# CO2-1 Quantitation of Gamma Ray Emission from Caputre Reaction of Uranium-238 (2)

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**INTRODUCTION:** Neutron induced  $\gamma$  ray spectroscopy has been conducted at the LINAC neutron source facility of KURNS to develop a quantification method of fissile enrichment in nuclear material. For that,  $\gamma$  ray emission from <sup>238</sup>U(n, $\gamma$ ) reaction has been focused on [1]. In 2019, the first experiment was performed with neutrons induced by pulsed electron of 1  $\mu$ s width [2]. In 2020, enhancement of the statistical accuracy was examined.

**EXPERIMENTS:** In the facility, accelerated electron is introduced on a tungsten target and the Bremsstrahlung ray is radiated. Photo-nuclear reaction is induced by the ray and then neutron is radiated. A metal uranium sample was irradiated by the neutron of white spectrum and

<sup>238</sup>U(n,γ) γ ray was measured with a HP-Ge detector. To determine the incident neutron energy, the time of flight technique was applied with periodically emitted (50 Hz) pulsed electron. The neutron fluence is almost proportional to the pulse width. In this work, neutrons of energy less than 50eV was focused on so that the width does not matter for the accuracy of neutron energy determination. Instead, intense pulse introduces dead time in HP-Ge detector by so-called γ-flash. We adjusted the width up to 3 μs so that we counted γ-rays induced by neutrons of 36.6eV as shown in Fig. 1.

**RESULTS:** In Fig. 2, the  $\gamma$  ray spectra measured with the electron pulses of 1 and 3  $\mu$ s are compared. The spectra for the thermal neutron are very similar each other and the counting statistics is enhanced by the pulse of 3  $\mu$ s width. For the measurement of events induced by the thermal neutron, the electron pulse of 3  $\mu$ s width is preferable.

On the other hand, for the events induced by the 6.67eV neutrons, the peak pulse height (energy) is shifted lower by ~40keV for the pulse of 3  $\mu$ s width compared to those for the pulse of 1  $\mu$ s width. One of the possible reasons of the shift might be undershoot of signal current from the HP-Ge detector short time after the  $\gamma$  flash. Thus, more detailed analyses are now being conducted to quantify the influence of the  $\gamma$  flash on the  $\gamma$  ray spectra induced by neutrons of energy from 5 to 50 eV.

Additionally, we also focused on the  $\gamma$  rays found in the time background region (see Fig, 1) where those emission is out of phase of the electron pulse. By comparison against the JENDL/FPY&FPD-2011[3], we have identified  $\gamma$  rays from fission products (FP) of which half-life is from 0.5 s to 10.4 min. We will quantify the radioactivity of those to validate isotopically FP yields and decay chains used for safety analyses of reactors.

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Fig. 1 Time of flight spectra of neutron which induces  $^{238}U(n,\gamma)$  reaction determined by  $\gamma$  ray detection.



Fig.2 <sup>238</sup>U(n, $\gamma$ )  $\gamma$  ray spectrum measured with pulsed neutrons of 3  $\mu$ s and 1  $\mu$ s width.



Fig. 3 Short lived fission products measured in time background region.

# CO2-2 Thermal-Neutron Capture Cross-Section Measurement of <sup>237</sup>Np Using Graphite Thermal Column

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**Introduction:** Neptunium-237 has a long half-life of 2.14×10<sup>6</sup> years, therefore it causes long-term radiotoxicity in nuclear waste management. A method of converting radionuclides to stable and/or short-lived nuclides has been studied, that is "nuclear transmutation". In nuclear transmutation of <sup>237</sup>Np with neutrons, accurate neutron capture cross sections of <sup>237</sup>Np are required to obtain transmutation rates. **Figure 1** shows the present status of reported data for the thermal-neutron capture cross section ( $\sigma_0$ ) of <sup>237</sup>Np. Contradicting data have been reported by many measurements for  $\sigma_0$  from 1956 to 2020. Thus, the  $\sigma_0$  of <sup>237</sup>Np was measured by a neutron activation method using graphite thermal column of the KUR.



Fig.1 Reported data for  $\sigma_0$  of <sup>237</sup>Np.

