

## SODIC CLAY-ZEOLITE ASSEMBLAGE IN BASALT AT BORON, CALIFORNIA<sup>1</sup>

WILLIAM S. WISE<sup>2</sup> AND WALLACE D. KLECK<sup>3</sup>

<sup>2</sup> Department of Geological Sciences, University of California  
Santa Barbara, California 93106

<sup>3</sup> Orange Coast College, Costa Mesa, California 92626

**Abstract**—An assemblage of clay minerals, Na-zeolites, Fe- and Sb-sulfides, and borates occurs in cavities in the basalt flow that underlies the main borax ore body exposed by the U.S. Borax open pit at Boron, Kern County, California. Analcime and saponite are widespread in cavities and fractures near the top of the basalt. Part of the basalt recently exposed, although restricted in distribution, contains a much more diverse assemblage of diagenetic minerals. Although only a few different minerals occur in each cavity, the composite order of deposition is: ferroan saponite, pyrrhotite, saponite, phillipsite, gmelinite, clinoptilolite, herschelite, analcime, greigite, rhodochrosite, searlesite, borax, calcite, and colemanite. In some cavities early-formed zeolites appear to have dissolved as later ones crystallized.

Microprobe analyses of the zeolites yielded the following compositions: phillipsite,  $\text{Na}_{4.28}\text{K}_{0.08}(\text{Al}_{4.36}\text{Si}_{11.64}\text{O}_{32}) \cdot x\text{H}_2\text{O}$  (Si/Al = 2.67); gmelinite,  $\text{Na}_{6.64}\text{K}_{0.10}(\text{Al}_{6.80}\text{Si}_{17.22}\text{O}_{48}) \cdot x\text{H}_2\text{O}$  (Si/Al = 2.53); herschelite,  $\text{Na}_{2.96}\text{K}_{0.24}(\text{Al}_{3.21}\text{Si}_{8.79}\text{O}_{24}) \cdot x\text{H}_2\text{O}$  (Si/Al = 2.75); clinoptilolite,  $\text{Na}_{6.48}\text{K}_{0.56}(\text{Al}_{7.15}\text{Si}_{28.77}\text{O}_{72}) \cdot x\text{H}_2\text{O}$  (Si/Al = 4.02); analcime,  $\text{Na}_{14.02}\text{K}_{0.09}(\text{Al}_{13.33}\text{Si}_{34.44}\text{O}_{96}) \cdot x\text{H}_2\text{O}$  (Si/Al = 2.58).

These zeolites formed by reactions between the basalt and evolving fluids, which were controlled by the development of an overlying, Na-borate lake. The wide distribution of saponite, analcime, and searlesite suggests that these minerals formed by the diagenetic reaction between basaltic glass and Na-borate water. The localized occurrence of the complex mineral assemblage, including some of the saponitic clay and the zeolites, phillipsite, gmelinite, and clinoptilolite, formed through a different process, perhaps deuteric or hydrothermal alteration of the basalt, before the lake developed. Because these zeolites are extraordinarily rich in Na, cation exchange may have taken place as the pore fluids became increasingly Na rich. In the latest stages of mineral growth, each of these zeolites was apparently partially dissolved and epitaxially overgrown by a second generation crystal, phillipsite on phillipsite, herschelite on gmelinite, and sodian heulandite on clinoptilolite.

**Key Words**—Analcime, Boron, Cation exchange, Clinoptilolite, Gmelinite, Heulandite, Phillipsite, Saponite, Sodium, Zeolite.

### INTRODUCTION

One of the characteristic properties of many zeolites and smectitic clays is their ability to exchange certain cations with those in a co-existing solution. In several earlier reports involving determination of the chemical composition of naturally occurring zeolites and clays, the authors suspected that even though these phases grew from one solution, they may have reached cation-exchange equilibrium with a later fluid. For example, dachiardite from Altoona, Washington, and from Agate Beach, Oregon, shows sharp compositional zoning from cores to the outer margins of fiber clusters (Wise and Tschernich, 1978). On the other hand detailed microprobe traverses of clay cavity linings or zeolite crystals commonly reveal compositional zoning, suggesting that exchange, if it occurred at all, was not particularly penetrative. In our experience few zeolite

occurrences show clear evidence of an earlier cation composition.

