Chapter 5 Appendix 15

STUDIES ON FISSION TRACKS AND DISTRIBUTIONS OF URANIUM AND RARE EARTHS IN GRANITE MATERIALS

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Many materials contain fossil records of the slow spontaneous fission of uranium they contain as an impurity. Fission fragments, heavy charged particles released in each fission event, produce microscopic trails of radiation damage that may persist over geological times and may be developed to a size observable under an optical microscope by a suitable etching treatment. Such tracks are also produced by fissions induced by thermal neutron irradiation of the uranium.

When the material is heated sufficiently, it anneals and the microscopic trails become shorter and narrower. The track density decreases, because the chemical etchant will not reach some of the shortened tracks.

Measurements of track densities before and after annealing can be used, along with laboratory studies of annealing rates, to determine the annealing temperature. Also, the track density of induced fissions is related to the concentration of uranium and the fluence of neutrons to which it was exposed. If the track density due to induced fissions can be distingusihed from that due to fossil tracks, estimates of either the concentration or the fluence can be made if the other is known.

Two such materials (one a fragment of a granite paving stone, the other a piece of stained glass from a cathedral window) that had been exposed to the atomic bomb at Nagasaki were used in the present work. The fossil record in zircons in the granite was used to estimate the temperature to which it had been exposed in the bombing. Induced fissions were used to estimate the concentration of uranium in the zircons. Nonuniform heating and cooling and nearly uniform exposure to the neutrons make the granite sample unsuitable for determining the neutron fluence from the bomb. Induced fissions in the stained glass were used to

Taken from a Master's thesis by H. Matsuda. The introductory material in the thesis has been shortened.

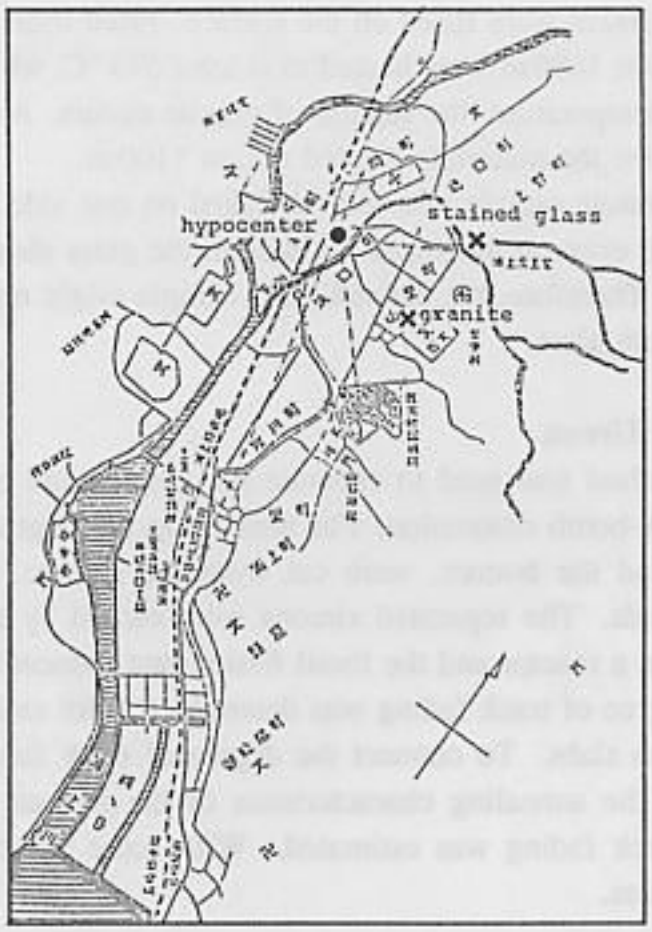

Figure 1. Map of the neighborhood of the hypocenter at Nagasaki. The sampling point for the granite is 415 m and that of the stained glass is 520 m from the hypocenter

estimate the concentration of uranium and the thermal neutron fluence from the A-bomb. Annealing of tracks in glass was also studied.

Samples and Geometrical Setting

A block of granite ($3 \times 6 \times 7$ cm, about 350 g) and three broken pieces of stained glass (3, 3, and 4 mm on the longest side) were the samples for this work. All of them had been exposed to the A-bomb in Nagasaki.

The granite block was from a paving stone that had been in front of the Takatani's at that time. There was no shielding of the paving stone from the radiation of the A-bomb. The distance from the hypocenter was 415 m as shown in Figure 1. Its slant range was calculated to be 655 m.

The pieces of stained glass were provided by the Culture Hall of Nagasaki. This stained glass was originally located at the upper side of the entrance of the Uragami Cathedral; it was on the west side of this structure and faced the hypocenter at the time of the detonation. The distances from the Cathedral to the hypocenter and to the detonation point were 520 and 730 m, respectively.

It has been reported that all the houses in the area inside 2000 m from the hypocenter were burned down. The surfaces of granite rocks found within 1680 m were flaked off, and

roof tiles within about 1100 m were fused on the surface. From these facts, it was assumed that all the materials within 1680 m were heated to at least 573 °C, which is the modification point of quartz. At this temperature, the flaking of granite occurs. A minimum temperature of 1200 °C is estimated for the materials located within 1100 m.

The surface of the granite sample was indeed flaked on one side. But the stained glass did not seem to be fused, even on the surface, although the glass should have been fused at such high temperatures. Therefore, the stained glass sample might not have been irradiated at the surface of the whole glass.

Fession-Tract Study of Zircon

The fission-track method was used to estimate the temperature reached by the granite surface exposed to the A-bomb detonation. For measuring the degree of track fading, two slabs, the top surface and the bottom, were cut from the granite. Zircon crystals were isolated with heavy liquids. The separated zircons were etched by a new technique. The zircons were irradiated in a reactor and the fossil fission and induced fission-track densities were measured. The degree of track fading was determined from ratios of fossil to fission-track densities in the two slabs. To connect the degree of track fading with the time and temperature of heating, the annealing characteristics of the zircons were studied and the activation energy for track fading was estimated. With some assumptions this permitted evaluating the temperatures.

Isolation of Zircons. The sample block of granite was cut parallel to the exposed surface with a diamond cutter (Figure 2) to obtain a slab from both the top and the bottom surface.