Experiments: Nitric acid solutions of <sup>237</sup>Np equivalent to 950 Bq were dropped onto a glass micro filter and dried with an infrared lamp. To monitor neutron flux components, a flux monitor set of Au/Al wire, Co, Sc, Mo and Ta foils was attached beside the Np sample. The samples were set on the upper edge of a polyethylene capsule. Neutron irradiation was performed using the graphite thermal column equipment (TC-Pn) outside the core of KUR. The structure of the core plane of the KUR is shown in Figure 2. In order to irradiate the capsule at the upper position, two dummy capsules were transferred to the TC-Pn in advance. After that, the capsule containing the samples was transferred and then irradiated for 30 minutes in 1-MW power operation. After irradiations, the samples were enclosed in a vinyl bag one by one, and then measurements were performed by the  $\gamma$ -ray spectroscopy.



Fig.2 Partial plane structure of the reactor core.

Gamma rays emitted from the sample were measured with a high-purity Ge detector installed in the hot laboratory. A measurement distance was appropriately taken as 100 mm from the front surface of the Ge detector to the sample position. **Figure 3** shows an example of  $\gamma$ -ray spectrum obtained by measurement of the irradiated <sup>237</sup>Np sample. Decay  $\gamma$ -rays emitted from <sup>238</sup>Np were measured with a good signal-to-noise ratio.



**Fig.3** An example of *γ*-ray spectrum of the irradiated <sup>237</sup>Np sample

**Analysis:** The reaction rates of neutron flux monitors were obtained from their  $\gamma$ -ray yields. The thermal-neutron flux component was analyzed with these reaction rates on the basis of Westcott's convention. The neutron field at the irradiation position was found to be well-thermalized. The <sup>237</sup>Np sample was quantified using 312-keV  $\gamma$  rays emitted from <sup>233</sup>Pa in in radiative equilibrium with <sup>237</sup>Np. The reaction rate of <sup>237</sup>Np was obtained with the sample amount and  $\gamma$ -ray yields given by <sup>238</sup>Np. The thermal-neutron capture cross section  $\sigma_0$  is derived straightforward by dividing the reaction rate of <sup>237</sup>Np by the thermal-neutron flux component. Currently, data analysis is in progress.

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# CO2-3 Measurement of Energy Resolution in the KURNS-LINAC Pulsed Neutron Facility

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**INTRODUCTION:** The electron linear accelerator at the Research Reactor Institute, Kyoto University (KUR-RI-LINAC) had been originally established in 1965 by the High Voltage Engineering Co., USA and started as a 23 MeV machine. In 1971, the machine power had been increased to 46 MeV. The KURRI-linac has two different operation pulse modes. One is a long mode with a maximum repetition rate of 120 Hz, a pulse width of 0.1-4.0 us and a peak current of about 0.5 A for the measurement at low energies below 10 eV. Another is a short mode with a maximum repetition rate of 300 Hz, a pulse width of 2-100 ns and a peak current of about 5 A for the measurement at high energies above 1 eV. It is worth noting that the peak current of short mode is ten times as large as that of long mode. In measurements of nuclear data, a water-cooled tantalum (Ta) target as a photo-neutron target and a light water moderator are used. There are two kinds of the moderator. One is a water tank type and another is an octagonal shape moderator called "pac-man type". In order to measure accurate nuclear data, it is very important to evaluate the energy resolution  $(\Delta E/E)$  of a moderator. For example, the energy resolution for pac-man type moderator had been calculated about 0.7 % between energy range of 0.1eV from 10keV [1]. However, measurement and detail evaluation of energy resolutions for the tank type moderator has not carried out in KURRI-LINAC, although the energy resolution has been evaluated by using the simplified evaluation formula [2]. Furthermore, the relationship between energy resolution and beam pulse width is not discussed in Reference [1] and [2].

In this study, the energy resolutions of neutron flux from a tank type moderator were obtained using the TOF technic in two operational mode with different pulse width.

**EXPERIMENTS:** In this study, the energy resolution of KURNS-LINAC was obtained by transmitted neutron flux via <sup>181</sup>Ta sample. In order to measure the transmitted neutron flux, a cylindrical moderator tanks which have 19 cm diameter and 30cm height was installed at the target room. The tanks were filled with light water.

