Estimation and Calibration of Thermal Neutron Flux for Neutron Activation Analysis

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Neutron activation analysis (NAA) is a very sensitive, powerful technique for identifying characteristics of many elements through performing both qualitative and quantitative analysis of major, minor, and trace elements in samples from almost every conceivable field of scientific or technical interest.^{1,2} Basically this technique is quite simple. A sample is irradiated by thermal neutrons and becomes radioactive. By measuring the β^+ s, γ s, β^- s, and half-life of the activated sample, the elemental constituents of the sample and their relative concentrations can be identified.

Industrial activation analysis is usually done with thermal neutrons from a reactor, where the neutron flux can be as high as ~ 10^{13} neutrons/cm²/s, or with an accelerator with fast neutron fluxes of ~ 10^{10} neutrons/cm²/s. When activation analysis is compared with other instrumental analytical methods such as gravimetric, calorimetric, spectrographic, or mass spectroscopy, its sensitivity is usually shown to be better over a factor of 10 than that of other methods. Activation analysis is used extensively in such fields as geology, medicine, agriculture, electronics, metallurgy, criminology, and the petroleum industry.^{3,4}

Theoretical Background

Assume that the sample has been activated in the accelerator. At the instant when the activation has been terminated, (t = 0), the activity of the sample is given by the following expression:

$$A_0 = \frac{\sigma m \eta \phi \alpha S}{w} \tag{1}$$

where

- A_0 : the number of disintegrations per second of the element in the sample at t = 0 (when irradiation stops),
- σ : cross section for the reaction, cm²
- m : mass of the target element, g,
- η : Avogadro's number, 6.023×10^{23} molecules/mole,
- ϕ : neutron flux, neutrons/cm²/sec,
- α : fraction of the target isotope in the sample [e.g., with an ordinary copper sample producing the ⁶³Cu(n, γ)⁶⁴Cu reaction, $\alpha = 0.69$ since 69% of all natural copper is ⁶³Cu],
- *S* : saturation factor, $1 e^{-\lambda t}$, where $\lambda = 0.693/T_{1/2}$, and $T_{1/2}$ is the half-life for the reaction,

w: atomic weight of the element.

After irradiation the sample is transferred immediately to the high purity germanium (HPGe) detector, and a spectrum is accumulated for a time, (t_1) , long enough to get reasonable statistics under the photo peak. The time is usually at least one half-life. The true number of disintegration, (N_d) , that occurred during t_1 can be determined from the following equation.⁵

$$N_d = \frac{\Sigma_p - \Sigma_\beta}{G\varepsilon_p f} \tag{2}$$

where

 Σ_p : sum under the photo peak,

- Σ_{β} : background for the same counting period under the photo peak,
- $G : A/2\pi s^2$, where $A = \text{area of detector in } cm^2$, and s = distance from source to detector in cm,
- ε_p : intrinsic peak efficiency for the γ energy and detector size used,
- f: decay fraction of the unknown activity, which is the fraction of the total disintegrations in which the measured gamma is emitted.

From the decay equation, N_0 can be calculated:

$$N_d = N_0 (1 - e^{-\lambda t}) \quad (t: \text{measurement time}) \tag{3}$$
$$A_0 = \lambda N_0$$

where *t* is the time for which the sample was counted.

Generally, radiation decay equation is following:

$$N = N_0 e^{-\mu_t t} \tag{4}$$

where

N : number of neutron with the absorber,

- N_0 : number of neutron without absorber,
- μ_t : linear absorption coefficient,
- *t* : thickness of absorber.

and definition of total cross section is

$$\sigma_T = \frac{\mu_t}{n} \tag{5}$$

where n is the number of nuclear in absorber such as following:

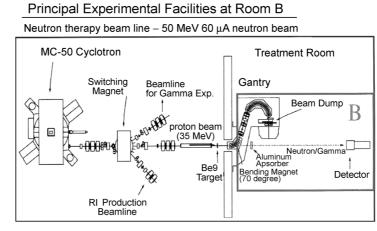


Figure 1. Diagram of MC50 cyclotron facility in KIRAMS.

$$n = \frac{\rho \times N_A}{A} \tag{6}$$

where ρ is density of absorber, N_A Avogadro's number, and A atomic weight.

