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Dating of Ancient Ceramics by Thermoluminescence. III

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Thermoluminescent (TL) dating of ancient ceramics was undertaken by a TL technique named the large grain method. Because the sources of alpha particles were embedded mainly in the clay fabric and the range of the particle is less than 23 μm for quartz, the contribution to TL from alpha particles could be neglected by the treatment of hydrofluoric-acid etching on the surface layer of the sample grain. The accumulated radiation dose from beta and gamma rays originating from the pottery itself and the surrounding soil was measured by means of high sensitive thermoluminescent dosimeter $CaSO_4$;Tm. In the present investigation, the colourless grains of greater than 250 μm in diameter obtained from Thai and Japanese potteries were used as samples. The experimental results showed fairly good agreements with the known ages for Japanese potteries.

The authors reported previously the investigations of thermoluminescent (TL) dating of ancient pottery sherds using separated mineral fractions.[1,2] In this case, TL dating was performed on the implicit assumption that natural uranium and thorium radioactive impurities in the clay fabric principally contribute to produce the trapped electrons and the contributions from potassium and the other external sources were negligibly small. In the measurements, most of the ceramic samples yielded good agreements with their archaeological ages, but TL of several samples which were rich in alpha contents showed the low specific glows for their known ages. The disagreement in the measurement was considered to be attributed to the method by which the effective radioactive content was evaluated.

Thereafter the characteristics of induced TL due to alpha, beta and gamma radiations emitted from the radioactive natural isotopes of uranium, thorium, and potassium and the cosmic rays have been investigated by several workers, and essentially important evidences for improving the accuracy of TL dating have been revealed. One of them is that a quartz grain embedded in a clay matrix has the different TL sensitivities for alpha, beta and gamma radiations. According to Aitken *et al.*, alpha particles are typically only 15% as efficient as beta or gamma radiations in the creation of TL.[3] In addition, it was revealed that alpha particles from disintegrations occurring at the clay matrix penetrate into the grain only about 23 μm on the average, and, therefore, only the surface layer of the sample grain can be experienced by alpha dosage.[4]

Considering the characteristics of TL mentioned above, was developed another technique of TL dating named the large grain method, in which only the ceramic grains of larger than 100 μm in diameter are used as crystalline samples and the surface of the grain is etched by hydrofluoric acid. By the hydrofluoric-acid treatment a surface layer which suffered alpha dosage is removed from the outer regions of the grain. When the sample

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grains with diameters larger than the penetrating range of alpha particle are used, the TL effect of alpha radiation can be neglected by the acid treatment.

In the present investigation, TL dating of a series of pottery sherds from Ban-Chiang in Thailand and from Furu in Nara prefecture of Japan was performed by the large grain method. In the experiment, the samples used were treated by the following procedure: The inclusions with diameters greater than 250 μm were obtained by sieving the ceramic grains after a 0.2-mm thick surface zone of the pottery sherd was removed. These inclusions were treated by hydrofluoric-acid etching for five minutes so that the grain surface layer corresponding to the alpha particle penetration depth was etched out. The colourless minerals, which were separated from the grains by means of magnetic separator after the surface etching, were used as TL samples.

The details of experimental apparatus used and the procedure of the measurements of accumulated dose were the same as those described in the previous paper.[1] The radiation dose accumulated in crystals was evaluated as follows: Each grain sample was exposed to gamma rays from ^{60}Co source by a quantity of 1000 rads. TL of this irradiated sample was recorded as a glow-curve of the sample which absorbed a known amount of radiation dose. In order to get a natural dose which has been absorbed by the sample throughout the burial time, the glow-curve of the sample was compared with that of the irradiated sample.

The method of estimation of annual dose from the clay fabric and the surrounding soil was quite different from that used in the previous experiment,[1] because TL originated from only beta and gamma radiations was measured in the present investigation. For the estimation of beta dose per year, the high sensitive TL dosimeter $CaSO_4$; Tm developed by Matsushita Electric Ind. Co., Ltd. was used. The TL dosimeter grain of the same size as the sample grain was enclosed in a 40-μm thick polyethylene bag which shielded alpha particles. This polyethylene bag with the dosimeter was embedded in a clad of clay matrix obtained from the interesting pottery sherd. This dosimeter set for the estimation of annual beta dose was put for fifty days inside a lead box of 5-cm thick in order to be shielded from the radiations emitted from the external sources. The annual gamma dose from the soil and the cosmic rays was evaluated by using the TL dosimeter confined in a 2-mm thick copper capsule which shielded beta radiations from the surrounding soil. The copper capsule was burried for fifty days in a sphere of the soil obtained from the site

