

## MODE OF OCCURRENCE, CHEMICAL VARIATION AND GENESIS OF MORDENITE AND ASSOCIATED ZEOLITES FROM THE MORDEN AREA, NOVA SCOTIA, CANADA

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### ABSTRACT

Selected zoned and homogeneous amygdaloids and veins of zeolite from the earliest Jurassic North Mountain Basalt, in Nova Scotia, have been examined in thin section and by SEM, XRD and electron-microprobe analysis. Zoned sequences of zeolites in amygdaloids and veins in the North Mountain basalt show a succession of clinoptilolite-Na → mordenite → heulandite, clinoptilolite-Ca, or epistilbite → stilbite, barrerite, or stellerite, and are interpreted as developing under falling temperatures. Locally, fractures within zeolites have been filled with smectite and at least three different Mg-Fe-Na-aluminosilicates. Barite, Cu, Ni and Au precipitates occur throughout some zoned amygdaloids. The high temperatures required for the precipitation of labradorite and mordenite, the repetitive zonation in some amygdaloids, and the occurrence of metal precipitates all suggest an origin for the zeolites by active hydrothermal circulation, not by burial metamorphism as implied by previous authors. Formation of mordenite was favored by hydrothermal circulation of alkaline lake waters and extraction of Na from underlying evaporites of the Blomidon Formation.

**Keywords:** zeolite, mordenite, North Mountain basalt, hydrothermal circulation, Nova Scotia.

### SOMMAIRE

Une sélection d'amygdules à zéolites, soit zonées, soit homogènes, et de veines provenant des coulées de basalte de North Mountain, en Nouvelle-Ecosse, d'âge jurassique précoce, a été examinée en lame mince, et caractérisée par microscopie électronique à balayage, diffraction X et microsonde électronique. Les séquences de zéolites suivantes ont été observées dans les amygdules et les veines: clinoptilolite-Na → mordenite → heulandite, clinoptilolite-Ca, ou epistilbite → stilbite, barrerite ou stellerite; elles seraient dues à la chute progressive de la température. À petite échelle, les fissures dans les zéolites ont été remplies avec une smectite et au moins trois aluminosilicates de Mg-Fe-Na distincts. La barite, et le cuivre, le nickel et l'or ont été précipités partout dans certaines amygdules zonées. La température élevée indiquée par la précipitation de la labradorite et de la mordenite, la zonation répétée dans certaines amygdules, et la présence de phases métalliques font penser à une origine des zéolites par circulation hydrothermale active, plutôt que par recristallisation métamorphique due à l'enfouissement, comme le préconisaient les auteurs antérieurs. La circulation d'eau alcaline d'origine lacustre et l'extraction de sodium des évaporites sous-jacentes de la Formation Blomidon auraient favorisé la formation de la mordenite.

(Traduit par la Rédaction)

**Mots-clés:** zéolite, mordenite, basalte de North Mountain, circulation hydrothermale, Nouvelle-Ecosse.

### INTRODUCTION

Morden (Fig. 1) is the type locality of the zeolite mordenite (How 1864, Walker & Parsons 1922). The hosting basalt flows are part of the middle unit of the North Mountain Basalt (Wark & Clarke 1980), a sequence of subaerial tholeiitic lava flows in the most northerly late Triassic – early Jurassic rift basin in the Appalachians. The basalts contain a variety of zeolites and quartz (agate, chalcedony, jasper, and amethyst), together with native copper and magnetite (Donohoe *et*

*al.* 1992). The North Mountain Basalt is overlain by lacustrine sediments of the Scots Bay Formation, which include siliceous hot-spring deposits (DeWet & Hubert 1989).

In the Morden area, the North Mountain Basalt is about 250 m thick, comprises 19 flows, and overlies the late Triassic Blomidon Formation (principally red shale, < 370 m). Zeolites are most abundant in amygdaloids in the basal and upper parts of flows. Zeolites also occur in irregular cavities, veins, and pipes and “bubble trains”. The samples studied come from between

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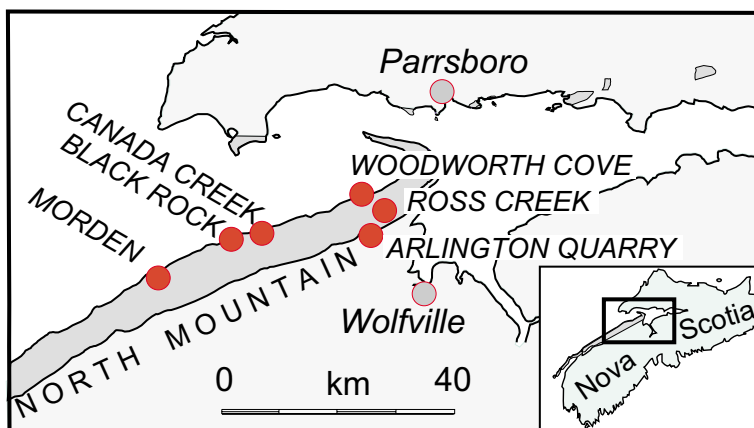


FIG. 1. Map showing the North Mountain basalt (dark shading) in Nova Scotia and the location of Morden and other sampling localities (cf. Table 1).

TABLE 1. LOCATION, MODE OF OCCURRENCE AND ASSOCIATED MINERALS, MORDEN AREA, NOVA SCOTIA

Sample	Location	Mode of occurrence	Zeolites	Associated minerals
W12*	Ross Creek	zoned amygdale	Cpt	Qtz
W26*	Ross Creek	zoned amygdale	Mor, Cpt, Stl, Heu	Qtz
W29	Black Rock	vein	Heu	Qtz
W30	Black Rock	vein	Heu, Stb	
W59	Morden	vein	Mor, Cpt, Heu	Qtz
Z1*	Morden	amygdale	—	Cal, Brr, Hem, copper
Z2*	Morden	amygdale	Mor, Heu, Cpt, Esb	Apo, Qtz, Cel
Z3*	Morden	amygdale	Mor	Qtz, Cel
Z26*	Arlington Quarry	zoned amygdale	Mor, Heu, Cpt	Cel
Z41	Black Rock	amygdale	—	Apo-Na, Qtz
Z247*	East of Ross Creek	zoned vein	Mor, Heu	
Z248	East of Ross Creek	vein	Cpt	Qtz
Z249	East of Ross Creek	zoned amygdale	Mor, Heu	Fe-Mg-Na aluminosilicates
Z250*	East of Ross Creek	vein	Cpt	Qtz
Z642*	Woodworth Cove	zoned amygdale	Mor, Brr, Stb	
Z643*	West of Canada Creek	zoned amygdale	Cpt, Heu	
Z645*	Woodworth Cove	zoned amygdale	Mor, Cpt, Heu	smectite
Z649*	Ross Creek	zoned amygdale	Mor, Heu, Cpt	smectite, Fe-Mg-Na aluminosilicates
Z650*	Ross Creek	zoned amygdale	Mor, Esb, ?Heu	Pl, smectite, Fe-Mg-Na aluminosilicates

\* Zeolite identification in these samples has been confirmed by X-ray diffraction and energy-dispersion spectra.

In many samples, in addition a thin layer of celadonite occurs at the rim of the amygdale or vein. Symbols: Apo: apophyllite, Apo-Na: natro-apophyllite, Brr: barrerite, Brr: barite, Cal: calcite, Cel: celadonite, Cpt: clinoptilolite, Heu: heulandite, Mor: mordenite, Pl: plagioclase, Qtz: quartz, Stb: stilbite, Stl: stellerite.

Morden and Ross Creek (Fig. 1), and include amygdaloids and veins, many of which show a zonal distribution of mineral species (Aumento 1966). Aumento also mapped a geographic sequence of zeolite zones within the North Mountain Basalt, with mordenite restricted to a small area around Morden. Neither Aumento nor previous workers such as Walker & Parsons (1922) were able to provide a definitive explanation of the geographic distribution of zeolites, although subsequent authors (*e.g.*, Adams 1980) have drawn parallels with the zonal sequence of zeolites in piles of basalt flows such as at Antrim, in Ireland (Walker 1960) and in Iceland (*e.g.*, Neuhoff *et al.* 1999).

The purpose of this paper is to establish the sequence and process of formation of the zeolites in this classic area and to determine if the zeolites are a product of hydrothermal mineralization or burial metamorphism. In this paper, I therefore focus on the critical assemblages of zeolites to address these issues.

#### SAMPLING AND ANALYTICAL METHODS

A large number of samples were collected in the field; eighteen have been studied in detail: three homogeneous amygdaloids, nine zoned amygdaloids and six veins (one of which is zoned) (Table 1). Zeolites in these samples were analyzed on polished thin section with a JEOL-733 electron microprobe having four wavelength spectrometers and a Tracor Northern 145 eV energy-dispersion detector. The beam was operated at 15 kV and 15 nA, with a beam diameter of 10–20  $\mu\text{m}$ . I used geological mineral standards and, as a control on the stoichiometry, I chose a sample of scolecite that had been extensively analyzed, thereby minimizing the consequence of not including structurally bound  $\text{H}_2\text{O}$  and hydroxyl in the matrix correction (Tingle *et al.* 1996).

Zeolites were also examined in back-scattered electron images. The data were reduced using a Tracor Northern ZAF matrix-correction program. A subset of about 200 microprobe analyses were chosen on the basis that their chemical balance error ( $E = \{(Al + Fe^{3+}) - Al_{theor.}\} \times 100/Al_{theor.}$ ; Passaglia 1975) is low ( $< 10\%$ ). These compositions are used in the various plots. Representative results of analyses are given in Table 2, and the nomen-

clature follows the IMA conventions (Coombs *et al.* 1997): a few compositions used to identify particular mineral occurrences have chemical balance errors in excess of 10%.

