OCTAHEDRAL CRISTOBALITE WITH QUARTZ PARAMORPHS FROM ELLORA CAVES, HYDERABAD STATE, INDIA

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Abstract

The vesicles found in a basalt contain layered quartz inverted from cristobalite with fibers of mordenite projecting from the basalt through and above the quartz. Attached to the free mordenite fibers are well-formed twinned octahedrons of cristobalite, some of which have inverted to quartz. The crystals contain either one or the other mineral. The cristobalite crystals are uniaxial negative, $\epsilon = 1.4862$, $\omega = 1.4841$; $G = 2.382$. The optic axes of cristobalite parallel one of the octahedral diagonals; those of quartz have a scattered orientation. Mordenite fibers formed first in the vesicles. Later, or contemporaneous with mordenite, a layer of massive cristobalite deposited, along with crystals attached to the mordenite fibers. Inversion of the massive cristobalite and some of the crystals followed. The unusual occurrence of cristobalite perched on mordenite indicates that cristobalite formed as an unstable mineral with temperatures not far from 300° or 400°C.

In 1941, B. F. Buie collected specimens of a vesicular basalt from the Deccan volcanics at Ellora caves situated in the northeast corner of Hyderabad State, India. These specimens are characterized by the occurrence in the vesicles of a cotton-like mass of the zeolite mordenite together with well-twinned octahedrons of cristobalite, some of which have inverted to quartz.

The material was studied in the laboratory by Van Valkenburg and the paper was written by him, as Buie is on active duty with the U. S. Army. The work was done under the direction of Professor Esper S. Larsen, and we wish to express our sincere thanks to him and also to Professor Cornelius S. Hurlbut, Jr., and Mr. F. A. Gonyer of Harvard University and to Mr. John C. Rabbitt of the U. S. Geological Survey for many helpful suggestions.

Description

The vesicles in the specimens range in size from 1 to 10 cm. across. A typical one has a wall covered with a layer of quartz about 5 mm. thick. This layer has a waxy luster and shows a pitted contact with the basalt. Fibers of mordenite project from the basalt through and above the quartz giving the quartz surface a cotton-like appearance. Octahedrons of cristobalite, some of which have inverted to quartz, are perched on the mordenite fibers (Fig. 2). The general relations are shown in Fig. 1.
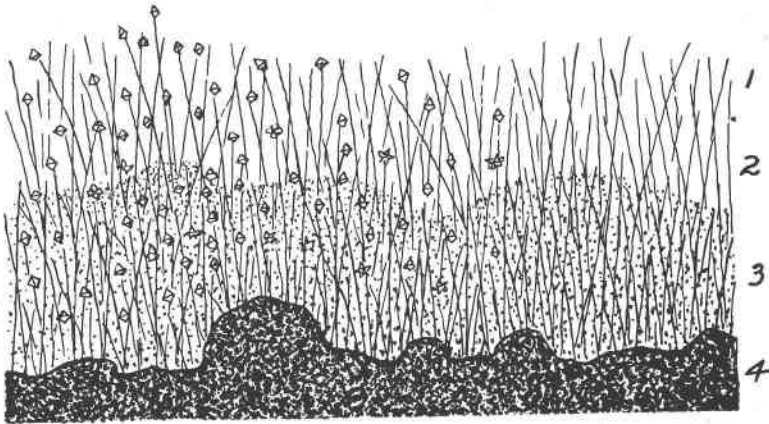
FIG. 1. An idealized cross section of a vesicle wall showing (1) crystals of cristobalite and quartz paramorphs perched on mordenite, (2) zone just above quartz layer where only quartz paramorphs occur, (3) layer of quartz inclosing crystals of quartz paramorphs perched on mordenite, (4) basalt with pitted contact. (Drawn by E. A. Schmitz.)

FIG. 2. Cristobalite and quartz perched on mordenite.

