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The Crystal Chemistry of Zeolites

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INTRODUCTION

Definition of zeolite

This chapter discusses the crystal chemistry, emphasizing observed chemical variations, for those zeolites that completely fulfill the requirements of Smith (1963) for a zeolite. These requirements include: (a) a three-dimensional framework of tetrahedra occupied more than 50% by Si and Al; (b) an "open" structure with a framework density (i.e. number of tetrahedral atoms per 1000 Å^3) lower than 20 (Brunner and Meier 1989) and hence enclosing cavities connected by windows larger than regular six-membered rings; and (c) an extraframework content represented by cations and water molecules. Thus, this chapter will not deal with those phases which are commonly classified as feldspathoids (leucite, pollucite) and those that can be classified as beryllo-phosphates (pahasapaite, weinebeneite), beryllo-silicates (chiavennite, hsianghualite), or zinc-silicates (gaultite). The requirements of Smith (1963) account for the characteristic properties of zeolites (molecular sieve, reversible dehydration, cation exchange), and although for some zeolite species, cation exchange is incomplete or is not yet reported, the presence of large windows (requisite b) reasonably assures its feasibility. Using the above criteria, all minerals known to date which can be classified as zeolites are listed in Table 1.

In the general formula $M_xD_y[Al_{x+2y}Si_{n-(x+2y)}O_{2n}]\cdot mH_2O$, where M are monovalent and D are divalent cations, it is possible to distinguish two parts, which although very different, are mutually dependent and form a homogeneous complex endowed with exclusive chemico-physical properties. The portion in square brackets represents the tetrahedral framework and is characterized by an overall negative charge which increases as the Si/Al ratio decreases. The part outside the square brackets consists of exchangeable extraframework cations, which neutralize the framework negative charge, and, finally, water molecules which often coordinate the extraframework cations.

Classification

Due to the large variability in chemical composition, a reliable classification in the zeolite family is possible only on the basis of structural considerations. In framework silicates (tectosilicates) such as zeolites, the primary building units (TO_4 tetrahedra) are linked so as to form a three-dimensional network where all oxygens are shared by two tetrahedra, the sharing coefficient (Zoltai 1960) being 2 or somewhat smaller. Although numerous theoretical networks can be obtained in this way, only a small number of complex structural units (secondary building units = SBU) formed by a finite (up to 16) number of tetrahedra have been recognized in silicate frameworks so far (Meier 1968, Meier and Olson 1970). The SBU are assembled in different ways to form frameworks

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Table 1. Schematic chemical formulae of natural zeolites. STC = Structure Type Code; R_a = Average Si/(Si+Al+Be); R_r = Range of Si/(Si+Al+Be); DEC = Dominant Extraframework Cations; SEC = Subordinate Extraframework Cations. *Italic:* Be-bearing species. Underlined: species with an "interrupted framework"

STC	**Zeolite**	**Schematic chemical formula**	**R_a**	**R_r**	**DEC**	**SEC**
ANA	Analcime Wairakite	$Na_{16}[Al_{16}Si_{32}O_{96}]\cdot 16H_2O$ $Ca_8[Al_{16}Si_{32}O_{96}]\cdot 16H_2O$	0.67 0.67	0.60-0.74 0.65-0.70	Na Ca	Ca,K,Mg,Cs Na,Cs
BEA	Tschernichite	$Ca[Al_2Si_6O_{16}]\cdot 8H_2O$	0.77	0.73-0.80	Ca	Na,Mg
BIK	Bikitaite	$Li_2[Al_2Si_4O_{12}]\cdot 2H_2O$	0.66	-	Li	-
BOG	Boggsite	$Ca_7(Na,K)_4[Al_{18}Si_{78}O_{192}]\cdot 70H_2O$	0.81	-	Ca	Na,K,Mg
BRE	Brewsterite	$(Sr,Ba)_2[Al_4Si_{12}O_{32}]\cdot 10H_2O$	0.74	0.73-0.74	Sr,Ba	Ca,Na,K
CHA	Chabazite Willhendersonite	$(Ca_{0.5},Na,K)_4[Al_4Si_8O_{24}]\cdot 12H_2O$ $Ca_2(Ca_{0.5},K)_2\,[Al_6Si_6O_{24}]\cdot 10H_2O$	0.67 0.50	0.58-0.81 -	Ca,Na,K Ca	Sr,Mg,Ba K
DAC	Dachiardite	$(Ca_{0.5},Na,K)_4[Al_4Si_{20}O_{48}]\cdot 18H_2O$	0.81	0.77-0.86	Na,Ca,K	Cs
EAB	Bellbergite	$Ca_5(Sr,Ba)_3(Na,K)_2[Al_{18}Si_{18}O_{72}]\cdot 30H_2O$	0.51	-	Ca	K,Sr,Na,Ba
EDI	Edingtonite	$Ba_2[Al_4Si_6O_{20}]\cdot 8H_2O$	0.60	0.59-0.61	Ba	K
EPI	Epistilbite	$Ca_3[Al_6Si_{18}O_{48}]\cdot 16H_2O$	0.75	0.73-0.77	Ca	Na,K
ERI	Erionite	$K_2(Ca_{0.5},Na)_7[Al_9Si_{27}O_{72}]\cdot 28H_2O$	0.75	0.68-0.79	Ca,Na,K	Mg
FAU	Faujasite	$(Na,K,Ca_{0.5},Mg_{0.5})_{56}[Al_{56}Si_{136}O_{384}]\cdot 235H_2O$	0.71	0.68-0.73	Na,Ca	K,Mg
FER	Ferrierite	$(Mg_{0.5},Na,K)_6[Al_6Si_{30}O_{72}]\cdot 20H_2O$	0.83	0.78-0.88	Mg,Na,K	Ca,Sr,Ba
GIS	Gismondine Garronite Gobbinsite Amicite	$Ca_4[Al_8Si_8O_{32}]\cdot 16H_2O$ $Ca_{2.5}Na[Al_6Si_{10}O_{32}]\cdot 13H_2O$ $Na_4Ca[Al_6Si_{10}O_{32}]\cdot 11H_2O$ $Na_4K_4[Al_8Si_8O_{32}]\cdot 10H_2O$	0.53 0.63 0.64 0.50	0.50-0.54 0.60-0.65 - -	Ca Ca Na Na,K	Na,K Na,K Ca,K,Mg Ca
GME	Gmelinite	$(Na,K,Ca_{0.5})_8[Al_8Si_{16}O_{48}]\cdot 22H_2O$	0.69	0.65-0.72	Na,Ca,K	Sr
GOO	Goosecreekite	$Ca_2[Al_4Si_{12}O_{32}]\cdot 10H_2O$	0.75	-	Ca	-
HEU	Heulandite Clinoptilolite	$(Na,K,Ca_{0.5})_7[Al_7Si_{29}O_{72}]\cdot 22H_2O$	0.81	0.73-0.85	Ca,Na,K	Mg,Sr,Ba
LAU	Laumontite	$Ca_4[Al_8Si_{16}O_{48}]\cdot 16H_2O$	0.67	0.65-0.69	Ca	Na,K

Table 1, continued

LEV	Levyne	$(Ca_{0.5},Na)_6[Al_6Si_{12}O_{36}]\cdot 18H_2O$	0.66	0.62-0.70	Ca,Na	K
LOV	*Lovdarite*	$Na_{13}K_4[Be_8AlSi_{27}O_{72}]\cdot 20H_2O$	0.75	-	Na	K
LTL	Perlialite	$K_9Na(Ca,Mg,Sr)[Al_{12}Si_{24}O_{72}]\cdot 16H_2O$	0.66	0.65-0.67	K	Na,Ca,Sr,Mg
MAZ	Mazzite	$Mg_{2.5}K_2Ca_{1.5}[Al_{10}Si_{26}O_{72}]\cdot 30H_2O$	0.72	-	Mg	K,Ca,Na,Ba
MER	Merlinoite	$K_6Ca_2Na[Al_{11}Si_{21}O_{64}]\cdot 22H_2O$	0.66	0.62-0.71	K	Na,Ca,Ba
MFI	Mutinaite	$Na_3Ca_4[Al_{11}Si_{85}O_{192}]\cdot 60H_2O$	0.88	-	Ca	Na,Mg,K
MON	Montesommaite	$K_9[Al_9Si_{23}O_{64}]\cdot 10H_2O$	0.70	-	K	Na
MOR	Mordenite Maricopaite	$Na_3Ca_2K[Al_8Si_{40}O_{96}]\cdot 28H_2O$ $Pb_7Ca_2[Al_{12}Si_{36}(O,OH)_{100}]\cdot n\ (H_2O,OH)$	0.83 0.75	0.80-0.86 -	Na,Ca,K Pb	Mg,Sr,Ba Ca
NAT	Natrolite Mesolite Scolecite Gonnardite-Tetranatrolite	$Na_{16}[Al_{16}Si_{24}O_{80}]\cdot 16H_2O$ $Na_{16}Ca_{16}[Al_{48}Si_{72}O_{240}]\cdot 64H_2O$ $Ca_8[Al_{16}Si_{24}O_{80}]\cdot 24H_2O$ $Na_{12}Ca_{2.5}[Al_{17}Si_{23}O_{80}]\cdot 20H_2O$ (Z=1/2)	0.60 0.60 0.60 0.57	0.58-0.61 0.58-0.62 0.60-0.61 0.52-0.63	Na Na,Ca Ca Na	Ca - Na Ca
NES	Gottardiite	$Ca_5Na_3Mg_3[Al_{19}Si_{117}O_{272}]\cdot 93H_2O$	0.86	-	Ca	Na,Mg,K
OFF	Offretite	$CaMgK[Al_5Si_{13}O_{36}]\cdot 16H_2O$	0.70	0.69-0.74	Ca,Mg,K	Na
PAR	Partheite	$Ca_8[Al_{16}Si_{16}O_{60}(OH)_8]\cdot 16H_2O$	0.51	0.50-0.52	Ca	Na
PAU	Paulingite	$K_{58}Ca_{42}Na_{10}Ba_{10}[Al_{172}Si_{500}O_{1344}]\cdot 490H_2O$	0.74	0.73-0.77	K,Ca	Na,Ba,Mg,Sr
PHI	Phillipsite Harmotome	$K_2(Na,Ca_{0.5})_3[Al_5Si_{11}O_{32}]\cdot 12H_2O$ $Ba_2(Na,Ca_{0.5})[Al_5Si_{11}O_{32}]\cdot 12H_2O$	0.69 0.71	0.57-0.77 0.68-0.76	K,Na,Ca Ba	Ba K,Na,Ca
ROG	Roggianite	$Ca_{14}(Na,K)[Be_5Al_{15}Si_{28}O_{90}(OH)_{14}](OH)_2\cdot 34H_2O$	0.58	-	Ca	Na,K
STI	Stilbite Stellerite Barrerite	$NaCa_4[Al_9Si_{27}O_{72}]\cdot 30H_2O$ $Ca_8[Al_{16}Si_{56}O_{144}]\cdot 56H_2O$ $Na_{16}[Al_{16}Si_{56}O_{144}]\cdot 52H_2O$	0.75 0.77 0.77	0.71-0.78 0.75-0.78 0.77-0.78	Ca,Na Ca Na	K,Mg Na,K,Mg K,Ca,Mg
	Terranovaite	$Na_4Ca_4[Al_{12}Si_{68}O_{160}]\cdot >29H_2O$	0.85	-	Na	Ca,K,Mg
THO	Thomsonite	$Ca_7Na_5[Al_{19}Si_{21}O_{80}]\cdot 24H_2O$	0.53	0.50-0.58	Ca,Na	K,Mg,Sr
YUG	Yugawaralite	$Ca_2[Al_4Si_{12}O_{32}]\cdot 8H_2O$	0.74	0.74-0.76	Ca	Na
Unk.	Cowlesite	$Ca_{5.5}(Na,K)[Al_{12}Si_{18}O_{60}]\cdot 36H_2O$	0.61	0.58-0.62	Ca	Na,K,Mg
*	Tschörtnerite	$Ca_6Cu_3SrK(OH)_9[Al_{12}Si_{12}O_{48}]\cdot 14H_2O$	0.50	-	Ca	Cu, Sr, K, Ba

* Not yet assigned by IZA

with different topologies (architectures) which generally are coded (Structure Type) after the name of the type material (Meier and Olson 1992). Each structure type is characterized by a topological (i.e. the highest space group) symmetry that is defined (Smith 1974) as the symmetry of a framework when idealized into its most regular shape by movements which leave intact the topologic relationship between nodes (i.e. tetrahedral centers).

Fifty-two zeolite species distributed in 38 different structure types have been identified to date. The topological symmetry (TS) of the framework may be reduced to the real symmetry (RS) of the mineral (Table 2) by one or more of the following causes (Gottardi 1979): (a) ordered distribution of tetrahedral cations; (b) ordered distribution of extraframework ions (cations or water molecules or holes); (c) squeezing of the framework; and d) repulsion of extraframework cations. When reading Table 2, an interesting question is whether phillipsite and harmotome with the same framework (PHI) and symmetry (P21/m) and heulandite and clinoptilolite with the same framework (HEU) and symmetry (C2/m) should be considered different zeolite species. The sharing coefficient of 2 is violated only in three minerals (maricopaite, partheite, and roggianite) which display a so-called "interrupted" tetrahedral framework in which one apex of some tetrahedra is occupied by an (OH) group instead of an O atom and hence is not shared by the adjacent tetrahedron.

Chemistry

The general formula of a zeolite permits a large chemical variability, the only constraint being Lowenstein's rule ($Si \geq Al$). A common feature of the chemical compositions is the presence of O, Si, Al, Ca, Mg, Ba, Na, K, and H as fundamental elements and of Fe, Sr, Li, Be, Cs, Cu, and Pb mainly as subordinate or occasional elements. The assignment of Fe to the tetrahedral framework, to extraframework sites, or as micro-impurities of Fe oxides and/or hydroxides is still the subject of open debate because of the very low percentages commonly detected in natural samples and because of the paucity of related studies. Different spectroscopic methods (optical absorption, Mössbauer, electron paramagnetic resonance) show that the Fe in red heulandite crystals from Val di Fassa (Italy) is primarily in hematite and marcasite inclusions and only subordinately as Fe^{3+} in extraframework sites (Bonnin and Calas 1978). X-ray diffraction, scanning electron microscopy, X-ray photoelectron spectroscopy, X-ray absorption spectroscopy, Mössbauer, and energy dispersive X-ray analyses carried out on a clinoptilolite- and mordenite-bearing sample from Cuba revealed that the Fe is present as an extraframework cation (octahedral coordination), and there was no evidence of Fe in tetrahedral coordi-nation (Marco et al. 1995). On the other hand, Fe in tetrahedral sites was detected in synthetic Fe-silicalite and in synthetic Fe-silicate analogs of mordenite (Chandwadkar et al. 1991, 1992). Most zeolites are white or pale colored and show negligible Fe contents, and in many occurrences, Fe-rich smectite formed before or after the crystallization of zeolites. These data suggest that Fe from zeolite analyses should be assigned to impurities and, hence, not considered in the calculation of the chemical formula.

The chemical parameters reported in Table 1 for each zeolite species are averages of compositions from the literature where: (a) data were obtained on crystals, the purity and mineralogical nature of which were previously tested by X-ray analyses; and (b) data are "reliable," i.e. with the content of tetrahedral cations (Si+Al) close to half of the oxygen atoms and the balance error $E = [(Al-Al_{theor.})/Al_{theor.}] \times 100$ lower than 10%, where $Al_{theor} = (Na+K+Li+Cs) + 2(Ca+Mg+Sr+Ba+Pb)$ (Passaglia 1970). A general overview of the chemistry of the zeolite family is presented in Table 1 in which the framework content is expressed by the ratio, $R = Si/(Si+Al+Be)$, which represents the percentage of tetrahedra occupied by Si, and the extraframework content expressed in terms of the dominant (DEC) and subordinate (SEC) cations.

Table 2. Symmetry and unit-cell parameters of natural zeolites.

