NUMERICAL INVESTIGATION OF THE RECRITICALITY OF THE MOLTEN CORIUM BASED ON HEAVY METALS DURING SEVERE ACCIDENT IN LIGHT WATER REACTOR

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ABSTRACT:

During a hypothetical severe accident with Light Water Reactor a molten corium pool would be formed in RPV bottom head containing large amount of fissile material as tons of Uranium and hundred kilograms of Plutonium. Extensive part of these are oxidized, but some part is transformed into pure heavy metals, due to high temperature chemical interactions between molten core components and with the wall of Reactor Pressure Vessel, as shown in some experimental investigations during the last decades in Russia and Europe. The question arises whether critical mass could be formed in this case. In the open literature, there was no quantification of the recriticality in conditions without water overflow of the molten corium in the bottom of reactor head. Such possible recriticality would be formed mainly on fast neutrons and dozen kilograms of Plutonium, if gathered in one place at the reactor bottom, would be enough to form a critical mass. To answer to this challenge, a numerical investigation of the effective coefficient of neutron multiplication k_{eff} for the molten corium pool during late in-vessel phase of severe accident was performed by a special methodology, proposed by us, which consists in a consecutive usage of several numerical codes – ORIGEN and MCNP for neutron-physics calculations, ASTEC for system calculations of some severe accident phenomena, THERMOCALC for thermo-chemical calculations of the system U-Pu-Zr-O and again MCNP for k_{eff} calculation of molten corium. VVER-1000, W-320 core was used as referent.

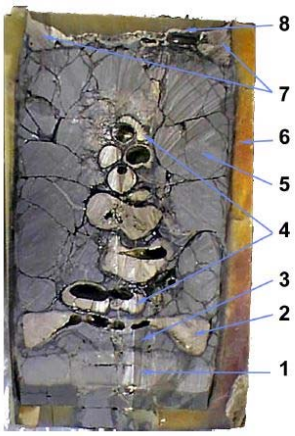
List of abbreviations:

ECCS – Emergency Core Cooling Systems
EOC – End Of Cycle
FP – Fission Product(s)
HM – Heavy Metal(s)
KTH – Royal Institute of Technology, Stockholm
LWR – Light Water Reactor
RPV – Reactor Pressure Vessel
SA – Severe Accident
TLFW – Total loss of Feed Water
MCNP – Monte Carlo N Pericles

1. Introduction and ground for the investigation

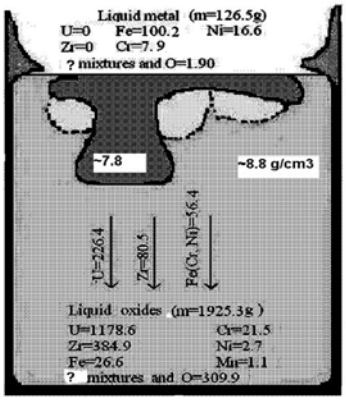
During a hypothetical severe accident with LWR the molten corium pool would be formed in RPV bottom head containing large amount of fissile material. A multiple layers with different coefficients of heat transfer can be formed. At the same time, as the experiments with the molten steel and corium from MASCA project have revealed [1], the interactions between molten steel and corium resulted in the mass exchange between phases when some amounts of metallic Uranium and Zirconium initially located in the sub-oxidized corium passed into metallic phase, what leaded to change of density and sinking of metal layer. At the bottom of the test facility Uranium in metallic form was found (Figure 1, a, b).

The discovered inversion in layers initiated extensive studies, e.g. [2], [3] and many others.



1. The corium lower non-melted briquettes;
2. The lower metallic part;
3. Zone of a partially melted corium;
4. Roundish metallic parts in the central zone;
5. The ingot oxidic part;
6. Epoxy;
7. The upper metallic part;
8. The ingot surface;

a) General view



b) Detailed view

Figure 1 Metals sink through oxydic layer in 1st stage of MASCA project, [1].