Powders of zeolites, extracted from most of the analyzed samples, were analyzed by X-ray diffraction using a packed powdered sample,  $CoK\alpha$  radiation and a scan from  $2^\circ$  to  $42^\circ 2\theta$  at a speed of  $0.25^\circ/2\theta/\text{min}$ .

TABLE 2. SELECTED RESULTS OF ELECTRON-MICROPROBE ANALYSES OF REPRESENTATIVE ZEOLITES, MORDEN AREA, NOVA SCOTIA

Sample Analysis Zeolite	Z2 1 Mor	Z2 2 Mor	Z2 3 Cpt	Z2 4 ?Esb	Z3 5 Mor	Z645 6 Mor	Z645 7 Mor	Z645 8 Heu	Z645 9 Cpt	Z645 10 Heu	Z649 11 Mor	Z649 12 Mor	Z649 13 Heu	Z649 14 Heu
SiO <sub>2</sub> wt. %	70.03	66.85	69.05	63.02	70.52	68.65	70.04	59.80	68.77	63.00	68.15	70.29	59.79	59.32
Al <sub>2</sub> O <sub>3</sub>	12.08	11.01	13.05	15.07	11.83	11.79	10.56	16.52	12.67	14.88	11.13	11.69	15.89	14.46
FeO <sub>i</sub>	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.51	1.08	n.d.	0.25
MgO	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.33	n.d.	0.30
CaO	2.27	1.42	5.68	7.35	0.63	1.52	0.58	7.21	6.08	5.47	0.69	1.48	6.53	3.96
Na <sub>2</sub> O	3.66	4.85	0.79	0.42	5.94	3.66	4.85	0.45	0.31	1.75	5.33	3.96	0.65	3.09
K <sub>2</sub> O	1.11	1.63	0.52	n.d.	2.80	1.92	2.37	1.19	0.66	1.45	2.14	1.65	0.80	1.44
Total	89.15	85.76	89.09	85.86	91.72	87.54	88.40	85.17	88.49	86.55	87.95	90.48	83.66	82.82
Oxygen	(96)	(96)	(72)	(72)	(96)	(96)	(96)	(72)	(72)	(72)	(96)	(96)	(72)	(72)
Si <i>apfu</i>	39.99	40.00	29.53	28.19	39.78	40.05	40.61	27.30	29.62	28.21	39.97	39.90	27.63	27.91
Al	8.13	7.77	6.58	7.95	7.87	8.11	7.22	8.89	6.43	7.86	7.70	7.82	8.66	8.02
Fe	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.25	0.51	n.d.	0.10
Mg	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.28	n.d.	0.21
Ca	1.39	0.91	2.60	3.52	0.38	0.95	0.36	3.53	2.81	2.62	0.43	0.90	3.23	2.00
Na	4.05	5.63	0.66	0.36	6.50	4.14	5.45	0.40	0.26	1.52	6.06	4.36	0.58	2.82
K	0.81	1.24	0.28	n.d.	2.02	1.43	1.75	0.69	0.36	0.83	1.60	1.20	0.47	0.86
Si/Al	4.92	5.15	4.49	3.55	5.06	4.93	5.63	3.07	4.60	3.59	5.19	5.10	3.19	3.48
E(%)	6.5	-10.6	7.1	7.2	-15.2	8.6	-8.9	9.2	3.2	3.4	-6.8	5.4	15.1	0.3

Sample Analysis Zeolite	Z650 15 Mor	Z650 16 Mor	Z650 17 Esb	W26 18 Cpt	W26 19 Cpt	W26 20 Esb	W30 21 Heu	W30 22 Stb	W30 23 Stb	W59 24 Mor	W59 25 Cpt-Na	W59 26 Cpt	W59 27 Cpt
SiO <sub>2</sub> wt. %	65.57	70.08	58.52	64.73	71.35	60.43	60.29	57.21	58.41	68.11	65.26	70.45	66.12
Al <sub>2</sub> O <sub>3</sub>	10.15	10.25	16.37	12.35	11.38	14.99	15.99	16.20	15.86	11.42	12.23	10.97	12.63
FeO <sub>i</sub>	n.d.	0.32	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
MgO	0.29	n.d.	n.d.	n.d.	0.18	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
CaO	1.28	1.24	8.20	5.90	4.86	7.19	7.49	8.20	7.75	0.86	0.35	5.45	5.11
Na <sub>2</sub> O	4.32	4.87	0.65	0.33	0.48	0.38	0.59	0.59	1.12	4.62	4.69	0.45	0.59
K <sub>2</sub> O	0.83	1.07	n.d.	0.28	0.45	0.59	0.98	n.d.	n.d.	1.99	6.73	n.d.	1.26
Total	82.44	87.83	83.74	83.59	88.70	83.58	85.34	82.20	83.14	87.00	89.26	87.32	85.71
Oxygen	(96)	(96)	(48)	(72)	(72)	(48)	(72)	(72)	(72)	(96)	(72)	(72)	(72)
Si <i>apfu</i>	40.45	40.70	18.08	29.48	30.39	18.62	27.46	27.02	27.26	40.09	29.12	30.45	29.49
Al	7.38	7.02	5.96	6.63	5.71	5.44	8.59	9.02	8.73	7.93	6.43	5.59	6.64
Fe	n.d.	0.16	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Mg	0.27	n.d.	n.d.	n.d.	0.11	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Ca	0.85	0.77	2.71	2.88	2.22	2.37	3.66	4.15	3.88	0.54	0.17	2.52	2.44
Na	5.17	5.49	0.39	0.29	0.40	0.23	0.52	0.54	1.01	5.27	4.06	0.38	0.51
K	0.65	0.79	n.d.	0.16	0.25	0.23	0.57	n.d.	n.d.	1.49	3.83	n.d.	0.72
Si/Al	5.48	5.80	3.03	4.45	5.32	3.42	3.20	3.00	3.12	5.06	4.53	5.47	4.44
E(%)	-8.3	-8.3	2.5	6.7	7.7	4.6	2.2	2.0	-0.4	0.9	-21.8	3.0	8.7

Symbols: Mor: mordenite, Heu: heulandite, Cpt: clinoptilolite, Stb: stilbite, Esb: epistilbite. n.d.: not detected.

Unit-cell parameters were refined using the algorithm of Appleman & Evans (1973). Natural and broken surfaces of zeolites were also examined with back-scattered electron images obtained by scanning electron microscopy (SEM), and individual features were characterized chemically by their energy-dispersion spectra.

# PETROGRAPHY

The host basalt consists mainly of labradorite and augite, with some pigeonite, orthopyroxene, and magnetite. Quartz is present in most holocrystalline varieties of the basalt, and the proportion of glass varies from

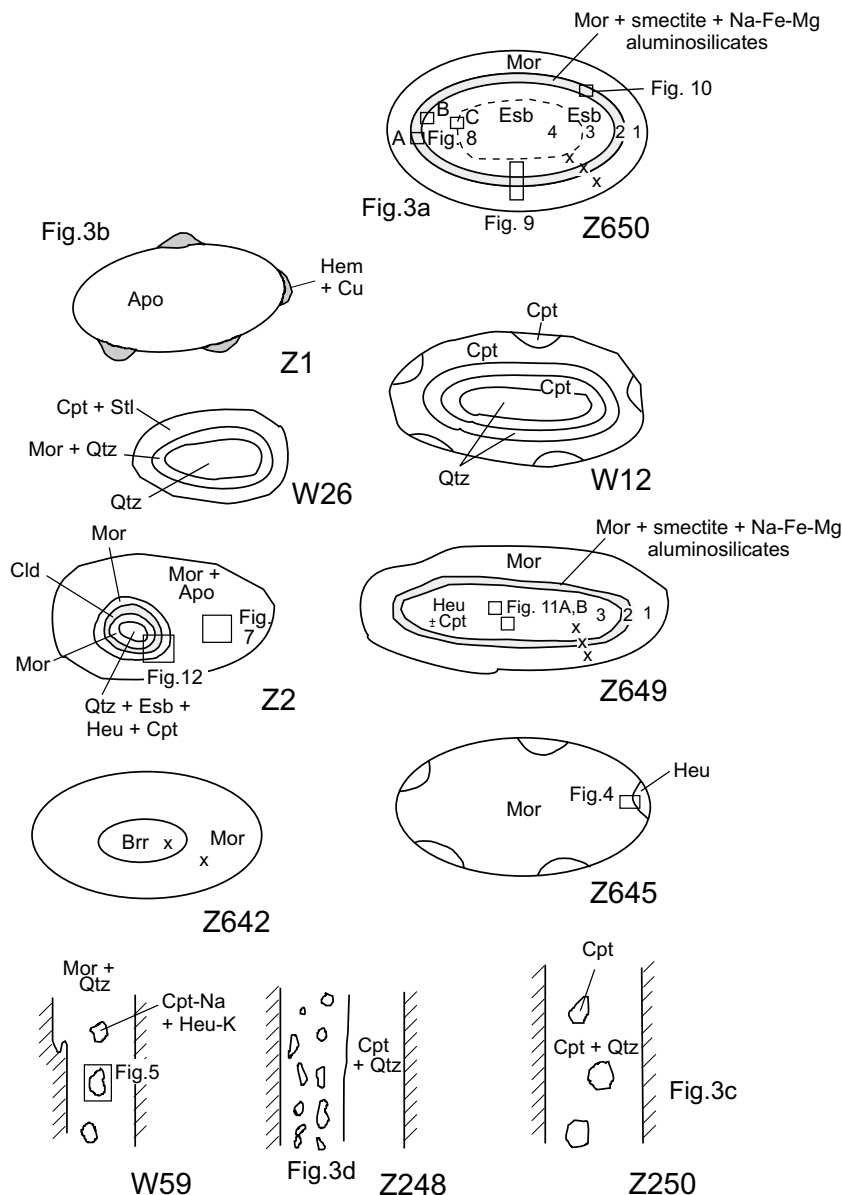


FIG. 2. Schematic representation of zoned amygdales and sheets, showing position of detailed compositions illustrated in subsequent figures. The outermost rim of celadonite is not shown. Mineral abbreviations in this and subsequent figures are given in the footnote to Table 1.

