RNC's initiatives in the ImPACT program

The ImPACT Program

Since 2007, Radioactive Isotope Beam Factory (RIBF)[1] has been one of the world-leading heavy-ion accelerator facilities for low-energy nuclear physics. RIBF has contributed to the expansion of the nuclear chart by producing more than 150 new isotopes and has provided a multitude of opportunities to discover new phenomena originating from high isospin asymmetry. In parallel to such pure nuclear physics activities, a new and national program of nuclear engineering was launched for social benefits. In the period of 2014–2018, the program entitled "Reduction and Resource Recycling of High-level Radioactive Wastes through Nuclear Transmutation"[2] was carried out as one of the ImPACT R&D programs conducted by the Cabinet of Japan.

Since Hideki Yukawa left the Japan Atomic Energy Commission in 1957, the nuclear physics and nuclear engineering communities have been separated in Japan. The incident of the Fukushima Dai-ichi Unit in 2011, however, impelled Japanese nuclear physicists to seriously consider possible contributions to nuclear engineering. In 2014, a demonstration experiment was performed with RIBF. In this experiment, the inverse kinematics with energetic radioactive-isotope (RI) beams of ^{137}Cs and ^{90}Sr was employed to study spallation reaction to find an efficient reaction pathway for reduction of the radioactivity.[3] The experiment provided an opportunity to the physicists and engineers to work together. The potentials of RI beams at RIBF were also demonstrated, and the fundamental reaction data obtained with RI beams were found highly useful for designing an accelerator-based transmutation system.

High-level radioactive waste has two main components: long-lived fission products (LLFPs) and minor actinides (MAs). The transmutation of MAs has been studied in detail for the purpose of reprocessing spent nuclear fuel under the concept of using fast-breeder reactors or accelerator-driven systems. The LLFP nuclides are supposed to create risks in the geological disposal strategy because their half-lives are much longer than those of MAs. However, the transmutation of LLFPs has not been studied extensively. Considering the present situation of R&D activities in the field of the radioactive waste, the ImPACT program was jointly proposed by the nuclear physicists and engineers, to focus on accelerator-based transmutation of LLFPs.

The ImPACT program emphasized on the "resource recycle" of radioactive waste. Thus, ^{107}Pd and ^{93}Zr were highly prioritized as objectives. The program consisted of five projects as follows:

Project-1: Development of separation and recovery technologies,

Project-2: Production of nuclear reaction data and new nuclear reaction control methods,

Project-3: Reaction theory modeling and simulation,

Project-4: Evaluation of the nuclear transmutation system and development of elemental technologies, and

Project-5: Process design concept by nuclear engineers.

All the projects were managed coherently, and Project-2 and Project-4 were conducted by RIKEN Nishina Center.

An overview of the activities in Project-2 and -4 is presented in this article.

Transmutation Reactions for Accelerator System

To design an accelerator system for the transmutation, the possible reactions for the transmutation were intensively discussed at an early stage of the ImPACT program.

First, we adopted nuclear reactions, not electromagnetic reactions. In the case of nuclear reactions induced by "hadron" particles, which interact strongly with LLFP nuclei, several reactions, such as spallation, knockout reactions, are applied with reaction cross sections of the order of 1 barn. Conversely, in the case of electromagnetic reactions, the reaction candidate is only the photoabsorption process (γ, n), and its cross section is approximately 0.2 barns for LLFPs. To induce the (γ, n) reaction, secondary high-energy photons, of which energy is more than 10 MeV, should be produced by high-energy electron beams through the bremsstrahlung process in a converter material or through the inverse Compton process with intense laser lights. However, the high-energy photons are wasted to create the electron-positron pairs, simply because the cross section of the pair creation is 10 barns, much higher than that of the photoabsorption. Thus, most of the electron-beam energy is wasted in creating the electron-positron pairs, not in transmuting LLFP nuclides significantly.