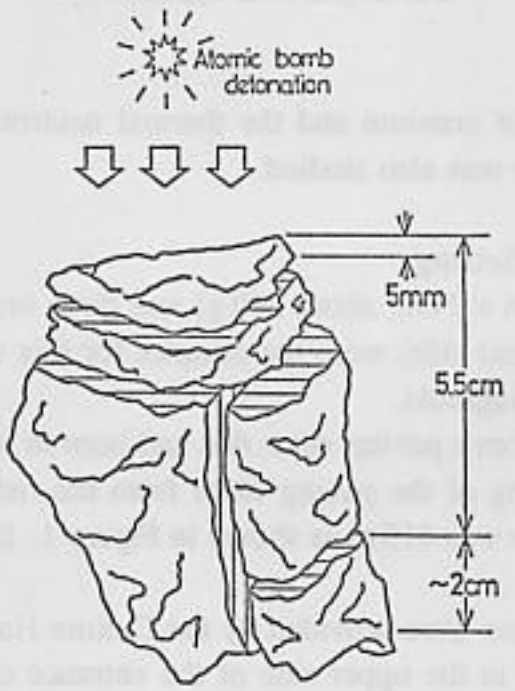

Figure 2. A drawing of the granite sample collected at the Takatani's in Nagasaki city. The top surface (5 mm thickness) and the bottom (about 2 cm thick, 5.5 cm from the irradiated surface) was used for the fission-track experiment. A slab (1 cm thick, 4.5 cm from the irradiated surface) was used for neutron activation analysis

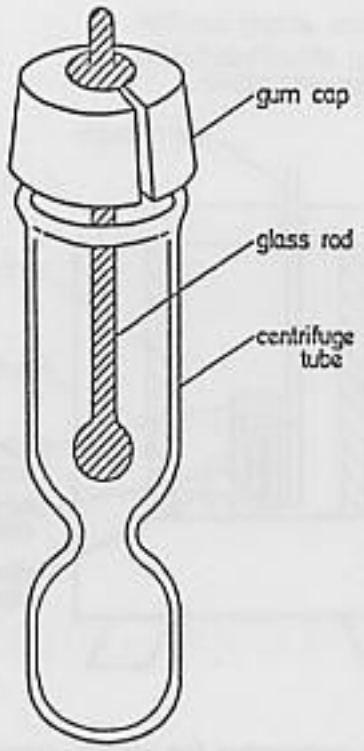

Figure 3. A drawing of the centrifuge tube with the constriction. A glass ball on the end of the glass rod is held by the gum cap during centrifugation

The thicknesses and weights were 0.5 and 1.5 cm and 14.1 and 24.8 g for the top and the bottom slabs, respectively.

The zircon content was determined as follows. The samples were crushed with an iron mortar to pass through a sieve of 60 mesh. The powder was put into water and stirred well. About one minute later, the muddy supernatant was discarded leaving the fine grains. The washing was repeated until the supernatant was transparent. After air-drying, the grains were transferred to a centrifuge tube with a constriction (Figure 3) and a heavy liquid, bromoform ($CHBr_3$, specific gravity 2.87) added. After stirring with an ultrasonic vibrator for about five minutes, a glass rod with a ball on one end was put in the tube as shown in Figure 3 and the tube centrifuged at 3000 rpm for five minutes. The tube was then closed with the ball on the rod and the liquid above the ball poured off. The grains left floating on the inside were washed with ethyl alcohol and poured out; the heavy grains were washed with ethyl alcohol and acetone and dried in the tube. Then methylene iodide (CH_2I_2, specific gravity 3.33) was added and the procedure repeated except that only acetone was used for washing. The heavy grains from this procedure were subjected to magnetic separation with a hand magnet. To the nonmagnetic fraction was added Clerici solution (an equivolume mixture of thallium formate and thallium malonate, each saturated at 20 °C, specific gravity 4.20); the result was washed with distilled water and dried. The remainder consisted of zircon and hornblende in a ratio of about one to one. The yield of zircon was about 40 mg per 20.3 g of sample rock (about 0.2%). The zircon crystals were very small (about $10 \times 10 \times 10\,\mu m$ to $50 \times 50 \times 300\,\mu m$). They were removed from the hornblende by picking them up with a small needle with a small amount of glue under a microscope ($\times 20$).

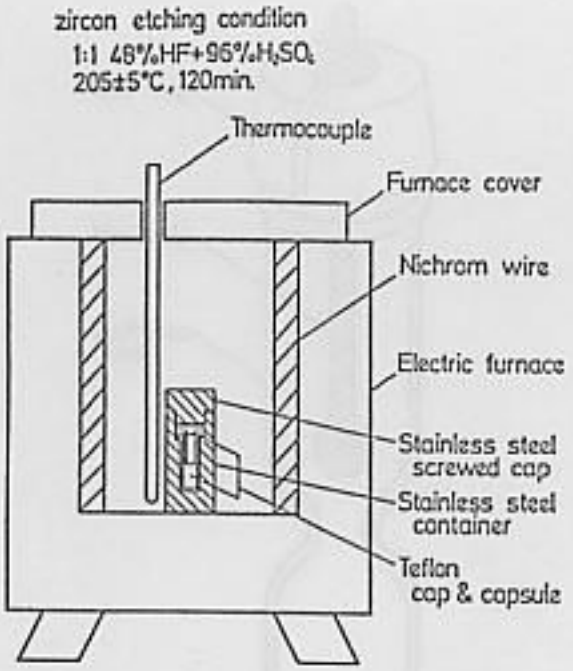

Figure 4. The etching apparatus for zircon crystals. The zircons are put into the teflon capsule together with etchant and etched with temperature and pressure

Chemical Etching of Zircon and Fossil Fission-track Counting. *Chemical Etching.* Because of the varied concentrations of minor impurities, even in the same kind of mineral, optimum etching conditions have to be determined for each mineral studied. The technique of Krishnaswami et al was used for the zircons in granite in the present work: The zircon grains were put in a teflon capsule and the capsule filled with etchant (1:1, HF and H_2SO_4) and covered with a teflon lid. The capsule was then fitted in a stainless steel container and a steel cap screwed on (Figure 4). The container was placed in an electric furnace as shown in the figure and heated as quickly as possible to about 200 °C. The temperature was measured with an alumel-chromel thermocouple and kept within ± 4.5 °C. The optimum etching time and temperature were estimated from several tens of trials to be 205 ± 2.5 °C for two hours from the time the required temperature was reached. Immediately after the etching, the container was thrown into a water bath. After cooling, most of the etchant was sucked from the teflon capsule with a glass capillary tube. The zircon grains were washed repeatedly with water and with ethyl alcohol and acetone in turn. Finally they were transferred onto a slide glass by sucking with acetone.

