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## ADVANCED ANALYSES OF MOLTEN CORE CORIUM INTERACTION WITH TOLBIAC-ICB AT FUKUSHIMA DAIICHI UNIT 1 IN THE FRAMEWORK OF THE ARC-F PROJECT

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### ABSTRACT

In the framework of the OECD/NEA ARC-F project, predictions of long-term MCCI progression by eight different computer codes have been compared for the configuration of the FUKUSHIMA DAIICHI UNIT 1 sump. Indeed, the severe accident at Fukushima Daiichi Nuclear Power Station occurred during the night between March 11th and 12th of 2011. Nevertheless, the effective cooling of the fuel debris was not restored before March 23rd. Therefore, Molten Core Concrete Interaction (MCCI) most likely occurred for more than 10 days without the presence of water (i.e., under dry conditions). Such a long period of MCCI is rarely studied.

This paper focuses on the detailed analyses of the results obtained with the TOLBIAC-ICB tool during the two steps of this benchmark. The first step consisted in a preliminary rough comparison of results allowing then to draw new converged axes of research. Results of TOLBIAC-ICB V3.4 mod2 is described and explained in relations with implemented physical models and results obtained from others codes. The second step presented simulations with more agreed initial data and boundary conditions. The detailed MCCI phenomenology is depicted and physically discussed based on the long-term evolutions of temperatures, detailed pool and crust compositions in term of chemical components, 2D ablation front, gas releases... All this advanced analysis raises the question of the validity of some models of current, intensively used, MCCI tool for very long term transients.

### 1. INTRODUCTION

The TOLBIAC-ICB code has been developed to simulate molten core concrete interaction (MCCI). The general approach used in this code is based on the segregation phase model, which assumes that a crust mainly composed of refractory oxides forms at the pool boundaries. The

development of the TOLBIAC-ICB (Spindler et al. 2005) code was started in 2002 in the frame of an agreement with EDF (Electricity of France) to provide a numerical tool for safety analysis related to MCCI in reactor situations. The main characteristic of this tool is that it couples thermal-hydraulics and physico-chemistry through a coupling with the GEMINI2 thermodynamic solver.

The main noteworthy TOBIAC-ICB feature is that the interfacial temperature between the crust and the pool is the liquidus temperature calculated from the evolving composition of the pool on the contrary of the other MCCI simulation tools where this interfacial temperature is the solidus temperature or is in-between the solidus and the liquidus temperature (Nuclear Energy Agency, 2017).

To simulate the proposed benchmark, the following mesh has been studied. After first preliminary calculations, a cylindrical geometry has been chosen to ensure the 2D axis-symmetrical ablation. The sump geometry is an equivalent geometry to the rectangular one given by the benchmark to conserve as well the power and the surfaces. The equivalent radius of the sump is 0.82m with a height of 1.35m. It should be noted that the volume is 1.13 time larger compared to the initial given rectangular geometry. Finally, for the two steps of the benchmark, the total simulated geometry, including the concrete thickness, was a complete cylinder of radius of 7.82 m (7m of concrete + 0.82m for the sump) and height of 10.35 (9m of basemat and 1.35m for the sump). The mesh sizes in the concrete are of 0.1m in radius and 0.1 m in height, the pool is calculated in 0D.

### 2. MAIN MODELS

This analytical benchmark (Pellegrini et al. 2022) is progressive in complexity, in order to distinguish all separated influences caused by each difference between the various

simulation tools. A dry concrete is firstly considered, standing for the accidental situation during the first days, with no water injection. So no water ingress model is considered as well as no water aspersion and melt ejected out of the pool, through the upper crust. To simplify this preliminary comparison of tool results, no stratification in the initial debris is also assumed.

