

POROSITY EVOLUTION AND MINERAL PARAGENESIS DURING LOW-GRADE METAMORPHISM OF BASALTIC LAVAS AT TEIGARHORN, EASTERN ICELAND

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ABSTRACT. Low-grade alteration of basaltic lavas at Teigarhorn, eastern Iceland, resulted in three distinct stages of mineral paragenesis that correlate to events in the burial and intrusive history of Icelandic crust. Metasomatism and brittle deformation during the paragenetic stages dramatically affected the paleo-hydrology of the lavas and formed temporally distinct mineral assemblages. The lavas initially contained up to 22 percent total porosity concentrated near the tops and bottoms of individual lava flows. Celadonite and silica (Stage I) precipitated along the walls of primary pores prior to deep burial of the lavas and occluded ~8 percent of the initial porosity. During burial (Stage II), hydrolysis of olivine and basaltic glass led to the formation of mixed-layer chlorite/smectite clays in the matrix of the lavas and as rims filling ~40 percent of the volume of primary pores. Chlorite contents in Stage II mixed-layer clay rims increased from ~20 to ~80 percent during the infilling of individual vesicles, reflecting increasing temperatures with time as the lavas were buried. The end of Stage II occurred after burial and is represented by filling of ~40 percent of total primary porosity by zeolites (scolecite or heulandite + stilbite + mordenite \pm epistilbite) and replacement of plagioclase by zeolites and albite. The Stage II zeolite assemblages are indicative of two regional metamorphic mineral zones in eastern Iceland, the mesolite + scolecite and heulandite + stilbite zones. The presence of the boundary between the mesolite + scolecite and heulandite + stilbite zones indicates that the maximum temperature during burial metamorphism was $90^{\circ} \pm 10^{\circ}\text{C}$. Localized areas of intense hydrothermal alteration associated with intrusion of basaltic dikes (Stage III) overprint Stages I and II. Extensive fracturing and hydrothermal brecciation during Stage III added 3 to 11 percent total porosity in which mm- to cm-scale museum-grade crystals of quartz, calcite (Iceland spar), stilbite, scolecite, heulandite, and laumontite precipitated. Estimates of the temperature during Stage III (based on fluid inclusion homogenization temperatures in calcite, chlorite geothermometry, and the zeolite assemblage) range from 120° to 180°C . Although the thermobarometric conditions during Stages II and III led to similar mineral assemblages, careful attention to textural and geologic relations permits seemingly complex, multi-stage parageneses in metabasalts to be interpreted in a petrotectonic context.

INTRODUCTION

The mineralogy of low-grade metabasalts is a sensitive indicator of the thermobarometric evolution of oceanic crust and continental flood basalts. Discrete temperature-, pressure-, and/or depth-controlled zones characterized by assemblages of silica minerals, phyllosilicates, and zeolites frequently serve as metamorphic indicators in low-grade metabasalts. Zones containing trioctahedral smectite, corrensite and/or mixed layer chlorite/smectite, and chlorite occur with increasing metamorphic grade in zeolite- and greenschist-facies metabasalts and active geothermal systems in basaltic rocks (Mehegan, Robinson, and Delany, 1982; Seki and others, 1983; Liou and others, 1985; Bevins, Robinson, and Rowbotham, 1991; Robinson, Bevins, and Rowbotham, 1993; Schmidt, 1990; Schmidt and Robinson, 1997; Kristmannsdóttir, 1979; Schiffman and Fridleifsson, 1991; Robinson and Bevins, 1999; Alt, 1999). In addition, as many as five separate depth-controlled "zeolite zones" have been described in regionally metamorphosed basaltic lavas and Icelandic geothermal systems (Walker, 1951, 1960a,b; Sukheswala,

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Avasia, and Gangopadhyay, 1974; Kristmannsdóttir and Tómasson, 1978; Jørgensen, 1984; Murata, Formoso, and Roisenberg, 1987; Schmidt, 1990; Neuhoff and others, 1997; Christiansen and others, 1999). Phyllosilicate and zeolite parageneses thus provide useful information for evaluating heat flow and structural evolution in large igneous provinces (Neuhoff and others, 1997) and exploration potential in fractured basaltic petroleum reservoirs (Christiansen and others, 1999). Complicating such interpretations, however, is the fact that basaltic provinces often undergo multiple stages of intrusion, extrusion, and deformation during which the lavas are altered through weathering, regional burial metamorphism, and local hydrothermal/contact metamorphism. Mineral assemblages formed during regional metamorphism and localized hydrothermal alteration can be nearly identical, complicating interpretation of mineral isograds associated with burial metamorphism (Neuhoff and others, 1997; Sukheswala, Avasia, and Gangopadhyay, 1974).

In the present study, we demonstrate that regional and local mineral parageneses in low-grade metabasalts can be distinguished on the basis of geologic and textural relations. These relations are difficult to establish from drillcores and areas of limited outcrop. Ideally one would like to investigate the textural and geologic relations between mineral parageneses in laterally continuous, unweathered outcrop exposures. Remarkable outcrop exposures created through coastal erosion at Teigarhorn, eastern Iceland (fig. 1), permit such detailed observations of geologic and textural relationships in basaltic lavas affected by burial metamorphism, brittle deformation, and hydrothermal alteration during dike emplacement. Geologic, petrographic, mineral chemical, and hydrologic information based on field and laboratory studies are presented in order to distinguish mineral assemblages and textures associated with three paragenetic stages of alteration at Teigarhorn: syn-eruptive, near surface alteration (Stage I), regional burial metamorphism (Stage II), and local hydrothermal alteration associated with dike emplacement (Stage III). Each stage is characterized by distinct mineral parageneses and textures, with Stage II resulting in formation of the regional zeolite zones described by Walker (1960b). Bulk porosity varied considerably during the metamorphic history of the lavas due to filling of pore spaces by secondary minerals and generation of secondary porosity through brittle deformation. The results provide an example of how observations of mineral paragenesis and porosity evolution can be integrated in a petrotectonic interpretation of low-grade metamorphism in basaltic terraines.

GEOLOGICAL SETTING

Geology.—Teigarhorn is located near the eastern edge of the volcanic plateau of present-day Iceland (fig. 1). Lavas erupted from fissures and central volcanoes during magmatism along a divergent plate boundary between the North American and Eurasian plates (Kristjánsson, Gudmundsson, and Haraldsson, 1995). The oldest exposed lavas are only about 13 Ma (Watkins and Walker, 1977). Upper crustal stratigraphy in eastern Iceland consists largely of subaerial basaltic lavas with minor intercalated sediments, silicic lavas, lignite layers, and pyroclastic units (Robinson and others, 1982; Gústafsson, 1992; Walker, 1960b; Wood, 1977; Sæmundsson, 1979). Continued volcanism and rifting led to pronounced tilting of the lavas that dominate crustal structure around Teigarhorn (Pálmason, 1973); dips increase from $<2^\circ$ to 6° southwest at the top of the lava pile to $\sim 6^\circ$ to 9° southwest at sealevel (fig. 1; Walker, 1974).

Disrupting the monoclinical structure of the lava pile are mafic and silicic intrusive centers and central volcanic complexes (fig. 1; Walker, 1963). Extrusive products of the extinct central volcanoes are interlayered with fissure-erupted lavas that make up most of the crust in eastern Iceland. Glacial exhumation has revealed an alignment of mafic dike swarms, central volcanoes, and intrusive centers that defines the geometry of paleo-rift zones. Teigarhorn lies within a prominent dike swarm (called the Álfafjörður dike

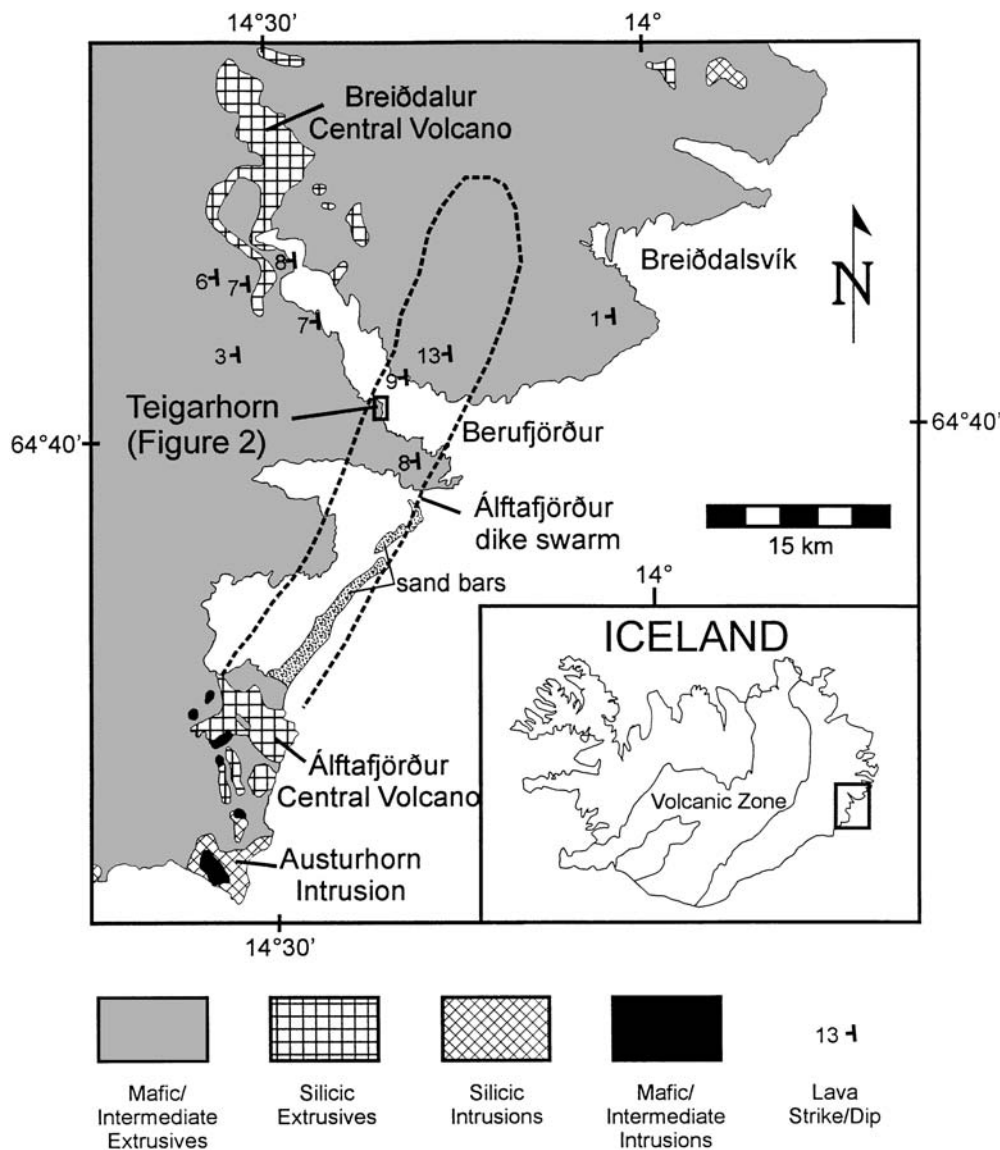


Fig. 1. Generalized geological map of a portion of eastern Iceland (after Jóhannesson and Sæmundsson, 1989) showing the distribution of major rock types, locations of intrusions and central volcanoes, and selected lava orientations from Walker (1963, 1974). Dashed curve outlines the Álftafjörður mafic dike swarm where dikes make up greater than 8 percent (by volume) of the crust (Walker, 1963).

swarm; Walker, 1963): the Álftafjörður volcanic center and the Austurhorn intrusion both lie within this swarm (figs. 1 and 2; Walker, 1963; Blake, 1970). The composition and mineralogy of these dikes are similar to the surrounding basalts discussed below (Walker, 1963). The dikes are subvertical, trend north-northeast (fig. 2), and vary in thickness from 0.25 to 3 m.

Lithology.—The volcanic stratigraphy exposed at Teigarhorn includes approx 150 m of subaerial mafic lava flows with minor intercalated pyroclastic units. The uppermost

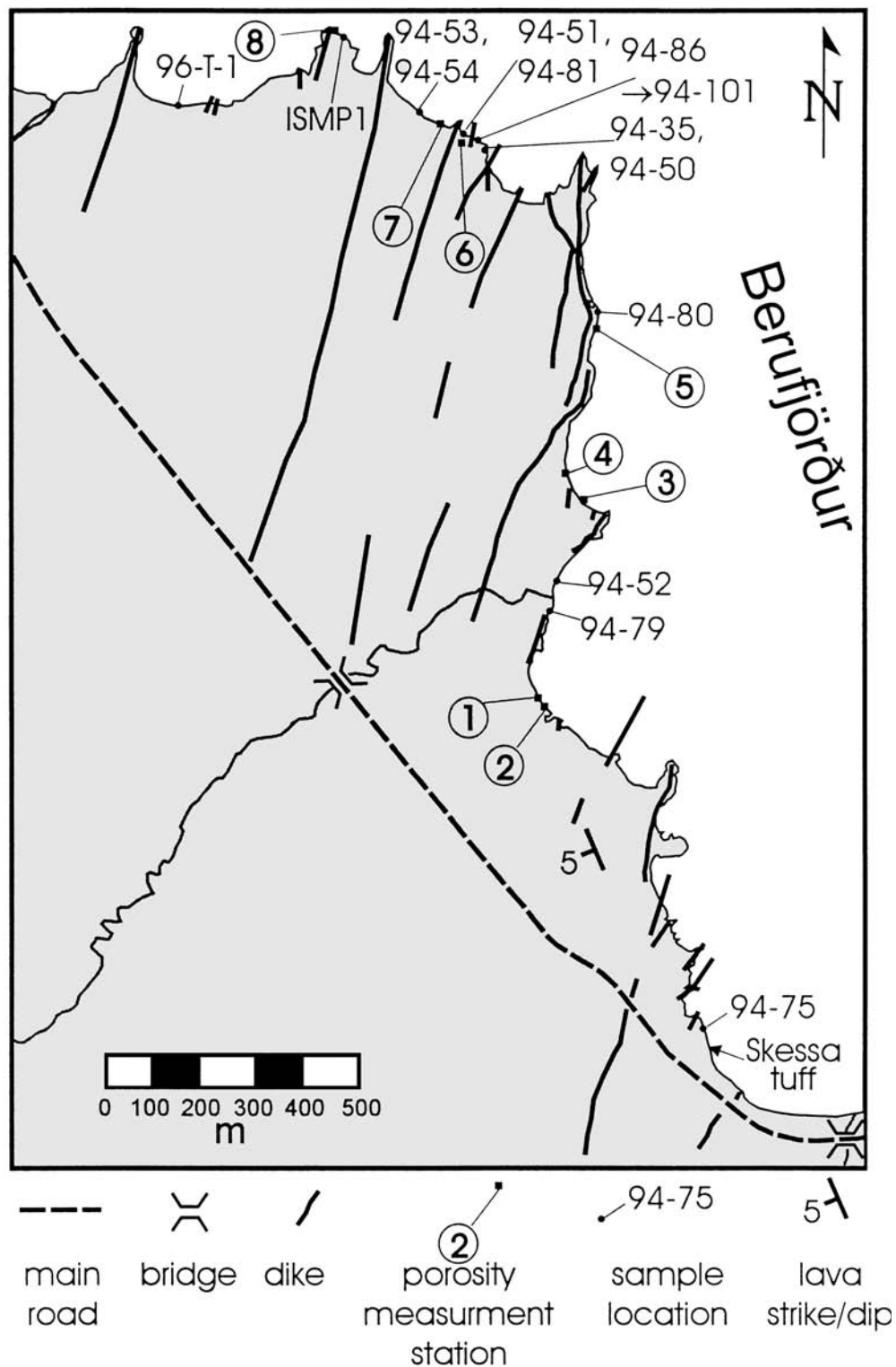


Fig. 2. Map of Teigarhorn showing the distribution of individual mafic dikes (determined by field mapping and from aerial photographs) and the locations of samples and porosity measurements. Also shown is the location of the Skessa tuff (Walker, 1963).

