*Transactions of the Korean Nuclear Society Autumn Meeting*
*Goyang, Korea, October 24-25, 2019*

# Fission Products Plate-out Experiments with Multi-Couponed Tube-type Test Section

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## 1. Introduction

The radionuclide containment system for an HTGR (High Temperature Gas-cooled Reactor) 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. As shown schematically in Fig. 1, the three release barriers considered within the scope of safety analyses are: (1) the particle coatings, particularly the SiC coating (2) the primary coolant pressure boundary; and (3) the reactor building/containment [1]. Table I summarizes the anticipated forms of the FP (Fission products) and their potential release behavior [1, 2, 3]. 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. Metallic FP, such as silver (Ag), cesium (Cs), and strontium (Sr), become stored either within carbon components of the reactor or in the carbon dust in the primary loop components. 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. KAERI prepares a lab-scale out-of-pile test apparatus to study plate-out characteristics of the major metallic FPs such as strontium, silver and cesium [4]. The apparatus is able to simulate HTGR core temperature at helium or argon flow condition. We invented multi-couponed tube type test section to get many test coupons with various temperature conditions. The results of Sr plate-out experiments with this test section are discussed.

## 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. 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 [5, 6].

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. 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.

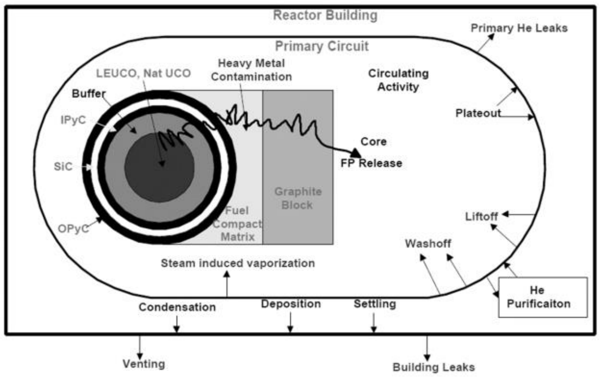


Fig. 1. Schematic of FP release barriers in HTGRs [1].

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<br>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 |

## 3. Experimental Apparatus

### *3.1. Description of Experimental Apparatus*

The experimental apparatus simulates the HTGR core temperature and reduced helium flow condition. The apparatus is an open loop that composed of a gas supply system, a preheater, a FP heater (Furnace), a test section, an air cooler and two kind of filters as shown in Fig. 2. The operating condition of plate-out test apparatus is as follows;

- Working Fluid — Helium or Argon
- FP Heater — ~ 2000 ℃

- Gas Temperature ~ 950 ℃
- Gas Flow ~ 65 liter/min
- Operating Pressure ~ 3 bar

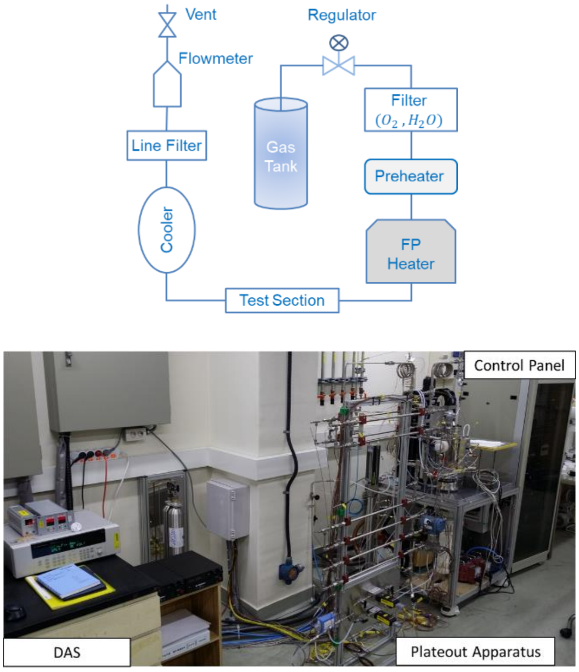


Fig. 2. Layout of the plate-out experimental apparatus.

