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Progress of prompt gamma activation analysis in Korea

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The new PGAA facility using diffracted neutron beam was developed in Korea. The basic characteristics of the facility were studied in detail. A general formalism of the k_0 factor as extended to non-1/v absorber and arbitrary neutron spectrum was discussed and the actual data for Cd, Sm, Eu, Gd have been measured and determined successfully owing to the simple nature of the diffracted neutron spectrum. The k_0 factors for B, N, Si, P, S and Cl were also determined and showed consistent results with previously reported ones. At an early stage, feasibility of boron concentration analysis and measurement of thermal neutron capture cross sections has been studied. The PGAA facility is now open to users. A considerable amount of beam time is already dedicated to studies on the elemental analysis.

Introduction

The study of neutron-induced prompt gamma activation analysis (PGAA) in Korea was initiated in the early 1980's at a former facility of the Korea Atomic Energy Research Institute (KAERI).[1,2] During the following ten years, the study came to a halt, mainly due to the removal of KAERI. In the mid 1990's, the request for boron analysis reactivated a development of PGAA facility at the site of the new research reactor, HANARO in KAERI. The whole task of developing the facility was undertaken by the author's group in Seoul National University (SNU) while KAERI provided the financial and various backing supports. In about 4 years, the development of SNU-KAERI PGAA facility was completed under the auspice from IAEA coordinated research project[3,4] with much valuable information and advice from the late Dr. Prof. G. MOLNÁR in Budapest, Hungary and from Dr. R. M. LINDSTROM in NIST, USA. In this paper the progress of PGAA studies in Korea during the last 6 years is summarized, including the most recent result on the measurement of prompt k_0 factors and the preparation of prompt gamma-ray spectrum analysis tools.

Development of SNU-KAERI PGAA facility

In the development of a PGAA facility at HANARO, the first problem was how to extract a neutron beam from an existing beam line (ST1) which was already dedicated to a previously planned device. To adapt into the existing plan with its space limitations, a novel method of using polychromatic neutrons was devised. All the high order diffracted neutrons obtained from pyrolytic graphite (PG) crystals set at a Bragg angle of 45° were collected and focused.[5] The mosaic spread of the PG crystal was optimized to 0.8° and focusing effect was realized by placing three pieces of flat crystal in a concave holder. A vertical diffracted beam line was installed due to space limitation at the horizontal ST1 line. The PGAA line consists of the crystal assembly, collimators, shields for background neutrons and γ-rays and a beam catcher. The LiF tiles are attached to the inner walls of lead shield to reduce the scattered neutrons. At the wall position towards the detector, 95% enriched ^{6}LiF tiles are used. By activating Au wires, the thermal equivalent neutron flux was determined to be $8.2 \cdot 10^7$ n·cm^{-2}·s^{-1} and the beam area was 2×2 cm^2 at the sample position with a beam uniformity of 12% in the central area of 1×1 cm^2. The Cd ratio for gold was 266. The details of the facility and performance are given in Reference 6. The detection system initially was comprized of a single HPGe detector (43%), signal electronics and a fast 16k ADC. In 2002, a NaI(Tl)-BGO guard detector and the associated electronics were installed, and the detection system was upgraded to Compton-suppressed and pair spectrometer with three modes of detection, single, Compton-suppressed and pair modes.[7] With several modifications suggested by Dr. MOLNÁR, including a reduction of the collimator aperture in front of the detector, the total background count rate has been reduced by a factor of 10 at the expense of about a quarter reduction in the full energy peak efficiencies. Hence, a better sensitivity of the spectrometer with a reduced dead-time in the ADC could be achieved.

Polychromatic neutron spectrum

Since little was known about the spectral details of the polychromatic neutrons obtained from Bragg diffraction by a PG crystal, several experimental and theoretical studies were undertaken to determine the beam characteristics.[8] For a direct study of the beam

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spectrum, a neutron time-of-flight (TOF) measurement was done with a 3 m TOF path length. Flight time peaks corresponding to diffraction orders 1–6 were resolved and their relative intensities were determined after corrections for detection efficiency, attenuation on the flight path and beam chopper transmission. The absolute neutron intensity at a position just after diffraction was determined for the same configuration by measuring the decay gamma's emitted from gold wire activation. About 90% of the diffracted beam flux consists of the neutrons from diffraction orders 2–4. Resolving the higher diffraction orders beyond 6 was not possible in the TOF spectra mainly due to the spectrometer's finite resolution. A theoretical approach was taken instead to predict the intensity for higher diffraction orders beyond 6. In the theory of higher order diffraction, the most crucial parts were the Debye-Waller factor and the thermal diffuse scattering, which describe the radical decrease of reflectivity for increasing diffraction order. The Debye-Waller factor for a PG crystal was available from X-ray studies even though their consistency was not good. The parameter for the thermal diffuse scattering from a PG crystal could be only predicted by a complicated and involved model calculation and hence was determined from a fit to the observed TOF intensities. The simple fit could describe closely the observed intensities of orders 3–6 and even predict those of higher orders beyond 6.[8]

