Transactions of the Korean Nuclear Society Spring Meeting
Jeju, Korea, May 19-20, 2022

Experiment and Analysis of Iodine Plate-out on the Stainless Steel Surface

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1. Introduction

The radionuclide containment system for an HTGR consists of multiple barriers to limit the release of radionuclides from the core into the environment to insignificant levels during normal operation and a spectrum of postulated accidents (Fig. 1). Table I summarizes the anticipated forms of the fission products (FP) and their potential release behavior [1]. The release of FP such as Cs, Sr, Cd, I, and Ag during operation or accident scenarios depends on their interaction with graphite. Following the release from the fuel during normal operation, FP will be transported in the coolant gas, and plate-out onto surfaces in the primary system. FP borne dusts are highly mobile and potentially reach the coolant circuit, leading to the introduction of radioactive impurities into many components of the reactor. This distribution of circulating and deposited activity is important when estimating maintenance doses and establishes the initial condition of an accident scenario.

It is usual to test the interaction of a specific FP with a specific material at different temperatures to generate data on interaction kinetics as a function of temperature. Each material, each FP and each of its compound form have different kinetics and so, potentially, very large number of experiments are necessary. Accordingly, in addition to related experiments, research on the development of analysis codes is steadily progressing, and the development of analysis codes for FP is also in progress in KAERI [2, 3]. In this study, an experimental device is established to confirm the deposition characteristics of iodine on stainless steel (STS316), which is important among non-metal FP, and experiments were performed according to temperature.

2. FP Interactions with Alloy Structures

The interaction process is not the same for each FP: there is the initial heterogeneous interaction occurring on the alloy surface which is generally highly temperature dependent and reversible (Fig. 2). This first step is governed by mass-transfer mechanisms and vapor-pressure driven or governed by absorptivity. The adsorption or condensation is followed, depending on the FP, by absorption into the bulk or diffusion and chemical reaction in the bulk. This subsequent interaction may be only partially reversible [4].

The major parameter effect on plate-out is the temperature of both coolant and alloy surface. The vaporized FP can be either condensed down or continued on vaporized state by coolant temperature. We can imagine this fact more well if we see Fig. 3 that represents the vapor pressure change to coolant temperature for the I, Cs, and Sr [5]. Surface temperature also allows either plate-out on their surface or not. The other parameters related on plate-out amount are alloy materials and surface oxidation condition. Generally, oxidized surface captures FP more.

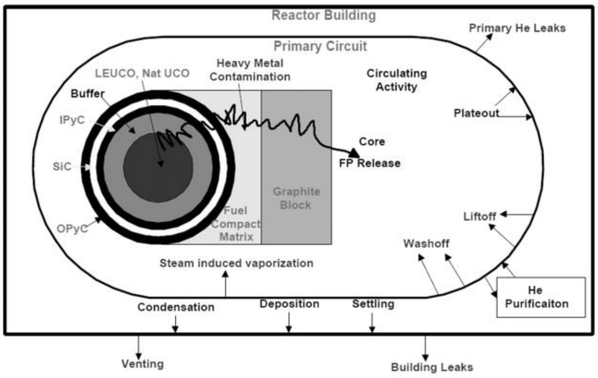


Fig. 1. Schematic of FP release barriers in HTGRs [1].

Table I: Interaction of FPs in Both Core and Primary Loop

Key FP	Interaction in core	Interaction in primary
I-131	Retained by PyC/SiC	Deposits on metals
Cs-137	Retained by SiC Matrix/graphite retention	Deposits on metals/dust
Sr-90	Matrix/graphite retention	Deposits on metals/dust
Ag-110m	Permeates intact SiC	Deposits on metals
H-3	Permeates intact SiC	Permeates heat exchangers
Xe-133	Retained by PyC/SiC	Removed by purification
Te-132	Retained by PyC/SiC	Deposits on metals/dust

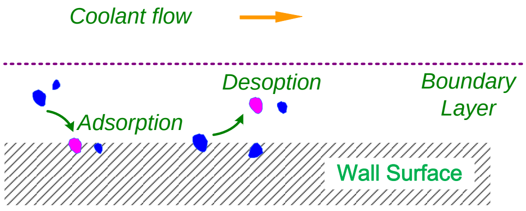


Fig. 2. Concept of FP interactions on the wall.

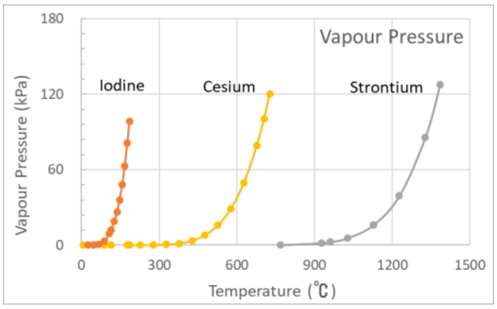


Fig. 3. Vapor pressure of FP [5].

