

A NEW METHOD FOR ORIENTING ELECTRON MICROSCOPE REPLICAS APPLIED TO TWINNED QUARTZ

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ABSTRACT

A technique which allows successive examination of specific areas of relatively rough surfaces by optical and then electron microscopy has been developed. The method utilizes the focusing action of a meniscus on thin carbon replicas floating on a liquid. The replica is positioned on a grid as the liquid is drawn down through a standard glass ball-and-socket joint. The method is an improvement over older ones since supporting films are not necessary, and therefore maximum resolution is realized. Preliminary studies with shadow-transfer replicas of cleavage surfaces of *d-l*-twinned quartz illustrate the method. Very fine striations (lower limit about 35 Å) were found within areas known to contain *d-l*-twins. The striations may be growth layers or possibly slip lines.

INTRODUCTION

In the course of studies on the morphology of cleavage surfaces of several particular types of quartz it was found desirable to relate specific areas of electron micrographs (up to about 250,000×) to structure on the surface itself visible to the naked eye. A new method for mounting replicas oriented on electron microscope grids was devised to accomplish this. We present here the method illustrated with some preliminary results on the structure of cleaved surfaces of Brazilian amethyst crystals.

Amethyst is characterized by fine *d-l*-twinning which extends throughout the crystal. At cleavage surfaces and along internal cracks ridges or lamella can be resolved with the unaided eye. Optical microscopy of such areas is usually sufficient to resolve some cross striations within the larger ridges. Figure 1 (taken with a Leitz "Ultrapak" at 50×) is an optical photomicrograph of the cleavage surface along the *c*-axis of "greened" amethyst from Brazil. From a petrographic microscopical examination it is known that each of the large ridges shown in Fig. 1 is either a *d*- or *l*-member of the alternating twin crystals. After heat bleaching, this amethyst appeared green in color. An optical examination indicated that a portion or perhaps all of the green color is due to another phase oriented between the laminar twins as discussed in an earlier paper (Cohen, 1956).

When we examined replicas of these surfaces in the electron microscope, striations as narrow as 100 Å or less were observed. The problem of relating the microstructure (100 Å striations) to the macrostructure

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(100μ ridges) was solved by use of the electron microscopical specimen mounting device.

Gay and Anderson (1954) have described a method for mounting serial sections on slotted types of electron microscope grids which requires a film of "Formvar" supported by a loop of wire. This works well with sections especially when high resolution is not required in subsequent elec-



FIG. 1. Photomicrograph of cleavage surface of "greened" amethyst approximately parallel to c axis. Magnification $50\times$.

tron micrographs. However, the supporting plastic film inevitably decreases the resolution attainable. We found it difficult to position *replicas* by picking them up from the surface of water with the "Formvar" covered loop. Various methods for examining identical areas of metallographic specimens by light microscopy and then electron microscopy have been described (McLauchlan, 1953). Since metallographic specimens are smooth and flat, these methods are based on placing an electron microscope grid in a given position on the film covered specimen and subsequently firmly attaching the replica to the grid by a relatively thick coat of plastic. Such methods cannot be used with rougher surfaces,

and high resolution is usually not attempted. Other methods which also protect the replica with thick plastic coatings require that the plastics be dissolved at the end of the process (Page, 1956). We have tried such techniques and found this last step damaged the replica beyond use more often than not. The surface tension of the solvents either breaks the delicate replica or the replica folds and curls while the plastic backing is being dissolved. A method described by Booker (1954) utilizes some of the principles we have used but his technique which was developed for wet stripped replicas requires a rather complicated assembly of apparatus and is useful only on rather smooth surfaces where orienting scratches can be made.

METHOD AND APPARATUS

Replicas of cleavage surfaces were prepared by a shadow-transfer technique (Hall, 1953). Platinum wire (0.005 in. diameter) was evaporated in various amounts from 0.025, 0.030 or 0.035 in. diameter tungsten wires bent in the form of a hairpin. A shadow angle of about 3 to 1 was usually used. The direction of shadowing was along the major axis of the ridges. In the same vacuum (10^{-4} to 10^{-5} mm. Hg) carbon was evaporated normal to the specimen surface in the manner of Bradley (1954), except that $\frac{1}{8}$ inch diameter spectroscopically pure carbon rods with one rod end squared off and the other sharpened to a fine point were used. The platinum and carbon film was immediately removed by slowly lowering the specimen at a 45° angle into 48% hydrofluoric acid. The replica usually floated free in one piece. The film was transferred to HCl (1:1 v/v) by picking it up on platinum gauze. After a time sufficient to dissolve any fluorides insoluble in HF or H_2O , the replica was transferred in the same manner to de-ionized water. We found the carbon films to be exceptionally sturdy and all but the very thinnest would withstand several such transfers.

