Neutron Yield Measurement of natCNeutron Yield Measurement of natC(d,n) Reaction for Radioisotopes Production and Development of PHITSbased Dosimetry System for Medical Application

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（放射性同位元素製造のための C(*d*,*n*) 反応からの中性子収量測定とその医療応用に向けた PHITS によるドシメトリ計算システムの開発）

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論 文 内 容 の 要 旨

Thesis Summary

In recent years, intensive accelerator-based neutrons sources with deuteron-induced reactions have been proposed for various applications, especially, production of medical radioisotopes (RIs). We focused on neutrons generated via the C(*d*,*n*) reactions, because they have advantages of neutron intensity and energy adjustability. Moreover, because high energy neutrons to be applied for RIs productions are strongly focused on forward emission angle, a simple irradiation system having a low quantity of shielding is possible in actual applications. To estimate the production amount and purity of produced RIs, accurate double-differential thick-target neutron yield (DDTTNY) is necessary. It is also required to design shielding of the irradiation system. For the RI productions, deuteron having a few to 50 MeV is used to generate the neutrons, but experimental DDTTNYs of the C(*d*,*n*) reaction in the range have not been measured systematically. Besides, the lack of experimental data leads to the uncertainty of the production amount and purity of produced RIs. For the RI production applications, neutron energy adjustment is promising to reduce byproducts' contribution, thus the uncertainty is sometimes cause of difficulty of optimization of deuteron energy for the neutron energy adjustment.

In the accelerator-based RI production method, new RIs are proposing for medical use for diagnosis and therapy. To avoid from negative effects of normal organs and tissues during the diagnosis or therapy, accurate prediction of absorbed dose is very important. External radiation doses like X-ray, CT, or IMRT therapy can be precisely predicted by using conventional models or the latest Monte Carlo based method, and also the internal dose of inhalation and ingestion can be determined by using dose coefficients listed in ICRP Publication 119. In contrast to that, however, the precise dose calculation method of "administrated medical RIs" has not been established yet. Nowadays, the dose prediction is given by Monte Carlo calculations using the ICRP/ICRU recommended AM and AF phantoms which are based on the organ masses according to the ICRP Publication 89 having statistics for Western Europeans and North Americans. In other words, because the phantoms are different from Asian reference personals, we cannot obtain precise dose prediction by using these estimations. Also, there is no established prediction method of the radiation dose of the clinical staff and the patient's family from the administrated patient. The dose can be measured by an expensive survey meter for the clinical staff, but after checking out of the hospital there is no way to measure the dose of the patient's family.

To overcome the above situations, this thesis has two purposes: First, DDTTNYs of the C(*d*,*n*) reaction for 12, 20 and 30 MeV deuteron were measured, because around 25 MeV, available experimental data are lacked even though the energy is considered promising for some medical RIs production, such as ^{64}Cu. Furthermore, the reproducibility of DEURACS was surveyed in the measured region to confirm the reliability of the code. Then, the DDTTNYs were used for estimation of RI production amount and purity of two applications, ^{132}Cs for environmental tracer and ^{47}Sc for medical use. Second, because some new RIs production is considered for the accelerator-based neutron method, a universal dose calculation system is proposed. In the calculation system, Monte Carlo simulation code, PHITS is used with realistic voxel phantom, JF-103, and JM-103, and the dose of an administrated patient is derived by using the latest ICRP recommendations. Furthermore, to establish a calculation method of clinical staff and patient's family dose, γ-ray sources from administrated patients are recorded. The source can be used in Monte Carlo simulation code such as PHITS or GEANT4, to predict the dose of them.

This thesis introduces the background of the study and motivation of this work in the first chapter.

After that, Chapter 2 is devoted to experimental measurements of the thick-target neutron yield by using deuteron induced accelerator-based neutron source and the multiple-foil activation method. The experiments were conducted at the tandem

accelerator facility of the Japan Atomic Energy Agency. Deuterons having 12, 20, or 30 MeV kinetic energy were separately incident on a thick target neutron converter made of ^{nat}C (Ø20 mm, 4 mm^{t}). So that neutrons were generated via the (*d*,*n*) reactions. Multiple-foil made of ^{nat}Ni, ^{nat}In, ^{27}Al, ^{59}Co, and ^{93}Nb were placed at 0º, 10º, 20º, 30º, and 45º from the deuteron beam direction and were exposed to neutron irradiation for different time durations with different beam currents. Neutron spectra were unfolded for each incident deuteron energy and each neutron emission angle by using GRAVEL code. The results were compared with the theoretical calculations with DEURACS, and they showed that the DEURACS calculation reproduces the experimental DDTTNYs at 12, 20, and 30 MeV of deuteron energy from 0° to 45° of neutron emission angle. Results showed that DEURACS can be used to interpolate the lacking experimental DDTTNY. Moreover, a detailed comparison of yields to other available published data are also explained in this chapter. It is showed that this research can full fill the lack of nuclear data within the mentioned deuteron energy range.

