

FY2021 (2021.4-2022.3) 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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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 2021. 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 are applying 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.

July 2022

FY2021 NSEC R&D Highlights

The following 6 highlights are selected among various outcomes of the R&D activities accomplished by the NSEC's 5 divisions in FY2021.

- ✓ **Nuclear and LWR Engineering Division:**
"Development of A Low-Cost Transportable Device for Detecting Nuclear Materials"
"Development of Nuclear Data Processing System FRENDY Version 2"
- ✓ **Fuels and Materials Engineering Division:**
"Radiation Damage Mechanism in Ceramics Irradiated with Swift Heavy Ions"
- ✓ **Nuclear Chemistry Division:**
"Experimental Approach to Oxidative Degradation of Nuclear Fuel Debris Induced by Water Radiolysis"
- ✓ **Environment and Radiation Sciences Division;**
"Development of Local-Scale High-Resolution Atmospheric Dispersion and Dose Assessment System"
- ✓ **Partitioning and Transmutation Technology Division:**
"Nuclear Fuel Cycle Simulator NMB4.0 Released"

Development of A Low-Cost Transportable Device for Detecting Nuclear Materials

KOMEDA Masao, TOH Yosuke

Research Group for Nuclear Sensing

The threat of terrorism is increasing worldwide, and security inspections for radioactive materials are being conducted at airports and ports. Notably, terrorism using nuclear materials is much concern because of its serious damage to society, and nuclear material detection equipment for nuclear security is becoming increasingly important. The active method is one of the promising detection methods. In the method, the object including nuclear materials is irradiated by the radiation, such as neutron, and induces nuclear reactions. It detects nuclear materials by measuring the radiation from the nuclear reaction in the object. While this method has superior detection sensitivity and accuracy compared to the passive method, the active device has significant drawback of being massive and expensive because it requires a radiation generator with periodically oscillating intensity.

In this study, we developed a new radiation generator to realize a low-cost and transportable device. In an active device, the intensity of the irradiated radiation must vary periodically. Therefore, we developed an irradiation device in which a radiation source is rotated in order to periodically change intensity. Californium-252 (Cf-252), which emits neutrons, was used as the radiation source.

The developed irradiation device (Fig.1) incorporates a 31-cm-diameter disk with a radiation source (Cf-252) attached to its periphery, which can rotate up to 3,000 times per minute. The irradiation device is much less expensive (about 4 million yen) than conventional radiation generators (which cost about 30 million yen or more because they use accelerators). In addition, its compact dimensions (43 cm in width, 35 cm in depth, and 57 cm in height) allow it to be easily moved to wherever it is needed.

In order to explain the basic principle of the detection using a rotating irradiation device, schematics of measurements are shown in Fig.2. Irradiation neutrons from the radiation source (blue line on the left side of Fig.2) pass through the object and are measured directly by the detector, whereas neutrons emitted by the fission reaction (red line on the left side of Fig.2) are measured by the detector with a slight "delay" at slow rotation speed because neutrons must be slowed down before a fission reaction occurs. As the speed

increases (e.g., 3,000 rpm), the "delay" becomes more noticeable, and the distortion of measured neutron spectrum can be observed (right side of Fig.2), which means nuclear materials can be found by detecting the distortion. A demonstration experiment was conducted using the developed irradiation device at Kyoto university, and it was confirmed that 57 g of uranium-235, which is well below the IAEA's significant quantity of uranium-235 of 25 kg, could be detected.

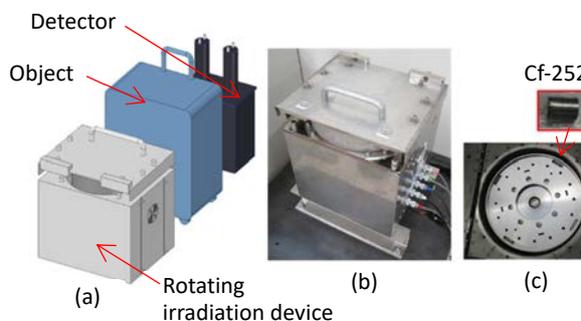


Fig.1 (a) Device arrangement, (b) Rotating irradiation device, (c) Disk and radiation source

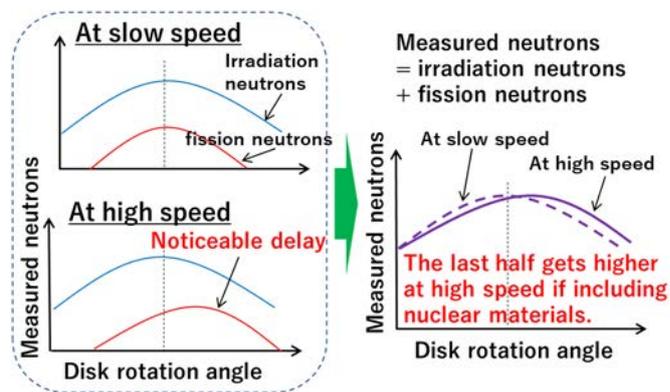


Fig.2 Schematics of measurements

Reference

- 1) M. Komeda, *et al.*, *Annals Nucl. Energy* 159, 108300 (2021).

Development of Nuclear Data Processing System FRENDY Version 2

TADA Kenichi

Research Group for Reactor Physics and Standard Nuclear Code System

The nuclear calculation codes have been used for many purposes, e.g., reactor design, and shielding analysis. The nuclear data processing system generates a cross section file which is used in these nuclear calculation codes. JAEA has developed the nuclear data processing system FRENDY to process the Japanese Evaluated Nuclear Data Library (JENDL) and other nuclear data libraries since 2013. The first version of FRENDY was released in March 2019 as an open source software under the 2-clause BSD license. FRENDY is widely used in the world and the OECD/NEA Data-Bank uses it to process their evaluated nuclear data library JEFF.

We continued to develop the new functions of FRENDY, e.g., multi-group cross section generation function¹⁾, statistical uncertainty quantification for the probability tables, and perturbation of the ACE file. Especially, the implementation of the multi-group cross section generation function for the reactor design code has been required by many processing code users. We released FRENDY version 2 including these functions in Jan. 2022 (https://rpg.jaea.go.jp/main/en/program_frendy/).

As shown in Fig.1, FRENDY version 2 can generate a multi-group cross section file from an ACE file¹⁾. The ACE file is used for many continuous energy Monte Carlo codes, e.g., MCNP, PHITS, and Serpent. Furthermore, many institutes release ACE files from their website. The users can generate the multi-group cross section files from these existing ACE files.

Critical benchmark calculations and a PWR pin-cell calculation were used for the verification of the multi-group cross section generation function in FRENDY. The multi-group cross section files were generated by FRENDY and NJOY using identical processing conditions. The comparison of the k-effective values using the multi-group cross section files processed by FRENDY and NJOY is shown in Table 1. As shown in Table 1, the k-effective values using cross section files processed by FRENDY show good agreement with those by NJOY.

We implemented unique functions in FRENDY version 2, e.g., automatic setting of the background cross section and explicit consideration of the resonance interference effect. These features will increase the value of FRENDY and attract not only current users but also new users.

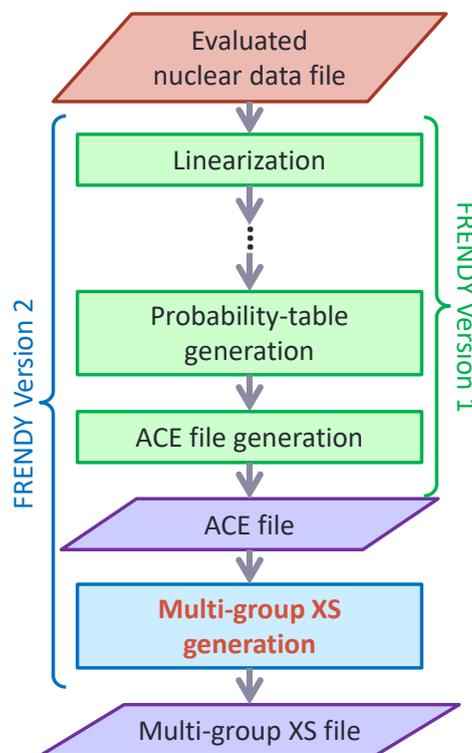


Fig.1 Processing flow of multi-group cross section (XS) generation in FRENDY version 2.

Table 1 Comparison of k-effective values using the multi-group cross section files processed by FRENDY and NJOY.

Experiments / Calculation case	FRENDY	NJOY	Relative difference
Jezebel	0.99832	0.99829	0.003%
Jezebel-Pu	0.99842	0.99838	0.004%
Godiva	0.99777	0.99776	0.001%
Jezebel-233	0.99932	0.99934	-0.002%
Flattop-U	0.99743	0.99723	0.020%
Flattop-Pu	0.99844	0.99827	0.017%
Flattop-233	0.99814	0.99801	0.013%
Thor	0.99785	0.99790	-0.005%
PWR pin-cell	1.40755	1.40751	0.003%

Reference

1) A. Yamamoto, K. Tada, et al., *J. Nucl. Sci. Tech.* 58(11), 1165 (2021).

Radiation Damage Mechanism in Ceramics Irradiated with Swift Heavy Ions

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When ceramics are irradiated with swift heavy ion beams (having more than tens of MeV energy), nanostructures called ion tracks are created along the ion paths. In many ceramics the size of the ion tracks can be predicted by the thermal spike theory. However, in some ceramics the size of the ion tracks observed by the experiment was found to be smaller than that predicted by the theory. Surprisingly, such size discrepancy was remarkable in ceramics having high radiation-resistance. What is the reason for the failure of the theory? Our systematic study of surface nanostructure (nanohillock) has revealed the reason¹.

According to our transmission electron microscopy observation (Fig.1)¹, the ion track size is smaller than the nanohillock size in CeO₂ which is one of the highly radiation-resistant ceramics, while the size difference is very small in ceramics having low radiation-resistance.

A likely process in ceramics having low radiation-resistance is described in Fig.2(a)². Firstly, (1) a part of the surface protrudes during local melting along the ion path and secondly, (2) the molten region turns into damaged region during rapid cooling. This process is very likely, since the nanohillock size is found to be similar to the ion track size in ceramics having low radiation-resistance³.

In contrast, in ceramics having high radiation-resistance (Fig.2(b)²), (1) a part of the surface protrudes during local melting along the ion path, (2) the molten region partially recrystallizes resulting in shrinkage of the damaged region and finally (3) the protruded part near surface recrystallizes. This hypothesis can be validated by Fig.1 showing that the ion track is smaller than the nanohillock. This means that ion tracks rapidly shrink in highly radiation-resistant ceramics owing to extremely fast recrystallization. It also shows that the recrystallization process is not assumed in the conventional theory^{1,4}. The well-aligned atomic layers inside nanohillocks (Inset of Fig.1) further support the recrystallization process which is depicted in the third process of Fig.2(b). In conclusion, the origin of the high radiation-resistance is high recrystallization capability.

