Chapter 8

ACTIVATION MEASUREMENTS FOR THERMAL NEUTRONS

Part H. ^{152}Eu and ^{36}Cl Intercomparison Study

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Introduction

Discrepancies have been observed between thermal neutron measurements in samples from Hiroshima and calculations based on DS86 (e.g., Roesch 1987; Straume et al. 1992; Shizuma et al. 1993). The discrepancies suggested a trend of lower measured than calculated thermal neutron activation near the hypocenter and larger measured than calculated activation at distances beyond about 1,100 m ground range. There appeared to be some differences in activation response between the ^{36}Cl and ^{152}Eu as well.

It was decided that direct intercomparison of measurements could provide better understanding of these discrepancies. Therefore, as part of the DS02 reassessment effort, an international intercomparison study of ^{36}Cl and ^{152}Eu measurements was carried out. The results are presented in this section. The methods of measurements and the data used here are described in further detail in other sections of this report, as follows: ^{152}Eu data from Komura et al. (Chapter 8, Part I), ^{36}Cl data from Straume et al. (Chapter 8, Part D), Rühm et al. (Chapter 8, Part E), and Nagashima et al. (Chapter 8, Part F).

The intercomparison study had the following objectives: (1) provide comparison of ^{36}Cl activation measured by three independent laboratories (Tsukuba, Livermore, and Munich) in granite samples from Hiroshima; (2) compare ^{36}Cl measurements with ^{152}Eu measurements in Hiroshima; and (3) compare both ^{36}Cl and ^{152}Eu with DS02 sample-specific calculations. Agreement between the independent methods and the different laboratories (which measured different parts of the same granite samples) would provide support for the reliability of the measurements. Also, if the independent sample-specific calculations are in agreement with the

measurements, one could conclude with some degree of confidence that both the measurements and the calculations are essentially correct.

It was understood that differences in sample aliquots, sample preparation, and neutron cross sections could potentially contribute to discrepancies. Therefore, mixtures of standard solutions were prepared of Eu and Cl and irradiated using a ^{252}Cf neutron source available at Hiroshima University (Hoshi et al. 1988). Following irradiation, coded aliquots of these samples were provided to the measurement groups. These efforts involved colleagues at Hiroshima University, Kanazawa University, Tsukuba University, the Technical University and the Ludwig Maximilians University of Munich, the Lawrence Livermore National Laboratory, and the University of Utah.

Selection of Granite Samples for Intercomparison

Granite Samples

We selected granite samples that were measured previously by Shizuma et al. (1993). These are sample numbers 1 through 9 in Table 1. Sample numbers 10 through 16 were large-distance granite samples from the Hiroshima area not exposed significantly to bomb neutrons and were used here to provide background values for the ^{152}Eu and ^{36}Cl measurements. These samples were collected specifically for this intercomparison study from temples in the Hiroshima city area. Various types of granite stones were collected. A stone manufacturer was asked to provide information on where these granite samples originally came from. Stone number 11 was obtained from the Kannonji temple, which seemed to come from Kurahashi Island or a similar place, because its color was rather white. The Iyo stone or Oshima stone in Table 1 (Number 12) was believed to be brought from Iyo or Oshima Island in Shikoku, which had a characteristic gray color. The local stones identified in Table 1 are believed to be very old tombstones taken from the Hiroshima area, which is on a large granite plate. The local stones appeared very old from the rough surfaces of the stones.

Sample number 17 of the Kikkawa Ryokan was selected as a long-distance (1,424 m ground range) exposed sample. Sample number 18 was obtained from a core taken from the former Faculty of Science building, Hiroshima University, to measure the depth profile of ^{36}Cl in granite at a distance where a substantial fraction of the measured ^{36}Cl would be from natural background (Chapter 8, Part G). One 16-cm diameter granite core sample was taken and cut into a 0-5 cm near surface sample and a 5-15 cm deep sample. Sample 18 was used for the measurement of ^{152}Eu only, but without success. That is, the ^{152}Eu in the sample was below the detection limit.