We used the neutron at "12 m room" which is located on 135 deg. axis from the electron beam line. The flight path of neutron flux between the <sup>181</sup>Ta sample which has 0.5 mm thickness and the moderator was 12.6 m as shown in Fig.1. In order to obtain the relationship between neutron beam pulse width and energy resolution, the KURNS-LINAC was operated under two different conditions as shown in Table 1. The beam frequency was ad-

justed to 50 Hz and the pulse width was 1  $\mu$ sec or 4  $\mu$ sec (nominally). The beam current was stabilized to about 27  $\mu$ A and 104  $\mu$ A. The transmitted neutron flux was measured by TOF technic with Li-glrass scintillation detector.

Table 1 KURNS-LINAC operational condition.

Table 1 ROR S-EI We operational condition.					
	Frequency	Beam pulse	Beam		
	(Hz)	width (µsec)	current		
Case A	50	1.0	26.68		
Case B	50	4.0	103.85		



Fig.1 Experimental geometry

**RESULTS:** Figure 2 shows the TOF spectra of transmitted neutrons with the two pulse widths. From the figure, it was obtained that the dips width of the transmitted neutron flux by the resonance were different between the 4  $\mu$  sec and 1  $\mu$  sec width. Table 2 shows the FHWMs of the neutron flux dips. The FHWM at the high-energy resonance is larger than that of the low-energy resonance.



Fig.2 Comparison of TOF spectrum between different beam pulse width.

Table FHWM of neutron flux dip

	1 $\mu$ sec	4 $\mu$ sec
10.36 eV	5.8 nsec	6.7 nsec
4.28 eV	25.3 nsec	25.5 nsec

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#### CO2-4 β–Decay Study of Fission Products around A=150 Using the On–Line Mass Separator KUR–ISOL

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**INTRODUCTION:** Short-lived neutron-rich nuclei of mass numbers around 150 have low fission yields of <sup>235</sup>U, so there are few decay information. In order to clarify the decay properties, it is useful to perform highly efficient measurements with large volume Ge detectors. So far, we had measured the  $\gamma$ -rays of the fission products with a clover type Ge detector installed at the on-line mass separator (KUR-ISOL). It has four large Ge crystals with a diameter of 80 mm and a length of 90 mm arranged in the shape of a four-leaf clover around a through hole with a diameter of 15 mm. In the present study, we aimed to measure the <sup>153,154</sup>Pr, which locate almost limit region available with the KUR-ISOL. In using the clover detector, the background  $\gamma$ -rays in the reactor room need to be reduced for precise measurements of  $\gamma$ -rays of the nuclei of interest, especially <sup>154</sup>Pr. Then,  $\beta$ -ray detectors were developed for  $\beta - \gamma$  coincidence measurement with the clover detector. The performance of the  $\beta$ -ray detectors were tested by measuring well evaluated some fission products of their decay schemes, and the detectors were adopted for the measurement of <sup>154</sup>Pr. In this report, the preliminarily results of <sup>153</sup>Pr as well as <sup>154</sup>Pr were described.

**EXPERIMENTS:** The experiment was carried out with the KUR-ISOL. A 72 mg of 93% enriched <sup>235</sup>UF<sub>4</sub> was irradiated with thermal neutrons flux of  $3 \times 10^{12}$  n/cm<sup>2</sup>/s under 5 MW operation. The fission products were transported to a surface-ionizing type ion sources by an He-N<sub>2</sub> gas jet. The ionized fission products were transported to the magnetic sector-type mass spectrometer to obtain the nuclei of interests. The radioactive beams were implanted in a thin aluminized Mylar tape, and were periodically transported to the center of the through hole of the clover detector by a computer-controlled tape transport system. For <sup>154</sup>Pr, two identical  $\beta$ -ray detectors made of plastic scintillator were installed in the through hole of the clover detector. Those were semi-cylindrical shape of 6 mm thick and 9 cm long. The MPPC (Multi-Pixel Photon Counter) module C13367 of  $3 \times 3 \text{ mm}^2$  made by Hamamatsu Photonics were set at the top ends of the scintillators. The  $\beta$ - $\gamma$  coincidence measurements between