*3.2. FP Heater Assembly and Flow Channel*

Fig. 3 shows the details of FP heater assembly and its major parts. IG-11 graphite which could withstand over 2000℃ in an oxygen free environment is used to compose of heater element, flow channels, channel liner and spacers, and FP crucible. The FP filled in crucible can be heated over 1500℃ by radiation emitted from the IG-11 liner. The liner is also heated by radiation emitted from heater element. The axial height of crucible can be adjustable up and down by using several adaptors at the bottom of lower flow channel.

Gas flow comes into from the bottom side of FP heater device and is heated at the lower channel inside and goes out through the many flow holes penetrated at the end of lower channel. The FP metal vapor entrained by flow comes into the crucible (crucible also has many flow holes). The gas flow containing FP vapor gets heated more when passing through the heating zone located in upper channel inside. The body temperature of the FP heater assembly can be controlled by a water supply system and the inside of assembly was filled with insulator to protect the body from very high-temperature. The body of the heater is designed to open and close automatically by a stepping motor.

*3.3. Measurement and Control*

The physical parameters which will be measured at the loop are the pressure, temperature, flow rate and the amount of plate-out metal on the tube surface. Three C-type thermocouples for monitoring graphite heater temperatures were installed near of graphite heater. K-type thermocouples were installed in the other locations. Gas flow rate is measured and controlled by mass flow controller connected between experimental apparatus and pressure regulator. The mass flow rate is measured again at the outlet of the experimental apparatus to confirm the flow rate. Gas entering into the coil test section is heated by a graphite heater. Power of the heater is controlled manually by using a potentiometer.

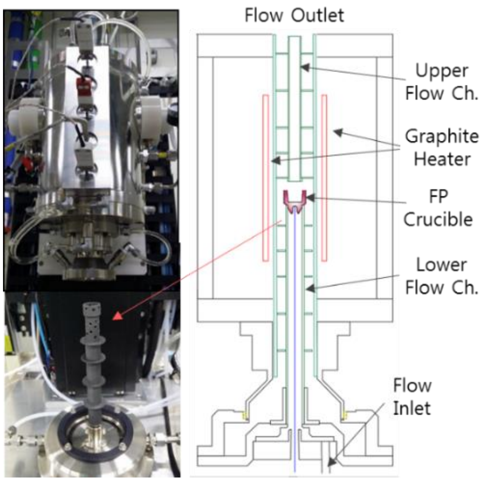


Fig. 3. Details of FP heater assembly.

## 4. Experiments

*4.1. Multi-couponed Tube-type Test Section*

A stainless steel 316 multi-couponed test section (MCTS) is invented to get sixteen test coupons at the four locations of Plate-out Observation Area (POA) as shown in Figure 4. The POAs where are blue colored parts in the figure 4 can be easily cut the coupons by a hand tube cutter after test. The POA is previously axially cut completely using EDM (electrical discharge machining). But the POA is previously radially cut incompletely in order to attach at the MCTS (keep the function of the flow channel in the MCTS itself) as shown in 3D layout of the figure 4.

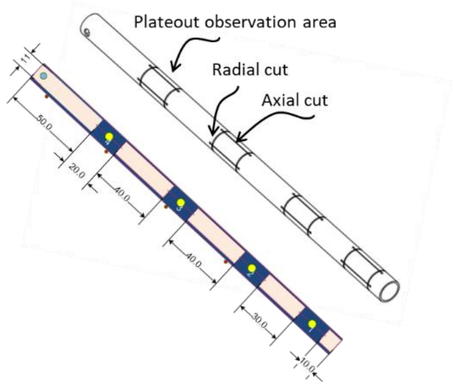


Fig. 4. Three dimensional layout of the multi-couponed test section and geometry of plate-out observation area.

There are four axial EDM cut at a POA, so we can obtain four test coupons at the same POA. Finally, we can get sixteen coupons at a MCTS because we located four POAs along to the gas flow direction in the MCTS.

