46. ALTERATION OF BASALTS FROM DEEP SEA DRILLING PROJECT LEGS 51, 52, AND 53, HOLES 417A AND 418A

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INTRODUCTION

The ability to sample material from Legs 51, 52, and 53, together with the high recovery of cores during each leg, has enabled a large selection of samples to be used to study the following: (a) the change in intensity of alteration with depth, (b) zones of alteration, (c) the change in alteration minerals with depth, (d) the gross elemental fluxes involved in their alteration, and (e) a comparison with material of approximately the same age but with markedly different degrees of alteration.

In general, the findings of this study are in line with those of previous work on low temperature alteration of basalts, published in previous *Initial Reports* (Volumes 17, 26, 34, 35, 37, and 49), as well as other reports in this volume and elsewhere (Andrews, 1977; Baragar, 1977; Peterson, 1978; Robinson et al., 1977; Scarfe and Smith, 1977; Scott and Hajash, 1976).

Noteworthy is the discovery of chlorite at two levels in Hole 418A, and the likelihood of an Mg feedback to the oceans, found by comparing the chemistry of the clay minerals in both holes.

SAMPLES AND METHODS

Around 100 samples chosen for study were taken from the following areas: (a) obvious alteration shown by green or brown coloration, (b) high density of amygdules, and (c) high vein density. The resulting bias of this sampling should be borne in mind when reading this report.

Some 70 thin sections were examined by petrological microscope, and subsequent analysis using microprobe equipment with energy-dispersive spectrometry (Department of Mineralogy and Petrology, Cambridge University, England) yielded around 750 14-element analyses (oxygen by difference). Dental tools were used to remove material from the whole rocks for X-ray diffraction studies by 114.6-mm Debye-Scherrer camera using Co $K\alpha$ radiation and an Fe filter. Ferrous iron was determined by normal colorimetric means.

SECONDARY MINERALOGY

Clays

In hand specimen, the clays are one of several shades of green, lining or completely filling vesicles and veins. In some cases, clays replaced feldspars or glass, until constrained by the crystallization of calcite or (in places) chalcedony.

In thin section, they are yellow, green, and pale or medium brown, with a fibrous or platy texture growing out from the walls (Plate 3, Figures 1 and 2). With cross-po-

larized light, one can see the spherical pattern of growth (Plate 1, Figure 5) where space permits. In general, green clay forms the rim of the vesicle and the yellow portion of the core; but sometimes this is reversed as observed by Robinson et al., 1977; or the sequence is green, yellow, and green once more. A good example of this zoning phenomena is shown in Plate 2, Figures 1 and 2. Here, K and Fe have migrated from the main vein (top) through the glass and not only altered the existing clay chemically (see Table 1), but also formed a green clay on the inner edge, before being truncated by calcite. It is interesting to note the presence of chlorine in the analyses, which is useful in forming soluble complexes, e.g., Mg(OH)Cl.

A plot of Hole 418A clay compositions in terms of their colors (not presented here) in thin section shows that, in general, the green clays have celadonite compositions whereas the yellow and brown clays have those of saponite.

It seems that the first clay formed was the yellow variety (trioctahedral saponite), produced by the interaction of hot fresh basalt with seawater and coating all available free surfaces. Later, once pervasive alteration of the interiors of the pillows had increased the ion activities sufficiently, green clay (dioctahedral, proto-celadonite, or 1 Md glauconite, according to Buckley, in press, started to form. The conditions must have been reducing at this time, maintaining a high activity of Fe ions and enabling both K and Fe to diffuse into the saponite. At first, only the chemistry is changed but structural alterations occur later, producing a dioctahedral celadonite (Figures 1, 2, 3, and 4; Tables 2 and 3), presumably via the "layer lattice theory" mechanism described by Elderfield (1976).

These basalts are very low in K (from shipboard studies of fresh rock), any excess being derived from the seawater.

TABLE 1
Microprobe Analysis of Clay Minerals in Elongate Zoned Vug (see Plate 2)

%	1	2	3	4	5	6
SiO_2	46.2	53.6	44.8	50.6	36.0	50.9
Al_2O_3	9.28	7.06	8.26	4.57	12.3	18.1
TiO_2	–	–	–	–	1.33	1.72
FeO*	12.0	14.5	14.3	21.8	27.1	4.39
MgO	11.2	6.27	11.2	4.22	1.24	1.82
CaO	0.60	0.36	0.54	0.36	2.98	6.02
Na_2O	0.53	–	–	–	1.41	1.86
K_2O	3.01	8.72	3.83	8.46	4.67	4.26
Total	82.98*	90.65*	83.13*	89.92	87.11	90.25

