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Author(s)	Miwa Shuhei, Yamashita Shinichiro, Ishimi Akihiro, Osaka Masahiko, Amaya Masaki, Tanaka Kosuke, Nagase Fumihisa
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Research Program for the Evaluation of Fission Product and Actinide Release Behaviour, Focusing on Their Chemical Forms

Shuhei Miwa[a], Shinichiro Yamashita[a,*], Akihiro Ishimi[a], Masahiko Osaka[a],
Masaki Amaya[b], Kosuke Tanaka[a], Fumihisa Nagase[b]

[a]*Oarai Research and Development Center, Japan Atomic Energy Agency,*
4002 Narita-cho, Oarai-machi, Higashi-ibaraki-gun, Ibaraki, 311-1393, Japan.
[b]*Tokai Research and Development Center, Japan Atomic Energy Agency,*
2-4, Shirane Shirakata, Tokai-mura, Naka-gun, Ibaraki 319-1195, Japan.

Abstract

We have launched a new research program for evaluation of fission product and actinide release behaviour under severe accident conditions, focusing on their chemical forms. In this research program, the fission product and actinide release models incorporated in the severe accident analysis code will be improved by implementing effects of chemical forms that are to be obtained by an experimental investigation on the fission product and actinide release behaviour from irradiated fuels. A novel in-situ measurement method of chemical forms of fission products and actinides just after their release as well as an off-line analysis method will be employed as the experimental means for evaluating the chemical forms at the release and their stability after release.

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Keywords: Fission products; actinides; release behaviour; chemical form

1. Introduction

After the 2011 nuclear accident at the Fukushima Daiichi (1F) Nuclear Power Plant (NPP) [1], Tokyo Electric Power Company (TEPCO) decided to decommission the reactors at 1F-NPP and the corresponding road map was

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* Corresponding author. Tel.: +81-29-267-4141; fax: +81-29-266-3713.
E-mail address: yamashita.shinichiro@jaea.go.jp

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drawn up [2]. Research and development (R&D) project to address the technological challenges which are needed to accomplish the 1F-NPP decommissioning were devised by the Agency of Natural Resources and Energy and TEPCO in collaboration with all of the related organizations and 1F manufacturers. The Japan Atomic Energy Agency (JAEA) has implemented the basic and fundamental researches on the highly radioactive materials in the R&D project, specifically the fuel debris, radioactive waste and spent fuel [2]. In the hot laboratory complex in the Oarai branch of JAEA, the following experimental-based research programs have been launched based on the significant knowledge and experience of post irradiation examinations.

- Evaluation of the seawater effects on corrosion behavior of component materials of irradiated fuel assemblies, reactor pressure vessels and primary containment vessels for the integrity assessment of their materials that experienced a diluted seawater exposure
- Basic study on analysis techniques of fuel debris for the establishment of the effective nuclear material accountancy methodology in the debris which has various form
- Evaluation of fission product (FP) and actinide release behaviour for better accuracy of the source term issues prior to the precise evaluation of the accident progress and the status inside the damaged 1F core

This article describes details of the third research program, the evaluation of FP and actinide release behaviour.

Continuing research on source term issues is also of special importance for improvement of accuracy of the Probabilistic Safety Assessment (PSA) method [3,4] for both safety evaluation of NPPs and severe accident management (SAM) measures [5,6]. Since its development and first application to a regulatory process by the Nuclear Regulatory Commission (NRC) in 1978 [3], the PSA method has been continuously improved through analyses of commercial NPPs. In particular, as the Three Mile Island Unit 2 (TMI-2) accident gave little information on FP behaviour after release from the bundle in spite of the large core melt; about 50% [7], source term issues have attracted much attention and been the subject of many experimental and analytical studies. Outcomes of these studies have been reflected as improvements of the severe accident analysis codes such as MAAP [8,9], MELCOR [10] and THALES-2 [11-13]. Presently, there are several on-going experimental and analytical programs about source term issues. For instance, the VERDON program [14] focuses on Ru behaviour in a reactor especially at the time of air ingression in a severe accident scenario under the framework of the Severe Accident Research NETwork of Excellence (SARNET) [15].

The present situation since the 1F-NPP severe accident [1], on the other hand, has strongly shown the need for better accuracy of the source term issues because NPP safety must be reinforced by improvement of accuracy and rationality of the PSA method. Since the PSA is evaluated through the use of the severe accident codes, the improvements of their accuracy are required for the precise evaluation of source term issues. Therefore, the decommissioning work of 1F-NPP is a matter of top priority in order to incorporate the lessons learned from the accident as soon as possible. The severe accident analysis codes are to be improved for the precise evaluation of the accident progress and the status inside the damaged 1F core prior to the actual removal of fuel debris in the core [2].

We have launched a new research program for the evaluation of FP and actinide release behaviour, which is one of the important source term issues, focusing on their chemical forms according to the above-mentioned viewpoints of reinforcing nuclear safety and carrying out the R&D project for the 1F-NPP decommissioning. Chemical forms are the key factor that affects not only the FP and actinide release behaviour but also their transport behaviour.

In this article, previous experimental and analytical research studies on FP and actinide release behaviour are reviewed in order to specify the subjects to be resolved; based on this specified subjects, we introduce a research plan.

