Ann. Rep., Inst. Geosci., Univ. Tsukuba, no. 27, pp. 25-31, Dec. 25, 2001.

Operating conditions for recognition of radiation-damage halo in quartz using cathodoluminescence microscope and cooled CCD camera

Kosei Komuro and Yoshitada Horikawa*

Introduction

Under cathodoluminescence (CL), radiation-damage halo is often found in quartz adjacent to radioactive minerals (*e.g.*, Smith and Stenstrom, 1965; Owen, 1988; Komuro *et al.*, 1995). As CL emission of quartz is not so strong, it is difficult to recognize faint CL halo under microscope. Photographic recording is widely conducted because it is able to register low-intensity CL with long time of exposure. Recently, cooled CCD camera is known to be useful and widely used in recording faint luminescence with shorter time of exposure, however, its application for observation of radiation-damage halo under CL has not been reported. In order to know the most suitable operating conditions for recognition radiation-damage halo in rock samples even of faint CL without distinct beam damage, we examine here the relation between operating conditions and appearance of CL halo in images taken by a cooled CCD camera for quartz from sandstone-type uranium ore samples.

Samples and apparatus

Samples used in the present study are thin sections of uranium ores from the Kaneyamba sandstone-type deposit, Zimbabwe, of Triassic to Jurassic in age. The ores consist of detrital grains mainly of quartz with feldspars and matrices of hydromica minerals with uraninite and coffinite. Radiation-damage halos are recognized in the marginal part of detrital grains of quartz and feldspars under CL. The petrographical, geochemical and CL descriptions are shown in Komuro and Koyama (1993) and Komuro *et al.* (1994, 1995).

The used CL unit was a Luminoscope® ELM-3R equipped with an Olympus BX-60 microscope with a Bitran BS-30C cooled CCD camera furnished in a darkroom. In this unit, an electric beam generated within a cold cathode electric gun passes through an aperture in an anode and a focus coil into a sample chamber. In the chamber, the beam is deflected downward onto the sample by the magnetic field produced by magnets on a yoke placed on the top of the chamber. Position, shape and bombardment area of the beam on the sample are adjusted with the focus coil current, and position and amount of magnets on the yoke. The beam current and voltage are strongly affected by the air pressure in the chamber that is adjusted with a current-controlled solenoid leak valve. Maximum beam current at every voltage in the unit is shown in Fig. 1.

Experimental

An appearance of CL is closely related with beam operating conditions such as voltage, current, and bombardment area on the sample. The best and used beam bombardment conditions were outlined by controlling the position and amount of magnets on the yoke with a method of trial and error. With changing the focus coil current under the conditions, a series of beam of ellipsoidal shape with different bombardment area can be obtained. In addition to beam operating conditions, imaging conditions especially of exposure time have a deep influence on the clearness of image. Here we examined the effect of (1) beam bombardment area, (2) beam voltage and current, and (3) exposure time, for the appearance of CL halo together with the beam damage of sample as follows:

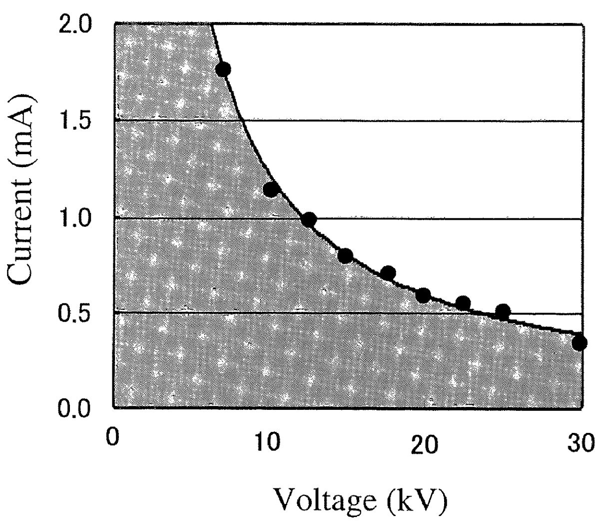

Fig. 1. The range of voltage and current obtained in the unit. The beam available is limited within the shaded area in the figure.

