

Hydroxyl-herderite from Brazil

and a Guide to Species Nomenclature for the Herderite/Hydroxyl-herderite Series

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In recent years two Brazilian localities, *Virgem da Lapa* and the *Golconda* mine, have produced remarkable crystals of hydroxyl-herderite. These crystals display an interesting variety of crystallographic features. Although they have commonly been labeled "herderite," they are, like most other specimens so labeled, actually the hydroxyl member of the series: hydroxyl-herderite.

INTRODUCTION

Herderite, $\text{CaBe}(\text{PO}_4)\text{F}$, and *hydroxyl-herderite*, $\text{CaBe}(\text{PO}_4)(\text{OH})$, are the end-members of an isomorphous series. Herderite was first discovered at Ehrenfriedersdorf, Saxony, Germany (Haidinger, 1828), and hydroxyl-herderite was first described from Paris, Maine, as *hydroherderite* by Penfield (1894). Members of the series are typically found in pockets in pegmatites and are among the last minerals formed.

Hydroxyl-herderite is the more common of the two species and commonly forms by the alteration of beryllonite (Palache and Shannon, 1928), or beryl (Yatsevich, 1935; Perham, 1964; Ginzburg and Shatskaya, 1964).

The paucity of recent and reliable analyses, coupled with the discovery of new occurrences not yet described in the literature, prompted us to analyze a large number of specimens to ascertain their fluorine content and thereby also to establish the correct nomenclature for specimens from specific localities.

PHYSICAL PROPERTIES

Most herderite and hydroxyl-herderite is colorless, brown, gray, or light blue in color and have a Mohs hardness to 5 to $5\frac{1}{2}$. There does not appear to be any direct correlation between the OH/F ratio and the color, hardness or other physical properties of these minerals. Likewise, there does not appear to be any direct relationship between the OH/F ratio and the weak or frequently absent fluorescence of these minerals in ultraviolet radiation. Members of the series are best characterized by optical methods and composition and refractive indices have been published by Leavens, Dunn and Gaines (1978).

CHEMISTRY

Thirty-three specimens in the Smithsonian Collection, plus three specimens in the Yale University collections, were analyzed by electron microprobe. The analytical data have been published in Leavens *et al.* (1978).

The most remarkable conclusion derived from the analytical data is that *herderite* is a very rare mineral! Of the 36 samples analyzed, all 36 are *hydroxyl-herderite*. A list of the localities of the studied specimens is given in Table 1. The number in parentheses after the locality represents the number of analyses performed on material from that locality. Samples from the Foote Mineral Company spodumene mine, near Kings Mountain, North Carolina, and from the Dunton Gem mine, Newry, Maine, were also studied and are probably also hydroxyl-herderite. However, these specimens have non-stoichiometric compositions and are discussed in more detail in Leavens *et al.* (1978). Also given in Table 1 is the range of the percentage of the (OH) end member for crystals from each locality studied. All contain more than 50% of the (OH) end member and are therefore hydroxyl-herderite.

The only valid herderite known to us is a green gem in the Smithsonian collection, previously reported by Dunn and Wight (1976).

HERDERITE

Concerning the fluorine end-member, *herderite*, $\text{CaBe}(\text{PO}_4)\text{F}$, only two complete analyses are known for which fluorine is greater than hydroxyl. These are by Hidden and Mackintosh (1884), and by Genth (1884), both on material from Stoneham, Maine.

In light of the above observation that, of 36 specimens labeled herderite, all 36 are hydroxyl-herderite, the following comments on the previously cited papers are pertinent.

Hidden and Mackintosh (1884) determined 11.32% fluorine and stated only that "the fluorine was calculated from the excess of lime." They state that they have no doubt that the formula they calculate from their analysis represents the true composition of the mineral. Genth (1884), however, in summarizing his analytical data, states "Somewhat doubtful is the exact quantity of fluorine which it contains." Genth thought his fluorine content (8.93%) too low, and it is very likely that he was doubtful of the accuracy of the analytical procedures for fluorine determination which were in use in 1884.

TABLE 1. Localities for Hydroxyl-Herderite.

	Percentage of the (OH) End-Member
Virgem da Lapa, Minas Gerais, Brazil (3)*	54–63%
Golconda mine, Minas Gerais, Brazil (6)	71–90%
Waldstein, Fichtelgebirge, Bavaria, Germany (1)	53%
Epprechtstein, Fichtelgebirge, Bavaria, Germany (1)	59%
Blue Chihuahua mine, San Diego County, California (2)	57–59%
Poland, Maine (3)	67–80%
Stoneham, Maine (6)	61–72%
Fletcher mine, Groton, New Hampshire (2)	76–97%
Bennett mine, Buckfield, Maine (2)	71–74%
Greenwood, Maine (1)	66%
Auburn, Maine (4)	64–86%
Palermo mine, North Groton, New Hampshire (2)	96–98%
Paris, Maine (1)	91%
Topsham, Maine (1)	63%
Keyes Mica mine, Orange, New Hampshire (2)	98%

*The number in parentheses indicates number of analyses performed.

