Acta Cryst. (1965). **19**, 757

Etching of Rhombohedral Cleavages of Quartz

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(*Received* 18 *March* 1965)

Synthetic quartz crystals obtained from Bell Telephone Laboratories, U.S.A. were cleaved carefully along $(10\bar{1}1)$ rhombohedral faces. The etch pattern consists of (*a*) individual isolated pits and (*b*) randomly distributed micropits interfering with each other. The stratigraphical pattern usually observed on cleavages of natural crystals is completely missing. Correspondence in the etch pattern has been established on matched cleavage faces by etching (i) both simultaneously in the same etchant and (ii) one in one etchant and the other in a different etchant. If the cleavages are etched in different etchants the shapes of the pits produced differ. By etching a cleavage face successively for three different periods and by removing an etch pattern once produced by polishing the face and then re-etching it, it is shown that the individual isolated pits are produced at the sites of dislocations.

A stratigraphical etch pattern is obtained by etching rhombohedral cleavages of natural quartz crystals. A shift in the stratigraphical pattern across a cleavage step is observed. The implications are discussed.

Introduction

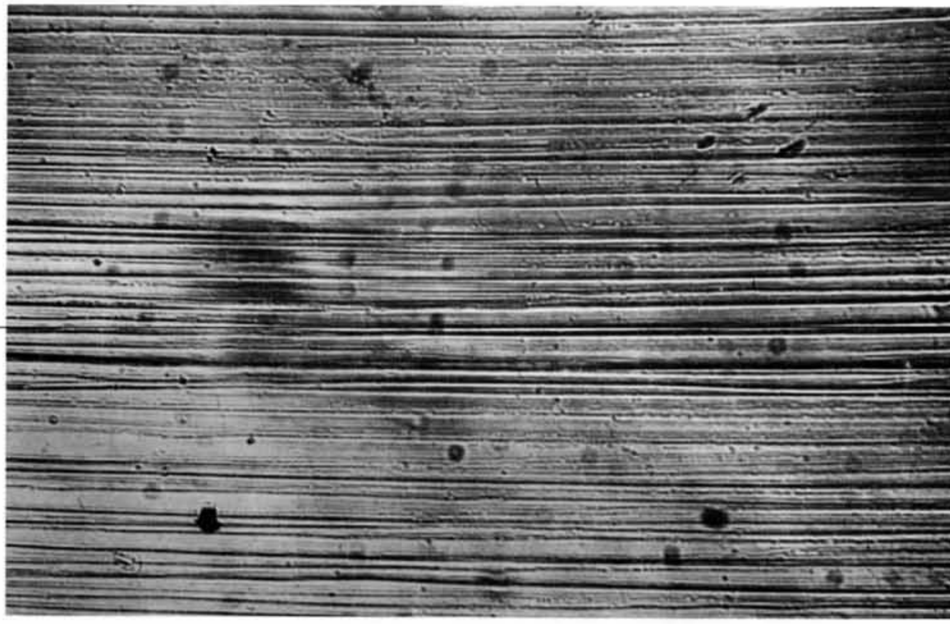
It is well known that studies of etch patterns on crystal cleavages give detailed information regarding the dislocation content and the history of growth of the crystals. Such investigations have been reported by Gilman & Johnston (1956) on lithium fluoride, Patel & Tolansky (1957) on diamond, Patel & Goswami (1962) on calcite and Patel & Ramanathan (1962) on mica. Generally for such investigations freshly cleaved faces are preferred to natural faces because cleavage faces are free from the usual complicated growth features. Because of the usual growth features on natural faces it is not convenient to make such investigations on non-cleavable or partially cleavable crystals. Tsinzerling & Mironova (1963) have reported that they have studied the etch patterns produced by hydrogen fluoride on the matched rhombohedral faces of quartz along which it is partially cleavable. We have studied in our laboratory the dislocation content and the history of growth by using etch methods of a number of cleavable mineral crystals such as diamond, mica, calcite, topaz, graphite, and fluorite. We thought it would be fruitful to extend similar studies first to partially cleavable crystals like quartz and then to non-cleavable mineral crystals. The present work takes its place in this series.