This work was supported by JSPS KAKENHI Grant Number 20K05389.

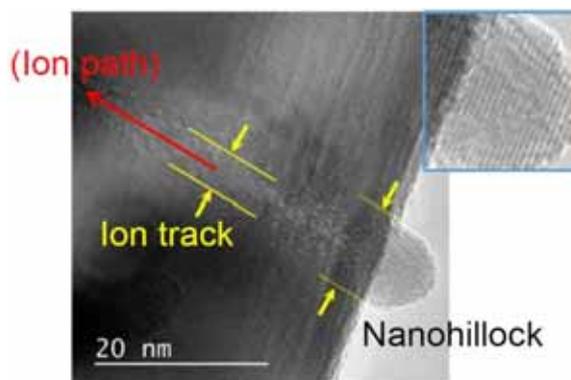


Fig.1 Nanostructure created in CeO₂ irradiated with swift heavy ions.² The inset shows the magnified image of a nanohillock.

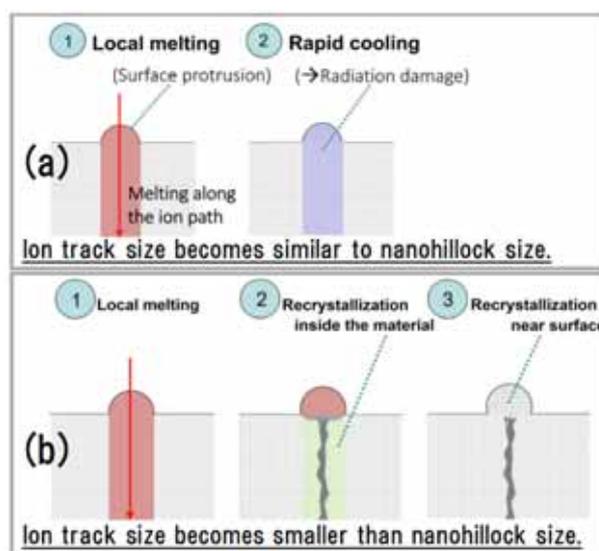


Fig.2 Nanostructure formation process by high energy heavy ions in (a) ceramics having low radiation-resistance and in (b) ceramics having high radiation-resistance.²

Reference

- 1) N. Ishikawa, et al., *しょうとつ J. Atomic Collision Res.* 18(3), 43 (2021) (in Japanese).
- 2) N. Ishikawa, et al., *Quantum Beam Sci.* 4(4), 43 (2020).
- 3) N. Ishikawa, et al., *J. Applied Phys.* 127, 055902 (2020).
- 4) N. Ishikawa, et al., *Nanotechnology* 28(44), 445708 (2017).

Experimental Approach to Oxidative Degradation of Nuclear Fuel Debris Induced by Water Radiolysis

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Fuel debris generated in the accident at TEPCO's Fukushima Daiichi Nuclear Power Station remains in the reactors, and substantial time and effort will be required until retrieval of the debris. The debris has been most likely exposed to water since the accident. The contact with water would bring on degradation of the debris matrix due to the radiolysis of water. The water radiolysis generates oxidants such as hydrogen peroxide (H_2O_2), which can oxidize uranium to hexavalent U(VI). According to previous studies of uranium(IV) oxide (UO_2) and spent fuels¹, the U oxidation proceeds at the interface to water and the matrix gradually dissolves because U(VI) has higher water-solubility than U(IV). Hence, to examine possible degradation processes of fuel debris, we carried out leaching experiments using simulated fuel debris².

The simulated debris samples were prepared from powders of UO_2 , stainless steel (SUS304), and zirconium metal (Zr) or oxide (ZrO_2) by heat treatments under various conditions (Table 1). After analyzing the phase composition of the samples, the simulated debris samples were immersed in aqueous H_2O_2 solution for up to 30 days. H_2O_2 was added since it is the oxidant generated by water radiolysis that has major impact on the U oxidation. After certain periods of immersion, the samples were analyzed by Raman spectroscopy as well as chemical analysis of the leached elements.

The analysis of the leached elements showed significant dissolution of U from the samples. The reaction of H_2O_2 concurrently induced precipitation of uranyl peroxides, $UO_2(O_2) \cdot nH_2O$ ($n = 2$ or 4). These two processes made the dissolved U concentration once increase and then decrease with leaching time (Fig.1(a)). The formation of uranyl peroxides was clearly confirmed by the Raman spectroscopy (Fig.1(b)). These results demonstrate that uranyl peroxides are possible alteration products of fuel debris from H_2O_2 reaction.

In contrast, the sample in which formation of a

U-Zr oxide solid solution proceeded to a remarkable degree (USZr O_2 1600-O) showed much less U dissolution and no Raman signal of uranyl peroxides. This finding indicates that formation of the oxide solid solution of Zr with UO_2 improves the durability of fuel debris against H_2O_2 reaction.

This work was supported by the Japan Atomic Energy Agency Nuclear Energy S&T and Human Resource Development Project through concentrating wisdom, Grant Number [JPJA18P18071886].

Table 1 Examples of simulated debris samples prepared in this study.

Sample	Heat treatment	Major U phase
USZr 1200-O	1200 °C, 2% O ₂	U ₃ O ₈
USZr 1600-O	1600 °C, 2% O ₂	UO ₂ (s.s.)** (low Zr)
USZr 1600-A	1600 °C, Ar	UO ₂
USZrO ₂ 1600-O*	1600 °C, 2% O ₂	UO ₂ (s.s.)** (high Zr)

* ZrO_2 was used as a starting material, while metal Zr for the other samples.

** "(s.s.)" stands for solid solution.

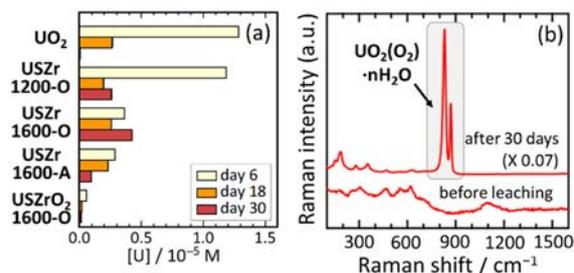


Fig.1 (a) U concentrations in the leachate and (b) Raman spectra of uranyl peroxides formed on USZr1600-O.

Reference

- 1) T.E.Eriksen, *et al.*, *J Nucl. Mater.* 420(1-3), 409 (2012).
- 2) Y. Kumagai, *et al.*, *J. Nucl. Sci. Tech.* 59(8), 961 (2022).

Development of Local-Scale High-Resolution Atmospheric Dispersion and Dose Assessment System

NAKAYAMA Hiromasa

Research Group for Environmental Science

We developed a local-scale high-resolution atmospheric dispersion and dose assessment system (LHADDAS)¹⁾ for safety and consequence assessment of nuclear facilities and emergency response to nuclear accidents or deliberate releases of radioactive materials in built-up urban areas in cooperation with Center for Computational Science and e-Systems (CCSE).

This system comprises of three parts: 1) Preprocessing of input files, and main calculation by a local-scale high-resolution atmospheric dispersion model using large-eddy simulation (LOHDIM-LES)²⁾. 2) A real-time urban dispersion simulation model based on a lattice Boltzmann method (CityLBM)³⁾. 3) Postprocessing of dose calculation by a simulation code powered by lattice dose-response functions (SIBYL)⁴⁾.

We examined the performance of LHADDAS using the following two application cases from our previous studies. The first case involved a controlled tracer plume dispersion in the vicinity of an actual nuclear facility and building arrays. The second was about tracer gas dispersion in an actual central urban district. Here, we showed the first application case of the LOHDIM-LES simulation of the actual tracer plume dispersion of radionuclides released from the nuclear fuel reprocessing plant in Rokkasho, Japan as an example. The simulation period was from 09:00–19:30 Japan Standard Time (JST) on 17 June 2008. Figure 1 shows an instantaneous shot of the tracer plume released into the atmosphere from a stack at 10:40 JST on 17 June. Figure 2 compares the LOHDIM-LES results with the monitoring post (MP) data. Consequently, the outcome demonstrated an intermittent increase in the air dose rates at MP2. These rates frequently showed higher peaks at MP3 than those at MP2. Therefore, the main part of the plume was transported above MP3. These time variation patterns at the different MPs were simulated well by LOHDIM-LES integrated with the SIBYL module. The model performance of LOHDIM-LES and CityLBM was also demonstrated with the data collected during the urban field experiments for the second application case.

LHADDAS has broad functionalities and performs extremely well in simulating turbulent flows, plume dispersion, and dry deposition under realistic meteorological conditions. It can simulate real-time tracer dispersion using a locally mesh-

refined lattice Boltzmann method. Moreover, it can estimate the air dose rates of radionuclides from air concentration and surface deposition taking into consideration the influence of individual buildings.

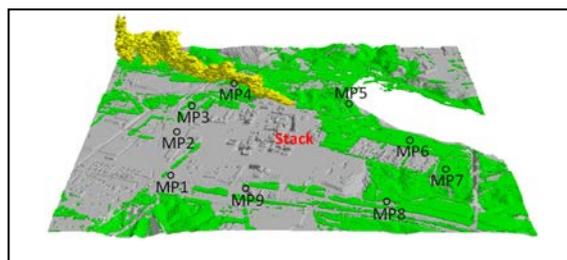


Fig.1 Instantaneous plume dispersion field at Rokkasho Reprocessing Plant at 10:40 JST 17 June.

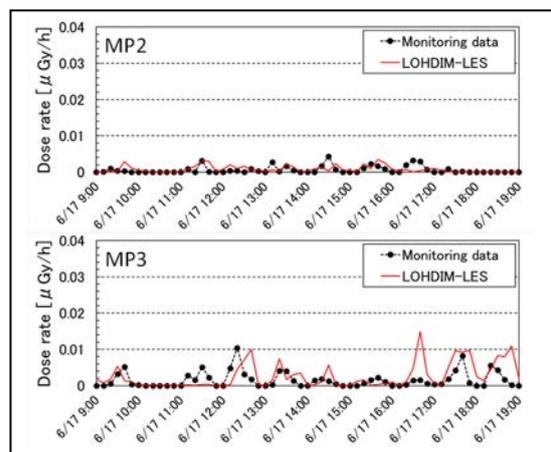


Fig.2 Time series of the air dose rate at the MPs during the period from 09:00 JST to 19:00 JST 17 June 2008.

Reference

- 1) H. Nakayama, *et al.*, *J. Nucl. Sci. Tech.* 59(10), 1314 (2022).
- 2) H. Nakayama, *et al.*, *J. Nucl. Sci. Tech.* 58(9), 949 (2021).
- 3) N. Onodera, *et al.*, *Boundary-Layer Meteor.* 179(2), 187 (2021).
- 4) D. Satoh, *et al.*, *PLoS ONE* 16(1), e0245932 (2021).

