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Development of ^{14}C Production Process by Utilizing HANARO

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1. Introduction

Feasibility study on the production of ^{14}C by using the HANARO has been performed. As an advantageous tracer, ^{14}C has been used in various chemical applications such as in the drug discovery and development [1], clinical studies [2], and environmental fields [3]. The advantageous properties of ^{14}C come from its long half-life (5745 years), which makes sure the ^{14}C-labeled compounds can be used for indefinite time in controlled conditions and its emission of only low-energy beta particles, which provides safer and simpler working conditions.

^{14}C as chemical forms such as barium carbonate ($Ba^{14}CO_3$), methane ($^{14}CH_4$), benzene ($^{14}C_nC_{6-n}H_6$), etc are produced in few countries, such as United States, Russia, and UK. Approximately 28.6Ci of ^{14}C was imported in 2005 and mostly consumed by the research or educational organizations [4]. Even though ^{14}C has not been produced in the nation, a local company has well established the labeling technologies of ^{14}C, and commercially produces the labeled compounds.

Hence, the feasibility of the production of ^{14}C from the HANARO is evaluated experimentally. In this paper, the experimental results will be compared with the results from a numerical analysis, which was reported previously [5] to support the experimental results.

3. Experiments

Fifty milligrams of aluminum nitride (AlN, 99.9%, Atlantic Equipment Engineers Co.) powder is employed as the target material. The powder is pelletized in 3.0mm diameter and by using a custom-made die. The pellet is calcined at 1,000℃ for 30minutes in a pure nitrogen stream (99.999% purity). After the calcination, the weight of the pellet is measured, introduced in a 5mm quartz tube, and then double encapsulated in the irradiation capsule, which is specially designed for the irradiation holes of the HANARO.

Irradiation is performed at the IP-5 hole, in which the average neutron flux is 4.84 x 10^{13} n/cm^2/s, for 2 cycles (total 41 days). After the irradiation, the target is cooled for sufficient time to make sure enough decays of impure radioisotopes.

The recovery system of ^{14}C is set up as shown in Figure 1. The system is kept pursing with N_2 gas (10ml/min) during the course of the processing. Oxidation catalyst bed is heated up to 750℃ and maintained until the process ends. In a digestion solution composed of 2g $SnCl_2$ dissolved in 30ml of concentrated H_3PO_4, the irradiated AlN pellet is introduced. The solution is heated up to 100℃, and 20ml of chromic acid is added. The gas from the catalyst bed is passed through a heat exchanger for cooling and scrubbed twice in vessels containing each 30ml of 2.5M NaOH solution.

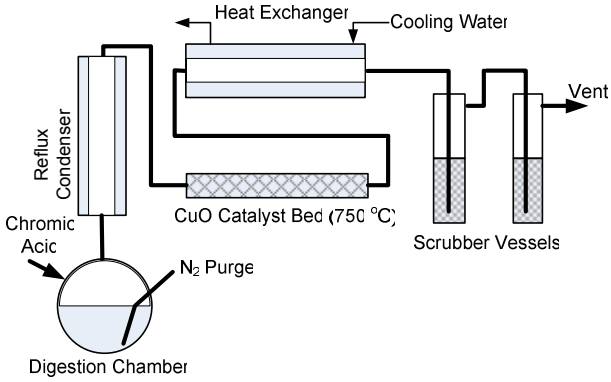

Figure 1. Recovery System of $^{14}CO_2$ Librated from Irradiated AlN Pellet

Activities of ^{14}C in the scrubber solutions and the digestion solution are measured by using a liquid scintillation counter (Perkin Elmer, Model TriCarb-2900TR). For the cocktails, 0.05ml of each active solution is added to each 15ml of Ultima Gold, well shaken, and stored at room temperature for at least 2 hours before the measurements.

4. Results and Discussions

^{14}C is generated by (n,p) reaction as follows;

$$^{14}N_7 + {}^{1}n_0 \rightarrow {}^{14}C_6 + {}^{1}p_1, \qquad \sigma = 1.81b \quad (1)$$

Several different target materials have been considered for the production of ^{14}C. The most concern in the production of ^{14}C is the thermal and radiological stabilities of the material because the formation of ^{14}C requires a long irradiation time (i.e. 1 ~ 3 years) to produce economically viable quantity as the cross-section of ^{14}N is relatively small. Aluminum nitride is usually employed as a thermal resistant ceramic material(disintegration temperature:2200℃). Aluminum nitride is palletized and calcined at 1,000℃ in a nitrogen stream to remove unwanted impurities. By the calcination, the weight loss is less than 1.0mg, which

means degradation of the ceramic or loss of nitrogen element is negligible.

The radioactivity of the pellet before the recovery processing is measured with a dose calibrator for any possible γ-emitting radioisotopes. However it is realized that the pellet does not emit noticeable γ radiations at all.

For the recovery of $^{14}CO_2$, a wet method is adopted from other's work [6] and modified to ensure better oxidative conditions. The liberated $^{14}CO_2$ from the digestion vessel is trapped in subsequent scrubber vessels. To ensure reasonably recovery efficiency, strong oxidizing agent (chromic acid) is introduced to the digestion vessel. Also, oxidation catalyst bed (copper oxide) is installed to further oxidize carbon monoxide to carbon dioxide since carbon monoxide is not readily trapped in the sodium hydroxide solution.

From the measurement of LSC, the activities in the first and the second scrubber solutions are 8.76 ± 0.03 and 0.73 ± 0.02 μCi, respectively. The activity of the digestion solution is approximately 0.07 ± 0.03 μCi. Neglecting the loss ^{14}C during the recovery process, total amount of ^{14}C produced by the irradiation followed by the processing is 9.5 μCi. It was estimated from our previous numerical study that approximately 8.92 ~ 10.2 μCi could be produced by the irradiation at IP-5 hole of the HANARO. Hence, the measured activity well agrees with the results from the numerical analysis.

Based on this comparison, it can be estimated that the production rates of ^{14}C at the holes with higher neutron flux (i.e. IR, OR) than that of IP-5 are 0.5 (IP-15, 1.52 x 10^{14} n/cm^2/s) ~ 1.5 (IR-1, 6.65 x 10^{14} n/cm^2/s) Ci per year when 100 grams of aluminum nitride is irradiated.

Hence, there is enough possibility to produce ^{14}C for the local uses, if a long period of irradiation (i.e. 1 ~ 3 years) at unoccupied positions of the holes with high neutron flux at the HANARO.

5. Conclusions

Experimental studies have been performed to estimate the possibility of ^{14}C by using HANARO. As the target material for the production of ^{14}C, aluminum nitride is used. Forty days of irradiation is performed for the 50mg of AlN pellet at IP-5 hole. After the recovery processing, 9.5 μCi of ^{14}C is produced. This activity is within the range of the predicted values by the numerical analysis from our previous work. It is concluded from this experimental study that there is enough potential to produce economically viable quantities of ^{14}C by using unoccupied positions of the irradiation holes at the HANARO.

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