* Doctoral Program in Geoscience, University of Tsukuba

(1) The effects of beam bombardment area from 8.4 to 106 mm^2 by changing the focus coil current for the beam of 15 kV and 0.8 mA were examined with the exposure time of 90 seconds. The smallest beam is focused on the samples.

(2) The effects of voltage ranging from 10 to 25 kV and current from 0.4 to the maximum value at each voltage in Fig.1 were examined with the beam bombardment area of 27 mm^2. The exposure time was also fixed as 90 seconds.

(3) The effects of exposure time from 1 to 300 seconds were examined for quartz samples of strong and faint CL halos with the beam of 15 kV, 0.8 mA, and 27 mm^2.

The appearance of CL halo in the examinations was recorded as image data by cooled CCD camera. The image data were firstly corrected by elimination of electric noises and background glimmer. The data originally denoted as the RGB space were finally converted to the popular L*a*b* color space, in which the value of L*, a* and b* represents lightness, redness (+)-greenness (–) and yellowness (+)-blueness (–), respectively. The following conditions were fixed throughout the examinations: objective lens: ×20, ocular lens: ×5 and cooling temperature of CCD: -5 ± 0.5 ℃.

It is well known that CL emission changes with time, especially in the start of bombardment, even in focusing or beam adjustment works. We examined previously the change of CL emission from a halo and the host quartz with time and found that the differences in color are not so large as those by beam operating conditions, even at the start of the bombardment. Our imaging in the present examinations was carried out after the bombardment of 240 seconds or more to minimize the effect of CL change at the start of bombardment.