Nuclear Fuel Cycle Simulator NMB4.0 Released

NISHIHARA Kenji¹, OKAMURA Tomohiro², KATANO Ryota¹, OIZUMI Akito¹, NAKASE Masahiko², ASANO Hidekazu², TAKESHITA Kenji²

1 Research Group for Nuclear Transmutation System

2 Laboratory for Zero-Carbon Energy, Institute of Innovative Research, Tokyo Institute of Technology

Currently in Japan, various reactor types and reprocessing technologies to reduce the waste load are being developed for future nuclear power utilization. In order to start full-scale development for the realization of these technologies, it is necessary to evaluate whether future introduction scenarios can be envisioned and how much the benefits and costs would be if they were introduced. For this evaluation, future scenarios with various technologies are set up, and quantity is evaluated assuming these scenarios, which are called nuclear fuel cycle simulations. So far, there are no publicly available general-purpose simulators in Japan, and many excellent simulators in other countries are not open. JAEA and Tokyo Tech have jointly developed NMB4.0¹⁾ (NMB stands for Nuclear Material Balance), a (1) fast, (2) versatile, and (3) flexible simulator, and we released it on the website (<https://nmb-code.jp>). The features of this simulator are described below.

(1) High speed: Conventional simulators have mainly analyzed the burnup changes of about 20 to 30 nuclear materials such as uranium in a nuclear reactor, but in order to accurately evaluate the generation and disposal of radioactive waste, the NMB can analyze about 150 fission products. To analyze the burnup changes of these materials, a fast solution method has been developed and equipped.

(2) Versatility: The NMB can analyze almost all processes in nuclear power generation (Fig.1). For reactors, the database has been developed to allow analysis of light water reactors and future-type reactors. For the nuclear fuel cycle, an extensive analytical model and database for temperature in geological disposal of radioactive waste have been developed, which is not included in most of the other simulators. This model allows to evaluate repository size.

(3) Flexibility: The future of nuclear power utilization in Japan is very ambiguous due to uncertainties in the replacement of light water reactors, delays in the development of fast reactors, and difficulties in selecting a geological disposal site. Therefore, we have designed the NMB capable of analyzing a variety of scenarios, such as withdrawal or continuation of nuclear power utilization, and changes in the timing of FBR introduction. (Analysis

example: Fig.2)

We are now calling for a wide range of users and developers to join Team NMB. The team is looking for users such as universities, research institutes, manufacturers, and electric power utilities, etc., to help us plan R&D and implementation strategies for future nuclear reactors and nuclear fuel cycle technologies for the sustainable use of nuclear energy.

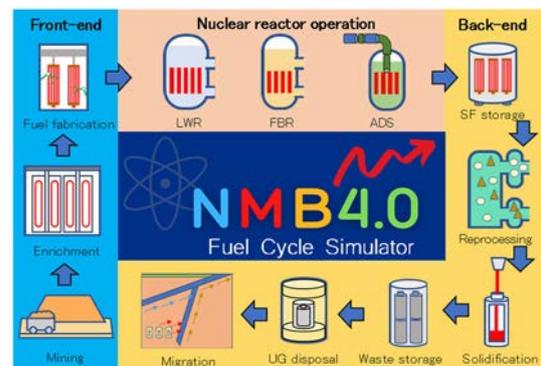


Fig.1 Nuclear fuel cycle handled by NMB4.0

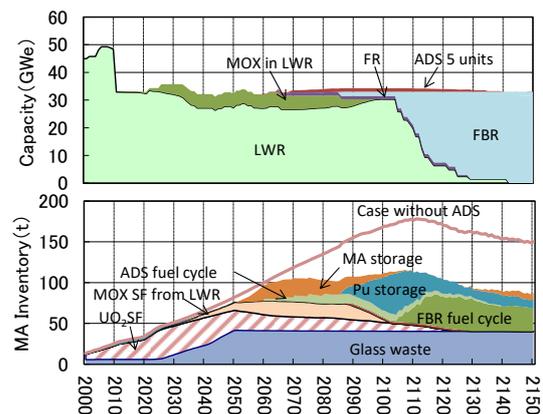


Fig.2 Example of analysis (reduction of minor actinides by ADS in FBR transition scenario)

Reference

- 1) T. Okamura, R. Katano, et al., *EPJ Nucl. Sci. Tech.* 7, 19 (2021).

FY2021 NSEC Group Activities

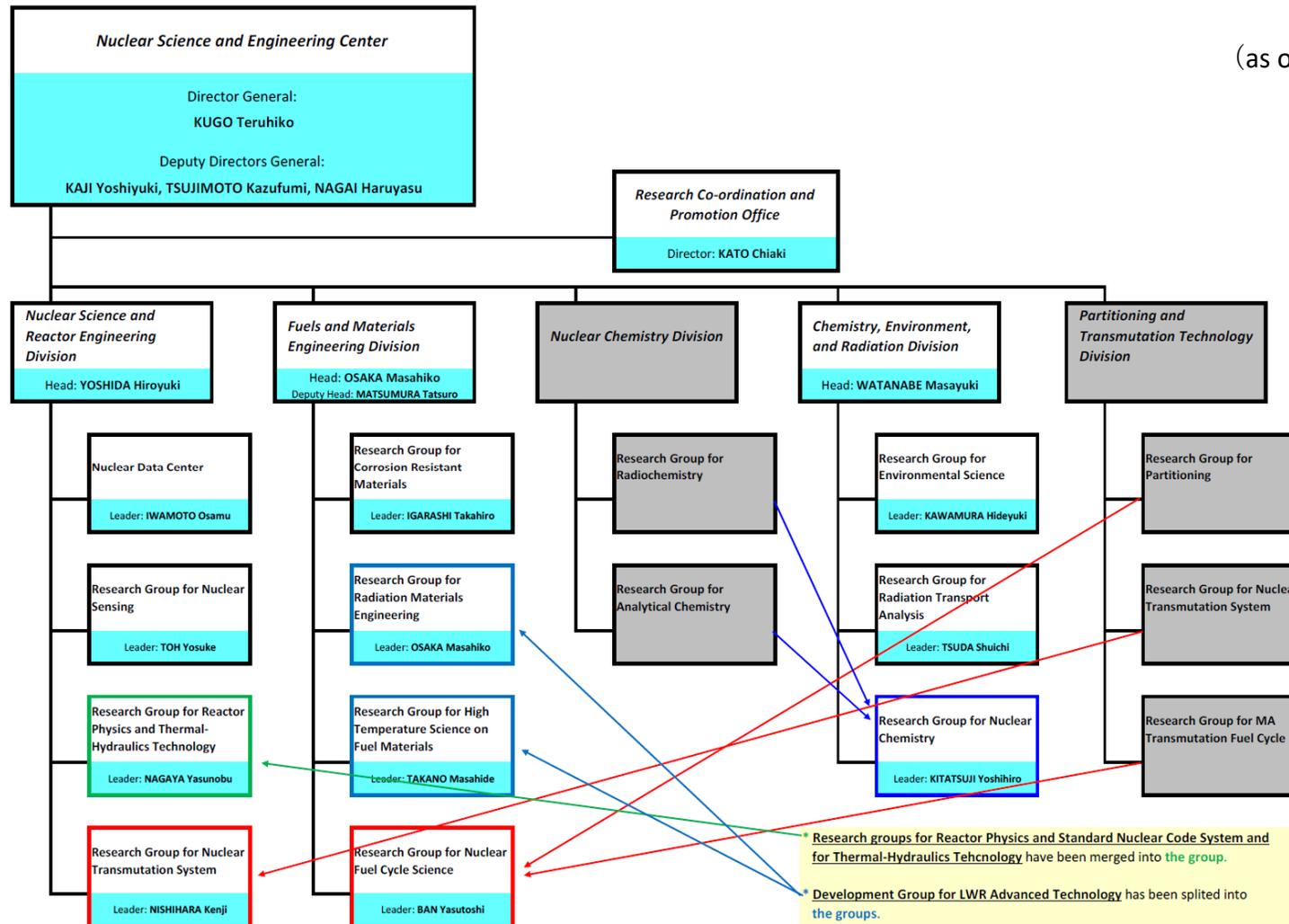
The NSEC of JAEA consisted of 15 Groups in FY2021.

Since April 2022, the NSEC has been re-organized from 15 Groups of 5 Divisions to 11 Groups of 3 Divisions:

- ✓ *“**Research Group for Reactor Physics and Standard Nuclear Code System**” and “**Development Group for Thermal-Hydraulics Technology**” have been merged into “Research Group for Reactor Physics and Thermal-Hydraulics Technology”; “**Nuclear and LWR Engineering Division**” has been renamed “**Nuclear Science and Reactor Engineering Division**”;*
- ✓ *“**Development Group for LWR Advanced Technology**” has been split and merged into “**Research Group for Radiation Materials Engineering**” and “**Research Group for High Temperature Science on Fuel Materials**”;*
- ✓ *“**Research Group for Radiochemistry**” and “**Research Group for Analytical Chemistry**” have been merged into “Research Group for Nuclear Chemistry”; “**Nuclear Chemistry Division**” and “**Environment and Radiation Sciences Division**” have been merged into “**Chemistry, Environment, and Radiation Division**”;*
- ✓ *“**Partitioning and Transmutation Technology Division**” has been abolished;*
 - *“**Research Group for Partitioning**” and “**Research Group for MA Transmutation Fuel Cycle**” have been merged into “Research Group for Nuclear Fuel Cycle Science” of “**Fuels and Materials Engineering Division**”;*
 - *“**Research Group for Nuclear Transmutation System**” has been transferred to “**Nuclear Science and Reactor Engineering Division**”.*

Organization of NSEC

(as of October 2022)



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 the mission, 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 (Japanese nuclear data library). The JENDL files are available from our website (<http://www.ndc.jaea.go.jp>).

Neutron Capture Cross Section Measurement with Activation Method

Neutron irradiated data using a fast neutron source reactor 'YAYOI' were revisited. By the analysis of gamma-rays from irradiated samples, the neutron capture reaction rates of technetium(Tc)-99 were deduced. The present measurement had a large sensitivity on the cross sections in the neutron energy region of 10-100 keV, where the recent nuclear data libraries of ENDF/B-VII.0, ENDF/B-VIII.0 and JENDL-4.0 provided rather inconsistent cross sections. The obtained data revealed that the evaluated cross section in JENDL-4.0 was the most favorable among them.¹⁾

TOF Measurement of Neutron Induced Reactions with ANNRI

Resonance parameters of niobium(Nb)-93 below 400 eV were deduced from measurements of neutron capture and transmission ratios with ANNRI installed at J-PARC with a time of flight (TOF) method. The Nb neutron capture cross section is important in activation evaluation because Nb is used in stainless steel for reinforcement and creates long-lived radioactive Nb-94 with half-life of 20 thousand years by neutron capture reaction. The present results suggested JENDL-4.0 evaluation of the neutron capture cross section at thermal neutron energy was overestimated by 18 %.²⁾ The obtained resonance parameters were adopted in JENDL-5.

