

## 51. CLAY MINERALS IN BASALTS FROM DEEP SEA DRILLING PROJECT SITES 417 AND 418

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### INTRODUCTION

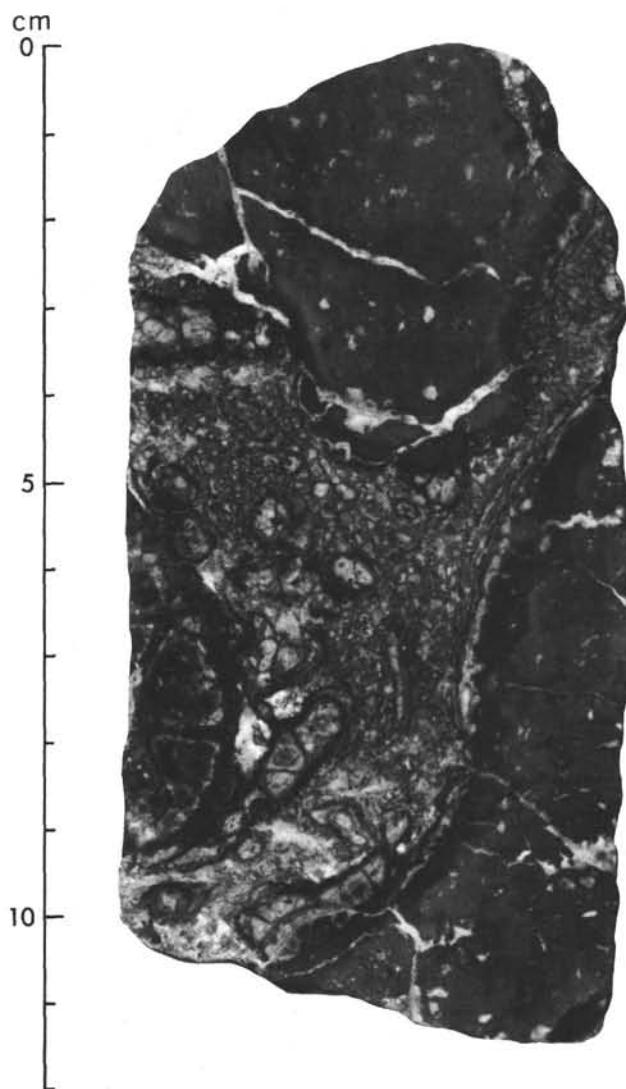
Clay minerals are common in altered rocks from Holes 417A, 417D, and 418A. Among the clay minerals<sup>1</sup> investigated in this study are smectites, celadonitic hydromica, mixed-layered silicates of mica-montmorillonite, and palagonite-like material. They are observed in the altered volcanic glass of hyaloclastites (Figure 1) and in basalts, as pseudomorphs after phenocrysts in basalts and as a fill in cracks and vesicles.

Minerals of the montmorillonite group and mixed-layered silicates were observed in nearly all samples investigated, even in samples of weakly altered rocks. Their mode of occurrence and mineral assemblages are described in this paper.

Under microscopic examination, three main types of smectites and mixed-layered silicates are distinguished: (1) colorless and light green (after plagioclase); (2) deep green (after olivine and hyaloclastite glass, rarely after pyroxene), and (3) brown (after basaltic glass). Pink montmorillonite was observed in two thin sections (Samples 417D-42-1, 3 cm and 417D-43-6, 3 cm). Type 1 is composed of fine-grained aggregates (Figure 2) which often form cross-fibrous ribbons, spherulites, and (rarely) long fibers. The mineral has the following characteristics:  $\alpha = 1.490$  to  $1.500$ ,  $\beta = 1.508$  to  $1.515$ , and  $\gamma = 1.510$  to  $1.518$ ; it is associated with K-feldspars, calcite, and more rarely with analcime and chalcedony. Type 2 occurs in various forms in the samples: as spherulites, crypto-grained masses, cross-fibrous layered bands alternating with light and dark colored layers (Figure 3), and as fibrous aggregates in pseudomorphs after olivine. Optically, this mineral is close to Type 1 with  $\alpha = 1.486$  to  $1.497$ ,  $\beta = 1.503$  to  $1.420$ , and  $\gamma = 1.508$  to  $1.525$ , but has slight pleochroism ( $y = \text{green}$ ,  $\alpha = \text{light green}$ ). It is associated with calcite, celadonite, and mixed-layered minerals. In some veins, with dark green smectite, pyrite crystals were observed (i.e., the lower portion of the cored interval in Hole 417A).

