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Abstract

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THE INFLUENCE OF RHOMBOHEDRON GROWTH RATES ON THE EXTERNAL APPEARANCE OF QUARTZ CRYSTAL TERMINATIONS

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Quartz crystals are characterized by a great variety of external forms ([5]). However, properly bounded crystals are encountered relatively rarely. The terminations of crystals are especially often distorted as a result of the uneven development of the principal rhombohedra ([3]).

In paper ([1]) we discussed the causes of asymmetry in the structure of quartz crystal terminations and, on the basis of experimental material, showed that it is determined to a large extent by the orientation of the crystals and of their individual faces during growth in space relative to the gravitational field and the third-order axis. This is connected with the fact that the deposition of various micro- and macroparticles on a crystal face induces the appearance of new crystallization centers and thus stimulates their more rapid growth ([3]). Therefore those among identically named faces which are directed upward during crystal growth are, as a rule, the more rapidly growing. The difference in the growth rates of identically named faces leads to distortion of the external appearance of the crystal. However, study of natural quartz crystals in pegmatites and crystal-bearing hydrothermal deposits shows that asymmetry in the structure of crystal terminations does not always appear, even if their orientation during growth was most favorable for distortion. This prompted us once again to turn to clarification of the principal conditions under which distortion of the terminations of natural quartz crystals occurs, and also to reproduce similar conditions experimentally.

The study of natural crystals was carried out on thin (2–3 mm) polished plates, cut parallel to the third-order axis and preliminarily subjected to γ-irradiation. In the overwhelming majority of crystals, under γ-irradiation the sectors and growth zones appear distinctly, making it possible to trace the character of the change in the ratios of the growth rates of individual faces. At the same time it is clearly established that in crystals with asymmetric terminations, the increase in the growth rates of the faces directed upward begins to appear most

Fig. 1

Figure 1: Fig. 1

contrastly in connection with the precipitation on the crystal surface of solid inclusions (sericite, hematite, goethite, fluorite, kaolinite, and other minerals) or of numerous quartz crystals (3). The precipitation of the latter generally testifies to a marked change in the physicochemical conditions of mineral formation, most likely connected with intramineralization tectonic movements. For us it is especially important here that such a change in the conditions of mineral deposition leads to an increase in the supersaturation of the solution with silica and, as a consequence, to an increase in the growth rate of the crystals; in this case the growth rates of the faces directed upward exceed (by 2–10 or more times) the growth rates of the identically named faces directed downward. The homogenization temperature in each subsequent growth zone (in the direction toward the crystal termination) is lower than in the preceding one. This testifies to a relative increase in the supersaturation of the solution with silica due to a decrease in the crystallization parameters.

In crystals growing in an inclined position and having an undistorted vertex habit, a "deposit" of foreign minerals by growth zones (especially in the form of continuous "jackets"), as a rule, is not observed. In those cases where "deposit" material is recorded in such crystals, the growth rates of faces of the same name remain constant, despite their different positions relative to the gravitational field. Such cases are especially often noted in the lowest-temperature crystals (according to data from homogenization of inclusions), i.e., under conditions of relatively lower concentrations in the silica solution. Very often the vertices of such crystals have an almost hexagonal habit, with equal development of negative $\{01\bar{1}1\}$ and positive $\{10\bar{1}1\}$ rhombohedra. It is interesting that, among artificial quartz crystals as well, those grown at very low (close to zero) supersaturations of the solution with silica tend toward such a habit (2, 6). In the case of large supersaturations, however, crystals with a typical trigonal habit are formed, and the ratio of the growth rates of the $\{10\bar{1}1\}$ and $\{01\bar{1}1\}$ faces is the greater, the higher the supersaturation.

Fig. 1. Dependence of the areas S of the $\{01\bar{1}1\}$ faces (a) and $\{10\bar{1}1\}$ faces (b) on the angle $g\hat{N}$ (explanations in the text)

Comparison of the facts presented above indicates that the distortion of the vertices of quartz crystals is largely determined (other conditions being equal) by the magnitude of supersaturation of the solution with silica (i.e., by the absolute and relative growth rates of the faces) and in some cases may not appear at all.

The proposition stated was tested by us experimentally in growing artificial quartz crystals in solutions with different supersaturation by silica. Recrystallization of quartz was carried out by the hydrothermal method in high-pressure vessels (⁴). The magnitude of solution supersaturation was estimated relatively from the temperature difference Δt between the zones of dissolution and crystallization of quartz.

Quartz was grown on cylindrical seeds of the same diameter cut along (0001), which eliminated the influence of the initial shape of the seeds on the ratio of the face areas of the crystals obtained. The recorded areas of the $\{01\bar{1}1\}$ and $\{10\bar{1}1\}$ faces in this case reflected the ratio of the growth rates of the pyramids $\langle 01\bar{1}1\rangle$, $\langle 10\bar{1}1\rangle$, $\langle 0001\rangle$, and $\langle 10\bar{1}0\rangle$. In the course of the experiments the growth pyramid $\langle 0001\rangle$ wedged out, and the crystal vertices were formed either by the $\{01\bar{1}1\}$ and $\{10\bar{1}1\}$ faces, or only by $\{10\bar{1}1\}$. The growth rate of the hexagonal prism under the experimental conditions was practically equal to zero. The position of the growing face during the experiment was set and recorded by the angle between the normal to the growth face and the direction of the vector of gravity $\widehat{gN}$. The initial orientations of the $\langle 01\bar{1}1\rangle$ faces were specified by values of the angle $\widehat{gN}$ of 20, 50, 90, and 160°.

