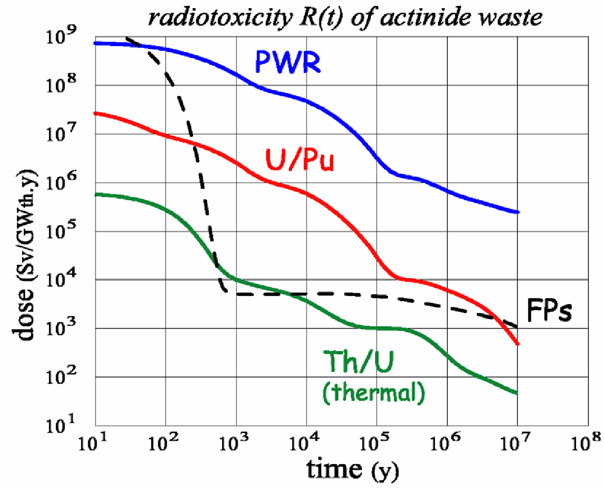

Figure 3:Radiotoxicityof the actinides wastes

6. Molten Salt Reactors to day

Starting from the MSBR project, many studies have been made in several countries concerning the possible uses of the MSR. There was an important renewal of interest when the transmutation of long life nuclear wastes became a big concern. There were studies about critical and sub- critical reactors loaded in some cases directly with spent fuel fluorides. A review concerning the various aspects of the MSR has been made during the EURATOM Concerted Action MOST [MOST 2005]. The future studies have been separated in five groups which are: design and safety, reactor physics, fuel salt chemistry, material- mechanics, fuel salt clean-up. Several concepts are under evaluation but a special emphasis is put on the simplest design, the TMSR (Thorium Molten Salt Reactor). In this concept [Merle- Lucotte 2005], the reactor is fuelled only with Th and ^{233}U, the required breeding ratio is close to one and the reprocessing has been simplified as much as possible. Only bubbling to extract gaseous products and some noble metals and salt property control are made on line, the remaining part of the reprocessing is being performed in a separated unit where the whole core volume is processed in at least six months. Some designs have been found to obtain good reactivity coefficients. The goal of the works undertaken in the five domains defined before is to demonstrate as soon as possible the scientific feasibility of molten salt reactors. Molten salt reactors are also studied as waste incinerators.

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