

A hydrated barium silicate in unmetamorphosed sedimentary rocks of central North Greenland

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Abstract

A new occurrence of barium silicates has been found in unmetamorphosed sedimentary rocks of central North Greenland. Two different barium minerals have been identified, the Ba-feldspar hyalophane, and an unknown and hitherto undescribed hydrated Ba-silicate with an anhydrous chemical composition identical to cymrite but with about 4 moles of water in the mineral structure. Both silicates are found in a black organic-rich chert sequence in close association with baryte. No replacement textures are observed between the three Ba-minerals.

The hydrated Ba-silicate in the unmetamorphosed rock sequence of the Navarana Fjord area is considered to represent a member in a sequence of Ba-silicates from the unknown Ba-silicate with four H₂O in the structure (BaAl₂Si₂O₈·4H₂O), through cymrite with one H₂O (BaAl₂Si₂O₈·H₂O) to celsian with none (BaAl₂Si₂O₈). This sequence might reflect the continuous release of water during prograde diagenesis and metamorphism. The association between chert and Ba minerals in the sedimentary rocks of the Navarana Fjord area is believed to reflect an addition of silica and barium by hydrothermal solutions.

KEYWORDS: Hydrated Ba-silicate, hyalophane, baryte, unmetamorphosed sedimentary rocks, chert, hydrothermal activity, diagenesis.

Introduction

DURING a geochemical study of unmetamorphosed sedimentary rocks from central North Greenland, a Ba-silicate with an anhydrous formula similar to that of cymrite (BaAl₂Si₂O₈) but with about 4 H₂O in the mineral structure was discovered in black organic-rich cherts (Jakobsen, 1987) together with hyalophane and baryte. The aim of the present study is to report the chemical characteristics of the unknown hydrated Ba-silicate and to discuss its possible genetic significance with respect to the other Ba minerals found.

Many described occurrences of Ba-silicates are from metamorphic rocks. Ba-silicates have been described in great detail from the Aberfeldy barium-zinc mineralization (Coats *et al.*, 1980; Fortey and Beddoe-Stephens, 1982) where observations indicated cymrite to be a diagenetic precursor mineral to celsian, an indication supported by the present work. In addition, cymrite has been reported from manganese mines (Smith *et al.*, 1949; Reinecke, 1982), from stratabound baryte deposits (Coats *et al.*, 1980), and from various other Ba-enriched rocks (Runnels, 1964; Essene, 1967; Bjørlykke and Griffin, 1973).

The occurrence of the Ba-silicates in the Navarana Fjord area, however, appears to be the first described example from unmetamorphosed rocks; however Kucha (1988) mentioned Ba-feldspars in Lower Carboniferous carbonates in Ireland. The observations from this study, when compared to those of other known occurrences of chemically related minerals in metamorphosed rocks permit the formulation of a model for the presence of various hydrated Ba-silicates.

Geology

Central North Greenland consists of a major sequence of sedimentary rocks (Fig. 1a) deposited in the eastern extension of the Franklinian basin during the Lower Palaeozoic (Dawes, 1976). The crystalline basement is only seen in small outcrops in the southeast (Henriksen and Jepsen, 1985). The sedimentary rock sequences can be divided into two east-west trending belts which, towards the south, are defined by a shelf sequence, mainly carbonate, and towards the north by a deep-water trough sequence, which is often seen to overlie the slope, outer and inner shelf sequences.