Further doubt of the accuracy of early fluorine determinations in herderite analyses was expressed by Penfield and Harper (1886), who gave a lengthy discussion of analytical techniques they used in herderite analysis.

Six samples from Stoneham, Maine, the only locality known to produce herderite ($F > OH$), were analysed in the present study. The fluorine content of the six samples varied from 3.29% to 4.52%. The OH—F midpoint in the series is at 5.82% F. The occurrence of herderite at Stoneham, Maine, is therefore suspect, if not doubtful. All six samples we analyzed have $OH > F$ and are therefore hydroxyl-herderite.

GENERAL OBSERVATIONS

Herderite and hydroxyl-herderite are minerals which have not, until recent times, attracted the attention of collectors interested in aesthetic specimens. Most specimens of these species are rather bland-looking, consisting of colorless to grey or white crystals on light colored matrices. The nature of the species is such that herderite crystals frequently have many vicinal faces and are usually poorly formed. The poor development of most crystals, coupled with the usual poor color contrast, and a paucity of excellent specimens, has not in past years endeared the species to many mineral collectors.

In recent years, however, two localities in Brazil have produced exquisite hydroxyl-herderite with well developed, sharp crystals, good color contrast, and a moderate abundance of specimens. These two "new" occurrences, at Virgem da Lapa and the Golconda mine, deserve detailed discussion.

MORPHOLOGICAL CRYSTALLOGRAPHY

The crystallography of hydroxyl-herderite presents special problems to the morphological crystallographer because, although monoclinic, the beta angle of hydroxyl-herderite is $90^\circ 06'$ and the resultant morphological development is therefore pseudo-orthorhombic. In addition, the crystals are frequently complexly twinned.

As Yatskevich (1935) pointed out in his detailed study of herderite, "herderite presents peculiar goniometric difficulties because of suprising variation in habit and similarity of angles in different zones." The zones to which he refers are [100] and [001]. For example, $(010) \wedge (011) = 37^\circ 57'$, whereas $(010) \wedge (120) = 38^\circ 24\frac{1}{2}'$ and $(010) \wedge (012) = 57^\circ 20'$ whereas $(010) \wedge (110) = 57^\circ 45\frac{1}{2}'$. Without optimum goni-

metric conditions, it would be impossible to distinguish between these two zones on the basis of these angles alone. Resolution of the problem is possible, however, if a third face occurs within either of these zones, namely (130) in the [001] zone or (021) in the [100] zone. When these intervening faces are present, the interfacial angles are very different in the two zones. For example, $(010) \wedge (130) = 27^\circ 51\frac{1}{2}'$ whereas $(010) \wedge (021) = 21^\circ 18'$. Furthermore, the common (*hkl*) fourth-order prisms {112}, {111}, $\{\bar{1}12\}$, and $\{\bar{1}11\}$ have completely different angles if the two zones are interchanged; and if these faces are present, as they are on the Brazilian crystals studied here, there is no ambiguity in the choice of orientation.

Several of the crystals studied here were too large for reflection goniometry and were measured by contact goniometry using a new technique, which is simple and more precise than the older *protractor and swiveled straight-edge* method. One can now acquire in hardware stores a device which measures the angle of deviation from horizontal by a direct reading on a dial. The instrument we used is called a *Level and Angle Finder* by the manufacturer (Pro Products Company, Rockford, Illinois). The circle is graduated in degrees and a gravity-controlled needle points directly to the angle of tilt of the instrument. It might be pointed out that the instrument is certainly the best inexpensive device on the market for demonstrating true and apparent *dips* in structural geology.

The largest hydroxyl-herderite crystal was easily measured, since the *m* {110} and *n* {111} forms are well-developed and quite large. The interfacial angles between these forms and the geometry of the crystal, relative to the orientation of Yatskevich (1935), could be rigorously determined. All of the angles could be measured to an agreement of $\pm 1^\circ$ of the angles in the angle table of Yatskevich (1935).

To assist the reader in visualizing the morphological development, the crystal drawings in Figures 1, 2a and 6a were generated by setting the *angle point* for the clinographic projection at a *phi* angle of 20° instead of 70° , which is conventional in most clinographic projections. In this manner, the reader's point-of-view is closer to the line-of-sight of the *b* axis.

Twinning in hydroxyl-herderite

In Friedel's (1911) treatment of twinning, he emphasized the basic influence of the geometry of the lattice upon the likelihood of twinning. In the case of hydroxyl-herderite, for which the unit cell is pseudo-orthorhombic, it is to be expected that twinning by pseudomerohedry would be extremely likely on the forms {100} and {001}. These are not planes of symmetry in the lattice but closely approach being so, since the beta angle of Yatskevich (1935) deviates only six minutes from rectangularity. The *a* and *c* axes of hydroxyl-herderite closely approach being two-fold axes of symmetry; therefore, twinning by pseudomerohedry by rotation of 180° about [100] or [001] is also extremely likely. There would be a rigorous difference in the appearance of these twins, ideally, but because of the very small deviation of the lattice from rectangularity in the case of the herderite series, no distinction can be made between twinning over these twin planes and twinning about these axes. Thus, out of the four cases of twinning by pseudomerohedry which are possible, only two can be considered: twinning on {001} and twinning on {100}. Definite evidence for twinning on {001} has been observed in crystals from the Golconda mine, and twinning on {100} occurs in all the large crystals examined.

