FY2018 (2018.4-2019.3) NSEC
Annual Report

Nuclear Science and Engineering Center
Nuclear Science Research Institute, Sector of Nuclear Science Research
Japan Atomic Energy Agency

September 2019

This report is issued by Japan Atomic Energy Agency.

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Preface

Akira ENDO

Director General, Nuclear Science and Engineering Center

It is our great pleasure to present to you the first annual report of the Nuclear Science and Engineering Center (NSEC) of the Japan Atomic Energy Agency (JAEA). This report provides our research highlights and an overview of the research groups' activities for Fiscal Year 2018.

The NSEC aims to conduct research and development to advance the science and technology that supports the use of nuclear energy and radiation. As such, we conduct varied research from elucidating various phenomena using our innovative techniques to developing computer simulation codes and databases. These cover nuclear data and reactor engineering, nuclear fuel and material engineering, nuclear chemistry, and environment and radiation sciences.

Through such knowledge and technologies, the NSEC contributes to solving various issues. Since the accident at the TEPCO Fukushima Daiichi Nuclear Power Plant, we have focused on research and development that contributes to the (1) accident response (promoting facility decommissioning and environmental restoration), (2) improvement of light water reactors' safety performance, and (3) steady implementation of the treatment and disposal of radioactive waste. Additionally, we disseminate our innovative technologies to resolve challenges in various fields, such as industry, environment and medicine.

We strive to become a leading center for research collaboration and use our fundamental research and development capabilities to contribute to advances in science and technology. We hope this annual report will increase your understanding of the NSEC.

September 2019

FY2018 NSEC R&D Highlights

The following 6 highlights are selected among various outcomes of the R&D activities accomplished by the NSEC's 6 divisions in FY2018.

- ✓ **Nuclear Data and Reactor Engineering Division**:
  "Theoretical prediction of transmutation by deuteron"
- ✓ **Fuels and Materials Engineering Division**:
  "Computer design of high toughness alloys – A study for Mg –"
- ✓ **Nuclear Chemistry Division**:
  "Chemical bonding between metal ion and separation reagent reveals Am/Cm selectivity"
- ✓ **Environment and Radiation Sciences Division**:
  "Prediction of scintillation light yield by micro-dosimetry"
- ✓ **LWR Key Technology Development Division**:
  "A numerical simulation code JUPITER for melting behavior in nuclear reactors based on computational fluid dynamics"
- ✓ **Partitioning and Transmutation Technology Division**:
  "Development of the "SELECT Process" to separate radioactive waste"

Theoretical prediction of transmutation by deuteron

Shinsuke Nakayama,[1] Naoya Furutachi,[1] Osamu Iwamoto,[1] and Yukinobu Watanabe[2]

[1] Nuclear Data Center, [2] Kyushu University

Among radionuclides produced in a nuclear reactor, those with long half-lives require long-term management and hence it is strongly desired to convert them into stable or short-lived ones. Spallation reaction is one of the candidates especially for transmutation of long-lived fission products (LLFPs). In recent years, it has been suggested that the use of deuteron as an incident particle improves the efficiency of transmutation, compared to other charged particles such as proton.

For the feasibility study of transmutation system with a deuteron primary beam, accurate cross section data of deuteron-induced reactions over a wide range of target mass numbers and incident deuteron energies are indispensable. However, the amount of currently available experimental data of deuteron-induced reactions do not necessarily meet the requirement. In such a case, reliable theoretical model calculations play a key role in providing the necessary cross section data by interpolating or extrapolating experimental ones.

Under the above situations, we have been developing a code system dedicated for deuteron-induced reactions, called DEURACS [1]. In the present study [2], we improved DEURACS and applied it to the calculation of deuteron-induced spallation reactions on typical LLFPs. In deuteron-induced reactions, three types of composite nuclei can be formed by the absorption of either neutron or proton in the incident deuteron, or the absorption of the incident deuteron itself. In DEURACS, a calculation considering this effect is performed. On the other hand, in the conventional codes for nucleon-induced reactions, such as CCONE, it is assumed that composite nuclei are formed only from the deuteron absorption.

Figure 1 shows the calculated and experimental isotopic production cross sections for the ^{93}Zr+*d* reaction at the incident deuteron energy of 210 MeV. For comparison, we also present the calculation results with the CCONE code, in which the deuteron breakup processes are not taken into account. As shown in the figure, obvious differences are seen between the two calculations, and DEURACS reproduces the experimental data well in the wide range of mass number. Moreover, we have confirmed that the calculated values of DEURACS are in good agreement with the experimental ones also for the ^{107}Pd+*d* reactions at the incident deuteron energies of 236 and 392 MeV.

From these results, it can be concluded that the framework of the improved DEURACS is applicable to deuteron-induced spallation reactions on LLFPs. These results also demonstrate that consideration of the breakup processes is essential to predict deuteron-induced spallation reactions.

Research and development on a transmutation system using a deuteron primary beam will make great progress by the present work. In addition, the result of the present work is expected to make a large contribution to various fields related to deuteron-induced reactions, such as production of medical radioisotopes, radioactivity evaluation in deuteron accelerator facilities, and so on.

This work was funded by ImPACT Program of Council for Science, Technology and Innovation (Cabinet Office, Government of Japan).

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1) S. Nakayama et al., Phys. Rev. C **94**, 014618 (2016).
2) S. Nakayama et al., Phys. Rev. C **98**, 044606 (2018).

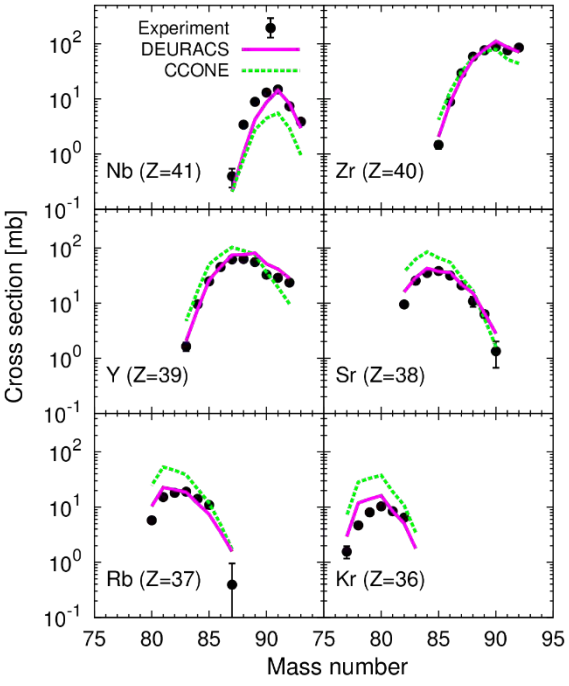

Fig.1 Calculated and experimental isotopic production cross sections for the ^{93}Zr+*d* reaction at 210 MeV.
The closed circles are the experimental data. The solid and the dashed curves represent the calculation results of DEURACS and CCONE, respectively.

Computer design of high toughness alloys –A study for Mg–

Tomohito Tsuru

Research Group for Radiation Materials Engineering

Mg alloys are promising candidates for next generation lightweight structural materials, especially for transportation equipment in terms of energy and environmental conservation. However, widespread application of wrought Mg alloys is limited because of their low formability at room temperature. The low formability of Mg alloys is caused by their low ductility and fracture toughness. The former is because of their strong plastic anisotropy originating from their hexagonal close-packed (HCP) structure and the latter is attributed to the intrinsic feature of their electronic structure.

An important property to improve the formability is the fracture toughness because pure Mg is intrinsically brittle, where local strain concentration occurs at twin boundaries (TBs) and cracks mainly propagate along these boundaries. We have systematically investigated the fracture toughness of Mg-M binary alloys from the viewpoint of the effect of solute elements. In the present study, we introduce an electronic-structure-based approach combined with fracture mechanics to understanding the effect of solute elements on interfacial fracture of Mg alloys. Rather than solely focusing on the segregation energy at the surface and interface, we focus on the comprehensive mechanism caused by the electronic structure, which dominates cohesion or decohesion of the interfacial fracture.

Figure 1 shows atomic models of typical TBs observed in experiments. We constructed several types of TB configurations to investigate the effect of various solute elements on the ideal work of interfacial separation. Three different TBs were constructed to investigate the difference in the type of boundary, including energetically stable and unstable boundaries. We calculated the surface energy of free surface was evaluated as well as the interfacial energy and applied the energy-based Griffith criterion for crack propagation [1]. The theory and computational procedure are described in our paper.

The difference in the ideal work of interfacial separation between the Mg-M alloys and pure Mg in terms of the site occupancy at the TB site for the (1012) TBs is summarized in Fig. 2 (a), where positive value corresponds to solute with toughness and negative to solutes with embrittlement. The values for Li, Ca, Sn, and Pb are very small or negative, which indicates that these elements cause segregation-induced embrittlement at the interface. On the other hand, the other solutes show positive values and Zr has the highest among them. Finally, we compare the calculated values with experiments in Fig. 2 (b), where our prediction shows good agreement with the experimental values and the toughness is indeed improved by Zr solute. We conclude that the electronic interactions of both the bulk and the surface and their difference characterize the fracture toughness of binary alloys [2]. We hope computational simulations plays more important role in the effective approach for new alloy design.

This work is supported by “JSPS KAKENHI (Grant Nos. 16K06714, 16K06783)” and “the U.S. Office of Naval Research under Grant N00014-16-1-2304”.

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1) A.A. Griffith, Philos. Trans. R. Soc. A, **221**, 582–593, (1921).
2) T. Tsuru, et al., Acta Mater., 151, 78–86, (2018).

Reprinted from JAEA R&D Review 2018-19, 47, (2019)

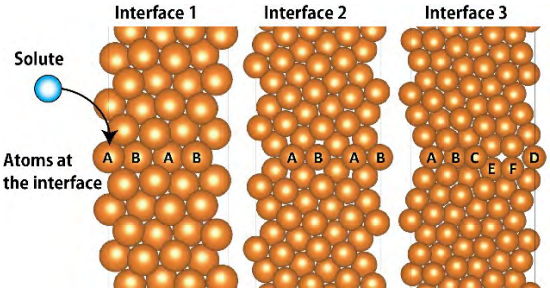

Fig.1 Atomic models of the interfaces. Atomic configurations of the various interfaces.

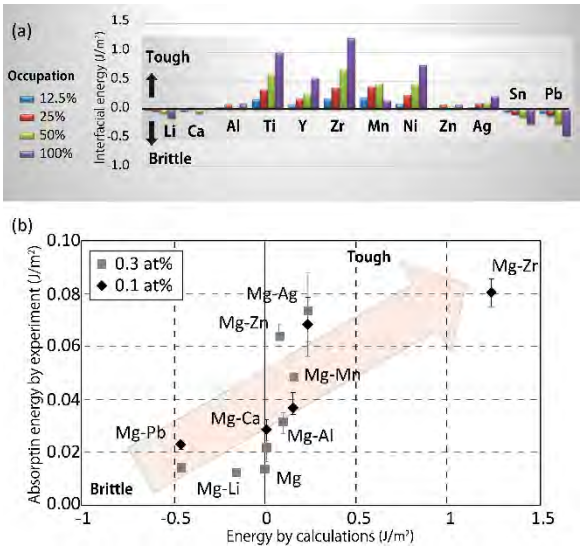

Fig. 2 (a) Energy of interfacial separation in terms of the site occupation. (b) Relationships between experiments and calculations.