lavas belong to the Grænavatn Porphyritic Group (Walker, 1959), and the oldest units are the Skessa tuff (see below) and surrounding undesignated lavas. These units were buried to approx 1500m at Teigarhorn (Walker, 1960b). Compositionally, the lavas exposed at Teigarhorn (fig. 2) are 3 to 25 m thick subaerial low-MgO tholeiites and ferrobasalts (Flower and others, 1982). Most flows are fine-grained, containing abundant groundmass plagioclase, olivine, pyroxene, and glass with minor olivine and pyroxene phenocrysts. Lavas with up to 30 modal percent plagioclase phenocrysts occur infrequently. The flows have scoraceous, brecciated flow tops. Vesicles are abundant near the top and bottom of the flows, but the flow centers are essentially non-vesicular and massive. A 20 m thick, green, welded, silicic tuffaceous unit (the Skessa tuff of Walker, 1959) is exposed near the southern portion of Teigarhorn (marked "Skessa tuff" on fig. 2). The Skessa tuff is a regionally extensive unit that appears to be the product of an eruption from an unexposed volcanic center approx 25 km north of Teigarhorn (Walker, 1962). Separating many flows are undulatory weathering surfaces that frequently are overlain by bright red basaltic tephra that are 1 to 15 cm thick and constitute less than 1 percent of the stratigraphy. The tephra are phreatomagmatic deposits produced by interaction between basaltic magmas and surface or groundwaters at eruption sites 10's to 100's of km away from Teigarhorn (Heister, ms).

The volcanic stratigraphy in eastern Iceland has been subjected to regional zeolite-facies metamorphism; higher grades of metamorphism are exposed around intrusive complexes and central volcanoes (Walker, 1960b). Walker (1960b) described depth-controlled zeolite-facies mineral zones (fig. 3) that parallel the paleosurface of the lava pile and transgress the volcanic stratigraphy. This implies that the zones formed after tilting of the lava pile and do not solely reflect variations in chemistry of the host lavas. The zeolite zones described by Walker (1960b) are underlain by zones characterized by stilbite \pm heulandite \pm mordenite, laumontite, and prehnite + epidote with increasing depth (Mehegan, Robinson, and Delany, 1982). Coastal exposures around Berufjörður (fig. 1) have provided museum-grade zeolite specimens for over 200 yrs (Betz, 1981). Teigarhorn, in particular, has been an important source for zeolites used in studies of crystal chemistry (Galli and Rinaldi, 1974; Passaglia, 1975; Passaglia and others, 1978; Alberti, Pongiluppi, and Vezzadini, 1982), crystal structures (Slaughter and Kane, 1969), reaction kinetics (Ragnarsdóttir, 1993), optical properties (Bauer and Hřichová, 1962), and piezoelectricity (Bellanca, 1940).

METHODS

Mapping and sample collection.—Representative samples of altered and unaltered lavas were collected throughout the coastal exposures at Teigarhorn in conjunction with mapping of dike distribution and outcrop porosity measurements (fig. 2). Estimates of total macroscopic porosity (herein defined as the percentage of a given volume of rock occupied by pore space) prior to mineral precipitation were conducted in situ during mapping following the methods of Manning and Bird (1991). In the present communication, a distinction is made between the total porosity at a given point in time (Φ) and the cumulative porosity that reflects pore abundance (Φ^*) but does not include the effects of pore occlusion by mineral precipitation (Manning and Bird, 1991). The contribution of primary pores to Φ^* (Φ_1^*) was estimated by placing a clear mylar sheet imprinted with a 54 by 41.5 cm grid (node spacing 1.5 cm) directly on the outcrop as a basis for point counting macroscopic pores. Estimates of Φ_1^* included vesicles and other primary structures as well as open space between flow units that could not texturally be linked to post-eruption processes. Although this technique only identifies macroscopic pores on the order of 1 mm or larger, the contribution of these pores to Φ_1^* is large, and uncertainties are on the order of 1.5 percent or less for grids of over 1000 points (Manning and Bird, 1991; Chayes, 1956). The contribution of secondary pore spaces to

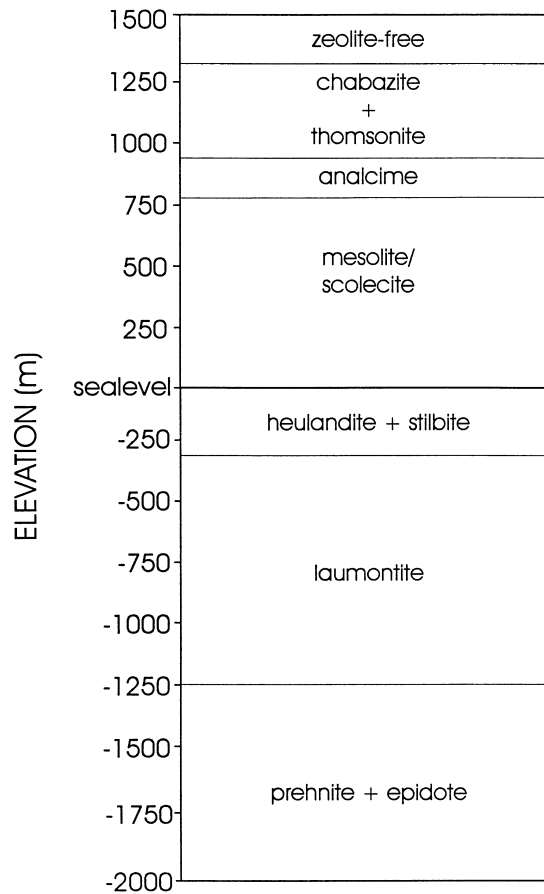


Fig. 3. Depth distribution of regional zeolite facies mineral zones in eastern Iceland. Zones above sealevel are from mapping in olivine normative tholeiites around Berufjörður by Walker (1960b) and below sealevel are from studies of the Reyðarfjörður drill core located ~40 km north-northwest of Teigarhorn (Mehegan, Robinson, and Delaney, 1982). Note that the outcrops at Teigarhorn are approximately at sealevel. Stage II alteration at Teigarhorn includes both the mesolite/scolecite and heulandite + stilbite mineral zones.

Φ^* (Φ_2^*) was estimated by measuring cross-sectional areas of fractures/veins, breccias, and dissolution features associated with late-stage alteration within 3 to 6 m² sections of outcrop. Although outcrop area measurements of pore space are scale dependent and not necessarily representative of bulk properties (Manning and Bird, 1991), they are useful for assessing the general magnitude and variability of Φ_2^* . Repeated area measurements indicate that uncertainties were ~1 percent.

Analytical techniques.—Petrographic and hand-sample observations were conducted on 20 samples representative of lithologies and mineral parageneses at Teigarhorn. Optical properties of minerals in transmitted light were used along with mineral habits in both hand specimen and thin section to augment field observations of mineral parageneses and phase relations. Estimates of the percentage of pore space occupied by various alteration minerals were conducted by digital analysis of photomicrographs. Images of discrete pores were digitized using the software package Scion Image (Scion Corporation, Frederick, Maryland) to calculate the areas of various secondary phase/assemblages and residual porosity relative to the total area of the pore. Repeat measurements indicate that errors in area measurements were less than 1 percent.

A battery of analytical techniques was used to confirm phase identities and compositions. Powder X-ray Diffraction (XRD) was conducted on mineral separates using a Rigaku theta-theta diffractometer with CuK α operating at 35 kV and 15 mA. Mineral separates for XRD identification were extracted from individual vesicles with a rotary drill. Mineral chemistry was determined by electron probe microanalysis (EPMA) on an automated JEOL 733A electron microprobe operated at 15 kV accelerating potential and 15 nA beam current. Calibration was conducted using natural geologic standards. Beam width for analysis of hydrous minerals (i.e., that is, clays and zeolites) was varied between 10 and 30 μ m depending on grain size. Raw counts were collected for 20 s and converted to oxide wt percents using the CITZAF correction procedure after accounting for unanalyzed oxygen following the methods of Tingle and others (1996). Raw counts for individual elements (including Na) were essentially constant with time, suggesting the elemental drift (loss) was negligible. Bulk rock compositions were determined on a Rigaku fully automated X-ray fluorescence (XRF) spectrometer. Fluid inclusion homogenization and freezing temperatures were determined on one sample of calcite (Iceland spar variety) using gas-flow heating/freezing stage adapted by Fluid, Inc.

RESULTS

Mineral Paragenesis

Alteration at Teigarhorn occurred during three paragenetic stages. The textural and mineralogical characteristics of these stages are summarized in table 1 and the sequence of mineral precipitation during each stage is shown in figure 4. Representative compositions and molar formulas for clays, zeolites, and feldspars are given in tables 2 through 5.

Stage I.—Paragenetic stage I is the earliest alteration at Teigarhorn and is characterized by linings of primary pore space by celadonite and silica minerals (table 1; figs. 4 and 5B). All other mineral assemblages are either precipitated on top of or cross cut Stage I alteration (fig. 5B). Stage I alteration is common but not present in all samples. Celadonite and silica minerals typically form layers along the walls of pore space with a layered or botryoidal habit. Occasionally Stage I silica forms layered geopetal structures

TABLE 1
Mineralogical and textural characteristics of paragenetic stages observed at Teigarhorn, eastern Iceland

Paragenetic Stage*	Mineral Assemblages in Pore Space	Mineral Assemblages in Matrix	Textures
Stage I	\pm celadonite \pm silica	minor celadonite	pores: lining primary pore space; geopetal structures matrix: minor celadonite replacing groundmass
Stage II	C/S ² + scolecite \pm chalcedony \pm quartz C/S + stilbite + heulandite \pm mordenite \pm epistilbite \pm quartz \pm chalcedony	C/S + albite + zeolites	pores: C/S pore linings partial or complete primary pore fillings by zeolites matrix: pseudomorphic replacement of groundmass and phenocrysts
Stage III	quartz \pm clinoptilolite \pm stilbite \pm heulandite \pm mordenite \pm scolecite \pm laumontite \pm calcite	quartz + C/S	pores: brecciation, fracturing, mineral precipitation matrix: discoloration (SiO ₂ metasomatism)

*Stage I was the earliest stage, Stage III the last. ²Mixed-layer chlorite smectite (see text).

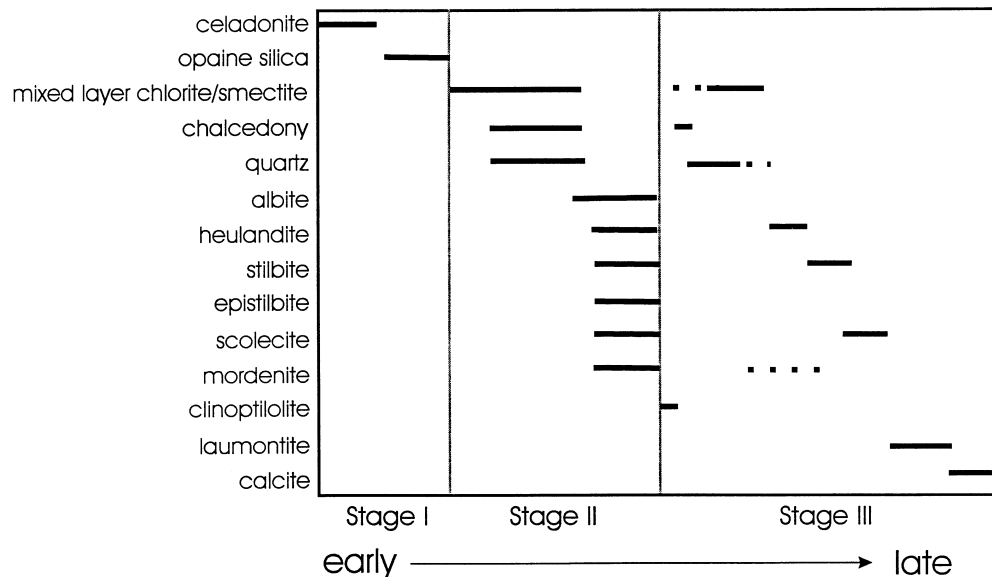


Fig. 4. Temporal sequence of secondary minerals developed in lavas at Teigarhorn. Events toward the right of the diagram occurred after events to the left.

on the bottom of vesicles that are oriented roughly parallel to the lava stratigraphy. Minor celadonite occurs in the volcanic groundmass.

Celadonite typically precedes silica in the crystallization sequence (fig. 5B). A representative celadonite composition from Teigarhorn is given in table 2, and 6 celadonite analyses are plotted in figure 6. Stage I celadonite is close to the endmember compositions as defined by Buckley and others (1978), with only slightly lower potassium content and higher aluminum than the ideal endmember composition $[K_2(Fe,Al)_2(Mg,Fe)_2Si_8O_{20}(OH)_4]$; table 2; fig. 6]. The slight deviation from ideal composition may be due to the presence of a small trioctahedral smectite component (either as intergrowths or interstratifications) in the celadonites, although smectite is not detected in the XRD patterns. Celadonite is distinguished from other phyllosilicates by its distinctive bright green color in both hand specimen and thin section (figs. 5B and 5D), its layered habit, and by its potassium-rich composition (table 2; fig. 6). Silica minerals consist of quartz and/or chalcedony. It appears likely that quartz and chalcedony replaced preexisting Stage I opaline silica during burial, as opaline silica is the only Stage I silica phase observed near the top of the lava pile above Teigarhorn (Neuhoff, unpublished data).

