## CO5-1 Study of Earth and Planetary Matters by Thermoluminescence

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TL OF ORDINARY CHONDRITES: Induced TL (thermoluminescence), the response of a luminescent phosphor to a laboratory dose of radiation, reflects the mineralogy and structure of the phosphor, and provides valuable information on the metamorphic and thermal history of meteorites. Especially the sensitivity of the induced TL is used to determine petrologic subtype of unequilibrated ordinary chondrites [1]. Natural TL, the luminescence of a sample that has received no irradiation in the laboratory, reflects the thermal history of the meteorite in space and on Earth. Natural TL data thus provide insights into such topics as the orbits of meteoroids, the effects of shock heating, and the terrestrial history of meteorites [2]. Natural TL properties are usually applied to find paired fragments [3-5]. This time we measured induced and natural TL properties of sixteen Yamato unequilibrated ordinary chondrites (LL3: 4, L3: 10, H3: 2) from Japanese Antarctic meteorite collection. Sampling positions of these chondrites were measured by GPS.

**PRIMITIVE ORDINARY CHONDRITES:** Most of the chondrites had TL sensitivities over 0.1 (Dhajala=1), corresponding to petrologic subtype 3.5-3.9. One chondrite, Y980324 (L3) was revealed to be a primitive ordinary chondrite, petrologic subtype 3.2-3.3. It is particularly significant in understanding the nature of primitive material in the solar system.



Fig. 1. Induced Peak Temperature vs. TL Sensitivity to search primitive ordinary chondrites

**PAIRING:** Natural and induced TL properties were also applied to find paired fragments, and we found no potential paired fragments by TL, satisfying the criteria of 1) the natural TL peak height ratios, LT/HT, should be within 20%; 2) that ratios of raw natural TL signal to induced TL signal should be within 50%; 3) the TL peak temperatures should be within 20°C and peak widths within 10°C.

SILICIFIED WOOD: A unique sample of petrified wood with bright CL emission was obtained from the conglomerate in the Omori Formation (middle Miocene), Kurihara, Izumo City, southwest Japan. It has concentric rings of clear silicified layer filled up the cracks among black layer including carbonaceous materials. CL color images were recorded, and shown in Fig.2. It is divided into two areas: red CL and blue CL parts. Raman spectra and CL spectra at A (center red CL), B (fringe, red CL), C (outside of silicified wood, red CL), D (center, blue CL), and E (fringe, blue CL) points were obtained in this silicified wood. We recognized quartz signal at all red CL points (A, B, C). We also recognize D and G bands of amorphous carbon at A, B, D, and E inside of silicified wood as shown in Fig.3. The red CL has a peak at 650 nm, and the blue CL exhibits a peak at 450 nm. These CL spectral features are related to agate [6].



Fig. 2. CL color image of a silicified wood.



Fig. 3. Raman spectra of the silicified wood.

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採択課題番号 21001 熱ルミネッセンスによる地球惑星物質の研究

-般通常

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**INTRODUCTION:** Soils contaminated with heavy metals can be remediated by chelating reagents [1] in the way that metals are complexed with such reagents to remove those metals from soil surface. The remediation would be affected by radiation irradiation where the following reactions would be expected: the activation of contaminated soil surface by radiolytic radicals, the redox reaction of heavy metals, and the radiolysis of chelating reagents. In this study the elusion of <sup>153</sup>Gd from sand samples and the irradiation field of <sup>60</sup>Co gamma-ray have been investigated.

EXPERIMENTS: The elution of <sup>153</sup>Gd which was initially loaded on sand was determined in several eluant with changing concentration and pH. The eluant was fulvic organic matter which was extracted from fallen cherry leaves with dilute NaOH, treated with concentrated HCl to remove humic component and to protonate fulvic one, and then purified with dialysis membrane to remove cationic and anionic ions [2]. The radiolysis of organic matter was determined by measuring the difference of total organic carbon concentration before and after <sup>60</sup>Co gamma-ray irradiation at KURRI.