Chemical bonding between metal ion and separation reagent reveals Am/Cm selectivity

Masashi Kaneko

Research Group for Radiochemistry

In order to reduce the toxicity of high-level radioactive liquid waste (HLLW), the partitioning of minor actinides (MAs), which are extremely long-lived, and the transmutation to short-lived nuclides, partitioning and transmutation, has been developed. In the partitioning process, the separation of americium (Am) from curium (Cm) has been required due to the heat generating property of Cm. However, the similarity of chemical properties between Am and Cm has made it difficult to separate them. JAEA has developed the separation process of Am from HLLW—SELECT process—to succeed in the selective separation of Am from Cm by using alkyldiamideamine (ADAAM) reagent[1]. This study revealed why the ADAAM reagent shows the high Am selectivity over Cm by using density functional theory calculation[2].

Am and Cm exist as a trivalent ion, Am^{3+} and Cm^{3+}, and form the complex with reagents. First, I modeled the complex, consisting of the metal ion (M^{3+}), nitrate ion (NO_3^-), and ADAAM reagent, as [M(ADAAM)$(NO_3)_3$] by referring to the result of the previous separation experiment[1] (**Fig. 1**). Second, I calculated the Gibbs energy difference of the formation of each complex. The comparison of the stability between the Am and Cm complexes indicated that the Am complex is more stable than the Cm complex. The calculated value of the separation factor of Am from Cm was 6.2 and reproduced the experimental value, 5.5.

I focused on the coordination bonds between the metal ion and ADAAM reagent to understand the Am selectivity over Cm of the ADAAM reagent. The ADAAM acts as tridentate ligand, one amine nitrogen atom and two carbonyl oxygen atoms. The comparison of the bond length between the metal ion and ADAAM reagent indicated that the metal-N bond length was 2.91 and 2.94 Å for Am and Cm complexes, respectively. Furthermore, I analyzed the covalent interaction of the coordination bonds by calculating the overlap of electron orbitals between the metal ion and nitrogen atom (in arb. unit). The orbital overlap of Am–N bond was larger than that of Cm–N bond (**Fig. 2**). This result indicated that the Am–N bond is stronger than the Cm–N bond. These results implied that the difference in the strength of the covalent bond between a metal ion and ADAAM reagent is a key to understand the Am/Cm selectivity. This study is expected to contribute to fundamental chemical aspects; such as understanding the bonding properties of f-block coordination compounds, as well as applied chemical aspects; such as the development of the MA partitioning process by theoretically designing extraction reagents with high and efficient MA selectivity.

This work was supported by JSPS KAKENHI Grant Number JP17K14915.

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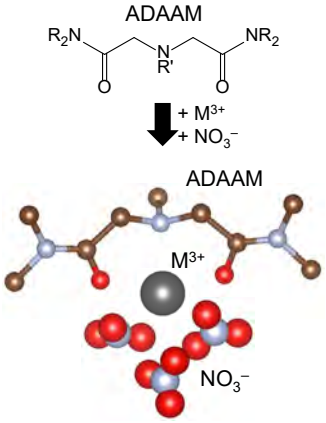

Fig. 1 Structure of model complex consisting of metal ion, ADAAM reagent, and NO_3^- ions.

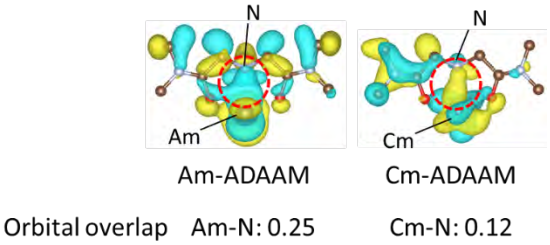

Fig. 2 Chemical bonding analysis of metal ion and ADAAM reagent.

Prediction of scintillation light yield by micro-dosimetry

Tatsuhiko Ogawa[1], Tetsuya Yamaki[2], Tatsuhiko Sato[1]

1 Research Group for Radiation Transport Analysis, 2 Takasaki Advanced Radiation Research Institute, National Institutes for Quantum and Radiological Science and Technology

Scintillators detect radiation by converting the deposited energy to photons (= scintillation light), which are subsequently converted to electric signals. One of the advantages of scintillators is linearity of the scintillation light yield to the deposited energy; therefore scintillators are applied to measure the particle energies or to identify the particle species.

However, it was known that the linearity is broken in case of high-LET (linear energy transfer) particle incidence because of the quenching effect. Birks' law [1] was commonly used to estimate the light output of scintillators with consideration for quenching effect. It assumes that light yield per unit particle flight path is described as a function of LET; therefore, it is not adequate for neutral particles, to which LET cannot be defined.

Since the quenching effect is a result of the interaction between excited fluorescent molecules, the following calculation procedures were combined in this study to estimate quenching effect universally [2]: 1, transport of radiation down to a few eV. 2, energy transfer between fluorescent molecules. 3, dissipation of the energy, which was excessively deposited to one molecule. In procedure 1, the delta-rays produced by the incoming radiation or its secondary particle were transported until they reach eV range by using RITRACKS code [3]. Procedure 2 was calculated on the basis of dipole interaction (Förster effect) between the excited fluorescent molecules. After procedure 2, all the excited molecules were assumed to emit one photon. Even if some molecules received excitation energy more than once, they deexcited non-radiatively to the 1st excitation state and emitted one photon likewise other molecules.

Figure 1 shows the light yield of NE-102A, one of the most common plastic scintillators, irradiated with protons or electrons of various energies. All calculated data were normalized for 1 MeV electrons (i.e. the light intensity owing to 1 MeV electrons is 1 MeV_{ee}). Both the calculated and the measured light yields due to electrons increase linearly with the incident energy because energy deposition by electrons is so sparse that quenching is weak. By contrast, the light yield due to protons is suppressed strongly by quenching.

The light yield was also calculated for heavy ions and compared with earlier measurement data [4] in Figure 2. Our model reproduced the light yield by various particles from protons to ^{81}Br in the low energy range, where quenching effect is strong.

The method developed in this study is useful to predict the response of scintillators as well as designing detector systems, such as telescope detector to distinguish particle species.

We wish to acknowledge Dr. Ianik Plante of NASA Johnson Space Center for the upgrade of RITRACKS for transport of ions heavier than Ni.

Reference

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3) I. Plante, *et al.*, 18. Monte-Carlo Simulation of Ionizing Radiation Tracks (in "Applications of Monte Carlo Methods in Biology, Medicine and Other Fields of Science (Edited by Charles J. Mode)"), IntechOpen (2011).

4) F.D. Becchetti, *et.al.*, Nuclear Instruments and Methods, 138, 93-104, (1976).

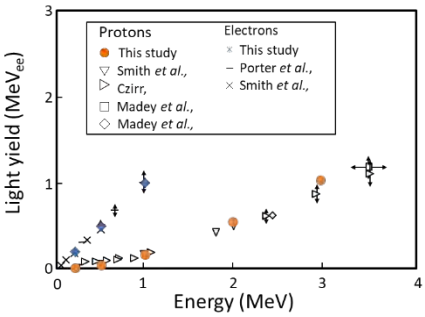

Fig.1 Comparison of scintillation light yield owing to MeV-class protons and electrons

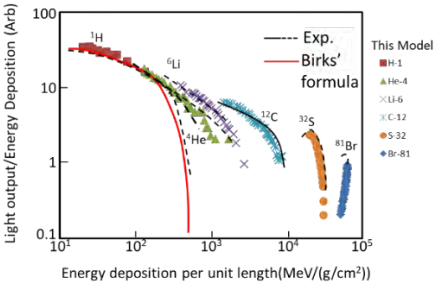

Fig.2 Comparison of scintillation light yield owing to heavy ions

A numerical simulation code JUPITER for melting behavior in nuclear reactors based on computational fluid dynamics

Susumu Yamashita[1], Kenichi Tada[2] and Hiroyuki Yoshida[1]

[1]Development Group for Thermal-Hydraulics Technology, [2]Research Group for Reactor Physics and Standard Nuclear Code System

Melting and relocation processes of reactor core structures (control rod, fuel assembly, etc.) are quite important to the accident scenario and debris distribution during severe accidents (SAs). However, melting behaviors still have a lot of uncertainties and are not yet fully understood. Although conventional SA-analysis codes have been attempting to understand melting behavior, detailed melting behavior of such structures was not revealed yet because of uncertainties of numerical models, e.g., assumptions about the phenomena and simplify the shapes of reactor-core structures, in SA analysis-codes. And there is no detailed melting behavior simulation code until now.

Therefore, we have developed a numerical-simulation code for melting and relocation behavior called JUPITER, based upon a mechanistic methodology. JUPITER can simulate melting and relocation without assumptions or simplification using basic equations governing the thermal-hydraulic behavior of the fluids in Eulerian grid system. Besides the fluid dynamics models, chemical reaction such as an eutectic reaction and oxidation model can also be taken into account, which is based on material science. Therefore, the melting behavior induced by oxidation and eutectic reaction is estimated mechanistically in the fully Eulerian grid system.

Because melting phenomena in a nuclear reactor are complicated, huge computational resources will be required. In order to realize the massively parallel computing in the state-of-the-art supercomputer, high-efficient parallel computing techniques were also implemented to JUPITER. Then, parallel computation using more than two hundred thousand cores was performed successfully.

By using JUPITER, we simulated melt-relocation from the bottom of reactor pressure vessel to the lower part of the primary containment vessel (pedestal) [1]. In this simulation, we assumed the highest density material, molten UO_2, should be poured first, followed by the lower density materials stainless steel, Zry, and B_4C. Fig.1 shows a cross-sectional view of the debris in the pedestal along its center to illustrate the accumulation state inside the sump pits. The melt near the wall of the pedestal is solidified by heat transfer to the wall. The highest density material, UO_2, is located at the bottom and the lowest density material, Zry, is located at the upper part. This distribution is given by mechanistic estimation for melt relocation and solidification based upon the physical governing equations.

In addition, we try to evaluate the re-criticality of fuel debris in the pedestal using the continuous-energy-neutron-transport Monte Carlo code MVP with this fuel-debris distribution by JUPITER. In an ordinary criticalicality analysis, it is assumed that the fuel debris is uniformly mixed, and effects of debris distribution are neglected. Here, with the distribution given by JUPITER, re-criticality can be calculated under more realistic conditions. As a result, we confirmed effects of nonuniform distribution of the debris on the re-criticality analysis.

Recently, JUPITER has been released at PRODAS [2]. We are continuously improving JUPITER to enhance the light water reactor safety. In addition, decommissioning of Fukushima-Daiichi NPS by evaluating the characteristics and re-criticality of fuel debris through updating the numerical models and calculating the melting behaviors under severe accident conditions.