2. Review of previous research studies on FP and actinide release behaviour

Single-effect studies and integral studies have been carried out for the evaluation of FP and actinide release behaviour. The former focuses on a single phenomenon under a controlled condition, which can give deeper insights into the specific phenomena. The integral studies cover topics from the FP and actinide release from the fuel to their transportation in a reactor. The data obtained by these experimental studies have been analyzed by FP and actinide release models for further improvement of these models.

2.1. FP and actinide release experiments

Representative integral tests are STEP [16], ACRR-ST [17], PBF-SFD [18], LOFT-FP [19] and Phebus-FP [20-23] tests. The single-effect tests for the evaluation of FP and actinide release behaviour are ORNL HI/VI [24], HEVA/VERCORS [25-27] and VEGA [28-30] tests. Those tests for the evaluation of FP and actinide transport behaviour were also reviewed; such as FALCON [31], STORM [32], and WIND [33] tests. These tests were performed in various systems and under different controlled conditions simulating severe accident conditions including high temperatures with a range of atmospheric conditions. Details of experimental conditions and results were described in [34]. A summary of the chemical forms of FPs and actinides together with the release kinetics are given in Table 1.

In STEP tests, it was found that the chemical forms of FP vapours and aerosols were complicated; however, Cs compounds in the aerosols were estimated to be CsI and CsOH. The analytical results of FP release rates by using the model at the time were found to overestimate the experimental values [16]. In ACRR-ST tests, the FP release and transport behaviour was investigated in a reductive atmosphere, under which the release of Te was suppressed [17]. In PBF-SFD tests, the FP release kinetics were evaluated in relation to the core degradation and H_2 generation [18]. LOFT-FP tests focused on effects of steam supply and re-flooding to the fuel assemblies on the FP release behaviour. It was found in the LOFT-FP tests that Cs compounds were deposited as the water soluble forms [19]. In Phebus-FP tests, more detailed information of the core degradation behaviour, the FP transport behaviour in the reactor cooling system and the containment vessel was evaluated [20-23]. Gaseous iodine as well as aerosol iodine were generated in the reactor coolant system and the containment vessel, and their amounts were increased by the presence of B_4C moderator [35].

In the most representative single-effect test series for the FP release behaviour, the ORNL HI/VI tests, the effects of temperature and atmosphere on the FP release behaviour were systematically investigated [24]. The test results gave the release kinetics of Kr, Cs, I and Sb from cladded fuel specimens as a function of temperature and atmosphere. The release kinetics of Cs and I were found to be almost the same as those of Kr. Te and Sb were first absorbed in zircaloy cladding, then they were released through cladding oxidation [24]. In another representative test series, the HEVA/VERCORS tests, the fuel specimens were re-irradiated in research reactors in order to investigate the release behaviour of short-lived FPs such as ^{131}I, ^{133}I, ^{132}Te, ^{99}Mo, ^{140}Ba and ^{140}La that were not accurately investigated in the ORNL HI/VI tests [25-27]. The HEVA/VERCORS tests gave the following results. The release behaviour of highly volatile FPs such as Cs and I depended on the atmosphere. Cs and I release rates were high, and had higher values in an oxidative atmosphere than in a reductive atmosphere. The release behaviour of semi-volatile FPs such as Mo and Ba was also influenced by the atmosphere. The release behaviour of Ru and Mo was promoted in the oxidative atmosphere, whereas those of Sr, Ba and Eu were enhanced in the reductive atmosphere. In the VEGA tests, effects of atmospheric pressure on the FP release behaviour were investigated [28-30]. The Cs release rate was suppressed by high pressure at a relatively low temperature condition.

Concerning the FP transport behaviour, the FALCON and WIND tests looked at the deposition and re-vapourization behaviour of FPs such as CsI and CsOH in the reactor cooling systems [32,34]. The FP deposition behaviour varied depending on their presence in gaseous or aerosol phase. The re-vapourization behaviour of Cs and Mo was influenced by the co-existence with H_3BO_3 which led to the formation of highly volatile B compounds.

From the review results summarized in Table 1, we divided FPs and actinides into five categories in the order of their volatility: FP gases (Xe, Kr), highly volatile FPs (Cs, I, Te, Sb), semi-volatile FPs (Ru, Mo, Sr, Ba, Pd, Rh), low-volatile FPs (Eu, Zr, Ce, Nb, Y, La), and finally actinides (U, Np, Pu).

Almost all of the FP gases in the fuel are released at high temperature. Although the highly volatile FPs (Cs, I, Te, Sb) are also nearly completely released at high temperature. The oxidative atmosphere and high burn-up of fuel promote the release kinetics of Cs and I at lower temperatures. The main chemical forms of Cs compounds have been predicted to be CsI and CsOH, while those of other chemical forms such as Cs_2MoO_4 and Cs_2Te were suggested [23,36]. Besides, as it was suggested that higher volatility Cs compounds would be formed by reaction with sea water [37], the analysis of Cs release from the 1F core should pay attention to this point since large amount of sea water was injected into the 1F core [1]. The chemical forms of I were also found to be complicated depending on the temperature and atmosphere [38]. Te and Sb, as mentioned before, reacted with Sn in zircaloy, and were released with its oxidation [26].

Table 1. Summary of FP and actinide release behaviour by element.