Sample Preparation

Sample preparation for the ^{36}Cl measurements is described in each of the sections on ^{36}Cl measurements (Chapter 8, Parts D, E, and F). Here we describe only the sample preparation for ^{152}Eu. All granite samples were cut from original samples into five pieces. The size of each piece was dependent on the sample. Three out of five pieces were sent to Rühm (Munich), Straume (Livermore), and Nagashima (Tsukuba) for ^{36}Cl measurements. For ^{152}Eu measurement, one piece from every granite sample was sent to Nichika (Nakagyo-ku, Kyoto) and powdered into greater than 100 mesh (less than 254 μm). The weight of these powder samples was at most 1 kg. Then these powder samples were sent to the Japan Chemical Analysis Center, where they were

Table 1. Samples used for intercomparison of Eu and Cl measurements

No.	Sample	Measure-ment	Stone type	Original weight (g)	Extracted weight (g)	Ground range (m)
1	Motoyasu Bridge. Railing	Eu, Cl		1000	22.1	134
2	Shirakami Shrine Fence	Eu, Cl		340	5.47	504
3	Myochoji Temple 7	Eu, Cl		800	26.3	639
4	Old Prefectural office	Eu, Cl		1000	19.7	877
5	Honkeiji Temple	Eu, Cl		500	10.7	896
6	Shingyoji Temple 1	Eu, Cl		600	19.4	915
7	Enryuuji Temple 5-1	Eu, Cl		440	16.5	925
8	City Office pavement	Eu, Cl		1000	17.2	1022
9	Kozenji Temple 6-1	Eu, Cl		1000	23.1	1177
10	Senngyoji temple	Eu, Cl	Iyo stone	1000	27.8	
11	Kannonji Temple	Eu, Cl		1000	21.0	
12	Senzoubo	Cl	Iyo or Oshima stone			8790
13	Senzoubo	Cl	local stone			8790
14	Senzoubo	Eu, Cl	local stone	962.1	20.8	8790
15	Myokenji Temple	Eu, Cl	local stone	914.6	14.6	7610
16	Myokenji Temple	Cl	local stone			7610
17	Kikkawa Ryokan	Eu, Cl		1300	40.4	1424
18	Surface core of Old Faculty of Hiroshima University (E-building)	Eu, Cl	0-5 cm depth			1385
	Deeper core of Old Faculty of Hiroshima University (E-building)	Eu, Cl	5-15 cm depth			1385

Note: All of the stone types are granite, including exposed samples and background samples. Ground ranges for samples 12-16 are original values.

dissolved and Eu extracted. The chemical separation method was identical to that used previously by Shizuma et al. (1993). The weights of the residual materials containing the extracted Eu were from 15 to 40 g. These samples were sent to Komura's laboratory and measured with Ge detectors in the very low background Ogoya tunnel (Chapter 8, Part I).

Preparation of Mixed Eu and Cl Standard Samples and Neutron Exposure

A Eu standard solution for atomic absorption analysis was obtained from Wako Pure Chemical Industries, Ltd. (Chyu-o-ku, Osaka). The solution included 1,000 ppm of Eu. Its chemical composition was $Eu(NO_3)_3$ in 1 mol/l HNO_3. A chloride ion standard solution for ion chromatography was obtained from Kishida Chemicals Co. Ltd. (Chyu-o-ku, Osaka). It included 10,000 ppm of Cl as a chemical component of NaCl. From 1 ml of both standard solutions, 2 ml of mixed standard solution was prepared in test tubes in which 1 mg of Eu and 10 mg of Cl was included.