Brown and dark olive-green smectites (Type 3) usually form crypto-grained or fine-grained masses after palagonitized glass in basalt. The color is variable: bright brown, dark brown, brown, light brown, and greenish brown; however, pleochroism was not ob-

<sup>1</sup>According to J. R. Cann (personal communication), chlorite-delessite in association with  $K_{sp}$  lined some veinlets in Hole 418A. The chemical composition of the chlorite is as follows (wt. %):  $SiO_2 = 36$ ,  $Al_2O_3 = 11$ ,  $FeO = 12$ ,  $MgO = 22$ ,  $CaO = 0.3$ , and  $K_2O = 0.5$ .



Leg 51A Hole 417A Core 24 Section 2 Sample No. 1

Figure 1. *Hyaloclastite with basaltic fragments. The glass is replaced by the mixture of green smectite, calcite, and celadonite.*

served. It has higher refractive indexes than either Types 1 and 2, with  $\alpha = 1.525$  to  $1.540$ ,  $\beta = 1.550$  to  $1.560$ ,  $\gamma = 1.556$  to  $1.566$ . It is sometimes associated with celadonite.

Clay minerals were identified optically and with X-ray analysis. Some powder patterns were studied

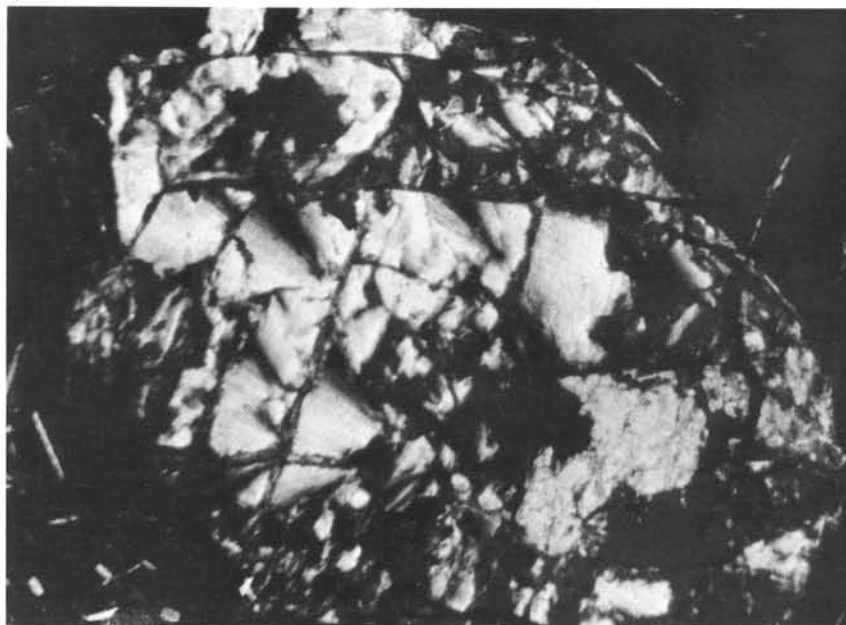


Figure 2. A part of the plagioclase phenocryst replaced by colorless fine-grained and spherulitic smectite in basalt. Sample 417A-37-3, 70 cm. Crossed nicols.



Figure 3. Fine-banded aggregate of green smectite in hyaloclastite. Sample 417-26-2, 58 cm. Crossed nicols.

(Table 1). Measurements were made with a diffractometer DRON-2 with Fe-radiation filtered with Ni; the speed of rotation was  $2^\circ/\text{min}$ .

#### CLAY MINERALOGY

According to the parameter  $d_{001}$  (Figures 4 and 5), two groups of smectites were distinguished: (1) montmorillonites and montmorillonite-like minerals ( $d_{001} = 11.9$  to  $14.0\text{\AA}$ ) and (2) hydromica ( $d_{001} = 10$  to  $10.2\text{\AA}$ ).