Reference

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2) PRODAS (PROgram and Database retrieval System), http://www.rist.or.jp/nucis/

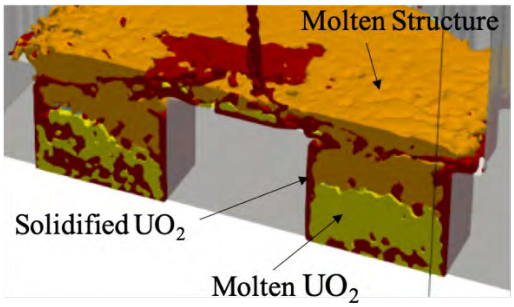


Fig.1 Interior distribution of accumulated debris (numerical result)

Development of the "SELECT Process" to separate radioactive waste

Tatsuro Matsumura

Research Group for Partitioning

Partitioning and transmutation (P&T) has been proposed as one of measures to reduce the radioactive toxicity and the volume of radioactive waste generated from the reprocessing of spent nuclear fuels by hydrometallurgical method. To this end, highly toxic radionuclides should be separated from high-level radioactive liquid waste, and studies for developing practical partitioning processes have been carried out. The former processes proposed to date demonstrated excellent performance to separate fission products (FPs), rare earths (REs) and minor actinides (MAs), however, some processes employed the reagents consisted of phosphorus or sulfur which cannot be decomposed into gases by incineration.

Thus, we have been developing the extractants consisting of carbon, hydrogen, oxygen, and nitrogen atoms (so-called CHON principle) in that such extractants can be decomposed into gases by incineration and this leads to decrease in the volume of the secondary waste. Recently, we have proposed a hydrometallurgical method called SELECT (Solvent Extraction from Liquid-waste using Extractants of CHON-type for Transmutation) aiming at recycle use of nuclear materials and separating actinides for transmutation. A conceptual flow sheet of SELECT process is shown as Fig. 1. SELECT process consists of the following four steps: (i) recovery of U and Pu from dissolution solution of spent nuclear fuels, (ii) recovery of Am, Cm, and rare earth elements (REs) from HLW, (iii) separation of REs from Am and Cm, and (iv) separation of Am and Cm.

We conducted a continuous counter-current experiment using mixer-settler extractors installed in a hot cell at NUCEF (NUclear fuel Cycle safety Engineering research Facility) at the Nuclear Science and Research Institute to evaluate the applicability of TDdDGA as an extractant for the 2nd step (Fig. 2). The experiment was performed successfully over a cumulative operation time of 10 h. TDdDGA effectively extracted Am and Cm. More than 98% of Am and Cm were recovered and not detected in the raffinate. The recovery of Y, La, Nd, and Eu were 94.0%, 99.9%, 99.9%, and 86.9%, respectively. Furthermore, the recovered MAs and REs had no remarkable contamination with fission products such as Sr, Cs, Zr, Mo, Ru, Rh, and Pd.

The present results demonstrated that TDdDGA can be used as the extractant in the 2nd step, and promoted the development of SELECT process.

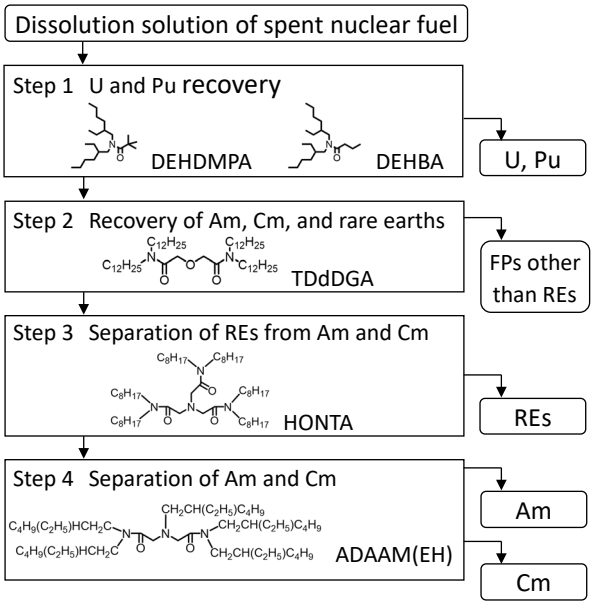


Fig.1 A conceptual flow sheet of SELECT process and proposed extractants in each step

Reference

1) Y. Ban, et al., Solvent Extraction and Ion Exchange, 37(1), 27-37, (2019).

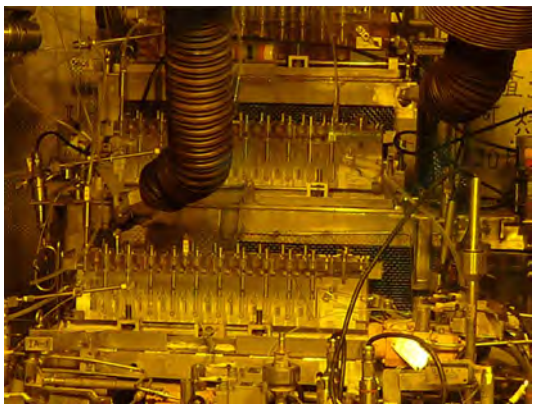
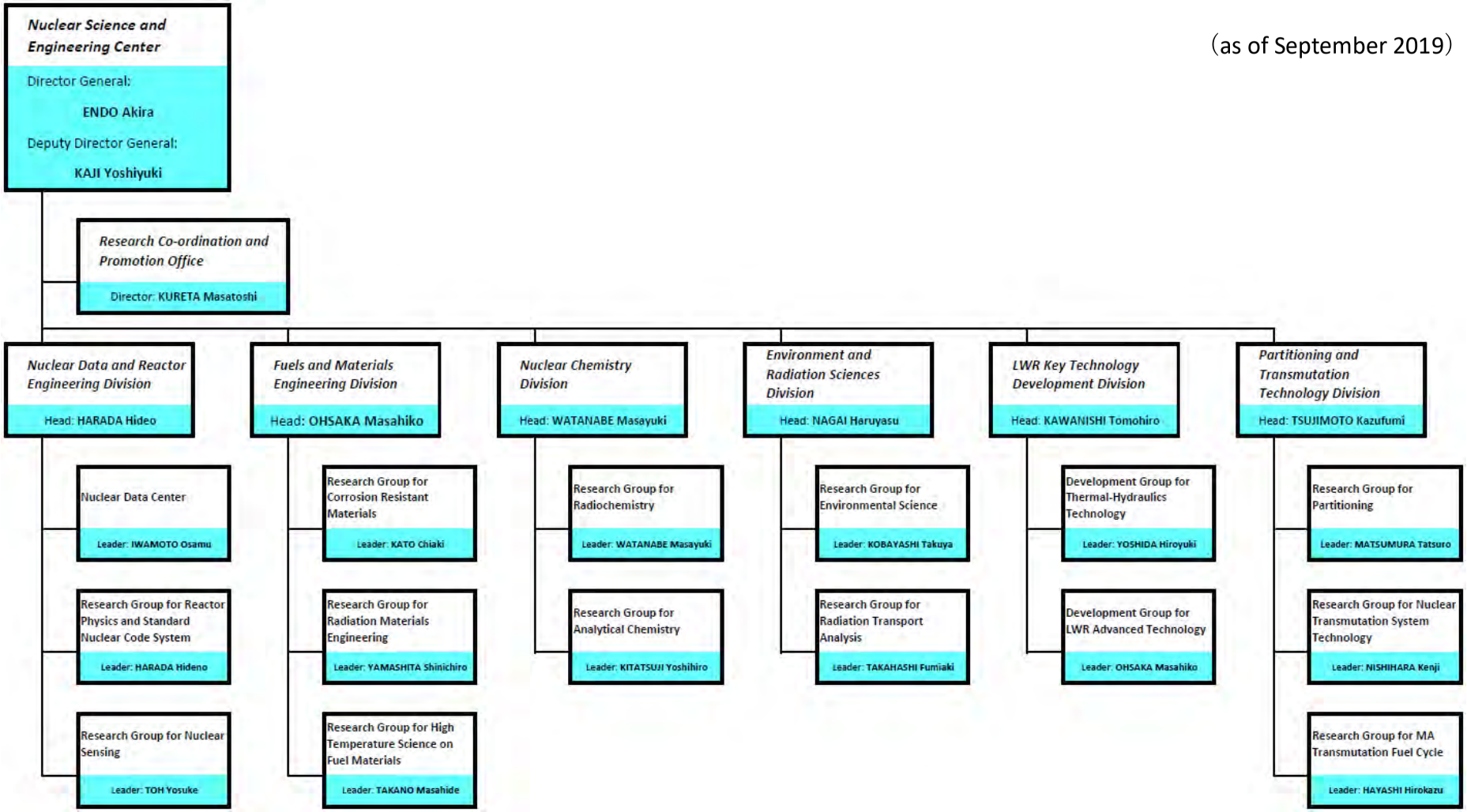


Fig.2 Mixer-settler extractors used in the continuous counter-current experiment to recover Am, Cm, and rare earths by TDdDGA.

FY2018 NSEC Group Activities

The NSEC of JAEA consists of 15 Groups.

Organization of NSEC



（as of September 2019）

NSEC URL: https://nsec.jaea.go.jp/en_index.html

Nuclear Data Center

The mission of Nuclear Data Center is to provide reliable nuclear data which are required in various applications such as nuclear reactors, accelerators, medical use of radiations, and so on. To achieve it, we engage in research works on nuclear data measurement, nuclear theories, and nuclear data evaluation related to nuclear reaction and structure collaborating with nuclear data researchers and organizations in Japan as well as in the world. The evaluated nuclear data have been compiled into databases called JENDL and are available from our website http://wwwndc.jaea.go.jp.

Measurement of MA cross sections

Cross sections of minor actinide (MA) are important in estimating production of MA in nuclear reactors and also needed in research on reduction of MA by nuclear transmutation. Total and capture cross sections of neutron reaction on ^{241}Am were measured in high accuracy with ANNRI installed at MLF in J-PARC[1]. In this work, uncertainty originated from amounts of the Am samples was significantly reduced by the utilization of a calorimeter which could measure decay heat of samples precisely. The total cross section was determined from 4 meV to 2 eV and capture cross section from 10 meV to 100 eV by time-of-flight method. The thermal capture cross sections were obtained with uncertainties of 4.1% which is nearly a half of current uncertainty of evaluated data of JENDL. Obtained capture cross section is shown in Fig.1.

Measurement of γ-ray emission probability of Am

The γ-ray emission probability I_γ is a basic property of nucleus and important to determine the amount of radiative material. The I_γ of ^{244g}Am were measured by the activation method using neutron capture reaction on ^{243}Am at Kyoto University Research Reactor Institute[2]. Using the known decay scheme, the absolute value of I_γ for the principal transition was precisely determined with the uncertainties of a few % which was remarkably reduced from almost 30% in the latest version of Table of Isotopes. Obtained emission probabilities are shown in Table 1.

Evaluation of Cross Sections

Next version of JENDL is under development. Revision of the data of structure material is one of main targets, because they are important for neutron transportation in material and also for their activations. Neutron cross sections were evaluated for Zr and Cu isotopes using the state-of-the-art nuclear reaction model code CCONE developed at JAEA. The available experimental data of various nuclear reactions, elastic scattering angular distributions, secondary neutron emission spectra were consistently analyzed by CCONE and deduced the reliable evaluated data[3,4].

