Optimization and simplification of the concept of non-moderated Thorium Molten Salt Reactor

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Abstract

Molten salt reactors, in the configuration presented here and called Thorium Molten Salt Reactor (TMSR), are particularly well suited to fulfil the criteria defined by the Generation IV forum, and may be operated in simplified and safe conditions in the Th$/^{233}$U fuel cycle with fluoride salts. The characteristics of the non-moderated TMSR based on a fast neutron spectrum are detailed in this paper: we aimed at designing an optimised TMSR with the simplest configuration. Using a simple kinetic-point model, we analyze the reactor's transient as the total reactivity margins are introduced in the core. We thus confirm, beyond the classical advantages of molten salt reactors, the satisfactory behaviour of the TMSR in terms of safety and the excellent level of stability which can be achieved in such reactors.

1. Introduction

To satisfy the criteria chosen by the Generation-IV International Forum for the new nuclear energy systems, we have developed a new concept of molten salt reactor based on the Thorium fuel cycle, and thus called Thorium Molten Salt Reactor (TMSR). The main advantages of this system are due to its liquid fuel and coolant and the Thorium cycle. The amounts of fissile and fertile matter can be adjusted without unloading the core, avoiding any initial reactivity reserve; the Fission Products which poison the core can be extracted without stopping reactor operation; nuclear waste production is minimized; and high temperatures may be reached without high pressure in the core.

Starting from the historical Molten Salt Breeder Reactor project of Oak-Ridge (Whatley et al., 1970), some parametric studies (Mathieu 2005; Mathieu et al., 2006; Nuttin et al., 2005) correlating the core arrangement and composition, the reprocessing performances, and the salt composition have been done in terms of safety coefficients, reprocessing requirements, and breeding capabilities.

Amongst all TMSR configurations, these studies have singled out the configurations with no moderator in the core as particularly simple and promising. Such a reactor presents indeed many intrinsic advantages, avoiding the deterioration of the moderator while ensuring excellent safety characteristics.

After the presentation of the design of such a non-moderated TMSR or 'TMSR-NM' and the

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description of the computational procedures, we first detail the results of an optimization study of this system based on numerical simulations, in terms of breeding ratio, feedback parameters and damage to structural materials. We then concentrate on a simplified design of the core, with no fertile blanket, to check the impact of such a modification on the performances of the reactor. We finally address the safety aspects, through a first assessment of the level of intrinsic safety (or stability) of the reactor based on a simple kinetic-point model, ending with some preliminary considerations necessary to define a safety approach specific to MSRs.

2. The non-moderated Thorium Molten Salt Reactor or TMSR-NM concept

2.1. Reactor design

The TMSR concept is a 2500 MWth reactor. The operating temperature is 630°C, corresponding to a thermodynamic efficiency of 40 %. As shown in Fig. 1, the core comprises a single cylinder whose internal diameter is approximately equal to its height (in our simulations: 2.6m high, 1.25m radius) and where the nuclear reactions occur within the flowing fluoride salt (shown in yellow in Fig. 1). We consider as part of the reactor itself all the components of the system containing the primary salt, i.e. the whole primary circuit (core, pipes, pumps and intermediate heat exchangers). The reactor represents the first barrier for the fuel in the defence-in-depth principle. It is enclosed in the reactor vessel which corresponds to the second barrier.
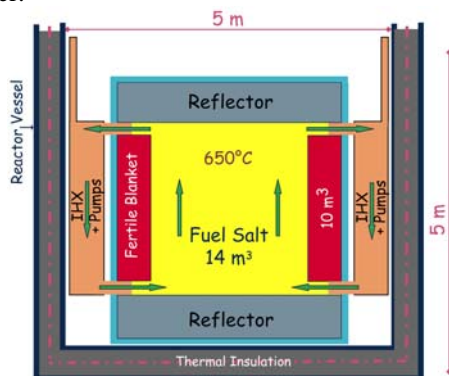

Fig. 1. To scale vertical representation of the TMSR, including pumps and intermediate heat exchangers (IHX) in the reactor vessel

The external core structures and the heat exchangers are protected by reflectors which have been designed to absorb 80% of the escaping neutron flux. The axial reflectors are made of a specific hastelloy currently being developed (Cury, 2007). In the general TMSR-NM concept, the radial reflector consists of a fertile blanket made of the same hastelloy and containing a binary fluoride salt LiF-ThF$_4$ with 28%-mole of ^{232}Th. Thanks to ^{233}U extraction within six months, this fertile blanket improves the breeding ratio.