Phases diagnosed optically as palagonite have diffractograms close to those of montmorillonite; however, the peaks are often diffuse due to a high content of amorphous material. In Figure 6, diffractograms of "palagonite" are noted from different zones of altered glass near a smectite veinlet. Some distance from the veinlet, a glass zone is less altered and has a diffuse peak around  $13$  to  $14\text{\AA}$ . In thin section, this altered glass is brown, isotropic with low indexes ( $n = 1.523 \pm 0.003$ ),



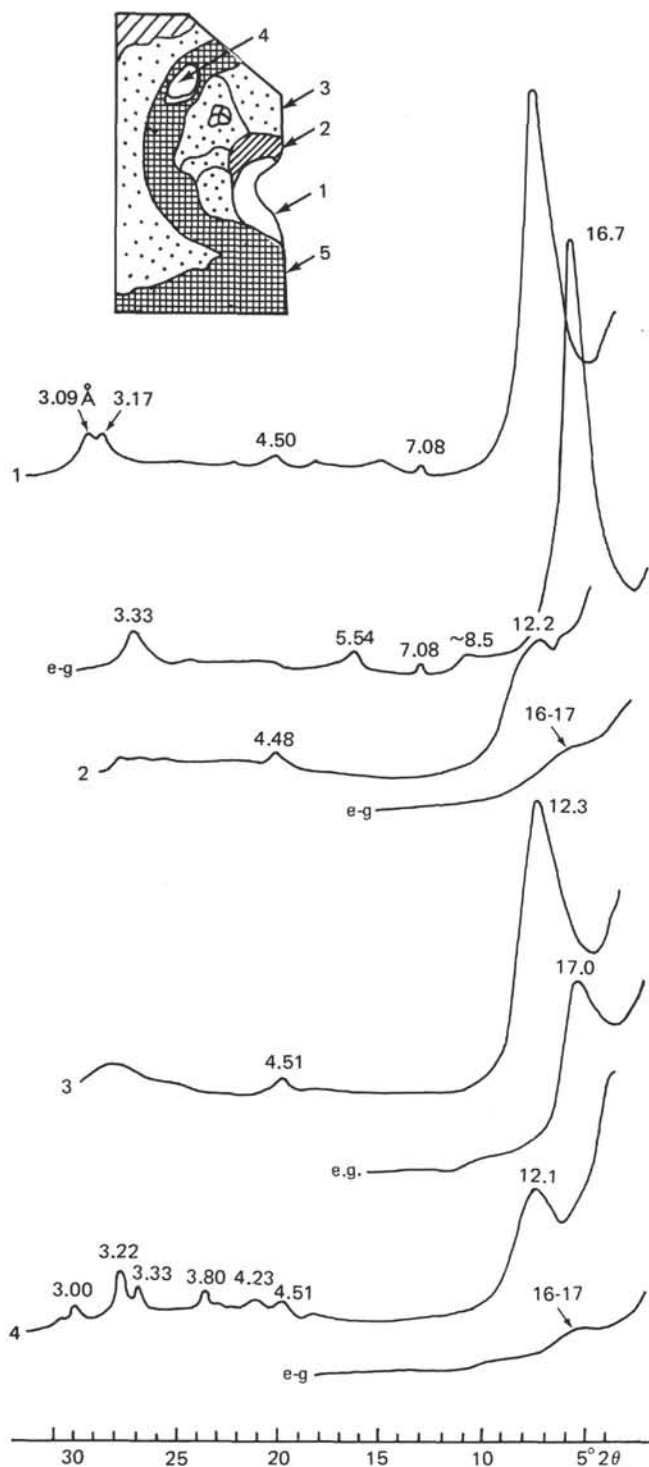


Figure 5. X-ray diffractograms of montmorillonites from Sample 417A-30-4, 122 cm. 1 = white smectite in a large cavity; 2 = white smectite in a calcite vesicle; 3 = green smectite replacing glass; 4 = white smectite in an amygdale; 5 = brown smectite with iron hydroxides.