The northernmost parts of the deep-water

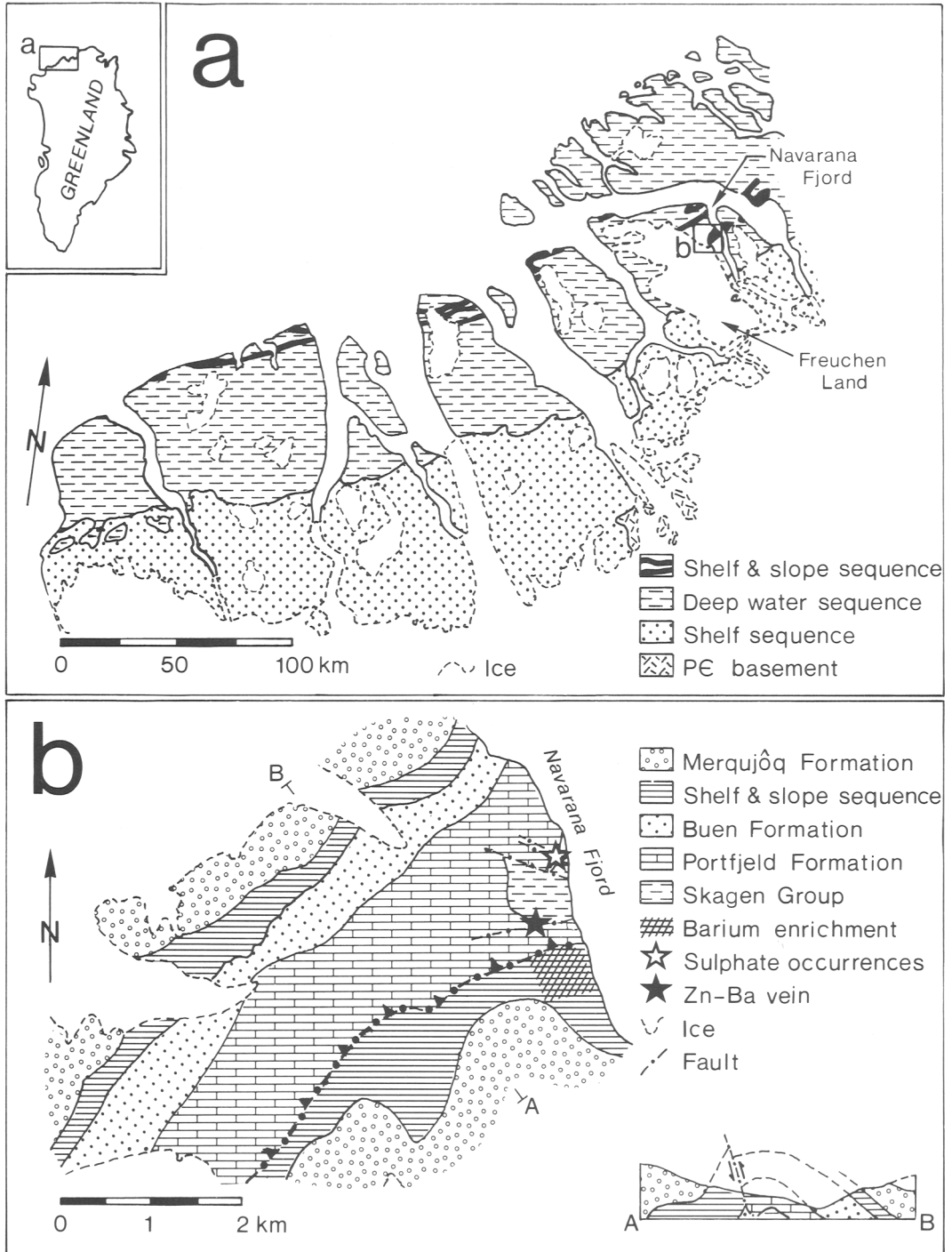


FIG. 1. Geological maps of North Greenland and the Navarana Fjord area. (a) Central and western North Greenland (modified after Henriksen, 1987). (b) The Navarana Fjord area. Mineralization in the area is situated close to the Navarana Fjord.

sequence were folded during the Late Palaeozoic Ellesmerian orogeny and today form part of the North Greenland fold belt.

The investigated Navarana Fjord area lies at the transition between the trough and shelf sequences. The area is weakly folded in one big anticline but no signs whatsoever of metamorphic minerals are found. Five rock sequences occur in the area (Higgins and Soper, 1985; Davis and Higgins, 1987) (Fig. 1*b*): the sandy Skagen Group of Late Precambrian/Early Cambrian age; the dolomitic Portfjeld Formation and the sandy to shaly Buen Formation, both of Lower Cambrian age; the 'unnamed shelf and slope sequence' (Henriksen, 1987) of Lower Cambrian to Lower Silurian age; and the turbiditic Merqujôq Formation of Lower Silurian age. Only the 'unnamed shelf and slope sequence' is of interest in connection with the presence of Ba-silicates.

On a regional scale the 'unnamed shelf and slope sequence' can be divided into four units (Higgins and Soper, 1985) of which probably only the uppermost two, units 3 and 4, are present in the Navarana Fjord area. Unit 3, the Kap Stanton Formation (Ineson and Peel, 1989), consists of a *ca.* 220 m thick sequence of black carbonates and shales with four major intercalations of intraformational conglomerates. Barium concentrations up to 1 wt.% are found in chert from this formation, but only baryte has been identified by X-ray diffractometry. Unit 4, the *slope* equivalent of the Amundsen Land Group deep-water sequence (Davis and Higgins, 1987), is a *ca.* 100 m thick sequence consisting of black organic-rich cherts with minor dark carbonates and shales. One sample consisting of black cherts with up to 6 wt.% Ba have been identified in this sequence reflecting the presence of baryte, hyalophane and a hydrated Ba-silicate.