3. Experiments

3.1. Description of Experimental Apparatus

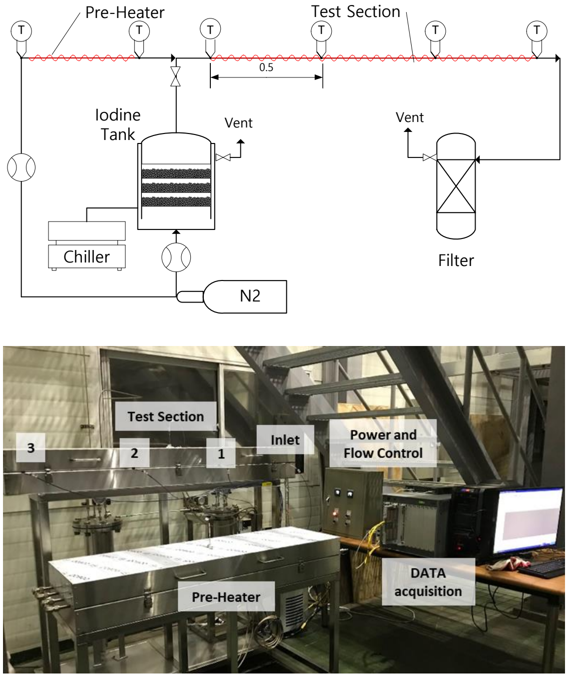
The non-metal deposition test apparatus was designed to determine the deposition characteristics of iodine on stainless steel (STS316). Nitrogen was used as the working fluid, and it was designed in such a way that the concentration of iodine entering the test section was controlled through iodine partial pressure control and flow rate control through temperature control in the iodine tank. The non-metal iodine deposition experiment apparatus consists of five parts: Iodine trapping apparatus (filter), iodine tank, preheater, and test section (Fig. 4).

In the experimental device, nitrogen, which is an iodine carrier gas, is supplied from the nitrogen tank, and passes through two flow meters through a branch of the gas flow path. Nitrogen gas controlled by the experimental flow rate is supplied to the preheater on one side and the iodine tank on the other side. The iodine vaporized in the iodine tank is transferred to the inlet of the test section along with nitrogen gas, passes through the preheater, meets and mixes with nitrogen gas heated to the experimental conditions, flows into the test section, deposits on the test section or exits the test section. The purified gas is discharged to the outside through the filter. The main specifications of the constructed device are as follows;

Working Fluid	Iodine
Iodine Flow	0~100 cc/min
Carrier Gas	Nitrogen
Nitrogen Flow	0~5 liter/min
Heater Power	5.0 kW
Design Temperature	500 °C
Design Pressure	1.0 MPa

3.2. Test Section

The test section for iodide deposition consisted of a 1.5m long 1/2" STS316 tube. The test section is pre-cut at 500mm intervals to facilitate the collection of test specimens after the test, and reassembled with tube fittings, and thermocouples are attached to each assembly location (Fig. 5). After the end of the experiment, specimens are taken from the test unit at 500 mm intervals. Iodine-deposited specimens are separated and extracted to a length of 4 cm near the temperature measurement site, and sealed in a tube filled with 10 ml of distilled water for ion chromatography analysis. The tube test section is installed inside the heating furnace and designed to maintain a constant test temperature condition during the experiment.



Note: number means location of thermocouples

Fig. 4. Plate-out experimental apparatus with diagram.

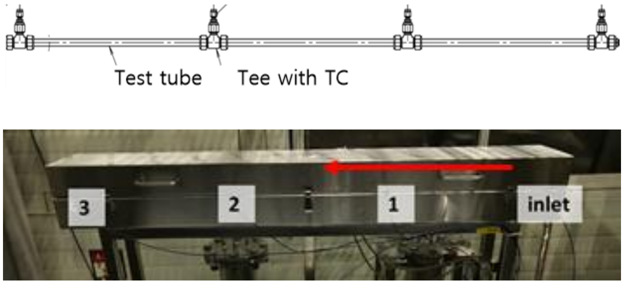


Fig. 5. Test section.

3.3. Test Results

As a result of carrying out a preliminary experiment to reach a steady state, it was confirmed that the temperature was maintained stable after about 4 hours. The experimental conditions are as follows.

○ Iodine tank temperature: 5℃, 36℃, 85℃ (3 cases)
○ Test section temperature: 50~60℃
○ Nitrogen (carrier gas) flow rate: 0.5 ~ 5.0 liter/min
○ Iodine flow rate: 5, 10, 100 cc/min

As a result of anion ion chromatography analysis, iodine is not detected in all specimens obtained at the iodine tank temperature of 5°C and 36°C. Even in the 85℃ experiment, only trace amounts are detected near 1ppm, the minimum detection limit of ion chromatography.

4. Analysis

4.1. Sorption Model

The adsorption (plate-out) is the accumulation of atoms or molecules onto the adsorbent's surface by either chemical or physical attraction (Fig. 2), while the absorption indicates a process that the substance being collected diffuses or penetrates into the other substance. For the adsorption, molecules are adsorbed just on the surface, but the molecules undergoing absorption are taken up by the volume, not by the surface. The experimental forms of sorption isotherms are used in order to evaluate the boundary layer concentration with ideal gas law, and it plays a major role in predicting the FP behavior near the surface of components within a gas-cooled system. In general, the boundary layer concentration can be correlated with the surface concentration. It is usually expressed as a non-linear form, but specific types of correlations based on experimental observations are available for typical species and specific surface materials at a certain condition.