Stage II.—Paragenetic Stage II alteration is characterized by continued infilling of primary pore spaces and partial replacement of groundmass and phenocryst minerals. As with Stage I, the distribution at the outcrop scale of Stage II alteration is largely a function of the distribution of primary porosity in the lavas (fig. 5A). Stage II mineral assemblages and paragenetic sequences are summarized in table 1 and figure 4. Mixed layer chlorite/smectite (C/S) is the first phase to precipitate in primary pore space during Stage II (fig. 5B, C). The term “mixed layer chlorite/smectite” and the abbreviation C/S are used in the present communication without specific inference of ordering among layers (Reynolds, 1988). Brown to brownish green radiating bundles of C/S for linings of primary pore space. The bundles nucleated on pore walls or on Stage I minerals where present and developed toward the center of the pores. As shown in figure 6, Stage II C/S

compositions (table 2) project near the binary compositional join between ferromagnesian trioctahedral smectite (saponite-ferrosaponite; Güven, 1988) to ferromagnesian chlorite (clinocllore-chamosite; Bailey, 1988). The percentage of chlorite (or smectite) interlayers was estimated from electron microprobe analysis of the total molar Si + Al + Mg + Fe content of the mineral by linear interpolation between smectite and chlorite endmembers (Schiffman and Fridleifsson, 1991). This interpolation requires calculation of molar formulas based on 28 oxygen charge equivalents (O + 2OH), for which the molar Si + Al + Mg + Fe contents of endmember chlorite and trioctahedral smectite are 20 and 17.82, respectively. By this analysis, Stage II clays contain between 12 and 85 mol percent chlorite. The most prominent interlayer cations in C/S clays are Ca^{+2} and Na^{+} (table 2). Potassium typically accounts for less than 5 mol percent of the total interlayer cations but in rare cases constitutes more than 25 mol percent with one sample as high as 60 mol percent. Mixed-layer chlorite-smectite with high K^{+} contents always occur in close proximity to celadonite, and textural evidence suggests that local replacement of Stage I celadonite by Stage II C/S may have occurred.

The compositions of C/S clays become progressively enriched in chlorite during Stage II alteration. Figure 7 shows the percentage of chlorite in a rim of Stage II C/S as a function of distance from the vesicle wall. In general, C/S near the wall of the vesicle precipitated earlier than that closer to the center of the vesicle. It can be seen in figure 7 that the chlorite content of the clay generally increases from ~20 percent at the vesicle wall to about 80 percent near the center of the vesicle, indicating that the chlorite content of Stage II C/S increased with time.

The last phases to crystallize during Stage II are minor quartz and chalcedony along with zeolites (table 1 and fig. 4) that serve as index minerals for the regional zeolite zones shown in figure 3. Zeolites partially or completely fill the pore space remaining after precipitation of C/S (fig. 5B). As indicated in table 1, two Stage II zeolite assemblages occur at Teigarhorn; the mesolite + scolecite zone described by Walker (1960b; fig. 3) is underlain by a zone characterized by heulandite + stilbite with occasional mordenite and epistilbite. The lower zone is similar to zones characterized by heulandite and/or stilbite identified in Icelandic geothermal systems (Kristmannsdóttir and Tómasson, 1978), East Greenland (Neuhoff and others, 1997), the Faeroe Islands (Jørgensen, 1984), the North Shore Volcanic Group (Minnesota, USA; Schmidt, 1990), and the Reydarfjörður drillhole (Mehegan, Robinson, and Delaney, 1982).

Mesolite + scolecite zone alteration at Teigarhorn is characterized by radiating bundles of near stoichiometric scolecite fibers (table 3) that fill nearly all pore spaces remaining after C/S precipitation (fig. 5D). Scolecite is white in hand specimen and often has a brownish hue in plane polarized light. Growth direction is very distinct; the bundles widen as a function of distance from the apex where nucleation first occurred at the edge of Stage II C/S pore linings.

Typical vesicle fillings in the heulandite + stilbite zone consist of intergrowths of stilbite, heulandite, and mordenite with occasional epistilbite (fig. 5B, C). Frequently only one or two of these minerals are present in a given pore (usually stilbite or heulandite). Mordenite is fibrous, occurring alone or as discrete fibers within heulandite or stilbite crystals (fig. 5C). Stilbite, heulandite, and epistilbite occur as euhedral blocky to lath shaped crystals. No evidence was found for heulandite + stilbite zone assemblages overprinting (replacing) mesolite + scolecite zone minerals. Representative compositions of Stage II stilbite, heulandite, and epistilbite are given in table 3 (Stage II mordenite is too fine-grained for reliable analysis). Epistilbite compositions are slightly richer in Al^{+3} and Na^{+} than the ideal formula ($\text{CaAl}_2\text{Si}_6\text{O}_{16} \cdot 6\text{H}_2\text{O}$). The ratio of Si to Al in Stage II heulandite and stilbite ranges from ~2.9 to ~3.4 and ~2.9 to ~3.5, respectively. Exchangeable cation sites in Stage II heulandite (fig. 8) and stilbite (fig. 9)

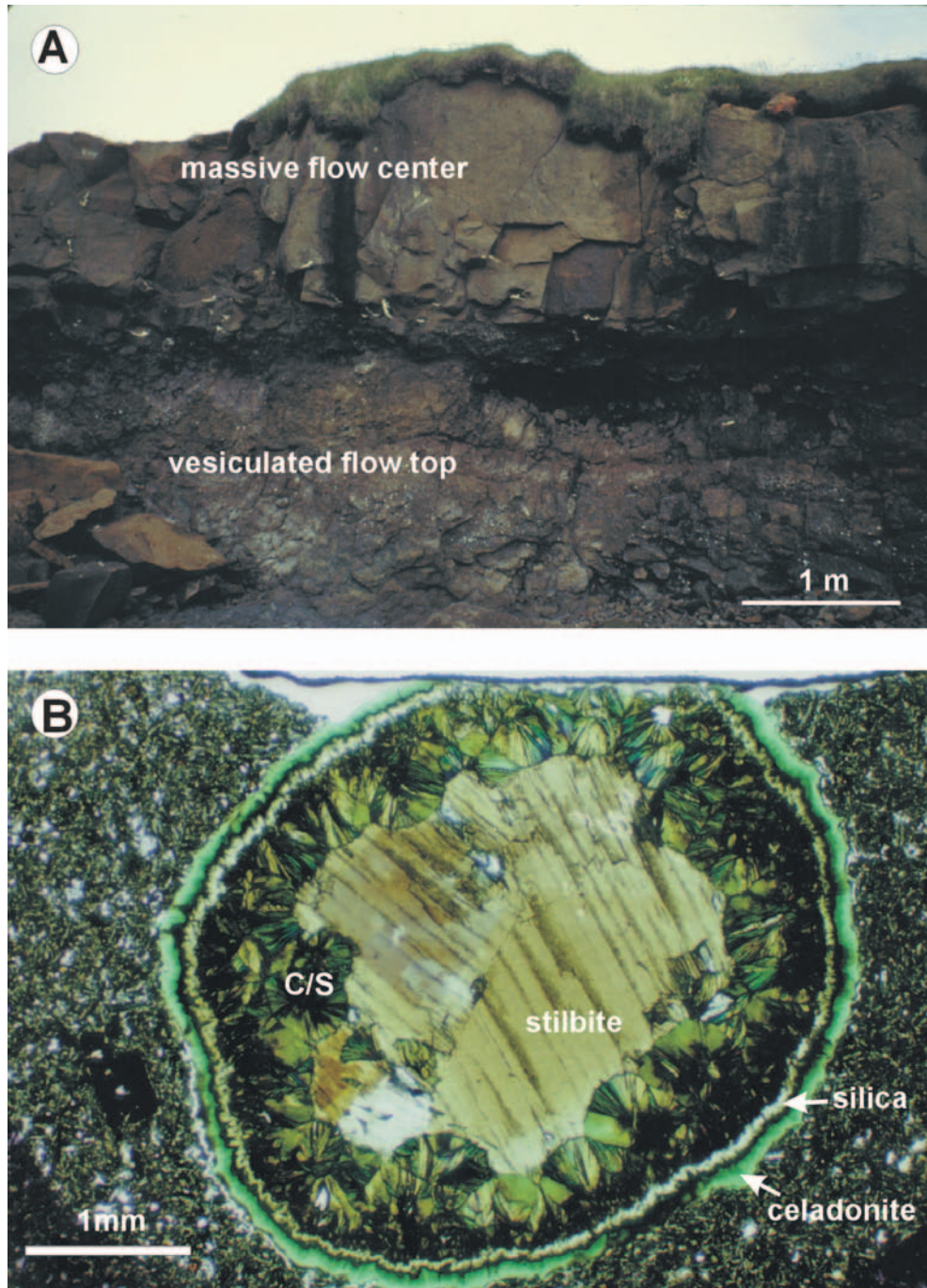
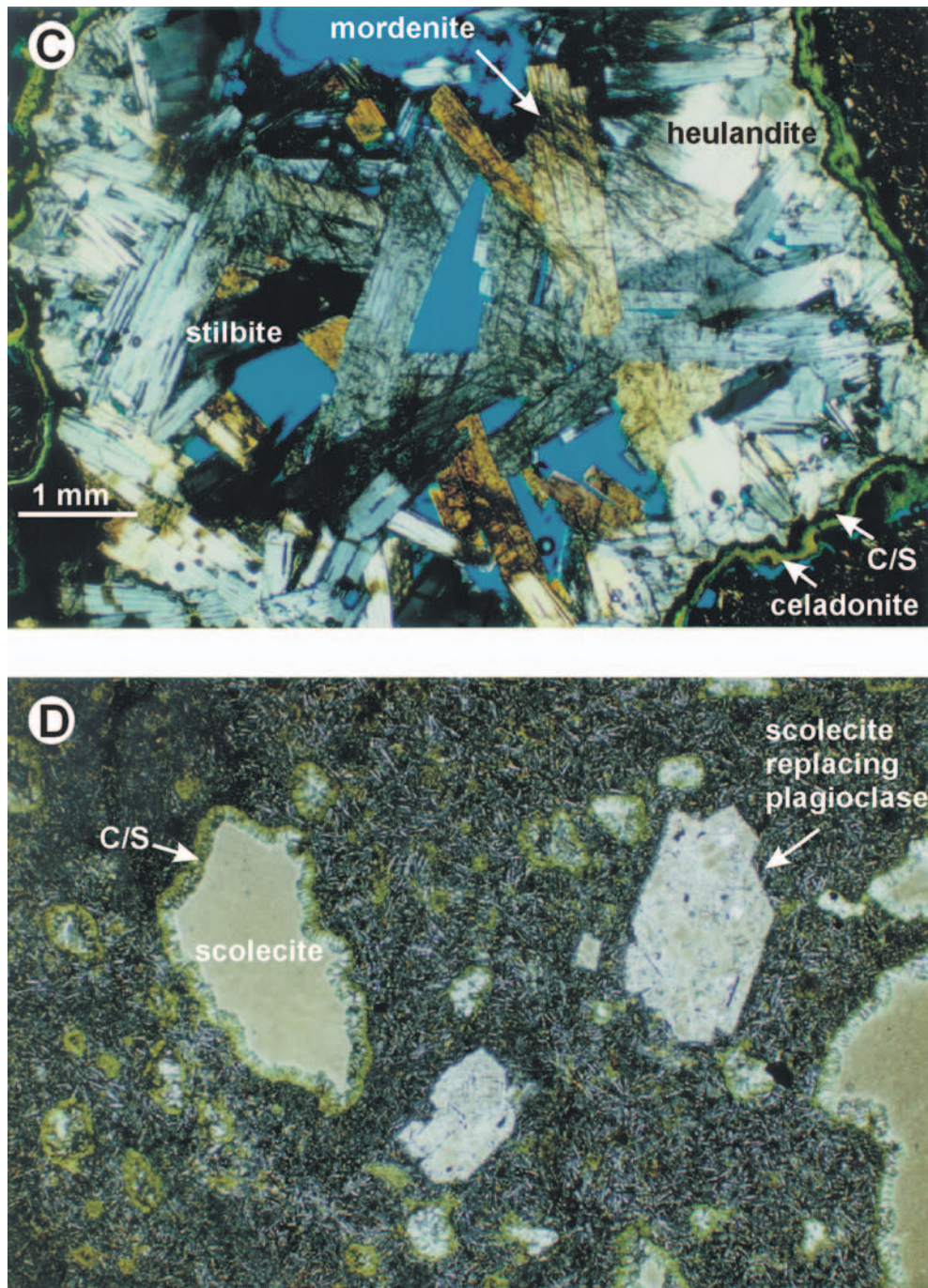


Fig. 5. Stage I and II alteration at Teigarhorn. (A) Outcrop view showing the flow scale distribution of primary pore spaces that host Stage I and II alteration. Two lava flows are visible separated by an undulating surface. The top of the lower flow is highly vesicular and contains abundant Stage I celadonite rims and Stage II scolecite visible as white pore fillings. The massive flow center of the upper flow contains only isolated, large vesicles filled with Stage II scolecite. (B) Stage I celadonite and silica rims preceding Stage II mixed-layer chlorite/smectite (C/S) and stilbite in sample ISMP-2. Photomicrograph taken with polars partially crossed. Analyses in columns 2 and 7 of table 2 were conducted on celadonite and C/S rims shown in photo.



(C) Stage II heulandite + stilbite zone assemblages (sample 94-53). Photomicrograph taken with polars partially crossed. Vesicle in rimmed with celadonite (Stage I) and Stage II C/S. Filling the pore are lath-shaped crystals of stilbite (birefringence phase) and blocky crystals of heulandite. The stilbite and heulandite crystals contain abundant mordenite fibers. Blue areas are residual porosity (color due to impregnated epoxy). (D) Stage II mesolite + scolecite zone assemblages (sample 94-77). Photomicrographs taken with polars partially crossed. Vesicles are rimmed with C/S and filled with scolecite. The large plagioclase phenocrysts have been replaced by scolecite.