TOLBIAC-ICB is provided by the concrete composition before the concrete decomposition (Table 1- composition at low temperature). From the benchmark input (Table 2), the initial concrete temperature is already the partitioning after the concrete decomposition (even if it is given at 300 K):

Compounds	Al <sub>2</sub> O <sub>3</sub>	Ca(OH) <sub>2</sub>	SiO <sub>2</sub>	H <sub>2</sub> O	Fe <sub>2</sub> O <sub>3</sub>	CaCO <sub>3</sub>
Weight (%)	15.4%	16.8%	62.5%	0%	6.0%	0%

**Table 1: Concrete composition before concrete decomposition**

Compounds	Al <sub>2</sub> O <sub>3</sub>	CaO	SiO <sub>2</sub>	H <sub>2</sub> O	Fe <sub>2</sub> O <sub>3</sub>
Weight (%)	15.4%	12.8%	62.5%	3.3%	6.0%

**Table 2: Concrete composition after concrete decomposition**

Due to the low conductivity of concrete (heat conductivity ~1 W/m/K), conduction is not considered in the concrete thickness in TOLBIAC-ICB (commonly made assumption). One can wonder whether this assumption is still realist for a long term accident of 10 days.

The water, issued from the concrete decomposition (3.3 wt%), is released inside the corium pool at vapor state. In case of free water would last after concrete decomposition, it would be vaporized between 100 and 400°C. The corresponding latent heat is of 2.2MJ/kg. The molten pool is modelled in 0D whereas the crust and concrete are meshed (§1). Regarding the pool energy balance closure laws on this pool, the heat convective at concrete/pool (bottom pool) is given by the BALI correlation (Bonnet et al., 2000) established for lower pool surfaces (which is the default correlation for this type of surface):

$$\dot{h} = 19.6 \lambda [\sigma/(\rho g)]^{-0.5} [\rho Jg^3/(\mu g)]^{0.146} [\mu Cp / \lambda]^{-0.22} \quad (1)$$

The convective heat transfer coefficient at concrete/pool (sides) was not investigated in the BALI tests. Thus, by default the same correlation as above is used (Bonnet et al., 2000).

For upward heat transfer, the BALI correlation for upper surface is considered:

$$\dot{h} = 24.55 \lambda [\sigma/(\rho g)]^{-0.5} [\rho Jg^3/(\mu g)]^{0.073} [\mu Cp / \lambda]^{-0.29} \quad (2)$$

As no stratification is assumed in the debris in the benchmark description, only a homogeneous pool is considered with normally a tiny crust at its uppermost location (even if sensibility studies have been performed on possible stratified pool configuration). This kind of crust generally appears at the upper surface of the pool because of radiative heat transfer towards the reactor cavities. Nonetheless, the crust density is larger than the melt density (crust composed of refractory materials), and the crust continually forms and sinks. Thus, in conclusion it is considered in the reference case calculation that a thin crust is located at the top of the corium pool in this configuration. The heat transfer at the pool upper pool crust interface is removed by radiation in this quasi-static TOLBIAC-ICB simulation. The radiation heat transfer is

simply calculated between two horizontal and infinite plane surfaces. As no information from the benchmark was given, the surroundings of the melt have firstly been considered at the constant temperature of 1573K (~steel melting point). The emissivity of the upper corium interface is taken to 1 and the emissivity of the surrounding to 0.8. As already written, the temperature at the pool/crust interface (surrounding the pool) is the liquidus temperature of the corium. Due to this features, the TOLBIAC-ICB solver does not deal with solid debris. Thus for the benchmark, the internal fuel energy release first heats-up the debris until the liquidus temperature before concrete ablation starts. Then, the pool/concrete interface structure is a stable macro-segregated crust whose composition is given at the various time steps by the GEMINI2 solver (NUCLEA data base) from the corium melt composition, in which the ablated concrete is incorporated at each time step.

Knowing the heat flux to the concrete, whose energy is used to heat, decompose, melt and heat-up this molten component of the concrete to the pool temperature (allowing their integration inside the corium pool), the erosion length is calculated (given here in 1D):

$$\frac{dx}{dt} = v_{ab} = \frac{\phi_{l \rightarrow c}}{\rho_s(H_{s,c} + H_{d,c} + H_{l,c}) + \rho_l C_{p,l}(T_p - T_l)} \quad (3)$$

Where  $v_{ab}$  is the ablation velocity. The denominator gathers all the energy contributions from the solid concrete at initial temperature  $T_0$  to the liquid at  $T_p$  (pool temperature):

- Specific heat to heat solid from  $T_0$  to  $T_l$  (expressed by the enthalpy  $H_{s,c} = C_{p,s}(T_l - T_0)$ )
- Decomposition enthalpy ( $H_{d,s}$  standing for the energy to chemically transform the components) which is not taken into account in this version of TOLBIAC-ICB.
- The latent heat of these components ( $H_{l,c}$ ) to melt them.

