

MIS028-01

Room:203

Time:May 25 10:45-11:00

## Luminescence and ESR for dating and earth/planetary science

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Electron spin resonance (ESR), thermoluminescence (TL), optically stimulated luminescence (OSL), and cathodoluminescence (CL) are methods based on solid state physics. They have quite long been used to study minerals, but not used to detect environmental change or large events quantitatively. ESR detects and quantify unpaired electrons, which are formed by radiation associated with lattice defects and with impurities. TL and OSL, based on the same principle, detects electrons formed by radiation and trapped at lattice defects and impurities, by observing the light which is emitted at the time of recombination. The major applications of these three methods have been dating. In particular, OSL dating method has become one of the most popular techniques in dating Quaternary sediments, like carbon-14 method. ESR dating method has succeeded in dating calcite, aragonite, and hydroxyapatite, however, still has many problems in dating quartz, which is one of the most popular minerals on the surface of the earth. CL is luminescence emitted when the mineral is irradiated by electrons, which is associated with lattice defects and impurities. CL has been used to check zoning which cannot be detected by SEM or by major element analysis when zircon is used for dating.

All these four methods detect small features in crystal such as lattice defects or impurities. New applications with these methods are now emerging such as identification of the source materials and detection and quantitative determination of shock events, in addition to conventional dating. In the presentation, the principle of the technique and the dating application, their current status, and these new application will be summarized.

Keywords: ESR, OSL, TL, CL, dating, luminescence

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## Use of ESR for study of geothermal activity

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ESR measurement is very useful in study of geothermal activity to date events, as well as to analyze thermal history. It can be applied other geological features, for example, to evaluate thermal influence at geological disposal sites.

### 1. Achievements

(1) ESR dating of quartz veins (Mizugaki, 2002)

ESR ages show that a younger hydrothermal event has been superimposed on older event products.

(2) ESR dating of quartz phenocrysts in hydrothermally altered rocks (Mizugaki, 2005)

In an active geothermal field, ESR ages of central silicified zone are younger, while those of peripheral zone are older. Both of them are much younger than original ages, then ESR ages show degrees of annealing by recent thermal activity. One sample shows younger age in peripheral zone, which located on a topographic lineament. It means that a fracture zone exists along the lineament, and hydrothermal fluid flow has been limited in it.

### 2. Future directions

Where very hot water springs out from a crack in quartz-rich rock, correlation between annealing and distance from the springing crack may be measured using ESR.

References: Mizugaki(2002) Advances in ESR Applications, 18, 181-186; Mizugaki(2005) Abst. 2005 Japan Earth and Planetary Sci. Joint Meeting, G018P-023

Keywords: ESR, electron spin resonance, geothermal activity, hydrothermal activity, thermal history

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## Relationship between OSL ages and water contents in the case of Japanese sediments

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There are a number of dating methods for Quaternary. Radiocarbon dating, for example, which can establish the time of formation of a wide range of organic materials, is predominant for late Quaternary ages, but for direct dating of the time of deposition of sediments the methods are relatively few (Aitken, 1998). The optically stimulated luminescence (OSL) dating (Huntley et al., 1985) is now used as an important tool for geoscientists concerned with sedimentary processes of the last million years or so, and also to some extent for archaeologists. It has been widely used for European, North American and Oceanian sediments. Though a number of OSL ages have been obtained from Indian and Chinese sediments in Asia also, OSL ages from Japanese sediments are not so many. In this study, the author reports several OSL ages of Japanese Quaternary sediments and compares them with sedimentary sequence and other estimated ages, to discuss the relationship between water contents and OSL ages.

Aitken, M. J. (1998): An introduction to optical dating. Oxford University Press.

Huntley, D. J., Godfrey-Smith, D. I. and Thewalt, M. L. (1985): Optical dating of sediments. *Nature*, 313, 105-107.

Keywords: OSL dates, water content, Quaternary, sediments, the Japanese Archipelago

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## Radiation dosimetric survey logger with a NaI scintillator

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NaI scintillators are widely used to investigate spatial dose rate in fields. Although, gamma-spectrometers are superior in spectral resolution, NaI scintillators have advantage in mobility in the field and should be meaningful devices for ESR/Luminescence dating. We have been successfully developed and tested as mobile dose survey loggers using NaI scintillators, including unfolding techniques of spectra, stabilization of environmental parameters for the device and calibration of spectra.