The replica floating on water was then examined through low power ($10\times$ to $30\times$) binocular viewers or an "Ultrapak" microscope using reflected surface lighting. The general areas of interest were cut out of the large replica into pieces about $\frac{1}{8}$ inch diameter by simply touching the film with the edge of wire mesh. These isolated portions were then transferred to the mounting device.

Figure 2 is a diagram of the apparatus we have used for several years to mount replicas and thin sections on electron microscope grids so that macrostructures are oriented with respect to grid wires. Distilled water or water with a wetting agent fills the syringe, rubber bulb, and ball-and-socket joint section as shown. The glass tube is clamped to a ring stand; another clamp holds the syringe. A grid is placed on the 100 mesh plat-

form held in the vertical position shown. In addition to 200 mesh rectangular grids, slotted grids such as Sjöstrand and Anderson types* have been used. The isolated piece of replica is now transferred to the surface of the liquid near the top of the assembly by means of a small appropriately shaped piece of wire mesh held in clamped forceps. The liquid is slowly removed from the column by the syringe until the replica is either just at the constriction of the joint or slightly below. The action of the meniscus during the lowering of the liquid level is that of focusing the floating replica to the center of the column. In the absence of wetting

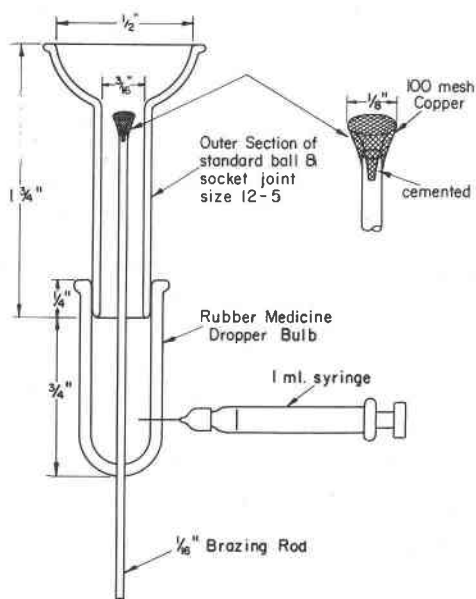


FIG. 2. Schematic diagram of replica orienting apparatus.

agents the apparatus must be thoroughly clean for focusing action to occur. The rod holding the grid can be moved vertically and horizontally and rotated while further liquid is removed through the syringe.

The operation is observed through a binocular microscope with a large working distance at $10\times$ to $30\times$. We have used Leitz binocular viewers with which many electron microscopes are now equipped. Surface lighting is used to illuminate the replica. Since the replicas are far too thin to allow any structure to be seen with transmitted light, no provision is made for transmission illumination. It was found helpful to apply black

* Obtained from E. F. Fullam, Inc., P. O. Box 444, Schenectady 1, N. Y.

paint to the inside and outside of the glass ball-and-socket joint; this reduces spurious reflections.

When the replica and grid are properly aligned so that macrostructure is oriented with the slots of the grid or with grid wires, the rod is gently but quickly pushed up. The replicas stick firmly to the grids and after a minute or two of drying the mounted specimen can be stored or examined immediately in the electron microscope. Cementing of the grid to the copper mesh platform is not necessary or desirable. If the piece of replica is cut large enough to span the gap at the ball-and-socket con-

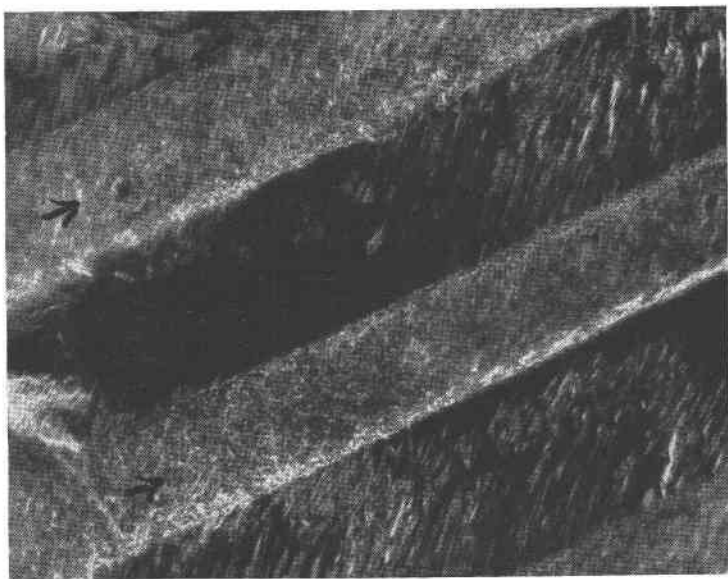


FIG. 3. Photomicrograph of carbon replica of "greened" amethyst cleavage surface mounted on a slotted electron microscope grid. The arrows point to two grid bars. The bands of striations were oriented with respect to the grid slots. Magnification 220 \times .

