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Experimental investigation on molten pool representing corium composition at Fukushima Daiichi nuclear power plant

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HIGHLIGHTS

- A configuration of molten core in the Fukushima Daiich NPP unit 1 is investigated.
- Corium ingot consists of metallic layer on the top and oxidic layer at the bottom.
- Boron carbide was more concentrated in the upper metallic layer.
- Two layered configuration would contribute to the post-accident recovery actions.

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ABSTRACT

A configuration of molten core in the Fukushima Daiichi NPP (nuclear power plant) was investigated by a melting and solidification experiment. About 5 kg of a mixture, whose composition in terms of weight is UO_2 (60%), Zr + ZrO_2 (25%), stainless steel (14%), B_4C (1%), was melted in a cold crucible using an induction heating technique. It was shown that the solidified melt consists of upper crust and lower solidified ingot. The solidified ingot was separated into two layers. A physical and chemical analysis was performed for the samples taken from the solidified melt to investigate the morphology and chemical characteristics. It was found that the solidified ingot consists of a metal-rich layer on the top and an oxide-rich layer at the bottom. In addition, the oxide layer at the bottom has composition close to the initial charge composition and surrounded by a thin crust layer. It turned out that B_4C was more concentrated in the upper metal-rich layer. These findings provide important insights for understanding the core melt progression and taking proper post-accident recovery actions for the Fukushima Daiichi NPP.

1. Introduction

In the accident of Fukushima Daiichi NPP (nuclear power plant), it is estimated that the fuel and structural materials in the core had been melted for unit 1, 2 and 3 [1,2]. The nuclear fuel in the core was heated up and melted by decay heat as the coolant in the reactor vessel was depleted due to steam release from a safety relief valve and leakage from the reactor vessel pressure boundary without additional water injection. The core melting progression is a stepwise process which results in melting and liquefaction of core materials at different temperatures, which consist of various materials such as UO_2, ZrO_2, Zr, stainless steel and B_4C as a control rod material [3].

The configuration of molten core, so-called corium, in the Fukushima Daiichi NPP is of interest, as it affects the failure mechanism of a reactor vessel and the information on in-vessel melt relocation process is crucial for taking the proper recovery actions. The molten core configuration during the severe accident progression provides the initial conditions for the corium discharge into the containment and subsequent corium concrete interaction, which will lead to an ablation of concrete basemat and release of a mixture of fission products, combustible gases, and steam into the containment. Consequently, it can lead to a failure of containment due to pressurization and subsequent release of radioactive materials to the environment as evidenced in the Fukushima accident [1,2].

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It was suggested that the compositions of corium pool formed at the late phase of a severe accident would be in the domain of miscibility gap of U–Zr–Fe–O phase diagram [4,5]. Two immiscible liquids with low and high content of oxygen will coexist. A chemical equilibrium in molten corium pool was studied experimentally in the OECD-MASCA program [6]. The results of STF [7] and MA series [8–10] tests in the OECD-MASCA program indicate the sensitivity of a pool structure and position of coexisting stratified layers to the corium composition, melt oxidation index, and U/Zr and corium/steel mass ratios.

To investigate the configuration of corium relocated into the reactor lower head for a pressurized reactor during a severe accident, a melting and solidification experiment was performed for a typical corium of $UO_2/ZrO_2/Zr$/stainless steel mixture [11]. It was shown that the molten corium was separated into two layers, of which the upper layer was oxide mixture while the lower layer was metal alloy. The upper layer consisted of mainly UO_2 and ZrO_2 while the lower layer mostly consisted of metal mixtures such as U, Zr and stainless steel. U was produced by reduction of UO_2 by Zr [11].

In this paper, the configuration of molten corium pool for a typical Fukushima Daiichi NPP unit 1 composition was investigated by a melting and solidification experiment using an induction heating technique in a cold crucible. The typical melt composition for a BWR (boiling water reactor) should be different from that of a PWR (pressurized water reactor) in that it consists of more metals and B_4C as a control rod material.

The physical and chemical analysis was conducted for the samples taken from the solidified corium ingot to investigate the morphology and chemical composition at each position. The purpose of the analysis is to provide useful insights for both understanding the accident progression and taking the proper actions during the decommissioning of the Fukushima Daiichi NPP.