Of possible pertinence to this study in the Navarana Fjord area (Fig. 1) is the occurrence of a carbonate-hosted Zn-Pb-Ba vein-type mineralization situated in the dolomitic Portfjeld Formation (Jakobsen and Steenfelt, 1985; Jakobsen, 1987; Jakobsen and Stendal, 1987; Jakobsen, 1989*a*) and several fault-related, rare, hydrated iron sulphate occurrences (Jakobsen, 1989*b*). These are situated 500 to 1000 m north of the outcrop of the Ba-enriched 'unnamed shelf and slope sequence' (Fig. 1*b*).

Geochemistry and petrography

The chert in which the different barium minerals was found is a *ca.* 100 m thick massive, black organic-rich rock sequence belonging to the Amundsen Land Group (the uppermost part of

the shelf and slope sequence). The actual sample, only one, where the barium minerals are found consists essentially of SiO₂, Al₂O₃, Fe₂O₃ and Ba.

Minerals found in the sample are, in order of decreasing amount, amorphous quartz, baryte, hydrated Ba-silicate, hyalophane, orthoclase and pyrite. Because of the microcrystalline habit and the black colour of the rock, only the pyrite (<1 µm euhedral crystals) and baryte occurring in cracks were visible during ordinary microscopy. The petrographic observations on Ba-silicates in the chert proper were made with the aid of the back-scattering facilities during microprobe analysis. The hydrated Ba-silicate (<100 µm in size) occurs partly as anhedral platy forms (Fig. 2*a*) and partly as fan-shaped aggregates with a pronounced cleavage (Fig. 2*b*). Baryte is often seen to be concentrated around the Ba silicate, either as isolated grains away from the silicate (Fig. 2*b*), or with a straight, sharp boundary to the silicate (Fig. 2*a*). No textural signs of replacement between the two minerals, or their dissolution, were observed.

Hyalophane occurs in the chert mainly as isolated grains with anhedral, platy forms but, when occurring together with baryte, baryte always occurs around and free of contact with hyalophane.

Chemical zonation was not observed in any mineral grains.

Analytical methods

Electron-microprobe analyses of the hydrated Ba-silicate and hyalophane were carried out by a JEOL 733 Superprobe, at the Institute of Mineralogy, University of Copenhagen. Analysis of Si, Al, Ba, Na and K were made on 34 grains of hydrated Ba silicate and on 8 grains of hyalophane from one sample, using albite (Na), orthoclase (K), wollastonite (Ca, Si), corundum (Al₂O₃) and baryte (BaSO₄) as standards. The analyses were performed using a beam diameter of 2 µm and a beam current of 10 nA. The JEOL program PACX was used for ZAF correction and instrumental control. Analyses of Ti, Fe, Mg and Ca were made on only a few grains, due to the low contents of these elements.

Attempts were made to further identify the hydrated Ba-silicate by X-ray diffraction methods but, due to difficulties with separation of the fine-grained minerals, no further information on the unknown mineral was obtained.

Results

Chemical analyses of the hydrated Ba silicate and hyalophane are listed in Tables 1 and 2,