The core contains a fluoride fuel salt, composed of LiF enriched in ^{7}Li (99.999 %) and heavy nuclei (HN) amongst which the fissile element, ^{233}U or Pu. The fraction of salt outside the core is assumed to be 1/3, flowing in the pipes, pumps and heat exchangers. For HN proportions ranging from 20 to 30 mole%, a binary salt LiF-(HN)F$_4$ has been chosen whose melting point is around 570°C. For lower proportions of HN, the calculations have been done with a salt containing 80 mole% of LiF completed with BeF$_2$ (other possible components like NaF or CaF$_2$ may be used) to lower the eutectic point temperature and to allow operation at 630°C. The salt density ranges from 3.1 to 4.6 according to the HN proportion, with a dilatation coefficient of 10^{-3}/°C (Ignatiev et al., 2005).

The salt management combines a salt control unit, on-line gaseous extraction and - off-line lanthanide extraction by pyrochemistry. With the on-line control and adjustment of the salt composition, the reactivity can be kept equal to one. A fraction of salt is periodically withdrawn and reprocessed off-line in order to extract the lanthanides before sending the salt back into the core. The actinides are sent back into the core as soon as possible to be burnt. The rate at which this off-line salt reprocessing is done depends on the desired reactor performance.

2.2. Computational tools and methods

Our numerical simulations rely on the coupling (see Fig. 2) of the MCNP neutron transport code (Briesmeister, 1997) with a home-made materials evolution code REM (Nuttin, 2002; Mathieu, 2005).

The probabilistic MCNP code evaluates the neutron flux and the reaction rates in all the parts (called cells) of the simulated system. This requires a precise description of the geometry and the characteristics of all materials involved (temperature, density, elements, isotopes,

proportions), together with the interaction cross-sections of each isotope constituting the reactor.

These calculations are static, for a given and fixed state of the system. Following the reactor operation all along its life also requires simulating the temporal evolution of the system. The neutronic code thus has to be coupled with an evolution code.
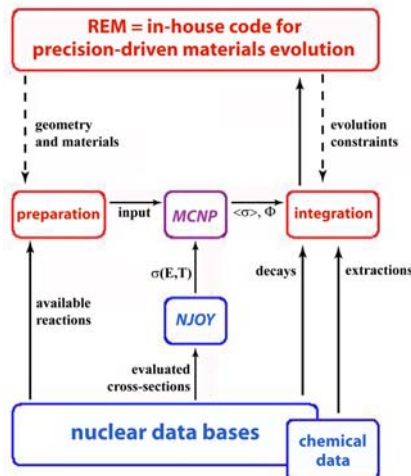

Fig. 2. Coupling of the MCNP neutron transport code with the in-house materials evolution code REM

The evolution code REM solves the Bateman equations to compute the evolution of the materials composition isotope by isotope within the cells as a function of the nuclear reactions and decays occurring in the system and of external parameters like reprocessing or fuel adjustment. These last parameters are implemented through specific removal constants equivalent to decay constants. Our simulations consider several hundreds of nuclei (heavy nuclei, fission products, structural materials…) with their interactions and radioactive decays.

The simulations of reactor evolution take into account the input parameters (power released, criticality level, chemistry...), by continuously adjusting the materials composition and thus the neutron flux of the system, via multiple interactions between the neutronic and the evolution tools. The REM code is indeed a precision-driven code, i.e. it has been designed to determine the reactor evolution while controlling the precision of the results at each step of this evolution. The resolution of the Bateman equations is constrained by several variables to keep the reactor's simulated physical parameters constant during the evolution. These include the total power

(with a one percent or so precision) and the reactivity (with a huge precision of some ten pcm, much smaller than the computational uncertainty of this parameter under MCNP). The numerical integration of the Bateman equations is done using a Runge-Kutta method.

