EXPERIMENT ON IODINE TRANSMUTATION THROUGH HIGH ENERGY GAMMA RAY

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Abstract
We carry out a research on nuclear transmutation through high energy gamma ray, to identify a way to reduce the hazards of long-lifetime radioactivity of nuclear waste. A laser Compton scattering gamma-ray facility was built on a storage ring at NewSUBARU and ~17 MeV gamma-ray photons were produced. An investigation on the reaction rate of radioactive Iodine waste was carried out. Based on the characteristics of laser Compton scattering gamma rays, a cylindrical target was adopted for the irradiation experiment. The radioactivity of the irradiated target was measured and the transmutation reaction rate was deduced.

INTRODUCTION
Some nuclear wastes have a long-lived radioactivity and are hard to dispose of. So, it is necessary to effectively eliminate the radioactivity of those nuclear wastes or convert them to less hazardous forms. Transmutation is considered as an approach to shorten the radioactive life of the nuclear waste by converting the nuclei of long-lived activity to corresponding isotopes of short-lived activity. High energy photons can induce nuclear reactions, which are regarded as an approach to get transmutation. Bremsstrahlung gamma rays are usually used to induce a photonuclear reaction, however, the poor coupling between the bremsstrahlung gamma rays and the giant resonance of nuclei is inevitable, since the energy spectrum of bremsstrahlung photons is very wide [1]. In order to improve the coupling between the gamma rays and the giant resonance of nuclei, Laser Compton scattering gamma rays seem a good alternative [2]. These gamma rays are generated from the collision of a laser light with a high energy electron beam, and there is a peak in the energy spectrum which can well overlap the peak of the nuclear giant resonance and hence realize good coupling.

As is known, 129I is a long-lived radionuclide present in nuclear waste; it has a very long life of more than 16 million years and a high chemical activity. Iodine is hard to include in the normal waste disposal packages due to its low temperature boiling point. So it is recognized as a discrete waste for transmutation.

Our aim in this paper is to experimentally investigate the transmutation reaction rate of iodine, which is defined as the number of transmuted nuclei per gamma-ray photon.

HIGH ENERGY GAMMA RAYS
A laser Compton scattering setup has been built on the NewSUBARU storage ring. The NewSUBARU ring is a racetrack shaped electron storage ring synchrotron radiation facility [3]. The circumference of the ring is about 118 m, and there are two 12 m long straight sections. An electron beam is injected from 1 GeV linac. An interaction vacuum chamber is built in one of the straight sections of the ring, where the electron beam collides with the incoming laser light in a head-to-head manner. The laser light with wavelength of 1064 nm comes from a Nd:YVO laser, consequently, the produced high energy photons are gamma-ray photons, and the maximum gamma-ray energy is 17.6 MeV.

The interaction point is designed at the center of the straight section, where both the electron beam and the laser light transverse profiles are focused onto the minimum area. A reflection mirror is located at the downstream end to guide the laser light travelling along the beamline through the interaction point, and the light is reflected out of the chamber by another upstream mirror [4]. The produced gamma-ray photons go through the downstream mirror, come into the hatch and reach the detector or irradiate the target.

A high purity germanium coaxial detector with the detection efficiency of 45% (relative efficiency) is used to measure the gamma rays. An example measured spectrum is shown in Fig. 1. The clear difference between the two signals of laser Compton scattering gamma rays and the background shows a good signal-to-noise ratio. The background signal is due to the bremsstrahlung gamma rays of the residual gases in the vacuum duct of the straight section. The maximum energy appears around 17 MeV, which is in agreement with the theoretical
prediction. After processing the experimental data, we achieved the actual gamma ray generation rate of $5 \times 10^3$ photons/mA/W/s.

NUCLEAR TRANSMUTATION

In accordance with the research on iodine transmutation, experiments have been developed in the NewSUBARU gamma ray facility to investigate the fundamental issues concerning photonuclear reactions. The final goal of this research is to understand the feasibility of transmuting the long-lived fission product $^{129}$I through the photonuclear reaction. As is known, $^{129}$I is transmuted into $^{128}$I by the $^{129}$I$(\gamma,n)$ $^{128}$I reaction. The generated $^{128}$I is unstable and is transmuted into the stable $^{128}$Xe with a short half-life of about 25 min by $\beta^-$ and $\epsilon$ decays. In such a way, $^{129}$I is transmuted to a shorter half-life nucleus.