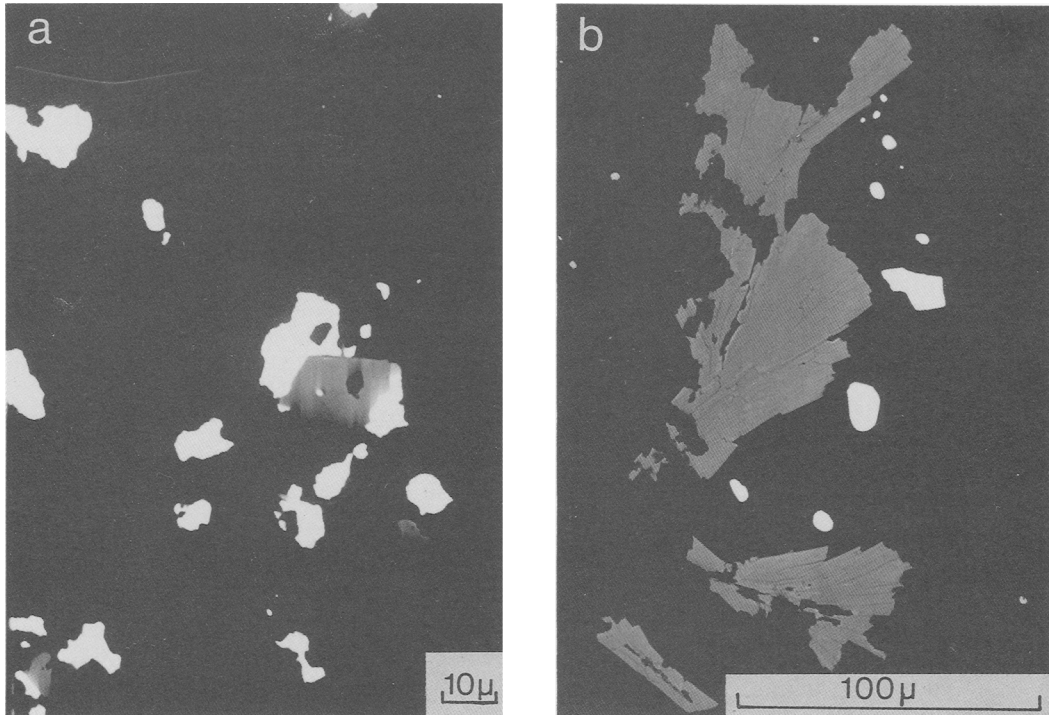


FIG. 2. Back-scattering images of hydrated Ba silicate and baryte. (a) Anhedrally platy Ba silicate (grey) surrounded by and showing a sharp boundary with baryte (white). (b) Fan-shaped aggregates of hydrated Ba silicates (grey), and blebs of baryte (white).

respectively. Aspects of their chemical characteristics are displayed in Figs 3, 4 and 5, together with data for celsian, cymrite and hyalophane from other localities.

The chemical composition of the hydrated Ba-silicate from the Navarana Fjord area (Table 1) compares favourably with published celsian and cymrite compositions (Carron *et al.*, 1964; Essene, 1967) from other localities in terms of cation ratios on an anhydrous basis. However, compared with cymrite proper (Table 1), the Navarana Fjord Ba-silicate has a significantly lower total of about 83 wt.% and a slight deficiency in Ba in the formula based on 8 oxygens. In order to further verify the correctness of these analyses, two samples from the Aberfeldy barium-zinc mineralization with previously analysed cymrite and celsian were provided by Dr N.J. Fortey of the British Geological Survey. The analyses obtained on both celsian and cymrite from these samples correspond closely with those previously published (Fortey and Beddoe-Stephens, 1982). Hence the analyses obtained on the Navarana Fjord Ba-silicate are considered correct

and the low totals are probably accounted for by a higher content of H₂O than in cymrite. Assuming H₂O to be responsible for the low totals obtained, calculations indicate contents from 3 to 5.5 (averaging 4) moles of H₂O in the mineral structure. Because of the fine-grained nature of the minerals these figures could not be substantiated further, nor could the presence of H₂O rather than OH be checked.

The contents of SiO₂, Al₂O₃ and BaO in the hydrated Ba silicate are all lower than in cymrites from other localities (Table 1) (Fortey and Beddoe-Stephens, 1982; Reinecke, 1982; Essene, 1967; Froehlich and Sandréa, 1973; Aye and Strauss, 1975).

The (BaO + Al₂O₃) versus (SiO₂ + K₂O + Na₂O) relations in Ba-silicates from Navarana Fjord, Aberfeldy and other localities are shown in Fig. 3. Included in the figure is the line of ideal solid solution between pure celsian and orthoclase, as well as lines showing the shift in composition due to the addition of 1–6 moles of H₂O to the mineral structure. The hydrated Ba-silicate from Navarana Fjord plots below the anhydrous

TABLE 1. ELECTRON-MICROPROBE ANALYSES OF HYDRATED Ba-SILICATE AND CYMRITE

	1 Hydrated Ba silicate Navarana Fjord N=34 s.d.	2 Cymrite Aberfeldy N=4	3 Cymrite California	4 Cymrite Hauts- Pyrénées	5 Cymrite Huelva Spain	6 Cymrite Greece N=5
SiO ₂	29.05 (1.69)	32.50	30.3	31.0	31.07	31.06
Al ₂ O ₃	23.49 (1.21)	24.82	25.9	26.0	25.45	25.80
MgO	—	—	—	0.2	—	—
CaO	—	—	0.0	0.3	—	—
BaO	29.66 (2.06)	36.52	38.7	38.0	40.18	38.72
Na ₂ O	0.18 (0.18)	0.18	0.0	—	—	—
K ₂ O	0.45 (0.28)	0.59	0.03	0.3	—	0.26
Total	82.83	94.61	94.93	95.80	96.70	95.86