Fossil Fission-track Counting. When the etched zircon was observed, it was picked up and transferred to another slide glass. The etched fission tracks on the zircon crystal were observed and photographed using oil immersion under an optical microscope (×1000). Figure 5 shows some microphotographs of the fossil tracks.

Thirty nine zircon grains separated from the uppermost part and 23 from the deepest part of the sample granite were etched and the fossil fission tracks photographed as described above. Two or three faces of each zircon crystal were observed to count the tracks. The counted area was measured by counting squares on the graph paper on which the photograph

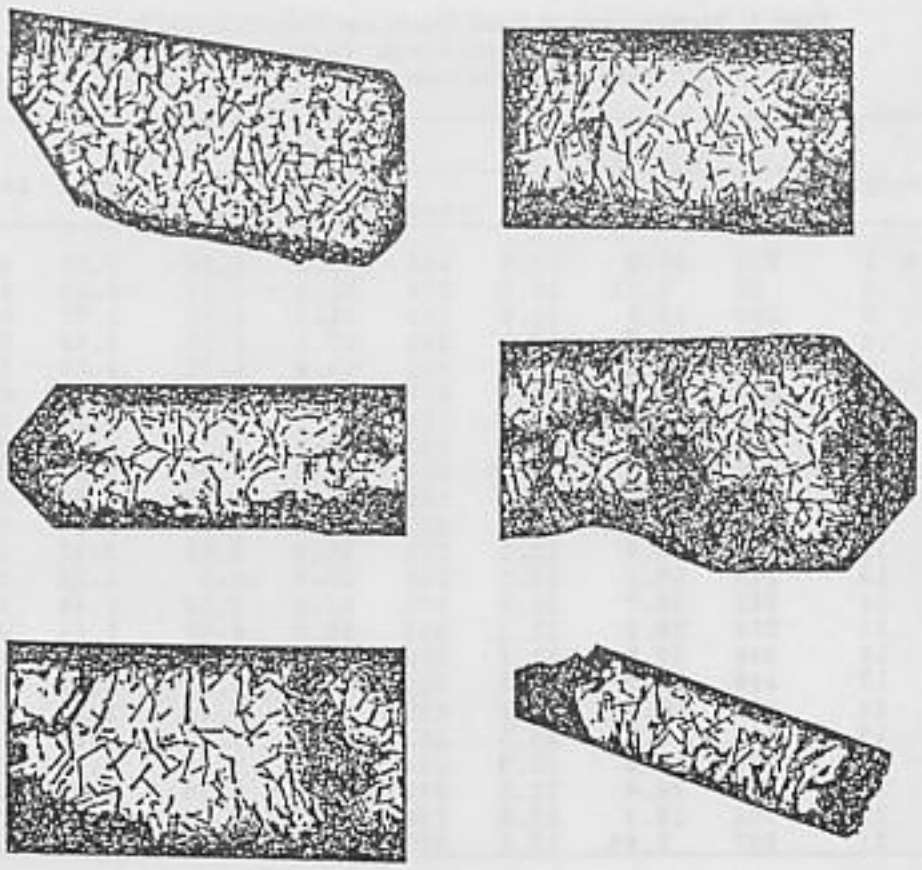

Figure 5. Naturally occurring fossil fission fragment tracks in zircon crystals isolated from granite, revealed by etching for 120 minutes in 1:1 48% HF/96% H_2SO_4 solution at 205 ± 5 °C. (× 700)

was drawn. The fossil fission-track densities are shown in Table 1 together with densities of the induced tracks, which will be described later.

Uranium Content of Zircon. Uranium was determined by exposing the sample to neutrons and counting the tracks due to thermal-neutron fission of the ^{235}U in the sample. To minimize interference from fossil tracks, the tracks were detected in a uranium-poor, external detector of mica or lexan placed in contact with the sample surface.

The uranium concentration, C, is proportional to the track density, ρ,

$$\rho = d\frac{^{238}A}{^{235}A}C\sigma\phi t \qquad (1)$$

where σ is the fission cross section of normal uranium, ϕ is the thermal-neutron fluence rate, t is the irradiation time, ^{235}A and ^{238}A are the abundances of ^{235}U and ^{238}U, respectively, and d is the density of the sample material. The probability that a fission fragment will reach the external detector decreases linearly with the distance from the surface of the sample and becomes zero for fissions beyond the range of the fission fragments from the surface. Therefore, the track density ρ can be represented by the average range of the fission fragments, R, and the surface area S as follows,

Table 1. Measurements of Fossil Tracks and Induced Tracks in Zircon Crystals Isolated from the Granite Sample. (a) Isolated from the Bottom Slab. (b) Isolated from the Top Surface

