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Molten Salt Reactor: Deterministic Safety Evaluation

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Molten Salt Reactors (MSRs) are one of the systems retained by Generation IV as a candidate for the next generation of nuclear reactors. This type of reactor is particularly well adapted to the thorium fuel cycle (Th-^{233}U) which has the advantage of producing less minor actinides than the uranium-plutonium fuel cycle (^{238}U-^{239}Pu) [1]. In the frame of a major re-evaluation of the MSR concept and concentrating on some major constraints such as feasibility, breeding capability and, above all, safety, we have considered a particular reactor configuration that we call the 'unique channel' configuration in which there is no moderator in the core, leading to a quasi fast neutron spectrum. This reactor is presented in the first section. MSRs benefit from several specific advantages which are listed in a second part of this work.

Beyond these advantages of the MSR, the level of the deterministic safety in such a reactor has to be assessed precisely. In a third section, we first draw up a list of the reactivity margins in our reactor configuration. We then define and quantify the parameters characterizing the deterministic safety of any reactor: the fraction of delayed neutrons, and the system's feedback coefficients that are here negative. Finally, using a simple point-kinetic evaluation [2], we analyze how these safety parameters impact the system when the total reactivity margins are introduced in the MSR. The results of this last study are discussed, emphasizing the satisfactory behavior of the MSR and the excellent level of deterministic safety which can be achieved.

This work is based on the coupling of a neutron transport code called MCNP [3] with a materials evolution code. The former calculates the neutron flux and the reaction rates in all the cells while the latter solves the Bateman equations for the evolution of the materials composition within the cells. These calculations take into account the input parameters (power released, criticality level, chemistry ...), by adjusting the neutron flux or the materials composition of the core on a regular basis. Our calculations are based on a precise description of the geometry and consider several hundreds of nuclei with their interactions and radioactive decay; they allow a thorough interpretation of the results. All the data discussed in this paper result from the evolution of the reactor over 100 years.

1. The Thorium Molten Salt Reactor

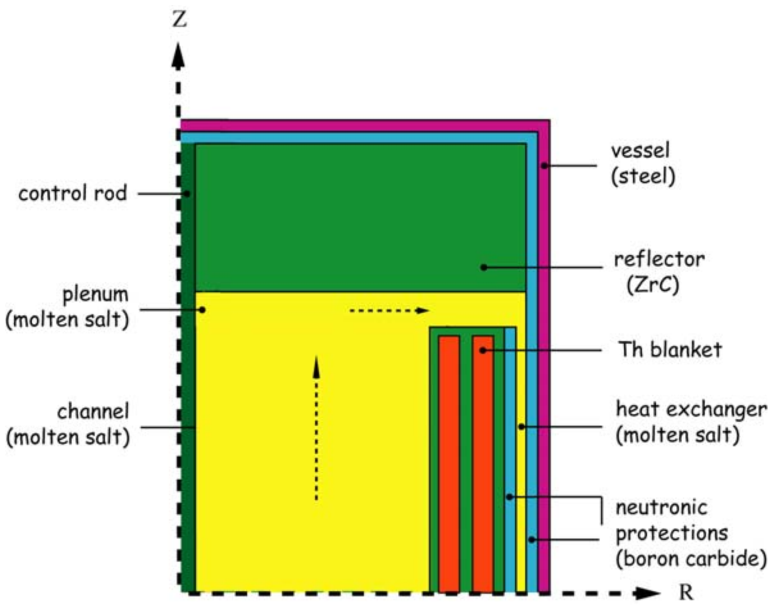

Figure 1: Vertical slice of a quarter of the TMSR

Our standard system, called Thorium Molten Salt Reactor or TMSR [4][5], is a 1 GWe Molten Salt Reactor based on the Thorium/^{233}U fuel cycle, associated to a chemical reprocessing unit, and based on a nearly fast neutron spectrum. Our reactor [6] is composed of a single big salt channel surrounded by a thorium and graphite radial blanket. The axial reflectors are composed of ZrC. One third of the 20 m^3 of fuel salt circulates in external circuits and, as a consequence, outside of the neutron flux. Its operating temperature is 630°C and its thermodynamic efficiency is 40%. A thorium and graphite radial blanket surrounds the core so as to improve the system's regeneration capability. The properties of the blanket are such that it stops approximately 80 % of the neutrons, thus protecting external structures from irradiation while improving regeneration. We assume that the ^{233}U produced in the blanket is extracted every 6 months.