Theoretical Model Prediction for Beta-delayed Neutron Emission

Beta-delayed neutron emission is important not only for nuclear reactors but also for nucleosynthesis in the universe. Experimental data for the delayed neutron emission have been

accumulated but their uncertainties are still large. With the theoretical models of relativistic quasiparticle random-phase approximation for beta-decay and a statistical model for neutron emission, the delayed neutron branching ratios were calculated for neutron rich unstable nuclei over the periodic table. The calculated results were in good agreement with available experimental data with reducing the root mean square deviation comparing with the preceding works.³⁾ This work is expected to contribute to further studies on the nucleosynthesis as well as the accuracy improvements of delayed neutron emission data for fission products.

Release of JENDL-5

JENDL-5, the latest version of JENDL, was successfully released in December 2021. It was a main target of the 3rd midterm (2015-2021) plan of Nuclear Data Center. JENDL-5 aimed to meet various needs in nuclear energy applications not only for nuclear reactor development but also for nuclear backend issues such as decommissioning of reactors and nuclear waste management. Large amount of neutron induced reaction data from light to heavy nuclides were revised and the number of nuclides nearly doubled. The performance for nuclear reactor and neutron shielding was improved by feedback of repeated benchmark tests. To meet additional needs of radiation applications, its previously released special-purpose libraries were revised and integrated into JENDL-5. JENDL-5 consists of 11 sublibraries that include data of neutron, charged-particle and photon induced reactions; neutron thermal scattering law, fission yield and decay data; and atomic related data.

Reference

- 1) S. Nakamura, *et al.*, *J. Nucl. Sci. Tech.* 58(12), 1318 (2021).
- 2) S. Endo, *et al.*, *J. Nucl. Sci. Tech.* 59(3), 318 (2022).
- 3) F. Minato, *et al.*, *Phys. Rev. C* 104(4), 044321 (2021).



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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 performing integral tests for the JENDL project with the codes. We are also developing an advanced neutronics/thermal-hydraulics coupling simulation system for enhanced safety of light water reactors (LWRs) and improvement of LWR design. The project started from 2020 JFY. This fiscal year we have developed an Interface Program to Accommodate Coupling Simulation for neutronics/thermal-hydraulics IPACS.

FRENDY

FRENDY is a nuclear data processing code.¹⁾ It enables processing of evaluated nuclear data such as JENDL. FRENDY version 1 was released in March 2019 as an open-source code from our website.²⁾ FRENDY version 1 generates the ACE files which are used for Monte Carlo codes such as PHITS and MCNP. FRENDY version 2³⁾ was released in January 2022. It implements new capabilities: the estimation of statistical uncertainty in probability table generation for unresolved resonance cross sections, the generation of multigroup constants, etc. FRENDY version 2 can be also downloaded from our website.³⁾

Integral Tests for JENDL-5

JENDL-5 was released in December 2021.⁴⁾ Since 2018, we have performed integral tests of preliminary and release versions of JENDL-5 for critical and shielding experiments. The test calculations were done for experiments mainly in the International Criticality Safety Benchmark Evaluation Project (ICSBEP)⁵⁾ handbook and conducted at JAEA. The test calculations for middle- and large-sized fast systems were done for integral experiments used for generation of the unified cross section data set of ADJ2017.⁶⁾ The shielding benchmark tests were done mainly for FNS experiments at JAEA and OKTAVIAN experiments at Osaka University. We have confirmed that JENDL-5

gives better than or the same prediction accuracy as JENDL-4.0⁷⁾ in many test cases for the critical and shielding experiments.⁸⁾

IPACS

IPACS⁹⁾ is an interface program for the advanced neutronics/thermal-hydraulics coupling simulation system. A prototype simulation system MVP/NASCA was developed by using IPACS. This system is the first step for the development of the advanced coupling simulation system. The objectives of this system are to find the issues of coupling simulations and to implement the current methodology used for neutronics (MVP) and thermal-hydraulics (NASCA) coupling. NASCA¹⁰⁾ is a subchannel analysis code for thermal-hydraulics analysis. As the next step, we will replace the subchannel code with a CFD code such as JUPITER¹¹⁾ developed at JAEA. IPACS can be obtained from RIST.

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- 1) K. Tada, *et al.*, *JAEA-Data/Code* 2018-014 (2019).
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- 3) <https://rpg.jaea.go.jp/main/en/program/frendy/> (accessed 2022-11-08)
- 4) <https://www.ndc.jaea.go.jp/jendl/j5/j5.html> (accessed 2022-11-08)
- 5) https://www.oecd-nea.org/jcms/pl_24498/international-criticality-safety-benchmark-evaluation-project-icsbep (accessed 2022-11-08)
- 6) K. Yokoyama, *et al.*, *JAEA-Research* 2018-011 (2019) (*in Japanese*).
- 7) K. Shibata, *et al.*, *J. Nucl. Sci. Tech.* 48(1), 1 (2011).
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- 11) S. Yamashita, *et al.*, *Mech. Eng. J.* 4(3), 16-00567 (2017).



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Research Group for Nuclear Sensing

Neutron is a powerful tool in the non-destructive analysis (NDA), since it has exceptional ability to penetrate high-density materials, and can induce fission and radiative capture reaction. Despite being a mature technique, further improvements in NDA are required and enable us to meet the distinctive demands in many fields. We have, therefore, developed the NDA techniques using the neutron interrogation to apply for nuclear and radioactive materials.

Development of an Active Neutron Non-destructive Analysis System: Active-N

The different analytical methods give us complementary information, which are particularly useful for the quantification of special nuclear materials and minor actinides (MA) in highly radioactive nuclear materials (HRNM), including spent fuel and MA transmutation fuel. JAEA and the Joint Research Centre of the European Commission are collaborating to develop an active neutron NDA system for nuclear non-proliferation and nuclear security. We have developed a novel NDA system: Active-N (Fig.1)¹⁾ by combining active neutron methods, as listed below, to deal with the difficult problem of measuring HRNM.

- (1) Differential Die-away Analysis (DDA)
- (2) Prompt Gamma-ray Analysis (PGA)
- (3) Neutron Resonance Transmission Analysis (NRTA)

The Active-N system was deployed at nuclear fuel cycle safety engineering research facility in JAEA. The Active-N system performance was evaluated by measuring of a simulated HRNM²⁾. The results indicate that the Active-N system has enough capabilities to detect and quantify of HRNM.



Fig.1 Active neutron non-destructive analysis system: Active-N

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

Non-destructive Quantitative Analysis of the Difficult-to-measure radionuclides: ¹⁰⁷Pd and ⁹⁹Tc

For radioactive material waste management, reliable quantification of radioactive nuclides is especially important. A non-destructive analytical scheme applicable to the difficult-to-measure nuclides of ¹⁰⁷Pd and ⁹⁹Tc, which emit no or almost no decay gamma-rays and whose half-lives are too long to be decayed out during a human lifetime, was developed. The analytical scheme is composed of a sophisticated instrument named ANNRI, which can perform measuring of gamma rays by a Ge detector array coupled with Time of Flight (TOF) measurement, and a high-intensity pulsed neutron beam generated at the Japan Proton Accelerator Complex (J-PARC) and can simultaneously perform TOF-PGA as well as PGA. The analytical capability was evaluated by applying to simulated samples of the Tc-platinum group metals obtained by the group partitioning process of spent nuclear fuels. It was confirmed that, although PGA cannot accurately analyze either of the nuclides in the simulated samples, TOF-PGA can analyze ¹⁰⁷Pd as well as ⁹⁹Tc³⁾. The TOF-PGA measurement technique could be widely used for the non-destructive analysis of ¹⁰⁷Pd and ⁹⁹Tc in nuclear wastes.

Reference

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- 2) K. Furutaka, *et al.*, *Proc. the INMM & ESARDA Joint Annual Meeting* (2021).
- 3) Y. Toh, *et al.*, *Anal. Chem.* 93(28), 9771 (2021).



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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, ACE-3D, and JUPITER, and measurement techniques to obtain thermal-hydraulics data. In addition, we are performing thermal-hydraulics experiments to construct validation databases. These advanced techniques are applied to the research activities for improving the safety of the LWRs and providing information on the decommissioning process of TEPCO's Fukushima Daiichi NPS.

Numerical Simulation of Oxidation under Severe Accident Conditions

JUPITER can simulate fuel debris melting and relocation behavior without assumptions or simplification. Besides, JUPITER can perform chemical reactions such as zirconium-water and eutectic reactions. As one of the examples of calculation by JUPITER, we performed a numerical simulation of oxidation simulation on the zircaloy surface (Fig.1)¹⁾. This simulation evaluated the time evolution of the oxide layer thickness on the zircaloy by the newly developed oxidation simulation model. In Fig.1, we visualized the growth of the oxide layer thickness. The oxide layer thickness increases as time proceeds, and the steam concentration decreases as the growth of the oxide layer. We confirmed that the modified JUPITER reasonably simulates the fundamental oxidation process.

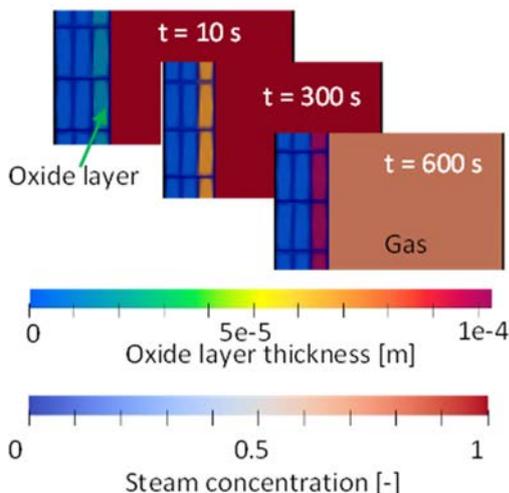


Fig.1 The time change of the oxide layer thickness

Research of Hydraulics Behavior in Venturi Scrubber

The filtered venting system is used to prevent the release of radioactive materials in severe accidents. We developed a numerical simulation method based on TPFIT to evaluate the performance of the Venturi scrubber, which is one of the components of the filtered venting system. In addition, we performed various experiments to validate the developed simulation method. Figure 2 shows an example of the predicted result in the Venturi scrubber²⁾. The result of visualized experiments is also shown. In the visualized result, the injected liquid formed a liquid bridge. We confirmed the formation of the same liquid bridge by the numerical simulation. This liquid bridge captures small radioactive particles effectively; therefore, the prediction of liquid bridge formation is important to evaluate the performance of the Venturi scrubber. We will evaluate the decontamination factor for the Venturi scrubber by using these results.