Experimental and observations

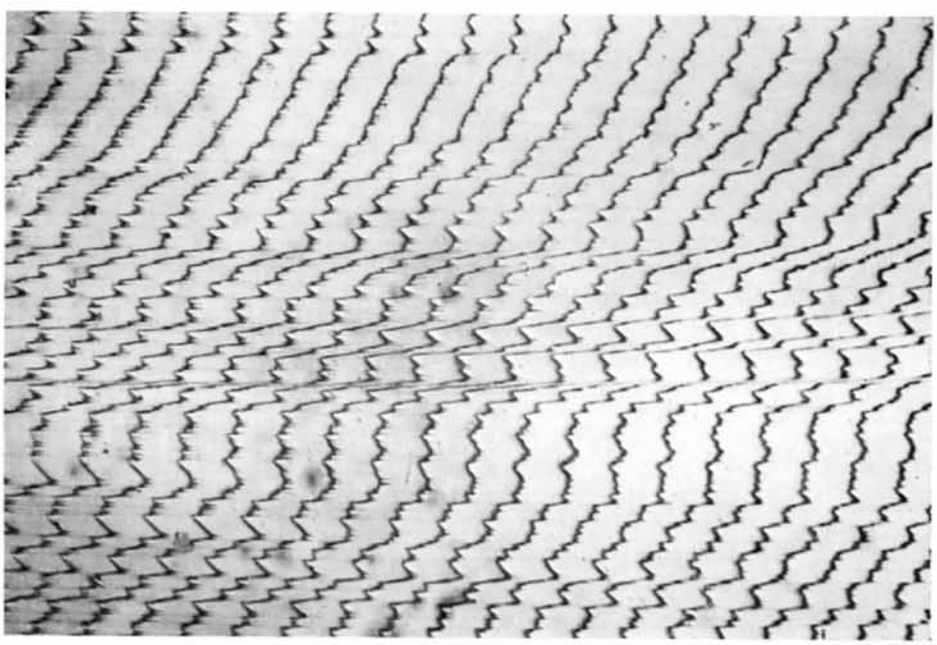
In order to produce better cleavage faces of quartz for experimental purposes, we selected synthetic quartz crystals, assuming that these crystals would have fewer flaws, since they are grown under controlled conditions and would therefore yield better cleavage faces than those obtained from natural quartz crystals. These crystals were supplied to us by Bell Telephone Laboratories, Incorporated (U.S.A.). In order to cleave the crystal, a kerf was made in it, and while a sharp steel blade was held in the kerf, a blow was given to the other end of the blade parallel to the rhombohedral faces. A cleavage fracture thus took place parallel to the rhombohedral faces producing two more or less matched faces. Thus Fig. 1(*a*) represents one of the cleavage faces and Fig. 1(*b*) represents the multiple beam interferogram taken on Fig. 1(*a*) revealing the topography of the cleavage face. The fringes indicate that the cleavage face is not quite flat as the crystal has only a partial cleavage. The sharp steps made by the cleavage lines on the cleavage face as seen on the perfectly cleavable crystals are also not observed on this face. In order to test whether the cleavage had taken place along the plane parallel to the rhombohedral face a natural rhombohedral face and the rhombohedral cleavage face were both etched simultaneously in sodium hydroxide solution (20 g NaOH + 3 ml distilled water) at 260°C for a few minutes. Thus Fig. 2(*a*) and 2(*b*) represent the etch patterns produced on the natural and the cleavage rhombohedral face respectively. It is clearly seen in these figures that the shape of the pits on both the faces is the same, thereby indicating that the cleavage is a rhombohedral cleavage. It may be noted that the etch pattern on the cleavage face appears more distinct than that on the natural face. It is conjectured, as usual, that the sites of etch pits may represent the sites of dislocations in the crystal. In order to test this, a cleavage face was successively etched for three different periods. Fig. 3(*a*), (*b*) and (*c*) represent the etch patterns on the same region produced after 15, 30 and 45 minutes respectively of etching at 260°C. It may be noted that:

(1) The shape of the etch pits resembles the shape of the pits on natural rhombohedral faces.

