Planning Lecture of Review Committee on Decommissioning of the Fukushima Daiichi NPS

Progress of R&D for the decommissioning of The Fukushima Daiichi NPP

(3) Topics on Decommissioning Researches: Analysis of sim-debris of the bundle degradation tests

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1. Introduction

This year was a 10 years memorial date after large-scale severe nuclear accidents at the TEPCO HD (Tokyo Electric Power Company Holdings Inc.) Fukushima Dai-Ichi Nuclear Power Station (further referred to as 1F) had happened. A cooling function of the Unit 1 got out of control after the tsunami strike and ended up by an explosion the next day. Increased dose rate and continuing aftershocks of high magnitude exacerbated by a questionable accident management [1] resulted in a cascade of three meltdowns occurred in the units 1, 2 and 3 and three explosions destroyed buildings of the units 1, 3, 4, even though the Unit 4 was shut down (under maintenance) [2]. This scale of the accident was beyond all assumptions made by promoters and antagonists of nuclear energy.

Now TEPCO is facing a challenge of a complete decommissioning of the 1F as a measure of the recovery of the lands of the Fukushima prefecture and prevention of the further leaking of radioactive substances out of the damaged reactors. However, decommissioning of 1F is an extremely difficult task, since a gap in understanding of the accidents propagation does not allow us to get the knowledge on the fuel debris final distribution and properties in the damaged units. The direct investigations were also minimal because of the harsh radioactive environment in the reactor buildings.

Collaborative Laboratories for Advanced Decommissioning Science (CLADS) was established in JAEA based on the acceleration plan for the 1F decommissioning to assist it by making R&D on the current challenging problems. The main objective of the studies was the investigation of a boiling water reactor (BWR) bundle degradation mechanisms in various atmospheres. Present test programme was focused on an early stage of a severe accident before melting of fuel materials such as UO_2-ZrO_2. Analysis of an accumulated data is going to be an input data for development of a multi-scale severe accident (SA) model.

2. Experiments

2-1. LEISAN facility

Large-scale control blade and bundle degradation tests were performed using a Large-scale Equipment for Investigation of Severe Accidents in Nuclear reactors (LEISAN) [3,4]. The facility was capable of going up to 1800 °C only in pure Ar firstly [5]. The heaters were placed in the upper furnace part making a gradient roughly 500 °C/m. Large-scale bundles of approximately 1.2 m-long with 20 claddings surrounded by channel boxes and one control blade were tested in Ar. An option of adding a steam+Ar mixture was added [6]. For this, a muffle capable to resist high temperatures in steam was developed. Outside of the muffle were heaters in Ar, inside was hot steam-argon mixture and a bundle.

In recent years, the facility had become additional thermocouples, which allowed measuring the temperature filed more precisely and a quadrupole mass-spectrometer for exhaust gas measurement [3,7]. One of the key features of the facility was the possibility of making in situ video, which helped to follow the features of melt progression (especially local exothermic reactions) and debris formation and relocation in situ right on the monitor. There is a further plan to equip the facility with an electronic low-pressure impactor for enhancement of aerosol study.

2-2. CR and CRFCB tests

One of the first tests CR-1 (CR stands for Control Rod) was performed with only one control blade rod [5]. In that case melting occurred without an influence of Zr. It was necessary to understand the pure behaviour of the control blade melting as a starting point of the bundle degradation. On the video the first place of the blade's sheath melt-through happened in the direct contact of the inner tubes, filled with B_4C granules, with the sheath of the control blade. No serious exothermic heat release was detected by the thermocouples (due to small number of them), only a small temperature increase was visible on the video. It gave the idea, that the energy of the Fe-B eutectic interaction is rather small to induce excessive heating damage. Oxidation of the Fe-rich melt was negligible because of Ar environment. That is why, in the next tests, Zircaloy-4 channel boxes together with one control blade were tested in CRFCB test (Control Rod Fuel Channel Box). The tests using the simple bundles revealed an impact of Zr on the melt formation and debris shape and relocation ability. It was clear that the debris chemical composition is important. Moreover, the degradation happened with a large exothermic energy release, even though the atmosphere was pure Ar, which was observed on in situ video [5].