The salt used is a binary salt, LiF – $(HN)F_4$ (where HN stands for Heavy Nuclei), whose $(HN)F_4$ proportion is set at 22 % (eutectic point). This corresponds to an initial fissile material (^{233}U) inventory of 5.3 metric tons. The salt density is set at 4.3 with a dilatation coefficient of 10^{-3}/°C [8].

The salt chemical reprocessing consists in two entities. The first one is a bubbling system in the reactor which extracts quickly the gaseous FPs and the noble metals. The second one is a slower and separate unit that extracts the other FPs. A fluorination removes the Uranium for immediate re-injection in core, and the remaining salt is then cleaned. A salt volume equal to the core volume is cleaned in six months.

2. Intrinsic Advantages of the TMSR

2.1 Differences between Molten and Solid Fuel

MSRs 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:

- Being liquid at the operating temperature, the fuel does not have to be kept under high pressure. It is homogeneous and very stable vis-à-vis irradiation. Any core melt risk is also eliminated.
- An on-line adjustment of the fertile and fissile matters is possible, doing away with the need for any initial reactivity reserve, for a double inventory or for the production of coated solid fuel elements.
- The consequences of coolant loss are mitigated since the fuel itself plays the role of coolant.
- The boiling point for the fluoride salts is 1800 K. In the event of abnormal heating, typically if the temperature increases beyond 1600 K, passive safety is secured thanks to a possible quick flush of the fuel in a separate tank.
- In the MSR configuration considered, the risk of internal structure loss due to graphite burning is eliminated since there is no graphite in the middle of the core. The risk for the graphite to collapse is thus suppressed. Some graphite is present in the Thorium blanket. Although its influence on the reactor is lower here, it may be interesting to replace it by another more resistant material (ceramics, carbides).
- This fuel presents of course some drawbacks, among which the problem of corrosion due to the fluoride salt and the presence of fissile matter in the pumps and the heat exchangers.

2.2 Advantages due to the Th/^{233}U fuel cycle: Resistance to Proliferation

One of the requirements for the next generation of reactors is proliferation resistance. This condition is met by the TMSR thanks to the ^{232}U present in the fuel. This ^{232}U is mainly produced through the (n,2n) reaction on ^{232}Th. This reaction has a high energy threshold and requires fast neutrons. 15 kg of ^{232}U are formed in the core at equilibrium. This results in a total amount in ^{232}U of around 1850 ppm, corresponding to a 2300 $GBq.kg_u^{-1}$ radioactivity in gamma rays of 2.6 MeV. Similarly, 30 g of ^{232}U, corresponding to 620 ppm, are produced in the salt contained in the Thorium blanket [4].

3. Deterministic Safety Parameters of the TMSR

3.1 Fraction of Delayed Neutrons

The fraction of delayed neutrons, β, is very important in reactor control and is an important safety parameter. 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. The value of β at equilibrium for the TMSR considered here is equal to 450 pcm.
We have considered in our study seven precursor families listed in Table 1 [7] for two fissile nuclei, ^{233}U and ^{235}U. Regarding the neutron spectrum of the TMSR, we take a value between those indicated for the precursor's abundances in a fast and in a thermal spectrum. We have considered two fissile nuclei since, in the TMSR, 90% of the fissions are due to ^{233}U and 10% to ^{235}U. The final values used in section 5 for the safety evaluation are summarized at the end of Table 1.

