

# THE ENDELLITE-HALLOYSITE NOMENCLATURE\*

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## ABSTRACT

The nomenclature of the minerals halloysite and endellite is reviewed, and the justification for the continued use of the names halloysite,  $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$ , and endellite,  $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4 \cdot 2\text{H}_2\text{O}$ , is given.

## INTRODUCTION

Recent advances in understanding the mineralogy of clays have quite naturally brought some conflicts in nomenclature. Such a conflict has existed since 1935 for two clay minerals that previously had been considered to be the single mineral, halloysite. In an attempt to resolve this difficulty, Alexander, Faust, Hendricks, Insley, and McMurdie (1943) suggested that the name halloysite be restricted to the mineral of composition  $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$  and proposed the new name endellite for the more highly hydrated related mineral,  $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4 \cdot 2\text{H}_2\text{O}$ . The validity of this nomenclature, however, has been questioned by MacEwan (1947). In this paper justification for the continued use of the names halloysite and endellite is more fully developed.

The various suggestions made to date are summarized below:

Author	$\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$	$\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4 \cdot 2\text{H}_2\text{O}$	
{ All authors, } { 1826-1934 }	—Halloysite— (treated as a single species)		
	Ross and Kerr (1934)	Halloysite	Not recognized
	Hofmann, Endell, and Wilm (1934)	Kaolinite	Halloysite
	Mehmel (1935)	Metahalloysite	Halloysite
	Hendricks (1938)	Halloysite	Hydrated halloysite
	Alexander, Faust, Hendricks, et al. (1943)	Halloysite	Endellite
MacEwan (1947)	—Halloysite— (to be used as a group name)		
	Metahalloysite	Hydrated halloysite	also "partly hydrated halloysite"
Brindley, G. W. and others (1951)	—Halloysite—		
	(Non-specific term for all forms of the mineral)		

## HISTORICAL SUMMARY

### (a) 1826-1934.

Berthier (1826) gave the name halloysite to a clay mineral from Angleur, Liège, Belgium. The pertinent observations<sup>1</sup> on the water in the

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<sup>1</sup> Translated by G. T. Faust.

mineral are given below:

- p. 332. "If powdered halloysite is exposed for a certain time to a temperature near  
 p. 333. 100° C., it loses water; beyond that it loses no more than 0.16 by calcination. The powder from the drying, but not the calcination, absorbs water rapidly if it is dipped into water, or if it is left in contact with humid air."  
 p. 333. "If one considered the water which remains after drying in the oven to be combined water, the analysis will show:

$$\begin{array}{r} \text{SiO}_2 = 0.4494 \\ \text{Al}_2\text{O}_3 = 0.3906 \\ \text{H}_2\text{O} = 0.1600 \end{array}$$

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- p. 333. "Indeed, it appears extremely difficult to determine with complete exactitude the portion of water which is in a combined state and that which is only absorbed by capillary attraction."  
 p. 334. "It is very probable that the true composition of halloysite is represented by the formula  $2\text{AlS}^2 + \text{AlAQ}^2$  which corresponds to the following numbers:

$$0.393 \left\{ \begin{array}{l} \text{SiO}_2 \dots\dots\dots 0.470 \\ \text{Al}_2\text{O}_3 \dots\dots\dots 0.262 \\ \text{Al}_2\text{O}_3 \dots\dots\dots 0.131 \\ \text{H}_2\text{O} \dots\dots\dots 0.137 \end{array} \right\} \begin{array}{l} 0.732 \\ 0.268 \end{array}$$

These statements indicate that Berthier considered the halloysite mineral from Angleur to have the composition  $2\text{AlS}^2 + \text{AlAQ}^2$ , which in present day usage is  $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$ , with excess water present as "absorbed capillary water" and not as a hydrate.

