



#### 4.43 **New Technique for Determination of long-lived radioisotopes, Iodine-129, Using Multiparameter Coincidence Spectrometry**

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### ABSTRACT

Multiparameter coincidence  $\gamma$ -ray spectrometry based on g-g coincidence is widely used in the field of nuclear structure studies, and has produced many successful results. In this study, feasibility of the method for neutron activation analysis of long lived iodine isotope,  $^{129}\text{I}$ , was investigated.

**KEYWORDS:** Iodine-129, Multiparameter coincidence spectroscopy, Neutron Activation Analysis

### 1. INTRODUCTION

Iodine-129 is an important radioisotope in environmental study, because its long half life ( $1.57 \times 10^6$  year). Iodine-129 is produced in nature as a result of spontaneous fission of heavy elements in the earth's crust and reaction of xenon with cosmic rays. Recently significant amount of radioactive iodine has been emitted to environment from nuclear reprocessing plants and nuclear test explosions. To study the contribution of the artificial radioactive iodine, the development of a reliable and practical method for the determination of  $^{129}\text{I}$  is required. In this study, we developed a new technique for the determination of  $^{129}\text{I}$  using multiparameter coincidence spectrometry.

In the case of neutron activation analysis, measurements of  $\gamma$ -rays from trace elements are strongly interfered by the  $\gamma$ -rays from major elements, e.g.,  $^{24}\text{Na}$ . So usually chemical separation processes are required to eliminate the major elements for detection of the small peak of gamma ray from  $^{129}\text{I}$ . Multiparameter coincidence spectroscopy using 2 Ge detectors was already applied to neutron activation analysis<sup>(1)</sup>. This method, however, has the disadvantage of low efficiency. In this study, to improve the low efficiency of multiparameter coincidence spectrometry, an array of 12 Ge detectors with BGO Compton suppressors was used. Iodine samples were irradiated at JAERI's research reactors with neutron flux  $3\text{-}5 \times 10^{13} \text{ ncm}^{-2}\text{s}^{-1}$ . Gamma-gamma coincidence of multiple  $\gamma$ -rays from the radioisotopes produced by neutron capture reactions was measured with a array of 12-Ge detectors with BGO Compton suppressors, GEMINI<sup>(2)</sup>, located at the tandem accelerator facility in JAERI. We already applied this method to the determination of trace elements in the rock samples<sup>(3)</sup>. In this study we also applied the multiparameter coincidence method for the determination of low level of  $^{129}\text{I}$ .

## 2. EXPERIMENTAL

### 2.1 Sample preparation

The ratio of  $^{129}\text{I}/^{127}\text{I}$  in the algae sample collected from Ibaraki prefecture in Japan was measured by neutron activation analysis using multiparameter coincidence measurement by making use of the  $^{129}\text{I}(n,\gamma)^{130}\text{I}$  reaction. About 100 grams of the algae sample was carbonized. After chemical purification, the iodine was precipitated as  $\text{PbI}_2$  (56mg). The scheme of the chemical procedure is shown in Fig. 1.

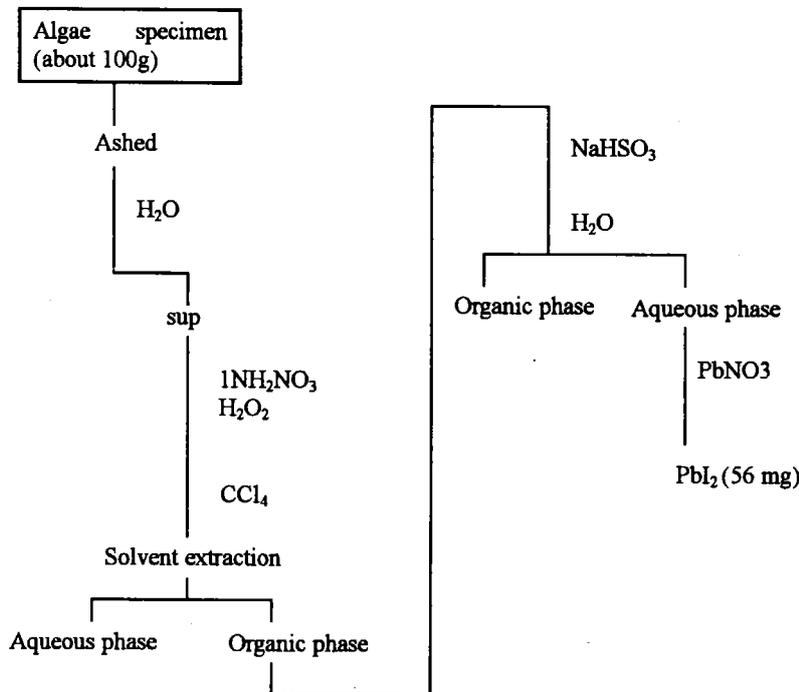


Fig.1 Scheme of the chemical separation procedure of iodine purification from algae sample

### 2.2 Irradiation

The sample was sealed in a quartz tube with 6 mm diameter. Two standard samples containing known amounts of natural iodine (20 mg) and  $^{129}\text{I}$  (12 ng) were also prepared. The samples were irradiated for 2 hours in the T-pipe of JRR-4 research reactor with thermal neutron flux of  $5 \times 10^{13} \text{ ncm}^{-2}\text{s}^{-1}$ .