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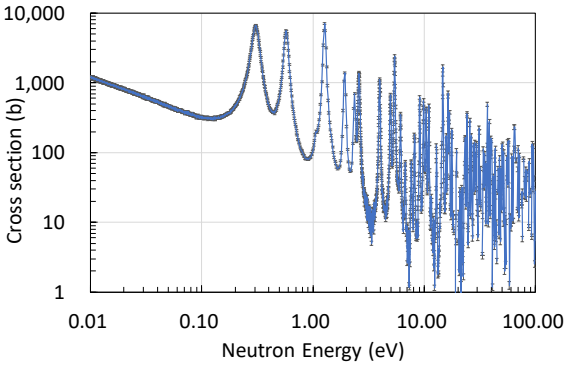

Fig.1 Am-241 neutron capture cross section

Energy Eγ (keV)	Emission probabilities (%)
153.863	17.1 ± 1.2
538.400	<0.42
743.971	66.5 ± 1.1
897.848	28.0 ± 1.0

Table 1 γ-ray emission probability of ^{244g}Am

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Research Group for Reactor Physics and Standard Nuclear Code System

The role of nuclear codes is increasing because of rapid advancement of computer technology. It enables to evaluate in details the energy and spatial distribution of neutrons and photons emitted after fission reaction in a nuclear reactor. Key properties such as criticality, decay heat, and radioisotope production rate etc. can be deduced from these neutronics calculations by these nuclear codes.

We are developing standard nuclear codes to support and advance the peaceful use of nuclear energy and technology. The main codes we are developing are **MVP**, **MARBLE**, and **FRENDY**. The advancements of each code in 2018 JFY are summarized below.

We are also collaborating tightly with nuclear data center to validate the next version nuclear data library JENDL-5, which will be processed and utilized in these nuclear codes.

MVP

MVP realizes fast and accurate Monte Carlo simulation of neutron and photon transport processes. The version 3 of MVP[1] was released in 2018 JFY and available from RIST in Japan and OECD/NEA data bank as NEA-1673 MVP/GMVP V.3. The cross section library for MVP3 was produced as MVPLIB2017 for user convenience from JENDL-4.0, 3.3, 3.2, JENDL/AC-2008, ENDF/B-VII.1, VII.0, VI.8, JEFF-3.2, 3.1.2, 3.1, 3.0, 2.2 and MVP photo atomic, photonuclear and dosimetry libraries. The MVPLIB2017 can be downloaded from our group's home page.[2]

MARBLE

MARBLE is a versatile reactor analysis code system. The version 2 of MARBLE enables one to execute almost all analysis functions of the JAEA's conventional neutronics code system for fast reactors with the unified user interface. By utilizing this function of the MARBLE-2, a new unified cross-section set ADJ2017 has been developed[3]. The ADJ2017 is based on JENDL-4.0 and adjusted using a total of 620 integral experimental data set. The ADJ2017 is expected to be utilized as a standard database of FBR core design.

FRENDY

FRENDY is a new nuclear data processing code. The role of the code is schematically shown in Fig.1. It enables to process nuclear data such as JENDL in ENDF-6 format. Users can easily utilize the FRENDY's functions in their own codes. The first version of the FRENDY treats the ENDF-6 format and generate the ACE files which is used for continuous energy Monte Carlo codes such as PHITS and MCNP. The FRENDY version 1 was released in March 2019 as an open source software from our group's home page.[5]

Reference

1) Y. Nagaya et al., JAEA-Data/Code, 2016-018, (2017).
2) https://rpg.jaea.go.jp/main/en/program_mvp/
3) K. Yokoyama et al., JAEA-Research, 2018-011, (2019).
4) K. Tada et al., JAEA-Data/Code, 2018-014, (2019).
5) https://rpg.jaea.go.jp/main/en/program_frendy/

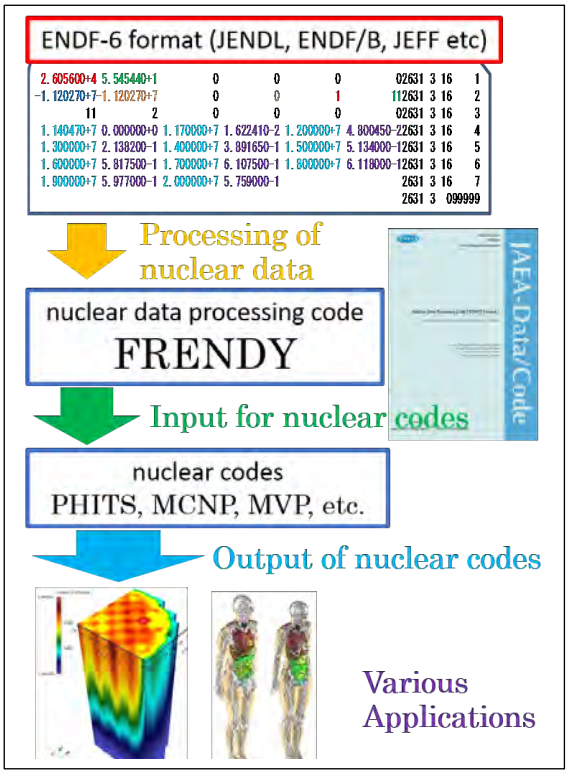

Fig.1 Role of FRENDY bridging nuclear data libraries such as JENDL and various nuclear calculation codes such as PHITS

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Research Group for Nuclear Sensing

Non-destructive analysis (NDA) is a key tool for nuclear safeguards, nuclear security, nuclear waste management and nuclear decommissioning. However, new and advanced NDA techniques are still required to address nuclear material accountancy of Special Nuclear Materials (SNMs) and Minor Actinides (MA) in highly radioactive nuclear materials, because high levels of radiation make it very difficult to measure the amount of SNMs and MA. Developments of low-cost, accurate and practical NDA systems are also needed for effective implementation. We have therefore developed NDA system and related methodologies. The following are a part of our group activities in 2018.

NDA system for the fuel debris

In decommissioning of Fukushima Daiichi Nuclear Power Plant, material accountancy of SNMs in fuel debris produced in the nuclear accident is required. Since the fuel debris contains many different materials, it is hard to measure SNMs. We have developed the Fast Neutron Direct Interrogation (FNDI) method that can measure a variety of nuclear materials nondestructively with reducing the influence of sample matrices. In order to quantify the amount of SNMs in the fuel debris, we designed a measurement system based on the FNDI method. In particular, we developed a fast response detector bank for fast neutron measurements using Monte Carlo simulations. The new detector bank has more than one order of magnitude faster response compared to the standard ones. Then we evaluated the nondestructive measurements of SNMs in the fuel debris. The results show that SNMs in the fuel debris can be measured accurately in many cases because the influence of the sample matrices can be reduced by using the designed system[1)].

NDA system for nuclear security

With the aim of establishing an active neutron NDA system for useful applications in the field of nuclear non-proliferation and nuclear security, active neutron NDA techniques: Differential Die-Away Analysis (DDA), Prompt Gamma-ray Analysis (PGA), Neutron Resonance Capture Analysis (NRCA), Neutron Resonance Transmission Analysis (NRTA) and Delayed Gamma-ray Analysis (DGA), have been studied and improved. By using the NDA techniques, we have been developing a new NDA system capable of quantifying fissile (U, Pu) mass contained in highly radioactive nuclear materials[2)]. In 2018, we mainly designed DDA and PGA system using Monte Carlo simulations.

The DDA is one of active neutron measurement technique based on active interrogation of nuclear material samples by external neutrons. The detection limit of the newly developed DDA system for the analysis of a large sample such as a MOX can container (2 liter) was investigated by the Monte Carlo simulation studies and experimental tests. As the results, it was found that the DDA system could detect less than 10 mg of ^{239}Pu, and the optimum thickness of the cylindrical HDPE polyethylene moderator is around 3cm-5 cm[3)].

Although one of the main targets is a spent fuel with a pellet shape, there is little knowledge of how well NRTA determines the amount of SNMs in a pellet fuel. In order to investigate the effect of the sample shape in the NRTA measurement, NRTA experiments were made with simulated spent fuel pellets (W, Ag, In, Rh) at a 10-m transmission station of the GELINA facility of EC-JRC in Geel. Measured transmission spectrum clearly shows resonance dips derived from the individual nuclei. The obtained abundances agree with expected ones within 7.5%, suggesting the potential of a compact NRTA system to measure the amount of SNMs in nuclear fuels[4)].

This research was implemented under the subsidiary for nuclear security promotion of MEXT.

Reference

1) M. Maeda et al., J. Nucl. Sci. Tech., Vol56, 617-628, 2019

2) Y. Toh et al., Proceedings of INMM 59th Annual Meeting, 2018

3) A. Ohzu, et al., Proceedings of the 2018 IEEE NSS & MIC conference record, Sydney, 2018

4) H. Tsuchiya, et al., Proceedings of INMM 59th Annual Meeting, 2018

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Research Group for Corrosion Resistant Materials

We have been researching on corrosion of metal materials used for nuclear facilities. If the corrosion occurs, the components of the nuclear facility will degrade, which may cause holes and cracks. Therefore, we are focusing on the corrosion mechanism to develop the methods of corrosion prediction and prevention. Through these research activities, we contribute to improving the reliability of nuclear facilities.

Researches on corrosion mechanisms of nuclear fuel reprocessing plant materials

Reprocessing plant materials will be corroded intricately by the reprocessing solution which containing severe corrosive nitric acid and several varieties of highly oxidizing metallic cations derived from spent nuclear fuels. The critical factor for the corrosion is "re-oxidizing" behavior of the cations. These cations cause intergranular corrosion in stainless steel because oxidizing cations shift the corrosion potential to noble (Fig.1). The re-oxidation rate constants of oxidizing cations, such as Cr, V, Pu, and Np, were analyzed and were discussed in terms of the effect on time dependencies of the corrosion rate. Our data showed that the cations with a high re-oxidation rate constant, such as Np, could keep the corrosion rate at a significant level continuously for the long immersion duration.

Researches on corrosion mechanisms of Boiling Water Reactor (BWR)

Stainless steels (SS), such as type 316L, show stress corrosion cracking susceptibility in boiling water reactor environments. It's important to understand the corrosion behavior of the crack tip in order to make the mechanism of SCC clear. The in-situ measurement of electrical conductivity of solution within crevice of type 316L in 288 °C water has been conducted with a newly developed electrochemical sensor system. The sensor consists of an electrode (ϕ ≈250 μm stainless steel) and an insulator (high purity alumina) directory imbedded into crevice former plate. The sensors were installed at different positions within tapered crevice of SUS316L(Fig.2). The sensor measures local electrical conductivity (k) of crevice solution beneath the electrode with electrochemical impedance method. At sensor-A position with gap ≈ 4.4 μm, k increased with time and localized corrosion occurred in the vicinity of this position (Fig.3). Thermodynamic equilibrium calculation based on oxide in the crevice showed that the pH value of crevice solution can reach to 3.53. We concluded that acidification occurred in a tight crevice and resulted in localized corrosion of stainless steel.