For irradiation of neutrons, we used a ^{252}Cf fission neutron source (Hoshi et al. 1988). Two types of neutron fields were prepared. In the first type of field, test tubes containing mixed

solutions were sandwiched with 5-cm thick nylon plate in front and 10-cm thick nylon plate as a backing and irradiated with ^{252}Cf neutrons. In the second type of field, test tubes were sandwiched with Newlite plate (polyethylene with 3.7% boron, see Hoshi et al. 1992) in the same manner as nylon. We call the former type, which used nylon, "thermal neutron irradiation" and the latter irradiation with Newlite "epithermal neutron irradiation."

Five test tubes of the 2 ml standard solutions were irradiated simultaneously. Neutron fluence of thermal and epithermal were monitored by gold foils of 50 μm thickness with and without Cd. Self-shielding correction factors of the gold foils for thermal and epithermal neutrons were experimentally determined using "thermal neutron irradiation" and "epithermal neutron irradiation" as shown in Figure 1. The irradiated thermal and epithermal neutron fluences were 3.19×10^{11} and 1.76×10^{10} (n/cm^2), respectively, for the "thermal neutron irradiation" and 1.13×10^{10} and 3.25×10^{10} (n/cm^2), respectively, for the "epithermal neutron irradiation." It required about 6 days and a few weeks, respectively, for the two kinds of irradiation.

In order to verify possible differences of the actual neutron fluence among irradiated tubes, all samples were measured using a Ge detector to evaluate ^{152m}Eu and ^{24}Na activity. After this measurement, three pairs of irradiated samples (one from "thermal neutron irradiation" and the other from "epithermal neutron irradiation") were sent to Livermore, Munich, and Tsukuba laboratories to measure ^{36}Cl. The results of the Ge measurements are summarized in Table 2. The activities of ^{152}Eu in aliquots of the liquid samples were also measured in Ogoya Laboratory, Kanazawa University. The obtained activities are consistent within the error. The ratios of these yields to the average values are plotted in Figure 2. There is no obvious difference among 5 tubes

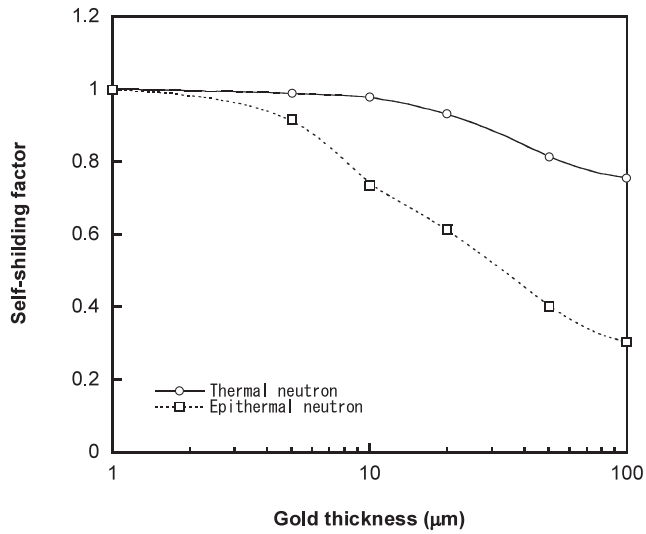

Figure 1. *Dependency of self-shielding factor of gold foil on thickness. The self-shielding factors were experimentally measured by changing the thickness of bare gold foils as shown in this figure. The correction factors obtained as the ratio of 1 μm thickness to the used 50 μm thickness are 0.813 and 0.402 for the "thermal irradiation" and "epithermal irradiation," respectively.*

Table 2. Results of the activity measurements of ^{152m}Eu and ^{24}Na for the standard solutions