Structure Type Zeolite	Topological Symmetry (TS)	Real Symmetry (RS)	*a* (Å)	*b* (Å)	*c* (Å)	β (°)
ANA	Ia3d					
Analcime		I4$_1$/acd, Ibca, I2/a	13.66-13.73	13.68-13.73	13.66-13.74	90.20-90.38
Wairakite		I2/a, Ia	13.66-13.70	13.64-13.66	13.55-13.59	90.20-90.55
BEA	P4$_1$22					
Tschernichite		P4/mmm (?)	12.88		25.02	
BIK	Cmcm					
Bikitaite		P2$_1$, P1*	8.61-8.63	4.95-4.96	7.60-7.64	114.42-114.57
BOG	Imma					
Boggsite		Imma	20.24-20.25	23.80-23.82	12.78-12.80	
BRE	P2$_1$/m					
Brewsterite		6.77-	6.77-6.82	17.45-17.60	7.73-7.75	94.27-94.54
CHA	R$\bar{3}$ m					
Chabazite[1]		R$\bar{3}$m	13.69-13.86		14.80-15.42	
Willhendersonite**		P$\bar{1}$	9.18-9.21	9.20-9.22	9.44-9.50	91.42-92.70
DAC	C2/m					
Dachiardite		C2/m	18.62-18.73	7.49-7.54	10.24-10.31	107.87-108.74
EAB	P6$_3$/mmc					
Bellbergite		P6$_3$/mmc	13.24		15.99	
EDI	P$\bar{4}$2$_1$m					
Edingtonite		P$\bar{4}$2$_1$m, P2$_1$2$_1$2	9.54-9.58	9.66	6.52	
EPI	C2/m					
Epistilbite		C2, C1***	9.08-9.10	17.74-17.80	10.20-10.24	124.55-124.68
ERI	P6$_3$/mmc					
Erionite		P6$_3$/mmc	13.19-13.34		15.04-15.22	

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* α = 89.89° γ = 89.96°

[1] Cell parameters of the pseudo-hexagonal cell

** α = 91.72-92.34° γ = 90.05-90.12°

*** α = 89.95° γ = 90.00°

Table 2, continued

FAU	Fd3m					
Faujasite		Fd3m	24.60-24.78			
FER	Immm					
Ferrierite		Immm(Pnnm), $P2_1/n$	18.90-19.45	14.12-14.28	7.48-7.54	90.0(1)
GIS	$I4_1/amd$					
Gismondine		$P2_1/c$	10.01-10.04	10.60-10.64	9.82-9.84	92.23-92.56
Garronite		I2/a	9.88	10.28	9.876	90.11
Gobbinsite		$Pmn2_1$	10.10	9.77-9.80	10.17	
Amicite		I2	10.23	10.42	9.88	88.32
GME	$P6_3/mmc$					
Gmelinite		$P6_3/mmc$	13.62-13.80		9.97-10.25	
GOO	$C222_1$					
Goosecreekite		$P2_1$	7.40-7.52	17.44-17.56	7.29-7.35	105.44-105.71
HEU	C2/m					
Heulandite		C2/m	17.62-17.74	17.81-18.05	7.39-7.53	116.13-116.90
Clinoptilolite		C2/m				
LAU	C2/m					
Laumontite		C2/m	14.69-14.89	13.05-13.17	7.53-7.61	110-113
LEV	$R\bar{3}m$					
Levyne		$R\bar{3}m$	13.32-13.43		22.66-23.01	
LOV	$P4_2/mmc$					
Lovdarite		Pma2	39.58	6.93	7.15	
LTL	P6/mmm					
Perlialite		P6/mmm	18.49-18.54		7.51-7.53	
MAZ	$P6_3/mmc$					
Mazzite		$P6_3/mmc$	18.39		7.65	
MER	I4/mmm					
Merlinoite		Immm	13.86-14.14	14.13-14.23	9.95-10.05	
MFI	Pnma					
Mutinaite		Pnma	20.22	20.05	13.49	
MON	$I4_1/amd$					
Montesommaite		$I\bar{4}2d$ ($I4_1md$)	7.14		17.31	
MOR	Cmcm					
Mordenite		$Cmc2_1$	18.05-18.25	20.35-20.53	7.49-7.55	
Maricopaite		Cm2m	19.43	19.70	7.54	

Table 2, concluded

NAT	$I4_1/amd$					
Natrolite		Fdd2	18.28-18.42	18.56-18.70	6.46-6.61	
Mesolite		Fdd2	18.34-18.44	56.52-56.69	6.52-6.56	
Scolecite		F1d1	18.50-18.55	18.90-18.97	6.52-6.53	90.44-90.65
Gonnardite-Tetranatrolite		$I\bar{4}2d$	13.04-13.27		6.58-6.64	
NES	Fmmm					
Gottardiite		Cmca	13.70	25.21	22.66	
OFF	$P\bar{6}m2$					
Offretite		$P\bar{6}m2$	13.27-13.32		7.56-7.61	
PAR	C2/c					
Partheite		C2/c	21.55-21.59	8.76-8.78	9.30-9.31	91.47-91.55
PAU	Im3m					
Paulingite		Im3m	35.05-35.12			
PHI	Bmmb					
Phillipsite[2]		$P2_1/m$	9.86-10.01	14.12-14.34	14.16-14.42	
Harmotome[2]		$P2_1/m$	9.81-9.92	14.10-14.17	14.26-14.34	
ROG	I4/mcm					
Roggianite		I4/mcm	18.33-18.37		9.16-9.19	
STI	Fmmm					
Stilbite[3]		C2/m	13.59-13.66	18.18-18.33	17.71-17.84	90.20-91.15
Stellerite		Fmmm	13.57-13.63	18.16-18.27	17.82-17.87	
Barrerite		Amma	13.59-13.64	18.18-18.20	17.79-17.84	
TER	Cmcm					
Terranovaite		C2cm	9.75	23.88	20.07	
THO	Pmma					
Thomsonite		Pncn	13.00-13.18	13.04-13.16	13.09-13.24	
YUG	C2/m					
Yugawaralite		Pc	6.70-6.73	13.97-14.01	10.04-10.09	111.07-111.20
UNKN.	?					
Cowlesite		Orthorhombic(?)	23.21-23.30	30.60-30.68	24.98-25.04	
?	$Fm\bar{3}m$					
Tschörtnerite		$Fm\bar{3}m$	31.62			

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[2] Unit-cell parameters of the pseudo-orthorhombic cell (Z=2) with β = 90°

[3] Unit-cell parameters of the pseudo-orthorhombic F2/m cell (Z=2)

Framework content. The tetrahedra in zeolites are exclusively occupied by Si and Al, except in roggianite and lovdarite where remarkable amounts of Be have been detected. The average R (R_a) uniformly ranges from 0.50 in amicite, willhendersonite and tschörtnerite to 0.88 in mutinaite; the minimum Si/Al ratio (1) allowed by Lowenstein's rule is observed in only three zeolite species. Mutinaite is the most Si-rich zeolite found to date. Zeolites with R_a of 0.50-0.60 (amicite, bellbergite, edingtonite, gismondine, gonnardite-tetranatrolite, mesolite, natrolite, partheite, roggianite, scolecite, thomsonite, tschörtnerite, willhendersonite) may be classified as Si-poor. Zeolites with $R_a > 0.80$ (boggsite, dachiardite, ferrierite, gottardiite, heulandite-clinoptilolite, mordenite, mutinaite, terranovaite) may be classified as Si-rich. All others can be considered as "intermediate." The degree of variability of R (R_r) varies greatly among zeolite species. The maximum range (0.23) is displayed by chabazite, and if the isostructural willhendersonite is considered, the structure type CHA exhibits R ranging from 0.50 to 0.81 ($R_r = 0.31$). High (≥ 0.10) R_r values are also shown by phillipsite (0.20), analcime (0.14), heulandite-clinoptilolite (0.12), erionite and gonnardite-tetranatrolite (0.11), and ferrierite (0.10). Intermediate (0.09-0.06) R_r values are shown by dachiardite, merlinoite (0.09), levyne, harmotome, thomsonite (0.08), tschernichite, gmelinite (0.07), stilbite and mordenite (0.06). Low (≤ 0.05) R_r values are shown by the other zeolite species.

Extraframework content . An average water content for each zeolite species is difficult to assess because a comparison of the data from the literature is frustrated by the use of a wide variety of analytical techniques, instruments, and experimental conditions. Furthermore, most recent chemical analyses were performed by electron microprobe and, when reported, the water content is the result of normalization of data to 100%. A representative number of water molecules given in the formulae (Table 1) is taken from Gottardi and Galli (1985). This number is known to be related to both structural and chemical parameters because it increases with the increasing void volume/total volume, Si/Al, and divalent/monovalent cation ratios.

Table 1, where the dominant cations (DEC) are in order of frequency and the subordinate cations (SEC) are in order of abundance, shows that Ca is the most common cation, being the DEC in all analyzed samples of many zeolitic species (wairakite, tschernichite, boggsite, willhendersonite, bellbergite, epistilbite, gismondine, garronite, goosecreekite, laumontite, scolecite, gottardiite, partheite, roggianite, stellerite, cowlesite, yugawaralite, mutinaite, tschörtnerite). It is also one of the DEC in many other species (chabazite, dachiardite, erionite, faujasite, gmelinite, heulandite-clinoptilolite, levyne, mordenite, mesolite, offretite, paulingite, phillipsite, stilbite, thomsonite). Na is the DEC in analcime, barrerite, gobbinsite, gonnardite-tetranatrolite, lovdarite, natrolite, and terranovaite and among the DEC in amicite, chabazite, dachiardite, erionite, faujasite, ferrierite, gmelinite, heulandite-clinoptilolite, levyne, mordenite, mesolite, phillipsite, stilbite, and thomsonite. K is the DEC in merlinoite, montesommaite, and perlialite and among the DEC in amicite, chabazite, dachiardite, erionite, offretite, ferrierite, gmelinite, heulandite-clinoptilolite, mordenite, paulingite, and phillipsite. Mg is the DEC in mazzite and among the DEC in ferrierite and offretite. Ba is the DEC in edingtonite, harmotome, and brewsterite, which also shows Sr as a DEC. Pb is the DEC in maricopaite. Li is the only extraframework cation in bikitaite. Cs has been found as a subordinate cation in some samples of analcime, wairakite, and dachiardite. Although not the DEC, Cu is extraordinarily abundant in tschörtnerite. The distribution of SE C (in order of frequency, Ca, Na, K, Mg, Sr, Ba, Cs, Cu) and the schematic formulae given for each zeolite species allow the definition of epistilbite, gismondine, goosecreekite, laumontite, partheite, roggianite, scolecite, stellerite, tschernichite, willhendersonite, and yugawaralite as Ca-zeolites; barrerite, natrolite, and gonnardite-tetranatrolite as Na-zeolites; montesommaite as a K-zeolite; bikitaite as a Li-zeolite; edingtonite as a Ba-zeolite; maricopaite as a Pb-zeolite; amicite as a (Na,K)-zeolite; analcime-wairakite, mesolite, mutinaite, and terranovaite as Na,Ca-zeolites; faujasite, ferrierite, gottardiite, mazzite, and offretite as Mg-rich zeolites; harmotome as a Ba-rich zeolite; and tschörtnerite as a Cu-rich zeolite. Chabazite, dachiardite, erionite, gmelinite, heulandite-clinoptilolite, mordenite, and phillipsite show Ca, Na, and K to be the DEC; Na, K, and Mg are the DEC in ferrierite; and Ca, Mg, K in offretite. Considering both the R_r values and the DEC of each zeolite species, the order of variability degree is: chabazite (R_r = 0.23; DEC = Ca, Na, K), phillipsite (R_r = 0.20; DEC = Ca, Na, K), heulandite-clinoptilolite (R_r = 0.12; DEC = Ca, Na, K), analcime-wairakite (R_r = 0.14; DEC = Na, Ca), erionite (R_r = 0.11; DEC = Ca, Na, K), ferrierite (R_r = 0.10; DEC = Mg, Na, K), gmelinite (R_r = 0.07; DEC = Na, Ca, K), levyne (R_r = 0.08; DEC = Ca, Na), and mordenite (R_r = 0.06; DEC = Na, Ca, K). The degrees of variability for the other species are very low and comparable.

Symmetry and unit-cell parameters

With the notable exception of cowlesite, the crystal structures of all zeolite species are known, and the symmetry (RS) is given in Table 2 along with the symmetry of the pertaining structure type (TS). The RS is the same as the TS for about half of the species. In all other species, the RS is slightly or markedly lower due to one or more of the causes described above. These factors explain the existence of crystals (also coexisting) of the same species with different symmetry: tetragonal, orthorhombic, and monoclinic in analcime; monoclinic and triclinic in bikitaite and epistilbite; tetragonal and orthorhombic in edingtonite; orthorhombic and monoclinic in ferrierite. The possible tetragonal $P4/mmm$ symmetry of tschernichite is higher than the TS ($P4_122$) of the pertaining structure type (BEA) and probably is due to an intergrowth of an enantiomorphic pair in the tetragonal space group $P4_122$ and $P4_322$ and a triclinic polymorph with space group $P\bar{1}$ (Boggs et al. 1993). The rhombohedral $R\bar{3}m$ symmetry given for chabazite represents an average structure actually formed by oriented aggregates of triclinic $P\bar{1}$ domains.

Table 2 emphasizes that: (a) the cubic (faujasite, paulingite, tschörtnerite), rhombohedral (levyne), and triclinic (willhendersonite and sometimes bikitaite and epistilbite) symmetries are scarcely represented; (b) the hexagonal (bellbergite, erionite, gmelinite, perlialite, mazzite, offretite) and tetragonal (tschernichite, montesommaite, gonnardite-tetranatrolite, roggianite, and locally analcime and edingtonite) symmetries are moderately represented; and (c) the orthorhombic and monoclinic symmetries are the most common.

Particularly large variations (in Å) in unit- cell parameters occur in chabazite (a = 0.17, c = 0.62), phillipsite (a = 0.15, b = 0.22, c = 0.26), ferrierite (a = 0.55, b = 0.16, c = 0.06), merlinoite (a = 0.28, b = 0.10, c = 0.10), laumontite (a = 0.20, b = 0.12, c = 0.08, β = 3°), gmelinite (a = 0.18, c = 0.28), erionite (a = 0.15, c = 0.18), mordenite (a = 0.20, b = 0.18, c = 0.06), heulandite-clinoptilolite (a = 0.12, b = 0.24, c = 0.14, β = 0.77°), and levyne (a = 0.11, c = 0.35). Crystallochemical studies show that unit-cell parameters are correlated with the framework contents (generally the unit-cell dimensions increase with R decreasing) in chabazite, ferrierite, erionite and mordenite, with the extraframework cations in gmelinite and levyne, and with both in phillipsite. The largest variations are shown by chabazite, which, because of its large variability in chemical composition, appears to be the most "flexible"zeolite species.

Occurrence

Zeolites occur in a wide variety of environments in two major types of occurrences as: (a) macroscopic and microscopic crystals, often euhedral, in veins of ore deposits or in fractures and vugs (amygdules) of plutonic rocks (pegmatite, granite, etc.), metamorphic rocks (gneiss, amphibolite, etc.), and mafic lavas (basalt); and (b) submicroscopic (<20-

30 μm) crystals more or less uniformly distributed in vitroclastic sediments which have undergone diagenetic or low-grade metamorphic processes. The former occurrences (hereafter named "amygdaloidal") include all the species known to date (52), and the zeolites are commonly confined to voids of the host rock, the walls of which are unaltered or locally lined by a thin layer of smectite and/or Fe hydroxides. Up to five or six zeolite species may be associated in such occurrences, also possibly including carbonates (calcite, strontianite, aragonite, siderite, ankerite, rhodochrosite, dolomite, and barytocalcite), sulfates (barite, celestine), phosphates (apatite, fairfieldite), silicates (apophyllite, gyrolite, pectolite, pumpellyite, prehnite, babingtonite, datolite, corrensite, vesuvianite, and eucryptite), oxides and hydroxides (goethite, gibbsite, cuprite, and manganite), sulfides (galena, pyrite, pyrrhotite, chalcopyrite, sphalerite, and cinnabar), silica minerals (quartz, opal, chalcedony, tridymite, and cristobalite), ettringite, strätlingite, mimetite, colemanite, borax, and native copper. Uniquely for these occurrences, the zeolite chemistry does not seem to correlate with that of the host rock. The latter occurrences (hereafter named "sedimentary") can best be detected by X-ray powder diffraction because of the very fine-grained nature of the rocks. Such materials are characterized by a limited number of species (generally analcime, chabazite, heulandite-clinoptilolite, phillipsite, mordenite, erionite, laumontite, and ferrierite; rarely harmotome and faujasite; also gonnardite, gismondine, and natrolite) and the coexistence of at least two different zeolite species and association with other authigenic phases (feldspar, calcite, quartz, opal, tridymite, cristobalite, thaumasite, gypsum, thenardite, dawsonite, searlesite, smectites, iron oxides, and hydroxides), fresh or palagonitized volcanic glass, and lithic fragments. Unlike the former occurrences, the chemistry of sedimentary zeolites is correlated with that of the parent glass and the pore waters, particularly the pH.

DETAILED CRYSTAL CHEMISTRY

The main crystallochemical features of each zeolite species are described below in detail. As noted above, TS refers to topological symmetry, RS to real symmetry, R to [Si/(Si+Al+Be)], DEC to dominant extraframework cations, and SEC to subordinate extraframework cations. Where not specifically reported, the content of extraframework cations is expressed in atoms per formula unit (p.f.u.). For additional information, the reader is referred to the monographs by Gottardi and Galli (1985) and Tschernich (1992); the latter includes a large number of occurrences and descriptions that have not been previously published.

Structure Type: ANA

Analcime: $Na_{16}[Al_{16}Si_{32}O_{96}]\cdot 16H_2O$

Wairakite: $Ca_8[Al_{16}Si_{32}O_{96}]\cdot 16H_2O$

The identification of several samples with intermediate chemical compositions and physical properties (refractive indices, density) reveals the existence of a continuous isomorphous series between these two end-members (Surdam 1966, Seki and Oki 1969, Seki 1971, Harada et al. 1972, Harada and Sudo 1976, Aoki and Minato 1980) and permits a classification of the zeolite with Na/(Na+Ca) > 0.5 as analcime and with Na/(Na+Ca) < 0.5 as wairakite. Analcime is much more abundant than wairakite and occurs: (a) in vugs of plutonic (pegmatite) and volcanic (basalt, dolerite) rocks; (b) as an alteration product of volcanic glass and "primary" leucite and nepheline from pyroclastic sediments (tuff, tuffite, ignimbrite) diagenetically altered in both continental and marine environments or metamorphosed under low-grade conditions; and (c) as phenocrysts in alkalic igneous rocks. The chemistry varies considerably from the schematic formula, where R = 0.67, Na/(Na+Ca) = 1, and with no other extraframework cations. R ranges from 0.60 in "primary" phenocrysts in theralite from the Square Top intrusion, Australia (Wilkinson and Hensel 1994), to 0.74 in sedimentary crystals in silicic ash-flow tuffs at Yucca Mountain, Nevada (Broxton et al. 1987). Sedimentary analcime samples are typically more siliceous than the schematic formula. The Na/(Na+Ca) ratio ranges from 1 in many samples to 0.60 in amygdaloidal crystals from Skye, Scotland (Livingstone 1989). The K content is generally lower than 1, but it is anomalously high (1.5-3) in analcime from phonolitic lavas (from a partial "analcimization" of leucite?) from Azerbaja n, Iran (Comin Chiaromonti et al. 1979), and from Eifel, Germany (Adabbo et al. 1994). Mg is generally absent or negligible and reaches a maximum (0.62) in analcime from low-grade metamorphosed basaltic tuffs from the Tanzawa Mts, Japan (Seki and Oki 1969). Sr and Ba are almost absent; but Cs is remarkably high in some amygdaloidal samples. An amygdaloidal sample associated with native copper from lava flows at Jalampura (India) contains 0.81% Cu (Talati 1978). Given these different occurrences, several generalizations can be made. Amygdaloidal analcime generally shows R values close to the average (0.67), although a lower value (0.64) has been reported for crystals from a basalt at Skye, Scotland (Livingstone 1989), and higher values (0.72-0.73) have been found in crystals from pegmatite at Bernic Lake, Canada (Cerny 1972) and from basalts at Husa u Marcinova, Bohemia (Pechar 1988), and Boron, California (Wise and Kleck 1988). The Na/(Na+Ca) ratio is generally very close to 1, but much lower values (0.60-0.70) have been reported for Si-poor crystals from Skye (Scotland) and slightly lower values (0.88) in crystals from Procida, Italy (de'Gennaro et al. 1977), and in Cu-bearing crystals from Jalampura, India (Talati 1978). Mg is generally absent, although in a sample from Sagashima (Japan) it reaches 0.42 (Harada et al. 1972). Samples associated with beryl, apatite, cookeite, and lithiophosphate in the Bernic Lake pegmatite show Cs contents up to 5.06 p.f.u, suggesting a possible existence of an almost continuous isomorphous series between analcime and pollucite (Cerny 1974).

