FY2020 (2020.4-2021.3) NSEC
Annual Report

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

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Table of Contents

Preface **1**

R&D Highlights **2**

Group Activities **8**

Organization of NSEC **9**

Nuclear and LWR Engineering Division **10**

Nuclear Data Center

Research Group for Reactor Physics and Standard Nuclear Code System

Research Group for Nuclear Sensing

Development Group for Thermal-Hydraulics Technology

Fuels and Materials Engineering Division **14**

Research Group for Corrosion Resistant Materials

Research Group for Radiation Materials Engineering

Research Group for High Temperature Science on Fuel Materials

Development Group for LWR Advanced Technology

Nuclear Chemistry Division **18**

Research Group for Radiochemistry

Research Group for Analytical Chemistry

Environment and Radiation Sciences Division **20**

Research Group for Environmental Science

Research Group for Radiation Transport Analysis

Partitioning and Transmutation Technology Division **22**

Research Group for Partitioning

Research Group for Nuclear Transmutation System

Research Group for MA Transmutation Fuel Cycle

Publication List **25**

Preface

KUGO Teruhiko

Director General, Nuclear Science and Engineering Center

The Nuclear Science and Engineering Center (NSEC) of the Japan Atomic Energy Agency (JAEA) aims to conduct research and development to advance the science and technology that supports the use of nuclear energy and radiation. This annual report provides research highlights and an overview of the research groups' activities in the NSEC for Fiscal Year 2020. We hope this annual report will increase your understanding of the NSEC.

The use of nuclear energy and radiation is supported by the underlying basic science and various technologies that link science and engineering. As such, we conduct fundamental research to elucidate various phenomena involving atomic nuclei, radiation, and radioactive materials using our innovative techniques for measurement and analysis. Based on modeling of the observed phenomena, we develop computer simulation codes and databases for predicting the behavior of energetic particles, heat and fluid in a reactor core, performance of nuclear fuel and reactor structural materials, properties and functions of radioactive materials related to their physical and chemical states, migration behavior of radionuclides in the environment, and the effects of radiation on the human body.

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 (1) the 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.

In 2020, JAEA formulated a vision for the future, JAEA2050+, in order to identify our direction moving forward. To realize JAEA2050+, the NSEC will serve as a leading center for research collaboration and use our research and development capabilities to contribute to advances in science and technology. We seek your understanding, support, and encouragement in our research and development activities.

September 2021

FY2020 NSEC R&D Highlights

The following 5 highlights are selected among various outcomes of the R&D activities accomplished by the NSEC's 5 divisions in FY2020.

- ✓ **Nuclear and LWR Engineering Division:**
  "Deuteron Nuclear Database for Design of Intensive Fast Neutron Sources"

- ✓ **Fuels and Materials Engineering Division:**
  "Experimental Evaluation of Sr and Ba Distribution in MCCI Products Formed under Temperature Gradient"

- ✓ **Nuclear Chemistry Division:**
  "Raman Microscopy Analysis of Uranyl Peroxides: Expectation to Analysis of Fuel Debris"

- ✓ **Environment and Radiation Sciences Division;**
  "Reassessment of organ doses received by A-bomb survivors by precisely reproducing body dimensions of the Japanese in 1945"

- ✓ **Partitioning and Transmutation Technology Division:**
  "Estimating nuclear data using a machine learning technique"

Deuteron Nuclear Database for Design of Intensive Fast Neutron Sources

NAKAYAMA Shinsuke

Nuclear Data Center

In the fields such as experimental nuclear physics, medicine, and fusion reactor development, intensive neutron beams with energies above 10 MeV are required. However, conventional neutron sources using nuclear reactors and electron or proton accelerators cannot satisfy these requirements as illustrated in **Fig.1**. On the other hand, neutron sources using deuteron accelerators efficiently generate neutrons with energies above 10 MeV. In **Fig.1**, neutron energy distribution obtained from a lithium target bombarded by a 40-MeV deuteron is presented.

For the design studies of such facilities, it is necessary to accurately predict the amount and energy distribution of neutrons generated from the nuclear reactions induced by deuterons. However, conventional calculation method does not satisfy it. This is because the quantum mechanical wave properties of deuterons are not considered appropriately.

In this work[1], we developed a new calculation method by integrating the several theoretical models taking quantum mechanical effects into account. Furthermore, the calculated results were compiled into a database applicable to the simulation code employed in the design studies of neutron sources. The database was named as JENDL/DEU-2020, and it stores the deuteron nuclear data on lithium, beryllium and carbon isotopes that are expected to be used as deuteron-beam targets in deuteron-accelerator neutron sources.

We present an example of the validation results of the simulation using JENDL/DEU-2020 in **Fig.2**. The figure shows the calculated and experimental neutron yields from a 7.5-mm thick lithium target bombarded by a 25-MeV deuteron. The red solid line and the blue dashed line are the simulation results with the PHITS code, but the red solid line represents the result where the data of JENDL/DEU-2020 are used as the deuteron-induced reaction cross-sections on Li-6,7. On the other hand, the blue dashed line is the result where the theoretical models implemented in PHITS is adopted for the calculation of the above mentioned cross-sections. As shown in **Fig.2**, the result using JENDL/DEU-2020 reproduce the experimental data better than that using the models in PHITS. Moreover, we have confirmed that the prediction accuracy of the simulation using JENDL/DEU-2020 for beryllium and carbon targets is as good as that for lithium target.

By using JENDL/DEU-2020, the reliability of the simulation is greatly improved and consequently the designs of various neutron sources suitable for their purposes will be easier. JENDL/DEU-2020 is expected to promote the use of intensive fast neutrons in various fields such as basic science, medicine, and so on. JENDL/DEU-2020 is available from the website of Nuclear Data Center[2].

This work was partially supported by JSPS KAKENHI Grant Number 19K15483.

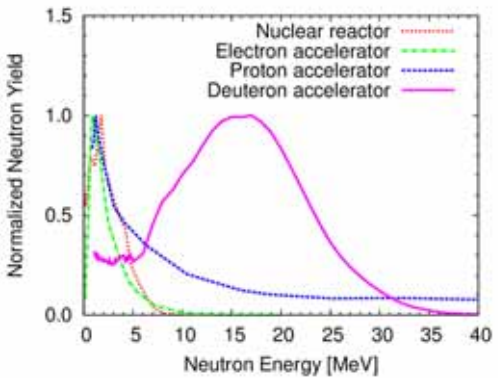

Fig.1 Neutron energy distributions obtained from typical neutron sources. Each distribution is normalized to have a maximum value of 1.0, and is presented in the range above 0.1 MeV.

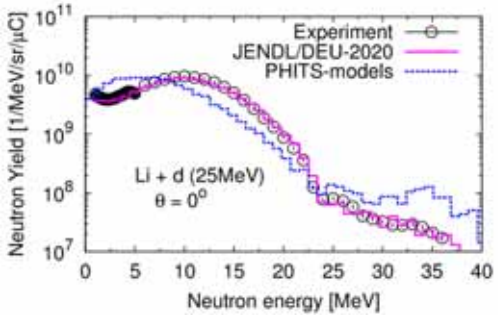

Fig.2 Neutron yields from a lithium target bombarded by a 25-MeV deuteron.

Reference

1) S. Nakayama, *et al.*, *J. Nucl. Sci. Technol.*, 58, 805-821 (2021).

2) https://wwwndc.jaea.go.jp/ftpnd/jendl/jendl-deu-2020.html (accessed 2021-09-07).

Experimental Evaluation of Sr and Ba Distribution in MCCI Products Formed under Temperature Gradient

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[b] Collaborative Laboratories for Advanced Decommissioning Science (CLADS)

In the severe accident at Fukushima Daiichi Nuclear Power Station (1F), the molten core materials were predicted to relocate from the reactor pressure vessel and spread over the concrete floor, resulting in molten core-concrete interaction (MCCI) at high temperatures. The MCCI products still continue to be exposed to water, and consequently the water-soluble radionuclides may be gradually dissolved from the MCCI products. The dissolution behavior is crucial for evaluating secondary source terms within 1F. In this study, we focused on Sr and Ba as the representative fission products. To obtain information on the distribution of Sr and Ba in MCCI products, the reaction test between simulated corium (containing non-radioactive Sr and Ba) and concrete was performed[1)].

Fig.1 (a) shows the schematic illustration of the sample before heating. The powder mixture of $U_{0.5}Zr_{0.5}O_2$, ZrH_2, stainless steel (SS), B_4C, Mo-Ru-Rh-Pd, $BaCO_3$, and $SrCO_3$ as a simulated corium was pressed into a tablet and placed on a piece of basaltic concrete. The light from a high-power Xe lamp was directed onto the tablet surface by a light-concentrating heating device to form a simulated MCCI product under vertical temperature gradient.

The cross-sectional image of the solidified sample is shown in **Fig.1** (b). The corium and concrete were interacted as liquid phases and solidified in the upper region of sample (once melted zone). Below the once melted zone, concrete was thermally dehydrated. **Fig.1** (c) and (d) show SEM images of the upper region and bottom region of once melted zone, respectively. Main phases are dark silicate glass containing 3 at%-Sr and 2 at%-Ba, and bright $(U,Zr)O_2$ particles in which Sr and Ba are not detected. $(U,Zr)O_2$ precipitated as fine particles (2–3 μm) in the upper region, while it precipitated as bigger particles (10–20 μm) in the bottom region. Additionally, content of $(U,Zr)O_2$ are much higher in the bottom region. During the heating tests, sedimentation of $(U,Zr)O_2$ particles may have occurred in the liquid silicate glass because of its higher density.

An image of the layered structure of MCCI product is drawn in **Fig.2**. Sr and Ba are likely to be concentrated in the upper silicate glass-rich zone. In the actual situation, the crust layer might form on the top surface with cracks and cavities. The water is possible to contact the silicate glass-rich zone, which may result in dissolution of Sr and Ba into water. Leaching behavior of Sr and Ba from the silicate glass will be the next issue to be investigated.

Reference

1) A. Sudo, *et.al*., *J. Nucl. Sci. Technol.*, 58, 473-481 (2021).

