HYDROBASALUMINITE AND ALUMINITE IN CAVES OF THE GUADALUPE MOUNTAINS, NEW MEXICO

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Hydrobasaluminite, like alunite and natroalunite, has formed as a by-product of the H_2S-H_2SO_4 speleogenesis of Cottonwood Cave located in the Guadalupe Mountains of New Mexico. This mineral is found as the major component of white pockets in the dolostone bedrock where clay-rich seams containing kaolinite, dickite, and illite have altered during speleogenesis to hydrobasaluminite, amorphous silica, alunite, and hydrated halloysite (endellite). Gibbsite and amorphous silica are associated with the hydrobasaluminite in a small room of Cottonwood Cave. Opalline sediment on the floor of this room accumulated as the cave passage evolved. Jarosite, in trace amounts, occurs in association with the opalline sediment and most likely has the same origin as hydrobasaluminite and alunite. The hydrobasaluminite was found to be unstable at 25ºC and 50% RH, converting to basaluminite in a few hours. Basaluminite was not detected in the cave samples.

Aluminite has precipitated as a secondary mineral in the same small room where hydrobasaluminite occurs. It comprises a white to bluish-white, pasty to powdery moonmilk coating on the cave walls. The bedrock pockets containing hydrobasaluminite provide the ingredients from which aluminite moonmilk has formed. It appears that recent cave waters have removed alumina and sulfate from the bedrock pocket minerals and have deposited aluminite and gypsum along the cave wall. Gypsum, amorphous silica and sulfate-containing alumina gels are associated with the aluminite moonmilk.

We report the occurrence of hydrobasaluminite as a by-product of speleogenesis and aluminite as a secondary deposit (moonmilk) in Cottonwood Cave of the Guadalupe Mountains in New Mexico. A description of the depositional setting and mineralogy, and a brief discussion on the origin of these two hydrated aluminum sulfate minerals are presented.

Hydrobasaluminite, $Al_4SO_4(OH)_{10}\cdot(12\text{-}36)H_2O$, has been previously reported only in Alum Cave, Sicily, Italy (Forti, 1997). Its origin is related to the oxidation of fumarole gas (containing minor H_2S) and acid weathering of volcanic ash and tuff (Hill & Forti, 1997). Two occurrences of basaluminite, $Al_4SO_4(OH)_{10}\cdot 7H_2O$, have been reported in caves of Japan and South Africa (Hill & Forti, 1997). Basaluminite in the South African cave was reported as a product of evaporation of ore-derived solutions (Martini et al., 1997). In non-cave environments, basaluminite and hydrobasaluminite in association with allophane, goethite, halloysite, gypsum, aragonite, and calcite (rock-milk) have been reported as a coating in fractures, cement in narrow fissures, and as a matrix constituent of brecciated rock in south-central England (Hollingsworth & Bannister, 1950). Srebrodol'skiy (1969) detected basaluminite and gypsum in the oxidized zone of a former U.S.S.R. sulfur deposit. It was reported as coatings in fractures of dolostone. Frondel (1968) noted a similar occurrence of basaluminite with meta-aluminite in association with gypsum along the periphery of a brecciated zone containing pyrite in Utah. Tien (1969) reported hydrobasaluminite and basaluminite associated with iron oxides between Pennsylvanian marine shale and coal in Kansas. Sunderman and Beck (1969) and Ambers and Murray (1995) describe a similar occurrence of hydrobasaluminite between Pennsylvanian limestone and siliclastic materials in Indiana. Basaluminite was reported with gypsum in clay masses within Cretaceous rocks in Maryland (Mitchell, 1970), and as coatings and joint infillings in Eocene shales of the Polish Flysch Carpathians (Wieser, 1974).