No.	Activity (Bq)									Neutron fluence (Au-equivalent)		
	^{152}Eu	% error	Ratio	^{152m}Eu	% error	Ratio	^{24}Na	% error	Ratio	Φ_{th}(n/c m^2)	Φ_{epi}(n/c m^2)	R_{Cd}
<Thermal neutron irradiation>												
1	4.82	0.95	1.04	3119	0.33	1.01	46.7	1.63	1.03			
2	4.57	0.98	0.986	3043	0.35	0.99	45.2	1.67	0.99			
3	4.65	0.95	1.00	3049	0.38	0.99	44.8	1.73	0.98	3.19E+11	1.76E+10	5.86
4	4.55	0.97	0.981	3062	0.39	0.99	44.7	1.75	0.98			
5	4.58	0.98	0.988	3136	0.21	1.02	46.4	0.86	1.02			
<Epithermal neutron irradiation>												
6	1.01	2.1	1.03	326.8	1.13	1.09	4.17	5.34	1.03			
7	1.06	2.1	1.08	305.8	1.20	1.02	4.05	5.28	1.00			
8	0.953	2.0	0.972	291.6	1.37	0.97	3.89	5.76	0.96	1.13E+10	3.25E+10	1.27
9	0.964	2.0	0.983	288.3	1.60	0.96	4.51	6.77	1.11			
10	0.914	2.0	0.932	277.8	0.88	0.96	3.14	3.04	0.89			

Note: Numbers 1 to 5 are for thermal neutron irradiations using nylon plates. Number 6 to 10 are for irradiations using Newlite plates. The ratio is that for the averages. The measurements of neutron fluence have been performed using 50-μm thick Au plates. For the neutron fluences, self-shielding is corrected for the thermal neutrons and epithermal neutrons. R_{Cd} indicates cadmium ratios for thermal and epithermal irradiation. Errors shown in this table are only statistical. Systematic uncertainties are estimated to be 4% for ^{152}Eu, and 7% for ^{152m}Eu and ^{24}Na measurements. The ^{152}Eu activities were cross-checked both at Hiroshima University and Ogoya laboratory. The results are consistent within the error.

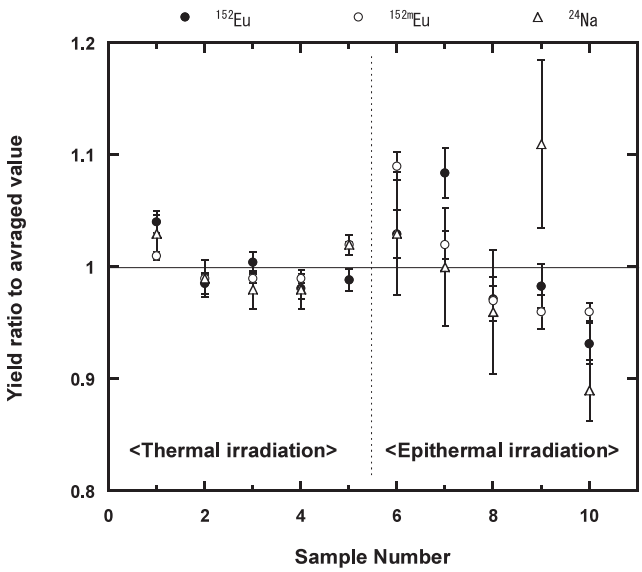

Figure 2. *Yield of the ^{152m}Eu and ^{24}Na for the standard solutions irradiated with thermal and epithermal neutrons. Samples from 1 to 5 are thermal neutron irradiations and from 6 to 10 are epithermal neutron irradiations. Samples No. 2 and 8, No. 4 and 7, and No. 3 and 9 were sent to Munich, Livermore, and Tsukuba, respectively. Remaining No. 1, No. 5, No. 6 and No. 10 samples were kept in Hiroshima University and Ogoya laboratory.*

of liquid simultaneously irradiated to “thermal neutron irradiation” or “epithermal neutron irradiation.”