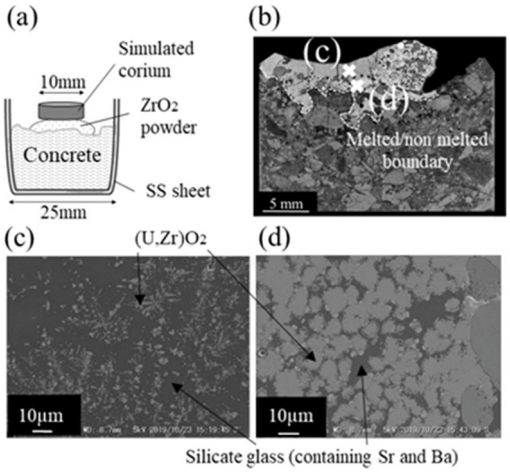

Fig.1 (a) Schematic illustration of the sample setup. (b) Optical micrograph of the sample cross section. (c) SEM image of upper region of once melted zone. (d) SEM image of bottom region of once melted zone.

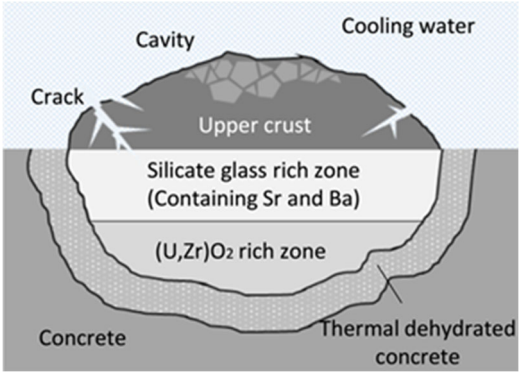

Fig.2 Image of the layered structure of the MCCI product.

Raman Microscopy Analysis of Uranyl Peroxides: Expectation to Analysis of Fuel Debris

KUSAKA Ryoji

Research Group for Radiochemistry

Fuel debris at the Fukushima-Daiichi nuclear power plants will be a complex solid mixture generated after melting and cooling of nuclear fuel and structural materials. Knowledge of the chemical state such as what kind of chemical substance the fuel debris is composed of will be one of important information that may influence the decommissioning process such as removal from the reactor, storage, processing and disposal of fuel debris. However, the method of clarifying the chemical state of fuel debris has not been determined.

Therefore, in this research[1], we focused on the use of Raman microscopy, which has many achievements in chemical analysis in many fields such as biology. Measurements by Raman microscopy do not require sample pretreatment and can be performed on very small samples, which will make it safer and easier to measure fuel debris with high radioactivity.

Since fuel debris is in contact with water in the reactor, it is expected to react with hydrogen peroxide generated by radiolysis of water and to be in a chemical state different from that at the time of the accident. In this study, in order to enable to analyze fuel debris based on the information of the chemical change, U_3O_8, which is one of basic uranium oxides, was reacted with hydrogen peroxide and the reacted U_3O_8 was analyzed with Raman microscopy.

As a result of Raman measurements, it was clarified that two types of uranyl peroxides, studtite and metastudtite, were formed on the surface of U_3O_8 by the reaction with hydrogen peroxide (**Fig.1**). Furthermore, the distribution of these products on the sample surface suggested that studtite was predominantly produced over metastudtite (**Fig.2**).

Uranyl peroxide is known to be produced by reaction with spent nuclear fuel and UO_2. Recently, it has been found that uranyl peroxide is also produced by reaction with simulated debris of uranium-stainless-zirconium system, which will be close to actual debris. Therefore, uranyl peroxide could also be produced in actual fuel debris, and the present study shows that Raman microscopy will contribute to the analysis of actual fuel debris.

This work was supported by JAEA Nuclear Energy S&T and Human Resource Development Project through concentrating wisdom, Grant Number JPJA18P18071886.

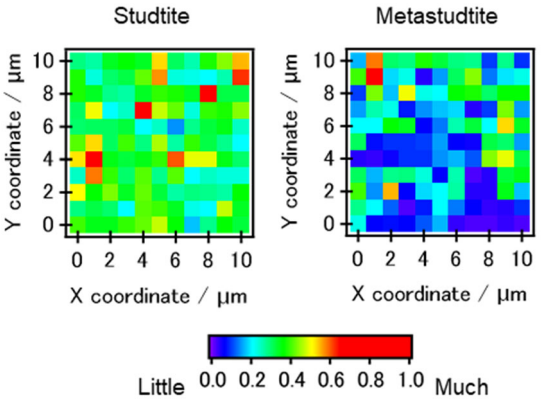

Fig.1 Raman spectra of U_3O_8 immersed in hydrogen peroxide aqueous solution.
Two types of uranyl peroxides (Studtite and Metastudtite) were observed.

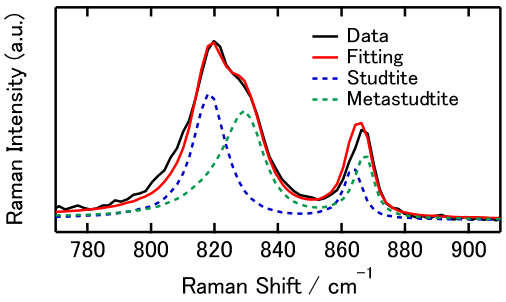

Fig.2 Distribution of studtite and metastudtite generated of the surface of U_3O_8.
These images indicate that studtite was predominantly produced over metastudtite.

Reference

1) R. Kusaka, *et al.*, *J. Nucl. Sci. and Technol.*, 58, 629-634 (2021).

Reassessment of organ doses received by A-bomb survivors by precisely reproducing body dimensions of the Japanese in 1945

Tatsuhiko SATO[1], Sachiyo FUNAMOTO[2], Colin PAULBECK[3], Keith GRIFFIN[4], Choonsik LEE[4], Harry CULLINGS[2], Stephen D. EGBERT[5], Akira ENDO[6], Nolan HERTEL[7], Wesley E. BOLCH[3]

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[2] Radiation Effects Research Foundation, Japan
[3] University of Florida, USA
[4] National Cancer Institute, USA
[5] No affiliation
[6] Nuclear Science and Engineering Center
[7] Georgia Institute of Technology, USA

Epidemiological studies of A-bomb survivors conducted by the Radiation Effects Research Foundation (RERF) are some of the most important resources for data on the health effects of radiation. Precise estimation of the organ doses is the key issue in the studies, and the dosimetry system for estimating doses received by individual organs for each survivor has devolved and improved over the years. For further refinements to the dosimetry system, a Japan-U.S. joint project team was formed, comprising Nuclear Science and Engineering Center of JAEA, RERF, the University of Florida, and the U.S. National Cancer Institute. Members of this project team developed a new set of models of the human body (called “phantoms”) for adults, children, and pregnant women by rigorously reproducing the standard body dimensions of Japanese people in 1945, which are planned to replace those in the current dosimetry system developed in the 1980s using primitive geometrical shapes. Examples of the newly developed phantoms are shown in **Fig.1**. The project team also developed a method for more accurately estimating the organ doses of A-bomb survivors with these phantoms by utilizing the latest computational techniques such as PHITS, which was developed mainly by JAEA. Based on idealized conditions in hypothetical survivors, our initial comparisons of doses using the new phantoms and methods with doses derived from the current dosimetry system found that doses were generally consistent, although for some organs dose estimates could change by approximately ±15 % at the maximum[1)]. Further studies are necessary for implementing the new phantom series in the dosimetry system.

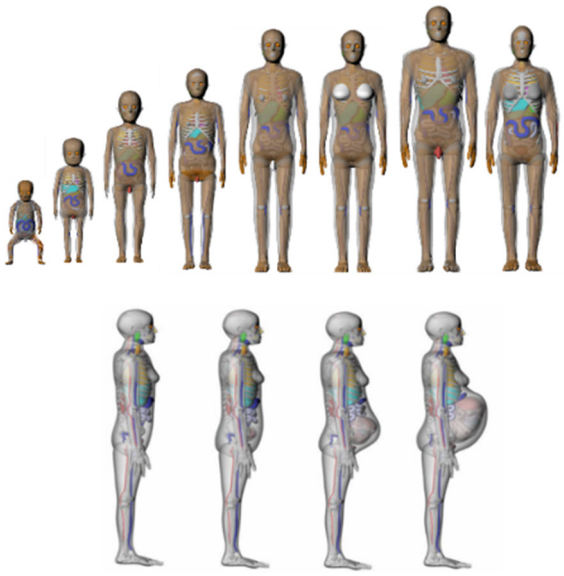


Fig.1 Adults, children, and pregnant women phantoms developed in this study.

Reference

1) T. Sato, *et.al.*, *Radiat. Res.* 194, 390-402 (2020).

Estimating nuclear data using a machine learning technique

IWAMOTO Hiroki

Research Group for Nuclear Transmutation System

Nuclear data are indispensable for the simulation of particle transport in estimating radiation doses, radioactive isotope production, and safety parameters of nuclear systems. The quality and quantity of nuclear data in the evaluated nuclear data libraries have been steadily enhanced owing to physicists' strenuous efforts; however, discrepancies between experimental data and the theoretical model calculations remain particularly at high incident energies. While it is important to improve the theoretical models, conventional approaches require substantial amount of time and effort. In such cases, a machine learning (ML) technique may serve a useful role in estimating nuclear data. In this study[1], we proposed a method to estimate nuclear data from experimental data using ML based on Gaussian processes[2].

Fig.1 shows the proton-induced sodium-22 (^{22}Na) production cross-section for silicon (^{nat}Si) nucleus which is a key material of the semiconductor devices. It is difficult to account for the experimental data even with the latest theoretical models. On the other hand, since the proposed method estimates the cross-section while learning from the experimental data, significant discrepancy between the estimated and experimental values does not occur in principle.

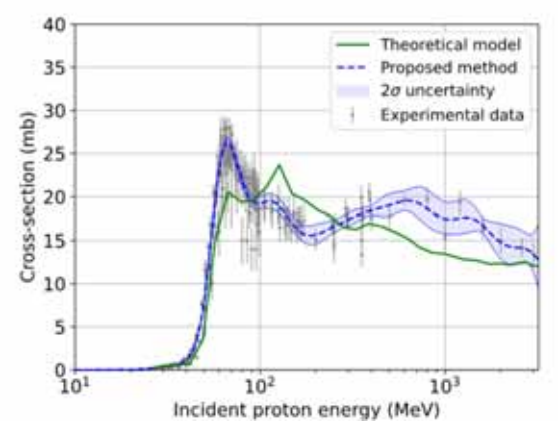


Fig.1 Comparison of the estimated ^{nat}Si(p,X)^{22}Na cross-section between the theoretical model and the proposed ML method.

The light blue band indicates 2σ uncertainty about the estimated cross-section.