The occurrence of aluminite, $Al_2(SO_4)(OH)_4\cdot 7H_2O$, in caves is apparently also rare. Hill and Forti (1997) note only one report by Martini et al. (1997) who describes the aluminite as white, chalky efflorescences on gypsum in an eastern Transvaal, South African cave as an evaporation product of ore-derived solutions. Reports of aluminite occurring in non-cave environments are moderately few as well. The mineral is found in bauxitic deposits and as fissure or joint fillings in brecciated zones. Deposits of aluminite were first noted around 1730 near Halle, Germany (Bassett & Goodwin, 1949). Gedeon (1954) identified aluminite associated with gypsum, calcite, boehmite, limonite, and quartz in a pyritic blue-mottled clay of a bauxite deposit in Hungary. Bárdossy and Sajgó (1968) also reported aluminite in a bauxite deposit in Hungary. Aluminite and basaluminite were reported with gibbsite, nord-

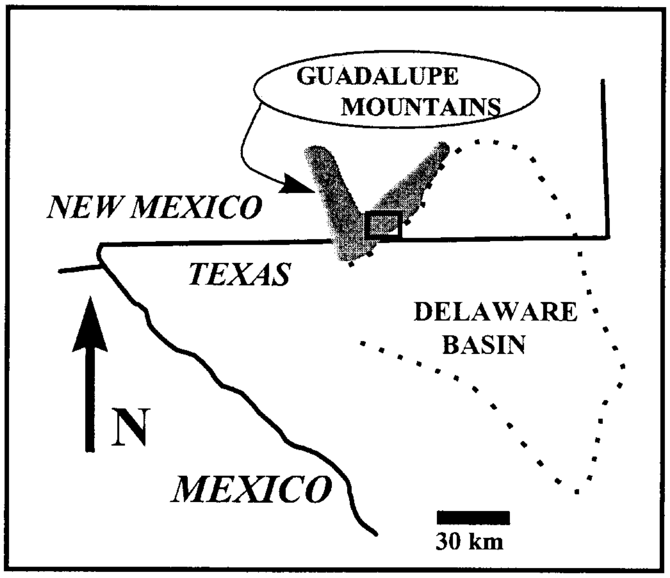

Figure 1. Study area in the Guadalupe Mountains is shown by an open rectangle.

strandite, and bayerite in sediments of solution pipes within Tertiary clays and chalk in southeast England (Wilmot & Young, 1985). The depositional environments of the brecciated rocks, fissures, and joints are similar to the cave environment.

DEPOSITIONAL SETTING

Hydrobasaluminite and aluminite occur in a small room of Cottonwood Cave in the Guadalupe Mountains, New Mexico. The study area is shown in Figure 1. This cave is located in the Permian dolostones and limestones of the Capitan Reef Complex, the same group of carbonate rocks in which Carlsbad Cavern is located. Hydrobasaluminite is found in areas of altered bedrock referred to as bedrock pockets by Polyak and Güven (1996). It resembles globules or blebs with the consistency of toothpaste, or loosely to moderately indurated materials with the consistency of a cookie. Aluminite is found as a bright to brilliant white and bluish-white, pasty to powdery, finely crystalline deposit on the cave walls. Carbonate speleothems which are numerous in Guadalupe Mountain caves are lacking in this room. The floor contains a relatively large block of gypsum. Figure 2 shows a profile of the small room where hydrobasaluminite and aluminite are reported. This section of the cave is located stratigraphically within a clay-rich pisolitic dolostone of the Permian Seven Rivers Formation (Jagnow, 1977). A 10-cm thick bed of kaolinite located near the ceiling appears to be a source bed for the hydrobasaluminite. Temperature and relative humidity in this room were measured as 11ºC and 95% during the spring of 1994.