A well-known empirical relation of FP concentration in boundary layer for sorption isotherms proposed by GA is as follows [1];

$$B = \frac{N_A p_{B.L}}{RT} \qquad (1)$$

Where,

$$p_{\mathrm{B.L}} = \sum_{l=1}^{3} b_l S_l^{n_l} \quad \text{for cesium,}$$

$$p_{\mathrm{B.L}} = \frac{S_l}{a_l \cdot (K - S_l)} \quad \text{for silver and iodine}$$

$$a_l = a_l^0 \cdot \exp\{-Q_l/(RT)\} \quad [\mathrm{Pa}^{-1}],\ l = 1, 2, 3$$

$$b_l = b_l^0 \cdot \exp\{-Q_l/(RT)\} \quad [\mathrm{Pa}/(\mu\mathrm{g/cm}^2)],\ l = 1, 2, 3$$

Here, the constants *a*, *b*, *n* and *K* are the parameters which depend on FP species, oxidized conditions, and pipe materials. The effects of the surface roughness are considered also.

4.2. Nodalization of POSCA Code

Analysis of the iodine deposition experiment was performed using the POSCA code [3] based on GA sorption correlation. Fig. 6 is a nodalized figure for POSCA code analysis of the iodide deposition experimental apparatus. It is simulated that nitrogen including iodine flows from the left. The total length of the test section was set to 1,500 mm, and the length of one node was set to 50 mm, and it consisted of a total of 46 nodes including 16 nodes from the iodine tank to the test section.

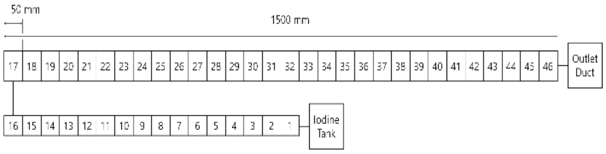


Fig. 6. POSCA model for the test section.

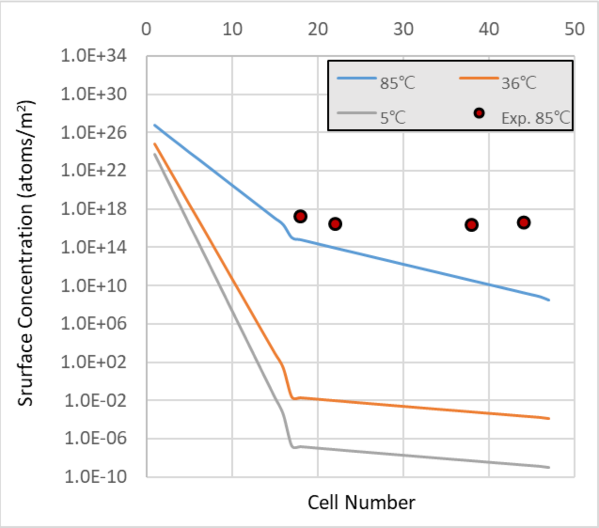


Fig. 7. Results of the POSCA analysis.

4.3. Results

Fig. 7 shows the results of the POSCA analysis for the iodine deposition experiment. As a result of the POSCA analysis, it was found that most of the iodine supplied from the iodine tank was deposited in the 800mm-long connecting pipe (nodes 1 to 16) before being injected into the test section. For this reason, in the case of the iodine tank temperature of 5 ℃ and 36 ℃ experiment with low iodine concentration, the amount of iodine deposited on the specimen is very small below the range of anion chromatography, so measurement data could not be obtained. Anion chromatography measurement range is very high in the range of 1.0ppm ~ 340ppm, that is, 3.70E+16 ~ 1.30E+19 atoms/m², unlike radioisotope measuring instruments that can measure at the atom level, making it impossible to measure a small amount of deposition and the measurement range It is also very narrow and can only be measured in a limited section. In the case of the iodine tank temperature 85℃ experiment with high iodine partial pressure, 0.6~4.3 ppm of iodine deposition was detected in the specimen, but the POSCA analysis evaluated that the deposition is 3 orders or more smaller than the experimental result (Fig. 7).

5. Conclusion

KAERI has prepare a lab-scale out-of-pile test apparatus to study plate-out characteristics of nonmetallic FP on the HTGR components. The amount of iodine deposition can be measured quantitatively with anion chromatography but its measurable range is limited as ppm level. In the case of the iodine tank temperature of 5 ℃ and 36 ℃ experiment, the amount of iodine deposited on the specimen is too small to exceed the measuring range of the anion chromatography.

From both the experiments and the POSCA analysis, it was possible to confirm the testable limit of the test apparatus for bare iodine without radioactivity. In the future, experiments below the range of ion chromatography need to measure the amount of deposition in real time using radioactive iodine and radioactivity measuring instruments.

ACKNOWLEDGEMENTS

This study was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (2019M2A8A1014757)

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