As illustrated in **Fig.2**, the proposed method provides uncertainty information of the estimated cross-sections, which takes a lot of effort to obtain in the conventional method. The obtained uncertainty information allows us to discuss effectiveness of experiments from the viewpoint of improving the accuracy of nuclear data, and is expected to is applicable to the quantification of the uncertainty due to nuclear data in research fields using nuclear data such as shielding analysis and reactor physics.

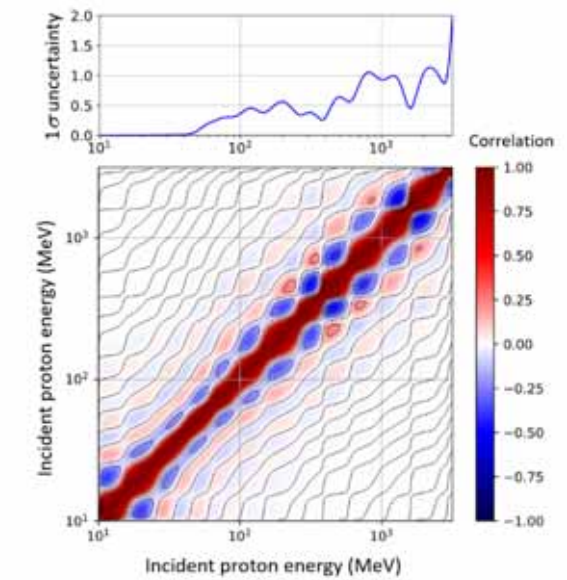


Fig.2 Uncertainty information of the ^{nat}Si(p,X)^{22}Na cross-section estimated by the proposed ML method.

Top panel shows one standard deviation, which gives the uncertainty of the evaluated cross-section at incident energy; bottom panel: correlation matrix, which allows us to understand the strength of the linear relationship between the cross sections between energy points.

Reference

1) H. Iwamoto, *J. Nucl. Sci. Technol.*, 57, 932-938, (2020).

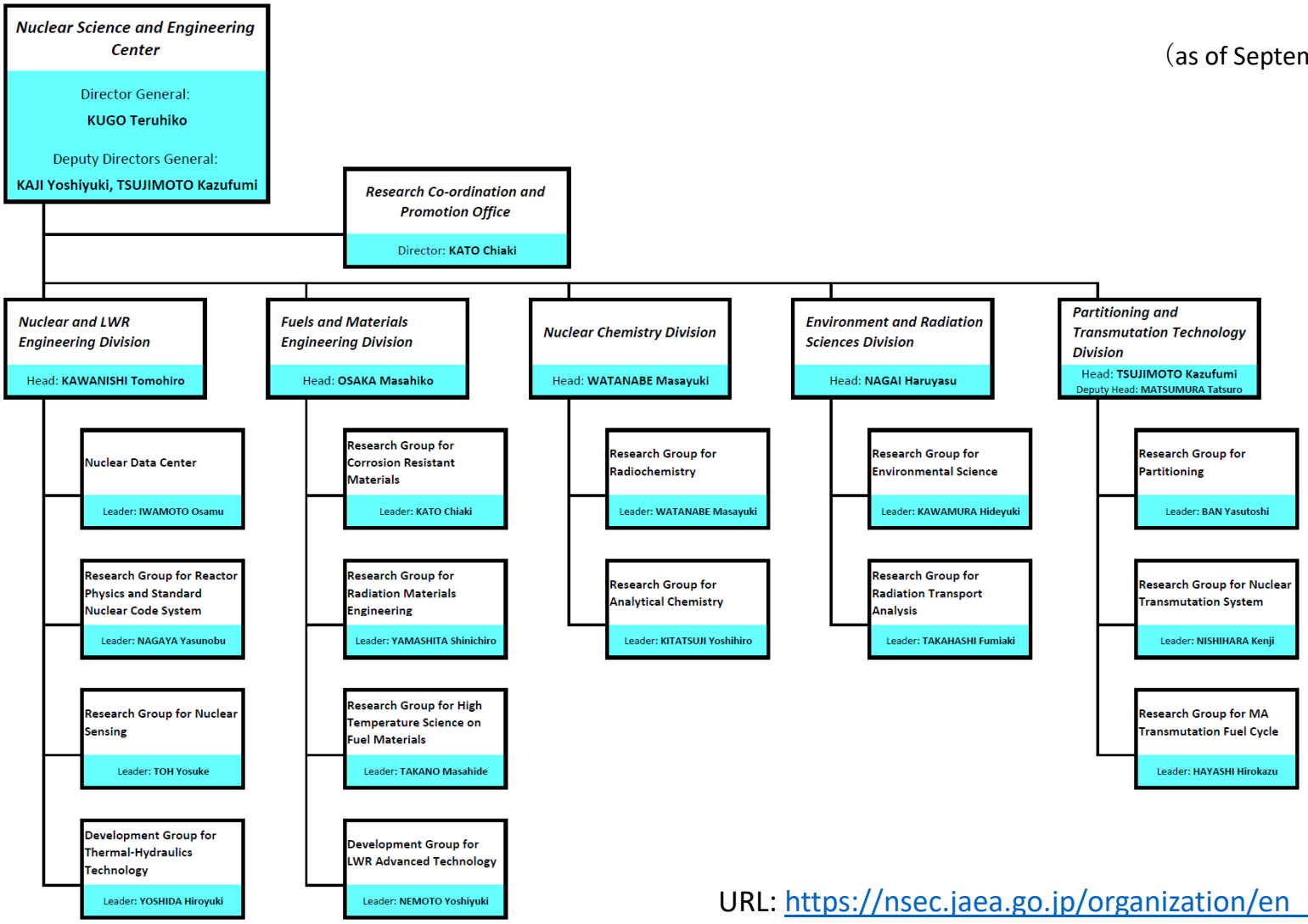
2) C.E. Rasmussen and C.K.I. Williams, *Gaussian Processes for Machine Learning*, the MIT Press (2005).

FY2020 NSEC Group Activities

The NSEC of JAEA consists of 15 Groups.

Organization of NSEC

（as of September 2021）



URL: https://nsec.jaea.go.jp/organization/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 that, 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. They are available from our website (https://wwwndc.jaea.go.jp).

Thermal Neutron Capture Cross Section Measurement

Thermal neutron capture cross sections are important for thermal reactors which are dominated in currently operating nuclear power plants. Our group has been targeting cross section measurements on minor actinides (MA) and fission products that are main components of nuclear wastes. The thermal cross sections were measured by activation method with the research reactor of Kyoto University (KUR).

For Cs-135, which is one of major long lived fission products, mass spectrometry was utilized to accurately quantify isotopes in Cs samples in combination with decay gamma-ray measurement from decay of contaminated Cs-137 in the sample. Resonance integral of neutron capture cross section, which indicates capture strength in resonance region, were also deduced. The obtained result revealed the current evaluated value of JENDL-4.0 was possibly overestimated by 30%.[1)]

The capture cross section of Am-243 affects the amounts of Cm isotopes production in reactors, which are major heat sources in spent fuels. In this work, the capture cross sections were measured for components of ground and isomeric state production. The present results agree with the recent measurement at ANNRI of J-PARC by time-of-flight method which shows possible underestimation of the current evaluation of JENDL by 10%.[2)]*

Neutron Filter System with ANNRI**

The accelerator driven system (ADS) of MA burner requires to improve accuracy of MA cross sections for fast neutrons. ANNRI has a large potential for the MA cross section measurement because of the large neutron intensity. However, double bunch structure of the neutron beam prevented the accurate cross section measurement for fast neutrons. To overcome this, we introduced the neutron filter system that produce quasi mono-energetic neutron beams. It was confirmed to work well via Au-197 measurement[3)], resulting in good agreement with the evaluated values which is accurately known.

Development of JENDL-5

Development of the next version of evaluated nuclear data library JENDL-5 is in progress. JENDL-5 aims to meet various needs from nuclear energy application including not only nuclear reactor development but also nuclear backend such as decommissioning and wastes management. Large amount of neutron induced reaction data from light to heavy nuclides have been revised and newly evaluated. The test libraries were created, and benchmarking for reactor and shielding has been done to validate and improve the evaluated data. JENDL-5 will include not only neutron data but also charged particle ones to meet the needs of radiation applications.[4)]

* This work includes the result of “Research and development for accuracy improvement of neutron nuclear data on minor actinides” entrusted by MEXT.

** This work is supported by the Innovative Nuclear Research and Development Program from MEXT, Grant Number: JPMXD0217942969.

Reference

1) S. Nakamura, *et al.*, *J. Nucl. Sci. Technol.*, 57, 388-400 (2020).
2) S. Nakamura, *et al.*, *J. Nucl. Sci. Technol.*, 58, 259-277 (2021).
3) G. Rovira, *et al.*, *Nucl. Instr. Meth. A*, 1003, 165318 (2021).
4) O. Iwamoto, *et al.*, *EPJ Web Conf.*, 239, 09002 (2020).

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Research Group for Reactor Physics and Standard Nuclear Code System

The role of nuclear codes is increasing with rapid progress in computer technology. It enables to evaluate in detail the energy and spatial distribution of neutrons and photons emitted after fission reactions in a nuclear reactor. Important properties such as criticality 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 nuclear energy technology. The main codes are MVP and FRENDY. We are also collaborating tightly with the JENDL project and preparing many cross section libraries. We produced a new multigroup library for ORIGEN in SCALE6.2 from JENDL/AD-2017 and developed a handy criticality analysis tool for fuel debris systems in JFY2020.

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 and available from RIST and OECD/NEA Data Bank as NEA-1673 MVP/GMVP V.3. The cross section libraries for MVP3 can be downloaded from our group's home page.[2]

FRENDY

FRENDY is a nuclear data processing code.[3] It enables processing of evaluated nuclear data such as JENDL. The first version of FRENDY treats the ENDF-6 format and generates the ACE files which are used for Monte Carlo codes such as PHITS and MCNP. FRENDY version 1 was released in 2019 as an open-source code from our group's home page.[2] Current developments include the estimation of statistical uncertainty in probability table generation for unresolved resonance cross sections,[4] and the generation of multigroup cross sections.[5] These capabilities are to be implemented in version 2.