If we put equation (5) in equation (4), neutron cross section is following:

$$N = N_0 e^{-\sigma_T nt} \tag{7}$$

$$T = e^{-\sigma_T n t} \tag{8}$$

$$\sigma_T = -\frac{\ln T}{nt} \tag{9}$$

Experiments

In this study, 50 MeV cyclotron (MC50) in Korea Institute of Radiological & Medical Science (KIRAMS) was used for neutron source by Be(p, n) reaction (Figure 1). And thermal neutron flux was generated using $10 \text{ cm}\phi \times 5 \text{ cm}$ cylindrical polyethylene. We used gold thin film to estimate the flux of thermal neutron by cylindrical polyethylene and measured the spectrum of the activated gold thin film using $2'' \phi$ HPGe detector (Figure 2). From the result of this Figure 2, we can calculate the thermal neutron flux. To get the activity exactly, we must also know the efficiency of HPGe detector. In this study, ²²Na, ⁵⁵Mn, ⁵⁷Co, ¹⁰⁹Cd, ¹³³Cs standard source were used for getting the efficiency and the energy calibration curve of detector (Figure 3). Because we used point source and the sample was not a same geometry, we corrected it using simulation program. To confirm the evaluated thermal neutron flux from activated gold thin film, we activated ⁵⁵Mn sample and got the cross section from 55 Mn(n, γ)⁵⁶Mn reaction.

Results and Discussion

We took the gamma-ray spectrum of the activated gold thin film using HPGe detector, and was showed in Figure 2. From the result of this Figure 2, we calculated the thermal

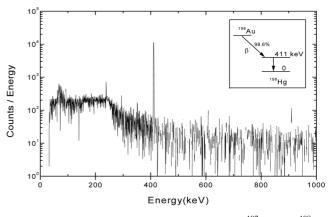


Figure 2. Gamma ray pulse height spectrum by ${}^{197}Au(n, \gamma){}^{198}Au$ reaction.

neutron flux in the 411 keV gamma ray peak area and its thermal neutron flux was 19426.68 as shown in Table 1. And also, detector efficiency of 411 keV gamma ray energy was estimated to about 0.24%. If we use the efficiency curve and the energy calibration curve of detector, we can know how

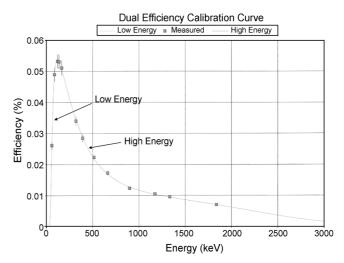


Figure 3. Absolute efficiency of HPGe detector (distance from detector to source: 10 cm).

γray energy	peak area	efficiency (%)	mass(g)	saturation factor $(1 - e^{-\lambda t})$	flux (n/cm ² ·sec)
411 keV	15086.15	0.24	0.072	0.869	19426.68 ± 101.62

Table 1. Evaluation of thermal neutron flux

Table 2. Thermal neutron capture cross section of ⁵⁵Mn

γray energy	peak area	efficiency (%)	mass (g)	saturation factor $(1 - e^{-\lambda t})$	cross section (barn)
847 keV	1004.34	9.53×10^{-2}	0.527	0.998	13.79 ± 0.45

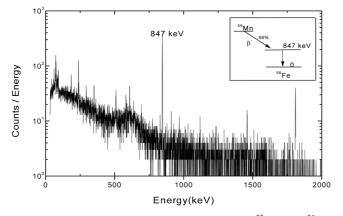


Figure 4. Gamma ray pulse height spectrum by ${}^{55}Mn(n, \eta){}^{56}Mn$ reaction.

many thermal neutron get into the activated sample. In the case of 198 Au, when gold thin film was exposed to thermal neutron flux during 18 hour, saturation factor was calculated to 0.869 with 2.696 days half-life for 0.072 g gold thin film mass in 1 cm diameter. From this data, we could evaluated the thermal neutron flux of gold thin film using MC50 in Be(p, n) reaction and the estimated data were listed in Table 1.

To confirm the above result of thermal neutron flux using MC50, we took the gamma-ray spectrum and calculated thermal neutron capture cross section of ⁵⁵Mn. The gamma-ray spectrum from ⁵⁶Mn produced by neutron activation of the ⁵⁵Mn was shown in Figure 4. Using the previous same method for gold thin film, we could calculated the capture cross section of ⁵⁵Mn from equation 1. The result of these

calculation was listed in Table 2. Thermal neutron capture cross section of ⁵⁵Mn was estimated about 13.79 barn whereas the reported was about 13.41 barn. This have a good agreement with the reported in table of isotope by other method.⁶

Our neutron activation method can be used to analyze very small amount of element in various sample. If unknown samples are exposed to thermal neutron from MC50 during selected time, we can measure the gamma ray spectrum from activated sample by (n, γ) reaction and can administer quantitative analysis of the sample. This is called by Nuclear Analytical Techniques (NAT). This technique is very advanced with HPGe detector and development of personal computer. So we can identify the unknown element through quantitative analysis using neutron activation method rapidly and exactly. Further, this technique will be helpful to background monitoring of very small amount element, source receptor model and controling air pollution.⁷

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