Actually, it is hard to use a $^{129}$I target directly because of radiotoxic. Instead, we considered $^{127}$I for the transmutation experiment. Pure $^{127}$I is generally unavailable, but its compound, sodium iodide $^{23}$Na$^{127}$I, is commercially available and from it we could make a practical target. The transmutation processes for $^{23}$Na and $^{127}$I are given in Fig. 2. By absorbing a gamma-ray photon, the $^{127}$I nucleus possibly releases a neutron to undergo $(\gamma,n)$ reaction, and is transmuted to $^{126}$I. Furthermore, $^{126}$I is unstable, and eventually is transmuted to $^{126}$Xe and $^{126}$Te in the course of radioactive $\beta^-$ and $\epsilon$ decays, respectively. The $\beta^-$ in this process radiates 388.63 keV photons with a half-life of 13.11 days and intensity of 34.1%, while $\epsilon$ decay radiates 666.33 keV photons with a half-life of 13.11 days and intensity of 33.1%. The unstable $^{22}$Na transmuted from $^{23}$Na also radiates photons with energy of 1274.53 keV through $\beta^+$ decay, and its half-life is 2.6 years. In the measurement, the radiations from $^{126}$I and $^{126}$Na can be separated by the differences in photon energy and half-life.

The transmutation reaction rate should be calculated so that we can compare the calculation results with the experimental result. The estimation of reaction rate is a complicated problem, since it is related to the gamma ray energy, the geometry of the target and the distance between the target and the originating point of gamma ray generation. A Monte Carlo code, MCNP5, is used to simulate the whole process [5]. MCNP5 is a general-purpose Monte Carlo N-particle code that can be used for neutron, photon, electron, or coupled neutron/photon/electron transport. The code treats an arbitrary three-dimensional configuration of materials in geometric cells. The simulation results are given in Fig. 3. The maximum reaction rate 1.32% appears at 0.5 cm radius.

![Fig. 2 Transmutation processes of $^{23}$Na and $^{127}$I.](image)

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![Fig. 3 Reaction rate by Monte Carlo simulation for $^{23}$Na$^{127}$I samples.](image)

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The gamma rays are introduced through the hatch from the tunnel to irradiate the target. An image plate is placed before the target. The image plate can record the injected gamma-ray photons, therefore, the total number of photons that enter the target can be deduced.

Through the measurement of radioactivity of the irradiated target, the number of transmuted nuclei at the terminus of irradiation can be deduced according to the decay law

$$N_i = \frac{\Delta N e^{\lambda t}}{1 - e^{-\lambda t}}$$

where $N_i$ is the number of undecayed nuclei, $\Delta N$ is the number of decays in the duration of $\Delta t$, $\lambda$ is the decay constant, and $t$ is the time interval from the end of irradiation to the beginning of radioactivity measurement.

The cylindrical $^{23}$Na$^{127}$I target, 5 cm long and 0.5 cm in radius, was irradiated for 8 hours. After irradiation, the...
activity of the target was measured. The emission photons mainly come from the $^{126}$I because its half-life is short in contrast to $^{22}$Na. The decay curve can be derived as shown in Fig. 4, and then the half-life of the decay could be deduced. Comparing to the acknowledged value, the half-life deduced from our experiment was ~12.6 days, a little bit shorter than the acknowledged 13.11 days. The loss of the radioactivity inside the target was estimated by the MCNP5 code, and we found that 72% of the emitted photons were absorbed in the target. Also by simulation with MCNP5, we learned that the possibility of transmuted $^{23}$Na nuclei was 10% of that of $^{127}$I.

By processing the imaging plate, we determined that the total number of injected gamma-ray photons was $8.41 \times 10^{10}$. From the data processing of the activity and taking into account the loss addressed above, we found the number of transmuted nuclei was $1.15 \times 10^9$. Therefore, we concluded that the reaction rate for the $^{23}$Na$^{127}$I target was about 1.37%, as shown in Fig. 5. The simulation curve is also plotted in the figure for comparison. The experimental result showed good agreement with the simulation findings. We concluded the present experimental result was valid though only one target (0.5 cm in radius) was used in this experiment, based on our successful determination of the reaction rate of Au sample for two cylindrical targets (0.25 cm and 0.5 cm in radius) obtained by using the same method [6].

**CONCLUSION**

Iodine transmutation was studied experimentally using the laser Compton scattering gamma ray facility based on the NewSUBARU storage ring. The transmutation reaction rate of the photonuclear reaction was calculated by Monte Carlo simulation and measured in experiment, and the results were in agreement with each other. According to this reaction rate, we concluded that several tens of kilograms of $^{129}$I could be transmuted through a gamma ray generation facility, which has an accelerator with several amperes of current and a MW laser with an accumulation cavity. Details of the system will be published in the future.

**REFERENCES**

[5] The user’s manual of Monte Carlo code MCNP5