Category	Element	Summary of release behaviour	Ref.	Expected chemical forms after release	Ref.
FP gas	Kr	• Almost complete release at high temperature (Burst release around 973-1073 K and 1373-1473 K)	[26]	Kr	-
	Xe	• Similar release behaviour as that for Kr	[26]	Xe	-
High volatile FP	Cs	• Almost complete release above 2623 K • Bursty release above 2800 K by liquid formation of fuel • Release enhanced by redistribution in the fuel (below 2310 K), high burn-up, debris configuration, oxidative atmosphere, presence of Pu • Release suppressed in high pressure atmosphere at relatively low temperature • Similar release kinetic as that for Kr • Retention in fuel as $CsMoO_4$, $Cs_2Cr_2O_4$, $Cs_2Fe_2O_4$, $Cs_2Si_4O_9$, $CsBO_2$ at high temperature • Possible formation of more volatile compounds by reaction with sea water	[25,26] [21,28] [26,34] [28] [24] [26] [37]	Cs, CsOH, CsI, Cs_2MoO_4, Cs_2Te, Cs_2TeO_3, Cs_2TeO_4, $CsBO_2$	[16, 19,23, 31,33]
	I	• Similar release behaviour as those of Cs • Release enhanced by high burn-up, debris configuration, oxidative atmosphere, presence of Pu	[18,24,26] [26]	CsI, I_2, Organic forms	[22,23, 33]
	Sb	• Almost complete release above 2573 K • Release after the oxidation of zircaloy cladding (reaction with Sn in zircaloy)	[26] [24,26]	Sb	[34]
	Te	• Similar release behaviour as those of Sb	[17,24,26]	Cs_2Te, Cs_2TeO_3, Cs_2TeO_4, SnTe	[16,32]
Mid volatile FP	Ru	• High volatility in oxidative atmosphere • Release enhanced by high burn-up	[14,27] [27]	RuO, RuO_2, RuO_3, RuO_4	[27]
	Mo	• Lower release kinetics than highly volatile FPs • Low release in reducing conditions • High volatility in oxidative atmosphere	[27] [27,29] [27]	Cs_2MoO_4, MoO_3	[23,27]
	Sr	• High volatility in reductive atmosphere • Release enhanced above 3130 K by chemical reduction • Possible formation of more volatile compounds by reaction with sea water	[27] [30] [37]	SrO	[34]
	Ba	• High volatility in reductive atmosphere • Reaction with zircaloy	[27] [22,27]	BaO	[27]
Low volatile FP	Eu	• Increased volatility in reductive atmosphere	[27]	Eu_2O_3	[27]
	Zr	• Small amount of release	[27]	ZrO_2	[27]
	Ce	• Increased volatility in reductive atmosphere • Small amount of release	[27] [27]	Ce_2O_3	[34]
Actinide	Pu	• Release enhanced by redistribution in the fuel (below 2310 K)	[28]	PuO	[24,28]

The release behaviour of the semi-volatile FPs (Ru, Mo, Sr, Ba, Pd, Rh) largely depends on the atmosphere [26,27]. The releases of Mo and Ru are promoted in the oxidative atmosphere, whereas those of Sr and Ba are promoted in the reductive atmosphere as described above. This behaviour is attributed to the fact that the FP release behaviour depends on the chemical forms of the FPs which are a function of the state of the surrounding system. It was suggested that higher volatility Sr compounds would also be formed by reaction with sea water [37].

The low-volatile FPs (Eu, Zr, Ce, Nb, Y, La) and actinides (U, Np, Pu) show low release rates. Nevertheless, it was predicted that the release behaviour of certain low-volatile FPs was also influenced by the atmosphere.

2.2. Analytical research studies for FP and actinide release behaviour

Table 2 shows the equations in the CORSOR model series. The CORSOR-M model is the basis of FP release model employed in the major severe accident analysis codes, MELCOR [10] and THALES-2 [11-13].

The data from the ORNL HI/VI, VERCORS and Phebus-FP tests were analyzed by the CORSOR model series [24,39,40]. The release rate equation in the CORSOR-M model can depict the fractional release of each FP using the individual release rate coefficient, *k* (min^{-1}). The coefficient is expressed as an Arrhenius equation which is composed of constants of the pre-exponential factor k_0 and activation energy *Q* [41] as shown in Table 2. The FP release rates calculated by the CORSOR-M model overestimated the ORNL HI/VI test results [24]. The CORSOR-M model was, therefore, improved by adjusting the k_0 considering effects of the atmosphere. The CORSOR-O model has three k_0 values for initial, oxidative and reductive conditions except for Kr, Cs and I [24]. The calculation results by using the CORSOR-O model well reproduced the ORNL HI/VI test results [24].

Although the analysis results obtained using the CORSOR-Booth model, in which the FP release process is subdivided and controlled by intra-granular diffusion, have good agreement for highly volatile FPs such as Cs and I [24,39,40], it could not reproduce the experimental data for semi-volatile FPs such as Mo and Ru [39,40]. One possible reason for the discrepancy between FP release rates estimated by the CORSOR-Booth model and the experiment arises from the hypothesis in the CORSOR-Booth model that FPs would be released from fuel as a single element. This result suggested that the semi-volatile FPs could be released as a compound. For example, the release rate of Mo is close to the experimental data based on the hypothesis that Cs would be released from the fuel as Cs_2MoO_4 rather than CsI and CsOH that have been considered as the chemical forms of Cs [39,40].