The lattice of the herderite series is pseudohexagonal as well as pseudo-orthorhombic. The $(010) \wedge (110)$ of $57^\circ 45\frac{1}{2}'$ deviates from $60^\circ 00'$ by only $2^\circ 14\frac{1}{2}'$, and this *obliquity*, as Friedel would call it, is such that twinning by pseudomerohedry on this pseudohexagonal plane of symmetry (110) would be possible. A multiple pseudohexagonal lattice cell with (130) as a possible twin plane by pseudorecticular merohedry with an *obliquity* of $2^\circ 8\frac{1}{2}'$ and an index of *three* would also exist. Another pseudohexagonal multiple cell in the herderite lattice with (012) as the twin plane would have an *obliquity* of $2^\circ 40'$ and an index of *two*; producing twinning by pseudorecticular merohedry. Thus, twinning on {012} would also be expected.

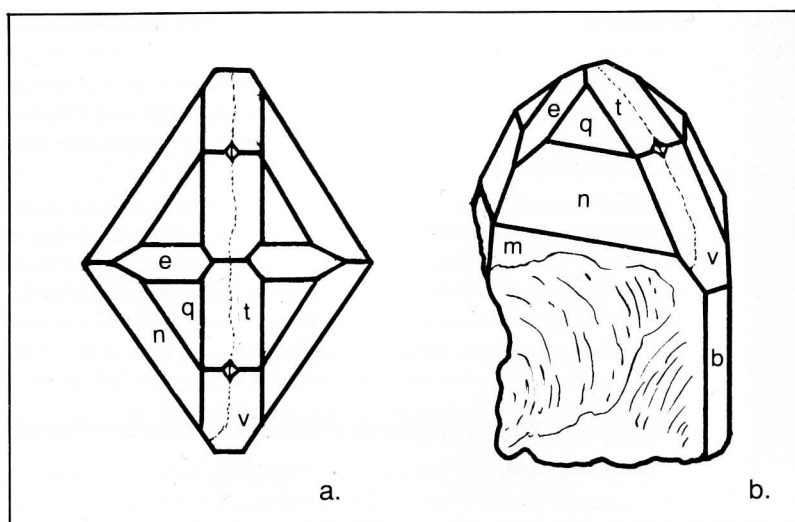


Figure 1. Sketches of a crystal of hydroxyl-herderite from Virgem da Lapa. The crystal, 15 mm in maximum dimension, is in the Smithsonian collection (NMNH # 143044). Form $m = \{110\}$, $n = \{111\}$, $e = \{302\}$, $t = \{012\}$, $v = \{011\}$.