Group	1	2	3	4	5	6	7
Precursor	^{87}Br	^{137}I	^{88}Br	^{93}Rb	^{139}I	^{91}Br	^{96}Rb
Half-Life	55.9 s	24.5 s	16.4 s	5.85 s	2.3 s	0.54 s	0.199 s
Abundances							
^{233}U (fast)	0.0788	0.1666	0.1153	0.1985	0.3522	0.0633	0.0253
^{233}U (thermal)	0.0787	0.1723	0.1355	0.1884	0.3435	0.0605	0.0211
^{235}U (fast)	0.0339	0.1458	0.0847	0.1665	0.4069	0.1278	0.0344
^{235}U (thermal)	0.0321	0.1616	0.0752	0.1815	0.3969	0.1257	0.0270
Mean Value	0.0742	0.1679	0.1209	0.1915	0.3533	0.0684	0.0240

Table 1: Abundances of seven delayed neutron precursors for two uranium isotopes

3.2 Feedback Coefficient

The feedback coefficient or temperature coefficient is the variation of the multiplication coefficient *dk* for a given variation *dT* in temperature of the whole or part of the core. This feedback coefficient has to be negative to ensure the intrinsic stability of the reactor. The practical evaluation of a feedback coefficient is done as follows. The multiplication coefficient *k* is first computed for our core with the matter compositions at equilibrium for a temperature of 900K. It is then re-calculated using the same compositions but at a different reactor's temperature. In practice, the modifications concern the temperature of the salt itself, the temperature of the graphite moderator, together with the density of the salt because of its dilatation (dilatation coefficient of 10^{-3} /°C in our case [8]). Other temperature variations like those of the reflectors or the blanket are not considered since these materials have a very low contribution and heat up very slowly.

The feedback coefficient $\frac{dk}{dT}$ can be broken down into three sub-coefficients related to the different modifications of the core presented above:

$$\left.\frac{dk}{dT}\right)_{total} = \left.\frac{dk}{dT}\right)_{salt\_heating} + \left.\frac{dk}{dT}\right)_{salt\_dilatation} + \left.\frac{dk}{dT}\right)_{graphite\_heating}$$

The third sub-coefficient is negligible in our case since there is no graphite moderator in core. The contributions of the heating of the salt and of its dilatation are of the same order of magnitude, as shown in Table 2. The total feedback coefficient for this TMSR configuration is equal to -5.37 pcm/K.

Total Coefficient	Salt Heating (Doppler)	Salt Dilatation
-5.37 ± 0.04 pcm/K	-3.14 ± 0.04 pcm/K	-2.02 ± 0.04 pcm/K

Table 2: Total feedback coefficient for the TMSR and break down in sub-coefficients [5][6]

The uncertainty on the coefficients comes first from statistical errors which are precisely estimated and also from systematic errors that are not quantified, like the evaluation of the cross-sections for example. Only the statistical uncertainties are given in Table 2. The systematic uncertainties are not precisely known but the value of the final feedback coefficient lies somewhere between -4 and -6 pcm/K.

4. Reactivity Margins in the TMSR

Beyond the safety aspects, 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 current PWR where this reactivity reserve amounts to 10 000 pcm. We detail in this section the reactivity margins of the TMSR which may be introduced in core involuntary because of perturbations of the reactor.

4.1 Insertion of reactivity

The impact of different kinds of reactivity insertions has to be studied. If the adjustment of fissile and fertile matters in the MSR is an important advantage, it may indeed present potential specific dangers which are evaluated below.

We first consider the unintentional introduction of ^{233}U in core. The reactivity increase of the TMSR is equal to 13.6 ± 0.2 pcm per extra kilogram of ^{233}U [4]. For an iso-breeder reactor, the core needs to be fed 2.6 kg/day in ^{232}Th. If ^{233}U is added, instead of ^{232}Th, this would lead to an increase of the reactivity of 35 pcm/day. In parallel, since ^{232}Th is a fertile element, the reactivity decreases by 0.50 pcm per kilogram of ^{232}Th added in core. As a consequence, if the 2.6 kg of ^{232}Th consumed per day are not replaced, the reactivity will increase by 1 pcm per day, which is negligible.