Next, several types of nuclear reactions were considered for transmutation and were categorized with respect to transmutation throughput and reaction controllability. The candidate reactions are spallation reaction, neutron knockout reaction, such as $(n, 2n)$, and muon capture reaction. The spallation reaction induced by high-energy proton- or deuteron-beams is very promising in terms of throughput, but it is not excellent in terms of controllability. When the spallation reaction is applied, the beams are irradiated directly to LLFP materials, and the target thickness is determined from the stopping range of the beams. Higher energy beams lead to thicker targets, and hence, the throughput is increased as a function of beam energy. However, according to the results obtained in Project-2, higher beam energy produces more nuclides as spallation products, because a higher energy beam deposits higher energy in the target nuclei, and the higher deposited energy in the nuclei is used to emit more protons and more neutrons from the nuclei. Thus, in the case of the spallation reaction, the optimal beam-energy should be carefully considered in terms of not only the throughput but also the costs necessary to treat radioactive nuclides produced in LLFP materials after long-term irradiation.

The neutron knockout reaction is induced by high-energy neutrons with energy more than 8 MeV. In the case of even-odd separated LLFP, especially for the Pd nuclides, the neutron knockout reaction has very high reaction controllability. After the even-odd separation, ^{105}Pd and ^{107}Pd are left as nuclear waste (^{105}Pd is a stable nuclide, and ^{107}Pd is a long-lived one). The $(n, 2n)$ reaction produces ^{104}Pd and ^{106}Pd which are both stable, and $(n, 3n)$ reaction also could be applied to produce ^{103}Pd and ^{105}Pd. The ^{103}Pd nuclide is not stable, but its half-life is as short as approximately 17 days. The neutron energy range appropriate for $(n, 2n)$ is 8–18 MeV, and it gives the cross section of approximately 2 barns. Higher energy neutron beams also induce $(n, 3n)$. The high energy neutron beams could be created by high energy deuteron beams via breakup reaction. The broad energy range of neutrons is very useful because this leads to the generation of high flux neutron for transmutation. The target thickness for the neutron knockout reaction is determined from the mean-free path of the reaction, which is of the order of 100 g/cm^2 in Pd metal.

The other reaction candidate is the negative muon capture reaction. The negative muon capture process also has high reaction controllability. A negative muon is a unique particle, which interacts electromagnetically and can form a muonic atom where, like an electron, the negative muon is trapped by a nucleus. Then, the muon is captured by a proton inside the nucleus by weak interaction, and the proton is converted to a neutron. The capture probability per muon is almost 100% for $Z > 40$ nuclides, when the negative muons are implanted to the material with $Z > 40$. Because s muon is approximately 100 times heavier than an electron, the energy transfer in the process is rather high, and a few neutrons are emitted from the nucleus. Hence, the muon capture has reaction controllability for the proton number and decrements the number by one. This feature of the muon capture is very useful for chemical separation. In the case of Pd with atomic number of 46, the products are only Rh with atomic number of 45. Although the negative muon is very promising in terms of controllability, the production of a muon is rather energy consuming; the production of one muon needs 5 GeV. A negative muon is created through decay of a negative pion, and the pion is created by a high-energy nuclear reaction, for example, proton induced nucleus collision, in which the proton energy should be higher than 300 MeV. The pion production cross section highly depends on the energy of beams and isospin of beams. Accelerator facilities for muons, such as TRIUMF, PSI, RAL, and J-PARC, deliver high-energy and intense proton beams with energy of more than 500 MeV, and the target thickness is limited to avoid pion capture in the target. These facilities are not optimized for negative muon productions. Instead of proton beams, deuteron beams can produce four times more negative muons than proton beams because of the isospin effects.

Negative muons are also useful to produce 14 MeV neutrons via muon catalyzed fusion of $d + t$. In the process, one muon can create approximately 100 14 MeV neutrons. Thus, the energy cost for a 14 MeV neutron is approximately 50 MeV. This value is smaller than that in case of an accelerated deuteron beam inducing $d + t$ fusion, which is approximately 2 GeV.

Regarding the neutron capture process, it is noted that neutron capture cross section for ^{107}Pd is higher than 1 barn at a neutron energy below 100 keV and 10 barns at 10 eV to 1 keV. Thus, neutrons produced via spallation reaction are also very useful for transmutation. In the case of the even-odd separation scheme for the Pd nuclides, the neutron capture changes ^{105}Pd and ^{107}Pd to ^{106}Pd and ^{108}Pd, respectively, which are both stable nuclides. Thus, a combination of the knockout reaction and neutron capture could lead to high throughput for transmutation. Considering all the above, we have decided to design an accelerator system for deuteron beams. The deuteron beams are utilized for a variety of applications: spallation reaction in direct irradiation as well as spallation neutron production, high energy neutron production via the deuteron breakup reaction, and efficient production of negative muons.