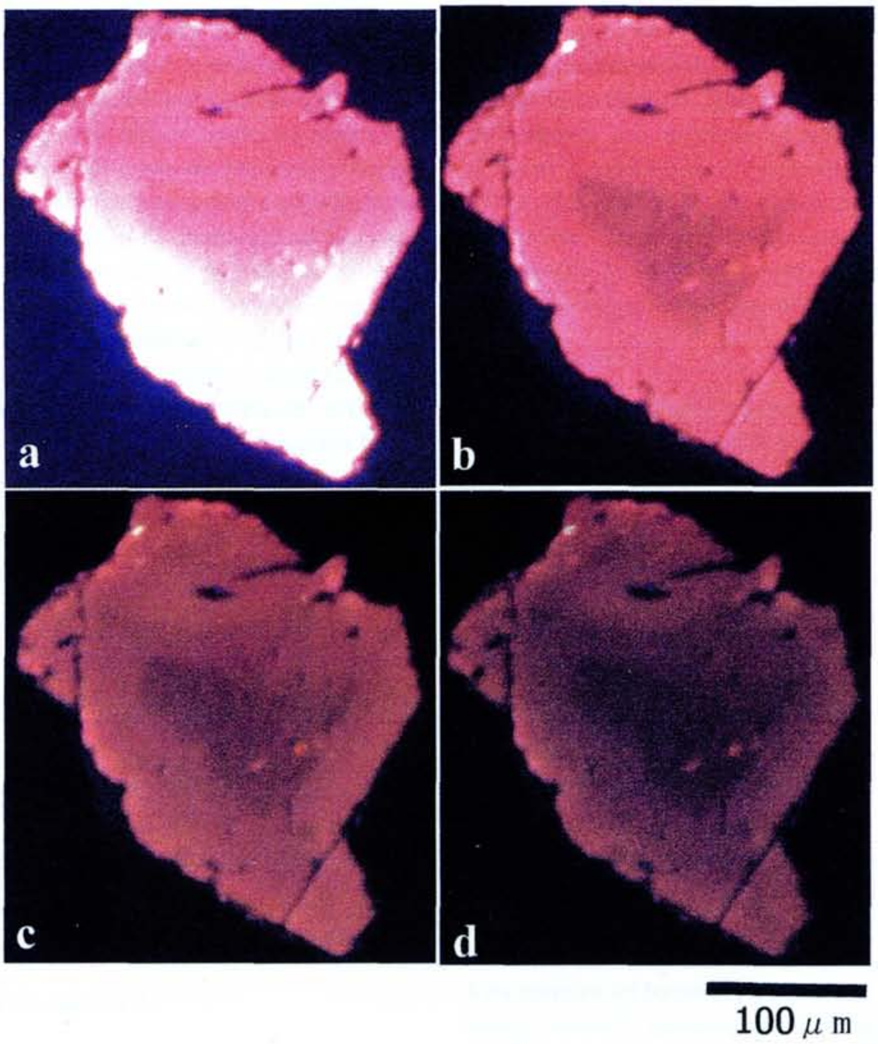

Fig. 2. CCD images of halo and the host quartz under different beam bombardment areas. The areas are (a) 8.4, (b) 27, (c) 75, and (d)106 mm^2. The brightness decreases with increasing of the bombardment area. Bright and dark portions are markedly found in (a).

Results and discussion

Figure 2 is the images of CL halo in quartz from the Kaneyamba uranium ores under the different conditions of beam bombardment area. The relationship between the color of halo and beam bombardment area for halo, host quartz and matrices at the same measurement position is shown in Fig. 3. This indicates that the values of L*, a* and b* of both halo and the host decrease with increasing the beam bombardment area, showing that the CL emission of quartz increase with beam density. In the case of smallest beam condition, however, the CL emission is not homogeneous with brighter and darker parts even in the same quartz probably due to beam structure (Fig. 2a). Such heterogeneous emission is not so obvious but partly recognizable under the larger beam conditions, suggesting that setting of the position of magnets on the yoke and changing the focus coil current must be made with paying attention to check the beam homogeneity in a visual field. Distinct beam damage was not so obvious in the mineral grains throughout the cases, but found in the mounting rasins under 27 mm^2.

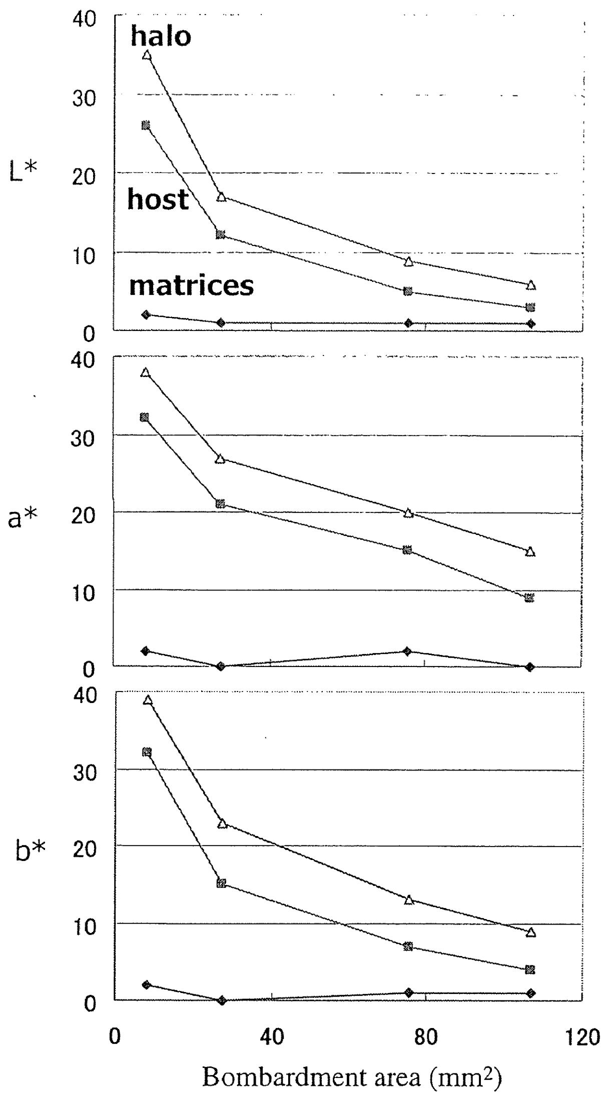

Fig. 3. The relationship between the L*, a* and b* values and bombardment area for halo (△), host quartz (■) and matrices (◆). The values of L*, a* and b* for halo and host decrease with increasing of bombardment area, while those for matrices are almost constant.

