Assay of a Ceramic Sample by Neutron Activation and Proton-Induced X-ray Emission

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Several techniques of multi-element analysis using reactors and accelerators have been developed. There are two techniques of neutron activation analysis: instrumental neutron activation analysis (INAA) and prompt gamma-ray analysis (PGA).[1-5] The conventional INAA requires the neutron-induced production of radionuclides emitting gamma-rays with adequate half-lives. INAA could not be used for radionuclides with typical half-lives of 10^{-14}- to 10^{-12} s. This limitation can be overcome by applying the technique of PGA. For a number of elements that do not produce good delayed products, the analytical sensitivity of PGA has been estimated to be better than that of INAA.[1] For the assay of the samples by PGA and INAA, the external standard samples need be prepared by matching their matrices and considering geometrical arrangement in order to reduce problems caused by neutron absorption and scattering[6,7] and gamma-ray absorption. Nakahara and his coworkers[8] have recently shown that those problems could be overcome using an internal mono-standard method. Multi-element analysis can be also performed by proton-induced x-ray emission (PIXE) using accelerator, which is based on the detection of characteristic x-ray. Ionization cross sections for K and L subshells were calculated and tabulated[9] for protons and helium ions. Hence, PIXE has become an effective technique in multi-elemental analysis of both thick and thin target samples.[10] In this work ceramic samples were assayed by PGA and PIXE and their results are compared.

Experimental Section

Ceramic samples provided by Boon Won Ceramics have been assayed by PGA and PIXE techniques. The PGA experiment was performed at the thermal neutron beam port (T1-4-1) of the JRR-3M reactor at Japan Atomic Energy Research Institute (JAERI), where the typical beam size on the sample was 20×20 mm^2 and the flux was 2.4×10^7 n cm^{-2} s^{-1}.[4,5] The measured amount of each sample was sealed in fluorinated ethylenepropylene resin (FEP) film, and then placed in an air-tight sample chamber made of polytetrafluoroethylene (PTFE, Teflon). The sample chamber was filled with He gas to reduce γ-rays from the background. The multi-mode gamma-ray spectrometer at the facility consists of a high purity Ge detector, BGO (bismuth germanate, $Bi_4Ge_3O_{12}$) anti-Compton shielding detectors, and a pulse height analyzer system controlled by a personal computer. The Ge detector was located 24.5 cm away from the sample and with its axis perpendicular to the beam. The prompt gamma-ray measurement was performed in three modes: singles, Compton suppression, and pair modes. The PIXE assay of the samples has been done by bombarding with protons at Korea Institute of Geology, Mining and Materials. The proton energy used in this study was 2.43 MeV.

Results and Discussion

The central portion of the ceramic sample has been assayed by PGA and PIXE techniques. Concentrations of nine elements, such as Na, Mg, Al, Si, K, Ca, Ti, Mn and Fe, have been deduced by PGA technique using the internal mono-standard method.[8] The assumption that the oxides of these observed elements comprise 100% of the composition of the sample was used in the analysis. In this method, the ratio of the photopeak areas of element 1 to element 2 is expressed by the following equation:

$$\frac{A_1}{A_2} = \frac{n_1 \cdot I_1 \cdot \int [\int \Phi(E_n, \boldsymbol{r}) \sigma_1(E_n) dE_n] \varepsilon_1(\boldsymbol{r}) d\boldsymbol{r}}{n_2 \cdot I_2 \cdot \int [\int \Phi(E_n, \boldsymbol{r}) \sigma_2(E_n) dE_n] \varepsilon_2(\boldsymbol{r}) d\boldsymbol{r}} \quad (1)$$

where A is photopeak area; n, number of atoms; I, number of photons emitted per neutron capture; Φ, neutron flux; σ, neutron capture cross section; E_n, neutron energy; $\boldsymbol{r}$, position vector from the origin in the detector to the point where neutron capture occurs.

Eq. (1) can be approximated by replacing $(\Phi(E_n, \boldsymbol{r}) \cdot \sigma(E_n))$ with $(\Phi_0 \cdot \sigma' \cdot w(\boldsymbol{r}))$ where Φ_0 is normalized neutron flux, σ', effective neutron capture cross section, and $w(\boldsymbol{r})$, normalized spacial density distribution of prompt gamma-ray source. As a result of this approximation, Eq. (1) can be represented by the following equation.

$$\frac{A_1}{A_2} = \frac{n_1 \cdot I_1 \cdot \sigma_1' \cdot \int w_1(\boldsymbol{r}) \cdot \varepsilon_1(\boldsymbol{r}) d\boldsymbol{r}}{n_2 \cdot I_2 \cdot \sigma_2' \cdot \int w_2(\boldsymbol{r}) \cdot \varepsilon_2(\boldsymbol{r}) d\boldsymbol{r}} \quad (2)$$

As shown in Eq. (2), the normalized neutron flux Φ_0 was canceled out between the numerator and the denominator, and the relative photopeak efficiency is expressed as the term $\int w(\boldsymbol{r}) \cdot \varepsilon(\boldsymbol{r}) d\boldsymbol{r}$. The relative photopeak efficiency

Table 1. Concentrations of elements observed in the ceramic sample

lement	concentrations (%)	
	PGA	PIXE
Na	0.57 ± 0.03	
Mg	1.00 ± 0.21	
Al	14.3 ± 0.5	16.5 ± 0.9
Si	31.1 ± 0.3	32.1 ± 1.7
K	1.97 ± 0.02	2.54 ± 0.13
Ca	0.34 ± 0.03	0.38 ± 0.02
Ti	0.12 ± 0.01	0.11 ± 0.01
Mn	0.021 ± 0.003	0.016 ± 0.002
Fe	0.72 ± 0.03	0.94 ± 0.05

was obtained by plotting an efficiency curve from the γ-rays of the elements Ti and Si. In this work the 341.7- and 1381.7-keV γ-rays of ^{49}Ti and 1273.3-, 2092.9- and 3539.1-keV γ-rays of Si were used. The values of the number of photons emitted per neutron capture were taken from the compiled data.[11] The relative efficiency curve was drawn on the assumption that it is linear in the energy range between 300 and 3000 keV on log-log graph paper. The linear tendency was observed even up to 3539.1 keV.

The most capture γ-rays used in the elemental analysis were the prompt ones, while the 1778.9-keV γ-ray used for the Al analysis the delayed one. The 1778.9-keV γ-ray is emitted in β^--decay of ^{28}Al isotope whose half-life is 2.25 min. The principal prompt γ-rays used in the elemental analysis for Na, Mg, K, Ca, Mn, and Fe were 870.1, 1808.9, 770.3, 1942.0, 314.4 and 352.4 keV, respectively. In order to check the neutron flux incident on the gamma-ray Ge detector during sample irradiation, the 596-keV peak induced by the capture of thermal neutrons by the Ge detector was counted.[12] The counts in this peak was measured to be negligible, implying that there was no significant capture of thermal neutrons by the detector. The results are shown in Table 1 along with those obtained by PIXE analysis. As shown in Table 1, the values of the concentrations of Si, Ca, and Ti obtained by PGA agree with those determined by PIXE analysis. Fair agreement is observed in the results for Mn, while there are discrepancies in the results for Al, K, and Fe. The discrepancy observed in the values of Fe concentration seems to be caused by its low detection sensitivity with PGA. However, the reason for rather smaller disagreement in the values of Al and K is not known except the fact that the broken pieces of the ceramic sample were used for PGA and the pulverized sample for PIXE analysis.

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