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THERMOLUMINESCENCE OF CHONDRULES IN PRIMITIVE ORDINARY CHONDRITES, SEMARKONA AND BISHUNPUR

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Abstract: The spatial distribution of the induced thermoluminescence (TL) and TL glow curves of the primitive ordinary chondrites, Semarkona (LL3.0) and Bishunpur (LL3.1), were investigated over a wide range of wavelengths using a TL spatial distribution readout system. Although bulk samples of Semarkona and Bishunpur have very low TL sensitivity, individual chondrules show a wide variety of induced TL intensity and glow curve shape. Chondrules with anorthite-normative mesostases have especially high induced TL intensity, and their TL is produced at wavelengths >480 nm, compared with <480 nm for the sensitivity range of the usual TL measuring systems. Some of the metamorphism-dependent TL sensitivity of type 3 ordinary chondrite therefore results in changes in the spectrum of the light produced.

1. Introduction

The induced thermoluminescence of ordinary chondrites provided unique insights into the metamorphic history of meteorites. For instance, the TL sensitivity of ordinary chondrites increases by a factor of 10^5 with metamorphism due to the crystallization of feldspar, and TL sensitivity is an effective means of classifying the type 3 ordinary chondrites into petrographic subtypes, 3.0-3.9 (SEARS *et al.*, 1980; SEARS, 1988; SEARS *et al.*, 1990). The primitive ordinary chondrites, which have low petrographic subtypes, have the lowest TL sensitivities. In the case of ALH-77214 (L3.4) with low petrographic grade, a large dose of irradiation, 13.2 kGy, made it possible to produce TL images. It was found that (1) a silica phase was responsible for the TL, as well as feldspar (MATSUNAMI *et al.*, 1992), (2) mesostases of normative-anorthite composition produced low TL peak temperatures and narrow peak widths, while a mesostasis of normative-albite composition showed high peak temperature and wide width (NINAGAWA et al., 1991). These peak shape characteristics are known to be associated with differences in thermal history. Primitive ordinary chondrites such as Semarkona and Bishunpur have TL sensitivity lower than ALH-77214 (L3.4) by one order of magnitude, making spatial measurements much more difficult. However, through such measurement we hoped to determine whether the low TL sensitivity of the meteorites was due to the TL of a few chondrules while the majority have little or none. We hope also to determine if the low TL sensitivity of feldspathic mesostases made it possible to detect TL from other minerals, and what kinds of chondrules or minerals were responsible for the TL in the primitive chondrites? Ultimately we hope that data on all these points will help clarify the origin and thermal history of these meteorites and their chondrules.

2. Samples and Equipment

The primitive ordinary chondrites, Semarkona (USMN 1805, 50.47 mg) and Bishunpur (BM 80340, 66.62 mg), were examined. Small chips were cut into slices by a wire saw and polished for TL image measurements and EPMA analysis. The TL spatial distribution readout system for measuring TL images was described previously (NINAGAWA *et al.*, 1990). The video image processor in this system was changed to a highly parallel array processor system (KOHATA, 1991). This highly parallel array processor system makes the two-dimensional photon counting possible without frame loss in analysis. The slices were exposed to γ -rays from ⁶⁰Co and received a dose of 13.2 kGy, similar to ALH-77214 (L3.4) (NINAGAWA *et al.*, 1991). The TL images were measured immediately after irradiation at the heating rate 0.25°C/s with a Corning 4–96 filter. The chemical compositions of mesostases responsible for the TL were analyzed by an electron probe X-ray microanalyzer [EPMA], JCXA-733 (JEOL LTD), operated at 15 kV accelerating voltage and 12 nA beam absorption on PCD (Probe Current Detector).

3. Results

Figures 1a and b show a backscattered electron image (BEI) of a sample of Semarkona (LL3.0) and an induced TL image at the temperature interval 40–440°C, respectively. A certain chondrule, squared in Fig. 1b (region I), shows intense TL. The local TL glow curve of region I including this chondrule is shown as a solid line in Fig. 1c. This glow curve has a $\sim 300^{\circ}$ C peak, which is different from those of mesostases in ordinary chondrites with high petrographic grades. It is the first time that the glow curve of the above type has been measured in ordinary chondrites. The dashed line will be discussed below. This chondrule is a porphyritic olivine chondrule (Fo_{99.5-99.7}) of type IA according to SCOTT and TAYLOR (1983) and group A1 of SEARS *et al.* (1992), and the mesostasis of this chondrule is responsible for the TL. The chemical composition of the mesostasis was determined by the EPMA and the result is shown in Table 1 with the C.I.P.W. norm. The mesostasis is characterized by high normative-anorthite composition.



