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Experimental study of transient phenomena in the three-liquid oxidic-metallic corium pool

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ABSTRACT

Non-steady physicochemical phenomena in the three-liquid molten pool of prototypic corium are studied in the context of in-vessel melt retention problem. Experiments are made on the Rasplav-3 test facility within the CORDEB program. Structure of the initial molten pool consists of the surface light melt of molten steel, the intermediate layer of oxidic melt separated from steel melt by the crust; and the bottom layer of heavy metallic melt. It is determined that the three-layer pool structure can stay stable for a certain period of time, but the partitioning of steel and oxidic melt components through the crust brings the possibility of transformation of the three-layer pool to a two-layer structure.

1. Introduction

One of the phenomena limiting the in-vessel melt retention (IVR) during a severe accident with core meltdown in a Light Water Reactor (LWR) is the focusing effect along the top metallic layer of the oxidic-metallic pool, which determines the maximum heat flux applied to the vessel wall. Comparison of this flux with DNB (departure from nucleate boiling) on the outside of the water-cooled reactor vessel surface gives the safety margin. The most systematic study of this problem was made in (Theofanous et al., 1997). In a general case the intensity of focusing effect increases, when the depth of top metal layer or stainless steel (SS) mass decreases.

An additional complexity in the problem solution was brought by the results of studies carried out within the OECD MASCA program. In (Asmolov et al., 2004, 2007), in particular, it was established that component partitioning in the system of the melted suboxidized $UO_2 + ZrO_2 + Zr$ corium and SS in the miscibility gap produces a two-layer oxidic-metallic pool, and its metallic liquid, beside SS components, includes U, Zr and a small amount of O. Depending on the system composition, the metallic liquid density can be either lower or higher than the oxidic liquid density. Therefore, the oxidic and metallic liquids can take both bottom and top position in the molten pool. Other conditions being equal, the lower SS fraction in the system is, the larger is U fraction in the metallic liquid, and the higher is its density. Experimental data on the metallic liquid composition provided in (Asmolov et al., 2004, 2007) are close to the data of (Barrachin and Defoort, 2004; Salay and Fichot, 2004) calculated using the NUCLEA thermochemical database and minimization of the Gibbs energy. The MASCA data are used for the modelling of molten pool structure and assessment of risks related to the focusing effect produced by the top metallic layer (Zhang et al., 2010; Le Tellier et al., 2015).

In principle, a multi-component system can have thermodynamically stable coexistence of two, three and more liquid phases. A most frequent case is the coexistence of metallic liquid with two oxidic liquid phases. Such stratification, for example, can be caused by the interaction of corium with certain oxidic materials (Gusarov et al., 2007; Asmolov et al., 2007). There are also publications on the stratification of metallic melt into three liquid phases (Konovalov et al., 2012).

The non-equilibrium melt having two, three and more liquid phases is also possible in presence of thermal gradient maintained in the system (Gusarov et al., 2006), or because of the spatial separation of

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light and heavy phases by gravity (Udalov et al., 2008), or due to the kinetically complicated mass transfer. The latter circumstance makes it difficult to establish equilibrium between liquid phases produced in different conditions (Lungu et al., 1973). The presence of several liquid phases having different composition and location, as well as the factor of their stability, can have a considerable influence on the in-vessel melt retention (Zhang et al., 2010; Le Tellier et al., 2015). In view of this, it is very important to analyze phenomena causing the transient evolution of liquid phases with different compositions in the UO_2-ZrO_2-Zr-SS system and to study their stability.

During the molten pool formation in the reactor vessel bottom a situation is possible, when only a part of SS eventually relocating to the bottom has the initial interaction with oxidic melt to form “heavy” metallic liquid in the bottom. In this case the remaining SS forms the surface layer of “light” liquid separated from the oxidic melt by the oxidic crust. In this way, the three-liquid three-layer molten pool is established. It is obvious that in comparison with the two-layer pool, the total SS mass being the same, the focusing effect of the surface metallic layer in the three-layer pool is higher.

The study in (Seiler et al., 2005) proposes a methodology for calculating the minimum possible fraction of the known total SS mass, which would form the top layer with maximum focusing effect corresponding to its depth. For the known oxidic melt mass and composition, the methodology uses calculations similar to (Barrachin and Defoort, 2004; Salay and Fichot, 2004) for determining the maximum SS mass, at which the produced metallic liquid would be heavier than the oxide, i.e. it would lie under the oxidic liquid. Consequently, the minimum possible remaining SS would form the top layer. It is assumed that the crust separating it from the oxidic melt prevents the physicochemical interaction between the SS and oxidic melt.

Beside the limitations of the conservative approach, which exclude the direct application of methodology (Seiler et al., 2005) in the calculations modelling transient processes, we also should note the assumption that the oxidic crust is impermeable. The mass transfer through the layer-separating crust really takes place as is shown in study (Bechta et al., 2006) on the interaction of suboxidized corium melt and cooled carbon steel (VVER vessel steel) carried out within the ISTC METCOR project, where steel components get into the oxidic melt and melt components get into the molten steel layer. This is accompanied by the formation of liquid or liquid–solid metallic interaction zone under the crust in this layer.

