Canadian Mineralogist
Vol. 31, pp. 467-470 (1993)

COMPOSITE NATROLITE–MESOLITE CRYSTALS FROM THE COLUMBIA RIVER BASALT GROUP, CLARKSTON, WASHINGTON

MICKEY E. GUNTER

Department of Geology and Geological Engineering, University of Idaho, Moscow, Idaho 83843, U.S.A.

CHARLES R. KNOWLES

Idaho State Geological Survey, University of Idaho, Moscow, Idaho 83843, U.S.A.

D. KATE SCHALCK

Department of Geology, Washington State University, Pullman, Washington 99164, U.S.A.

ABSTRACT

Chemical composition, optical properties, and X-ray-diffraction photographs confirm the occurrence of "single" crystals of natrolite capped by mesolite in cavities in the Columbia River Basalt Group near Clarkston, Washington. The indices of refraction for the natrolite tip are α 1.4781(3), β 1.4814(3), and γ 1.4895(3), with a measured $2V_z$ of 62.9°, and the index of refraction for the near-isotropic mesolite tip is 1.5055(3). Chemical analysis by electron microprobe shows the natrolite and mesolite portions to be near their ideal compositions. Single-crystal X-ray diffraction shows a tripling of *b* for the mesolite portion of the composite crystal. Calcium and sodium do not readily exchange in natrolite and mesolite; thus, the formation of Ca-free natrolite at the beginning of growth and Ca-rich mesolite at the end of growth indicates a change in the environment during crystal growth.

Keywords: natrolite, mesolite, physical properties, Columbia River Basalt Group, Clarkston, Washington.

SOMMAIRE

Les données chimiques, les propriétés optiques, et les clichés de diffraction X confirment la présence de cristaux "uniques" de natrolite avec surcroissance de mésolite dans les cavités d'un basalte du groupe de Columbia River, près de Clarkston, au Washington. Les indices de réfraction de l'extrémité composée de natrolite sont: α 1.4781(3), β 1.4814(3), γ 1.4895(3), $2V_z$ = 62.9°. L'indice de réfraction pour l'extrémité presque isotrope composée de natrolite est 1.5055(3). Des analyses à la microsonde électronique montrent que natrolite et mésolite possèdent une composition presque idéale. Une étude par diffraction X sur cristal unique démontre un triplage de *b* pour la portion du cristal composite faite de mésolite. Le calcium et le sodium ne sont pas facilement échangeables dans ces deux minéraux. La formation d'abord d'une natrolite sans Ca, suivie d'une surcroissance de mésolite riche en Ca, résulterait d'un changement du milieu de croissance.

(Traduit par la Rédaction)

Mots-clés: natrolite, mésolite, propriétés physiques, groupe de basaltes de Columbia River, Washington.

INTRODUCTION

The zeolites used in this study were originally collected for use in an optical mineralogy class. A crystal observed in cross-polarized light exhibits retardation and indices of refraction indicative of natrolite; however, one end of the crystal is nearly isotropic (Fig. 1A). The index of refraction also is higher for the isotropic end, nearer to the accepted value for mesolite. To confirm these observations, we collected both chemical and X-ray-diffraction data. In the course of this study, we discovered that this is not the only reported occurrence of intergrown natrolite-group zeolites. Tschernich (1992) provided a review of the known occurrences. The structure of both natrolite and mesolite have been refined in space group *Fdd*2, respectively by Artioli *et al.* (1984) and Artioli *et al.* (1986). Chemically, natrolite $[Na_{16}(Al_{16}Si_{24}O_{80}) \cdot 16H_2O]$ differs from mesolite $[Na_{16}Ca_{16}(Al_{48}Si_{72}O_{240}) \cdot 64H_2O]$ by an ordered substitution of $2\,Na = Ca + H_2O$ (Gottardi & Galli 1985), which causes a tripling of *b* for mesolite.