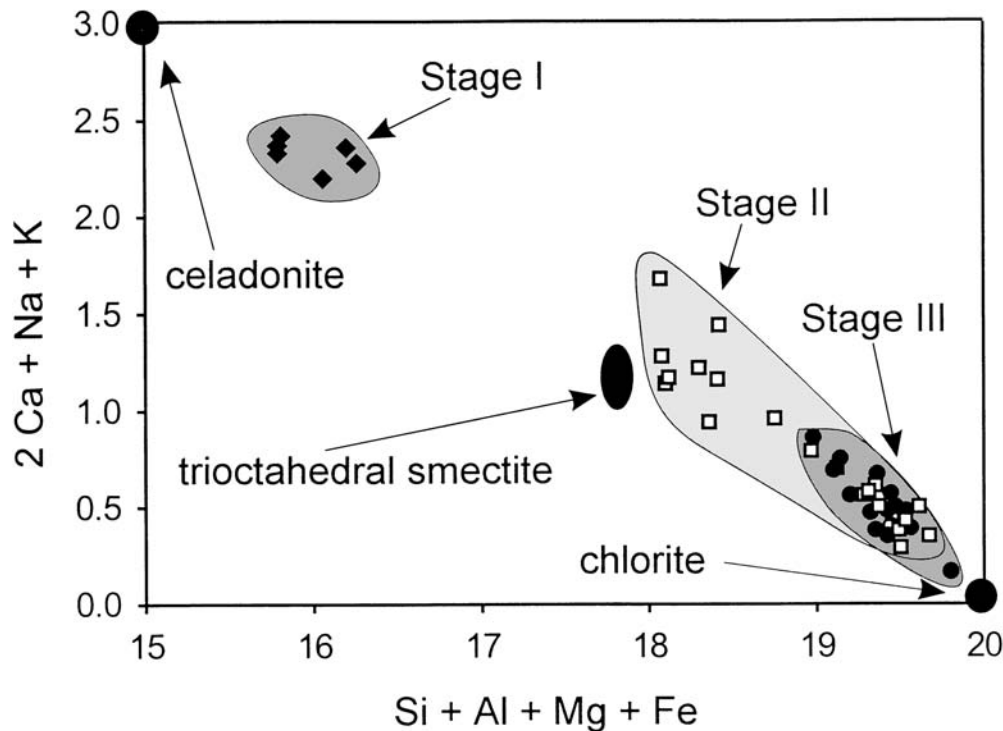


Fig. 6. Chemistry of phyllosilicates from Teigarhorn (normalized to 28 O charge equivalents) as a function of total mols of charge associated with exchangeable ions ($2\text{Ca} + \text{Na} + \text{K}$) and total molar $\text{Si} + \text{Al} + \text{Mg} + \text{Fe}$ content. Stage I phyllosilicates are shown as solid diamonds, Stage II phyllosilicates as open squares, and Stage III phyllosilicates as solid circles. Ideal, endmember celadonite, chlorite, and trioctahedral smectite are plotted as large black ovals. Note restricted composition ranges for Stage I and III phyllosilicates and higher smectite contents of Stage II phyllosilicates.

are dominantly occupied by Ca^{+2} , although in some samples Na^{+} and K^{+} constitute up to 30 percent of the exchangeable ions.

Stage II minerals in the matrix of the lavas occur predominantly as pseudomorphic replacements of primary phenocryst and groundmass phases (table 1). Groundmass glass, pyroxene, and olivine as well as pyroxene and olivine phenocrysts are partially replaced by C/S with compositions similar to that lining primary pore space. Groundmass and phenocryst plagioclase is pseudomorphically replaced by scolecite and/or completely albitized (table 4).

Stage III.—The latest alteration at Teigarhorn (Stage III) is developed in secondary pore space (fractures, breccias, et cetera) and primary pores intersected by this porosity (figs. 10, 11). Stage I and II minerals filling vesicles and other primary pore spaces are cross cut by Stage III porosity. Stage III breccias commonly entrain Stage I and II minerals (fig. 11C). This is frequently visible in outcrop as rafts of vesicle walls coated with celadonite and Stage II heulandite within Stage III breccias. Development of fracture porosity during Stage III is associated with intrusion of mafic dikes that cross cut the lavas. The near-vertical dikes make up more than 8 percent of the volume of rocks at Teigarhorn (fig. 2) and vary in thickness between 0.5 and 3 m. Spacing between dikes is uneven, ranging from less than 1 to over 30 m. Lava flows within a few meters of the dikes are extensively fractured, but the fractures generally do not contain secondary

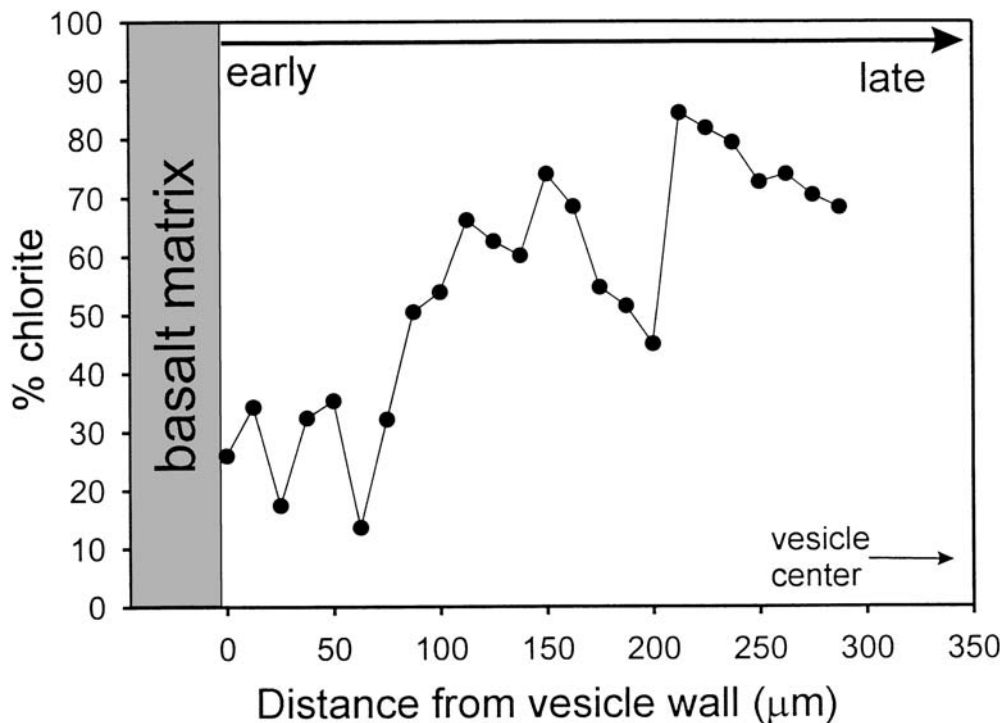


Fig. 7. Temporal changes in the compositions of Stage II mixed-layer chlorite-smectite (C/S) depicted as a function of percent chlorite (calculated using method of Schiffman and Fridleifsson, 1991). Analyses are from a 300 μm linear traverse through a Stage II clay rim of a vesicle in sample 94-50. The compositions of C/S trend from early smectitic clays (near the vesicle wall) to relatively late chloritic clays (towards the center of the vesicle).

minerals. At distances greater than 3 m from some dikes, brittle deformation features host Stage III mineral assemblages. This alteration is only locally developed.

Stage III mineral assemblages fill 0.2 to 2 cm thick veins oriented parallel to the dikes and irregular fractures and breccias up to 0.8 m across. Veins and fractures can be traced for up to 10 m. The margins of veins are occasionally lined with 5 micron equant crystals of clinoptilolite (compositions given in fig. 8 and table 5) and usually marked by one or more layers of chalcedony. Filling the veins is quartz that is an- to subhedral near the margin and euhedral in the center with the c axis oriented normal to the vein walls. Clusters of euhedral quartz crystals (individual crystal size two to four mm) frequently form stalactiform deposits hanging vertically from the roofs of large cavities in the breccias. Generally chlorite-rich C/S occurs intergrown with quartz and as discrete fracture fillings (table 2). The last assemblages to fill the veins and breccias are euhedral stilbite, heulandite, or stilbite + heulandite + mordenite (fig. 11A, B) with local development of later scolecite, laumontite, and calcite in large, open cavities (fig. 11B). Stage III zeolites frequently occur as vein fillings and as large (cm-scale) euhedral crystals for which Teigarhorn is famous. Representative compositions of Stage III zeolites are given in table 5. The compositions of heulandite, stilbite, and scolecite formed during Stage III are similar to those formed during Stage II (table 5; figs. 8 and 9).

Fluid inclusion heating/freezing experiments were conducted on a 3 cm crystal of Iceland spar type calcite that formed late in Stage III along with scolecite and laumontite. Two-phase inclusions (liquid with vapor bubbles constituting ~5 percent of the inclusion

TABLE 3

Representative compositions of Stage II zeolites from vesicles determined by EPMA

	scolecite 96-T-1	scolecite 94-80	heulandite 94-95	heulandite 94-35	stilbite 96-T-1	stilbite 94-96	epistilbite 94-93	epistilbite 96-T-1
SiO ₂	45.35	44.41	60.50	60.06	61.96	58.94	60.36	55.83
Al ₂ O ₃	25.77	24.44	16.36	16.50	16.48	17.02	18.02	16.97
Fe ₂ O ₃	0.05	0.06	0.00	0.04	0.00	0.00	0.00	0.03
MnO	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00
MgO	0.01	0.00	0.03	0.01	0.03	0.07	0.00	0.04
BaO	0.02	0.00	0.18	0.22	0.24	0.32	0.13	0.25
SrO	0.00	0.00	0.30	0.90	0.60	0.56	0.00	0.24
CaO	13.12	13.72	7.32	7.22	5.96	7.34	8.04	6.90
Na ₂ O	1.13	0.04	0.74	0.60	1.26	0.84	1.35	1.00
K ₂ O	0.00	0.00	0.92	0.95	2.01	1.00	0.07	1.03
total	85.45	82.68	86.33	86.50	88.53	86.09	87.98	82.28
Anhydrous formula unit composition								
Si	3.00	3.02	6.84	6.81	6.87	6.72	5.94	5.72
Al	2.01	1.96	2.18	2.19	2.16	2.28	2.09	2.29
Fe	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Mn	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Mg	0.00	0.00	0.00	0.00	0.00	0.02	0.00	0.00
Ba	0.00	0.00	0.00	0.00	0.00	0.02	0.00	0.00
Sr	0.00	0.00	0.02	0.07	0.04	0.05	0.00	0.00
Ca	0.92	1.01	0.88	0.88	0.71	0.88	0.86	0.98
Na	0.15	0.01	0.16	0.14	0.28	0.19	0.26	0.27
K	0.00	0.00	0.14	0.14	0.28	0.14	0.00	0.00
O	10	10	18	18	18	18	16	16

volume) occur throughout the crystal. Most inclusions are up to 0.25 mm in diameter with ovoid, elongate, or triangular morphologies. Primary inclusions homogenized at $115^{\circ} \pm 8^{\circ}\text{C}$ and frozen at -0.1° to 0°C .

Stage III fractures and breccias are frequently surrounded by aureoles (1-40 cm wide) of extensive SiO₂ metasomatism (fig. 10). These zones are lighter in color than the surrounding lavas, and in extreme cases are pale yellow. The lava is extensively replaced by fine-grained aggregates of quartz with about 5 modal percent C/S (fig. 11C and D). Stage III C/S (table 2, fig. 6) is more chloritic than in Stage II, ranging from 55 to 90 percent chlorite. Where this alteration is most intense the original texture of the lava is completely obliterated. The replaced lava is highly porous with a spongy texture (fig. 11D) and has approx 20 percent secondary porosity.

Bulk Rock Chemistry

Bulk rock compositions of samples from two successive lava flows (and an intervening mafic volcanoclastic unit) from Teigarhorn (samples 94-86, 94-87, 94-90, 94-91, 94-96, and 94-101; fig. 2) are given in table 6. Samples 94-86 and 94-101 are from the overlying (younger) flow, 94-87 is the mafic volcanoclastic horizon, and 94-90, 94-91, and 94-96 are from the underlying (older) flow (see footnote to table 6 for sample descriptions). The least altered samples of both flows (94-86 and 94-90) are similar in composition and belong to the low MgO tholeiite lavas described by Flower and others (1982). The mafic volcanoclastic unit is compositionally similar to the unaltered lavas except for higher Fe, Si, K, Rb, Cu, Cr, and Ni and lower Ca, Na, P, Sr, and V contents in the oxide-rich, more oxidized volcanoclastic horizon.

The effects of alteration on bulk rock composition can be assessed by comparing these relatively unaltered samples with examples of Stages I and II (94-91 and 94-96) and Stage III (94-101) alteration from the same flows. This is illustrated by the isocon plots

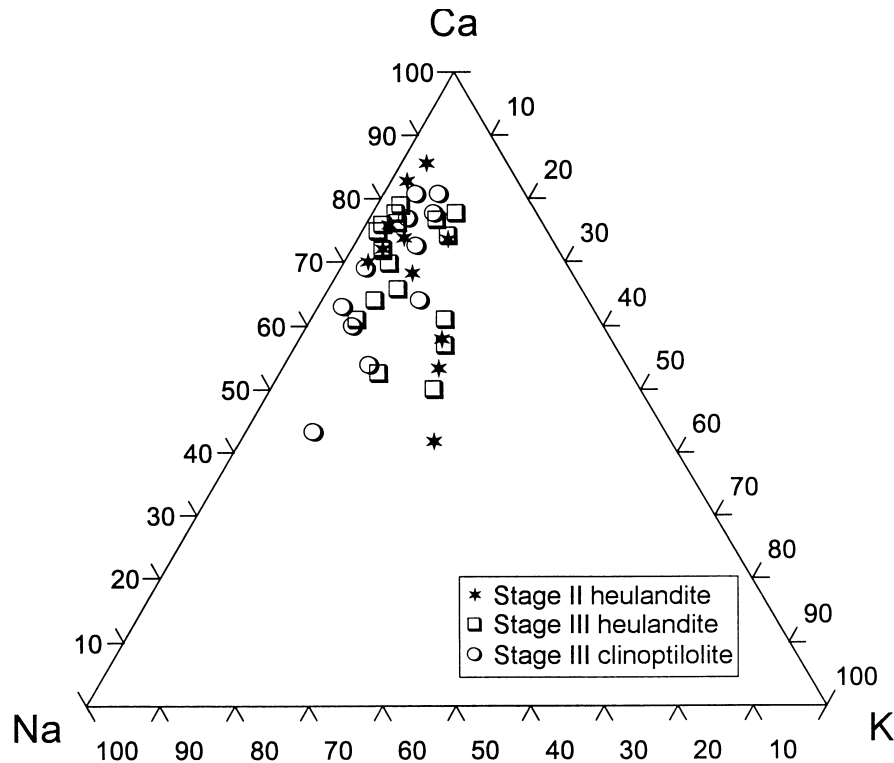


Fig. 8. Molar proportions of Ca, Na, and K in heulandite and clinoptilolite from Teigarhorn.

(Grant, 1982) in figure 12, which compare the compositions of sample 94-96 (which exhibits Stage I and II alteration) and sample 94-101 (extreme Stage III SiO_2 metasomatism) to those of the relatively unaltered samples 94-90 and 94-86, respectively, of the same flows. The linear trends (labeled “constant mass isocon” in fig. 12) represent no elemental loss or gain during alteration. It can be seen in figure 12A that Stage I and II alteration together lead to only minor changes in bulk rock composition, as illustrated by the fact that the major element data plot along the isocon. With the exception of Rb and La in sample 94-96, the other trace element data listed in table 6 have been omitted for clarity, but they all plot along the constant mass trend as well. The most significant bulk chemical signature of Stages I and II is an increase in La, Rb, and to a lesser extent K_2O concentration; Rb and K_2O were likely concentrated due to precipitation of celadonite in 94-96. The fact that most of the data in figure 12A plot along the constant mass isocon reflects the isochemical nature of the hydration reactions that typify Stage I and II alteration. In contrast, the extreme metasomatism associated with Stage III is clearly reflected in sample 94-101. Quartz along with minor stilbite and chlorite have entirely replaced the lava (94-86) resulting in a bulk rock composition that is ~ 92 percent SiO_2 . As can be seen in figure 12B, this sample is clearly enriched in SiO_2 and depleted in all other analyzed elements relative to the unaltered lavas.