Nuclear Reaction Data with RIBF

Project-2 aimed to produce reaction data of the LLFPs that could be evaluated and utilized as input for a simulation code as part of Project-3. As introduced in the previous section, all the possible reactions for transmutation were discussed: proton-, deuteron-, and neutron-induced reactions and muon capture reactions.

In the case of direct irradiation of charged-particles into an LLFP material, an energy-dependence study of the reaction is quite important because of the stopping power in the material. This argument is also applied for the high-energy neutron-induced spallation reaction. A challenge for the energy-dependence study is to build a bridge over the Fermi energy; at a lower energy, the collective natures of the reaction are dominant and at a higher energy, the nucleon-based natures become dominant.

According to the experiences in the previous experiment of ^{137}Cs and ^{90}Sr,[3] the reaction study for proton-, deuteron-, and neutron-induced reactions was conducted in inverse kinematics at RIBF under a large domestic collaboration comprising the University of Tokyo, Tokyo Institute of Technology, Kyushu University, Miyazaki University, Niigata University, and RIKEN. Compared with the traditional activation method via normal kinematics

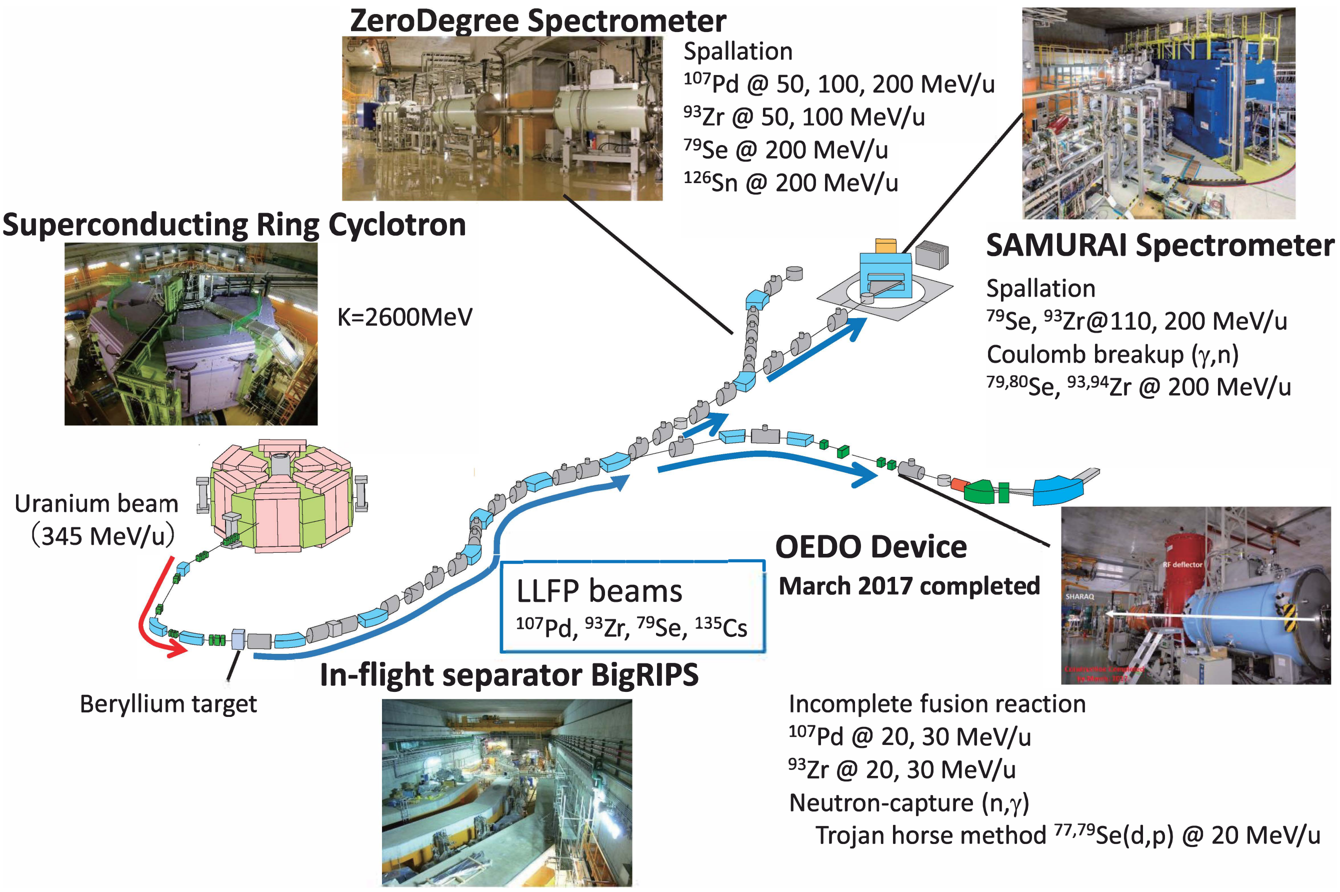

Fig. 1. Reaction study for long-lived fission products at RIBF in the ImPACT program. Major spectrometers such as, ZeroDegree, SAMURAI, and SHARAQ, were utilized.[4)]

with stable beams and RI targets, the inverse kinematics technique gives clear particle identification for reaction products and easy control of RI beam energies for the study of energy-dependence. In addition, we do not have to prepare RI targets but stable-isotope targets, such as protons and deuterons. All these advantages of inverse kinematics result in reaction data of excellent quality at RIBF.

As shown in Fig. 1, intense beams of LLFPs, such as ^{107}Pd and ^{93}Zr were produced at the in-flight separator "BigRIPS" via fission reactions with a uranium beam accelerated at Superconducting Ring Cyclotron (SRC), and then were delivered to three spectrometers (ZeroDegree, SAMURAI, and SHARAQ), which were utilized to identify and analyze the reaction products.