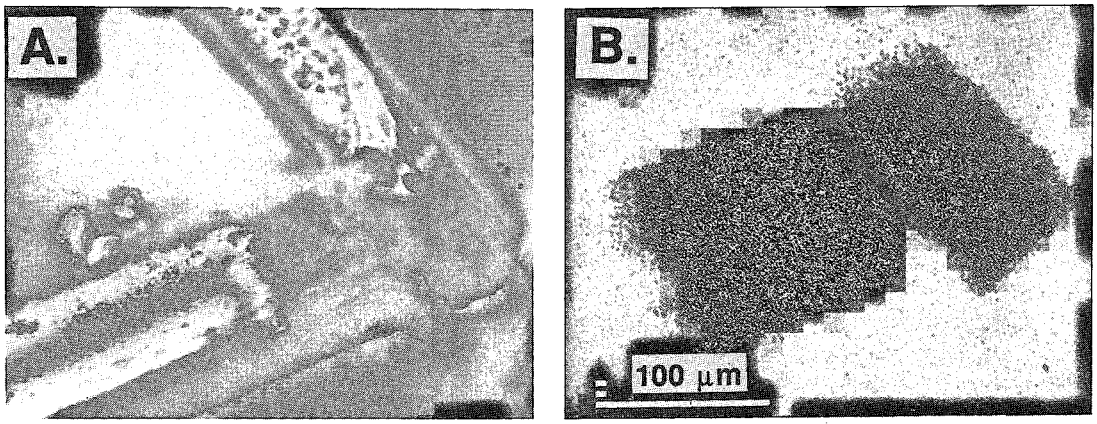

FIG. 1. A. Thin-section photograph in cross-polarized light showing birefringent natrolite terminated with near-isotropic mesolite tips. Chemical analyses were performed on the natrolite and mesolite portions of both of these crystals and yielded near-ideal end-member chemical formulas (Table 1). B. Concentration of Ca at the near-isotropic (mesolite) tip showing an abrupt change in Ca content at the suture between natrolite and mesolite.

OCCURRENCE

The faults and folds that disrupt the basalts in the vicinity of Lewiston, Idaho, define what is referred to as the Lewiston structure (Hooper *et al.* 1985). This structure brought the Imnaha Basalt, the oldest formation in Columbia River Basalt Group, to the surface on the Snake River approximately 10 kilometers west of Clarkston, Washington (on the north side of the river, at river mile marker 131). In the area of these structural disturbances, the Imnaha Basalt contains zeolites (natrolite, mesolite, chabazite, and erionite), which occur in filled vesicles. No thorough mineralogical study has been done in this area. The zeolite-containing amygdules weather out of the basalt and are easily collected. The single crystals of natrolite capped by mesolite occur in the amygdules. The crystals are approximately 50 μm in cross-section and 400 μm in length, with the first 350 μm composed of natrolite, and the tip, approximately 50 μm, composed of mesolite. The amygdules are approximately 20 mm in diameter with 3 mm walls.

OPTICAL PROPERTIES

The composite crystals were first noticed with a polarizing-light microscope. When viewed in cross-polarized light, the natrolite tip is birefringent (first-order red to second-order blue), and the mesolite tip seems near-isotropic. Precise optical data were collected by spindle-stage methods (Bloss 1981). Extinction data collected for the natrolite tip of the crystal yielded a $2V_z$ of 62.9(5)°, as determined by the computer program EXCALIBR (Bloss 1981, Gunter *et al.* 1988). Indices of refraction were determined using the double-variation method (Bloss 1981, Su *et al.* 1987). For the natrolite tip, the indices are α 1.4781(3), β 1.4814(3) and γ 1.4895(3). The mesolite tip is nearly isotropic, and its single index of refraction is 1.5055(3).

CHEMICAL COMPOSITION

Chemical composition (Table 1) was determined with an ARL–EMX–SM electron microprobe operating at 15 kV and 20 nA. The electron beam was defocused to approximately 20 μm. Amelia albite and synthetic anorthite were used as standards. All data were corrected for drift and dead time, and Bence & Albee (1968) matrix corrections were applied. Three spots were analyzed on each of the natrolite and mesolite portions

TABLE 1. CHEMICAL COMPOSITION OF NATROLITE-MESOLITE CRYSTALS SHOWN IN FIGURE 1A

	large crystal		small crystal	
	natrolite	mesolite	natrolite	mesolite
SiO_2 wt.%	47.8	48.4	49.0	45.5
Al_2O_3	25.3	24.4	24.9	24.1
CaO	0.0	9.5	0.0	9.6
Na_2O	15.5	5.2	15.0	5.2
H_2O*	9.5	12.4	9.5	12.4
number of cations based on 80 (and 240) atoms of oxygen				
Si	24.6	25.0 (75.0)	25.0	24.4 (73.2)
Al	15.4	14.8 (44.4)	15.0	15.3 (45.9)
Ca	0.0	5.2 (15.6)	0.0	5.5 (16.5)
Na	15.5	5.2 (15.6)	14.9	5.4 (16.2)
H_2O	16.0	21.3 (64.0)	16.0	21.3 (64.0)

* Weight percent H_2O calculated from ideal formula (Ross et al. 1992).

of the two crystals shown in Figure 1A; each chemical composition reported is the average.