CRISTOBALITE AND INVERTED QUARTZ

Both quartz and cristobalite occur in twinned octahedrons that are identical macroscopically. In a paper following this Dr. Wolfe describes the crystallography of the two minerals. Crystals that occur near the outer mordenite ends are clear and have few inclusions; those near the

walls of the quartz are milky and contain abundant inclusions of mordenite. The layered quartz has inverted from cristobalite, as the crystals on the drusy surface are twinned octahedrons. A thin section shows octahedral quartz crystals attached to mordenite but encased in massive inverted quartz. Adjacent to the quartz surface, paramorphs of quartz after cristobalite are perched on the mordenite. In this part all the crystals have inverted and no cristobalite was seen; near the mordenite ends,
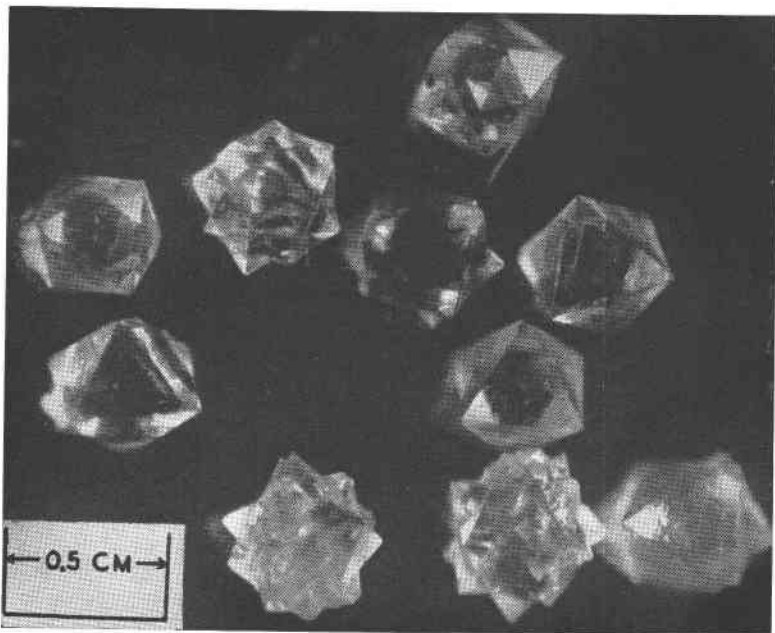

FIG. 3. Cristobalite octahedrons and twins shown in reflected light.

however, cristobalite and the quartz octahedrons occur together. One crystal may be cristobalite and the next may have inverted. Under the microscope it was seen that every crystal is composed entirely of one mineral or the other. No evidence was found to explain this unusual occurrence. It seems probable that inversion proceeded rapidly after a nucleus of quartz had inoculated the cristobalite. Inversion from cristobalite to quartz involves a volume change of 10 per cent and one might expect fracturing during inversion but careful examination of the quartz paramorphs shows no fractures.
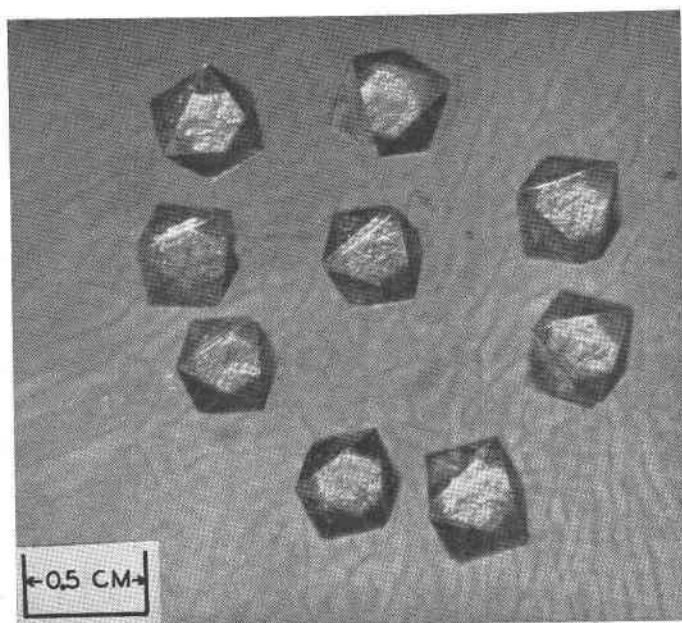
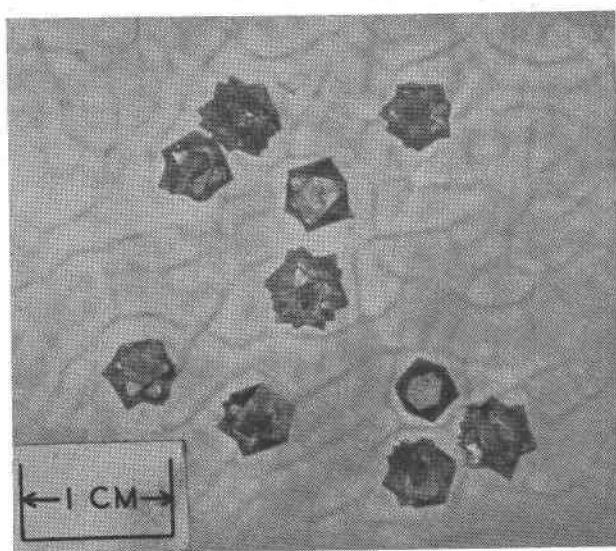FIG. 4. Cristobalite octahedrons in transmitted light showing etching on faces.