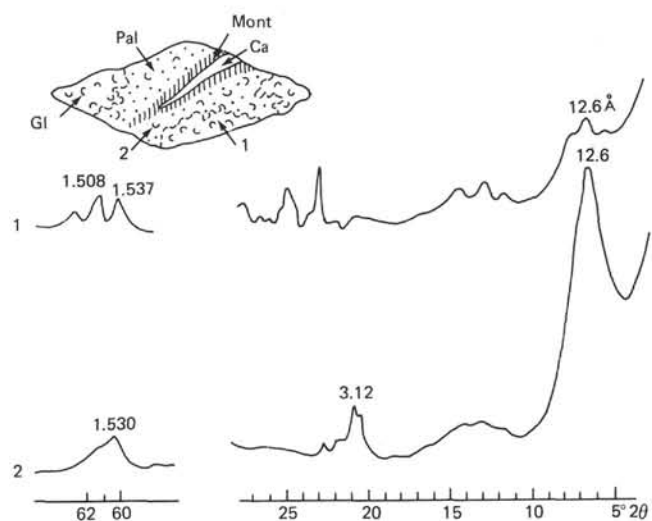


Figure 6. X-ray diffractograms of a palagonite and smectite from Sample 417D-30-5, 67-73 cm. Ca = calcite vein; M = smectite (montmorillonite) rimming the vein; Pal = palagonite; Gl = glass; 1 and 2 = sample locations, correspond to numbers of diffractograms.

Recalculation of analyses was conducted, assuming that the total charge of cations in smectites equals 22. We attempted to differentiate the ferric and ferrous iron using crystal-chemical formulas. In smectites with a high iron content, most octahedral positions are occupied by Fe, and the Al content is low. In some samples, Fe occupies a part of the tetrahedral positions as well. The probability that Fe appears as ferric ions (which are isomorphous with  $Al^{3+}$ ) is a question discussed in detail in the description of celadonite. Therefore, we interpret all Fe as ferric in iron-rich smectites and ferrous in low-iron ones. In some samples, a high content of Mg is noted, sometimes having octahedral cations  $>2.00$ . This may be the result of the following: (1) some  $Mg^{2+}$  cations are in interlayers, and (1) the mineral is more or less trioctahedral. In some cases, the predominance of one cause over the other remains unclear; thus, in Table 2, all Mg is placed in octahedral positions.

On the basis of chemical data (Table 2), smectites and mixed-layered minerals are divided into three groups corresponding to their optical properties. Colorless and light green minerals have Al as the main octahedral cation (with admixture of Mg) and Ca in the interlayers. The Si:Al ratio (in terms of atoms per unit cell) varies from 1.49 to 1.97 (3:2 to 4:2); according to Ross and Hendricks (1945), this corresponds to the range of beidellyite to beidellyite-montmorillonite. The  $Al^{VI}$  content is rather high (1.59 to 1.82),  $Al^{IV}$  is 0.29 to 0.52. A small admixture of  $K^+$  is observed in the interlayers. The  $K_2O$  content is sometimes as high as 4 per cent and is



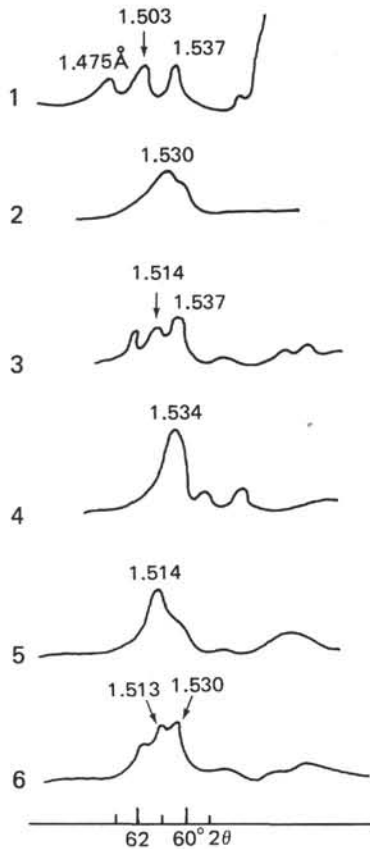


Figure 7. Portions of diffractograms of smectite within the reflection of (060). 1 and 2 = 417D-30-5, 67-73 cm (see Figure 6, corresponding numbers 1 and 2); 3, 4, and 6 = green smectite from altered glass (3 = Sample 417D-59-3, 101-106 cm; 4 = Sample 417D-68-4, 10-12 cm; 6 = Sample 418A-40-2, 79-84 cm); 5 = white smectite from the vesicle (Sample 418A-40-2, 43-48 cm).