Keywords: NaI scintillator, Radiation, Dosimetry, logger

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## Thermoluminescence Study of Chondrites

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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 new twenty-four Yamato chondrites from Japanese Antarctic meteorite collection. Sampling positions of these chondrites were measured by GPS.

Most of the chondrites had TL sensitivities over 0.1 (Dhajala=1), corresponding to petrologic subtype 3.5-3.9. One chondrite, Y981221 (H3), 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.

Natural and induced TL properties were also applied to find paired fragments. We found eight potential paired fragments, i) Y981275 - Y981285(L3), ii) Y981175 - Y981186(H3), iii) Y981278 - Y981283(L3), iv) Y981275 - Y980593(L3), v) Y981283 - Y980453(L3), vi) Y981301 - Y981285(L3), vii) Y981140 - Y980660(H3), viii) Y980593 ? Y981285(L3) 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.

### References

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- [2] P. H. Benoit et al. 1991. *Icarus* 94: 311-325.
- [3] K. Ninagawa et al. 1998. *Antarctic Meteorite Re-search* 11:1-17.
- [4] K. Ninagawa et al. 2002. *Ant-arctic Meteorite Re-search* 15:114-121.
- [5] K. Ninagawa et al. 2005. *Antarctic Meteorite Re-search* 18:1-16.

Keywords: Thermoluminescence, Chondrite, Subtype, Pairing

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## Detection of radiation-induced defect center in plagioclase using cathodoluminescence spectroscopy

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Cathodoluminescence (CL) techniques have been used as an effective method to visualize radiation halos in quartz. Recent scientific interests focus on the CL of quartz as a geodosimetry indicator to clarify the formation process of CL halos, whereas no method has yet been developed for a quantitative estimation of the radiation dose on quartz from disintegration of natural radionuclides. Feldspar is one of the most important rock-forming minerals in the Earth's crust, of which CL depends on the genetic conditions such as temperature, pressure, and radiation dose during various geologic processes. Geoscientific applications of the CL of feldspar minerals, therefore, have been extensively explored for the study of mineral genesis interpretation. No investigation of radiation effects on CL of feldspar has been performed to date, although the visible halos can be easily found in the feldspar directly attached to radioactive minerals. In this study, CL of plagioclase implanted by He<sup>+</sup> ion has been conducted to quantitatively estimate the radiation dose from disintegration of natural radionuclides possibly to be applied to geodosimetry.

Single crystals of albite (Or<sub>1</sub>Ab<sub>99</sub>) from Minas Gerais, Brazil, oligoclase (Or<sub>2</sub>Ab<sub>82</sub>An<sub>16</sub>) from Inabu, Japan, andesine (Or<sub>1</sub>Ab<sub>53</sub>An<sub>46</sub>) from Bekily, Madagascar; and anorthite (Ab<sub>5</sub>An<sub>95</sub>) from Yoichi, Japan were selected for CL and Raman measurements. He<sup>+</sup> ion implantation (dose density:  $2.18 \times 10^{-6}$  to  $6.33 \times 10^{-4}$  C/cm<sup>2</sup>) on the samples was performed using a 3M-tandem ion accelerator at 4 MeV corresponding to the energy of alpha-particles from <sup>238</sup>U. A scanning electron microscopy-cathodoluminescence (SEM-CL) was used to obtain CL spectra of these plagioclase. Operating conditions were set at 15 kV (accelerating voltage) and 1.0 nA (beam current).

CL spectra of unimplanted plagioclase consist of emission bands at 350, 420, 580 and 700-740 nm which are assigned to Ce<sup>3+</sup> impurity, Ti<sup>4+</sup> impurity or Al-O<sup>-</sup>-Al defect, Mn<sup>2+</sup> impurity and Fe<sup>3+</sup> impurity centers, respectively. Similar emission bands were recognized in CL spectra of the implanted plagioclase. The intensities of these CL emissions decrease with an increase in radiation dose of He<sup>+</sup> implantation. Raman spectroscopy on the plagioclase reveals that Raman peak at 505 cm<sup>-1</sup> shows a decrease in the intensity and an increase in the full width at half maximum (FWHM) with increasing radiation dose of He<sup>+</sup> ion implantation. He<sup>+</sup> implantation destroys a linkage of T-O-T bond in the framework structure of the plagioclase, resulting in a reduction of CL emissions caused by impurity centers. CL spectra of implanted samples of albite and oligoclase exhibit characteristic red emissions at 700 nm, of which intensities increase with an increase in the radiation dose. It suggests that CL emissions at 700 nm are assigned to radiation-induced defect center formed by He<sup>+</sup> ion implantation. Deconvolution of CL spectra obtained from the implanted albite and oligoclase can successfully separate emission bands into six Gaussian components at 3.05, 2.82, 2.10, 1.86, 1.67, and 1.56 eV, where the component at 1.86 eV is undetectable in unimplanted samples. Integral intensity of the component at 1.86 eV linearly correlates with the radiation dose of He<sup>+</sup> ion implantation. The deconvoluted CL spectra of andesine and anorthite, however, have no component at 1.86 eV. These results suggest that the component at 1.86 eV might be assigned to O<sup>1- / 27</sup>Al x <sup>23</sup>Na center. The component intensity at 1.861 eV correlates with the radiation dose as a function of O<sup>1- / 27</sup>Al x <sup>23</sup>Na center, but does not depend on the concentration and distribution of the emission center, degree of Si-Al order and on the presence of microstructures or texture. CL spectral deconvolution, therefore, should be applied to quantitatively evaluate radiation dose of alpha particles from natural radionuclides on Na-rich feldspar as a geodosimetry indicator.