(2) Most of the pits are point bottomed; however, attention is drawn to the shallow pits nucleated along surface scratches.



(*b*)



(*a*)

Fig. 1. (*a*) Quartz cleavage parallel to the rhombohedral face (× 150). (*b*) Multiple beam interferogram taken on (*a*).

(a)

(b)

Fig. 2. Patterns produced by etching for a few minutes in sodium hydroxide on rhombohedral faces of quartz (× 150). (a) Natural face. (b) Cleavage face.

(a)

(b)

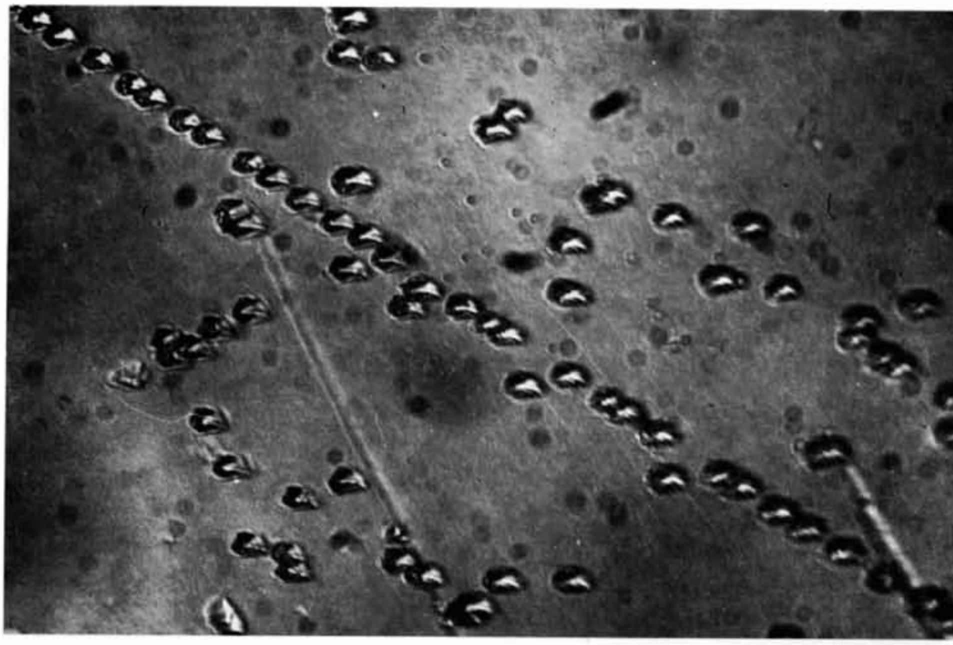
(c)

Fig. 3. Etch patterns produced by sodium hydroxide on the same cleavage face after (a) 15 min, (b) 30 min, (c) 45 min (×125).

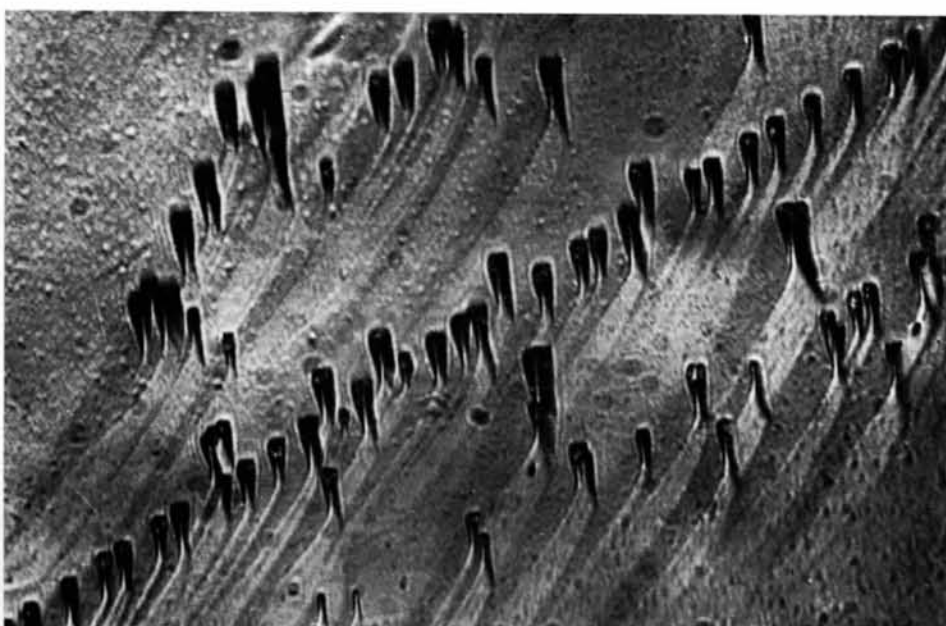
(a)

