Determination of Sodium and Copper in Ohasari Sea Shell By Radiochemical Neutron Activation Analysis

Sudi Ariyanto

Abstract

Determination of Sodium and Copper in Ohasari Sea Shell By Radiochemical Neutron Activation Analysis. Elemental analysis of Sodium and Copper in Ohasari sea shell has been performed by radiochemical NAA. The concentration of Na in Ohasari shell is found to be 4745.00 ± 119. 29 ppm. The concentration of Cu in Ohasari shell is found to be 17.29 ± 1.76 ppm. Ohasari shell contains some elements such as Ta-182, Pt-l97, 1-131, 1-132, 1-133, Ce-144, Ce-141, Sc-47, Te-132, Hg-203, Au-198, Ru-103, Cu-64, As-74, As-76, Sb-122, Na-24.

Abstract

Determination of Sodium and Copper in Ohasari Sea Shell By Radiochemical Neutron Activation Analysis. Telah dilakukan pengukuran Sodium dan Tembaga dalam kulit kerang jenis Ohasari dengan menggunakan NAA dan radiokimia. Ditemukan bahwa konsentrasi Na dalam kerang 4745.00 ± 119.29 ppm, sedangkan konsentrasi Cu be 17.29 ± 1.76 ppm. Beberapa unsur yang terkandung di dalam kulit kerang Ohasari adalah Ta-182, Pt-197, 1-131, 1-132,1-133, Ce-144, Ce-141, Sc-47, Te-132, Hg-203, Au-198, Ru-103, *Cu-64*, As-74, As-76, Sb-l22, Na-24.

Introduction

Neutron Activation Analysis (NAA) is a nuclear analytical technique for quantitative multi-element analysis. The principle of NAA is to transform stable nuclides into radionuclides by nuclear reaction. This nuclear reaction is induced by the incident neutron. After the irradiation, the characteristic gamma rays emitted by the radionuclides are quantitavely measured by gamma spectroscopy. At present, NAA is done automatically using a computer assisted gamma-ray spectrometer and high performance Ge detector. Sometimes radiochemical treatment is necessary for some reasons:

1. The photopeak to measure overlaps with other peak when irradiated sample contains nuclides that also emits gamma-rays with the same energy.
2. When peak area is too small to detect and when it is influenced by spectra of major nuclides. This condition will become more serious when the interfering nuclides have similar or longer half-lives.

Basic Principle

In general, when a reaction takes place in neutron activation it is written as belows:

$$_{n}X(n,ry+IX \quad .$$

If there are Na and Cu in the same sample, then it will be difficult to analyze Cu. Activated sodium (^{24}Na) emits 2 gamma rays with the energy of 1.369 MeV and 2.754 MeV. On the gamma-ray spectrum of 24Na, the annihilation radiation at 0.511 Me V appears as a the result of gair production effect. On the other hand, Cu nuclide emits mainly 0.511 MeV of annihilation radiation(38%) and 1.34 MeV gamma ray (0.5%). The half-lives of 24Na is slightly longer than that of 64Cu. Due to this condition, the presence of sodium will interfere with the detection of 64eu and it then becomes necessary to remove 24Na from the sample in order to get a reliable result for MCU.

In order to get solid copper from the Ohasari sample is dissolved in hydrochloric acid. Into this solution is also added cupric ion carrier and also Zn ribbon with 99.9% zinc content.. The reaction can be given as follows:

$$Zn(solid) + 2W \sim Zn^{2+} + H2 \ (gas)$$
$$Cu^{2++} + Zn \ (solid) \sim Cu \ (solid) + Zn^{2+}$$

In this experiment, a comparative method is utilized to define the concentration of the sample. Comparative method is a method in which an unknown sample is compared to the amount and the count rate of an identical element which is used as standard. For this purpose, both the sample and the standard are irradiated under the same irradiation condition. The quantity of each element in the sample is shown by the below relationship,

$$W_x = W_o \cdot \frac{n_x}{n_o}$$

Simple manipulation of the above equation leads to the equation below,

$$\frac{W,}{n_x} = \frac{W_o}{n_o}$$

Wx and W_o are the amount of the unknown element and the standard respectively, while n, and fie, are the counting rates from the unknown element and the standard.