The VEGA test results were also analyzed by the CORSOR model from the viewpoint of the effects of atmospheric pressure on the FP release behaviour [28-30]. The CORSOR-M model was, thus, improved as the CORSOR-P model which implemented the dependence term of FP diffusion in open porosity on atmospheric pressure [28]. The CORSOR-P model could better reproduce the test results under high pressure, such as in the PBF-SFD tests [28]. In addition, the data of release rates in the fuel melt and liquid formation by the high temperature reaction between fuel and zircaloy cladding for the VEGA tests were analyzed using the Booth model, the diffusion model by fuel oxidation and the fuel dissolution model [30]. This analysis result could reproduce the Cs release behaviour with the high temperature reactions between fuel and cladding material as well as fuel oxidation.

The VERCORS test results were analyzed by the ELSA module in the ASTEC code system [42,43]. In the ELSA module, the chemical forms of FPs and actinides were analyzed by the chemical equilibrium calculation [42]. Estimated values for Mo and Ba, which could not be reproduced by the CORSOR model series, showed good agreement with experimental values in the VERCORS tests [43].

3. Establishment of important issues

In this section, we describe two important issues identified in the present research program, based on the review described in the previous section. The first issue is concerned with the elements to be investigated. And the second is about the key factor for the improvement of CORSOR-M model incorporated in THALES-2 [11-13] which has been chosen as the target code to be improved with in JAEA.

3.1. Priority of elements to be investigated

The priority of FP and actinide elements to be investigated in this program was selected considering the following two viewpoints: “the contribution to public exposure”, and “decay heat”. While the public exposure is the most important aspects for the safety evaluation of NPP, i.e. for the source term issues, the decay heat is believed to be of critical importance from the viewpoint of debris-removal in the decommissioning work of 1F-NPP. Another important factor, the toxicity, is not considered since the release fraction into the environment is negligible for the most toxic elements, actinides [44].

From the viewpoint of public exposure, Cs and I are very important because almost all Cs and I in the fuel are released in a severe accident, which significantly impacts on the off-site exposure in the immediate and in the long term for I and Cs, respectively [26,45-47]. Sb, Te, Mo, Sr and Pu are of intermediate importance [45]. Although Te, Mo and Pu have a small impact from the viewpoint of external exposure [27], they are important because Te and Mo are reported to form compounds with Cs, such as Cs_2Te and Cs_2MoO_4 [23,36], and Pu results in significant internal exposure [48].

Table 2. Equations in CORSOR model series [24,39-41].

Release model	Integral Code	Equation Intra-granular	 Grain boundary, open porosity	Remarks
CORSOR		$k = A\exp(BT)$ k: release rate coefficient, A: pre-exponential factor, B:constant , T: temperature		
CORSOR-M	MELCOR THALES-2	$k = k_0 \exp(-\frac{Q}{RT})$ k_0: pre-exponential factor, Q: activation energy, R: gas constant		The value of k_0 in CORSOR-M was adjusted considering effects of the atmosphere.
CORSOR-O	MELCOR			
CORSOR-P	THALES-2	$k = k_0\sqrt{\frac{P_0}{P}}\exp(-\frac{Q}{RT})$ P_0: atmospheric pressure, P: pressure		
CORSOR-Booth	MELCOR	$f = 6\left[\frac{\tau}{\pi^3}\right]^{\frac{1}{2}} - 3\frac{\tau}{\pi^2} \quad \tau < 1$ $f = 1 - 6\frac{\exp[-\tau]}{\pi^2} \quad \tau \geq 1$ $\tau = \frac{\pi^2 D_G[t - t_0]}{a^2} \quad D_G = D_0\exp\left[-\frac{\vartheta}{T}\right]$ f: release fraction, a: equivalent sphere radius for the fuel grain, D: effective diffusion coefficient D_0: pre-exponential factor	$release\ rate_{Cs} = \frac{f_{t+\Delta t} - f_t}{F\Delta t}$ $\dot{m}_k = \left[\frac{A_{fuel}NuD_{k\cdot gas}}{D_{fuel}RT}\right](P_{k,eq} - 0)$ $DIFF_{Cs} = \left[\frac{1}{release\ rate_{Cs}} - \frac{1}{\dot{m}_k}\right]^{-1}$ $DIFF_k = DIFF_{Cs} \cdot S_k$ D_{fuel}: diameter of fuel pellet, A_{fuel}: fuel rod flow contact area, $D_{k,gas}$: diffusivity of Class k in the gas mixture, Nu: Nusselt number, $P_{k,eq}$: equilibrium vapour pressure of Class k at temperature T, S_k: scaling factor for each radionuclide of Class k	The values of D_0, Q, S_k in CORSOR-Booth were adjusted
ORNL-Booth	MELCOR			

From the viewpoint of decay heat, the priorities of Cs, Sr, Ba, Eu and Pu are high [47]. In particular, since ^{137}Cs (^{137m}Ba) and Pu will release decay heat for long periods [47], the evaluation of inventories of Cs and Pu that remain in the fuel debris are considered to be important in terms of the long-term storage of fuel debris. The priorities of the elements were thus determined considering the above-mentioned two viewpoints: Cs and I have the highest priority, followed by Te, Mo, Sr, Ba and Pu.