Note: FeO* = total iron; – = not detected; 1 = saponite at right end of vug; 2 = proto-celadonite at left; 3 = saponite at left; 4 = proto-celadonite far left; 5 = devitrified glass; 6 = glass; * by total indicates chlorine (< 0.1%) included; Sample 418A-15-4, 46-53cm.
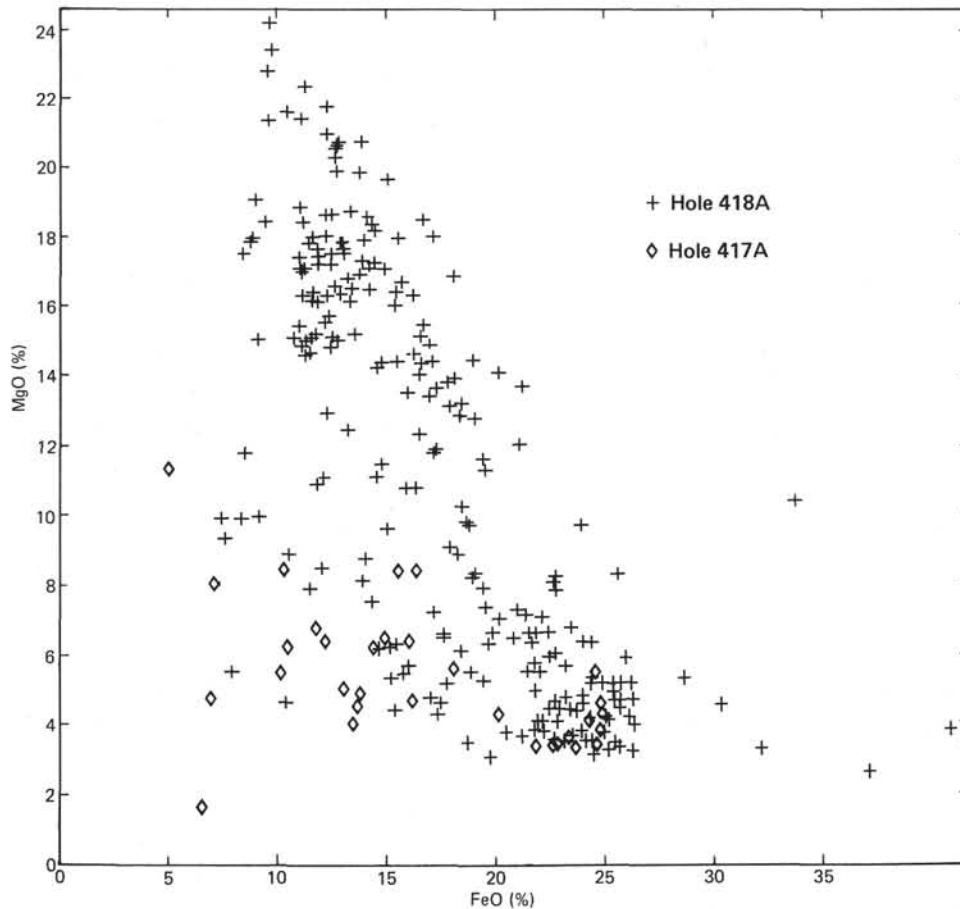
Figure 1. *Clays from Holes 417A and 418A. Those with high iron probably contain an iron oxyhydroxide impurity. Notice all clays from Hole 417A have low MgO.*

Thus, its removal in such large quantities in clays, feldspars, and zeolites must be included when considering its geochemical budget, a point already mentioned by Scarfe and Smith (1977). In contrast, Fe comes from both devitrifying glass and the altering of titanomagnetites (Peterson, 1978), as iron is of low activity in the oxidizing seawater.

Saponite and proto-celadonite occur in both Holes 417A and 418A, but celadonite only occurs in the former, where the most altered rocks were drilled. If, as Donnelly (1978) suggests, Hole 417A is in an area affected by an "upside-down drain" or a hydrothermal exhaust for the surrounding area, then celadonite is the end-member of an alteration sequence for clays in this instance. A comparison of all the clay analyses from both holes (Figures 1, 2, 3 and 4) and some representative analyses (Table 3) shows that a drastic decrease in magnesium occurs and, as it does not form another phase, must return to the ocean.

As areas typified by Hole 417A are considered to be unusual, "the magnesium problem" envisaged by Drever (1974) will continue to be solved by the formation of saponite, proto-celadonite, and chlorite (Elderfield, 1977; Perry, 1975).

Chlorite

Chlorite was found only in two areas in Hole 418A, an upper level (Sections 33-1 to 37-1), and a lower level (Sections 85-2 to 86-5). Representative microprobe

analyses and X-ray diffraction data are given in Tables 2 and 4.

FeO in the one sample analyzed so far was 4.9%. As this presently must be considered a minimum, the chlorite may be a delessite or a later oxidized penninite or talc-chlorite (Deer et al., 1962, 1963). It occurs in veins (together with potassium feldspar) pseudomorphs, and plagioclase, especially in the upper areas of the hole.

In hand specimen, it is pale green and, thus, difficult to distinguish from clay minerals. In thin section, it is easily recognized by its radial extinction (Plate 1, Figure 6) and anomalous low birefringence colors. Spatially, this chlorite occurs after saponite and before calcite and associated iron oxyhydroxide.

Other occurrences of chlorite in oceanic basalts have usually been in dredged rocks from transform fault zones (Humphris, 1976) and Icelandic drill holes (Kristmannsdöttir, 1975). These studies suggest a formation temperature of about 230°C. These areas in Hole 418A may have been ducts for hydrothermal and later, cooler oxidizing fluids. No other hydrothermal minerals are to be found, and celadonite is notable by its absence, perhaps having been converted to chlorite (Deer et al., 1962).

Carbonates

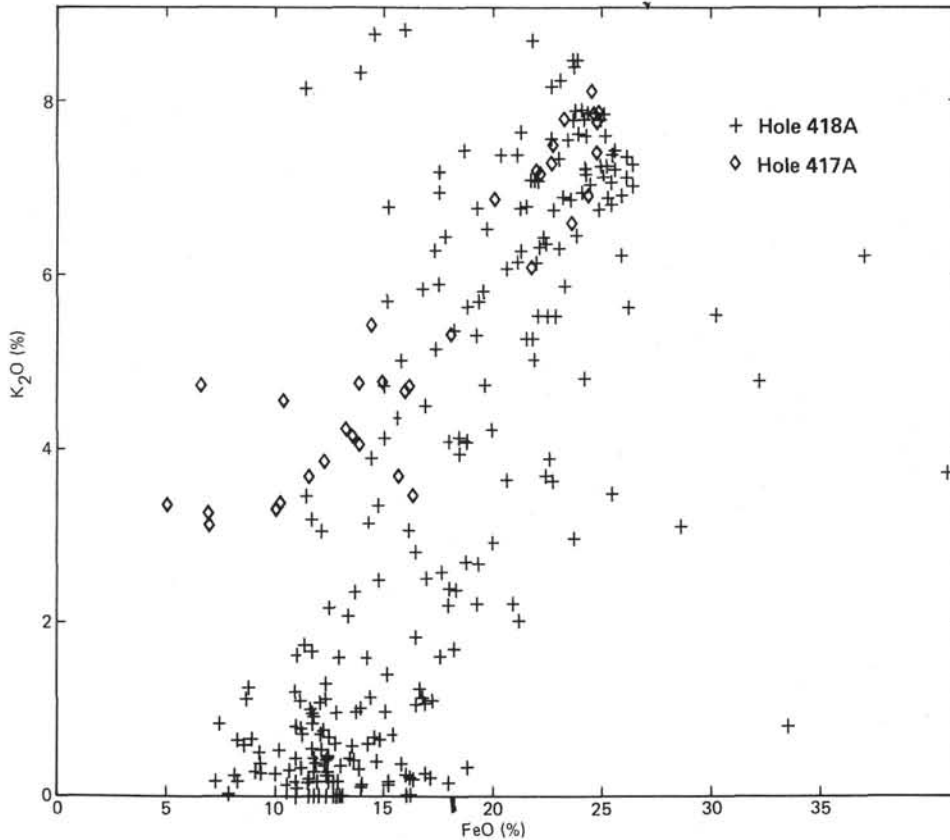The only carbonate in the samples is calcite, although it consistently contains small quantities (<1%) of Ni, Mn,

