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Gamma Irradiation Effect on Rock Crystals

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Abstract

Gamma irradiation effect on rock crystals from various geologic origins was studied. Gamma irradiation produced smoky color and sometimes citrine color in rock crystals, but a few of rock crystals were not affected.

The pattern of smoky coloration is classified into three types which are related to the geologic origin; 1. normal type, 2. lamellar type, 3. irregular type.

The degree of blackening of rock crystals from pegmatite is roughly related to the temperature at which the crystal was formed, and the thermal history of the crystal can be well presumed.

On absorption spectra of rock crystals, A_2 peak position is variable in 440-500 mμ range. Rock crystals with A_2 peak at about 440-460 mμ have high intensity in smoky color and mostly occur in pegmatite, whereas crystals with A_2 peak at about 500 mμ have low intensity in smoky color and mostly occur in hydrothermal vein.

The wide variation (440-500 mμ) of A_2 peak position is thought to be caused by the difference of interstitial ion in Al defect.

Citrine color has absorption peak at 390-410 mμ and is often coexistent with smoky color. Rock crystals with citrine color occur in hydrothermal vein or skarn.

Introduction

Many works regarding color center of quartz by X-ray, γ-ray or neutron irradiation are well known (BAMBAUER, 1961; COHEN, 1956; MITCHELL and PAIGE, 1956; ÓBRIEN, 1955).

The studies of blackening by X-ray or gamma irradiation on rock forming quartz from various geologic origin have been made in recent years, and it has been found that the degree of blackening is roughly related to the temperature at which quartz was crystallized (HAYASE, 1961; RITTER and DENNEN, 1966). According to previous reports, it is stated that blackening of rock forming quartz is homogeneous but that of rock crystal is mostly heterogeneous (HAYASE, 1961; RITTER and DENNEN 1966).

In the present work, blackening by gamma ray was studied on rock crystals from various geologic origins and the grade of blackening in single rock crystal was clarified, and also their optical absorption spectra in the visible region were studied.

Experimental procedure

About forty rock crystals obtained from pegmatite, hydrothermal vein and skarn were examined. They were usually cut perpendicular to the *c*-axis at the basal portion or sometimes cut parallel to the *c*-axis with the thickness of 1 mm, the surfaces were well polished.

The slices perpendicular to the *c*-axis were about 0.7–3.5 cm in diameter. The slices of rock crystals from hydrothermal vein and skarn were transparent, but those from pegmatite usually showed slightly smoky color.

The slices were irradiated by gamma rays of the dosage of 10^7 roentgen. The gamma rays used were from cobalt 60 gamma ray source, and the dose rate was about 1.18×10^5 roentgen per hour. The 10^7 roentgen of cobalt 60 irradiation is nearly the saturation dosage for blackening of quartz. The slices which show smoky color caused by natural radioactivity from the rock were irradiated without decoloring by heating, as heat treatment affects the degree of blackening of quartz by artificial gamma irradiation (RITTER and DENNEN; 1966). The blackening of the crystal by natural radioactivity is usually weak in intensity and therefore almost negligible in comparison with the blackening by the artificial irradiation of 10^7 roentgen.

The degree of their blackening after gamma irradiation was measured with Jarrel Ash recording microphotometer scanning from the edge toward the center perpendicular to *m* plane and the curves of the variation of blackening were recorded automatically. Influences of inclusions and fractures to light transmission were carefully checked.

Measurements of the absorption spectrum were done at the most colored part of the slices by Shimazu spectrophotometer using normal light within the range of 210–800 mμ at the room temperature.

Results and discussion

After gamma irradiation, most samples showed smoky color, some samples showed citrine color, and a few samples were not colored. Rock crystals from the same druse are similar in intensity and their patterns of coloration, but those from different druse, a considerable difference is usually detectable.