New ORIGEN library

The SCALE6.2 code system[6] was released in 2016 (the latest version is SCALE6.2.4). The ORIGEN code[6] in SCALE6.2 used for activation calculations in decommissioning is completely different from the previous ORIGEN-S code.[7] JENDL Activation Cross Section File for Nuclear Decommissioning 2017 (JENDL/AD-2017)[8] was released in 2018. Thus we have produced a new multigroup library (200 group, 300 K) for ORIGEN in SCALE6.2 from JENDL/AD-2017 with the AMPX-6 code[9] in order to popularize JENDL/AD-2017 widely. JPDR activation analyses with ORIGEN and the new library demonstrate that the new library has a performance equivalent to the ORIGEN library in SCALE6.2.

HAND

HAND[10] is a handy criticality analysis tool for fuel debris systems generated from the severe accident at the Fukushima Daiichi Nuclear Power Station. HAND is designed for the use in planning fuel debris retrieval. HAND enables users to perform quick criticality analysis without expert knowledge of reactor physics. The input form of HAND is very simple and users can intuitively understand the calculation results. This tool is expected to be the effective tool to estimate the criticality of fuel debris systems. HAND is an open-source code and can be downloaded from our group's home page.[2]

Reference

1) Y. Nagaya, *et al.*, *JAEA-Data/Code*, 2016-018 (2017).
2) https://rpg.jaea.go.jp/main/en/program/ (accessed 2021-09-06).
3) K. Tada, *et al.*, *JAEA-Data/Code*, 2018-014 (2019).
4) K. Tada and T. Endo, *2021 Annual Meeting of AESJ*, 1B01 (2021). (*in Japanese*)
5) A. Yamamoto, *et al.*, *2020 Fall Meeting of AESJ*, 2H03 (2020). (*in Japanese*)
6) W.A. Wieselquist, *et al.*, ORNL/TM-2005/39 Version 6.2.4 (2020).
7) I.C. Gauld, *et al.*, ORNL/TM-2005/39 (2006).
8) https://wwwndc.jaea.go.jp/ftpnd/jendl/jendl-ad-2017.html (accessed 2021-09-06).
9) D. Wiarda, *et al.*, ORNL/TM-2016/43 (2016).
10) K. Tada, *JAEA-Data/Code*, 2020-014 (2020). (*in Japanese*)

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Research Group for Nuclear Sensing

Non-destructive analysis (NDA) is one of the most essential and fundamental methods in many fields, such as nuclear material accountancy, nuclear safeguards and security, decommissioning and many other scientific fields. Despite being a mature method, further improvements in NDA are required and enable us to meet the distinctive demands of researchers and engineers. We have therefore developed an advanced NDA system for highly radioactive nuclear materials, and an analytical method of At-211 for radiotheranostics.

Developments of Prompt Gamma-ray Analysis Apparatus in Active-N

No established non-destructive method exists to measure the amount of highly radioactive nuclear fuel materials such as spent fuels, and it is one of the urgent issues in nuclear material accountancy. Therefore, we have started a research on development of innovative non-destructive analysis (NDA) system, named "Active-N", which provides the following active-neutron NDA methods: Differential Die-away Analysis, Neutron Resonance Transmission analysis, and Prompt Gamma-ray Analysis (PGA). To reduce neutrons that induce radiation damage to the germanium detector for PGA, its extra shielding was developed. Through a series of Monte Carlo simulations, the ability of several common materials in reducing the neutrons was investigated, and the reduction of fast neutrons and suppression of 2.22-MeV gamma rays originated from hydrogen were optimized. The results of the simulations show that by using 1:1 mixture of HDPE and LiF with natural isotopic composition, the same levels of fast neutron reduction and 2.22-MeV gamma suppression can be achieved with a shielding in which ^{6}LiF plates of 1cm-thickness are inserted every 3 cm. It is found that by adding the extra shielding, the fast neutron fluence at the germanium crystal position can be reduced approximately an order of magnitude, from 1.9×10^{-5} to 2.7×10^{-6} /cm2/source, which is low enough to avoid the severe damage of PGA detector. [1)]

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

Developments of an Analytical Method of At-211

The α-emitting radio halogen At-211 has drawn great interest because of its potential use in the treatment of cancers. After At-211, produced by a nuclear reaction, is separated from the irradiated targets in the chemical procedures, basic researches related to the chemical properties of At-211 and radiolabeling reactions for pharmaceuticals have been performed for various purposes. At present, the radioactivity and chemical forms of At-211 have been determined separately. The former has been determined by γ-ray spectroscopy using semiconductor detectors. The latter has been measured by imaging plates (IPs) visualizing thin-layer chromatography. However, IPs need typically more than 10 h for the visualization. In addition, these are sensitive to the x-rays emitted from radioactive nuclides, such as Po-207 biproducts of Rn-211, as well as At-211, so that not all the chemical forms of At-211 are analyzed with the visualization of IPs. To prevent severe loss of At-211 due to its short half-life of 7.2 h, and to obtain accurate analytical results, a rapid analytical method for both the radioactivity and all chemical forms of At-211 was required. We propose a new method by means of an α-scintillation-camera system comprising of thin-layer chromatography and a high-sensitive CCD camera. The performance of the system was experimentally verified: low-radioactivity At-211 of 56-672 Bq was measured in a short time of 1,000 sec and all chemical forms of At-211 were able to be evaluated. The results reveal that the method enables us to analyze two features of At-211 within a significantly short time.[2)]

Reference

1) K. Furutaka, *et al.*, *Proc. Joint Inter. Conf. Monte Carlo 2020,* 297-394 (2020).
2) M. Segawa, *et al.*, *J. Radioanal. Nucl. Chem.*, 326, 773-778, (2020).

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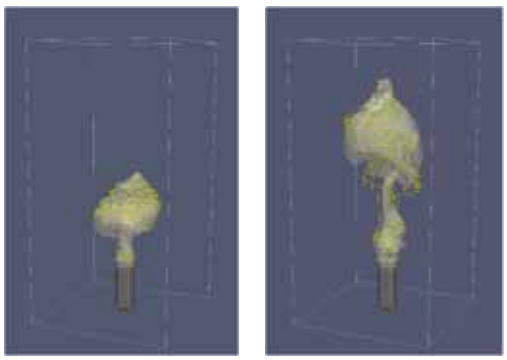
Development Group for Thermal-Hydraulics Technology

For the establishment of thermal-hydraulics numerical simulation methods for multi-phase flow, we are developing multi-phase CFD numerical simulation codes, TPFIT and JUPITER, measurement techniques to obtain thermal-hydraulics data and performing thermal-hydraulics experiments to construct validation databases. These advanced 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.

Multi-phase CFD Codes

TPFIT is developed for two-phase flow in the nuclear reactor. TPFIT uses compressible fluid equations and can be applied to the numerical simulation of two-phase flow in any system.
JUPITER can simulate melting and relocation behavior without assumptions or simplification. Besides, JUPITER can perform the massively parallel simulation of multi-phase flow with more than two hundred thousand cores, by adapting high-efficient parallel computing techniques.
As one of the examples of CFD codes, we performed a numerical simulation of air-water two-phase flow with microparticles in a pool (Figure 1)[1]. In this simulation, microparticles' motion in two-phase flow was simulated by TPFIT with the Lagrangian particle tracking method. In Fig.1, we visualized interfaces as white surfaces, and yellow spheres show microparticles. A two-phase flow in the water pool is obtained, and we can simulate two-phase flow with microparticles by TPFIT.
TPFIT and JUPITER were released at PRODAS[2]. We are continuously improving TPFIT and JUPITER to enhance the safety of the LWRs.



(a) t=0.03[s] (b) t=0.06[s]

Fig.1 Numerical simulation of two-phase flow with microparticles in water pool.

Experimental Studies

We perform various experiments to validate those simulations and construct basic physical models. As one of the experiments, we performed optical measurement of aerosol particles (microparticles) capturing behavior near the gas-liquid interface (Figure 2)[3]. Microparticles' capturing at interface is utilized in water filtering industrially. The capturing is also used in filtered venting systems and pool scrubbing in the nuclear reactors. By this experiment, we could visualize aerosol particle behavior. Based on experimental results, we constructed a physical model to express microparticle behavior vicinity and on the interface. We introduced the constructed model to TPFIT to improve numerical simulation accuracy.

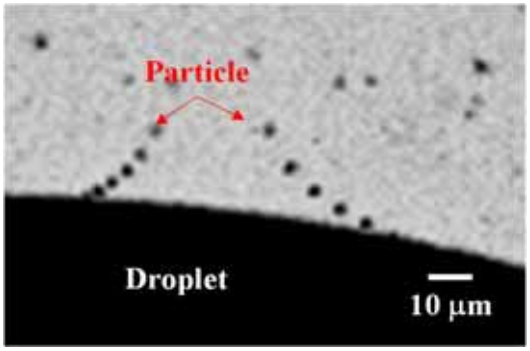


Fig.2 Continuous images of the particle moving in the vicinity along the gas-liquid interface.

Reference

1) H. Yoshida and S. Uesawa, *Proceedings of ASME's Nuclear Engineering Conference powered by ICONE (ICONE2020)*, ICONE2020-16393 (2020).
2) PRODAS (PROgram and DAtabase retrieval System), https://prodas.jaea.go.jp/ (accessed 2021-09-06).
3) S. Uesawa and H. Yoshida, *Mech. Eng. J.*, 7, 19-00539 (2020).

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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 localized corrosion mechanisms in stainless steels[1)]

Stainless steel (SS) is popular material frequently used in nuclear facilities, but the localized corrosion often occurs due to chloride ions (Cl^-). Cu alloyed with SS is known to be both beneficial and detrimental to localized corrosion. To understand the role of Cu in localized corrosion, the developed extra-high-purity (EHP) SS, which is Cu-free, were used and anodic polarization measurements were conducted in two solutions, one with Cu^{2+} and one without Cu^{2+}, to investigate the effects of Cu^{2+} in the bulk solution on the localized corrosion resistance. (Fig. 1) At first, the specimens were polarized to 0.4 V in 0.1 M NaCl-1mM $CuCl_2$ (condition a1 and b1) to deposit metal Cu and Cu compound onto the surface. Subsequently, the solution was replaced to 0.1 M NaCl (a2) or 0.1 M NaCl-1mM $CuCl_2$ (b2). Shown as rapid increase in current density at ca. 0.8 V, pitting occurred in b2, but no pitting occurred in a2. In conditions of a1 and b1, deposition of Cu compound on the surface was confirmed. These results suggest that both deposited Cu on the surface and Cu^{2+} in the bulk solution were necessary for the pitting to occur.
Therefore, the decrease in pitting corrosion resistance of 316 EHP SS in 0.1 M NaCl with 1 mM $CuCl_2$ is mainly due to precipitated Cu compounds and continuous Cu^{2+} supply to the surface.

