CO5-1 Study of Earth and Planetary Matters by Thermoluminescence

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INTRODUCTION: Sears et al. have made a great progress in quantitative classification of type 3 ordinary chondrites, which have been classified into petrologic subtypes, ranging from 3.0 to 3.9, using the thermoluminescence (TL) sensitivity [1,2,3]. The minerals responsible for the TL have been interpreted to be mainly feld-spars that recrystallized in chondrule mesostases owing to metamorphism. However Matsunami et al. has found that silica phases of ALH-77214 (L3.4) which is type 3 ordinary chondrites with low TL sensitivity, were also responsible for the TL, using a TL spatial distribution readout system combined with microscope [4]. This year we confirmed that the silica phases were cristobalite by micro-raman spectroscopy.

TL of ALH-77214 (LL3.4): Figure 1 shows BSE image, induced TL images at 100° C and at 250° C, and CL image of the ALH-77214 (L3.4). There are one region in which TL is emitted at 100° C, and another region in which TL is emitted at 250° C.



Fig. 1. ALH-77214 (L3.4). Long dimension is 2.9 mm. The slice sample was irradiated by γ -rays and received a dose of 13.2kGy. The squares show the regions where local TL glow curves were analyzed. (a) BSE image; (b) induced TL image through Corning 4-96 filter at 100 °C and (c) at 250 °C. (d) CL image.

Figure 2 shows local glow curves of the ALH-77214 (L3.4). There are two types of glow curves corresponding to figure 1 (b) and (c). One had a peak at 100°C. Another had a peak at 250°C. Analysis of chemical composition revealed that the minerals responsible for these TL were

silica phase (I), anorthite-normative mesostasis (II), silica rich mesostasis (III), silica phase (IV), anorthite-normative mesostasis (V), and albite-normative mesostasis (VI). Silica phase had a peak at 250°C, and albite- and anorthite-normative mesostasis both had a low temperature peak at 100°C.



Fig. 2. Local glow curves of square region I-VI.

RAMAN: Induced TL glow curves of five terrestrial silica phases, hydrothermal quartz, volcanic quartz, tridymite, cristobalite, and silica glass, were utilized to identify the silica phase in ALH-77214. From similarities of the shape of glow curve, the silica phase was identified to be tentatively cristobalite [4]. Micro-Raman spectroscopy is useful for investigating crystal structure. This time Micro-Raman spectra confirmed that these silica phase and silica rich mesostasis were cristobalite as shown in Fig.3 [5].



Fig. 3. Raman spectra of region I, III, and IV.

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CO5-2

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Introduction: Many researches have been conducted using high energy electron (Max. 46 MeV) at KURRI-LINAC [1-4]. In addition a new irradiation system where chemical reaction occurs without inducing radioactivity has been investigated. We have reported here the energy reduction methods of accelerated electron for generating low-energy bremsstrahlung photon.

Generation of lower-energy electron: In this study microwave was introduced into only one of two acceleration tubes to generate lower energy electrons. Electrons were accelerated in the case that microwave was introduced into the first acceleration tube. In the contrary case, electrons were not accelerated because they were diffused before entering the second acceleration tube. The results described below were obtained in the former case.

Microwave was generated by klystron whose output was regulated by pulse forming network voltage. When the voltage was decreased from 27 kV (standard operation) to 20 kV (lower limit for stable operation), the peak energy of the electron beam and its current decreased to 6.8 MeV (Fig. 1) and half of standard, respectively.

The acceleration of electron requires energy supply from microwave. In acceleration tube the transit time of electron is quite shorter than the microwave filling time. The injection of electron from the electron gun with the optimum timing maximizes electron acceleration energy. On the contrary, electrons flying through the region unfilled microwave lose their own energy by emitting microwave. Electrons with higher energy fly faster and lose more energy, thereby injecting them faster than the optimum timing. As a result the higher energy component of electron beam decreases (Fig. 2).

Measurement of photon and neutron flux: Photon flux, $\varphi_g,$ and neutron flux, $\varphi_n,$ were evaluated from gold-foil activation method (Au-196 and Au-198, respectively). For the pulse forming network voltage of 22 kV and 20 kV, $\phi_g < 1.1 \times 10^5$ (detection limit) (cm⁻²s⁻¹) and $\phi_n = 9.2 \times 10^2$ (cm⁻²s⁻¹) and both fluxes were lower than detection limit, respectively, at an irradiation point where ϕ_g = 3.1×10^{11} and $\phi_n = 1.7 \times 10^7$ for standard operation. In comparison with results of the activation method and the response of a BF₃ filled proportional counter, neutron flux was assumed to 30 (cm⁻²s⁻¹) at 20 kV. Operation modes in which low-energy electrons were generated demonstrated irradiation with non-activating photon at KURRI-LINAC.



Fig. 1. The pulse forming network voltage regulates the peak energy of electron beam.