Type of smoky colored rock crystal

1. Normal type: In this type, the samples are colored almost uniformly, or the central part is deeply colored and the degree of blackening is decreased toward the edge. Rock crystals from pegmatite accompanied by microcline and those from a few hydrothermal veins belong to this type (Figs. 1 and 2).

2. Lamellar type: As already mentioned by BAMBAUER *et al.* (1961), a mimetic

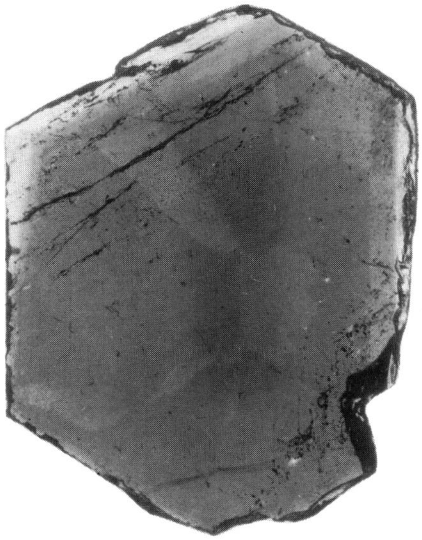

Fig. 1.

Fig. 2.

Figs. 1,2. Rock crystal from Tanakami district, Shiga Prefecture. Section (Fig. 1. 14 × 17 mm, Fig. 2. 14 × 28 mm.) perpendicular to the *c*-axis. The difference of blackening between two individuals of the twin is observed.

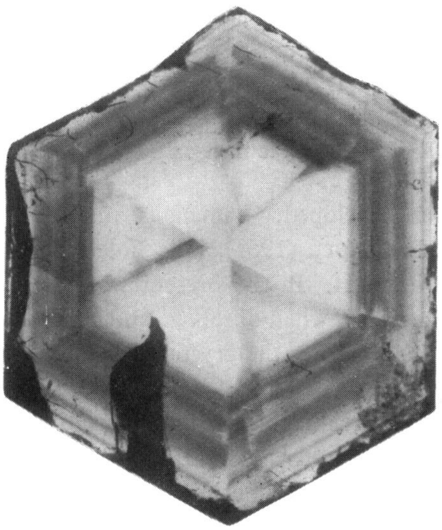

Fig. 3. Rock crystal from Obira mine, Ooita Prefecture. Section (10 mm in diameter) perpendicular to the *c*-axis.

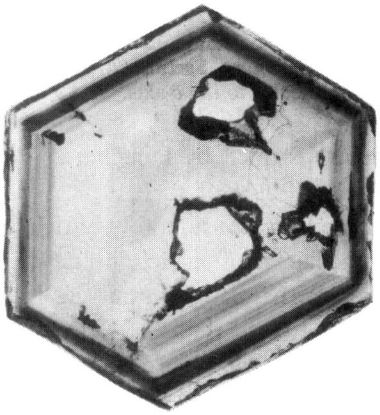

Fig. 4. Rock crystal from Tsukide, Shiga Prefecture. Section (8 mm in diameter) perpendicular to the *c*-axis.

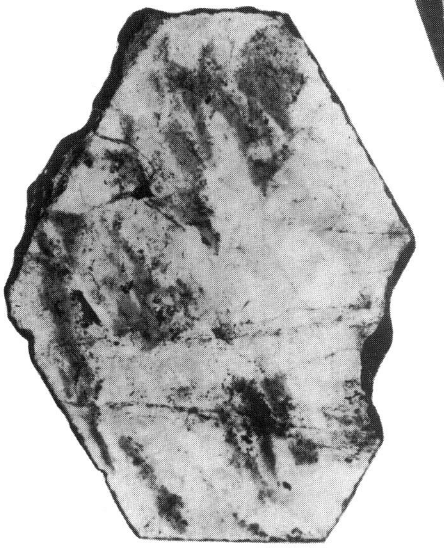

Fig. 5. Rock crystal from Kurobira, Yamanashi Prefecture. Section (20×25 mm) perpendicular to the *c*-axis.