2.3. Systematic studies of the concept

Our studies evaluate many parameters of the reactor, such as the influence of the chemical reprocessing on neutronic behaviour, the burning capabilities, the evaluation of the intrinsic safety level, and deployment capabilities including electronuclear deployment scenarios (Merle-Lucotte et al., 2006, 2007b).
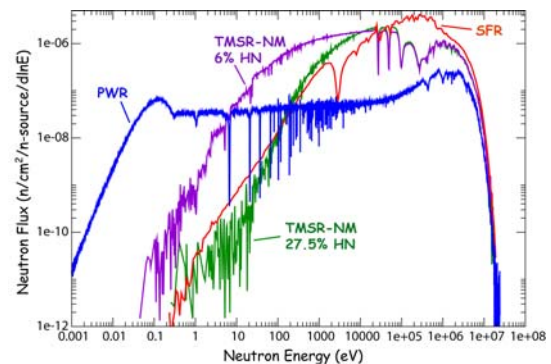

Fig. 3. Neutron spectrum of two TMSR configurations (6% and 27.5% of heavy nuclei) compared to the neutron spectrum in a Pressurised Water Reactor (PWR) and in a Sodium cooled Fast Reactor (SFR)

A systematic study has thus shown that the salt composition plays a role in neutron energy moderation, in actinide solubility, and initial fuel inventory. The role of moderator assumed by the fuel salt in such reactors is illustrated in Fig. 3: the neutron spectrum shifts from epithermal for the smaller heavy nuclei content, to fast for the salts with larger proportions of heavy nuclei.

2.3.1 Breeding capacities

The results of the parametric study are presented in Fig. 4 where the reprocessing capacity required for a TMSR-NM system is shown.

The ordinate is the weight of heavy nuclei reprocessed per day while the abscissa corresponds to the initial fissile (^{233}U) inventory. Heavy nuclei proportions in the MSR fuel are indicated (x %mole HN). For each proportion of heavy nuclei in the salt,

ranging from 6 mole % to 27.5 mole %, we evaluate by simulation the breeding ratio of each reactor configuration as a function of the daily amount of heavy nuclei reprocessed. Under-breeder reactor configurations, which are located under the red line (bottom of the figure), cannot be considered in the frame of Generation IV systems since they do not allow sustainable reactor deployment.
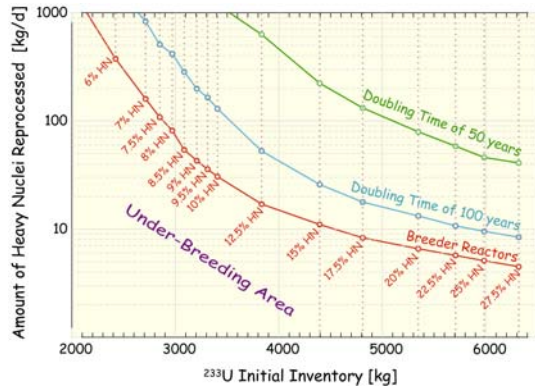

Fig. 4. Amount of heavy nuclei reprocessed per day versus initial fissile (^{233}U) inventory, for different heavy nuclei proportions in the fuel

2.3.2 Feedback parameters

The practical evaluation of the feedback coefficients is done as follows. The multiplication coefficient k is first computed for the core with the matter compositions at equilibrium for a temperature of 900 K. It is then re-calculated using the same compositions but at a different reactor temperature. In practice, the modifications concern the temperature of the salt itself, together with the density of the salt because of its dilatation (dilatation coefficient of 10^{-3} /°C in our case). Other temperature variations like those of the reflectors or the blanket are not considered since these materials have a very small contribution and heat up very slowly.

The total feedback coefficient at equilibrium is displayed in Fig. 5, together with its components, the contributions of the salt heating and salt density, as a function of the HN proportion in the salt. All these safety coefficients are largely negative for all TMSR-NM configurations, ranging from -10 pcm/K to -5 pcm/K.