Researches on corrosion mechanisms under air-water interfacial environment[2)]

After the accident of Fukushima Daiichi Nuclear Power Station (1F), seawater had been injected and purified cooling water is circulated into the primary containment vessel (PCV) in the present. To understand the corrosion mechanism in the air/solution alternating condition that is the air/solution interfacial environment, the corrosion rate of carbon steel under air/solution alternating conditions was investigated in artificial seawater components. It is clear that the corrosion rate of carbon steel in the alternating condition accelerates with increasing concentration in the concentration region between deionized water to 200 times diluted artificial seawater (ASW), and the corrosion rate decreases with increasing concentration in the concentration region between 20 times diluted ASW to undiluted ASW. (Fig. 2) It can be considered that the reason why the carbon steel corrosion was suppressed in highly ASW would be caused by Mg ions and Ca ions in the artificial seawater precipitate and cover on the surface due to the increase in pH near the surface by oxygen reduction reaction.

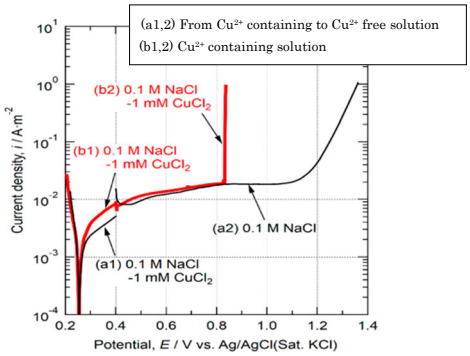


Figure 1 Anodic polarization curves of EHP SS.

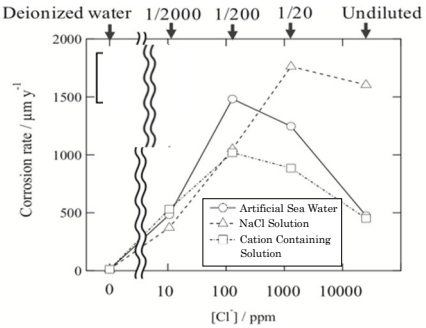
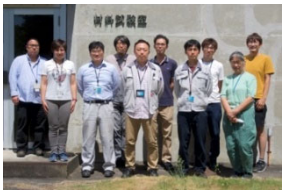


Figure 2 Corrosion rate of carbon steel in various test solutions with different concentration.

Reference

1) T. Aoyama, *et.al.*, *Metals*, 11, 511, (2021).
2) K. Otani, *et.al.*, *Zairyo-to-Kankyo*, 69, 246–252, (2020).



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Research Group for Radiation Materials Engineering

In our group, a wide variety of nuclear materials, including the materials which are under the consideration of future potential use, have been studied for the purpose of deepening the understanding on their radiation tolerance. A tandem accelerator is one of the most familiar experimental tools for material irradiation study of our group. Especially, as for ceramic materials, systematic irradiation studies have been done, and a review paper of the studies[1] has been published recently. In order to explore a new advanced material not only for nuclear use but also non-nuclear one, we are also making the most use of a shared supercomputer system which was renewed recently.

Systematic understanding of characteristic damage structures in ceramics irradiated with SHI

Irradiation with high energy is known to create various radiation damages in a target material. Due to continuous effort over a lengthy period of time, we have gradually learned more about characteristic damage structures, such as ion tracks and hillocks, in a variety of ceramics irradiated with swift heavy ions (SHI). In our recent work, we have summarized the formation processes of ion tracks and hillocks in various ceramics irradiated with SHI. Figure 1 presents one of typical high-resolution images of a transmission electron microscope (TEM), and formation processes of ion tracks and hillocks in non-amorphizable and amorphizable ceramics with a 200 MeV Au ion, giving us the information such that the difference in shape of hillocks possibly comes from the difference in volume of protrusion and that the material dependence of nanostructure formation can be ascribed to the intricate recrystallization process.

Computational materials science based on atomistic simulation

High entropy alloy (HEA) has a lot of fascinating features as a structural material not only for nuclear industry but also non-nuclear one. Many types of HEAs are proposed, and experimental and theoretical studies on these alloys are now aggressively proceeding all over the world. In our group, we have recently started experimental and theoretical works in a collaboration program with some of the domestic universities. Figure 2, for example, shows atomic structure models of CoCrFeNiMn alloy (one of the most representative HEAs) which was created by using special quasi-random structures (SQSs). SQSs are an important tool in modeling disordered alloys with atomic resolution. By using these atomic structure models, it is expected that local lattice distortion, short range ordering, and a stacking fault energy of this HEA will be evaluated.

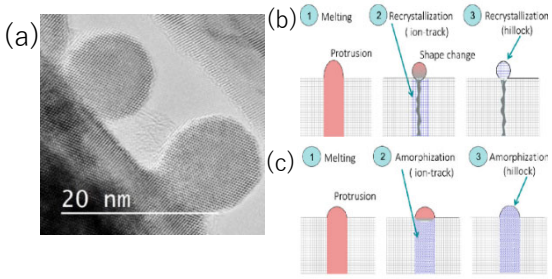

Fig.1 (a) High-Resolution TEM image of hillock observed in CeO_2 with irradiated a 200 MeV Au ion, and schematic formation processes of ion tracks and hillocks in (b) CeO_2, (c) amorphizable ceramics (YIG, $LiNbO_3$, and $ZrSiO_4$).

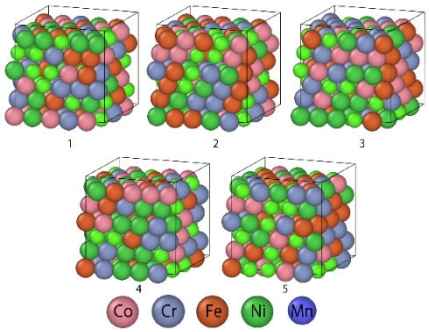

Fig.2 Atomic structure models of FCC alloy created by using Special Quasi-random Structure (SQSs). Five different atomic structure models were produced as a CoCrFeNiMn random alloy[2].

Reference

1) N Ishikawa, *et al.*, "Comprehensive Understanding of Hillocks and Ion Tracks in Ceramics Irradiated with Swift Heavy Ions", *Quantum Beam Sci.* 4(4), 43 (2020).
2) "Development of new reduced activation HEAs by additive manufacturing method", *JFY2020 progress report*. This work was supported by MEXT Innovative Nuclear Research and Development Program Grant Number JPMXD0220354336.

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Research Group for High Temperature Science on Fuel Materials

We treat issues on nuclear fuel materials for both light water reactors and advanced reactors in the future. Experimental works on uranium and non-radioactive materials are carried out in the Research building No. 4, with various heating devices and analytical apparatus. Those on transuranium elements (TRU; Pu and minor actinides) are performed at TRU-HITEC, consisting of specially designed hot cells and a glove box with highly purified argon atmosphere. We also take advantage of computer science to understand or predict phenomena concerning the fuel materials at high temperatures and under the irradiation conditions.

Nitride fuel for MA transmutation

For fabrication of the actual nitride fuels for minor actinide (MA) transmutation, the N_2 gas highly enriched with N-15 is needed to avoid the accumulation of C-14 in the fuel from $^{14}N(n,p)^{14}C$ reaction. To realize the availability of $^{15}N_2$ gas in several hundred kg – 1 ton/year is an important issue. In cooperation with the gas plant industry, we have designed basic specifications of a N_2 cryogenic distillation plant, based on the existing O-18 commercial plants with the similar concept. Fig. 1 shows an example of distillation simulation results and a CAD image of the plant. The annual production of the designed plant is 1 ton of $^{15}N_2$ gas enriched with 99 % N-15.[1] The product cost is estimated to be 1/30 of the current distribution price, produced from the existing NO cryogenic distillation plants.

Another technical issue is the economical use of $^{15}N_2$ gas in the fuel fabrication process. MA nitride is obtained by carbothermic reduction of the oxide under highly-pure N_2 gas flow, with evolution of CO gas. We have designed and assembled a "nitrogen circulation refining system" for laboratory-scale demonstration of N_2 gas use in closed cycle without exhaust. This prototype system consists of the CO removal unit to purify N_2 gas with nickel catalyst, and the circulation unit that can automatically keep the system pressure and the N_2 gas flow rate, as is shown in Fig. 2. Comparative experiments on the nitridation of lanthanide oxide as surrogate of MA by using this system or ordinary "once-through" N_2 gas flow resulted in that the impurity oxygen in both nitride products are the same level. Especially, CO in N_2 gas flow at the furnace outlet (several thousand ppm) was completely removed to zero ppm through the well-designed CO removal unit.[2] These results means that the concept of the prototype system can be applied to the actual fuel fabrication.

By the R&D activities described above, we have considerably advanced in solving the 15N2 issues for the nitride fuel fabrication.

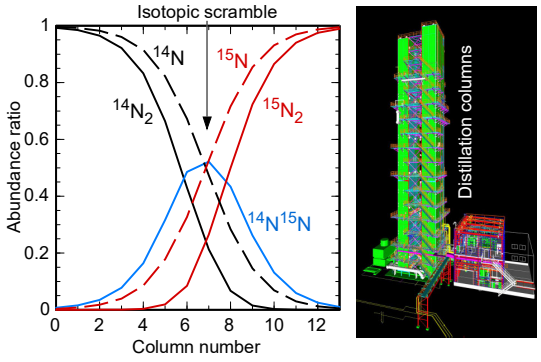

Fig.1 Example of the N-15 distillation simulation results (left) and CAD image of the plant (right). 13 distillation columns with 70 m height.

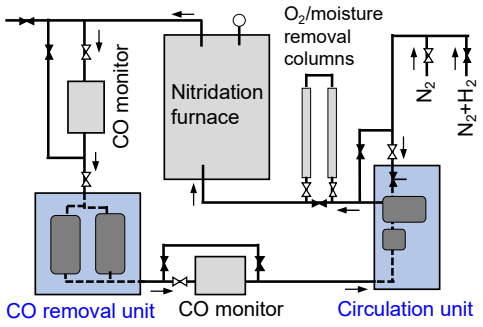

Fig.2 Schematic diagram of the prototype "nitrogen circulation refining system."