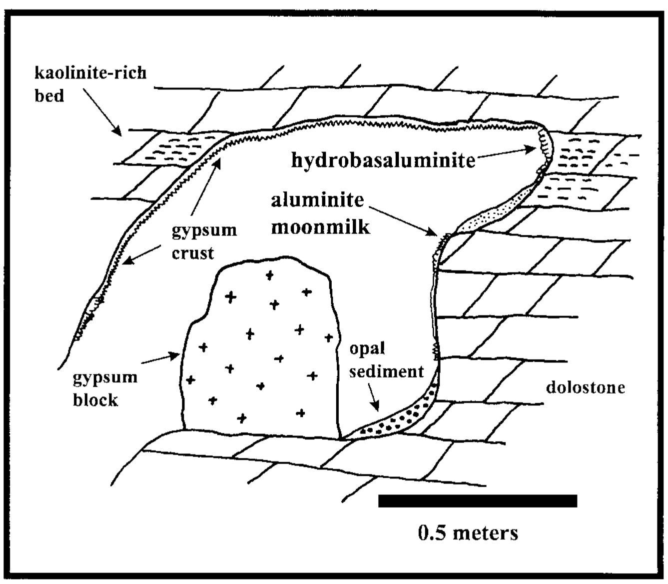

Figure 2. Profile of the small room in Cottonwood Cave where hydrobasaluminite and aluminite occur.

METHODS

X-ray diffraction (XRD), electron microscopy, and energy dispersive spectroscopy (EDS) were used to identify all mineral phases. XRD of random powders and suspended mounts were performed using Cu-Kα radiation. Samples were kept moist until XRD analyses were performed. Moist hydrobasaluminite was packed into a 2-mm deep sample chamber and x-rayed immediately at 1°2θ per minute. The 2-mm thickness prevented rapid dehydration of the hydrobasaluminite. Aluminite moonmilk was examined scanning electron microscopy (SEM) on a Hitachi S-570 at Texas Tech University. Scanning transmission electron microscopy (STEM) and EDS were used to determine semiquantitative elemental compositions on a JEM-100CX analytical electron microscope at Texas Tech University. Transmission electron microscopy (TEM) on a JEM-1200EX at Sandia National Laboratory was used to determine crystal morphology, single crystal quality, and existence of other fine-grained components such as opal. Hydrobasaluminite and aluminite samples were prepared for TEM by suspending powders in deionized water and settling powders onto holey carbon, copper grids. Monoclinic unit-cell parameters reported by Clayton (1980) were used with the Appleman and Evans (1973) indexing and least-squares powder diffraction computer program revised for the PC by Benoit (1987) to index our XRD data. The optical binocular polarizing microscope was used for direct observations of the mineral morphologies.

RESULTS

Hydrobasaluminite occurs in bedrock pockets in dolostone. The mineralogy of the bedrock consists of dolomite, quartz, mica, illite, kaolinite, and dickite. The dolostone is locally rich

Table 1. List of X-ray diffraction data for hydrobasaluminite from Cottonwood Cave.

hkl	*dmeas* (nm)	*dcalc* (nm)	*Imeas*
001	1.259	1.266	100
110	0.808	0.808	4
-111	0.766	0.762	3
002	0.633	0.633	11
-202	0.5920	0.5924	13
-112	0.5623	0.5630	9
012	0.5328	0.5342	3
201	0.5265	0.5272	15
020	0.5002	0.4981	1
120	0.4705	0.4686	24
112	0.4520	0.4516	3
121	0.4218	0.4220	16
-113	0.4152	0.4139	2
202	0.3969	0.3970	19
013	0.3893	0.3885	5
212	0.3690	0.3688	44
-411	0.3477	0.3474	8
-322	0.3409	0.3404	3
130	0.3226	0.3229	19
302	0.3162	0.3187	1
-323	0.3094	0.3095	7
131	0.3062	0.3064	8
312	0.3040	0.3036	2
-230	0.3004	0.2992	1
-422	0.2954	0.2957	14
-511	0.2817	0.2820	15
-513	0.2753	0.2752	1
not indexed	0.2734		5
223	0.2640	0.2638	2
232;-333	0.2544	0.2547	25
-613	0.2385	0.2379	21
240	0.2345	0.2343	9
403	0.2276	0.2279	2
610	0.2242	0.2241	7
340;-621	0.2187	0.2190	7
242	0.2114	0.2110	8
620	0.2090	0.2088	5
432	0.2064	0.2065	12
-711	0.2028	0.2028	18
621	0.1953	0.1954	17
250	0.1914	0.1914	1
612	0.1905	0.1902	2
433;-543	0.1878	0.1879	4
-813; 720	0.1833	0.1833	5
-253; 343	0.1816	0.1817	13
252;-353	0.1780	0.1781	14
153;-823	0.1749	0.1750	1
721	0.1729	0.1729	16
-821	0.1696	0.1696	1
253	0.1677	0.1678	2
542	0.1664	0.1664	2
-902	0.1640	0.1640	1
641;-743	0.1616	0.1616	22
-741; 353	0.1594	0.1593	1
-831; 261	0.1585	0.1585	1
-362	0.1564	0.1564	5
732	0.1518	0.1518	3
-460;-843	0.1494	0.1496	11
362	0.1472	0.1473	6
-752, 651	0.1454	0.1454	7
911	0.1442	0.1442	5

