# Transmutation study for reduction of high level radioactive wastes

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#### Abstract

We are challenging to nuclear transmutation study for reduction of radioactive wastes, as a project in ImPACT program organized by Cabinet office, Government of Japan. The mission we are assigned is to determine the optimal path for transmuting nuclear wastes. For this purpose, we develop a microscopic effective reaction theory (MERT) and thereby generate reaction data needed to find the reaction path. In this paper, we report the present status and future perspective of the theoretical study on the nuclear transmutation at RCNP.

## 1 Introduction

#### 1.1 Nuclear transmutation study

High level radioactive wastes produced in nuclear power plants are disposed in a deep geological repository. Such a way may cause, however, safety issues of long-term storage of the nuclear wastes. Furthermore, the so-called NIMBY (not-in-my-backyard) syndrome makes it seriously difficult to determine the disposal site. As a possible solution to these issues, nuclear transmutation is an important and challenging subject in the field of nuclear physics and nuclear engineering. In spite of long-term and worldwide studies, nuclear transmutation technique has not been established.

The research project, "Reduction and Resource Recycle of High Level Radioactive Wastes with Nuclear Transmutation", in Impulsing Paradigm Change through Disruptive Technology program (ImPACT) organized by Cabinet office, Government of Japan [1], has newly started in fiscal year 2014. The aim of this project is to convert long-lived fission products (LLFP) included in high level radioactive wastes into short-lived or stable nuclides through nuclear transmutation. Nuclear scientists and engineers are collaborating on this project and a key feature is that we can measure nuclear reaction data including LLFP by using the Radioactive-Ion Beam Factory (RIBF) in RIKEN. Those data are crucial to design an optimal path for transmuting LLFP.

The nuclear transmutation study in ImPACT is classified into the five projects: (i) developments of separation and recovery technologies, (ii) acquisition of nuclear reaction data, (iii) reaction theory modeling and simulation, (iv) evaluation of nuclear transmutation system and development of element technologies, and (v) process concept for design. We are developing a reliable model for reactions involving LLFP, as a part of the project (iii).

#### **1.2** Reaction theory modeling and simulation

To determine the optimal path for conversion of nuclear wastes, we have to consider a whole view of macroscopic reaction systems, as an aggregate of microscopic reactions of various kind inside a target materials. For this purpose, we use the particle and heavy ion transport code system (PHITS) [2]. The PHITS code is proven to work for facility design, medical physics, radiation production, and geoscience. However, it is not sure whether the reaction models used in PHITS are valid for reactions of LLFP and other unstable nuclei. In addition, comprehensive data set is not available although a part of reaction data including LLFP will be obtained in RIBF. Even more seriously, it is impossible to directly measure neutron reaction data on LLFP. The mission of the RCNP theory group is to verify and/or improve the reaction models implemented in PHITS, and generate genuine reaction data of LLFP with a nuclear reaction theory.

# 2 Microscopic Effective Reaction Theory (MERT)

### 2.1 Overall picture of MERT

For reactions of stable nuclei, thanks to the richness of existing data, phenomenological approaches work well. On the other hand, for the description of nuclear reactions of LLFP, we cannot follow that way, because even elastic scattering data do not exist for LLFP. Therefore we need to construct a microscopic reaction theory that has a predictive power for various direct reactions. The microscopic description of nuclear reactions is one of the most essential subjects for not only the transmutation study but also nuclear physics itself.

It is difficult to solve a nuclear many-body scattering problem exactly in most cases. Fortunately, however, we do not need to treat all the degrees-of-freedom of the many-body system, if we choose an appropriate model-space in describing a reaction process with a required accuracy. According to the multiple scattering theory [3, 4], we can use an effective nucleon-nucleon interaction instead a realistic nucleon-nucleon force. Then, a g-matrix interaction, which is constructed by solving the Bruckner-Bethe-Goldstone equation in

infinite nuclear matter, is used as a simplified effective interaction. It is demonstrated that the nucleonnucleus elastic scattering at intermediate energies is well described by a g-matrix folding model, where the wave function or density of the target nucleus is given by some structural calculation. If some effects induced by a specific feature of a nucleus, which are not included in g matrix, are expected, we should treat them explicitly. Deuteron-induced reaction is a typical example. As known well, deuteron is a weakly bound nucleus and easily breaks up into p and n in the scattering process. Then we should include that property in the reaction model, and the importance of a p + n + A three-body model (A is a target nucleus) is suggested. The deuteron breakup effects can be treated by, for example, the continuum-discretized coupled-channels method (CDCC)[5]. As an input of CDCC, we can apply the g-matrix folding potential to the scattering potential between p(n) and the target nucleus. Thus we can merge a reaction model and the microscopic framework. This theoretical framework is referred to as the microscopic reaction theory [5] or the microscopic *effective* reaction theory (MERT) emphasizing that we adopt an effective model-space.

As mentioned above, in the ImPACT program we must generate nuclear reaction data including LLFP. Our basic strategy is 1) we determine the model-space of MERT by the analysis of reaction data measured, and 2) MERT generates the objective reaction data. We show an example of the procedure in Fig. 1.



Figure 1: Schematic explanation of extracting (n,2n) data from (p,pn) data.