This paper describes an assemblage of clay minerals and zeolites from the cavities in a basalt flow underlying the borax ore body of the U.S. Borax mine at Boron, California. Both the clay minerals and zeolites are characterized by extremely Na-rich compositions. Evidence is presented that some of these zeolites and associated clay minerals formed with Ca-bearing compositions but were later exchanged with Na-rich water, when later Na-zeolites and the borosilicate, searlesite, also crystallized.

### GEOLOGIC SETTING

The clay minerals and zeolites occur in vesicles and fractures in basalt flows that underlie the Kramer borate deposit, Boron, California. The basalt and overlying lacustrine shales and borate deposits comprised the Miocene Kramer Beds (Barnard and Kistler, 1966). The Saddleback Basalt Member forms the basal unit of the formation and consists of at least two flows having a total thickness of about 60 m. These flows overlie a thin sequence of sediments and volcanic breccias that filled a shallow basin formed by an early phase

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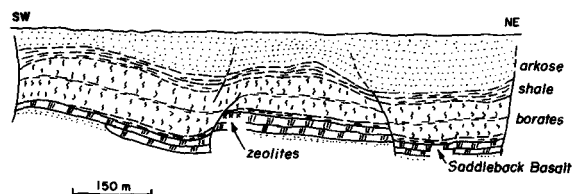


Figure 1. Cross section of the Kramer borate deposit, Boron, California, showing the Saddleback Basalt Member of the Kramer Beds. Location of the zeolitic part of basalt is shown near one of the faults that cut the deposit (after Barnard and Kistler, 1966).

of Basin and Range faulting. During Miocene time, the basin continued to develop through further faulting and was kept filled with various playa sediments, including silt, tuff, arkosic sand, and chemically precipitated borax. The origin of the borate is not precisely known, but is presumed to have come from thermal springs in the vicinity (Siefke, 1985).

A representation of the structure of the Kramer borate deposit is given in Figure 1. Because the open-pit mine was designed to extract only the borate ore, little of the underlying basalt is exposed, except near faults, and the base of the flows has never been exposed by the mining operation. The chemical analysis and normative mineralogy of the Saddleback Basalt (Table 1) shows that it is sub-alkaline, typical of basalts in calc-alkaline volcanic provinces (Basaltic Volcanism Study Project, 1981). The basalt flows were buried to depths of 200–330 m by the overlying sediments. Temperature in the buried basalt and overlying sediments could not have been above 60°C, which is the upper stability limit of borax. Analcime, searlesite, and iron sulfides have been known from the upper vesicular zone of the basalt since the opening of the pit in 1957 (Morgan and Erd, 1969). In 1983, a section of the basalt was exposed (Figure 1) that contains an unusually diverse assemblage of zeolites, iron sulfides, carbonates, and borates.

## METHODS

Minerals from the basalt cavities were identified by a combination of visual, optical, and X-ray powder diffraction (XRD) techniques. Most crystals are several millimeters in diameter, allowing easy separation of phases for further study. The identity of all phases was initially verified by XRD, but with experience, each phase could be recognized visually, even in complex epitaxial overgrowths. As is common with cavity (or vein) filling minerals, successive phases overgrow earlier ones, and none contain all the observed minerals. Therefore, a composite paragenetic sequence was constructed through the compilation of sequences observed in several tens of samples.

The identity of the clay minerals and zeolites was determined by standard techniques. XRD traces were

Table 1. Composition of the Saddleback Basalt, Boron, California (Higgins, 1973).

Normative minerals			
SiO <sub>2</sub>	49.50		
TiO <sub>2</sub>	1.20		
Al <sub>2</sub> O <sub>3</sub>	17.55	Quartz	0.0
Fe <sub>2</sub> O <sub>3</sub>	1.56	Orthoclase	2.11
FeO	7.90	Albite	24.13
MnO	0.17	Anorthite	34.92
MgO	6.55	Nepheline	0.0
CaO	10.40	Diopside	13.56
Na <sub>2</sub> O	2.80	Hypersthene	16.12
K <sub>2</sub> O	0.35	Olivine	4.17
P <sub>2</sub> O <sub>5</sub>	0.16	Magnetite	2.30
H <sub>2</sub> O	1.82	Ilmenite	2.32
Total	99.96	Apatite	0.38

Analyst: R. E. Higgins, X-ray fluorescence analysis.

obtained for clay preparations that were untreated and then successively saturated with ethylene glycol, and heated to 250° and 500°C. The samples were not sufficiently well crystallized to yield definitive 060 peaks. Therefore, full characterization of these samples depended on microprobe analyses.

