



## Chemical form consideration of released fission products from irradiated fast reactor fuels during overheating



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### ABSTRACT

Experiments simulating overheating conditions of fast reactor severe accidents have been previously carried out with irradiated fuels. For the present study, the chemical forms of the fission products (FPs) included in the irradiated fuels were evaluated by thermochemical equilibrium calculations. At temperatures of 2773 K and 2973 K, the most stable forms of Cs, I, Te, Sb, Pd and Ag are gaseous compounds, and their fractions are about 100% in an oxygen partial pressure range between  $10^{-5}$  and  $10^3$  Pa. Cs and Sb detected in the thermal gradient tube (TGT) in the experiments can take gaseous chemical forms of elemental Cs, CsI, Cs<sub>2</sub>MoO<sub>4</sub>, CsO and elemental Sb, SbO, SbTe, respectively. The chemical forms of Cs and Mo are sensitive to the oxygen partial pressure. By comparing experimental results and the estimations, it is seen CsI thermochemically behaves in a manner that traps it in the TGT, while elemental Cs trends to move as fine particles. The moving behavior of the gaseous FPs will obey not only thermochemical principles, but also those of particle dynamics.

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### 1. Introduction

During severe accidents in nuclear plants, the reactor core may become overheated and melt down, and then the radioactive materials within reactor containment functions will be released into the environment. To secure severe accident management with appropriate accuracy, the release behavior for radioactive materials must be understood well. There are some complicated processes from the reactor core to the environment depending on the type of reactor, temperature history, the atmospheric conditions, and so on. Aerosol materials formed from fission products (FPs), actinide elements, cladding materials, etc. play important roles in movement processes through the air to the environment. These aerosol materials will be generated by gaseous FP species growing as fine particles in an initial process. Therefore, it is important to comprehend aspects of the chemical form for FPs in evaluating the aerosol behavior since the chemical forms are thought to decide the aerosol properties such as the particle size (Buckle, 1991).

The chemical forms of gaseous FPs generated from overheated irradiated fuels have been investigated by a number of researchers with experimental methods (Collins et al., 1988; Boer and Cordfunke, 1997) and thermochemical methods (Götzmann, 1993; Cordfunke and Konings, 1993). In experimental methods, thermal gradient tubes (TGTs) for which the inner surface is linearly kept between one temperature and another temperature were utilized to determine the chemical forms of volatile FPs such as Cs and I. However, it is not easy to evaluate the other FP species than Cs and I, because they do not have appropriate radioactive nuclides for  $\gamma$ -ray measurements or they are less volatile compared with Cs and I. Therefore, thermochemical simulations are used to consider the chemical forms in overheated irradiated fuels including FPs.

A few experimental studies of fast reactor fuels with abundant Pu content have been conducted under an inert atmosphere expected to be in the initial release process (Sato et al., 2003, 2011, 2013). One of the features of FP release in fast reactor severe accidents is thought to happen under low oxygen partial pressure (Strain et al., 1993; Mignanelli and Potter, 1984) unlike in light water reactors. In the field of source term researches of light water reactors, the chemical forms of FPs will be taken into consideration as functions of oxygen potential and/or water vapor pressure.

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However, the considerations for gaseous chemical forms of FPs released due to overheating have not been made especially for under fast reactor fuel atmosphere.

The authors previously carried out experiments representing overheating conditions of irradiated fast reactor fuels (Sato et al., 2011). For the present study, the chemical forms of the FPs included in the experimentally irradiated fast reactor fuels were evaluated by thermochemical equilibrium calculations. By comparing experimental results with calculated estimations, considerations were made on how the gaseous chemical forms of FPs might influence the FP release behavior.

## 2. Summary of the procedures and results for the heating experiments conducted in 2008–2009 (Sato et al., 2011)

Three heating experiments were conducted to obtain helium release and retention behavior in mixed oxide fuels irradiated in the experimental reactor JOYO (Sato et al., 2011). To obtain information on the volatile FP release behavior, the trapping system for released materials (Sato et al., 2013) was used in addition to a TGT and filters.

### 2.1. Heating experiments

Details of the experimental device were shown in Sato et al. (2003, 2013). A  $(U_{0.8}, Pu_{0.2})O_2$  fuel pin irradiated to about  $65 \text{ MWd kg}^{-1}$  was cut for preparation of about 3 g specimens without the cladding material. Each specimen was put into a W crucible and heated to 2773 K and 2973 K for 500 s in an induction furnace (heating rate: 15 K/s).

The inner surface temperature of the TGT, which was installed vertically above the induction furnace, was kept between 1023 K (at the inlet side, the lower position) and 423 K (at the outlet side, the upper position). The temperature varied linearly between the inlet and outlet sides. There were separable sampling tubes along the inside of the TGT and filters were included in a stainless steel case at the top end of the TGT. Ar including impurity oxygen content of about 1 ppm was used as a carrier gas for gaseous FP chemical species; the gaseous FPs deposited on the inner surface of the sampling tubes depending on their volatility. The other released materials that passed through the sampling tubes could be captured with the top end filters.