0 to 30%. In most samples, the original glass has devitrified, but plagioclase and pyroxene are commonly almost pristine, with very little alteration, even in zones with abundant amygdales. In some samples, however, there is partial alteration of augite to amphibole or chlorite and of labradorite to albite.

A variety of textural relationships among different mineral phases are distinguished in amygdales and veins. The following generalizations can be made from a study of many zoned amygdale and vein samples.

The outer parts of many amygdales consist of a thin layer of bluish green celadonite (*e.g.*, Z2, Z3 and Z26). In some cases, hematite rosettes with crystals of native copper at their center form the outermost part of the amygdale (Z1, Figs. 2, 3B).

Some amygdales lack concentric zoning inside the celadonite layer. Phases present include mordenite, apophyllite, quartz, heulandite and epistilbite. However, many amygdales are zoned, consisting of concentric

layers (Fig. 3A), commonly showing white → pink → dark green → pink → colorless zones from rim to core. They are generally microcrystalline, except for large crystals in some cores.

In most samples, the outermost zeolite zone consists of mordenite, but in a few samples, rosettes at the margin of amygdales and veins consist of either heulandite (Z250: Fig. 3C and Z645: Fig. 4) or clinoptilolite (W12: Fig. 2), and texturally these predate mordenite (Fig. 4). In W26, the outer clinoptilolite is more continuous and occurs with massive stellerite, rimming a zone of mordenite and quartz. Clinoptilolite is particularly common in veins, where it generally seems to have been the first mineral to crystallize (Fig. 3D). In vein sample W59, large crystals of clinoptilolite-Na have strongly embayed margins and are surrounded by mordenite and lesser amounts of quartz (Fig. 5). These textural relationships suggest that clinoptilolite-Na was replaced by mordenite and quartz.

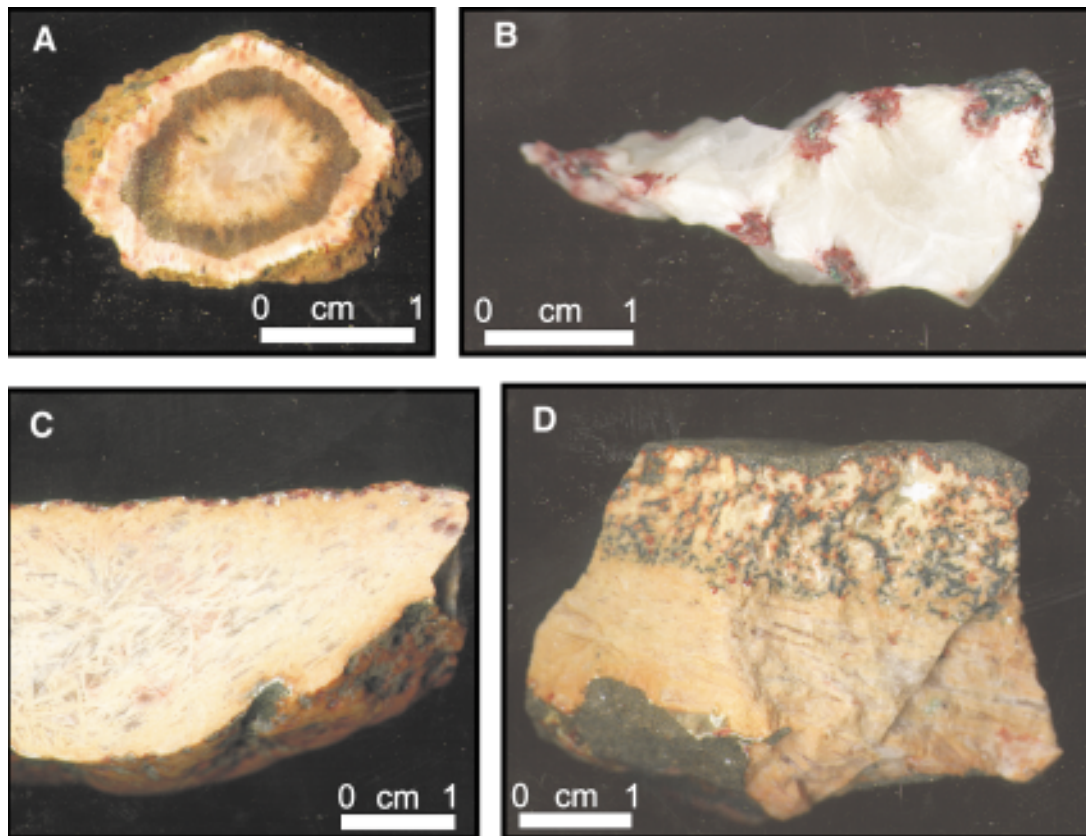


FIG. 3. Photographs of hand specimens of amygdales and veins (*cf.* Fig. 2). (A) Cut face of zoned amygdale Z650, showing outer zone of mordenite and inner zone of epistilbite, separated by a dark zone with Fe–Mg–Na aluminosilicates and mordenite. (B) Amygdale Z1 of apophyllite, with hematite rosettes at the rim. Native copper is found in the middle of the rosettes. (C) Cut face of vein Z250 of clinoptilolite and quartz, with a few heulandite hemispheres at the margin of the vein. (D) Vein Z248 of clinoptilolite and quartz with fragments of host basalt.

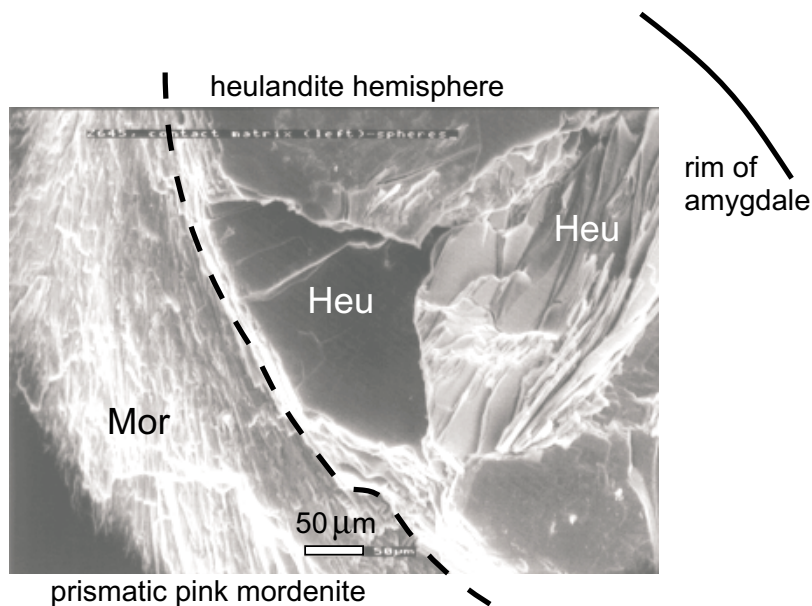


FIG. 4. Scanning electron micrograph of sample Z645 showing large crystals of heulandite forming hemispheres at the margin of the amygdale, with the interior consisting of prismatic mordenite. In this and subsequent figures, identifications are by X-ray diffraction, confirmed by energy-dispersion spectra.

The mordenite that generally forms the outermost zone of amygdales (Fig. 6A) commonly contains minor heulandite or epistilbite (or both). In sample Z649, EDS analyses and SEM images reveal the presence of various particles rich in elements such as Ni or Cu in the outer zone of mordenite. These particles are of the order

of 1–2  $\mu\text{m}$  in diameter and seem to be late precipitates. In sample Z650, small anhedral microlites of labradorite (Table 3, anal. 32), typically 40  $\mu\text{m}$  in length, are surrounded by mordenite within the outer zone of mordenite.

In some samples, mordenite forms euhedral plates, oriented in all directions, surrounded by matrix apophyllite that is generally anhedral but shows some euhedral terminations (Fig. 7). These relationships suggest that mordenite was the first mineral to precipitate.