Sample		Fossil			Induced			Ratio	Error
		tracks	area	density	tracks	area	density		
a	1	235	16.8	13.9	166	83.7	1.98	7.07	0.71
	2	98	5.83	16.8	254	51.1	4.97	3.38	0.40
	3	180	12.9	14.0	185	31.3	5.90	2.37	0.25
	4	263	20.7	12.7	342	65.1	5.25	2.42	0.20
	5	734	55.6	13.2	409	64.8	6.31	2.09	0.13
	6	124	8.37	14.8	418	42.4	9.86	1.50	0.15
	7	515	37.7	13.7	162	50.8	3.19	4.29	0.39
	8	338	24.1	14.0	243	34.5	7.05	1.99	0.17
	9	168	16.8	9.99	188	29.2	6.44	1.55	0.16
	10	338	29.0	11.6	166	23.0	7.23	1.61	0.15
	11	215	16.1	13.4	313	44.9	6.97	1.92	0.17
	12	131	10.8	12.2	175	35.6	4.91	2.47	0.29
	13	240	19.1	12.5	298	29.5	10.1	1.24	0.11
	14	362	33.7	10.8	471	64.9	7.26	1.48	0.10
	15	254	20.7	12.3	393	58.9	6.67	1.84	0.15
	16	348	27.5	12.7	366	27.2	13.5	0.94	0.07
	17	295	28.7	10.3	301	50.4	5.97	1.72	0.14
	18	632	50.8	12.4	451	69.8	6.46	1.93	0.12
	19	136	8.75	15.5	162	21.0	7.70	2.02	0.23
	20	224	18.2	12.3	244	38.7	6.30	1.95	0.18
	21	332	26.4	12.5	371	54.7	6.78	1.86	0.14
	22	204	16.4	12.5	736	92.3	7.97	1.56	0.12
	23	107	7.85	13.6	357	35.5	10.1	1.33	0.12
b	24	641	63.0	10.2	174	59.3	2.93	3.47	0.30
	25	80	6.62	12.1	342	38.0	9.01	1.34	0.17
	26	197	15.6	12.7	440	50.0	8.97	1.44	0.12
	27	289	25.4	11.4	247	40.2	6.15	1.85	0.16
	28	762	71.7	10.6	301	47.4	6.35	1.67	0.11
	29	201	19.9	10.1	320	52.6	6.09	1.66	0.15
	30	214	22.7	9.43	272	59.5	4.58	2.06	0.19
	31	63	5.90	10.7	199	29.0	6.86	1.56	0.23
	32	263	19.9	13.2	380	45.8	8.30	1.59	0.13
	33	87	10.5	8.26	423	50.8	8.32	0.99	0.12
	34	289	25.4	11.4	203	27.8	7.31	1.56	0.14
	35	280	27.5	10.2	230	69.4	6.20	1.64	0.13
	36	409	48.8	8.38	490	88.4	5.55	1.51	0.10
	37	213	22.5	9.48	437	48.6	9.00	1.05	0.09
	38	578	58.6	9.87	290	42.7	6.79	1.45	0.10
	39	114	14.5	7.89	262	27.8	9.44	0.84	0.09

$$\rho = \frac{N_{tr}}{S\frac{R}{2}} = \frac{2\,N_{tr}}{S\,R} \tag{2}$$

where N_{tr} is the number of tracks counted. So the uranium concentration in ppm is found by combining Equations (1) and (2), as follows

$$C = \frac{2 \times 10^6}{\sigma\,\phi\,t}\,\frac{^{235}A}{^{238}A}\,\frac{M_u}{d\,N_{av}}\,\frac{N_{tr}}{S\,R} \tag{3}$$

where M_u is the mass number of uranium and N_{av} is Avagadro's number.

Experimental. The zircon crystals were exposed on a mica sheet that had been annealed sufficiently and fixed with triacetyl cellulose film by dissolving its surface with ethyl acetate. The mica sheet was then sandwiched between polycarbonate plates held with cellophane

tape. About 10 grains were put onto each mica sheet. These samples were placed in an irradiation tube and exposed in the rotary sample rack of the TRIGA Mark II reactor at the Atomic Research Institute of Rikkyo University to a thermal-neutron fluence rate of $4.9 \times 10^{11}\ cm^{-2}\ s^{-1}$ for 30 minutes. After several days to permit decay of induced radioactivities, the mica sheet was removed and etched with 48% hydrofluoric (HF) acid at room temperature for 30 minutes. Examples of the etched tracks are shown in Figure 6. Frequent "stars" are seen. The photograph was taken with a microscope at 500 × and the track density measured as described above. The track densities are listed in Table 2 and a frequency distribution of the uranium concentrations found is given in Figure 7.

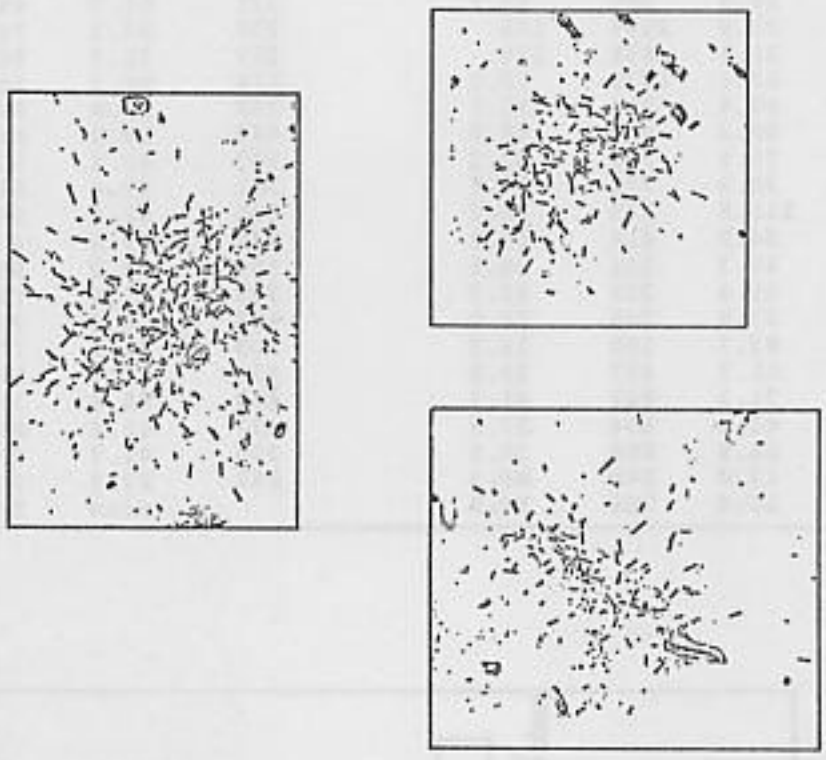

Figure 6. Induced fission tracks in mica sheets, revealed by etching for 30 minutes in 48% HF acid at room temperature. (× 500) An external surface of the mica in contact with a zircon crystal exhibits a "map" of the uranium distribution in the zircon. The track density is proportional to the uranium concentration

Activation Energy for Total Track Fading. *Activation Energy.* The repair of a damaged crystalline structure occurs by a complicated series of atomic processes. Nevertheless, some simple inferences can be drawn from the results of measurements of the kinetics of track repair in solids. The most useful type of information comes from heating a series of duplicate samples, each for a different time, t, and temperature, T, combination, and plotting the results on a diagram of ln(t) versus T^{-1}, an Arrhenius plot. It is usually (but not always) true that a single straight line separates the fading and no-fading regions on the diagram, indicating that a relation of the following form holds

