# Effect of Dissolved Hydrogen Concentration on Fuel Crud in a PWR Primary Environment

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## 1. Introduction

Dissolved hydrogen (DH) has been specified to be controlled in a range of 25-50 cc/kg-H<sub>2</sub>O (STP) in the EPRI guidelines to avoid any type of corrosion of the primary structural materials of PWRs [1]. Especially, a main issue regarding DH concentration in the primary environments of PWRs is to minimize primary water stress corrosion cracking (PWSCC) and general corrosion of Ni-based alloys. Many experimental studies have consistently reported that the PWSCC decreases as hydrogen concentrations deviate from a maximum peak point corresponding to the Ni/NiO phase transition [2,3]. In addition, the general corrosion rate decreases with the increase of the DH concentration [4]. However, research for a relation between DH and crud deposition on fuel assemblies still remains to be investigated. This is an important challenge to be resolved because fuel crud deposition is closely related to axial offset anomaly, power reduction, and accelerated cladding corrosion.

Therefore, the purpose of this work is to elucidate the effect of DH concentration on the fuel crud deposition in simulated primary water of a PWR at  $328^{\circ}$ C. We conducted crud deposition tests in the DH concentration range of 0-70 cc/kg-H<sub>2</sub>O for 10 days. In all tests, the sub-cooled nucleate boiling (SNB) behavior on the cladding surface was also periodically monitored using the acoustic emission (AE) technique and correlated with the deposition behavior.

#### 2. Experimental Methods

Crud deposition tests were performed using a circulating loop reported in our previous work [5]. The loop system consisted of the following main components: a primary solution tank, chemical source tank, high pressure pump, pre-heater for solution inlet temperature control, metering pump for mixed ion injection, back pressure regulator for pressure control, heat exchanger, a test section equipped with a cladding tube, and an AE acquisition system.

Table 1 summarizes the main experimental conditions for the crud deposition tests. Coolant temperature of the test section was maintained at 328°C and the system pressure was regulated at 130 bar. In order to provide SNB condition on the cladding tube surface, the temperature of internal heater inserted into the cladding tube was maintained at 380°C. Dissolved oxygen concentration was maintained below 5 ppb to eliminate the effects of oxygen on the crud deposition. The DH concentration was controlled to 0, 35, and 70  $cc/kg-H_2O$ . Crud sources were injected as two different concentrations with DH concentrations. In test-1, mixed Fe and Ni ions (each 500 ppb) were injected into the test section, and the crud source injected in test-2 was only Ni ion (1 ppm). However, in the case of test-2, the Fe ion source (2 ppm) was added to the primary solution circulating in the loop system.

Crud deposition tests were conducted for 10 days. During the crud deposition tests, the acoustic sounds of SNB emitted from the heated cladding surface were monitored using the AE technique for 300 s every 24 h. After the deposition tests, the cladding tube was cut into tubular segments for the measurement of deposit mass and microscopic analysis of the deposits. The concentrations of Ni and Fe were analyzed using an inductively coupled plasma-atomic emission spectroscope (ICP-AES). The morphology and composition of the deposits were analyzed using a scanning electron microscope (SEM) and an energy dispersive spectroscope (EDS).

Table 1. Main conditions of the loop system for crud deposition tests.

Test	Solution		DH	T(°C)/
	Primary water (ppm)	Crud source (ppb)	(cc/kg)	P(bar)
1	1500 B + 3.5 Li	500 Fe + 500 Ni	0, 35, 70	328/ 130
2	1500 B + 3.5 Li + 2 Fe	1000 Ni		

#### 3. Results and discussion

Because the crud deposition rate is accelerated through evaporation and drying-out of vapor bubbles on the cladding surface under SNB conditions, the SNB behavior is an important factor affecting crud deposition [6]. Therefore, it is necessary to determine whether DH concentration affect SNB behaviors or not. Figs. 1 (a) and (b) show the AE hit number for test-1 and 2 monitored in the DH concentration range from 0 to 70 cc/kg during the crud deposition tests, respectively. Here, the AE hit number means boiling-AE events for bubble dynamics on the fuel cladding surface, such as bubble formation, growth, departure, travel and collapse. Thus, it can be useful to compare quantitatively the relative amounts of vapor bubbles emitted on the cladding tube with DH concentration. At a given DH concentration, the AE hit number increased as the test time passed. However, there was no significant difference in the variation of the AE hit number with DH concentrations. This indicates that the SNB behavior on the fuel cladding surface was not affected by DH concentration.



Fig. 1. Variation of the boiling-AE hit number on fuel claddings with various DH concentrations during the deposition tests: (a) test-1 and (b) test-2.

Fig. 2 shows the SEM micrographs of the deposits on the cladding tubes with DH concentrations. When the Fe and Ni ions were injected (Test-1), polyhedral particles were deposited regardless of the DH concentration. In the case of Test-2, polyhedral deposits were formed in the DH concentration of 35 cc/kg and 70 cc/kg, whereas needle deposits as well as polyhedral deposits were observed at 0 cc/kg. In this result, the polyhedral deposit was nickel ferrite (NiFe<sub>2</sub>O<sub>4</sub>) and the needle-like deposit was analyzed as nickel oxide (NiO). It seems that the structure of Ni oxide is affected by DH concentration when the crud source is only Ni ion.

Fig. 3 shows the amount of cruds deposited on the fuel cladding tubes with DH content after deposition tests. The amount of deposits increased in the test-1 than the test-2. This implies that the crud deposition mass increases when the mixed Fe and Ni ions was injected. However, within the DH concentration of 0-70 cc/kg, the change in the amount of deposits was minute in both test-1 and test-2. This indicates that DH concentration has no meaningful effect on the amount of crud in the range of 0-70 cc/kg. Therefore, we need not consider

fuel crud deposition when we determine the optimal DH concentration for mitigation of PWSCC and general corrosion.



Fig. 2. SEM micrographs of the deposits in each DH concentrations after the deposition tests: (a) test-1 and (b) test-2.



Fig. 3. Amount of deposits on fuel claddings with various DH concentrations after deposition tests.

#### 4. Conclusions

This paper provides the effect of DH on the fuel crud deposition and sub-cooled nucleate boiling behavior in simulated primary water of a PWR. We conducted crud deposition tests in DH concentration ranges of 0-70 cm<sup>2</sup>/kg-H<sub>2</sub>O under a sub-cooled flow boiling condition at  $328^{\circ}$ C for 10 days. AE monitoring results revealed that the sub-cooled nucleate boiling on the cladding surfaces was not affected by DH concentrations. The effect of DH concentration on the amount of deposits was also minute in the DH concentrations of 0-70 cm<sup>2</sup>/kg-H<sub>2</sub>O. This indicates that the option of DH in the upper part of the 25-50 cc/kg range or beyond the upper limit of 50 cc/kg could be chosen to mitigate PWSCC

and general corrosion without a concern of fuel crud deposition.

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