Sedimentary analcime exhibits a mean R value of 0.71 with a large range (0.66-0.74). The lower values (0.66-0.68) have been reported for analcime from ferruginous mudstone (Utada and Vine 1984), palagonitic tuff (Iijima and Harada 1968), altered pyroclastic rocks (Barnes et al. 1984, Noh and Kim 1986), and phonolite (Adabbo et al. 1994). The higher values (0.72-0.74) have been reported for analcime from diagenetically altered silicic (mainly rhyolitic) tuffs (Djourova 1976, Walton 1975, Sheppard and Gude 1969a, 1973; Gude and Sheppard 1988, Broxton et al. 1987). The Na/(Na+Ca) ratio is always very close to 1.

In analcime from low-grade metamorphic rocks, R has a mean value of 0.69 and ranges from 0.66 in crystals from metabasites (Cortesogno et al. 1976) to 0.72 in crystals from rhyolitic sediments (Iijima 1978) and from vitric siltstone and sandstone (Vitali et al. 1995). The Na/(Na+Ca) ratio is about 1, although it is 0.66 in a sample associated with wairakite (Seki and Oki 1969).

Analcime "phenocrysts" from alkalic igneous rocks (phonolite, lamprophyre, blairmorite, tephrite, analcimite, theralite, and tinguaite) have been described from at least a dozen occurrences (Coombs and Whetten 1967, Aurisicchio et al. 1975, Pearce 1970, Woolley and Symes 1976, Wilkinson 1977, Comin-Chiaromonti et al. 1979, Giret 1979, Luhr and Kyser 1989, Avdeev 1992, Viladkar and Avasia 1994, Wilkinson and Hensel 1994). The origin ("primary" or "secondary" as replacement pseudomorphs of leucite or nepheline) is uncertain and is still debated in the literature (Wilkinson 1977, Karlsson and Clayton 1991, 1993; Pearce 1993). For these analcimes, R fluctuates near the mean value (0.67); but in analcime from theralites, R is 0.60, and in analcime from tinguaites, R is 0.73 (Wilkinson and Hensel 1994). The Na/(Na+Ca) ratio is close to 1.

Wairakite is much less common than analcime and is found in geothermal areas and in low- to medium-grade metamorphic rocks (Seki and Oki 1969, Seki 1973, Steiner 1955, Takéuchi et al. 1979, Aoki and Minato 1980, Utada and Vine 1984, Bargar and Beeson 1981). The mean value of R is 0.67, ranging from 0.65 to 0.70; the Na/(Na+Ca) ratio ranges from 0.03 to 0.47. The lowest values of R (0.65) and Na/(Na+Ca) ratio (0.03) are in wairakite from Toi, Japan (Aoki and Minato 1980) and the highest values (0.70 and 0.47, respectively) have been found for wairakite from the Tanzawa Mountains, Japan (Seki and Oki 1969). K and Mg are negligible or absent, and Sr and Ba are absent. Samples from active geothermal areas selectively concentrate Cs, ranging up to 4,700 ppm in crystals from Yellowstone National Park, Wyoming (Keith et al. 1983), and up to 80 ppm in crystals from Wairakei, New Zealand (Steiner 1955).

Optical observations (Akizuki 1980 1981), structure refinements (Mazzi and Galli 1978, Takéuchi et al. 1979, Pechar 1988), and crystallochemical studies (Aoki and Minato 1980, Papezik and Elias 1980) demonstrated that the cubic TS of both zeolite species is lowered to tetragonal, orthorhombic, or monoclinic RS in analcime, and to monoclinic RS in wairakite depending on different Al ordering schemes in the tetrahedral sites and related occupancy of Na and Ca in the extraframework cation sites. The cubic unit-cell parameter of the analcime-wairakite series is 13.66-13.73 Å. The Si/Al ratio correlates with the *a* parameter in analcime (Saha 1959, Coombs and Whetten 1967); correlations between Si, Na, and H_2O contents and monoclinic unit-cell parameters have also been determined for wairakite (Aoki and Minato 1980).

Structure Type: **BEA**

Tschernichite: $Ca[Al_2Si_6O_{16}]\cdot 8H_2O$

Only two occurrences of this zeolite have been described, one at the Neer Road pit, Goble, Oregon, in vugs of basalt associated with boggsite, analcime, offretite, erionite, mordenite, thomsonite, heulandite, levyne, chabazite, okenite, calcite, opal, and native copper (Boggs et al. 1993); and the other at Mt. Adamson, Antarctica, in vugs of basalt, rarely associated with heulandite (Galli et al. 1995). At both localities, tschernichite occurs as large individual crystals (tetragonal dipyramids) and as drusy radiating hemispherical groups. The large crystals are poorer in Si (R = 0.73-0.75), and the Ca content is close to the schematic formula. The drusy crystals are richer in Si (R = 0.79-0.80) and poorer in Ca because the Na and, in the sample from Antarctica, Mg contents are appreciable. The structure is tetragonal $P4/mmm$ with a = 12.88 Å and c = 25.02 Å (Boggs et al. 1993).

Structure Type: **BIK**

Bikitaite: $Li_2[Al_2Si_4O_{12}]\cdot 2H_2O$

Bikitaite occurs in lithium-rich pegmatite dikes at Bikita (Southern Zimbawe) where small transparent crystals are associated with eucryptite, stilbite, quartz, calcite, and allophane (Hurlbut 1957, 1958) and at King's Mountain, North Carolina, where intergrowths of bladed crystals (like jackstraws) are associated with eucryptite, albite, quartz, apatite, and fairfieldite (Leavens et al. 1968). The empirical chemical formulae of the crystals from both occurrences (Kocman et al. 1974, Leavens et al. 1968) are very similar and are close to the schematic formula: R is 0.66, and Li is the only extraframework cation. Crystals from Bikita are monoclinic $P2_1$ with a partially ordered (Si,Al) distribution (Kocman et al. 1974) or triclinic $P1$ with a fully ordered (Si,Al) distribution (Bissert and Liebau 1986). The monoclinic unit-cell dimensions are: a = 8.61-8.63 Å, b = 4.95-4.96 Å, c = 7.60-7.64 Å, β = 114.42-114.57°. In the triclinic crystals: α = 89.89° and γ = 89.96°.

Structure Type: **BOG**

Boggsite: $Ca_7(Na,K)_4[Al_{18}Si_{78}O_{192}]\cdot 70H_2O$

Boggsite occurs with tschernichite from the Neer Road pit, Goble, Oregon (Howard et al. 1990), and from Mt. Adamson, Antarctica (Galli et al. 1995). It occurs in vesicles of basalt as small, colorless, zoned hemispheres embedded in drusy tschernichite at Goble and on a thin layer of smectite at Mt. Adamson. Crystals from the two occurrences display the same tetrahedral content (R = 0.81) and extraframework cations in the same order of abundance (Ca > Na » K > Mg). Nevertheless, in comparison with the holotype sample, where the Ca/Na ratio is 2.6, boggsite from Antarctica is poorer in Ca, richer in Na (Ca/Na = 1.1), and contains more K and Mg. Boggsite is orthorhombic *Imma* (Pluth and Smith 1990) with a = 20.24-20.25 Å, b = 23.80-23.82 Å, c = 12.78-12.80 Å. Optically, it is biaxial negative (Howard et al. 1990).

Structure Type: **BRE**

Brewsterite: $(Sr,Ba)_2[Al_4Si_{12}O_{32}]\cdot 10H_2O$

Brewsterite has been described from five localities: Strontian, Scotland (Mallet, 1859); Burpala pluton, Siberia (Khomyakov et al. 1970); Yellow Lake, Canada (Wise and Tschernich 1978a); Cerchiara, Italy (Cabella et al. 1993); and Harrisville, New York (Robinson and Grice 1993). It occurs in hydrothermal ore veins cutting metamorphic (gneiss, metachert), plutonic (granite, syenite, aplite), and volcanic (porphyritic trachyte) rocks in association with other zeolites (analcime, harmotome, stilbite, heulandite, thomsonite, and mesolite), calcite, barite, strontianite, galena, and pyrite. R is nearly constant (0.73-0.74), and the extraframework content is chiefly Sr and Ba. Ca, Na, K, and Mg are absent or negligible. The Sr/Ba ratio is highly variable and allows the distinction of two types of samples, those with Sr > Ba (Sr-dominant) from Strontian (Schlenker et al. 1977), Burpala pluton, and Yellow Lake (Artioli et al. 1985), and those with Ba > Sr (Ba-dominant) from Cerchiara and Harrisville. Samples from Yellow Lake and Cerchiara are nearly Ba-free and Sr-free, respectively; the remaining samples are intermediate between these two end-members. Brewsterite is monoclinic $P2_1/m$ (Perrotta and Smith 1964, Schlenker et al. 1977, Artioli et al. 1985, Cabella et al. 1993). Unit-cell parameters are: a = 6.77-6.82 Å, b = 17.45-17.60 Å, c = 7.73-7.75 Å, β = 94.27-94.54° .$c > a$ was assessed by Weissenberg X-ray analysis and elongation a was determined by an optical study (Nawaz 1990).

Structure Type: **CHA**

Chabazite : $(Ca_{0.5},Na,K)_4[Al_4Si_8O_{24}]\cdot 12H_2O$

Willhendersonite: $Ca_2(Ca_{0.5},K)_2[Al_6Si_6O_{24}]\cdot 10H_2O$

The TS $R\bar{3}m$ of the tetrahedral framework of chabazite and willhendersonite is lowered to triclinic $P\bar{1}$ as a result of ordering of the (Si,Al) distribution and of the position of the cations and water molecules. Ordering in the tetrahedral sites can be either in domains or complete, depending on the Si/Al ratio of the crystal. Triclinic domains in chabazite (Si/Al > 1) are oriented in such a way that the average structure of the mineral is pseudo-trigonal $R\bar{3}m$ (Smith et al. 1964, Mazzi and Galli 1983). The real triclinic symmetry $P\bar{1}$ in willhendersonite results from a Si/Al ratio of one, with resultant ordering (Tillmanns et al. 1984).

Chabazite is one of the most widespread natural zeolites and has been reported from a variety of occurrences: in vugs of plutonic rocks (granite, syenite, granodiorite, gabbro, and pegmatite), volcanic rocks (tholeiitic and alkalic basalts, including leucitite, and andesite), and metamorphic rocks (gneiss, serpentinite, metabasite, and cornubianite), where it occurs in association with many other zeolites (natrolite, mesolite, thomsonite, phillipsite, stilbite, gismondine, analcime, heulandite-clinoptilolite, mordenite, ferrierite, gonnardite, levyne, gmelinite, laumontite, offretite, and paulingite), apophyllite, quartz, fluorite, barite, carbonates (calcite, aragonite, and siderite), searlesite, colemanite, and pyrite.

Sedimentary chabazite occurs both in palagonitic basalt associated with phillipsite, gonnardite, gmelinite, analcime, thaumasite, apophyllite, and gyrolite and in pyroclastic rocks (phonolitic and trachytic ignimbrites, and rhyolitic and nephelinitic tuff) diagenetically altered in continental environments ("open," "closed," and "geoautoclave" systems) where it coexists with phillipsite, analcime, clinoptilolite, erionite, mordenite, harmotome, dawsonite, and clay minerals.

Chabazite generally exhibits pseudocubic rhombohedral morphology, but lens-shaped aggregates more ("phacolitic" habit) or less ("herschelitic" habit) rounded due to complex twinning are common (Akizuki and Konno 1987, Akizuki et al. 1989). Epitaxial intergrowths with gmelinite are common (Passaglia et al. 1978a, Wise and Kleck 1988), and samples epitaxially overgrown by offretite have also been observed (Passaglia and Tagliavini 1994, Passaglia et al. 1996).

Reliable chemical compositions, including 78 amygdaloidal samples, 5 samples from palagonitic basalt, and 28 sedimentary samples from "open" (22 samples) and "closed" (6 samples) systems, revealed that the schematic formula given above is only an average. R ranges from 0.58 in amygdaloidal chabazite from Kaiserstuhl, Germany (Livingstone 1986), to 0.81 in sedimentary chabazite from the John Day Formation near Monument, Oregon (Sheppard and Gude 1970). Ca, Na, and K are, in order of frequency, the DEC; Sr is generally present in low amounts, but it is remarkable (0.50-0.80) in several amygdaloidal samples (Passaglia 1970, Livingstone 1986, Robert 1988, Birch 1989, Passaglia and Tagliavini 1994). Where present, Ba contents are very low and reach their highest values (ca. 0.13) in some amygdaloidal samples (Passaglia 1970, Argenti et al. 1986). Mg is consistently negligible, but considerable contents (0.5-0.8) have been recognized in the amygdaloidal samples from Narre Warren, Australia (Birch 1989), from Adamello, Italy (Passaglia and Tagliavini 1994), and in a sedimentary sample from the Big Sandy Formation, Arizona (Sheppard and Gude 1973).

In amygdaloidal crystals, R shows a mean value of 0.67 and ranges from 0.58 in the sample from Kaiserstuhl (Livingstone 1986) to 0.75 in the sample from Narre Warren, Australia (Birch 1989). Ca commonly is the DEC, but Na dominates in samples displaying "herschelitic" and "phacolitic" habits (Passaglia 1970, Birch 1988, 1989; Wise and Kleck 1988, Akizuki et al. 1989, Vezzalini et al. 1994). K is dominant in chabazite from Vallerano, Italy (Passaglia 1970), and from Ercolano, Italy (de'Gennaro and Franco 1976). Sr-rich (0.50-0.80) samples are not rare, Ba is negligible, and Mg reaches the highest value (0.79) in the first occurrence of chabazite ("herschelite" habit) epitaxially overgrown by offretite (Passaglia and Tagliavini 1994).

In sedimentary samples from "open systems" and "geoautoclaves," the mean value of R is 0.71, ranging from 0.63 to 0.81. The lower values (0.63-0.67) have been found in crystals from phonolitic, leucititic, and melilitic ignimbrites; intermediate values (0.68-0.74) were determined for crystals from trachytic ignimbrites (de'Gennaro et al. 1980; Passaglia and Vezzalini 1985, Passaglia et al. 1990, Adabbo et al. 1994); and the highest value (0.81) was found in chabazite from a rhyolitic tuff (Sheppard and Gude 1970). K and Ca are, in order of frequency, the DEC, being commonly in similar amounts; Na, Mg, and Sr are consistently subordinate. In sedimentary samples from "closed systems," the mean value of R is 0.77, and it is close to 0.78 in crystals from rhyolitic tuff (Gude and Sheppard 1966, 1978; Sheppard and Gude 1973, Sheppard et al. 1978). It is definitely lower (0.72) in crystals from a nephelinite tuff (Hay 1964). Na is dominant in most "closed-system" samples.

Chabazite formed from the interaction of mafic glass with sea water in palagonitic basalt (Noack 1983, Passaglia et al. 1990) shows R ranging from 0.65 to 0.69 and Na prevailing over K and Ca. The sample from Palagonia (Italy) is an exception, with R of

0.62, Ca dominating, and a remarkable Sr content (0.47) (Passaglia et al. 1990). A sample occurring in "marine volcanics" from the Waite mata Group (New Zealand) has an R value of 0.71 and Na as the DEC (Sameshima 1978).

The unit-cell parameters (pseudo-hexagonal cell) are: a = 13.69-13.86 Å, c = 14.80-15.42 Å. A negative correlation between R and the c parameter was found in natural samples (Passaglia 1970). The unit-cell parameters of cation-exchanged samples showed significant variations primarily in c, dependent on R and on the occupancy of the extraframework sites (Passaglia 1978a, Alberti et al. 1982a).

Willhendersonite has been described as "trellis-like" twinned aggregates from a mafic potassic lava at S. Venanzo, Terni, Italy, in a limestone xenolith from a basalt at Mayen, Eifel, Germany (Peacor et al. 1984), and as vitreous rectangular laths from the breccia ridge of a melilitite plug at Colle Fabbri, Terni, Italy, (Vezzalini et al. 1997). It is associated with phillipsite, thomsonite, apophyllite, chabazite, gismondine, tobermorite, ettringite, thaumasite, calcite, aragonite, and vaterite. For all samples, R is near 0.50, and Ca is the DEC. The K content is very high in the samples from S. Venanzo and Mayen (1.80 and 1.48, respectively) and very low in the sample from Colle Fabbri (0.41). The structure is triclinic $P\bar{1}$ with a = 9.18-9.21 Å, b = 9.20-9.22 Å, c = 9.44-9.50 Å, α = 91.72-92.34°, β = 91.42-92.70°, γ = 90.05-90.12° (Tillmanns et al. 1984, Vezzalini et al. 1997).

Several other aspects of the crystal chemistry of chabazite and willhendersonite are noteworthy. For example, among the natural structure types, the tetrahedral framework CHA shows the highest variation in Si/Al ratio (R = 0.50-0.80), and there is only a narrow gap in R between willhendersonite (0.50) and the lowest value found in chabazite (0.58). Interestingly, crystals with R = 0.50 are entirely triclinic (Tillmanns et al. 1984), whereas crystals with R ranging from 0.66 to 0.72 showed submicroscopic triclinic domains (Mazzi and Galli 1983). These observations lead one to wonder about the degree of triclinicity of chabazite crystals with R lower than 0.66 and of hypothetical crystals with R lower than 0.58.