Table I. Dating Results of Pottery Sherds from Furu

Pottery Fragment	Accumulated Dose (Rad)	Annual Dose (Rad/y)	TL Age (y)	Archaeological Age (y)
Jomon 1	492	0.192	2560	B.C. 590
Jomon 2	487	0.213	2290	B.C. 310
Haji 1	342	0.214	1600	A.D. 380
Haji 2	358	0.212	1690	A.D. 280
Haji 3	351	0.214	1640	A.D. 330
Haji 4	375	0.230	1630	A.D. 340
Haji 5	387	0.231	1680	A.D. 300
Haji 6	350	0.225	1560	A.D. 420
Haji 7	369	0.217	1700	A.D. 270

Table II. Dating Results of Pottery Sherds from Ban-Chiang

Pottery Fragment	Burial Depth (cm)	Accumulated Dose (Rad)	Annual Dose (Rad/y)	TL Age (y)	Archaeological Age (y)
1-3	50 - 60	237	0.196	1210	A.D. 760
1-1	50 - 60	559	0.220	2540	B.C. 570
1-4	50 - 60	467	0.179	2610	B.C. 640
1-5	50 - 60	654	0.198	3300	B.C. 1330
1-2	50 - 60	653	0.181	3610	B.C. 1640
2-3	80 - 90	424	0.179	2370	B.C. 400
2-4	80 - 90	599	0.196	3060	B.C. 1090
2-5	80 - 90	781	0.194	4030	B.C. 2060
2-1	80 - 90	863	0.202	4270	B.C. 2300
2-2	80 - 90	873	0.182	4800	B.C. 2830
3-4	110-120	855	0.193	4430	B.C. 2460
3-1	110-120	951	0.197	4830	B.C. 2860
4-3	190-200	1111	0.207	5370	B.C. 3400
4-1	190-200	1010	0.177	5710	B.C. 3740
4-2	190-200	1176	0.188	6260	B.C. 4290
4-4	190-200	1266	0.198	6390	B.C. 4420

where the sample pottery sherd was excavated. The gamma dose suffered from the soil sphere of 30 cm and 7 cm in diameters was 95% and 50% of that which the pottery shered had been suffered in the site, respectively. Using the TL glow-curves of these $CaCO_4$; Tm dosimeter for beta and gamma radiations, the total annual dose which the sample grains suffered in the burial situation was evaluated. In the case that a sufficient amount of soil from the site could not be obtained, a rough estimation for the gamma dose from the surrounding soil per year was presumed from the alpha counting rate of the soil adhered to the surface of the sherd measured by means of a scintillation counter.

In Table I, are tabulated the results of the present experiments of TL dating for the pottery sherds from the site of Furu in Nara prefecture. These results by the large grain method showed fairly good agreements with the known ages.

Table II shows the results of the measurements for the pottery sherds from the site of Ban-Chiang in Thailand. Because the archaeological ages of the samples are not known, the experimental results cannot be compared with the known ages. But, taking into consideration the depth of each soil layer from which the pottery sherd was excavated, the results obtained are considered to show the proper ages.

The experimental error in the present measurements seems to be less than $\pm 10\%$ of each TL age. Among several sources of the experimental error in TL dating, the following two ones may have the extremely important roles. One of them exists in the way of annual dose evaluation. The principle of TL dating can be substantiated on the assumption that annual radiation dose received by the pottery sherd is constant through the burial time which has elapsed since the pottery was last heated. But the uncertainties, for instance, the variation of the concentration of ground water in the site soil, are there in the actual burial situation. Another source of error is due to the fact that reproducibility of the glow-curves of a pottery fragment is rather poor because of the complication of TL materials embedded in the clay fabric. In the case when a sufficient amount of the sample

grain can not be obtained, the ubiquity of high sensitive TL substances often causes a poor reproducibility of glow-curves.

It is noted here that an experiment is now in progress in order to improve the accuracy of the archaeological dating. The experiment is undertaken by such a way as the results obtained by TL measurements are compared with those by the other sicentific dating technique, *e.g.,* archaeolo-magnetic technique, neutron fission track method and so on.

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