Reference

1) M. Takano, *JAEA-Review* 2020-080 (2021).
2) T. Iwasa, *et al.*, *JAEA-Technology* 2020-024 (2021).

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Development Group for LWR Advanced technology

This group was founded to conduct a research program to improve LWR (Light Water Reactor) safety by evaluating the rationality of regulation standard, safety margin estimation, accident management, etc., from a scientific point of view. Through this, the group aims to advance the safety technology in LWR, and also contribute to accelerate the decommissioning and dismantling of Fukushima Dai-ichi Nuclear Power Station (1F). This group mainly conducts studies on fission product (FP) behaviors under severe accident (SA) condition, and development of accident tolerant fuel (ATF) cladding.

Fission product (FP) behavior

We are updating a database of FP chemistry named ECUME (Effective Chemistry database of fission products Under Multiphase rEaction). ECUME covers important phenomena revealed after the 1F SA.

Radioactive cesium (Cs) is one of the principal radiation sources and information on the Cs distribution within reactor pressure vessel (RPV) is of crucial importance to make a reasonable safety assessment for the fuel debris retrieval and the reactor decommissioning at 1F. Especially, a large amount of Cs can be present in the upper region of the RPV by chemical reactions of Cs vapor species with structures, known as chemisorption. Thus, a chemisorption model is incorporated into current SA analysis codes. However, the existing chemisorption model cannot accurately reproduce experimental results and is considered not to be suitable for the estimation of the Cs-chemisorbed amounts under various conditions experienced in 1F. Therefore, a modified Cs chemisorption model which accounts for effects of chemically affecting factors (i.e., silicon content in stainless steel and gaseous Cs concentration) was constructed. As a result, modified model called ECUME model was able to describe these effects and more accurately reproduce the experimental data than the existing model (Fig.1). Thus, the Cs distribution in various chemical condition in 1F accident is expected to be estimated with higher reliability by implementing ECUME model into SA analysis code SAMPSON, which is developed under the collaboration with the Institute of Applied Energy (IAE).

Accident Tolerant Fuel (ATF)

For further improvement of safety of the existing LWRs, ATFs are being developed. In Japanese development program, FeCrAl-ODS steel for BWR, SiC fiber reinforced SiC matrix composite (SiC/SiC) for BWR and PWR, and also Cr and CrN coated zircalloy for PWR, were selected as major ATF cladding concepts. We are conducting fundamental studies on ATF cladding candidates to promote the developments in industries. To investigate the detail oxidation behavior of the FeCrAl-ODS material during hypothetical accidental condition which may affect the integrity of the fuel, we conducted oxidation tests in high temperature steam flow in thermobalance in the temperature range from 600 to 1150 °C, and mass gain during oxidation was visibly observed over 750 °C. Followed parabolic law, the oxidation rate was the highest at 1150 °C (Fig.2), while which at 750 °C was unexpectedly higher than that in the range from 800 to 950 °C. Raman spectroscopy on the sample surface indicated that oxides such as Al_2O_3, Fe_2O_3, Fe_3O_4, etc. were formed, and ratio of Al_2O_3 increased as the testing temperature rises. Oxidation resistance of FeCrAl-ODS during accident is considered that it is ensured by the surface Al_2O_3 layer, however this study suggested that it is not the case at lower temperature such as 750 °C.

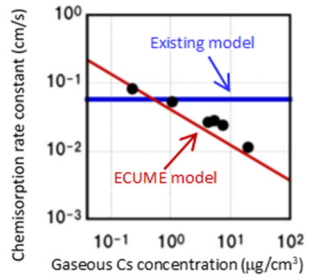

Fig.1 Dependence of gaseous Cs concentration for the chemisorption rate constant[1)]

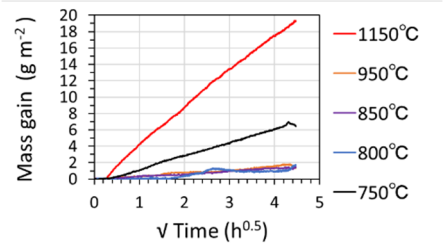

Fig.2 Profile of mass gain on FeCrAl-ODS during oxidation test in high temperature steam[2)]

Reference

1) K. Nakajima, *et al.*, *Mech. Eng. J.* Vol.7, No.3 (2020).
2) Y. Nemoto, *et al.*, *abstract for the AESJ Fall meeting in 2020*, 1E07 (2020) (*in Japanese*).

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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 the nuclear industry by developing new technique of measurement and computational method. Two remarkable achievements of JFY2020 would be introduced as followings.

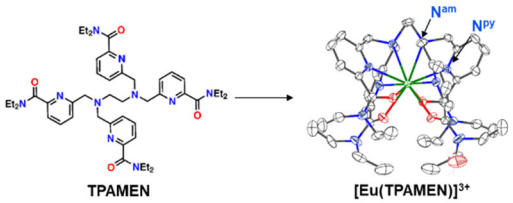
Ligand Design for Minor Actinides Separation from Lanthanides

Partitioning and transmutation strategy of minor actinides (MAs) has been developed to reduce radiotoxicity of high-level liquid waste (HLLW). We have designed ligands with high separation ability of trivalent MAs (Am^{3+}, Cm^{3+}) from lanthanides by the aid of density functional theory (DFT) calculation.

DFT calculation designed a novel ligand, TPAMEN, which is predicted as a potential candidate for MAs separation from lanthanides, and synthesized the complex with Eu^{3+}, $[Eu(TPAMEN)]^{3+}$, as well as the single crystal (Fig. 1).[1] X-ray crystallography revealed a 10-fold coordination sphere of $[Eu(TPAMEN)]^{3+}$. Detailed geometry optimizations of $[M(TPAMEN)]^{3+}$ (M = Eu^{3+}, Am^{3+}) using the obtained crystal structure as the starting coordinate reproduced the coordination geometry of $[Eu(TPAMEN)]^{3+}$ and indicated the shorter Am^{3+}-N bond lengths than the Eu^{3+}-N bond lengths (Fig. 1). Further electron density analysis suggested that the potential high performance of TPAMEN in Am^{3+}/Eu^{3+} separation attributed to the stronger covalency in the Am^{3+}-N bonds than the Eu^{3+}-N bonds.

Speciation of Platinum-Group Metals in High-Level Liquid Waste

Light platinum-group metals (Ru, Rh, Pd), existing as fission products in HLLW, are known to precipitate with coexisting ions. This is considered to hinder the vitrification process of HLLW. As the first step to understand the chemical behaviors of PGMs in HLLW we have discussed stepwise nitration reaction of ruthenium-nitrosyl ion, $Ru(NO)^{3+}$, using DFT calculation.[2] We compared the relative stability among the coordinative isomers of the nitrated products, $[Ru(NO)(NO_3)_x(H_2O)_{5-x}]^{(3-x)+}$ (x = 1-5). This indicated that the stepwise nitration reactions proceed via the NO_3^- coordination to the equatorial plane toward the Ru-NO axis (Fig. 2). We also simulated the fraction of the nitrated species based on the Gibbs energies in the stepwise nitration reactions (Fig. 3). Correction of association energy between the nitrated species and a substituting ligand increased the fractions of the complexes with x = 2, 3 in high HNO_3 concentration, being consistent with the experimental observation.



Bond length/ Å	Calc/Exp	Bond length/ Å	Calc
Eu^{3+}-N^{am}	2.789/2.78(1)	Am^{3+}-N^{am}	2.736(4)
Eu^{3+}-N^{py}	2.76(4)/2.66(6)	Am^{3+}-N^{py}	2.75(4)
Eu^{3+}-O	2.44(2)/2.42(2)	Am^{3+}-O	2.50(2)

Fig.1 Molecular structures of TPAMEN and $[Eu(TPAMEN)]^{3+}$ (upper) and bond lengths of $[M(TPAMEN)]^{3+}$ (down).

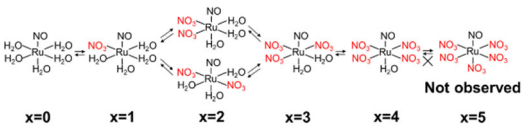


Fig.2 Stepwise nitration reaction of $Ru(NO)^{3+}$.

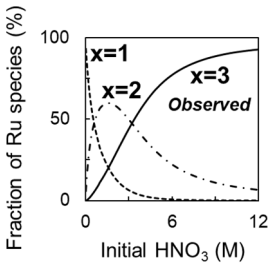


Fig. 3 DFT simulation of Ru speciation depending on HNO_3 concentration.

Reference

1) Schnaars, K. *et al.*, *Inorganic Chemistry*, 60, 2477-2491, (2021).
2) Kato, A. *et al.*, *RSC Advances*, 10, 24434-24443, (2020).

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Research Group for Analytical Chemistry

Accurate analyses of radioactive substances are fundamental technologies supporting nuclear development. Control of the chemical state of analytes and removal of interference elements prior to instrumental analysis are key techniques for the high precision chemical quantitative analysis. We developed practical analytical methods for radionuclides and traces of irradiation based on studies of elucidation of chemical reactions and separation phenomena such as adsorption, precipitation, aggregation, etc. It is engaged on instrumental analyses such as electrochemical method, mass spectrometry, and so on.

Speciation analysis is necessary to understand the behavior of radioactive elements such as actinides. Our trace-detection and microscopic analysis techniques were applied to the measurement of real samples. In the decommissioning of the TEPCO's Fukushima Dai-ichi Nuclear Power Station (1F), there is concern about internal exposure due to ingestion of particulate matter containing alpha radionuclide into the body. Our micro-analysis technique was applied to elucidation of forms of particulate alpha emitter in a stagnant water in 1F.

Spin-off research using analytical techniques of actinides is also our key issue. Electrochemical analysis and uranium handling techniques were utilized to reaction kinetics study for a large capacity battery.

External Exposure Dose Estimation using ESR Technique

Electron spin resonance (ESR) dosimetry using tooth is one of the powerful tools for the individual external exposure dose estimation. The principal of ESR dosimetry is to measure the long-lived CO_2 radicals in tooth enamel as a trace of radiation. Organic dentine and metal components in tooth interfere the ESR measurement, therefore, it is necessary to remove them and prepare "pure" inorganic enamel. We have established the enamel preparation methodology by centrifugation and appropriate chemical treatments, it enables us to improve the detection limit of ESR dosimetry to detect the exposure dose below 100 mGy. We have been estimating the exposure dose of wild animals captured in Fukushima Prefecture[1]; our future aim is to assess the exposure dose for children in Fukushima Prefecture.

