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Core severe accidents with cermet fuels – a specific study for pressurized water reactors

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Abstract

Cermet fuels, as a result of the specificity of composites whose components have highly varied thermal-mechanical properties, present very different types of behaviour with respect to transients and accident conditions. In the first part of this paper, an analysis is made of the prospective behaviour of such a composite under accident conditions. The second part focuses on the determination of orienting parameters that could effect a possible return to the criticality of the corium. © 1999 Elsevier Science B.V. All rights reserved.

1. Introduction

The French nuclear power plants (NPPs) currently comprise 55 pressurized water reactors (PWRs), 20 of which are authorized to use uranium/plutonium mixed (MOX) fuel for one-third core loading. The delay in the implementation of the fast breeder reactor development programme has resulted in the accumulation of large stocks of plutonium. The storage capacities of the NPPs and the reprocessing centres of La Hague and Marcoule, although not saturated, are overloaded. In addition, the French utility, EDF ('Electricité de France'), requires COGEMA (the French fuel cycle operator) to reprocess only the quantity of irradiated fuel needed for the fabrication of the MOX fuel authorized to be loaded into the reactors.

A certain number of studies [1–5] have resulted in the definition of cermet (CERamic-METal) fuels, with the aim of increasing the fraction of plutonium fuel that can be loaded into PWRs and reducing conversion by elimination of ^{238}U at least in the cermet rods or in the metallic matrix replacing the uranium. The physical characteristics of this type of fuel are sufficiently different from those of classic uranium oxide fuel (UO_2) to require giving specific thought to the severe accidents liable to affect this composite.

2. The specificity of cermet

The idea of making a composite in which the fissile ceramic is closely bonded to a metallic matrix is underpinned by two objectives related to the metallic nature of the matrix. Metal is an efficient barrier against the diffusion of gases (except, perhaps, with respect to helium, which is not a major problem here). In this case, improved retention of fission gases would be a favourable factor for increased burnup [6], and, in addition, metals have much better thermal conductivity than that of ceramics. Simple studies have shown that, if the proportion of fissile ceramic does not exceed 60% of the volume of the compound, the latter's thermal conductivity is still closer to that of a metal than of a ceramic [7]. For further research, it was decided, rather arbitrarily, to associate equal quantities of the two components. The improvement in thermal conductivity resulted in a much lower operating temperature than in the case of UO_2. Thus, the first series of irradiation tests using the TANOX device in the SILOE reactor (Grenoble) [8,9] showed that, with an Mo–UO_2 cermet, the core temperature was 800 K (530°C) for a power of 350 W cm^{-1}. Consequently, under PWR conditions, a temperature of about 690 K (420°C) would be expected,

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which has been confirmed by our Russian colleagues [10].

Studies of a neutronic character subsequently led to a definition [4,11] of potential candidate metals for the fabrication of the matrix: these were Zircaloy4 (Zr4), which is also the cladding metal, and an aluminium (88%) and silicium (12%) alloy (% in volume), silumin, in view of its transparency to neutrons and its very good thermal conductivity (five times greater than that of Zr4) (Table 1).

Although the melting temperature of uranium oxide is about 3100 K (for a fresh fuel), and its operating temperature about 1273 K, there remains 'a melt margin' of about 1827 K. If we make highly simplifying assumptions, the failure of the fuel would be total in the event of melting, and this molten fuel would mix with the melted metallic parts of the cladding and structures (corium). In the case of a cermet, the problem is very different, since the oxide retains its melting temperature (3100 K), but the metallic matrix has a much lower melting temperature ranging from 1873 to 2093 K. In view of the reactor operating temperature of 420°C, the 'melting margin' of the matrix (which leads to the failure of the rod, but not of the fuel) is the same as, if not better than, in the previous case. However, the kinetics, the scenario and its consequences (as well as the final state) are not the same in the two cases. The matrix and its cladding would fail and melt, but the ceramic of the fuel would preserve its integrity (a margin of about 1300 K). It can be assumed that the molten metal would carry part of the structures along with it and to avoid this, it would be advisable to develop an 'early melting' concept for the matrix by further reducing its melting point and seeking an eutectic in the region of 1373 K with an additive. The melt margins would thus be reduced, but with these 'low' temperatures, the kinetics of structure and vessel aggression would be slowed (the melting temperature of the vessel is about 1623 K). In addition, by remaining at these temperature levels, the kinetics of the very exothermal Zr oxidation reaction, which induces an acceleration of UO_2 melting in the classic case, would be considerably slowed since it accelerates significantly around 1273 K and becomes explosive around 1873 K (it should be noted, however, that the specialists do not at all agree on the same values for these temperature thresholds). As a result, the hydrogen hazard could be managed using conventional methods (ignitors, recombiners, etc). It is obvious that in the case where the melting of the matrix is favoured, it would be necessary to ensure a rapid flow of the metal towards the lower part of the core and to avoid V- or U-shaped structures that could retain the metal and the ceramic particles.