Clinoptilolite crystals were sufficiently large for the usual XRD techniques, including heating experiments (Alietti, 1972); however, epitaxial overgrowths of heulandite were too small for any technique other than the determination of the optical orientation and microprobe analysis.

For electron microprobe analysis, crystals or masses of clay were selected from several cavities and embedded in epoxy resin. The mount was then polished and coated with 250 Å of carbon. Analyses were made on an ARL EMX electron microprobe having wavelength and energy dispersive X-ray detectors (Tracor). An accelerating voltage of 15 kV for all elements was used with a sample current of 7.5 nA. Standards used were andesine (Ca and Al), K-feldspar (K and Si), albite (Na), olivine (Mg), hematite (Fe), and sanbornite (Ba). To overcome some difficulties in analyzing zeolites by electron microprobe methods, large beam diameters (20 μm) were used, except for the heulandite overgrowths, which were too small for accurate analysis. Emission data were reduced by a ZAF program that combines signals from both detector systems.

## MINERALS IN THE BASALT CAVITIES AND FRACTURES

Cavities in the basalt were partly to completely filled with clay minerals, zeolites, borosilicates, carbonates, borates, and sulfides over a range of time, pore-fluid compositions, and possibly, temperature. Table 2 illustrates the composite paragenetic sequence coupled with events that may have affected the pore-fluid composition. As with most zeolite occurrences in mafic flows, the mineral phases vary from cavity to cavity,

Table 2. Paragenetic sequence of clay-zeolite-sulfide minerals, Saddleback Basalt, Boron, California.

	Influx of Na-borate water		Post-lake faulting	
Ferroan saponite	xxxxxx			
Pyrrhotite	xxxx	xxxxxx	xxx	
Saponite	xxxxx	xxxx	xxx	xx
Phillipsite		xxxxx	ooo	xx
Gmelinite		xxxxxx	oo	
Clinoptilolite		xxxxx	oo	
Heulandite			xx	
Herschelite			xxx	
Analcime			xxxxxxxx	
Greigite			xxx	
Rhodochrosite			xx	
Searlesite			xxxxxx	
Borax				xx
Calcite				xx
Colemanite				xxx

xxx = phase crystallizing; ooo = phase dissolving.

and no cavity contains all or even most of the phases listed in Table 2. A description of these phases in the context of the paragenetic sequence follows.

#### Clay minerals

All vesicle walls are coated by a layer of clay, 1–2 mm thick; however, about 10% of the cavities are completely filled with clay. The clay varies in appearance, but the earliest material is pale green and waxy. Later growths are ball-like masses, curved rods, curled leaf-like flakes, or earthy, moss-like masses. When cavities

are first exposed to the atmosphere the clay is pale to apple-green, but it rapidly (within 30 min) darkens to greenish-black, and within one day becomes brown. We interpret these changes to mean that the clay crystallized with iron only in the ferrous state, which rapidly oxidized upon exposure to the atmosphere (Kohyama *et al.*, 1973).

XRD analyses show that these clays have a 14-Å basal spacing, which expands to 17 Å with glycolation. Microprobe analyses (Table 3) show that all samples are rich in Mg and Fe; cell contents are consistent with a trioctahedral smectite. Ferrous iron appears to fill nearly half the octahedral sites in the saponite that forms the earliest cavity linings and about 30% of the sites in the latest masses. The early saponite contains significant amounts of Ti as well as Fe in octahedral sites and contains Ca, K, and Na in interlayer sites. The later saponites contain less Fe and no Ti and have mostly Na in interlayer sites.

#### Iron sulfides

Pyrrhotite occurs in nearly half the vesicles and forms single, isolated crystals (0.1–0.5 mm in size), rosettes, crusts, veins, or masses associated with saponite. Where fresh, the pyrrhotite has a bronzy color, but it alters to iridescent red. Pyrrhotite is commonly altered to goethite. The crystallization of pyrrhotite apparently accounts for the depletion of iron in the later saponite.