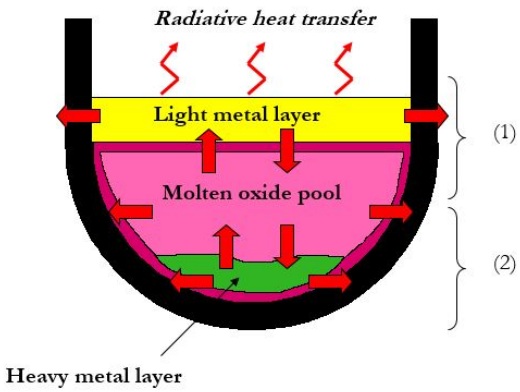
The results from MASCA project and other similar projects have shown that in late stage of in-vessel phase of SA the formed pool would be stratified at least at three levels:

- upper – metallic – layer
- medium – oxidic – layer
- lower – heavy metals (including U and Zr) – layer

The oxidic layer is the largest one among them. It also can be sub-stratified depending on the density of some oxides, which is shown in Table 1 and was introduced in ASTEC v.2.0 (Figure 2):

Table 1

Compound	Melting point, °C	Density, [g/cm^3] (at 20 °C)
UO_2	2846	10.6 {8.9 g/cm^3 (at 2000 °C)}
ZrO_2	2715	5.89
Different Iron (incl. non-stoichiomeric) oxides	1100-1370	5.17 – 5.74



(1) Layers out of thermodynamic equilibrium

(2) Layers in thermodynamic equilibrium

Figure 2 Stratification of molten core in the pool during late in-vessel phase of SA, [6].

The inversion of HM and their concentrating at the bottom of RPV provoke another question – what happens with the Plutonium and is there a potential for recriticallity of the corium or in some of its parts. Several kilograms of pure inversed Pu gathered at one place of the bottom of RPV would be enough to form a critical mass. This was the focus of our interest of this investigation. As a referent plant was used unit 5 of NPP Kozloduy, VVER-1000, B-320.

The topic of recriticallity for LWR and especially for VVER-1000 was already studied, but at thermal neutrons, i.e. when re-flooding the melted core by water, e.g. [4].

A preliminary assessment with the industrial code ORIGEN for the quantity of this fissile material was done in the end of 2005 for the unit 5 of NPP Kozloduy with the former assemblies TVS-M type for the end of 11th cycle. The code has given the following quantities of different isotopes of Plutonium:

362 kg of Pu-239 at EOC, T=261.4 efpd,

6 kg of Pu-238

113 kg of Pu-240

72 kg of Pu-241

17 kg of Pu-242

In the open literature, there was no many information on the criticality of Plutonium dealt in civil facilities [5]. There were studies on plutonium physics and chemistry ([7] ÷ [12]), but related to severe accidents, i.e. reactions at very high temperatures (T>2000 °C) – not at all at that time, but later it appeared in [13]. In Table 2 the melting points of Plutonium compounds that can be found in the reactor molten core are presented.

Table 2

Compound	Melting point, °C	Density, $[g/cm^3]^*$
PuO	1017	19.84
PuO_2	2390	11.5
Pu_2O_3	2085	11.48

The main assumption from chemical point of view was that at high temperatures the Plutonium is more active than the Zirconium and can replace it in the oxide compounds:

$PuO_2 + Zr \rightarrow ZrO_2 + Pu$ …(1)

and

$2Pu_2O_3 + 3Zr \rightarrow 3ZrO_2 + 4Pu$ …(2)

This promoted to look in the thermochemistry at high temperatures to find out through the solving of Gibbs equations what quantity of Plutonium could be formed in the reactor core in conditions of full molten core in RPV.