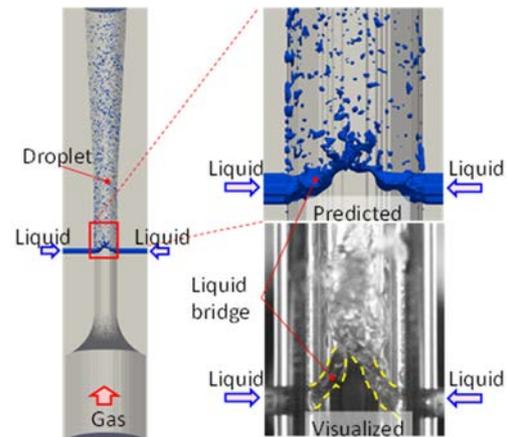


Fig.2 Predicted and Visualized Hydraulic Behavior in Venturi Scrubber

Reference

- 1) S. Yamashita, *et al.*, *Proc. NURETH-19* 35516 (2021).
- 2) S. Uesawa, *et al.*, *2022 Annual Meeting of AESJ* 1E15 (2022) (in Japanese).



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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 focus on the corrosion mechanism to develop the methods of corrosion prediction and prevention. Through these research activities, we contribute to the improvement of the reliability of nuclear facilities.

Researches on Corrosion Behavior of Tantalum in Sodium Hydroxide Solution

The corrosion behavior of tantalum used in nuclear fuel reprocessing equipment in sodium hydroxide (NaOH) solution, which is expected to be used in decontamination to reduce worker radiation exposure, was investigated by immersion corrosion tests, and the mechanism of aging degradation was studied. The immersion corrosion tests showed that the corrosion rate (CR) increased with NaOH concentration. Moreover the CR peaked and then decreased with immersion time (Fig.1). The time to peak was shorter for higher NaOH concentrations. Scanning electron microscopy, Raman analyses and X-ray diffraction patterns of the surface of the specimens after the tests showed the corrosion product ($\text{Na}_8\text{Ta}_6\text{O}_{19}$) film formation and the film growth with immersion time. And our electrochemical measurement suggested that the change in CR with time should be explained by the film growth and concluded that the film growth inhibits corrosion reactions.

Researches on Corrosion Behavior of Stainless Steel in High-Temperature Water

The effects of electrochemical corrosion potential (ECP) on the water chemistry within a crevice of Type-316L stainless steel was investigated. In-situ measurements of the electrical conductivity of the solution within a 15- μm -gap crevice (σ_{crev}) were conducted in high temperature water (288 °C containing 10 ppb of Cl^- as major anionic impurities). An increase in the ECP at the external surface of the crevice specimen (E_{ext}) from approximately -0.49 V to 0.15 V resulted in an increase in σ_{crev} (measured at 21 mm from the crevice mouth) from 12 $\mu\text{S}/\text{cm}$ to >300 $\mu\text{S}/\text{cm}$ (Fig.2). A further increase in E_{ext} led to a decline in σ_{crev} . Finite element model analysis of the crevice water chemistry taking into account the electrochemical reaction and transportation

phenomena of ions quantitatively reproduced this behavior. It was indicated that in the low potential range (see Fig.2), Cl^- was considered to be the major anionic species transported into the crevice and σ_{crev} increased with E_{ext} . In high potential range, transpassively generated HCrO_4^- began to migrate into the crevice. Since HCrO_4^- act as strong oxidizer and oxidize metal cation to precipitate them as metal oxide eventually decreased σ_{crev} value.

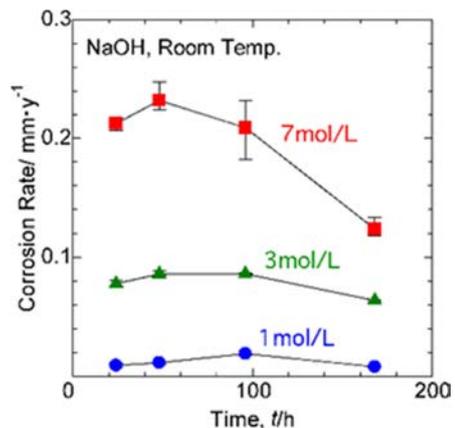


Fig.1 Corrosion rate of Ta in NaOH solution.

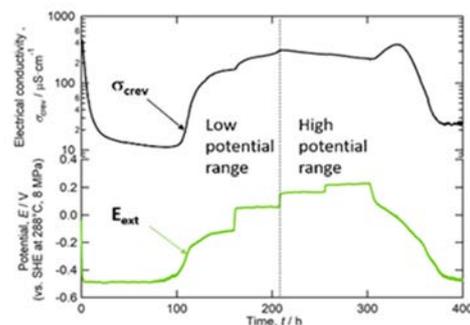


Fig.2 Change in electrical conductivity within the crevice and potential with time.

Reference

- 1) Y. Ishijima, *et al.*, *Mater. Trans.* 63(4), 538 (2022).
- 2) Y. Soma, *et al.*, *Corrosion* 78(6), 503 (2022).



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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 use, have been studied for the purpose of deepening the understanding on their radiation tolerance. In-situ observation is one of the most useful techniques for material irradiation studies. Especially, for modeling of the microstructural evolution of irradiated materials, systematic irradiation studies have been performed^{1),2)}. In addition, in order to apply for advanced nuclear systems, we are exploring not only a novel advanced material but also an innovative technology for material fabrication³⁾.

Point-defect migration investigated using in situ high-voltage electron microscopy

Modeling the microstructural evolution of irradiated materials requires knowledge of the migration energies of point defects, which are induced by the impact of energetic particle radiation. In our group, we have estimated the migration energies of interstitials and vacancies formed as point defect pairs by observing the evolution behavior of self-interstitial atom (SIA) clusters in electron-irradiated α -iron using an in-situ high-voltage electron microscopy. According to a rate-theory analysis, the interstitial or vacancy migration energy can be estimated from the temperature dependence of the peak cluster number density or the cluster growth rate, respectively. Figure 1 (a) and (b) show the estimated interstitial¹⁾ and vacancy²⁾ migration energies, respectively. The values are concordant with those calculated from the first-principles studies.

Ion irradiation study on additive manufactured low activation medium entropy alloys

Some concentrated solid solution alloys (CSAs) and medium entropy alloys (MEAs) have attractive properties such as a high radiation tolerance and corrosion resistance at elevated temperatures. These new alloys are, therefore, expected to be a promising candidate as structural materials for advanced nuclear systems. On the other hand, additive manufacturing (AM) is an emerging technology that can provide many benefits such as freedom of design, waste minimization and the ability to manufacture complex structures, and the active use of AM in the nuclear fields is now under consideration. In this study, we focused on MEAs

which were fabricated by AM in order to understand the effect of high dose ion irradiation on microstructural evolution. Figure 2 (a) and (b) show the cross-sectional TEM images after irradiation at 773 K up to over 300 dpa, indicating that small cavities with the size of less than 10 nm formed in both arc-melted and selective laser melted (SLMed) MEAs. By comparing this result with that of arc-melted and SLMed 316L SSs, the cavity formation was suppressed significantly.

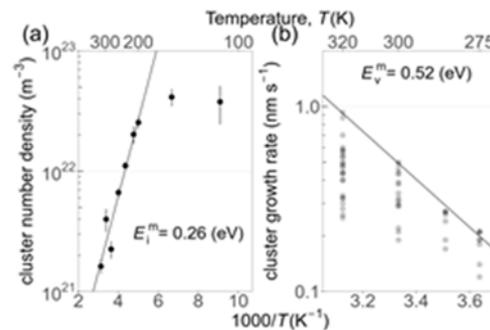


Fig.1 Estimation of migration energies of (a) interstitials (E_{im})¹⁾ and (b) vacancies (E_{vm})²⁾ in α -iron. Fitting conditions: (a) above 200 K, where interstitials are thermally mobile, and (b) the maximum growth rate of isolated SIA clusters at each temperature.

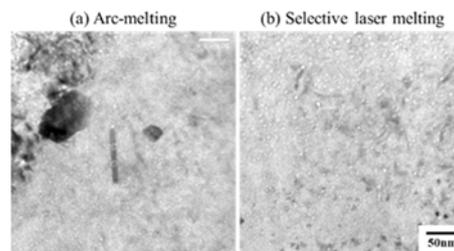


Fig.2 Cross-sectional TEM image of ion-irradiated MEAs fabricated by (a) arc-melting, (b) selective laser melting.

Reference

- 1) Y. Abe, *et al.*, *Philos. Magazine* 101(14), 1619 (2021).
- 2) Y. Abe, *et al.*, *Philos. Magazine* 102(12), 1173 (2022).
- 3) "Development of new reduced activation HEAs by additive manufacturing method", JFY2021 progress report.

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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 work on uranium and non-radioactive materials is carried out in the Research building No.4, with various heating devices and analytical apparatus. That on transuranium elements (TRU: Pu and minor actinides) is performed at TRU-HITEC, consisting of specially designed hot cells and 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 irradiation conditions.

Advanced Technique for Melting Temperature Measurement

For the safety analysis of fuel materials, melting point (T_m) is one of the essential properties. In case of the nitride fuel for minor actinide transmutation containing ZrN or TiN as inert matrices, its T_m is expected to be extra high (over 3,000 °C) and cannot be measured by the conventional method using induction furnace and tungsten crucible because of the mutual reaction between the crucible and nitride specimen. In addition, melting (liquefaction) of the nitrides is not congruent but decomposition into liquid metal saturated with nitrogen and N_2 gas. Therefore, the liquefaction temperature strongly depends on the nitrogen partial pressure of the system. Recent advanced technique for T_m measurement using laser spot heating and high-speed infrared spectrometer is a unique method that can solve the above problem. The measurement system was assembled in Research building No.4 for optimization of measurement conditions and analysis method.¹⁾ Schematic of the system and the appearance are shown in Fig.1. YAG laser shot forms a liquid phase in the limited area (a few mm in diameter) on the surface of disk-shaped specimen, and the radiation spectrum of this area is recorded at high speed. Semiconductor laser is used for preheating the specimen to avoid fracture by thermal shock.

The key technique of this method is to determine the emissivity and temperature simultaneously by analyzing the spectrum. The monochromatic radiation thermometer is used only to determine the timing of liquid phase formation (thermal arrest on the temperature curve). After optimization of the analysis, we successfully demonstrated the T_m measurement of tantalum and molybdenum, known as high T_m metals, within the uncertainty of several ten degrees Celsius.

Example of the temperature curve and the laser power pattern of a sintered $Er_{0.3}Zr_{0.7}N$ specimen is shown in Fig.2. The advantages of this method are very short measurement time (0.1 sec) and the small specimen size (3 mm in diameter), as well as no crucible. Now we apply this system to the lanthanide-zirconium nitride solid solutions to determine the liquefaction temperature as functions of composition and nitrogen partial pressure, aiming for application to the TRU nitride fuel in near future.

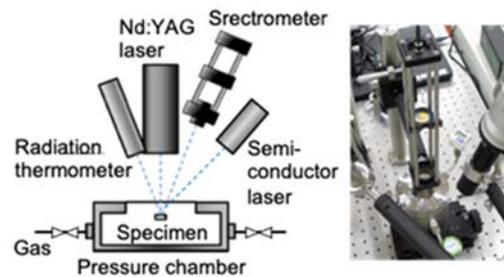


Fig.1 Schematic and appearance of the melting point measurement system.