We also measured cadmium ratios, which are 5.86 and 1.27, respectively, for “thermal neutron irradiation” and “epithermal neutron irradiation.” The thermal and epithermal neutron fluences can be estimated from the cadmium ratio and resonance integral as (Beckurts and Wirtz 1964);

$$R_{CD} - F_{CD} = \frac{\phi_{th}}{\phi_{epi}} \frac{\sqrt{\pi}}{2} g(T_n) \sqrt{\frac{293.6}{T_n}} \sigma_{act}(v_0) / \int_{E_{CD}}^{\infty} \sigma_{act}(E) dE \quad (1)$$

where F_{CD} is the absorption correction in 0.5 mm-thick cadmium foil, E_{CD} is the energy cut-off for cadmium, $g(T_n)$ is the correction factor for non-1/v absorber, and the denominator of equation (1) is the resonance integral of gold foil (1,535 barns). This approximation is used to estimate averaged flux of thermal and epithermal neutrons. For the “thermal neutron irradiation” the fluence of thermal neutrons was 18 times the epithermal neutrons, while for the “epithermal neutron irradiation” that factor was only 0.35 times. For the atomic-bomb neutron spectrum in Hiroshima, this factor is 0.5 at 100 m ground range and 0.3 at 2,000 m ground range. Therefore, the epithermal to thermal ratio of the atomic-bomb neutrons is similar to that for the “epithermal neutron irradiation” field.

Results and Discussions

Results of the new measurement of ^{152}Eu and ^{36}Cl in Hiroshima granite samples are shown in Table 3 and Figure 3 together with the values obtained by the DS02 calculation. Calculated values in Table 3 are corrected by the thermal neutron transmission factors as discussed in Chapter 8, Part J. The ^{36}Cl calculation includes a small contribution of ^{39}K(n,α)^{36}Cl reaction that amounts 0.3 to 3% of the total production, depending on distance from the hypocenter. In Figure 4, ^{152}Eu data are converted into ^{36}Cl-equivalent using the ratio of thermal activation reactions of ^{151}Eu(n,γ)^{152}Eu to ^{35}Cl(n,γ)^{36}Cl (5,900 vs. 46.3 barn). For the ^{36}Cl data plotted in Figure 4, backgrounds are subtracted from the measured values in Table 3 in the following way. For the Tsukuba data, the background of (1.6 ± 0.5) × 10^{-13} is assumed for all samples based on the

Table 3. Results of nine exposed granite samples in Hiroshima

Sample	GR (m)	DS02	^{36}Cl/Cl			^{152}Eu/Eu (Bq/mg)	
			Tsukuba	Livermore	Munich	DS02	Ogoya
Motoyasu	134	1.77E-10	1.89E-10 ± 1.30E-11	1.67E-10 ± 1.14E-11	1.67E-10 ± 1.20E-11	92.89	99.4 ± 5.5
Shirakami	504	3.02E-11	2.22E-11 ± 1.80E-12	2.18E-11 ± 6.57E-13	1.88E-11 ± 1.50E-12	15.71	15.2 ± 0.89
Myochoji	639	1.17E-11	1.07E-11 ± 9.90E-13	9.72E-12 ± 3.38E-13	1.14E-11 ± 1.00E-12	6.14	6.2 ± 0.38
Old Prefect.	877	1.95E-12	2.45E-12 ± 1.50E-13	4.15E-12 ± 2.23E-13	2.21E-12 ± 2.10E-13	1.00	1.57 ± 0.10
Honkeiji	896	1.68E-12	1.43E-12 ± 1.00E-13	1.39E-12 ± 4.18E-14	1.77E-12 ± 2.00E-13	0.87	0.99 ± 0.07
Shingyoji	915	1.45E-12	1.17E-12 ± 1.60E-13	1.32E-12 ± 6.48E-14	9.90E-13 ± 1.10E-13	0.75	0.78 ± 0.06
Enryuuji	925	1.34E-12	1.78E-12 ± 2.60E-13	1.33E-12 ± 1.27E-13	1.68E-12 ± 2.10E-13	0.69	1.06 ± 0.09
City Office	1022	6.33E-13	4.06E-13 ± 6.10E-14	-	3.30E-13 ± 4.00E-14	0.32	0.27 ± 0.09
Kozenji	1177	1.94E-13	2.13E-13 ± 4.20E-14	4.35E-13 ± 2.60E-14	2.20E-13 ± 4.00E-14	0.10	0.15 ± 0.03

Note: DS02 calculated results are *in situ* values corrected by transmission factors at the sample location (see Chapter 8, Part J). Backgrounds for ^{36}Cl measurements are not subtracted. GR (m) means DS02 ground range, which was corrected according to the new hypocenter.