^{233}U is produced in core as follow:

$$^{232}\text{Th} + \text{n} \longrightarrow {}^{233}\text{Th} \xrightarrow{\beta} {}^{233}\text{Pa} \xrightarrow{\beta} {}^{233}\text{U}$$

The radioactive period of ^{233}Pa being 27 days, it decays rather quickly in core - around 2.55% per day - giving ^{233}U. This corresponds to (1) a disappearance of neutron capturing matter and (2) an appearance of fissile matter. If the reactor were to stop operating, the first effect would increase the reactivity by
33 ± 11 pcm/day, the second effect by 27 ± 1 pcm/day, giving a total reactivity margin of 60 ± 11 pcm/day.

Finally, the total reactivity margin due to feeding mistakes or uncontrolled ^{233}Pa decay is equal to
96 ± 11 pcm/day. The TMSR core is only moderately sensitive to these effects, since it contains a large amount of fissile matter compared to the amount consumed per day.

4.2 Loss of Salt Circulation

As detailed in section 3.1, the value of β for the whole salt at equilibrium is equal to 450 pcm. As two thirds of the salt is in the core and one third outside in the heat exchangers during normal operation, only 300 pcm of the delayed neutrons are emitted in core. If the salt circulation is stopped, all the delayed decays will occur in the reactor, since all the fission products will stay in core. This will represent an addition of 150 pcm to the multiplication coefficient in some ten seconds.

4.3 Movement of the Control Rod

As shown on figure 1, the hexagon in the centre of the core does not contain fuel salt but represents a central control rod. In this paragraph, we study the impact of the movements of this control rod on the global reactivity of the reactor. Since things are not definitely set in the TMSR, two cases are considered: a graphite control rod and a ZrC control rod [4].

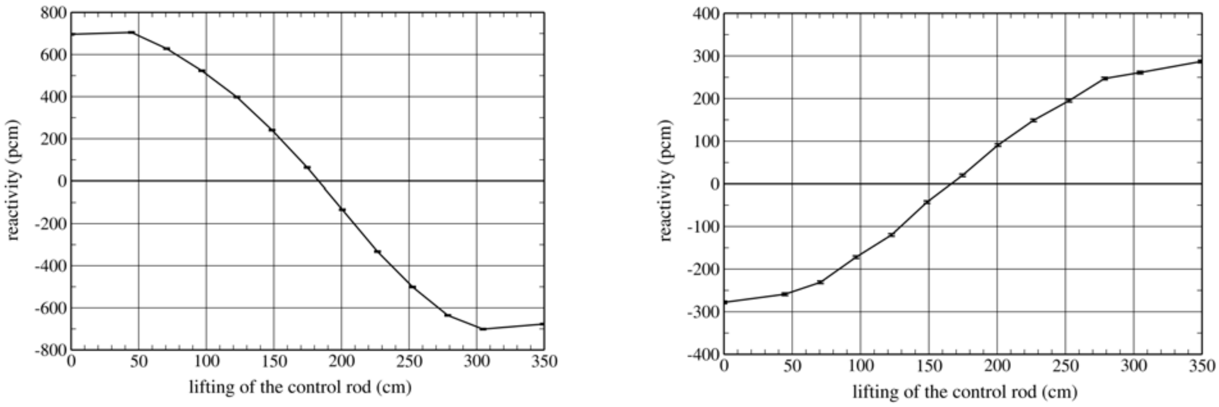

Figure 2: Impact of the lifting of the graphite (left) or ZrC (right) control rod on the reactor's reactivity