TOLBIAC-ICB, there is no model of fission product release. The energy of chemical product reactions (such as concrete decomposition) is not taken into account in this version of TOLBIAC-ICB; the heat of concrete decomposition as well as the latent heat of vaporization are not considered in the global energy balance as it has supposed negligible in front of the melting latent heat and specific energy of concrete.

### 3. FIRST STEP OF BENCHMARK

The TOLBIAC-ICB results of the first step of the benchmark have been presented in December 2020 and reported in the global summary results (Pellegrini et al., 2020). Two different calculation results were provided: the first one obtained with an assumption of isotropic ablation and the second one considering a quite more realistic anisotropic ablation. Indeed, the ablation is known to be isotropic for a limestone rich concrete and strongly anisotropic for a basaltic concrete (Fukushima concrete like). In the benchmark, the concrete composition rather looks like a basaltic (62% of SiO<sub>2</sub> and 0% of released CO) concrete so that, based on the observations VULCANO VF-U1 test representative of 1F1 1st day of MCCI (Bouyer et al, 2019; Boulin et al., 2020) which highlighted a mean axial ablation depth to be about 6 times greater compared to mean radial ablation depth, we decided to performed a sensibility study to the 2D geometry of the ablation. However VF-U1 test was a short-term MCCI test compared to this benchmark test. So, we are not completely

convince that its result is representative for this ARC-F benchmark. The reality might be in-between our two sensibility calculation results (isotropic or anisotropic). Both results were in the same ranges of magnitude with an ablated depth significantly larger than the radius in the anisotropic case.

In generally, it was noted that the pool temperature was higher than the average one obtained by the other participants and that, even if the eroded depth and radius were quite higher than other participants results, the ablated mass obtained with TOLBIAC-ICB was in the range of other participants. Interpretation work of these discrepancies has led to the conclusion that the pool temperature is high because of the higher thermal boundary condition of the pool (which is the liquidus temperature, quite higher than the solidus temperature considered by some other solver). Also, it appeared that the TOLBIAC-ICB given depth and radius were the maximum value and the ablation was not uniform (Pellegrini et al., 2020). That was not necessarily the case for other participants. Furthermore, from a mass balance, the eroded volume well coincides with the eroded shape. We noticed that the density of the concrete was of 2140 kg/m<sup>3</sup>, below the average 2400 kg/m<sup>3</sup> used by most of the other MCCI tools.

We further investigated the isotropic case results to highlight the remaining main discrepancies with the other tools. The Tliquidus was given by the NUCLEA data base (via GEMINI2) (Pellegrini et al., 2020). It is very close to the pool temperature evolution. The Tsolidus is not used in TOLBIAC-ICB and thus was not calculated. The corresponding evolution of the percentage of incorporated concrete inside the pool during the transient reaches an asymptotic value at 85% corresponding to a range of low liquidus temperatures (2000-1700K).

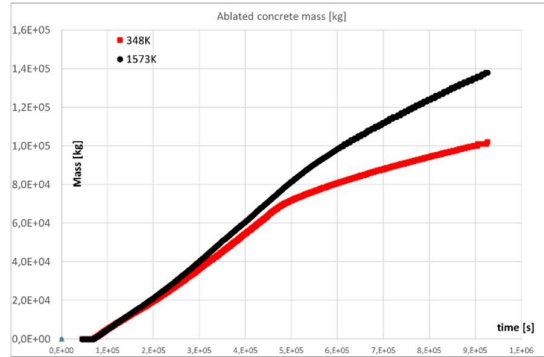
The evolution of the crust and pool compositions have been studied (Table 3). Whereas heavy oxide metal first were incorporate inside the crust (surrounding the pool), their percentage in weight then diminished and oxide of iron and silica were predominant on the very long term of the transient. On the same manner, the main species inside the pool at the end of the transient were SiO<sub>2</sub> (38,11 wt.%) and CaSiO<sub>3</sub> (29,38 wt.%).