In the series of experiments conducted at relatively small values of Δt (3–4°), the $\{01\bar{1}1\}$ and $\{10\bar{1}1\}$ faces were present. In this case, the growth rates of the $\{10\bar{1}1\}$ face practically did not depend on orientation in the field of gravity, while the $\{01\bar{1}1\}$ faces, having a growth rate 2–3 times greater than the rate of the $\{10\bar{1}1\}$ face, showed a clear dependence on the angle $\widehat{gN}$. Variations in the area of the $\{01\bar{1}1\}$ faces under these conditions are unambiguously connected with changes in its growth rate. This makes it possible to judge the relative change in the growth rates of differently oriented faces from the ratio of their areas. The $\{01\bar{1}1\}$ faces had the smallest area (and, consequently, the greatest rate) at $\widehat{gN} = 160^\circ$ (Fig. 1). As can be seen,

already at $\Delta t = 3\text{-}4^\circ$, with a decrease in the angle $\widehat{gN}$, the growth rate of the face $\{01\bar{1}1\}$ decreases continuously. If the area of the face characterized by the largest value of the angle $\widehat{gN}$ (160°) is conventionally taken as unity, then at angles $\widehat{gN}$ equal to 90, 50, and 20°, the areas of the faces $\{01\bar{1}1\}$ increase, respectively, by factors of 1.7, 2.1, and 2.9. But since the apex of the crystal is composed mainly of the faces of the positive rhombohedron, whose growth rate under these conditions is extremely low, visually distorted crystal apices are almost not recorded (Fig. 2).

Two other series of experiments were carried out at comparatively identical temperatures and pressures, but at higher supersaturations (respectively $\Delta t = 6^\circ$ and $\Delta t = 23^\circ$). Under such conditions, as noted in [6], the faces $\{0001\}$ and $\{01\bar{1}1\}$, owing to higher growth rates, wedge out, and the apices of the crystals are bounded only by the faces $\{10\bar{1}1\}$. As a result, even slight differences in their areas lead to a clearly visually recorded distortion of the shape of the crystal apex. Thus, at $\Delta t = 6^\circ$ and $\widehat{gN} = 10^\circ$, the area of the face $\{10\bar{1}1\}$ is 1.2 and 1.4 times greater than at $\widehat{gN}$ equal to 75 and 160°, respectively. The apices of the crystals have a clearly distorted shape (Fig. 2). Still greater distortions are observed in experiments with $\Delta t = 23^\circ$. In these cases the faces $\{10\bar{1}1\}$ facing

Fig. 2. Quartz crystals grown at values of the temperature difference: a –23°, b –6-7°, c –3-4°

Figure 2: Fig. 2. Quartz crystals grown at values of the temperature difference: a –23°, b –6-7°, c –3-4°

upward wedge out practically completely, and the crystals acquire a shape with sharply beveled apices (Fig. 2).

Fig. 2. Quartz crystals grown at values of the temperature difference: *a* –23°, *b* –6-7°, *c* –3-4°

Thus, the investigations carried out showed that distortions of the external form of quartz crystal apices reflect not only the position of the growing crystal in space, but also the magnitude of supersaturation of the solutions with silica, which determines the absolute and relative growth rates of different faces. Quartz crystals grow in any position in space without distortion of the apices if the growth rates of $\{01\bar{1}1\}$ and $\{10\bar{1}1\}$ are close to one another and tend toward zero. This is achieved at extremely low supersaturations of the solution with silica. In nature such conditions are realized either in very low-temperature processes (for example, in the formation of ideally faceted crystals of Marmarosh diamonds, quartz crystals in mercury deposits, silicified limestones, agate geodes, etc.), when appreciable supersaturations cannot arise because of the very low overall solubility of silica; or at higher parameters (early stages in chamber pegmatites and, more rarely, in hydrothermal crystal-bearing veins), during the formation of crystals under very calm tectonic conditions.

A disturbance of the physicochemical parameters of crystallization, usually associated with intramineralization tectonic movements, leads to appreciable, sometimes substantial, supersaturation of the solutions with silica, as well as with other components. This promotes an increase

absolute values of the growth rates of the $\{01\bar{1}1\}$ faces and an increase in their ratio to the growth rates of the $\{10\bar{1}1\}$ faces, as well as the deposition of various minerals (including numerous quartz crystals) on the surfaces of the faces turned upward. Both of these factors, as was shown above, also determine, along with the orientation of the crystals in space during growth, the degree of distortion of their vertices.

In connection with the foregoing, it appears possible to judge more definitely the absolute growth rates of quartz crystals in nature. Most probably they grew considerably faster than is generally assumed at present (6), since the supersaturations of solutions with silica at which distortion of crystal vertices begins to appear provide growth rates of the principal rhombohedra on the order of thousandths of a millimeter per day.

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Note: Figure translations are in progress. See original paper for figures.