The left part of figure 2 illustrates the effect of the ascent of the graphite control rod on the reactivity. The reactivity decreases when the control rod is extracted out of the core, to reach a maximum at -1400 pcm. In the middle of the core, the variation of reactivity reaches -7.6 ± 0.2 pcm/cm. This evolution originates from the high moderating power of the graphite, which increases the fission cross-sections near the control rod. The extraction of the rod entails a slight hardening of the neutron spectrum resulting in a reactivity decrease. Since graphite has a lower density than the salt (1.86 for graphite and 4.3 for the salt), the control rod will spontaneously be extracted by gravitational force, leading to a drop of reactivity. If the control rod is inserted in the core by accident, the velocity of the movement will be around 1cm/s, which corresponds to an increase of reactivity of 7.6 pcm/s. The total insertion of the control rod requires 3 mn for the total reactivity margin of 1400 pcm.

The evolution of the reactivity as a function of the lifting of the ZrC control rod is shown on the right part of Figure 2. In this case, the reactivity increases when the control rod is extracted from the core, to reach a maximum at 560 pcm with a maximal reactivity variation of +2.7 ± 0.2 pcm/cm. Compared to the graphite, the ZrC has a lower power of moderation and much higher neutron capture cross-section. The extraction of the control rod is followed by a minimization of the parasitic captures, increasing the reactivity of the system.

This time, the ZrC has a higher density than the salt (6.73 for the ZrC and 4.3 for the salt). This means that the control rod will spontaneously be inserted under the gravity effect, which leads to a decrease of the reactivity. If the control rod is accidentally extracted, its velocity is still 1 cm/s, which corresponds here to a 2.7pcm/s addition of reactivity.

4.4 Draining of the Core

As mentioned in section 2.1, the reactor is equipped with a draining system through stoppers made of solid salt and designed to melt at a given temperature. It is thus important to check that the salt evacuation will not worsen an accident, since it occurs mainly in abnormal situations.

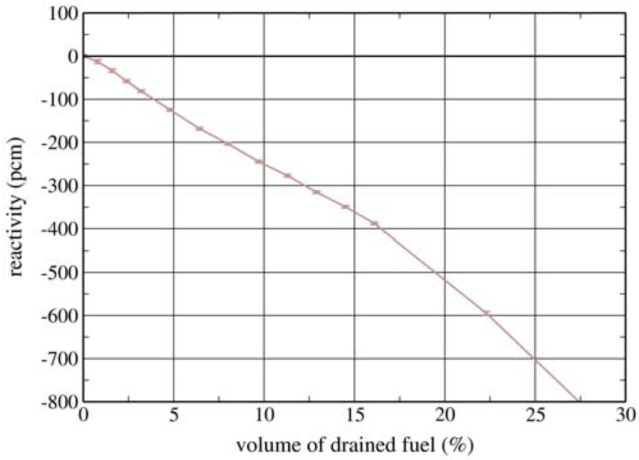

Figure 3: Draining of the Core

As indicated on figure 1, the axial reflectors are made of ZrC and not graphite. This improves the distribution of fissions in the reactor. As shown on Figure 3 which plots the variation of the multiplication coefficient as a function of the fraction of salt volume extracted, the effect of draining is to decrease the reactivity, i.e. it does not generate a positive reactivity margin.

4.5 Loss of the Thorium Blanket

Our reactor is surrounded by a thorium blanket so as to optimize the breeding ratio of the system. The structure of this blanket is made of graphite, containing a salt composed of LiF (78%) – ThF_4 (22%). This salt may be solid or liquid. The second solution simplifies the recovery of the fissile matter but the blanket could then be accidentally drained. If the salt is circulating in a unique channel, the draining might be complete. The extraction of the entire blanket leads to a 500 pcm increase of the reactivity [4]. The reactivity increase due to the draining of the blanket comes from the matter constituting the structure, the graphite. In normal operation, neutrons entering the blanket are captured by Thorium. If the fertile salt is no longer present, these neutrons are only moderated in the graphite and then return to the core.
The blanket could also be composed of several independent vials of salt; an involuntary draining of the blanket will then be partial. If only one channel or vial is emptied, the multiplication coefficient variation is only of the order of a few pcm and is thus negligible.