Porosity

Lava flows at Teigarhorn exhibit values of Φ_1^* (volume percent of primary pores) up to 22 percent with subsequent processes during Stage III leading to values of Φ_2^* of up to 11 percent (table 7). The range of values of Φ_1^* given in table 7 is typical of basaltic lavas

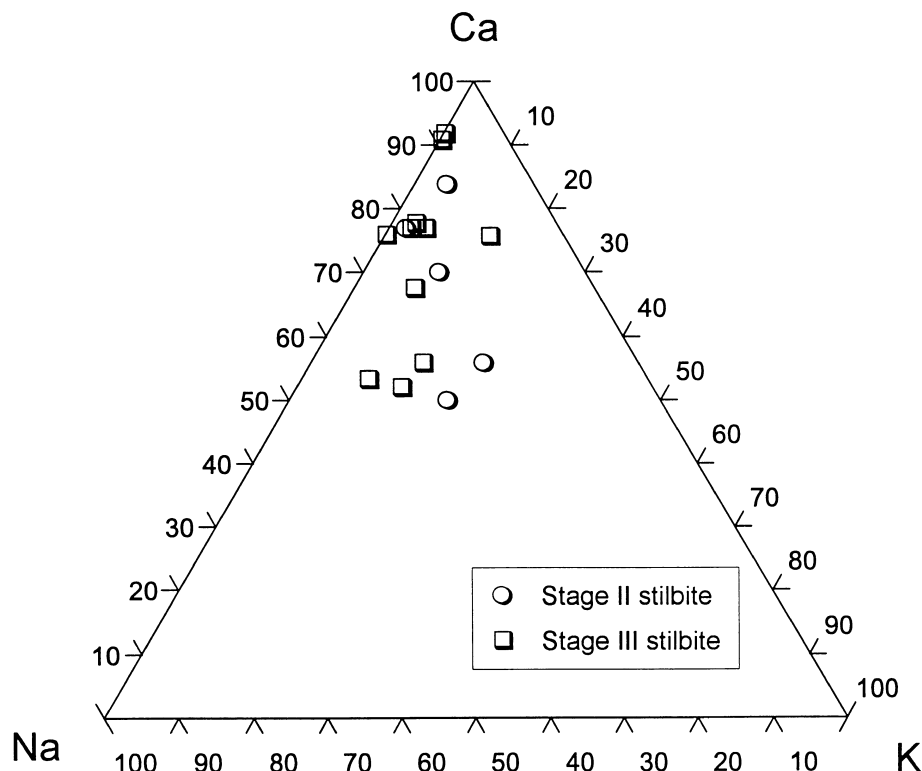


Fig. 9. Molar proportions of Ca, Na, and K in stilbite from Teigarhorn.

reported by Christensen and Wilkens (1982) from the Reydarfjörður drillhole in eastern Iceland. Variations in Φ_1^* (table 7) reflect differences in pore density both between flows and within various portions of a given flow. The estimates of Φ_2^* (volume percent of secondary pores) are similar to those obtained by Manning and Bird (1991) for fractured basalts near the margin of the Skaergaard intrusion in East Greenland.

Percentages of Φ_1^* occupied by Stage I celadonite and silica and Stage II clays and zeolites along with the percent of Φ_1^* remaining after precipitation of Stage I and II minerals are given in table 8. Inspection of the table shows that the extent of porosity reduction during surficial alteration and burial metamorphism is heterogeneous between samples and is a strong function of paragenetic stage. Precipitation of Stage I minerals, where developed, leads to a 10 to 30 percent reduction in Φ_1^* . Chlorite/smectite interlayer clays are more common than Stage I minerals and fill 0 to 80 percent of Φ_1^* . Zeolites, the last minerals to precipitate during Stage II, fill an additional 0 to 90 percent of Φ_1^* . The percent of Φ_1^* remaining after Stage II varies between ~0 and 35 percent; that is, Φ after Stage II ranges from about 0 percent to nearly 8 percent.

DISCUSSION

Time-space development of mineral parageneses.—Mineral parageneses developed at Teigarhorn present a time-integrated view of the petrogenesis of zeolite-facies basaltic aquifers in eastern Iceland. In this context, the temporal development of mineral assemblages can be directly related to the geologic evolution of the crust in eastern Iceland. This is summarized in figure 13, which depicts the history of the lavas at

TABLE 4
*Representative compositions of altered and fresh plagioclase
determined by EPMA*

	fresh plagioclase 94-91	albitized plagioclase 94-79	albitized plagioclase 96-T-1
SiO ₂	50.51	66.60	67.80
Al ₂ O ₃	30.37	19.60	19.47
Fe ₂ O ₃	0.80	0.29	0.05
MnO	0.00	0.04	0.00
MgO	0.16	0.09	0.02
BaO	0.04	0.00	0.04
SrO	0.08	0.00	0.04
CaO	13.43	0.28	0.51
Na ₂ O	3.72	11.40	11.44
K ₂ O	0.07	0.10	0.07
total	99.19	98.40	99.44
Atoms per formula unit (8 O)			
Si	2.33	2.97	2.99
Al	1.66	1.03	1.01
Fe	0.01	0.01	0.00
Mn	0.00	0.00	0.00
Mg	0.03	0.01	0.00
Ba	0.00	0.00	0.00
Sr	0.00	0.00	0.00
Ca	0.66	0.01	0.02
Na	0.34	0.99	0.98
K	0.01	0.00	0.00
% Albite	33.6	98.1	97.2
% Anorthite	65.9	1.3	2.4
% Orthoclase	0.5	0.6	0.4

Teigarhorn. The horizontal axis in figure 13 represents time after eruption and the vertical axis is crustal depth. Also illustrated in figure 13 are petrotectonic events (lava eruption, burial during crustal accretion, dike intrusion, glacial exhumation) and mineral parageneses experienced by the lavas.

Celadonite and silica rims (Stage I) formed at shallow depths (fig. 13). Stage I geopetal structures at Teigarhorn have orientations similar to the lava flows, and in other localities exhibit progressively more shallow dips between silica bands due to progressive tilting (Walker, 1974). This relationship suggests that the geopetal structures formed prior to tilting. Near surface genesis for Stage I minerals is consistent with the low temperatures ($< \sim 50^\circ\text{C}$) at which celadonite forms in natural systems (Odin, 1988) and the supersaturation of Icelandic surface waters with respect to chalcedony (Gíslason and Eugster, 1987b; Gíslason, Arnórsson, and Ármannsson, 1996). Silica and potassium are among the most mobile elements in near-surface environments in Iceland (Gíslason, Arnórsson, and Ármannsson, 1996), providing components necessary for formation of celadonite and opal.

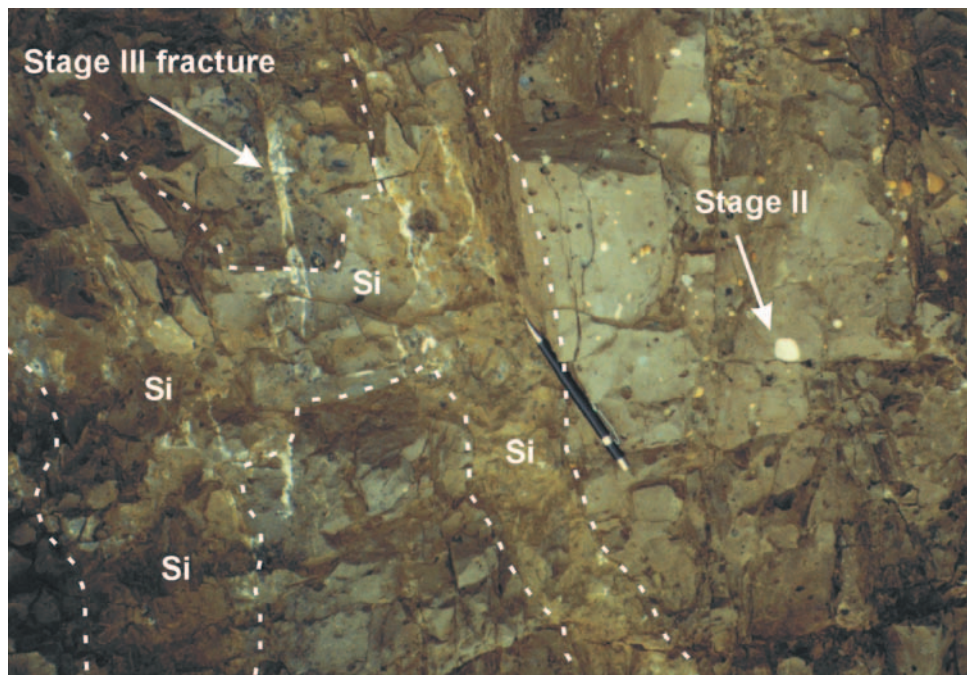


Fig. 10. Stage III fractures filled with quartz (outcrop view). Pencil for scale. The dashed white lines are boundaries of metasomatic zones containing quartz + minor C/S that replace the basalts (see fig. 11C and D). The aureoles are visible as areas of yellow alteration marked by the symbol "Si." Also visible are vesicles containing Stage II scolecite.

Burial metamorphism resulted in formation of Stage II mineral assemblages. Mixed-layer chlorite/smectite clays most likely formed during burial (fig. 13). Chemical components necessary for C/S precipitation were derived from hydrolysis of olivine and basaltic glass. We propose that the increase in chlorite content of C/S pore linings with crystallization time (fig. 7) is a consequence of progressive growth of C/S as temperature increased during burial of the lavas. The trend toward endmember chlorite composition shown in figure 7 is analogous to prograde zones characterized by smectite, C/S, and chlorite observed with increasing temperature in active geothermal systems in Iceland (Kristmannsdóttir, 1979; Schiffman and Fridleifsson, 1991). The early, more smectite-rich C/S formed at relatively low temperatures (shallow depths) and as burial proceeded C/S became progressively more chlorite-rich as temperature increased during burial.

Precipitation of Stage II zeolites occurred after burial of the lavas. Regional mineral isograds in eastern Iceland characterized by Stage II zeolites, such as the boundary between the mesolite + scolecite zone and the heulandite + stilbite zone, transgress (crosscut) the tilted volcanic stratigraphy in eastern Iceland (Walker, 1960b). The zeolite isograds thus postdate tilting of the lava pile, as observed in the Early Tertiary plateau basalts of East Greenland (Neuhoff and others, 1997). In East Greenland, the sequence of regional zeolite mineral zones with depth did not develop successively as the lavas were buried. Instead, it appears that regional zeolite formation occurred after the main stage of Early Tertiary volcanism in East Greenland ceased. The notable lack of scolecite precursors to heulandite + stilbite zone mineral assemblages at Teigarhorn suggest a similar model for the timing of zeolite zone development in eastern Iceland. Zeolites did not precipitate during burial of the lavas. The pervasive albitization observed at

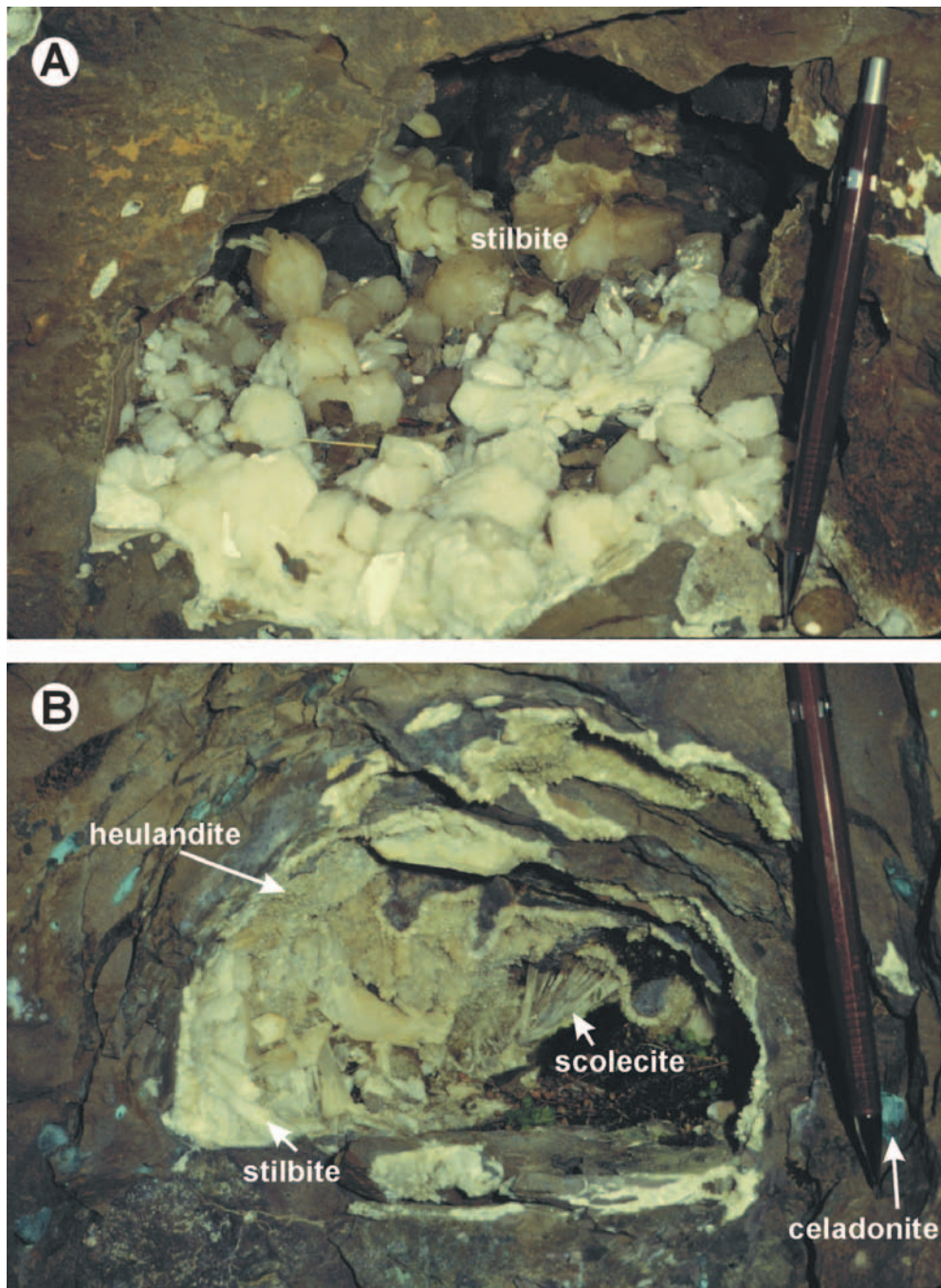


Fig. 11. Texture and mineralogy of Stage III alteration at Teigarhorn. (A) Cavity in Stage III breccia lined with euhedral stilbite. Pencil for scale. (B) Stage III breccia lined with heulandite and later stilbite and scolecite. Primary vesicles in lava around cavity are lined with green celadonite. Pencil for scale. (C) Timing and textural relationship between Stages I and III (sample 94-94). Photo taken with polars slightly crossed. The wall of a primary pore space is lined with celadonite; Stage III stilbite fills a vein along the wall of the pore. A patchy aggregate of quartz (with minor C/S) is replacing the lava on the other side of the vein. The “pie slice”-shaped celadonite in the center of the photo has been dislodged from the wall of the pore space during Stage III brecciation. Note the vuggy texture of the quartz aggregate replacing the lava (blue areas are porosity filled with impregnated epoxy). (D) Hydrothermally altered lava (sample 94-97) that has undergone extensive SiO_2 metasomatism. Photo taken with polars slightly crossed. The matrix of the lava is pervasively replaced by quartz and C/S. Stilbite filled cavities appear to represent both partially replaced Stage II vesicle fillings and late Stage III veins. Abundant pore space is present between quartz grains (blue areas).