Structure Type: **DAC**

Dachiardite: $(Ca_{0.5},Na,K)_4[Al_4Si_{20}O_{48}]\cdot 18H_2O$

Dachiardite has been described from a dozen localities in fractures of an aplite pegmatite, in vugs of massive volcanic rocks (basalt, andesite, and porphyrite), and of a hydrothermally altered rhyolite. It is generally associated with mordenite, commonly with heulandite-clinoptilolite, and rarely with epistilbite, stilbite, ferrierite, erionite, phillipsite, yugawaralite, analcime, silica minerals (quartz, chalcedony), and carbonates (calcite, siderite, and ankerite). Most samples occur as simple fibers along b; but simple twinnings are not rare, and the holotype "octagonal beakers" from Elba Island, Italy, are polysynthetic twins of eight individuals (Berman 1925). Svetlozarite, a proposed new zeolite (Maleev 1976), has been interpreted as a multiply twinned and highly faulted dachiardite and was, hence, rejected (Gellens et al. 1982).

R has a mean value of 0.81 in dachiardite but ranges from 0.77 in crystals from Yellowstone National Park, Wyoming (Bargar et al. 1987), to 0.86 in crystals from Altoona, Washington (Wise and Tschernich 1978b). Ca and Na are the common DEC; the K content is highly variable ranging from 0.01 to 1.02 in crystals from Yellowstone National Park where it slightly prevails over Na and Ca (Bargar et al. 1987); Mg, Sr, and Ba are absent or negligible; Cs occurs (0.12) in crystals from Elba Island, Italy (Bonardi 1979, Vezzalini 1984). The Na/Ca ratio uniformly ranges within two extremes. Na-dominant (Na > Ca) crystals are from Alpe di Siusi, Italy (Alberti 1975a), Ogasawara Island, Japan (Nishido et al. 1979), Altoona, Washington, Agate Beach, Oregon (Wise and Tschernich 1978b), Montreal Island, Canada (Bonardi et al. 1981), and Yellowstone

National Park, Wyoming (Bargar et al. 1987). Ca-dominant (Na < Ca) crystals are from Elba Island, Italy (Gottardi 1960, Bonardi 1979, Vezzalini 1984), Cape Lookout, Oregon (Wise and Tschernich 1978b), Kagoshima and Ogasawara Island, Japan (Nishido and Otsuka 1981), Yellowstone National Park, Wyoming (Bargar et al. 1987), and in the rejected "svetlozarite" from Rhodopes, Bulgaria (Maleev 1976). Different samples from the same occurrence may exhibit very different extraframework-cation contents. For example, crystals from Ogasawara Island (Japan) may be either Na-dominant or Ca- dominant, and crystals from Yellowstone National Park (Wyoming) are generally Ca-dominant but are rarely Na-dominant or K-dominant.

The structure is monoclinic $C2/m$ (Vezzalini 1984) with a = 18.62-18.73 Å, b = 7.49-7.54 Å, c = 10.24-10.31 Å, and β = 107.87-108.74° . No correlations between Na/Ca and Si/Al ratios were found, but β increases with Si/Al ratio, and refractive indices, $2V_x$, and $c \wedge Z$; all increase with Ca (Nishido and Otsuka 1981).

Structure Type: **EAB**

Bellbergite: $Ca_5(Sr,Ba)_3(Na,K)_2[Al_{18}Si_{18}O_{72}]\cdot 30H_2O$

Bellbergite has been described only from Bellberg, Eifel (Germany), in cavities of Ca-rich xenoliths in a leucite-tephrite lava (Rüdinger et al. 1993). It occurs as dipyramids in association with thomsonite, ettringite, pyrrhotite, sanidine, and clinopyroxene. The empirical formula shows an R value of 0.50, Ca as the DEC, high amounts of K and Sr, and subordinate Na and Ba. The structure is hexagonal ($P6_3/mmc$, $P\bar{6}2c$, or $P6_3mc$) with a = 13.24 Å and c = 15.99 Å. Optically, it is uniaxial negative.

Structure Type: **EDI**

Edingtonite: $Ba_2[Al_4Si_6O_{20}]\cdot 8H_2O$

Edingtonite has been described from five occurrences in hydrothermal veins in altered mafic rocks and metasediments, associated with sulfides (pyrite, chalcopyrite, galena, and sphalerite), carbonates (calcite and barytocalcite), hematite, hydroxides (goethite and manganite) and, in one occurrence, with analcime and harmotome (Novak 1970, Mazzi et al. 1984, Grice et al. 1984, Belitsky et al. 1986).

R is nearly constant (0.59-0.61), and Ba is the only DEC [Ba/(Ba+Ca+Mg+Na+K) = 0.91-0.99]. K is negligible (<0.10); Na, Ca, and Mg are absent or present in trace amounts; Sr is absent.

Structure refinements of crystals from different localities systematically show two different symmetries. Crystals from the type locality of Old Kilpatrick, Scotland, and from Ice River, Canada, have a disordered (Si, Al) distribution and are tetragonal $P\bar{4}2_1m$ with a = 9.58 Å, c = 6.52 Å (Mazzi et al. 1984). Crystals from Bohlet, Sweden, and Brunswick, Canada, have a nearly ordered (Si,Al) distribution and are orthorhombic $P2_12_12$ with a = 9.54 Å, b = 9.66 Å, c = 6.52 Å (Galli 1976, Kvick and Smith 1983). The (Si,Al) ordering and symmetry (tetragonal or orthorhombic) seem to vary from crystal to crystal from the same occurrence; different crystals from Ice River, Canada, are orthorhombic (Grice et al. 1984) or tetragonal (Mazzi et al. 1984).

Structure Type: **EPI**

Epistilbite: $Ca_3[Al_6Si_{18}O_{48}]\cdot 16H_2O$

Epistilbite has been described in vugs of massive rocks along with many other zeolites (mordenite, heulandite, dachiardite, stilbite, scolecite, levyne, laumontite, and chabazite), gyrolite, pumpellyite, quartz, pyrite, and sphalerite. The characterized occurrences (about 20) are mainly from basalt in Iceland and Japan but are also from an aplite pegmatite at

Elba Island (Italy), gneiss at Gibelsbach, Switzerland, and dolerite at Mt. Adamson, Antarctica (Galli and Rinaldi 1974, Mehegan et al. 1982, Akizuki and Nishido 1988, Nishido 1994, Vezzalini et al. 1994).

The chemical composition shows only a slight variability. R has a mean value of 0.75, ranging from 0.73 to 0.77; Ca is the DEC; Na is present in subordinate amounts, reaching a maximum value (~1.0) in a few samples from Iceland. K is occasional or negligible (≤0.30), and Mg, Sr, and Ba are absent.

The structure is monoclinic *C*2 (Alberti et al. 1985) with a = 9.08-9.10 Å, b = 17.74-17.80 Å, c = 10.20-10.24 Å, and β = 124.55-124.68° . A crystal from Gibelsbach shows triclinic *C*1 symmetry (a = 9.08Å, b = 17.74Å, c = 10.21Å, α = 89.95° ,β = 124.58° , andγ = 90.0°) due to partial (Si,Al) ordering accompanied by a preferred distribution of the extraframework cations (Yang and Armbruster 1996). Negative correlations between Ca and β and between R and Ca were reported (Galli and Rinaldi 1974). Data on twinnings and optical properties of crystals from Iceland and Japan are reported in Akizuki and Nishido (1988).

Structure Type: **ERI**

Erionite: $K_2(Ca_{0.5},Na)_7[Al_9Si_{27}O_{72}]\cdot 28H_2O$

Erionite from vugs in volcanic rocks (basalt, andesite, limburgite, dolerite, and latite-trachyte) and diagenetically altered vitroclastic rocks (sedimentary) have been investigated. Amygdaloidal crystals are associated with many other zeolites (chabazite, mordenite, stilbite, heulandite, phillipsite, analcime, paulingite, and harmotome), epitaxially intergrown with offretite (Rinaldi 1976, Wise and Tschernich 1976a), or overgrown on levyne (Passaglia et al. 1974). Sedimentary erionite coexists with analcime, chabazite, clinoptilolite, mordenite, and phillipsite, and it has formed in both continental (Staples and Gard 1959, Sheppard et al. 1965, Sheppard and Gude 1969b, Surdam and Eugster 1976, Boles and Surdam 1979, Gude and Sheppard 1981) and marine (Sameshima 1978) environments. In comparison with offretite, erionite is more siliceous, more alkalic, and optically positive (Sheppard and Gude 1969b, Wise and Tschernich 1976a, Rinaldi 1976). R shows a mean value of 0.75 and ranges from 0.68 to 0.79; K, Na, and Ca are the DEC; Mg is generally subordinate but highly variable (0.10-2.0); and Sr and Ba are negligible.

Amygdaloidal specimens show a mean R value of 0.75 which ranges from 0.70 in a sample from Yeongil area, Korea (Noh and Kim 1986), to 0.77 in a sample from Campbell Glacier, Antarctica (Vezzalini et al. 1994). Ca and K are generally the DEC, but Na may also be remarkably high (1.70); Mg is dominant (2.13) in a sample from Sasbach, Germany (Rinaldi 1976). Sedimentary specimens show a mean R value of 0.78 with small variations (0.76-0.79), but a sample from marine sediments (Sameshima 1978) is anomalously Si-poor (R = 0.74); Na and K are the DEC; and Ca and Mg are highly variable.

A general study on the crystal chemistry of erionite (Passaglia et al. 1998) reached the following conclusions: (1) R ranges from 0.68 to 0.79; (2) Ca is the most common DEC, but crystals with Na as the DEC are not rare; K is always present in considerable amounts (1.6-3.4) but is predominant only in a sample from Ortenberg, Germany; Mg is subordinate, reaching a maximum value (0.82) in a sample from Agate Beach (Oregon); (3) crystals epitaxially overgrown on levyne are quite frequent and are substantially poorer in Si and Mg than those associated with other zeolites; (4) the optic sign depends on the Si/Al ratio of the sample (positive in the Si-rich crystals, negative in the Si-poor crystals); and (5) epitaxial intergrowths with offretite are rare. The structure is hexagonal *P*6$_3$/*mmc* with a = 13.19-13.34 Å and c = 15.04-15.22 Å. The unit-cell volume is negatively cor-related with the Si/Al ratio (Sheppard and Gude 1969b, Passaglia et al. 1998).

Structure Type: **FAU**

Faujasite: $(Na,K,Ca_{0.5},Mg_{0.5})_{56}[Al_{56}Si_{136}O_{384}]\cdot 235H_2O$

Faujasite has been described from only a limited number of occurrences, including four in vugs in limburgite from Germany (Rinaldi et al. 1975, Wise 1982), three in vesicles of palagonitic mafic tuffs from San Bernardino County, California (Wise 1982), and an occurrence in basaltic tephra at Jabal Hannoun and Jabal Aritayn, Jordan (Ibrahim and Hall 1995). Faujasite commonly occurs with phillipsite but also occurs with offretite in Germany and chabazite in Jordan.

R has a mean value of 0.71 and generally ranges from 0.70 to 0.73; only a sample from Hasselborn, Germany (Wise 1982) is less siliceous (R = 0.68). Ca (average content: 14.2 atoms) and Na (average content: 13.7 atoms) are the DEC, and Mg (average content: 5 atoms) and K (average content: 3.8 atoms) are generally subordinate. All of these cations are quite variable even in different crystals from the same occurrence. For example, Ca ranges from 3.93 to 21.12 and is dominant in a sample from Germany analyzed by Wise (1982) and in three samples from Jordan (Ibrahim and Hall 1995); Na ranges from 7.12 to 22.00 and is dominant in two samples from Sasbach, Germany (Rinaldi et al. 1975), in the sample from San Bernardino County, California (Wise 1982), and in five samples from Jordan. Mg ranges from 1.53 to 11.12, and K ranges from 0.54 to 10.66. Sr is very low (0.32) and has been detected only in a couple of samples from Germany (Wise 1982), and Ba is absent. For both tetrahedral and extraframework contents, no differences are obvious between sedimentary and amygdaloidal specimens, although in the former, Na dominates more commonly. Two occurrences of amygdaloidal Na-rich crystals from basalts of Aci Castello and Aci Trezza (Italy) are reported in Gottardi and Galli (1985, R. Rinaldi's communication) and in Wise (1982), respectively. Unfortunately, no paper describing these occurrences has been published. The structure is cubic $Fd3m$ with a = 24.60-24.78 Å.

Structure Type: **FER**

Ferrierite: $(Mg_{0.5},Na,K)_6[Al_6Si_{30}O_{72}]\cdot 20H_2O$

Ferrierite has been described mainly in vugs of volcanic rocks (basalt, andesite, porphyrite, and latite) coexisting with other zeolites (heulandite, dachiardite, mordenite, analcime, chabazite, and harmotome), carbonates (calcite, aragonite, and siderite), barite, apatite, pyrite, cinnabar, and chalcedony (Alietti et al. 1967, Wise et al. 1969, Yajima and Nakamura 1971, Wise and Tschernich 1976b, Birch and Morvell 1978, England and Ostwald 1978, Passaglia 1978b, Orlandi and Sabelli 1983, Sameshima 1986, Birch 1989). It has also been found in sedimentary rocks coexisting with mordenite, clinoptilolite, and quartz (Regis 1970, Noh and Kim 1986). Amygdaloidal samples are generally associated with Fe-hydroxides and calcite.

R shows a mean value of 0.83 and ranges from 0.78 in crystals from Unanderra, New South Wales, Australia (England and Ostwald 1978), to 0.88 in crystals from Agoura, California (Wise et al. 1969). Mg, Na, and K are, in order of frequency, the DEC. Ca is consistently subordinate, reaching a maximum value (~1.0) in a sample from Albero Bassi, Italy (Alietti et al. 1967); Sr and Ba are commonly absent and reach, respectively, maximum values of 0.23 in a sample from Unanderra, New South Wales, and of 0.45 in a sample from Silver Mountain, California (Wise and Tschernich 1976b). Compared with amygdaloidal samples, sedimentary ferrierite samples show a smaller range of R (0.81-0.84), have K as the DEC, and have low Ca and Mg contents.

Structure refinements of Mg-dominant samples from Silver Mountain, California (Gramlich-Meier et al. 1984), and Monastir, Italy (Alberti and Sabelli 1987), and of a Na-dominant, Mg-poor sample from Altoona, Washington (Gramlich-Meier et al. 1985), reveal that where Mg is dominant, the zeolite is orthorhombic *Immm* or *Pnnm*. Where Na is dominant, ferrierite is monoclinic $P2_1/n$. The orthorhombic unit-cell parameters are a = 18.90-19.45 Å, b = 14.12-14.28 Å, and c = 7.48-7.54 Å. The a parameter is negatively correlated with the Si content and positively correlated with the Mg content, which, in turn, is positively correlated with the Al content (Wise and Tschernich 1976b, Gramlich-Meier et al. 1985, Sameshima 1986). A decrease in R is correlated with an increase in Mg and a lengthened a parameter.

Structure Type: **GIS**

Gismondine: $Ca_4[Al_8Si_8O_{32}]\cdot 16H_2O$
Garronite: $Ca_{2.5}Na[Al_6Si_{10}O_{32}]\cdot 13H_2O$
Gobbinsite: $Na_4Ca[Al_6Si_{10}O_{32}]\cdot 11H_2O$
Amicite: $Na_4K_4[Al_8Si_8O_{32}]\cdot 10H_2O$

These four zeolite species share the same tetrahedral framework with TS $I4_1/amd$ which is lowered to the RS of the various species as a consequence of: (a) slight distortion of the framework; (b) tetrahedral Si-Al ordering; and (c) the distribution and coordination of the Ca, Na, and K cations and water molecules in the extraframework sites (Alberti and Vezzalini 1979, McCusker et al. 1985, Rinaldi and Vezzalini 1985, Artioli 1992, Artioli and Marchi 1999). These four zeolites all occur in vugs of volcanic rocks in association with phillipsite, in some cases as intimate intergrowths (Walker 1962a,b; Cortesogno et al. 1975, Vezzalini and Oberti 1984, Pöllmann and Keck 1990, Artioli and Foy 1994, Howard 1994). The DEC in all four species are Ca, Na, and K, with Mg, Sr, and Ba in very subordinate amounts or even absent. On the basis of the tetrahedral framework content, gismondine is related to amicite, both having R ≈ 0.50 and, hence, an ordered (Si,Al) distribution. Garronite is related to gobbinsite, both having R ≈ 0.63 and a disordered (Si,Al) distribution. On the basis of extraframework cation content, gismondine is related to garronite, both having Ca as the DEC; and amicite is related to gobbinsite, both having Na as the DEC.

Gismondine (the most common of the group) occurs in vugs of mafic volcanic rocks (basalt, leucitite, nephelinite, and tephrite), but also in fractures at the contact between basalt and metamorphosed serpentinite (Cortesogno et al. 1975, Argenti et al. 1986) and in a palagonitic nephelinite tuff (Iijima and Harada 1968). It occurs in association with thomsonite, chabazite, calcite, aragonite, apophyllite, gyrolite, but mainly with phillipsite, commonly in intimate intergrowths. Morphology, twinning, and optical orientation are reported by Nawaz (1980). R shows a mean value of 0.53 with a small range (0.50-0.54). Only one anomalously high value of R (0.58) has been found in crystals from Hoffil, Iceland (Walker 1962b). Ca is the DEC; Na and K are present in very subordinate amounts, commonly with K > Na in samples from leucitite and Na > K in samples from basalt and nephelinite. The highest Mg content (0.38) found in a sample from Bavaria, Germany, is possibly due to smectite impurities (Pöllmann and Keck 1993). Sr, as traces in samples from leucitite, reaches its maximum value (0.20) in a sample from Lubam, Poland (Vezzalini and Oberti 1984). The sample from Montalto di Castro, Italy, in association with Ca-silicates (vertumnite and tobermorite), has a composition which is very close the schematic formula (R = 0.51, only Ca as the extraframework cation) (Vezzalini and Oberti 1984), whereas the sample from Hofill, Iceland (Walker 1962b) has an unusually high R value (0.58) and Na content (0.90). The structure of gismondine is monoclinic $P2_1/c$ with unit cell dimensions nearly constant: a = 10.01-10.04 Å, b = 10.60-10.64 Å, c = 9.82-9.84 Å, and β = 92.23-92.56° .