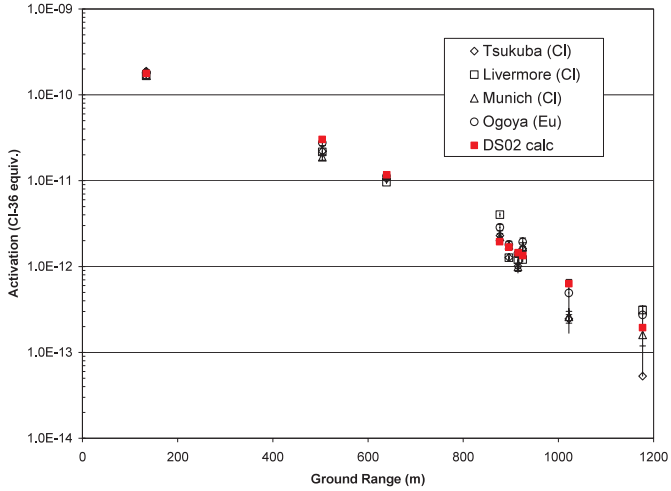

Figure 3. *Comparison of the ^{152}Eu and ^{36}Cl measurement data with sample-specific calculations based on DS02. The sample modeling calculations accounted for sample self-shielding and the effect of local environment. Backgrounds of ^{36}Cl are subtracted, and ^{152}Eu is converted into ^{36}Cl equivalent (see text).*

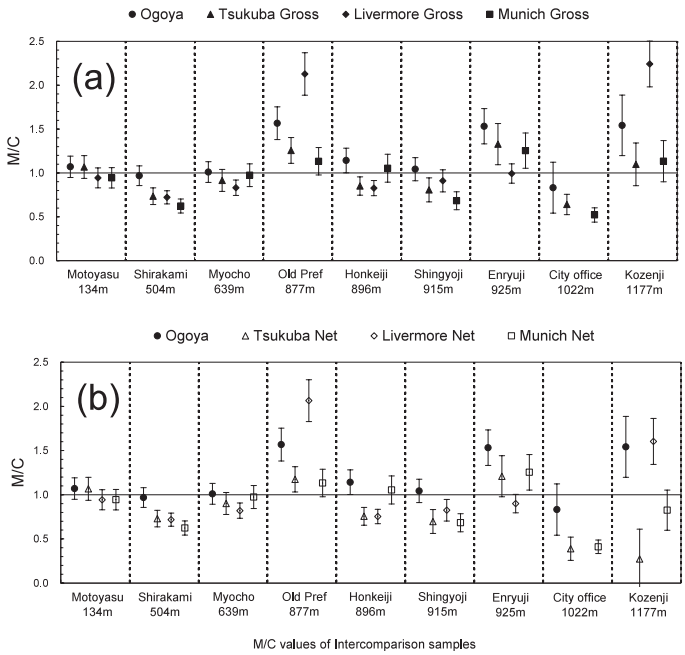

Figure 4. *The measured to DS02-calculated (M/C) ratio for intercomparison samples. (a) Without background subtraction for ^{36}Cl measurement. (b) With background subtraction for ^{36}Cl measurements (for details see text).*

measurement of distant control samples. For the Livermore data, $(1.24 \pm 0.3) \times 10^{-13}$ obtained from deep concrete sample measurements is used for the background. For the Munich data, the background values of (0.7 ± 0.09) and $(0.6 \pm 0.09) \times 10^{-13}$ estimated by the original method (see Chapter 8, Part G) are used for City Office and Kozenji samples, respectively, while the background is not subtracted for other samples. The contribution of natural neutrons to the formation of ^{152}Eu is considered negligibly small (see Chapter 10, Part A).