[5–9)] ZeroDegree is suitable for inclusive measurements with relatively heavy fragments. SAMURAI has a wide acceptance in both momentum and scattering angle for reaction products. Hence, semi-exclusive measurements were performed to detect reaction products as well as neutrons in the projectile frame. At the SHARAQ spectrometer, CNS, the University of Tokyo and RIKEN worked together to develop an efficient deceleration scheme for RI beams. A new device "OEDO"[10)] was installed in the SHARAQ beamline to control emittance growth by transverse RF electric fields. An operation mode of BigRIPS was developed to realize energy-decelerated beams.[11,12)] The combination of OEDO and energy-decelerated beams successfully led to a reaction study at an energy of 20 MeV/nucleon.

Enhancement of element production by incomplete fusion reaction with weakly bound deuteron was observed at 50 MeV/nucleon,[9)] as shown in Fig. 2, and the incident energy was much higher than the energy scale of complete fusion reaction. The reaction mechanism could be applied to produce a higher Z element by increasing the atomic number by 2 via heavy loosely bound nuclei such as ^{9}Be and ^{6}He. Another highlight is a deduction of neutron capture reaction via the Trojan horse method with a low energy RI beam at 20 MeV/nucleon.[13)] In future, the new method can be widely applied not only for nuclear engineering but also for nuclear astrophysics.

All these data stimulated theoretical works, and the JAEA nuclear data group developed a new data library "JENDLE/ImPACT–2018" for proton- and neutron-induced reactions up to 200 MeV/nucleon.[14)] The new library has been built in as a package of PHITS simulation code.[15)]

A test of ^{107}Pd transmutation with macroscopic quantities was conducted with normal kinematics by utilizing a low energy deuteron beam at the AVF cyclotron at RIBF.[16] Almost 100% enriched ^{107}Pd was prepared as a target and irradiated for several days. After the irradiation, the reaction products were identified from gamma-ray and ICP-MS measurements. The production yields were found consistent with PHITS predictions.[15]

Accelerator System

Project-4 proposes an accelerator-based transmutation system for LLFPs, and focuses on three objectives: (1) to design an accelerator system with a possible transmutation scenario, (2) to develop essential technologies for a high-power accelerator system, and (3) to encourage ideas for new high-power and energy-saving accelerators. Four activities by the RIKEN Nishina Center are selected and briefly introduced.