The positive correlation (Vezzalini and Oberti 1984) in gismondine between (Na+K)/(Na+K+Ca) and Si/(Si+Al) (i.e. the monovalent cations, mainly Na, increase as the

Si content increases) may yield the average chemical composition of *garronite*, where the increase in Si (R ≈ 0.63) is accompanied by a higher Na (and K) and a lower Ca content (~1 and 2.5, respectively). *Garronite* has an R value consistently higher than 0.60 (0.60-0.65), but its Ca content varies from 2.33 in a sample from Island Magee, County Antrim, Ireland, to 2.88 in a sample from Tardree Forest, County Antrim (Nawaz 1982), and its Na(+K) content varies from 0.10-0.30 in samples from Fara Vicentina, Italy (Passaglia et al. 1992), Goble, Oregon (Artioli 1992), and Table Mountain, Colorado (Kile and Modreski 1988), to 1.10-1.40 in samples from Ireland, Iceland (Walker 1962a, Nawaz 1982), and Halap Hill, Hungary (Alberti et al. 1982b). Therefore, a compositional gap between gismondine and garronite seems to exist in the tetrahedral framework content, but not in the extraframework cations.

Garronite occurs only in vugs of basalt in association with phillipsite, gonnardite, chabazite, thomsonite, analcime, and levyne. The morphology and twinning of the sample from Fara Vicentina, Italy, are reported in Howard (1994). The structure is monoclinic $I2/a$ (pseudotetragonal $I\bar{4}m2$) with a = 9.88 Å, b = 10.28 Å, c = 9.876 Å, and β = 90.11° (Artioli and Marchi 1999).

Gobbinsite occurs in vugs of basalt at Gobbins and Island Magee, Ireland, associated with gmelinite, chabazite, gonnardite, garronite, and phillipsite (Nawaz 1982, Nawaz and Malone 1982) and at Iki Island, Japan, associated with cowlesite (Kuwano and Tokumaru 1993). In another occurrence at Magheramorne Quarry, Ireland, it is intimately associated with phillipsite (Artioli and Foy 1994). R is approximately constant (0.64); Na is consistently dominant and ranges from 3.61 in a sample from Island Magee (Nawaz 1982) to 5.63 in a sample from Iki Island, Japan. Consequently, Ca ranges from 0.93 to 0.17; Mg is appreciable (0.31) only in the sample from Gobbins (Nawaz and Malone 1982); and K is generally very low, although it is very high (2.11) in a sample from Island Magee (Mc Cusker et al. 1985) where both R (0.62) and Na (2.50) are anomalously low. The structure is orthorhombic $Pmn2_1$ (Mc Cusker et al. 1985) with a ≈ 10.10 Å, b = 9.77-9.80 Å, and c = 10.17 Å

Amicite occurs in vugs of Na-rich rocks such as nephelinite in association with merlinoite, calcite, and aragonite at Hegau, Germany (Alberti et al. 1979), and ijolite-urtite pegmatite and apatite-bearing nephelinite associated with natrolite at Kola Peninsula, Russia (Khomyakov et al. 1984). The chemistry of specimens from the two occurrences is almost the same: R is about 0.50, Na and K are comparable (~4), and Ca is very low. The structure is monoclinic $I2$ (Alberti and Vezzalini 1979) with a = 10.23 Å, b = 10.42 Å, c = 9.88 Å, and β = 88.32° .

Structure Type: **GME**

Gmelinite: $(Na,K,Ca_{0.5})_8[Al_8Si_{16}O_{48}]\cdot 22H_2O$

Gmelinite typically occurs in vugs of basalt mainly associated with analcime and natrolite but also with chabazite, phillipsite, heulandite, and stilbite. R shows a mean value of 0.69 and ranges from 0.65 in a sample from White Head, County Antrim, Ireland (Passaglia et al. 1978a), to 0.72 in a sample from Boron, California (Wise and Kleck 1988). Na is consistently abundant and is commonly the DEC. Ca is highly variable (0-2.5) and is dominant in the samples from Montecchio Maggiore, Italy, and from Great Notch, New Jersey (Passaglia et al. 1978a). K is generally the SEC, but it is the DEC in the samples from Fara Vicentina, Italy (Vezzalini et al. 1990), and from an alkalic rock at Kola Peninsula, Russia (Malinovskii 1984). Mg and Ba are absent or negligible. Where determined, the Sr content is appreciable (0.2-0.5) and reaches 1.35 in the Ca-rich, Na-poor sample from Montecchio Maggiore, Italy (Passaglia et al. 1978a). Large dipyramidal crystals show compositional zoning and are locally intergrown with subordinate chabazite

(Passaglia et al. 1978a). Na-rich chabazite crystals from Hayata, Japan, have a "herschelite" habit and a gmelinite rim whose (0001) surface consists of a spiral growth pattern (Akizuki et al. 1989).

The structure is hexagonal $P6_3/mmc$ with a = 13.62-13.80 Å and c = 9.97-10.25 Å, the smaller a values and the larger c values occurring in the K-dominant samples (Malinovskii 1984, Vezzalini et al. 1990). The unit-cell parameters are correlated with the extraframework contents, with a increasing and c decreasing with increasing divalent cations (Passaglia et al. 1978a). Structure refinements of Na-, Ca-, and K-rich natural crystals (Galli et al. 1982, Vezzalini et al. 1990) show that the tetrahedral framework undergoes a deformation involving a lengthening of c and a shortening of a which follows the bond strengths of the extraframework cations (Ca > Na > K) in the large channels (C2 site). Structure refinements of Ca-, Na-, and K-exchanged crystals (Sacerdoti et al. 1995) show a marked lengthening of c and a minor shortening of a in the K-Na-Ca series ($c_K > c_{Na} > c_{Ca}$), correlated with both occupancy factor (100% in the K and Na forms and ca. 50% in the Ca form) and ionic radius of the cation in the gmelinite cage (C1 site).

Structure Type: **GOO**

Goosecreekite: $Ca_2[Al_4Si_{12}O_{32}]\cdot 10H_2O$

The only fully characterized goosecreekite sample is from the type locality at Goose Creek Quarry, Loudoun County, Virginia, where it occurs as euhedral, but highly curved, colorless crystals in vugs of an altered diabase in association with stilbite, quartz, chlorite, actinolite, albite, prehnite, apophyllite, babingtonite, and epidote (Dunn et al. 1980). The empirical chemical formula is very close to the schematic one and shows an R value of 0.75. Ca is the only extraframework cation. The structure is monoclinic $P2_1$ with a = 7.40 Å, b = 17.44 Å, c = 7.29 Å, and β = 105.44° from single-crystal measurements (Rouse and Peacor 1986) and with a = 7.52 Å, b = 17.56 Å, c = 7.35 Å, and β = 105.71° from powder X-ray diffraction data (Dunn et al. 1980).

Structure Type: **HEU**

Heulandite-Clinoptilolite: $(Na,K,Ca_{0.5})_7[Al_7Si_{29}O_{72}]\cdot 22H_2O$

Based on crystallochemical data (X-ray single-crystal photographs, chemical composition, and optical properties), Hey and Bannister (1934) demonstrated that clinoptilolite and heulandite are members of an isomorphous series and, hence, suggested that the "name clinoptilolite is unsuitable and should not be used." Numerous structural studies have revealed that heulandite and clinoptilolite share the same tetrahedral framework and monoclinic $C2/m$ symmetry (Alberti 1972, 1975b; Koyama and Takeuchi 1977). In the past, many efforts were spent to identify chemical and/or physical parameters to justify heulandite and clinoptilolite as two distinct zeolitic species. Clinoptilolite was proposed as a zeolite of the heulandite group with (Na+K) > Ca by Mason and Sand (1960) and with Si/Al > 4 by Boles (1972). A zeolite of the heulandite group with a structure which survives an overnight heating at 450° C was proposed tobe named clinoptilolite by Mumpton (1960). The first proposed definition, the only one consistent with mineralogical nomenclature practice, was invalidated by the description of many samples with Ca > (Na+K) and Si/Al > 4 (Kirov 1965, Boles 1972, Sheppard and Gude 1973, Alberti 1975b, Wise and Tschernich 1976b, Alietti et al. 1977, Minato and Aoki 1978, Barrows 1980, Noh and Kim 1986, Altaner and Grim 1990, Tsolis-Katagas and Katagas 1990, Rice et al. 1992, Münch and Cochemé 1993). Studies of several heulandite-clinoptilolite samples revealed three distinct thermal behaviors that were correlated with the Si/Al and (Na+K)/(Ca+Mg+Sr+Ba) ratios (Alietti 1972, Boles 1972, Alietti et al. 1977). Such results, along with those obtained on cation-exchanged samples (Shepard and Starkey 1964, 1966; Alietti et al. 1974), clearly demonstrated that the thermal stability of a sample increased mainly with an increase in the (Na+K)/(Ca+Mg+Sr+Ba) ratio and, to a lesser extent, an increase of the Si/Al ratio. Although a statistical analysis showed the existence of at least five contiguous subgroups that can be distinguished on the basis of Ca (Hawkins 1974), a wealth of chemical data for heulandite-clinoptilolite samples corroborate the pioneering statement by Hey and Bannister (1934) because a consideration of both tetrahedral framework and extraframework cations shows a continuous solid solution along the join between the schematic formulae given in Gottardi and Galli (1985) for heulandite $(Na,K)Ca_4[Al_9Si_{27}O_{72}]$ and for clinoptilolite $(Na,K)_6[Al_6Si_{30}O_{72}]$. Many samples from the literature have been described as heulandite or clinoptilolite on the basis of chemical parameters and/or thermal behavior. Indeed, several definitions are based only on the occurrence, the name heulandite generally being adopted for samples in vugs of igneous rocks and clinoptilolite being used for samples in diagenetically altered vitroclastic sediments. In this review, heulandite and clinoptilolite will be described as a single species (referred to as heulandite-clinoptilolite), although the recommended nomenclature approved by the International Mineralogical Association (Coombs et al. 1997) distinguishes two species on the basis of the framework Si/Al ratio: heulandite has Si/Al < 4 and clinoptilolite has Si/Al > 4. A thorough discussion of clinoptilolite-heulandite nomenclature and chemistry is presented in the chapter by Bish and Boak (this volume).

Heulandite-clinoptilolite is a very common zeolite that has been described from a variety of environments. The zeolite occurs in vugs of massive plutonic (granite, pegmatite, and gabbro), volcanic (basalt, andesite, porphyrite, tephrite, latite, and dolerite) and metamorphic (gneiss) rocks where it is often associated with other zeolites (mordenite, dachiardite, ferrierite, chabazite, stilbite, epistilbite, laumontite, mesolite, scolecite, phillipsite, erionite-offretite, analcime, levyne, gmelinite, thomsonite, and natrolite), calcite, rhodochrosite, prehnite, apophyllite, datolite, quartz, chalcedony, opal, pyrite, chalcopyrite, searlesite, and borax. It is a common alteration product of vitroclastic sediments that have been diagenetically altered in subaerial ("open" and "closed" systems) and marine (deep-sea) environments in association with analcime, mordenite, ferrierite, erionite, phillipsite, chabazite, opal, cristobalite, clay minerals, and carbonates. Heulandite-clinoptilolite also occurs in the "zeolite facies" of low-grade metamorphic rocks.

R ranges from 0.73 in amygdaloidal crystals from basalt of Verandowana, India (Sukheswala et al. 1974), to 0.85 in a sedimentary sample from rhyolitic tuff of Cañadon Hondo, Argentina (Mason and Sand 1960). The monovalent (M = Na+K) and divalent (D = Ca+Mg+Sr+Ba) cations show all possible ratios: M/(M+D) ranges from 0.07 in sedimentary crystals from silicic volcanic sandstone of the Báucarit Formation, Mexico (Münch and Cochemé 1993) to 1.0 in crystals from a burial diagenetic clay-rich horizon of the chalk soil of Sangstrup Klint, Denmark (Nørnberg 1990). The Na/(Na+K) ratio ranges from 0 (Na-free) in the amygdaloidal crystals from Malpais Hill, Arizona (Wise and Tschernich 1976a), to 0.94 (almost K-free) in crystals from Yellowstone National Park, Wyoming (Bargar and Beeson 1981). Ca is generally the most common divalent cation. The Mg content is negligible or low depending on the genetic environment and reaches a high value of 1.46 in the sample from Maldon, Australia (Birch 1989). Sr is generally 0.40-0.50 and is especially high (2.10 and 1.24, respectively) only in amygdaloidal samples from Campegli, Italy (Lucchetti et al. 1982) and Kozakov Hill, Czechoslovakia (Cerny and Povondra 1969). Ba is commonly absent or negligible, but it reaches a value of 1.03 in a sedimentary sample from non-marine sandstone of the Blairmore Group, Canada (Miller and Ghent 1973).

Reliable chemical compositions of a variety of samples from different genetic environments (49 amygdaloidal, 73 subaerial and 39 deep-sea diagenetically altered sediments, and 18 from 'burial diagenesis') show the following average values: R = 0.81, M/(M+D) = 0.60, Na/(Na+K) = 0.50, and Ca/(Ca+Mg+Sr+Ba) = 0.70. Amygdaloidalal samples are generally Si-poorer (R = 0.78) than samples from the other genetic environments (R = 0.81-0.82) but they show a high variability in the framework content (R = 0.73-0.83) as do samples from diagenetically altered subaerial sediments (R = 0.74-0.85). The framework contents of the samples from diagenetically altered deep-sea sediments and “burial diagenesis” environments are less variable (R = 0.77-0.84). Monovalent cations commonly are slightly prevalent over divalent cations but are slightly subordinate in amygdaloidal samples, although many crystals are very rich in alkalis (Wise et al. 1969, Alberti 1975b, Yoshimura and Wakabayashi 1977, Barnes et al. 1984, Wise and Kleck 1988). Moreover, M ≈ D in samples from diagenetically altered subaerial sediments, and M »D in samples from both deep-sea (Boles and Wise 1978, Sameshima 1978, Stonecipher 1978, Ogihara and Iijima 1990, Vannucci et al. 1992, Ogihara 1994) and “burial diagenesis” sediments (Iijima 1978, Minato and Aoki 1978, 1979; Minato et al. 1980, Ogihara and Iijima 1989, Tsolis-Katagas and Katagas 1990). Na and K are comparable, but Na is slightly prevalent in amygdaloidal crystals and slightly lower in the other samples where, however, Na-rich samples occur (Sheppard and Gude 1969a, Honda and Muffler 1970, Utada et al. 1972, Minato and Aoki 1978, Bargar and Beeson 1981, Ogihara and Iijima 1989, Vannucci et al. 1992, Ogihara 1994). Both the frequency and abundance of Mg are higher in sedimentary samples, but Sr and Ba are higher in amygdaloidal crystals. Unit-cell parameters show the following ranges: *a* = 17.62-17.74 Å, *b* = 17.81-18.05 Å, *c* = 7.39-7.53 Å, and β = 116.13-116.90°. Hawkins (1974) reported that an increase in R causes *b* to lengthen and *c* to shorten.

Structure Type: **LAU**

Laumontite: $Ca_4[Al_8Si_{16}O_{48}]\cdot 18\text{-}14H_2O$

Laumontite is a widespread zeolite that occurs in many environments, including (a) in vugs of plutonic (granite, syenite, monzonite, and gabbro), volcanic (basalt, andesite, and porphyrite), metamorphic (gneiss, amphibolite, hornfels, marble, and skarn) and sedimentary (mudstone, and sandstone) rocks; (b) as a cement of plagioclase-rich sandstone; (c) as a precipitate from spring waters at atmospheric pressure and temperature (McCulloh et al. 1981); and (d) as an alteration product of vitroclastic sediments (tuff, and tuffite) in “burial diagenesis” or in low-grade metamorphism (zeolite facies). Laumontite coexists with other zeolites (natrolite, stilbite, heulandite, scolecite, and thomsonite), calcite, quartz, fluorite, plagioclase, thenardite, gypsum, corrensite, and chlorite.

The chemical composition is generally very close to the schematic formula. R shows a mean value of 0.67, ranging from 0.65 to 0.69. Ca is mainly the DEC inasmuch as the average content of Na and K is usually 0.3 and 0.2, respectively. A sample, originally designated “primary leonhardite” and occurring in fissures of porphyritic andesite at Kurtsy, Crimea, Ukraine, shows low Ca (2.15) and H_2O (13.5) contents and high Na (1.85) and K (1.85) contents (Stolz and Armbruster 1997). The notable content of Be (0.36) in a sample from an altered diabase at Toggiano, Italy (Gallitelli 1928), requires confirmation. Mg, Sr, and Ba are absent or negligible.

The structure (Artioli et al. 1989, Artioli and Ståhl 1993) is monoclinic *C*2/*m* with *a* = 14.69-14.89 Å, *b* = 13.05-13.17 Å, *c* = 7.53-7.61 Å, and β = 110-113°; unit-cell variations depend mainly on the degree of hydration. The uncertainty in the water content given in the schematic formula is due to partial reversible dehydration of the zeolite at room temperature. The resulting phase (named “leonhardite”) shows slightly shorter unit-cell parameters and lower refractive indices (Coombs 1952, Pipping 1966) but retains the original symmetry (Artioli et al. 1989) and, hence, must be considered a variety of laumontite. The term “leonhardite” should be avoided as a species name.