The relationship between the color of halo and beam voltage and current at the same measurement position is shown in Fig. 4. In each voltage, the L*, a* and b* values of both halo and host quartz increase with beam current, showing the CL emission increases with beam current. Among the conditions of maximum current of each voltage, the case of 15 kV has the highest L*, a* and b* values, showing the condition of the strongest CL emission. Distinct beam damage of the sample was not found throughout the examined conditions.

The effects of exposure time for quartz samples of strong and faint CL halos are in Figs. 5 and 6, respectively, in which brightness of the images is corrected to be clearly visible with keeping color balance. It is noted that the image with longer exposure is clearer. The exposure time over 90 seconds is good enough to reveal fine CL structure even for faint CL. Taking the upper limit of the range of CCD imaging also into consideration, exposure time from 60 – 90 seconds is appropriate for recording CL halo in quartz.

We conclude here that the beam condition with 15 kV, 0.8 mA and bombardment area of 27 mm^2 is the most suitable for recognition of radiation-damage halo in quartz clearly in the unit. For natural samples after mounting, bombardment area of about 75 mm^2 is practical. The descriptive works of halos with various occurrences are necessary for better understanding of the development of halos in nature.

Acknowledgements

We are thankful to Prof. Y. Kajiwara of the Univ. of Tsukuba and Dr. S. Toyoda of Okayama University of Science for their continuous encouragement, and Messrs. H. Okada and H. Numazawa of Seishin Co. for technical supports. This study was partly supported by the resources of REIMEI Research of JAERI (Japan Atomic Energy Research Institute) and by the JNC (Japan Nuclear Cycle Development institute) Cooperative Research Scheme on the Nuclear Fuel Cycle.