Fig. 1. BEI, induced TL image, and glow curves of Semarkona (LL3.0), including a type IA chondrule. Long dimension is 2.9 mm. The position of high TL intensity is put by deep black points. The square shows the region where local TL glow curves were analyzed and chemical compositions of mesostases were analyzed by EPMA.
(a) BEI.

- (b) Induced TL image through Corning 4-96 filter at the temperature interval 40-440°C.
- (c) Local glow curves through Corning 4–96 filter (line) and through Corning 4–69 & 7–59 filters (dashed line).

where molar ratio of normative minerals, ab/(ab+an/2), is less than 0.7. This TL image shows that the TL intensity of the marginal portion is stronger than that of the central one.

Figures 2a and b show a BEI of another slice sample of Semarkona (LL3.0) and its induced TL image at the temperature interval 40–440°C. Some chondrules also show the TL. The local TL glow curve of square region I in Fig. 2b is shown as a solid line in Fig. 2c and has two peaks of about 140°C and 260°C. This glow curve is a new type, which is also different from those of mesostases in ordinary chondrites with high petrographic grades. This chondrule is classified as a porphyritic olivine-pyroxene chondrule (Fo_{99.0}, Wo_{2.7}Fs_{1.0}En_{96.3}) and as type IB of Scott and

TAYLOR (1983) and group A1 of SEARS *et al.* (1992). The mesostasis of this chondrule is also characterized by normative-anorthite composition (Table 1). The marginal portion of square region II shows also the TL. The local TL glow curve shown as a solid line in Fig. 2d has a peak of ~160°C. This chondrule is revealed to be classified as a porphyritic olivine chondrule (Fo_{65.0-81.5}) and as type II of SCOTT and TAYLOR (1983) and group B1 of SEARS *et al.* (1992). The chemical composition of the mesostasis is shown in Table 1 with the C.I.P.W. norm. The mesostasis is characterized by high normative-albite mesostasis" is used for the mesostasis where molar ratio of normative minerals, ab/(ab + an/2), is more than 0.7. The



Fig. 2. BEI, induced TL image, and glow curves of Semarkona (LL3.0). Long dimension is 2.9 mm. The position of high TL intensity is put by deep black points. The squares show the regions were local TL glow curves were analyzed and chemical compositions of mesostases were analyzed by EPMA.
(a) BEI.

- (b) Induced TL image through Corning 4-96 filter at the temperature interval 40-440°C.
- (c) Local glow curves of square region I through Corning 4–96 filter (line) and through Corning 4–69 & 7–59 filters (dashed line).
- (d) Local glow curves of square region II through Corning 4–96 filter (line) and through Corning 4–69 & 7–59 filters (dashed line).

glow curve of normative-albite mesostasis is the same as those of mesostases in ordinary chondrites with high petrographic grades.

Figures 3a and b show a BEI of a slice sample of Bishunpur (LL3.1) and its induced TL image at the temperature interval 40–440°C. The local glow curve for square region I as shown in Fig. 3c has a 110°C peak and the TL becomes stronger in a higher temperature region. The texture of this chondrule is a barred olivine chondrule (Fo_{98,6-98,7}) and this chondrule has normative-anorthite mesostasis (Table 1). It is classified as group A1 in the scheme of SEARS *et al.* (1992). The glow curve of square region II in Fig. 3b has a 170°C peak. This chondrule is a porphyritic olivine chondrule (Fo_{77,1-87,4}) of type II or group A5. The glow curve



Fig. 3. BEI, induced TL image, and glow curves of Bishunpur (LL3.1). Long dimension is 2.9 mm. The position of high TL intensity is put by deep black points. The squares show the regions where local TL glow curves were analyzed and chemical compositions of mesostases were analyzed by EPMA.
(a) BEI.

