

Interference of the Hydrogen Analysis using PGAA

Hyun-Je Cho, Young-Sam Chung and Young-Jin Kim

Korea Atomic Energy Research Institute, P.O.Box 105 Yusong, 305-600, Daejeon, Korea
hjcho@kaeri.re.kr

1. Introduction

The prompt gamma neutron activation analysis (PGAA) technique has been proven as a most useful method for an elemental analysis like boron, carbon, nitrogen, silicon, phosphorus, sulfur, chlorine, cadmium and especially hydrogen, because these materials are difficult to measure nondestructively by other methods [1-2]. The simplest example of a prompt gamma-ray measurement method is a neutron capture by hydrogen. The single hydrogen peak is easily measured because the signal is independent of the chemical form of the hydrogen present. Hydrogen captures a neutron as it goes to a higher level of the excited state *via* the $^1\text{H}(n,\gamma)^2\text{H}$ reaction, the product nucleus then immediately de-excites to the ground state by an emission of a characteristic 2223 keV gamma-ray.

Since all of the materials used in the PGAA setup produce prompt gamma rays by the capture of scattered neutrons from sample and sample holder, a careful analysis of the background in the PGAA becomes inevitable. The contribution of the background prompt gamma rays to a sample spectrum varies depending on the composition of the matrix under irradiation. In the case of this background where the gamma rays interfere with the gamma rays of the analyte, a background correction becomes very crucial. When determining the hydrogen content at trace levels, it is necessary to measure a blank. The blank have closely to be matched to the sample matrix and geometry, but to be free of hydrogen. Measurement of a blank allows for a correction of background generated from the neutron capture by the environmental hydrogen and also for the background due to spectral interferences.

2. Experimental

The present study measured the hydrogen concentrations for metal and biological samples using the HANARO PGAA facility. In the sample position, the neutron beam size has a 2 cm diameter, the neutron flux measured from irradiation of the gold foils is $1.4 \times 10^8 \text{ ncm}^{-2}\text{s}^{-1}$ and a gold-cadmium ratio of thermal/fast neutrons of about 300. A sample for an irradiation is usually suspended in the neutron beam by means of Teflon strings at a 45° angle to both the detector and the neutron beam. The Teflon sheet is used to suppress the background appearing from the sample case. The variations in the neutron flux are monitored by a periodic irradiation of a Ti foil, which has a simple gamma-ray

spectrum and its 1382 keV gamma ray was used. The count rates are corrected for the variations in the neutron flux over the course of an analysis.

The background affects from the inside shielding composition materials and the preparing sample condition can weakly appear in spectrum, as shown in Fig. 1.

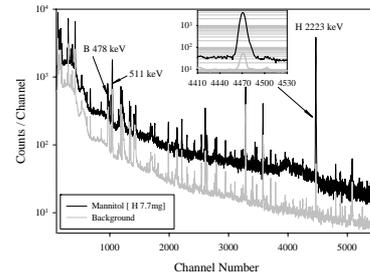


Fig. 1. Comparison of the spectrum obtained from sample and blank.

To confirm a background on the determination of hydrogen, the blank spectrum is necessary to measure a prior to every sample measurement. Teflon is used as a packaging and mounting material because it contains no hydrogen. Measurement of a blank allows correction for background generated from neutron capture by environmental hydrogen and also for background due to spectral interferences. The background count rate in the hydrogen peak area arising from the air and blank was estimated about 0.08 cps that is corresponding to approximately $20 \mu\text{g}$ of hydrogen. The samples for the hydrogen concentration analysis is examined the moisture content of before and after weighing and then samples keep in the oven for drying as same temperature as sample irradiation position.

The background in the hydrogen region (2220~2225 keV) of a prompt gamma-ray spectrum measured for a hydrogen free material arises from a few possible sources. Neutron capture by hydrogen in the atmosphere and shielding materials gives rise to 0.08 ± 0.02 counts per second of a gamma radiation. The interference is emitted by other elements in the sample matrix which represents a second possible source of background, which is distributed within about 5 keV of the hydrogen peak energy. The interference peaks which appeared from 2220 ~ 2222 keV, 2224 ~ 2225 keV give rise to an influence for a hydrogen analysis. The main background region appears from tail of high energy region in hydrogen peak area. Also, a possible source of background arises from Compton scattering of high

energy gamma-rays. This results in decrease in the signal to noise ratio by raising the continuum baseline under the hydrogen peak. A few interference nuclides appear inside hydrogen peak and near the tail energy of the hydrogen peak but the influences of the interferences peaks relatively weak about the hydrogen count rate. Because of the complexity of prompt gamma-ray spectra, spectral interferences often constitute a major source of uncertainty in PGAA measurements.