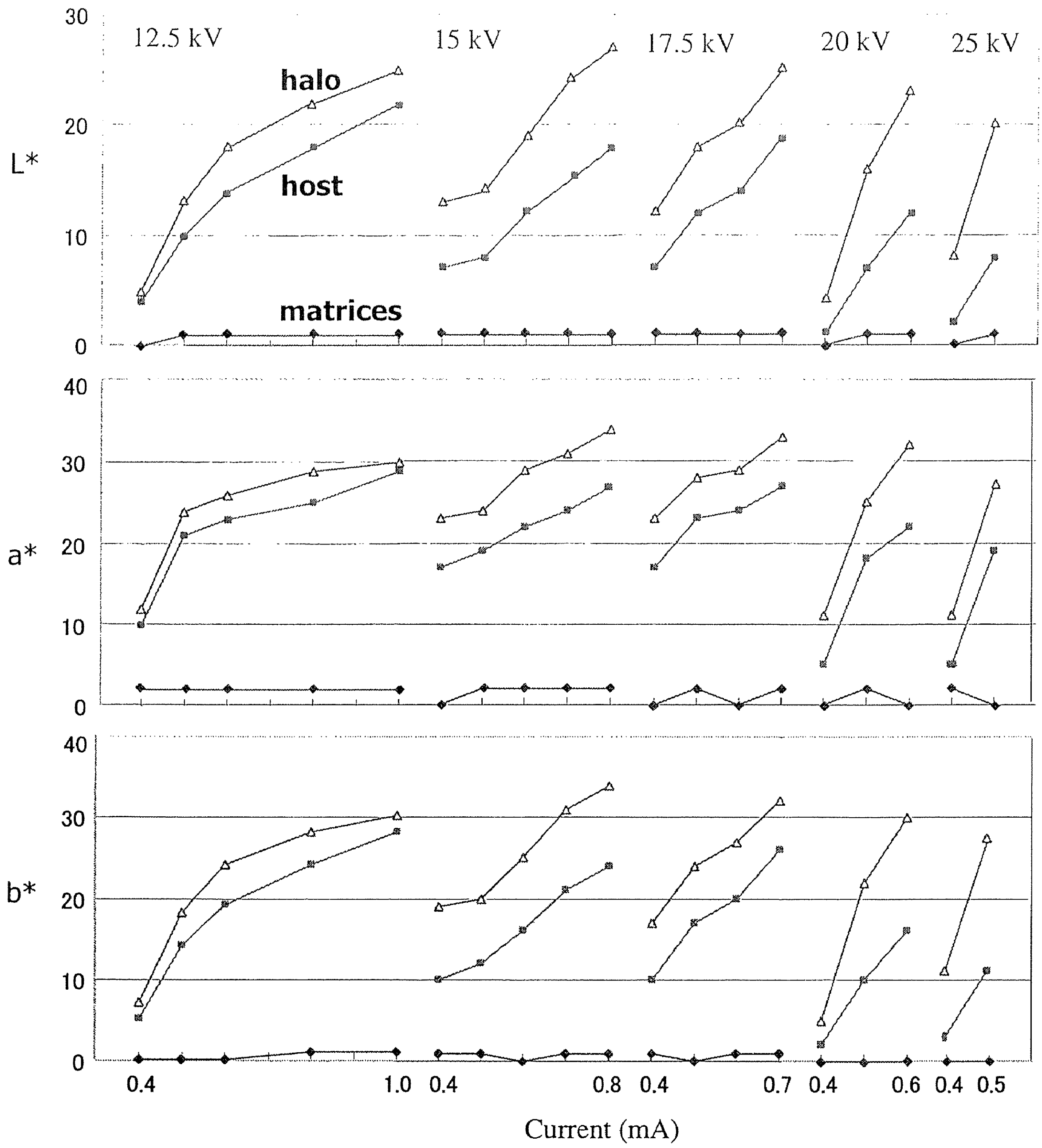

Fig. 4. The relationship between the L*, a* and b* values and beam voltage and current for halo (△), host quartz (■) and matrices (◆). The values of L*, a* and b* for halo and host increase with increasing of current in each case of voltage, while those for matrices are almost constant. Among the conditions of maximum current of each voltage, the case of 15 kV has the highest L*, a* and b* values.