Electrochemical Studies of Uranium(IV) to Build a Redox Flow Battery using Uranium

As one of the perspective candidates of the application of depleted uranium, we are studying a redox flow battery (RFB) using uranium (U) as an electrode active material. To develop the RFB with U, U^{III}, and U^{V} species which are unstable in an aqueous solution must be stabilized. We conceive to use an ionic liquid (IL) for their stabilization. Also, an organic solvent was mixed to solve high viscosity which was a disadvantage of an IL. The electrochemical behavior of uranium(IV) tetrachloride in [EtMeIm][Tf_2N]–DMF (1:1 v/v) mixture was studied at 298 K.[2] We found a quasi-reversible U^{III}/U^{IV} couple by using glassy carbon as a working electrode in an IL–DMF mixture having a viscosity that could be pumped. Since the mixture system has low resistance, it is suitable for building an RFB which must handle a large current. The reversibility of the U^{III}/U^{IV} couple and the improvement of the electromotive force in an RFB system by addition of Cl^-. We believe that these results will help build a novel URFB system.

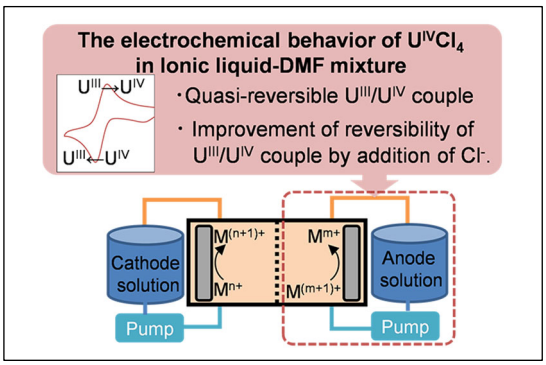

Fig.1 Concept of the U redox flow battery.

Reference

1) T. Oka, *et al.*, *Radiat. Meas.*, 134, 106315 (2020)

2) K. Ouchi, *et.al.*, *Chem. Lett.*, 50, 1169-1172 (2021).

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Research Group for Environmental Science

We research dynamics of radioactive materials in the atmospheric, terrestrial, and oceanic environments to improve assessment technology of environmental impact of radioactive materials. Computer models are developed and validated to predict the movement of radioactive materials in the environment. In particular, we developed atmospheric prediction systems (WSPEEDI-II and WSPEEDI-DB) and an oceanic prediction system (STEAMER). We also obtain observational data by field survey and analyze the data with advanced analysis technology. The data are used to clarify dynamics of radioactive materials and validate accuracy of the computer models.

Development of a Combined LES/RANS Model

We developed a large-eddy simulation (LES) model to predict the atmospheric dispersion of radioactive materials in local areas. The LES model was used in many studies to predict turbulent flows in urban areas. However, it takes much time to perform atmospheric dispersion simulations using the LES model, because it precisely calculates unsteady flow fields. On the other hand, a Reynolds-averaged Navier-Stokes (RANS) model solves averaged equations to compute mean flow fields. The RANS model has the advantage of low computational costs, although the RANS model is generally less accurate than the LES model for predicting atmospheric fields.

To reduce the calculation time in emergency response, our previous study proposed a practical and quick atmospheric dispersion simulation method for a fixed source location based on atmospheric dispersion fields precalculated by the LES model[1]. This study aims to develop a combined LES/RANS model to perform atmospheric dispersion simulations for changeable source locations with low computational costs and high accuracy[2]. In the combined model, the flow fields are pre-calculated using the LES model. Then, the atmospheric dispersion simulations are performed using the RANS model based on the pre-calculated LES data. The empirical parameters in the turbulent flux were adjusted by comparison with results of wind-tunnel experiment (WD)[3]. The combined model successfully calculated horizontal dispersion fields consisting with those calculated by the LES model (Fig. 1).

In addition, we applied the combined model to predict the real plume dispersion in an urban area, the Central Business District of Oklahoma City. It took 60 hours to predict the atmospheric dispersion fields in the area using the LES model by a single Intel CPU core. On the other hand, it took 1.5 hours to predict those using the combined model (RANS calculation with pre-calculated LES data). It was also demonstrated that the combined model results quantitatively agree with the LES results. These results suggested that the combined model can predict the real plume dispersion in the urban area with low computational costs and high accuracy.

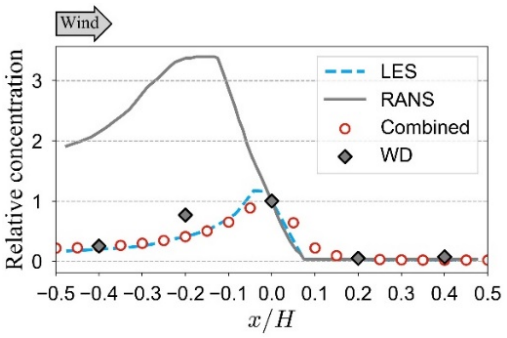

Fig.1 Horizontal profiles of the passive scalar concentrations in the street canyon at the surface. The concentrations are normalized by the values at release point (x = 0). The profiles of LES, RANS, and WD were obtained from the previous study[3].

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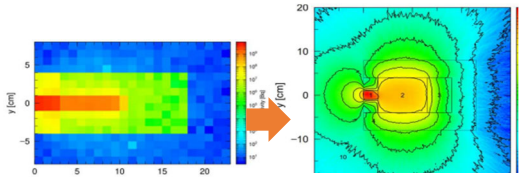
Research Group for Radiation Transport Analysis

Nowadays, computer simulation techniques that can analyze radiation transport in materials are essential tools for research and development in the field of nuclear and radiation sciences. We have developed a radiation transport simulation code, the Particle and Heavy Ion Transport code System (PHITS)[1], and apply it to industrial, scientific, and medical studies. PHITS has been upgraded to improve its higher reliability and to its wider areas to be applied. In addition, we also conduct studies on radiological protection and radiation effects. This paper presents our progresses in FY2020: upgrades of PHITS and development of a radiation dose database for public exposure in Japan.

Upgrades of the PHITS Code

DCHAIN-PHITS[2]

DCHAIN-PHITS is the time-dependent radio-nuclide production, buildup, burnup, and decay code which is coupled to PHITS. Hybrid activation cross section libraries are newly available for DCHAIN-PHITS, utilizing various combinations of evaluated data. We improved DCHAIN-PHITS to output the statistical uncertainties by propagating those of the PHITS simulation. Support for mesh tallies has also been added, allowing for induced activity calculations in tetrahedral geometries as well as with rectangular coordinate mesh tallies (xyz mesh) superimposed on any geometry, utilized in the connected induced activity and dose calculation depicted in Fig. 1.



(A) Induced activities (by DCHAIN-PHITS)

(B) Dose distribution (by PHITS)

Fig.1 Estimation of doses from induced activities by calculations of DCHAIN-PHITS and PHITS

Implementation of SCINFUL-QMD Algorithm

A new algorithm was developed to calculate responses of an organic scintillator based on a computer code SCINFUL-QMD that has been utilized in many experimental studies. PHITS can reproduce well the neutron responses and detection efficiencies of an organic scintillator (e.g., light outputs from an organic scintillator) with the new algorithm. Thus, the improved PHITS is a powerful tool for various research and development efforts in the field of radiological science.

New Calculation Method for DPA[3]

DPA is an average number of displacement atoms per atom of a material and is widely used as an exposure unit to predict the operating lifetime of materials in radiation environments. In FY 2020, we improved PHITS to derive the Athermal Recombination Correction-dpa (arc-dpa), because the arc-dpa model provides more physically realistic descriptions of primary defect creation in materials compared to a former standard model (NRT-dpa model). In our analysis, the arc-dpa cross sections are smaller than the NRT-dpa cross sections by a factor of about 3 for the displacement cross sections of Cu and W under proton irradiations with energies above 100 MeV.

Radiation Dose Database for Public Exposure

In radiological protection, radiation dose criteria are usually set with the effective dose that is derived from organ doses calculated with the ICRP phantoms (Male: RCP-AM, Female: RCP-AF). The ICRP phantoms are constructed by referring to body physiques of standard Caucasian adults. The body physiques can affect shielding of radiation within body tissues for external exposure. So far, studies have been made on the dosimetric characteristics of adult Japanese for photon irradiations. In FY 2020, we analyzed the impact of body sizes on organ doses due to external neutron irradiation. Here, the male and female phantoms with different body physiques were constructed by changing busts, chests, waists and hips of Japanese adult standard phantoms; JM-103 (male) and JF-103 (female). After that, the radiation dose database for public exposure is developed based upon the results of our analyses.

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Research Group for Partitioning

Partitioning and transmutation has been proposed as a strategy for managing high-level radioactive waste (HLW) generated by reprocessing of spent nuclear fuels via a hydrometallurgical method. HLW consists of many elements of different radiotoxicities and chemical properties. Thus, it is reasonable to separate the elements into groups instead of treating them together from the viewpoint of increasing transmutation efficiency and reducing disposal area.

To this end, our research group has been developing several extractants and has investigated their extraction properties. Recently, we proposed a novel hydrometallurgical process called SELECT (Solvent Extraction from Liquid waste using Extractants of CHON-type for Transmutation) to recycle nuclear materials and separate actinides for transmutation. A conceptual flow sheet of SELECT is shown in Figure 1. The extractants used in this process consist of carbon, hydrogen, oxygen, and nitrogen, and thus can be decomposed into gases by incineration. This contributes to reducing the volume of secondary solid waste.

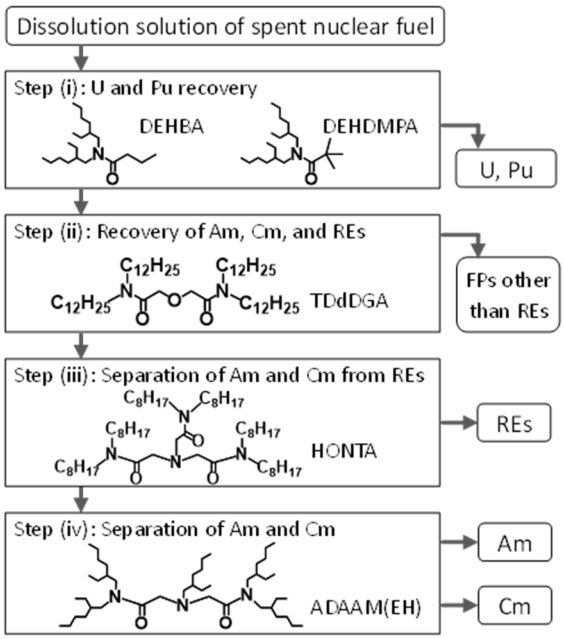

Fig. 1 A conceptual flow sheet of SELECT process and proposed extractants in each step

Radiation Tolerance Evaluation of SELECT Solvents

Because the extraction solvents will receive radiation energy from various kinds of radioactive nuclides during the separation, radiation tolerance is a critical requirement for the process feasibility. Therefore, our research group has also been investigating radiation physics and chemistry research in conjunction with the SELECT development.