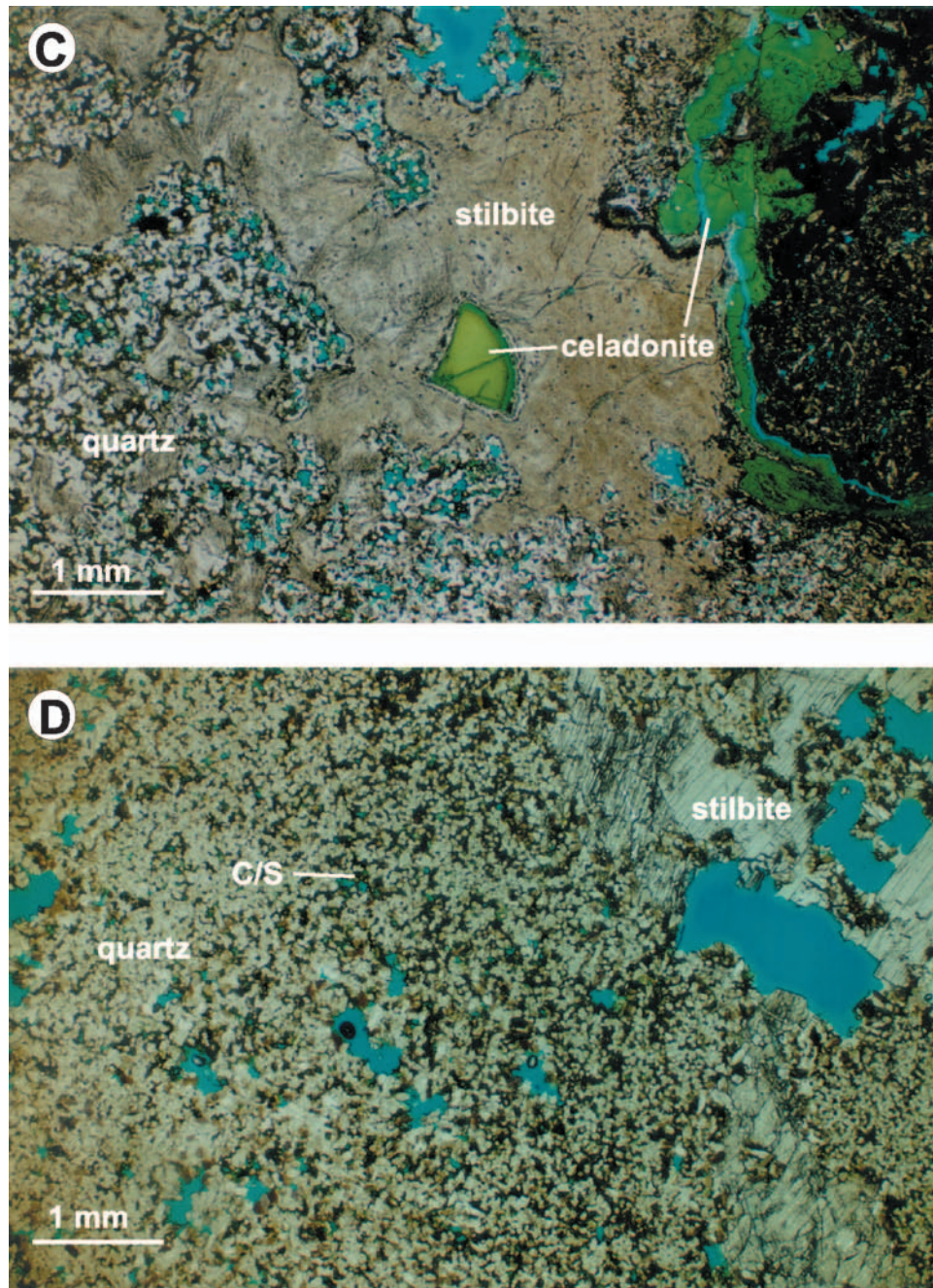


Fig. 11 (continued)

Teigarhorn is not present at elevations $>100\text{m}$ in lavas exposed in the mountains around Berufjörður (Neuhoff, unpublished data). This suggests that albitization occurred concurrently with Stage II zeolite formation. The paucity of absolute age dates in eastern Iceland do not permit concise constraints on the timing and duration of zeolite

TABLE 5
Representative compositions of Stage III fracture-filling zeolites determined by EPMA.

	clinoptilolite 94-52	clinoptilolite 94-54	heulandite 94-50	mordenite 94-54	stilbite 94-97	stilbite ISMP2	scolecite ISMP1	laumontite 94-81
SiO ₂	67.70	63.901	59.42	72.67	58.83	55.75	46.13	50.58
Al ₂ O ₃	13.18	14.801	15.64	10.81	16.00	17.42	25.58	21.98
Fe ₂ O ₃	0.05	0.000	0.00	0.00	0.00	0.00	0.00	0.03
MnO	0.01	0.000	0.00	0.02	0.01	0.00	0.02	0.00
MgO	0.01	0.006	0.02	0.01	0.01	0.01	0.00	0.00
BaO	0.01	0.087	0.18	0.04	0.25	0.41	0.03	0.01
SrO	0.10	0.115	0.26	0.05	0.25	0.31	0.07	0.04
CaO	5.67	7.007	6.63	4.25	7.51	7.72	13.87	11.32
Na ₂ O	0.69	0.713	1.37	1.35	0.66	1.19	0.13	0.27
K ₂ O	0.51	0.342	0.77	0.32	0.75	0.26	0.00	0.18
total	87.95	86.97	84.29	89.52	84.27	83.07	85.82	84.39
Anhydrous formula unit composition								
Si	14.72	14.15	6.88	5.11	6.81	6.59	3.03	3.98
Al	3.46	3.89	2.13	0.90	2.19	2.42	1.98	2.04
Fe	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Mn	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Mg	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Ba	0.00	0.00	0.00	0.00	0.00	0.02	0.00	0.00
Sr	0.00	0.00	0.02	0.00	0.02	0.02	0.00	0.00
Ca	1.30	1.66	0.82	0.49	0.94	0.99	0.97	0.95
Na	0.32	0.32	0.31	0.09	0.14	0.27	.001	0.05
K	0.14	0.11	0.11	0.06	0.12	0.05	0.00	0.02
O	36	36	18	12	18	18	10	12

TABLE 6

Bulk rock compositions from Teigarhorn, eastern Iceland

Samples [‡]	94-86	94-87	94-90	94-91	94-96	94-101
SiO ₂ [*]	46.844	48.145	46.021	47.414	47.058	92.210
TiO ₂ [*]	3.710	3.642	3.457	3.379	3.396	0.326
Al ₂ O ₃ [*]	13.533	13.068	13.895	13.531	13.305	2.558
Fe ₂ O ₃ ^{*,†}	17.004	20.301	17.077	16.632	16.616	2.574
MnO [*]	0.329	0.140	0.253	0.234	0.251	0.024
MgO [*]	5.212	5.746	5.750	5.520	5.399	0.939
CaO [*]	9.261	5.738	9.991	10.076	9.676	0.987
Na ₂ O [*]	3.163	1.812	2.946	2.868	3.113	0.551
K ₂ O [*]	0.763	1.633	0.322	0.280	0.519	0.236
P ₂ O ₅ [*]	0.442	0.289	0.398	0.382	0.387	0.043
Total [*]	100.261	100.514	100.110	100.316	99.720	100.448
H ₂ O ^{-§}	1.372	6.257	1.955	2.092	1.029	0.755
H ₂ O ^{+§}	0.134	3.169	0.483	0.605	0.527	1.640
Nb [†]	24.5	20.3	23.2	22.4	22.6	2.6
Zr [†]	238	208.3	217.9	210.3	212.7	26.1
Y [†]	47.2	39.3	42.6	41	42.5	3.8
Sr [†]	291	111.8	301.9	305.2	283.9	205.8
U [†]	1.6	0.8	1.4	1.3	2.2	0.9
Rb [†]	17.1	56.9	0.9	1.2	15.9	2.6
Th [†]	4.1	1.2	0.2	3.6	3.3	B/D
Pb [†]	2.7	2	2.8	0.4	3.5	0.1
Ga [†]	28	23.7	25.1	23.4	24	6.1
Zn [†]	148.6	163.1	143.4	134.5	128.4	22.6
Cu [†]	173.6	241.9	154.7	104.1	134.9	39
Ni [†]	29.6	52.4	38.7	39.5	37.4	5.9
Cr [†]	6	55.4	33.2	38.3	29.6	2.8
V [†]	434.6	354	454.4	439.4	428	46.4
Ce [†]	47	24.1	31.7	30.9	6.3	19
Ba [†]	180.1	111.7	120.2	126.7	103.3	107.7
La [†]	24.3	14.8	16.1	27.2	21.3	B/D

*Sample descriptions:

94-86: compact lava with minor Stage III heulandite veins

94-87: bright red mafic volcanoclastic unit marking boundary between two lava flows

94-90: compact, massive lava with minor small vesicles filled with smectite

94-91: compact basalt with minor vesicles containing Stage II clay minerals

94-96: vesicular lava with abundant Stage I celadonite, Stage II heulandite and smectite

94-101: yellow, SiO₂-metasomatized lava from aureole around Stage III breccia*Weight percent of oxide in anhydrous (calcined) sample. †Total Fe reported as Fe₂O₃. §Determined by loss on ignition at 110°C (H₂O⁻) and 900°C (H₂O⁺). †Parts per million of element in untreated sample. B/D: below detection limit.

precipitation during Stage II; however, it seems likely that it did not last more than a few Ma, as observed in East Greenland (Neuhoff and others, 1997).

Pore structures filled with Stage III alteration cross-cut Stage II mineralization. Stage III is spatially and, presumably, temporally linked to dike intrusion (fig. 13). The spatial association of Stage III with late dikes indicates that this last paragenetic stage is a local, and not a regional, phenomenon. The close association with dike intrusion indicates that Stage III occurred while the region around Teigarhorn was still undergoing intrusive

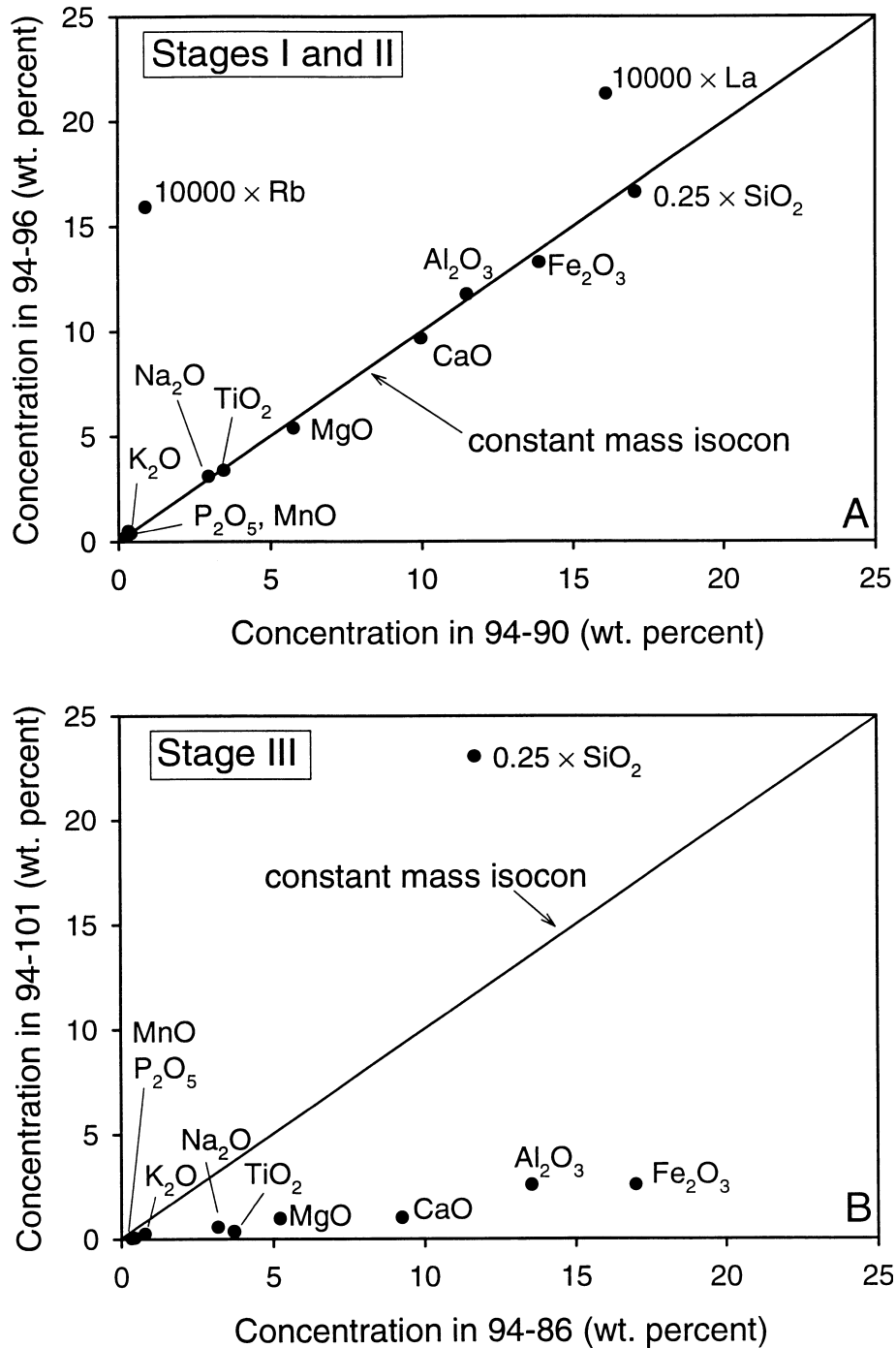


Fig. 12. Isocon plots (Grant, 1982) depicting the variation in bulk rock chemistry accompanying Stages I and II (A) and Stage III (B) alteration at Teigarhorn. The lines labeled "constant mass isocon" depict the theoretical distribution of data assuming that the mass and compositions of the lavas do not change during alteration. The SiO_2 , Rb, and La analyses have been multiplied (by factors of 0.25, 10000, and 10000, respectively) to facilitate comparison with data for other major oxide components. Data used to construct this plot are listed in table 6.