The dark zone found in samples such as Z649 and Z650 (Fig. 3A) is composed of prismatic crystals cut by fractures, along which a variety of minerals have precipitated. The fractures are most clearly visible in SEM images (Fig. 8A). XRD analysis shows the presence of mordenite and a mineral that shifts to peaks at 1.68 nm and 0.83 nm upon glycolation (Fig. 6C), probably a smectite. A trace amount of chlorite, with a peak just above background at 0.7 nm, may be present (Fig. 6C). Electron-microprobe analyses of the bright spots seen in back-scattered electron images in this zone (Figs. 9, 10) show variable compositions: some appear to be calcite, but most appear to be Fe–Mg aluminosilicates with considerable amounts of Na and consistently about 1% Cl. Four distinct compositions are recognized (Table 3, anal. 33–36): (1) samples with electron-microprobe totals of about 85% and 3–8%  $\text{Na}_2\text{O}$ ; (2) samples with totals of about 92% and 12–18%  $\text{Na}_2\text{O}$ ; (3) a sample

TABLE 3. SELECTED RESULTS OF ELECTRON-MICROPROBE ANALYSES OF MINERALS COEXISTING WITH ZEOLITES, MORDEN AREA, NOVA SCOTIA

Sample	Z2	Z1	Z41	Z41	Z650	Z650	Z650	Z650	Z650
Analysis	28	29	30	31	32	33	34	35	36
Mineral	Apo	Apo	Apo	Apo	Pl	(1)	(2)	(3)	(4)
$\text{SiO}_2$ wt. %	51.36	52.08	56.47	55.29	53.52	39.66	40.52	48.85	36.76
$\text{Al}_2\text{O}_3$	0.83	0.17	n.d.	n.d.	28.57	7.9	7.12	8.27	6.73
FeO	n.d.	n.d.	n.d.	n.d.	0.96	15.98	11.17	14.12	11.99
MgO	n.d.	n.d.	n.d.	n.d.	14.68	16.72	17.81	15.34	
CaO	24.83	25.13	26.66	26.14	12.47	0.55	0.73	0.82	0.70
$\text{Na}_2\text{O}$	0.48	n.d.	2.17	2.21	4.18	3.95	14.93	9.62	26.91
$\text{K}_2\text{O}$	4.04	4.76	0.51	0.54	0.29	2.72	0.19	0.27	0.23
Cl	n.d.	n.d.	n.d.	n.d.	n.d.	0.78	1.09	1.09	1.01
Total	81.54	82.14	85.81	84.18	99.99	86.22	92.47	100.85	99.67
Spot in Fig. 10					5	10	9	4	

Symbols: Apo: apophyllite, Pl: plagioclase; (1) to (4) are compositions of Fe–Mg–Na aluminosilicates.



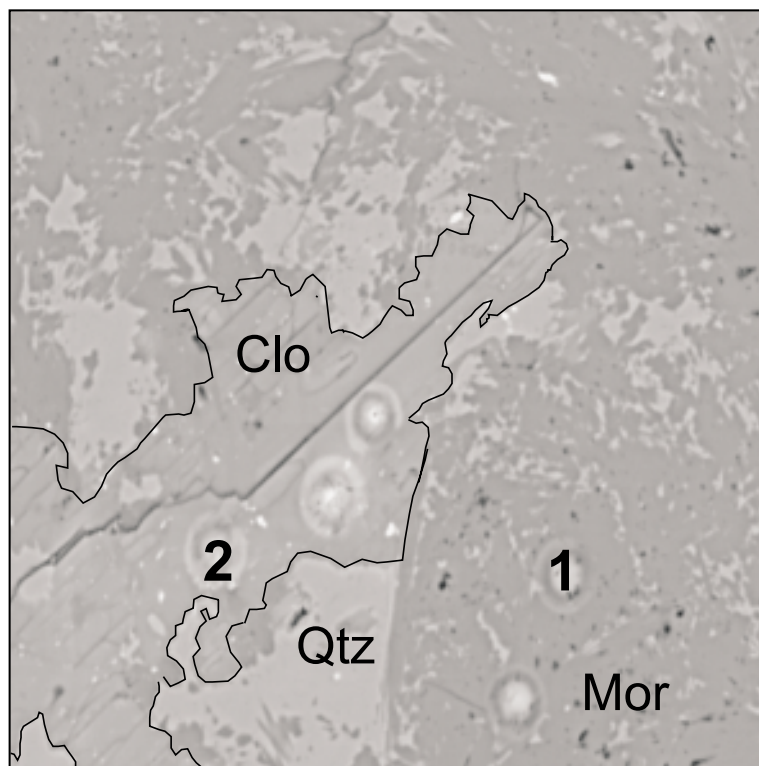


FIG. 5. Back-scattered electron image of sample W59 showing clinoptilolite-Na with strongly embayed margins, surrounded by mordenite and quartz. 1: composition 24; 2: composition 25 in Table 2.

with a total of ~100% and 9.6%  $\text{Na}_2\text{O}$  that appears to be a sodic amphibole (similar to that reported by Kontak 2000), and (4) a sample with 27%  $\text{Na}_2\text{O}$ . None of these compositions correspond to smectite, although type (1) might be a smectite-chlorite mixed-layer clay. In general, similar results were obtained whether a  $10\text{ }\mu\text{m}$  or a  $1\text{ }\mu\text{m}$  spot was analyzed, suggesting that the reported analytical results either pertain to individual minerals or very fine-grained mixtures. SEM images revealed small masses ( $< 20\text{ }\mu\text{m}$ ) of various compositions, including phases that are: 1) Ba- and S-rich (probably barite), 2) Ca- and S-rich (probably gypsum or anhydrite), and 3) Fe-, Mg- and Cl-rich, presumably the minerals described above. In some samples, the 1-mm-wide transition between zones (Fig. 9) show numerous small voids, which in SEM images appear to be residual space between subhedral crystals (Fig. 8B). Alternatively, these voids may result from dissolution along crystal faces.

The innermost zones are commonly composed of flat or tabular crystals becoming larger toward the center of the amygdale (Fig. 8C). In many samples, these inner zones consist mainly of epistilbite (Z650), or heulan-

dite (*e.g.*, Z649), or minerals of the stilbite group: X-ray diffractograms indicate the presence of stilbite, stellerite and barrerite (Table 1). In some samples, quartz forms the core of amygdaloids (W12, W26) (Fig. 2). In sample Z649, EDS analyses and SEM photos reveal the presence in the innermost zone of barite crystals forming rosettes (Fig. 11B) and of various micrometric metallic particles, such as Cu and Au (Fig. 11A), which appear to be late precipitates. The central zones of veins that consist principally of heulandite enclose some stilbite or quartz.

Exceptionally, there is evidence of repetitive zonation. A vug within a larger amygdale of mordenite + apophyllite (Z2) is zoned: celadonite  $\rightarrow$  mordenite + quartz  $\rightarrow$  epistilbite + heulandite + quartz (Fig. 12). Sample W12 shows an alternation of quartz and clinoptilolite.

This study has shown that zeolite assemblages in the North Mountain Basalt are extremely variable. This variability is illustrated by comparing pairs of samples from the same outcrop, such as Z649 with Z650, Z642 with Z645, and Z1 with Z2 (Fig. 2).

## CHEMICAL COMPOSITION OF THE MINERALS

Representative electron-microprobe analyses were made of zeolite-group minerals, generally using back-

scattered electron images in order to select a one-phase spot. Results are presented in Table 2, and the whole set of chemical data for cases with  $E < 10\%$  are plotted in Figures 13 and 14 to show variation in cation abundances.

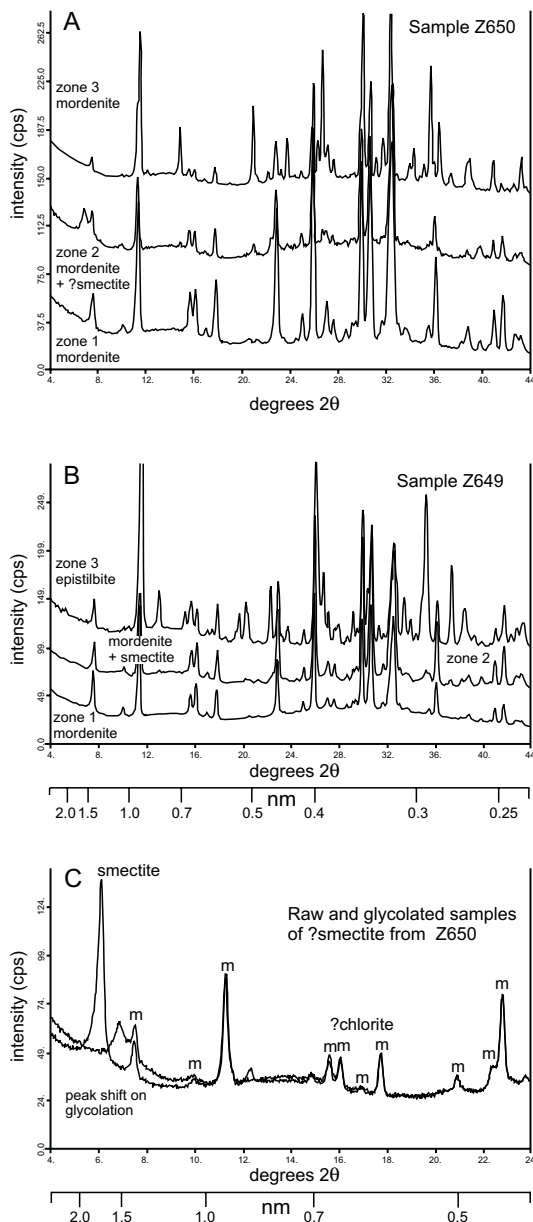


FIG. 6. X-ray diffractograms of samples from zoned amygdals Z650 and Z649 (CoK $\alpha$  radiation). (A) zones 1, 2 and 3 of Z650 (cf. Fig. 2). (B) Zones 1, 2 and 3 of Z649. (C) Raw and glycolated samples from zone 2 of Z650 showing characteristic peaks of mordenite (m), smectite and possible chlorite.