Figure 2. *Clays from Holes 417A and 418A. Notice the low $K_2O\%$ and FeO% group of Hole 417A clays.*

Mg, Si, and Fe. It occurs late in the alteration sequence, and occupies any remaining space in the samples, e.g., vesicle and vein centers, as well as replacing altered olivine or feldspars. Occasionally, one vein is seen on close examination to consist of two, side by side, separated by fragments of clay, pyrite, and silica (Sample 418A-24-2, 38-40 cm).

The rapid alteration on the basalt glass at a late stage (Scott and Hajash, 1976) seems to provide not only calcium in addition to that supplied by feldspar breakdown, but also hydroxyl ions (Bischoff and Ku, 1970). These hydroxyl ions displace the normal bicarbonate-carbonate equilibrium such that large quantities of calcite are able to form. This process is probably assisted by the removal of Mg ions from solution (into clays) which normally inhibit calcite nucleation.

Feldspars

These are of two generations, i.e., large phenocrysts and small microlites. The microlites usually contain inclusions of original melt at their centers, and it is the phenocrysts which are altered, especially in the higher calcium centers. Unaltered, the feldspars are labradorite which tend toward a clay composition, or very pure potassium feldspar containing only K, Si, and Al in the microprobe analyses. Potassium feldspar occurs occasionally (down to Section 418A-41-1) and is especially abundant in association with chlorite. In Hole 417A, the majority are potassium feldspar

(adularia by XRD) with the remaining few altered toward a clay composition. At the time of this writing, more samples await examination.

Zeolites

Phillipsite (Table 2) and analcite occur in both holes, and natrolite occurs in Sample 418A-83-3, 75-80cm (Table 5, Figure 5). They usually occur at pillow margins, in large veins, and (space permitting) in the center of vesicles. They are usually found fairly late in the alteration sequence, when the permeability has fallen such that the ion activities are sufficient either to inhibit the formation of smectites or convert them to zeolites (Velde, 1977). Alkaline conditions prevail due to the continued alteration of basalt glass in the vicinity. From the rule of Cornu and the parallel reasoning of Elderfield (1976), the presence of the less-hydrated zeolites (natrolite and analcite) would seem to support the occurrence of higher temperatures in Hole 417A (Donnelly, 1978) and some levels of Hole 418A. A red euhedral zeolite, as well as a red powdery vein filling from Sample 418A-33-1, 2-8 cm, were discovered to be phillipsite; no mordenite, seen by shipboard scientists, has been analyzed as yet.

Opaques

Few opaques were examined, but those that were proved to be pyrite or chalcopyrite. They seemed primary, except fragments of pyrite seen in a calcite vein in Sample 418A-24-2, 38-43 cm.
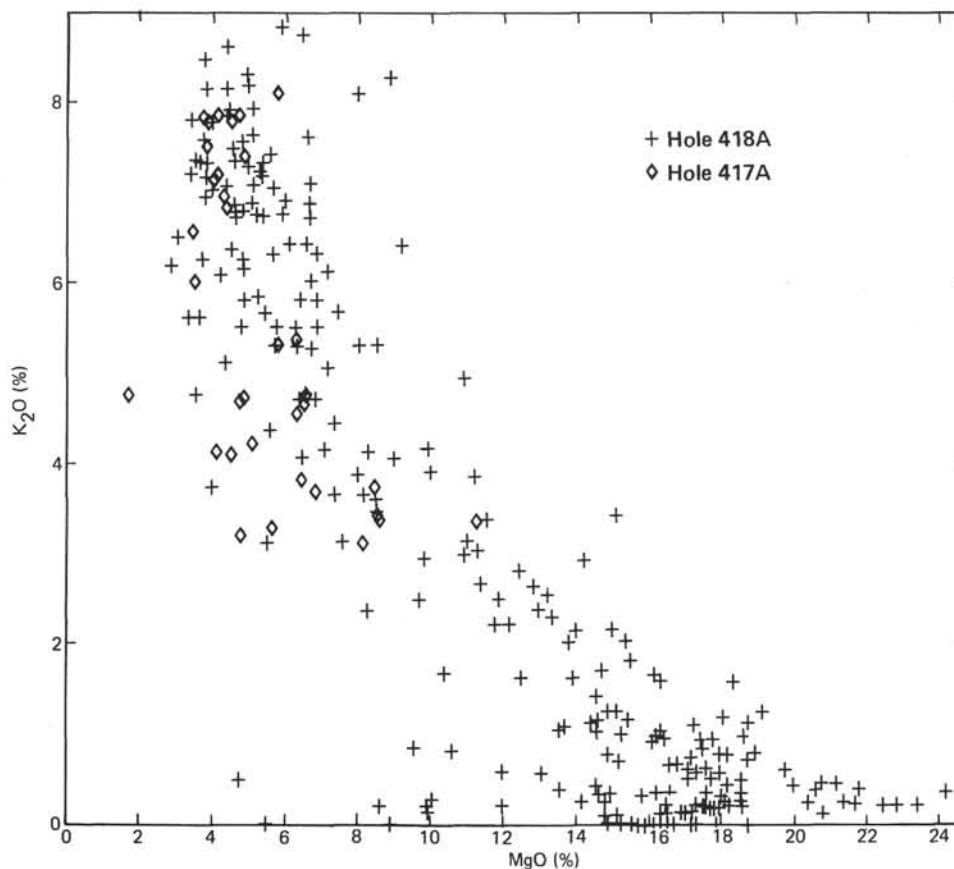
Figure 3. *Clays from Holes 417A and 418A. Notice all clays from Hole 417A contain > 3% K$_2$O.*

Silica

Silica is found only in Hole 418A from Sections 56-6 to 73-6, and occurs as a vein filling and pseudomorphing feldspars. It is clear in plane light with second-order birefringence colors and radial extinction. The analyses are close to 100 per cent silica and, thus, the mineral is probably chalcedony (as opal usually contains a few per cent water).