TABLE 7

Outcrop porosity measurements

Station ¹	Primary Porosity (Φ_1^* ; %)	Secondary Porosity (Φ_2^*)		
		Survey Area (cm ²)	Pore Area (cm ²)	Porosity (%)
1	22	-	-	-
2	4	60000	6894	11
3	12	-	-	-
4	4	-	-	-
5	6	-	-	-
6	17	-	-	-
7	4	56000	1696	3
8	12	34000	1046	3

¹Station locations given in figure 2. Descriptions: (1) flow top; (2) flow center with abundant Stage III fractures developed ~10 m from dike; (3) flow top; (4) flow center; (5) flow center; (6) flow top; (7) flow center with abundant Stage III fractures developed ~15 m from dike; (8) flow bottom with abundant Stage III fractures developed about 5 m from dike.

TABLE 8

Percent of pore space occupied by Stage I and II minerals based on thin section area measurement. All samples are from flow tops

Sample	N ^a	Percent of Total Primary Pore Space (Φ_1^*)				
		celadonite ^b	silica rim ^b	C/S clay ^c	zeolites ^d	residual porosity ^e
94-35	1	0	0	27	73	0
94-79	2	0	0	74	23	3
94-80	12	0	0	63	37	0
94-88	15	0	0	0	91	9
94-89	2	31	0	0	69	0
94-91	1	0	0	66	0	34
94-92	1	0	0	0	71	29
94-93	12	0	6	70	13	11
94-96	2	23	0	77	0	0
ISMP2	1	13	5	48	35	0
AVERAGE	49	7	1	42	41	9

^aNumber of individual pores measured. ^bRims of paragenetic Stage I celadonite or silica on vesicle walls. ^cIncludes all paragenetic Stage II trioctahedral smectite and chlorite interlayer clays. ^dStage II zeolite assemblages, dominantly mesolite + scolecite and heulandite + stilbite + mordenite. ^ePrimary pore space (Φ_1^*) remaining after paragenetic Stage II.

activity related to rifting. Increased heat flow and permeability due to fracturing during dike emplacement initiated hydrothermal fluid circulation that led to the extensive wall-rock replacement and fracture filling characteristic of Stage III mineralization. No age dating has been conducted on the dikes at Teigarhorn that would permit constraints on the absolute timing of Stage III alteration.

Thermobarometric conditions during Stages II and III.—Barometric conditions during paragenetic Stages II and III were complex functions of the burial and hydrogeologic

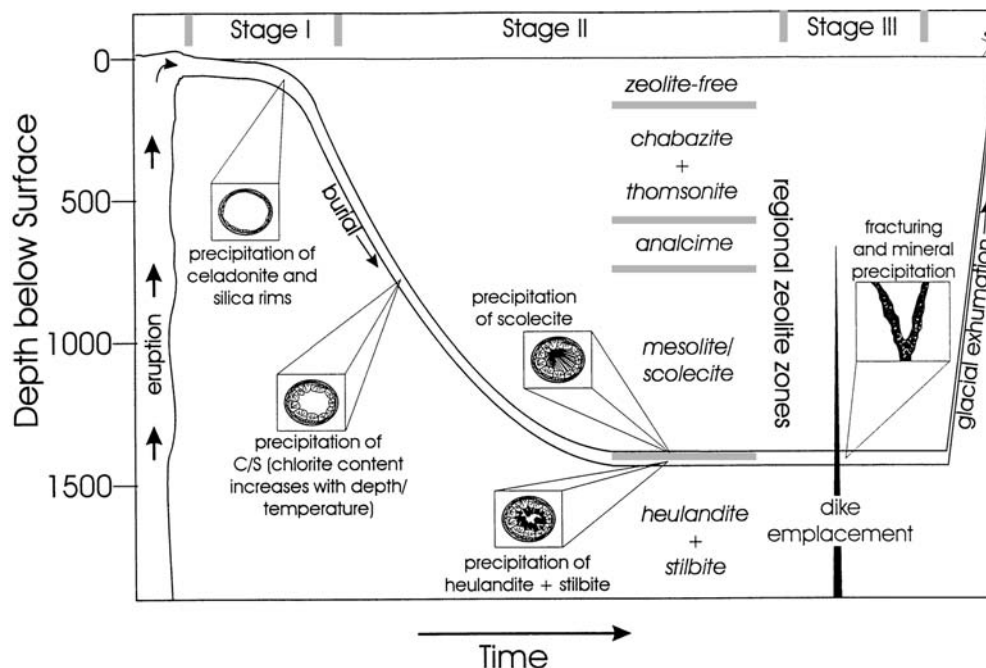


Fig. 13. Spatial and temporal development of pore-filling mineral assemblages at Teigarhorn. The vertical axis depicts depth below land surface at the time of each event depicted in the figure. Time elapsed after eruption increases to the right. No scale is implied on the horizontal axis. The parallel curves on the figure are boundaries of the lavas exposed at Teigarhorn in space and time. Note that the timing of dike emplacement is meant to infer only the timing of dikes associated with Stage III alteration at Teigarhorn.

history. Pressures during precipitation of Stage II C/S increased from a minimum of 1 bar (atmospheric pressure) to pressures between ~ 170 and ~ 500 bar, corresponding, respectively, to hydrostatic and lithostatic pressures at the maximum burial depth of 1500 m for the lavas at Teigarhorn (Walker, 1960b). Occlusion of macroscopic porosity by Stage II clay and zeolite minerals (table 8) likely resulted in near-lithostatic pressures. Pressures during Stage III were probably closer to hydrostatic conditions due to the higher permeability of continuous fractures (see below).

Although the systematic changes in Stage II C/S compositions appear to record the thermal history of the lavas (see above), the occurrence of this phase along with scolecite, stilbite, heulandite, and other Stage II zeolites is in contrast with the distribution of these minerals in active Icelandic geothermal systems. Smectite is present at low temperatures in Icelandic geothermal systems (below 200°C), with C/S occurring between 200° and 220°C (Kristmannsdóttir, 1979) and in some fields up to 270°C (Schiffman and Fridleifsson, 1991). At higher temperatures chlorite is present. Thus, C/S in Icelandic geothermal systems occurs at temperatures $> 100^{\circ}\text{C}$ higher than the boundary between the mesolite + scolecite zone and stilbite zones ($90^{\circ} \pm 10^{\circ}\text{C}$; Kristmannsdóttir and Tómasson, 1978).

In regionally metamorphosed basalts, the boundary between zones characterized by smectite and C/S is generally observed under zeolite-facies conditions (Schmidt and Robinson, 1997; Robinson, Bevins, and Rowbotham, 1993; Bevins, Robinson, and Rowbotham, 1991). For example, in the North Shore volcanic group of Minnesota, smectite coexists with heulandite and stilbite; C/S is associated with higher grade laumontite-albite assemblages (Schmidt and Robinson, 1997). Based on the association of C/S with scolecite and heulandite + stilbite at Teigarhorn, C/S formed at even lower

grades during Stage II in eastern Iceland. The relatively high chlorite contents of C/S associated with the zeolite assemblages at Teigarhorn may be a consequence of albitization of primary plagioclases, which promotes chlorite formation through release of aluminum (Shau and Peacor, 1992; Schmidt and Robinson, 1997). As noted by Schiffman and Fridleifsson (1991), the temperatures at which C/S occurs in geothermal systems are often higher than in regional diagenetic systems. In contrast, temperature estimates based on zeolite assemblages and fluid inclusion thermometry are similar (see below). This suggests that estimation of temperatures from observations of zeolite assemblages in geothermal systems may be more reliable than similar estimates based on phyllosilicate distribution.

Assuming a temperature of formation for Stage II zeolite assemblages of $90^\circ \pm 10^\circ\text{C}$ (see above) leads to an estimated thermal gradient of approx $53^\circ \pm 7^\circ\text{C/km}$ (given a surface temperature of 10°C). A thermal conductivity for water-saturated basalt of 1.5 to 2.0 W/(mK) (Oxburgh and Agrell, 1982) gives an estimated heat flow during Stage II zeolite precipitation of 1.9 to 3.2 heat flow units ($\text{HFU} = 10^{-6} \text{ cal} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$). This value is consistent with conditions on the flank of the present-day active volcanic zone in Iceland (Flóvenz and Sæmundsson, 1993).

Mineral assemblages and fluid inclusion measurements indicate that temperatures during Stage III were slightly higher than in Stage II. Empirically-based chlorite geothermometry (Cathelineau, 1988; Cathelineau and Nieva, 1985) suggests that the relatively early, chlorite-rich Stage III C/S formed at temperatures between 140° and 180°C . It should be noted that temperatures estimated using chlorite geothermometry generally disagree with measured temperatures in Icelandic geothermal systems (Schiffman and Fridleifsson, 1991). Late Stage III zeolite assemblages are broadly similar to those in Stage II but may have formed at slightly higher temperatures based on the presence of laumontite. Laumontite becomes prominent at temperatures above 120°C in Icelandic geothermal systems (Kristmannsdóttir and Tómasson, 1978), consistent with pressure corrected homogenization temperatures for late-Stage III calcite ($125^\circ \pm 8^\circ\text{C}$ assuming hydrostatic pressures). The apparent decrease in temperature during Stage III may reflect dissipation of heat flow associated with crystallization of the dikes or may be the result of errors associated with the chlorite geothermometer noted above.

Chemical fluxes during alteration.—The widespread precipitation of phyllosilicates, silica minerals, and zeolites in pore spaces during Stages I and II indicates that Si, Al, Ca, Na, K, Fe, and Mg were mobile over scales of a millimeter or more. This is further illustrated by the textural preservation of pore walls and by successive lining of pore spaces during mineral paragenesis, which require that chemical components necessary for precipitation of secondary minerals were transported into the pores. All components necessary to form the mineral assemblages described above were readily available in the primary phases in the lavas (glass, olivine, plagioclase, pyroxene) with the exception of H_2O , which was provided by the metamorphic fluids. The similarity between the bulk rock compositions of lavas altered during Stages I and II and those of unaltered lavas (table 6; fig. 12A) indicates that many elements were relatively immobile (that is, conserved) on the scale of individual lava flows during regional alteration. An exception is potassium, which is concentrated during Stage I in celadonite (table 6). As noted above, potassium is very mobile during near-surface alteration of basaltic lavas (Gíslason, Arnórsson, and Ármannsson, 1996). The concentration of Rb, K_2O , and La apparent in figure 12A and table 6 is consistent with the observations of Wood (1976) that these elements were mobilized during zeolite-facies metamorphism.

In contrast, high fluid fluxes through some Stage III (see below) fractures resulted in widespread leaching at the outcrop scale of all elements except silicon (table 6; fig. 12B). Extensive replacement of the lavas by quartz (plus minor chlorite-rich C/S) is suggestive of interaction with acidic fluids. The low pH of these fluids may reflect the presence of

relatively large amounts of dissolved magmatic gases. The solubilities of primary phases in the lavas as well as C/S and zeolites are all enhanced under very acidic conditions. In contrast, quartz solubility is insensitive to pH under acidic conditions, and it formed early during Stage III from SiO_2 released during hydrolysis/dissolution of primary phases. The large modal abundance (>90 percent) of quartz in Stage III aureoles suggests that some SiO_2 may have been added during alteration. Aside from minor C/S precipitated in the aureoles and later in veins, no mineralogical sinks for iron and magnesium formed during Stage III, suggesting that these elements were transported over distances of meters (the scale of outcrops at Teigarhorn) away from their sources. Late precipitation of zeolites and calcite in Stage III veins most likely occurred as the aqueous solutions became more alkaline as protons were consumed by hydrolysis.

Both Stages II and III exhibit a pronounced temporal change during paragenesis from phyllosilicates + quartz to assemblages dominated by Ca-zeolites. In both cases, this paragenetic trend was probably coincident with changes in temperature and pressure. However, the abrupt shift in the mineral composition from C/S to zeolites must reflect the chemical evolution of the aqueous electrolyte solutions from which these minerals precipitated, as schematically illustrated in the phase diagram in figure 14. The arrows A and B in figure 14 represent two possible trends in fluid composition that would result in mineral paragenesis of Ca-zeolites after C/S (figs. 4, 5B and C). The decrease in $\log [(a_{\text{Mg}^{+2}}/a_{\text{H}^+})^2]$ suggested by paragenetic trends A and B would result from decreasing the activity of Mg^{+2} in the pore fluid as a consequence of precipitation of C/S (represented by clinocllore in the phase diagram). Mineralogical sources of Mg^{+2} (olivine, basaltic glass) in basaltic lavas are generally less stable than plagioclase under low-grade metamorphic conditions (Gíslason and Eugster, 1987a; Gíslason and Arnórsson, 1993) and are often hydrolyzed before plagioclase. Albitization of plagioclase was likely contemporaneous with precipitation of zeolites in pores during Stage II (see above), adding Ca^{+2} to aqueous solution and increasing $\log [(a_{\text{Ca}^{+2}}/a_{\text{H}^+})^2]$ as suggested by trend A in figure 14. Instability of zeolites relative to C/S during early Stage II alteration provides an explanation for the apparent lack of zeolite formation during burial of the lavas inferred from our petrographic observations that heulandite + stilbite zone assemblages do not replace mesolite/scolecite zone assemblages. These observations, combined with the phase relations depicted in figure 14, suggest that the lavas were buried before olivine and basaltic glass were exhausted, resulting in fluid compositions during burial in which C/S was stable relative to zeolites.