2-3. CRFCBF tests

Control Rod Fuel Channel Box Fuel rod (CRFCBF) tests were performed in Ar and in steam atmosphere to grasp the differences of the bundle behaviour under steam-rich and under steam-starved conditions [6]. The tests showed a severe degradation of the channel boxes under pure Ar and steam-starved conditions. It happened because the oxidation layer on the surface of channel boxes tend to self-dissolve in the bulk of alloy by oxygen diffusion into the bulk. In the absence of the oxide layer, nothing prevented low-temperature Zr-Fe eutectic formation. Relocating melt solidified and created a blockage in the lower half of the bundle. Due to such mechanism of relocation and solidification, it could not penetrate far in the lateral direction. Probably, to propagate laterally two special conditions should be fulfilled: a secondary liquefaction should happen and the way down should be blocked completely.

In the framework of CRFCBF tests, only two kinds of debris were analyzed. The first material emerged in the region of high temperatures from the top to the middle of the bundle. The second material was able to relocate right to the bottom of the furnace, though the temperature there was low to expect any molten material. For Zr-Fe containing melt agglomerates it was easier to relocate in the axial direction so deep. Steam-rich conditions, according to the conclusion, may significantly change the course of the accident, however starting from which threshold of preoxidation and steam flow rate, was not clear.

2-4 CLADS-MADE

The recent tests in the framework of the CLADS Mock-up Assembly DEgradation (CLADS-MADE) programme were developed in close connection to the situation in the units of 1F. The test scenarios were developed taking into account the plant data widely published by TEPCO [8,9], IAEA [10], INPO [11] and the others. For the Unit 2 the developed scenario included a transient phase with 0.4 K/s followed by a steam starvation phase. For the Unit 3 there was a constant oxidation during transient heating with 0.6 K/s. For the Unit 1 a scenario had a constant steam flow rate and 1 K/s heating rate. More details on the scenarios can be found in [4].

A series of complementary post-test investigations favorably distinguished CLADS-MADE work from the previous preliminary studies. Such methods as optical microscopy, scanning electron microscopy (SEM) with energy-dispersive X-ray spectroscopy (EDS), X-ray diffraction analysis (XRD) and Raman spectroscopy were used. These investigations advanced our understanding of the accident progression at the beginning phase of the 1F accidents. Let us discuss the main findings of the post-test analysis of the sim debris.

It was established that an oxide layer significantly influenced the interaction of molten Fe-rich control blade melt with Zircaloy-4 of the channel boxes. It prevented lateral melt propagation and promoted individual blockage formation in each bypass channel. Melt-through of the channel box do not occur until the zirconium oxide surface layer is dissolved in the bulk. In case of local damage to the oxide layer a local melt-through may easily occur involving materials under oxide layer into formation of eutectics [12].

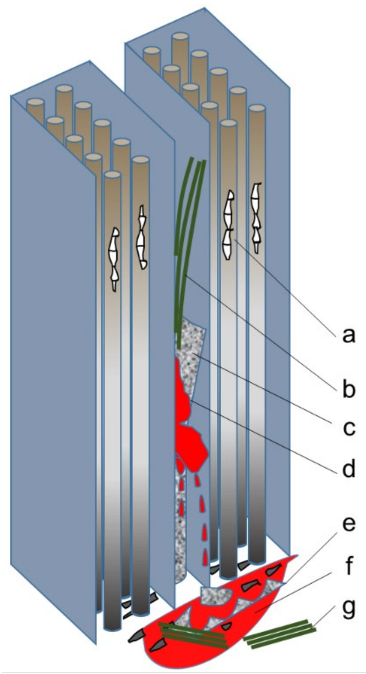

Figure 1. Scheme of a model bundle degradation: a – fully oxidized claddings, b – remaining control blade debris, c – remaining sheath, d – melt created blockage, e – relocated oxidized debris, f – relocated melt, g – relocated control blade debris.

The investigation by SEM EDS gave the first ideas on the phase compositions of the degraded materials at different elevations. Debris were consisting of oxidized materials, and still unchanged B_4C, surrounded and protected from direct contact with environment by a metallic part. The metallic part of debris consisted of two major volume fractions – Fe+C-rich and Cr+B-rich. The metallic part at the hottest elevations contained mostly solid solutions of B in (Fe,Ni) and borides of mixed composition (Fe,Cr)B. At the elevations where the reaction of B and C with melt was lower due to lower temperature, logically borides with higher Fe content $(Fe,Cr)_2B$ were identified. In the colder areas of blockage, almost unchanged stainless steel (SS) composition was detected with very small amount of dissolved B (<0.5 wt.% overall). However, this very small B concentration may play a crucial role as it makes Fe-rich materials extremely robust by precipitation of a small-scale B-rich domains. Those domains had local B enrichment up to 2.8 wt.% and was always assotiated with locally high Cr content. Obviously, Cr compounds with B were favourably stabilized in the liquefied melt [7].