Time	12.5h	13.9h	19.4h	26.4h	33.3h	40.3h	81.9h	151.4h	220.8h	290.3h
Comp[wt.%]										
U	0	0	1,21	0	0,77	0,62	0,44	0,25	0,16	0,14
UO2	0	0	70,04	59,21	65,18	62,79	45,97	40,71	26,4	22,66
Zr	0	0	1,15	0	0,81	0,65	0,47	0,27	0,17	0,15
ZrO2	0	0	27,6	40,4	33,1	31,04	22,5	13,09	8,49	7,29
CaO	0	0	0	0,1	0,08	0,06	0,05	0,03	0,02	0,01
FeO	0	0	0	0,29	0,06	3,87	25,73	25,62	16,61	14,26
Cr2O3	0	0	0	0	0	0,97	4,84	5,55	3,6	3,09
NiO	0	0	0	0	0	0	0	0,09	0,06	0,05
SiO2	0	0	0	0	0	0	0	14,39	44,48	52,34
Al2O3	0	0	0	0	0	0	0	0,01	0,01	0,01
wt.% of concrete in pool	0	0	0,001	0,183	0,366	0,526	0,806	0,856	0,851	0,851
Ablated Vol [m <sup>3</sup> ]	0	0	0,017	2,015	4,06	6,39	20,07	43,07	60,56	69,5

**Table3: The evolution of the crust composition and percentage in weight of concrete in the pool (isotropic case)**

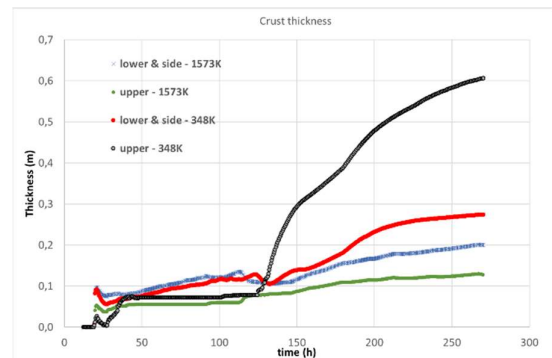
Regarding more in details the power released by the upper plate, it appeared that the considered boundary upper temperature considered in TOLBIAC-ICB was quite higher than those considered in other tool; 1573K instead of 348K. Sensibility calculations have been carried out to this boundary

condition in temperature considering the isotropic case. The maximum eroded depth and radius were lower (of about 0.5m) in case of the lower temperature (348K) as well as the ablated mass (Figure 1) which is also close to other participants results. In this new simulation, the percentages in weight of SiO<sub>2</sub> inside the crust and the pool reached 67%. The upper boundary condition has thus a large impact on the results.



**Fig 1: Ablated masses**

Regarding more precisely to the crusts evolution (Figure 2), although the crust thicknesses around the pool were already quite thick in the reference calculation with 1573K as upper structures temperature (~10cm above the pool and ~20 cm at the bottom and side walls), it becomes even more physically questionable in the sensibility simulation; with at the end ~30 cm crust thickness at the side and the bottom and 60 cm at the top.



**Fig. 2: crust thickness evolution – isotropic case with various upper boundary conditions**

This surprising result found, however, an explanation in the coupling of thermal-hydraulics of TOLBIAC-ICB with the physico-chemistry results of GEMINI2. Indeed this upper crust thickness is controlled by its conductivity times the difference of temperature between the liquidus temperature and the upper crust temperature divided by the power transmitted through the crust (power from the pool and internal power of the crust due to the Uranium presence). As already explained, at the beginning U element is incorporated to the crust and after 100h, its quantities decrease and thus does the transmitted power. The upper crust surface temperature tends to about 1580K in the reference case (boundary condition of 1573K) and 698K in the sensibility case (boundary condition of 348K). This is this increasing difference of temperatures in the sensibility case which leads to the important upper crust

thickness. This difference de temperature remains limited in case of the bottom and side crusts to the difference between the liquidus temperature and the concrete melting temperature (~1540K). So, the thicknesses of these crusts do not reach so large value as at pool top.