Validation of the determined neutron spectrum has been done by comparing calculated quantities with measured ones for effective cross section and Cd ratios. The calculation has been done for the effective ^{10}B(n,α) cross section and Cd ratios by using the spectral information of the beam and the cross section dataset from the evaluated data library. Measurement of the effective ^{10}B(n,α) cross section by using both a thin and a "black" sample was performed based on the principle in Reference 4. The measured effective ^{10}B(n,α) cross section is 3987±40 b. Accordingly, other effective quantities to characterize the beam are derived: effective temperature 269±5 K, effective velocity 2120±20 m/s. To access the epithermal component in the diffracted neutron beam, Cd ratios were measured for ^{35}Cl, ^{113}Cd, ^{149,150,152,154}Sm, ^{157}Gd and ^{179}Au. The Cd ratio for gold was obtained in the typical way by detecting decay gamma from an activated wire with the diameter of 0.1 mm. Measuring Cd ratios for the rest of the listed nuclides was performed by comparing the count rates of prompt capture gamma's during the irradiation of the target in a normal diffracted beam and in a Cd-filtered diffracted beam. To prepare the Cd-filtered beam, a Cd plate of 0.5 mm thickness was placed at the position of the beam collimator. The observed Cd ratios were in the range 370–400, except for that of Au (266). The calculated Cd ratios indicated an almost negligible portion of epithermal neutrons in the diffracted beam. The main origin of epithermal neutrons in the diffracted beam was traced to the background neutrons scattered from the structural materials of the PG mount and goniometer which were inevitably immersed in the strong white beam. Hence, a smooth spectrum of background neutrons was added to the diffracted neutron spectrum and its magnitude was fitted to explain the observed Cd ratios. The spectral shape of background neutrons was assumed to be close in shape to the white beam of 308 K Maxwellian with a $1/E$ tail above 0.3 eV. The assumption is based on the nearly constant and isotropic cross section of the main scatterers. The amount of background neutrons was less than 0.5% of total neutrons. With this refined beam spectrum, excellent consistency was achieved between the calculated and measured Cd ratios for various non-$1/v$ nuclides. The final comparison between the calculated effective temperature and the measured one showed consistency within ~3%.[8] Therefore, it has been shown that owing to its simple nature, the polychromatic neutron beam has a well-defined spectrum for PGAA, which is a strong advantage for k_0 standardizing and measuring non-$1/v$ absorbers.

Gamma-ray spectrometer

An n-type HPGe main detector is placed at 25 cm distance from sample. The energy resolution (FWHM) is typically 2.2 keV at 1332.5 keV. The energy and efficiency calibrations were carried out based on the Budapest prescriptions.[9,10] To calibrate the efficiency of the detector in the low energy region of 60–1408 keV, standard radioactive isotopes ^{60}Co, ^{133}Ba, ^{134}Cs,^{137}Cs, ^{152}Eu and ^{241}Am were used. The efficiency calibration was extended up to 11 MeV by normalizing the relative efficiency data determined from nitrogen and chlorine prompt gammas to the absolute efficiency curve in the low energy region.[9] The efficiency curve function was fitted with an 8th order polynomial. For the proper isotopic identification, the channel-energy non-linearity in the system was fit with a 6th order polynomial and corrected to determine the accurate gamma-ray energies.[10] The efficiency calibration was also performed by Monte Carlo simulation and the well-known local minimum usually identified around 600 keV[11] on the efficiency curve had been analyzed.[12] The events of the full energy absorption peak formation are classified by interaction mechanism and checked. Finally, it was confirmed that the Compton scattering events mainly contribute to the formation of the local minimum.

Gamma-ray spectra are acquired and analyzed by a commercial software. Detailed off-line analysis is carried out using new software developed by the author's group. It is based on the iterative Hypermet peak analysis routine[13] which has multi-parameter

fitting ability. The graphical user interface part of the software was coded in MATLAB, and is running on a Windows platform. In order to fully cover the peak shapes encountered in the prompt gamma-ray spectrum, various peak analysis routines for a single peak or the Doppler-broadened peak were inserted as selectable analysis options.

Boron measurement and k_0 study

Boron analysis was the primary motivation of developing SNU-KAERI facility as described above and this work was begun soon after completing characterization of the diffracted neutron beam.[14] In order to estimate the reliability of the analysis, boron concentrations were measured for standard reference materials at first by the relative method. When the Doppler-broadened boron peak was overlapped with the sodium peak, the boron peak area was determined by the decomposition method. The shape of the interfering sodium peak was set as a simple Gaussian, while the boron peak shape was represented by a difference between two error functions.[14] The measured boron concentrations were consistent with the certified or the information values within 6%. The practical application of boron analysis for biological samples is under way.

