Effects of oxidation states of fuel cladding surface on crud deposition in simulated primary water of PWRs

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ABSTRACT

The purpose of this work is to understand the pre-oxidation effect of fuel cladding surface on crud deposition in simulated primary water of PWRs at 328 °C. Crud deposition tests were performed using ZIRLO™ (ZIRLO™ is a trademark of Westinghouse Electric Company LLC in the United States of America) claddings with three different oxidation states under sub-cooled nucleate boiling conditions. Boiling characteristics on the cladding surfaces were also simultaneously monitored using acoustic emission technique. The deposited amount on the pre-oxidized claddings was increased by up to 45%, compared with that on the as-received clean cladding. The number of boiling events also showed higher values on the pre-oxidized claddings. The effect of the pre-oxidized layers is discussed in the view point of the degree of sub-cooled nucleate boiling, zeta potential and wettability.

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1. Introduction

Corrosion products are released from the surfaces of the structural materials contacting with primary coolant in pressurized water reactors (PWRs) and are transported to the reactor core. These corrosion products are deposited on the fuel assemblies and are activated by a neuron flux in the core. The activated corrosion products are the major source of the radiation build-up and occupational radiation exposure. The corrosion products deposited on the surface of fuel assemblies are referred to as crud (an acronym for chalk river unidentified deposits) (Deshon et al., 2001; Byers and Deshon, 2004).

The deposition of the crud on fuel assemblies becomes a major concern with respect to material integrity and core performance: The crud on fuel assemblies can increase fuel cladding temperatures due to increased thermal resistance, resulting in accelerated fuel cladding corrosion (Cox, 2005; Edsinger et al., 2011). Above all, the thick crud may shift the core neutron flux from the top of the core to the lower half of the core along the axis of the fuel assemblies, called crud induced power shift or axial offset anomaly (EPRI, 2004). This phenomenon forces plant personnel to reduce the power output, owing to a decrease in core shutdown margin.

Laboratory simulation researches show that the rate of the corrosion product deposition is affected by several factors as follows:

heat flux, particle concentration, flow velocity, pH, and sub-cooled nucleate boiling (SNB), etc. The deposition rate of hematite particles increases linearly both particle concentration and heat flux (Kawaguchi et al., 1983; Asakura et al., 1978). The lower flow velocity along the fuel rod results in more deposition (Uchida et al., 1987). Several researches have reported that corrosion product is mainly composed of nickel ferrite, and its solubility has a minimum value at pH = 7.4 (Robertson, 1989; Song and Lee, 2003). Therefore, elevated pH operation from pH = 6.9 to pH = 7.4 has been implemented to minimize nickel ferrite precipitation in the core (Kim et al., 2012; EPRI, 2007). Especially, the crud deposition is accelerated on the wall surfaces under a heat transfer condition of SNB (Buongiorno, 2014; Byers, 2010).

The crud deposition has been a growing issue, as most utilities have moved to apply an extended fuel cycle and a high burn-up operation program. This is because the operation of a long-term fuel cycle has increased the difficulty of pH optimization (Song and Lee, 2003). Internal oxidation of zirconium-based fuel cladding materials may also limit these operation strategies. As the operation time passes, the thickness of internally oxidized layer in a fuel cladding material increases, which in turn increases the temperatures of the fuel cladding and may change the degree of SNB. Consequently, it is expected that the crud deposition behavior is affected by the oxidation state of the fuel cladding surface.

The purpose of this work is to elucidate the effect of the pre-oxidized layer in fuel cladding on the crud deposition behavior in simulated primary water at 328 °C and 130 bars. To achieve this

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goal, crud deposition tests are performed under a sub-cooled nucleate flow boiling condition. The boiling signals on the cladding surface are simultaneously monitored using the acoustic emission (AE) technique, and correlated with the deposition behavior.

2. Experimental methods

2.1. Specimen and solution preparation

ZIRLO™ cladding tubes, which are commonly used as a fuel cladding material, were used as the heat transfer tube. The cladding specimen has a dimension of a 9.5-mm outer diameter (OD) and an 8.3-mm inner diameter with a 55-cm length. One end of the cladding tubes was welded with a zirconium disc to give a leak tight joint. Some of tubes were pre-oxidized at 400 °C in the air of an electric chamber furnace for 3 days and 33 days, respectively. In this case, only the 25-cm long part from the closed tube end was heated in the furnace at 400 °C, and the remaining part of the tube (the open end side of the tube) was located outside the furnace. To prevent the oxidation on the inner surface of the tube, a vacuum was created on the inside of the tube. The tube specimens were degreased in acetone and washed by distilled water before the tests. The cladding tube was heated using an internal heater to provide the condition of SNB on the surface of the tube specimen during the crud deposition test. The gap between the cladding tube and the internal heater (OD = 8.2 mm) was filled with MgO paste.