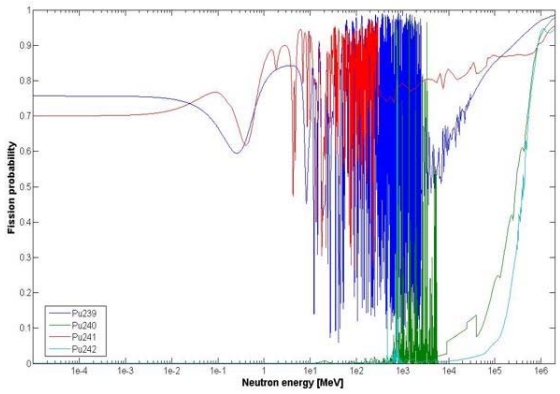


Figure 3 Fission probability of Plutonium isotopes.

It is obvious that this “potential of recriticality”, in the conditions of non-over-flooding of the molten core, means potential of criticality taking into account the entire spectrum of the Plutoniumhe heavy metals, but especially in the field of fast neutrons (Figure 3).

2. Methodology

To answer to above questions, a numerical investigation of k_{eff} of the molten corium pool was performed by a special methodology proposed by the authors, consisted in a consecutive usage of several numerical codes MCNP for neutron-physics calculations for EOC.

The Methodology consisted in four steps;

1. Run MCNP calculations for a typical core loading at 40 MWd/tU with 4,4% enrichment and new Zirconium content for TVSA assemblies (that were ahead to loading) to calculate the fission products.

2. Run ASTEC (v.1.3 at that time) for:

– assessment of Pu decrease in form of FP release of Plutonium oxides

– quantitative assessment of the molten pool components.

It was used the scenario with TLFW with all ECCS unavailable during all the time of the accident.

3. Run THERMOCALC code (for which the neutron-physics dept. of KTH had license for use) for Thermochemical equilibrium calculations (through solving the Gibbs equation) to assess the chemical form of Plutonium and the mixture of heavy metals in the bottom layer.

4. Run again MCNP with the Pu and other heavy metals formed after achievement of thermochemical balance in the pool to calculate k_{eff}.

3. Results from calculations

The main goal of the first step – MCNP – was to assess the quantity of fission products for EOC of VVER-1000 (i.e. to repeat the preliminary calculations made by ORIGEN, but at the new conditions, described in in step 1 of the Methodology).

The main goal of the second step – run ASTEC– was to assess, besides the other components of the corium, how much Plutonium (in form of oxides) evaporates from the pool, respectively how much rests in the pool. The latter is shown on Figure 4.

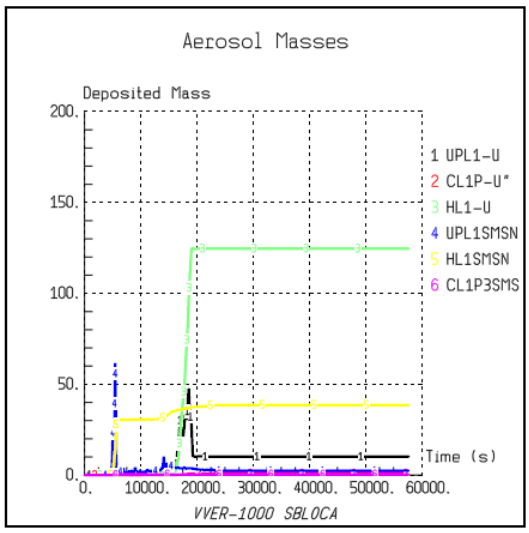
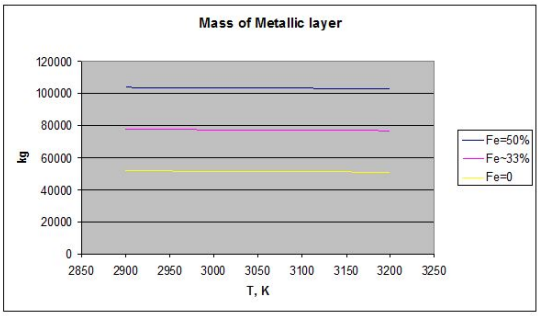