Berthier's view that the excess water in the mineral that he examined was not essential, but was rather held mechanically by adsorption or through capillarity continued to be generally accepted, as was natural for this period. Le Chatelier (1887) referred to the excess water as "Eau hygrometrique" (hygroscopic water) and he was followed in this by Lacroix (1893). Le Chatelier (1887) states<sup>1</sup> in addition:

"The water is very sharply divided into two parts—the one part goes off at 150° C. after 24 hours of heating, or at 250° C. in one quarter of an hour; the other part begins to go off only at about 400° C. The proportions of this latter water is always very exactly two equivalents of water to one equivalent of alumina."

Dana (1892) also follows Le Chatelier and writes concerning "halloysite:"

"A silicate of aluminum, like kaolinite, but amorphous and containing more water; the amount is somewhat uncertain but, as shown by Le Chatelier, the formula is probably to be taken as  $\text{H}_4\text{Al}_2\text{Si}_2\text{O}_9 + \text{Aq}$ , or  $2\text{H}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 + \text{Aq}$ ."

Larsen and Wherry (1917)<sup>2</sup> studied the dehydration of "halloysite" and the change in the optical properties of the mineral with loss of water. Their conclusions follow:

"Obviously, the water in halloysite is in part only mechanically held. This part is given off very readily and the resulting partially dehydrated material has a composition near that of kaolinite. The formula should therefore probably be written  $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O} \cdot \text{Aq}$ .

"The close approach of this and many other analyses of halloysite to the composition  $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O} \cdot \text{Aq}$ , combined with the results of optical examination given above, indicates that the material called halloysite is the amorphous mineral corresponding to crystalline kaolinite, holding through capillarity or adsorption more or less excess water."

Larsen and Wherry (1917) found that material from Wagon Wheel Gap, Colorado, had an index of refraction of  $1.470 \pm 0.010$  two months after it was collected. This material they regarded as halloysite with unessential water. They found that when this mineral was dehydrated at  $65^\circ \text{C}$ . for four hours, its index of refraction rose to  $1.555 \pm 0.003$ .

In the first edition of Larsen's Tables (1921) he lists the following data for "halloysite."

<i>Page</i>	<i>N</i>	<i>Formula</i>
172	$1.470 \pm$	$\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot n\text{H}_2\text{O}$
173	$1.52 \pm$	$\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 + n\text{H}_2\text{O}$
174	$1.542 \pm$	$\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot n\text{H}_2\text{O}$
174	$1.555 \pm$	$\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$

It is clear from the foregoing that the general opinion was that stated in Dana-Ford (1932), "It is probable that halloysite is an amorphous mineral corresponding to kaolin, but holding by capillarity or adsorption varying amounts of excess water." It is then evident that various investigators have observed the hydrated material without recognizing that it was a distinct mineral. It is obvious, however, that all of them considered that the name "halloysite" should apply to the dehydrated form, that is to  $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$ . MacEwan (1947) has shown that many of the analyses of "halloysite" made before 1934 were probably made on the more highly hydrated material we now call endellite, or on mixtures of endellite and halloysite. It is now known that sub-microscopically crystalline kaolinite has been mistaken for halloysite. This in itself shows that the  $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$  formula was commonly believed to represent halloysite.

The important point is that prior to 1934 the water content in excess of  $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$ , was universally considered to be nonessential

<sup>2</sup> MacEwan (1947) on page 40 states that "Larsen and Wherry in their paper put forward the formula  $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O} + 2\text{Aq}$ ," but no such proposal is to be found in their paper. They merely compared their analysis of the mineral with a hypothetical formula of that composition.

water. Mineralogists actually used the term halloysite to refer indiscriminately to  $\text{Al}_2\text{Si}_2\text{O}_7 \cdot 2\text{H}_2\text{O}$ ,  $\text{Al}_2\text{Si}_2\text{O}_7 \cdot 4\text{H}_2\text{O}$ , and to mixtures of the two, but considered that they were referring to  $\text{Al}_2\text{Si}_2\text{O}_7 \cdot 2\text{H}_2\text{O} + n\text{H}_2\text{O}$  (unessential). This was true not only of American mineralogists (see quotation from Dana-Ford above), but also of British and German mineralogists. For example, Read (1936) gives, "Halloysite—Hydrated aluminum silicate, near kaolinite,  $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$ ," and Ramdohr (1936) gives, "Halloysite,  $\text{H}_4\text{Al}_2\text{Si}_2\text{O}_9 + \text{Aq.}$ "

I, therefore, believe that application of the term halloysite to  $\text{Al}_2\text{Si}_2\text{O}_7 \cdot 2\text{H}_2\text{O}$  is in accord with past usage.