striction so that edges of the replica are supported by the glass when the liquid is withdrawn to slightly below this position, the orientation can be accomplished by use of the rod movements alone without withdrawing further amounts of water. Photographs through the "Ultrapak" or binocular viewer serve as a permanent record of the orientation, or sketches may be made for reference when the replica is examined in the electron microscope.

Figure 3 is an optical micrograph of a replica of a cleavage surface of "greened" amethyst mounted on a slotted grid so that the major ridges are roughly parallel to the slots. The replica dips between some portions

of the grid openings, and this prevents the entire area from being illuminated (with the oblique lighting of the Ultrapak), and being in focus. This is not, of course, a serious difficulty with the electron microscope.

With this method no plastic supporting films are necessary provided the replica is strong enough to support itself on the grid. Most carbon replicas have this strength; the very thinnest films can be mounted on 400 or 1000 mesh grids to give the carbon more support. We prefer to eliminate supporting plastic substrates so that all of the resolution inherent in the replica can be realized.

RESULTS AND DISCUSSION

Although the extremely complicated microstructure of amethyst quartz evinced by this study cannot as yet be fully explained, it seems

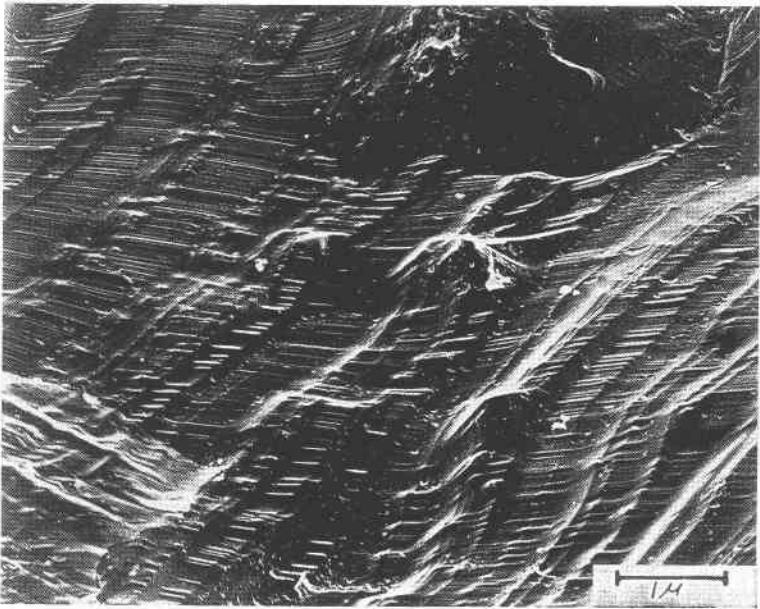


FIG. 4. Electron micrograph of a carbon replica of "greened" amethyst cleavage surface. A portion of the thin replica has been folded over itself at the lower left.

worth while to comment on several features of the electron micrographs of cleavage surfaces prepared as described above. Cleavage was usually effected by heating the quartz above its transition (α to β) temperature and quenching in air at room temperature. This procedure allows a good deal of plastic flow to take place and therefore may complicate the interpretation of the structure. Some samples were cleaved by percussion to see if changes in structure on cleavage surfaces would be obtained. No

important differences were noted. The "greened" amethyst is imported by the jewelry trade in the form of chips which are prepared by heating. Consequently the samples studied here have a prior history with the complications discussed above.

