Out-of-Pile Strontium Plateout Experiments

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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. 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 FPs and their potential release behavior [1, 2, 3]. The release of FPs 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, FPs will be transported in the coolant gas, and plateout onto surfaces in the primary system. Metallic fission product (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 prepare 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.

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].

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 both Table II and Fig. 2 that represents the vapor pressure change to coolant temperature for the Cs, Sr and Ag. 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.

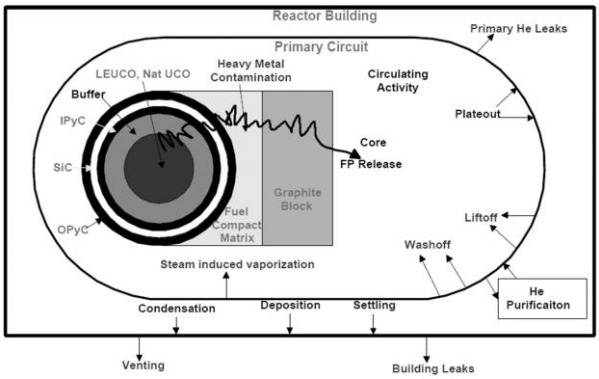

Fig. 1. Schematic of FP release barriers in HTGRs [1].

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

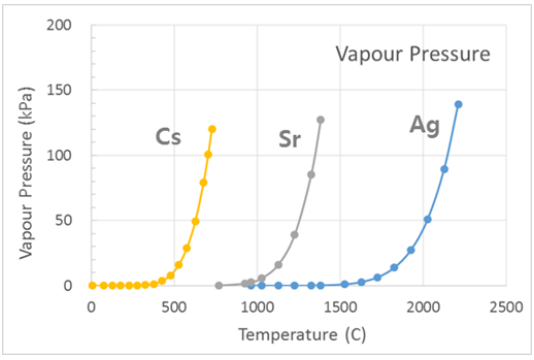

Fig. 2. Vapor pressures of FP metals [6].

Table II: Melting/boiling Points of the Major Metallic FPs.

Metal	Melting Point (℃)	Boiling Point (℃)
Silver (Ag)	961	2212
Strontium (Sr)	769	1384
Cesium (Cs)	28.5	705

3. Experimental Apparatus

3.1. Description of Experimental Apparatus

The experimental apparatus simulates the VHTR 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 a filter as shown in Fig. 3. 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

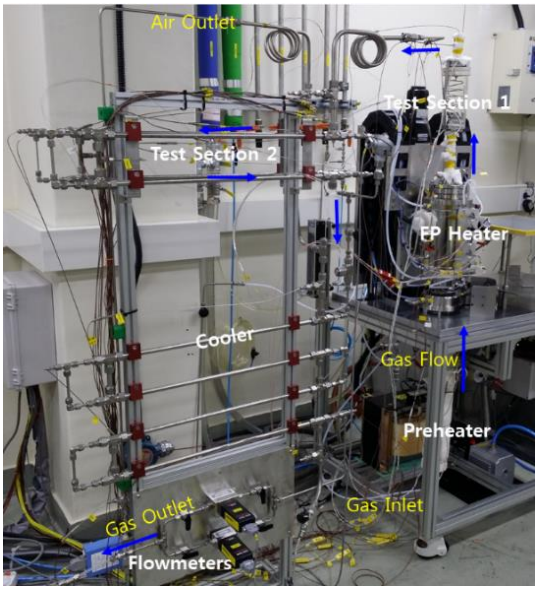

Fig. 3. Plate-out experimental apparatus in KAERI.

3.2. FP Heater Assembly and Flow Channel

Fig. 4 shows the details of FP heater assembly and its major parts. IG-11 graphite which could be withstand over 2000℃ in an oxygen free environment are 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 are entrained by flow come into the crucible (crucible also has many flow holes). The gas flow containing FP vapor get 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. Surroundings of both FP crucible and graphite heater are prepared four C-type thermocouples, one for measuring crucible bottom temperature and the others for monitoring graphite heater temperatures. All the other locations are installed K-type thermocouples. Gas flow rate is measured and controlled by mass flow controller connected between experimental apparatus and pressure regulator. The pressure regulator keeps the outlet pressure of gas supply tank less than 5 bar. 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 both a pre-heater and a graphite heater. Power of both heaters are controlled manually by using the potentiometers.