0.5mm diameter K-type thermocouples are imbedded to the both outer wall and internal flow channel of MCTS. The thermocouples are fixed by steel wires at every POA location as shown in Figure 5-(a). The assembly of MCTS is inserted in the internally insulated housing that connected to the top of furnace (gas outlet) as shown in both Figure 5-(b) and 5-(c).

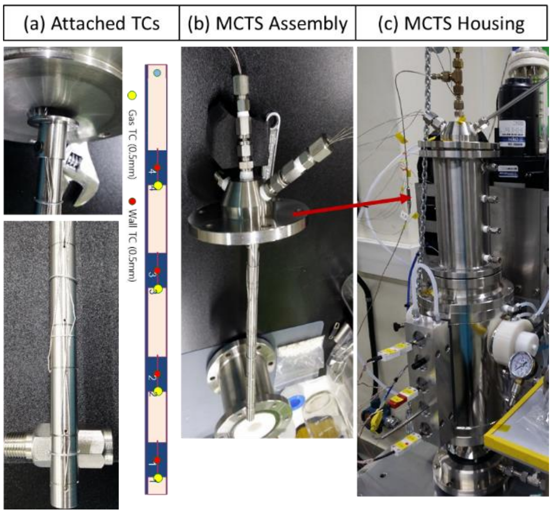


Fig. 5. Assembly of a MCTS with thermocouples attached both on the wall and in gas channel of all the POAs.

### *4.2. Experimental Condition*

The test condition is as follows,

- FP metal: pure strontium (Sr)
- Initial mass of Sr: 251.8 mg
- Sr temperature: 886 ~ 901 ℃ (liquid state)
- Test duration: 6.0 hours
- Argon flow rate: 3.6 liter/min
- System pressure (exit): 1.07 bar
- Heater power: 1065~1080 W

Crucible temperature is a major concern in the Sr plate-out experiment because it determines the amount of Sr release at the crucible. Liquid Sr is released with increasing temperature and the releasing amount of Sr greatly depends on vapor pressure. The temperature window of the liquid state Sr is from 769℃ (melting point) to 1384℃ (boing point).

We set the crucible temperature to 900 ℃. After reach of the crucible temperature near 900 ℃, steady-state is maintained during over 6 hours (Fig. 6).

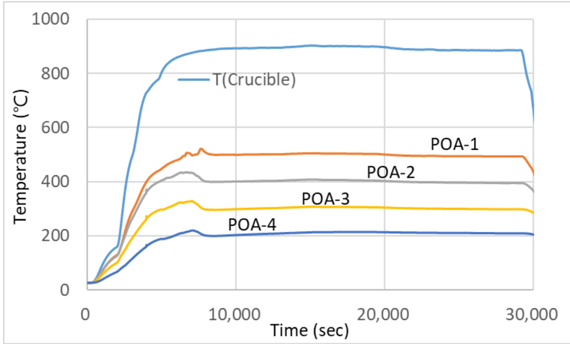


Fig. 6. History of gas temperatures in the MCTS internal channel.

## 5. Results and Discussion

After 6.0 hours steady-state test, 83% of initial strontium (Sr) in the crucible is entrained by argon gas flow and the Sr in argon gas flow is plated out on the inner surface of the MCTS. We collected 4 plate-out coupons at every POA location by cutting of the MCTS using a potable tube cutter. Three of four coupons are submerged in liquid Kerosene immediately after cut. Other one is exposed in air to check the Sr plate-out visually as shown in Figure 7-(c). The color of the inner surface of the coupons was dark gray at first when visual investigation soon after test, but the color is changed to white after longer time of exposure to air as shown in the figure. Most of outer surface of plated out layer is carbonized after exposed to air. This carbonized Sr compound is confirmed by FTIR (Fourier Transform Infra-red) analysis on the Sr compound exposed a month in air [7]. The FTIR analysis revealed the white colored Sr compound (created in air) as $SrCO_3$.