Reference

1) E.Irisawa et.al., *J. Nuc. Sci. and Tech*., 56, 337–344, (2018).
2) Y.Soma et.al., *Zairyou-to-Kankyo*, 67, 381-385, (2018).

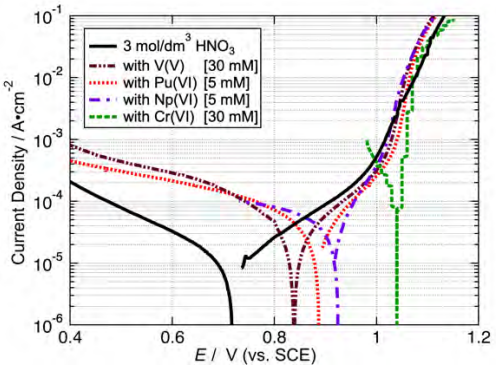


Fig. 1 Polarization curves of SS (SUS310Nb) in the boiling 3 M-HNO_3 containing oxidizing cations[1].

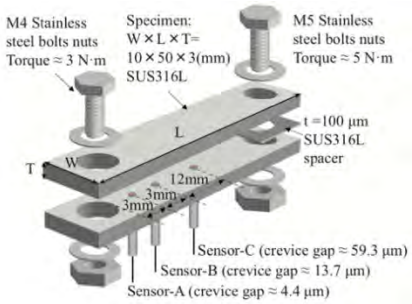


Fig. 2 Schematic illustration of crevice specimen with sensors.

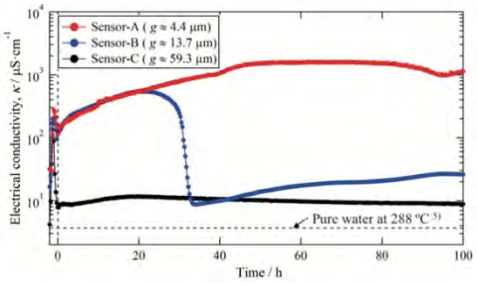
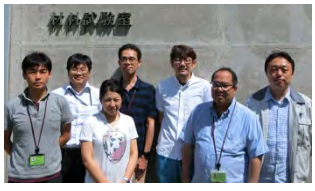


Fig. 3 Change in κ as a function of time in 288 °C pure water with 32 ppm concentration on dissolved oxygen.[2]



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Research Group for Radiation Materials Engineering

We are conducting several kinds of researches to deepen the understanding of material degradation mechanism under radiation environment. A study on stress corrosion cracking (SCC) of low carbon austenitic stainless steel (SS) is one of the representative research topics in our research group. This research subject is still recognized as a significant issue on material degradation because some crucial questions remain unanswered. Besides, we are also making the most use of computational atomistic simulations to clarify the unrevealed mechanism of material deterioration.

Unanswered questions on SCC

In spite of extensive studies on SCC in low carbon SSs, the initiation and propagation mechanisms have not been fully understood yet. The depletion of Cr concentration in the vicinity of grain boundaries (GBs) is considered to be the main cause of the standard grade of SS. However, such sensitization phenomenon was not confirmed irrespective of the investigation of a Type 316L SS sample extracted from a cracked core shroud of a Japanese boiling water reactor. In order to approach the degradation mechanism such as cracking occurred in low carbon SSs, the effect of long-term thermal aging (LTA) on SCC initiation susceptibility was investigated using Type 316L SS with different material parameters such as cold working (CW) and LTA. Figure 1 showed the number of surface cracks initiated during creviced bend beam (CBB) testing at 288 °C for 1000 hours. Type 316L CW demonstrated resistance to cracking, whereas Type 316L CW + LTA indicated the high SCC initiation susceptibility [1].

Computational materials science based on atomistic simulation

A mechanism of anomalous tension-compression (T-C) asymmetry in ultrafine-grained (UFG) aluminum (Al) and copper (Cu) is proposed using large-scale atomistic simulations and dislocation theory. Unlike coarse-grained metals, UFG Al exhibits remarkable T-C asymmetry of the yield stress. Figure 2 showed stress-strain curves and T-C asymmetry in UFG Al and Cu with different dislocation densities under uniaxial stress. The yield stress σ_y was defined as the maximum stress in these curves. The yield stress decreased with increasing dislocation density under both tension and compression [2]. The atomistic simulations reveal that the yield event is not related to intergranular dislocations but caused by dislocation nucleation from the GBs.

Reference

1) S. Aoki, et al., "Effect of long-term thermal aging on SCC initiation susceptibility in low carbon austenitic stainless steels", Proc. 18th International Conference on Environmental Degradation of Materials in Nuclear Power Systems –Water Reactors, p.1879, Springer (2018)-Published 7 October 2017.
2) T. Tsuru, "Origin of tension-compression asymmetry in ultrafine-grained fcc metals", Phys. Rev. Materials 1, 033604-Published 9 August 2017.

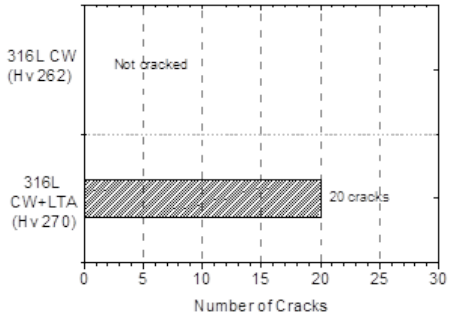

Fig.1 Number of surface cracks in Type 316L SS initiated during CBB testing.

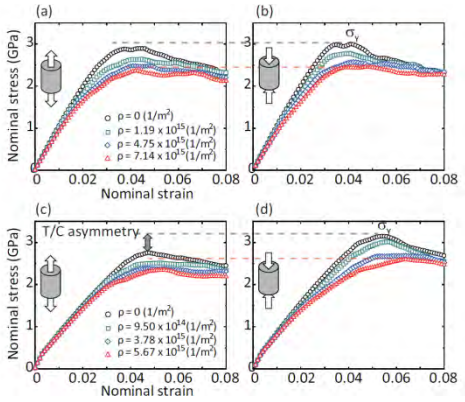

Fig.2 Stress-strain curves and T-C asymmetry in UFG metals: (a) Cu under tension, (b) Cu under compression, (c) Al under tension, and (d) Al under compression.

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Research Group for High Temperature Science on Fuel Materials

We treat issues on nuclear fuel materials for light water reactors and advanced reactors in the future. Experimental works on uranium and non-radioactive surrogate materials are performed in the Research Building No. 4 with various heating devices, analytical equipment, and material property measurement apparatus. Those on transuranium elements (TRU: Pu and minor actinides) are performed at the TRU-HITEC in NUCEF, consisting of specially designed hot cells and glovebox with highly purified argon atmosphere. We also take advantage of computer science to understand or predict phenomena concerning the fuel materials. Fuel performance code and thermodynamic calculation code are strong tools, so we make sustained efforts to upgrade the fuel material properties database and thermodynamic database having high practicality.

R&D on MA transmutation fuel

For the volume and radiotoxicity reduction in the high-level radioactive wastes, partitioning and transmutation of minor actinides (MA: Np, Am, Cm) are key technologies. We have been engaged in R&D on the MA-bearing nitride fuel for accelerator driven system (ADS) as our main mission in these two decades. The potential fuel forms are (MA,Pu,Zr)N single phase solid solution pellet or (MA,Pu)N/TiN dispersion pellet, in which ZrN and TiN are inert matrices to dilute TRU. Since 2016 a big project "R&D on nitride fuel cycle for MA transmutation to enhance safety and economy" has been carried out by a competitive fund of the MEXT through FY2019. This project covers the elemental technologies needed for engineering scale fuel fabrication and the fuel behavior data needed for safety analyses under irradiation. The annual topic on fuel fabrication in 2018 is the sol-gel process to obtain sphere particles of the oxide and fine carbon powders as precursor for carbothermic nitridation. By using Dy as surrogate material, the appropriate process parameters such as feed solution composition, viscosity, dropping nozzle size, and frequency were determined. These results enabled us to obtain excellent shape of particles with controlled diameter (see Fig. 1). Details of the R&D project and progress were presented at NuMat 2018 international conference as an invited lecture.[1)]

Thermodynamics of fuel materials

In order to contribute to decommissioning of the Fukushima Daiichi Nuclear Power Station, we have been studying on characteristics of the fuel debris from the aspects of laboratory scale experiments and thermodynamic evaluation. The interaction tests between UO_2 and Zr were performed at around 2173 K using UO_2 crucible and Zr metal pellet. From the results of the surface microanalysis, we explained the mechanisms of uranium diffusion and fuel degradation from thermodynamic perspective (Fig. 2).[2)]

Reference

1) M. Takano, "Material challenges for future nuclear fuel cycles –R&D on nitride fuel cycle for MA transmutation," NuMat 2018, Oct. 2018, Seattle.
2) N. Shirasu, M. Kurata et al., "High-temperature interaction between zirconium and UO_2," in proc. FDR 2019, paper No. FDR2019-1067.

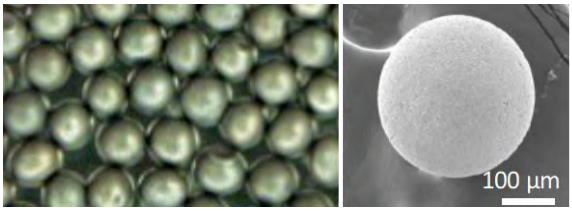

Fig.1 Dy_2O_3+C mixed particles (ϕ 0.33 mm) calcined from the gel particles (ϕ 0.47 mm).

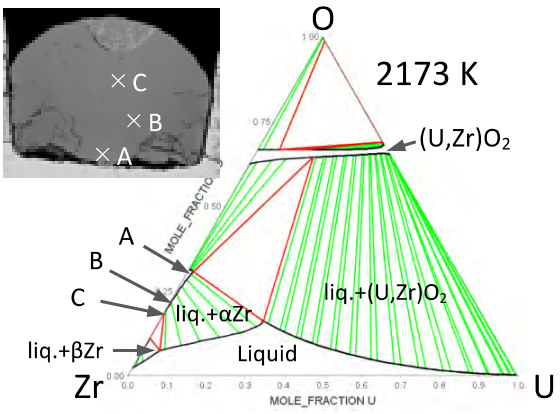

Fig.2 Cross section of test piece and ternary phase diagram of U-Zr-O system at 2173 K.

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Research Group for Radiochemistry

Radiochemistry provides fundamental information about the essential materials in developing nuclear technology by identification of the chemical reaction of radioactive materials by measurement and chemical separation. We chemically resolve various issues arisen from nuclear industry by developing new technique of measurement and computational method.

Chemical Reactivity of Nuclear Fuel

We have shown that uranium dioxide (UO_2) becomes chemically stable by forming solid solution containing zirconium (Zr). The stabilization by Zr indicates that the molten fuel debris in TEPCO's Fukushima Dai-ichi Nuclear Power Plants (1F) would be stable against dissolution by chemical reactions and would remain in the reactors until it is retrieved.