FORMULAE BASED ON 8 OXYGENS

Si	2.06	2.10	1.994	2.010	2.023	2.019
Al	2.00	1.89	2.009	1.987	1.953	1.976
	4.06	3.99	4.003	3.997	3.976	3.995
Mg	—	—	—	0.0	—	—
Ca	—	—	0.0	0.021	—	—
Ba	0.84	0.92	0.997	0.966	1.025	0.985
Na	0.02	0.02	0.0	—	—	—
K	0.04	0.05	0.003	0.025	—	0.022
	0.90	0.99	1.000	1.012	1.025	1.007

(1) This work, (2) Fortey & Beddoe-Stephens (1982), (3) Essene (1967),
 (4) Froehlich & Sandrea (1973), (5) Aye & Strauss (1975), (6) Reinecke (1982).
 s.d. Standard deviation

celsian-orthoclase line in positions corresponding to 3–5.5 (averaging 4) moles of H₂O in the mineral structure, and on a straight line which includes cymrite and celsian from Aberfeldy. Most cymrites from other localities fall off this line and plot close to pure celsian with 1 mole of H₂O.

On the (Ba + Al) versus (Si + Na + K) cation diagram (Fig. 4), the hydrated Ba-silicate of Navarana Fjord plots on a straight line between orthoclase and celsian, close to the latter end member and close to celsian and cymrite from Aberfeldy. Cymrite from other occurrences plots close to pure celsian. On the Ba versus Al cation diagram (Fortey and Beddoe-Stephens, 1982) shown in Fig. 5, however, the hydrated Ba-silicates of Navarana Fjord plot below the celsian-orthoclase substitution line.

The analyses of hyalophane reach a total of 100 wt.%, but generally the SiO₂ content is higher and the BaO and K₂O content lower than in other published analyses (Table 2). On the (BaO + Al₂O₃) versus (SiO₂ + K₂O + Na₂O) wt.% diagram (Fig. 3), hyalophanes of Navarana Fjord and Aberfeldy plot on the line between celsian and orthoclase although the high SiO₂ and low BaO content displace the analyses toward orthoclase. The formula based on 8 oxygens does not fit those calculated from the Aberfeldy area (Table 2). The sum of Ba, Na and K is far too low, and on Fig. 4 the analyses of hyalophane from Navarana Fjord scatter below both the Na + K + Si = Ba + Al substitution line and the

hyalophanes from Aberfeldy. The ratios between Ba and Al (Fig. 5) seem on the other hand to match those of Aberfeldy, and the high SiO₂ content (Table 2) is most probably artificial, due to the very fine grain size of the hyalophane and the incorporation of SiO₂ in the analysis from the surrounding chert.

Discussion

The chemical analysis of the Ba-silicate from the Navarana Fjord area is compatible with cymrite, but with a theoretical average H₂O content corresponding to 4 molecules (Table 1; Figs 3 and 4). The slight displacement of the hydrated Ba-silicate toward orthoclase is due to small contents of Na₂O and K₂O. Furthermore the overall chemistry fits that of cymrite and celsian from the Aberfeldy area, except for the presumed higher H₂O content of the Navarana Fjord hydrated Ba-silicate. Cymrite from other localities is more celsian-pure (without K₂O and Na₂O), probably reflecting a difference in chemistry of the depositional environments as suggested by Fortey and Beddoe-Stephens (1982).

A comparison of the chemical data of this study and those on Ba-silicates from other localities lead to the suggestion that the hydrated Ba-silicate of Navarana Fjord is either an intermediary phase in a sequence from, e.g. harmotome (Ba₂Al₃Si₁₁O₃₂·12H₂O), as suggested by Fortey and Beddoe-Stephens (1982), through the