Figure 4 is an electron micrograph of a replica of the cleavage surface of "greened" amethyst showing very fine striations within bands which cross the figure diagonally. Many such striated bands were seen on surfaces cleaved approximately parallel to the c axis of the crystal. The

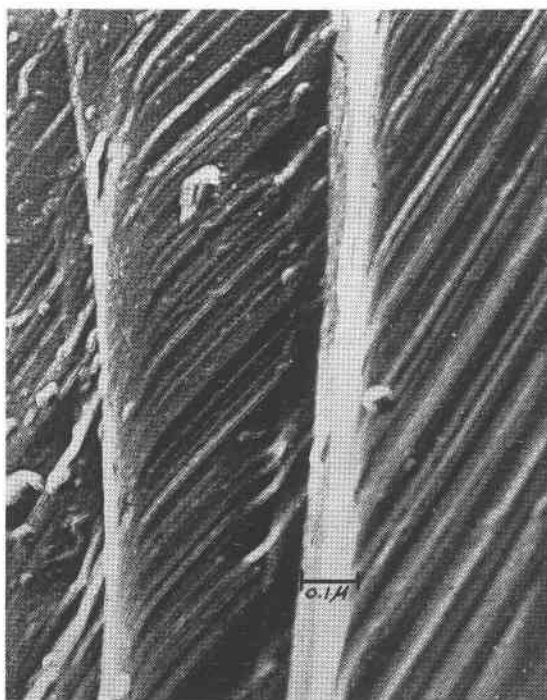


FIG. 5. Electron micrograph of a carbon replica of another area of the same surface shown in Fig. 4.

bands run in the same direction as do the major ridges visible in the light microscope such as can be seen in Fig. 1. The bands shown in Fig. 4 are too narrow to be resolved in the "Ultrapak" microscope but they parallel the larger ridges. Since the presence of optical twinning can be shown only by a petrographic microscopical examination (with inherent low resolution), it is not possible to state whether the bands in Fig. 4 are portions of the d - or the l -components of the lamellar twins.

Figure 5 is an enlargement of another area with similar bands and cross striations. Many areas show the cross striations separated by relatively

large distances. For example, some of the most prominent cross striations along a rather narrow band in the lower center of Fig. 4 are separated by as much as 1μ ; in other electron micrographs, distances between striations approach 10μ . In light micrographs such as Fig. 3 the cross striations (of very wide bands) that are just resolved, are most likely the more prominent striations observed in the electron micrographs. At several locations in Fig. 4 the striations cross over to adjacent bands but many areas such as in Fig. 5 give no evidence of the linking of adjacent bands. Figure 6 is a still greater enlargement of the upper right portion of Fig. 5. This electron micrograph shows examples of the very fine striations. Two ridges separated by about 35 \AA are indicated by a fork. The ridges appear to be made up of aligned roundish protuberances.

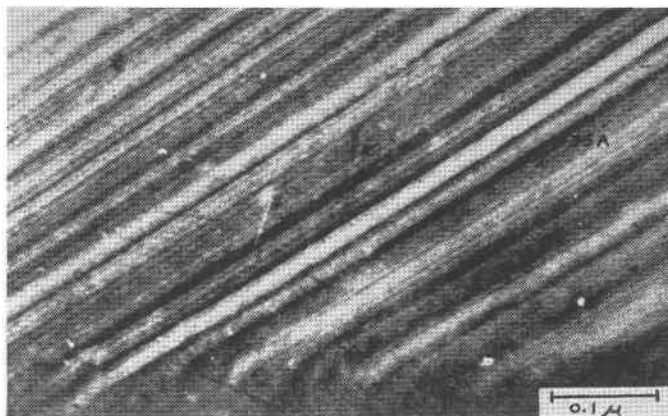


FIG. 6. Electron micrograph of a carbon replica of the upper right area of Fig. 5.

Negatives of several areas similar to Fig. 6 were scanned with a microdensitometer to see if there was any regularity of the striations. None was found.

There are several other features that merit attention. Regular arrays of pits are found at the apparent edges of several bands; however a few pits are also distributed at random over the surface. Conchoidal fractures as might be expected were found in the quartz. These fracture ridges did not have any fine cross striations. None of the various structures seen in these electron micrographs can be definitely assigned to the separate phase found by light microscopy.

A tentative explanation of these cleavage surface structures would be that the striations within the bands are growth layers in one of the twins comprising the amethyst. An alternate explanation is that these are slip lines produced by deformation. Many of the structures seen in Figs. 4,

5 and 6 and in other similar electron micrographs cannot as yet be interpreted. Since the replicas are from *cleavage* surfaces, they may show deformations resulting from cleavage forces. The interplay of these forces with the complex twinned structures of the crystal would be complex itself and difficult to recognize and discuss.

Undoubtedly it would be more fruitful to study natural faces of the "greened" amethyst but we have been unable to procure such specimens. However, further work is contemplated on the $10\bar{1}1$ faces of ordinary amethyst.

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