The results on the dissolution of U in the simulated fuel debris by H_2O_2 are shown in Fig.1[1]. It is significantly inhibited by the incorporation of Zr. In this experiment, U oxide containing Zr was used as simulated fuel debris, because the fuel debris is expected to contain Zr by melting with the fuel-cladding material. When the simulated debris was exposed to H_2O_2, the reaction of H_2O_2 proceeded at a rate comparable with that of UO_2. However, the dissolution of U was significantly inhibited (Fig.1).

Interfacial Spectroscopy for Nuclear Engineering Chemistry

We have been developing advanced laser spectroscopy, so-called Sum Frequency Generation (SFG) spectroscopy which can identify the chemical structure on the surface and interface in the materials. In the nuclear industry, solvent extraction is one of the promising chemical techniques for isolating specific elements in such as radioactive waste or spent nuclear fuel. We address the SFG to improve the efficiency of solvent extraction. As a representative example of common solvent extraction system, for instance, the interface vibrational spectra of extraction of europium ions (Eu^{3+}) using di-2-ethylhexyl phosphate extractant (HDEHP) were obtained (Fig.2)[2]. As the concentration of Eu^{3+} increases, the positive sign of the water signal becomes negative. Positive and negative indicates that interfacial water molecules pointing upward (hydrogen-up) change to a downward (hydrogen-down) orientation as Eu^{3+} is adsorbed to the interface. That is, interfacial water molecules are bonded to Eu^{3+} with hydrogen-down orientation (Fig.2). The structure of Eu^{3+} sandwiched between HDEHP and water molecules has not been reported so far in the organic or aqueous phases, so that the Eu^{3+} structure at the interface is unique.

Reference

1) Kumagai, Y. et al., Journal of Nuclear Materials, vol.497, p.54–59, 2017.
2) Kusaka, R. et al., Physical Chemistry Chemical Physics, vol.20, issue 4, p.2809–2813, 2018.

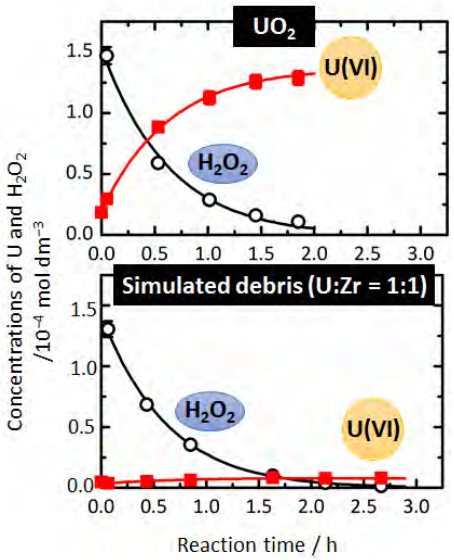

Fig.1 Comparison of the H_2O_2 reaction kinetics between UO_2 and the simulated fuel debris

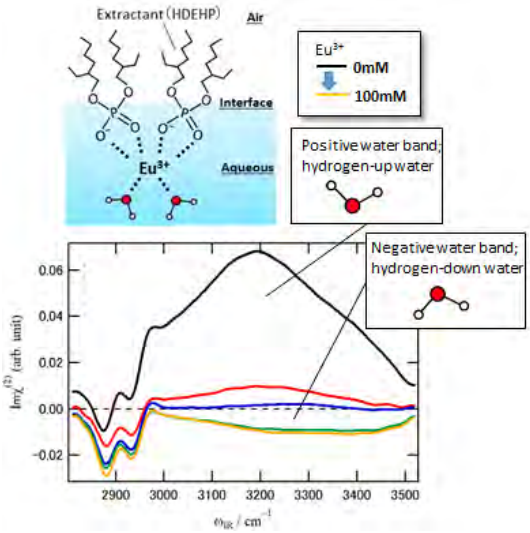

Fig.2 Vibrational spectra of the interface obtained by advanced laser spectroscopy: heterodyne-detected vibrational sum frequency generation spectroscopy

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Research Group for Analytical Chemistry

Accurate analyses of radioactive substances are fundamental technologies supporting the nuclear usage. Removal of interference elements prior to instrumental analysis and control of the chemical state of the analyte are key techniques for the high precision chemical analysis. The research group for analytical chemistry is developing practical analytical methods for actinide and fission products based on studies of elucidation of chemical reaction and ion separation phenomena such as adsorption, precipitation, aggregation, etc.

Radioanalytical chemistry

We have been committed to the development of a wide range of analytical techniques for the determination of long-lived radionuclides. Estimating the risks associated with radiation from long-lived radionuclides in radioactive waste is essential to ensure the long-term safety of potential disposal sites. Our research focuses on developing analytical techniques based on inductively-coupled plasma mass spectrometry (ICP-MS) along with its preparation procedures using solid phase extraction (SPE) materials. We utilized an in-house microvolume SPE cartridge for the sample preparation of ICP-MS determination of ^{93}Zr[1], achieving rapid and reliable measurement (Fig. 1). Another research interest is laser ablation (LA), one of the hyphenation techniques to ICP-MS, allowing direct measurement of a solid sample. We employed LA-ICP-MS to ^{107}Pd determination[2] (Fig. 2). The solid Pd sample was obtained as precipitate by laser-induced photo reduction. These unique techniques have a capability to reduce operating time, radiation exposure to workers, and secondary waste.

Electrochemistry of Actinide

In the decommissioning of Fukushima Daiichi Nuclear Power Plant, there is concern about internal exposure due to scattering of alpha nuclides contained in debris. In order to predict transfer of the radionuclides into the contaminated stagnant water, it is necessary to accurately understand the chemical state and reaction of the radionuclides in the water of environmental condition. Redox of actinide ions in weakly acidic solution is complex reaction due to the formation of precipitate. We tried to elucidate their reaction mechanism by electrochemical impedance measurement. Electric resistances of the electrode surface covered with the uranium and neptunium deposits exhibit a change in chemical form from amorphous hydroxide to crystalline oxide. As chemical form affect the aggregation of particles, it is important factor for elucidation of the migration behavior in environmental water.

Reference

1) Asai et.al., Talanta, 185, 98-105, 2018
2) Asai et. al., Anal. Bioanal. Chem., 411, 973–983, 2019

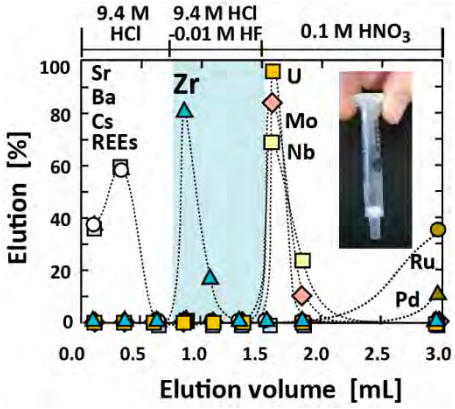

Fig. 1 Microvolume SPE cartridge
The Zr separation can be completed by 1.2 min. Almost all other elements originally contained in the spent nuclear fuel sample have been removed.

Fig. 2 Direct measurement of ^{107}Pd by LA-ICP-MS

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Research Group for Environmental Science

We are conducting research and development in order to upgrade the migration and assessment technology of environmental radioactive materials, required for environmental impact assessment of various radionuclides releases and environmental pollution events. As an application to the global environmental problem, we participate in the project "Climate Change Research Using Nuclear Science and Technology" of the Forum for Nuclear Cooperation in Asia (FNCA), and leading the research in Japan.

Prediction of the dispersion of radionuclides released into the ocean

Cesium-137 was released to the ocean by the accident at the Fukushima Daiichi Nuclear Power Station (1F) of the Tokyo Electric Power Company Holdings, Inc. In this study, we applied the oceanic-dispersion model (SEA-GEARN) developed at JAEA. Input oceanic data were calculated by oceanic general-circulation models of the Japan Marine Science Foundation (JMSF), the Japan Agency for Marine-Earth Science and Technology (JAMSTEC), the Meteorological Research Institute (MRI) of the Japan Meteorological Agency (JMA), and the National Oceanic and Atmospheric Administration (NOAA). The data-assimilation method was applied to these oceanic general-circulation models to assimilate observed data into numerical simulations. The direct-release rate of ^{137}Cs into the ocean was estimated using the observed sea surface ^{137}Cs concentrations near the northern and southern discharge channels of 1F. To provide the ^{137}Cs-deposition amounts at the sea surface, we conducted an atmospheric-dispersion simulation using the latest-estimated ^{137}Cs release rate into the atmosphere.

Compared to the simulation with a lower horizontal resolution, the higher-resolution simulation reproduced well the ^{137}Cs concentration observed in the coastline and offshore of Fukushima prefecture. The ^{137}Cs was suggested to have been spread along the coast in the north–south direction during the first few months after the 1F accident. The simulations for the western part of the North Pacific and the whole North Pacific reproduced the main ocean current favorably using the data-assimilation method, despite the relatively low resolution. This suggests that the Kuroshio Extension plays a large role in the transport process of ^{137}Cs from the coast to the outer ocean (Fig.1).

Analysis of the depth distribution of ^{137}Cs by the dispersion simulation showed that most of that transferred to the ocean by direct release or deposition from the atmosphere existed in the surface layer (0 to 211 m from the sea surface) shortly after the 1F accident. However, as time passed, it was transported from the surface layer to deeper layers (Fig.2). The ^{137}Cs amounts in the surface, intermediate (211 to 510 m), deep (510 to 1,050 m), and bottom layers (deeper than 1,050 m) one year after the 1F accident were 71, 19, 4, and 0.8 % of the total release amount, respectively.

This study suggests that the accident-derived ^{137}Cs was dispersed widely into the North Pacific and gradually dispersed from the surface to the deeper layers, regardless of the different ocean-current data.

Reference

1) Kawamura, H. et al., Journal of Environmental Radioactivity, **180**, 36–58, (2017).

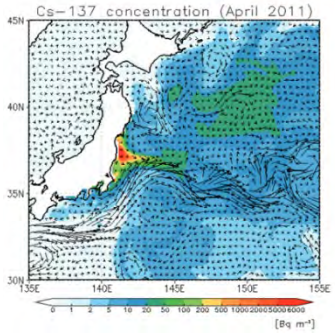

Fig.1 Averaged surface ^{137}Cs concentrations in April 2011

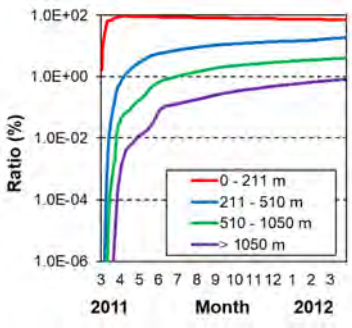

Fig.2 Time series of the ratios (%) of ^{137}Cs amounts at the calculation domain

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Research Group for Radiation Transport Analysis

Nowadays, a computer simulation technique that can analyze radiation transport in a material is essential for research and development in the field of nuclear and radiation sciences. We develop a radiation transport simulation code, Particle and Heavy Ion Transport code System (PHITS), and apply it to industrial, scientific, and medical studies. In addition, we also conduct studies on radiation protection and effects.