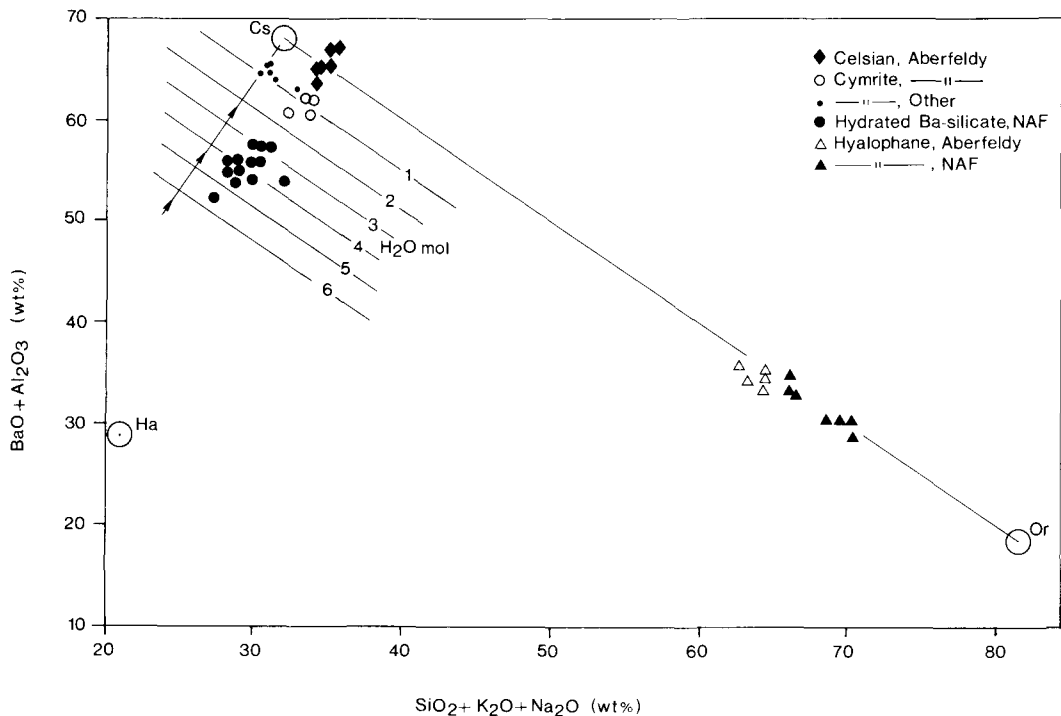


FIG. 3. $\text{BaO} + \text{Al}_2\text{O}_3$ vs. $\text{SiO}_2 + \text{K}_2\text{O} + \text{Na}_2\text{O}$ (wt.%) for hydrated Ba silicate and hyalophane from the Navarana Fjord area, and from other localities. Additional data are from Forthey and Beddoe-Stephens, 1982; Reinecke, 1982; Essene, 1967; Froehlich and Sandra, 1973 and Aye and Strauss, 1975. Also indicated in the figure are calculated lines representing $(\text{BaAl}_2\text{Si}_2\text{O}_8)_x$, with contents of one to six moles H_2O in the mineral structure. The line with arrows indicates the change in composition of a hydrated celsian to pure celsian with release of water. The position of harmotome is also indicated. Abbreviations: NAF, Navarana Fjord area; Cs, pure celsian; Or, pure orthoclases; Ha, harmotome.

TABLE 2. ELECTRON-MICROPROBE ANALYSES OF HYALOPHANE

	1 Navarana Fjord N=8		2 Aberfeldy area N=6	
		s.d.		
SiO_2	59.59	(1.63)	53.44	
Al_2O_3	21.37	(0.70)	21.04	
BaO	10.06	(2.33)	13.76	
K_2O	7.68	(1.84)	9.06	
Na_2O	0.19	(0.11)	1.40	
Total	98.89		98.70	
FORMULAE BASED ON 8 OXYGENS				
Si	2.85		2.73	
Al	1.21		1.27	
	4.06		4.00	
Ba	0.19		0.28	
K	0.47		0.59	
Na	0.02		0.14	
	0.68		1.01	

(1) This work,
(2) Forthey & Beddoe-Stephens (1982).
s.d. Standard deviation

unknown hydrated Ba-silicate to cymrite and ending with the water-free phase celsian, or the first-formed authigenic phase (instead of harmotome). These sequences could thus parallel release of water during prograde diagenesis and metamorphism. The experimental results of Nitsch (1980) on the reaction $\text{cymrite} \rightarrow \text{celsian} + \text{H}_2\text{O}$ with increasing temperature are in agreement with this suggestion. In the Navarana Fjord area diagenesis has not been sufficiently intense to lead to the formation of cymrite. The relative deficiency in Ba of the hydrated Ba silicate (Fig. 5) probably reflects this less intensive diagenesis, with only limited Ba diffusion within the sedimentary sequence.