Figure 4 Masses of volatile aerosols detached from the pool

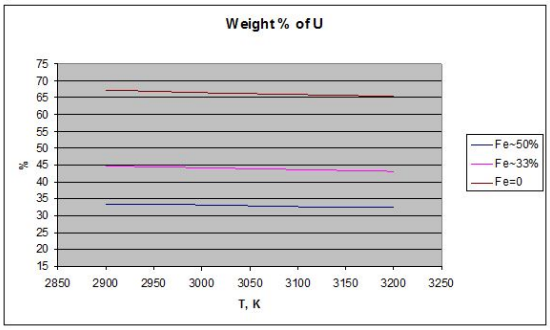
That calculation has shown that the rate of PuO_2 release as FP is $<1.10^{-9}$ kg/s, so one could estimate that all Pu remains in the pool.

For the transition from ASTEC to THERMOCALC there were quite recently published solution models for the U-O-Zr and the Pu-O-Zr systems and we embedded them in the third step, so they were used for thermochemical calculations to estimate the composition of the different layers. The time scale has allowed the system to reach equilibrium.

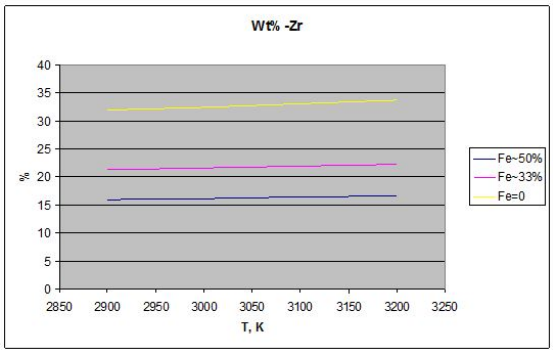
Three sensitivity calculations were made for different amount of Fe in the thermodynamic equilibrium metallic layer of the lower part of pool *before* oxidation and minimization of Gibbs energy, by applying the THERMOCALC code, and they are shown on Figure 5.



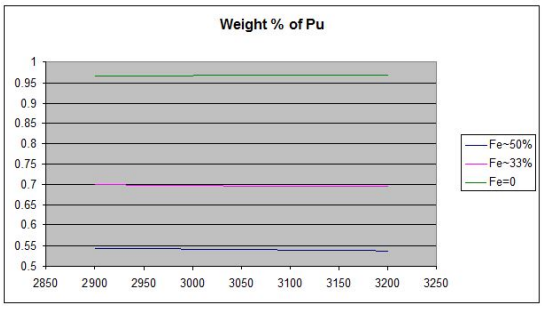
a) Mass of metallic layer



b) Weight percent of Uranium in the oxide layer



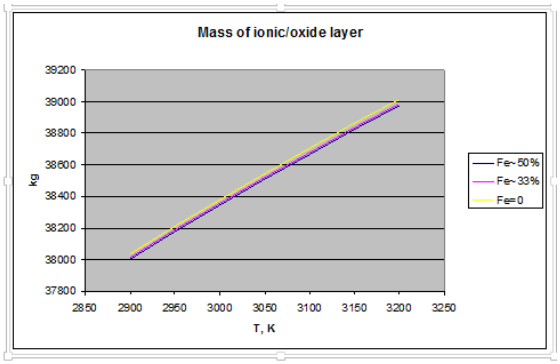
c) Weight percent of Zirconium in the oxide layer



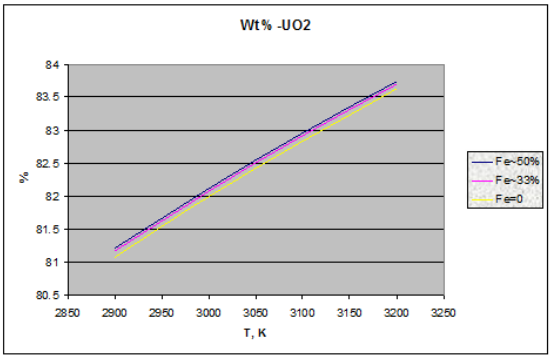
d) Weight percent of Plutonium in the oxide layer

Figure 5 Assessment of the Zirconium, Uranium and Plutonium in the pool before oxidation at different levels of Fe content.

