Canadian Mineralogist,
Vol. 12, pp. 188-192 (1973)

LAUMONTITE AND BARIAN-STRONTIAN HEULANDITE FROM THE BLAIRMORE GROUP (CRETACEOUS), ALBERTA.

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ABSTRACT

Laumontite and barian-strontian heulandite occur with other authigenic minerals as pore-filling cement in the non-marine standstones of the Lower Cretaceous Blairmore Group in the southern Alberta foothills, Canada. Heulandite contains up to 6.9 wt % BaO and 4.1 wt % SrO. Distribution coefficients for Ba, Sr, Na and K for coexisting laumontite and heulandite indicate that Sr and Ba are strongly favored in the heulandite structure. This element distribution is thought to be largely controlled by crystal chemical factors and the limited thermal stability of the zeolites.

INTRODUCTION

Zeolites occur with other authigenic minerals as pore-filling cement in the non-marine sandstones of the Lower Cretaceous Blairmore Group in the southern Alberta Foothills, Canada (Mellon & Wall 1963; Mellon 1967, Fig. 1). The Blairmore Group is essentially a wedge, 600 to 6,500 feet thick, of non-marine sandstones, shales, and conglomerates (Norris 1964). A variety of authigenic minerals, including calcite, quartz, chlorite, kaolinite, dolomite, and zeolites occur in different parts of the Blairmore Group. Zeolites are restricted to plagioclase-rich sandstones of the Beaver Mines Formation (Mellon 1967) and are more common in the medium- to coarse- grained sandstones. Zeolite distribution is irregular, both on the scale of an outcrop and a hand specimen.

FIG. 1. Index map showing the locations of stratigraphic sections sampled in the present study. 1. Burnt Timber Creek; 2. Sheep River; 3. Ma Butte; 4. Mill Creek.

Because of the occurrence of the petrologically significant, chemically equivalent assemblages, laumontite and calcite—kaolinite—quartz (plus H_2O and CO_2 to balance the reaction, Zen 1961; Coombs *et al.* 1970) in the Blairmore Group, an investigation of the stratigraphic distribution, petrography, and mineralogy of these minerals and associated mineral assemblages was undertaken. In this paper, we report the results of an electron microprobe and *x*-ray diffraction study on coexisting laumontite and barian-strontian heulandite. A second paper will provide an interpretation of the P(load)-T-fluid composition conditions of crystallization of the authigenic minerals.

PETROGRAPHY

Laumontite occurs chiefly as an intergranular pore-filling, but in some samples it occurs within plagioclase (now albite) and plagioclase-rich volcanic rock fragments. The distribution of laumontite is irregular on the scale of a thin section. In some samples, laumontite and chlorite cement occur in one-half a thin section and chlorite, illite, quartz or calcite occur as cements in the other half of the section. Laumontite usually formed after chlorite was deposited as a pore-lining.

Heulandite occurs only as intergranular cement, with single crystals, or less commonly, two large crystals or a mass of small, interlocking crystals, filling pores. Deposition of heulandite follows deposition of chlorite as a pore-lining. Heulandite completely fills the remaining pore space, except in one case where calcite occurs with heulandite and was apparently deposited later.

Heulandite and laumontite often occur in the same hand samples, but are rarely observed in direct contact. In a few cases, electron beam scanning revealed single euhedral heulandite crystals in the center of pores, with laumontite rimming the heulandite.

X-RAY DIFFRACTION STUDY

Heulandite was detected by x-ray diffraction in six samples. One sample which contained very little laumontite (RS-SC-1) [1] was chosen for a detailed x-ray diffraction study and a heulandite concentrate was separated by heavy liquids.

X-ray diffraction data for Blairmore heulandite (Table 1) are compared with data reported by Černý & Povondra (1969). Agreement of the Blairmore data with that reported by other workers is reasonably good.

The unit cell dimensions reported for heulandite were kindly obtained by R. C. Erd of the U.S. Geological Survey using single crystal measurements for the initial parameters and x-ray powder diffraction data for the least squares unit cell refinement (using the program of Evans *et al.* 1973).

Boles (1972) has demonstrated a correlation between the area of the *ac* plane in heulandite and the Σ Mg + Ca + Sr + Ba ions per unit cell and the Al ions per unit cell. A plot of the Blairmore unit cell data and composition on these diagrams shows good agreement with other heulandites (Table 2).

Laumontite

Laumontite was detected by x-ray diffraction in 15 samples. One sample, which contained little or no heulandite, (RS-SC-6), was separated by heavy liquids.