3.2. Key factor for the improvement of FP and actinide release models

Although systematic data for FP and actinide release behaviour as a function of temperature, atmosphere, burn-up and so on had been obtained by the single-effect tests such as the ORNL HI/VI, HEVA/VERCORS and VEGA tests as well as the integral tests, there remain relatively large uncertainties in the FP and actinide release behaviour causing as yet unconsidered factors in the models.

The source term program in SARNET is now being developed, based on PIRT established by EURSAFE to reduce the uncertainties of the release and transport behaviour of FPs and actinides [49]. This program covers the following phenomena: effect of oxidative conditions on FPs, especially Ru; release and transport behaviour under an air ingress scenario; volatility of I in the primary circuit; control rod aerosol release (Ag-In-Cd) that affects I transport; aerosol retention in containment cracks; aerosol remobilization in the primary circuit [50]; and I and Ru behaviour in the containment, especially concerning the volatile fraction in the atmosphere [51]. Thus, the evaluation of I and Ru behaviour in a reactor are still recognized as important research issues in the SARNET.

Concerning the release behaviour of I, whose chemical form has been taken to be CsI [16], however it is now clear that whatever form it takes, it can react with other FP's and the changes of the chemical form are of crucial importance. For instance, from the result of Phebus-FPT3 [35] test in which the B_4C moderator was injected into the system, it is suggested that the chemical form of I was changed by the high-temperature reaction between CsI and B_4C because the yield of gaseous iodine was higher than that for Phebus-FPT2 [35]. Since larger gaseous iodine formation could directly lead to the increase of I release fraction into the environment, precise evaluation of the chemical forms of Cs and I is important.

In the FP and actinide release tests including the VERDON program in SARNET, the release kinetics of FPs and actinides have usually been evaluated based on the results of on-line monitoring of released gamma-emitting isotopes such as ^{137}Cs by gamma-ray spectrometry [24-28]. The release amount was estimated from the total deposited FPs and actinides in thermal gradient tubes (TGTs) and filters in the experimental device that was determined by a combination of alpha-spectrometry, beta-spectrometry and chemical analysis. Although such radiochemical analyses are appropriate measures for the estimation of release amounts of FPs and actinides, it is nearly impossible to make direct measurements for the chemical forms. The difficulties in measurement of the chemical forms mean there are only limited data at the present for chemical forms of released FPs and actinides in spite of their significance for the FP and actinide release behaviour. Information available for the evaluation of chemical forms is only the results of measurement of deposited FP and actinide compositions with the aid of the chemical equilibrium calculation. Such predicted chemical forms should have large uncertainties since the chemical forms are affected by the state of the local environment from inside the fuel to the deposited surface.

In the analytical research, the FP and actinide release rates are only calculated as a function of temperature in the CORSOR model series. The fact that any CORSOR model series can hardly reproduce the release behaviour of, in particular, semi-volatile FPs such as Mo [39,40] suggests a large dependency of the release behaviour on the chemical forms. Specifically, Kissane et al. [23] suggested that the chemical form of released Cs would be Cs_2MoO_4 rather than CsOH, which leads to the importance of Mo release behaviour in relation to Cs. Thus, it is believed that the FP and actinide release behaviour can be evaluated more precisely if FP and actinide release model would be improved by implementing the effects of chemical forms based on the investigation of the FP and actinide release behaviour. Such an improvement could give deeper insights into FP and actinide transport behaviour that finally could lead to the precise evaluation of source term issues. In this research program, therefore, the chemical forms of FPs and actinides will be focused on as the factor to be investigated for the improvement of FP and actinide release models.

4. Research program for the evaluation of FP and actinide release behaviour

In this section, we give details of the research program in response to the important issues identified for the effects of chemical forms on the FP and actinide release behaviour. In our research program, the CORSOR-M model and the subsequent FP and actinide compound categorization process employed in THALES-2 [13,41] are to be improved by implementing the effects of chemical forms into the models. In addition, CORSOR-Booth model were selected and will also be improved in order to accurately evaluate the FP and actinide release kinetics in the complex phenomena of a severe accident, in which the evaluation of FP release kinetics is difficult by the improved CORSOR-M model with only different k_0 and Q sets according to the atmospheres. The experiments will be performed in order to accumulate a database related to FP and actinide release behaviour, focusing on the chemical forms observed in irradiated fuel specimens. These research contents are summarized in Table 3.

4.1. Improvement of FP and actinide release models

Fig. 1 shows the overall plan for improvement of the CORSOR-M model and the subsequent FP and actinide compound categorization process employed in THALES-2 [13,41]. In THALES-2, release rates of each FP and actinide are calculated first, and then predetermined chemical forms are assigned to the released FPs and actinides.

In this research program, optimization of the constant values in the FP and actinide release models, such as k_0 and Q in the CORSOR-M model, is carried out based on the experimental database of FP and actinide release kinetics that includes the effect of atmosphere and the chemical reaction among fuels and zircaloy and/or B_4C. The experimental database is composed of refined existing data and data that will be newly obtained in this research program.