Upgrade of PHITS code

PHITS has been upgraded to improve its reliabilities and expand areas to be applied. In FY2018, we released PHITS versions 3.08 and 3.10. The major upgraded features for these versions are as follows. Please access PHITS homepage [1], if you have an interest in further information on the latest edition.

/ High energy nucleus-nucleus interaction model (JAMQMD2) is developed and implemented.
/ Neutrino interaction model is newly developed and implemented.
/ We establish a procedure for coupling PHITS with thermal analysis software such as ANSYS.

In recent years, "Track-structure Mode" has been developed to consider all ionization and excitation events explicitly, in addition to averaged energy deposition that has been calculated by many users. Track-structure Mode is available for electron and positron in the current version of PHITS. In FY2018, we have updated the mode to be applied to proton and all kinds of carbon ions. The updated mode can explicitly simulate carbon ion track in water and plot the position where ionization and excitation events might occur (Fig.1). We will implement the updated mode to a future version of PHITS.

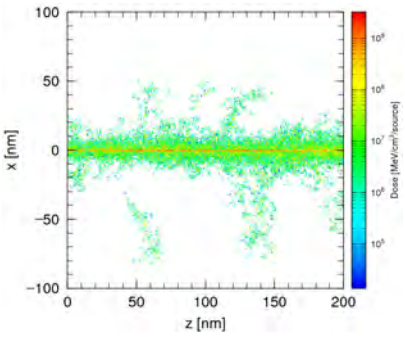

Fig.1 C^{6+} ion track around 1 MeV/u in water.

Development of WASAVIES

We improved WASAVIES (WArning System for AVIation Exposure to Solar energetic particles (SEP)) to be capable of real-time and automatic analyses of radiation dose to an aircraft crew due to a solar flare event [2]. The improved system can determine the model parameters to characterize the profiles of ground level enhancement (GLE) event, when GLE event is detected. The performance of the system was examined by analyses of major GLE events in the 21st century. This study is carried out under collaboration with other institutes and universities and partially supported by JSPS KAKENHI grants (26106006, 15H 05813, 15H05815, 16H01180, and 17K05671).

Internal dosimetry for insoluble cesium-bearing particles

An insoluble cesium-bearing particle with radioactivity can move as a single particle in a human body after intake of these particles. In this case, it is impossible to estimate the number of radioactive cesium disintegrations in the body by an existing method in which the average behavior of countless nuclei in the body is taken into account. Then, a new method was developed to simulate the behavior of the particle in the body stochastically [3]. We derived internal doses based upon the number of disintegrations calculated by the developed method. As a result, uncertainty originating in the insolubility of cesium particles was negligibly small for the exposure level of 1 mGy in lung absorbed dose (Fig.2).

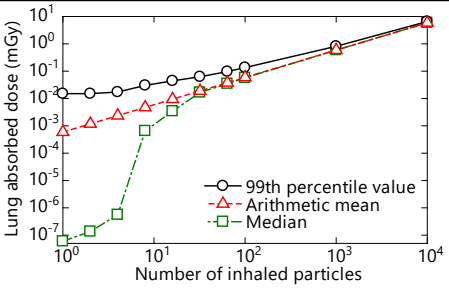

Fig. 2 Variation of lung dose with the number of inhaled particles

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Development Group for Thermal-Hydraulics Technology

For the establishment of thermal-hydraulics numerical simulation method for large-scale two-phase flow and multi-phase flow, we are developing two-phase flow and multi-phase flow numerical simulation codes for large-scale computers, TPFIT and JUPITER (please see the highlight), performing thermal-hydraulics experiments to construct validation databases and developing measurement techniques to obtain detailed thermal-hydraulics data. These developed techniques are applied to the research works for improving the safety of the LWRs and considering the decommissioning process of the Fukushima-Daiichi nuclear power station.

TPFIT

TPFIT (Two-Phase Flow with Interface Tracking) is developed in JAEA for two-phase flow analysis in reactor systems, and can be applicable to computational fluid dynamics simulation including two-phase flow in any systems. TPFIT adopts the advanced interface tracking method, and can be applied to the detail numerical simulation of two-phase flow including movement and deformation of the interface.

As one of examples, the process of bubble coalescence in boiling phenomena for light water reactors was simulated in this method (Fig.1) [1]. This numerical study contributed the establishment of a physical model to predict the critical heat flux.

TPFIT can be also applicable to high-speed compressible two-phase flow, such as two-phase flow in Venturi scrubber, whose maximum velocity is more than 150 m/s. In the snapshot of numerical results, small droplets in high velocity gas flow were simulated directly (Fig.2). Recently, TPFIT has been released at PRODAS[2].

Experiment and measurement

In addition to developing TPFIT and JUPITER, our research group performs various experiments to validate those simulations and construct basic physical models. As one of experiments, we perform the thermal-hydraulics experiment of Venturi scrubber[3]. The Venturi scrubber is one of the components of the filtered venting systems and removes fission products in venting gas under sever accidents. We have performed the visualize experiment of two-phase flow and the numerical simulation with TPFIT to understand the detailed decontamination mechanism (Fig.2). In addition, to elucidation of the removal mechanism of radioactive aerosol by water droplets, the direct observation method of the capturing behavior was developed, and observed the capturing behavior of the particles to consider the removal mechanisms. We proved that an inertial impact of aerosol particles to a droplet occurs as one of physical mechanisms of removal radioactive aerosol by the above Venturi scrubber.

Reference

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2) 2) PRODAS (PROgram and Database retrieval System), http://www.rist.or.jp/nucis/
3) S. Uesawa et al., Proceedings of 12th International Topical Meeting on Nuclear Reactor Thermal-Hydraulics, Operation and Safety (NUTHOS-12), No.964,

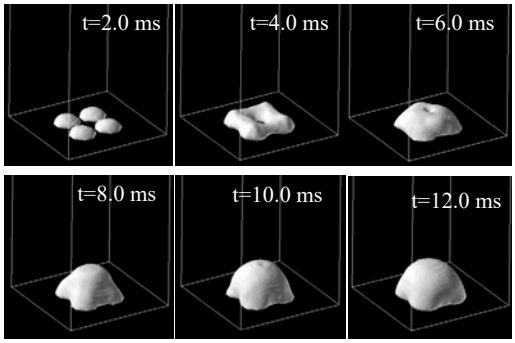

Fig.1 SIMULATION OF BUBBLE COALESCENCE

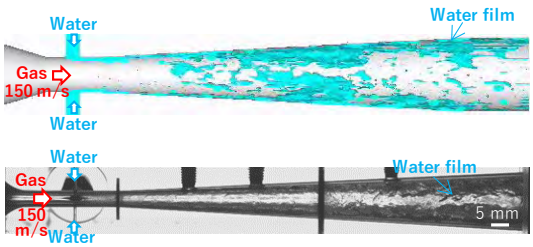

Fig.2 TWO-PHASE FLOW BEHAVIOR IN VENTURI SCRUBBER
(TOP: SIMULATION, BOTTOM: EXPERIMENT)

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Development Group for LWR Advanced technology

We are conducting a fundamental research on LWR advanced technology to provide fundamental knowledge for the improved safety performance of LWR by contributing to improving the scientific rationality for regulation standard, safety margin enlargement for accident management design, and decommissioning and dismantling of Fukushima Dai-ichi Nuclear Power Station (1F). Main research themes are the fission product (FP) behaviors, and fuel/material behaviors under conditions from normal operation to severe accident (SA).

Fission product behavior

We are conducting a fundamental research on FP behavior in a reactor during LWR SA. The objective is to construct a database of FP chemistry named ECUME (Effective Chemistry database of fission products Under Multiphase rEaction)[1]. ECUME consists of dataset for chemical reaction kinetics, elemental model set, and thermodynamic dataset. ECUME is being constructed based on systematic experiments and analyses by using unique experimental apparatus (TeRRa, CREST) and analysis tools (CHASER) (Fig. 1). The TeRRa can reproduce temperature profiles and steam environment in a reactor during LWR SA. The CHASER is based on the 3D-CFD analysis tool, which can make the analysis of FP transportation with fine space resolution possible. These experiments and analyses can provide data on detailed and consecutive FP chemistry during transportation from reactor core to environment in LWR SA.

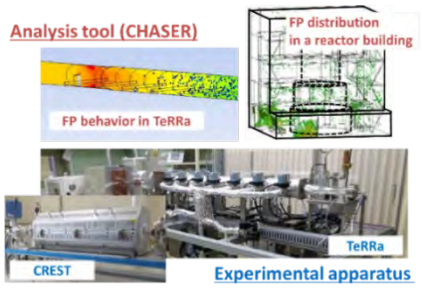

Fig.1 Experimental apparatus and analysis tool for more detailed simulation of FP behaviors

ECUME can be reflected to the improvement of physical/chemical models applied to a SA analysis code. ECUME covers important phenomena revealed after the 1F SA, namely effect of BWR control material boron on cesium (Cs) and iodine chemistry, chemical reaction of Cs with structural materials (Cs chemisorption), and Cs aerosol behavior at reactor building. As an example of practical application, we incorporated the Cs chemisorption model in ECUME to the SA analysis code. It is expected to result in more accurate estimation of Cs amount retained in reactor pressure vessel of 1F.

Accident tolerant fuel (ATF)

For the reduction of the accident risk and the further improvement of safety of the existing LWRs, ATFs with higher reliability are being developed with a focus on enhanced tolerance to high temperature steam under SA. FeCrAl steel strengthened by the dispersion of fine oxide particle for BWR and SiC fiber reinforced SiC matrix composite (SiC) for BWR and PWR were selected as major ATF cladding concepts. SA analyses are performed for PWR with SiC fuel cladding in order to confirm the advantage of SiC fuel cladding compared to current fuel cladding in terms of time to core melt and hydrogen generation[2]. In the SA analyses, small break LOCA sequences are selected because of dominant sequences of PWR. According to the SA analyses using the MELCOR code, the time of the core melt for SiC fuel cladding is delayed a few hours compared to current fuel cladding. The hydrogen generation for SiC fuel cladding is also reduced compared to current fuel cladding as shown in Fig.2. Hence the merit of SiC fuel cladding is shown for small break LOCA sequences.

*This study was funded by Ministry of Economy, Trade and Industry (METI) of Japan.

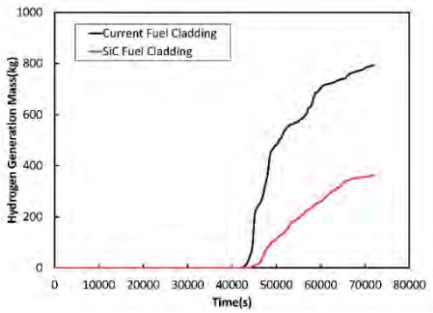

Fig.2 Hydrogen generation mass for current fuel cladding and SiC fuel cladding[2]

Reference

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2) Y. Yamakoshi et al., Proc of TOPFUEL2018, A0076 (2018).

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Research Group for Partitioning

Our research group has been developing partitioning processes by hydrometallurgical method as one of measures for managing high-level radioactive liquid waste (HLLW) generated from reprocessing of spent nuclear fuels.