At this stage of reflection, and always on the basis of very simplistic scenarios, the question of the future of this ceramic can be posed. The molten metal would carry along intact spheres, which, a priori, would only release very small quantities of gas, towards the lower part of the vessel where they would form a metallic pool in which the ceramic spheres would be dispersed. The melted metal has a density of about 6 g cm^{-3} for Zr4, whereas, the fuel has a density of about 10 g cm^{-3}, three assumptions should therefore be examined:

(a) through gravity, the spheres fall to the bottom of the pool and there is a total segregation of the two phases (probable);

(b) the mixture, due to convection currents, remains homogeneous or near-homogeneous (a stage of this type will certainly occur, even if the final state is probably that described in (a));

(c) the convective currents are so strong that the spheres, although denser, float to the surface (magmatic currents where dense rocks float to the surface of a less dense magma; however, gas release plays an important role in this type of phenomenon, a common place example is dense effervescent medicine that floats to the surface of a glass of water), an unlikely scenario that could perhaps occur during a transient, since this case does arise in nature.

In these three cases, since melting is favoured, a corium catcher should be provided at the bottom of the vessel, to be cooled, preferably, by a redundant circuit independent from the primary system.

It is clear that, according to the hypothesis selected, the sizing and design of such a corium catcher will be

Table 1
Some characteristics of candidates as metallic inert matrix

	T Melt. (K)	Density (kg m^{-3})	Thermal Cond. (W m^{-1} K^{-1})				c_p (J kg^{-1} K^{-1})
		293 K	293 K	373 K	673 K	873 K	293 K
Mo	2850	9590	138	135	123	116	255
Al	940	2710	236	240	228	215	902
SiAl	–	2260	162	173	180	–	871
Zr4	2120	6570	23	21.8	21.4	22.3	276
Inox (304)	1680	7820	15.2	16.6	20.8	23.5	460
UO_2	3100	11 000	5	–	–	3	235

Table 2
Volumes and masses of the corium: UO_2 and cermet cases

Components		Standard 900 MWe		Cermet 900 MWe	
Region	Material	Mass (kg)	Volume (m^3)	Mass (kg)	Volume (m^3)
Fuel	U/MOX/Pu	88 400	8.8	44 200	4.42[a]
Matrix	Zr4	0	0	26 500	4.42[a]
Cladding	Zr4	16 300	2.72	16 300	2.72
Structures	Steel total	84 000	12	84 000	12
Lower part	Assumed to melt	26 900	3.84	0	0
Corium		13 1600	15.4	87 000	11.6

[a] Since the cermet is a 50/50% volumic fraction.

very different. In case (a), the decay heat will be concentrated into a small space (the bottom of the vessel), which will contain the fissile ceramic, therefore 50% of the volume compared to classic UO_2, i.e. a power density that is twice as large. In case (b), the distribution being homogeneous, the power density is the same as in the classic case, although it should be noted that as a result of the low temperature, the structures will not be carried away and the volume of the corium will be smaller, hence a slightly higher density. In case (c), the spread of the heating material would permit better cooling by radiation, since the surface area would be greater than in (a) and the material more concentrated than in (b). However, hypothesis (a) gives rise to a certain number of concerns, since, as 50% of the volume of cermet fuels is in an inert matrix, the fissile material content has to be increased in order to obtain the same cycle time. If segregation of the phases occurs at the bottom of the vessel, there is a considerable risk of recriticality, particularly with a high content of plutonium fuel (Table 2).

Table 3 gives some approximate dimensions of the final geometry, as regards the chosen hypothesis. The magnitude order of the thickness of the different zones compared to the neutron mean free path well explain the core catcher and borated water inefficiency. Then it becomes necessary to define a core catcher which splits the corium into subcritical volumes between two very absorbing ceramic walls (see Fig. 1 the different configurations).