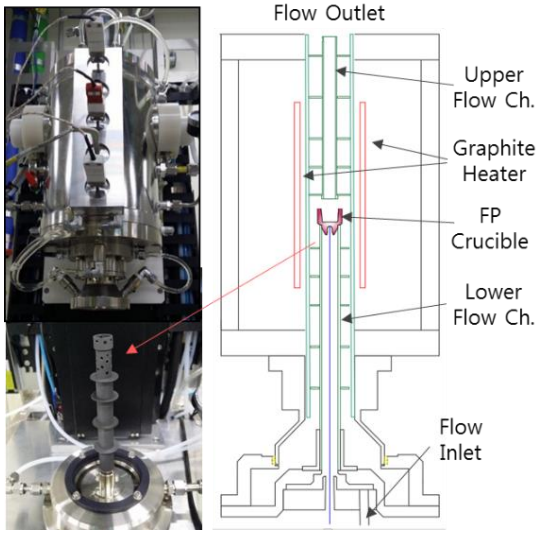

Fig. 4. Details of FP heater assembly.

4. Strontium Experiments

4.1. Strontium in RCS

Strontium which has a melting point of 769 ℃, it can be liquefied and slightly separated from the dust when

passing through the core. Some chemical reactions can occur if Strontium collides with impurities in the coolant, such as oxygen or hydrogen as well as FP-dust suspended through the coolant in a chemical compound form [7]. It can adhere to any component of the RCS or become suspended and circulate in the RCS according to the mass conservation law, either until all strontium are decayed or until it is captured by a filter in a helium purification system.

4.2. Quartz Visualization Test

A quartz visualization test section is installed at the top of upper channel that is easy to get the HTGR test condition (Fig. 5). The test condition is as follows,

- Initial mass of Sr : 209 mg
- Sr temperature : 1034 ℃ (liquid state)
- Test duration : 3.0 hours at 1034 ℃
- Argon velocity : 1.0 m/sec
- Gas temperature in the test section
  - Internal : 815 ~ 930 ℃
  - Outlet : 291 ℃

After reach the target temperature of 1034 ℃, steady-state was maintained for during 3 hours (Fig. 6). After test, 70% of the weight of initial strontium was lost and strontium and graphite dust plated out on the inner surface of the quartz test section (Fig. 7). The color of the inner surface of the quartz tube was black or dark brown at first when visual investigation soon after test, but it changed to white after longer time of exposure to air as shown in Fig. 7. It is also found that the condensed Strontium compound at the channel outlet is dropped on the top spacer of the upper channel (Fig. 5).

Fig. 8 shows the results of surface analysis of coupon long time exposed to air by using SEM-EDS (Scanning Electron Microscopy – Energy Dispersive Spectrometer). The SEM-EDS detected both oxygen (26 wt%) and carbon (12 wt%) with much of strontium (48 wt%) on the surface. The white colored material is postulated to be $SrCO_3$compound through the analysis of SEM-EDS.

It is very interesting that the carbon amount are reduced immediately at the monoatomic depth profile of the plateout surface of quartz coupon with XPS (X-ray Photoelectron Spectroscopy) point analysis as shown in Fig. 9. It means that during the experiments, either plateout pure strontium is oxidized on quartz surface or oxidized strontium is plated out on quartz surface. Then most of outer surface of plated out layer is carbonized after exposed to air. This carbonized strontium compound is confirmed by FTIR (Fourier Transform Infra-red) analysis on the strontium compound exposed a month in air as shown in Fig. 10. The FTIR analysis revealed the white colored strontium compound (created in the air) as $SrCO_3$.

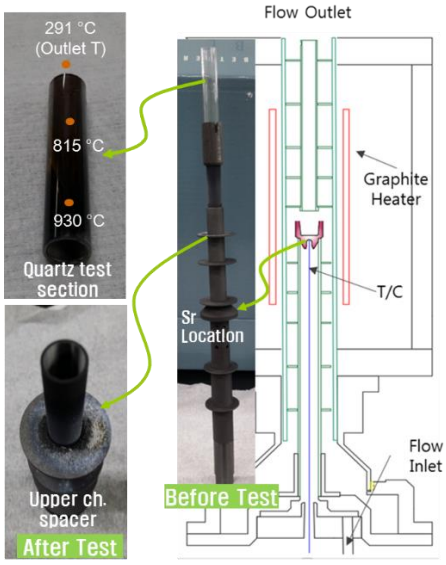

Fig. 5. Test section of before/after test (Quartz visualization test).