A Monte-Carlo particle-transport code called PHITS was employed to calculate the radiation dose absorbed by the SELECT solvents. A droplet model combined with an approach using a refraction boundary was proposed to simplify and reduce the computational time needed to simulate radiation-energy transfer in the heterogeneous structures seen in the solvent-extraction process, such as emulsions forming in the mixture of the organic (i.e., SELECT solvents) and aqueous (e.g., HLW) solutions.[1)] The simulation results indicated that the dose absorbed by the extraction solvent from alpha radiation depends upon the emulsion structure, and that from beta and gamma radiation depends upon the size of a mixer-settler apparatus.

The subsequent radiation impacts on the extractants degradation and extraction ability were examined using a solvent test loop in conjunction with cobalt-60 gamma irradiation as collaborative research with Idaho National Laboratory (US). *N,N,N′,N′,N″,N″*-Hexaoctyl-nitrilotriacetamide (HONTA) was tested as an extractant to separate MAs from REs in SELECT process, and was found to decay exponentially with increasing dose, affording a dose coefficient of $d = (4.48 \pm 0.19) \times 10^{-3}$ kGy^{-1}.[2)]

Using the calculated absorbed dose and the obtained dose coefficient, the radiation tolerance of the extractant in SELECT process could be evaluated quantitatively.

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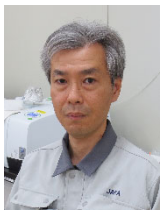

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Research Group for Nuclear Transmutation System

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, structure analysis and plant behavior using many computational codes.

ADS Conceptual Design Based on Computational Science

Beam window design

A design of a beam window which is a boundary between an accelerator and a subcritical core is one of the most important issues in the R&D of ADS. We have developed the coupled analysis system consisting of particle transport (PHITS), thermal-hydraulics (FLUENT) and structure analyses (ANSYS) for the efficient beam window design. Figure 1 presents calculation results by the coupled analysis system. The system employs the same calculation geometry and shares calculation results such as heat density calculated by PHITS or temperature calculated by FLUENT. By using the temperature distribution shown in Fig. 1 (b), the structural analysis is performed for the calculation model of the beam window. The system enables the efficient beam window design and more feasible concept will be created.

Natural convection in reactor vessel

It is supposed that the ADS is safer than conventional critical reactors because it is operated in a subcritical state. Among transient event for the typical ADS, all events except loss of heat sink (LOHS) satisfy the no-damage criteria[1]. To avoid the damage by LOHS, the ADS equips Direct Reactor Auxiliary Cooling System (DRACS) to remove the decay heat. The most important points of a DRACS operation are its reliability and to ensure the flowrate in a natural circulation state.

So, the CFD (Computation Fluid Dynamics) analysis of the natural circulation was performed to clarify the flowrate in the ADS reactor vessel. The calculation results showed that the flowrate in the natural circulation state became about 4.3 % of the nominal value and the decay heat was removed adequately (Fig. 2). It was also investigated the effect of an inner cylinder in the natural circulation state. The inner cylinder sets above an ADS core to rectify the LBE (Lead-Bismuth Eutectic) flow. By using the computational science, we could discuss the requirement of the inner cylinder.

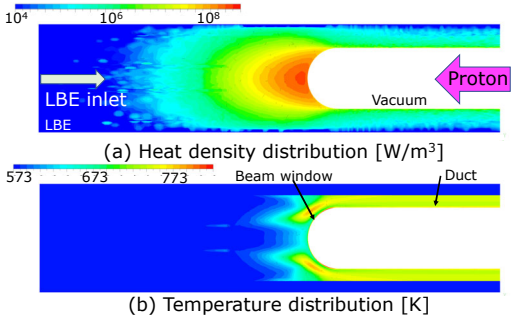

Fig.1 Calculation results by beam window coupled analysis system

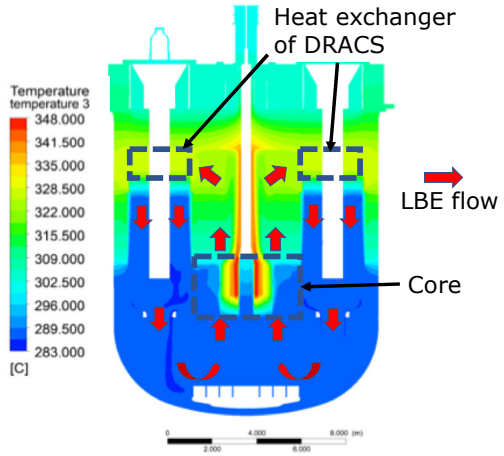

Fig.2 Temperature distribution in reactor vessel under natural circulation state

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Research Group for MA Transmutation Fuel Cycle

Feasibility study on transmutation system shows that 20 % of minor actinides (MAs) in the fuels can be transmuted in one burning cycle of about 2 years. Reprocessing of the spent fuels is necessary to transmute MAs remaining in the spent fuels. Repeating fuel fabrication, transmutation, and reprocessing can transmute most of MAs coming to the dedicated MA transmutation fuel cycle in the double-strata fuel cycle concept. We have been carrying out research and development of the technology on reprocessing of spent MA transmutation fuels. One of our concerns is on pyroprocessing technology, in which molten salts and liquid metals are used as solvents; this technique is suitable for reprocessing spent MA nitride fuels.

Fission Product (FP) Compounds in the Spent MA Nitride Fuels

Uranium(U)-free MA nitride fuels have been chosen as the first candidate for transmutation of MAs using accelerator drive system (ADS) in JAEA. To develop reprocessing technology for high burnup spent fuels, the influences of the fission product elements (FPs) should be considered. Data on the chemical states of FPs in the spent U-free MA nitride fuels are very limited, however, those for uranium nitride (UN) and uranium-plutonium mixed nitride ((U,Pu)N) fuels have been studied[1]. Rare earth elements (REs), the most abundant group in FPs, are soluble to the fuel to form solid solutions of nitrides[1]. Existence of REs scarcely affect the conditions of pyroprocessing (i.e., molten salt electrorefining), because the chemical stability of RE nitrides are similar to those of actinide nitrides[2]. On the other hand, the platinum group elements (Ru, Rh, Pd), the second most abundant group, tend to precipitate out as the intermetallic compounds such as (U,Pu) (Ru, Rh, Pd)$_3$[1]. Their chemical stability can affect the conditions of pyroprocessing. It was reported that UPd_3 did not dissolve by the molten salt electrorefining of a burnup-simulated UN sample[2].

Therefore, we investigated chemical behavior of platinum group elements coexistent with MA nitride samples in this study[3]. The chemical reaction of NpN synthesized via the carbothermic reduction of $^{237}NpO_2$, with Pd powder at 1323 K in Ar gas flow made a Np-Pd alloy, in which $NpPd_3$ was a main product. Chlorination of Np-Pd alloy was attempted using $CdCl_2$ which is considered to be used for dissolving the residue of the electrorefining into molten salts. $NpCl_3$ was obtained by the reaction of Np-Pd alloy with $CdCl_2$ at 673 K in vacuum. Figure 1 shows the X-ray diffraction (XRD) profiles of the products. These experimental results suggest that (1) $MAPd_3$ phase can be formed in spent MA nitride fuels, (2) MAs existing in $MAPd_3$ phases can be converted to chlorides, which is soluble in molten salts, by the chemical reaction with $CdCl_2$.

This study contains the results of "R&D on Nitride Fuel Cycle for MA Transmutation to Enhance Safety and Economy" entrusted to Japan Atomic Energy Agency by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

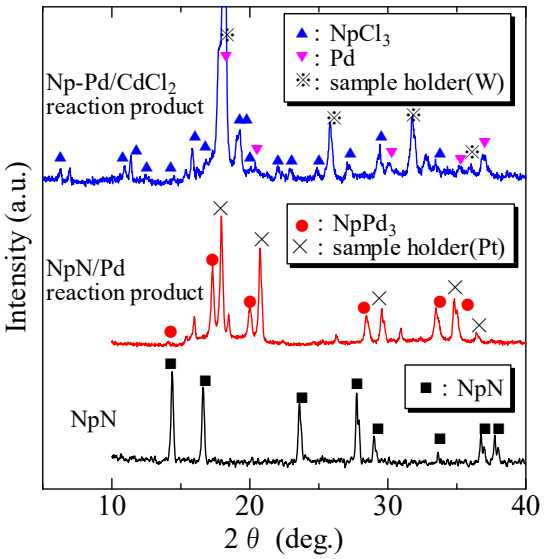

Fig.1 XRD profiles of the products

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Nuclear and LWR Engineering Division

Papers

1) Overview of the OECD-NEA Working Party on International Nuclear Data Evaluation Cooperation (WPEC), M. Fleming, D. Bernard, D. Brown, M. Chadwick, C. de Saint Jean, E. Dupont, Z. Ge, H. Harada, A. Hawari, M. Herman, O. Iwamoto, I. Kodeli, A. Koning, F. Malvagi, D. McNabb, R.W. Mills, G. Noguère, G. Palmiotti, A. Plompen, M. Salvatores, V. Sobes, M. White & K. Yokoyama, *EPJ Web of Conf.* 239, 15002 (2020).
2) HPRL – International cooperation to identify and monitor priority nuclear data needs for nuclear applications, E. Dupont, M. Bossant, R. Capote, A.D. Carlson, Y. Danon, M. Fleming, Z. Ge, H. Harada, O. Iwamoto, N. Iwamoto, A. Kimura, A.J. Koning, C. Massimi, A. Negret, G. Noguere, A. Plompen, V. Pronyaev, G. Rimpault, S. Simakov, A. Stankovskiy, W. Sun, A. Trkov, H. Wu & K. Yokoyama, *EPJ Web of Conf.* 239, 15005 (2020).

Nuclear Data Center

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