Iron Oxyhydroxide

This is found throughout samples from both holes. It is usually dark red in thin section and isotropic, with an amorphous XRD pattern. It is derived from ferrous iron in solution, which is precipitated as the system becomes oxidizing. This explains why it is found mainly in or near breccia regions and large veins, usually predating calcite.

SUMMARY

This report can be summarized as follows:

a) The initial high temperature alteration product is a high Mg, trioctahedral saponite.

b) This is followed by, or changed to, a low-temperature dioctahedral "proto-celadonite" which eventually forms a celadonite *sensu stricto*.

c) The decreased permeability in some areas results in the formation of zeolites, and chlorite associated with

potassium feldspar forms where higher temperatures prevail.

d) The system goes oxidizing and iron oxyhydroxide precipitates, closely followed by calcite.

In some areas the above processes are not sequential and mixtures of alteration minerals are obtained.

e) Potassium, magnesium, and bicarbonate ions are removed in large quantities from seawater during the alteration process. Under extreme conditions, magnesium may be returned to the oceans, as shown by comparing the chemistry of the clays in Holes 418A and 417A (Figure 1).

f) The clays do not seem to change in any systematic manner downhole. Both types of clays are found throughout, with the intensity of alteration concentrated in the breccia zones.

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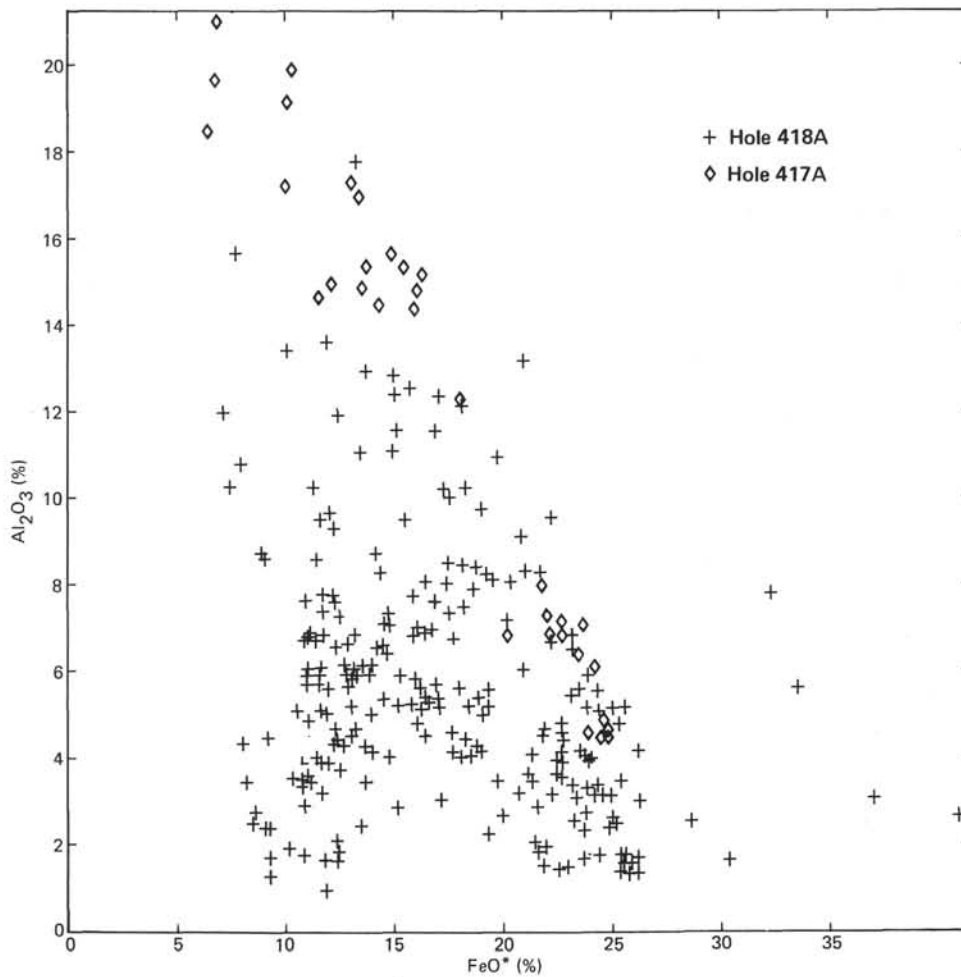
Figure 4. *Clays from Holes 417A and 418A. Notice the high alumina group from Hole 417A.*

TABLE 2
Representative X-Ray Powder Camera Patterns

Phillpsite		Saponite		Proto-Celadonite		Celadonite		Chlorite	
418A-33-1, 2-8 cm		418A-57-1, 68-69 cm		418A-33-1, 2-8 cm		417A-41-3, 2-11 cm		418A-37-1, 63-65 cm	
d-Å	I	d-Å	I	d-Å	I	d-Å	I	d-Å	I
8.22	m	15.8	s	14.3	s	10.0	vs	14.3	vs
7.19	vs	11.7		9.96		4.53	vs	7.19	s
6.43	m	4.58	s	4.55	s	4.36	m	5.11	s
5.40	m	4.13	s	4.29	m	4.12	m	4.55	s
5.08	m	3.17	m	2.58	s	3.67	w	4.13	w
5.01	m	3.05	d	2.40	m	3.33	m	3.58	s
4.32	w	2.64	d	1.51	m	3.09	w	2.87	m
4.14	s	2.48				2.68	w	2.58	w(d)
3.96	w	2.29	w			2.58	vs	2.41	m(d)
3.69	w	1.74	w			2.43	w	1.53	s
3.48	w	1.53	s			1.51	s	1.50	m
3.27	m								
3.26	m								
3.19	vs								
3.17	s								
3.10	w								
2.95	m								
2.75	m								
2.70	m								
2.55	m								

Note: Produced by Co K radiation, using Fe filter, vs = very strong, s = strong, m = medium, w = weak, d = diffuse.