This method is quite simple and gives a reliable result.. Cumulative errors can be minimized because there are fewer factors involved in the calculation or analysis.

Standard deviation

Standard deviation for concentration obtained in the comparative method is calculated as the following:

where C is the calculated concentration, ax is the standard deviation of the sample and ao is

$$\sigma_c = C\sqrt{\left(\frac{\sigma_x}{n_x}\right)^2 + \left(\frac{\sigma_o}{n_o}\right)^2}$$

sea shell, firstly the irradiatied the standard deviation of the standard. ax and c, are given as follows:

or

$$\sigma_x = \frac{\sqrt{N_x}}{t_x}; \quad \sigma_0 = \frac{\sqrt{N_o}}{t_o}$$
$$\sigma_x = \sqrt{\frac{n_x}{t_x}}; \quad \sigma_0 = \sqrt{\frac{n_o}{t_o}}$$

where N, and No are the counts for the sample and the standard respectively, while tx and t0 is the measuring time for the sample, and the standard, n, and fie are the counts rates for the sample and the standard.

The objective of this experiment is to analyze Na and Cu in the Ohasari Sea Shell by incorporating radiochemical separation into NAA

Experiment

Standard Sample Preparation

Sodium standard sample is prepared as the following:

1. Weigh purified sodium chloride which is equivalent to 0.25 g of sodium and place it into a 500 ml glass beaker. Put 200 ml of purified water into the same beakar and dissolve it.
2. Transfer the sodium chloride solution into a 250 ml volumetric flask and adjust the solution to the 250 mllino of the flask with purified water.
3. Transfer the sodium salt solution into a glass bottle marked as "1000 ppm Na"
4. Take 50 J.ll of the solution with a micro-pipetter and put it onto 25 mmo filter paper
5. Dry the filter paper under a 250 W infrared lamp

6. Put the dried filter paper into a plastic bag and then seal the plastic bag with an electric plastic sealer

The above procedure is also implemented for Cu standard. In this case metallic copper is dissolved into nitric acid and then 10 µl of the solution is put onto filter paper. This sample is dried using infrared lamp.

Both the standard sample and the sample to be analyzed are put into a plastic tube and then irradiated simultaneously with neutrons using a pneumatic transfer tube of JRR-3 research reactor at Tokai Research Establishment of JAERI. These samples are irradiated for 2 minutes.

Experimental Procedures

In this experiment, the following reagents and apparatuses are needed:

Hydrochloric acid (4M HCl), concentrated nitric acid (RN03), nitric acid (2M HN03), copper carrier solution (5 mg *Cu2+/ml),* ethanol (C2H50H), metallic zinc ribbon, hot plate, infrared lamp, filtration apparatus, chemical balance, tall beaker (100 ml), ordinary beaker (100 ml), plastic beaker (l l.), funnel, volumetric pippete (5 ml), Pi-pump, measuring cylinder (50 ml), watch glass, filter paper (5C, 9cm~), glass filter paper (24 mmo), glass rod, tweezers, spoon, plastic tube, thick paper (5x5 em).

The procedure of this experiment is outlined as follows. The first step is sampling and sample preparation(including the standard). The prepared samples are irradiated in JRR-4 with neutron flux of 6.10^{13} ern" s-I. In the next step, non-destructive analysis (leaching) is performed on the sample and then Na spectrum is measured and then its concentration is calculated. Radiochemical analysis is performed to exclude Na from the samples. In this radiochemical process Cu solid is obtained and its spectrum is analyzed and its concentration is calculated.