Table 2. Results for the Uranium Concentration in Zircons Isolated from the Granite Sample, Determined by the Induced Fission-track Method

Flux n/cm^2	Tracks	Area $x10^{-6}cm^2$	U-conc. ppm	Error	Flux n/cm^2	Tracks	Area $x10^{-6}cm^2$	U-conc. ppm	Error
$10x10^{15}$	37	88.8	600	98.6	$8.8x10^{15}$	243	34.5	676	43.4
	33	52.5	905	158		188	29.2	618	45.1
	9	18.7	693	231		166	23.0	693	53.8
	31	65.0	687	123		313	44.9	669	37.8
	32	89.3	516	91.2		175	35.6	472	35.7
	67	67.3	1434	175		298	29.5	970	56.2
	12	49.3	351	101		471	64.8	698	32.2
	13	50.1	374	104		393	58.9	641	32.3
	15	29.0	745	192		366	27.2	1292	67.5
$9x10^{15}$	235	97.1	697	45.5		301	50.4	573	33.0
	293	151.6	557	32.5		451	69.8	620	29.2
	195	100.3	560	40.1		162	21.1	737	57.9
	249	81.1	884	56.0		244	38.7	605	38.7
	89	28.5	869	93.7		371	54.7	651	33.8
	254	33.8	2164	136		736	92.3	766	28.2
	62	21.5	831	106		357	35.5	965	51.1
	226	62.7	1038	69.0		174	59.3	282	21.4
	105	96.4	314	30.6		342	37.9	866	46.8
	186	80.3	667	48.9		440	50.0	845	40.3
	74	75.9	281	32.7		247	40.2	590	37.5
	81	30.3	770	85.6		301	47.4	610	35.2
	91	111.5	235	24.6		320	52.6	584	32.6
	79	54.9	414	46.6		272	59.5	439	26.6
	50	40.7	354	50.1		199	29.0	659	46.7
	69	55.9	355	42.7		380	45.8	797	40.9
	100	37.9	760	76.0		423	50.8	799	38.8
$8x10^{15}$	166	83.7	190	14.7		203	27.8	701	49.2
	254	51.1	477	29.9		430	69.4	595	28.7
	185	31.3	567	41.7		490	88.4	532	24.0
	342	65.1	504	27.3		437	48.6	863	41.3
	409	64.8	606	30.0		290	42.7	652	38.3
	418	42.4	946	46.3		262	27.8	905	55.9
	162	50.8	306	24.0			mean	545	4.8

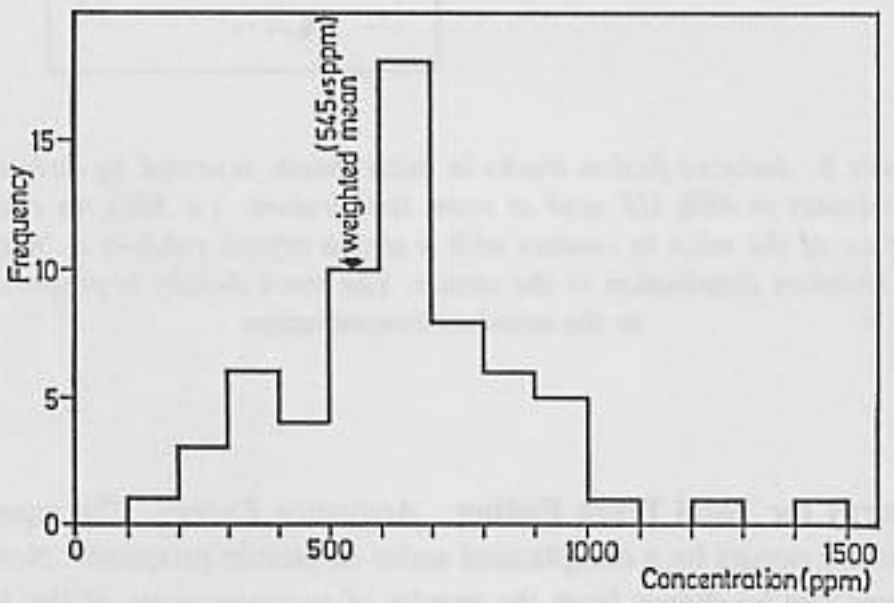

Figure 7. The frequency distribution of the uranium concentrations measured in zircon crystals isolated from the granite sample

$$t = A f \left[\frac{\rho_i}{\rho_o}\right] e^{\left[\frac{E}{kT}\right]} \tag{4}$$

where k is Boltzmann's constant, A is another constant that describes the time and temperature for fading, and ρ_o and ρ_i are the track densities before and after annealing, respectively. Here E is the activation energy for fading.

Experimental. The zircon grains were transferred into a platinum dish with a needle under a microscope (× 10). The dish was then placed in an electric furnace that had been heated and kept at an operating temperature. The temperature was measured with an alumel-chromel thermocouple. Heating was carried out at 650 °C for 480 and 960 minutes, at 700 °C for 30, 60, and 120 minutes, at 750 °C for 8 and 19 minutes, and at 800 °C for 1, 2, and 4 minutes. The zircon grains were etched as described above and the track density determined. The Arrhenius plot of the experimental data is shown in Figure 8. Here a solid circle indicates no fading of tracks; a semisolid circle, partial fading; and open circles, total fading. A straight line separating the partial fading and total fading regions was drawn. From its slope, see Equation (4), the activation energy for total fading was estimated to be 3.4 eV.