### 2.3 Measurement

After irradiation,  $\gamma$ - $\gamma$  coincidence measurement of the algae sample was performed without any radiochemical purification using GEMINI. The energy resolutions of the Ge detectors were 2.0-2.3 keV FWHM at 1.3 MeV, and the typical detector efficiencies were 40-70 % relative to 3" x 3" NaI detector. The double folded  $\gamma$ - $\gamma$  coincidence events were recorded on a magnetic tape event by event. The Iodine sample collected from algae was measured for 22 hours, while the standard samples of  $^{129}\text{I}$  and natural iodine were measured for 10 hours and 3 hours, respectively. All the samples were measured together with a  $^{133}\text{Ba}$  standard source to perform dead-time calibration for the detector system. The total amount of iodine in the algae sample was calculated using the  $^{127}\text{I}(n, 2n)^{126}\text{I}$  reaction of the standard sample of natural iodine. A 4096 x 4096 channel matrix was created by off-line sorting of the experimental data. Fig.2 shows a partial decay scheme of  $^{130}\text{I}$ , which includes some dominant gamma ray energies. In this study, the 536-740 keV pair was analyzed because of the low background.

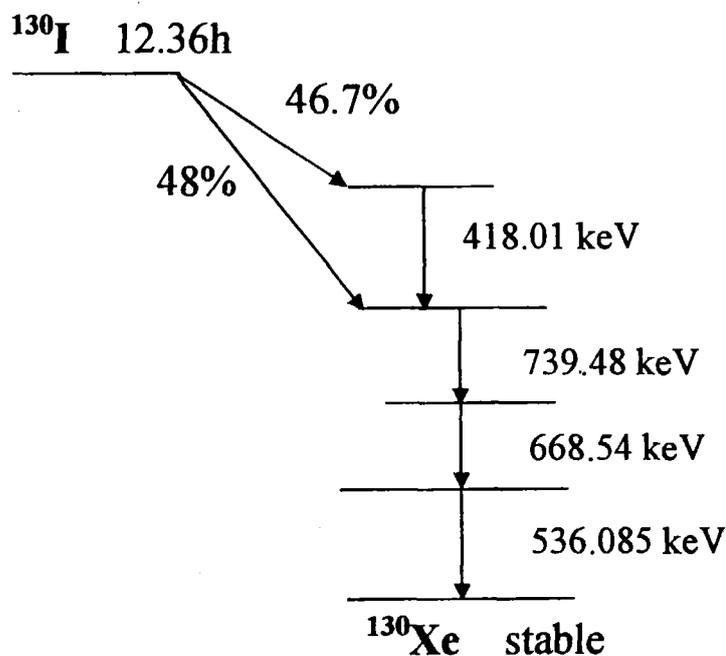


Fig.2 Partial decay scheme of  $^{130}\text{I}$ .

### 3 Results and Discussion

Figure 3 shows a coincidence  $E\gamma$ - $E\gamma$  matrix obtained from the algae sample. In the matrix the  $\gamma$ - $\gamma$  coincidence peak can be observed at 536-740 keV. The isotopic ratios of  $^{129}\text{I}/^{127}\text{I}$  is determined to be  $3.5 \times 10^{-10}$  with the reported data<sup>(4)</sup>. The ratio measured in this work was consistent statistically

with that of algae from other regions of Japan. Detection limit of this method is estimated to be about  $^{129}\text{I}/^{127}\text{I} = 10^{-13}$

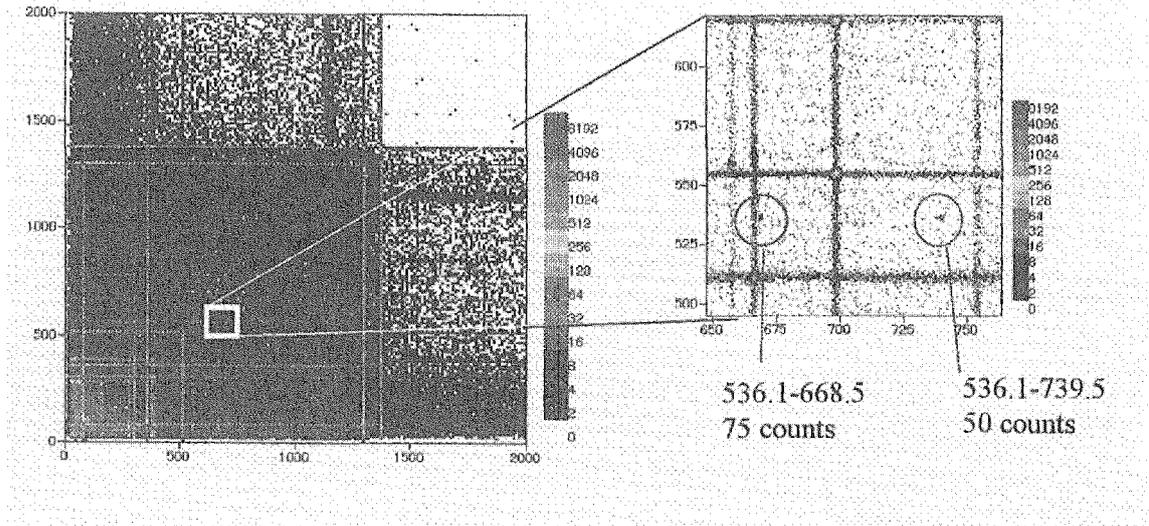


Fig. 3 Two-dimensional matrix obtained from  $\text{PbI}_2$  obtained from the algae sample.

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