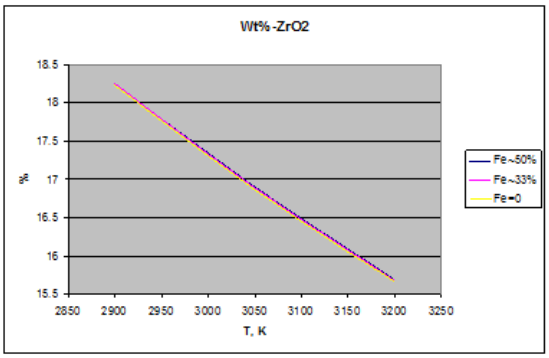
On Figure 6 (a) ÷ e)) are shown the results from Thermocalc calculations – mass and – weight percent of metals in the thermodynamic equilibrium ionic/oxide layer *after* oxidation and minimization of Gibbs energy.



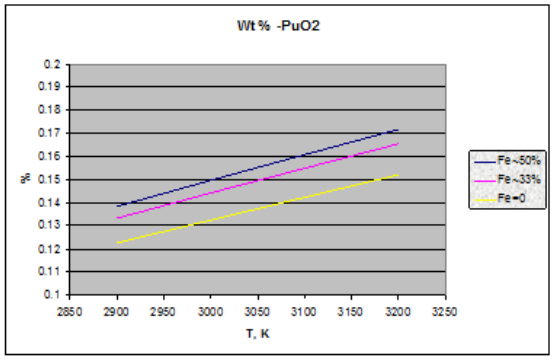
a) Mass of ionic/oxide layer



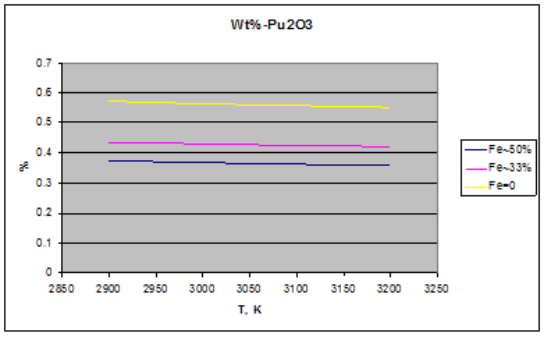
b) Weight percent of Uranium dioxide in the ionic/oxide layer



c) Weight percent of Zirconium dioxide in the ionic/oxide layer



d) Weight percent of Plutonium dioxide in the ionic/oxide layer



e) Weight percent of Plutonium trioxide in the ionic/oxide layer

Figure 6 Assessment of the oxidation of Pu, Zr and U at the expense of the internal Oxygen contained in the pool

Obtaining the results form THERMOCALC calculations, we have reached to step 4 and applied again MCNP to calculate k_{eff} for this system.

The final result is shown on Figure 7.

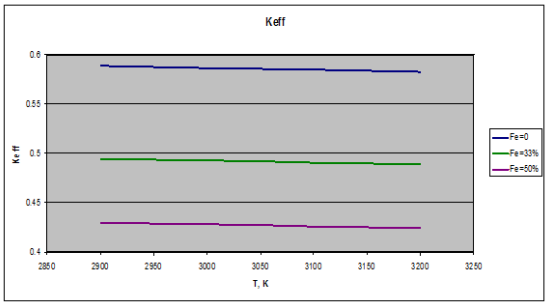


Figure 7 Sensitivity calculation of k_{eff} for the molten pool

4. Conclusion

This result shows that the maximum values of k_{eff} for the chemical system of U-Pu-Zr-O in the molten pool are far below 1 and recriticality in “dry” conditions in the late in-vessel phase of severe accident is not possible.

5. Acknowledgements

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6. References:

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