Greigite, less common than pyrrhotite, occurs as brassy coatings on earlier clay linings and on some zeolites, such as analcime. Very high magnification using a scanning electron microscope (not illustrated here) shows that the mineral forms framboids consisting of cubes, about 1 µm in size. Greigite is most easily recognized by its strong magnetism.

Smythite, present in lower parts of the Saddleback Basalt (Morgan and Erd, 1969), was not recognized in any of the zeolite-bearing samples.

Table 3. Electron microprobe analyses of saponitic clays in the altered part of the Saddleback Basalt, Boron, California.

	Earliest cavity lining	Intermediate layer	Latest filling
SiO <sub>2</sub>	43.08	43.70	47.51
TiO <sub>2</sub>	3.43	3.07	0.0
Al <sub>2</sub> O <sub>3</sub>	6.21	6.02	3.79
FeO <sup>1</sup>	19.78	19.52	14.57
MgO	10.92	11.78	17.36
CaO	0.58	0.47	0.20
Na <sub>2</sub> O	1.04	2.15	1.66
K <sub>2</sub> O	0.86	0.69	0.17
Total	85.92	87.44	85.00
<b>Unit-cell contents</b>			
<b>Tetrahedral layer</b>			
Si	6.90	6.90	7.39
Al	1.10	1.10	0.61
<b>Octahedral layer</b>			
Al	0.07	0.02	0.09
Mg	2.60	2.77	3.95
Fe	2.65	2.58	1.90
Ti	0.41	0.37	0.0
<b>Interlayer cations</b>			
Ca	0.10	0.07	0.04
Na	0.32	0.66	0.50
K	0.21	0.17	0.05

<sup>1</sup> Total Fe as FeO.

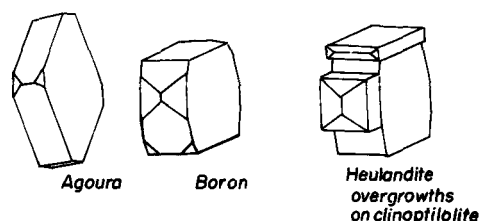
### Zeolites

Phillipsite was the first zeolite to form, and it occurs as isolated, blocky, complexly twinned groups as much as 4 mm in size. Where fresh, it is transparent and colorless to translucent white, but it is very commonly pseudomorphed by saponite. A few samples exhibit overgrowths of clear, glassy phillipsite on partially dissolved and clay-coated phillipsite. Microprobe analyses of the early phillipsite (Table 4) show that the zeolite is Na rich and contains minor K. These phillipsites are far more Na rich than those from saline, alkaline lakes (Sheppard and Gude, 1968) or those from the seafloor that were analyzed by Boles and Wise (1978).

Gmelinite occurs as single crystals or as groups of intergrown crystals. It is typically translucent and colorless to beige. Crystals (as large as 6 mm in size) exhibit the typical hexagonal dipyramid modified by a first order prism and basal pinacoids (Figure 2). Many crystals exhibit dissolution that appears to have started from the prism faces and to have removed much of the interior of the crystal (Figure 2). These gmelinites (Table 4) contain only Na and are richer in Si compared to all other gmelinites listed by Passaglia *et al.* (1978).

Many gmelinite crystals are epitaxially overgrown by flat herschelite plates, attached by respective (0001) faces. Although herschelite is the Na analogue of chabazite, the predominant prism and *c*-pinacoid forms give it a distinctive morphology. The Boron herschelite occurs as single crystals in addition to overgrowths on gmelinite. Both occurrences have about the same com-

### CLINOPTILOLITE



### GMELINITE

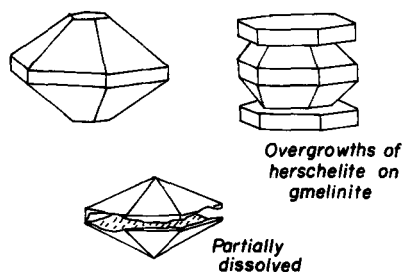


Figure 2. Drawings of clinoptilolite and gmelinite crystals from the Saddleback Basalt, Boron, California. Agoura clinoptilolite (Wise *et al.*, 1969) has typical heulandite form, but clinoptilolite crystals at Boron have much large (001) face. Heulandite overgrowths are typically on (001) and (100) faces of the clinoptilolite. Gmelinite crystals are dipyramidal and commonly have centers dissolved. Rarely, herschelite overgrowths are epitaxially attached by respective (0001) faces.