R & D on SELECT process

Separation performance of partitioning processes depends largely on the extractants used in each process. Many extractants have been developed and/or studied in our research group and some of them extracted target metals effectively. Among these extractants, we have focused on the extractants that comply with CHON principle (extractants consisted of carbon, hydrogen, oxygen, and nitrogen atoms), and have proposed a hydrometallurgical method called SELECT (Solvent Extraction from Liquid-waste using Extractants of CHON-type for Transmutation) aiming at recycle use of nuclear materials and separating actinides for transmutation. We have performed experiments with genuine HLW and radioactive nuclides such as U, Pu, Am, and Cm in a hot cell and/or glove boxes. The obtained results have supported the validity of SELECT process from the viewpoint of the separation performance of metal elements. The extractants used in SELECT process are exposed to high radiation dose, and thus we have been conducting the studies on radiation effect to develop practical separation processes.

Radiation effect on SELECT process

Extraction solvents will receive radiation energy from various kinds of radioactive nuclides during the separation. To evaluate the radiation effects in SELECT process, the radiation energy transfer to the extraction solvent should be evaluated in terms of each radiation type emitted from the radioactive nuclides. Monte Carlo Particle Transport code (PHITS) was employed to calculate the dose absorbed by an extraction solvent containing a diclycolamide which was proposed as an extractant to recover minor actinides (MAs) and rare earths (REs) from HLLW in SELECT process. The dose rate to the extraction solvent during the separation was estimated up to 3.16 kGy/h by the calculation, of which 2.60 kGy/h was due to energy deposition by primary and secondary electrons corresponding to low LET radiations. It is well known that diglycolamide compounds also extracts beta-ray emitters such as Y and Pm, however, the dose rates of 1.07 kGy/h in mixing part and 0.301 kGy/h in settling part were due to the radiation came from the outside of the extraction solvent. The calculation indicated that the influence of beta-ray and gamma-ray transmitted from the nuclides in the feed solution contacted to the extraction solvent could not be disregarded.

N,N,N',N',N'',N''-Hexaoctyl-nitrilotriacetamide (HONTA) is one of the extractants complied with CHON principle. HONTA was proposed as an extractant to separate MAs from REs in SELECT process, and the effect of gamma-ray irradiation on HONTA was studied. Dioctylamine, dioctyl-formamide, and dioctylacetamide were observed as the main degradation products after irradiation of gamma-ray by a ^{60}Co source. From the concentration change of HONTA with respect to the absorbed dose (Fig. 1), the decay constant and the G-value for the degradation of HONTA were obtained as 2.8×10^{-6} Gy^{-1} and 0.37 μmol/J, respectively.

Using the calculated absorbed dose and the obtained G-values, the radiolysis of the extractant in SELECT process could be predicted quantitatively.

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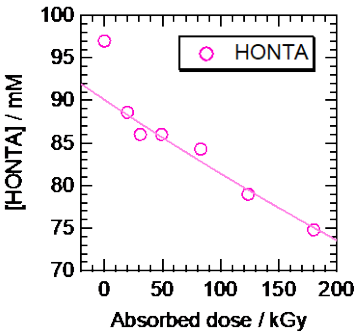

Fig.1 Effects of absorbed dose on concentration of HONTA.

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Research Group for Nuclear Transmutation System

Research Group for Nuclear Transmutation System is developing Accelerator Driven System (ADS) devoted to transmutation of Minor Actinide (MA) into short-lived or stable nuclide. Present R&D level on ADS is at maturing of conceptual design and testing by small-scale equipment for each component and material. To improve ADS design, we are developing analysis system consisting of particle transport, thermal-hydraulics, material and plant behavior. As small-scale R&D, we are conducting validation of nuclear data for particle transport analysis using zero-power reactor.

Validation of nuclear data for ADS

Nuclear data of probability of reactions between Pb and neutron is important for ADS design in which huge amount Pb is used as coolant coupled with Bi. It can be validated by measuring difference of criticality between two core configurations: with and without Pb region. The difference is called lead void reactivity worth. To measure it, a series of integral experiments are conducted as part of a collaboration with the Los Alamos National Laboratory in the United States. In this series experiment, several different experimental systems of fuel composition were examined on the Comet (Fig.1) critical assembly of the National Criticality Experiments Research Center. To provide systematical data for validation, a high-enriched uranium (HEU)/Pb and a low-enriched uranium (LEU)/Pb systems was constructed[1]. The validation in LEU/Pb system was successfully done as shown in Fig.2, while that in HEU/Pb system needs further investigation. HEU/Pb system was re-examined with a new device that can guarantee the gap reproducibility with a higher accuracy and precision. Furthermore, construction of plutonium (Pu)/Pb system has been completed using two kinds of plutonium plates from the Zero Power Physics Reactor (ZPPR). We will valuate Pb cross sections systematically using these data obtained in various systems and confirm availability of present Pb nuclear data for ADS design.

Reference

1) M. Fukushima, et al., “Lead Void Reactivity Worth in Two Critical Assembly Cores with Differing Uranium Enrichments”, Nuclear Sci. Eng. 189, 93-99 (2018).

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Fig.1 Comet

Comet is zero-power reactor consisting of thin disks of fuel (U, Pu) or moderator (Pb, Al, etc.). Disks are replaced to each other and criticality differences, i.e. reactivity were measured.
(National Criticality Experiments Research Center (https://www.nnss.gov/pages/facilities/NCERC.html))

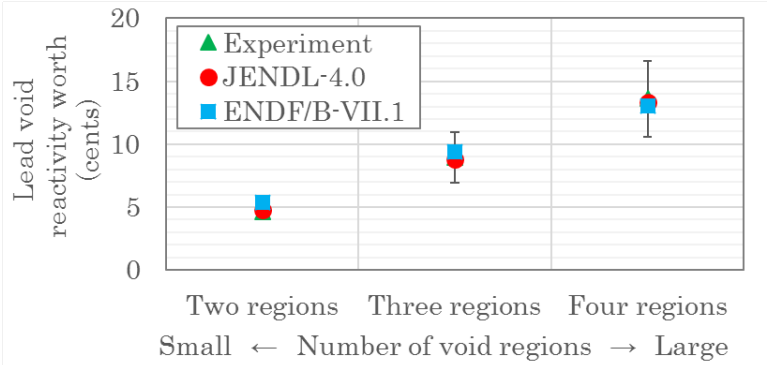

Fig.2 Comparison of experimental and calculated lead-void-reactivity worth in the LEU/Pb systems

The results of this series experiment appear as green triangles（▲）. The analytical results using the evaluated nuclear-data library developed by JAEA (Japanese Evaluated Nuclear Data Library, JENDL-4.0) and the United States (Evaluated Nuclear Data File/B-Versions, ENDF/B-VII.1) are indicated as red circles（●） and blue squares（■）, respectively. In the LEU/Pb systems, the experimental and analytical results overlap, indicating good agreement.

Research Group for MA Transmutation Fuel Cycle

Feasibility study on transmutation system shows that only 20 % of minor actinides (MA) in the fuel can be transmuted in one burning cycle of about 2 years. To increase the transmutation ratio, reprocessing of spent MA transmutation fuels and reuse of MA elements existing in the spent MA transmutation fuels are necessary. Therefore, fuel fabrication, transmutation, and reprocessing should be repeated in the MA transmutation fuel cycle. We have been carrying out research and development of the technology on reprocessing of spent MA transmutation fuel to recover and reuse MAs in the spent fuels. One of our concerns is on pyrochemical treatment technology, in which molten salts and liquid metals are used as solvents; this technique is suitable for treatment of MA transmutation nitride fuels.

Electrorefining process

Detailed design study of the flowsheet for the electrorefining process of spent MA nitride fuel was carried out [1]. The flowsheet contains the recycling process of the molten salt used in the electrorefiner (Fig.1). Multistage counter current reductive extraction using molten salt/Cd phases and zeolite treatment of the molten salt are included in the salt recycling process which aims at removing FP elements from the molten salt bath to keep the purity of the actinides recovered in Cd cathode. The process parameters were determined to meet the target recovery yield of MAs (> **99.9 %**) which is one of the condition enable to transmute **99 %** of MA contained in high level waste (HLW). The other target is the acceptable impurity level of rare earth (RE) fission products in the recovered material (RE/MA < 5 wt.%). Our study shows that it is possible to meet the targets by purifying **2-3 %** from 1000 kg molten salt in the electrorefiner.

Renitridation process

The nitridation-distillation combined method was developed for renitridation process. Actinides (An) recovered in a Cd phase by electrolysis or reductive extraction, are converted to nitride by heating the An-Cd alloys in N_2 gas stream. Technical feasibility had been proved with small scale experiments using TRU elements. Our recent studies contain the scale-up of the apparatus for this process and decreasing the impurities existing in the product. Nitride formation reaction of 2 wt.% Gd-Cd alloys was studied using an apparatus developed for 100 g-Cd scale nitridation tests. Experiments were carried out by heating the alloy sample under vacuum at 693 K to distill Cd, followed by heating in N_2 gas flow at 1073 K for nitridation. The product having small amounts of an intermediate compound $GdCd_2$, was crashed and heated again in vacuum followed by in N_2 gas flow to obtain pure GdN (Fig. 2), in which the content of Cd impurity decreased to 0.07 wt.% [1].

Reference

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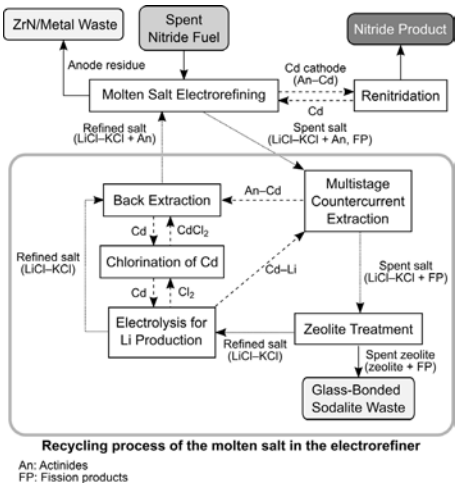

Fig.1 A detailed flowsheet for the electrorefining process.

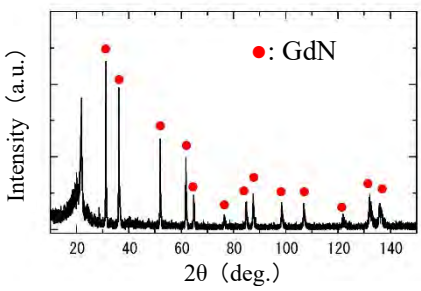

Fig.2 XRD profile of the GdN product.

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Publication list

（added in March 2020）

Nuclear Data and Reactor Engineering Division

Nuclear Data Center

Papers

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Research Group for Reactor Physics and Standard Nuclear Code System

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LWR Key Technology Development Division

Development Group for Thermal-Hydraulics Technology

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Development Group for LWR Advanced Technology

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Partitioning and Transmutation Technology Division

Research Group for Partitioning

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Research Group for MA Transmutation Fuel Cycle

Paper

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Nuclear Science and Engineering Center

Papers

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