## Mordenite

Mordenite has a very low  $(Ca + Mg):(Na + K)$  ratio (Fig. 13). Sample Z650 (■ in Fig. 14) has the highest Na content of any other sample of mordenite. The exchangeable cations in the mordenite structure are Ca, Na, and K. Within a single sample of amygdale or vein, cation variation is principally the result of the exchange of Ca for K (Fig. 14). Compared with samples from numerous localities considered in the detailed study of mordenite by Passaglia (1975), the Nova Scotian material shows a more limited range of substitution of Ca for alkalis, but a larger range of substitution of Na for K. These data suggest that all three cations are equally exchangeable, whereas Passaglia (1975) concluded that the role of K was only minor.

The number of K and Ca atoms per formula unit (*apfu*) for the mordenite of this study ranges from 0.65 to 2.02 and from 0.36 to 1.29, respectively. These are outside the ranges of 0.1–0.8 and 1.6–2.5 *apfu* given by Gottardi & Galli (1985), but within the range shown by Passaglia (1975, his Fig. 2) drawn from the literature data. The mordenite in this study has limited Si/Al variation: 5.0 to 5.8, with only four values (6.4, 4.2, 4.92 and 4.93) outside this range from a total of 55 analyses.

## Clinoptilolite – heulandite

Most samples containing clinoptilolite contain clinoptilolite-Ca (*e.g.*, Z645, Z649); however, clinoptilolite-Na is found in sample W59 and has a subequal abundance of Na and K. It is similar to some clinoptilolite-K from pelagic sediments (Boles 1977).

Heulandite shows large variation in the  $(Ca + Mg):(Na + K)$  ratio (Fig. 13) and is generally heulandite-Ca. The proportion of K and Na is subequal and relatively low (Fig. 14), except in sample W59, where heulandite-K has a chemical composition similar to that of the clinoptilolite-Na described above. Within a single amygdale or vein, variation in heulandite composition is mainly due to the variation in Na content (Fig. 14), whereas variation among samples is largely in K content.

## Stilbite and epistilbite

Epistilbite was identified with certainty only in the core of the zoned amygdale (Z650) with Na-rich mordenite, whereas all the zoned amygdals with K-rich mordenite contain heulandite in their cores. Epistilbite contains extremely low K and variable extent of exchange of Na for Ca; stilbite has a similar range of



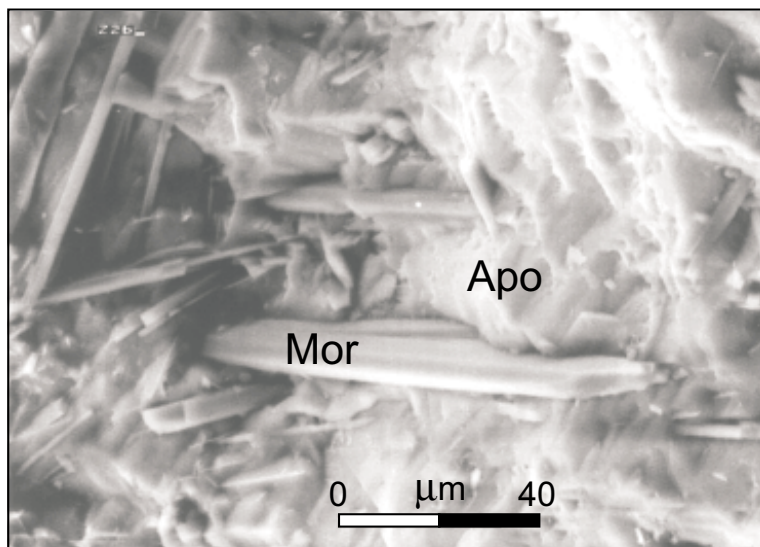


FIG. 7. Scanning electron micrograph of euhedral crystals of mordenite surrounded by anhedral apophyllite (sample Z2).

exchange of Na for Ca and has no K. In one of the zoned amygdals (Z26: ○ in Fig. 14), two distinct groups of epistilbite occur, one with no Na and higher Ca, and the other with some Na and lower Ca.

#### *Apophyllite*

Two members of the apophyllite family are distinguished chemically (Table 3). In samples Z1 and Z2, K-rich apophyllite occurs with mordenite. In sample Z41, natro-apophyllite occurs with quartz.

### DISCUSSION

#### *Burial metamorphism versus hydrothermal origin of the zeolites*

The geographic zonations recognized by Aumento (1966) have led previous investigators to compare zeolite zonation in the North Mountain Basalt with that produced by burial metamorphism (*e.g.*, Colwell 1980). Comparisons can be made with mineral assemblages found at depths of 1500 m in Iceland (Neuhoff *et al.* 1999) and in thick piles of basalt flows in the Proterozoic of Minnesota (Schmidt & Robinson 1997).

The maximum depth of burial of the North Mountain Basalt is unknown. On the north side of the Bay of Fundy, it is overlain by 3–4 km of younger sediment, but this thins to less than 1 km on the south side of the bay (Keen *et al.* 1991). The Fundy basin is a half graben with greatest subsidence in the north (Withjack *et al.* 1995). It is therefore probable that the maximum

thickness of sediment overlying the North Mountain Basalt on the south side of the bay was less than one kilometer. There is no evidence for much greater thicknesses of sediment that were subsequently eroded. Apatite thermochronology from three sites respectively 5, 20 and 40 km southeast of the basal contact of the basalt on the south side of the Bay of Fundy show track ages of 160 to 270 Ma (Ryan & Zentilli 1993) that date the time at which the rocks cooled through 100°C. Given the emplacement age of 200 Ma for the North Mountain Basalt, it is clear that there is no geothermal evidence for deep burial on the south side of the Fundy graben.

Furthermore, unlike the host basalts in Iceland and Minnesota, the North Mountain Basalt shows no pervasive alteration except for devitrification of glass. In many samples, phenocrysts appear almost pristine. Augite has altered to chlorite and labradorite to albite in some restricted zones, such as parts of the middle flow unit of North Mountain (Papezik *et al.* 1988) and in places near Parrsboro (Greenough *et al.* 1989). Only in basalts now buried >500 m in the Chinampas well, in the northern Bay of Fundy, is there pervasive metamorphism, with albitized feldspar, minor prehnite, some alteration of augite, and chlorite linings in vesicles (Greenough & Papezik 1987, Pe-Piper *et al.* 1992). The presence of hot springs in the lacustrine sediments immediately overlying the North Mountain Basalt near Morden (DeWet & Hubert 1989) suggests that shallow circulation of a hydrothermal fluid may have been responsible for zeolite formation.

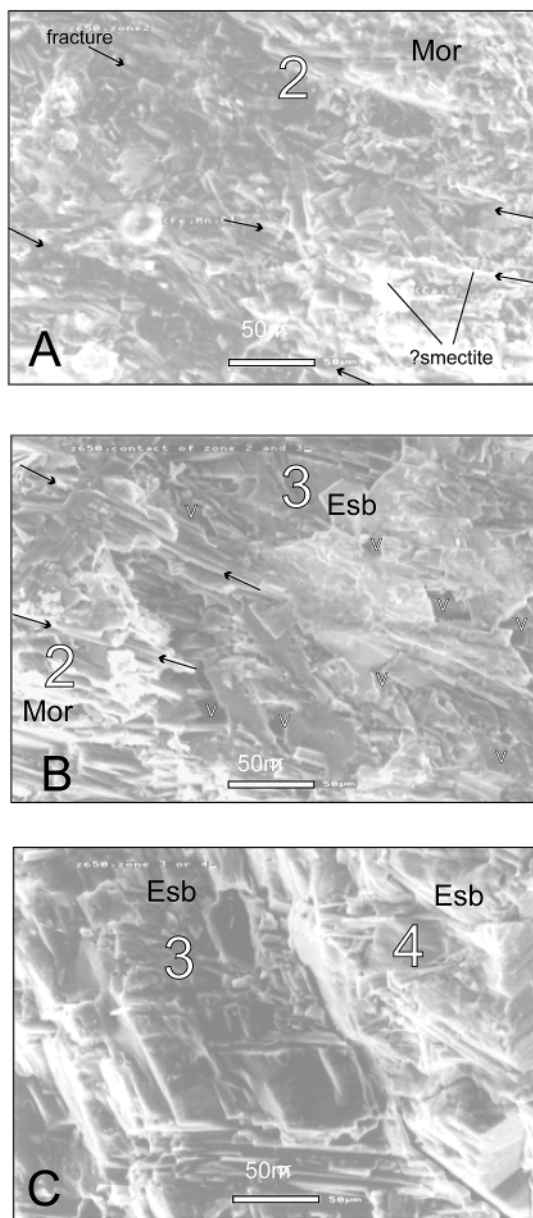


FIG. 8. Scanning electron micrographs from zoned amygdale Z650. A. Zone 2 (cf. Fig. 2) showing mordenite cut by curvilinear fractures (arrows), in some cases filled by a bright mineral (?smectite). B. Transition from zone 2 to zone 3. Fractures (arrows) are visible in zone 2. Voids (V) appear to have formed between subhedral crystals of epistilbite. C. Contact of zones 3 and 4 showing coarsely crystalline epistilbite.

