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Prediction of chemical effects of Mo and B on the Cs chemisorption onto stainless steel

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Abstract

Chemical effects of Mo and B on the Cs chemisorption onto stainless steel (SS) were predicted using a chemical equilibrium calculation based on an assumed model. A step reaction model for the Cs chemisorption process considering surface oxide layer formed on SS was assumed based on a literature review. Resultant major Cs compounds by Cs chemisorption were calculated for the two types of SS having different oxide surface layer structures. It is seen that Mo has induced the formation of Cs_2MoO_4 as a major Cs compound. On the other hand, little effects were observed for B. Cs-Si-O compounds were major resultant compounds regardless of Mo or B existence, indicating the stability of Cs-Si-O compounds. The results suggest that Cs-Mo-O in addition to Cs-Si-O compounds should be considered for further investigation on Cs chemisorption.

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Keywords: Cesium; Boron; Molybdenum; Chemisorption; Severe Accident; Stainless steel; Chemical equilibrium calculation.

Nomenclature

1F	Fukushima Daiichi Nuclear Power Station
BWR	Boiling Water Reactor
FP	Fission Products
LWR	Light Water Reactor

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SA	Severe Accident
SS	Stainless Steel

1. Introduction

Cs chemisorption is of crucial importance for the research and development of decommissioning of Fukushima Daiichi Nuclear Power Station (1F) and improved source term assessment, since it can significantly affect radionuclide retention in LWR under severe accidents. We have therefore started a fundamental study on the Cs chemisorption onto reactor structural materials such as stainless steel (SS) at relatively high temperatures around 1,000 K. The goal of this study is to depict a process model for Cs chemisorption based on the elucidation of its mechanisms. However, little is known so far about this Cs chemisorption phenomenon. Our first step was therefore to reproduce the formation of Cs chemisorbed compounds on the surface of SS that appears as results of Cs chemisorption and chemical reaction with SS components by a basic experimental test. It has been confirmed that the previously reported Cs chemisorbed compounds of Cs-Si-O were indeed formed and it turned out that the compound was a new and unexpected one, $CsSiFeO_4$ [1-2]. We are then entering into the next step, to investigate dependences of Cs chemisorption on temperature and atmosphere. On the other hand, as prediction of chemical effects of Mo and B on the Cs chemisorption is important and urgent matter, it should be investigated in parallel to the temperature and atmosphere dependences. This is because the BWR-1F decommissioning requires the B chemical influences on the Cs behavior to be certified and Mo has been evaluated as the most likely element that can be combined with Cs [3-4]. The chemical effects of B are also important in terms of its strong chemical affinity and comparable amounts to Cs in a severe accident (SA) at a LWR.

We have therefore carried out a prediction of chemical effects of Mo and B on the Cs chemisorption with the aid of chemical equilibrium calculation. A step reaction model for the Cs chemisorption process on SS was assumed based on the literature review and appropriateness of the model was confirmed by the chemical equilibrium calculation in terms of resultant stable Cs compounds by the chemisorption. Effects of Cs compounds in the case of Mo and B inclusion were then evaluated based on the assumed process model.

2. Assumed model for the Cs chemisorption process

A step reaction model for the Cs chemisorption onto SS was hypothesized for the prediction of chemical effects of Mo and B on the Cs chemisorption. For this purpose, a review on the related literatures to the Cs chemisorption was carried out.

Previous investigations for both laboratory-scale simulating experiments of CsOH chemisorption onto SS [1-2,5] and analysis of real samples taken from the Three Mile Island-2 (TMI-2) SA [6-7] were reviewed and Cs chemisorbed compounds are summarized in Table 1. It was found that Cs-Cr-O and Cs-Si-O compounds were major ones. Therefore, these two Cs compounds were implemented into the assumed process model for Cs chemisorption. Although our previous studies have shown the formation of a Cs-Si-O compound including Fe, $CsFeSiO_4$ [1-2], this compound was not taken into consideration since the thermodynamic data for $CsFeSiO_4$ is not present. Thus a minor component of SS, Si, was included for the assumed model for the confirmation of stable compounds by a chemical equilibrium calculation, in addition to major components of SS such as Fe and Cr.

Table 1. Summary of previous studies related to Cs chemisorption.

	Experiments with simulant FP			Analysis of TMI-2 real samples
	Elrick and Power [5]		Di Lemma et al. [1-2]	Baston et al. [6] Lorenz and Collins [7]
Temperature (K)	< 873	> 1,073	> 1,073	N.A.
Chemisorbed Compound	Cs-Cr-O	Cs-Si-O	Cs-Fe-Si-O	Cs-Si-O