(b) 1934—present.

Halloysite was first definitely differentiated from kaolinite by Ross and Kerr (1934), who made chemical, optical,  $x$ -ray, and dehydration studies. The form of the dehydration curves and the ease with which water was lost led them to state, "It is quite evident that the water lost up to  $110^\circ$  (1.44 to 4.75 per cent) is not significant, but represents adsorbed water. In fact, the water lost up to  $200^\circ$  (2.5 to 5.20 per cent) is no doubt adsorbed." This study justified them in following generally accepted and long-established usage by applying the name halloysite to the microcrystalline compound  $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$ .

Spencer (1935) in his abstract of the paper by Ross and Kerr (1934) writes:

"Twelve analyses of halloysite (including 'indianite,' 'newtonite,' and 'lithomarge') from various localities show a  $\text{SiO}_2:\text{Al}_2\text{O}_3$  ratio ranging from 206:100 to 165:100, the agreement with the usually accepted formula  $\text{H}_2\text{Al}_2\text{Si}_2\text{O}_8$  being only approximate."

This formula given by Spencer may also be expressed as  $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot \text{H}_2\text{O}$ . This formula does not agree with the analyses in the literature, and has not been accepted by mineralogists.

Hofmann, Endell and Wilm (1934) were the first to show that the  $x$ -ray pattern of the fully hydrated mineral differed from that of material heated to  $50^\circ$  or dried over  $\text{H}_2\text{SO}_4$  or  $\text{P}_2\text{O}_5$ . They erroneously concluded that the dehydrated material was kaolinite. This may have influenced them to use the name halloysite for the fully hydrated material which they state to be approximately  $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 4\text{H}_2\text{O}$ .

Mehmel (1936) confirmed their findings that the fully hydrated mineral was distinct (characteristic  $x$ -ray line at  $10.1 \text{ \AA}$ ), but showed by optical and  $x$ -ray study that the dehydrated material (characteristic  $x$ -ray line at  $7.46 \text{ \AA}$ ) was not kaolinite. Mehmel does not give the basis for his nomenclature. Perhaps following Hofmann, Endell, and Wilm (1934), he called the fully hydrated mineral halloysite and referred without comment to the dehydrated mineral as metahalloysite.

The work of Ross and Kerr (1934) was apparently not known to Mehmel when he proposed the name metahalloysite for the same mineral that they had characterized so completely and named halloysite. It is clear that their name has priority over his, which is in itself sufficient basis for dropping the name "metahalloysite." We believe that the historical summary above disproves MacEwan's statement, "Hofmann and co-workers, and Mehmel, were historically correct in assuming that the hydrated halloysite was the true halloysite . . ." Furthermore, MacEwan (1947, p. 36) has pointed out that it is not clear whether Mehmel knew that his "metahalloysite" occurred naturally, all of Mehmel's samples having been prepared by dehydration. Mehmel used the name metahalloysite only in reference to an artificial product, and a name having such a basis has no standing in the nomenclature of mineralogy. The discoverer of the natural mineral may accept, if he wishes, the name applied to the artificial compound, but he is not bound to do so.

For all these reasons, the term *metahalloysite* should be dropped from mineralogical nomenclature.

Hendricks (1938) used halloysite for  $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$  and hydrated halloysite for  $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 4\text{H}_2\text{O}$ . He writes, "The great ease with which this dehydration takes place has not generally been recognized and as a consequence there is some confusion in the literature, but as a rule Mehmel's "metahalloysite is the usual halloysite of others." MacEwan questions the validity of this statement, and remarks that no references were cited to support it. The detailed historical summary above warrants our having felt in 1938 that the statement was so much in accord with accepted usage that it needed no defense.