The textural relationships between the hydrated Ba-silicate, hyalophane and baryte are not unambiguous. Neither replacement relationships between sulphate and silicates nor dissolution of any pair of the minerals was observed. Likewise, baryte often surrounds the silicates. These obser-

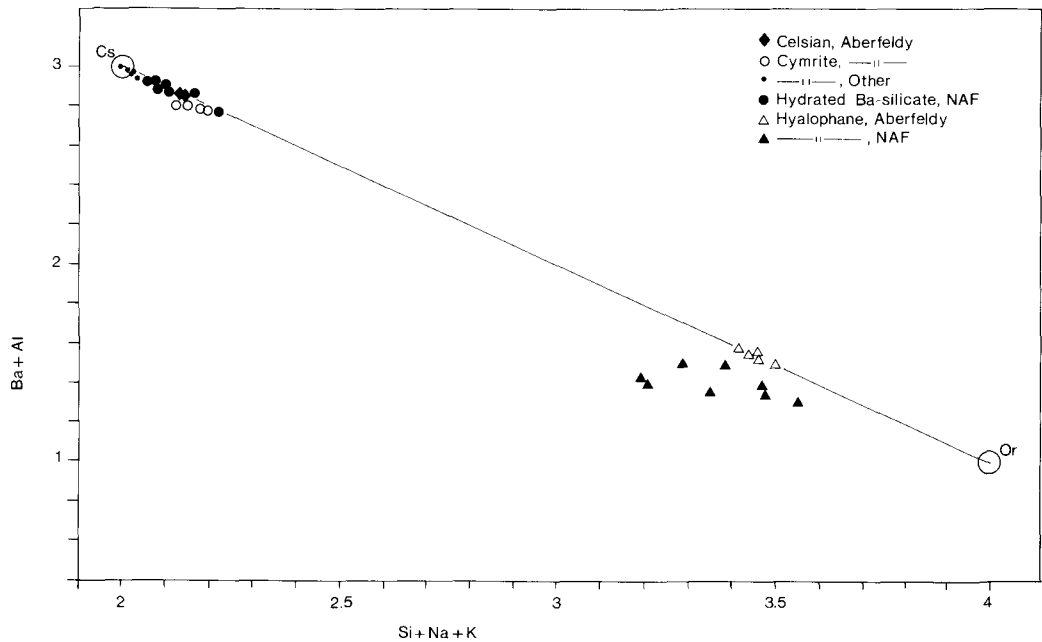


Fig. 4. Cation Ba + Al vs. Si + Na + K for the hydrated Ba silicate and hyalophane of the Navarana Fjord area (this study), and of other localities. Source of data and abbreviations as in Fig. 3.

vations are not in agreement with observations in similar rock types, e.g. Bjørlykke and Griffin (1973), of dissolution and replacement of baryte under reducing conditions, even though such reducing conditions in the sediments from the Navarana Fjord area are indicated by the presence of pyrite and up to 2 wt.% organic material. It would be expected that baryte was unstable (Garrels and Christ, 1965) and that Ba was released and incorporated in the silicates formed during diagenesis (Bjørlykke and Griffin, 1973). However, from textural evidence it seems most probable that both minerals grew contemporaneously in the sediment during diagenesis. According to Garrels and Christ (1965), baryte can be stable under reducing conditions if the environment is calcareous and alkaline, such as the overall case in the sedimentary rocks of the 'unnamed shelf and slope sequence' of the Navarana Fjord area.

High contents of barium in cherts associated with shale-hosted Zn–Pb–Ba mineralizations are well known from e.g. Aberfeldy, Scotland (Coats *et al.* 1980); Selwyn Basin, Yukon, and North West Territories; Silvermines, Ireland (Large, 1980). According to Russell *et al.* 1984) the Ba and SiO₂ in the Aberfeldy mineralization is of exhalative hydrothermal origin, whereas the alu-

minium necessary for the formation of the great amount of Ba silicates present was added to the sea from the deeply weathered Greenland–North-American shield. According to Boström *et al.* (1973) and Brongersma-Sanders (1966), an alternative mechanism for Ba enrichment in pelagic sediments relates accumulation of Ba-containing siliceous organisms. For the chert in the slope equivalent of the Amundsen Land Group it is considered most probable that both the silica and barium are of hydrothermal origin as the water column at the time of deposition was too shallow to accumulate radiolaria (as opposed to the deep-water sequence of the Amundsen Land Group (Surlyk and Hurst, 1984)). Moreover, no textural evidence is in favour of a pelagic origin, whereas both the SiO₂ and Ba could have been dissolved from the underlying Buen Formation which is weakly enriched (0.1–0.2 wt.%) in Ba.

