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Development of radiation-damage halos in low-quartz: cathodoluminescence measurement after He^+ ion implantation

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Summary

In cathodoluminescence (CL) images of synthetic low-quartz samples after He^+ implantation at 4 MeV with a dose density over $1.14 \times 10^{-4}\,C\,cm^{-2}$, bright CL halos of about 14 μm in width from the implantation surface are recognized. These widths are consistent with the theoretical range. This confirms experimentally that the CL halos in low-quartz found in geological samples are formed by alpha radiation. It also shows that CL colour continuously changes with dose density, demonstrating that it is possible to use the CL halo as a new dosimeter that is useful for dating and analysis of radionuclide migration in natural geological media.

Introduction

Under cathodoluminescence (CL), halos of 30–40 μm in width are often found in low-quartz adjacent to radionuclide-bearing minerals, similar to pleochroic halos in biotite visible under transmitted light. First found by *Smith* and *Stenstrom* (1965), these CL halos are believed to be formed by radiation-damage, because the widths are identical with the calculated traversed distances (range) of alpha particles emitted from natural radionuclides in radionuclide-bearing minerals. Recently *Götze* et al. (2001) found that CL halos in low-quartz from the Witwatersrand conglomerate have a 650 nm emission which is associated with a non-bridging oxygen bond. These researchers considered that the emission is formed by bond breaking due to alpha particles. However, until now the formation of CL halos has not been confirmed experimentally.

The contrast between a halo and the host low-quartz is generally known to increase with age and concentration of radionuclides in adjacent radionuclide-bearing minerals (*Owen*, 1988; *Closel* et al., 1992; *Komuro* et al., 1995). *Owen* (1988) suggested that the CL halo might be a new dating tool if the contrast can be quantified. However, this is difficult as can be appreciated from the fact that pleochroic halos in biotite have not been used as a dating tool. Recently, a new dosimetric tool for determining ancient radionuclide migration has been required by natural analogue studies related to radioactive waste disposal. *Meunier* et al. (1990) and *Komuro* et al. (1995) found fossil radiation-damage halos remaining after radionuclide removal in some uranium deposits. It is expected that CL halos could be a new dosimetric tool that register ancient accumulative dose if a relationship between accumulative dose and a property of a halo such as contrast can be identified. However, the relationship has not been identified so far. In order to investigate the development of CL halos by alpha radiation experimentally, particularly the relationship between dose and CL halo development, CL colour measurements were carried out on synthetic low-quartz samples after He^+ ion implantation.

Experimental

Synthetic low-quartz samples cut into thin chips of about 1 mm in thickness were used in this experiment. The low-quartz samples contained Al – 130 ppm, Li – 25 ppm, Na – 14 ppm, Fe – 5.9 ppm, B – 0.14 ppm, and Ge – 0.001 ppm.

He^+ implantation in the synthetic low-quartz samples was performed with a 3M-tandem ion accelerator in the Takasaki Research Center of Japan Atomic Energy Research Institute. Taking the degree of activation of the samples into consideration, the acceleration energy of He^+ was set at 4 MeV, corresponding roughly to the energy of alpha particles emitted from the decay of ^{238}U (4.18 MeV). The homogeneous He^+ ion beam was irradiated perpendicularly on the surface of the quartz chip. The implantation was made under seven sets of conditions with different dose densities shown in Table 1.

CL measurements on polished sections of low-quartz samples cut vertically for irradiation were carried out by a Luminoscope® ELM-3R equipped with an Olympus® BX-60 microscope furnished in a darkroom. The brightest and most

Table 1. *Conditions for He^+ implantation experiments*

Sample No.	Dose (C)	Average current (μA)	Bombardment area (cm^2)	Dose density ($C\,cm^{-2}$)
3α-K	1.71×10^{-2}	1.25	3.75	4.55×10^{-3}
3α-Q	4.26×10^{-3}	1.25	3.75	1.14×10^{-3}
3α-C	1.13×10^{-3}	1.1	2.25	5.02×10^{-4}
3α-P	1.28×10^{-3}	1.25	3.75	3.41×10^{-4}
3α-O	8.52×10^{-4}	1.25	3.75	2.27×10^{-4}
3α-M	5.68×10^{-4}	1.25	3.75	1.51×10^{-4}
3α-N	4.26×10^{-4}	1.25	3.75	1.14×10^{-4}

suitable beam condition that could not cause distinct beam damage was adopted, namely beam voltage of 15 kV, beam current of 0.8 mA, and beam area of 61 mm^2. The generated CL was recorded as an image taken by a Bitran® BS-30C cooled CCD camera. The images obtained were expressed in the Red–Green–Blue (RGB) colour space as 16-bit values. They were first corrected by elimination of electric noise and background glimmer.