Unfortunately, C was a material, which was difficult to detect by SEM/EDS. The investigation by XRD uncovered, that the debris contained not only Fe, Ni solid solutions with B and Fe, Cr borides, but also some small amount of mixed carboborides of $(Fe,Cr)_{23}(B,C)_6$ [3]. Raman spectroscopy was used to understand B and C compounds, which formed in the debris after reaction of SS melt and B_4C. It turned out that much of C after degradation and dissolution of B_4C remained unused. The traces of C, surrounding the reacted granules were detected by Raman spectroscopy.

Regarding the data obtained by the above-mentioned methods, a new mechanism of B_4C granules degradation by a graphitization was identified both under steam-starved and steam-rich conditions. In an oxidative environment remaining graphite was consumed by oxygen and C escaped from the melt in the form of carbon oxide gases (bubbles on the liquefied metal surface were confirmed in situ). In the case of steam-starved conditions C remained in the structure, which promoted $(Fe, Cr)_{23}(B, C)_6$ carboboride mixed phases formation.

2-5. Possibility of three types of debris

Let us try to find the analogue debris on the video of the PCV internal investigation performed by TEPCO [13]. A lot of stone-like debris was observed on the video. Visual appearance and post-test investigation data plus data provided by TEPCO on the primary containment vessel (PCV) investigation suggested that after an accident, debris can be divided into three general types: a) metallic, b) oxidic, and c) original parts, degraded by partial melting, mechanically slumped [14].

Metallic debris are forming especially in large amounts under reducing conditions. Having the lowest melting temperature among the core materials, they tend to relocate deeper into the lower elevations and block the water supply channels, thus making local conditions steam-starved. Metallic melt, depending on composition and temperature, had different ability to be oxidized on the way down. Fe, Cr, Ni - bearing melt has lower affinity to oxygen, thus less oxidizes during relocation. Additional to that, the temperature at which this melt becomes liquid is decreasing fast by addition of B and C. Zr-bearing melts have the highest possibility to be oxidized during relocation. But at the same time, Zr-bearing melts have tendency to relocate as big agglomerates, which make the ratio of surface to volume small. Having a bigger volume, cooling of the big agglomerates are slower. Thus, big agglomerates can reach deeper elevations of the core and can be the first in the lower head of the reactor.

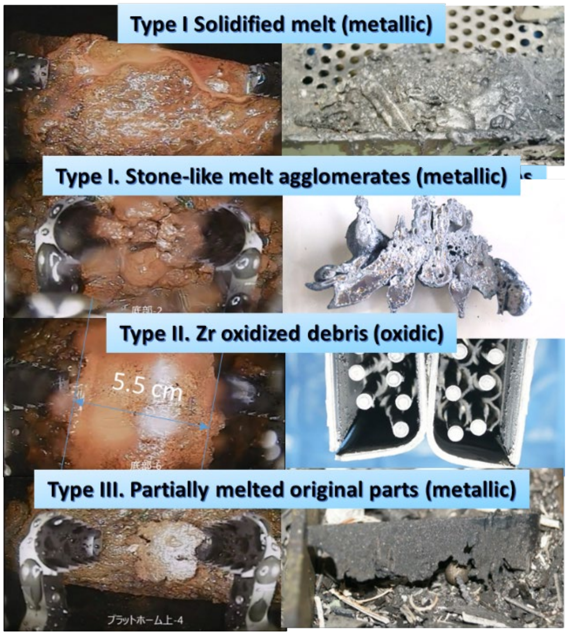

Figure 2. Three types of debris

After all alloys and eutectics had already relocated to colder elevations, there is a turn of oxides to start melting and creating a large pool of oxidized materials. Usually it is a mixture of UO_2 and ZrO_2. For example, there is no doubt that when a part of the oxidic pool in TMI-2 accident reached the RPV lower head, there was some layer of debris already there. It was not confirmed what kind of debris were there, but considering our observations it may be metallic debris.