In the present study, interferences for the hydrogen peak region, as shown in Fig. 2, appear with a few peaks. The energies of the main interference peaks are 2221 keV and 2225 keV within the hydrogen region. But, in case of long time measurement, the energies of interference peaks have a few like 2220.3 keV (Gd), 2221 keV, 2224.4 keV (Ge), 2224.8 keV (Ba) and 2225.3 keV, where remainder peaks except for 2221 and 2225 keV have extremely small peak area.

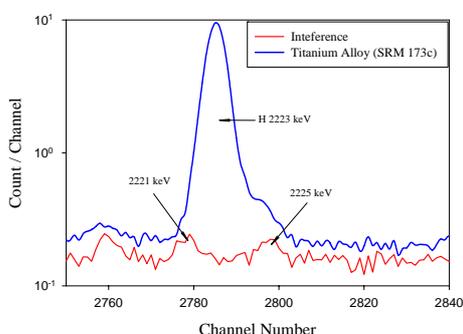


Fig. 2. A portion of the prompt gamma-ray spectrum obtained from thermal neutron capture using NIST(SRM 173c) sample.

The temperature of the PGAA system was 24~25 °C at sample position and continuously controlled a moisture and composition materials such as the sample device circumstances within the system. Also, the neutron flux monitoring is checked by using blank and Ti on the before and after the measurement of the sample. The biological samples are used the power type but metal samples are used the bead type. In the case of powder sample, the sample geometry is better than the beads that are difficult to homogeneity for sample preparation. The sample vial is made of the Teflon that has no influence on the hydrogen peak region, but appears a little of moisture influence for the powder type. In the case of these, the present measurement has the phase of dry using deccicate during a few minutes. After the dry step, the sample is inserted in the sample device and measured.

3. Results and Discussion

The CRMs of Titanium-base alloy (173c), Hydrogen in Titanium alloy (2453), Peach leaves (1547), Spinach (1570) and Oyster tissues (1566b) were analyzed in the present work. The samples of approximately 3~50 mg were taken in a 0.5 mm thick Teflon cylinder of 6 mm i.d.

and 1 cm height. The combined uncertainties of the relevant materials were in the range of 2.7% for the hydrogen analysis. The main sources of the uncertainties are due to statistical errors (0.25%), the detection efficiency (2.3%), the background subtraction (1.2%), and error sources including some other corrections. The count rates for the hydrogen energy region were measured with on- and off-beam and blank, as shown in Table 1. In the case of neutron beam block by beam shutter, the count rate appears about 0.1 cps. Generally, the count rate of the long time measurement is lower than that of short measurement. The count rate for the blank in the hydrogen energy region was about 1.5 times higher than air condition of the on-beam stage. The hydrogen concentration of each sample was determined from the measured count rate of the hydrogen peak, sample mass and hydrogen sensitivity. The results are summarized in Table 2 together with the certified values. The relative errors of the measured values are about 1 % for a high hydrogen concentration such as the spinach, about 2 % for the 1000 ppm level like the SRM 2453 and about 7 % for the below 100 ppm level like the titanium-base alloy. The analysis of the hydrogen content using the present PGAA facility is in good agreement with the certified value, but at the below a 10 ppm, the measured values are lower than the certified values.

Table 1. Comparison of the counts rate for hydrogen energy region.

Classification	Measuring Time(sec)	Hydrogen Region (cps)
Off-Beam	10,000	0.107 ± 0.012
On-Beam	10,000	0.214 ± 0.013
Blank+(on-beam)	10,000	0.348 ± 0.015

Table 2. Comparison with the measured and certified values for the biological and metal samples [In the A ± B, B is S.D].

Standard Reference	Hydrogen Content [mg/kg]	
	Certified Value	Present Work
NIST SRM		
1570 (Spinach)	54500 ± 800	53950 ± 1200
1566b (Oyster Tissue)	7.20 ± 0.40	5.76 ± 0.50
173c(Titanium-base Alloy)	60	55.6 ± 1.5
2453 (H in Titanium Alloy)	1140	1115 ± 10

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