 $\beta$ -detectors and the clover detector were carried out. The period of collection–measurement cycle was set at two times of each half–life of the nuclide to be measured, approximately. The whole detectors were shielded with 10 cm thick lead bricks and 10 cm thick polyethylene bricks which contain boron. The data was recorded in the APV8008 and APV8016 DAQ systems made by Techno AP with event by event mode including time information. Measurements of <sup>153</sup>Pr (T<sub>1/2</sub>= 4.3 s) and <sup>154</sup>Pr (T<sub>1/2</sub>= 2.3 s) were performed for 13.5 and 15.5 hours, respectively. The APG-1500 amplifiers (Techno AP) was adopted to match the signals from the MPPC module to the DAQ systems.

**RESULTS:** The decay schemes were proposed based on the X- $\gamma$  and  $\gamma$ - $\gamma$  coincidence relations and the  $\gamma$ -ray singles and add-back spectra. For <sup>153</sup>Pr, the proposed decay scheme has excited levels up to 3436 keV and includes observed 72  $\gamma$ -rays and 25 excited levels. These results are more detailed than the previous result [1]. For <sup>154</sup>Pr, the part of the measured  $\beta - \gamma$  coincidence spectrum is shown in Fig.1. The statistics of the  $\gamma$ -rays are more than one order of magnitude of those of the previous ones [2]. As shown by the arrow in the inset, the  $\gamma$ -ray of 1294 keV by the  ${}^{40}$ Ar(n, $\gamma$ ) ${}^{41}$ Ar reaction was clearly reduced in the  $\beta$ - $\gamma$  coincidence spectrum. The efficiency of the developed  $\beta$ -ray detector was evaluated to be 20% for each for the energy of  $\beta$ -ray at 2 MeV from the calibration measurements of <sup>94</sup>Rb, <sup>94,95</sup>Sr, <sup>94</sup>Y, and <sup>146</sup>La. The decay scheme up to 2940 keV including the newly observed 43  $\gamma$ -rays and 18 excited levels was proposed compared to the previous work [2]. Detailed analysis is in progress.



Fig.1 A portion of  $\beta - \gamma$  coincidence spectrum. The open and closed circle indicate the  $\gamma$ -ray of <sup>154</sup>Nd and <sup>154</sup>Pr, respectively. The inset shows the comparison of singles spectrum and  $\beta - \gamma$  coincidence one.

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# CO2-5 Measurements of thermal neutron total and scattering cross section of moderator materials

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**INTRODUCTION:** The high temperature gas-cooled reactor is a Generation IV reactor concept that use a graphite moderator. The design takes advantage of the inherent safety characteristics with specific design optimizations. The treatment of thermal scattering caused by crystalline is important for graphite to improve the prediction accuracy of graphite moderated core analysis. There is sufficient thermal scattering law for single crystal graphite, but there is no method has been established to reconstruct thermal scattering cross section for polycrystalline graphite. On the other hand, a new concept of small modular reactor using CaH<sub>2</sub> as solid moderator has been developed [1]. The experimental and evaluated data of the thermal scattering law for CaH<sub>2</sub> are insufficient. In order to provide basic data for thermal neutron scattering law evaluation, the total and scattering cross sections of moderator materials were measured.

**EXPERIMENTS:** The total and scattering cross sections of moderator materials were measured in the incident neutron energy region from 0.001 to 10 eV by transmission and scattering experiments at the KURNS-LINAC. An experimental arrangement is shown in Fig. 1. Pulsed neutrons were produced from a water-cooled Ta-target by  $(\gamma, n)$ reaction with a pulsed electron beam. The incident neutron spectrum on a sample, the transmitted and scattering neutron spectrum were measured by means of a time-of-flight (TOF) method with a <sup>6</sup>Li-glass scintillation detector. A 5.0 mm diameter by 5.0 mm thick <sup>6</sup>Li-glass was located 12.0 m from the neutron source. The scattering neutrons were measured by a 50 mm diameter by 5.0 mm thick <sup>6</sup>Li-glass scintillation detector. The scattering neutron spectra were observed at angle of 30 degrees and 45 degrees with respect to the neutron beam direction.