The uncertainties indicated are a quadratic combination of the statistical and systematic uncertainties. The calculations are precise enough to

lead to negligible statistical errors. Concerning the systematic uncertainties on the contribution of salt heating, the cross-sections concerned are well known, inducing only negligible uncertainties. The uncertainties on the salt density and its dilatation lead to systematic errors less than 20% on the contribution of salt dilatation.
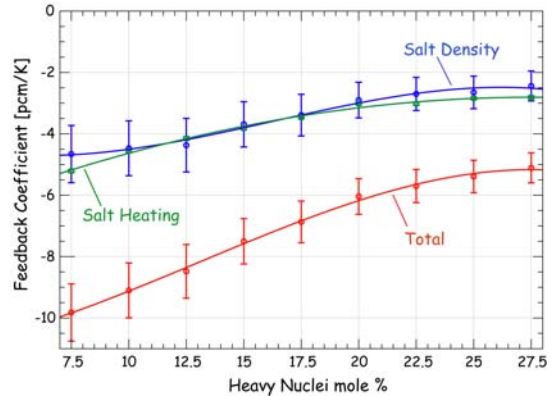

Fig. 5. Feedback coefficients of the TMSR-NM, as a function of the HN proportion in the fuel salt.

2.4. Characteristics of a typical TMSR-NM configuration

The TMSR-NM configurations with large amounts of heavy nuclei in the fuel salt, corresponding to the fast neutron configurations, are particularly interesting, because of their better deployment capacities (Merle-Lucotte et al., 2007b) and because they can be operated without Beryllium, which is known for its high chemical toxicity. As an illustration, we will detail the characteristics of the TMSR-NM containing 22.5%-mole of heavy nuclei. This configuration will be called 'typical configuration' in the following.

2.4.1 Reactor evolution

With such a salt composition, the initial heavy nuclei inventory comprises 5700 kg of ^{233}U and 46 tons of ^{232}Th. Fig. 6 illustrates the evolution of the fuel salt composition all along the operation of this reactor, up to equilibrium. The proportion of minor actinides in the salt remains low, around one percent at equilibrium.

The feedback coefficient of this configuration, equal to -5 pcm/K, is largely negative. A good breeding ratio is obtained with the daily off-line

reprocessing of a quantity of this salt containing 50 to 200 kg of heavy nuclei, i.e. a reasonable reprocessing rate.
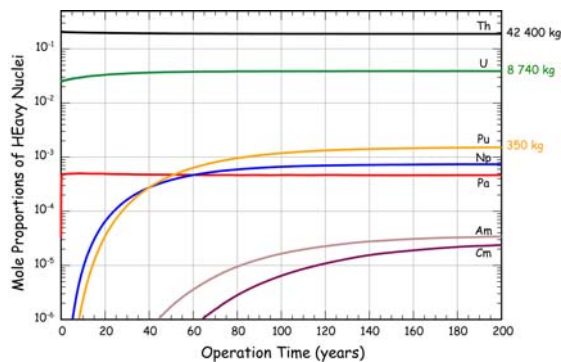

Fig. 6. Heavy nuclei inventory in mole proportions for the [233]U-started TMSR configuration with 22.5 mole% in the fuel salt

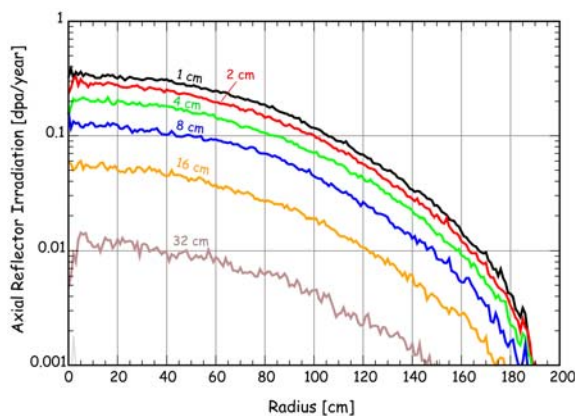### 2.4.2 *Damage to structural materials*