# 2.2 Recent progress of MERT

Elastic scattering is one of the most basic processes in nuclear reactions. For nucleon-nucleus elastic scattering, MERT well succeeds to reproduce the measured cross sections and spin observables [6]. It is successful for describing elastic scattering of light ions, deuteron [7] and <sup>3,4</sup>He [8, 9]. For heavier projectile, the results of MERT are not perfect at large scattering angles [10], even if the three nucleon force effect and the coupled-channels effect are included. Further investigation is needed to understand the whole picture of the nucleus-nucleus elastic scattering.

In addition to elastic scattering, very recently, we have solved a long-standing problem on the description of  $\alpha$ -<sup>12</sup>C inelastic scattering to the 0<sup>+</sup><sub>2</sub> state of <sup>12</sup>C [11]. The 0<sup>+</sup><sub>2</sub> state of <sup>12</sup>C, the Hoyle state, is well known to have a three- $\alpha$  cluster structure. There are many experimental and theoretical studies on the Hoyle state but the monopole transition strengths of the Hoyle state obtained by structural calculations are different from that extracted from reaction observables. This inconsistency between the structural calculations and reaction analyses is called a missing monopole strength [12]. To solve this problem, we analyze the  $\alpha$ -<sup>12</sup>C scattering with MERT.

Figure 2 shows the  $\alpha$ -<sup>12</sup>C inelastic scattering into the 0<sup>+</sup><sub>2</sub> state of <sup>12</sup>C at 172.5, 240, and 386 MeV. The red lines show the results of microscopic coupled-channels calculation with the Melbourne g-matrix interaction and the transition densities of <sup>12</sup>C obtained by the 3 $\alpha$  resonating group calculation with the Volkov force [13]. The theoretical calculations well reproduce the measured cross sections at forward angles, except for 172.5 MeV. Thus we show that there is no room for the missing monopole strength. This achievement is not relating the transmutation study directly but is important to validate the reliability of MERT for elastic and inelastic scattering.



Figure 2: Angular distributions of  $\alpha$  inelastic scattering on <sup>12</sup>C into the  $0_2^+$  state of <sup>12</sup>C at 172.5, 240, and 386 MeV, as a function of the center-of-mass scattering angle. This figure is taken from Ref. [11].

# 3 A new model for deuteron induced reaction

We are considering proton, deuteron, <sup>12</sup>C, and muon beams as a tool of nuclear transmutation. It is found by the PHITS simulation that deuteron beam is more effective to convert LLFP than proton and heavy ion beams. Deuteron has an advantage taking into account its cross sections, its beam intensity, and stopping power. However, it was found that NASA's formula of the total reaction cross sections of deuteron induced reaction [15, 16, 17], which is used in PHITS, is less reliable at low incident energies. We should examine the reliability of the model by comparing with the results of MERT and the experimental data.

The microscopic CDCC is applied to the analysis of deuteron induced reactions [14]. In the CDCC calculation, we adopt optical potentials between p(n) and target constructed with the *g*-matrix folding model. Figure 3 shows the total reaction cross sections of deuteron induced reaction calculated with CDCC (thick lines), as a function of the deuteron incident energy. It can be seen the good agreement between the



Figure 3: Total reaction cross sections of deuteron induced reaction for <sup>12</sup>C (red solid line), <sup>58</sup>Ni (green dashed line), <sup>116</sup>Sn (blue dotted line), and <sup>208</sup>Pb (purple dash-dotted line) targets, as a function of the deuteron incident energy. The four thin lines represent the results of NASA's formula [15, 16, 17] implemented in PHITS [2]. This figure is taken from Ref. [14].

results of CDCC and the experimental data. For heavier target, there is a difference between the results of CDCC and the NASA's formula even at high incident energies.

Furthermore, it is found that the results of CDCC can be parametrized with a simple functional form. We thus suggested a new parametrization of the total reaction cross sections for deuteron induced reaction up to 1 GeV, and then, we have made the theoretical simulation of transmutation more reliable.

#### 4 Summary and perspective

We have introduced the nuclear transmutation study in ImPACT program for reduction of nuclear wastes. In this project, we should make the simulation of an optimal path by PHITS more reliable from the theoretical point of view. For this purpose, we are developing the microscopic effective reaction theory (MERT).

We have mentioned the high reliability of MERT for nucleon, deuteron, and light ions induced elastic scattering. Also, MERT demonstrated the predictive power for the  $\alpha^{-12}$ C inelastic scattering. Furthermore, we proposed a new parametrization of the total reaction cross sections of deuteron induced reactions by the microscopic CDCC analysis. As a result, the simulation by PHITS will be improved partly.

(n,xn) reactions are also effective for transmutation. However, reactions between neutron and LLFP can not be measured directly. It should be extracted with MERT by utilizing proton and deuteron data. We are analyzing the new (p,xn) and (p,xp) data measured at RIBF. Such spallation cross sections consist of multinucleon knockout reactions as direct processes and decay of the residue described as pre-equilibrium (and evaporation) processes. We are now trying to connect the two reaction processes to establish a theoretical description of the spallation cross sections.

The nuclear transmutation study with muon beams is ongoing in the Muon Science Innovative muon beam Channel (MuSIC) project in RCNP. The theoretical description of transmutation with muon is in the scope of our research plan. We expect that MERT will play an important role also in muon physics.

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