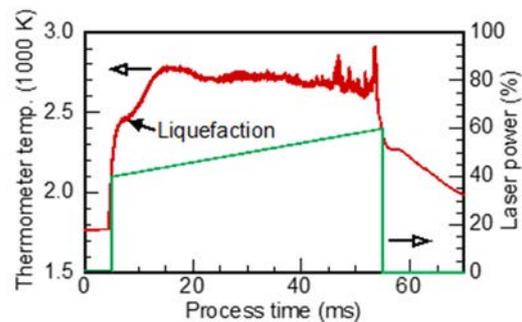
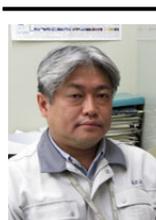


Fig.2 Example of temperature curve and laser power pattern as a function of time. Vertical axis does not show the actual specimen temperature.

Reference

- 1) T. Iwasa & T. Arima, *JAEA-Tech.* 2021-036 (2022) (in Japanese).



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Development Group for LWR Advanced technology

Our group has been conducting research programs to improve LWR safety by evaluating the rationality of regulation standards, safety margin estimation, accident management, etc., from a scientific point of view. Through this, we aim to advance the safety technology in LWR, and also contribute to acceleration of the decommissioning of Fukushima Dai-ichi Nuclear Power Station (1F). We mainly focus on fission product (FP) behaviors under severe accident (SA) conditions, and development of accident tolerant fuel (ATF) cladding.

Fission product behavior

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 owing to 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 models cannot accurately reproduce experimental results. Then, an improved model called ECUME model was constructed by clarifying effects of chemically affecting elements (i.e., silicon content in stainless steel and gaseous Cs concentration). Furthermore, ECUME (Effective Chemistry database of fission products Under Multiphase rEaction) model was incorporated into SA analysis code, SAMPSON (Severe Accident analysis code with Mechanistic, Parallelized Simulations Oriented towards Nuclear Fields), which is developed under the collaboration with the Institute of Applied Energy (IAE), and the experiment in the TeRRa (Test bench for FP Release and tRansport) facility with the temperature gradient tube to simulate SA conditions such as temperature decrease and aerosol formation was analyzed. Consequently, the SAMPSON with ECUME model successfully reproduced the experimental results (Fig.1). This result indicates that ECUME model and the analytical method such as models of aerosol formation and the calculation method of saturated CsOH vapor pressure can be applicable to the analysis of Cs chemisorption behavior. From these results, the amount of chemisorbed Cs can be estimated according to the SA analysis conditions.

Accident Tolerant Fuel (ATF)

For improvement of safety of the conventional LWRs, we are conducting fundamental studies on ATF cladding candidates to promote the developments in industries. Chromium (Cr)-coated zirconium (Zr) alloy cladding for PWR is one of the major ATF cladding concepts. In this research, bare cladding and Cr-

coated cladding with 10 micrometer thickness of Cr phase on its outer surface, were cut into 20 mm length and applied for the tests*. To investigate the oxidation behavior after cladding burst in the hypothetical loss of coolant accident (LOCA) condition, we conducted oxidation tests in high temperature (650-1,150 °C) steam flow using thermobalance.

Figure 2 shows an example of the mass gain (oxidation) profile during oxidation tests at 1,150 °C and cross section of the samples after 1,200 s of the tests. Oxidation on the Cr-coated outer surface was suppressed and macroscopic mass gain was halved compared to the bare sample.

This result indicates that Cr coating on the Zr alloy cladding outer surface may suppress the oxidation in the accidental condition, and would be beneficial to enhance the accident tolerance of the cladding in this temperature range, even after burst of the cladding.

*The samples were provided by Mitsubishi Nuclear Fuel Co., Ltd.

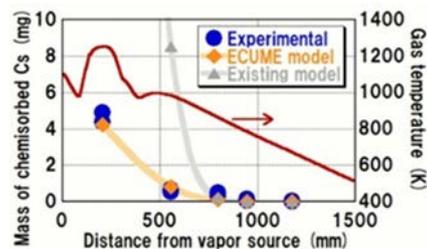


Fig.1 Comparison of experimental and calculated masses of chemisorbed Cs

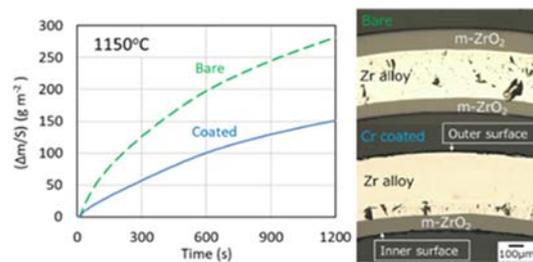


Fig.2 Oxidation profiles during the test, and cross section of the samples after the tests

Reference

- 1) H. Karasawa, *et al.*, 2021 Fall Meeting of AESJ 3E03 (2021) (in Japanese).
- 2) Y. Nemoto, *et al.*, 2022 Annual Meeting of AESJ, 2J05 (2022) (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. We chemically resolve various issues arisen from the nuclear industry by developing new technique of measurement and computational method. The following two topics are our remarkable achievements of 2021.

Solvent Extraction Mechanism of Lanthanide Ions

The transport mechanism of metal ions through the liquid–liquid oil/water interface in solvent extractions remains unclear. Since the transport of metal ions between the oil and aqueous phases is closely related to the extraction kinetics, the understanding of the transport mechanism of metal ions can contribute to the improvement of the solvent extraction process. In our recent study¹⁾, the adsorption process of trivalent lanthanide ions from the aqueous phase to the interface in the solvent extraction with the di-(2-ethylhexyl)phosphoric acid (HDEHP) extractant was investigated through a model interface: water surface covered with HDEHP (air/HDEHP/aqueous interface) by using vibrational sum frequency generation (VSFG) spectroscopy. The symmetric POO– stretch vibrational signals of HDEHP at the interface observed by VSFG spectroscopy and density functional theory (DFT) calculations demonstrated that 1:1 lanthanide-HDEHP complexes were formed at the air/HDEHP/aqueous interface. The formation of the interfacial complex could be an elementary chemical process occurring just before the transfer of lanthanide ions to the side of the organic phase (Fig.1).

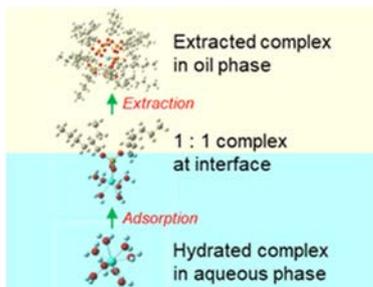


Fig.1 Proposed extraction mechanism of lanthanide ions by di-(2-ethylhexyl)phosphoric acid (HDEHP) extractant.

Dissolution of U_3O_8 in Bicarbonate Solution with H_2O_2 Addition

In the event of nuclear waste canister failure, the

release of radioisotopes is predicted to proceed via interaction of groundwater with spent fuel resulting in dissolution. The dissolution will depend on the nature of the spent fuel surface, water chemistry of the groundwater, and oxidants generated by radiolysis. In this study, the dissolution of an oxidised form of uranium, U_3O_8 , in bicarbonate solution upon H_2O_2 addition was investigated.

The dissolution of U was found to increase with bicarbonate concentration (Fig.2)²⁾. This was attributed to an increase in the oxidative decomposition of H_2O_2 at the U_3O_8 surface, where the oxidation reaction $U^{(IV)} \rightarrow U^{(VI)} + e^-$ is driven by the reduction of H_2O_2 . The oxidation reaction destabilises the U_3O_8 lattice leading to dissolution: $U_3O_8 + H_2O_2 \rightarrow 3UO_2^{2+}_{(aq)}$. As bicarbonate forms stable complexes with $U^{(VI)}$, increasing the bicarbonate concentration drives the dissolution of U_3O_8 .

The 2nd topic was performed as part of "Project on Research and Development of Spent Fuel Direct Disposal as an Alternative Disposal Option (2020FY)" under the contract with Ministry of Economy, Trade and Industry (METI) (Grant Number: JPJ007597).

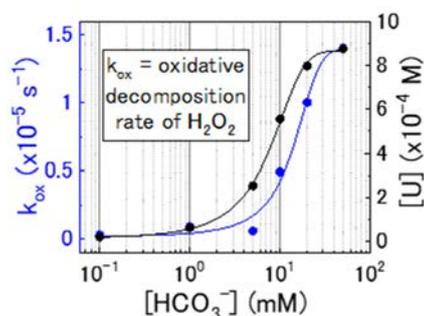


Fig.2 The relationship between the oxidative decomposition of H_2O_2 on U_3O_8 and the dissolution of U into bicarbonate solution.

* Reproduced from Ref.2 with permission from the Royal Society of Chemistry.

Reference

- 1) R. Kusaka, *et al.*, *J. Phys. Chem. B* 125(24), 6727 (2021).
- 2) J. McGrady, *et al.*, *RSC Adv.* 11(46), 28940 (2021).



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Research Group for Analytical Chemistry

Accurate analyses of radioactive substances are fundamental technologies supporting nuclear science. We are developing practical analytical methods for radionuclides based on studies of elucidation of chemical reactions and separation phenomena such as adsorption, precipitation, aggregation, etc. The separation techniques were also applied to the development of radioactive isotope isolation for alpha therapy. In the decommissioning of the TEPCO's Fukushima Daiichi Nuclear Power Station (1F), our micro-analysis technique was applied to elucidation of forms of particulate alpha emitter in a stagnant water in 1F.

Design of a Resin-packed Micro-Channel for Uranium Separation

To reduce the radiation exposure risk and production of secondary radioactive wastes during uranium (U) quantification, we are studying scaled-down separation method using a microchip. Microchannels were designed to densely pack with resins, and effect of channel shape was examined¹. An ideal separation performance could be obtained by arranging more than approx. 10 resins along the depth and width of the microchannels as shown in Fig.1. Then, the separation performance of the microchannel for the seawater standard added 431 ppb Cs-133 (simulated 1F retained water) was evaluated. The measured value of U in the seawater sample was in good agreement with the certified value. It indicates that the microchip column is sufficiently practical. The size of the separation site was scaled down to $1/5000$ compared to commonly used counterparts. The radiation exposure risk and secondary radioactive wastes can be reduced to 1/10 and 1/800, respectively, using a resin-packed microchannel.

Selective Pd Separation from Simulated Radioactive Liquid Waste by Precipitation using Xenon Lamp Irradiation

Inventory estimation of ^{107}Pd , which is one of the long-lived fission products in high-level radioactive liquid waste (HLLW) is indispensable for the long-term safety assessment of geological repository. We have developed a simple and one-step Pd separation technique based on photoreduction with Xe lamp irradiation for the determination of radioactive ^{107}Pd in highly radioactive samples². A simulated HLLW solution was used to evaluate the separation performance. The Pd precipitate was formed by Xe lamp irradiation and recovered by

centrifugation (Fig.2). The Pd recovery from the simulated HLLW solution reached up to 50 %, while 99.5 % of the other 13 elements were separated. The Pd precipitate could be separated from radioactive elements (Sr, Cs, and Ba) and the interfering elements for the ICP-MS determination of ^{107}Pd (Zr and Ru). These results show the applicability of the proposed separation technique to HLLW samples.