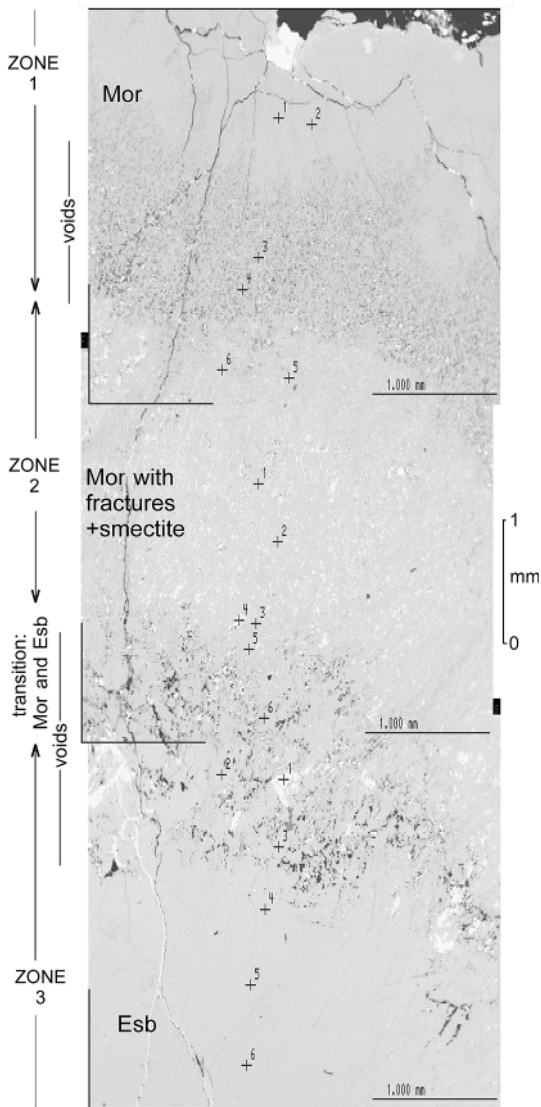


FIG. 9. Composite of back-scattered electron images of thin section through zones 1 to 3 of amygdale Z650 (cf. Figs. 2, 8), showing inhomogeneous character of zone 2, the presence of voids and fractures, and the location of the spots analyzed.

On the north side of the Bay of Fundy near Parrsboro, direct evidence for shallow precipitation of zeolites is found in polymictic conglomerates that immediately overlie the North Mountain Basalt and developed as a result of erosion of fault scarps immediately following basalt extrusion (Withjack *et al.* 1995). A few basalt clasts in the conglomerate contain zeolites in amygdales (Miller 1997).

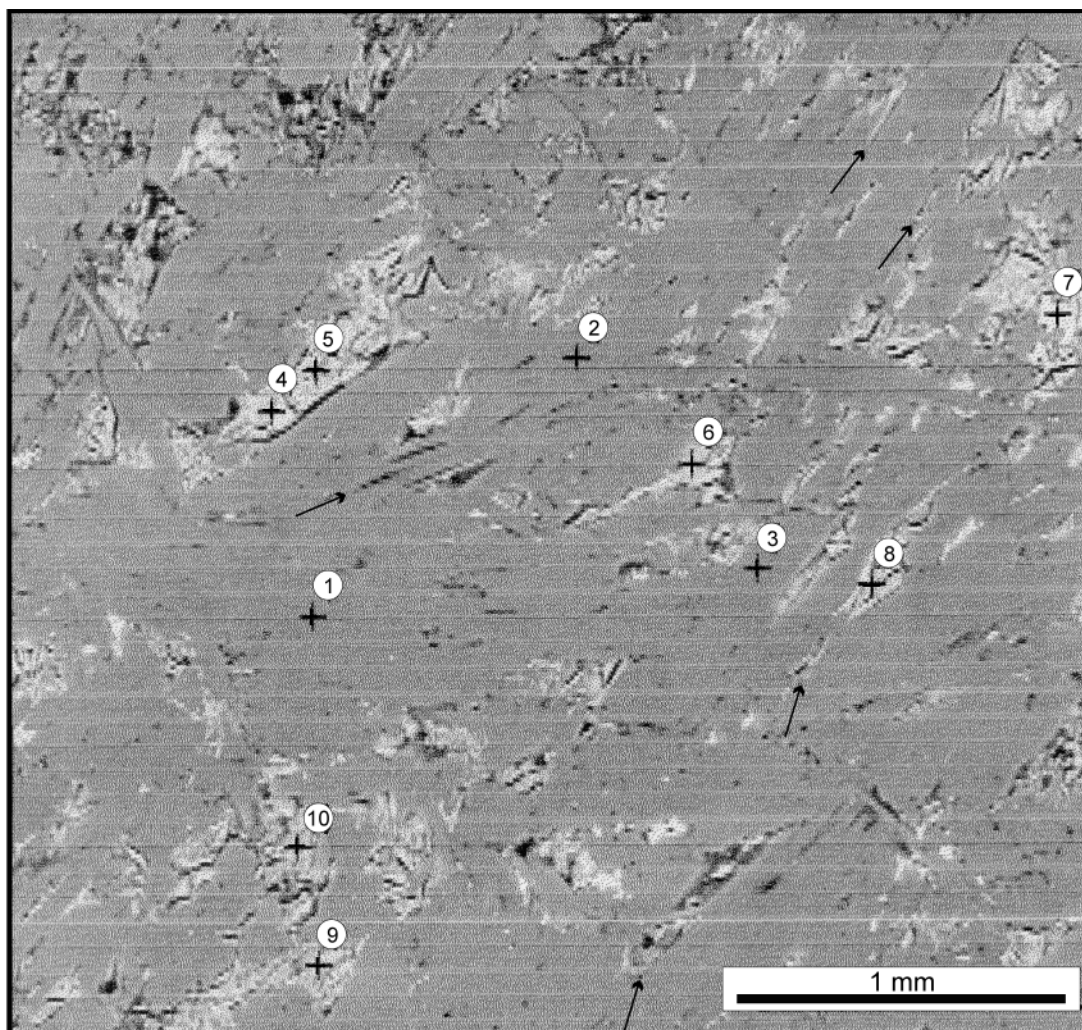


FIG. 10. Detailed back-scattered electron image of zone 2 in sample Z650. Principal mineral is mordenite. Light areas are other minerals discussed in text; compositions 35 and 36 in Table 3 are from positions 9 and 4, respectively. Arrows indicate examples of fractures.

### *Zoning sequences*

The zoning sequence and textural evidence suggest the following general sequence of crystallization: first celadonite, then clinoptilolite-Na, then mordenite, then heulandite-K, then heulandite-Ca, clinoptilolite-Ca and epistilbite, and finally stilbite-type minerals such as stilbite, barrerite and stellerite, or apophyllite. The general fields of stability of the zeolites are known from modern hydrothermal systems (Kristmannsdóttir & Tómasson 1978), and the observed sequence in the samples studied conforms in a general manner to a progressive de-

crease in temperature [*i.e.*, to the rule of Cornu (1908) that increased hydration is favored by lower temperatures].

However, this simple sequence is not found in all samples. For example, in Z2 two zones of mordenite are separated by a zone of celadonite, and in Z643 there are two distinct zones of heulandite. In some samples, mordenite is not the first zeolite to crystallize, following heulandite and stilbite in W26 and following heulandite in Z645. It has not been possible to determine whether such changes reflect changes in pore-water composition or temperature (or both).

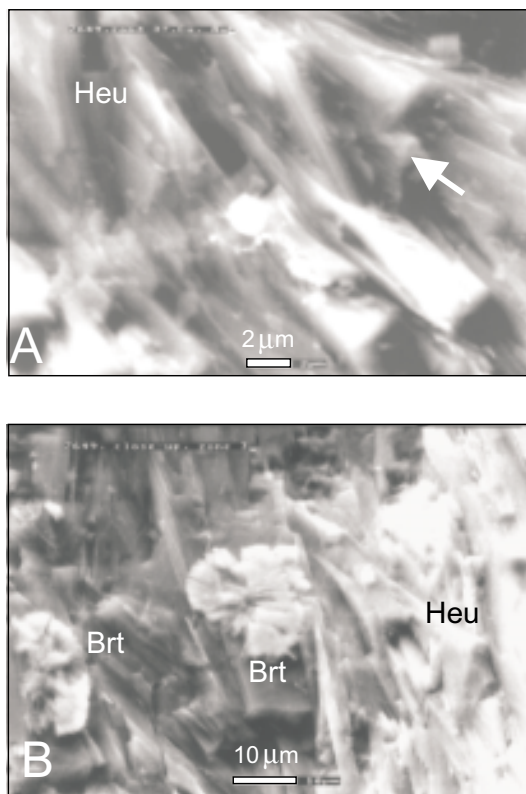


FIG. 11. Scanning electron micrographs of heulandite, zone 3 of amygdale Z649 (cf. Fig. 2). A. Particle rich in Cu and Au (indicated by arrow). B. Barite rosettes.

Comparison with the literature permits some estimates of temperature conditions. In the experimental work of Barth-Wirsching & Holler (1989), plagioclase formed from basaltic glasses at about 250°C. This temperature estimate may be applicable to the labradorite microlites in mordenite in zone 1 of sample Z650. The crystallization of the celadonite rims requires a temperature of only about 50°C (Odin *et al.* 1988). In hydrothermal systems in Iceland, a smectite-group mineral is present at temperatures of <200°C, and stilbite forms at temperatures above 90°C (Kristmannsdóttir & Tómasson 1978).