The CL emission is known to change continuously with time, in particular, at the start of bombardment (e.g., *Marshall*, 1988). We recognized such dynamic phenomena, even during focusing or beam adjustment. This indicates that CL colour depends strongly on the irradiation time of electron beam before imaging. Taking the results of preliminary time-dependent CL colour change measurements for CL colour changes of both a halo and the host low-quartz into consideration, we took images with exposure times of 120 s after 240 s irradiation.

Results and discussion

The CL images of synthetic low-quartz samples after He^+ implantation are shown in Fig. 1. In all of the images with a dose density greater than 1.14×10^{-4} C cm^{-2},

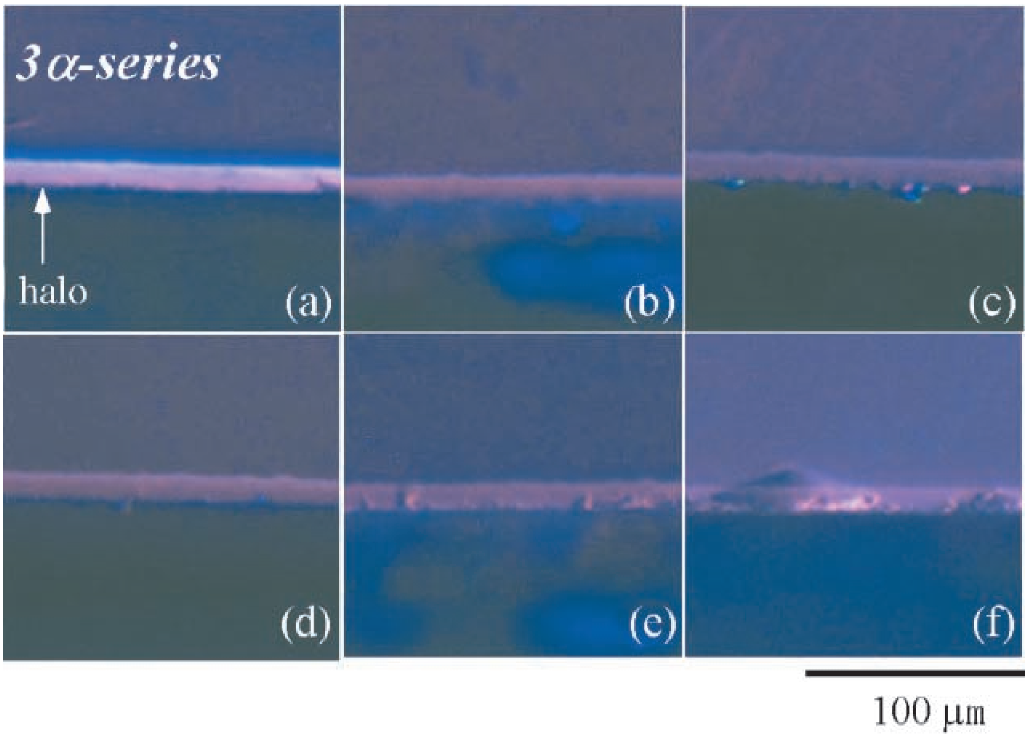

Fig. 1. CL images of synthetic low-quartz samples after He^+ implantation. **a** 3α-K (4.55×10^{-3} C cm^{-2}), **b** 3α-C (5.02×10^{-4} C cm^{-2}), **c** 3α-P (3.41×10^{-4} C cm^{-2}), **d** 3α-O (2.27×10^{-4} C cm^{-2}), **e** 3α-M (1.51×10^{-4} C cm^{-2}) and **f** 3α-N (1.14×10^{-4} C cm^{-2}), in order of decreasing dose density. CL halos with a width of about 14 μm are recognized in all the samples. In each of the halos, a brighter zone can be found at 11–14 μm from the surface

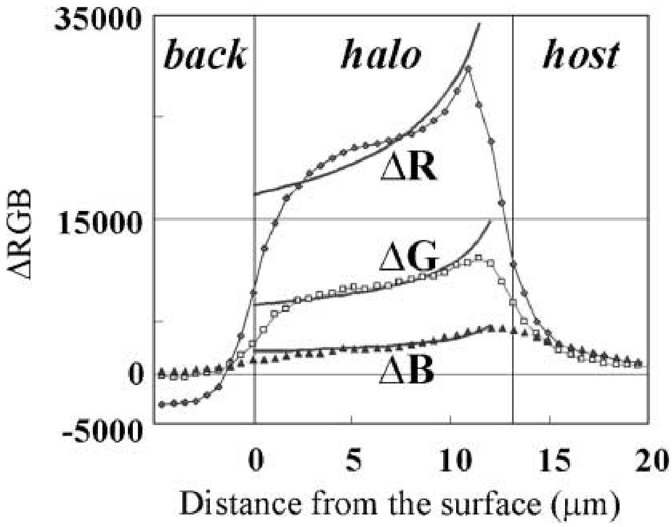

Fig. 2. Colour profile for the sample of 3α-K (4.55×10^{-3} C cm^{-2}). Colour values are denoted as the difference between halo and the host. All the ΔR, ΔG and ΔB values increase exponentially from the surface to the inside, showing a good correlation with the Bragg's curve, the approximation of which is shown by thick curves