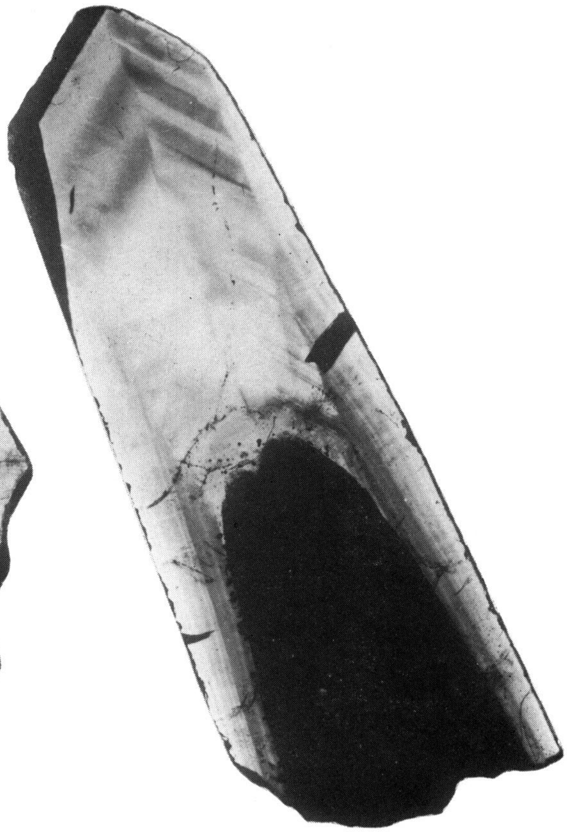

Fig. 6. Rock crystal from Obira mine, Ooita Prefecture. Section (12×40 mm) parallel to the *c*-axis.

structure formed by lamellae of symmetry are developed. Lamellae* are parallel to $r(10\bar{1}\bar{1})$, $z(01\bar{1}1)$ or $m(10\bar{1}0)$. According to BAMBAUER (1961), it is noted that such lamellae* are considerably enriched in Al, Li and H. In the present work, the part of lamellae* is often rich in fine twins after Brazil law when observed under the microscope. Perhaps high impurities in lamellae* may deform the crystal lattice and easily form the twins.

Such lamellae* are thought to be formed at relatively low temperature because they are often observed in rock crystals from hydrothermal vein, skarn and quartz pegmatite, and especially they are often developed at the peripheral part of a rock crystal. BAMBAUER *et al.* (1961) thought the lamellae* to be of secondary formation

* Lamellae mean the blackened lamellae which are produced by X-ray or gamma ray irradiation.

from their observation, but in the present work, it is not determinable whether the lamellae* are of secondary formation or not (Figs. 3, 4 and 6).

3. Irregular type: Rock crystals were rarely colored irregularly. One of them (Kurobira, Yamanashi Prefecture) is colored around the inclusions and numerous fine twins after Brazil law are observed in the colored part under the microscope (Figs. 5). Rockcrystals which belong to this type occur in hydrothermal vein.

Difference of smoky coloration between the two individuals of the twin

It is well known that the twin is differently colored by the radiation. In the writer's samples, the difference of coloration occuring in Dauphiné twin is often observed.

In the twinned crystal, the larger individual of the twin is generally more deeply colored than the smaller one as shown in Figs. 1 and 2. In rare cases twin boundaries are colored.

As FRONDEL (1945) mentioned, the origin of the differential coloration of twinned quartz is problematical. In general, the difference of smoky coloration is correlated to the difference of the content of Al substituted for Si in tetrahedron. However, in the case of Dauphiné twin, the two individuals of the twin are thought to be crystallized in the same condition. So FRONDEL (1945) imagined that there has been a diffusion of the sensitizing factor within the crystal which was dammed up at the twin boundaries.