FIG. 5. Quartz and cristobalite octahedrons shown in transmitted light.

The cristobalite octahedrons and twins are about 0.5 mm. in diameter and are transparent to milky white, depending on the amount of mordenite inclusions present (Fig. 3). Etching on the octahedral faces of both quartz and cristobalite is common (Fig. 4). The crystals of cristobalite are optically uniaxial negative with $\epsilon = 1.4862$ and $\omega = 1.4841 \pm 0.001$. The specific gravity as determined in heavy liquids is 2.382 ± 0.003. The optic axis is parallel to one of the diagonals of the octahedron. A thin section
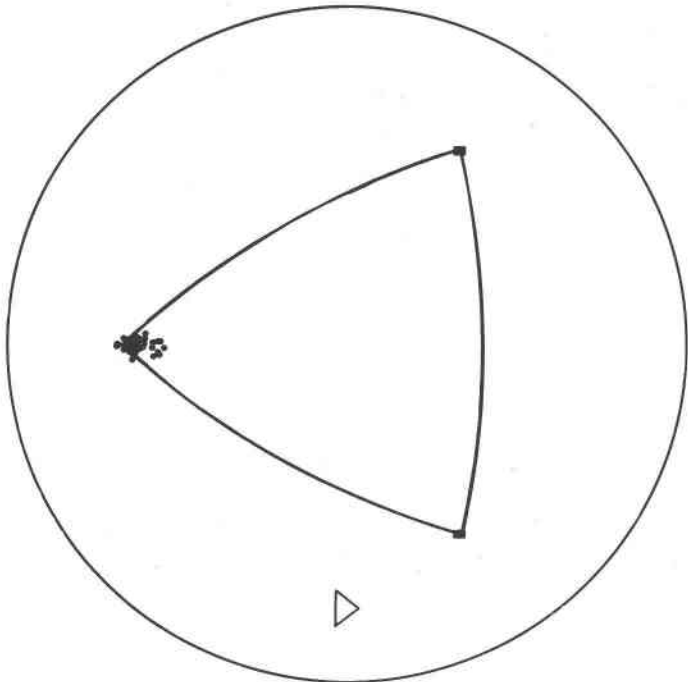

FIG. 6. A stereographic plot of 30 untwinned cristobalite crystals showing concentration of optic axes about an octahedral axis.

was made of 30 untwinned cristobalite crystals with each crystal lying on an octahedral face. In thin section the faces have the outline of a triangle. Using the Fedorov stage for each grain, the vertical plane including the optic axis was turned until its trace on the stage (in 0 position) was normal to the N-S cross hair. For reference position the edge making the smallest angle with the N-S cross hair and with the opposite corner of the triangle to the right was used. The optic axis was located in the usual way. The angle between the optic axis and an octahedral edge was found to average 53°. Figure 6 shows the stereographic plot of the optic axes

with respect to the crystal axes. The squares represent the octahedral axes. One of the octahedral axes was chosen to show the concentration of the optic axes as any one of the octahedral axes could have been used as a reference to show the concentration. The triangle at the bottom of the diagram shows the orientation of the face with respect to the vertical cross hair.

X-ray powder photographs using $Cu/Ni(CuK\alpha = 1.539$ Å) radiation were taken of known critobalite from the Miocene lavas of the San Juan Mountains of Colorado and of the crystals described in this paper. The two photographs are identical and their "d" spacings are given below. For comparison the "d" spacings of an alpha cristobalite (1) are given.