Keywords: cathodoluminescence, plagioclase, radiation-induced defect center, radiation halo, deconvolution

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## Coastal transportation process of sands presumed from OSL intensity of alkali feldspar around the Sinano river mouth

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In this research recent coastal sand grain transportation process around the Shinano river mouth is presumed using the bleaching percentage (BLP) estimated from OSL intensities of alkali feldspar grains contained in sand samples in addition to grain size distribution and the minerals ratio that had been used previous researches. The BLP distribution has the possibility of being useful for estimating recent coastal transportation process of sand grain which is not understood enough by the traditional method of grain size distribution and the mineral ratio. Generally, it is expected that bleached grain increase if transportation distance increase. Whereas sand grains eroded from beach probably obtained OSL intensity during its burial at beach.

As a result of research, it was presumed the sand grains which supplied from Ohkouzu Diversion Channel were transported to about 23 km northeasterly the Yotugouya-hama beach. The BLP distribution indicates the sand eroded from beach transportation process at the Nozumi beach, northern side of the Ohkouzu Diversion Channel mouth.

Keywords: transportation process of sands, OSL, alkali feldspar, grain size distribution, beach erosion, Ohkouzu Diversion Channel

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## Study of ESR signals and TLCIs from natural quartz for sediment provenance

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### Introduction

Quartz is one of the most abundant minerals on the surface of the earth. ESR dating of quartz, therefore, has been applied to a wide variety of samples, such as fault gouge, volcanic tephra, sediments, and flint artifacts [1]. Another direction of studies is to utilize the ESR signals as a marker of the samples like isotope analysis. The intensity of the E1' center in quartz is shown to be a useful parameter to investigate the provenance of Aeolian dust as well as of sediments [2]. As the intensity correlates with the age of the host granite in the range 10 Ma to 1 Ga, it is possible to distinguish quartz grains of Tertiary origin from those of Cretaceous. The Al, Ti-Li and E1' center signal intensity of natural quartz grains were irradiated with 2.5kGy gamma doses, as a means of estimating sediment provenance [3]. Samples were prepared various the host rocks and the sediments of Tertiary and Quaternary around Kizu River and Saho River. In this study, it will be discussed to estimate those sediments provenance. If this technique is established, it will be useful to elucidate the provenance of river basin and the encroachment of mountain.

### Sample preparation and experiments

Quartz grains for ESR measurements and TLCIs were extracted from the host rocks and the sediments. The samples were irradiated 2.5kGy gamma dose, then heated at 270 degree Celsius for 15minutes to ensure that the intensity of E1' center signals approaches to the maximum. ESR spectra recorded on an ESR spectrometer (JEOL TE-100, FA200; X-band), operating amplitude of field modulation was 0.1mT at 100kHz modulation frequency. The ESR signals of E1' center signal were observed using a microwave power of 0.01mW at room temperature. The ESR intensities of the Al and Ti-Li centers were measured with a microwave power of 5mW at 77K.

Temperature range of TLCIs measurement were 200-400 degree Celsius to provide a heating rate 30 degree Celsius/s.

### Result and discussion

The ESR signal intensities were found useful to distinguish the sediment provenance. The results in TLCIs were well correlated to the ESR results. The combination use of the ESR signal intensities and the characteristics of TLCIs were effective to investigate the fine-grained sediment provenance.

### References

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Keywords: Electron Spin Resonance, Thermoluminescence, quartz grain, sediment, provenance