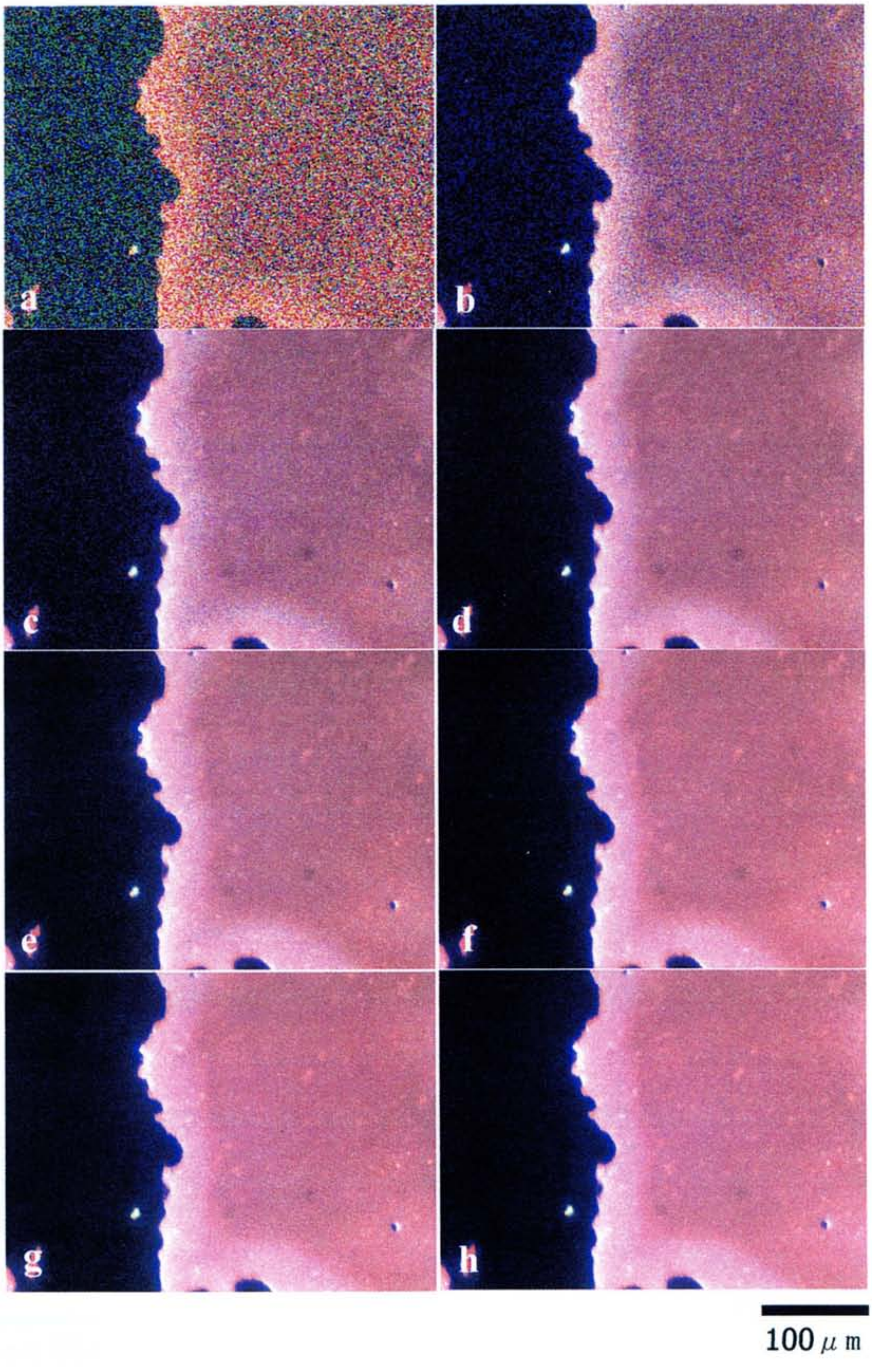

Fig. 5. CCD images for quartz with strong CL halos from the Kaneyemba ore of 1.42 wt% U_3O_8 for different exposure time. The exposure times are (a) 1, (b) 10, (c) 30, (d) 60, (e) 90, (f) 120, (g) 180 and (h) 300 seconds. The exposure time of over 60 seconds is good enough to reveal fine CL structure.