(b)

Fig. 4. Etch patterns produced in sodium hydroxide solution at 260°C on matched cleavage faces (× 175).



(a)



(b)

Fig. 5. Etch patterns on matched cleavage faces. The patterns were produced, in (a) by sodium hydroxide, in (b) by hydrogen fluoride (× 350).

Fig. 6. Rhombohedral cleavage face etched for 15 min in sodium hydroxide solution (× 175).

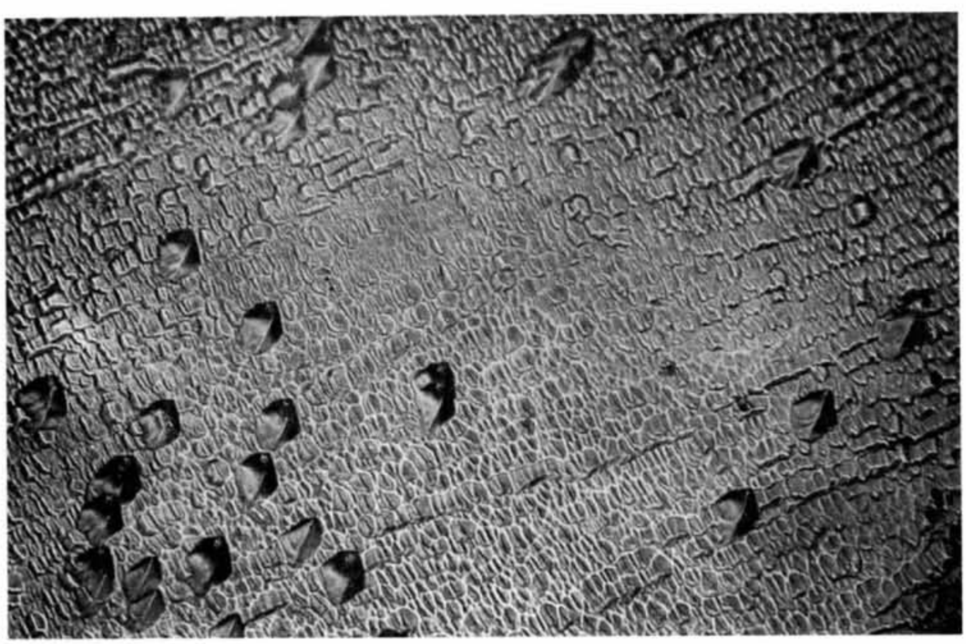


Fig. 7. Area shown in Fig. 6 after polishing and re-etching (× 175).

Fig. 8. Natural quartz crystal cleaved along a rhombohedral plane and etched in sodium hydroxide solution (× 250).

(3) Successive etching does not produce any new pits, but the original pits grow in extension and depth.

(4) The pits nucleated at surface scratches are completely washed out in the third etching indicating that the dislocations introduced at these sites are only superficial.

