

Chapter 10

FACTORS AFFECTING NEUTRON MEASUREMENTS AND CALCULATIONS

Part E. Hydrogen Content in Granite

**Tetsuro Komatsubara, Kimikazu Sasa, Hiroyuki Ohshima,
Hiromi Kimura, Yoshikazu Tajima, Tsutomu Takahashi, Satoshi Ishii,
Yoshihiro Yamato, Masanori Kurosawa, Kohei Furuno**

Introduction

For evaluation of radiation doses from the atomic bomb at Hiroshima, many systematic measurements have been made of the residual activities of activation products in rocks and concrete. For the Motoyasu Bridge, which is located close to the bomb hypocenter, the depth profile of ^{152}Eu was measured in a granite core (Hasai et al. 1987; Shizuma et al. 1997). In order to reproduce the depth profile of the activities, it is important to calculate the neutron scattering and absorption (Endo et al. 1999). In this section, the first result of hydrogen analysis by proton-proton elastic recoil coincidence spectrometry for the granite samples is described.

Material and Methods

The granite samples were taken from a pillar of the Motoyasu Bridge. The place of origin of the granite was Kurahashi, in the south part of Hiroshima City. The major components of minerals in the granite are quartz, plagioclase, orthoclase and biotite. The quartz, which is clear crystal, possesses a very small amount of hydrogen. The orthoclase is white in color and includes clay minerals with some content of hydrogen. The biotite has hydrogen as a component of its crystal, in which the corresponding water content is about 1.8% wt (H_2O). Furthermore, small amounts of chlorite, which is a metamorphic mineral of biotite, exist with more than 5 times the hydrogen content of biotite.

Since the hydrogen distribution in granite is not uniform, a number of measurements were repeated, and the average was calculated to evaluate the water content in the whole granite sample. A piece of the core sample was labeled U4-1, which denotes the upper quarter of the

fourth slice from the north end. The distance from the north surface is between 6 and 8 cm. Eleven slices of the granite of 0.2 mm thickness were prepared. Five of the eleven slices were irradiated in the proton beam.

The proton-proton elastic recoil coincidence measurements (Cohen et al. 1972) were performed at University of Tsukuba Tandem Accelerator Center. The granite slices were irradiated in a 20 MeV proton beam accelerated by the 12UD Pelletron Tandem Accelerator. For the detection of hydrogen in the granite sample, the coincidence method is quite selective to reduce elastic and inelastic scatterings by silicon, oxygen or other elements. This method of hydrogen analysis is applicable for a very wide range of hydrogen content, from several ppm to more than 10 percent. The analysis is not destructive. Furthermore, this method can provide the depth profile of the hydrogen content.

In Figure 1, three sample holders are shown. The size of the beam spot was 2 mm in diameter, as shown by the small circles. One measurement was performed with 0.8 nA beam intensity to collect coincidence events for about 10 min. At the end of the measurement, the sample holder was moved up by 2.5 mm to change the irradiation point, and the measurements were repeated.

Two Si detectors were located at ± 45 degrees with respect to the beam axis. Solid angles of the detectors are 3.4 msr, defined by 4 mm slits located at 61 mm from the irradiation point. In order to reduce edge scatterings from the slits, buffer slits were installed in front of the Si detectors. The distance from the slit to the buffer slit was 66 mm. Coincidence measurements were performed with standard NIM modules. Logical signals of the coincidence were created by a time-to-amplitude converter, with the resolving time period adjusted to 20 nsec. For correction of dead time, pulser signals of 2 Hz were also accumulated in the same spectra as proton events. In all measurements, the numbers of counts were corrected for the dead time deduced from the pulser signals. A typical dead time was 1.5%.