Reflections obtained from Blairmore laumontite (Table 3) compare well with those reported by Lapham (1963). Unit cell dimensions for Blairmore laumontite (Table 2) agree well with those given by Lapham (1963), Coombs 1952), Liou (1971), and Schramm & Fisher (1971).

ELECTRON MICROPROBE ANALYSES

Introduction

TABLE 1. X-RAY DIFFRACTION DATA FOR Ba-Sr HEULANDITE

Ba-Sr heulandite* Blairmore Group				Sr heulandite** Cerny & Povondra (1969)			
hkl	d_{calc}Å	d_{meas}Å	*I*	*hkl*	d_{calc}Å	d_{meas}Å	*I*
110	11.900						
020	8.993	8.99	100	020		8.952	3
200	7.935	7.93	4	200		7.949	7
$\bar{1}11$	6.860						
$\bar{2}01$	6.797			$20\bar{1}$		6.831	1
001	6.650	6.67	3	001		6.652	1
220	5.950	5.96	3	220		5.930	1
130	5.608			130		5.598	0.5
$\bar{2}21$	5.423						
021	5.347			021		5.334	0.5
$\bar{3}11$	5.259	5.26	7	$31\bar{1}$		5.268	3
111	5.123	5.12	8	111		5.131	7
310	5.075			310	5.0743	5.073	3
$\bar{1}31$	4.664	4.664	17	$13\bar{1}$	4.6515	4.656	3
040	4.496	4.496	17	040	4.4641	4.461	5
$\bar{4}01$	4.367	4.363	7	$40\bar{1}$	4.3697	4.368	5
201	4.250			201	4.2635	4.270	3
$\bar{3}31$	4.053			$33\bar{1}$	4.0421	4.044	3
131	3.989	3.989	12				
400	3.967	3.976	12	400,421		3.940	10
330	3.967						
$\bar{4}21$	3.928						
240	3.912	3.914	15	240	3.8911	3.892	1
221	3.843			221	3.8473	3.853	1
$\bar{2}41$	3.750	3.752	7				
041	3.724						
$\bar{2}02$	3.711	3.709	6	$20\bar{2}$	3.7276	3.731	3
420	3.630			420	3.6271	3.626	0.5
$\bar{3}12$	3.564	3.567	6	$31\bar{2}$	3.5762	3.576	5
$\bar{1}12$	3.521			$11\bar{2}$	3.5366	3.534	0.5
150	3.508			150	3.4842	3.490	1
$\bar{5}11$	3.474						
$\bar{2}22$	3.430	3.427	5	$22\bar{2}$	3.4398	3.437	5
$\bar{4}02$	3.399			$40\bar{2}$	3.4085	4.413	3
311	3.396						
002	3.325	3.324	5				
$\bar{1}51$	3.237						
$\bar{4}22$	3.179	3.184	8	$42\bar{2}$	3.1843	3.181	5
$\bar{4}41$	3.133	3.132	9				
510	3.126			510	3.1263	3.126	3
022	3.119						
$\bar{3}32$	3.109						
241	3.089			241	3.0832	3.079	1
$\bar{1}32$	3.080	3.079	4				
$\bar{5}31$	3.049			$53\bar{1}$	3.0439	3.046	1
$\bar{5}12$	3.035	3.034	8				
$\bar{3}51$	3.010						
060	2.998	2.997	22				
331	2.995			331	2.9955	3.002	0.5
151	2.984						
440	2.975	2.976	19	440		2.971	7
350	2.975			060			
$\bar{6}21$		2.800	5	401		2.888	0.5
$\bar{2}61$		2.742	11	530		2.801	8
170		2.536	7	$26\bar{1}$		2.724	1
$\bar{4}62$		2.247	7	042		2.672	1
$\bar{1}91$		1.929	3	$62\bar{2}$		2.633	0.5
$\bar{7}71$		1.797	8	$35\bar{2}$		2.557	0.1
$\overline{10}.02$		1.770	2	620		2.534	0.5
462		1.734	3	$55\bar{1}$		2.509	0.5
				351		2.487	1
				441		2.423	5
				312		2.365	1

*Calculated spacings and indices from least squares refinement of powder data using program of Evans *et al.* (1973). All calculated spacings ≥2.975Å listed. Measured values from split of RS-SC-1; sample contained small amounts of laumontite and quartz. X-ray diffractometer data are: Cu/Ni radiation, $\lambda CuK\alpha_1$=1.54051Å, silicon metal used as internal standard; scanned at 1/8° 2θ per minute from 4°-70°. Chart speed 1/8" per minute.