Line	"d" 1	"d" 2	I 1	I 2
1	4.03	4.04	10	10
2	3.34	3.13		7
3	3.11	2.89	7	8
4	2.81	2.48	8	9
5	2.46	2.11	9	
6	2.10	2.02		
7	2.01	1.93		
8	1.92	1.87		
9	1.86	1.69		
10	1.681	1.61		
11	1.602	1.57		6
12	1.526	1.53		
13	1.483	1.494		
14	1.431	1.430		
15	1.415	1.400		
16	1.394	1.370		
17	1.359	1.339		
18	1.326	1.300		
19	1.293	1.273		
20	1.273	1.235		
21	1.198	1.203		
22	1.092	1.181		
23		1.095		

1. Ellora Caves and San Juan cristobalite.
2. Alpha cristobalite.
"d" = spacings in Å units, I = relative intensity.

Powder photographs were also taken of the quartz paramorphs and no cristobalite was seen to exist in the crystals. The crystals have the optical properties of quartz but retain the octahedral habit of cristobalite. Undulatory extinction is common in many of the crystals and their twins.

The optic axes of the quartz paramorphs have a scattered orientation with respect to the octahedral axes. Figure 7 shows an equal-area plot of about 100 optic axes measured on the Fedorov stage in the same way as were the cristobalite axes. Figure 8 shows a contoured diagram of Fig. 7
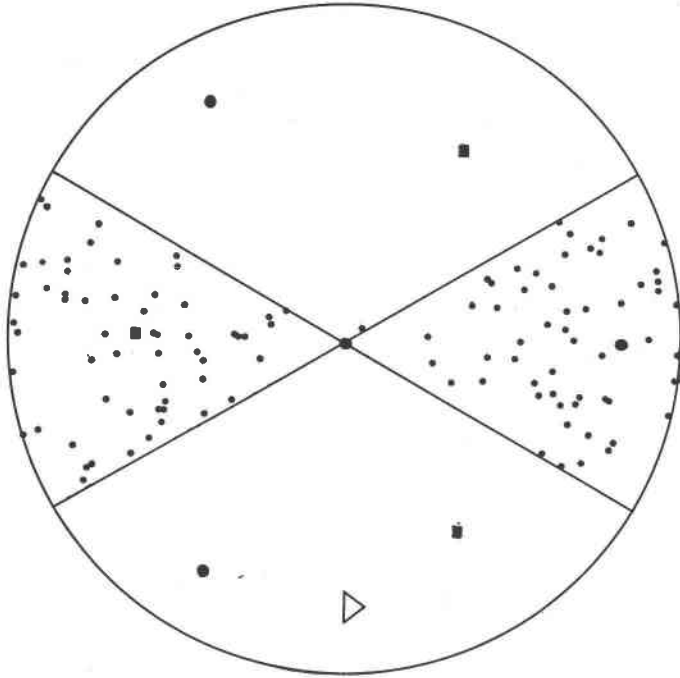

Fig. 7. An equal-area plot of about 100 optic axes of quartz paramorph lying on an octahedral face shows a scattered orientation of the axes. The squares represent octahedral axes and the circles the octahedral faces. The triangle indicates the orientation of the crystals with respect to the vertical cross hair of the microscope. The edge is vertical that gives the smallest extinction angle.

in which the segment has been rotated through two 60° arcs. The numbers represent relative concentrations of the optic axes; the squares are the octahedral axes and the circles the poles of the positive faces. The area with concentration 8 shows significant concentration of axial points, but the point location has no simple relation to the octahedral axes or faces. There is a suggestion of slight concentration in a zone midway between the center and the circumference of the diagram but not enough to say that a preferred orientation exists. On the inversion of alpha to beta cristobalite, an inversion that takes place rapidly with a small adjustment in cell structure, one of the octahedral axes becomes the axis of the

inverted crystal; however, on the inversion of beta cristobalite to quartz, an inversion which is commonly sluggish and takes place with a large volume change, the orientation of the quartz is not closely controlled by the old structure.
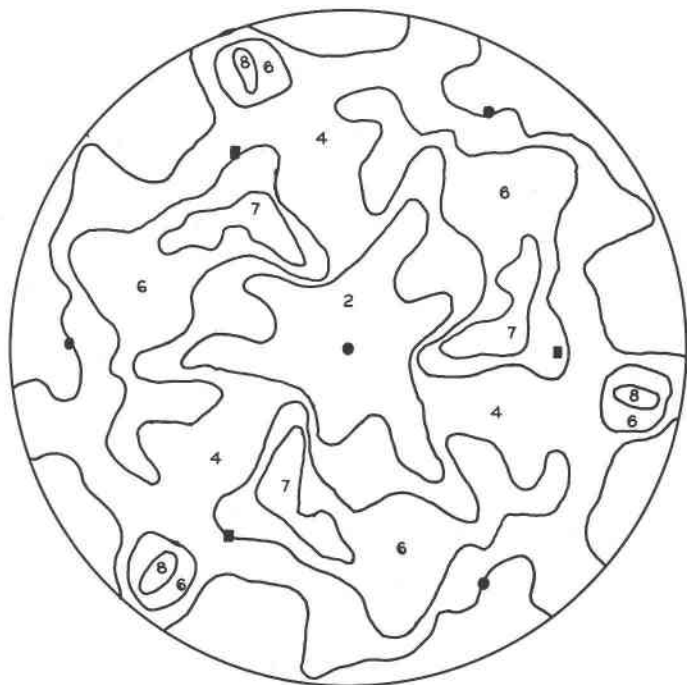