**RESULTS:** The distribution of <sup>153</sup>Gd without organic substances, which was obtained for the investigation of the effect of organic substances, shows that the remaining ratio is almost one in a natural pH region (Fig. 1), decreases with acidity, and is almost constant in the basic pH region. It is remarkable that the curves of the remaining ratio treated with the fulvic solution show a dip in the weakly acidic region, pH of around 5-6. This dip, shown in Fig. 1, can be held accountable for the competition between the site of complexation with fulvic acid and that of the adsorption on the sand surface.

As shown in Fig.2, natural organic substances were irradiated with <sup>60</sup>Co gamma-ray up to the dose of 12 kGy. The resulting decomposition to  $CO_2$  was not observed by TOC measurement. Much more irradiation dose is required to significantly decompose organic substances.

Irradiation with photon generated by KURRI-LINAC has been also planned, in which irradiation we require to measure dose and its rate depending on the mode of linac operation. The irradiation dose will be obtained by the cylindrical air-filled ion chamber calibrated at the <sup>60</sup>Co gamma-rays filed. The response of the chamber is shown in Fig. 3. The measurement of irradiation dose yielded the same response between vertical and horizontal alignment at further than 30 cm from the 60Co gamma-ray source.



Fig. 1. Elution property of <sup>153</sup>Gd from sand with fulvic acid extracted from fallen cherry leaves



Fig. 2. Effect of radiation to natural organic substances at different initial total organic carbon concentrations



Fig. 3. Variation of response of the air-filled ion chamber between vertical and horizontal alignment

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# CO5-3 Thermoluminescence Colour Image Analyses of Lake Sediments

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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 aim of this study is to reconstruct paleoclimate change using TLCI method.

**EXPERIMENTS:** The boring core from Lake Hovsgol is used for the experiment. Lake Hovsgol is the largest fresh water lake in Mongolia and is located in Baikal Rift Zone at an elevation of 1645m on the southern fringes of the East Siberian permafrost zone and connected to Lake Baikal through Egiin River, a tributary of the Selenga River. Because of its location in the middle of Asian continent and high altitude, the area is likely to be more sensitive to climate change, less affected by human activities. The sediments recovered from the lake have preserved important proxy records of past environmental changes.

The drilling operation was conducted in March 2004 at  $50^{\circ}$  57' 19" N 100° 21' 32" E in a water depth of 239.3 m by Hovsgol Drilling Project Members and HDP-04 core was obtained. Twenty-six samples were taken from the HDP-04 core for every 2 m starting from 2.46m to the depth of 56.47m.

Each sample was treated with  $H_2O_2$  to remove organic matter that emits no luminescence. Samples were then irradiated by gamma-ray from Co-60 with dose rate of about 1.467kGy/h at Kyoto University Research Reactor Institute. The total gamma dose was 36 kGy given for 24.5 hours on the irradiation stage, which stood off 20cm from the radiation source.

Afterwards the photograph of TLCI was taken under a constant temperature of about  $230^{\circ}$ C. Colored pixels were picked up by the software programmed in this study and then converted to numerical values. Based on [2], color index was calculated from these numerical values and then were divided into five color ranges; blue (B, 450 <= B <= 495nm), green (G, 495 < G <= 565nm), yellow

(Y, 565 < Y <= 580nm), red (R, 580 < R <= 700nm) and others (Gap).

**RESULTS:** Four photos of TLCI were taken for each sample. The average number of picked up pixels of each sample is shown in Fig. 1. The reproducibility was not so good, but the result of TLCI reflected the characteristic of the sampling depths. Number of colored pixels changes dramatically and color combination also changes a little, suggesting that TLCI analyses may provide important information on the source of sediment supply which can be the environmental indicator of the region. In the next step, this change must be correlated to other environmental proxies such as grain sizes or chemical compositions to understand what caused this variation in TLCI characteristics.



Fig. 1. The average number of picked up pixels for each color range. Sample number corresponds to depth (every 2 m) where sample is collected.