4.6 Final Result

Table 3 summarises the reactivity margins detailed in the previous paragraphs.

Origin	^{233}U Addition	^{233}Pa Decay	Salt Circulation	Control Rod	Core Draining	Blanket Draining
Reactivity Margin	35 pcm	60 pcm	150 pcm	1400 pcm	-	500
Time of Evolution	Some s	1 day	Some 10 s	Some mn	-	Some mn

Table 3: Reactivity margins of the TMSR

5. Deterministic Safety Evaluation of the TMSR

Using a single kinetic-point evaluation presented in sub-section 5.1, we analyze how the safety parameters presented above impact the system when the total reactivity margins are simultaneously introduced in the TMSR. Deterministic safety implies the definition of a viability domain, corresponding to the range of acceptable core parameters. As internal pressure is very low in a TMSR, only phenomena following a temperature increase of the fuel salt could endanger the reactor. As a consequence, the viability region is limited by the fuel solidification temperature with $T_{\min} = 800K$ as lower limit, and by the salt dissociation temperature with $T_{\max} = 1600K$ as upper limit.

5.1 The kinetic-point model

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

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

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

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

where N represents the neutron flux within the core,
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),
λ_i : $\ln 2/(t_{1/2})_i$ where $(t_{1/2})_i$ is the decay period of the delayed neutron precursors of the i^{th}-group,
C_i : the proportion of delayed neutron precursors of the i^{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),
d : the density of the fuel salt (4.3 g/ cm^3).

This kinetic-point model implies:

- A uniform distribution of the fissions within the core
- No heat propagation
- No follow-through of the precursors of the delayed neutrons

The goal of this section is to give an idea of the reactor's global behavior during accidental transients due to reactivity insertion or heat evacuation loss. Further calculations taking in consideration heat propagation, a realistic distribution of the fissions and the propagation of the precursors would lift the limits of the kinetic-point model.

5.2 Safety evaluation of the reference configuration

From Table 3, we can conclude that the insertion of the total reactivity margins of the TMSR corresponds to the addition of around 2000 pcm in some minutes (around 100 seconds). This insertion of 2000 pcm, in 0.001 to 100 seconds, is thus displayed on figure 4 in terms of temperature, power and reactivity evolutions. The calculations have been done using a feedback coefficient equal to -5 pcm/K, close to our reactor's coefficient value.

As expected, the final temperature reached at equilibrium is equal to $T_o + \frac{2000\,pcm}{dK/dT}$ = 1300 K.

We can conclude from Figure 4 first that the prompt criticality is reached for an insertion time shorter than one second, since the maximal reactor reactivity is higher than the fraction of delayed neutrons. If we detail for example the insertion time of 1 ms (figure 4, black dashes), we first have to consider the evolution of the reactivity (figure 4, top) since we modify it directly. In this case, the whole 2000 pcm reactivity is added before the reactor begins to react. The reactivity insertion then leads to an increase of the reactor power (figure 4, middle) which reaches a maximum at 1 MW/cm^3. Finally, the reactor temperature (figure 4, bottom) begins to increase after some milliseconds. A significant temperature increase triggers the reactor feedback, and as a consequence reactivity and power decrease. The heat in excess has finally to be evacuated to recover normal operating conditions.

However, even in the case of the prompt reactivity regime, a huge and dangerous increase of temperature could be avoided thanks to the reactor's safety parameters, as seen for the insertion time of 0.1 second. In this case (figure 4, green curves), the thermal feedback is fast enough to lower the reactivity back to zero before the end of the 2000 pcm reactivity insertion. A bounce could thus be observed, especially on the power evolution, when the remaining reactivity margins are inserted. A second feedback leads to the final reactivity and power decreases, and normal running conditions are reached again when the accumulated heat is evacuated.