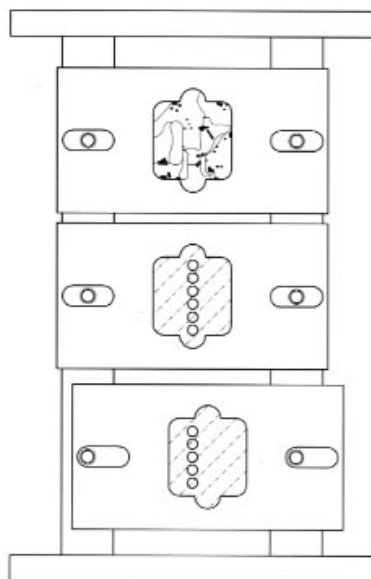


Figure 1. Three sample holders and irradiation spots on granite samples.

Result and Discussions

As an example of the coincidence measurements, a two-dimensional spectrum is shown in Figure 2. A number of coincidence events appearing in this figure can be derived for the biotite component. The two-dimensional spectrum of Figure 2 is projected to a diagonal line to produce the sum-energy spectrum shown in Figure 3(a). This spectrum indicates the depth profile of hydrogen, since the stopping powers for scattered protons are larger than those of the incident protons. Furthermore, the spectrum is distorted by secondary scattering. When an incident proton scatters near the entrance surface, the scattered and recoiled protons can be scattered again in the rest of the thick granite sample. The reduction rate was experimentally determined by additional measurements of proton scattering from Mylar foils attached on the surface of the granite. The corrected spectrum assuming a simple exponential function is shown in Figure 3(b).

Since the hydrogen content is not uniform, random sampling measurements were performed at 44 sampling points. The total charge of the beam irradiation was 5.0×10^{-7} coulomb for each measurement. Counts of scattered protons calculated from the projection spectra, as shown in Figure 3a, are widely distributed from a few hundred to ten thousand counts. The distribution of the measured counts is shown in Figure 4. Lower count peaks could correspond to quartz. Higher counts can be explained by the existence of chlorite.

In order to calculate the hydrogen content from the number of scattered protons, thin standard materials were irradiated to measure coincidence events. The standard materials were a polyethylene film of 30 μm thickness, Mylar films of 0.9, 2.5 and 5.7 μm . A glass T-1030 of 100 μm in thickness and hydrogen content of 135 ppm wt (H_2O) (Yurimoto et al. 1989) was also irradiated. A calibration factor was experimentally deduced as $k = 25.7 \pm 2.3$ counts for the 5.0×10^{-7} coulomb irradiation at 1 $\mu\text{g}/\text{cm}^2$ water (H_2O) content. The average value of the measured results for the 44 irradiation points was calculated as $Y = 1,900 \pm 700$. When we assume the possibility of more local concentration of hydrogen, this uncertainty is increased by a factor of 2. The correction factor for self-absorption was deduced as $f = 2.07 \pm 0.04$. The average of proton scattering yields was converted to weight of corresponding water content and calculated to be $d = Y \times f / k = 153 \pm 58 \mu\text{g}/\text{cm}^2$ (H_2O). If we use a reported density of granite of 2.64 g/cm^3 (Hasai et al. 1987), the weight ratio between the water content and granite is evaluated as $0.29 \pm 0.11\%$ wt (H_2O).

In the reference (Hasai et al. 1987), the water content is reported as four different terms: penetration water, adsorptive water (I), adsorptive water (II) and bound water. Our new result of the water content would correspond to the adsorptive water (I and II) and bound water. The penetration water, which is derived from rain and dew, may have escaped during the slice preparation. This new result for the water content, which is the water content from the sum of the three terms, agrees with the previous result of $0.51 \pm 0.10\%$ wt (H_2O) (Hasai et al. 1987) within the uncertainty.