Calcium Scan and BSE Image

To verify further the chemical variation within these minerals, Ca scans and back-scattered electron (BSE) images were performed. The BSE images and Ca scans were produced on a Cameca Camebax automated electron-microprobe operating at 15 kV and 20 nA using an 8-μm beam. The same samples were used for the chemical analysis (Fig. 1A), Ca scans (Fig. 1B), and BSE

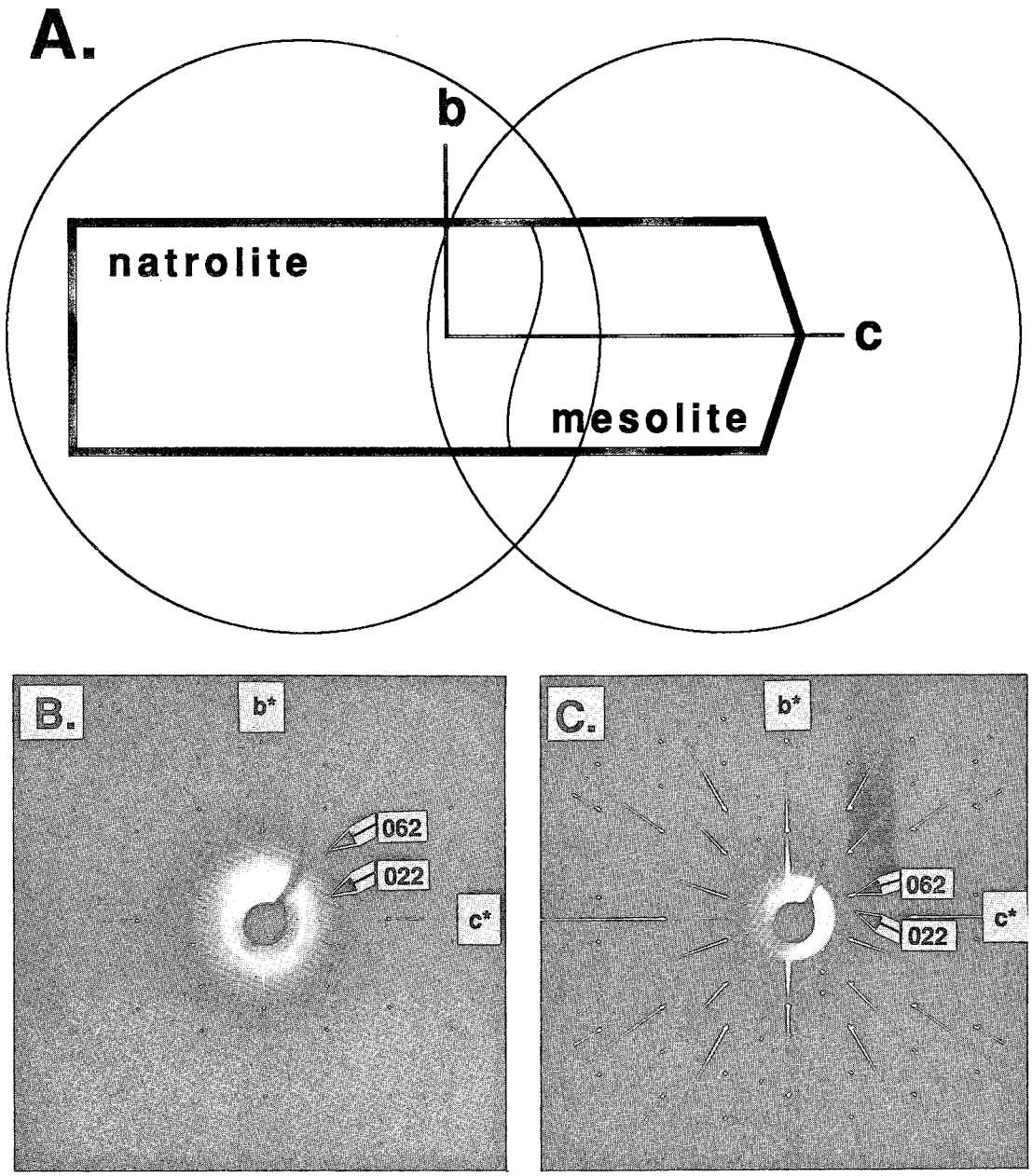

FIG. 2. A. Sketch showing the crystal orientation with respect to the incident X-ray beam, which is parallel to *a*, the precession axis. B. Diffraction pattern b^*c^* showing the (022) and (062) spots of natrolite. C. Diffraction pattern b^*c^* showing the (022) and (062) spots of mesolite, confirming its relationship to natrolite by a tripling of *b*.

images. Both show the chemical changes occurring in these samples. From Figure 1B, there seems to be very little Ca in the natrolite, and the BSE images show brighter areas corresponding to the higher atomic number of mesolite.

X-Ray Diffraction

As a structural verification, precession X-ray-diffraction photographs were obtained on a composite crystal mounted with *c* parallel to the dial axis and *a* as the precession axis (Fig. 2A) in order to compare the (0*kl*) of the two parts. The natrolite tip of the crystal was first placed in the X-ray beam, followed by the mesolite tip. (The circles on the crystal drawing represent the approximate diameter of the X-ray beam). The results confirm the optical and chemical data. Extra diffraction spots occur in the mesolite photo owing to the tripling of its *b* axis. The position of the (022) natrolite reflection (Fig. 2B) is occupied by the (062) reflection of mesolite (Fig. 2C), and a new reflection for (022) in mesolite appears at a smaller 2θ value (Fig. 2C).

Summary

Natrolite and mesolite are easily distinguished by their optical properties. Routine optical microscopy led to the discovery of composite natrolite–mesolite crystals in the Imnaha Basalt of the Columbia River Basalt Group. The crystals began growth as natrolite, and ended as mesolite. Natrolite and mesolite deviate only slightly from their ideal formulas (Tschernich 1992, Ross *et al.