a CL halo of brighter colour is recognizable in the area about 14 μm from the implantation surface. This confirms experimentally that the CL halo in low-quartz is formed by alpha radiation. The width is consistent with 13.2 μm, which is the theoretical range of an alpha particle of 4 MeV (*Owen*, 1988). In these images, brighter zones can be found at 11–14 μm from the surface.

A colour profile for the sample of 3α-K (4.55×10^{-3} C cm^{-2}) shows that all of the ΔR, ΔG and ΔB values, which are the differences between halo and the host, increase exponentially from the surface to the inside (Fig. 2). This shows that the colour profiles correlate well with the Bragg's curve, i.e., specific ionization in a material as a function of depth from the surface, formulated approximately as follows:

$$i = C(R - x)^{-1/3}$$

where i = specific ionization; C = constant (proper to a material); R = range of alpha particle, and x = depth from the surface. This indicates that the difference of CL colour is closely related to specific ionization, i.e., the damage by irradiation, and can possibly be used as a dosimeter. In the profiles for smaller dose density, all of the ΔR, ΔG and ΔB values increase from the surface to the inside, but the correlations with the Bragg's curve are not so obvious.

Although the colour of the host hardly changes with dose density, that of the halo changes notably. The relationship between dose density and the CL colour of a halo, which is denoted as the difference between the averaged value throughout a halo and the host, is illustrated in Fig. 3. The ΔR, ΔG and ΔB values increase with dose density, but the gradients gradually decrease, showing a clear relationship as is expected in a dosimeter. The ΔR, ΔG and ΔB values seem to reach constants or to be saturated when dose density reaches over a certain value.

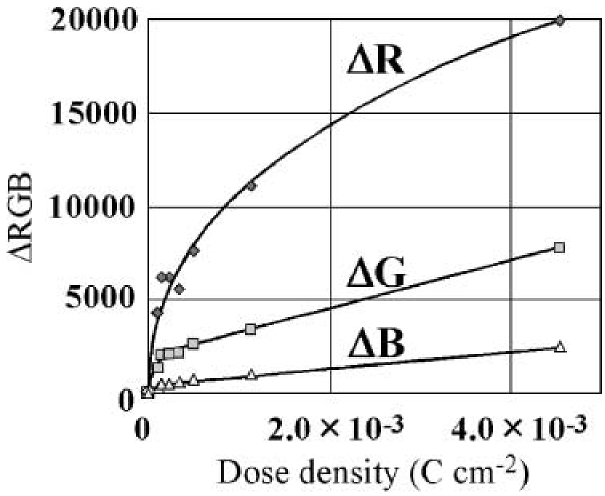

Fig. 3. The relationship between dose density and the CL colours of halos. The differences of RGB between the averages for whole halo and the host are plotted against dose density. All the ΔR, ΔG and ΔB values increase with increasing dose, but the gradients gradually decrease. The ΔR, ΔG and ΔB values seem to reach constants or to be saturated when dose density reaches over a certain value. Curves indicate general trends

The preliminary results of CL measurement after He^{+} implantation for samples of different origin suggest that the relationship between colour change and dose density is different among the samples. The reason is not clear but might be related to the numbers of non-bridging oxygen (*Götze* et al., 2001) or some other kind of lattice defects induced by irradiation. The analysis of CL spectra may be useful for understanding this phenomenon. Further CL measurement after He^{+} implantation of low-quartz samples of various origins, combined with other characterization such as spectroscopic, chemical and XPS analysis, is also necessary to develop a practical dosimeter for geological samples.

Conclusion

CL measurement of the synthetic low-quartz samples after He^{+} implantation showed that bright CL halos of about 14 μm in width from the implantation surface are recognized for those samples where dose density was greater than $1.14 \times 10^{-4}\,C\,cm^{-2}$. It has been shown experimentally for the first time that a CL halo can be formed by alpha radiation. It has also been shown that CL colour changes continuously with dose density, demonstrating that the CL halo may be used as a new dosimeter. Combined with petrographic observation and chemical mapping of radionuclides, the dosimeter could be useful not only for dating but also for analysis of radionuclide migration in natural geological media, especially for studying removal or immobility of radionuclides in ancient uranium deposits. This would be a very valuable application in natural analogue studies relevant to radioactive waste disposal. Before the dosimeter can be applied confidently to natural samples, more work will be required on the relationship between CL colour and dose for low-quartz of various origin, supported by other analytical methods.

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