**From a Guinier-de Wolff photograph, quartz internal standard, Cu/Ni radiation, λ=1.5418Å. Intensities estimated visually.

Polished, carbon-coated mounts of 16 zeolite mineral separates were analyzed for Ca, Na, K, Ba, Sr, Si, Al, Fe, and Mg, using an ARL-EMX electron probe microanalyzer. Operating conditions and standards used were: beam spot size — 15 μm; beam current — 0.1 μamp; Ca, Mg — clinopyroxene; Si, Al, Na — albite; K — ortho-

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[1] Refers to samples held at Department of Geology, University of Calgary.

clase; Ba — hyalophane; Sr — strontianite; and Fe — hematite. Data were corrected using factors given by Albee & Ray (1970). Because water cannot be determined by electron microprobe, water content was estimated by subtracting the sum of all other oxides from 100 per cent.

Heulandite

Analyses for 5 Blairmore heulandites are reported in Table 4. The prominent feature of the Blairmore heulandites is their notable content of Ba and Sr, which was first detected by non-dispersive *x*-ray analysis using the solid state detector on the electron microprobe (Jones & Ghent 1971).

The ideal formula of heulandite is $(Ca,Na)_2Al_2Si_7O_{18}\cdot 6H_2O$ (Merkle & Slaughter 1968). Černý & Povondra (1969) suggest that a more realistic formula, based on 72 oxygens, is $(Na,K)_x(Ca,Sr,Ba)_y(Al,Fe)_{x+2y}Si_{36-(x+2y)}O_{72}\cdot nH_2O$, where $x = 0$ to 2.6, $y = 2.7$ to 4.3, $x + y = 6.8$ to 9.7, and $n = 20$ to 30. Considering the difficulty of obtaining accurate analyses of zeolites by electron microprobe, the structural formulas of Blairmore heulandites show reasonable agreement with those reported by other workers. Si/Al ratios of Blairmore heulandites are <4.0, which fits the definition of heulandite proposed by Boles (1972).

TABLE 2. HEULANDITE AND LAUMONTITE CELL DIMENSIONS (Å)

Heulandite	1	2	3	4
a	17.717±.009	17.71	17.73	17.722±.012
b	17.986±.006	17.84	17.82	17.856±.012
c	7.424±.003	7.46	7.43	7.458±.007
β	116°24'±2.2'	116°20'	116°20'	116.37±.06°
V Å³	2119±1	2111.4	2103.36	2114.5

1. Heulandite, RS-SC-1. 2. Heulandite, Deer *et al.* (1963).
3. Heulandite, Merkle & Slaughter (1968).
4. Polycationic, strontian heulandite, Černý & Povondra (1969).

Laumontite	1	2	3	4	5
a	14.712±.003	14.75±.05	14.75±.03	14.737±.004	14.740±.015
b	13.090±.002	13.083±.02	13.10±.02	13.066±.002	13.072±.012
c	7.555±.002	7.57±.05	7.55±.01	7.550±.003	7.549±.008
β	111°42.8'±1.6'	112°	112°±.2°	111.97°±.2°	111.9°±.1°
V Å³	1351.7±.4	1344.7	1342.9	1348.2	1349.6

1. RS-SC-6. 2. PDF card 15-276 (Lapham 1963).
3. Coombs (1952) cited by Lapham (1963). 4. Liou (1971).
5. Schramm & Fischer (1971).

TABLE 3. LAUMONTITE *X*-RAY DIFFRACTION DATA

	1		2	
hkl	*d*(Å)	*I*	*d*(Å)	*I*
110	9.45	100	9.49	100
200	6.84	75	6.86	35
020	6.55	1	6.54	2
201,→011	6.17	8	6.19	2
111	5.05	13	5.052	6
220	4.73	25	4.731	20
221	4.49	23	4.500	8
130,→201	4.17	100	4.156	60
131	3.77	8	3.768	2
401	3.65	38	3.667	14
002,→221	3.51	58	3.510	30
400,→131	3.42	17	3.411	8
312,→012	3.35	17	3.367	4
040	3.27	48	3.272	20
331,→311	3.20	25	3.205	8
330	3.15	23	3.152	16
420,→112	3.03	40	3.033	25
240,→041	2.95	5	2.950	4
060	2.18	9	2.180	6

1. RS-SC-6 2. PDF card 15-267 (Lapham 1963)

The BaO content of Blairmore heulandites, up to 6.9 wt %, is the highest value reported. Černý & Povondra (1969) have reported a heulandite with SrO = 4.44 wt % and BaO = 2.44 wt %; Ogawa (1967) has reported a heulandite with 4.8 wt % BaO (SrO was not reported). Thus, the Blairmore heulandites show the highest combined BaO + SrO content known to us.

