Progress in Nuclear Energy 106 (2018) 345–356

Contents lists available at ScienceDirect

Progress in Nuclear Energy

journal homepage: www.elsevier.com/locate/pnucene

An investigation on the physical and chemical behaviors of fuel debris during severe accident progression

JinHo Song*, BeogTae Min, SeongWan Hong, SangMo An, SeongHo Hong

Korea Atomic Energy Research Institute, 989-111 Daedeok-daero, Yueong-gu, Daejeon, 34057, South Korea

ARTICLE INFO

Keywords:
Corium
Severe accident
Physical and chemical behavior
Oxidation index
U/Zr ratio

ABSTRACT

Present paper investigates the potential configuration of post-accident fuel debris for a severe accident in a nuclear power plant. The physical and chemical behaviors of molten core material are investigated by experimental efforts. A series of melting and solidification experiments utilizing a cold crucible technique was performed using corium of 4.5–15 kg at various compositions. Considering that U/Zr atom ratio and Zr oxidation index could vary substantially during the accident progression, corium melts at compositions with U/Zr in the range of 0.94–1.14 and Zr oxidation index in the range of 36.3–100% were investigated. It was found that the solidified corium separated into a metal rich and an oxide rich layer in cases of partially oxidized corium. Depending on the compositions, metal rich layer was placed either on top or at the bottom of the oxide rich layer. Inductively Coupled Plasma-Atomic Emission Spectroscopy was used to estimate the chemical compositions of the layers and X-Ray Diffraction was used to find the crystal structure of the layers. In the second series of experiments, a molten corium of 13 kg in the form of liquid jet is poured into a pool of water. Melt water interaction resulted in fragmented and quenched particles with a range of size distributions. Scanning Electron Microscope and Electron Probe Micro-Analysis were used to analyze the physical and chemical behaviors of the particles. Partially oxidized corium melt resulted in a quenched particles with irregular shape. The composition of the particles varied among different particles and also within a particle. Fully oxidized melt resulted in quenched particles with homogenous composition among different particles and homogenous distribution of composition within a particle. The shape of these particles was spheroid in most of cases.

1. Introduction

In the Fukushima Daiich Nuclear Power Plant (FDNPP) accident, it is estimated that the nuclear fuels in the core and structural materials are molten and relocated to the lower head of the reactor and some of them is discharged to the floor of the pedestal area through a breach in the reactor vessel in unit 1, 2 and 3 reactors (IAEA, 2015). The configuration of solidified molten core material in various shapes in the Fukushima Daiich nuclear power plants is of interest, as it will give us the knowledge for better understanding of core melt progression and it will provide us crucial information necessary for the decommissioning process. Until recently, TEPCO had difficulties in assessing the configuration of solidified molten core material which is called fuel debris, due to a high dose rate. As an example, use of cosmic rays for an imaging of the reactor core gave only partial view of the fuel debris location (TEPCO, 2015). It is estimated that completion of decommissioning process will take another decades (TEPCO, 2018).

The decay heat generated by the nuclear fuel results in a fuel failure in combination with the inability to supply cooling water to the reactor. The failure of the fuel is characterized by hydrogen generation from an oxidation of cladding surrounding the fuel, and subsequent melting of the fuel. During the melting process, the molten fuels interact with neighboring structure including the control rods and internal structures. Therefore, the molten material inside the reactor consists of various materials including UO_2, ZrO_2, Zr, stainless steel, inconel, and control rod materials such as B_4C or Ag-In-Cd. The amount of ZrO_2 depends on the amount of oxidation of Zr cladding of the fuel. Likewise, the amount of stainless steel and control rod material would depend on the accident progression.

The core melting progression is a stepwise process which results in a melting and liquefaction of core materials at different temperatures, which consists of UO_2, ZrO_2, Zr, stainless steel, control rod materials like B_4C (Hofmann, 1999). Therefore, the composition of molten core material, which is called corium, would vary during the progression of the accident. In the case of the TMI accident, nuclear fuels consisted of 94,000 kg of UO_2 fuel, 24,000 kg of zirconium. The UO_2 fuel was

* Corresponding author.
E-mail address: dosa@kaeri.re.kr (J. Song).

https://doi.org/10.1016/j.pnucene.2018.02.028
Received 28 June 2017; Received in revised form 15 November 2017; Accepted 28 February 2018
Available online 05 April 2018
0149-1970/

dissolved by zirconium and form (U, Zr)O_2 melt. It was estimated that about 43% of Zr was oxidized and some of structural metallic material was also oxidized (Akers et al., 1990). The end-state configuration of TMI-2 core was examined by the mechanical probing and core boring operations.

An international research program of OECD/BSAF for an analytical prediction of accident progression and configuration of fuel debris has been on-going and the first results of predictions are reported. In the case of FDNPP unit 1, which was considered most damaged among three damaged reactors, it is estimated that the reactor core was severely damaged and a major portion of the core materials melted and relocated to the lower head of the reactor vessel and farther to the pedestal area. The composition of the relocated debris predicted by the OECD/BSAF participants for unit 1 were UO_2 at 50 to 60 weight percent, zirconium metal at 10% weight percent, ZrO_2 at 10%–40% weight percent, and stainless steel metal at 10%–30% weight percent. It is noted that the range of the amount of zirconium oxide had big uncertainly (Nagase et al., 2016).