Structure Type: **LEV**

Levyne : $(Ca_{0.5},Na)_6[Al_6Si_{12}O_{36}]\cdot18H_2O$

Levyne has been described only from vugs of massive volcanic rocks (mainly basalt and andesite but also dolerite and tephritic-leucitite) associated with many other zeolites (analcime, gmelinite, natrolite, thomsonite, chabazite, gismondine, cowlesite, phillipsite, and heulandite). It can be epitaxially overgrown by erionite (Galli et al. 1981, Passaglia et al. 1974, Mizota et al. 1974, Wise and Tschernich 1976a) or by offretite (England and Ostwald 1979, Ridkosil and Danek 1983, Birch 1987, 1989; Kile and Modreski 1988). R shows a mean value of 0.66 and ranges from 0.62 to 0.70. The lowest value is in a sample from Oki Island, Japan (Tiba and Matsubara 1977), and the highest R is in a sample from Milwaukie, Oregon (Wise and Tschernich 1976a). Ca is generally the DEC, although Na-dominant samples are not rare (Birch 1979, 1989; Galli et al. 1981). Na is consistently present and prevails over K which is lower than 0.5. Mg, Sr, and Ba may be present in negligible amounts.

The structure is rhombohedral $R\bar{3}m$ (Merlino et al. 1975) with a = 13.32-13.43 Å and c = 22.66-23.01 Å The monovalent-cation content (mainly Na) is positively correlated with the a parameter and negatively correlated with both the c parameter (Galli et al. 1981) and refractive indices (Tiba and Matsubara 1977).

Structure Type: **LOV**

Lovdarite: $Na_{13}K_4[Be_8AlSi_{27}O_{72}]\cdot20H_2O$

Lovdarite occurs in association with natrolite in a pegmatite vein at Mount Karnasurt, Lovozero massif, Russia (Men'shikov et al. 1973). Crystallochemical data (Khomyakov et al. 1975) match those given in the original description showing: (a) in the tetra- hedral framework, the presence of Be is remarkable, but (Si+Al) are prevalent, with R [Si/(Si+Al+Be)] equal to 0.75; (b) the extraframework content is almost exclusively Na and K in the ratio shown in the schematic formula; and (c) negligible amounts of Ca, Mg, and Ba were reported only in the original description. The structure is orthorhombic *Pma*2 with a = 39.58 Å, b = 6.93 Å, and c = 7.15 Å (Merlino 1990).

Structure Type: **LTL**

Perlialite: $K_9Na(Ca,Mg,Sr)[Al_{12}Si_{24}O_{72}]\cdot16H_2O$

Only two occurrences of perlialite, both from Russian alkalic rocks, have been described from the Khibiny massif, Kola Peninsula (Men'shikov 1984) and from the Murun massif (Konev et al. 1986). The crystallochemical data for samples from both occurrences are very similar. R shows a mean value of 0.66 and ranges from 0.65 to 0.67 (Khibiny massif); K dominates (~9); Na is close to 1.0; and divalent cations add up to about 1.0, with Ca prevailing over Mg and Sr. Ba was determined in noticeable amounts (0.37) only in the sample from the Murun massif.

The crystal structure, determined on a Tl-rich (following heavy-liquid separation) sample, revealed hexagonal (*P*6/*mmm*) symmetry with a = 18.54 Å and c = 7.53 Å (Artioli and Kvick 1990). The unit-cell parameters given in the original descriptions are: a = 18.49-18.54 Å and c = 7.51-7.53 Å

Structure Type: **MAZ**

Mazzite : $Mg_{2.5}K_2Ca_{1.5}[Al_{10}Si_{26}O_{72}]\cdot30H_2O$

Mazzite occurs at Mt. Semiol (France) as clear, transparent hexagonal prisms that form radiating bundles, associated with offretite, phillipsite, chabazite, calcite, and siderite in vugs of an olivine basalt (Galli et al. 1974). The chemical composition reported in the original description is apparently unreliable because it has a large E value (+13.6%). A new electron microprobe analysis carried out on holotype crystals (G.Vezzalini, pers. comm.) is more reliable (E = +6.5%) and corresponds to: $Mg_{2.25}K_{2.18}Ca_{1.30}Na_{0.18}Ba_{0.02}$-$[Al_{10.13}Si_{26.03}O_{72}]\cdot30H_2O$. R is 0.72, Mg is the DEC, K is high, and Ca is moderate. The structure is hexagonal *P*6$_3$/*mmc* (Galli 1975) with *a* = 18.39 Å and *c* = 7.65 Å.

Structure Type: **MER**

Merlinoite: $K_6Ca_2Na[Al_{11}Si_{21}O_{64}]\cdot22H_2O$

Merlinoite occurrences have been reported from massive volcanic rocks (Passaglia et al. 1977, Alberti et al. 1979, Khomyakov et al. 1981, Hentschel 1986, Della Ventura et al. 1993, Yakubovich et al. 1999), in silicic tephra that was diagenetically altered in a lacustrine environment (Hay and Guldman 1987), and in marine volcanic sediments (Mohapatra and Sahoo 1987). Nevertheless, reliable crystallochemical data have been obtained only from the samples at the type locality (Cupaello, Italy), where merlinoite is associated with phillipsite, chabazite, apophyllite, and calcite in vugs of a nepheline-melilitite (Passaglia et al. 1977), at Sacrofano, Italy, in cavities of a fassaite-rich ejectum (Della Ventura et al. 1993), and at Kola Peninsula, Russia in a pegmatite lode that slices nepheline syenite associated with cancrinite, sodalite and nepheline (Yakubovich et al. 1999). Samples from the three occurrences show different tetrahedral contents (R = 0.71 in the sample from Cupaello, 0.66 in the sample from Sacrofano, and 0.62 in the sample from Kola Peninsula). The extraframework content shows K as DEC. Ca and Ba are present in subordinate amounts only in the samples from Cupaello and Sacrofano. Na content is remarkable (3.16) in the sample from Kola Peninsula, otherwise quite low (0.55-0.67). Negligible amounts of Mg, Sr, and Mn were found only in the sample from Sacrofano. The structure is orthorhombic *Immm* (Galli et al. 1979) with *a* = 13.86-14.14 Å, *b* = 14.13-14.23 Å, and *c* = 9.95-10.05 Å.

Structure Type: **MFI**

Mutinaite: $Na_3Ca_4[Al_{11}Si_{85}O_{192}]\cdot60H_2O$

Mutinaite is a newly found zeolite in a few spheroidal cavities of Ferrar dolerite at Mt. Adamson, Antarctica (Galli et al. 1997a). It occurs as subspherical aggregates of tiny radiating lath-like fibers or as aggregates of transparent, colorless to pale-milky, tiny tabular crystals, in association with heulandite and, very rarely, with terranovaite and tschernichite. The empirical formula $(Na_{2.76}K_{0.11}Mg_{0.21}Ca_{3.78})[Al_{11.20}Si_{84.91}O_{192}]\cdot60H_2O$ shows the highest R value (0.88) found in a natural zeolite to date, Ca fairly prevalent over Na, and Mg and K very subordinate. The structure is orthorhombic *Pnma* with *a* = 20.22 Å, *b* = 20.05 Å, and *c* = 13.49 Å.

Structure Type: **MON**

Montesommaite: $K_9[Al_9Si_{23}O_{64}]\cdot10H_2O$

Montesommaite has been described from Pollena, Monte Somma, Italy, where it occurs as colorless, dipyramidal crystals associated with chabazite, natrolite, dolomite, and calcite in vesicles of volcanic scoria (Rouse et al. 1990). The chemical composition is very close to the schematic formula, showing an R value of 0.70 and a very low Na content (0.16). The structure is orthorhombic *Fdd*2, pseudo-tetragonal ($I\bar{4}2d$ or *I*4$_1$*md*) with *a* = 7.14 Å and *c* = 17.31 Å. On the basis of symmetry, unit-cell parameters, and chemical composition, the zeolite is presumed to be related to merlinoite and members of the gismondine group (Rouse et al. 1990).

Structure Type: **MOR**

Mordenite : $Na_3Ca_2K[Al_8Si_{40}O_{96}]\cdot28H_2O$

Maricopaite : $Pb_7Ca_2[Al_{12}Si_{36}(O,OH)_{100}]\cdot n(H_2O,OH)$

Mordenite and maricopaite share the same orthorhombic structure type, but they have different space groups (*Cmc*2_1 in mordenite, *Cm*2*m* in maricopaite). Moreover, maricopaite has a so-called "interrupted" framework (see Introduction).

Mordenite has been described in vugs of volcanic (andesite, basalt, dolerite, porphyrite, and rhyolite) and intrusive (granite) rocks (Sukheswala et al. 1974, Passaglia 1975, de'Gennaro et al. 1977, Wise and Tschernich 1978b, Nativel 1986, Vezzalini et al. 1994), and as a diagenetic product of silicic tuff (Sheppard and Gude 1969a, Noh and Kim 1986, Sheppard et al. 1988, Tsolis Katagas and Katagas 1989, Pe-Piper and Tsolis Katagas 1991, Passaglia et al. 1995), pitchstone (Harris and Brindley 1954), and volcanic marine sediments (Bushinsky 1950, Vitali et al. 1995). Amygdaloidal samples are associated with other zeolites (analcime, chabazite, heulandite, dachiardite, stilbite, natrolite, scolecite, epistilbite, ferrierite, erionite, and thomsonite), silica minerals (quartz, cristobalite, and chalcedony), calcite, apophyllite, okenite, and barite. In diagenetically altered volcanic sediments, mordenite occurs in association with clinoptilolite, chabazite, erionite, phillipsite, ferrierite, analcime, tridymite, cristobalite, opal, K-feldspar, and smectite. It is also commonly associated with silica minerals. Many amygdaloidal samples are described as red compact masses with a radial, fibrous structure, the fibers being surrounded by microcrystalline quartz and hematite (Passaglia 1975).

Amygdaloidal and sedimentary samples show the same mean value of R (0.83) with almost the same ranges (0.81-0.86 in amygdaloidal samples, 0.80-0.85 in the sedimentary ones). The lowest R value (0.80) was found in a sample from volcanic marine sediments at Tonga Trench, Southwest Pacific (Vitali et al. 1995); the highest R value (0.86) is in a sample from basalt at Kirkee, India (Passaglia 1975). The extraframework cations are mainly Na, Ca, and K, generally with Na > Ca > K. Ca dominates in the samples from porphyrite at Mt. Civillina, Italy (Passaglia 1975), from basalt at Deccan, India (Sukheswala et al. 1974), and from a diagenetically altered rhyolite at Samos, Greece (Pe-Piper and Tsolis-Katagas 1991). K dominates in an amygdaloidal sample from a pegmatite at Elba Island, Italy (Alberti et al. 1986), and in sedimentary samples from silicic tuffs at Yucca Mountain, Nevada (Sheppard et al. 1988), and near Yeongil, Korea (Noh and Kim 1986). Ca and Na are the same (2.41) in a sample from basalt at Cilaos, Ile de la Reunion (Nativel 1986). The Mg content is commonly low, showing higher values in the sedimentary samples. Sr is generally absent or negligible, although it is relatively high (0.45) in samples from basalts at Penticton, British Columbia ("ashtonite" of Reay and Coombs 1971), and at Cilaos, Ile de la Reunion (Nativel 1986). Ba is generally absent or in traces, but it reaches a remarkable value (0.43) in a sample from basalt at Leucois Paulista, Brazil (Passaglia 1975). The amygdaloidal samples have a mean R comparable with that of the sedimentary samples but are slightly richer in Na and Ca and poorer in K and Mg.

The structure is orthorhombic *Cmc*2_1 (Alberti et al. 1986) with a = 18.05-18.25 Å, b = 20.35-20.53 Å, and c = 7.49-7.55 Å. The b parameter is negatively correlated with R (Passaglia 1975).

Maricopaite has been found only at the Moon Anchor Mine, Tonopah, Maricopa County, Arizona (Peacor et al. 1988). It occurs as sprays of translucent, white, acicular crystals coating and filling fractures in quartz, with mimetite in a vein of a calcite-fluorite gangue. The tetrahedral framework is only partially occupied by Si and Al (R = 0.75), and the extraframework cations are exclusively Pb and Ca, showing amounts close to those given in the schematic formula. Both density (2.94 g/cm^3) and refractive indices (α = 1.56, β = 1.58, γ = 1.59) are higher than those generally found in zeolites but are consistent with the Pb content of the mineral.

The structure (Rouse and Peacor 1994) is orthorhombic *Cm*2*m* with *a* = 19.43 Å, *b* = 19.70 Å, and *c* = 7.54 Å. The tetrahedral framework is "interrupted" by (OH) groups with 17% of TO_4 three-fold connected. $Pb_4(O,OH)_4$ tetrahedral clusters occupy and obstruct cruciform channels. Maricopaite is similar to roggianite and partheite in that it exhibits a framework interrupted by (OH), but it is the only zeolite having Pb as the extraframework cation.

Structure Type: **NAT**

Natrolite: $Na_{16}[Al_{16}Si_{24}O_{80}]\cdot 16H_2O$
Mesolite: $Na_{16}Ca_{16}[Al_{48}Si_{72}O_{240}]\cdot 64H_2O$
Scolecite: $Ca_8[Al_{16}Si_{24}O_{80}]\cdot 24H_2O$
Gonnardite-Tetranatrolite: $Na_{12}Ca_{2.5}[Al_{17}Si_{23}O_{80}]\cdot 20H_2O$ ($Z = 1/2$)

These zeolites share the same tetrahedral framework with TS $I4_1/amd$ which lowers to $I\bar{4}2d$ by an overall rotation of the structural chains by an angle of about 24° . The RS of the zeolite species is determined by the (Si,Al) distribution and occupancy of the extraframework cations and water molecules. The completely ordered (Si,Al) distribution in mesolite, scolecite, and most of the natrolite samples lowers their RS to orthorhombic *Fdd*2, which, in turn, is lowered to monoclinic *F*1*d*1 in scolecite because the sites occupied by Na in natrolite are alternatively occupied by Ca and H_2O in scolecite (Peacor 1973, Pechar 1981, Pechar et al. 1983, Artioli et al. 1984, 1986; Kirfel et al. 1984, Smith et al. 1984). An orthorhombic structure is retained in a few natrolite samples with a partially disordered (Si,Al) distribution (Alberti and Vezzalini 1981, Hesse 1983, Krogh Andersen et al. 1990, Alberti et al. 1995). A strongly disordered (Si,Al) distribution in gonnardite-tetranatrolite prevents their RS from departing from tetragonal $I\bar{4}2d$ (Chen and Chao 1980, Mazzi et al. 1986, Mikheeva et al. 1986, Pechar 1989, Artioli and Torres Salvador 1991). No sound chemical or crystallographic parameter was found to discriminate gonnardite from tetranatrolite; thus, because of time priority, it was suggested to retain gonnardite as a mineral species and to discredit tetranatrolite (Artioli and Galli 1999). Alberti et al. (1982c) assessed the compositional fields of natrolite, mesolite, and scolecite, and found a strong correlation between the unit-cell parameters and the degree of disorder in the Si-Al distribution. A crystallochemical classification of the minerals of the natrolite group, including the non-isostructural mineral thomsonite, was proposed on the basis of new chemical data and data taken from the literature (Nawaz 1988, Ross et al. 1992). Unusual crystallization conditions (high temperature and H_2O pressure) favor the formation of disordered gonnardite-tetranatrolite instead of ordered natrolite. In this context, paranatrolite (Chao 1980) is interpreted as a highly hydrated phase, which under room conditions dehydrates into disordered gonnardite-tetranatrolite (Alberti et al. 1995).

Natrolite is the most common among the zeolites of this group and has been described mainly from vugs of massive plutonic (nepheline syenite, pegmatite, and diorite), volcanic (basalt, phonolite) and metamorphic (gneiss, marble) rocks. It occurs in association with analcime, phillipsite, chabazite, garronite, gismondine, stilbite, gmelinite, pectolite, apophyllite, prehnite, calcite, and aragonite, and also in intergrowths with other "fibrous" zeolites (mesolite, thomsonite, and gonnardite). The chemical composition is very close to the schematic formula. R generally ranges from 0.60 to 0.61, being slightly lower (0.58-0.59) in a few samples (Cortesogno et al. 1975, Pechar et al. 1983, Kile and Modreski 1988, Pechar and Rykl 1989, Ross et al. 1992, Alberti et al. 1995, Meneghinello et al. 1999), which, according to Lowenstein's rule, are partially (Si, Al) disordered (Alberti et al. 1995). The negative charge of the framework is balanced by Na and very small amounts of Ca. K, Mg, Sr, and Ba are absent or negligible (<0.5). Crystals with appreciable $Ca \Leftrightarrow Na_2$ substitutions are not rare. In fact, relatively high Ca contents (1-3) and consequently relatively low Na contents (14-10) were found in samples from Voltri, Italy (Cortesogno et al. 1975), and from several localities in Czechoslovakia (Pechar et al. 1983, Pechar and Rykl 1989). Crystals closely intergrown with gonnardite from Palagonia, Italy, and from Uzabanya, Hungary, have the highest degree (~50%) of disordered (Si,Al) distribution ever found in orthorhombic natrolites (Alberti et al. 1995). The structure is orthorhombic *Fdd*2 and the unit-cell parameters (a = 18.28-18.42 Å, b = 18.56-18.70 Å, and c = 6.46-6.61 Å) do not show any correlations with the chemistry. The (b-a) difference decreases and c increases with increasing (Si,Al) disorder (Alberti and Vezzalini 1981). Correlations between optical properties and crystal chemistry are reported in Gunter and Ribbe (1993).

Mesolite occurs mainly in vugs of massive volcanic rocks (andesite, basalt, and porphyrite) in association with analcime, natrolite, scolecite, thomsonite, chabazite, levyne, laumontite, erionite, and heulandite. R generally shows only small deviations (0.60-0.62) from the theoretical value (0.60) and is slightly lower (0.58-0.59) in crystals occurring in fissures of contact metamorphosed marls in the proximity of a phonolitic intrusion at Horni Jilové, Czechoslovakia (Rychly and Ulrych 1980), and in samples from Carlton Peak, Minnesota (Nawaz et al. 1985), Dunseverick, Ireland (Nawaz 1988), Table Mountain, Colorado (Kile and Modreski 1988), and Coastal Ranges, Oregon (Keith and Staples 1985). Na and Ca are the DEC, and K, Mg, Sr, and Ba are absent or negligible. Most samples have a Na/(Na+Ca) ratio (0.48-0.52) close to the theoretical value (0.50), although some samples with Ca > Na [Na/(Na+Ca) = 0.45-0.47] also have been described (Sukheswala et al. 1974, Alberti et al. 1982c, Artioli et al. 1986). A sample with Ca » Na [Na/(Na+Ca) = 0.41] occurs at Horni Jilové, Czechoslovakia (Rychlý and Ulrych 1980). Crystals with Na > Ca have been found in tholeiitic basalt of the Coastal Ranges, Oregon (Keith and Staples 1985), and in altered volcanogenic sediments of Baja California, Mexico [Na/(Na+Ca) = 0.56 and 0.61, respectively] (Barnes et al. 1984). Inasmuch as the structure of mesolite is built by the alternation of one natrolite plane with two scolecite planes, the b parameter is three times larger than that for natrolite and scolecite. The structure is orthorhombic *Fdd*2 with unit-cell parameters showing narrow ranges (a = 18.34-18.44 Å, b = 56.52-56.69 Å, and c = 6.52-6.56 Å) and uncorrelated with chemistry (Alberti et al. 1982c).