Etch pattern on matched cleavage faces

Fig. 4(*a*) and (*b*) represents the etch patterns produced on matched cleavage faces in sodium hydroxide solution at 260 °C. Just as in the case of perfectly cleavable crystals, it is interesting to find here also that there is an exactly one to one correspondence in the number and positioning of the etch pits on matched cleavage faces, when etched one in one etchant and the other in a different etchant. Thus (*a*) and (*b*) in Fig. 5 represent the etch patterns produced on matched faces by sodium hydroxide solution and hydrogen fluoride respectively. It is indeed interesting to find that there exists a one to one correspondence in the number and position of the etch pits on matched faces, even when they are etched in two different etchants. It may be pointed that the shapes of the pits on the two faces are different because they are formed by different etchants. These observations suggest that the pits reveal the sites termination of dislocation lines in the crystal faces. In order to know how deep the dislocation lines run in the body of the crystal, a rhombohedral cleavage was etched for 15 minutes in sodium hydroxide solution at 260 °C. Fig. 6 shows the etch pattern produced. This face was then heavily polished to remove a layer 20 microns thick, the etch pattern thus being completely removed. The polished face was then re-etched in the same etchant. Fig. 7 represents the etch pattern produced on the polished face in the same region as shown in Fig. 6. It may be noted that the etch pattern consists of somewhat bigger isolated pits and small crowded pits interfering with each other. Comparison of the etch patterns of Figs. 6 and 7 reveals that there is a one to one correspondence in the bigger pits; however, no correspondence exists in the small crowded pits. The small crowded pits in Fig. 7 are numerous in comparison those in Fig. 6, because in Fig. 7 they are nucleated at scratches developed during polishing. It is observed that the small crowded pits are washed out on further etching of the crystal.

It may be pointed out that in numbers of rhombohedral cleavages of synthetic quartz that were studied by etching, the stratigraphical etch pattern, as reported by Patel & Tolansky (1957) on diamond, Patel & Goswami (1962) on calcite, was completely absent. This may be because the conditions in which the synthetic quartz crystals grow do not change until the growth is completed. In order to find whether the stratigraphical etch pattern can be produced on rhombohedral cleavages of natural quartz crystals, a natural crystal was cleaved along the rhombohedral plane and etched in sodium hydroxide solution. Fig. 8 represents the etch pattern. The stratigraphical etch pattern is clearly seen. The thick black line running across the photograph is a large cleavage step. Careful observation reveals that the stratigraphical pattern is shifted on crossing the cleavage step. The shift in the pattern and the corresponding depth of the step were measured at different places along the cleavage step. These observations are given in Table 1.

Table 1. *Displacement of rectilinear etch patterns across cleavage steps*

Observation No.	Step height (h)	Shift in the pattern (d)	Magnification (m)	Angle calculated $\theta=\tan^{-1}$ (mh/d)
1	$2\cdot5\times10^{-4}$ cm	8×10^{-3} cm	33	45°0′
2	3	9	33	46 42
3	4	12	33	46 48
4	2·5	8	33	45 0
5	3	10	33	44 48

The inclination of the plane which gives rise to the stratigraphical pattern is calculated from the depth and the corresponding shift as reported by Patel & Goswami (1964) and is given in the last column. Now in quartz the angle that one rhombohedral face makes with the other rhombohedral face is 46° 16′, which is very near to the observed values of θ mentioned in the table. It may therefore be conjectured that the stratigraphy reveals the edges of the layers of one $(10\bar{1}1)$ face on the neighbouring $(01\bar{1}1)$ face.

Conclusions

That the number and position of the individual isolated pits do not change (i) on successive etching and (ii) even after polishing the face heavily, suggests that these pits may represent the sites of dislocations in the crystal. This is also confirmed by the matching of the individual isolated pits on matched faces when etched with different etchants. The stratigraphical pattern observed on rhombohedral cleavage faces of natural quartz crystal can be explained in the same manner as explained by Patel & Tolansky (1957) in the case of diamond. That the stratigraphical etch pattern appears on the rhombohedral cleavages of natural quartz crystal and is completely absent on synthetic quartz crystals suggests that the conditions under which the quartz grew in nature might not have remained constant during the entire period of its growth while these conditions are controlled in the case of quartz grown in the laboratory.

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