The characteristics of the samples are shown in Table 1. The graphite and polyethylene sample are used as a moderator in the KUCA facility.

**RESULTS:** The total cross sections of moderator materials were derived in the incident neutron energy region from 0.001 to 10 eV. Figure 2 shows the scattering neutron specta for CaH<sub>2</sub> sample. The room scattering neutron background could be properly evaluated by using the measurement without sample. Figure 3 shows the scattering neutron spectrum with the resonance filter (Cd, In, Ag, Mn). It was shown that it is possible to measure quasimonochromatic neutrons from a white neutron source.

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Fig 1. Experimental arrangement

Table 1 characteristics of samples					
Sample	Polyethylene	Graphite	CaH <sub>2</sub>		
Structure	High molecu-	Polycrystalline	Powder		
Sample shape	Square plate	Square plate	Disk in Al		
Density [g/cm <sup>3</sup> ]	0.949	1.754	1.200		
Size [mm]	$50.8 \times 50.8$	$50.8 \times 50.8$	<i>\$</i> 50		
Thickness [mm]	3.18	12.7	5.0		
[atoms/b]	$1.26 \times 10^{-2}$	$1.12 \times 10^{-1}$	$8.63 \times 10^{-3}$		



Fig. 2 Scattering neutron energy spectra



Fig. 3 Scattering neutron energy spectrum with resonance filter (Cd, In, Ag, Mn)

# CO2-6 Development of a neutron sintillator for a compact NRTA system

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**INTRODUCTION:** The Japan Atomic Energy Agency (JAEA) is developing a compact Neutron Resonance Transmission Analysis (NRTA) [1] system for nuclear material (NM) accountancy applications. NRTA is a neutron TOF technique [2] that utilizes a pulsed neutron source. To realize a compact NRTA system for high accuracy measurements, generation of a short-pulse neutron beam is required [1]. A newly arising laser-driven neutron source (LDNS) is considered potentially to be useful for short-pulse neutron beam generation without using a conventional accelerator. We, therefore, started a feasibility study of a NRTA system with an LDNS [3,4].

Amongst the study, requirements on neutron detector development arose: i.e., neutron detector with high efficiency, applicability to high counting rate, and insensitivity to gamma rays. It should be noted that the 2.2-MeV gamma ray induced by  $n({}^{1}\text{H}, \gamma){}^{2}\text{H}$  neutron capture reaction in a moderator is an origin of background in an NRTA measurement. Although the gamma-ray background dies away as time goes by, it will interfere measurement of epithermal neutron transmission with a compact system. Therefore, we started development of a gamma-ray insensitive multi-layer <sup>6</sup>Li glass scintillation (MLS) detector [4].

**EXPERIMENT AND RESULT:** Performance tests of a MLS detector ( $50 \times 50 \times 5$  mm in MLS size) and a 1-cm <sup>6</sup>Li-glass scintillation detector ( $100 \times 100 \times 10$  mm) were performed at the 12-m TOF measurement room of the KURNS-LINAC. Pulsed electron beams of about 30 MeV were used for neutron generation with a repetition rate of 50 Hz. The pulse width and the average current were 2  $\mu$ sec and about 65  $\mu$ A for the experiments of the MLS detector. Those were reduced to be 0.5 µsec and about 15 µA for the experiments of the 1-cm <sup>6</sup>Li-glass detector to prevent detector saturation. The output signals from the detectors were recorded using a 1-GS/s flash waveform digitizer (CAEN V1751). 800-µsec-long waveforms starting with each electron shot were stored in the memory of the digitizer and transferred to a computer. Dynamically moving baseline of measured waveforms were removed off line. Neutron induced signals were counted disregarding noise signals from their pulse shapes.

A reference material filter consisting of In, Ag, Co and Mn (referred to as notch-filters) were placed in the neutron beam path. These materials were thick enough to prevent neutrons with the neutron energy of large resonances (i.e., In-115 (1.46 eV), Ag-109 (5.19 eV), Co-59 (132 eV) and Mn-55 (336 eV)) arriving the detectors.