Note: the unit-cell parameters for hydrobasaluminite are: a=1.4930(5)nm, b=0.9963(3)nm, c=1.3695(11)nm, β=112.43(3)º, **V**=1.882(1)nm^3, indexed and compared to the monoclinic unit-cell of Clayton (1980). The numbers in parentheses for the values represent the uncertainties of the last digits.

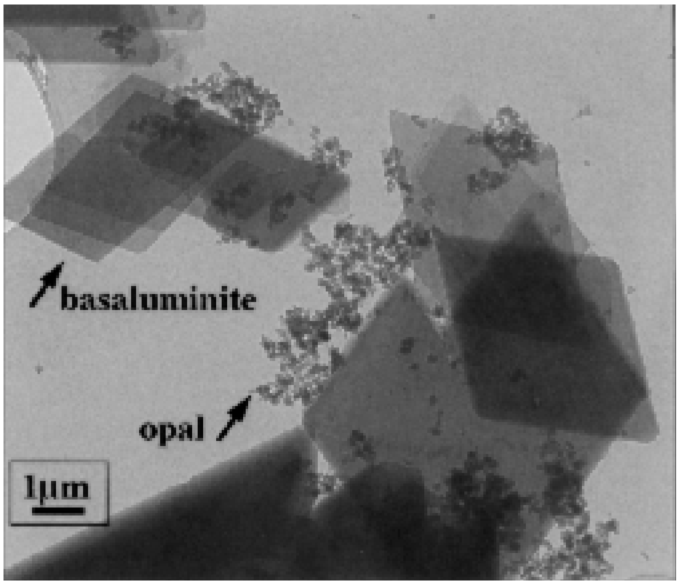

Figure 3. TEM micrograph showing rhomb-shaped platelets of hydrobasaluminite. The platelets have dehydrated to probably basaluminite due to the vacuum of the electron microscope.

in kaolinite in the small room in which hydrobasaluminite and aluminite are found. Weathered bedrock in this area contains pockets that are commonly blackened by manganese mineralization. In other caves of the Guadalupe Mountains, these areas of altered bedrock, usually associated with manganese mineralization, are referred to as bedrock pockets by Polyak and Güven (1996) and contain significant amounts of alunite and hydrated halloysite. In Cottonwood Cave, gibbsite and amorphous silica are abundant in the bedrock pockets containing hydrobasaluminite.

Powder diffraction data for hydrobasaluminite from Cottonwood Cave are listed in Table 1. Unit-cell parameters and powder data for hydrobasaluminite were compared with those of Clayton (1980), and the parameters were determined to be a=1.4930(5)nm, b=0.9963(3)nm, c=1.3695(11)nm, β=112.43(3)°, **V**=1.882(1)nm^3. We note, however, that hydrobasaluminite is insufficiently described in the literature, and further study is needed to find the correct structure and unit-cell for this mineral. The hydrobasaluminite was allowed to dehydrate at 25ºC and 50% RH for 30 hours. XRD results showed that the sample completely converted to basaluminite and the conversion was irreversible when the sample was remoistened. Basaluminite was not found in the cave samples.