Table 3
General data for (a) cermet UO_2 and cermet MOX corium, (b) cermet PuO_2, Zr4 corium

	Cermet UO_2, Cermet MOX corium	Cermet PuO_2, Zr4 corium
Corium vol.	11 m^3	11 m^3
CER (fuel) vol.	4 m^3	0.3 m^3
MET vol.	7 m^3	10.7 m^3
Heterogeneous down		
Fuel elevation	92 cm	22 cm
Metal thickness	58 cm	129 cm
Heterogeneous up		
Fuel elevation	40 cm	2.3 cm
Metal thickness	111 cm	148 cm

3. Corium criticality

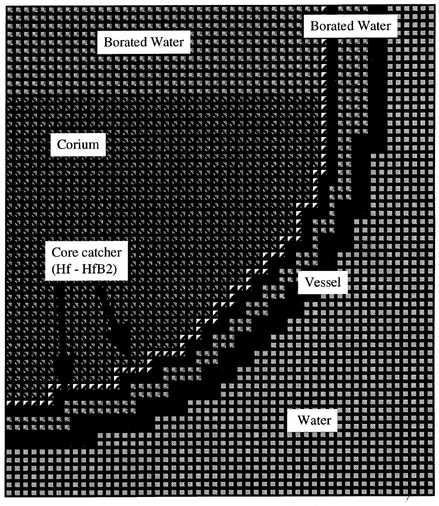
The possibility of reaching a critical configuration led us to carry out a neutronic study. The aim here is not to perform reference calculations, but to analyse the evolution of the corium, according to a few possible scenarios and to examine the main lines of system in response to the disruptions brought about by the melting of the core.

The three hypotheses described above are analysed here:

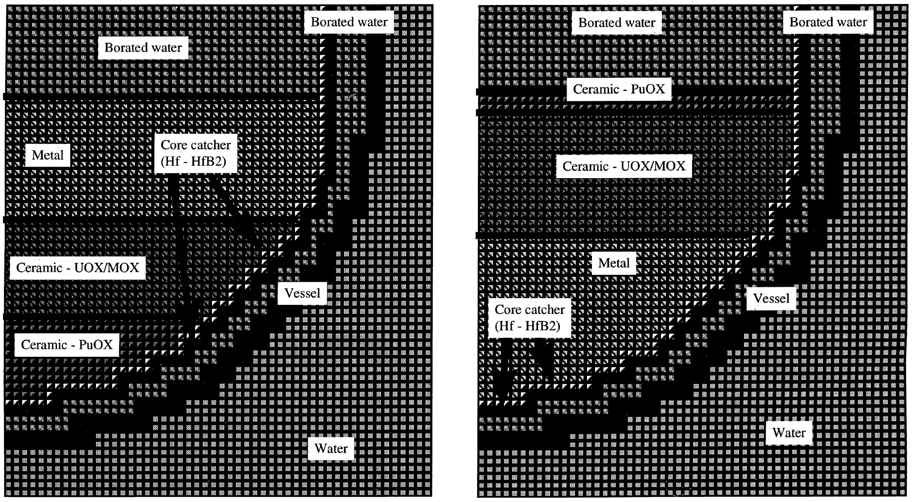
corium with segregation of the phases, the ceramic phase fills the bottom of the vessel (or corium catcher). The density of the fuel is assumed to be equal to that of the maximum stack of spheres with a diameter of about 100 μm. The filling of interstices by the molten metal is not taken into account (it should be noted, however, that in the case of an absorbent metal, such as Mo or steel, the proportion of matrix bonding the ceramic of the corium would be a factor reducing the reactivity). The molten metal covers the ceramic. The volumes taken into account are those encompassing all the ceramic of the fuel, and all the metals of the cladding and the matrix (100% core melting). The overall temperature is assumed to be 1473 K, which corresponds to the higher limit of the cross-section tabulations for the isotopes of the fuel (at this temperature, the cladding is assumed to be melted; this means that the alloy has been 'spiked' to obtain a lower melting point), (heterogeneous down configuration, 'Het.down'),

homogeneous corium, (homogeneous configuration, 'Hom'), and

inverse segregation of the phases, metal at the bottom and ceramic at the surface, (heterogeneous up configuration, 'Het.up').



Homogeneous



Heterogeneous down

Heterogeneous up

Fig. 1. Three corium modelizations.

In the first stage, the calculations were performed for cylindrical geometry dimensions, then generalized to two RZ dimensions in order to take into account the geometry at the bottom of the vessel, using an SN method (APOLLO2 using JEFF 2.2 library). The bottom of the vessel is considered either with a corium catcher made of hafnium di-boride (HfB_2), which is a refractory ceramic and very absorbent (efficiency similar to that of B_4C), or without a corium catcher.