Scolecite occurs in vugs of mainly massive volcanic rocks (basalt, andesite, and dolerite), rarely of metamorphic rocks (gneiss, and skarn), and amphibole gabbro. It has been described in association with other zeolites (natrolite, mesolite, thomsonite, heulandite, stilbite, laumontite, chabazite, levyne, and epistilbite), prehnite, calcite, quartz, and pyrite (Kuwano 1977, Alberti et al. 1982c, Johnson et al. 1983, Yamazaki and Otsuka 1989, Ross et al. 1992, Vezzalini et al. 1994). The chemistry is close to the schematic formula: R ranges from 0.60 to 0.61; Ca is by far the DEC; Na is consistently subordinate; and K as well as Mg, Sr, and Ba are negligible or absent. Exceptionally Na-rich (0.87-1.54), and consequently Ca-poor (7.10-7.35), crystals occur in the skarn area of the Italian Mountains, Colorado, basalt of Antrim, Ireland (Alberti et al. 1982c), dolerite of Mt. Adamson, Antarctica (Vezzalini et al. 1994), and andesite of Makinokawa, Japan (Kuwano 1977). The structure is monoclinic *F*1*d*1 with unit-cell dimensions (a = 18.50-18.55 Å, b = 18.90-18.97 Å, c = 6.52-6.53 Å, and β = 90.44-90.65°) correlated with the extra-framework cation contents (Alberti et al. 1982c).

Gonnardite-Tetranatrolite. On the whole, 26 occurrences from 22 different localities have been reported. Out of these, eleven samples have been described as tetranatrolites (Krogh Andersen et al. 1969, Guseva et al. 1975, Chen and Chao 1980, Alberti et al. 1982b, Mikheeva et al. 1986, Nawaz 1988, Pechar 1989, Ross et al. 1992), the other ones as gonnardites (Meixner et al. 1956, Harada et al. 1967, Iijima and Harada 1968, Alberti et al. 1982b,c; Mazzi et al. 1986, Nawaz 1988, Ciambelli et al. 1989, Passaglia et al. 1990 1992, Ross et al. 1992). This zeolite occurs in fractures of pegmatite dikes and in cavities of both massive volcanic rocks (tholeiitic and nepheline basalt, and ijolite) and palagonitic tuff and basalt usually in association with other fibrous zeolites (thomsonite, natrolite), but also with analcime, garronite, phillipsite, chabazite, and calcite. R displays a mean value of 0.57, ranging from 0.52 in the sample from Magnet Cove, Arkansas (Ross et al. 1992) to 0.63 in the sample from Lovozero massif, Russia (Guseva et al. 1975, Rastsvetaeva 1995). Only four samples which were all described as tetranatrolites (Krogh Andersen et al. 1969, Guseva et al. 1975, Chen and Chao 1980, Pechar 1989) show R > 0.60, i.e. Si/Al > 1.5. Na is the DEC and Ca the SEC. The Na/(Na+Ca) ratio has a mean value of 0.84, ranging from 0.65 in the sample from Aci Castello, Sicily (Meixner et al. 1956) to 1 in the sample from Lovozero massif, Russia (Guseva et al. 1975). K is usually found in negligible amounts (<0.10), although it surprisingly reaches a value of 0.88 in the sample from the Khibinsk alkali massif, Russia (Mikheeva et al. 1986). Mg and Ba are absent or negligible. Sr, usually absent, show the maximum value (0.11) in a sample from Mont Saint-Hilaire, Canada (Ross et al. 1992). The water content is on average about 20 molecules, but is higher (about 23 molecules) in the samples described as gonnardites than (about 17 molecules) in the samples described as tetranatrolites. A positive correlation between Na/Ca and Si/Al ratios has been postulated (Passaglia et al. 1992). The structure is tetragonal $I\bar{4}2d$ with a = 13.04-13.27 Å, and c = 6.58-6.64 Å.

Structure Type : **NES**

Gottardiite: $Ca_5Na_3Mg_3[Al_{19}Si_{117}O_{272}]\cdot 93H_2O$

Gottardiite occurs as thin pseudo-hexagonal lamellae in subparallel or wedge-shaped aggregates in vugs lined by Fe-smectite in dolerite at Mt. Adamson, Antarctica (Galli et al. 1996). The empirical formula $(Na_{2.5}K_{0.2}Mg_{3.1}Ca_{4.8})[Al_{18.8}Si_{117.2}O_{272}]\cdot 93H_2O$ shows a high R value (0.86) and Ca, Mg, and Na contents close to the schematic formula. The structure is orthorhombic *Cmca* with a = 13.70 Å, b = 25.21 Å, and c = 22.66 Å (Alberti et al. 1996).

Structure Type: **OFF**

Offretite: $CaMgK[Al_5Si_{13}O_{36}]\cdot 16H_2O$

Offretite has been characterized from a dozen occurrences in vugs of massive volcanic rocks (basalt, limburgite, and cornubianite) in association with mazzite, chabazite, levyne, erionite, phillipsite, and faujasite. Epitaxial intergrowths with erionite (Pongiluppi 1976, Rinaldi 1976, Wise and Tschernich 1976a), and overgrowths on levyne (Sheppard et al. 1974, Wise and Tschernich 1976a, England and Ostwald 1979, Kile and Modreski 1988, Birch 1989) and chabazite (Passaglia and Tagliavini 1994, Passaglia et al. 1996) have been described. Compared with erionite, offretite is typically less siliceous, richer in alkaline earths, and optically negative (Sheppard and Gude 1969b, Rinaldi 1976, Wise and Tschernich 1976a). R shows a mean value of 0.70 with small variations (0.69-0.72), and Ca is the DEC in most samples apart from two occurrences in Australia (Birch 1988 1989) which are Na-dominant. Crystals from Mont Semiol, France (the type locality), and from Sasbach (Germany) are Mg dominant. Mg and Ca are comparable in samples from Adamello, Italy (Passaglia and Tagliavini 1994), and Fittà, Italy (Passaglia et al. 1996). K is consistently close to 1. Sr and Ba are absent. A study of the crystal chemistry (Passaglia et al. 1998) allowed the following conclusions: (1) R ranges from 0.69 to 0.74; (2) Ca, Mg and K are in comparable amounts (~1), and Na, Sr and Ba are generally negligible or absent; (3) in clear disagreement with previous reports, epitaxial overgrowths are found only on chabazite and not on levyne; (4) epitaxial intergrowths with erionite are rare; and (5) the optic sign depends on the Si/Al ratio of the sample (positive in Si-rich crystals, negative in Si-poor crystals). The structure is hexagonal $P\bar{6}m2$ with a = 13-27-13.32 Å and c = 7.56-7.61 Å.

Structure Type: **PAR**

Partheite: $Ca_8[Al_{16}Si_{16}O_{60}(OH)_8]\cdot16H_2O$

Partheite has been described from the Taurus Mts, Turkey, occurring in rodingitic rocks associated with thomsonite and prehnite (Sarp et al. 1979), and from Denezhkin Kamen (Urals), occurring in a gabbro-pegmatite (Ivanov and Mozzherin 1982). The chemical compositions from the two occurrences are very similar and are close to the schematic formula: R averages 0.51, and Ca is essentially the only extraframework cation inasmuch as Na, K, and Mg are negligible. The structure is characterized by an interrupted tetrahedral framework with unshared (OH) vertices at every second AlO_4 tetrahedra. The symmetry is monoclinic $C2/c$ with a = 21.55-21.59 Å, b = 8.76-8.78 Å, c = 9.30-9.31 Å, and β = 91.47-91.55° (Sarp et al. 1979, Engel and Yvon 1984).

Structure Type: **PAU**

Paulingite: $K_{58}Ca_{42}Na_{10}Ba_{10}[Al_{172}Si_{500}O_{1344}]\cdot490H_2O$

Paulingite has been described only in five localities: three in the USA, one in British Columbia, Canada (Tschernich and Wise 1982), and one in the Czech Republic (Lengauer et al. 1997). It occurs in vugs of basalt mainly associated with phillipsite, erionite-offretite and smectite, but also with chabazite, heulandite, harmotome, calcite, and pyrite. The schematic formula was derived from the average of the available chemical compositions which have a very large variability. The tetrahedral framework content is almost constant (R = 0.73-0.74 in four samples and 0.77 in the holotype sample from Rock Island Dam, Washington). As far as the extraframework content is concerned, K > Ca except in the samples from Ritter, Oregon, and Vinarická Hora, Czech Republic, where Ca is the dominant cation. Na ranges from 6.08 to 15.2, and Ba ranges from 1.60 to 22.24. Mg and Sr were found in crystals from Riggins, Idaho, and in relatively high amounts (1.74 and 1.44, respectively) in crystals from Chase Creek, British Columbia. The structure is cubic $Im3m$ (Gordon et al. 1966) with a = 35.05-35.12 Å.

Structure Type: **PHI**

Phillipsite: $K_2(Na,Ca_{0.5})_3[Al_5Si_{11}O_{32}]\cdot12H_2O$

Harmotome: $Ba_2(Na,Ca_{0.5})[Al_5Si_{11}O_{32}]\cdot12H_2O$

On the basis of structural and crystallochemical data, phillipsite and harmotome can be described as a single zeolite species. In both structures, the orthorhombic $Bmmb$ TS is lowered to monoclinic $P2_1/m$ as a result of a slight distortion induced on the tetrahedral framework by the presence of larger cations (K and Ba) in the two-fold extraframework sites. The other extraframework site is four-fold and accommodates the smaller Ca and Na cations (Rinaldi et al. 1974). The variation in Si/Al, K/Ba, and Na/Ca suggests the absence of a compositional gap, and the unit-cell parameters vary as a function of the Si/Al and (Na+K)/(Na+K+Ca+Ba) ratios. R ranges from 0.57 to 0.77, K/(K+Ba) ranges from 0.03 to 1, Na/(Na+Ca) ranges from 0.02 to 1, and other extraframework cations (Mg, Sr) are absent or very low. The unit-cell parameter (pseudo-orthorhombic setting) ranges are: a = 9.81-10.01 Å, b = 14.10-14.34 Å, and c = 14.16-14.42 Å. The existence of a continuous isomorphous series between phillipsite and harmotome, suggested by Cerny et al. (1977), has been reinforced by later descriptions of samples with intermediate compositions (Tschernich and Wise 1982, Passaglia and Bertoldi 1983, Robert 1988, Hansen 1990, Armbruster et al. 1991). The names phillipsite and harmotome are retained in present nomenclature, and, in light of the above considerations, samples with K/(K+Ba) > 0.50 (i.e. K > Ba) will be descibed as phillipsite, whereas samples with K/(K+Ba) < 0.50 (i.e. Ba > K) will be described as harmotome. The term 'wellsite,' commonly utilized for Ba-rich phillipsites, is not used. On the basis of structural data, the extraframework two-fold site can be almost completely occupied by K and Ba, and the four-fold site can be partially occupied by Na and Ca. These trends occur in all harmotomes and in most phillipsites, although some K-rich (3-4 atoms p.f.u) phillipsites must accommodate some K in the four-fold site, and the existence of many K-poor (<1 atoms p.f.u.) phillipsites may indicate the presence of Ca and Na also in the two-fold site.

Phillipsite is much more common than harmotome and has been described from vugs of massive volcanic rocks (basalt, nephelinite, leucitite, and melilite), as an alteration product of volcanic glass in palagonitic basalt and tuff (“hyaloclastites”), and in vitroclastic sediments diagenetically altered in continental (“closed,” “open,” and “geoautoclave” systems) and deep-sea environments. R has an average value of 0.69 and ranges from 0.57 to 0.77. K/(K+Ba) has an average value of 0.99 but ranges from 0.53 to 1, and Na/(Na+Ca) shows an average of 0.6 with a range from 0.02 to 1. Amygdaloidal samples occur as twinned pseudo-orthorhombic prisms or radiating fibrous aggregates with many other zeolites, carbonates, and silica minerals. Intergrowths with gismondine are common (Vezzalini and Oberti 1984, Pöllmann and Keck 1990) but intergrowths with garronite (Alberti et al. 1982b) and with gobbinsite (Artioli and Foy 1994) are rare. R has a mean value of 0.66 and ranges from 0.57 in crystals associated with gismondine and Si-poor chabazite from the leucitite of Vallerano, Italy (Galli and Loschi Ghittoni 1972), to 0.76 in crystals associated with erionite, clinoptilolite, mordenite, and dachiardite in basalt of Cape Lookout, Oregon (Wise and Tschernich 1978b). The K/(K+Ba) ratio shows a mean value of 0.95 and ranges from 1 in most samples to 0.53 in crystals associated with heulandite, gmelinite, analcime, laumontite, and prehnite from igneous rocks of Kurtzy, Crimea (Cerny et al. 1977). The Na/(Na+Ca) ratio averages 0.40 and ranges from 0.02 (almost Na-free) in crystals associated with chabazite from basalt of Ardeche, France (Robert et al. 1988), to 1 (Ca- and K-free) in crystals associated with Na-rich chabazite, gmelinite, clinoptilolite, searlesite, borax, and calcite in basalt of Boron, California (Wise and Kleck 1988). Phillipsites from “hyaloclastites” display an average R value of 0. 65, varying from 0.62 in crystals associated with gismondine and chabazite in palagonitic tuff of Oahu, Hawaii (Iijima and Harada 1968), to 0.73 in crystals associated with chabazite in palagonitic trachybasalt of Vivara, Italy (Passaglia et al. 1990). All samples are almost Ba-free [K/(K+Ba) = 0.98-1], and the Na/(Na+Ca) ratio is high (0.8) in crystals associated with analcime, gmelinite, chabazite, gyrolite, thomsonite, and thaumasite in palagonitic basalt of the Mururoa Atoll, South Pacific (Noack 1983), and in crystals from Vivara (Italy). The Na/(Na+Ca) ratio is intermediate (0.3) in crystals from Hawaii and low (0.1) in crystals associated with chabazite and gonnardite in palagonitic basalt of Palagonia, Sicily (Passaglia et al. 1990). Diagenetic phillipsites from “closed systems” have R values ranging from 0.76 to 0.77 for crystals in altered rhyolitic tuff (Hay 1964, Sheppard and Gude 1968, 1969a; Surdam and Sheppard 1978) and lower values (0.71-0.73) in crystals from nepheline phonolitic and trachytic tuffs at Olduvai Gorge, Tanzania (Hay 1964 1980). Ba is absent, and the K content is close to 2, except for crystals from China Lake, California (Hay 1964), and near Barstow, California (Sheppard and Gude 1969a), where it is about 1. Na greatly prevails over Ca [Na/(Na+Ca) = 0.83-1.0]. Diagenetic phillipsites from “open systems” (mainly “geoautoclaves”) exhibit an R (average value = 0.72) ranging from 0.70 to 0.74 in crystals from phonolitic-trachytic pyroclastite. A lower value (0.68) is for the sample from trachytic ash-flow tuff of the Canary Islands (Garcia Hernandez et al. 1993), and a higher value is for the sample from rhyodacitic cinerite of Garbagna, Italy (Passaglia and Vezzalini 1985). Ba is absent or negligible [K/(K+Ba) = 0.95-1]. The Na/Ca ratio is extremely variable: Na›Ca in samples from the Neapolitan Yellow Tuff, Italy (Passaglia et al. 1990), from the Canary Islands, and in most specimens from Eifel, Germany (Adabbo et al. 1994); Ca » Na in samples from “phonolitic tephritic ignimbrite with black pumices,” Italy (Passaglia et al. 1990); and Na ≈

Ca in sample from Garbagna, Italy. Diagenetic crystals in pelagic volcanic sediments of the Pacific, Indian, and Atlantic Oceans ("deep-sea" phillipsites) have compositions that are very similar. R values are 0.71-0.74, K contents are close to 2, Ba is absent, and Na greatly prevails over Ca (Sheppard et al. 1970, Kastner and Stonecipher 1978, Stonecipher 1978).

The unit-cell parameters are: a = 9.86-10.01 Å, b = 14.12-14.34 Å, and c = 14.16-14.42 Å. An increase in Al (i.e. a decrease in R) is responsible for an increase in the b and c parameters but does not influence the a parameter which, in turn, is positively correlated with the monovalent cations (Galli and Loschi Ghittoni 1972).

Harmotome [K/(K+Ba) < 0.50] occurs primarily as an amygdaloidal zeolite in metalliferous veins and vugs of plutonic (pegmatite and gabbro) and volcanic (basalt and andesite) rocks (Kostov 1962, Cerny and Povondra 1965, Pokorny 1966, Black 1969, Hoffman et al. 1973, Rinaldi et al. 1974, Cerny et al. 1977, Tschernich and Wise 1982, Passaglia and Bertoldi 1983, Hansen 1990, Armbruster et al. 1991). It also occurs (two occurrences) as a diagenetic product in a lacustrine tuff and a basaltic volcaniclastic sandstone (Sheppard and Gude 1971, 1983). Amygdaloidal samples are associated with other zeolites (analcime, heulandite, laumontite, paulingite, erionite, and ferrierite), apophyllite, prehnite, pectolite, hyalophane, gibbsite, pyrite, galena, quartz, and calcite. R shows an average value of 0.71 and ranges from 0.68 in crystals from trachyandesite of Iskra, Bulgaria (Kostov 1962), to 0.76 in crystals from basalt of Weitendorf, Austria (Armbruster et al. 1991). The (K+Ba) content is close to or slightly higher than 2, and the K/(K+Ba) ratio averages 0.22 and ranges from 0.03 in crystals from Korsnas, Finland (Cerny et al. 1977), to 0.45 in crystals from Selva di Trissino, Italy (Passaglia and Bertoldi 1983). Na generally prevails over Ca [Na/(Na+Ca) = 0.64-1], although it is subordinate in crystals from Selva di Trissino and Chase Creek, Canada (Tschernich and Wise 1982). The only analyzed sedimentary sample is from Wikieup, Arizona, where it is associated with analcime, chabazite, clinoptilolite, and erionite (Sheppard and Gude 1971). R is 0.74, Ba slightly prevails over K [K/(K+Ba) = 0.45], Na prevails over Ca [Na/(Na+Ca) = 0.90], and the Mg content (0.51) is anomalously high for this zeolite species.