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採択課題番号 21004 熱ルミネセンスカラー画像解析を用いた湖沼堆積物特性の分析 一般通常 (金沢大・環日本海域セ)長谷部徳子(金沢大院・自然)伊藤一充、稲垣亜矢子、山田浩史

## CO5-4

## Photon Activation Analysis Using KUR-Linac

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**INTRODUCTION:** Neutron activation analysis (NAA) has high sensitivity and accuracy especially for siderophile elements represented by iridium [1]. However, sensitivity and accuracy for nickel in NAA using the research reactor which is dominated by thermal neutron are not so higher than those for iridium because nickel is determined by NAA using fast neutron induced reaction <sup>58</sup>Ni(n,p)<sup>58</sup>Co. In this work, the photon activation analysis (PAA) was adopted for the determination of trace amount of nickel. As a preliminary experiment, trace amount of nickel as an impurity in high-purity metal samples was determined by PAA to confirm that PAA is applicable to the determination of trace amount of nickel. The motivation of nickel-determination in this work is also described elsewhere in terms of cosmochemistry [2].

**EXPERIMENTAL:** The nickel contents in the high-purity iron sample (JSS003-5), iron oxide (JSS009-2), and cobalt foil were measured by PAA. As the reference standard samples, JB-1b, JG-1 and JMn-1 whose nickel contents are  $148\pm2$  mg  $\cdot$  kg<sup>-1</sup> [3],  $7.47\pm2.56$  mg  $\cdot$  kg<sup>-1</sup> [4, 5], and  $12632\pm828$  mg  $\cdot$  kg<sup>-1</sup> [6], respectively, were used. 30-100 mg of JSS003-5, JSS009-2, and the reference standards were sealed into high-purity aluminum foil and shaped into pellet of 9 mm diameter. The high-purity cobalt foil which was cut into square with about  $3\times3$  mm<sup>2</sup> was also sealed into high-purity aluminum foil.

Those samples, reference standards and gold foils (9 mm\u00f6 × 0.02 mm) as the fluence rate monitor of bremsstrahlung photons were enclosed in a quartz tube for irradiation with bremsstrahlung photons. The irradiation was carried out using the Electron Linear Accelerator at Research Reactor Institute, Kyoto University. The accelerator was operated at electron energy of 30 MeV with a mean current of around 0.08 mA for 33 hours. The quartz tube including irradiation samples was placed in the back of a platinum converter and cooled with running tap water. After the irradiation,  $\gamma$  rays of <sup>57</sup>Ni (1377 keV) produced by the photon induced reaction of  ${}^{58}$ Ni( $\gamma$ , n)  ${}^{57}$ Ni in each sample were measured by a low background Ge detector. The correction of bremsstrahlung photon fluence rate using gold foil monitors in the irradiated quartz tube was referred to Oura et al.(1999)[7].

**RESULTS and DISCUSSION:** The contents of nickel in the high-purity metal samples measured by PAA are listed in Table 1. The reference values of nickel contents in these samples are also listed.

| Table | 1. | Measured  | contents        | of Ni | in | individual | samples |
|-------|----|-----------|-----------------|-------|----|------------|---------|
|       |    | (in mg·kg | <sup>-1</sup> ) |       |    |            | -       |

| (  | m mg kg /           |             |                   |  |  |  |  |
|--|---------------------|-------------|-------------------|--|--|--|--|
| Sample   | Ni contents         | Ref. Std.   | Reference         |  |  |  |  |
|  |                     |             | value             |  |  |  |  |
| JSS003-5   | $0.542\!\pm\!0.072$ | JG-1, JMn-1 | $0.4 \pm 0.1^{a}$ |  |  |  |  |
| JSS009-2   | $0.352\!\pm\!0.058$ | JB-1b       | $< 2^{b}$         |  |  |  |  |
| Co-foil  | $478\!\pm\!19$      | JMn-1       | 95 °              |  |  |  |  |
| $a \mathbf{D}_{-} \mathbf{C} \mathbf{O} = \mathbf{D}_{-} \mathbf{C} \mathbf{O} \mathbf{C} \mathbf{D}_{-} \mathbf{C} \mathbf{I} \mathbf{O}$ |                     |             |                   |  |  |  |  |

<sup>a</sup>Ref. 8. <sup>b</sup>Ref. 9. <sup>c</sup>Ref. 10.

For JSS003-5 and JSS009-2, the PAA results are consistent with the reference values [8-10]. Since about 30 mg of JSS009-2 was subjected to PAA, it is possible to determine about 0.010  $\mu$ g of Ni within uncertainties of 20% using PAA in this work. It was confirmed that PAA used in this work made it possible to determine 1% of Ni at least in a few  $\mu$ g of cosmochemical samples like cosmic spherules [14]. For Co-foil, the reason that the PAA result has been about five times higher than the certified values is investigated at the present stage.