Edelman and Favejee (1940) objected to the terminology of Hendricks (1938) on the basis that this nomenclature gives the erroneous impression that the mineral rich in water originates from the mineral poor in water. The objection is valid, and Alexander, Faust, Hendricks, Insley and McMurdie (1943) proposed the name endellite for  $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 4\text{H}_2\text{O}$  and retained the name halloysite for  $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$ .

To summarize,

- (1) The use of the name halloysite for  $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$  is consistent with the usage generally accepted over a long period of years;
- (2) The mineral of this composition was first adequately characterized by Ross and Kerr, who called it halloysite;
- (3) The name metahalloysite for the mineral of this composition should be dropped, from consideration of priority, and for other valid reasons.

There remain to be discussed MacEwan's suggestion of the use of halloysite as a group name and his objections to the name endellite for  $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 4\text{H}_2\text{O}$ .

## USE OF HALLOYSITE AS A GROUP NAME

MacEwan (1947) states, "thus the name 'halloysite' would be taken to cover all the naturally occurring halloysite minerals (hydrated, non-hydrated, and intermediate) as well as artificially produced complexes; and for particular forms the precise terms 'hydrated halloysite,' 'glycerol-halloysite,' etc. would be used. . . ." In essence, MacEwan proposes to return to the practice, used before the distinction between the two hydrates was known, of grouping them together under a single term, together with artificial organic complexes.

His justification for this is chiefly his belief "*that the two halloysite minerals are only two members of a series of substances which includes various intermediate forms, . . .*" (Italics G.T.F.), and "I wish to suggest that recent research has shown that halloysite and metahalloysite (hydrated halloysite and halloysite; or endellite and halloysite) are so closely connected that they are more conveniently regarded as two aspects of a single substance than as entirely separate substances."

The weight of evidence seems to point almost certainly to a discontinuity in the series. Such a discontinuity appears in the optical work by Mehmel (1937) and by Alexander, Faust, Hendricks, *et al.* (1943), and in the dehydration studies by Hofmann, Endell and Wilm (1934) and by Mehmel (1937). Optical work at the U. S. Geological Survey is also in agreement with this earlier work.

Brindley and Goodyear (1948) made an *x*-ray study of the transition endellite to halloysite (halloysite to metahalloysite in their terminology). They found that endellite showed spacings of 9.5 to 10.1 Å, and halloysite 7.5 to 7.9 Å, but were "unable to obtain evidence for the existence of intermediate stages with spacings between 9.5 and 7.9 Å" (p. 411). The variations in spacings found corresponded to variations in the formula  $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot n\text{H}_2\text{O}$ , with *n* varying from  $3\frac{1}{2}$  to 4 for endellite,  $2\frac{1}{3}$  to  $2\frac{2}{3}$  for halloysite.

Very convincing evidence for the invalidation of MacEwan's chief argument for the use of halloysite as a group name, quoted above, has come from the recent phase-rule studies on the system  $\text{Al}_2\text{O}_3\text{-SiO}_2\text{-H}_2\text{O}$  and the subsystem  $\text{H}_2\text{O-halloysite}$ . Bates (1952) has published a diagram delimiting the field of stability of halloysite and endellite as a function of temperature and the vapor pressure of water in the system. He found no evidence of phases intermediate between endellite and halloysite. Roy and Osborn (1954) in their study of the system  $\text{Al}_2\text{O}_3\text{-SiO}_2\text{-H}_2\text{O}$  extended these studies and their comments are especially pertinent to this discussion. On page 861 they write:

"Brindley and Goodyear (1948) have stated that halloysite has an 001 spacing of 7.4 or 7.5kX. Such a spacing was encountered in several of our runs and it was at first consid-

ered feasible that the 7.4 Å reflection—characteristic of a hydrate with more than 2 H<sub>2</sub>O—represented the true value for halloysite. This would have meant that halloysite and kaolinite are not polymorphs. However, a study of the spacings of halloysite dehydrated at temperatures from 100° C. to 450° C. shows that after the attainment of equilibrium the spacing is constant at 7.17 Å. Apparently in the work of Brindley (1951, p. 52) and also in our runs which yielded a structure with a 7.4 Å spacing, dehydration was not carried to equilibrium at the temperatures used. In hydrothermal runs at temperatures and pressures in the region near the equilibrium decomposition curve, endellite may dehydrate only partially to yield the 7.4 Å phase. However, we have found that the spacing of halloysite derived by decomposition of endellite under temperature and pressure conditions existing to the right of Curve II (Fig. 6) is invariably 7.17 Å. Moreover, halloysites dehydrated at 110° C., 300° C. and 400° C. would not rehydrate to the 7.4 Å phase, even under high water pressures at appropriate temperatures. While the above shows that halloysite is a true polymorph of kaolinite with a 7.17 Å spacing, it also does suggest that the interlayering proposed by Brindley and Goodyear has some unique stability, since no other intermediate spacings have been consistently found."

MacEwan suggests that there are advantages in the use of "halloysite" as a vague general term but this seems dubious to me. I believe that mineralogic nomenclature should not be burdened by devising mineralogical names for such organic complexes of clay minerals as "glycol-halloysite."

MacEwan (1947) states that the name endellite has no obvious connection with halloysite. This point is really an advantage as there is no ambiguity about the meaning of the name. The name endellite is being used by many mineralogists in the United States, and also is being rapidly adopted by economic geologists. Callaghan (1948) has described the economic geologic relations of endellite in the Gardner Mine Ridge area, of Lawrence County, Indiana.

In Germany the name endellite also is being used by mineralogists. Professor Strunz (1949) is using these names in his *Mineralogische Tabellen*. Professor Ulrich Hofmann in a letter to Dr. Clarence S. Ross<sup>3</sup> writes as follows:

"Questions of nomenclature always appear to me as something difficult and I am very grateful to you and Dr. Hendricks that you have undertaken this difficult question so fundamentally. I myself have, as you know, first used the name halloysite for the mineral with the higher water content, 4H<sub>2</sub>O·Al<sub>2</sub>O<sub>3</sub>·2SiO<sub>2</sub> and Mehmel has followed this usage. The essence of this is, as you have already emphasized, what did mineralogists previously understand the name halloysite to mean? If the name halloysite refers to the water-poor mineral 2H<sub>2</sub>O·Al<sub>2</sub>O<sub>3</sub>·2SiO<sub>2</sub>, then Endell, Wilm, and I have in our paper, *Angewandte Chemie*, 47, (1934) 541, for the first time recognized correctly the existence of the water-rich mineral. It is accordingly correct to give this new mineral a name and I am very grateful to you, that you have chosen for it the name of my lifelong and unfortunately deceased friend and coworker, Endell."

and in 1955 in a letter to the author<sup>4</sup> he states:

"My associates and I find the names endellite and halloysite useful and justified."

<sup>3</sup> Personal communication, June 26, 1949.

<sup>4</sup> Personal communication, April 19, 1955.

## SUMMARY

For nearly a hundred years, the name halloysite was applied by many mineralogists indiscriminately to both  $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$  and  $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4 \cdot 2\text{H}_2\text{O}$  in the mistaken belief that the additional water in the more highly hydrated mineral was unessential and adsorbed. Halloysite was never clearly defined or characterized until the work of Ross and Kerr on the whole kaolin group. These authors were clearly justified in following the long accepted usage and restricting the name halloysite to the mineral with only  $2\text{H}_2\text{O}$ .

The acceptance of the name endellite for the mineral with  $4\text{H}_2\text{O}$  will obviate ambiguity and leave no doubt about the identity of the mineral under consideration as the proposal of MacEwan does not do.

Hofmann, who first discovered and described the mineral, has agreed to the name endellite for this mineral with higher water content.

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