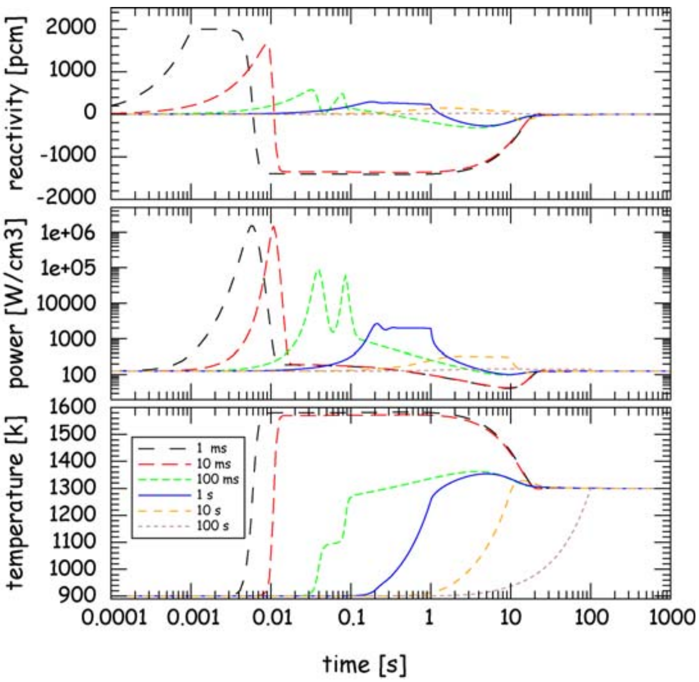

Figure 4: Evolution of the reactivity, the reactor's power and the salt temperature following a 2000 pcm reactivity insertion for a feedback coefficient of -5 pcm/K

For insertion times greater or equal to one second, and a fortiori in the realistic case of 100 seconds, the reactor feedback is fast enough to avoid the prompt reactivity regime. Thus the reactor succeeds in absorbing a 2000 pcm reactivity insertion in one second and behaves safely.

Concerning the validity domain of our results, our simulations are reliable for the longer insertion times and are pessimistic for the shorter insertion times, since we consider a uniform power distribution within the core and we do not take into account the time to reach the densities equilibrium (equilibrium considered as immediate). Larger local powers are indeed reached within the core, leading to a faster feedback to reactivity insertion than shown on figure 4. For the insertion times between 0.01 and 10 seconds, things are more complex and precise calculations have to be done to confirm our results.

We now study the reactor's behavior following a reactivity insertion as a function of the feedback coefficient value and with a simultaneous complete or partial loss of the heat evacuation.

5.3 Parametric study of the deterministic safety

As detailed in section 3.2, the feedback coefficient value of our system lies between -4 and -6 pcm/K when considering the total errors on its determination. The evolution of the reactor to a reactivity insertion of 2000 pcm in one second is displayed on figure 5 (left) for different values of the feedback coefficient. The reactor's behavior is safe for a feedback coefficient better than -4 pcm/K.

A significant temperature increase could also lead to the loss of some heat exchangers. The influence of such a complication during the accidental transient is studied on figure 5 (right). The remaining extracted power decreases from 100% (normal operation) to 5% (residual evacuation with no heat exchanger). This corresponds to having, during some seconds, more power produced in the reactor than during normal operation, together with a lower extracted power.
With a 2000pcm reactivity insertion within 1 second, the situation is dangerous if more than three quarters of the heat exchangers are lost, i.e. if only 25% of the power extraction capability is still available: the temperature increases too much because the power produced is larger than the power extracted. Moreover, in this case, the time left for an active feedback (like the draining of the core) is not sufficient. However, in the more realistic case of a reactivity insertion in 100 seconds represented by the thick purple line on figure 5 (right), the loss of the heat exchangers doesn't lead to a catastrophic situation.