- (b) Induced TL image through Corning 4-96 filter at the temperature interval 40-440°C.
- (c) Local glow curves of square region I through Corning 4–96 filter (line) and through Corning 4–69 & 7–59 filters (dashed line).
- (d) Local glow curves of square region II through Corning 4–96 filter (line) and through Corning 4–69 & 7–59 filters (dashed line).

of normative-albite mesostasis in Bishunpur (Table 1) is also the same as those of mesostases in ordinary chondrites with high petrographic grades.

The TL spectra of the Semarkona (LL3.0) including the chondrule in Fig. 1a were measured by a time-resolving spectroscopy system (NINAGAWA *et al.*, 1986) and are shown in Fig. 4. These spectra show that TL has a spectral peak at \sim 570 nm

| Meteorite | Semarkona | Semarkona | Semarkona | Bishunpur | Bishunpur |
|--------------------------------|-------------|-------------|--------------|-------------|--------------|
| Texture ^{#1} | PO | POP | PO | BO | PO |
| Type ^{#2} | type IA | type IB | type II | | type II |
| #3 | A1 | A1 | B1 | A1 | A5 |
| Region | I in Fig. 1 | I in Fig. 2 | II in Fig. 2 | I in Fig. 3 | II in Fig. 3 |
| N ^{#4} | 12 | 2 | 11 | 3 | 2 |
| SiO ₂ | 53.0 | 50.2 | 69.8 | 56.8 | 63.5 |
| TiO ₂ | 0.92 | 0.16 | 0.15 | 0.68 | 0.51 |
| Al_2O_3 | 20.8 | 27.2 | 18.9 | 16.9 | 14.9 |
| Cr ₂ O ₃ | 0.40 | 0.13 | #5 | 0.65 | 0.15 |
| FeO | 0.32 | 0.43 | 1.35 | 0.90 | 4.07 |
| MnO | 0.14 | 0.07 | | 0.31 | 0.12 |
| MgO | 6.65 | 4.82 | 0.21 | 8.53 | 2.05 |
| CaO | 16.6 | 16.2 | 1.78 | 11.8 | 3.02 |
| Na ₂ O | 0.93 | 0.50 | 6.60 | 3.13 | 10.0 |
| K ₂ O | — | | 0.40 | 0.16 | 0.09 |
| NiO | 0.07 | | | 0.06 | |
| S ₂ O | _ | — | | 0.08 | |
| P_2O_5 | 0.09 | 0.18 | 0.76 | 0.05 | 1.64 |
| Q | 8.41 | 6.87 | 24.71 | 5.51 | _ |
| С | | | 4.37 | | |
| or | 0.24 | 0.12 | 2.36 | 0.95 | 0.53 |
| ab | 7.87 | 4.23 | 55.85 | 26.49 | 72.91 |
| an | 52.46 | 71.91 | 8.83 | 31.59 | — |
| ne | | | _ | | 1.75 |
| di-wo | 11.69 | 3.53 | _ | 11.25 | 6.26 |
| en | 10.11 | 2.94 | _ | 9.50 | 2.32 |
| fs | | 0.15 | | 0.29 | 4.05 |
| hy-en | 6.46 | 9.07 | 0.52 | 11.74 | _ |
| fs | — | 0.45 | 2.29 | 0.36 | |
| cm | 0.59 | 0.19 | 0.03 | 0.96 | 0.22 |
| il | 0.72 | 0.30 | 0.29 | 1.29 | 0.97 |
| $ab/(ab+an/2)^{\#6}$ | 0.14 | 0.06 | 0.87 | 0.47 | 1.00 |

 Table 1.
 Average chemical compositions and the C.I.P.W. norms of mesostases of five chondrules with intense TL in the Semarkona (LL3.0) and Bishunpur (LL3.1) chondrites, obtained by EPMA. Recalculated to total 100 wt%.

#1: PO (Porphyritic Olivine chondrule).

POP (Porphyritic Olivine-Pyroxene chondrule).

BO (Barred Olivine chondrule).

#2: according to Scott and TAYLOR (1983).

#3: according to SEARS et al. (1992).

#4: number of averaged analyses.

#5: not detected.