2. Experimental methods

2.1. Experimental setup

The VESTA-S (Verification of Ex-vessel corium STAbilization-Small) facility was used for a melting and solidification experiment, which employs an induction heating technique with a cold crucible. This technique has been optimized for melting of various reactor materials ranging from a metallic mixture of U, Zr and Fe to an oxidic mixture of UO_2 and ZrO_2 which are the prototypic reactor materials [12,13]. The cold crucible consists of a water distribution channel at the bottom and palisade-like fingers on the water distribution channel. The fingers and distribution channel are made of copper and inside of which water is forced to circulate to provide cooling. Outside the fingers, an induction coil is located to provide induction heating by high frequency generator. The basic principle for the cold crucible technique is described in detail in Refs. [12–14]. The facility was originally constructed to investigate the interaction behavior between corium melt and the structural materials such as a sacrificial material for a core catcher [13]. For this facility, a high frequency generator was designed to supply power up to 225 kW with 100 kHz. However, the frequency could change depending on several factors, such as crucible dimensions, induction coil turns, and melting materials and so on.

A schematic diagram of the facility is given in Fig. 1. Two cold crucibles are installed in the test chamber. A melt crucible is for melt generation and another crucible, an interaction crucible, is for the interaction of melt with a structural material specimen. The melt crucible has a dimension of 100 mm in diameter and 150 mm in height. If melt is generated in the melt crucible and the melt temperature reaches a desired temperature, it can be delivered into the interaction crucible by a remote-controlled rotating system for the melt-material interaction. This melt delivery process is available only for metallic melt not for oxidic melt because of crust formation at the top of the oxidic melt.

There are two optical pyrometers installed at the top of the test chamber to measure the melt temperatures in the melt and interaction crucibles. The whole process is monitored by CCD cameras. Argon gas is purged into the test chamber through a tube at the bottom of the test chamber and/or through a guide tube installed between the top of the crucible and the optical pyrometer to maintain inert atmosphere inside the chamber and secure the optical path by removing aerosols produced during the melting process.

2.2. Melting and solidification experiment

It is to be noted that only the melt crucible was used for the present melting and solidification experiment without melt delivery. The melt crucible was filled with a mixture of UO_2 pellets (2.7 kg), ZrO_2 powder (0.675 kg), stainless steel pellets (0.630 kg), B_4C powder (0.045 kg), and Zr in the forms of pellets (0.3855 kg) and ring (0.0645 kg) as an initiator for induction heating. The composition in terms of weight is UO_2 (60%), Zr + ZrO_2 (25%), stainless steel (14%) and B_4C (1%), which corresponds to the melt composition at Fukushima Daiichi NPP unit 1 when the molten core is relocated into the reactor lower head [15,16]. In addition, dummy ZrO_2 powder was added at the bottom of the crucible (0.2 kg) and on the top of the charging layer (0.4 kg) to protect the crucible during the melting process and reduce heat loss from the melt to the environment, respectively. A charging pattern is shown in Fig. 2 below. Two holes are initially made for gas venting and provision of an optical path for the pyrometer.

Melting of the charged materials is performed by increasing the power of high frequency power generator. The power was increased gradually while the melt temperatures were monitored by an optical pyrometer (1500–3500 °C within ±0.6% error) on the top and two C-type thermocouples (0–2320 °C, ±1% error). Fig. 3 shows the melt generation behavior along with the supplied electrical power. A Q-factor is a measure for the coupling of melt and induction heating [12]. That is, Q-factor decreases as the amount of melt increases, and thus the melt generation process can be monitored in real time. The electrical power was increased gradually up to 67 kW. As the Q-factor and melt temperature reached a plateau and was maintained long enough at this value, it was judged that the charged materials were completely melted. Then, we stopped the power supply and terminated the experiment.

2.3. Chemical analysis of solidified corium samples

For the destructive and non-destructive chemical analysis, nine samples were taken from the solidified corium ingot (#1-#9) after the experiment.

The elemental composition of the nine solidified corium samples was determined by Jobin-Yvon Ultima2 inductively coupled plasma atomic-emission spectrometry (ICP-AES) and Eltra CS800 elemental analyzer (EA). Further information on the chemical composition and structure was obtained from X-ray diffraction (XRD) analysis using Bruker D8 Advance X-ray powder diffractometer with CuKa radiation and scattering angles of 10°–70°. For ICP-AES measurement, the solid samples were dissolved using HCl, HNO_3, and HF. Furthermore, microwave-assisted digestion was used to dissolve any insoluble residues of the samples completely. The elemental concentration of U, Zr, Fe, Ni, Cr, and B was measured using ICP-AES, and the carbon concentration was measured using EA.