Non-destructive analysis of sodium is outlined as the following:

1. Put 2 rnl of 2M nitric acid, distilled water and ethanol each into 100 rnl beaker separately. Put all 3 beakers on a tray and then placed in a ventilated draft. chamber
2. Wear rubber gloves. Put the irradiated shell sample into nitric acid and leave it for about 20 seconds to remove surface contamination by etching. Then using tweezers wash the sample with water and ethanol successively, and dry the shell specimen under an infrared lamp for about 10 minutes.
3. Put the sample into a plastic tube that is already weighed then weigh it using an electronic balance. Write down the sample net weight Ws.
4. Place the sodium standard sample on a sample holder at a distance of about 30 em from the Ge detector.. Collect the gamma ray spectrum for 30 seconds using 4096 channels analyzer (MCA). Adjust ROI to the photo-peak corresponding to 1.369 MeV gamma-ray of 24Na and read the peak area counts.
5. Replace the standard sample with the ohasari sample and collect data as was done in Step 4 above.

To analyze the concentration of Cu, the same sample used to analyze Na is processed chemically to get solid Cu and to remove all Na. The procedure of radiochemical separation process is outlined as the following:

l. Put 20 ml of 4 M HCl in a 100 ml tall beaker, add 5.00 ml of copper carrier solution (25 mg Cu) using a 5ml volumetric pipette, and place the beaker on a tray in a draft. chamber
2. Put the irradiated shell sample into the solution. Cover the beaker immediately with a watch glass
3. After the gas generation ceases, add 1-2 drops of concentrated HN03 to preserve the copper ions in the oxidized

form of Cu2+, then filter it with 5C 9 erne filter paper to remove any insoluble substances still remaining in the solution. Collect the filtrate in a 100 ml tall beaker. Discard the used filter paper..

Result and Discussion

The energy that is used to analyse Na in the sample and standard is 1.369 MeV.. The measurement results and the data to calculate Na concentration in Ohasari shell is shown in Table 1.

Table 1 Experiment data and results for Na

	W,mg	count	time,s	count/s
Standard	0.5	1933.00 ± 43.97	30	64.43 ± 1.47
Sample	475.7	8727.00 ± 93.42	30	290.90 ± 3.11

The comparison between sample and the standard results in the value of Na concentration in Ohasari shell is about 4745 ± 119.29 ppm.

Table 2 Experiment data and result for Cu

	W,mg	count	time,s	count/s
Standard	0.01	2451± 49.51	20	122.55 ±2.48
Sample	475.7	100±10.00	200	0.50 ±0.05

The Table 2 shows the data and the measurement results for Cu concentration in the Ohasari shell.. The energy of Cu that is used for calculation is 0.511 MeV and the carrier recovery is 49.7%.. The comparison between sample and the standard results in the value of Cu concentration in Ohasari shell is about 17.29 ± 1.76 ppm.

A qualitative analysis is also performed to detect elements in the shell. Several samples prepared by participants of a domestic training course are also included in the measurement.. The result on the gamma spectrum is shown in Figure 1. It can be known from this spectrum that there are many elements in Ohasari shell: Ta-182, Pt-197, 1-131,1-132,1-133, Ce-144, Ce-141, Sc-47, Te-132, Hg-203, Au-198, Ru-103, Cu-64, As-74, As-76, Sb-l22, Na-24. The existence of Na-24 in the gamma spectrum. shows that the elimination process of Na in some samples is not performed well.

Conclusion

The measurement of Na and Cu in Ohasari shell have been performed and the following are clarified:

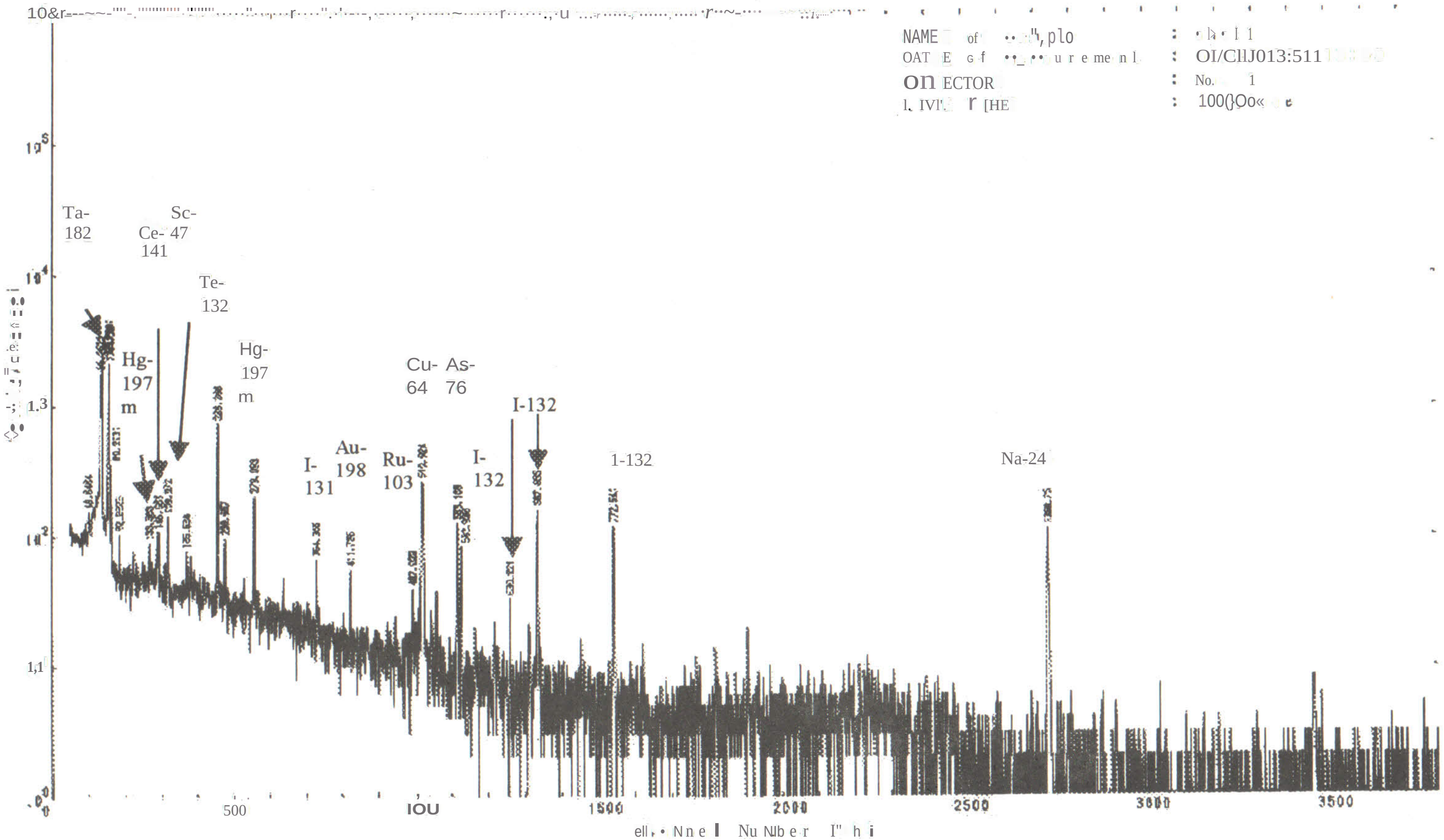
1. The concentration of Na in Ohasari shell is found to be 4745 ± 119.29 ppm.
2. The concentration of Cu in Ohasari shell is found to be 17.29 ± 1.76 ppm.
3. Ohasari shell contains some elements such as Ta-182, Pt-197, 1-131, 1-132, 1-133, Ce-144, Ce-141, Sc-47, Te-132, Hg-203, Au-198, Ru-103, Cu-64, As-74, As-76, Sb-I22, Na-24.

Reference

I. Ko Noguchi, Radiochemical Neutron Activation Analysis: Determination of Sodium and Copper in Sea Shell, Lecturing Note ITP 2001, NuTEC, Tokyo.

Acknowledgement

The author would like to express his acknowledgement to Dr. Hiroshi Kamioki, Dr. Kouhei Kushita, Dr. Ko.Noguchi for their assistance in doing experiment in NuTec Laboratory of JAERl.


NAME
ECTOR
No. 1
Ta-182
Ce-141
Sc-47
Te-132
Hg-197m
Hg-197m
I-131
Au-198
Ru-103
Cu-64
As-76
I-132
I-132
1-132
Na-24
500
1500
2000
2500
3000
3500

15