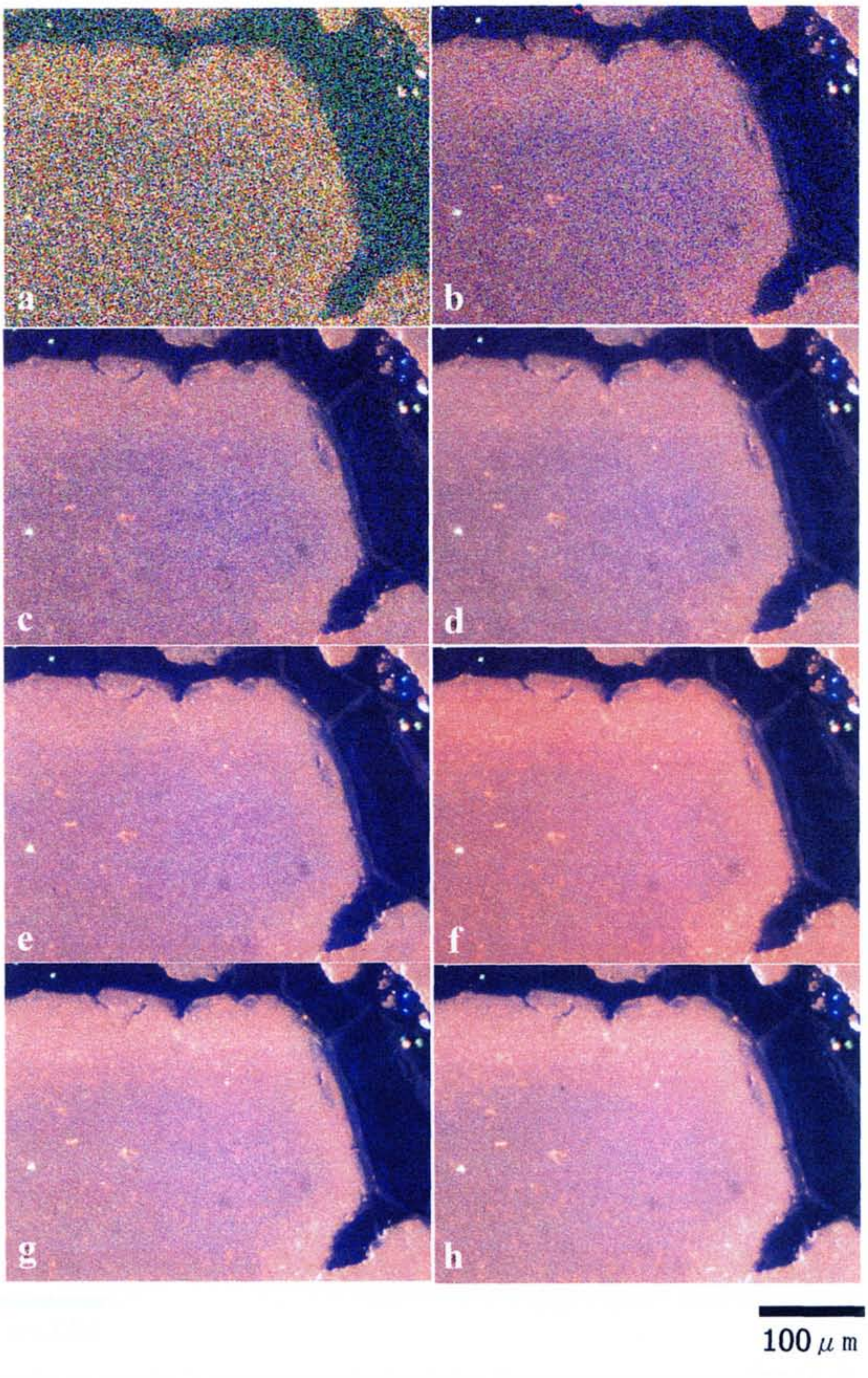

Fig. 6. CCD images for quartz with faint CL halos from the Kaneyemba sample of 0.06 wt% U_3O_8 for different exposure time. The exposure times are the same as in Fig. 5. The exposure time of over 90 seconds is good enough to reveal fine CL structure.

References

Komuro, K. and Koyama, K. (1993): *Ann. Rep., Inst. Geosci., Univ. Tsukuba*, **19**, 73-78.

Komuro, K., Suzuki, S., Yamamoto, M., Ohtsuka, Y. and Koyama, K. (1994) *Ann. Rep., Inst. Geosci., Univ. Tsukuba*, **20**, 67-72.

Komuro, K., Yamamoto, M., Fujiki, S., Fukushima, T. and Koyama, K. (1995): *Ann. Rep., Inst. Geosci., Univ. Tsukuba*, **21**, 65-69.

Owen, M. R. (1988): *Geology*, **16**, 529-532.

Smith, J. V. and Stenstrom, R. C. (1965): *J. Geol.*, **73**, 627-635.

Key words: radiation-damage halo, radiation damage, cooled CCD camera, cathodoluminescence, quartz and beam operating conditions.