#6: molar ratio of normative minerals.

in the temperature interval between 200–350°C. These spectra agree with those of some plagioclase feldspars in which Mn^{2+} ions act as luminescence centers (HUNTLEY *et al.*, 1988) and correspond to yellow cathodoluminescence of the A1 and A2 chondrule mesostases (SEARS *et al.*, 1992).

Usual TL measuring systems have used Corning 4-69 & 7-59 filters (SEARS



Fig. 5. Spectral response of TL spatial distribution readout system. Quantum efficiency of a photon imaging head with a bialkali photocathode, Hamamatsu Photonics Co. C2166–01 (line I) and detection efficiencies of a TL spatial distribution readout system with Corning 4–96 filter (line II) and with Corning 4–69 & 7–59 filters (line III). Photons of the wavelengths shorter than 400 nm cunnot transmit the lens of microscope (dashed line).

et al., 1990) or Corning 5-60 & 7-98 filters (MELCHER, 1981) to suppress blackbody radiation emitted at high temperature. These filter combinations limit the detection wavelength from 320 nm to 480 nm or from 360 nm to 480 nm, respectively. The "low TL sensitivity" of the primitive ordinary chondrites is in part due to limiting the detection wavelengths to ultraviolet-blue region. The Corning 4-96 filter, which is used in the present apparatus, transmits 360 nm to 580 nm, so that the TL from the group A1 and A2 chondrules can be detected in our TL system. The quantum efficiency of a photon imaging head with a bialkali photocathode, Hamamatsu Photonics Co. C2166-01 and detection efficiencies of the TL spatial distribution readout system with Corning 4-96 filter or with Corning 4-69 & 7-59 filters are shown in Fig. 5.

The TL data for the present chondrules, as seen through Corning 4–69 & 7–59 filters, are shown by the dashed lines in Figs. 1c, 2c, 2d, 3c, and 3d. For the group A1 chondrules (Figs. 1c and 2c) and the high temperature peak in the barred olivine chondrule (Fig. 3c) the TL of the dashed line is much weaker than that of the solid line, while for the group B1 and A5 chondrules (Figs. 2d and 3d) the TL intensity expressed by the dashed line is the same order of magnitude as that of the solid line. Thus, in the primitive ordinary chondrites the TL of the chondrules with albite-normative mesostases is produced mainly in the wavelengths shorter than 480 nm, while the TL in chondrules with anorthite-normative mesostases is produced mainly at wavelength >480 nm.

4. Discussion

Although Semarkona and Bishunpur would have the extremely low TL sensitivity, we could measure intense TL images for certain chondrules at a dose of 13.2 kGy. The anorthite-normative mesostases in the group A1 chondrules have high induced TL intensities and a variety of glow curve shapes. Measurements with different filters show that the light is produced at the wavelengths longer than 480 nm. In the case of the particularly bright chondrule in Semarkona (LL3.0), the TL spectra were measured directly by a time-resolving spectroscopy system and the TL spectra had a dominant peak at about 570 nm.

On the other hand, chondrules in Semarkona and Bishunpur with albitenormative mesostases have the TL with about 160°C peak temperature and measurements with different filters show that the light is produced at wavelengths shorter than 480 nm.

The above results mean that the TL detection wavelengths are important for the very low petrologic type meteorites. The TL sensitivity measured by usual TL measuring systems has been limited to the range of wavelengths shorter than 480 nm, detecting not all the TL emissions but those from the albite-normative mesostases of the B1 and A5 (or type II) chondrules and some of anorthite-normative mesostases. The TL emission in the wavelengths longer than 480 nm may be as useful in classifying the meteorites as that at the wavelengths shorter than 480 nm.

The TL sensitivity is the reflection of the crystallization of mesostasis from glass to feldspar (SEARS, 1988). The high TL sensitivity of normative-anorthite

mesostases in these primitive chondrites means that they are not glassy and have probably crystallized. The chondrules, which include these crystallized mesostases, were presumably made by slow cooling in their formation. The variety of glow curves in the normative-anorthite mesostases is the reflection of chondrule formation processes and the content of minor element like manganese because their textures are various (type IA chondrule, porphyritic olivine pyroxene chondrule and barred olivine chondrule) and Mn^{2+} ions would act as emission centers.

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