During heating,  $\gamma$ -ray spectrometry measurements were made for the filters attached at the top end of the TGT to obtain trend data of FP release. After heating, the fractional releases for radioactive nuclides such as Cs-137 were evaluated by  $\gamma$ -ray spectrometry measurements of residual specimens in the crucibles. Moreover, the sampling tubes were separately measured to obtain FP profiles along the TGT.

### 2.2. Release rate and FP profiles along the TGT

From  $\gamma$ -ray spectra for the filters obtained during heating, Cs-134 and Cs-137 could be detected, and then the count rates for Cs-137 as a function of heating time were obtained for the 2773 K and 2973 K temperature experiments. Although the measuring time for each spectrum differed for these experiments, 98% and 100% release percentages of Cs-137 were obtained at 500 s and 450 s for 2773 K and 2973 K experiments, respectively. Here, the fractional release percentages of 98% and 100% for both experiments were evaluated from measurements for specimens before and after heating. Therefore these measured count rates could be evaluated into the fractional release as seen in Fig. 1. These time dependence profiles were not linear, but the average release rates of Cs-137 for these times

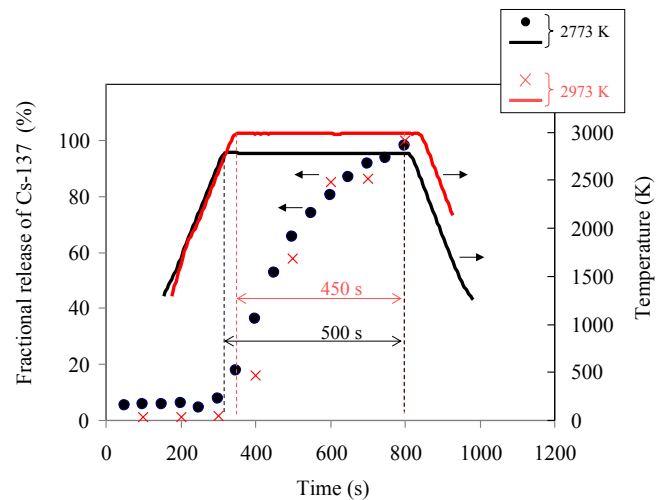


Fig. 1. Fractional release for Cs-137 as a function of heating time in the 2773 and 2973 K experiments.

were evaluated as 11.8%/min and 13.3%/min for 2773 K and 2973 K experiments, respectively. These average release rates were in good agreement with literature values (United States Nuclear Regulatory, 1981).

Three nuclides, Cs-137, Cs-134 and Sb-125, were detected in  $\gamma$ -ray spectra for sampling tube contents. Fig. 2 shows the count rates for Cs-137 and Sb-125 along the TGT obtained from both experiments. Only the Cs-137 profiles need to be used to understand physicochemical properties of Cs, because the profiles of Cs-134 are the same. In both Cs-137 profiles, the count rates above a TGT temperature of 750 K were higher than those below that. There was no clear difference in adhering behavior between the heating temperatures of 2773 K and 2973 K. This implied that some of the chemical species containing Cs could adhere on the inner surface of the sampling tubes below 750 K. As seen in Fig. 2, Sb-125 could adhere below 750 K, and then it could be detected above that temperature only for the 2773 K experiment. Although all of the counts rates were too low to identify any clear features, this difference between both tests may result from the inventory difference as seen in Table 1.

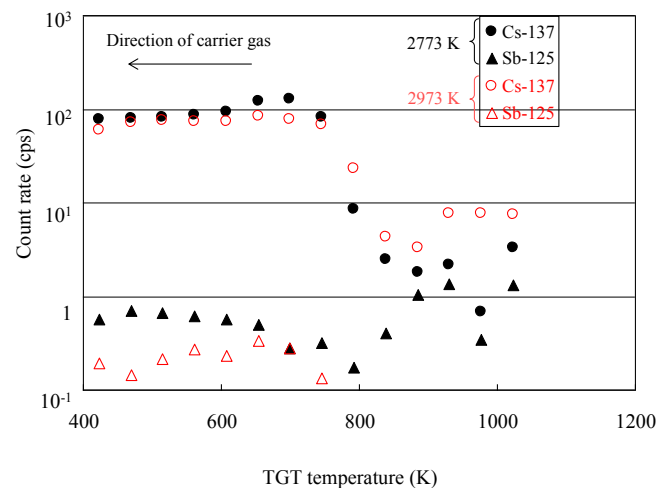


Fig. 2. Count rate profiles for Cs-137 and Sb-125 along the TGT in the 2773 and 2973 K experiments.

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