#### *The variability of zeolite assemblages*

The veins and amygdales illustrated show a wide range of variability in zeolite minerals present (Fig. 2). The irregular distribution of zeolites and related minerals in pyroclastic rocks has been attributed to factors such as localized heat-flow, ionic activities in pore waters, and permeability of the host rock. The high initial permeability decreases subsequently due to deposition

of secondary minerals, leading to the formation of individual closed systems (Keith & Staples 1985). Although there is evidence of open-system hydrothermal activity for the formation of the North Mountain Basalt zeolites, restricted domains may have developed as pore space was filled by precipitates of quartz, celadonite and zeolites. As a result, the hydrothermal pathways may have been substantially different, even for quite closely spaced amygdales. Samples such as Z649 and Z650, from the same outcrop, show a similar early history, with the precipitation firstly of mordenite and then of a greenish layer with Na–Fe–Mg-rich aluminosilicates. Their later history differs, with the interior of one containing epistilbite, the other, heulandite.

#### *Variations in exchangeable cations*

Within a single amygdale or vein, variation in the nature of the exchangeable cations in mordenite is principally the result of the exchange of Ca for K, whereas the exchange of Na for K is more important in variation among amygdales (Fig. 14). In heulandite and clinoptilolite–Ca, variability within an amygdale or vein is principally the result of changing Na content, whereas exchange of K for Ca predominates among samples from different amygdales. A similar trend is seen in the clinoptilolite–Na. These observations suggest that fluid composition may be important in defining the type of zeolite that occurs in a particular amygdale or vein, whereas temperature variations may influence chemical variation within that individual amygdale or vein. It seems probable that fluids rich in Ca and poor in K would precipitate epistilbite and a zeolite of the stilbite group, whereas fluids poor in Ca and rich in K would precipitate heulandite and clinoptilolite. Formation of the clinoptilolite–Na probably resulted from fluids with particularly elevated concentrations of Na<sup>+</sup> and K<sup>+</sup>. Bowers & Burns (1990) found that the composition of alkali-rich clinoptilolite correlates with groundwater with elevated Na<sup>+</sup> and K<sup>+</sup> but depleted Ca<sup>2+</sup> concentrations. Using textural evidence from zeolitized tuffs at Yucca Mountain, Utah, Sheppard *et al.* (1988) suggested that in some instances, mordenite has formed at the expense of clinoptilolite during late-stage dissolution; circumstantial evidence points to elevated temperatures as the cause of this conversion. Such a conversion is in agreement with the activity diagrams presented by Bowers & Burns (1990). Sturchio *et al.* (1989) also proposed that K–Na-rich clinoptilolite formed at elevated temperatures. Thus, it may be significant that the clinoptilolite–Na in this study was found only in a vein where there was clear textural evidence for early clinoptilolite partly replaced by mordenite.

#### *The origin of fractures and voids*

The mordenite – smectite – Fe–Mg–Na aluminosilicate zone in samples Z650 and Z649 contains abundant

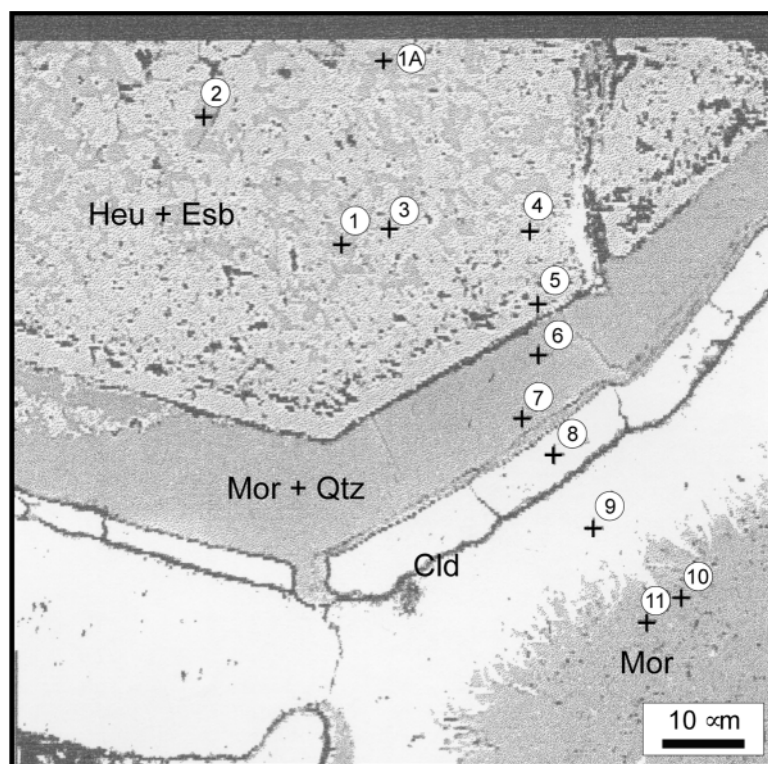


FIG. 12. Back-scattered electron image showing minerals growing within a small vug within a larger amygdale (Sample Z2). Numbered positions indicated on the image are sites of the spots analyzed. 10, 11: mordenite in the host amygdale (anal. 2 and 1, respectively, in Table 2). Remaining spots are from the vug: 8, 9: celadonite, 7: quartz, 6: mordenite, 5: heulandite (anal. 3 in Table 2), 1A: ?epistilbite (anal. 4 in Table 2).

fractures (Figs. 8, 9). These might be the result of degassing and an abrupt drop in pressure (*i.e.*, “explosive” hydrothermal venting). Such changes in pressure might cause not only hydraulic fracturing but also abrupt chemical changes in percolating fluids.

The voids between subhedral crystals seen at the boundaries between zones in the same samples appear to have a different origin. The SEM images (Fig. 8B) suggest that they result from either the cessation of crystallization of mordenite or possibly from later dissolution.

#### *Precipitation of the Na-rich Mg-Fe aluminosilicates*

Mg-Fe-Na-rich minerals, including some amphibole species, occur in a few zoned amygdals as a dark zone. This zone is fractured and is up to 0.5 cm wide. To account for the high Na and Cl, it is suggested that precipitation of these minerals was a consequence of hydration reactions, under the following scenario. As

porosity was reduced, the migration of pore fluid may have been slowed. Some of the slowly percolating fluids may have been consumed in hydration reactions, so that the fluid:rock ratio decreased and the remaining fluid was equilibrated at the new conditions. Saturation (*e.g.*, in NaCl) has been shown to be possible for systems undergoing hydration reactions (Trommsdorff & Skippen 1987). Cations and  $\text{Cl}^-$  may have become concentrated by such a process, and the Mg-Fe-Na-rich minerals could have precipitated directly from solutions of appropriate composition. After the precipitation of these minerals, the precipitation of mainly heulandite or epistilbite continued. Such a process may also be responsible for the later precipitation of barite, Cu, Ni, and Au, which occur sparsely in these zoned amygdals. As an alternative, as discussed below, hydrothermal waters may have intermittently dissolved stratigraphically underlying evaporites, resulting in high concentrations of  $\text{Na}^+$  and  $\text{Cl}^-$ .



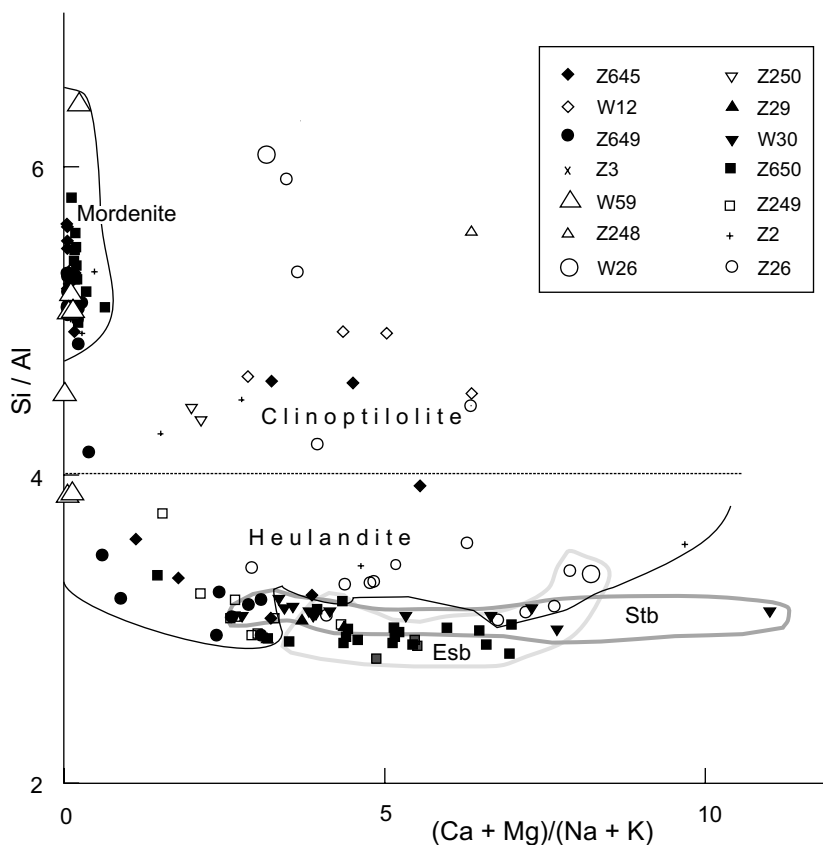


FIG. 13. Plot of Si/Al against  $(Ca + Mg)/(Na + K)$  for zeolite-group minerals (electron-microprobe data for which E is less than 10%).