Linear accelerator system ImPACT2017

Delivering high-energy and intense deuteron beams is essentially important for the nuclear transmutation as described in the second section. The next question is the intensity of deuteron beams. In the existing facilities in operation, the maximum intensity of proton beams is of the order of 1 mA. As discussed in the ImPACT project, the beam intensity necessary for the transmutation is at least 1 A, which is three orders of magnitude higher than that of the present facilities. It should be noted that 1 A corresponds to approximately 1 mol/day. If all the beams contribute to the transmutation, the throughput would be approximately 100 g/day for the mass number of 100, which corresponds to 36 kg/year.

To achieve a 1-A deuteron beam, we have made a conceptual design for a linear accelerator system called "ImPACT2017,"[17] as shown in Fig. 3. There are a few key aspects of ImPACT2017. First, the system does not start with a radio-frequency quadrupole (RFQ). Second, the RF cavity of the linear accelerator is made as a single cell. The RFQ device is convenient and widely employed because a single RFQ device has three functions: beam bunching, acceleration, and focusing. However, a small beam aperture of RFQ (∼1 cm) limits the beam current due to the space charge effect. Thus, we must develop other types of acceleration scheme, which is capable for a large size beam. To accelerate a high-intensity beam, we must supply high RF power to the cavities. A standard configuration has several cells in one cavity, and the RF power is supplied to the cavity through RF couplers. Each RF coupler has a limitation for transmitted power; hence, many RF couplers should be attached to the cavity. In this case, a multi-cell configuration may cause problems in operation. Decrease the number of RF couplers is significant for reasonable operation of the linear accelerator, and we have decided to employ a single cell configuration for the cavity. Further details of the new idea are found in Ref. 17).

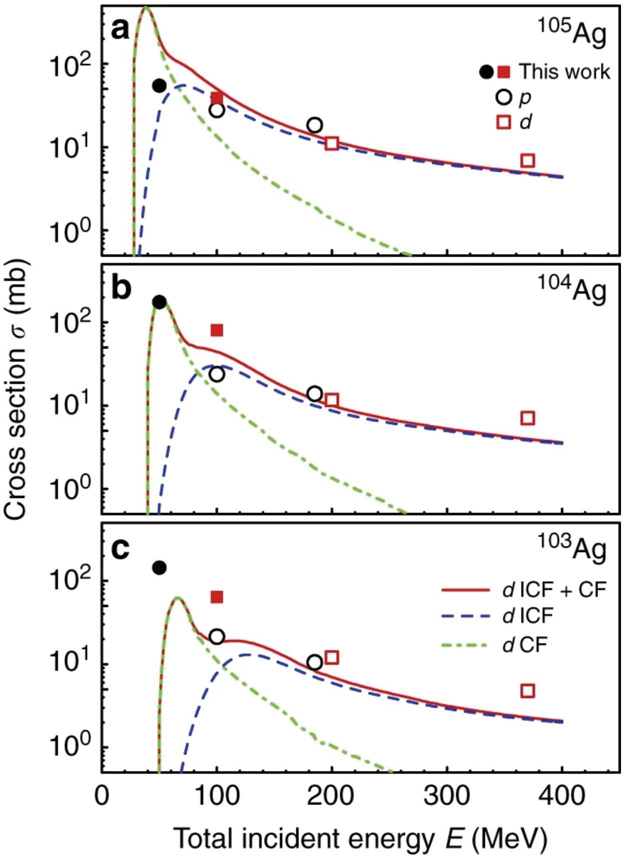

Fig. 2. Proton-induced and deuteron-induced cross sections for the ^{103,104,105}Ag isotopes produced by ^{107}Pd as a function of total incident energy, taken from Ref. 9). Cross sections on proton and deuteron are displayed by circles and squares, respectively. Enhancement of the ^{103,104}Ag cross sections with deuterons is clearly observed at the incident energy of 100 MeV, *i.e.*, 50 MeV/nucleon for deuterons.