Hydrobasaluminite (which dehydrates to basaluminite quickly under the vacuum of the TEM) interspersed with submicron, gel-like, amorphous silica was observed by TEM from powder samples. The rhomb-shaped platelets of hydrobasaluminite, shown in Figure 3, are characteristic of this mineral (Sunderman & Beck, 1969; Tien, 1969; Clayton, 1980; Ambers & Murray, 1995). The amorphous silica occurs as accumulations of submicron gel-like spheres (Fig. 4), or as

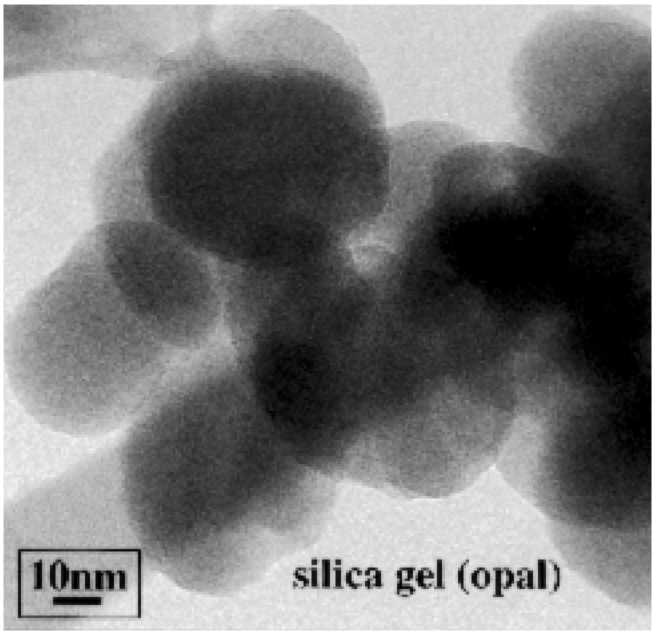

Figure 4. TEM micrograph showing the amorphous silica gel-like material that is associated with hydrobasaluminite.

deci-micron single spheres.

Aluminite occurs as a moonmilk on cave walls where bedrock is exposed. The walls of this small room, however, are mostly covered with a macrocrystalline coating of secondary gypsum. Associated with the aluminite and gypsum coatings are globules of dull white gypsum moonmilk. The floor consists of opal sediment mixed with an accumulation of fallen wall deposits. Faint blue-green to green nodules of opal also occur in the aluminite moonmilk.

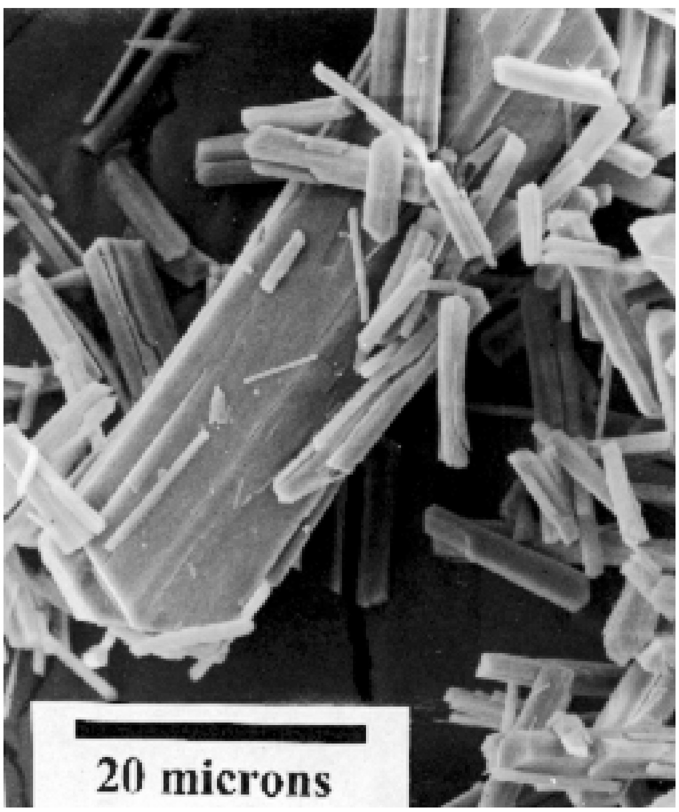

Figure 5. SEM micrograph of aluminite laths.