Table 4. Selected electron microprobe analyses of zeolites from the Saddleback Basalt, Boron, California.

	Phillipsite	Gmelinite	Herschelite	Clinoptilolite	Heulandite	Analcime
SiO <sub>2</sub>	58.26	57.65	54.88	68.38	63.94	58.21
Al <sub>2</sub> O <sub>3</sub>	18.52	19.32	16.99	14.42	15.79	19.11
Fe <sub>2</sub> O <sub>3</sub>	0.02	0.0	0.02	0.05	0.19	0.09
MgO	0.0	0.0	0.0	0.20	0.63	0.0
BaO	0.0	0.0	0.0	0.37	1.59	0.0
CaO	0.05	0.0	0.02	0.02	0.01	0.02
Na <sub>2</sub> O	11.05	11.46	9.52	7.94	8.19	12.22
K <sub>2</sub> O	0.32	0.27	1.17	1.04	1.19	0.12
Total	88.22	88.70	82.60	92.42	91.53	89.77
<b>Unit-cell contents</b>						
Oxygen	32	48	24	72	72	96
Si	11.64	17.22	8.79	28.77	27.69	34.44
Al	4.36	6.80	3.21	7.15	8.06	13.33
Fe	—	—	—	0.02	0.06	0.04
Mg	—	—	—	0.13	0.40	—
Ba	—	—	—	0.06	0.27	—
Ca	0.01	—	—	0.01	—	—
Na	4.28	6.64	2.96	6.48	6.88	14.02
K	0.08	0.10	0.24	0.56	0.66	0.09
Si/Al	2.67	2.53	2.75	4.02	3.44	2.58
E%	-0.4	+0.9	+0.2	-3.6	+9.5	-5.7

E% = 100(Al - Al<sub>theor</sub>)/Al<sub>theor</sub>; Passaglia *et al.* (1978).

position as gmelinite (Table 4). Herschelite shows no dissolution features.

One of the most abundant zeolites is clinoptilolite, which forms crystals as long as 3 mm and occurs as single crystals or clusters. Some cavities are lined with a crust of interlocking crystals. The crystals generally have the form of members of the heulandite group, but have a much larger (001) face (see Figure 2). Most are transparent and colorless, but some show partial dissolution along cleavage planes and replacement by saponite. The composition of the clinoptilolite (Table 4) is more Na rich than any previously reported, especially those from saline, alkaline lakes (Sheppard and Gude, 1968) or from deep-sea deposits (Boles and Wise, 1978). Rare crystals of clinoptilolite have oriented overgrowths of water-clear Na-heulandite (see Figure 2 and Table 4). The name heulandite is used even though the crystals are Na rich because the Si/Al ratio (3.44) is within the range of heulandites (Boles, 1972). Furthermore, the orientation of the optic plane in these crystals is perpendicular to {010}, whereas it is parallel to {010} in the clinoptilolite (Wise *et al.*, 1969).

The most widespread zeolite in the Saddleback Basalt is analcime, which occurs as glassy, colorless to slightly yellow trapezohedra and complex clusters of crystals. The composition is unusually rich in Si (Table 4), especially for occurrences in basaltic rocks (Saha, 1959); the Si/Al ratio is 2.58, well above the ideal ratio of 2. Analcime has not been affected by dissolution.

#### *Searlesite*

The sodium borosilicate, searlesite, occurs in veins, crusts, and clusters of radiating crystals as much as 1 cm long. Searlesite is glassy, transparent, and colorless. Small crystals of analcime locally have grown on the searlesite, indicating an overlap of crystallization. Although it is not abundant in the vesicles and fractures of the basalt, searlesite is not restricted to the zeolite occurrences, indicating that it is a persistent reaction product of the basalt and Na-borate waters.

#### *Borax and colemanite*

Transparent, tabular crystals of borax are rare in vesicles and more commonly coat fracture walls. Borax generally does not occur with minerals other than saponite, but it obviously formed only after the establishment of the overlying Na-borate lake. Colemanite rarely occurs in some cavities and seems to have formed very late because it covers all other minerals.