The general paragenetic trend from C/S to zeolites, as well as fluid composition trends A and B in figure 14, is consistent with the results of experimental simulations of basalt-meteoric water interaction at low temperatures. Ghiara and others (1993) conducted experiments on closed-system dissolution of basaltic glass in deionized water at 200°C that resulted in a mineral paragenesis of early smectite + phillipsite followed by analcime. Total Mg concentrations in solution during these experiments initially rose rapidly, and then declined to a steady state level as Mg-rich smectite precipitated. Calcium concentrations also rose rapidly during the initial stages of these experiments, and continued to increase until the last stages of the experiments. Thus, $\log [(a_{\text{Ca}^{+2}}/a_{\text{Mg}^{+2}})]$ increased with time in response to precipitation of Mg-rich smectite and the fluids evolved toward the stability fields of zeolites as suggested in figure 14. Similar experiments by Gíslason and Eugster (1987a) on dissolution of basaltic glass and crystalline basalt at 25°, 45°, and 65°C resulted in nearly identical solution composition trends. Subsequent analysis of the solid phases produced in these experiments (Gíslason, Veblen, and Livi, 1993) identified several magnesium-bearing phases (kerolite, chrysotile, talc, and smectite) along with quartz, calcite, kaolinite, and amorphous precipitates. Early formation of magnesian minerals in these experiments (including smectite) con-

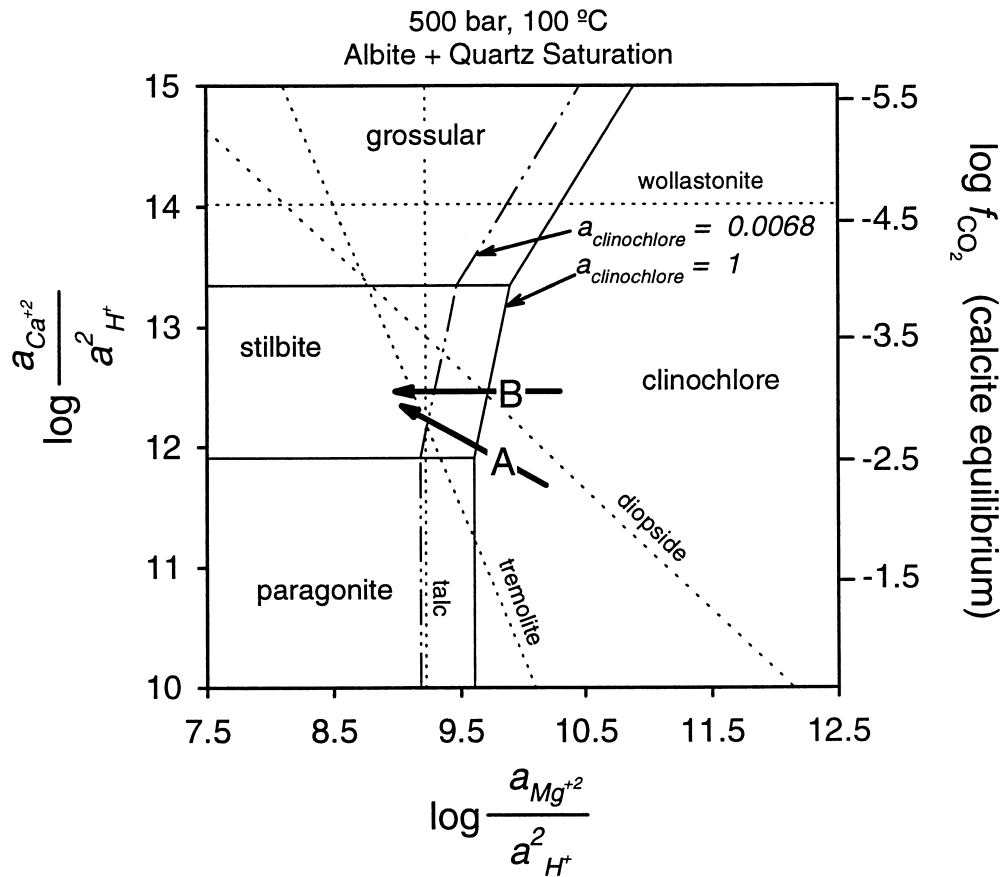


Fig. 14. Stabilities of minerals in the system $\text{MgO-CaO-Na}_2\text{O-Al}_2\text{O}_3\text{-SiO}_2\text{-H}_2\text{O-HCl}$ at 100°C , 500 bar in equilibrium with quartz and albite as functions of $\log [(a_{\text{Ca}^{+2}})/(a_{\text{H}^+})^2]$ and $\log [(a_{\text{Mg}^{+2}})/(a_{\text{H}^+})^2]$ in an aqueous solution with $a_{\text{H}_2\text{O}} = 1$. Diagram calculated using thermodynamic properties and algorithms in the computer code SUPCRT92 of Johnson, Oelkers, and Helgeson (1992), with addition of data for stilbite ($\text{CaAl}_2\text{Si}_7\text{O}_{18} \cdot 7\text{H}_2\text{O}$; ΔG_f° (cal/mol) = -2381234; ΔH_f° (cal/mol) = -25588306; S° (cal/molK) = 191.41; V° (cm³/mol) = 332.61; C_p° (cal/molK) = $154.12 + 0.168 \text{ T(K)} - 10860000 \text{ T(K)}^{-2}$; Neuhoﬀ, unpublished data). The diagram was constructed for conditions approximating the maximum burial temperature and lithostatic pressure at Teigarhorn (see text). The right-hand axis gives values of $\log f_{\text{CO}_2}$ in equilibrium with calcite. Phase boundaries between Al-bearing minerals are shown as solid lines. Saturation surfaces for stoichiometric diopside, talc, wollastonite, and tremolite are depicted as dotted lines. Two sets of phase boundaries involving clinochlore are depicted in figure 12: one for endmember clinochlore ($a_{\text{clinochlore}} = 1$; solid lines) and one corresponding to $a_{\text{clinochlore}} = 0.0068$ (dash-dot-dot lines), the activity of clinochlore in sample 94-94 (table 2) assuming ideal mixing (Holland, Baker, and Powell, 1998). Prehnite is not depicted on the diagram as the thermodynamic properties reported for this phase in Johnson, Oelkers, and Helgeson (1992) imply that it is stable relative to all Ca-zeolites, conflicting with experimental and field evidence indicating that Ca-zeolites are stable relative to prehnite at this temperature and pressure. Trends A and B are discussed in the text.

summed Mg, causing the value of $\log [(a_{\text{Ca}^{+2}})/(a_{\text{Mg}^{+2}})]$ to increase as inferred above from natural parageneses.

Fluid compositions in local equilibrium with Stage II mineral assemblages were predicted using Version 8 of the computer code EQ3NR (Wolery, 1991) assuming heterogeneous equilibrium at 100°C , 500 bar between clinochlore, stilbite, quartz, and albite. Additional constraints were provided by fixing the concentration of Cl^- to 0.001 (representative concentration in meteoric-water dominated geothermal systems in Ice-

land; Arnórsson, Sigurdsson, and Svavarsson, 1982; Arnórsson, Gunnlaugsson, and Svavarsson, 1983) and assuming that the activity of Ca^{+2} was constrained by the absence of grossular and paragonite in the assemblage (fig. 14). Resulting fluid compositions were basic (pH 8.78–9.20) with activities of Ca^{+2} ($\log a_{\text{Ca}^{+2}} = -5.59$ to -4.99) that are 2 to 3.5 orders of magnitude higher than the activity of Mg^{+2} ($\log a_{\text{Mg}^{+2}} = -8.44$ to -7.89). Calculated values of $\log [(a_{\text{Ca}^{+2}})/(a_{\text{Mg}^{+2}})]$ agree well with values computed using compositions of Icelandic geothermal fluids at temperatures near 100°C (Arnórsson, Sigurdsson, and Svavarsson, 1982; Arnórsson, Gunnlaugsson, and Svavarsson, 1983). The absence of calcite during Stage II requires that $\log f_{\text{CO}_2}$ was less than -2.5 bars (fig. 14).

Porosity evolution.—Hydrogeochemical modeling of reactive fluid transport (Rose, 1995; Manning, Ingebritsen, and Bird, 1993) and field observations (Manning and Bird, 1991, 1995; Robert and Goffé, 1993; Bevins, Rowbotham, and Robinson, 1991; Schmidt, 1990; Schmidt and Robinson, 1997) demonstrate the importance of porosity (Φ) and permeability (k) in controlling mineral parageneses in low-grade metabasalts. In particular, the magnitude of k influences the extent and distribution of chemical alteration during hydrothermal and metamorphic processes (Norton, 1988; Rose, 1995). In regions of large Φ and k , high fluid fluxes promote disequilibrium conditions that can influence the locations of mineral isograds (Lasaga and Rye, 1993; Lasaga, 1989; Bolton, Lasaga, and Rye, 1999; Dewers and Ortoleva, 1990) and promote metastable mineral formation (Steefel and Van Cappellen, 1990; Steefel and Lasaga, 1994). Consequently, measurements of Φ and k are critical for modeling mineralogical alteration during fluid flow (Norton, 1988; Norton and Knapp, 1977). In lavas, these properties are controlled by complex sequences of pore forming events (for example, degassing of the lava to form vesicles during eruption, brittle deformation after extrusion, dissolution of primary minerals during metasomatism) and reduction of Φ through mineral precipitations. Typical laboratory measurements of Φ (Norton and Knapp, 1977) in altered rocks reflect the cumulative effects of these processes. Laboratory measurements on altered rocks typically underestimate k during active stages of hydrothermal and metamorphic flow systems because much of the Φ is occluded by secondary minerals (Norton, 1988). Although laboratory measurements of initial Φ on unaltered rocks or outcrop measurements of pore abundance (such as those conducted in this study) can in part mitigate these problems, it is clear that Φ and k change dramatically with time (generally decreasing) during progressive infilling of pore spaces during mineralogical alteration (Ague, 1995). In addition, systematic decreases in k with depth in the crust (Manning and Ingebritsen, 1999) can at least partially be attributed to mineral precipitation. Generation of secondary Φ associated with mineral dissolution during alteration (Ortoleva and others, 1987) also complicates assessment of Φ and k at various stages in the alteration history of crustal rocks.

The data in tables 7 and 8 provide quantitative estimates of the amount of porosity created during volcanism and brittle deformation (Φ_1^* and Φ_2^* , respectively) and of the amount of Φ_1^* destroyed through mineral precipitation. These data are summarized in figure 15, which depicts the evolution of Φ at Teigarhorn as a function of the three stages in mineral paragenesis. Two main pore forming events influenced the lavas at Teigarhorn: creation of primary Φ during extrusion and solidification of the lavas and brittle deformation during dike emplacement. Vesiculation of the lavas resulted in a heterogeneous distribution of Φ_1^* , with flow tops having as much as 22 percent Φ_1^* and flow centers being essentially non-porous. Sporadic development of Stage I alteration decreased the value of Φ by ~ 8 percent on average. Precipitation of Stage II C/S during burial resulted in a loss of an additional 40 percent of Φ . Stage II zeolites filled most of the remaining volume of Φ_1^* . Development of Stage III alteration required addition of 3 to 11 percent total porosity (Φ_2^*) through brittle deformation. Quantitative measurements of the degree of filling of Φ_2^* by Stage III minerals were not conducted. However, outcrop observations

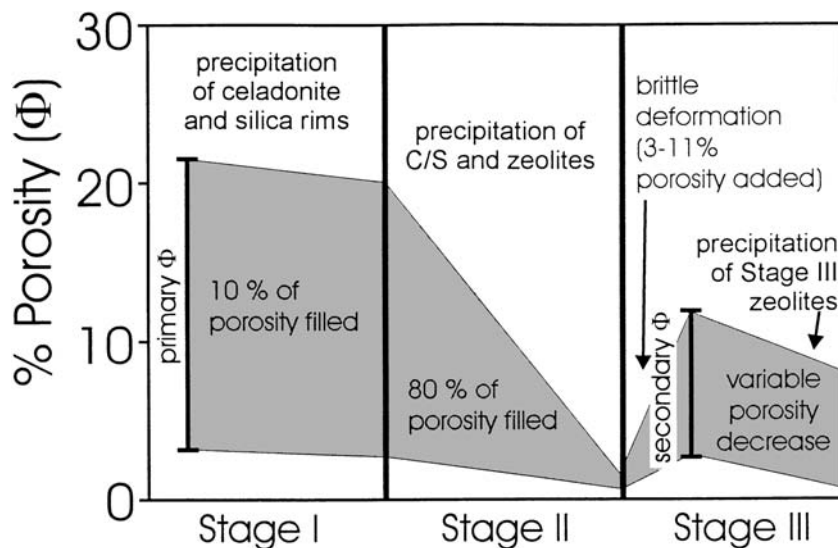


Fig. 15. Variation of bulk porosity (percent) in lavas at Teigarhorn as a function of paragenetic stage (based on the measurements in tables 7 and 8). Time elapsed after eruption increases to the right. Vertical brackets illustrate the ranges in total primary and secondary porosity determined by outcrop and thin section measurements. The gray region illustrates changes in the range of bulk porosity values realized through geologic time, with the relative height and absolute position of the boundaries of this region illustrating the variation in and magnitude of porosity.

suggest that the percent of Φ occluded by mineral precipitation during Stage III was smaller than that in Stage II as evidenced by the abundant open cavities remaining in breccias and fractures that host museum-grade zeolite, quartz, and Iceland spar crystals.

The textures and extent of infilling of primary pore spaces during Stages I and II suggest that although Φ decreased during alteration, the matrix of the lavas was sufficiently permeable to permit transport of chemical components necessary for precipitation of observed mineral assemblages. As noted above, mass transfer must have occurred from the matrix of the lavas to primary pore spaces. This apparently was true even after pore walls were coated with Stage I silica and celadonite and Stage II C/S. With the exception of scoreaceous zones, where connections between pore spaces are readily observable, much of the Stage I and II pore filling occurred in effectively isolated vesicles. Vesicles hosting Stage II minerals were thus only partially sealed by precipitation of Stage I minerals and Stage II C/S. The nearly complete infilling of Φ_1^* during Stage II was particularly acute because of the large molar volumes of zeolites and smectites. Subsequent alteration was limited by the lack of pore space for storage and migration of fluids, except where brittle deformation locally increased Φ , resulting in Stage III alteration.

Although Φ was larger in flow tops at the onset of Stage II than in Stage III (table 7), fractures conducting fluid flow during Stage III are often laterally continuous in outcrop for as much as 10 m and associated with irregular porous metasomatized aureoles (figs. 10, 11C and D). Thus, k during Stage III was probably much greater initially than in porous zones occupied by Stage I and II minerals, despite lower values of Φ (table 7). The extensive metasomatism producing the vuggy SiO_2 -rich zones near Stage III pores was likely a consequence of high water/rock ratios possible in large, conductive fractures. Late precipitation of zeolites closed the fractures, resulting in a decrease of both Φ and k . The duration of convective fluid and heat transport during Stage III was probably limited by declining k due to fracture filling.

CONCLUDING REMARKS

The mineral parageneses observed in eastern Iceland and other low-grade metabasalt localities often result from multiple stages of alteration that can be difficult to distinguish. However, as shown in figure 13, the textural and mineralogical signatures of low-grade metamorphism can be interpreted by relatively simple paragenetic schemes that account for the tectonic history of the lavas. It is also readily apparent that mineral precipitation and tectonic events accompanying the paragenetic stages depicted in figure 13 profoundly influence the hydrogeologic properties of basaltic crust (fig. 15).

An example of the importance of petroctectonic interpretations of zeolite-facies mineral parageneses is the “zone of abundant zeolites,” described by Walker (1960b) as the deepest exposed mineral zone developed in non-olivine normative tholeiitic lavas in eastern Iceland. The assemblage described by Walker (1960b) in this zone (“magnificent specimens” of heulandite, stilbite, scolecite, epistilbite) is similar to the integrated mineralogy of Stage II and III alteration found at Teigarhorn, which Walker (1960b) cited as a type locality for the “zone of abundant zeolites.” Inspection of figures 3 and 4 in Walker (1960b) suggests that the vertical extent of the “zone of abundant zeolites” is correlated to dike distribution along the northern shore of Berufjörður just as the distribution of Stage III alteration at Teigarhorn is associated with dikes. Based on these comparisons, it appears that the “zone of abundant zeolites” reported by Walker (1960b) represents hydrothermal aureoles around dike swarms and is not a signature of burial metamorphism.

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