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TABLE 3
Representative Microprobe Analyses of Clay Minerals

%	417A-24-1, 86-90	417A-24-1, 86-90	417A-28-4, 54-57	417A-41-3, 2-11	418-A-33-1, 2-8	418A-46-5, 30-31	418A-46-5, 30-31	418A-70-2, 96-101	418A-60-4, 52-56	418A-86-5, 86-88	418A-16-2, 6-10	418A-36-2, 97-101	418A-42-4, 126-130	418A-73-6, 87-92
SiO_2	45.1	48.0	45.6	55.2	41.4	52.7	50.7	43.9	49.2	45.1	49.8	48.5	50.7	48.5
Al_2O_3	15.1	19.9	6.79	0.63	6.86	3.29	4.37	5.85	2.19	5.40	3.88	8.22	3.03	3.13
TiO_2	0.27	–	–	–	–	–	–	–	–	–	–	–	–	0.17
FeO*	16.3	10.3	22.1	20.8	11.2	10.8	12.3	12.8	19.3	16.3	24.0	21.7	24.1	22.2
MgO	8.46	6.28	4.01	6.65	17.2	22.4	20.3	17.7	7.49	12.4	3.62	3.94	4.36	6.78
CaO	0.39	0.87	0.75	–	0.97	1.66	2.18	2.91	0.94	2.41	0.26	1.19	0.38	1.92
Na_2O	–	0.30	–	–	1.81	–	–	1.15	–	–	–	–	0.50	–
K_2O	3.43	4.54	7.16	9.12	0.72	0.20	0.23	0.15	5.66	2.78	8.44	7.05	7.90	6.28
TOTAL	89.05	90.19	82.40	92.40	80.16	91.05	90.08	85.56	84.78	84.39	90.00	90.60	90.97	88.98

Cations per 22 O, OH

Si	6.79	6.90	7.54	8.40	6.84	7.46	7.33	6.92	8.07	7.29	7.95	7.55	8.00	7.76
Al	1.21	1.10	0.46	–	1.16	0.54	0.67	1.08	–	0.71	0.05	0.45	–	0.24
Al	1.47	2.26	0.86	0.11	0.17	0.00	0.07	0.01	0.42	0.31	0.68	1.06	0.56	0.35
Ti	0.03	–	–	–	–	–	–	–	–	–	–	–	–	0.02
Fe	2.06	1.23	3.05	2.64	1.55	1.27	1.49	1.69	2.64	2.21	3.20	2.83	3.18	2.97
Mg	1.90	1.34	0.99	1.51	4.23	4.72	4.38	4.15	1.83	2.98	0.86	0.91	1.03	1.62
Ca	0.06	0.13	0.13	–	0.17	0.25	0.34	0.49	0.17	0.42	0.05	0.20	0.06	0.33
Na	–	0.08	–	–	0.58	–	–	0.35	–	–	–	–	0.15	–
K	0.66	0.83	1.51	1.77	0.15	0.04	0.04	0.03	1.18	0.57	1.72	1.40	1.59	1.28

Note: – = not detected, * indicates all Fe calculated as FeO. FeO (colorimetric) analysis on a saponite and a celadonite gave 4.4% and 4.0%, respectively.

TABLE 4
Representative Microprobe Analyses of Chlorites

%	418A-33-1, 2-8	418A-36-5, 27-34	418A-36-5, 27-34	418A-37-1, 63-65	418A-83-3, 75-80	418A-85-2, 12-17	418A-85-2, 12-17
SiO_2	34.7	32.5	34.4	33.7	31.0	33.1	35.7
Al_2O_3	11.9	11.4	12.1	11.7	11.0	10.9	13.7
TiO_2	–	–	–	–	0.59	0.41	–
FeO*	16.1	12.2	12.8	10.0	9.68	13.5	8.95
MgO	20.9	22.1	23.1	25.4	19.2	23.8	24.4
CaO	0.81	0.41	0.27	0.24	1.10	0.40	0.99
Na_2O	–	–	–	0.60	–	–	–
K_2O	–	0.28	0.29	–	–	0.08	0.21
Total	84.41	78.61	82.96	81.71	72.57	82.19	84.15

Cations per 28 O, OH

Si	7.15	7.05	7.09	6.98	7.22	6.95	7.12
Al	2.88	2.93	2.95	2.86	3.03	2.69	3.21
Ti	–	–	–	–	–	0.07	–
Fe	2.77	2.22	2.20	1.74	1.88	2.37	1.49
Mg	6.42	7.15	7.10	7.84	6.65	7.45	7.21
Ca	0.18	0.10	0.06	0.05	0.28	0.09	0.21
Na	–	–	–	–	0.28	0.02	0.05
K	–	0.08	0.08	–	0.28	0.02	0.05
Total	19.40	19.53	19.48	19.71	19.33	19.64	19.29

Note: * indicates Fe calculated as FeO; FeO (colorimetric) on one sample was 4.0%.