Simulated primary water was prepared using high purity demineralized water with the resistivity above 18 MΩ·cm and nuclear grade chemicals of LiOH and H_3BO_3. The prepared solution was 3.5 ppm Li and 1500 ppm B in weight, which was used to simulate a primary water environment in PWRs. This solution of 200 L was stored in the solution tank and used as the simulated primary water recirculating the test loop system.

The sources of Ni ions and Fe ions for crud deposition were prepared using Ni- and Fe-ethylene diamine tetraacetic acid, respectively. The mixed chemical solutions of 1000 ppm Fe + 40 ppm Ni in weight were stored in the injection tank for injection to the test section.

2.2. Test loop system

Crud deposition tests were performed in an autoclave connected to a primary water recirculating loop system, which was shown schematically in Fig. 1. The loop system consists of the following main components: solution tank, high pressure pump (HP pump), pre-heater for solution inlet temperature control, chemical source tank, metering pump for mixed ion injection, back pressure regulator (BPR) for system pressure control, heat exchanger, test section equipped with a cladding tube.

A cladding tube was vertically located in the test section, as shown in Fig. 1. The inlet solution into the test section was pre-heated and the temperature of the flowing water adjacent to the cladding tube was maintained at 328 °C. The temperature of the internal heater was maintained at 380 °C to provide the condition of SNB on the surface of the cladding tube during the crud deposition test. The pressure of the test section was regulated at 130 bars. Dissolved oxygen was controlled to be less than 5 ppb and dissolved hydrogen was maintained at 35 cm^3/kg (STP) by controlling the hydrogen overpressure of the solution tank. The flow rate adjacent to the cladding tube in the test section was controlled at 5 m/s. After all these conditions are stabilized, we started to inject the mixed Fe and Ni ions into the test section through the metering injection pump. The mixed precursor ions were injected with a flow rate of 1.1 ml/min from the injection tank directly to the downstream of the preheater. This precursor solution is diluted in the simulated primary water stream and then its final chemistry is calculated to be 4.0 ppm Fe and 0.16 ppm Ni in the test section. Each deposition test was conducted for 120 h.

2.3. Boiling signal monitoring

The acoustic emissions generated from the sub-cooled nucleate boiling on the heated cladding surface were monitored using the acoustic emission technique for 5 min every 24 h during the deposition tests. An AE sensor was directly coupled to the upper end of the cladding tube, as shown in Fig. 1. The AE sensor was connected to a pre-amplifier, which was connected to the AE signal acquisition system. A low frequency AE sensor (type R3a, 30–75 kHz resonant frequency, MISTRAS from Physical Acoustic Corporation, USA) was chosen to collect the AE-boiling signals. In this test, the AE signals from the sensor were pre-amplified by a gain of 40 dB. The threshold value was set at 47 dB to eliminate the background noises. The obtained AE signals were analyzed using the AE-win software (Model: PCI-2, Physical Acoustic Corporation, USA).

2.4. Analysis for surface property

To characterize the wettability of the different specimen surfaces, the static contact angle was measured at 25 °C in air. A 3-μl droplet of deionized water was first placed by a syringe vertically down onto the specimen surface. The image of the droplet was then captured by a high resolution camera, and the static contact angle was finally determined by using image analysis software. Measurements were made at three different points on a specimen surface and were repeated two times at each point. In this paper, the mean value was reported together with the standard deviation.

The zeta potential measurements were performed using a Malvern Zetasizer Nano ZS system equipped with a surface zeta potential cell kit. For magnetite, the nanoparticles with an average size of 5 nm were diluted in high purity deionized water to be a concentration of 5 mg/l. After stabilization for 24 h, the prepared samples were transferred to a measurement cell. The surface zeta potentials of the three different claddings were also measured using the Malvern surface zeta potential cell kit. The flat sample was mounted between two electrodes and immersed in the solution containing tracer magnetite nanoparticles. This solution was the same as used for zeta potential measurement of magnetite nanoparticles. The apparent tracer mobility was measured at a number of different distances from the sample surface. Every measurement was conducted at 25 °C and repeated 3 times.
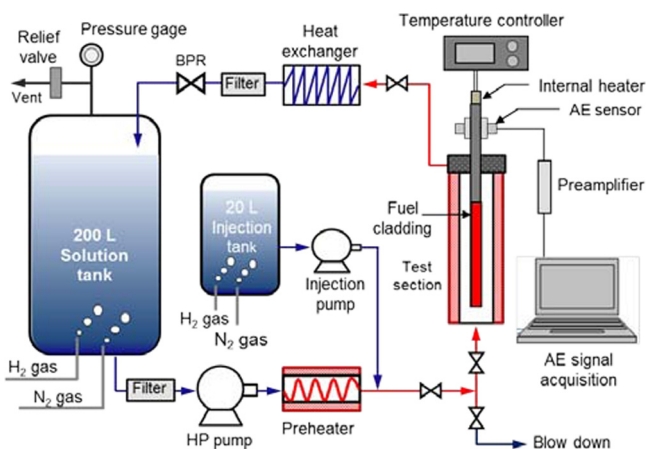

Fig. 1. Schematic of test system for crud deposition and acoustic emission measurement.