Fig. 7. Radiation damages of the axial reflector as a function of the core radius (zero corresponding to the centre of the core), for different depths of the reflector

Neutron radiations are modifying the chemico-physical properties of the structural materials inside the reactor. These radiation damages in neutron-irradiated materials, dependent on many factors like the irradiation dose and the neutron spectrum, are expressed in dpa (displacements per atom), corresponding to the number of times an atom is displaced for a given fluence. We have calculated these damages for the axial and radial reflectors, which are the most irradiated elements of the core. Figure 7 presents the radiation damages calculated for different depths of the axial reflectors (the damages to the radial reflectors being of the same

order), ranging from 0.01 to 0.4 dpa per year in the middle of the reactor.

3. Simplified design of the TMSR-NM

This concept is really very reliable and robust, allowing further simplifications to make it even more attractive without losing its advantages. A major simplification consists in replacing the fertile blanket surrounding the core by a passive reflector fully made of hastelloy, without any fertile matter inside. A new set of systematic studies has allowed us to evaluate the impact of such a modification on the performances of the reactor, in terms of breeding capabilities and safety performance.
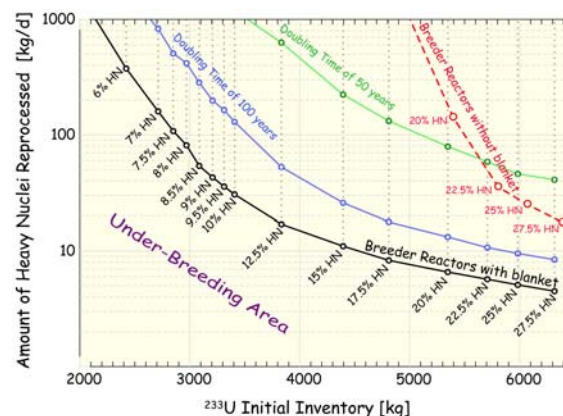

Fig. 8. Amount of heavy nuclei reprocessed per day versus initial fissile ([233]U) inventory, for different heavy nuclei proportions in the fuel, by considering TMSR-NM configurations with and without fertile blanket

The breeding configurations of such a TMSR-NM with no fertile blanket are located on the red curve in figure 8 (upper right third of the figure). The reactor configurations containing proportions of heavy nuclei larger than 20 mole% succeed in producing at least their own fissile inventory while considering realistic reprocessing capacities, i.e. less than some hundred kilograms of heavy nuclei reprocessed per day.

A worldwide nuclear deployment up to 0.7% per year could be achieved using such simplified non moderated TMSRs.

4. Safety studies

Molten salt reactors are based on a liquid fuel, so that their technology is fundamentally different from the solid fuel technologies currently in use. Some of the advantages of the MSR in terms of safety originate directly from this characteristic, during regular operation as well as in accidental situations. These include no possible core melt, absence of initial reactivity reserve, low proportion of fission products in the fuel salt during operation… The risks associated to such reactors have to be assessed, taking into account the really specific features of these systems, for example due to the presence of fissile matter in the pumps and the heat exchangers.

A new safety assessment would require the cross-over of many disciplines like thermal hydraulics, neutronics, material and engineering sciences. Here, we focus on our field of competence: the calculation of the feedback coefficients and delayed neutron fraction, and the impact of these parameters on the reactor's behaviour and its safety parameters.

4.1. Safety parameters and reactivity margins

The fraction of delayed neutrons, β, is very important for reactor stability. The total number of fission product atoms giving rise to delayed neutron emissions will depend on the fissile composition of the reactor and, to a lower extent, on the type of neutron spectrum. We have considered in our study seven precursor families detailed in reference (Merle-Lucotte et al, 2007a). We have considered two fissile nuclei since, in the TMSR-NM, 90% of the fissions are due to ^{233}U and 10% to ^{235}U. Calculating the value of β at equilibrium for a typical TMSR-NM configuration, we have found it to be 450 pcm, one third of the delayed neutrons being emitted outside the core. This leads to a fraction of delayed neutrons inside the core of 300 pcm.