Unit-cell parameters and powder data for aluminite were compared with those of Sabelli and Ferroni (1978) and Farkas and Werner (1980), and the parameters were determined to be a=0.7419(4)nm, b=1.5791(7)nm, c=1.1650(4)nm, β=110.32(2)°, **V**=1.2783(6)nm^3.

TEM reveals that the aluminite moonmilk is made up of aluminite laths, gypsum prisms, submicron amorphous sulfate-containing alumina spheres (gels), and some amorphous silica spheres. Aluminite crystals are micron to decimicron-sized slender, rod-like to slightly flattened, euhedral laths (Fig. 5). Spheres of sulfate-containing amorphous alumina were observed as branching chains of gel-like spheres with an outer sheath as shown in Figure 6a. The outer sheath is probably an artifact caused by the tertiary butylamine which was used in sample preparation. When the gels dehydrated without the organic coating, they crystallized as fibrous radiating clusters as shown in Figure 6b. Gypsum crystals in the moonmilk are decimicron-sized, stubby, euhedral prisms.

Alunite, gibbsite, hydrated halloysite, and jarosite were identified by XRD, electron microscopy, and EDS. Alunite and hydrated halloysite in caves of the Guadalupe Mountains have been described by Polyak and Güven (1996). Black to dark brown, nearly pure gibbsite was located along a ledge directly below or adjacent to the hydrobasaluminite. Gibbsite was also found to occur in bedrock pockets below Wonderland in Cottonwood Cave, and at the top of the Four O'clock Staircase in Virgin Cave. Gibbsite crystals are decimicron-sized laths. The jarosite was found as small (<mm-sized) yellow pods in the floor opal. Jarosite crystals are micron-sized cube-like rhombs, similar in morphology to alunite.

DISCUSSION

Bedrock pockets in this area of Cottonwood Cave are identical to those of Carlsbad Cavern and Lechuguilla Cave described by Polyak and Güven (1996) which formed by the H_2S-H_2SO_4 speleogenesis mechanism. Pockets of alteration in Cottonwood Cave differ from those noted by Polyak and Güven (1996) by consisting mostly of hydrobasaluminite with only minor amounts of alunite and hydrated halloysite, rather than predominantly alunite and hydrated halloysite. The bedrock pockets are 5 to 50 cm in diameter and show a black (probably hydrous) manganese oxide that stains the pocket margins. The most obvious evidence of H_2S-H_2SO_4 speleogenesis in Cottonwood Cave are remnant "primary" gypsum blocks in the area of the bedrock pockets and other areas of the cave (Hill, 1987; Buck et al., 1994). These blocks are rarely observed in direct contact with the bedrock pockets. Hydrated halloysite (endellite) has also been reported as a H_2SO_4-indi-

