Development of the Separation System of ^{203}Tl Stable Isotope

[a]Do-Young Jeong, [a]Kwang-Hoon Ko, [a]Gwon Lim, [a]Tak-Soo Kim, [a]Hyunmin Park, [a]Sipyo Rho
[a]Cheol-Jung Kim
a Laboratory for Quantum Optics, Korea Atomic Energy Research Institute, Yusong, Daejeon, Korea,P.O.Box 105
dyjeong@kaeri.re.kr

1. Introduction

Thallium has two naturally occurring isotopes, ^{203}Tl and ^{205}Tl, with abundances of 30% and 70%, respectively. ^{203}Tl is an indispensable raw material for the production of ^{201}Tl radioisotopes by a proton bombardment in a cyclotron. ^{201}TlCl is a radiopharmaceutical used widely to diagnose a heart disease.

Development of the isotope-selective photoionization technique of Tl has been attractive, but an isotope selective ionization of thallium has never been achieved so far because of its small isotope shift as well as the lack of an available autoionization state. We have proposed a new method to separate the thallium isotopes, which is based-on an isotope-selective optical pumping followed by infrared photoionization [1].

Many photoionization methods are available, such as the two photon excitation followed by the direct ionization in a high electric field. But, other ionization methods do not have the sufficient selectivity for a single stage. Two or three stages have to be applied for obtaining the sufficient selectivity. Moreover, they need strict experimental conditions and are expected that the efficiency decreases in the ionization step. However, our method is expected to overcome these drawbacks.

With this background, we developed the laser isotope separation system to have high isotopic abundance of ^{203}Tl. The system configuration and characteristics are represented in this paper.

2. Theoretical Background

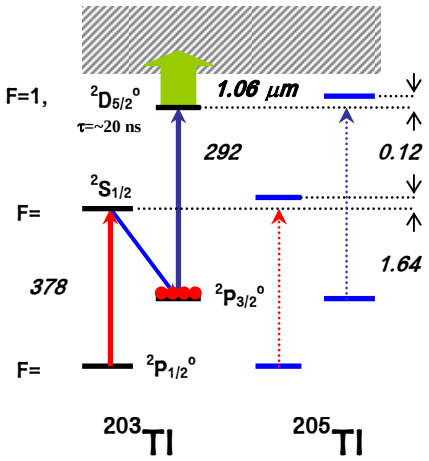

Figure 1. Isotope selective optical excitation and ionization pathway.

The atomic structure and the ionization pathway for the separation of ^{203}Tl isotope are shown in figure 1. Ground state (6 $^2P_{1/2}$) thallium atoms were optically and isotope-selectively pumped into a metastable level (6 $^2P_{3/2}$) by a continuous wave laser (378 nm). The metastable atoms were then photoionized with no additional selectivity to continuum via an excited state (7 $^2D_{5/2}$) by a pulsed UV (292 nm) laser and a Nd:YAG laser. The isotope selectivity of the scheme was calculated by the density matrix equation and we compared it with other laser based methods.

3. System Configuration

The separation system is shown in figure 2. It consisted of an optical pumping laser system, a pulsed UV laser system for the optical excitation, an ionization laser system, and a separation chamber.

The Optical pumping laser system consists of a Ti:S laser, a second harmonic generator, and a frequency locking unit. The second harmonic generator created the 378 nm UV laser up to 1 W. The frequency locking unit makes it possible to select only one isotope in spite of the thermal drift of the reference cavity of the laser by controlling actively the Ti:S laser's frequency. The optical pumping laser frequency could be locked to the corresponding transition line for 20 hours with the frequency stability of less than 1 MHz. It was incident on the interaction chamber after expanding the beam diameter up to 1 ~ 2 cm.

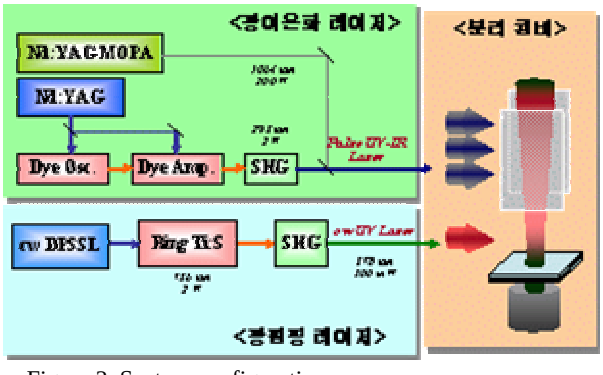

Figure 2. System configuration.