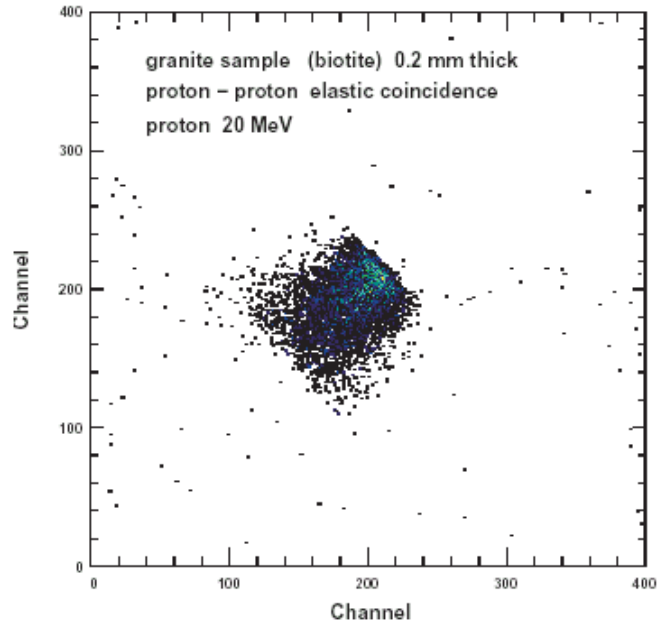


Figure 2. Two-dimensional spectrum of proton-proton elastic recoil coincidence measurement. The sample composition is assumed to be biotite.

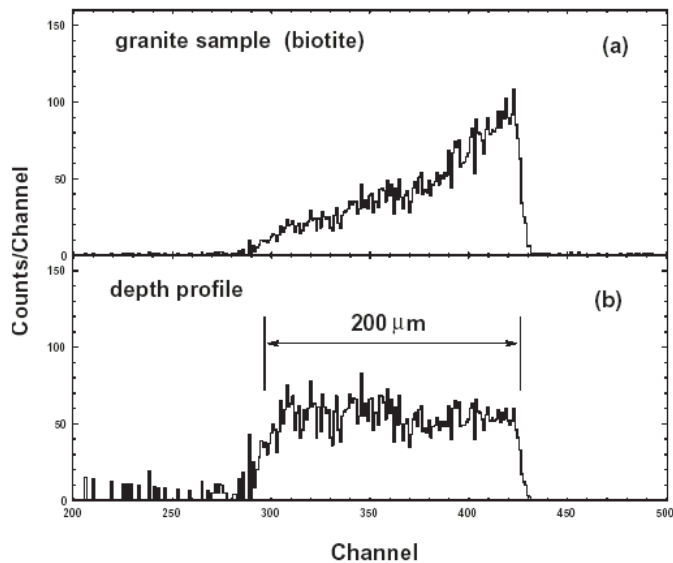


Figure 3. (a) Projection spectrum deduced from the two-dimensional spectrum shown in Figure 2. The yield is shown as a function of the sum of proton energies. (b) Depth profile of hydrogen content converted from the spectrum (a) by assuming an exponential function.

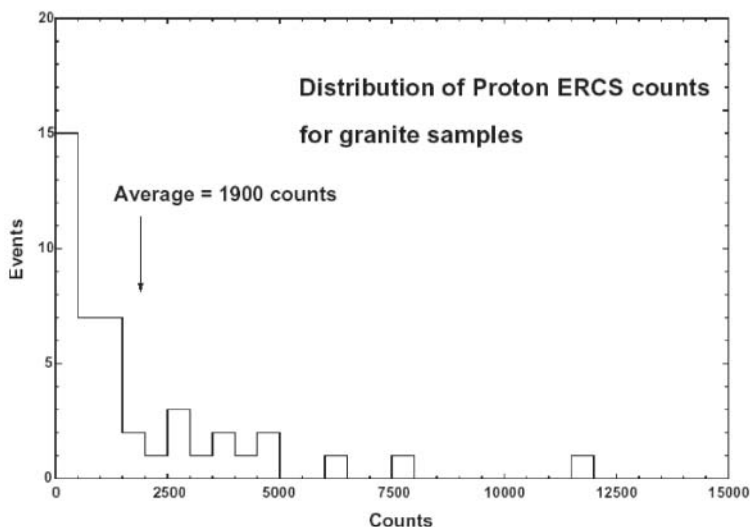


Figure 4. Distribution of proton-proton coincidence counts measured for the 44 irradiation points on granite samples.

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