Fig. 2. Electron injection timing affects the energy spectra of electron beam; (A): optimum for highest energy, (B): 200 ns faster and (C) 400 ns faster.

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Luminescence Characteristics of Lake Sediment

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INTRODUCTION: Luminescence dating observes the natural accumulated radiation damage caused by radioisotopes such as U and Th as the form of glow after stimulation by heating or lightening. The luminescence is observed at various wavelength bands and their characteristics have been studied mainly on quartz and feldspar.

The emission color of luminescence can be recorded easily by using thermoluminescence color image(TLCI) analysis for samples artificially irradiated with gamma-rays [1]. When TLCI is applied to polymineral from lake sediments, the luminous mineral compositions of samples may reflect the regional characteristic and climate of the studied area. The validity of TLCI method for boring core samples from 3 lakes is investigated.

EXPERIMENTS: The boring core from Lake Baikal (in Russia), Lake Khuvsgul (in Mongolia) and Byeokgolje (in South Korea) is used for experiment. Each sample was etched step-by-step [2] with H_2O_2 (step B), HCl (step C), H_2SiF_6 (step D) and HF (step E), finally resulting in 5 types of aliquots including untreated sediment (step A). Samples were then iradiated by gamma-ray from Co-60 with dose rate of about 1.423kGy/h at Kyoto University Research Reactor Institute. The total gamma dose were 36 kGy given for 23.5 hours on the irradiation stage, which stand off 30cm from the radiation source.

Afterwards the photograph of TLCI was taken under a constant temperature of about 230°C. Colored pixels were picked up by the software programmed in this study and then converted to numerical values. Based on [3], color index were calculated from these numerical values and then were divided into five color ranges; blue (B, 450 <= 8 <= 495nm), green (G, 495 < G <= 565nm), yellow (Y, 565 < Y <= 580nm), red (R, 580 < R <= 700nm) and others (Gap).

RESULTS: Twelve photos of TLCI were taken for each step (A-E) of samples. The reproducibility was not so good, but the result of TLCI reflected the characteristic of the sampling areas. The average number of picked up pixels of each sample is shown in Fig. 1. Step B samples were most luminous for all lake sediments. The change in the TLCI through chemical treatment was obvious for Lake Khuvsgul, therefore the Lake Khuvsgul may caontain minerals delivered from many sources.





Fig. 1. The average number of picked up pixels of each sample.



Fig. 2. Color ratio of each sample.

studied lakes, but the Lake Baikal and Khuvsgul show similar color (red, yellow and green in addition to blue one) because these lakes are close. The blue luminescence probably came from quartz.

Even though there are some problems to be overcome, we found that TLCI may become efficient method to obtain the information of regional characteristics as same as the photonic instrumentation.

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Analysis of Rare Earth Elements in Ivory Samples

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INTRODUCTION: There are many Japanese traditional art works made by ivory; ornaments, accessories, parts of Japanese music instruments, and so on. Foreign trade of ivory has been inhibited from 1989. Since then, Japanese ivory arts and craft associations have been subject to restriction for importing ivories from African countries. For trying to keep the number of African elephants, it is important to find out the marker which show the origin of ivory. Takeuchi et al. analyzed several elements by NAA and stable isotopes for distinguish the origin of ivory [1], and they could not find good marker for origin. In thins work, we have separated rare earth elements (REEs) in ivory by using chelating cartridge, and analyzed REEs in the eluent by inductively coupled plasma spectroscopy-mass spectroscopy (ICP-MS).

EXPERIMENTS:

(1) Sample solution

Ivories from three different origin (the Republic of Zaire, Gabon, and the Republic of Congo) were chosen for analysis. Approximately 0.5 g of small tip of ivory was heated at 90°C for 2 hours with 2 ml of conc. HCl and 5 ml of distilled water. After cooling to room temperature, the solution was filtered by 0.45 μ m membrane filter, then the solution was diluted to 25 ml with ditilled water.

(2) Chelating cartridge

Chelating cartridge (3MTM EmporeTM Disk Cartridge Chelate, 10mm/6ml) was washed with 5 ml of 3 M HNO₃, 5 ml of distilled water, then 5 ml of 0.1 M Ammonium acetate before use.

(3) Separation of REEs from sample solutions

Five ml of sample solution was diluted to 50mlwith 0.1 M Ammonium acetate solution, and pH was adjusted around 5.5 by 6M ammonia solution. Thus obtained solution was loaded onto chelating cartridge. After washing with 5 ml of 0.5 M Ammonium acetate solution, REEs collected on the chelating cartridge was eluted by 2 ml of 3M HNO₃ and 1 ml of distilled water. The eluate was distilled to 10 ml with distilled water.