In both cases, reflooding is considered:

with primary system, water at 573 K (300°C) and with 1200 ppm of boron, (borated water: B-water),
with water at 573 K (300°C), but without boron,
without water (vacuum), and
with highly borated water at 406 K (133°C).

3.1. PuO_2, Zr4

The mass composition of the corium is 5% PuO_2, 93% Zr4 and 2% Er_2O_3; this fuel corresponds to a 100% IMF (Inert Matrix Fuel) core for three cycles of 12 months.

Tables 4 and 5 show that, in the case of total draining, supercriticality is very high for the two heterogeneous configurations, particularly for the most likely configuration involving ceramic at the bottom. The homogeneous configuration is supercritical, but only just, which, considering the very simplified calculations performed here, can be considered as not posing a problem. If the case of reflooding with highly borated water is considered, it can be observed that the homogeneous

Table 4
Reactivity of the core after melting (%) (corium without core catcher)

	Void (%)	5000 ppm B-water $T = 406$ K (133°C) (%)
Hom	+6.1	+0.5
Het.up	+21.9	−22.6
Het.down	+54.0	+53.9

Table 5
Reactivity of the core after melting (%) (corium with core catcher)

	Void (%)	5000 ppm B-water $T = 406$ K (133°C) (%)
Hom.	−0.3	−0.9
Het.up	+16.5	−9.1
Het.down	+53.3	+53.3

Table 6
Reactivity of the core after melting (%) (corium with water and slightly borated water)

	Water (300°C) (%)	1200 ppm B-water. $T = 573$ K (300°C) (%)
Hom.	+0.6	−0.3
Het.up	+11.4	+1.4
Het.down	+53.3	+53.3

Table 7
Reactivity of the core after melting (%) (corium with core catcher, 8% Pu fuel)

	Void (%)	5000 ppm B-water $T = 406$ K (133°C) (%)
Hom.	+34.5	+34.3
Het.up	+46.9	+37.6
Het.down	+57.6	+57.0

configuration remains supercritical (this is not significant; in a preliminary approach, this configuration can be said to be only slightly sensitive to reflooding). On the other hand, the differential effect is very strong for the two heterogeneous configurations: the reflooding effect is nil in the ‘ceramic at the bottom’ (down) configuration, but very strong in the ‘ceramic at the surface’ (up) configuration. This can be accounted for by the fact that, in view of the volumes involved and the different distances, the mass of Pu is supercritical in the rapid spectrum at the bottom of the vessel and that the neutrons in this configuration do not ‘see’ the borated water ‘very far above’, whereas in the ‘up’ configuration, the whole surface area of the fuel is in direct contact with the highly poisoned water.

Table 5 demonstrates the efficiency of the corium catcher. It is significant, although low, for both the homogeneous configuration and up heterogeneous configuration as the neutrons are highly likely to encounter the absorbent material, whereas in the ‘down’ heterogeneous configuration, for the same reasons as above, the crucible is neutronically inefficient.

Table 6 presents the results for non-borated water and primary system water (1200 ppm), while Table 7 presents the results for the case of vacuum and water at 5000 ppm, for a Pu at 11.8% mass (8% in volume), which is the fuel used for an 18 month cycle.

The results are not very encouraging, especially for the down configuration, which is the most probable, and therefore brings up the question of redefining the core-catcher crucible, whose principle is the splitting of the critical mass, which, on the one hand, reduces the critical mass by geometrical division, and, on the other, increases the surface area in contact with the absorbent material to permit increasing the capture cross-section. This new configuration is presented in Fig. 2.

3.2. UO_2, Zr4

Table 8 without ambiguity shows in the most penalizing case, i.e without core catcher, without highly borated water, the UOX cermet (50/50% with 7.5% ^{235}U) is always subcritical.

3.3. MOX, Zr4

In the case of a cermet MOX (18% Pu content) devoted to 18 months' cycle, we observe an important positive reactivity for all the final corium configurations. The positive reactivity is nevertheless lower by a factor of two than in the Pu case (Table 9). Here again, the core catcher and the highly borated water injection do not affect the reactivity in an important way (Table 10).