Good agreement can be seen in Figure 3 between the measurements and the sample-specific calculations based on DS02. Although there is a tendency that scattering of measured values becomes larger at distances beyond 1,000 m, the ^{152}Eu and ^{36}Cl measurements made as part of this intercomparison study support the DS02 calculations (see also Chapter 12, Part D).

Figure 4 shows a comparison among four laboratories of the measured to DS02 calculated ratio (M/C) for each sample: (a) plotting without background subtraction and (b) with background subtraction. The latter M/C values are listed in Table 4. To compare the sample-specific calculations with the measurements, the calculations were converted here from the DS02 fluence into values that can be compared directly with the measurement results. Briefly, it is estimated that the largest random uncertainties are transmission factors (~7%) and sample distances (~5%). The largest systematic uncertainty is from the activation cross section (<5%). It is estimated that the total random uncertainty associated with the calculation for each sample derived from the DS02 free-field fluence is about 10%. This uncertainty has been propagated together with the experimental uncertainties, to the calculated uncertainties of the M/C ratios given in Table 4 and shown in Figure 4.

The effect of the background subtraction for the ^{36}Cl data is significant at the most distant sample, Kozenji, while the background subtraction does not show a significant effect within 1,000 m. It is clear that most M/C values are around unity, and the measured values from all involved laboratories are consistent each other.

Figure 5a shows the ^{36}Cl/^{35}Cl ratios obtained at Tsukuba, Livermore, and Munich, normalized to the corresponding ^{152}Eu/^{151}Eu ratios obtained by gamma spectrometry at Ogoya, and to the corresponding to (n,γ) cross section for the ^{35}Cl(n,γ)^{36}Cl and ^{151}Eu(n,γ)^{152}Eu reactions of 43.6 b and 5,900 b, respectively. The ^{36}Cl data used to calculate the ratio in Figure 5a are corrected for the background and ^{39}K(n,α)^{36}Cl reaction. As can be inferred from Figure 5a, the ratios are

Table 4. Measured to calculated ratio based on DS02

Sample	GR (m)	^{36}Cl/Cl			^{152}Eu/Eu
		Tsukuba	Livermore	Munich	Ogoya
Motoyasu	134	1.07 ± 0.13	0.94 ± 0.11	0.94 ± 0.12	1.07 ± 0.12
Shirakami	504	0.73 ± 0.09	0.72 ± 0.08	0.62 ± 0.08	0.97 ± 0.11
Myochoji	639	0.90 ± 0.12	0.82 ± 0.09	0.97 ± 0.13	1.01 ± 0.12
Old Prefect.	877	1.17 ± 0.14	2.06 ± 0.24	1.13 ± 0.16	1.57 ± 0.19
Honkeiji	896	0.76 ± 0.10	0.75 ± 0.08	1.05 ± 0.16	1.14 ± 0.14
Shingyoji	915	0.70 ± 0.13	0.82 ± 0.10	0.68 ± 0.10	1.04 ± 0.13
Enryuuji	925	1.21 ± 0.23	0.90 ± 0.12	1.25 ± 0.20	1.53 ± 0.20
City Office	1022	0.39 ± 0.13	- -	0.41 ± 0.08	0.83 ± 0.29
Kozenji	1177	0.27 ± 0.34	1.60 ± 0.26	0.82 ± 0.23	1.54 ± 0.34

Note: Backgrounds for ^{36}Cl measurements are subtracted (see text). GR (m) means DS02 ground range, which was corrected according to the new hypocenter.

scattered around a value of 1, indicating good agreement between the ^{36}Cl and ^{152}Eu results, even for the large-distant samples. An exception might be the results obtained for the City Office, which are consistently lower for the ^{36}Cl data obtained at Tsukuba and Munich, compared to those obtained for ^{152}Eu. Other outliers might be the ^{36}Cl results for the samples from the Old Prefectural Office and Kozenji obtained at Livermore, which appear to be much higher than those obtained at Tsukuba and Munich for ^{36}Cl. The ratio averaged over all measured samples in Figure 5a is 0.87 ± 0.03, and the average obtained by excluding the two higher data at Livermore is 0.86 ± 0.03.