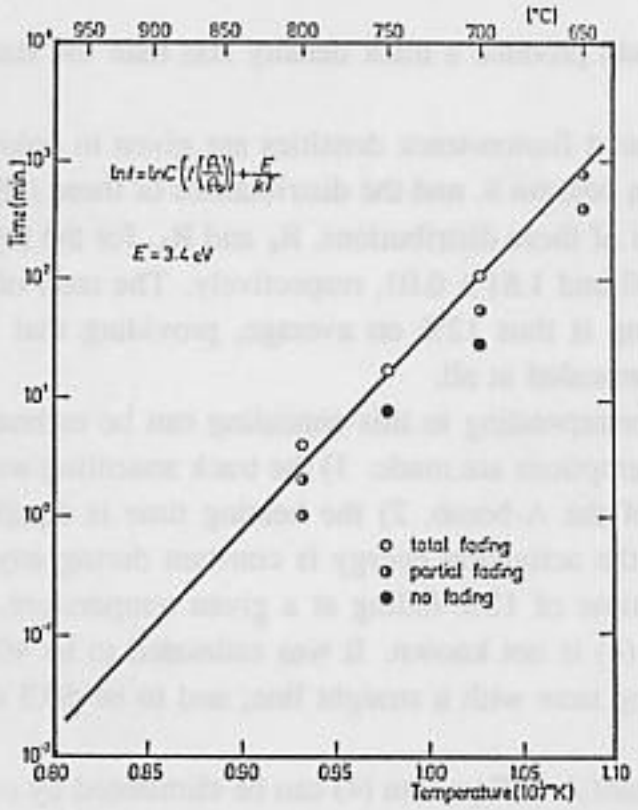

Figure 8. An Arrhenius plot showing the zircon annealing results. Open circles represent total fading, half-shaded circles partial fading, and solid circles no fading

Results and Discussion. The fission tracks induced by the neutrons of the A-bomb are neglible in number and, hence, need not be corrected for in these experiments. The fluence of thermal neutrons from the bomb at the sample site was about 6.4×10^{12} cm^{-2} as shown

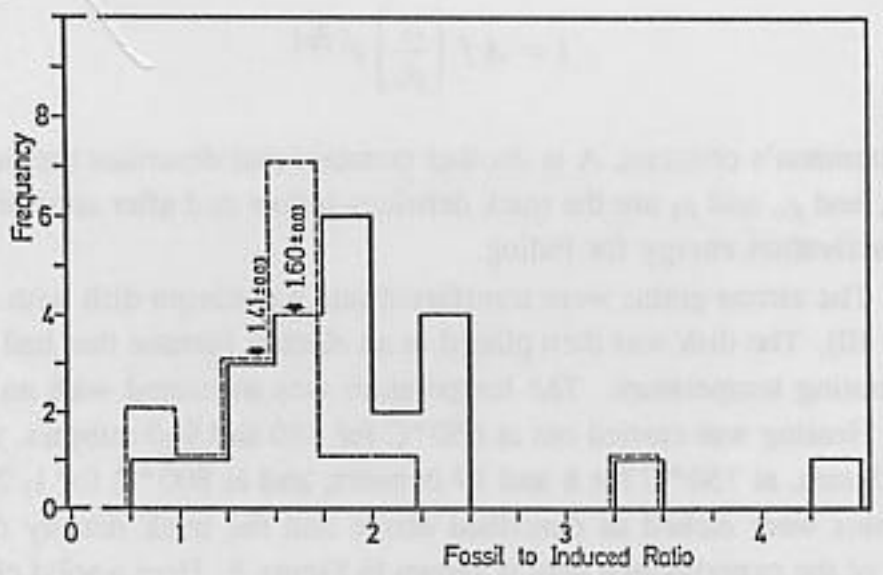

Figure 9. The frequency distribution of the ratio of fossil tracks to induced tracks. The solid line is for the top surface of the granite sample; the broken line is for the bottom surface

later. This fluence would produce a track density less than the statistical error in the track counting.

The fossil and induced fission-track densities are given in columns 4 and 7 of Table 1. Their ratios are given in column 8, and the distributions of these ratios is presented in Figure 9. The weighted means of these distributions, R_t and R_b, for the top (slab 1) and the bottom (slab 2), are 1.41 ± 0.03 and 1.61 ± 0.03, respectively. The ratio of R_t to R_b is 0.88 ± 0.03. The degree of annealing is thus 12% on average, providing that the tracks in the zircons from slab 2 were not annealed at all.

The temperature corresponding to this annealing can be estimated from Equation (4) if the following three assumptions are made: 1) the track annealing was caused only by heating due to the explosion of the A-bomb, 2) the heating time is equal to the brightening time of the fireball, and 3) the activation energy is constant during any reduction process. It is difficult to obtain the time of 12% fading at a given temperature, because the function of ρ_i and ρ_o in Equation (4) is not known. It was estimated to be 40.8 minutes by fitting the annealing versus heating time with a straight line; and to be 50.3 minutes by fitting it with a sine curve.

The unknown constant A in Equation (4) can be eliminated by combining the expressions for two different times as follows,

$$\ln \left[\frac{t_1}{t_2}\right] = \frac{E}{k}\left[\frac{1}{T_1} - \frac{1}{T_2}\right] \tag{5}$$

where t_1 is the heating time of the granite sample by the A-bomb and T_1 is the temperature produced by the bomb and 3.4 eV, 1.38×10^{16} erg K^{-1}, 1.4 s, and 973 K (700 °C) are substituted for E, k, t_2, and T_2, respectively. T_1 is calculated to be 950 °C when t_1 is 40.8 minutes and 919 °C when t_1 is 50.3 minutes. Figure 10 shows a curve of T_1 as a function

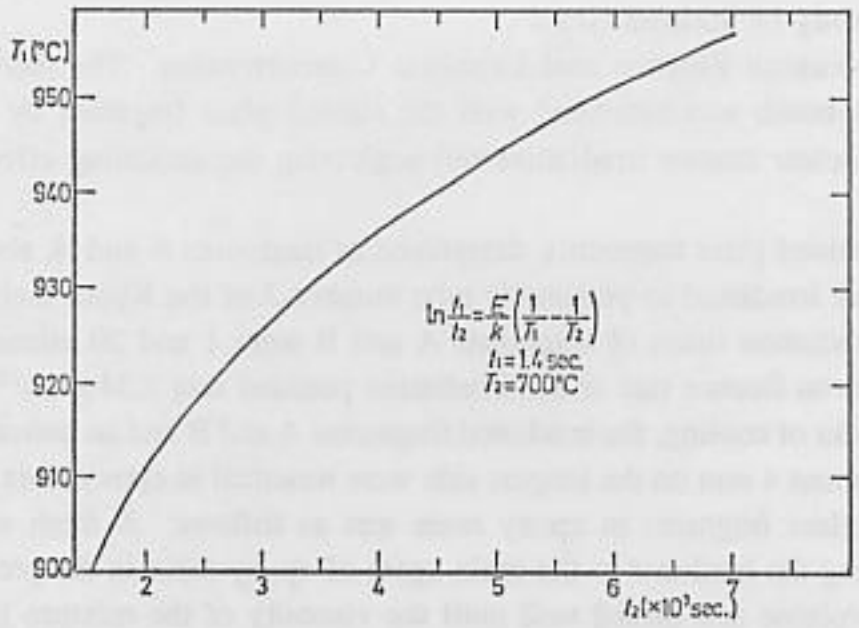

Figure 10. The relation of the surface temperature of the granite sample to the heating time for 12% track fading at 700 °C