TOF spectra measured by the two detectors are shown in Fig. 1. The ordinate was roughly adjusted making so as the integrations of the 5.19-eV Ag resonance dips are the same. Time dependent backgrounds were evaluated by connecting the bottom of the dips using the following function [5]:

$$B(t) = b_1 e^{-\lambda_1 t} + b_2 e^{-\lambda_2 t}$$

The first term explains the 2.2-keV gamma-ray background from a moderator, and the second term the background neutron scattering around the detector. The time constant  $\lambda_1$  is shorter than  $\lambda_2$ . Time dependent total backgrounds, B(t), and gamma-ray backgrounds,  $b_1 e^{-\lambda_1 t}$ , are also plotted in Fig. 1 (a). Gamma-ray backgrounds are found to be dominant in the early time frame of the TOF spectra. The gamma-ray background of the MLS detector is about half of that of the 1-cm <sup>6</sup>Li-glass detector at the Co-59 and Mn-55 resonance region, while the shape of neutron resonance spectra does not change much, as seen in Fig. 1 (b). This describes that an MLS type scintillator can reduce the sensitivity of 2.2-MeV gamma rays without changing much the neutron counting efficiency.



Fig. 1 (a) TOF-spectra taken with the MLS and the 1-cm <sup>6</sup>Li-glass neutron detector. (b) Expanded spectra with a linear ordinate scale.

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# CO2-7 Feasibility study on a current-mode <sup>3</sup>He gas neutron detectors for thermal and epi-thermal neutron measurements

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INTRODUCTION: In the present study, we have developed a current-mode neutron detector [1] that can be used in intense neutron flux fields such as boron neutron capture therapy (BNCT) fields. It is necessary to measure the neutron flux for a large dynamic range because of con-nection between a BNCT field in a hospital and the neutron calibration field in the National Institute of Advanced In-dustrial Science. The difference of neutron fluxes be-tween the BNCT field and the calibration field is more than 5 orders of magnitude. We have tried to extract neutron signals using a current mode <sup>3</sup>He gas detector (propor-tional counter and ionization chamber). The current mode gas detector is expected to be high radiation re-sistance in comparison with a photo-multiplier tube in the scintillation detector.

**EXPERIMENTS:** A collimated neutron beam with 30 mm diameter was obtained by the photo neutron reaction using a tantalum target with a water moderator at the KURRI Linac [2]. Figure 1 shows the experimental setup. In the experiments, we used a <sup>3</sup>He proportional counter (Reuter Stokes RS-P4-0806-278). The <sup>3</sup>He proportional counter was set at 12 m away from the target. A BF<sub>3</sub> pro-portional counter was used as a neutron monitor. All data were normalized using counts obtained with the BF<sub>3</sub> pro-portional counter. Possibility of a current-mode <sup>3</sup>He gas detector was experimentally evaluated by means of the time-of-flight (TOF) method. The measurements were performed for both pulse and current In the pulse mode, signals from the <sup>3</sup>He modes. proportional counter were ob-tained using a preamplifier (ORTEC 142PC) and main amplifier (ORTEC In the current mode, the signals were obtained 570). using a current integrator (ORTEC 439). Finally, TOF data were extracted using a multi-stop time to digital converter.

**RESULTS:** Figure 2 shows results of TOF spectra of pulse and current modes for the <sup>3</sup>He proportional counter. In the current mode, thermal bump is successfully observed in the TOF spectrum.

The <sup>3</sup>He proportional counter has high neutron sensitivity because of high gas pressure. The count rate was 6309 cps in the pulse mode. In this case, the pulse shape after the main amplifier is distorted by the pile-up effect because of 2  $\mu$ s of shaping time in the amplifier. Moreover, the dis-tortion due to gamma flash is added for the epi-thermal neutron range. On the other hand, TOF spectrum in the current mode is not affected by the distortion of pulse shape in the current mode. However, the present experimental setup has large time resolution caused by the

In the next step, the gamma-ray subtraction will be tried to use a <sup>4</sup>He proportional counter that is not sensitive to neutrons. Relation among the gas pressure, detection efficiency in the pulse and current modes and linearity in the large flux range in the current mode will be also investigated.



