# A case of epitaxic replacement.

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Summary.—Quartz from a granite pegmatite near Kalkovo, Bulgaria, has undergone partial replacement by biotite along cracks parallel to  $(10\overline{10})$ ; the biotite is in regular orientation relative to the quartz, and appears to constitute an instance of epitaxic replacement, defined as a replacement in which the replacing mineral develops in a regular, structurally controlled orientation relative to the mineral replaced.

ALTHOUGH the term 'metasomatism' was originally used by C. F. Naumann to denote processes leading to the formation of mineral pseudomorphs, the term has since acquired a wider sense, being loosely applied when a rock undergoes partial or complete replacement. The process is thus interpreted mainly in a petrological manner, attention being paid primarily to the general physico-chemistry and mobility of the rock components under the action of metasomatizing solutions. Not much is said, however, about the way the original mineral and the later product contact each other—that is, about their structural relationship. And this is an important detail from a mineralogist's point of view.

According to the way solutions penetrate the rocks, diffusion and infiltration metasomatism are differentiated (Korzhinsky, 1955). Further, metasomatic processes include the introduction of potash, soda, boron, chlorine, sulphides, &c., whereas pneumatolitic, hydrothermal, vadose, and similar adjectives are used to give an idea of the nature of the metasomatizing solutions and the environment in which the metasomatic process is operative. All these could equally well be applied for the replacement of rocks and minerals, attention being paid usually to textural and structural controls (in the petrological sense) for the rocks. When, however, replacement is restricted to one individual mineral only, the structural character (in the crystallochemical sense) of this mineral in relation to that of the newly formed replacement product should be considered as an additional factor in that particular replacement. Such considerations could also throw light on the intimate mechanism of replacement.

The contact between two minerals formed in succession must be

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either fortuitous or controlled by the lattice structures. When the later mineral is formed at the expense of the earlier in a regular manner then such replacement could conveniently be termed epitaxic metasomatism. This term should only be applied to the replacement of minerals, but through the latter details could also be gathered about the general trend of the metasomatic process in rocks.

## A case of epitaxic replacement.

During certain morphological studies on quartz crystals from various occurrences in Bulgaria, regularly oriented lamellae of mica were found in cracks of quartz that has suffered post-crystallization deformation



FIG. 1. Crack along  $(10\overline{1}0)$  of quartz along which epitaxic replacement has taken place (slightly enlarged).

(figs. 1, 2, and 3). This quartz is found in granite pegmatites that cut Mesozoic (?) granodiorite near the village of Kalkovo, to the south of the town of Sofia. The pegmatites are coarse-grained and include, apart from quartz, plagioclase (oligoclase and albite), microcline, biotite, allanite, and minor quantities of tourmaline and sulphides (Vergilov, 1955; Nikolov, 1918). The quartz has been formed intermittently throughout the pegmatitic stage, the first formed being grey in colour and then successively smoky, again grey, colourless, and finally milky; the last generation frequently forms an outer rim on the crystals (fig. 2). In contrast to these large quartz crystals are minute waterclear crystals of quartz denoting the last stage of quartz formation, following patchy replacement of the former quartz crystals by gilbertite (Vergilov, 1955).

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#### EPITAXIC REPLACEMENT

Biotite, which is of interest in our case, is formed in two stages at higher and lower temperatures. The low-temperature stage obviously follows the post-crystalline fracturing of the quartz crystals referred to above. The mica lamellae which are found along the cracks of the quartz



FIG. 2. Epitaxic growth of biotite lamellae on  $(10\overline{1}0)$  of quartz.  $\times 4$ .



FIG. 3. The same as in fig. 2, the area of the biotite lamellae enlarged.

crystal are a few millimetres in size and invariably elongated along the *a*-direction. They are almost completely transformed into iron hydroxide and some clay substance, but certain less decomposed lamellae display a small biaxial figure that opens in the direction of the elongation:  $\gamma > 1.6$ . Obviously these mica lamellae represent strongly baueritized and hydrated biotite, and appear to correspond to the second generation of the biotite in the pegmatitic veins. The cracks are parallel to the (1010) face of the quartz crystal, which, as seen in fig. 1, is zoned and sectorial. The biotite lamellae are oriented with (001) parallel to (1010) of the quartz and their elongation (*a*) follows or is at 60° to the horizontal striations on the quartz prism zone. Such regular orientation between quartz and biotite is not referred to in the literature so far as it is known to the author, though regular orientation of quartz and rutile is well known (Royer, 1928; Shafranovsky, 1940; von Vultee, 1955). Having in mind the frequent regular intergrowth between rutile and altered biotite (sagenite) and between quartz and rutile it seems quite logical to expect the observed regular orientation between biotite and quartz. According to M. L. Royer (1928), in order to have epitaxic growth the difference between the corresponding parameters of the two crystal structures should be less than about 15 %. In the observed case the difference is as follows:

 Quartz
 .
 .
  $a 5 \cdot 300 \text{ Å.}, c 5 \cdot 393 \text{ Å.}$  

 Biotite
 .
 .
  $a 4 \cdot 903 \text{ Å.}, \frac{1}{2}b 4 \cdot 605 \text{ Å.}$ 
 $\Delta 0 \cdot 397 \text{ Å.}, \Delta 0 \cdot 788 \text{ Å.},$ 

or 8 % for a and 17 % for the  $c(\frac{1}{2}b)$  parameters. The latter percentage slightly exceeds the maximum value given by Royer, and possibly this is the reason why such cases are not frequent in nature.

From fig. 4 it is clear that epitaxis could easily be realized because of like dimensions and similarity of the structures of the two minerals. Attention should also be paid to the mainly covalent bonds in the structures of the two minerals.

The main difference that could be drawn between this case of epitaxis and the ones so far described in the literature concerns the manner of formation of both minerals. While most of the known cases of epitaxis are due to exsolution, simultaneous growth, or later overgrowth, in this case it accompanies replacement. The crack in the quartz crystal represents in fact a crushed micro-zone, which is thickly interspersed with regularly oriented biotite lamellae, formed by solutions which had penetrated the crystal after its deformation and caused its partial replacement, thus cementing the broken pieces. So one could define *epitaxic metasomatism* as a process leading to partial or complete replacement of one mineral by another, the latter developing along preferred directions. As in the case described this process is provoked by lattice adjustments of mobilized ions from the host mineral and ions introduced by the metasomatizing solutions.

Other examples. Epitaxic replacement seems to be a fairly common process although little or no attention is being paid to it. The important processes of sericitization, chloritization, uralitization, pinitization, and others are in fact essentially epitaxic in character. Careful examination of thin sections of partially or completely altered plagioclases often

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FIG. 4. The structural scheme of quartz projected on (1010) and a superimposed net of the silicon atoms of the (001) planes of mica.

reveals regularly disseminated sericite lamellae. A. I. Ginsburg and S. I. Berkhin (1953) describe regular replacement of muscovite by polylithionite which D. P. Grigoriev (1955) rightly explains in terms of epitaxis. In granitized areas formation of muscovite at the expense of biotite is most probably on epitaxic lines, as are also certain cases of graphic granite, formed through replacement of microcline by silicarich solutions (Wahlstrom, 1939).

The significance of the epitaxic replacement would perhaps stand out more clearly when the degree of mobility of ions is considered in greater detail.

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