* 1992). Thus, a change in the petrogenetic conditions occurred during crystal growth, probably a change in fluid chemistry to higher levels of Ca. Laboratory experiments would be required to verify this, but natrolite is difficult to synthesize, and mesolite has not been synthesized (Tschernich 1992).

Acknowledgements

The authors thank the two reviewers and R.F. Martin for improving this manuscript. The Ca scan and the BSE image were provided by Scott Cornelius and Diane Johnson of The GeoAnalytical Laboratory, Department of Geology, Washington State University, Pullman, Washington. MEG thanks the Idaho Mining and Mineral Resources and Research Institute for partial support of this project.

References

Artioli, G., Smith, J.V. & Kvick, Å. (1984): Neutron diffraction study of natrolite, $Na_2Al_2Si_3O_{10}\cdot 2H_2O$, at 20 K. *Acta Crystallogr.* **C40**, 1658-1662.

———, ——— & Pluth, J.J. (1986): X-ray structure refinement of mesolite. *Acta Crystallogr.* **C42**, 937-942.

Bence, A.E. & Albee, A.L. (1968): Empirical correction factors for the electron microanalysis of silicates and oxides. *J. Geol.* **76**, 382-403.

Bloss, F.D. (1981): *The Spindle Stage: Principles and Practice.* Cambridge University Press, Cambridge, U.K.

Gottardi, G. & Galli, E. (1985): *Natural Zeolites.* Springer-Verlag, Berlin.

Gunter, M.E., Bloss, F.D. & Su, Shu-Chun (1988): EXCALIBR revisited. *Am. Mineral.* **73**, 1481-1482.

Hooper, P.R., Webster, G.D. & Camp, V.E. (1985): Geologic map of the Clarkston 15 minute quadrangle, Washington and Idaho. *Wash. Div. Geol. Earth Resources*, **GM-31**.

Ross, M., Flohr, M.J.K & Ross, D.R. (1992): Crystalline solution series and order–disorder within the natrolite mineral group. *Am. Mineral.* **77**, 685-703.

Su, Shu-Chun, Bloss, F.D. & Gunter, M.E. (1987): Procedures and computer programs to refine the double variation method. *Am. Mineral.* **72**, 1011-1013.

Tschernich, R.W. (1992): *Zeolites of the World.* Geoscience Press, Phoenix, Arizona.

Received April 21, 1992, revised manuscript accepted September 9, 1992.