The writer's opinion is as follows; during or after crystallization, minor elements continue to diffuse into the individuals of the twin from outside. In Dauphiné twinned rock crystal, it is thought that there are two types of the barrier against the diffusion from outside into the crystal; one is the crystal boundaries, the other is twin boundaries in the crystal. In this case, the specific area of barrier per unit volume of the smaller individual of the twin is greater than that of the larger individual of the twin. From this reason, the diffusion to the smaller individual of the twin is less than that to the larger one, therefore the larger individual of the twin is more deeply colored than the smaller one.

The curve of blackening in rock crystal as a geothermometer

It is generally known that quartz formed at higher temperature is more deeply blackened. The writer collected rock crystals from druse in pegmatites located in Tanakami district, Shiga Prefecture. The druse consists of rock crystal and microcline, or rock crystal, microcline, topaz and muscovite, or rock crystal only.

For comparison of the curves of blackening, rock crystals similar in size were selected from each pegmatite to prepare specimens of slices perpendicular to the *c*-axis and they were used for the experiments.

The results are shown in Figure 7. Curve I in the figure is high in intensity, and Curve II is relatively high in intensity continuing from the center to the edge. Curves

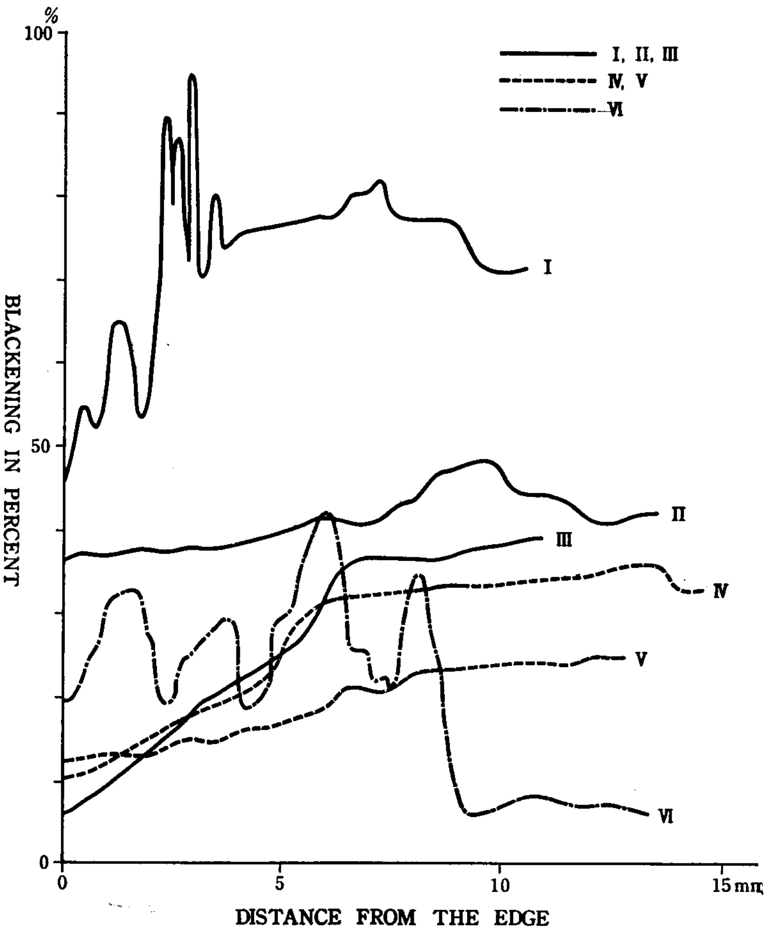

Fig. 7. Blackening curve in rock crystal from druse of pegmatite in Tanakami district.
I, II, III Rock crystal accompanied by microcline.
IV, V Rock crystal accompanied by microcline and topaz.
VI Rock crystal only.