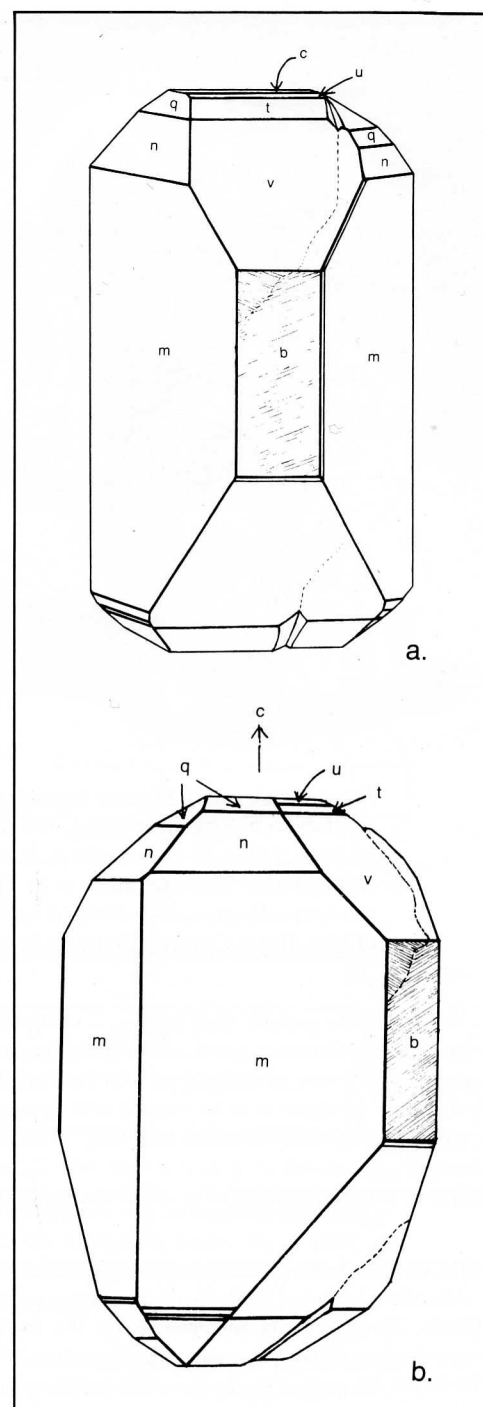
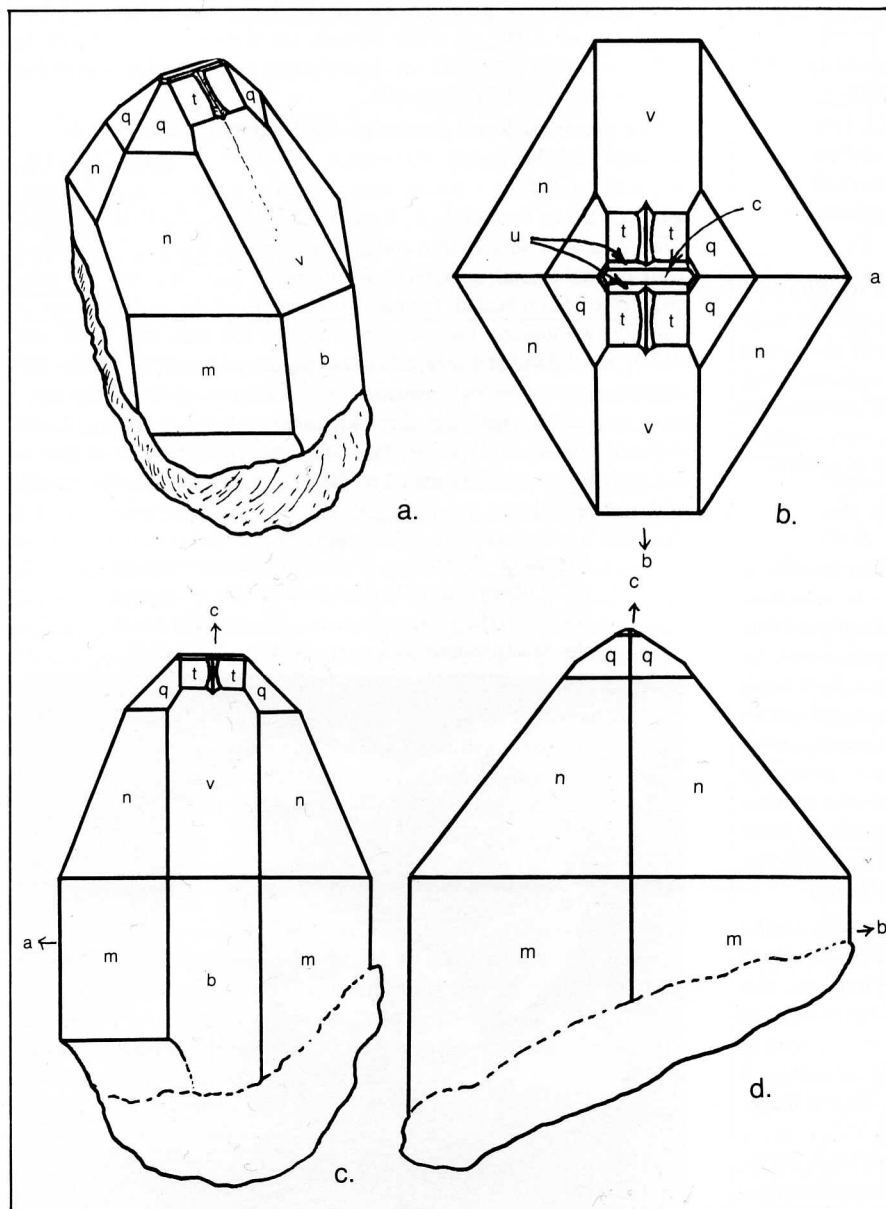


Figure 3. Sketches of a hydroxyl-herderite crystal from Virgem da Lapa. The crystal is in the collection of one of the authors (WEW), and measures 25 mm.

Figure 2. Sketches of a crystal of hydroxyl-herderite from Virgem da Lapa. The crystal, 44 x 55 x 78 mm, is in the Smithsonian collection (NMNH # 143032). Form $q = \{112\}$, $b = \{010\}$, $c = \{001\}$, $u = \{021\}$.