The excess aluminium in the Navarana Fjord area could be similar in origin to that in the Aberfeldy mineralization. However, as the amounts of Ba silicates in the Navarana Fjord area are very much smaller than in the Aberfeldy area, it is as likely that the small amount of dissolved aluminium needed for the precipitation of Ba-silicates simply was the normal content of dissolved aluminium in seawater.

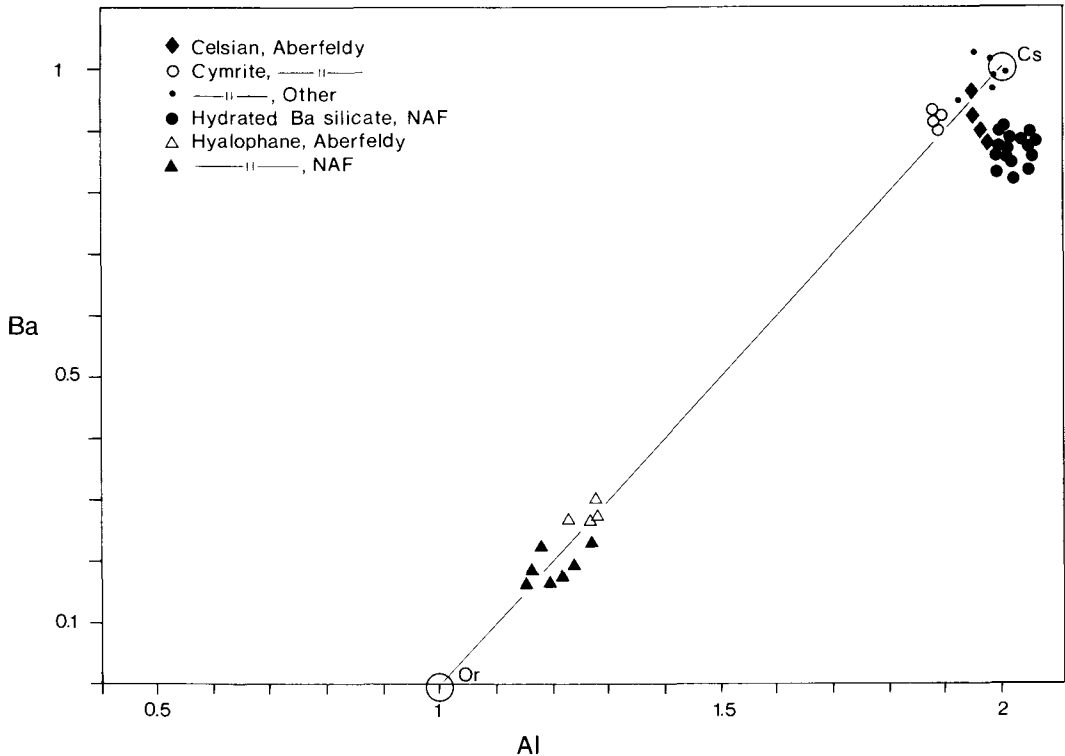


FIG. 5. Ba vs. Al (recalculated on the basis of 8 oxygens) for hydrated Ba-silicate and hyalophane of the Navarana Fjord area and other localities. Diagram modified after Fortey and Beddoe-Stephens (1982). Source of data and abbreviations as in Fig. 3.

Conclusion

The hydrated Ba-silicate discovered in unmetamorphosed organic-rich cherts from the Amundsen Land Group in central North Greenland agrees with the formula $\text{BaAl}_2\text{Si}_2\text{O}_8$ on an anhydrous basis, but a total of only 83 wt.% oxides suggests the presence of 4 moles H_2O in the mineral structure. These results indicate that the unknown mineral may be related to cymrite ($\text{BaAl}_2\text{Si}_2\text{O}_8 \cdot \text{H}_2\text{O}$) and represent an intermediate stage in a prograde diagenetic sequence of minerals from e.g. harmotome, through hydrated Ba silicate, to cymrite and ending with celsian on increasing P - T conditions. During deposition of the Amundsen Land Group, SiO_2 and Ba were dissolved most probably from the underlying Buen Formation and supplied to the starved shelf and slope basin by hydrothermal activity. Barium and silica was most probably kept as a silica gel on the sea floor (Large, 1980) and during later diagenesis, under relatively reducing and calcareous conditions, baryte and Ba-silicates were formed simultaneously.

The diagenetic process was not very intensive. This resulted in a deficiency in Ba of the Ba silicates, and in the lack of the transformation of the highly hydrated Ba silicate to cymrite.

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