III, IV and V are of moderate intensity in the central part, and gradually become weaker toward the edge. Curve VI shows low intensity at the center, from which intensity varies alternatingly high and low toward the edge. This variation depends on the lamellae parallel to *r* and *z* plane. Such lamellae are thought to be formed at relatively low temperature, so the variation of the curve VI is not so directly related to the temperature as other curves are.

From these curves, it is thought that rock crystal I was crystallized at a considerably high temperature, and rock crystal II began to crystallize at a relatively high temperature and the temperature was preserved until the end of crystallization. Rock crystals III, IV and V began to crystallize at a moderate temperature, and the temperature gradually decreased. Rock crystal VI began to crystallize at a low temperature as in a hydrothermal vein.

From these results, it is conclusive that a druse which consists of microcline and rock crystal (Curves I, II and III) was generally formed at higher temperature than the temperature at which druse containing topaz (Curves IV, V) was formed, and druse which consists of rock crystal only (Curve VI) was formed at lower temperature than the others.

Absorption spectrum of rock crystal from various geologic origin

As already described, gamma irradiation usually produced smoky or citrine color in rock crystals. Measurements of absorption spectra were done on the slices perpendicular to the *c*-axis, having the thickness of 1 mm.

Smoky color : It is well known that smoky color is represented by an absorption band with a pronounced peak at about 460 mμ (A_2 peak) and with also less pronounced peak at about 620 mμ (A_1 peak). As the previous data (COHEN, 1956), measured on the slice perpendicular to the *c*-axis, the peak at about 620 mμ (A_1 peak) is obscure, but the peak at about 460 mμ (A_2 peak) is considerably clear, so the writer preferred the A_2 peak for the measurements of rock crystals from various geologic origins.

The variation of A_2 peak position in rock crystal is shown in Figure 8. The A_2 peak position is variable in the 440–500 mμ range. The rock crystals with A_2 peak at about 440–460 mμ mostly occur in pegmatite; they were relatively high in blackening. But the rock crystals with A_2 peak at about 500 mμ occur in hydrothermal vein, skarn and quartz pegmatite; they showed relatively low degree of blackening. Thus, two types of smoky colored quartz are distinguished from the A_2 peak position. This variation may be caused by the difference of interstitial ion in Al defect.

According to HALPERIN and RALPH (1963), synthetic quartz containing the alkali ion (Na^+, Li^+) showed the main peak at 450 mμ in the visible region of absorption spectrum after X-ray irradiation, then the alkali ions in this synthetic quartz were replaced by hydrogen by hydrolysis and after X-ray irradiation of this sample at extreme low temperature (77 °K), main peak appeared at about 500 mμ instead of 450 mμ.

Similar results were obtained by the above authors in the case of quartz crystal into which potassium was diffused. Such experimental data show that the different interstitial ion may affect the A_2 peak position.

In rock crystals which have A_2 peak at about 440–460 mμ, the color is perhaps caused by Al-Na or Al-Li defect, but those which have A_2 peak at about 500 mμ, the color may be caused by the Al defect in which the interstitial ion consists of positive ion except Na and Li. Hydrogen is perhaps not possible because the color is not