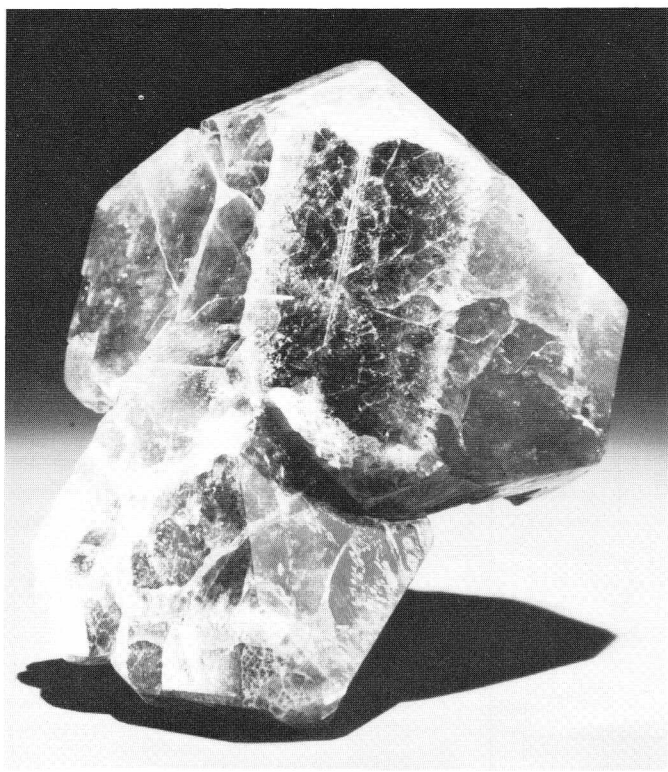


Figure 4. A group of large hydroxyl-herderite crystals from Virgem da Lapa. The largest crystal is 60 x 80 x 75 mm on the *a*, *b*, and *c* axes, respectively. The specimen is in the duPont mineral collection, University of Delaware (Gift of Mrs. David Craven). Photo by Dane Penland.

To complicate matters even further, a multiple cell with *b* as the unique axis is pseudotetragonal, and twinning by pseudoreticular merohedry on {102} with an obliquity of 0°31' and an index of *two* is very likely. Thus, herderite must be grouped with cryolite as a species which is apt to twin on many different twin laws. It is entirely probable that during crystal growth several, if not all, of these twin laws would be operative in the positioning of atoms on the growing crystal edifice.

VIRGEM DA LAPA, MINAS GERAIS, BRAZIL

Abundant hydroxyl-herderite began coming from this locality in 1975/6. Many crystals encountered on the market are euhedrons unattached to matrix but matrix specimens have also been available. The matrix for most of the hydroxyl-herderite crystals is an etched, tan to cream-colored microcline, although some crystals have been found perched on green tourmaline (elbaite) crystals. Other associated species include topaz and lepidolite. The microcline crystals are severely etched to a honeycomb texture. Crystals of hydroxyl-herderite on microcline are emplaced in random orientations suggestive of no epitaxial relationship, and tend to penetrate the microcline honeycomb in some instances. The latter feature suggests that the etching of the microcline was completed before crystallization of the hydroxyl-herderite.

The crystals have interestingly variable color. Most crystals exhibit an "alexandrite effect" in that they appear to be blue-green in daylight and lavender in incandescent light. One crystal (Fig. 3), however, was found to appear blue-green under either type of light. This crystal was among the first discovered, and also has an atypical development of faces in which *m* is less well-developed and the pseudo-pyramidal faces are larger. Some crystals appear blue in daylight, light violet in incandescent illumination, and have a peculiar yellow or gray "cap" at the end of the *c*-axis which is bordered by the forms {001} and {012}. Many crystals are colorless or pale straw-yellow and some are colorless

throughout. In hand specimen the habit resembles that of topaz, and the fracturing and luster resemble those of apatite.

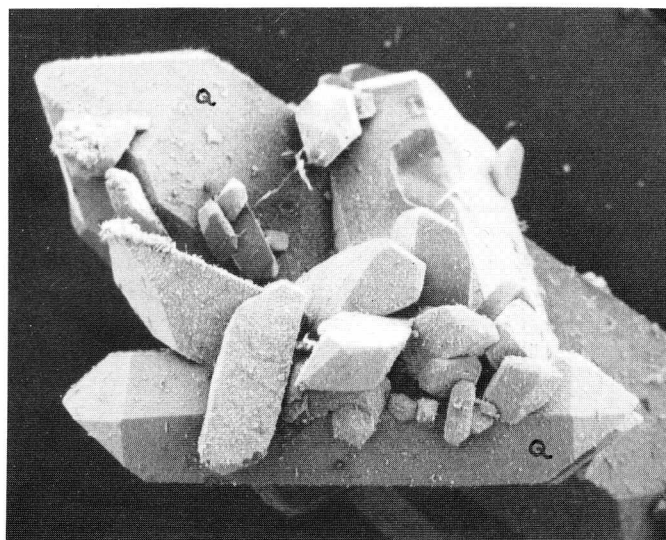
An analysis of hydroxyl-herderite from Virgem da Lapa is given in Table 2. Microprobe analyses of three additional crystals from Virgem da Lapa indicate that they have compositions between 54 and 63% of the hydroxyl-herderite end member.

According to Jack Lowell, of *Colorado Gem and Mineral Company*, during the summer of 1977 some 200 more hydroxyl-herderite specimens were found at Virgem da Lapa, including many on matrix. The largest crystal found in this recent mining operation was only 5 cm in length, compared with a maximum of 17 cm for the largest crystal found in the 1976 discovery. These new crystals are associated with albite instead of the buff-colored microcline of the 1976 discovery and are also notable in that there is less tourmaline associated with the hydroxyl-herderite, but much tantalite.