TABLE 5
Representative Microprobe Analyses of Zeolites

% Sample (Interval in cm)	418A-79-4, 57-60 Phillipsite	417A-28-1, 25-28 Phillipsite	418A-41-1, 75-80 Analcite	418A-83-3, 75-80 Analcite	417A-30-5, 28-31 Analcite	418A-83-3, 75-80 Natrolite
SiO_2	57.7	61.3	55.3	52.6	54.1	42.0
Al_2O_3	15.2	17.8	24.8	22.3	22.3	27.1
TiO_2	1.48	–	–	–	–	–
FeO*	2.45	–	–	–	–	–
MgO	0.28	–	–	–	–	–
CaO	1.01	0.08	0.16	0.10	–	3.98
Na_2O	–	3.60	12.3	12.7	13.4	11.0
K_2O	11.4	7.37	–	–	0.09	–
Total	89.52	90.19	92.64	87.73	90.90	84.12
Cations per 72 O, OH						
Si	26.6	27.2	23.9	24.1	23.9	20.7
Al	8.28	9.29	12.1	12.1	12.2	15.7
Ti	0.54	–	–	–	–	–
Fe	0.90	–	–	–	–	–
Mg	0.01	–	–	–	–	–
Ca	0.54	0.03	0.07	0.05	–	2.16
Na	–	3.08	10.3	11.3	11.5	10.4
K	6.66	4.16	–	–	0.07	–
Z	34.88	36.49	36.50	36.20	36.10	36.40
R	9.19	7.30	10.44	11.40	11.57	14.72

Note: – = not detected; * indicates all Fe calculated as FeO; Z = Si + Al; R = 2Ca + Na + K.

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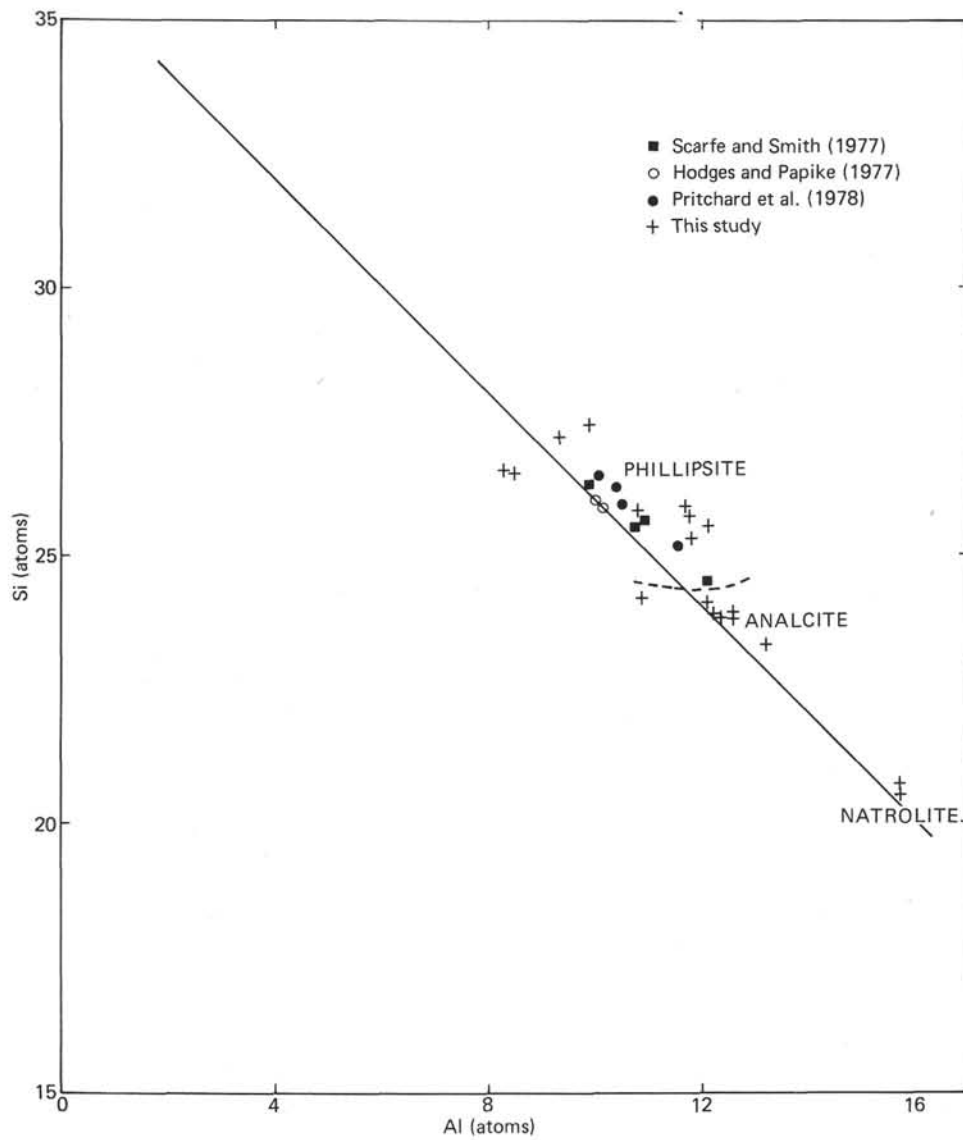
Figure 5. *Plot of zeolites from Holes 417A and 418A.*

PLATE 1

Length of bar is 0.3 mm

Figure 1   The growth of calcite in this vein changes to a more magnesium-rich (~1%) type with time. The interstices between the green clay (light) and the calcite (white) are filled by orange-brown iron oxyhydroxide (black). Sample 418A-19-7, 111-114 cm.

Figure 2   Vug lined with green clay, filled with dark red-brown iron oxyhydroxide, and truncated by later calcite. Sample 417A-28-4, 54-57 cm.

Figure 3   Two segregation vesicles. The larger filled with green clay having a brown alumina rich center. The cores of the feldspars (top right) are altering towards material with a clay chemistry. Sample 417A-28-4, 54-57 cm.

Figure 4   Vesicle rimmed with green clay which changes inward to yellow clay with a dark center. Sample 418A-59-6, 106-110 cm.

Figure 5   Vesicles with calcite hemispheres on the wall and filled with green clay with a spherical growth pattern. The center of the left vesicle is filled with calcite rimmed by yellow clay. Crossed polarized light. Sample 418A-60-4, 52-56 cm.