#### The formation of mordenite

Mordenite is the most siliceous zeolite found in the North Mountain Basalt. Experiments to synthesize mordenite were pioneered by Barrer (1948). More recent syntheses of mordenite from natural silicic glasses have been reported by Kirov *et al.* (1979), Phillips (1983), and Barth-Wirsching & Holler (1989). These experiments have shown that mordenite is favored over clinoptilolite by higher temperatures and higher Na:K ratios. Experiments by Kusakabe *et al.* (1981) have demonstrated the conversion of clinoptilolite to mordenite (*cf.* sample W59) under conditions of increased temperature ( $170 \rightarrow 365^\circ\text{C}$ ), pH ( $5.3 \rightarrow 12.2$ ), and  $\text{Na}^+$  concentration in fluids ( $40 \rightarrow 2,300$  ppm). If  $\text{Na}^+$  concentration or pH decreased, higher temperatures were required for the conversion. There is no direct evidence as to which of the above factors (T, pH,  $\text{Na}^+$ ) might have caused the conversion of the clinoptilolite to mordenite in vein W59.

In natural systems, the crystallization of mordenite is favored by: 1) a highly silicic composition of the original glass, clearly not the case in the North Mountain Basalt, and 2) high pH of pore water, which influences the Si:Al ratio of the pore water and the Si:Al ratio of the zeolite precipitate (Mariner & Surdam 1970). This ratio is higher in mordenite and lower in heulandite, epistilbite and stilbite, so that a mildly alkaline pH of 7–9 will favor mordenite or clinoptilolite rather than phillipsite (Phillips 1983). The solutions that precipitated mordenite in the North Mountain Basalt can be inferred to have had high concentrations of Na, Ca and Si.

#### The geological setting of mordenite formation

The North Mountain Basalt zeolites are well known for their chemical and textural variability. This may be the result of initial open-system hydrothermal activity, followed by creation of closed domains as porosity was



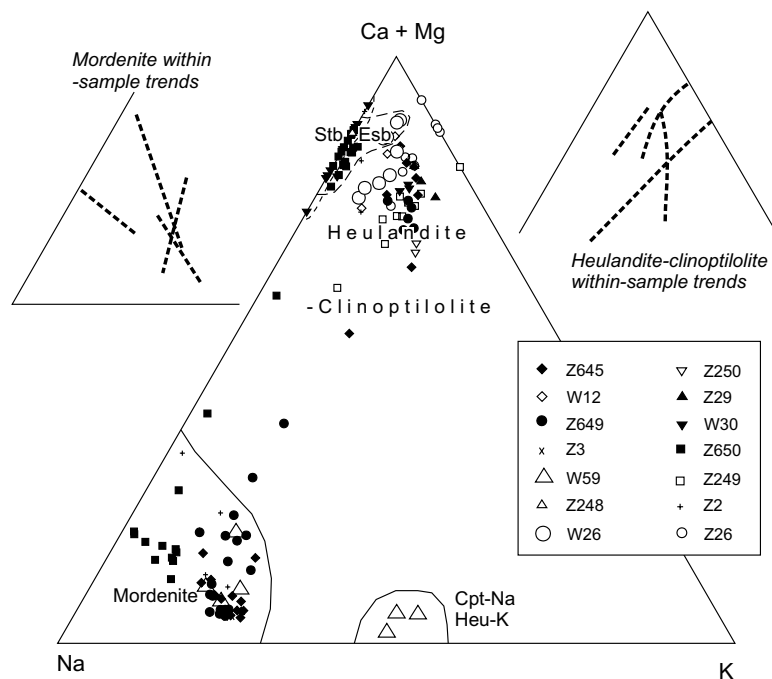


FIG. 14. Plot of zeolite compositions in terms of  $(Ca + Mg) - Na - K$  (electron-microprobe data for which E is less than 10%). Insets show trends of compositional variation of heulandite and mordenite within individual amygdales or veins.

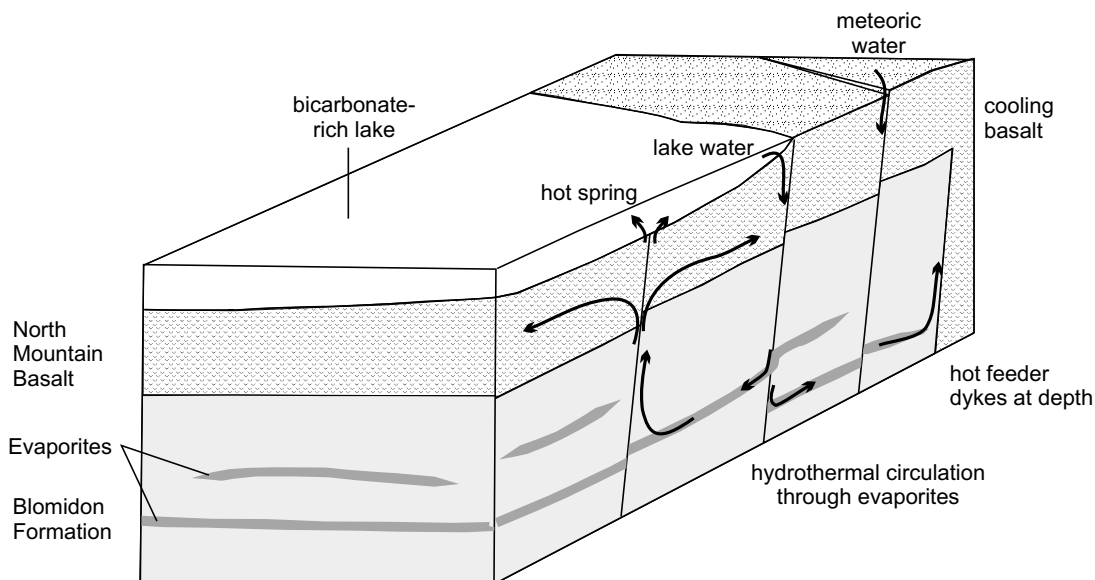


FIG. 15. Inferred geological setting leading to hydrothermal circulation of an alkaline fluid rich in Na.

reduced. In an open system, many variables such as Si migration, starting material, initial solution, material transport as a function of chemical gradient, percolation velocity and reaction time may affect the kind of zeolite being formed. Of all these variables, solution chemistry and reaction time may be particularly important.

A hydrothermal origin for the zeolites is indicated by several lines of geological evidence: the lack of deep burial, the evidence from conglomerates that zeolite precipitation was early, and the presence of siliceous hot springs in the overlying lacustrine sediments. All the new petrographic evidence, such as the presence of porous zones, repeated episodes of zeolite crystallization, late precipitation of compounds such as barite, gypsum, Au, Cu, Ni, and Fe–Mg–Na-rich minerals, all favor an origin for the zeolites by repeated hydrothermal pulses in an open system. The temperatures required or suggested by the identified zeolites are as high as 250°C. Such a temperature is difficult to achieve under conditions of burial metamorphism.

The alkaline conditions necessary for the dissolution of silica and the formation of mordenite may have been favored by the development of an alkaline lake, from which the carbonate-rich sediments of the Scots Bay Formation were deposited, immediately following extrusion of the North Mountain Basalt (Fig. 15). Evidence of hydrothermal precipitation of silica minerals is seen within the lake sediments (DeWet & Hubert 1989). Mertz & Hubert (1990) have described residual halite and gypsum in the Blomidon Formation immediately below the North Mountain Basalt. West of Wolfville (Fig. 1), deeper but much thicker evaporites are found in the Carboniferous Windsor Group of the Minas sub-basin (Clifton 1967), which, in the opinion of Ackermann *et al.* (1995), experienced post-late Triassic, pre-Cretaceous dissolution. Hydrothermally circulated fluids may have dissolved these evaporites, gaining access along faults that were synchronous with basalt extrusion (Schlische & Ackermann 1995). Hydrothermal circulation through beds of evaporites could have contributed to the variable Na and Ca contents of fluids inferred from the composition of the zeolites.

#### CONCLUSIONS

1. Zoned sequences of zeolites in amygdaloids and veins in the North Mountain Basalt show a succession inferred to result from falling temperature: clinoptilolite-Na → mordenite → heulandite, clinoptilolite-Ca or epistilbite → stilbite, barrerite or stellerite. Individual complexly zoned amygdaloids show evidence of multiple fluid-circulation events.

2. Locally, mordenite has been hydrofractured and the fractures filled with small patches of at least three different Mg–Fe–Na-rich aluminosilicate minerals. Barite, Cu, Ni and Au precipitates occur throughout some zoned amygdaloids.

3. In an individual amygdale, mordenite varies mostly in K:Ca ratio and heulandite – clinoptilolite-Ca, in Na:Ca ratio. These changes are interpreted to result from changes in temperature. The formation of epistilbite and the stilbite group of minerals or of heulandite and clinoptilolite is also dependent on the proportions of Ca, Na and K in the fluid phase.

4. The high temperatures required for the precipitation of mordenite and labradorite, the repetitive zonation in some amygdaloids, and the occurrence of metal precipitates all suggest an origin of the zeolites by active circulation of a hydrothermal fluid, not by burial metamorphism. This interpretation is supported by the lack of geological evidence for deep burial and the absence of pervasive alteration in the basalts.

5. Hydrothermal circulation was aided by faulting synchronous with basalt extrusion. An alkaline lake formed on top of the basalt flows and circulating hydrothermal fluids probably flushed through evaporites of the underlying Blomidon Formation and perhaps from the Windsor Group, thus providing the alkaline Na-rich fluids necessary to precipitate mordenite.

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