Table 3. Research items for the evaluation of FP and actinide release behaviour in this program

Research items	Research contents	Expected outcomes
Improvement of FP and actinide release models	・Improvement of CORSOR-M model, CORSOR-Booth model and the subsequent process for chemical form categorization in THALES-2 by implementing the effects of atmosphere and chemical forms ・aa	・ Improved model implementing the dependence on atmosphere and the chemical reaction among fuels and zircaloy and/or B_4C − Improved constant values of CORSOR-M model and CORSOR-Booth model − Improved categorization table and/or chemical reaction rate coefficient of each FP
Evaluation of the release kinetics from fuel	・Investigation of the correlation between FP and actinide release kinetics and chemical forms of FPs and actinides at release by TG-DTA-MS ・Investigation of the FP release kinetics using a FP release test device	・Database of release kinetics with the dependence on atmosphere and the chemical reaction among fuels and zircaloy and/or B_4C −Release kinetics data −Data of chemical form at release
Evaluation of the stability of chemical forms after release	・Investigation of the chemical forms of deposited FPs and actinides using a FP release test device ・Analysis of chemical forms by chemical equilibrium calculation	・Improved categorization table and/or database of chemical reaction rate coefficient with the dependence on atmosphere and the presence of structural material such as zircaloy and B_4C −Data of chemical form of deposits

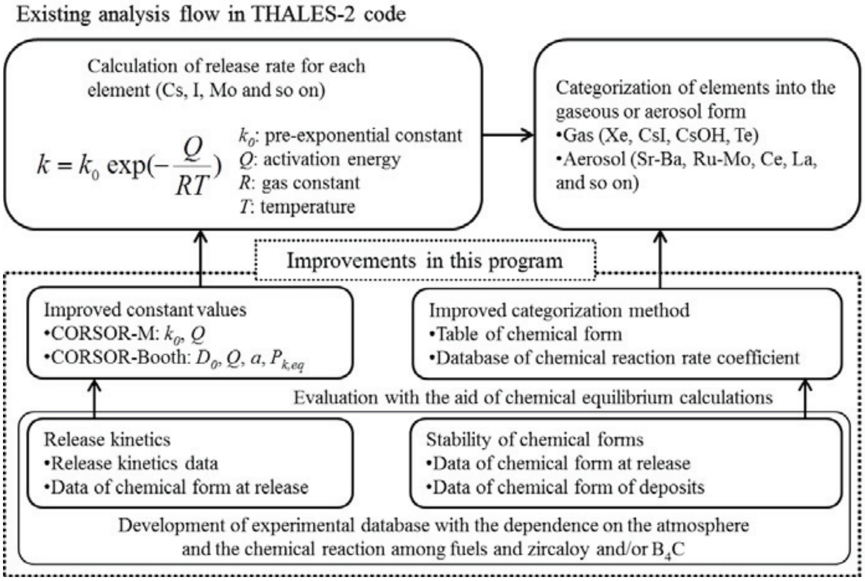

Fig. 1. The overall plan toward improvement of the CORSOR-M model and the subsequent FP and actinide compound categorization process in this program [41].

In order to see the expected effects of atmosphere on Cs release kinetics by CORSOR-M model adjusted value sets of the constant k_0 and Q for Cs, in reductive, inert and oxidative atmospheres were calculated by the least squares fitting of the existing data set for the Cs release rate coefficient obtained in ORNL HI/VI [24], VERCORS [26] and VEGA [28] tests with the Arrhenius equation shown in Table 2. The oxygen partial pressures in each test were estimated by the chemical equilibrium calculation from the test conditions using Thermo-Calc software [53] with SSUB4 database [54]. Test results were divided into the 3 groups on the basis of the oxygen partial pressure, specifically reductive ($< 1\times10^{-11}$ atm), inert (1×10^{-11} - 1×10^{-4} atm) and oxidative ($> 1\times10^{-4}$ atm) atmospheres. Table 4 lists the calculation results of k_0 and Q sets for each atmosphere. It is seen that the constants increased with decrease of oxygen partial pressure. Fig. 2 compares the calculation results of Cs release rates by the CORSOR-M model with the adjusted atmosphere-dependent k_0 and Q sets (hereafter, CORSOR-MR model) with those by the original CORSOR-M model [41] as shown in Table 4. The calculation condition of maximum temperature and the heating rate were set to about 2,400 K and 15 K/min, respectively, based on the calculation results of MELCOR in 1F Unit 1 [52]. The Cs release fraction by CORSOR-M model attained the total release faster than any other, followed by CORSOR-MR model in the reductive atmosphere. The Cs release fraction by CORSOR-MR model in the oxidative atmosphere is the highest at the beginning, when the temperature is low, but that above 2,000 K (100 min. in Fig. 2) is lower than CORSOR-M. The Cs release fraction for CORSOR-MR model in reductive, inert and oxidative atmospheres at about 135 min, the point at which the Cs release fraction for CORSOR-M model achieved 1.00, were 0.97, 0.67 and 0.78, respectively.

Table 4. Calculation conditions of Cs release rates [24,26,28,41]

Models	Atmosphere	k_0 [min^{-1}]	Q [kJ/mol]
CORSOR-M	-	2.0×10^{5} [a]	2.7×10^{2} [a]
CORSOR-MR	Reductive	8.6×10^{2} [b]	1.8×10^{2} [b]
	Inert	2.9×10^{0} [b]	9.3×10^{1} [b]
	Oxidative	8.4×10^{-1} [b]	6.5×10^{1} [b]

[a] Values employed in CORSOR-M model [41]

[b] Values calculated in this research program by the squares fitting to the release rate coefficients obtained in ORNL HI/VI [24], VERCORS [26], VEGA [28] test.