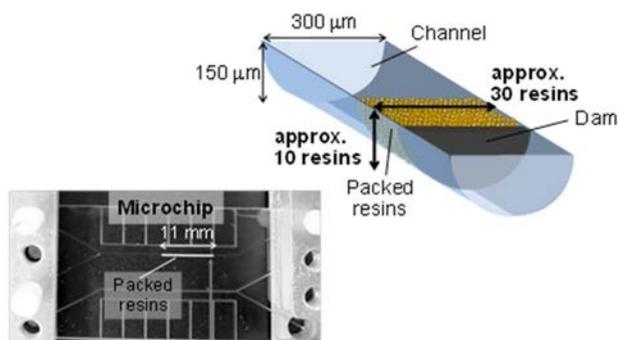


Fig.1 Diagrams of resin-packed microchannels with an ideal separation performance.

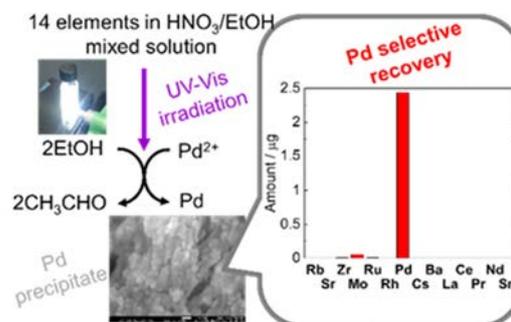


Fig.2 Formation of Pd precipitate by Xe lamp irradiation.

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- 1) K. Ouchi, *et al.*, *Anal. Sci.* 37(12), 1789 (2021).
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Research Group for Environmental Sciences

We research the dynamics of radionuclide transfer in the atmospheric, terrestrial, and oceanic environments to improve assessment technology of environmental impact of radionuclides. Computer models are developed and validated to predict radionuclide transfer in the environments. In particular, we develop the atmospheric prediction systems (WSPEEDI-II and WSPEEDI-DB) and an oceanic prediction system (STEAMER). We also conduct field survey to obtain environmental data, and analyze the data with advanced technology. The data is utilized to understand the dynamics of radionuclide transfer and to validate computer models.

Simulation on the dispersion and sedimentation of radionuclides in the ocean off Fukushima

We developed a sophisticated oceanic dispersion model considering interactions of radionuclides in seawater with multiscale suspended particles and seabed sediments¹⁾.

Oceanic dispersion hindcast was conducted by adopting a double-nested downscaling configuration for high resolution calculation in the coastal ocean²⁾ to validate the model's accuracies and analyze the ^{137}Cs transfer caused by the TEPCO's Fukushima Daiichi Nuclear Power Plant (FNPP1) accident. The direct release of ^{137}Cs into the ocean was imposed in the model based on ^{137}Cs measurement data in the seawater. In addition, the atmospheric ^{137}Cs deposition was also incorporated into the sea surface, which was calculated by WSPEEDI-II. Three size classes (sand, silt, and clay) were considered as suspended particles and seabed sediments.

Comparison between model and observation demonstrated that oceanic structure and the ^{137}Cs concentrations in seawater and seabed sediments were appropriately reproduced in the coastal ocean off Fukushima. The study suggested that ^{137}Cs was mainly distributed along the coastal bottom bathymetry (Fig.1). Shear stress effecting the resuspension of the seabed sediments was mainly generated by waves in the nearshore area and by ocean currents in the offshore area. The ^{137}Cs distribution in the seabed sediment off Fukushima was mainly formed due to adsorption from the dissolved phase over several months since the accident when the direct release of ^{137}Cs was remarkable. On the other hand, ^{137}Cs transfer from the suspended phase resulted in a small effect on the distribution in the seabed sediment of ^{137}Cs one

year after the accident. In terms of the ^{137}Cs adsorption on the seabed sediments, there was a difference among particle size classes, where clay was the most effective in adsorption. The study also suggested that the seabed sediments were a probable source releasing ^{137}Cs into the seawater after several months since the accident.

We will further improve the oceanic dispersion model developed in this study. We should continue the environmental assessment of radionuclides caused by the accident by utilizing this model in the future.

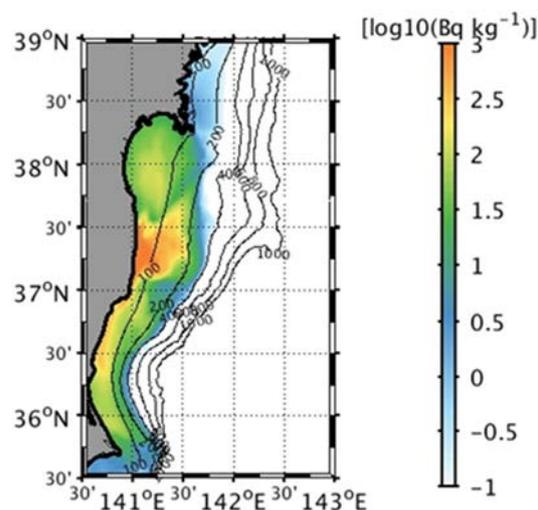


Fig.1 Simulated annual mean ^{137}Cs concentration in the seabed sediments one year after the FNPP1 accident. Dotted lines indicate the depth of bottom bathymetry.

Reference

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- 2) Y. Kamidaira, *et al.*, *J. Geophys. Res.: Oceans* 123(4), 2808 (2018).



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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), and applied it to industrial, scientific, and medical studies. PHITS has been upgraded to improve its higher reliability and to extend its area to be applied. In addition, we also conduct studies on radiological protection and radiation effects. This paper presents our progresses in FY2021: upgrades of PHITS and the development of a radiation dose estimation system for public exposure.

Upgrades of the PHITS Code

The major upgraded features are as follows in the newest version (v 3.27) that was released in FY 2021. Please check the PHITS homepage¹⁾.

Generalized track structure mode

ITSART (Ion Track Structure calculation model for Arbitrary Radiation and Targets) has been implemented to simulate nano-scale transport of protons and arbitrary ions in arbitrary matter.

Treatment of nuclear data

We upgraded the treatment of nuclear data in the newest version of PHITS.

- The memory for containing nuclear data is changed from static to dynamic one so that PHITS can read extremely large nuclear data without increasing a parameter for total memory usage of PHITS.
- Functions to read nuclear data libraries in the A Compact Evaluated nuclear data file format (ACE format) are implemented for deuteron, alpha particle, and photon. Here, the data library for deuteron, JENDL/DEU-2020 is contained in PHITS. Deuteron will be applied to intensive fast neutron sources using deuteron accelerators.
- A new function is introduced to automatically determine the maximum energy of nuclear data library for each nucleus from an address file. The users do not have to define the maximum energy for each nucleus and can easily use high energy nuclear data libraries, such as JENDL-4.0/HE.
- A new function (named as 'Exfor2frag') is introduced to convert the experimental cross section data in the EXFOR (Experimental Nuclear Reaction Data) to a data file in PHITS.

The users can exactly reproduce the data of the EXFOR in radiation transport analyses by PHITS.

RT-PHITS

(User support function - β -version -)

RT-PHITS (RadioTherapy package based on PHITS) has been developed under the support of the National Institutes for Quantum Science and Technology (QST). DICOM (Digital Imaging and Communications in Medicine) data, such as PET-Image and CT-Image, can be converted to an input file for the dose calculation by RT-PHITS. We have newly introduced a β -version of Python Tkinter GUI. The users can interactively define a condition of radiation dose analysis through the GUI, as depicted in Fig.1.

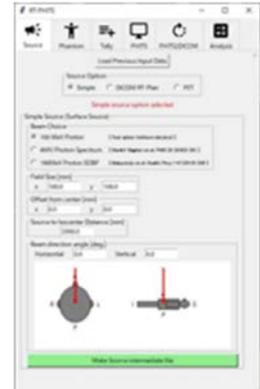


Fig.1 Interactive setting through GUI in RT-PHITS

Radiation Dose Estimation System for Public Exposure

In FY 2020, we analyzed the impact of body sizes on organ doses due to external neutron irradiation by human models. After that, a radiation dose database was prepared by the analyses. In FY 2021, radiation dose estimation system was developed based upon the radiation dose database for public exposure. Effective dose can be rapidly obtained for neutron (1×10^{-9} MeV to 1×10^4 MeV) and photon (1×10^{-2} MeV to 2×10^1 MeV) exposures by taking account for physique of adults, ages (6 age groups) of infants or children and an irradiation geometry.

Reference

- 1) PHITS homepage, <https://phits.jaea.go.jp/> (accessed 2022-11-08).



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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 with different radiotoxicities and chemical properties, and thus it is reasonable to separate the elements into groups. To this end, our research group has been developing several extractants and 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. A conceptual flow sheet of SELECT is shown in Fig.1. This process consists of the following four steps:

- recovery of U and Pu from dissolution solution of spent nuclear fuels by monoamides,
- recovery of Am, Cm, and rare earth elements (REs) from HLW by tetradodecyl diglycolamide (TDdDGA),
- separation of Am and Cm from REs by hexaoctyl nitrilotriacetamide (HONTA), and
- separation of Am and Cm by alkyl diamide amine (ADAAM).

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.

Studies on a Novel Extractant, TDdPTDA

Although the former studies showed the applicability of the extractants used in the current SELECT process, we have been developing novel extractants for further improvement in the process. We focused on phenanthroline diamides (PTDAs) as alternative extractants to HONTA for the separation of REs from Am and Cm, and single-stage batch experiments were carried out using TDdPTDA (Fig.2) which is one of PTDAs^{1,2}.

According to the time dependencies of Am, Cm, and Eu extraction, the extraction reaction reached equilibrium within 30 seconds indicating that the extraction rate of TDdPTDA exhibited was fast enough for practical use rather than that of HONTA, and this leads to improvement of the separation process. Effects of diluents such as 3-nitrobenzotrifluoride (F-3), nitrobenzene, and n-dodecane on the extraction behavior of Am, Cm, and lanthanides were evaluated with various concentrations of TDdPTDA and nitric acid. The results indicated that the structure of metal-

TDdPTDA complex depended on the polarity of the diluents. Furthermore, TDdPTDA showed sufficient separation performance not only in nitrobenzene and F-3 but also in n-dodecane. This property also supports the practicality of TDdPTDA in that commercially operating reprocessing plants have used aliphatic hydrocarbons such as n-dodecane. The studies on PTDAs are ongoing, and we will synthesize novel PTDAs with different side chains, investigate the extraction properties, and evaluate the separation performance to enhance the SELECT process.