Morphologically, the Virgem da Lapa hydroxyl-herderite is remarkable and the crystals are undoubtedly the best ever found. Indeed, when compared with the poor quality of most herderite crystals found elsewhere, these are the most euhedral crystals known for the species. Virgem da Lapa hydroxyl-herderite is predominantly prismatic in habit, although many crystals have been found which are rather equant. All of the crystals from this locality which we have examined are twinned on {100}, although it must be mentioned that the re-entrant angle associated with the {100} twinning is always small and sometimes absent. We have observed no re-entrant indicative of twinning on {001}, but the luster of {010} on some crystals (as shown in Fig. 3 and 4) suggests that this law may also be operative in the development of the Virgem da Lapa twinned crystals.

The dominant forms present on the Virgem da Lapa crystals are, in order of decreasing dominance, *m* {110}, *v* {011}, *n* {111}, *b* {010}, *q* {112}, *t* {012}, and *c* {001}. Representative crystals are depicted in Figures 1, 2, 3 and 4. One crystal, NMNH #143022 (Fig. 2) is 44 x 58 x 78 mm and grew on a microcline base. The lower fifth of the crystal underwent fracturing at one time and has subsequently been rehealed, but the two segments of the crystal are not in parallel orientation. Two re-entrant angles are seen on *t* {012} and {0 $\bar{1}$ 2}, as shown in Figure 2a. The crystal surfaces within the re-entrants are significantly curved and are not rational planes. The junctures or sutures between the components of the twin are barely definable on *m* {01 $\bar{1}$ } and {011}. The positive end of the *a* axis is defined by curved striations on *q* {112}, as compared with the smooth surface of { $\bar{1}$ 12}.

Figure 5. Scanning electron photomicrograph (20x) of crystals of hydroxyl-herderite on quartz (Q) from the Golconda mine. The specimen is in the Smithsonian collection (NMNH # 121026).



A slightly different habit observed on another Virgem da Lapa crystal is shown in Figure 1. This crystal was not as large as the former one and measured only: $b = 15$ mm, $c = 14$ mm, $a = 10$ mm, and was susceptible to good goniometric measurement with the two-circle reflecting goniometer. Re-entrant angles could only be seen along the edges between $t \{012\}$ and $m \{011\}$, and between $t \{012\}$, and $m \{011\}$. Sutures between the two units of the twin on $a \{100\}$ could be seen down the full length of these faces, but no suture was observable on $b \{010\}$.

In addition, one of the authors (PJD) examined a crystal at the Tucson Gem and Mineral show in 1978 which had *five* distinct fourth-order prisms on a crystal 12 x 8 x 6 cm. However, it was not possible to measure the forms on this crystal.

TABLE 2. Partial Microprobe Analysis of Hydroxyl-herderite from Virgem da Lapa, Minas Gerais, Brazil

CaO	34.25%
P ₂ O ₅	44.14%
F	5.31%
H ₂ O*	3.22%
BeO**	15.45%
Total	102.37
Less O $\equiv$ F	2.34
Total	100.03%

NMNH #135016

*Determined by the Penfield method.

**Theoretical BeO by difference

Si, Al, Fe, Mg, K, S are absent.

Accuracy of data = $\pm 2\%$ relative.

THE GOLCONDA MINE, MINAS GERAIS, BRAZIL

This pegmatite has produced what are probably the most dramatically twinned hydroxyl-herderite crystals ever found. The crystals from this locality have been observed in two different types: colorless to very light brown microscopic crystals which appear to be untwinned (Fig. 5) and associated with quartz, and larger brown crystals which are superbly twinned (Fig. 6.). Crystal drawings and sketches of some of these crystals are shown in Figures 6 and 7. A photograph of one is shown in Figures 8 and 9. The larger brown twinned crystals are frequently white on the composite $b \{010\}$ face where the twin plane (100) is observed, but the rest of the crystal is usually an even brown color with a hue similar to that of typical Brazilian eosphorite. When composed of two individuals (Fig. 6c), the crystals have a triangular appearance when viewed along the b axis. When composed of four individuals, the crystals have a bow-tie appearance when viewed along the b axis, as is shown in the sketch of the "fourling" shown in Figure 6b.

Twinning on $\{100\}$, as seen in the Golconda crystals composed of two individuals (Fig. 6a), is quite common in herderite and is the dominant twinning in the previously described Virgem da Lapa crystals. However, crystals which show twinning on $\{100\}$ and $\{001\}$ are not common, and it is the existence of crystals with both twin laws operative which make these brown bow-tie hydroxyl-herderites so very distinctive. Without doubt, they are the most beautifully twinned hydroxyl-herderite crystals ever seen and described.