Before the accident at 1F it was assumed, that no materials could survive such high temperatures as exist during the UO_2 molten pool relocation. A molten pool was considered to be uniform and no possibility for initial core materials survival was assumed. But a temperature gradient is absolutely normal situation even during severe accident inside of RPV. As a result, the PCV investigation in the Unit 2 had detected a top tie-plate came out of the RPV (Reactor Pressure Vessel) only partially melted. Such surprize was not predicted by any expert worldwide. However, in CLADS-MADE tests an upper part of the control blade had relocated in the same way to the bottom of the furnace. After this finding it became clear, that sim-debris from SA tests can be helpful in interpreting the real debris origin, ways of their relocation sometimes may give a rough idea about their chemical composition. Such attempt was made in [14].

2-6. Comparison with the internal investigation by TEPCO

In CLADS-MADE-01 test, control blade melt solidified on the surface of a channel box. Then, it was easily detached from it. One of the surfaces of such stone-like piece was flat. This process seemed to take place in the case of stone-like debris of the Unit 2. As soon as hot metallic melt came out of RPV through minor breaches, it solidified on the massive metallic beams of CRD supporting structures. No damage to those structures was possible because of the volume of melt. However, the mass of the accumulated solidified material would gradually increase, and finally some of agglomerates may simply fall down. Thus, stone-like debris is a result of the gradual process of the melted material coming out from the minor breaches, solidifying on the CRD massive stainless steels structures and then falling from above to the bottom of the pedestal [14].

The partially destroyed top tie plate was found lying on the top of the debris in the lower part of the PCV [14]. There is no guarantee that the other core parts like channel box or a control blade or even highly radioactive fuel pellets, did not relocate to the PCV. Indeed, some features of the observed debris remained on the round-shaped part of a control blade, 90° walls of the channel box with flat surfaces. Unfortunately, the opposite side of the debris was not investigated. That is why we cannot confidently conclude, that it was a part of a channel box or just a melt agglomerate solidified at a 90° angle in the corner between two massive CRD supporting beams. Additional investigations of the PCV of all units is a key factor of increasing our knowledge on the materials remaining there.

Solidified melt was found in the PCV, which looked much like the investigated debris of CLADS-MADE-01. In the test they were consisting of only metallic debris due to relatively low maximum temperature of the test. In the case of the Unit 2 debris, they can be a layered mixture of both: poured metallic and oxidic melt. In both cases, a large amount of the poured melt would have enough ability to propagate along the platforms and relocate to the lower elevations. In contact with platform, there was no deformation of the underlying platform beams. Cooling was obviously fast, probably the melt was nearly quenched by the difference in temperatures. It happened in the test as well. The melt in the test could not penetrate even through a thin 1 mm grating, because it had immediately solidified on it without any reaction occurring. After the test, it was easily detached. Probably, it can be expected in the case of Unit 2 PCV debris. The only concern is how to cut them into small pieces for withdrawal. The latest update of withdrawal technology was published in [15].

The presence of the large amount of accumulated molten material suggests two RPV–PCV relocation paths in BWRs – a) minor breaches near CRD joints with the gradual melt release in small amounts but in many places simultaneously and b) one major breach with the release of a large amount of melt and the top tie plate. The former mechanism was the cause for the stone-like debris; the latter explains the molten debris, which TEPCO was not able to move by the robot during PCV inspection. The top tie plate could penetrate obviously through a major breach. That is why if the major breach exist, degraded assembly parts can be observed on the top of the debris. From the author's point of view, such degraded assembly parts should be the first large-scale sample for a thorough investigation in the laboratory. It is going to give ideas on the accident progression of a particular unit by detection of changes in microstructure in this debris. Special interest is the molten edge of the degraded part. It may give the idea on the core composition at the moment of contact with this debris.

3. Conclusion

The work on analysis of metallic sim-debris have shown similarity between the test and real debris, which allowed the interpretation of the Unit 2 PCV debris origin and uncovered control blade degradation mechanism with formation of boron compounds in the metallic part of melt. Though the information was surely lacking, a rough estimation of the chemical composition of metallic debris and the place of the first solidification was made [14]. This can be a justification for the future accurate application of sim-tests for the understanding of the real situation inside the damaged reactors of 1F, which is expected to be an important method for the 1F forensics.

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