R&D of key technologies for high-power accelerator system

As essential components for high power accelerator systems, a superconducting RF cavity for low-velocity charged-particles, material-free window-system for vacuum sealing, and liquid-target system for production targets have been developed by the RIKEN accelerator group.

R&D of a quarter-wave superconducting RF-cavity for low-velocity charged-particles[18] is essential to achieve low power consumption and space-saving. A prototype of the cavity was designed, constructed, and tested. The cavity was successfully cooled down to 4 K with liquid He, and acceptably high values of Q ($\sim 10^9$) and acceleration electric field (up to 9 MV/m) were achieved. Based on the experiences in this R&D process, the linear accelerator RILAC has been recently upgraded by installing a similar type of superconducting RF-cavities.[19]

A plasma-window with a diameter of up to 20 mm was developed as the material-free window system for vacuum sealing. The achieved pressure and the V-I characteristics as a function of diameter were investigated.

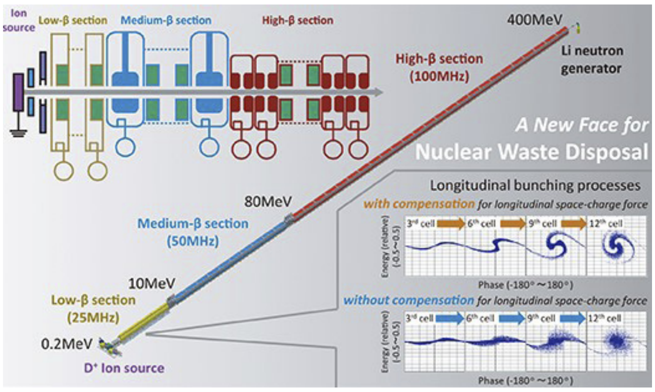

Fig. 3. Schematic of the 1-A Linac, taken from the cover page of Proc. Jpn. Acad. Ser. B **95**(7).17)

Detailed information can be found in Ref. 20).

The new idea of a target accepting the high intensity beam was proposed and published as patent.21)

Summary and Future Perspectives

The ImPACT program ran from 2014 to 2018 as the first step in the nuclear transmutation of LLFPs and successfully stimulated the interest of the nuclear chemists, nuclear physicists, accelerator physicists, and nuclear engineers in Japan. The program created new data as well as new ideas. In the second step, it is necessary to identify R&D items and proceed. For example, the transmutation scenario with high-energy neutrons still needs nuclear reaction data with ^{99}Tc and other nuclides. In addition, 1-A Linac would be highly desirable. R&D for a target to accept the very intense deuteron beam must be conducted in the future. It should be noted that these R&D can be linked to our future of pure nuclear physics, where high intensity beams may become necessary. The methodology of inverse kinematics can also be extended to a reaction study for MAs.

In the program, as an alternative scenario, a transmutation with 14 MeV neutrons produced via muon-catalyzed fusion was also discussed; for which, an efficient production of negative pions has been proposed with an FFAG accelerator.22) To promote further the muon scenario, production cross sections of negative pions with a deuteron beam would be highly desirable. A negative muon was also recognized as a charming particle that causes muon capture reaction and changes the atomic number of nuclei capturing the muon. Based on the muon capture reaction, a new application of muons was proposed to produce the isotope ^{99}Mo for medical use, where ^{99}Tc in the waste is utilized as the substance for ^{99}Mo production via muon capture.23) Experiments of muon capture were organized at the facility of RCNP Osaka University, RAL muon facility, and J-PARC. The results are being published.24,25)

The future role of nuclear reactors is being intensively discussed in conjunction with the realization of a carbon-free society to avoid global warming. However, risk management in reactor operation has become a serious concern since the Fukushima incident, and the cost of nuclear energy has increased. Regardless of the promotion of or opposition to nuclear energy, the problem of high-level radioactive waste needs to be solved because the waste already exists. In Japan, approximately 18,000-tons of spent fuel exists, and the location for the geographical disposal has not been decided. Mitigating the risk of the waste is an important issue because this negative legacy should be left to posterity. Pure nuclear physics is now playing an important role in realizing a radioactive-waste free society.

Acknowledgment

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