Observations of growth defects in berlinite by high-temperature X-ray topography. By A. ZARKA and B. CAPELLE, Laboratoire de Minéralogie–Cristallographie, Université P. et M. Curie, 4 place Jussieu, 75230 Paris CEDEX 05, France

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Abstract

Growth defects in a synthetic berlinite sample were studied at high temperature (300–500 K) by X-ray topography. Preliminary observations of contrast changes with temperature are reported.

During the past few years there has been a resurgence of interest in berlinite (AlPO₄₃) as a possible substitute for quartz in piezoelectric device applications. Berlinite has a similar structure (Schwarzenbach, 1966) to quartz at room temperature (RT) and at higher temperature exhibits the same type of polymorphism. In particular, AlPO₄₃ shows an α–β transition occurring at 853 K instead of 846 K for quartz (Van Tendeloo, Van Larduyt & Amelinckx, 1976; Bachheimer, Berge, Dolino, Saint-Gregoire & Zeyen, 1984). In evaluating berlinite as a replacement for quartz in high-frequency acoustic-wave devices, anomalies were observed in its propagation loss–frequency–temperature characteristics (Detaint, Poignant & Toudic, 1980; Steinberg, Roy, Estes, Chai & Morris, 1984). In an attempt to explain these electrical anomalies, we shall present in this paper some results concerning the defects in a berlinite sample observed by X-ray topography above RT.

Fig. 1. X-ray transmission topographs of the Y-cut berlinite sample obtained with a rotating-anode X-ray tube. Ilford L4 nuclear plate. 110 reflection, Mo Kα. (a) RT. The growth-defect contrast is essentially due to dislocations and growth bands barely visible. (b) 330.5 K. The contrast of growth band A is less pronounced. (c) 382.8 K. The contrast of growth band A vanishes while the intensity contrast increases in the B zone. (d) 322.5 K. Inversion of the contrast located in zones A and B in comparison with Fig. 1(c). (e) RT. The crystal shows a very similar contrast to the topograph in Fig. 1(a).
For this study a Y-cut plane about 0.7 mm thick was cut from a synthetic crystal grown in the Laboratoire de Chimie Minérale et Matériaux at Montpellier (France). The X-ray topographic furnace used was built in our laboratory and operates between RT and about 500 K with a precision of about ±0.3 K at the sample. Temperature gradients between the top and bottom of the sample may also be adjusted but in our case the gradient was minimized to less than 0.5 K. During the first cycle of temperature change (RT–383 K–RT) fifteen topographs were obtained at different temperatures. The 110 reflection was always used and the exposure time was 4 h on 100 μm L4 Ilford nuclear plates. During the second cycle two topographs at 396.2 K (Fig. 2a) and 423.5 K were obtained under the same conditions. A selection of all the topographs are presented in Figs. 1 and 2.

In the first RT topograph (Fig. 1a) the crystal exhibits a high density of defects. However, it is possible to distinguish various types of contrast that differ by their temperature behaviour.

Dislocation-related contrasts vary very little with temperature but they appear more strongly, especially after the second thermal cycle (Figs. 1a and 2b). On the contrary, some contrasts related to growth bands parallel to (211) show a significant evolution during cycles. This evolution is obvious at 382.8 K (Fig. 1c): the contrast of the growth band labelled A vanishes and the intensity in the area labelled B is clearly enhanced. On decreasing the temperature the sample shows contrast behaviour very similar to that observed during the temperature increase. We note in Fig. 1(d) (322.5 K) the reversal of contrast between areas A and B (in comparison with the contrast displayed in Fig. 1c). During the second cycle the topographs show the same phenomenon with contrast reversal between regions A and B. At the end of this cycle (Fig. 2b) the contrast of the growth band A is more accentuated.

A third type of contrast appears on the whole sample, its evolution is non-reversible during the cycles. This last type is typical of localized stresses and is strongly decreasing after two thermal cycles. The whole sample seems to present a more perfect aspect than at the start of the study. This effect is well known; the stresses were created during growth and could not be relaxed in the bulk, but they can do so during annealing (thermal cycles) of slices.

The variations of contrast for the growth bands can be interpreted in terms of localized variations of the deformation in the sample with temperature. These variations appear in the same temperature range as the anomalies observed in propagation loss–frequency–temperature characteristics. These anomalies have been attributed (Detaint et al., 1980; Steinberg et al., 1984) to water impurities in these hydrothermally grown crystals. This explanation is qualitatively valid and in good agreement with the changes of the deformation contrast observed during the thermal cycles. The contrast due to the water impurity content is observed in selected areas and is predominantly located at growth bands that correspond to the adsorption of water on some growing faces during the crystal growth. The contrast changes observed during the cycle are roughly similar on recycling the crystal.

A combination of selected growth conditions reducing the water content and heat treatment controlling the water distribution could be of great interest for improving the piezoelectric device applications of berlinite.

References