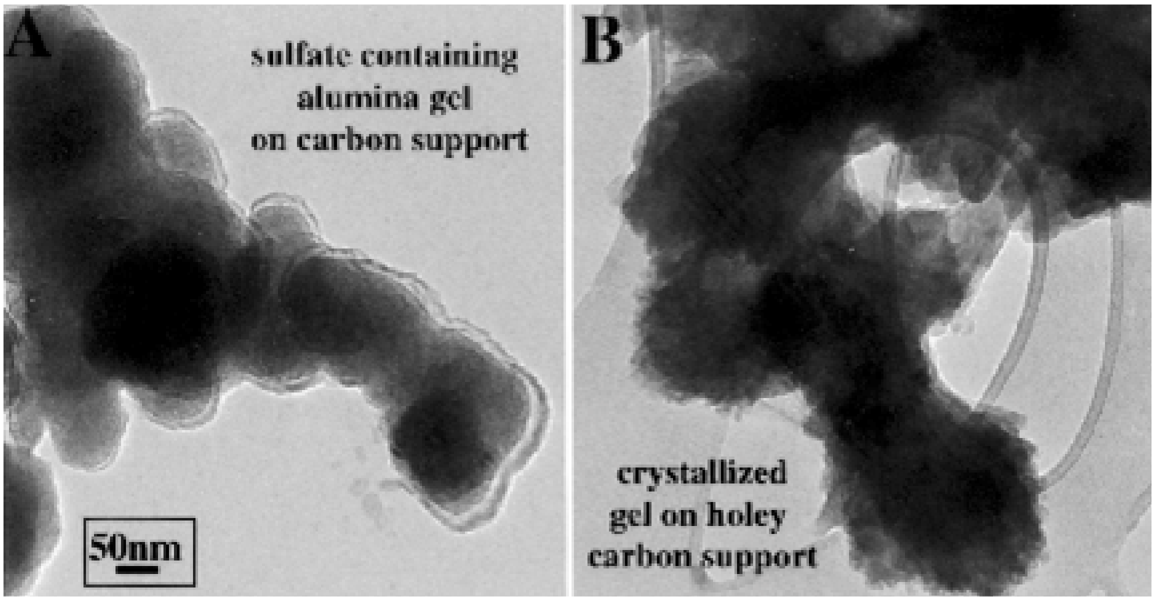

Figure 6. TEM micrograph showing sulfate-bearing alumina gel. (a) A sheath has formed on the alumina gel from tertiary butylamine which is used to disperse small particles during sample preparation. The tertiary butylamine apparently prevents further crystallization of the gel materials during preparation. (b) The alumina gel, without tertiary butylamine, crystallizes upon preparation.

cator mineral in Cottonwood and other caves of the Guadalupe Mountains (Hill, 1987). The bedrock pockets containing alunite, hydrated halloysite (Polyak & Güven, 1996), and hydrobasaluminite are additional evidence of H_2S-H_2SO_4 speleogenesis.

Hydrobasaluminite, like alunite, is an aluminum sulfate and commonly forms from the interaction of kaolinite with H_2SO_4 (Hollingsworth & Bannister, 1950; Adams & Hajek, 1978; Bassett & Goodwin, 1949; Ambers & Murray, 1995; Beecroft et al., 1995). The Cottonwood Cave hydrobasaluminite probably formed in the same way where H_2SO_4-bearing waters altered the kaolinite-rich bed in the dolostone bedrock by the reaction

$$H_2SO_4 + 2[Al_2Si_2O_5(OH)_4] + nH_20 \rightarrow$$
$$Al_4SO_4(OH)_{10} \cdot (n\text{-}4x)H_20 + 4[SiO_2 \cdot xH_20],$$

where n≈16-40 and x≈1-5. This reaction suggests that, other than hydrobasaluminite, silica should also be a by-product of the alteration process. Sparsity of K-bearing clay minerals such as illite and smectite favored the production of hydrobasaluminite over alunite as demonstrated experimentally by Adams and Hajek (1978).

Gibbsite and amorphous silica, which are both commonly associated with the hydrobasaluminite, probably have more complex origins. Gibbsite could have formed during H_2S-H_2SO_4 reactions; however, Adams and Hajek (1978) showed that gibbsite is favored by low SO_4/Al and high OH/Al ratios. During speleogenesis the SO_4/Al ratio was probably too high for production of gibbsite. It is possible that the gibbsite formed by the alteration of hydrobasaluminite during higher pH conditions (Ambers & Murray, 1995 and Beecroft et al., 1995). In Cottonwood Cave gibbsite was found close to the kaolinite bed. It was also identified in another area of Cottonwood Cave and in nearby Virgin Cave where it appears to be a product of altered bedrock pockets containing alunite and hydrated halloysite. Formation of gibbsite in this setting would therefore more likely be secondary from seepage and condensation cave waters.

Amorphous silica in Cottonwood Cave probably formed with the hydrobasaluminite during speleogenesis from excess silica produced by the reaction noted above. Amorphous silica spheres were observed with the hydrobasaluminite platelets under the optical and electron microscopes. Silica in the form of chert in Endless Cave and in another area of Cottonwood Cave probably formed in a similar way. In these other locations the chert is near occurrences of alunite and hydrated halloysite rather than hydrobasaluminite. The opal sediment found on the floor immediately below the aluminite moonmilk is indicative of the removal of silica from clays during the development of the cave passage.