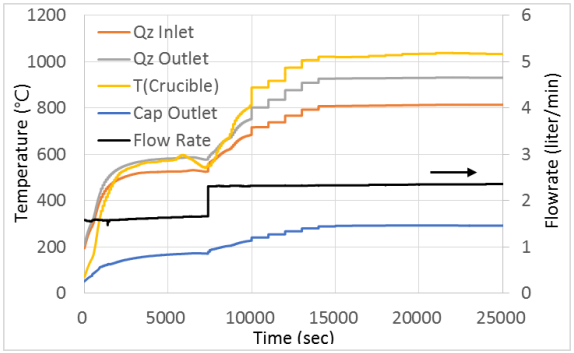

Fig. 6. Operating data of quartz visualization test (Sr/Gas temperatures and flow rate).

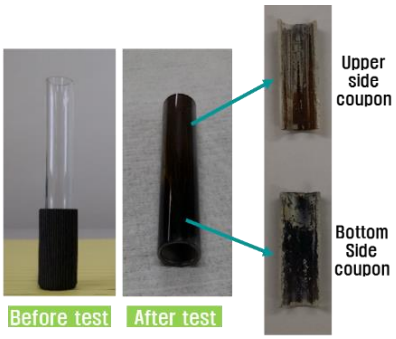

Fig. 7. Quartz test coupons.

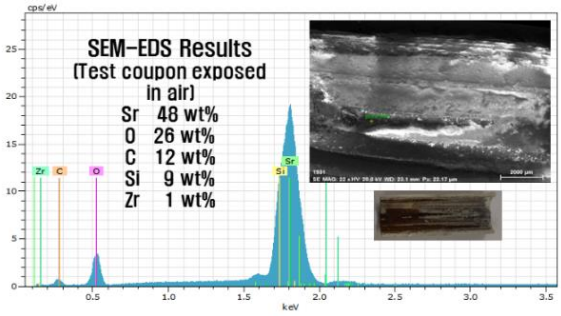

Fig. 8. SEM-EDS results of the upper side coupon.

5. Conclusion

KAERI has prepare a lab-scale out-of-pile test apparatus to study plate-out characteristics of metallic FP on the HTGR components. Strontium which has a melting point of 769 ℃, it can be liquefied and slightly separated from the dust when passing through the core. Chemical interactions of strontium are observed visually and quantitatively from the results of a plateout experiment with the analysis of SEM-EDS, XPS and FTIR. Either plateout pure strontium is oxidized on quartz surface or oxidized strontium is plated out on quartz surface during the plate out experiment. Then most of outer surface of plated out layer is carbonized after exposed to air. This carbonized strontium compound is confirmed by FTIR analysis as $SrCO_3$

ACKNOWLEDGEMENTS

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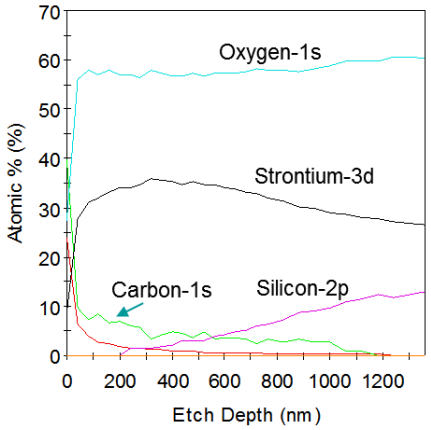
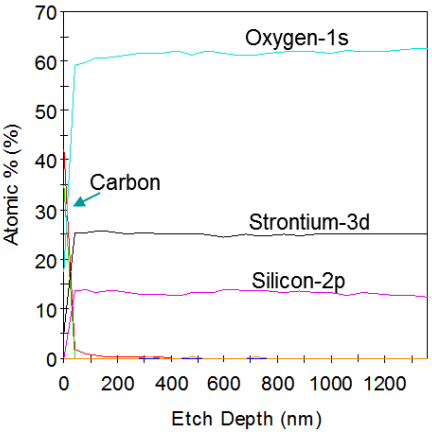


(a) Upper side coupon



(b) Bottom side coupon

Fig. 9. Monoatomic depth profile of the Sr plateout surface of quartz coupon (XPS point analysis).

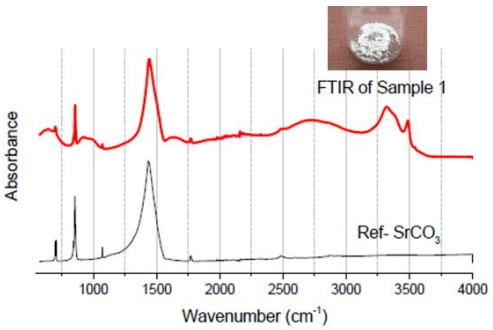


Fig. 10. FTIR analysis on the strontium compound exposed a month in air.