Laumontite

Electron microprobe analyses of 11 laumontites from the Blairmore Group and 3 laumontite-leonhardites (Coombs 1952) are presented in Table 5.

The general formula for laumontite is $Ca_x(Na,K)_yAl_{2x+y}Si_{6-(2x+y)}O_{12}\cdot 4H_2O$, where ideally $x = 1$ and $y = 0$ (modified from Coombs 1952). Analyses of Blairmore laumontites show excellent agreement with this general formula.

Substitutions of pairs of alkali ions for Ca without variation in the Si-Al ratio may occur in laumontite (Coombs 1952). Amirov *et al.* (1967) report that the laumontite structure has ordered Si-Al tetrahedra. For coupled substitutions such as Na, Si for Ca, Al, it is possible

TABLE 4. ELECTRON MICROPROBE ANALYSES OF HEULANDITES FROM THE BLAIRMORE GROUP, ALBERTA

wt %	RS-SC-1[a]	MC-10	RS-SC-4	RS-SC-5	OG-SC-6	1	2
SiO_2	56.1	57.9	59.2	55.6	55.9	55.77	57.66
Al_2O_3	15.5	14.7	15.1	15.7	14.8	15.88	16.12
FeO*	0.1	0.2	0.1	<0.1	<0.1	---	1.44[b]
MgO	<0.05	<0.05	<0.05	<0.05	<0.05	0.085	0.02
CaO	4.4	3.3	3.0	3.1	3.2	3.04	1.59
SrO	3.3	1.5	2.9	4.1	2.8	4.44	---
BaO	5.6	6.9	6.0	4.6	5.7	2.44	4.80
Na_2O	0.7	0.5	0.3	0.6	0.4	1.54	1.98
K_2O	0.6	0.4	1.8	1.8	2.0	1.34	0.30
SUM	86.3	85.4	88.4	85.5	84.8	12.45+	11.48+
H_2O**	13.7	14.6	11.6	14.5	15.2	3.07-	3.48-
						$+=H_2O^+$,	$-=H_2O^-$
	Number of ions on anhydrous basis of 72 oxygens						
Si	27.01	27.83	27.70	27.01	27.40	26.97	27.46
Al	8.79	8.33	8.33	8.99	8.55	9.05	9.04
Fe	0.04	0.08	0.04	0.00	0.00	---	0.52
Ca	2.27	1.70	1.50	1.61	1.68	1.58	0.81
Sr	0.92	0.42	0.79	1.16	0.80	1.25	---
Ba	1.06	1.30	1.10	0.88	1.09	0.46	0.90
Na	0.65	0.47	0.27	0.57	0.38	1.44	1.83
K	0.37	0.25	1.07	1.12	1.25	0.83	0.18
Si/Al	3.07	3.34	3.33	3.00	3.20	2.98	3.04

1. Strontian heulandite (Černý & Povondra 1969)
2. Barian heulandite (b=includes 0.027% MnO; Ogawa 1969)
--- not reported; FeO*=total Fe as FeO;
H_2O** is water content estimated from 100-sum.
[a] Refers to samples held at the Dept. of Geology, University of Calgary.

that ordering of the Al-Si tetrahedra makes this type of substitution more difficult.

DISTRIBUTION OF ELEMENTS BETWEEN COEXISTING LAUMONTITE AND BARIAN-STRONTIAN HEULANDITE

The distribution of Ba, Sr, Na, and K between coexisting laumontite and heulandite, summarized as distribution coefficients with respect to Ca, is given in Table 6. The data indicates that (a) there is more total substitution of Ba, Sr, Na, and K in the heulandite structure; (b) Ba and Sr are strongly favored in the heulandite structure; and (c) K and Na are slightly favored in the heulandite structure, with the exception of one sample.

Two explanations for this distribution seem worthy of discussion. First, the element distribution shows no crystal-chemical control and is entirely a function of the different pressure-temperature-fluid composition conditions under which the two minerals crystallized. This implies that laumontite could also show extensive Ba-Sr substitution under appropriate conditions.

A second possibility, which we favor, is that the heulandite structure is capable of more extensive Ba-Sr substitution than is laumontite. Published chemical analyses of laumontites and heulandites suggest that heulandites generally show more extensive solid solution of Ba and Sr than do laumontites. We feel that these differences in amount of Ba-Sr solid solution shown by laumontite and heulandite can be explained by a comparison of the atomic structures of the two minerals.