The existence of twinning on both $\{100\}$ and $\{001\}$ on the same crystal was first noted by Penfield (1894) on crystals from Stoneham, Maine. However, this type of double twinning in hydroxyl-herderite was discernible only by the well-trained eye of a morphological crystal-

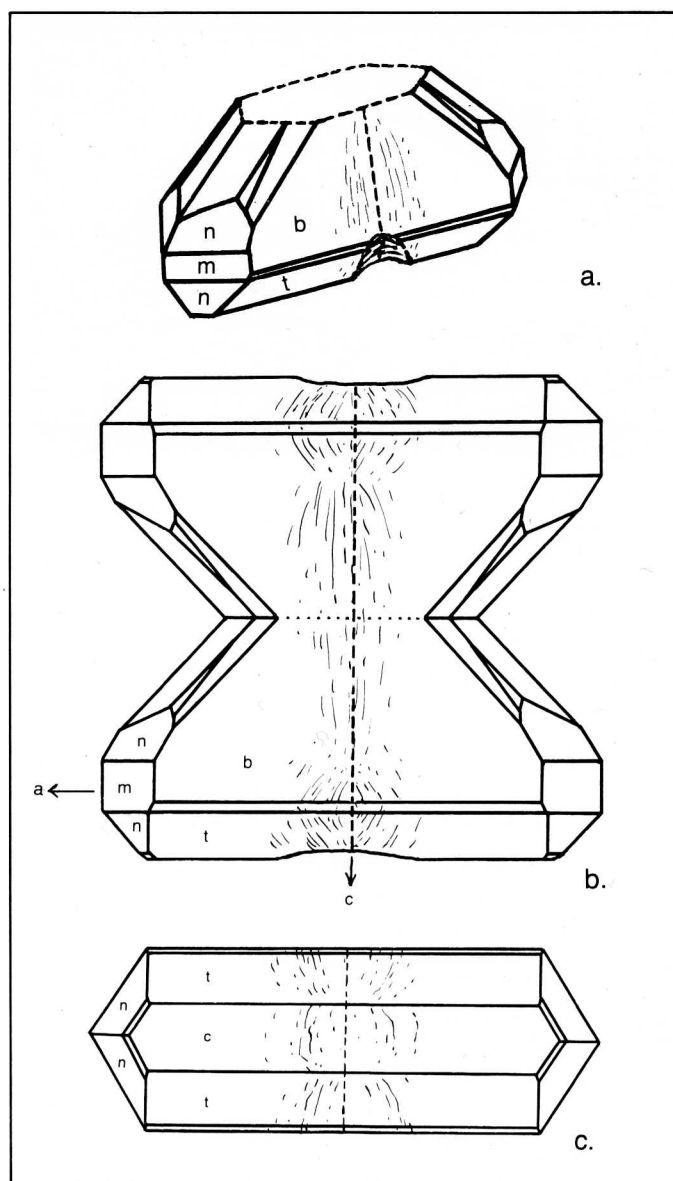
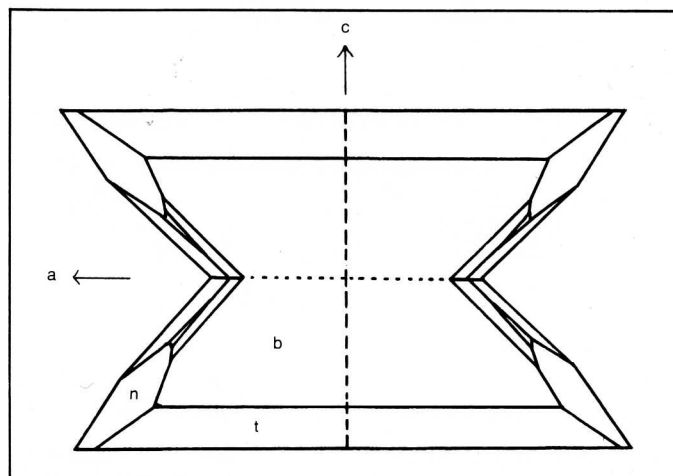


Figure 6. Sketches of hydroxyl-herderite crystals from the Golconda mine. (a) A twin on $\{100\}$; (b) a twin on $\{100\}$ and $\{001\}$; (c) the same crystal viewed down the c axis.

Figure 7. An idealized sketch of a crystal of hydroxyl-herderite from the Golconda mine, showing twinning on $\{100\}$ and on $\{001\}$. The viewpoint is looking down the b -axis.



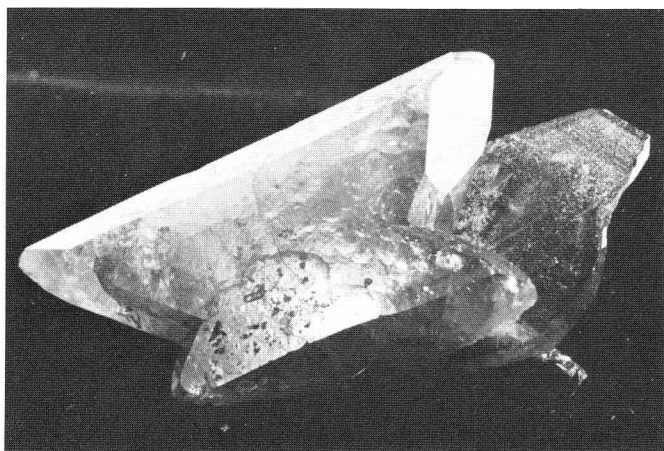


Figure 8. Photograph of a "fourling" twin of hydroxyl-herderite on smoky quartz from the Golconda mine. Smithsonian collection. (Photo by W.E.W)

lographer and it was not until the discovery of the Golconda mine crystals that this twinning could be easily observed.