The feedback coefficients have been detailed in the reactor design section: these coefficients are significantly negative for all heavy nuclei proportions, including the density coefficient which can be seen as a void coefficient.

Using a liquid fuel allows the adjustment of fertile and fissile matter without unloading the core, doing away with the need for any initial reactivity reserve, contrary to the case of a light water reactor where this initial reactivity reserve amounts to 10 000 pcm. However, some reactivity margins may be introduced involuntarily in the core of the TMSR through three possible perturbations:

- A direct insertion of reactivity, mainly an unintentional introduction of ^{233}U in core instead of the 2.6 kg/day of ^{232}Th necessary to ensure breeding. With an insertion time of some seconds, this represents 27 pcm of additional reactivity,.
- The loss of salt circulation: this results in 2.55% per day of the ^{233}Pa already produced in the core decaying to ^{233}U, and the decay in the core of all the delayed neutron precursors instead of 2/3 of them during normal operation. The loss of circulation globally represents a reactivity reserve of 60 + 150 = 210 pcm per day.
- The draining of the fertile blanket, either into the core where it mixes with the fuel salt, or outside of the reactor. We have demonstrated by simulation that both accidents lead to a decrease of reactivity.

The draining of the core itself, a first, results in a decrease of reactivity of -2.5 pcm per percent of fuel salt drained, thanks to the negative void coefficient. The total reactivity margin of the TMSR-NM amounts to 250 pcm. We will consider an upper limit of 1000 pcm in the next paragraph.

4.2. Insertion of reactivity transient

We have analyzed how the safety parameters presented previously impact the system after the insertion of reactivity at least equal to the total reactivity reserves of the reactor. This safety analysis implies the definition of a viability domain, corresponding to the range of acceptable core parameters. As the internal pressure is very low in a TMSR, only phenomena resulting from a temperature increase in the fuel salt could endanger the reactor. As a consequence, the viability region is limited by the fuel solidification temperature with $T_{\min} = 800 K$ as lower limit, and by the salt volatilization temperature with $T_{\max} = 1600 K$ as upper limit.

Transient simulations are carried out using a simple mathematical model which includes the following system of three equations of point reactor kinetics with seven groups of delayed neutrons:

$$\frac{\partial N}{\partial t} = \frac{\rho - \beta}{l} N + \sum_i \lambda_i C_i \qquad (1)$$

$$\frac{\partial C_i}{\partial t} = \frac{\beta N}{l} - \lambda_i C_i \qquad (2)$$

$$\frac{\partial T}{\partial t} = (P - P_o)/(C_p d) \qquad (3)$$

With N the neutron flux, t the time since the beginning of the transient, ρ the reactivity, β the proportion of delayed neutrons (300 pcm in the TMSR), l the mean time between two fissions (8.46 µs here), $\lambda_i = \ln 2/(t_{1/2})_i$ where $(t_{1/2})_i$ is the decay period of the delayed neutron precursors of the i$^{\text{th}}$-group, C_i the proportion of delayed neutron precursors of the i$^{\text{th}}$-group, T the mean core temperature (T_o = 900K), P the instantaneous power per cm^3, P_o the extracted power per cm^3 (125 W/cm^3, constant), C_p the specific heat (1.05 J/g/K) and d the density of the fuel salt (4.3 g/ cm^3 for the typical TMSR-NM configuration with 22.5% of heavy nuclei).

The limitations of this model lie in a uniform distribution of the fissions within the core, the lack of heat propagation simulation, and the absence of follow-through of the precursors of the delayed neutrons. Concerning the validity domain of our results, our simulations are reliable for insertion times longer than some ten seconds and are pessimistic for the insertion times shorter than one second, since we consider a uniform power distribution within the core and we do not take into account the time to reach the density equilibrium (equilibrium considered as immediate). Larger local powers are indeed reached within the core, leading to a faster feedback. For insertion times between 0.01 and 10 seconds, things are more complex. Further calculations taking in consideration heat propagation, a realistic distribution of the fissions and the propagation of the precursors would loosen the limits of the kinetic-point model. Because the insertion of the total reactivity margins of the TMSR corresponds to the addition of less than 1000 pcm in more than some minutes, as previously detailed, our model is valid. It is indeed impossible to insert such a reactivity reserve in a shorter time.