Figure 6   Large vein with a small vein passing through a pseudomorphed feldspar. Both veins are lined with a thin yellow clay layer with a thicker pale green chlorite layer (black-white) having a radial fibrous habit. The original feldspar is replaced by chlorite and K-feldspar. Crossed polarized light. Sample 418-37-1, 63-65 cm.
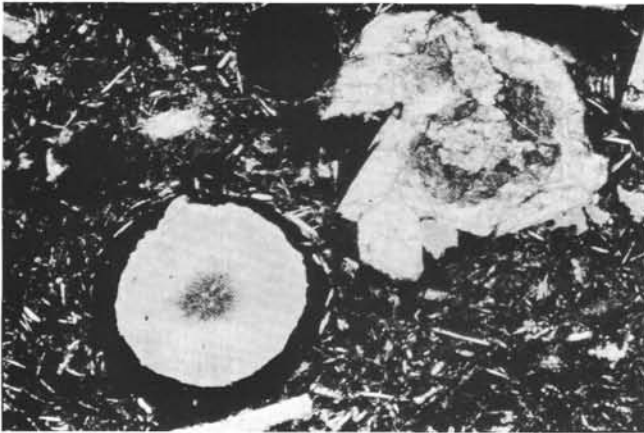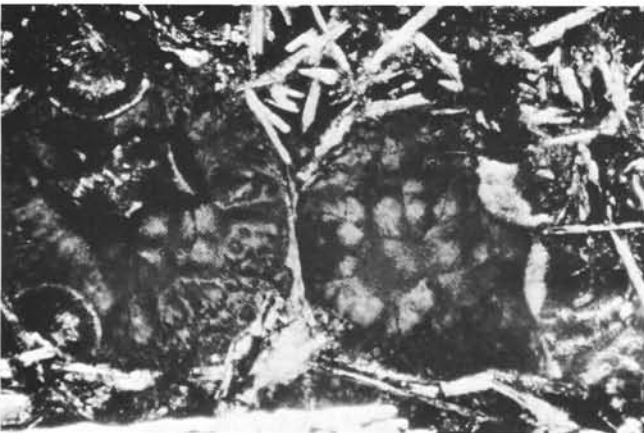
PLATE 1
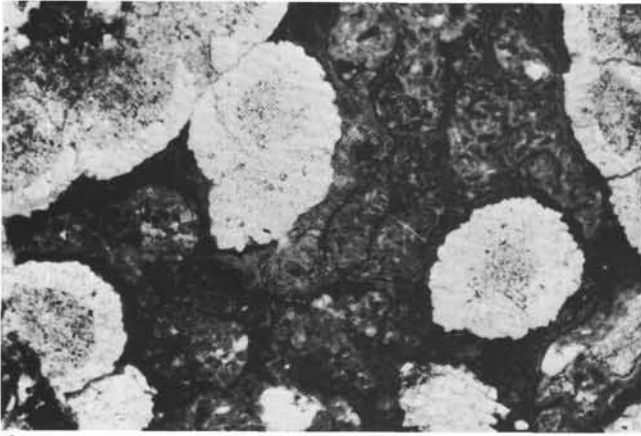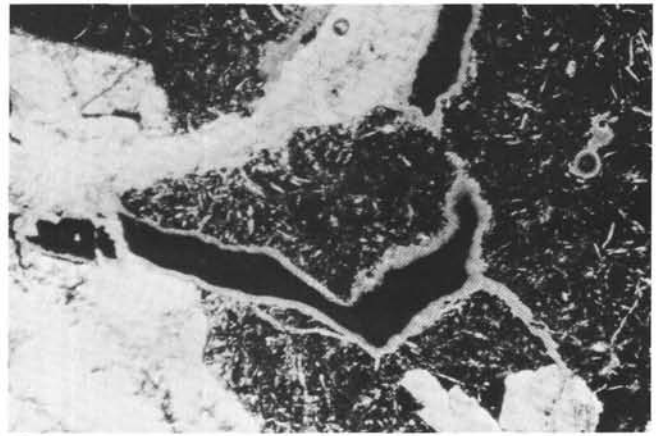
0.3mm

PLATE 2
Sample 418-A-15-4, 46-53 cm
(Scale bar is 0.3 mm)

Figure 1   Photomicrograph of an area of glass (G), and
              devitrified glass (DVG), wholly or partially engulfing
              vugs lined with yellow saponite(s), (light), which has
              been modified to green proto-celadonite (pc), (dark).
              See Table 1. Centers are calcite (c).

Figure 2   Sketch of photograph above, clarifying the layering in
              the vugs. Notice the thickening of proto-celadonite
              towards the source of the diffusion (top).
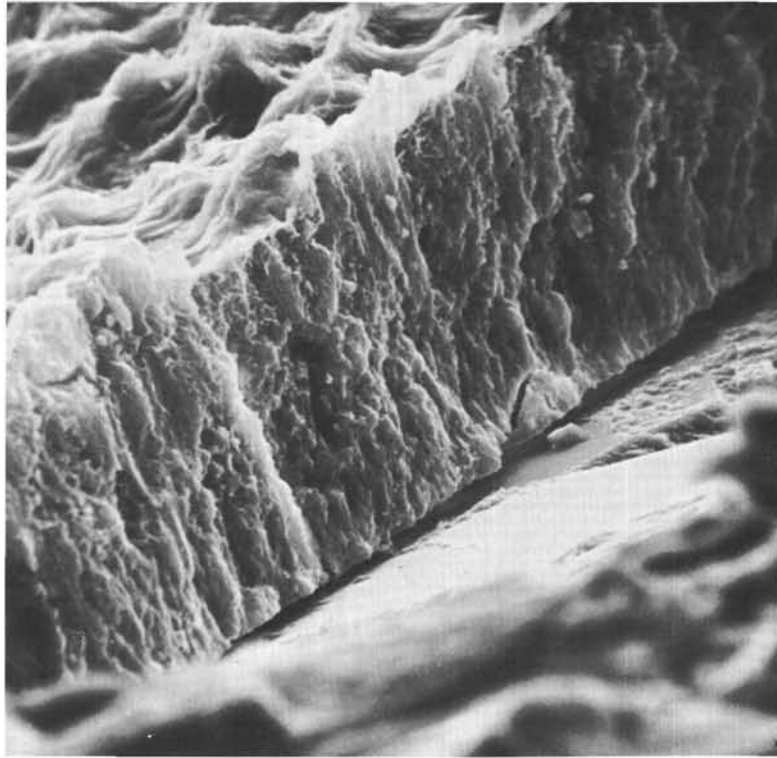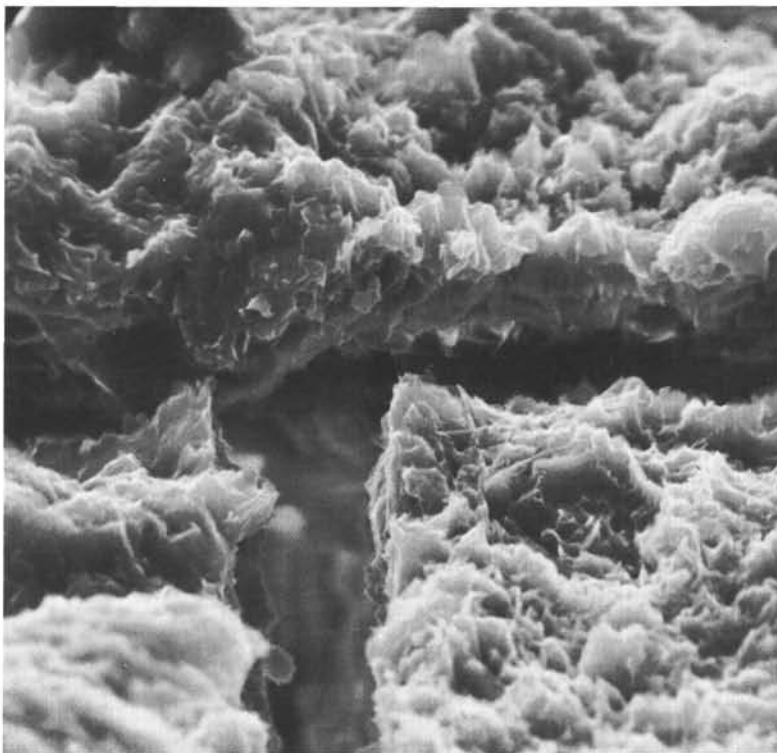
PLATE 2