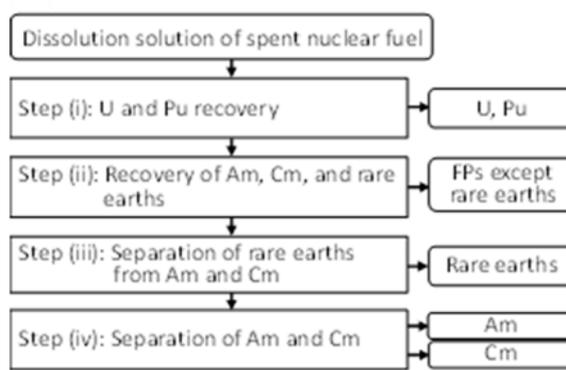


Fig.1 Conceptual flow sheet of SELECT process

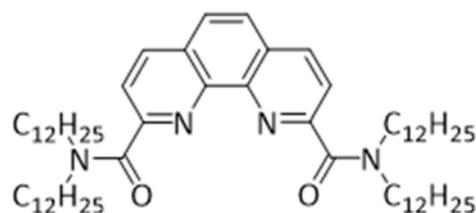
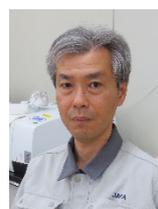


Fig.2 Structure of N,N,N',N'-tetradodecyl-1,10-phenanthroline-2,9-dicarboxamide (TDdPTDA)

Reference

- 1) N. Tsutsui, *et al.*, *Anal. Sci.* 36(2), 241 (2020).
- 2) N. Tsutsui, *Doctoral Dissertations (Tokyo Institute of Tech.)* (2022).



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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 embodying the upper radiation shield design and the subcriticality monitoring system.

Radiation shielding design

Since the ADS core is connected with beam transport equipment via a beam duct and backscattering of high-energy particles becomes a significant issue, the design of ADS upper structure requires a detailed assessment of radiation doses around the beam duct and radioactivity of the equipment for radiation protection. We have analyzed them using the Monte Carlo transport code system PHITS and proposed the upper structure of ADS¹⁾. Figure 1 illustrates the proposed structure with the upper shield to reduce backscattered neutron, the neutron dump to receive high energy neutron and the additional shield to mitigate neutron and gamma ray in upper area. This structure enables to reduce the radiation dose to sufficiently low level at the site boundary of ADS.

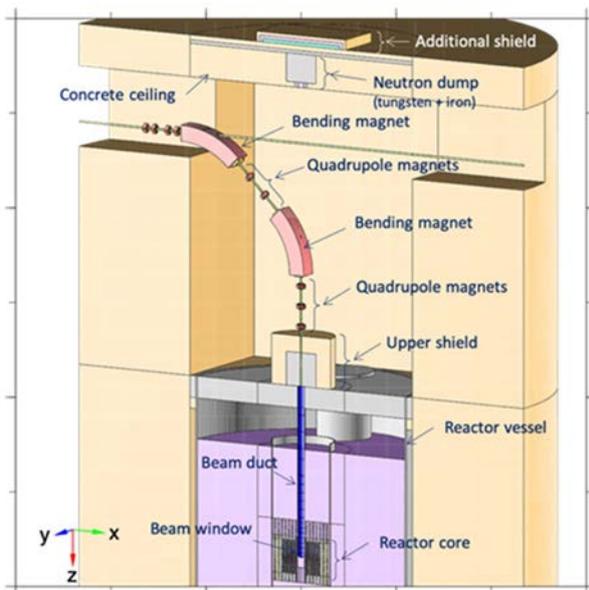


Fig.1 Proposed design of ADS upper structure

Subcriticality monitoring

We have proposed a safe and efficient methodology to make the ADS approach a target subcriticality during fuel loading²⁾. In this methodology, we need to perform the pulse neutron source (PNS) measurement for the absolute value of the subcriticality (Fig.2) at the initial state to give a calibration point for subsequent subcriticality monitoring.

As a feasibility study, we have explored practical parameters of the accelerator with 1.5 GeV protons for the PNS measurement. Considering uncertainties derived from the dead-time, the statistical error and the practical measurement time (<10 h), we have deduced accelerator parameters with which the measurement uncertainty satisfies a given upper error bound (=2%): 200 Hz of pulse repetition with 10 μs of pulse width and 3 mA of peak current.

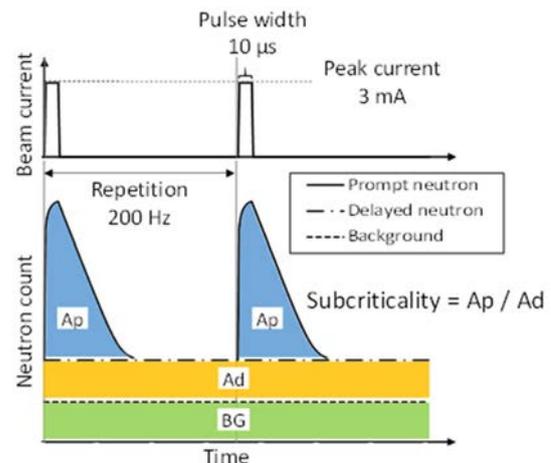


Fig.2 Schematic view of PNS measurement

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- 1) H. Iwamoto, et al., *JAEA-Res.* 2021-012 (2022) (in Japanese).
- 2) T. Okamura, K. Nishihara, et al., *JAEA-Data/Code* 2021-016 (2022) (in Japanese).



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Research Group for MA Transmutation Fuel Cycle

Since the ratio of minor actinides (MAs) which can be transmuted in the accelerator-driven system (ADS) core is limited, residual MAs in the spent transmutation fuels should be separated and reused to transmute MAs effectively. Therefore, we have been carrying out research and development of the technology for reprocessing MA transmutation fuels. Regarding U-free inert matrix MA nitride fuels, studies on pyrochemical reprocessing and nitridation of recovered transuranium elements (TRU: MAs and Pu) have been carried out.

Fission product (FP) compounds in the spent MA nitride fuels

Recovery yield of MAs should be high to minimize the amounts of MAs in the radioactive waste which is discharged by reprocessing MA transmutation fuels¹⁾. Target recovery yield is 99.9 % to transmute 99 % of MAs received into the transmutation fuel cycle¹⁾. To develop the technology for reprocessing the spent fuels, stability of the FP compounds formed in the fuels should be considered.

In UN and (U,Pu)N fuels, platinum group metals (PGMs: Ru, Rh, Pd) tend to precipitate out as the intermetallic compounds such as (U,Pu)(Ru,Rh,Pd)₃²⁾. Molten salt electrorefining experiments using burnup-simulated UN samples showed that thermodynamically stable UPd₃ was not dissolved at the anode³⁾. To treat such anode residues, we have proposed a chlorination method using CdCl₂. This method was proved to be effective for surrogates of TRUPd₃ (e.g., GdPd₃) and NpPd₃⁴⁾. Solid solution type ZrN-based nitride fuel, (TRU,Zr)N has been chosen as the first candidate for MA transmutation fuel for ADS in JAEA¹⁾. But even the chemical compounds formed by the reaction of (TRU,Zr)N with PGMs have not been studied. Therefore, chemical reaction behavior of (Gd,Zr)N with PGMs have been investigated regarding Gd as a surrogate of TRU. The mixture of Gd_{0.25}Zr_{0.75}N and Pd (molar ratio 4:3.1) heated at 1,323 K in Ar gas atmosphere was analyzed⁵⁾. The X-ray diffraction (XRD) profile showed the existence of a phase which was isostructural to cubic GdPd₃ and its lattice constant was smaller than that of GdPd₃ (Fig.1)⁵⁾. The difference should be due to the substitution of Zr atoms for Gd atoms to form (Gd,Zr)Pd₃. Figure 2 shows a SEM image of the sample; (Gd,Zr)Pd₃ phase was identified by EDS analyses as well⁵⁾. Based on the results, we presume that compounds like (TRU,Zr)Pd₃ can be

formed in ZrN-based MA transmutation nitride fuels. In our future work, stability of such intermetallic compounds will be clarified to develop the technology for reprocessing the MA transmutation nitride fuels.

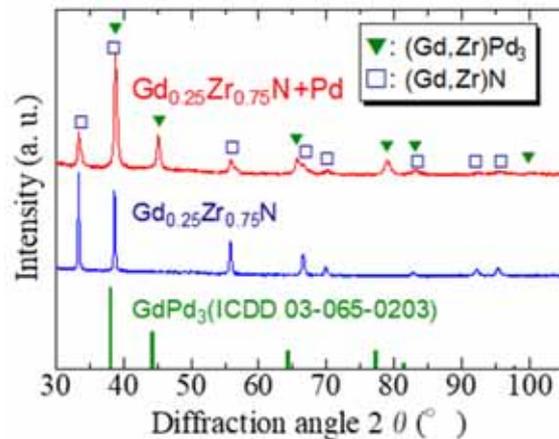


Fig.1 XRD profiles of the samples

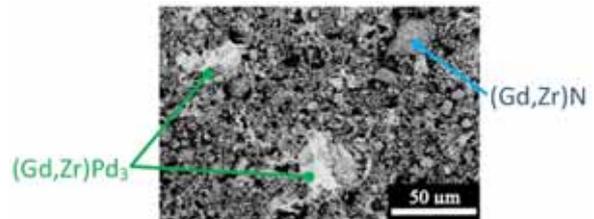


Fig.2 SEM image of Gd_{0.25}Zr_{0.75}N+Pd sample after the reaction

Reference

- 1) H. Tateno, T. Sato, *et al.*, *J. Nucl. Sci. Tech.* 57(3), 224 (2020).
- 2) Y. Arai, "3.02 Nitride Fuel" in *Comprehensive Nucl. Mater. Vol.3* (Elsevier), 41 (2012).
- 3) T. Satoh, *et al.*, *J. Nucl. Sci. Tech.* 46(6), 557 (2009).
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- 5) R. Mishima, *et al.*, *2022 Annual Meeting of AESJ 2I11* (2022) (*in Japanese*).



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Publication list

* Works with underlined numbers were published before FY2021 (not included in the lists of previous NSEC reports).

Nuclear and LWR Engineering Division

Nuclear Data Center

Papers

- 1) β -delayed neutron-emission and fission calculations within relativistic quasiparticle random-phase approximation and a statistical model, F. Minato, T. Marketin & N. Paar, *Phys. Rev. C* 104(4), 044321 (2021).
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Research Group for Reactor Physics and Standard Nuclear Code System

Papers

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- 2) Adaptive setting of background cross sections for generation of effective multi-group cross sections in FRENDY nuclear data processing code, A. Yamamoto, T. Endo & K. Tada, *J. Nucl. Sci. Tech.* 58(12), 1343 (2021).
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- 4) Benchmark study of particle and heavy-ion transport code system using shielding integral benchmark archive and database for accelerator-shielding experiments, Y. Iwamoto, S. Hashimoto, T. Sato, N. Matsuda, S. Kunieda, Y. Çelik, N. Furutachi & K. Niita, *J. Nucl. Sci. Tech.* 59(5), 665 (2022).
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