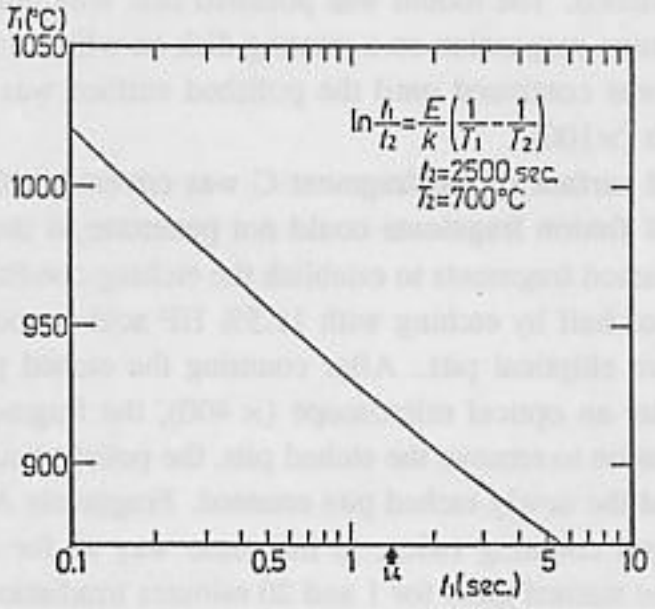

Figure 11. The relation of the surface temperature of the granite sample to the heating time produced by the A-bomb

of t_1 in the range of about 30 to 120 minutes for the latter.

It may be that the heating time, t_1, is longer than the brightening time of the fire ball. In this case, the temperature T_1 must be lower than 950 °C. Figure 11 shows T_1 as a function of t_1 for t_1 between 0.1 and 6 seconds and for t_2 = 2500 s, i.e., 42 minutes. T_1 decreases as t_1 increases but is about 900 °C at 1.4 seconds. This temperature is smaller than the temperature (573 °C at 1600 m from the hypocenter) derived from the flaking of rocks, when they are compared at the same ground range. It is believed the reason for the difference to be that the sample was too thick to obtain only zircons exposed to the highest temperature in the granite.

Fission-track Study of Stained Glass

Estimation of Neutron Fluence and Uranium Concentration. The thermal-neutron fluence from the A-bomb was estimated with the stained glass fragment by extrapolation of results from a nuclear reactor irradiation and neglecting the annealing effect in the stained glass.

Two small stained glass fragments, designated as fragments A and B, about 3 mm on the longest side, were irradiated in pneumatic tube number 3 of the Kyoto University Research Reactor. The irradiation times of fragments A and B were 1 and 20 minutes, respectively. The thermal-neutron fluence rate at the irradiation position was $2.34 \times 10^{13}\,\mathrm{cm^{-2}\,s^{-1}}$.

After two weeks of cooling, the irradiated fragments A and B and an unirradiated fragment (fragment C) of about 4 mm on the longest side were mounted in epoxy resin. The procedure for mounting a glass fragment in epoxy resin was as follows: A fresh epoxy resin was prepared by adding the hardener to the main agent of epoxy resin in the proportion eight to one hundred in volume and mixed well until the viscosity of the mixture became as small as possible. The mixture was centrifuged for one minute at 3000 rpm to eliminate foam. The sample to be mounted was set on a silicone oil-coated polyvinyl chloride plate with a polyethylene frame. The mixture was poured into the polyethylene frame. On standing overnight, the resin hardened. The mount was polished first with emery paper, second with an 0.3 μm diameter alumina suspension on a rotating disk on which a felt polishing cloth was pasted. The polishing was continued until the polished surface was a smooth plane under microscopic observation ($\times$100).

Half of the polished surface of the fragment C was covered with cellulose nitrate film thick enough that ^{252}Cf fission fragments could not penetrate to the glass. The other half was exposed to ^{252}Cf fission fragments to establish the etching conditions. Clear tracks were revealed on the irradiated half by etching with 11.5% HF acid at room temperature for one minute. The tracks were elliptical pits. After counting the etched pits on the nonexposed half of fragment C under an optical microscope ($\times$ 400), the fragment was polished again with the alumina suspension to remove the etched pits, the polished surface was etched again in the same manner, and the newly etched pits counted. Fragments A and B were subjected to polishing, etching, and counting twice, in the same way as for fragment C. The track densities obtained for the stained glass for 1 and 20 minutes irradiation were 3336 ± 204 and $67990 \pm 1206\,\mathrm{cm^{-2}}$, respectively.

The uranium content of this stained glass was determined by substituting the specific gravity of the stained glass and the measured track density into Equation (3). The specific gravity of this stained glass was measured to be 2.67 by drifting it in a density solution of bromoform and ethyl alcohol. The concentrations obtained for the 1- and 20-minute irradiations were 141 ± 3 and 143 ± 3 ppb, respectively. The two agree within the statistical errors. The weighted mean of the two is 143 ± 3 ppb.

The relationship between track density and neutron fluence is shown in Figure 12. The straight line of slope 1 was drawn to fit the two points for 1- and 20-minute irradiations and extrapolated to the track density of the nonexposed surface of fragment C to obtain the neutron fluence of the A-bomb. It is here assumed that the track density on the nonexposed surface of C is produced only by the neutrons from the A-bomb. The neutron fluence for the A-bomb was thus estimated to be less than $6.4 \times 10^{12}\,\mathrm{cm^{-2}}$.