The evolution of the cave passage where hydrobasaluminite occurs in Cottonwood Cave, offered in Figure 7, is based on mineralogy and locations of deposits. We propose that gypsum replacement of dolostone bedrock occurred initially along joints or zones of higher permeability. Ascending H_2SO_4-bearing water reacted with kaolinite-rich seams in the dolostone bedrock. Replacement of dolostone by gypsum probably occurred by a process similar to the subaqueous replacement process described by Buck et al. (1994). As replacement of the dolostone by gypsum progressed, so did conversion of kaolinite to hydrobasaluminite and amorphous silica. We envision there was little open cave passage when the gypsum replacement and clay alterations took place. After descent of the water table, seepage and condensate waters began to slowly remove the replacement gypsum, hydrobasaluminite, and

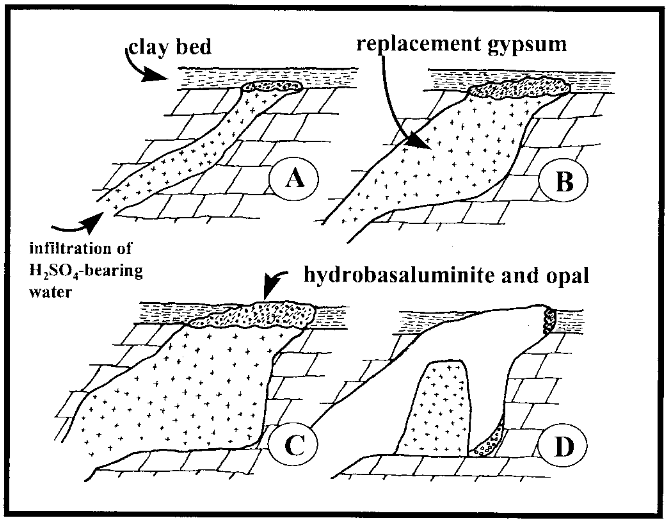

Figure 7. Proposed progression of cave development showing the origin of the cave passage and associated minerals. (A) Initial gypsum replacement of dolostone bedrock occurs along joints or zones of higher permeability. (B) The kaolinite-rich bed serves as a temporary barrier, but is eventually altered to hydrobasaluminite and amorphous silica. (C) Maximum replacement has occurred. (D) After sulfuric acid speleogenesis, seepage and condensate water removed most of the gypsum, hydrobasaluminite, and amorphous silica, forming the open cave passage as it is seen today.

amorphous silica, and eventually formed open cave passage. The absence of drip waters in the area of the hydrobasaluminite has resulted in the protection of these primary features (the remnant gypsum block and hydrobasaluminite globules) since the opening of the cave passage. Except at the bedrock pocket in the clay bed, the alumina phases have since been removed; but the opal has fallen and accumulated on the floor directly below the clay bed. This progression of cave development is illustrated in Figure 7 (A-D).

Aluminite is a secondary deposit of moonmilk on the bedrock surface. The aluminum and sulfate ions for formation of aluminite were supplied by the weathering of hydrobasaluminite, alunite, and gibbsite which are located above in bedrock pockets.

CONCLUSIONS

Hydrobasaluminite is a H_2SO_4-indicator mineral like the primary gypsum, alunite, natroalunite, and hydrated halloysite (endellite) in the caves of the Guadalupe Mountains. Hydrobasaluminite is a product of the interaction of H_2SO_4-bearing waters with a kaolinite-rich bed and other clay minerals in the dolostone bedrock during the formation of Cottonwood Cave.

Aluminite makes up a pasty to powdery deposit (moonmilk) on the cave wall immediately below the hydrobasaluminite and gibbsite deposits. It is a late-stage precipitate, and not a direct by-product of sulfuric acid speleogenesis.

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