The current work carried out within the CORDEB project presents the experimental study on the behavior of the three-liquid prototypic molten corium and SS pool. The study reproduces the above situation (except the scale and method of heat deposition), which can take place during the molten pool formation in IVR conditions.

2. Experimental procedure and macroanalysis results

Experiments CD1-01 and CD1-02 were performed on the Rasplav-3 test facility of the Rasplav platform. Corium was heated and molten using the technology of induction melting in the cold crucible in the argon atmosphere. Fig. 1 shows schematics of the induction furnace and oxidic-metallic pool at the moment, when the formation of initial three-layer structure was completed.

At the initial experimental stage, the molten pool of prototypic $UO_2 + ZrO_2 + Zr$ corium was produced; it had mass M_{cor}, oxidation index C-30, $(U/Zr)_{at} = 1.2$, $T_{cor} \approx 2500\,^{\circ}C$. After that SS having mass M_{ss} was introduced into the pool and the relative SS mass ($M_{ss}/(M_{ss} + M_{cor})$) became ≈ 0.16 for CD1-01 and 0.17 for CD1-02, the conditions were maintained for the period of time sufficient for the system to reach equilibrium (30 min). As determined in (Asmolov et al., 2004, 2007) and repeated in the discussed experiments, in such conditions the produced metallic melt is located in the pool bottom. After that, by changing the inductor power, position of pool (13) and shield (5) versus inductor (1), an oxidic crust (15) was formed on the oxidic melt surface; it had the temperature of approx. 1600 °C. After that an additional portion of SS was supplied above the oxidic crust surface; and it formed the melt surface layer (16): 466.1 g in CD1-01 and 490.4 g in CD1-02. The total relative mass of SS (together with the previously introduced material) was approximately 29% for CD1-01 and CD1-02. The melt surface temperature measured by the pyrometer and submersible thermocouple was approximately 1500 °C.

The established three-layer pool was exposed to the unchanged furnace parameters in CD1-01 for approximately 1 h, and in CD1-02 – for three hours. After completion the inductor was disconnected and molten pool was crystallized in the argon atmosphere.

CD1-01 ingot was taken from the crucible and enclosed in epoxy resin. After that ingot was cut along the axis. The resulting section is shown in Fig. 2a. Fig. 2b shows the schematics of metallic and oxidic ingot parts.

Three layers can be distinctly seen in the cross section; they were in the liquid state during the experiment: “light” metallic layer on top (LM), in the bottom “heavy” metallic layer (HM) and intermediate oxidic layer (Ox). Metallic “lens” found in the ingot bottom part was likely to be formed after the first SS portion was supplied into the molten pool, and after partitioning of the oxidic and metallic melts components. Heavy metal, which went down to the bottom, got partially crystallized on the water-cooled calorimeter to form the metallic “lens” Figs. 2 and 3).

CD1-02 ingot was cut along the axis, see Fig. 3a. The top oxidic-metallic layer (section, Fig. 3a) lies on the metallic part of underlying layer, which was cut into two parts. It is on top of oxidic part of the bottom layer, which also was cut into two parts. In this way, the ingot has two macrolayers, which were in the liquid state during the experiment: top oxidic-metallic layer and bottom layer consisting of the upper metallic and lower oxidic parts (oxides on the HM sides partially spilled out during disassembly). Similar to CD1-01 the pool bottom has a metallic “lens”. Locations of metallic and oxidic fragments in the CD1-02 ingot are shown in Fig. 3b. Ingot parts were weighed; their compositions were determined by the XRF analysis, except for LM. Its composition was determined by averaging of SEM/EDX data because of shape’s “irregularity”. The results are presented in Table 1.

To determine bottle density, samples were cleaved from the metallic ingot parts, and samples from the oxidic parts were produced by their crushing to the particle size < 100 µm and sampled by quartering. Ethyl alcohol was used as the picnometric liquid.

Table 2 gives the measured bottle density.

The final ingots and analyses results bring us to the conclusion that the crust separating Ox and LM is permeable for their components. Masses and compositions of all three layers of the molten pool undergo changes versus time: LM mass decreases, HM mass increases along with the decrease of U fraction and density.

The “irregular” shape of the LM surface layer, which is observed in Fig. 2 and, especially, in Fig. 3 is can be explained not only by the shrink cavity formation and by the bend of the Ox – LM interface surface at crystallization process. Mainly, the LM component transport into Ox is carried out in the hotter middle LM part, which is not in contact the cold crucible surface. This can explain the formation of large surface crust and gas cavity in the LM layer in CD1-02. Probably these phenomena result from the crucible wall’s influence due to the small scale of the test facility.

3. SEM/EDX analysis data

The qualitative results of the SEM/EDX analyses of CD1-01 and CD1-02 ingots are similar. For this reason only CD1-02 results are discussed below.

Top parts of both ingots Figs. 2 and 3 show the metallic layer in close contact with oxidic crust. Its characteristic microstructure and phase analysis are given in Fig. 4 (locations 3, 4) and in Table 3 (points P3, P4, P5). Average composition of the layer is given in Table 3