Application of In-beam Activation Analysis in Elemental Distribution Analysis

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Abstract. The analysis of elemental distribution in key parts of samples is of importance in the fields of material science. A non-invasive method utilizing neutrons has been presented. The elemental distribution of materials can be obtained by measuring the gamma spectra with collimated neutron beams using in-beam neutron activation analysis. In order to verify the feasibility of neutron imaging and activation analysis in elemental distribution studies, the experimental model was calculated and verified by Monte Carlo simulations. Using the CARR reactor neutron imaging and activation analysis system, the elemental distribution experiment of the flat sample was carried out. The neutron radiography results of the flat sample and the gamma spectrum of each measurement area were analyzed. The distribution of In, Gd, Eu, Au, and B were in good agreement with the sample. Neutron radiography and in-beam activation analysis were effective tools for conducting elemental distribution studies.

1. Introduction

Neutron is free of charge, thus it can intrude deeply into materials unaffected by the atomic electron-shells. Neutrons are ideal probes for studying the internal structure and composition of materials. Neutron beams for material analysis can be created by research reactors or isotopic neutron sources. Neutron radiography and neutron activation analysis are two commonly used analytical methods. These two methods are non-destructive, sensitive to light elements, do not require sample pretreatment, and can be used to analyze the internal structure of the sample and its elemental composition¹⁻³.

In neutron radiography, transmitted neutrons interact with the converter screen to produce radioisotopes, alpha particles or lights which can be recorded by films, imaging plates or cameras. In neutron activation analysis, the nuclear reactions occur between nuclei and neutrons. The reaction products to be measured are either the radiation, released instantaneously upon neutron capture; or, if the resulting new nuclei are radioactive, the induced radioactivity by which they decay. Conventional activation analysis is a bulk method. Through collimation or focusing of the neutron beams, the active measuring area can be pointed. By performing a two dimensional scan of the sample using collimated neutron beams, and a dataset of gamma spectra can be obtained, from which the elemental distribution can be analyzed⁴⁻⁶.

China Advanced Research Reactor (CARR) is a 60 MW tank-in-pool inverse neutron trap-type research reactor. The pressurized light water is used as the moderator and the primary cooling water. The undermoderated core is surrounded by heavy water reflector, where the maximum unperturbed thermal neutron flux would be expected to be $8 \times 10^{14}$ n/s cm² at 60 MW. The reflective layer is equipped with 9 horizontal channels. Since the horizontal holes are tangential channels, the fast neutron background and gamma
background in the beam current are greatly reduced, and the beam throughput is very high, which is very suitable for neutron radiography and in-beam activation analysis for elemental distributions.

In this work, the experimental model was calculated and verified by Monte Carlo simulations, and the experimental model is optimized according to the simulation calculation results. The experiments of a flat sample had been carried out at CARR reactor to analyze element distributions.

2. MCNP simulations

Monte-Carlo N-Particle Transport Code (MCNP) was a general-purpose code to simulate the transport process of neutrons, photons, etc, and was first developed by Los Alamos National Laboratory in 1957. The simulations were performed using the MCNP5 code to verify the experimental system parameters. The energy distributions of neutron beams from CARR reactor and parameters of neutron collimator were used as input data. Four measuring areas were set in the sample, and the gamma spectra of measuring areas were calculated separately using the ENDF/B-VII library.

According to the gamma energy spectrum of each measurement area calculated by the MCNP simulations, elements in each measurement area can be recognized based on the characteristic peak energy of the gamma energy spectrum. The results showed that the background of the low-energy gamma was high and indicated that it was necessary to optimize the shielding design for low energy gamma. Therefore, shielding devices for the detection system had been added.

![Gamma spectrum of each measuring area of experimental samples calculated by MCNP5](image)

Figure 1. Gamma spectrum of each measuring area of experimental samples calculated by MCNP5

3. Experiments

In order to reduce the effects of neutron-absorption and gamma self-shielding effects, a flat-type sample was used to verify the feasibility of the elemental distribution analysis. In$_2$O$_3$, Gd$_2$O$_3$, Eu$_2$O$_3$, B$_4$C powder and Au foil were made into a flat sample with the size of 2.5cm*5cm, and the upper and lower surfaces were covered with aluminum sheets and iron sheets respectively, and the outer layers were wrapped with aluminum foil.
The neutron radiography devices and in-beam activation analysis devices were located at the end of the CNGB neutron beam guide at CARR reactor. When CARR reactor power was 15 WM, the mean neutron wavelength was 4 angstroms (about 5.1 meV) at sample position about 1m away from the end of the neutron guide, and the cold neutron flux was up to $4.8 \times 10^8 \text{cm}^{-2}\text{s}^{-1}$.\textsuperscript{17-19}

When performing neutron radiography, the neutron image of the sample was converted into a visible image by a scintillation screen, and then converted into a digital image by the camera through the lens. The scintillation screen was made of LiF and ZnS(Ag) material with a short response time to neutrons (attenuation constant of 0.2μs) and the detection efficiency of the scintillation screen was more than 35%. In order to increase the reflection efficiency of light and reduce the activation by neutron irradiation, a 0.2 μm aluminum film was plated on a 5 mm thick pure aluminum substrate, and a 10 nm SiO\textsubscript{2} protective layer was further plated on the surface as a mirror. The visible light images were converted into digital images by using sCMOS sensor chip.

In order to meet the spatial resolution requirements of the sample elements, a neutron collimator was used with 1.2 cm * 2.5 cm size made by the $^{10}\text{B}$-enriched polymer. The neutron collimator was placed at the end of the neutron guide, about 30 cm from the sample chamber. An N-type electrically cooled coaxial HPGe γ detector was used to collect the γ spectrum, and the HPGe detector was surrounded by Compton-suppressed spectrometer and a circular lead shield with a thickness of 10cm to achieve low detection limits. The detection system had been calibrated using $^{152}\text{Eu}$ source before experiments.

4. Results
The gamma spectra of 4 measuring parts with the size of 1.2cm*2.5cm have been collected by the HPGe detector for in-beam activation analysis. The neutron irradiation and gamma measurement were performed simultaneously. Since the thickness of the flat sample was less than 2 mm, the 2D distribution of elements in the sample had been analyzed and the neutron self-absorption effect of this sample would not be discussed.
The elemental composition can be obtained from the gamma spectra of 4 measuring parts based on the characteristic peak energies of the measured gamma-rays. In (1861.2 keV, 416.86 keV, 1097.3 keV) has been recognized in part a, and Gd(79.5 keV, 181.9 keV, 199.9 keV) has been recognized in part b, Eu (89.8 keV) and Au (411.8 keV) have been recognized in part c, B (478 keV) has been recognized in part d. It turned out that the elemental distribution of 4 measuring parts were basically consistent with the actual sample. The count rate of part b was relatively high due to the large cross section of Gd. The Doppler-broadened peak with of B was larger than normal gamma rays since the 478 keV gamma rays were emitted by recoiled 7Li particles.

5. Conclusions
The effectiveness of in-beam activation analysis system used for elemental distribution analysis had been validated by MCNP simulations. The elemental composition of the flat sample had been analyzed by neutron radiography and in-beam activation analysis at CARR, and the elemental distribution results were in good agreement with the actual sample. Neutron imaging techniques and in-beam activation analysis methods can be used for elemental distribution studies of flat materials.

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