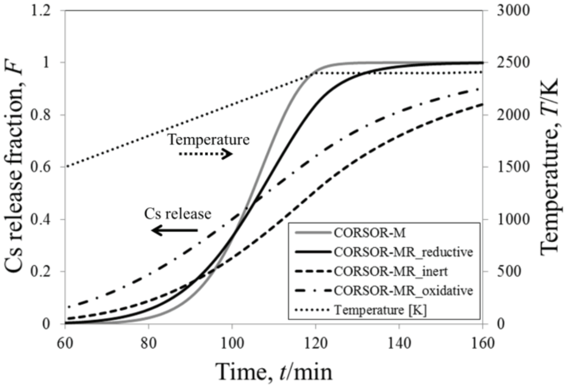

Fig. 2. The comparison of Cs release rates by using CORSOR-M and CORSOR-MR models (CORSOR-M: calculation results using CORSOR-M model [41], CORSOR-MR: calculation results using improved CORSOR-M model with adjustment of k_0 and Q by the results of ORNL HI/VI [24], VERCORS [26] and VEGA-4 [28] tests).

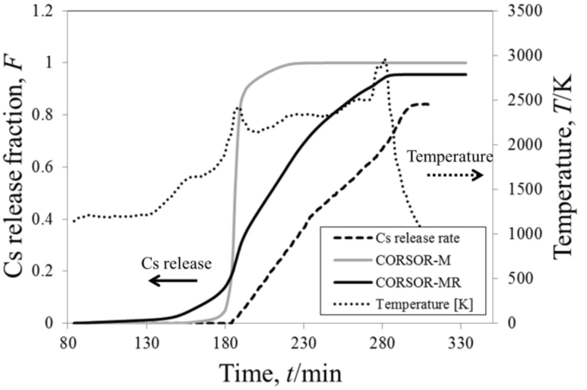

Fig. 3. The calculation results of Cs release rates of Phebus-FPT1 [39] (CORSOR-M: calculation results using CORSOR-M model [41], CORSOR-MR: calculation results using CORSOR-MR model in inert atmosphere).

Fig. 3 shows the calculation results for CORSOR-M and CORSOR-MR model for an integral test, Phebus-FPT1 test [39]. The atmosphere for Phebus-FPT1 was steam [39], which corresponded to the inert atmosphere in the CORSOR-MR model. The Cs release rate calculated by the CORSOR-M model was higher than the experimental data as well as the results in [24,39,40]. Although the Cs release rate calculated by the CORSOR-MR model shows the closer value to experimental data than that calculated by the CORSOR-M model, it could not reproduce the experimental data with high accuracy. This result implies a lack of accuracy of the evaluation by using CORSOR-MR model for the complex phenomena such as degradation of fuel and interaction between fuel and structural material in a severe accident. However, the extensive database for the dependence of FP and actinide release behaviour on atmosphere and zircaloy and/or B_4C injection could lead to increase of the accuracy of the CORSOR-MR model in some degree.

As another means for the precise estimation of the FP and actinide release behaviour, the CORSOR-Booth model is employed and will also be improved in this research program. The constant values in the CORSOR-Booth model, D_0, Q, a, $P_{k,eq}$ as shown in Table 2 will also be improved by using the anticipated experimental database of FP and actinide release behaviour. The diffusion coefficient D including D_0, Q will be improved by the application of the fuel oxidation model proposed by Lewis et al. [55]. In this fuel oxidation model, the diffusion coefficient was expressed as a function of oxygen stoichiometry deviation [55]. The vapour pressure of FP compounds will be evaluated by using newly obtained data of FP and actinide chemical forms just after their release from fuel, and the vapour pressure constant $P_{k,eq}$ in the CORSOR-Booth model will be improved.

The chemical form categorization is also to be improved by using newly obtained data from both in-situ measurements of the chemical forms of FPs and actinides just after their release and evaluation of the deposited FPs, as shown later, with the aid of the chemical equilibrium calculations as shown in Fig. 1. In addition to the categorization table, the categorization method in the model will be improved by evaluating the chemical reaction rate coefficient of each FP as a function of the atmosphere and the presence of structural material such as zircaloy and B_4C from the chemical form studies.

The evaluations by using the mechanistic models incorporated in VICTORIA [56] or FASTGRASS [57] are also to be performed in order to reflect the detailed FP and actinide release behaviour obtained by these codes. These results will be used to improve the FP and actinide release model and the subsequent FP and actinide compound categorization process.

4.2. Evaluation of the release kinetics from fuel

In order to improve the CORSOR-M and CORSOR-Booth models, FP and actinide release kinetics from the fuel, the chemical forms just after their release will be investigated by using a thermogravimetry (TG) – differential thermal analysis (DTA) – mass spectrometry (MS) device and a FP release test device [58,59].