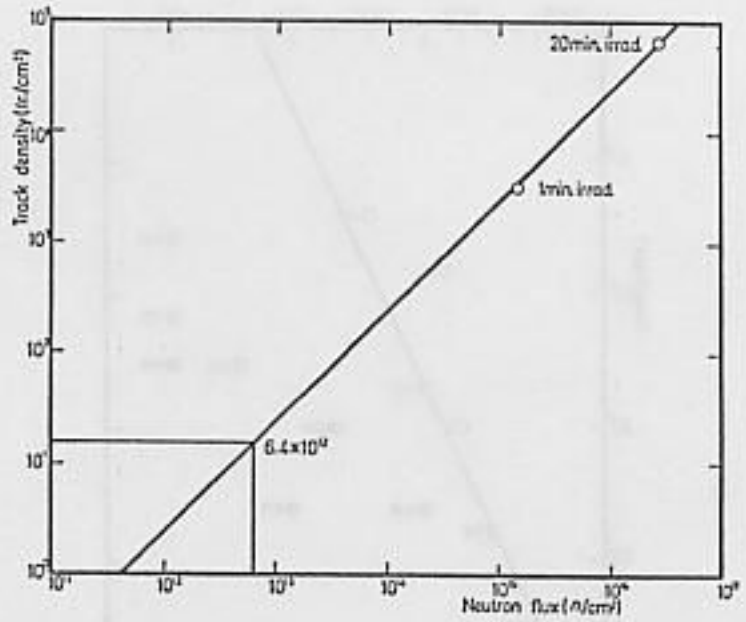

Figure 12. The relation of the track density to the thermal-neutron fluence. Extrapolation of the two measured points to the track density observed on an unirradiated sample of glass gives an upper limit to the fluence of $6.4 \times 10^{12}\ cm^{-2}$

Annealing Experiment. The annealing of the stained glass from the Uragami Cathedral was studied for different temperatures and durations. A large stained glass fragment D (about 1.5 cm on the longest side) was exposed to fission fragments from a ^{252}Cf source. The irradiated sample was heated to temperatures above 150 °C. Fragment C, mounted in epoxy resin, was used for the annealing at 100 °C; the epoxy resin is decomposed by heating over about 150 °C.

In the case of the small fragment C, the tracks were revealed by etching with the standard condition (11.5% HF, 15 °C, one minute). The diameters of the etched pits were measured with an ocular sliding-type micrometer under a microscope (× 400). Then the surface was polished to rub off the etched tracks and exposed to ^{252}Cf fission fragments again. Heating was carried out at 100 ± 2.5 °C for 3, 7, and 27 hours in an electric furnace. The diameters of the annealed pits etched in the standard manner were measured and the ratios in diameter of the annealed to the nonannealed pits were calculated. An average of about 40 observed pits was used for each stage of annealing.

In the case of fragment D, handling was easy because of its large size. The fragment was exposed to ^{252}Cf fission fragments. One side of the sample was etched and the size of the pits measured. Then the sample was placed in the electric furnace. After heating, the other side was etched in the same way and the pit diameters on the two sides were compared under the microscope (× 400). The heating was repeated at higher temperatures. The annealings were carried out at 150 °C for 3 hours, at 200 °C for 15 minutes, at 280 °C for 40 hours, at 400 °C for 15 minutes and 2 hours, at 450 °C for 15 hours, at 500 °C for 1 hour, and 600 °C for 10 minutes.

Figure 13 shows the Arrhenius plot of these data. The number beside a circle is the

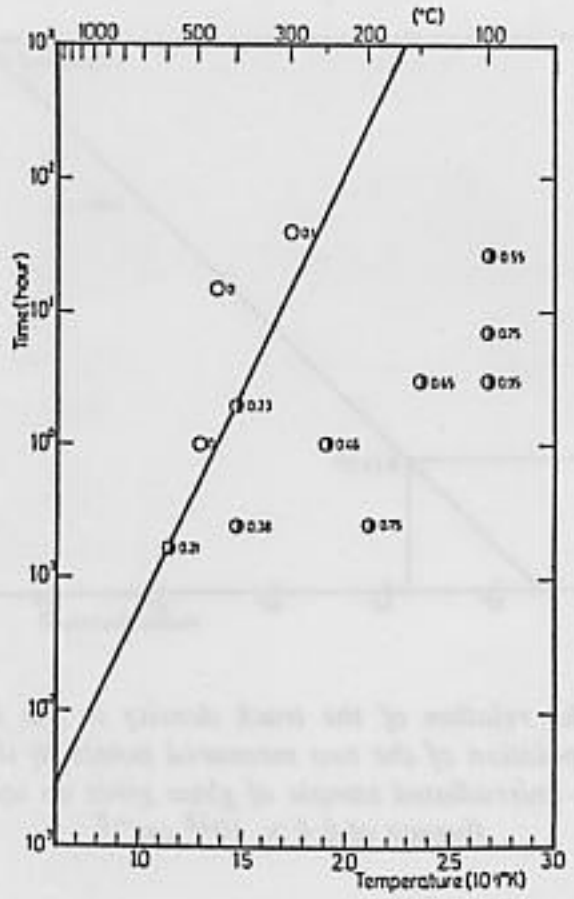

Figure 13. An Arrhenius plot showing the results of annealing the stained glass. The numbers beside the circles give the degree of annealing

degree of annealing. The activation energy for 20% fading in diameter was deduced to be 0.63 eV from the slope in Figure 13.

Results and Discussion. The thermal-neutron fluence of the A-bomb at the Uragami Cathedral was estimated to be less than $6.4 \times 10^{12}\,\mathrm{cm}^{-2}$. This value is consistent with the published value.

The distance from the detonation point to the Uragami Cathedral, where the stained glass was obtained, is 727 m; it is 655 m to Takatanis, where the granite sample was collected. These two distances are so close that the temperatures at the two points must have been close to one another. If a stained glass was heated to about 900 °C, it would melt, at least on the surface. But the stained glass sample used in this experiment had no indication of melting; so this sample fragment may not have faced the detonation point.

In the case of approximating the relation of the annealing of tracks versus heating time, the following equation fits the experimental data in Figure 13.

$$\frac{R_i}{R_o} = a\ log\{log(t)\} + b \qquad (6)$$

where R_o and R_i are the diameters of the etched pits before and after annealing, respectively, and a and b are constants. For example, the degree of track annealing of the four points at 100 °C (experimental: 0.95, 0.75, 0.55, and 0.22) are calculated to be 0.95, 0.749, 0.563, and 0.20.