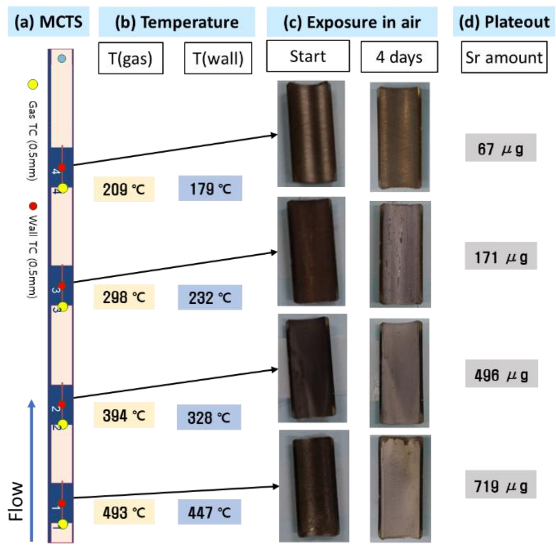


Fig. 7. Key test results (temperature profile of the MCTS, photos of the test coupons, and amount of Sr plate-out).

The amount Sr plate-out is measured by using the ICP-AES (Inductively Coupled Plasma-Atomic Emission Spectroscopy). ICP-AES results represent the Sr amount of plate-out is decreased along to the MCST outlet direction (Figure 7-(d)). Sr of 719 μg is plated out on the first POA, but Sr of 67μg is plated out on the forth POA. It means that the Sr vapor is immediately plated out to the channel wall. This explains why Sr is treated as perfect sink material in the FP plate-out model in most of design codes [8].

## 6. Conclusion

It is confirmed that the multi-couponed tube-type test section (MCTS) can provide many test coupons with various temperature conditions successfully.

83% of initial strontium (Sr) in the crucible is entrained by argon gas flow during 6.0 hours steady-state operation condition (Sr temperature of 900 ℃). The Sr vapor in argon gas flow is plated out on the inner surfaces of 16 coupons attached at a MCTS. Chemical interactions of Sr are observed visually from the various coupons. The color of the inner surface of the coupons was dark gray at first when visual investigation soon after test, but the color is changed to white after longer time of exposure to air. This white colored Sr compound is confirmed by FTIR analysis as $SrCO_3$. Meanwhile, ICP-AES analyses of other coupons represent that the Sr amount of plate-out is decreased along to the MCST outlet direction. It means that the Sr vapor is immediately plated out to the channel wall.

## ACKNOWLEDGEMENTS

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

## REFERENCES

[1] INL/EXT-10-17997, Mechanistic Source Terms White Paper, U.S. DOE, Idaho Falls, 2010.
[2] IAEA-TECDOC-978, Fuel Performance and Fission Product Behavior in Gas Cooled Reactors, IAEA, 1997.
[3] T. Burchell, R. Bratton, W. Windes, NGNP Graphite Selection and Acquisition Strategy, ORNL, 2007.
[4] S. D. Hong, N. I. Tak, B. H. Park, E. S. Kim, M. H. Kim, High Temperature Experimental Apparatus to Study Plate-out of HTGR Metallic Fission Product, Transactions of the KNS Autumn Meeting, Yeosu, Korea, 2018.
[5] R. Moormann, and K. Hilpert, Chemical Behavior of Fission Products in Core Heatup Accidents in High-Temperature Gas-Cooled Reactors Nuclear Reactor Safety, Nuclear Technology, Vol. 94, 1991.
[6] M. P. Kissane, A Review of Radionuclide Behavior in the Primary System of a Very-high-temperature Reactor, Nuclear Engineering and Design, Vol. 239, pp. 3076-3091, 2009.
[7] S. D. Hong, Y. K. Kim, J. B. Hwang, E. S. Kim, M. H. Kim, Out-of-Pile Strontium Plateout Experiments, Transactions of the KNS Spring Meeting, Jeju, Korea, 2019.
[8] D. L. Hanson, N. L. Baldwin, D. E. Strong, Fission Product Behavior in the Peach Bottom and Fort St. Vrain HTGRs, IWGGCR/2 (IAEA), Specialists Meeting on Coolant Chemistry, Plate-out and Decontamination in Gas-cooled Reactors, Page 49, 1980.