#### *Carbonates*

Rhodochrosite occurs as reniform clusters and rosettes of minute crystals in a few vesicles. Calcite forms massive vein material and cavity fillings that appear to cover all other minerals, except colemanite.

## ORIGIN OF CLAY MINERALS AND ZEOLITES IN THE BASALT

The composition of zeolites (Si/Al, as well as extra-framework cations) that occur in cavities of basalt is commonly controlled by the composition of the host rock (Keith and Staples, 1985; Nasher and Davies, 1960; Wise, 1982). The Saddleback Basalt is sub-alkaline and relatively Ca rich, but the zeolites are Na rich. Geologic relationships (Figure 1) show that the basalt flowed into a shallow basin that contained a playa lake. As the basin continued to deepen, lacustrine sediments covered the basalt. Local hot springs fed Na-borate waters into the lake, and the compositions of the zeolites probably are related to the Na-borate lake. The clay minerals and zeolites could have originated by: (1) diagenetic reaction of the basalt glass with Na-rich ground water after the flow was covered by the Na-borate lake; (2) a hydrothermal mechanism, possibly related to the hot springs feeding the Na-borate lake; or (3) deuteric alteration of basalt, followed by extensive Na exchange, when pore fluids became Na rich.

In Australia, diagenetic reaction of basaltic glass with cold ground water was considered by Nasher and Davies (1960) and Nasher and Basden (1965) as the likely mechanism to account for the widespread mineral assemblage. At Boron, the only minerals that have a wide distribution in the basalt are analcime, searlesite, and saponite. We interpret the origin of these minerals as resulting from reaction of Na-borate water from the overlying lake, reacting with Al and Si from the glass of the basalt.

The restricted distribution of zeolites in the basalt must be due to the process that localized fluids and/or heat. Hydrothermal systems commonly provide fluids at a temperature that is elevated above that of ground water to a restricted volume of host rock. Keith and Staples (1985) presented field and isotopic evidence that is consistent with such an origin for zeolites in basalts of the Coast Range of Oregon. Without evidence of hydrothermal activity, such as wall-rock alteration, deuteric alteration of the basalt flow has been postulated by Nasher and Davies (1960) to explain the occurrence of zeolites. In rocks with restricted circulation of fluids, the composition of the glass will control the composition of the zeolites (Wise, 1982). Because hydrothermal systems typically have high fluid/rock ratios, there may be no control of fluid compositions by the host rocks.

The occurrence of phillipsite, gmelinite, and clinoptilolite in the Saddleback Basalt is presently known only from a small area within the open pit. The crystallization of these zeolites probably resulted from some localized process—an alteration-related deuteric process or a small hydrothermal system, such as a hot spring. Because the 10-m vertical exposure of the ba-

salt, containing the complex assemblage of zeolites, exhibits no evidence of hydrothermal alteration, we appeal to deuteric alteration for the origin of the zeolites. All these zeolites, however, are Na rich, unusually so for zeolites in a sub-alkaline basalt (Keith and Staples, 1985). The phillipsite and clinoptilolite are more Na rich than those crystallizing from saline environments, such as sea water or saline, alkaline lakes. If these zeolites are deuteric alteration products, extensive cation exchange must have taken place.

One line of evidence supporting this conclusion is the clay-mineral compositions (Table 3). The earliest saponitic clays contain metal ions in proportion more closely matching the basalt glass than those formed later. The later saponites, containing very little Ca, were either cation exchanged by, or grew from Na-rich water.

Epitaxial overgrowths of phillipsite on earlier phillipsite, herschelite on gmelinite, and heulandite on clinoptilolite occurred following partial dissolution of the host mineral (Table 2), indicating a change in fluid composition. The close spatial association of analcime and searlesite with these overgrowths suggests that they resulted from the influx of Na-borate water.

The high silica content of the analcime was controlled by the activity of dissolved silica in the pore fluids. Thermodynamic modeling by Wise (1984) suggested that these fluids should be oversaturated with respect to silica, but a silica phase has not been found at Boron. Apparently, the excess silica was consumed by reaction with Na-borate to form searlesite (Bonnie Williamson, Department of Geological Sciences, University of California, Santa Barbara, California, personal communication, 1987).

It has long been recognized that the lake waters had a high sulfide content (Morgan and Erd, 1969). In part, this sulfur accounts for the extensive crystallization of pyrrhotite and greigite.

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