The unit-cell parameters are: a = 9.81-9.92 Å, b = 14.10-14.17 Å, and c = 14.26-14.34 Å. The correlations between unit-cell parameters and chemical composition found for phillipsites (Galli and Loschi Ghittoni 1972) also apply to harmotome (Cerny et al. 1977).

Structure Type: **ROG**

Roggianite: $Ca_{14}(Na,K)[Be_5Al_{15}Si_{28}O_{90}(OH)_{14}](OH)_2\cdot34H_2O$

Roggianite has been described in two adjacent occurrences from Val Vigezzo, Italy (Passaglia 1969, Vezzalini and Mattioli 1979), both in albitite dikes only rarely associated with thomsonite. The holotype sample from Alpe Rosso was tested later (Passaglia and Vezzalini 1988) for zeolitic behavior and was re-analyzed by electron microprobe and by atomic absorption spectrometry for Be, which was not determined for both the original occurrence and the second occurrence (Pizzo Marcio). The results indicate that roggianite is a Be-bearing zeolite with R [Si/(Si+Al+Be)] equal to 0.58 and Ca as the DEC. Structure refinements show an imperfect tetrahedral framework with some (OH)-vertices not shared by two tetrahedra ("interrupted framework") pl us some extraframework (OH) groups (Galli 1980) and one tetrahedral site occupied by Be (Giuseppetti et al. 1991). The structure is tetragonal $I4/mcm$ with a = 18.33-18.37 Å and c = 9.16-9.19 Å.

Structure Type: **STI**

Stilbite: $NaCa_4[Al_9Si_{27}O_{72}]\cdot30H_2O$

Stellerite: $Ca_4[Al_8Si_{28}O_{72}]\cdot28H_2O$ (Z = 2)

Barrerite: $Na_8[Al_8Si_{28}O_{72}]\cdot 26H_2O$ (Z = 2)

The tetrahedral framework of this group has an orthorhombic *Fmmm* TS (Galli and Gottardi 1966) and shows a minimum anionic charge corresponding to an Al content of 8 atoms (half of the unit-cell of stellerite and barrerite and the full unit cell of stilbite) that is generally neutralized by Ca(+Mg) atoms. Any Al increase is counterbalanced by Na(+K) atoms (Passaglia et al. 1978b). Where Al is very close to 8 and Na ≈ 0 (stellerite), the RS remains orthorhombic *Fmmm* because the framework geometry is unaltered and Ca, completely surrounded by water molecules, fully occupies the site on the mirror plane normal to *a* (Galli and Alberti 1975a). Where Al > 8 atoms and Na > 0 (stilbite), the RS is monoclinic *C*2/*m* because the framework is rotated clockwise around binary diads and Ca is pushed from the mirror plane by Na located in a site near the framework oxygens (Galli 1971). In the case of Al ≈ 8 and Na(+K) ≈ 8 (barrerite), the mineral is orthorhombic because the lower charge of Na (in comparison with Ca) and its distribution with a low occupancy in the extraframework sites of stellerite and stilbite allows the mirror plane normal to *a* to be maintained. The resulting RS (*Amma*) is lower than the RS of stellerite (*Fmmm*) because some Na atoms occupy a site that has no counterpart in stellerite and stilbite, and they force the framework to rotate around a screw diad parallel to *a* (Galli and Alberti 1975b). Artificially Na-exchanged stellerite has an average orthorhombic *Fmmm* symmetry due to the statistical distribution of Na on both sides of the (001) mirror plane (Passaglia and Sacerdoti 1982), whereas artificially Ca-exchanged barrerite has the expected *Fmmm* symmetry (Sacerdoti and Gomedi 1984). The strong positive correlations between Al and Na(+K) and between Na(+K) and the β angle allow consideration of stellerite (orthorhombic) and stilbite (monoclinic) as a continuous series in which the Ca(+Mg) content is nearly constant and the degree of monoclinicity (the β-angle) increases with Na(+K) and Al content (Passaglia et al. 1978b). Anomalous Na-rich and Ca-poor stilbites were classified as intermediate members between stilbite and barrerite (Passaglia et al.,1978b), and their monoclinicity, markedly lower than expected, was explained on the basis of the structural features (Quartieri and Vezzalini 1987). As a result of the large dependence of the β-angle on chemistry, many samples are obviously heterogeneous, being composed of a number of crystals with different β-angles (Passaglia et al. 1978b, Akizuki and Konno 1985), and other crystals show an association of stilbite and stellerite (Morad et al. 1989). Thus, a reliable classification should be done through an accurate X-ray diffraction analysis inasmuch as the chemical composition and morphology do not precisely define these species.

Stilbite is the most common among the three zeolites and has been described in vugs of plutonic (granite and quartz monzonite), volcanic (basalt, andesite, dacite, and porphyrite), and metamorphic (gneiss, mica schist, and meta-ophiolite) rocks and as sandstone cement (Ueno and Hanada 1982). It occurs in association with other zeolites (heulandite, laumontite, chabazite, mordenite, mesolite, analcime, scolecite, erionite, levyne, natrolite, thomsonite, and stellerite), apophyllite, datolite, prehnite, pectolite, calcite, pyrite, quartz, and opal. Many reliable chemical compositions (if not otherwise quoted in Passaglia et al. 1978b) show that R is in the range 0.73-0.78, very close to the mean value of 0.75, being significantly lower (0.71) only in crystals from Montresta, Italy. Ca(+Mg) contents are commonly very close to 4 but are lower (1-2) in Na-rich crystals from rocks weathered in a foreshore environment at Capo Pula, Italy, Faroer Islands, Kii Peninsula, Japan (Harada and Tomita 1967), Phillip Island, Australia (Birch 1988), Tsuyazaki, Japan (Ueno and Hanada 1982), and Rocky Pass, Alaska (Di Renzo and Gabelica 1997). In samples with Ca(+Mg) close to 4, the Na(+K) content uniformly ranges from ~0.40 in crystals from Oravita, Romania, to 1.75 in crystals from Flodigarry, Sky. A value as high as 2.72 has been found in crystals from Montresta, Italy. Na greatly prevails over K [Na/(Na+K) = 0.70-0.98], except for samples from granite gneiss of some Swiss localities and from Prascorsano, Italy; Mg is negligible or very subordinate, and the only relatively high Mg content (1.13) has been found in a sample from the granite of the Siljank ring structure, Sweden (Morad et al. 1989). Sr and Ba are typically absent or negligible, and Sr is appreciable (0.31) only in a sample from the Siljank ring structure, Sweden. The unit-cell parameters of the pseudo-orthorhombic $F2/m$ cell are a = 13.59-13.66 Å, b = 18.18-18.33 Å, c = 17.71-17.84 Å and β = 90.20-91.15° .

Stellerite is less common than stilbite, and it has been described in vugs of the same rock types (granite, granodiorite, dolerite, basalt, andesite, diabase, gneiss, hornfel, and metadolerite) in association with other zeolites (heulandite, epistilbite, chabazite, laumontite, and stilbite), fluorite, prehnite, babingtonite, sphene, fluorapophyllite, calcite, quartz, tridymite, and native copper (Galli and Passaglia 1973, Passaglia et al. 1978b, Alberti et al. 1978, Birch 1989, Morad et al. 1989). It has also been described in fractures in devitrified, non-zeolitic rhyolitic tuffs from Yucca Mountain, Nevada (Carlos et al. 1995). R varies (0.76-0.78) little from the average value (0.77), being slightly lower (0.75) in some samples. Ca(+Mg) is generally very close to 4 with Mg negligible (max. 0.17). Na(+K) ranges from 0.02 in crystals from the metadolerite of Corop, Australia (Birch 1989), to 0.40, but it is higher (0.40-0.60) in samples with Ca(+Mg) slightly lower than 4, and it is anomalously high (1.74) in crystals with Ca(+Mg) equal to 3.46 from andesite of Capo Santa Vittoria, Italy (Passaglia et al. 1978b). K is very subordinate, and Sr and Ba are absent. The unit-cell parameters are: a = 13.57-13.63 Å, b = 18.16-18.27 Å, c = 17.82-17.87 Å, and β = 90° or with statistically insignificant deviations from orthogonality.

Barrerite has been described in vugs of altered andesite that crops out in the foreshore of Capo Pula, Italy, associated with heulandite (Passaglia and Pongiluppi 1974, 1975), and in fractures of basalt exposed in the tidal area of Rocky Pass, Alaska, associated with Na-rich stilbite and epistilbite (Di Renzo and Gabelica 1997, Sacerdoti et al. 1999). R is 0.77-0.78, and the extraframework contents are different from that given in the schematic formula showing Na = 3.92-5.45, K = 1.06-1.73, Ca = 0.84-1.12, and Mg = 0.11-0.18. The unit-cell parameters are within the ranges given for stellerite, being a = 13.59-13.64 Å, b = 18.18-18.20 Å, and c = 17.79-17.84 Å. The X-ray powder pattern does not show any peaks indexable in *Amma* (barrerite) but forbidden in *Fmmm* (stellerite), and, hence, the distinction between the two zeolite species requires careful X-ray single-crystal analyses.

Structure Type: **TER**

Terranovaite: $Na_4Ca_4[Al_{12}Si_{68}O_{160}]\cdot{>}29H_2O$

Terranovaite occurs as transparent, small spheres closely associated with heulandite or as globular aggregates of small prismatic crystals associated with tschernichite in vugs of dolerite at Mt. Adamson, Antarctica (Galli et al. 1997). The empirical formula, $Na_{4.2}Ca_{3.7}K_{0.2}Mg_{0.2}[Al_{12.3}Si_{67.7}O_{160}]\cdot{>}29H_2O$, has a high R value (0.85). Na and Ca are the DEC (with Na slightly prevailing over Ca), and K and Mg are very low. The structure is orthorhombic *C2cm* with a = 9.75 Å, b = 23.88 Å, and c = 20.07 Å and *Cmcm* TS.

Structure Type: **THO**

Thomsonite: $Ca_7Na_5[Al_{19}Si_{21}O_{80}]\cdot 24H_2O$

Thomsonite has been described from many localities where it occurs in vugs of igneous rocks (basalt, andesite, dolerite, and nepheline syenite). It occurs rarely in metamorphic (meta-ophiolite and gneiss) rocks and as a late-stage hydrothermal alteration of lazurite (Hogarth and Griffin 1980). It is associated with other zeolites (analcime, mesolite, laumontite, gonnardite, edingtonite, harmotome, chabazite, phillipsite, gismondine, and natrolite), calcite, aragonite, celestine, pectolite, vesuvianite, and epidote. The schematic formula calculated from reliable chemical analyses in the literature differs from the one which is normally reported, $Ca_8Na_4[Al_{20}Si_{20}O_{80}]\cdot24H_2O$, and it is slightly richer in Si and Na and poorer in Ca. The mean R value is 0.53 (instead of 0.50), and the mean contents of Na and Ca are 4.5 (instead of 4) and 6.8 (instead of 8), respectively. The chemical composition is variable: R ranges from 0.50 in a few samples to 0.58 in a sample from Hills Port, Antrim, Ireland (Nawaz 1988); the Na content varies from 3.02 in a sample from Procida, Italy (de'Gennaro et al. 1977) to 7.24 in a sample from Mazé, Japan (Harada et al. 1969), and the Ca content ranges from 8.43 in a sample from Shinshiro, Japan (Matsubara et al. 1979) to 3.55 in a sample from Tyamyr, Russia (Yefimov et al. 1966). Samples with chemical compositions close to the schematic formula reported in the literature do exist but are very rare. Ca generally prevails over Na, but samples with Na > Ca have been described from Mazé and Iragawa, Japan (Harada et al. 1969). Samples with comparable Ca and Na have been described from Howitt Plains, Australia (Birch 1988), Table Mountain, Colorado (Kile and Modreski 1988), and Taymyr, Russia (Yefimov et al. 1966). Where present, K and Mg are very low, but a sample in nepheline syenite at Taymyr, Russia (Yefimov et al. 1966) is K and Mg rich (1.54 and 0.83, respectively), and a sample from Ile de la Réunion (Nativel 1972) is the most Mg rich (0.84) known. Where detected, Sr can be appreciable and reaches values as high as 2.12 in the sample associated with brewsterite at Yellow Lake, Canada (Wise and Tschernich 1978a), 2.18 in a sample from Honshu, Japan (Ross et al. 1992), and 3.15 in the K- and Mg-rich and Ca-poor sample from Taymyr, Russia. Ba is absent or negligible.

The different habits and forms (blocky, complex crystals, bladed crystals, botryoidal growths, and waxy balls) of thomsonite have been correlated with the Si content (Wise and Tschernich 1978a). The geometry of the unit cell ($a < b < c$) and optical orientation ($\alpha = b$, $\beta = c$, and $\gamma = a$) have been proposed by Nawaz and Malone (1981). The structure is orthorhombic *Pncn* with a = 13.00-13.18 Å, b = 13.04-13.16 Å, and c = 13.09-13.24 Å. The (Si,Al) distribution in the tetrahedra is ordered in samples with Si/Al ≈ 1 and partially disordered in samples with Si/Al > 1 (Alberti et al. 1981, Pechar 1982, Pluth et al. 1985).

Structure Type: **YUG**

Yugawaralite: $Ca_2[Al_4Si_{12}O_{32}]\cdot8H_2O$

Yugawaralite has been described from six occurrences in fractures and vugs of volcanic breccia (Wise 1978), effusive rocks (basalt, andesite, trachyandesite, and rhyolite), and pyroclastites (andesitic tuff), all highly altered in active (Seki and Haramura 1966, Bargar and Beeson 1981, Kvick et al. 1986) and possible (Eberlein et al. 1971, Pongiluppi 1977) geothermal areas. It is typically associated with laumontite, calcite, and quartz, but some associations with stilbite, heulandite, stellerite, wairakite, and, rarely, mordenite have also been observed.

The composition is nearly constant. R ranges from 0.74 to 0.76, Ca is the dominant and almost exclusive extraframework cation, Na, K, and Mg are negligible, and Sr and Ba are absent. A sample from a drill hole at Yellowstone National Park (Bargar and Beeson 1981) is unusually poor in Si (R = 0.67), rich in Na (0.17), and it is the only sample documented to occur in association with analcime and dachiardite.

The structure is monoclinic *Pc* (Kvick et al. 1986) with a = 6.70-6.73 Å, b = 13.97-14.01 Å, c = 10.04-10.09 Å, and β = 111.07-111.20°. Optical properties and orientation were given by Harada et al. (1968) and Akizuki (1987).

Structure Type: **Unknown**

Cowlesite: $Ca_{5.5}(Na,K)[Al_{12}Si_{18}O_{60}]\cdot36H_2O$

Cowlesite has been described from about 20 localities mainly in County Antrim, Ireland (Nawaz 1984, Vezzalini et al. 1992), but also in Australia (Birch 1989), USA, Canada (Wise and Tschernich 1975, Kile and Modreski 1988), and Japan (Fujimoto et al. 1990, Kuwano and Tokumaru 1993). It occurs invariably in vugs of basalt, alone or associated with other zeolites (levyne, erionite-offretite, phillipsite, thomsonite, garronite, chabazite, scolecite, and gismondine). The crystals are very small ($<0.1 \times 0.002$ mm) so that single-crystal studies have failed to date. Such crystals appear to be good candidates for structure study with synchrotron X-rays.

The composition is very close to the schematic formula. R shows a mean value of 0.61 and generally ranges from 0.60 to 0.62 but it is slightly lower (0.58) in samples from Monte Lake, Canada, and Spray, Oregon (Wise and Tschernich 1975). Ca is the DEC, and Na and K are consistently present in subordinate amounts (<1.0) even though Na reaches noticeable values in samples from Flinders, Australia (1.13), Dunseverick, County Antrim (1.31), and Iki Island, Japan (1.86). The sample from the latter occurrence is also richer in K and Mg (0.24 and 0.60, respectively) and is the only one associated with gobbinsite (Kuwano and Tokumaru 1993). The symmetry, derived from powder data and from very faint precession photographs, is orthorhombic (Nawaz 1984) with a = 23.21-23.30 Å, b = 30.60-30.68 Å, and c = 24.98-25.04 Å.

Structure Type: **Not yet assigned**

Tschörtnerite : $Ca_6Cu_3SrK(OH)_9[Al_{12}Si_{12}O_{48}]\cdot 14H_2O$

Tschörtnerite has been found in cavities of a Ca-rich xenolith in leucite tephrite lava at the Bellberg volcano near Mayen, Eifel, Germany (Effenberger et al. 1998). It occurs as light blue, transparent cubes (maximum size of 0.15 mm) in association with chalcopyrite, cuprite, willhendersonite, phillipsite, gismondine, strätlingite, and bellbergite. The empirical formula $(Ca_{5.60}Cu_{2.90}Sr_{1.04}K_{0.70}Ba_{0.30})(OH)_{8.44}[Fe_{0.09}Al_{11.85}Si_{12.06}O_{48}]\cdot 14.01H_2O$ shows an R value very close to 0.50, with Ca as the DEC, and noticeable amounts of Cu and extraframework (OH) groups. The structure is cubic $Fm\bar{3}m$ with a = 31.62 Å and contains a new extremely large cage with 96 tetrahedra and a Cu(OH)-bearing cluster.

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