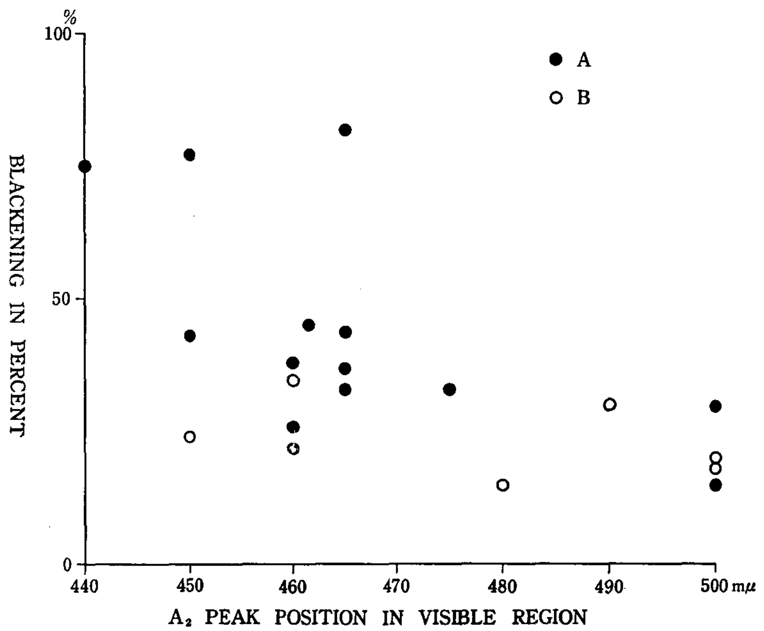

Fig. 8. Relation between A_2 peak position and blackening.
A; Normal smoky rock crystal
B; Rock crystal with smoky lamellae

produced by Al-H defect at room temperature, though the color is produced at extreme low temperature (BAMBAUER, 1961; HALPERIN and RALPH, 1963; etc.), but potassium ion may be probable.

Citrine color : Citrine color has absorption peak at 390-410 mμ and is produced in rock crystals from hydrothermal vein and skarn.

It is known that citrine color is produced by gamma irradiation in quartz synthesized at relatively low temperature and pressure in a strong solution of sodium carbonate (GORDIENKO *et al*, 1967).

From the datum mentioned above and mineralogical occurrence, citrine colored quartz is thought to have grown at relatively low temperature.

Citrine color and smoky color are often coexistent; in one case, smoky lamellae develope in citrine colored crystals, in another case, citrine color is masked by smoky color in rock crystals belonging to normal type. In the latter case, the existence of citrine color is confirmed by the measurement of absorption spectrum.

Summary

1. Smoky or citrine color is produced in most of rock crystals by gamma irradiation and the pattern of coloration of rock crystal is related to the occurrence.

2. Difference of blackening between two individuals of the twin is often observed.

3. The curves of blackening were tentatively used as a geothermometer and the thermal history of various rock crystals were presumed.

4. In the absorption spectra of smoky colored crystals, it is found that A_2 peak positions are variable in a wide range (440–500 mμ) and its cause was considered. Rock crystal with A_2 peak at about 440–500 mμ mostly occurs in pegmatite and has high intensity in smoky color, whereas crystal with A_2 peak at about 500 mμ mostly occurs in hydrothermal vein and has low intensity.

5. Absorption spectrum of citrine color was measured, and from various data, citrine colored quartz is thought to be formed at relatively low temperature.

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References

BAMBAUER, H.U., G.O. BRUNNER and F. LAVES (1961): *Zeit. Krist.*, **116**, 173–181.

BAMBAUER, H.U. (1961): *Schweiz. Min. Petr. Mitt.*, **41**, 335–369.

COHEN, A.J. (1956): *Jour. Chem. Phys.*, **25**, 908–914.

FRONDEL, C. (1945): *Amer. Mineral.*, **30**, 432–446.

GORDIENKO, L.A., V.G. LUCHNIKOV., M.I. SAMOILOVICH., V.E. KHADZHI., L.I. TSINOBER and E.N. TSYGANOV. (1967): *Growth of crystals* (translated from Russian, Consultants Bureau, New York), 7, 297–300.

HALPERIN, A. and J.E. RALPH. (1963): *Jour. Chem. Phys.*, **39**, 63–73.

HAYASE, I. (1961): *Inst. Chem. Research. Bull. Kyoto Univ.*, **39**, 133–137.

MITCHELL, E.W.J. and E.G.S. PAIGE. (1956): *Phil. Mag.*, Ser. 8, **1**, 1085–1115.

ÓBRIEN, M.C.M. (1955): *Proc. Roy. Soc. London*, **A231**, 404–414.

RITTER, C.J. and W.H. DENNEN. (1966): *Amer. Mineral.*, **51**, 220–228.