In Chapter 3, the application of accelerator-based neutron to RIs production is discussed. This chapter is categorized into two parts. At first, an environmental tracer ^{132}Cs ($T_{1/2}$ = 6.5 d) production experiment is described. This is produced by irradiating a 12-g of Cs_2CO_3 sample using accelerator-based neutrons by 1.2 μA of 30-MeV deuterons. The ^{132}Cs production experiment was conducted at the Cyclotron and Radioisotopes Center (CYRIC) of Tohoku University, Japan. The ^{133}Cs(*n*,*2n*)^{132}Cs reaction, initiated by irradiating 30 MeV deuterons on a thick ^{nat}C (Ø40 mm, 4 mm^{t}), yielded 102.2 kBq/g of ^{132}Cs with a radioactive purity of 98% after 2-hour irradiation. Also, a feasibility study on andosol, haplic fluvisol, and gleyic fluvisol soil explains that ^{132}Cs can be used as an alternative tracer of ^{137}Cs ($T_{1/2}$ = 30y) for environmental radioactive monitoring. Moreover, the production yields and purities of an emergent theranostic medical RI, ^{47}Sc is estimated by PHITS simulation by using measured DDTTNYs of 12, 20, and 30 MeV deuterons. When irradiating 5 g of ^{47}Ti powder for 5 hours, ^{47}Sc were produced via ^{47}Ti(*n*,*p*) reaction and yielded a maximum 4.59×10^{-5} MBq/g/μC for neutron yields of 30 MeV deuterons. Based on the results from this proof-of-concept study, ^{47}Sc has the potentiality to produce in a suitable quality for theranostic clinical applications.

PHITS-based internal dosimetry for organ/tissue absorbed dose estimation is reported in Chapter 4. An improved internal dosimetric calculation with the realistic Japanese reference voxel phantoms, JM-103 and JF-103 based on CT images of the real patients, have been implemented in PHITS. The novelty of our calculation system is applicability to any administrated RIs and any RIs using or to be used in the research field (e.g. tracer). The program uses the radionuclide decay database of ICRP Publication 107 and considers the ICRP recommended 83 different source regions irradiating 47 target tissues, defining the effective dose as presented in both of the ICRP Publications 60 and 103. This calculation system applies to all of the RIs. In this thesis, we calculated the dose for many RIs including ^{132}Cs and ^{47}Sc which is newly proposed. This is the case study of the adsorbed dose calculation with RIs having prospects. An extension of the developed dose calculation method is applied to estimate the sex-averaged effective dose based on the tissue weighting factors in ICRP 2007 recommendations; are presented for ^{47}Sc with other frequently used medical RIs e.g., ^{18}F, ^{99m}Tc, ^{90}Y, etc. Dosimetric results were showed that our developed dosimetry calculations can be recommended for the Japanese averages and also for the Asian personal. Also, this chapter presents the dose coefficients for intravenous administration for several emergent medical RIs (e.g., ^{64,67}Cu, ^{152,161}Tb, ^{166}Ho, etc.), and compared with data published in the ICRP Publication 53 and 128. From these aspects, the present results can play an important role as a guideline for patient-specific dosimetry.

Chapter 5 and Chapter 6 are designated for the PHITS-based external dosimetry with our developed computational framework. In Chapter 5, an operational quantity for the area or workplace monitoring called the ambient dose equivalent, $H^*(10)$, is expressed according to the latest ICRU definition. This is done by using the universal γ-ray source calculation. This procedure successfully applied to $H^*(10)$ estimation for a post-treated patient with ^{47}Sc-, ^{64}Cu-, ^{64}Cu/^{67}Cu-theranostics. The calculation results showed significance to the radiation safety issues especially for the clinical staff who works in the nuclear medicine department, and as well as the person who will take care of the patient after therapeutic or theranostic applications. The universal γ-ray source is one example of applications of our developed dose calculation system. If the calculation system is updated, we can calculate the universal γ-ray source with the same method using the updated one. Next, Chapter 6 set out the estimation of the absorbed dose coefficients for organs/tissues of a researcher for the occupational exposure from an external radiation source. This is also an application of our improved PHITS-based dosimetric calculation. As of practical experience, during dealing with radioactive ^{132}Cs an occupational radiation worker may expose to external radiation emitted by Cs radionuclides. This chapter explains the absorbed dose to all individual body parts of the researcher rather than an overall dose estimation with a personal dosimeter. Therefore, an occupational radiation worker can take necessary precautions to save the critical body parts by ensuring personal safety.

Finally, Chapter 7 summarized the present thesis work and pointed out the possible prospective future outlook.