Figure 5b shows the similar results for the standard solution measurements. Both "thermal neutron irradiation" and "epithermal neutron irradiation" are indicated. The results seem to be closer to unity than were the granite samples (Figure 5a). The averaged Cl/Eu ratios for thermal neutron irradiation are 1.08 ± 0.05 and for epithermal neutrons 0.91 ± 0.05, respectively. These ratios show consistency of the measurements at three laboratories.

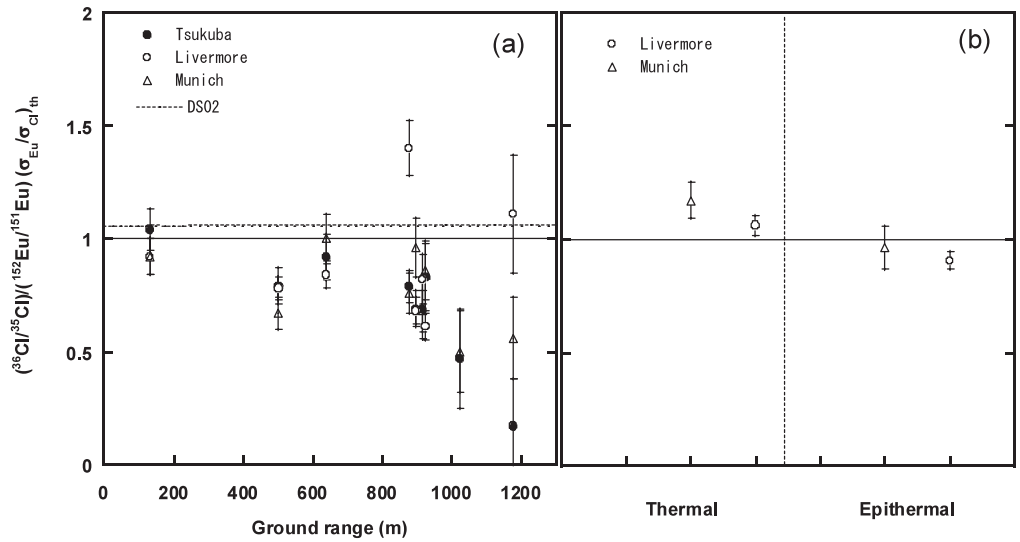

Figure 5. *Comparison of the Cl measurements against Eu measurements. Ratios of the specific activities of ^{36}Cl to ^{152}Eu are plotted as the values divided by corresponding thermal neutron cross-section ratios. Figure 5a indicates the results for the actual granite samples in Hiroshima. Backgrounds and contributions of ^{39}K(n,α)^{36}Cl reaction are subtracted for ^{36}Cl data. Figure 5b indicates measurements using standard Eu-Cl samples irradiated with ^{252}Cf neutron source. "Epithermal" indicates the ^{252}Cf irradiation sandwiched with Newlite, which includes 3.7% boron, and "thermal" indicates irradiation sandwiched with nylon. See text for more explanation.*

Conclusions

An intercomparison study was completed using parts from the same granite samples collected within 1,200 m ground range from the Hiroshima hypocenter. Also, liquid standard samples that contained the same amount of Eu and Cl were irradiated by thermal and epithermal neutrons and measured by the independent laboratories. The ^{152}Eu data were obtained at Ogoya and the ^{36}Cl data were obtained at Tsukuba, Livermore, and Munich. The results of this intercomparison study demonstrate that the ^{152}Eu and ^{36}Cl measurements agree well with each other and also with the sample-specific calculations based on DS02.

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