Fig. 1. Experimental setup for the  ${}^{3}$ He gas detector. The detector was set at 12 m away from the target.



Fig. 2. TOF spectra of (a) pulse and (b) current modes.

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#### CO2-8 Technique of Transferring Radioactive Atomic Nuclei Implanted in Dry Ice Film

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**INTRODUCTION:** The nuclear charge radius is one of the fundamental parameters that gives information about the effective interactions on nuclear structure. Muonic atoms are ideal tools for the study because the interaction between negative muons and nuclei can be accurately described by electromagnetic interaction. The energies of the X-rays emitted from the muonic atoms are measured for the investigation [1]. The study of muonic atoms for unstable nuclei, however, is little progress mainly because a large amount of radioactivity is needed. We have recently developed a new method using solid hydrogen film to produce muonic atoms with high efficiency. In this method, negative muons  $(\mu^{-})$  are injected to solid deuterium film (D) in which nuclei of interest (A) are implanted beforehand, thereby muonic atoms being formed through the highly-efficient muon transfer reaction:  $\mu^-D + A \rightarrow D + \mu^-A$ . The feasibility of this method was demonstrated for some stable isotopes and promising results were obtained [2]. In this study, the technical difficulties to be expected in applying this method to unstable nuclei, in particularly concerned with high-efficient recovering of residual radioactivities in deuterium film after experiments, is being approached using a radioactive-isotope beam from KUR-ISOL and dry ice film instead of solid hydrogen film.

**EXPERIMENTS:** An apparatus capable of implanting radioactivities to dry ice film was installed at the beam line of KUR-ISOL [3]. This apparatus consists of two coolable copper blocks and one CO<sub>2</sub> gas diffuser in the vacuum chamber. Dry ice film is formed on the surface of one cooled block (catcher) by sprayed CO2 gas through the diffuser and radioactivities are implanted into it. The amounts of radioactivities transferred from the catcher to the other block (trap) are examined under the different conditions of the film formation. In the recent experiments, in order to improve the reproducibility of the transfer efficiency, the film forming parameters were controlled more finely by recording the vacuum degree in the chamber and the temperatures of the copper blocks using a data logger. Moreover, "standing collars" were attached on the four sides of the film formation surface so that more CO<sub>2</sub> gas could retain near the surface during film formation, and during transfer, the collars covered the catcher-trap gap so that more CO2 gas could agglomerate on the trap block again. About 10<sup>6</sup> ions of <sup>146</sup>LaO<sup>+</sup> were implanted into dry ice film on the catcher in every run. After the implantation, the pre-cooled trap was moved to the frontal vicinity of the catcher, and then the catcher was warmed by stopping LN<sub>2</sub> flow. With this procedure, the atoms of <sup>146</sup>Ce and <sup>146</sup>Pr, daughter and grandchild nuclides of <sup>146</sup>La, were released from the catcher and were re-trapped on the trap together with  $CO_2$  gas. The transfer efficiency was measured by detecting the  $\gamma$ -rays emitted from each of the blocks after the transfer procedure.

**RESULTS AND DISCUSSION:** The four runs for the two types of films with different thicknesses were performed in two-days of machine time, and their transfer efficiencies were measured. The higher efficiency was obtained although the reproducibility cannot be mentioned due to the small number of runs. (see Fig.1). As in previous reports, the transfer efficiency was derived from the peak counts of 454 keV gamma-ray of <sup>146</sup>Pr for the catcher and for the trap by taken into account of the detection efficiency of each Ge detector. The ratio of the counts for the trap to those for the catcher was 12.1:1, while the previous best value was 8.8:1 (=880%) [4].

The experiments to obtain the absolute transfer efficiency are planned while searching for the better conditions for dry ice film formation. In addition to developing a highly efficient recovery method, the experiments focusing on the scattering of radioactivities during the implantation are also planned hereafter.

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Fig.1 Transfer of radioactivities using a cold trap type recovery apparatus. Gamma-ray spectra of (a) the catcher and (b) the trap after the transfer. Gamma-rays of <sup>146</sup>Ce ( $T_{1/2} =$ 14 m) and <sup>146</sup>Pr ( $T_{1/2} =$  24 m) were observed.