TABLE 5. ELECTRON MICROPROBE ANALYSES OF LAUMONTITES FROM THE BLAIRMORE GROUP, ALBERTA

wt %	RS-SC-5	RS-SC-6	RS-SC-1	RS-SC-7	RS-SC-8	RS-SC-10	OG-SC-6	OG-SC-8	MB-8	MC-10	MC-11
SiO_2	51.8	52.3	53.7	52.9	52.8	51.5	52.8	52.4	52.0	51.8	52.0
Al_2O_3	20.8	20.5	20.6	20.7	20.9	21.1	20.5	20.6	21.0	21.2	21.4
CaO	10.5	9.7	10.3	10.6	10.5	10.3	9.4	10.0	10.6	9.1	10.4
SrO	0.2	0.3	0.2	0.3	0.3	0.3	0.2	0.3	0.2	0.3	0.2
BaO	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
Na_2O	0.2	0.7	0.1	<0.05	<0.05	0.2	0.2	<0.05	0.2	1.9	0.8
K_2O	1.3	1.2	1.0	1.2	1.3	1.5	1.7	1.8	0.8	1.0	0.6
SUM	84.8	84.7	85.9	85.7	85.8	84.9	84.8	85.1	84.8	85.3	85.4
H_2O*	15.2	15.3	14.1	14.3	14.2	15.1	15.2	14.9	15.2	14.7	14.6
Number of ions on anhydrous basis of 12 oxygens											
Si	4.07	4.11	4.14	4.10	4.09	4.05	4.13	4.10	4.07	4.05	4.05
Al	1.93	1.90	1.87	1.89	1.91	1.95	1.89	1.90	1.94	1.95	1.96
Ca	0.88	0.82	0.85	0.88	0.87	0.87	0.79	0.84	0.89	0.76	0.87
Sr	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
Ba	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Na	0.03	0.11	0.01	0.00	0.00	0.03	0.03	0.00	0.03	0.29	0.12
K	0.13	0.12	0.10	0.12	0.13	0.15	0.17	0.18	0.08	0.10	0.06

H_2O*is water content estimated from 100-sum.

TABLE 6. DISTRIBUTION COEFFICIENTS FOR Ba, Sr, Na, and K BETWEEN COEXISTING HEULANDITE AND LAUMONTITE

Sample	*Kd* Ba*	*Kd* Sr	*Kd* Na	*Kd* K
RS-SC-1	>65.5	51.5	16.4	1.4
RS-SC-5	>77.9	69.5	10.2	4.7
OG-SC-6	>83.7	41.2	5.9	3.5
MC-10	>95.1	13.8	0.7	1.1

$$*Kd\text{Ba} = \frac{\text{moles BaO}}{\text{moles CaO}}\ \text{heulandite} \ / \ \frac{\text{moles BaO}}{\text{moles CaO}}\ \text{laumontite}$$

Other *Kd*'s are defined similarly.

Laumontite has one of the highest specific gravities of any zeolite (2.3) and this can be correlated with the absence of large diameter channels in its structure (Amirov *et al.* 1967). Ca ions in laumontite are bonded to four oxygens and two water molecules having the geometry of a slightly distorted trigonal prism. This type of bonding is not observed in any other zeolite (Amirov *et al.* 1967). The Ca ions are situated under silica tetrahedra and not on the axes of ring channels.

In contrast to laumontite, heulandite has a lower specific gravity (2.1) and larger-diameter, more numerous channels in its structure (Merkle & Slaughter 1968). Merkle & Slaughter (1968) report that Ca ions occur both near the sides of open ring channels and at the intersections of the ring channels. Ca ions in heulandite are bonded to three oxygens from alumina tetrahedra and five water molecules. According to Černý & Povondra (1969), some cationic sites seem to be preferentially occupied by bivalent cations, and alkalies (Na + K) tend not to substitute for bivalent cations but occur in addition to them to compensate for Si-Al substitution.

Because the Ca ions in laumontite are in smaller diameter sites and are strongly bonded to four oxygens from silica tetrahedra as compared to three oxygens in heulandites, one might predict that substitution of Ba and Sr for Ca would be favored in the heulandite structure. Because of the limited thermal stability of the two minerals, one would also predict that these fractionation trends should hold for other coexisting laumontites and heulandites.

ACKNOWLEDGEMENTS

The writers wish to thank R. C. Erd for his help in obtaining, processing, and interpreting x-ray diffraction data. K. J. Murata of the U.S. Geological Survey made several helpful comments on the manuscript. The writers are responsible for any errors. Ghent acknowledges financial support from NRC — Operating Grant A-4379.

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Manuscript received July 1973.