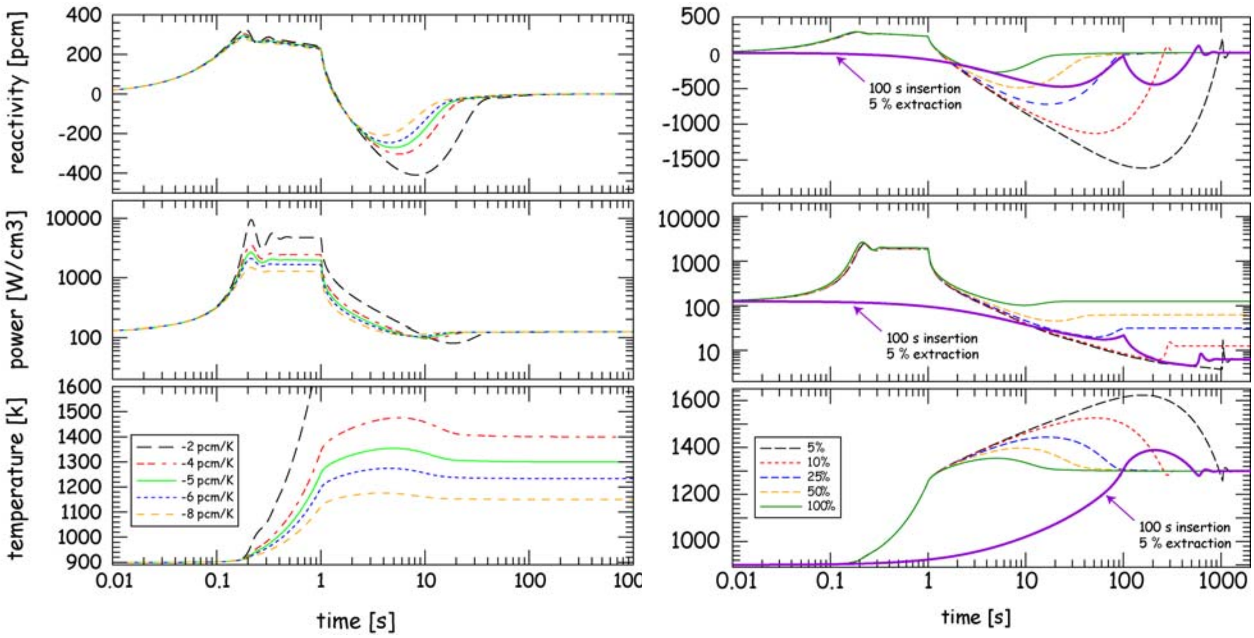

Figure 5: Evolution of the mean reactor's reactivity, the reactor's power and the salt temperature following a 2000 pcm reactivity insertion within 1 second (black, red, green, blue and orange curves) or within 100 seconds (purple curves), for several feedback coefficient values (left) and for several extracted power values using dK/dT = -5 pcm/K (right)

Conclusions

In the frame of a major re-evaluation of the MSR concept, we have considered a particular Thorium Molten Salt Reactor (TMSR), in which there is no moderator in the core, leading to a quasi fast neutron spectrum.
Beyond the classical advantages of the MSR, our goal was the evaluation of the deterministic safety level of such a reactor. We have first reviewed all the sources of reactivity margins of our system: a maximum of 2000 pcm could be accidentally introduced in the reactor in around 100 seconds, mainly due to the loss of salt circulation, the movement of the control rod and the draining of the fertile blanket. The safety parameters of the TMSR are then quantified, leading to a feedback coefficient of -5 pcm/K.

Using a simple kinetic-point model, we have analyzed the reactor's behavior when the total reactivity margins are introduced in the TMSR. For insertion times greater or equal to one second, and a fortiori in the realistic case of 100 seconds, the reactor succeeds in absorbing the reactivity insertion and behaves safely.
In the case of a simultaneous loss of the heat exchangers during the reactivity insertion, a dangerous increase of temperature is observed, except for the longer insertion times which correspond to the more realistic situation.
The results of our studies confirm the satisfactory behavior of the TMSR and the excellent level of deterministic safety which can be achieved in such reactors.

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