4. Conclusions

Cermet fuels have potential advantages related to the fact that they enable very high burnups as a result of their good thermal conductivity, which gives them a very low operating temperature compared to standard ceramic fuel. However, the behaviour of this composite under accident conditions could lead to difficult situations to model, as experience in this domain is lacking. Using a few simplifying ‘common sense’ assumptions, we have shown that, depending on the possibilities envisaged, their behaviour is highly varied and leads to classes of problems belonging to quite different areas of

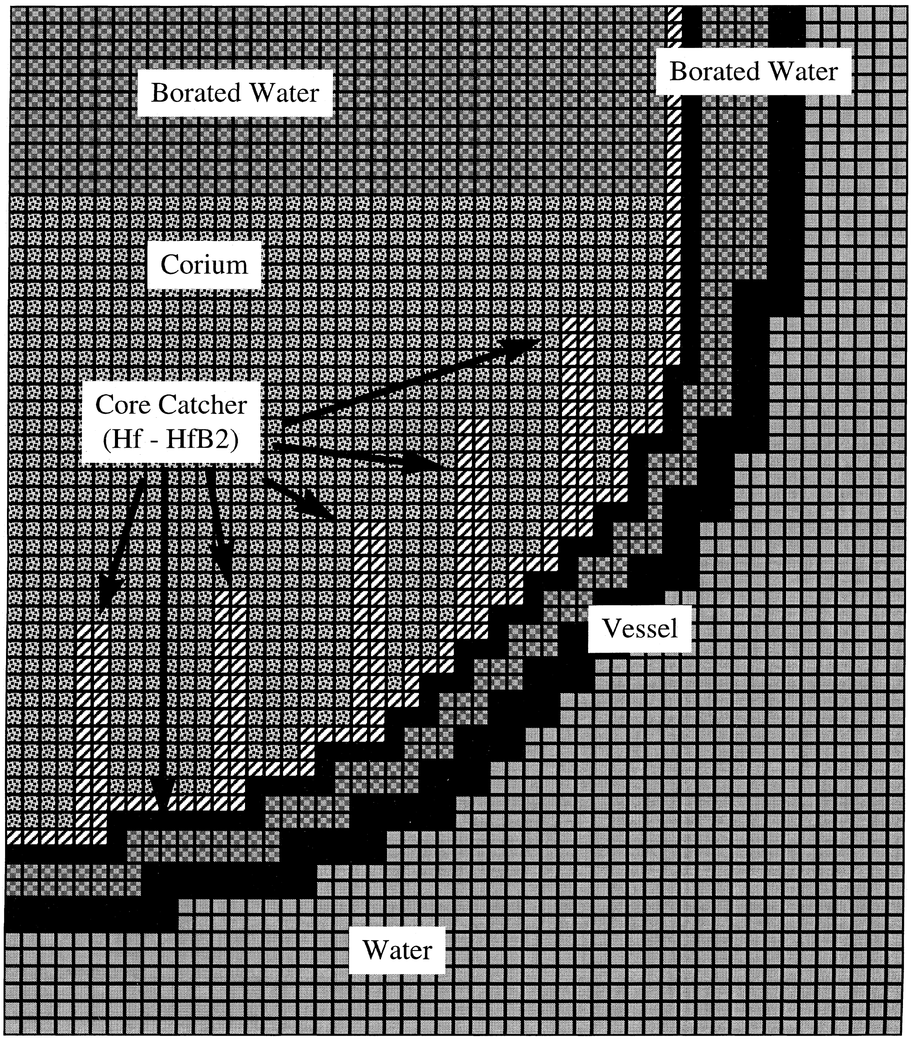


Fig. 2. New core-catcher configuration.

physics. Thus, in order to improve our understanding in this field, a theoretical and experimental programme will have to be conducted with respect to the materials and the behaviour of a composite corium. As regards metallurgy, research aiming to perfect metals having the desired characteristics for the matrices would be welcome. As regards re-criticality of the corium, we have shown that the assumptions made on the phases obtained weigh greatly on the consequences of a melting accident. However, the main areas of sensitivity were highlighted during this study, and various accident management concepts using a corium catcher appropriate to the problems posed, together with reflooding management using borated water, were also refined. All

Table 8
Reactivity of the core after melting (%) (corium cermet – UO_2 (7.5% ^{235}U) without borated water and without core catcher)

	Void (%)
Hom.	−37.1
Het.up	−15.7
Het. Down	−10.5

Table 9
Reactivity of the core after melting (%) (corium MOX (18%) without core catcher and borated water)

	Void (%)
Hom.	+8.0
Het.up	+22.6
Het.down	+25.5

Table 10
Reactivity of the core after melting (%)

	5000 ppm B-water $T = 406$ K (133°C) (%)
With highly borated water	
Hom.	+6.4
Het.up	+18.7
With a core catcher	
Hom.	+6.2

these tendencies have, of course, to be validated by means of criticality calculations using more sophisticated modelling in order to take into account the more realistic scenarios that will result from the modelling of 'composite' corium behaviour.

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