1

0.3mm
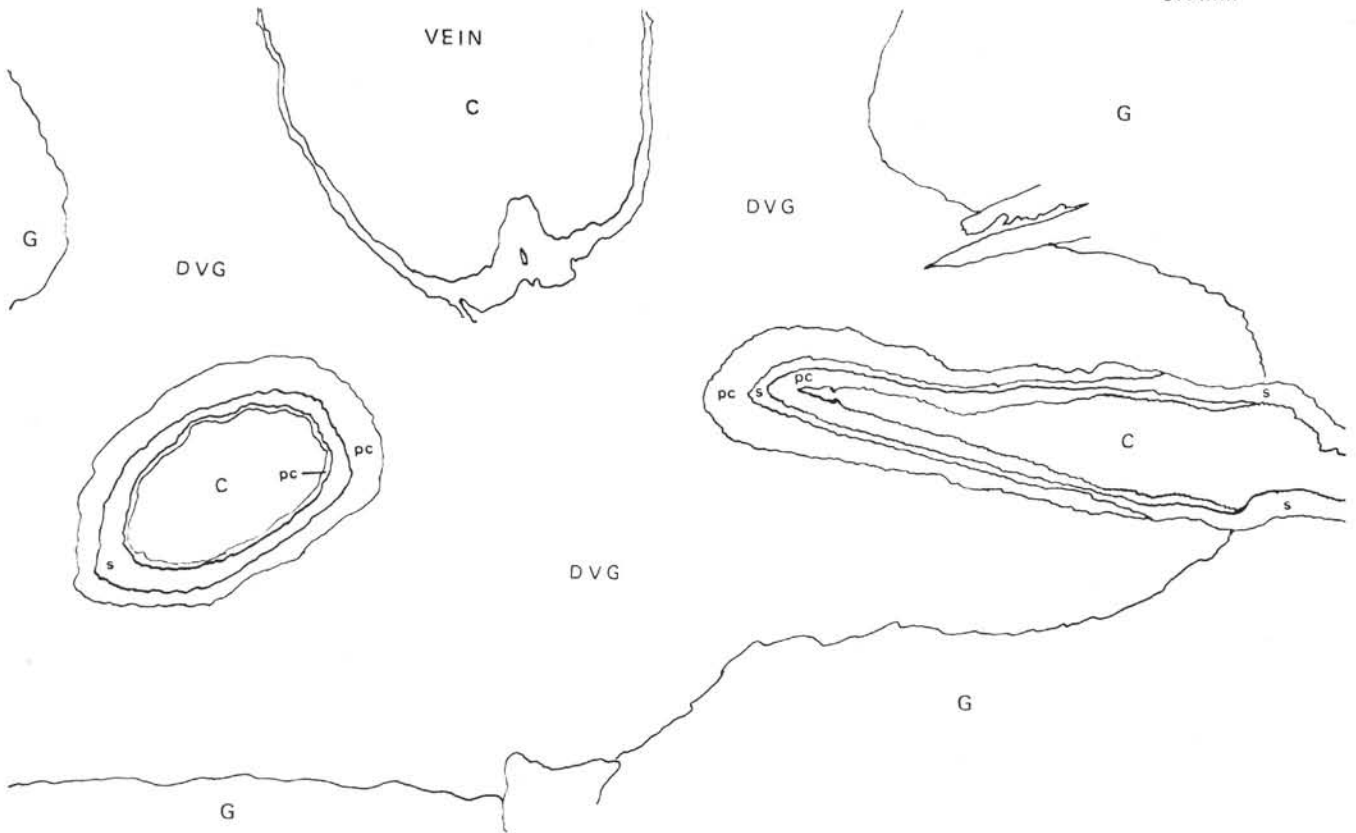
2

PLATE 3
Scanning Electron Micrographs of Clay-Lined Vesicle in Sample
418A-81-2, 51-56 cm

Figure 1   Clay growing out from smooth vesicle wall, showing
            ''egg-box'' surface structure (Andrews et al., 1977).
            1190×.

Figure 2   Cracks in clay due to dehydration under the electron
            beam, showing platy structure. 2450×.

PLATE 3

1

2