The TG-DTA-MS device (ThermoMass Photo, Rigaku Co. Ltd. [60]) is able to use a standard thermal analysis (TG-DTA) for the specimen and quadrupole-MS of released materials from the specimen [60]. Thanks to the skimmer interface structure in this MS device, novel in-situ measurements of chemical forms of released FPs and actinides are expected to be possible under high temperature and multi atmosphere conditions. Effects of atmosphere on the FP and actinide release behaviour including release kinetics, chemical forms and fuel oxidation, can be evaluated with this TG-DTA-MS device. The measurement results can be directly used to make the improvements of the CORSOR-M and CORSOR-Booth models, i.e. determination of k_0, Q, oxygen stoichiometry deviation and $P_{k,eq}$. Heating tests of simulated volatile FP compounds such as CsI and Cs_2MoO_4, and simulated semi-volatile FPs such as Mo and Ru will be carried out to investigate the correlation between FP and actinide release kinetics and chemical forms of FPs and actinides just after their release.

The FP release kinetics will also be investigated by using the FP release test device that is installed in a hot cell [58,59]. Fig. 4 shows a schematic diagram of the FP release test device and trapping system for released FPs. The FP release test device consists mainly of a high-frequency induction furnace for the specimen heating, two sets of thermal gradient tubes (TGTs) and filters onto which released FPs and actinides deposit, an on-line gamma-ray monitoring system for deposited FPs in one set of the filters, and an on-line mass spectrometer and an off-line gas chromatograph for the FP gas measurements. The irradiated fuel specimen, set in a crucible, is inductively heated to a maximum temperature of 3273 K under an inert or reductive atmosphere. The temperature of the specimen is measured by a pyrometer at the bottom of the heating container in which the crucible for the fuel specimen is loaded.

The FP release kinetics will be investigated by using on-line mass spectrometry for FP gases and the monitoring system of gamma-rays for the highly volatile FPs such as Cs. The heating furnace will also be improved to allow heating tests in an oxidative atmosphere. The heating tests of irradiated fuel will, then, be carried out to investigate the FP release kinetics, focusing on the effects of the atmosphere and the chemical reaction among fuels and zircaloy and/or B_4C.

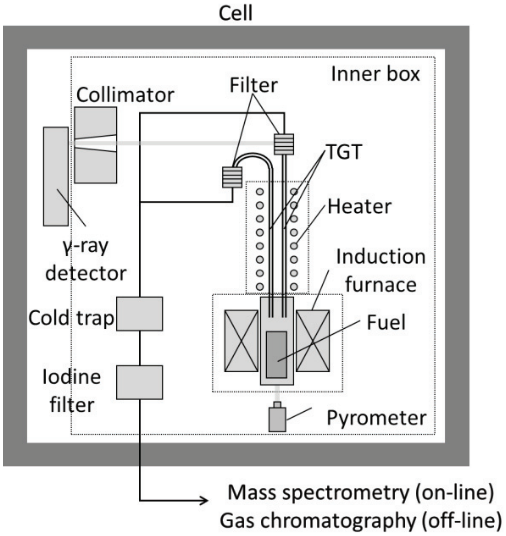

Fig. 4. Schematic diagram of the FP release test device and trapping system for released FPs [58,59].

4.3. Evaluation of the stability of chemical forms after release

In order to improve the chemical form categorization, stability of the FPs and actinides will be evaluated by an experimental investigation of chemical forms of deposited FPs and actinides by using the FP release test device [58,59]. The trapping system for FPs and actinides consists of sampling tubes in TGTs and filters as mentioned before. The inner surface temperature of the TGTs is kept between 1023 K (at the inlet side) and 423 K (at the outlet side) with a linear temperature gradient between the inlet and outlet. Fourteen separable sampling tube units are put into each casing tube. The released FPs and actinides are deposited according to their temperature-dependent properties.

The chemical forms of FPs and actinides deposited onto the sampling tubes in the TGTs and filters will be investigated by X-ray diffraction analysis combined with elemental analysis by gamma-ray spectrometry and chemical analysis. The heating tests of the irradiated fuels will be carried out after the systematic tests with simulated FPs such as CsI and Cs_2MoO_4.

From the chemical forms of FPs and actinides just after their release in addition to the results for the deposited FPs and actinides, the stability of the chemical forms of FPs and actinides will be evaluated as a function of the atmosphere and the presence of structural material such as zircaloy and B_4C with the aid of chemical equilibrium calculations. The evaluation results of the stability of the chemical forms can be directly used to make the improvements of the categorization method in the model.

5. Summary

We have launched a new program for the evaluation of FP and actinide release behaviour in severe accident conditions; we have focused on the chemical forms of FPs and actinides as a means to improve the severe accident analysis code for the reinforcement of nuclear safety as well as for implementation of the 1F-NPP decommissioning R&D project.

In our research program, the CORSOR-M model and the subsequent process for chemical form categorization in THALES-2 will be improved in terms of effects and stability of chemical forms. The calculation result of Cs release rate by the improved CORSOR-M model using existing experimental data set of Phebus-FPT1 shows an improved comparison with the experimental data. For further improvement with high accuracy, the constants in the CORSOR-M model and CORSOR-Booth model will be improved by using an experimental database, which will be constructed in our research program. This will include the dependence of release kinetics, the chemical forms of FPs and actinides just after their release and the stabilities of their chemical forms with atmosphere and chemical reaction. Systematic experimental data will be acquired by both the novel in-situ measurement device for chemical forms just after the release, based on TG-DTA-MS, and the FP release test device installed in a hot cell.

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