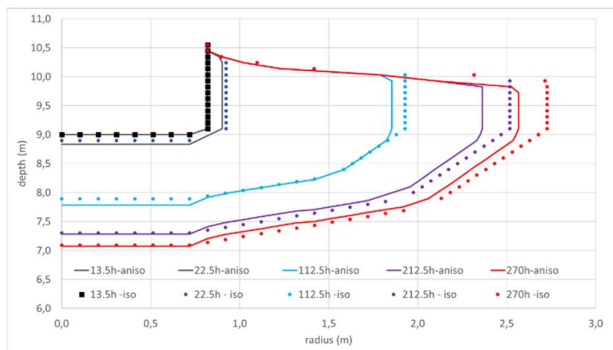
This last point underlines a weakness of TOLBIAC-ICB. The classical values of heat fluxes during a MCCI are about 200 kW/m<sup>2</sup> and the associated crusts are of the order of mm or cm. The heat fluxes reach at the end of this benchmark very long-term transient are almost 50 lower. This explains very thick crusts, which end by controlling the energy transfers. One can wonder if these configurations are expected to be representative of the long term phase of MCCI i.e. during an interaction between an oxide/metal stratified corium melt and a concrete structure, and about the validation status of the modelling for these very long term configurations.

#### 4. SECOND STEP OF BENCHMARK

Following the lessons of the first step, it was decided for the second step to impose the concrete density to 2400 kg/m<sup>3</sup> and the evolution of the upper release power (given by SNL based on BSAF results).

As previously an isotropic and an anisotropic (with a ratio of 6 on the axial heat coefficient) cases have been computed.

This time, TOLBIAC-ICB results were in great agreement with other MCCI tools results in terms of eroded depth, radius and mass but also of H<sub>2</sub> et H<sub>2</sub>O gas generation and of oxide masses (ZrO<sub>2</sub>, Cr<sub>2</sub>O<sub>3</sub>, FeO) formation (here reference of the section where global comparison of second step is made).



**Fig. 3: 2D concrete erosion (isotropic and anisotropic cases)**

Figure 3 highlights the discrepancies in the ablated concrete geometry in case of isotropic and anisotropic cases; the maximum ablated radius being far less important in anisotropic case whereas the maximum ablated depth is a little bit higher (owing to the respective difference of surfaces, bottom surface being far larger than side surface). The evolution of resulting ablated concrete mass reaches 1.1 10<sup>5</sup> kg in the isotropic case compared to 10<sup>5</sup> kg in the anisotropic case. The bottom and side crust thickness were about 20 cm at the end of the transient.

#### 5. CONCLUSIONS

Detailed analyses of results obtained with the TOLBIAC-ICB tool during the ARC-F project on the simulation of MCCI in the Fukushima Daiichi Unit1 accident have been discussed. They underlined the importance of the thermochemical interactions between corium and concrete elements, the need of simulate realistically the crusts thicknesses and the evolution of their composition. Current models of crust and

assumption regarding the conduction inside the concrete and their negligible heat decomposition energy seem not to be representative of the long-term MCCI phase. This also raises the issues of the interfacial temperature model and of the volumetric power in TOLBIAC-ICB, which is proportional to the UO<sub>2</sub> concentration. This power takes not into account the fission products and the crust composition evolutions. Furthermore, in TOLBIAC\_ICB, this evolution is not a progressive crusting.

Next work could focus on wet MCCI taking a particular attention to the long-term thermochemistry with salt water

#### NOMENCLATURE

C <sub>p</sub>	corium heat capacity of corium (J/kg/K-1)
g	gravitational acceleration (m.s <sup>-2</sup> )
H	enthalpy (J/kg)
h	convective heat coefficient pool/concrete (W/m <sup>2</sup> /K)
J	superficial velocity (bottom and sides) (m/s)
v	velocity (m/s)
x	concrete erosion length (m)

#### Greek letters

φ	heat flux (W/m <sup>2</sup> )
λ	corium conductive heat coefficient (W/m/K)
μ	dynamic viscosity (kg/m/s)
ρ	corium density (kg/m <sup>3</sup> )
σ	corium surface tension (N/m)

#### Subscripts

ab	ablation
c	concrete
g	gas phase
i	interface
l	liquid
s	solid

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