Fig. 8. A contoured diagram of Fig. 7 with the segment rotated through two 60° arcs, showing relative concentrations of optic axes.

Mordenite

Crystals of mordenite completely cover the outer surfaces of the vesicle. The needles of mordenite are white to transparent and have a maximum diameter of about 0.08 mm. and an average diameter of about 0.02 mm. Under the microscope the needles show faint birefringence, parallel extinction, and negative elongation. A thin amorphous film coats the needles, making it difficult to determine the indices. One index was found to be 1.481 ± 0.003. The amorphous film has an index lower than typical mordenite. A formula has been derived from the following chemical analysis by F. A. Gonyer on mordenite fibers which are slightly contaminated by the amorphous film. The unit cell contains 24 oxygen atoms (2).

	%	Mol. number	Oxygen	Bases	Atomic numbers
SiO_2	64.24	1.070	2.140	1.070	9.59
Al_2O_3	14.47	.142	.426	.284	2.52
Fe_2O_3	1.23	.008	.024	.016	0.14
CaO	4.49	.080	.080	.080	0.71
Na_2O	1.55	.027	.026	.052	0.46
H_2O	13.84	.769		1.538	14.01
Total	99.82		2.696		

Formula $= Ca_{0.71}Na_{0.46}Al_2(Al_{0.52}Si_{9.59})O_{24}+14.01H_2O.$

An x-ray powder picture of mordenite, using $Cu/Ni_9(CuK\alpha = 1.539$ Å) radiation shows the following "d" spacings in Å units, with a few line intensities:

Line	"d"	Relative Intensity
1	8.25	10
2	6.47	7
3	5.72	6
4	4.48	8
5	3.87	9
6	3.84	
7	3.43	
8	3.35	8
9	3.18	
10	2.85	
11	2.67	
12	2.55	
13	2.50	
14	2.03	
15	1.945	
16	1.86	
17	1.792	
18	1.652	
19	1.515	
20	1.435	

CONDITIONS OF FORMATION

This unique association of cristobalite perched on mordenite fibers indicates that temperatures were not high when the cristobalite formed. The mordenite has a fresh appearance and shows no alteration on contact with quartz and cristobalite. The basalt next to the quartz-mordenite filling is altered for only about two millimeters. Fenner (3) points out that cristobalite can form below its inversion point (1470°C.) and cites several laboratory experiments in support of this view. He says further that "any set of conditions which will bring together quantities of un-

grouped SiO_2 molecules in such a manner as to favor their rapid assemblage in definite groupings without giving time for perfect equilibrium to be reached (as in the reaction of vapors); or which will suddenly bring a system in which equilibrium prevails into new conditions, at the same time introducing obstacles to the establishment of a new equilibrium (as in the sudden chilling of a melt), will favor the deposition of unstable forms." Larsen (4) has shown that cristobalite found in gas cavities of the San Juan lavas of Colorado formed much below its inversion point and he believes the lavas were extruded at a temperature below 900°C.

In the material here described the cristobalite was deposited later than, or contemporaneous with, mordenite and both were deposited in the vesicles of a basalt. The cristobalite must have formed at a temperature well below its range of stability (above 1473°C.) and the association with mordenite indicates a temperature not far from 300° or 400°C.

PARAGENESIS

After the basalt solidified, fluids in the vesicles, some of which may have moved upward from the magma, altered the walls and deposited mordenite, followed and probably accompanied by cristobalite. Massive drusy cristobalite formed on the walls and on the mordenite needles after all or most of the mordenite had formed. Later, the massive cristobalite on the walls, those crystals near the walls, and part of the crystals near the outer ends of the mordenite, inverted to quartz. Alteration of the walls has furnished the silica and alumina for the mordenite, but most of the silica for the cristobalite must have been brought in as the amount of silica deposited is too great to have been derived by the small amount of alteration of the walls.

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