Two crystals from the Golconda mine (NMNH #121031 and NMNH #121033, measuring 16 x 20 x 40 mm and 14 x 22 x 38 mm, respectively) were just within the size limits for precision measurement on the two-circle reflecting goniometer. They were measured with this technique and the results are rigorous. Figure 6a represents NMNH# 121033, which is twinned on {100}. Since the angular measurements of plus and minus $\{hkl\}$ equivalent forms are too close to be differentiated on large crystals such as these, we oriented the crystals to make the striated $q\{112\}$ form positive. The $\{112\}$ form is smooth and free of striations. In the region of the $q\{112\}$ form on these crystals, there is a series of vicinal faces in a zone with $\{010\}$ which produce a zone-plane



Figure 9. Photograph of a twinned crystal of hydroxyl-herderite on prismatic green tourmaline from Virgem da Lapa. Pala Properties International specimen. (Photo by Harold and Erica Van Pelt.)

of reflections which lies at right angles to the zone $\{001\}$ - $\{110\}$. Reflections from these somewhat curved surfaces do not fall into rational positions in the hydroxyl-herderite lattice, and the interfacial angles between them do not conform to interfacial angles within any zones in hydroxyl-herderite. Whether these vicinal faces formed in response to other twin laws than $\{100\}$ or $\{001\}$ could not be ascertained. A series of striations on $b\{010\}$ parallel to the intersection with $a\{100\}$, clearly betokens twinning on $a\{100\}$, with a major re-entrant angle on $b\{010\}$, and on $t\{012\}$, produced by twinned $m\{110\}$ planes. In the last stages of growth of this crystal, deposition was uniform, without twinning and the earlier twinning by the twin law $a\{100\}$ seen on the $b\{010\}$ and part of the $t\{012\}$ forms was almost completely covered. The late growth did not obscure the striations on the $\{0kl\}$ forms, however. These striations, like those on $b\{010\}$, were produced by the alternate growth of $m\{110\}$ and the $\{0kl\}$ faces of intersection, producing a herringbone pattern across the twin plane.

The paper by Yatsevich (1935) is recommended for the reader who wishes to delve further into the crystallography of the herderite series. Yatsevich's work involves a reorientation of the crystal setting used by Penfield (1894) and should be read prior to reading the older literature.

Six crystals having the morphological characteristics described above, all from the Golconda mine, were analyzed with an electron microprobe and found to contain from 71-90% of the hydroxyl-herderite end member.

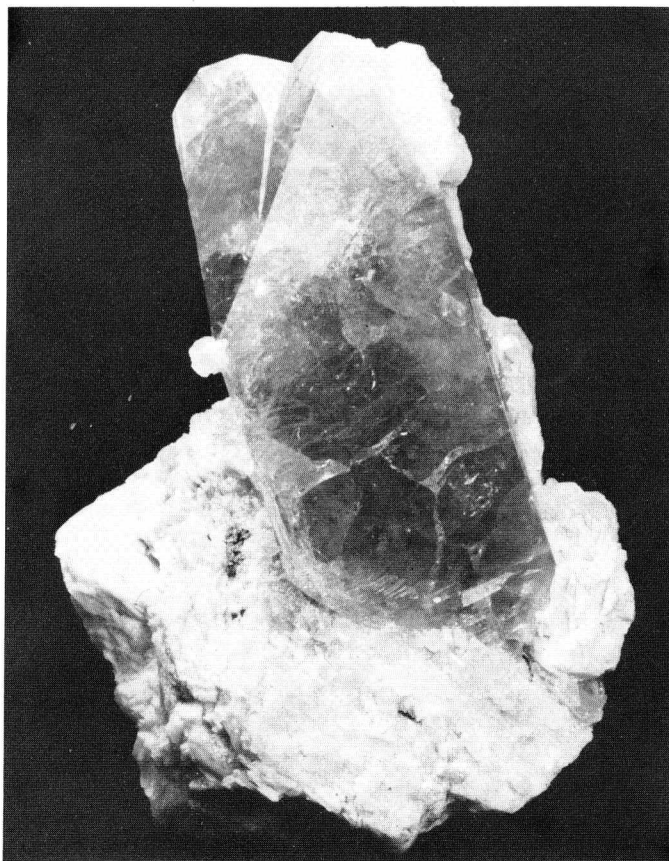


Figure 10. A hydroxyl-herderite crystal 9 cm tall, on matrix, from Virgem da Lapa. (Photo by W.E.W.)

SUMMARY

In summation, herderite is a very rare mineral. All specimens of purported herderite examined in this study are hydroxyl-herderite. The superbly twinned crystals from the Virgem da Lapa pegmatite and the Golconda mine, in Minas Gerais, Brazil, represent the best hydroxyl-herderite ever found and offer excellent examples of the two types of twins possible on herderite and hydroxyl-herderite.

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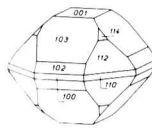
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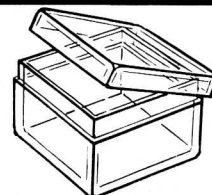
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