We have calculated the insertion of this upper limit of 1000 pcm in ten seconds in the TMSR-NM with 22.5% of heavy nuclei. The impact of this transient on the reactivity, power and temperature of the reactor is presented in Fig. 9 (blue curve) as a function of the time since the beginning of the insertion.

As expected, the final temperature reached at equilibrium is equal to $T_o + \frac{1000\,pcm}{dK/dT} = 1100$ K. The reactivity insertion leads to an increase of the reactor power (Fig. 9, middle) which reaches a maximum at 225 W/cm^3. Finally, the reactor temperature (Fig. 9, bottom) begins to increase after some milliseconds. A significant temperature increase triggers the reactor feedback, and as a consequence, the reactivity and power decrease. Finally, the heat in excess has to be evacuated to return to normal operating conditions. Thanks to its excellent safety parameters, the TMSR-NM succeeds in absorbing at least a 1000 pcm reactivity insertion in one second and behaves safely.
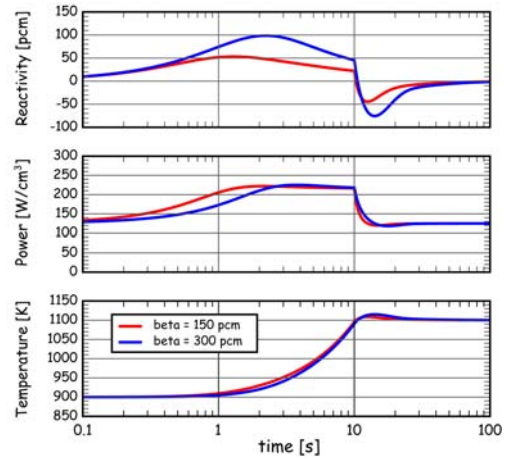

Fig. 9. 1000pcm reactivity insertion (in ten seconds) transient in the TMSR-NM with 22.5% of heavy nuclei with a feedback coefficient of -5 pcm/K and for two different values of the fraction of delayed neutrons

The design of the TMSR-NM is not yet finalized since many studies are ongoing, mainly concerning the thermal hydraulics, heat exchanges and safety fields, the portion of fuel salt in/out the core may vary. For example, it could be necessary to have a up to half of the fuel salt out of the core instead of the 1/3 considered here, depending on the heat exchangers and the energy conversion system. This will significantly modify the fraction of delayed neutrons in the core, and thus impact the safety level of the system. In the case of a smaller fraction of delayed neutrons, for example half of the

previous value (Fig. 9, red curve), we have found that the prompt reactivity regime is reached earlier and the reactor feedback occurs considerably faster, the reactor still behaving safely.

5.   Conclusions

The Thorium Molten Salt reactor (TMSR) presented here without moderator in the core appears to be a very promising, simple and viable molten salt reactor concept. More precisely, the non-moderated TMSR configurations with large heavy nuclei proportions, leading to a fast neutron spectrum, present particularly interesting characteristics concerning their performances in terms of breeding and safety. The main topics that need further investigation to demonstrate the scientific feasibility of the concept deal with the on-line control of the salt composition and of its chemical and physical properties, with experimental validations of materials, and with more complete safety evaluations. The innovative core design and the specificities of MSRs (motion of delayed neutron precursors) call for an analysis of events due to non-conventional perturbations in the reactor's operation.

Such studies are ongoing in the frame of the French concerted research program 'Molten Salt Reactors' (PCR-RSF) and of European programs dedicated to such systems. Finally we want to stress the hardiness and the flexibility of this TMSR-NM concept, allowing it to be adjustable without loosing its advantages in the event of any technological problem or modification, as we illustrate here through the simulation of a possible design simplification. All these properties put the TMSR in a very favourable position to fulfil the conditions defined by the GEN IV International Forum.

6.   Acknowledgement

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