The dye laser system for generating 292 nm laser by the second harmonics consisted of an oscillator and three amplifiers for creating the average power of 25 W and the pulse duration of 30 ns. This system was pumped by two pulsed green lasers with the total powers of about 100 W. The frequency double laser beam of about 2 W

was created by passing through a SHG crystal and was applied for the optical excitation from the metastable state to the highly excited state.

The direct ionization technique to above the ionization limit was chosen for an ionization step. The ionization cross section was measured for determining the sufficient laser pulse energy condition [2]. The pulse energy is needed 40 mJ in order to obtain the ionization efficiency of 80%. In our experiment, the IR laser was normally operating at 20 mJ (200 W) which was corresponding to the ionization efficiency of 50%. The ionization laser pulses had to be temporally and spatially overlapped with the ultra-violet laser pulses.

The separation chamber was composed of a vacuum pump, an atomizer, an extractor and multi-pass optics. The ions that were created by the lasers were extracted by the electric field in the interaction region. The multi-pass optics was set up for the high interaction efficiency between the metastable atoms and UV/IR lasers. The corresponding interaction region was lengthened to 5 cm by the multipass mirrors. The atomizer was heated up to 920 C and the electric field was provided over 1 kV/cm for an extraction.

4. Experimental Results

The frequency stabilized 378 nm continuous laser was incident on the separation chamber. It induced the fluorescence from the atoms which was used for the adjustment of the mirrors in the chamber. The ultraviolet and infrared lasers were then combined and incident on the chamber at the above the optical pumping beam. They didn't have to overlap with the CW laser beam. The wavelength of the UV laser was controlled by tilting a grating in an oscillator at the position on which the ion signal became maximized. In result, the ion current as shown in figure 3 was monitored in an oscilloscope.

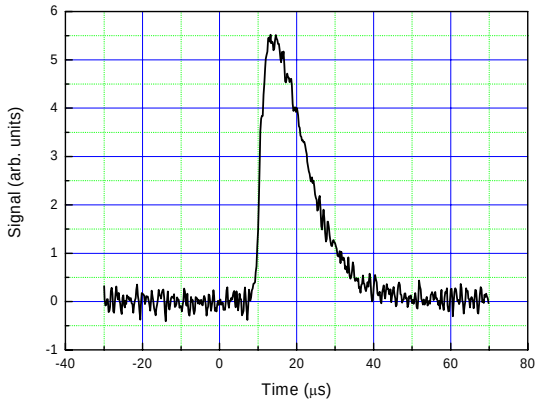

Figure 3. Ion signal monitored by an oscilloscope. The repetition rate of the signal is 10 kHz.

The thallium coated extraction plate was dissolved in the 10% nitric acid for about 1~3 days. The resultant abundance of ^{203}Tl was measured as 91% by a TIMS and an ICP-MS. The abundances of ^{203}Tl are depicted in figure 4 before and after the separation.

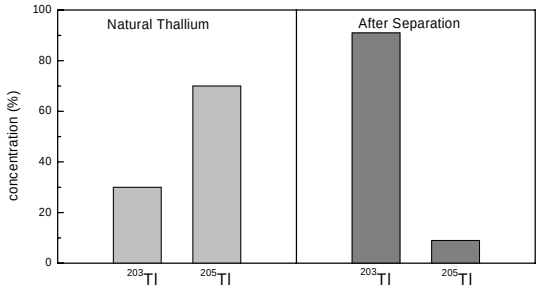

Figure 4. The abundances of the thallium sample after the separation process.

5. Conclusion

The thallium separation system has been developed to enrich ^{203}Tl isotope which is useful for the radiopharmaceutical after the collision in cyclotron. The abundance of ^{203}Tl has been obtained up to 91% after the single stage separation. This value is higher than that of other laser based techniques at the single stage separation. We expect that our ionization becomes a very useful method to separate ^{203}Tl isotope.

REFERENCES

[1] D.Y. Jeong, K.H. Ko, G. Lim, C.J. Kim, "Method for Separation of Thallium", Patent no. 478533, Korea.

[2] K.H. Ko, G. Lim, T.S. Kim, J. Kim, S. Rho, H. Park, C.J. Kim and D.Y. Jeong, "Selective Ionization of Thallium Stable Isotopes based on Optical Pumping followed by Infrared Photoionization", International Conference on Laser Probing, (2004)