The strong influence of material composition on the configuration of core material mixtures has been demonstrated (Kim and Olander, 1988; Gúeneau et al., 1998). Experimental investigations including MASCA have shown that when a sufficient amount of zirconium metal is available, then metallic uranium migrates to the metal layer. The transfer of species between the U, O, Zr, and the steel resulted in a density increase of the metallic phase which led to an inverse stratification with a metallic layer relocating below the oxidic pool (Bechta et al., 2008). The separation of metal rich layer and oxide rich layer is explained by the liquid immiscibility in an O, U, Zr, Fe model corium in a thermodynamic phase equilibrium (Gúeneau et al., 1998; Bechta et al., 2008; Seiler et al., 2003).

Solidified molten corium could reside in the reactor core region or it could relocate to the lower head region. If the reactor vessel had breached, the molten corium would be discharged on the floor of the pedestal area. These fuel debris could be in the form of solidified ingot or particulate debris. If there has been molten corium water interaction, the molten corium could have been fragmented and quenched into particles.

The configuration of fuel debris would give a clue to the core melt progression process which occurred in the reactor. Also, depending on the configuration and composition of the fuel debris in the reactor and on the floor of pedestal area, proper tools and method of retrieval process have to be developed and used. In this paper, configurations of fuel debris after a severe accident are investigated experimentally for corium melts at partially oxidized compositions and at full oxidation compositions to provide an insight on the configuration of fuel debris during the severe accident progression in a typical nuclear power plant including FDNPP.

2. Methods of investigation

Two kinds of experiments were performed for an investigation of potential configuration of fuel debris. The first series of experiment was a melting and solidification experiment similar to that of MASCA to investigate the configuration of solidified core material inside the reactor. The second series of experiments is a melt water interaction test to investigate the morphology of fuel debris during molten core and water interaction. A prototypic reactor materials at various compositions were used for these experiments. Physical and chemical analyses of the fuel debris were performed.

2.1. A melting and solidification experiment

A cold crucible melting technique is used to generate corium melts at various compositions. A cold crucible used in the experiment is shown in Fig. 1.

A cold crucible consists of a water distribution chamber and palisades like fingers on top of the chamber. The fingers and distribution chamber are made of copper and inside of which water is forced to circulate to provide cooling. Outside the finger, induction coil is located to provide induction heating by radio frequency generator. A mixture of UO_2 pellets, ZrO_2 powder, metal chips are used to fill the cold crucible in several layers. The center hole is for the measurement of melt temperature by a pyrometer. A more detailed description of cold crucible can be found in Hong et al. (2003). The power from radio-frequency generator is imparted among the induction coil, the cold crucible, and the charged material. A molten liquid pool is formed inside the crucible with thin crusts at the interface of molten liquid pool and the inner boundaries of the crucible. There is a convective motion in the liquid pool due to a electromagnetic force and an internal heat generation. This is similar to the prototypic reactor situation, where the convective motion in the molten corium pool is induced by the volumetric heat source of decay heat. After melting is completed, which can be monitored by the temperature measurement by the pyrometer and power supply curve, the power supply to the induction coil is stopped. Then, the corium melt in the cold crucible is solidified by the heat loss.

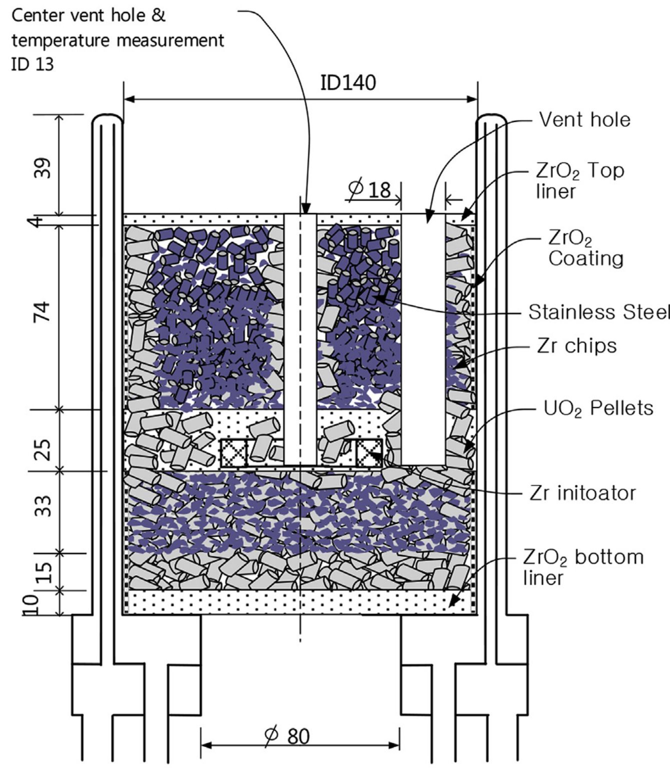

Fig. 1. Typical charging pattern for the cold crucible.

Typical shape of solidified melt after the experiment is illustrated in Fig. 2. Top crust in the upper part of the crucible consisted of un-melted UO_2 pellets and zirconia powder. Since charged material in Fig. 1 has void spaces, the liquid pool of molten material is formed below the top crust. Between the molten liquid pool and inner boundaries of the cold crucible, a thin crust layer is formed which became an insulation layer between the hot molten liquid and fingers of cold crucible at room temperature. Depending on the compositions of corium, the solidified corium ingot were at different configurations. The solidified ingot could be in the form of homogenous mixture, or stratified layers at different compositions. In Fig. 2, it is shown that the solidified melt consisted of two separated layers. Upper part of the crucible is occupied with top crust which consisted of un-melted UO_2 pellet and zirconia powder. Lower part consisted of two layers. The top part of the solidified ingot has cracks on top, which could be due to the contraction during the solidification process. This configuration of separate layers will be