(4) Analysis of REEs by ICP-MS

Sample solutions, elution, and the eluate were distilled in 10, 100, and 1000 times by 0.1 M HNO_3 , and

elements were analyzed in them by ICP-MS. Elements analyzed were; Sc, Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, and Lu.

RESULTS: Since Sc and Gd were found in 0.1 M Ammonium acetate solution and 0.1 M HNO₃, these elements were eliminated from analysis. The results obtained for the eluate are shown in Table 1. In the ivory from the Republic of Congo 12 elements were con-In this work, small tips of ivory were used for tained. analysis. And there were no information of the position for the tip. Usually outside of the ivory has brown colour, and it is supposed that the concentrations of the elements related to soil are rather high comparing inside of ivory. Before investigating the marker for the origin, several positions of whole one ivory should be analyzed. For this purpose, the REEs separation method which were used in this work has proved to be effective one for further work.

Table 1 Levels of elements in ivory (unit: 10^{-9} g/g)

Elements	Origin		
	the Republic of Zaire	Gabon	the Repub- lic of Congo
Y	ND*	ND	110
La	20	6	430
Ce	50	ND	900
Pr	6	1	100
Nd	20	8	350
Sm	ND	ND	50
Eu	5	ND	20
Tb	ND	ND	4
Dy	2	ND	30
Но	ND	ND	4
Er	3	ND	20
Yb	ND	ND	1

*ND: not detected

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CO5-5 Investigation for Environmental Radioactivity in Relic Obtained from Salt-production Site, Aichi, Japan

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INTRODUCTION: The intake of salt is necessary and indispensable to human life. The production of salt from other than seawater is dependent on resources such as rock salt and salt lakes. About 180 million tons of salt are produced annually throughout the world, with the salt from seawater making up about 1/4 of the total. In Japan, which has no rock salt and salt lakes, the source of almost all salt is seawater.

The salt that is called "Moshio" was first made in an-It has been thought that Moshio had been cient Japan. made from seaweed and seawater. One of the famous salt-production sites where Moshio was manufactured is Matsusaki site, Aichi Prefecture, Japan. A large quantity of salt manufacturing earthenware was discovered at this site. In addition, a wooden tubing vessel was found that was thought to have been used to collect brine. A white substance often discovered in other salt-production sites was found stuck to both the salt manufacturing earthenware and the wooden tubing vessel at the Matsusaki site. Mori [1] analyzed the number of diatom valves of a sample of deposition that adhered to the wooden tubing vessel at the Matsusaki site. The most prevalent diatom was Cocconeis scutellum, a marine epiphytic diatom. The detection of marine epiphytic diatoms suggested the manufacture of salt at the Matsusaki site. Furthermore, as the same marine epiphytic diatoms in the earthenware had been discovered in the wooden tubing vessel, it was deemed that the wooden tub had contained liquid that originated from fractionated marine particles. However, there is no chemical evidence of salt manufacture at the Matsusaki site other than the results of the marine epiphytic diatom analysis.

In our study, environmental radioactivity in the substance that adhered to the wooden tubing vessel was measured to investigate the origin of the substance. Then, there was a search for evidence based on the seaweed or sea grass at the Matsusaki site. The judgment as to whether the substance that adhered to the wooden tubing vessel was of marine origin is discussed from a comparison of environmental radioactivity and chemical analysis. **EXPERIMENTS:** The excavation situation of the substance that adhered at a container is assumed to be a ring poor that straightens brine water in the Matsusaki ruins was reported by Tatematsu [2]. The ring pool divided the log into half, and it was buried in the sand on the coast. The thick calcareous adhered to the log (container that straightens brine water). A part of the adhesion thing was used for the analysis.

An attempt to determine the origin of the substance was made by examining the environmental radioactivity in the substance:

1) 87 Sr/ 86 Sr ratio, 2) activity ratio of 234 U/ 238 U, 3) δ 13 C, and 4) 228 Ra/ 226 Ra activity ratio.

RESULTS: The ⁸⁷Sr/⁸⁶Sr ratio in the origin of the adhesion substance obtained at the Matsusaki site was 0.70920 -0.70928. This value agreed well with the estimated value of seawater from shell, showing that the adhesion could originate from seawater and/or marine products. The activity ratio of $^{234}U/^{238}U$ showed the values of the seawater and/or marine products as well as the ⁸⁷Sr/⁸⁶Sr ratio, and this explanation based on environmental radioactivity supported that the origin of the adhesion substance was seawater and/or a marine product. Furthermore, the δ^{13} C in the adhesion substance was in a range that was smaller than the value of seawater and larger than that of a land plant. It was suggested that the adhesion substance could originate from not only seawater but also seaweed and/or sea grass. The ²²⁸Ra/²²⁶Ra activity ratio in the substance showed that the adhesion substance originated not only from seawater but also from seaweed and/or sea grass.

The detailed data for the origin of the relic obtained form Matsusaki site, Japan, will be published by Ohta et al. [3]

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