The second important application field of the facility was the production of elemental PGAA data like the prompt k_0 factors and partial capture cross sections. First, the elements of non-1/*v* strong absorbers, Cd, Sm, Eu and Gd, have been measured.[15] Chlorine was used as the comparator element.[16] For the most strongly detected γ-ray line, the analytic sensitivity was determined and the prompt $k_{0,\mathrm{Cl}}$ factors relative to Cl 1951.14 keV line were obtained. These were converted to the $k_{0,\mathrm{H}}$ values relative to H 2223.25 keV line by using the $k_{0,\mathrm{H}}$(Cl) value determined in a parallel measurement with an ammonium chloride sample. The partial cross sections were calculated from the latest capture cross section for ^{1}H, 332.6(7) mb.[17]

Throughout the existing literature, confusion exists with regard to the definition of the prompt k_0 factor. In the following definition, the k_0 factor is a composite nuclear constant which is related to the capture rates of two elements irradiated simultaneously and is given by:[16]

$$k_{0,c}(E_{\gamma,x}) = \frac{P_x(E_{\gamma,x})}{P_c(E_{\gamma,c})} \cdot \frac{\sigma_{0,x}}{\sigma_{0,c}} \cdot \frac{\theta_x / M_x}{\theta_c / M_c} \qquad (1)$$

where x and c denote the analyte and comparator elements, P is the absolute emission probability, E_γ is the γ-ray energy, σ_0 is the neutron capture cross section of the isotope measured at 2200 m·s^{-1}, θ is the natural abundance of the isotope absorbing neutron and M is the atomic mass of the element, respectively. For the case of 1/*v* absorption, the above equation is equivalent to:

$$k_{0,c}(x) = \frac{A_{sp,x}(E_{\gamma,x}) / \varepsilon(E_{\gamma,x})}{A_{sp,c}(E_{\gamma,c}) / \varepsilon(E_{\gamma,c})} \qquad (2)$$

where A_{sp} is the specific count rate and ε is the full energy absorption peak efficiency for the relevant γ-rays. Equation (2) consists solely of measurable quantities and, hence, it has been taken by some experimentalist as the definition of the prompt k_0 factor. For the non-1/*v* absorption, these two equations are no longer equivalent to each other. Hence, Equation (1) has been taken as the definition of the prompt k_0 factor in our study, which enables a direct intercomparison of values from different laboratories. Whether a given isotope absorbs neutrons in 1/*v* or non-1/*v* manner depends not only on the capture cross section of the isotope, but also on the spectrum of the neutron beam. Conventionally, in reactor neutron activation, the non-1/*v* absorption has been parametrized by Westcott *g*-factor. For a neutron spectrum widely different from Maxwellian, a more general approach to the problem is required. A more detailed discussion of this topic is dealt with in the latest IAEA TECDOC.[18] In the most general situation of neutron spectrum and target nuclei, two correction factors including the effective *g*-factor and Cd ratio are introduced. By using the actual spectrum of neutron beam inside the target, the effective *g*-factor is defined by adopting the Høgdahl convention. It has been shown that the effective *g*-factor is calculable by using the incident neutron spectrum, and the influence of epithermal neutrons on the neutron capture rate is corrected by measuring the Cd ratio for each non-1/*v* isotope. Hence the general form of the prompt k_0 factor is given by:[15]

$$k_{0,c}(x) = \frac{A_{sp,x}(E_{\gamma,x}) / \varepsilon(E_{\gamma,x})}{A_{sp,c}(E_{\gamma,c}) / \varepsilon(E_{\gamma,c})} \cdot \frac{\hat{g}_c}{\hat{g}_x} \cdot \frac{\left(\frac{R_{cd}}{R_{cd}-1}\right)_c}{\left(\frac{R_{cd}}{R_{cd}-1}\right)_x} \qquad (3)$$

where $\hat{g}$ is the effective *g*-factor, and R_{cd} is Cd ratio. The general formalism and the measured data for ^{113}Cd, ^{149}Sm, ^{151}Eu and ^{155,157}Gd are reported in Reference 15. Intercomparison of k_0 factors for non-1/*v* isotopes shows better consistency between measurements of different laboratories after correction with *g*-factors, as pointed by MOLNÁR.[18]

The 1/*v* absorbers, B, N, Si, P, S and Cl, have been measured[19] after the upgrading of the detection system to Compton suppressed and pair spectrometer. Hydride samples were prepared for B and N to use hydrogen as comparator, and for other elements, chlorine compounds or mixtures, were used to give a chlorine comparator.

The powder samples were cold-pressed into disk (10 mm dia.) and then wrapped by Teflon film (25 μm thickness). The analytic sensitivity, $k_{0,H}$ factor and partial cross sections were determined by the simplified approach based on Eq. (2). Results showed good consistency with reported ones from other laboratories.[19]

Conclusion

The new PGAA facility using diffracted neutron beam was developed in KOREA, and the basic characteristics were described. Researches on boron analysis and k_0 factors were performed at the new facility. After August 2002, the facility is managed by NAA group in KAERI[20], and open to users. A considerable amount of beam time is being allocated to studies on elemental analysis.

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