**CONCLUSION:** The amount of nickel in high-purity metal samples was determined by PAA. It was confirmed that PAA was able to determine about 0.010  $\mu$ g of nickel in the high-purity iron oxide sample within uncertainties of 20 %.

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採択課題番号 21025 京大炉(KUR)における光量子放射化分析法を用いた
共同通常
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## CO5-5 Cathodoluminescence Study of Nanodiamond Formation in Meteorites

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**INTRODUCTION:** Primitive chondritic meteorites contain up to ~1500 ppm of nanometer-sized diamonds. These nanodiamonds were recognized as presolar because of the isotopically anomalous noble gases that they involve. However, the origin of meteoric nanodiamonds is still very much an open issue. One early idea was that nanodiamonds were produced by high-pressure shock metamorphism of graphite through grain-grain collisions during the passage of intersteller shocks, whereas this process is not expected to be efficient because of not accompany of a large amount graphite. In recent years, other ideas have suggested that graphite grains might be induced to transform to diamond by intense particle irradiation, of which neutrons could be released from deep in the exploding star.

We conduct neutron implication experiments on graphite to clarify the formation mechanism of nanodiamonds by assuming neutron irradiation process at KURRI. Although no instrumental currently available can determine nanometer-sized diamonds, cathodoluminescence (CL) could detect characteristic emissions from nanodiamonds if they would aggregate up to several hundreds nanometers. The main purpose of this study is to establish a CL experimental data on the synthetic nanodiamond particles that could be used for the astrophysical interpretation of the planetary nebulae.

**EXPERIMENTS:** K2 (Ultradispersed Detonation Diamonds-UDD) nanodiamonds were mounted in the brass holder with non-luminescent epoxy resin. Their surfaces were finished using silicon colloidals with a grain size of 100 nm, and coated with amorphous carbon.

CL spectra were obtained using a scanning electron microscope-cathodoluminence (SEM-CL), which comprises the SEM (JEOL: JSM-5410) combined with a grating monochromator (Oxford: Mono CL2). The SEM-CL system was operated at 15 kV accelerating voltage and a probe current of 1.5 nA. CL spectra were recorded at the liquid nitrogen temperature (LNT) in the wavelength range of 350-800 nm with 1 nm spectral resolution and a dwell time of 1 second per step by photon counting. All CL spectra were corrected for total instrumental response.

**RESULTS & DISCUSSION:** Size dimensions of nanodiamonds were divided into the following groups: less than 3nm (Diamond: A/1 and A/2), between 3-7 nm (Diamond: B/1 and B/2), bigger than 7 nm (Diamond: C/1 and C/2), and a mixture of (Diamond: D/1 and D/2) all of them. A dominant broad peak centered at 535 nm appears at all of the UDD samples (Fig.1), which is re-

lated to the defect center at LNT [1]. It is important to note there is no systematic change in the peak intensities of the CL spectra as a function of the increasing grain sizes. A diamond-related 535 nm broad peak was also observed at NGC 7026 and NGC 7027 planetary nebulae, which are C-rich objects [2,3]. This indicates the presence of nanodiamond dust particles in the dust matter of these nebulae, which may be originated due to ejection of the outer parts of the Red Giants during planetary nebula formation.

CL spectra from these nanodiamonds show substantial similarities but also some slight differences, whereas the assignment of emission bands at around 535 nm is not always straightforward, but corresponding to dislocation defects associated with nitrogen atoms. Although it was not possible to define exactly the aggregation status of nitrogen atoms, the CL spectra suggest a low concentration of nitrogen in the nanodiamonds and give some indications for distinguishing impact diamonds from terrestrial diamonds. A nitrogen content typically less than 20 ppm can be sufficient to provide the luminescence of diamond [4]. In particular, this characteristic spectral features are uncommon in impact diamonds, implying an indicator of astronomical diamond. Therefore, CL analysis provides us useful information on defects or trace amount of impurities in nanodiamonds.



Fig. 1. CL spectral features of the UDD nanodiamonds (size: 2-20 nm) dominated by a broad band at 535 nm.

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- 隕石ナノダイアモンドの形成に関する研究
- 一般通常

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