caused by the presence of some mica layers in smectites. The Si content decreases with increasing K, to the approximate level for K-rich micas. Green and light brown smectites are characterized by high Mg content.  $Mg^{2+}$  is the main cation in interlayers and is embedded in octahedral positions. Al is low, with  $Al^{VI}$  varying from 0.21 to 1.00, and  $Al^{IV}$  varying from 0.20 to 0.48. Octahedral positions are also rich in iron, but tetrahedral positions are very rich in Si; thus, the Si:Al ratio ranges from 2.8 to 7.1, corresponding to montmorillonite.

The minerals belong to a nontronite-montmorillonite series because of their high content of Fe. Additionally, they have a type of octahedral layer intermediate between di- and trioctahedral layers, i.e., they are intermediate phases between the nontronite-montmorillonite and saponite series.

Brown smectites are distinctly different, having extremely high Fe content. Iron enters not only octahedra, but also the tetrahedra, to compensate for insufficient contents of Al.  $Mg^{2+}$  dominates in the interlayers; smectites of this type are similar to nontronite or saponite. The Si:Al ratio is very high (6.70 to 11.45) because of

the low Al content:  $Al^{VI} = 0$  to 0.11 atoms/unit cell,  $Al^{IV} = 0.22$  to 0.57 atoms/unit cell. However, in one sample (418A-85-1, 32-34 cm) no Al was detected.

Among minerals formerly described as smectites (Table 2), there are numerous phases that belong to mixed-layered silicates. Some contain rather high  $K_2O$  (see Table 2). Sample 418A-59-5, 58-61 cm is very close to celadonite, but contains too much CaO in its interlayers.

Titanium is low in most of the analyzed smectites, but is rather high (up to 5.86 wt. %  $TiO_2$ ) in four samples, two of which also contained  $V_2O_5$  (0.13 and 0.24 wt. %).

Celadonite contains excess water and can be termed "hydroceladonite" with transitions to mica-montmorillonite. Celadonite, while observed in all the investigated holes is less abundant than smectites. Celadonite decreases downhole until it disappears near the bottom of Holes 417A and 417D. Detailed investigations of celadonite are difficult because of its common association with smectite; it is impossible to separate these minerals from each other. In the diffractogram (Figure 4), peaks of celadonite and smectite are not clearly distinguishable from those of amorphous material. Celadonite replaces olivine (often with green smectite or calcite), plagioclase phenocrysts (with colorless smectite, K-feldspar, or zeolites), and basaltic glass (with calcite and brown smectite). It also rims some amygdules, filled with green or brown smectite, calcite, and iron hydroxides. In some veinlets of analcime + celadonite, the celadonite forms fine-grained aggregates which are bright green with strong pleochroism ( $\gamma$  = bright green,  $\alpha$  = light greenish yellow) and has  $\gamma = 1.580$  to  $1.596$ , and  $\gamma = 1.603$  to  $1.625$ . In thin sections, the birefringence seem low because lamellae are quite small. Electronographic study reveals that the mica under investigation is a single-layer polytype IM with a varying degree of crystallinity. Celadonites differ from smectites with high K and low Ca in the interlayers. However, these values vary significantly and there is a gradual transition between these mineral series. The absence of discontinuity is caused by formation of the range of mixed-layered phases (mica-montmorillonite type). That all the celadonites are hydromica is confirmed by the relatively low sum of oxides in analysis (Table 3), which means a high content of water. We conditionally place the boundary between celadonite and smectite at 4 per cent  $K_2O$  (mineral with  $K_2O < 4\%$  = smectite). Calculations of the composition per unit cell are made assuming that the total charge of cations equals 22. All Fe is calculated as ferric iron, but it is possible (with calculated formulas) to estimate approximately the real value for the ratio of  $Fe^{3+}:(Fe^{3+} + Fe^{2+})$ . In Mg-rich dioctahedral micas, the Al content is as low as 1.5 atoms; but, in analyzed celadonites, only 0.0 to 0.28 atoms/unit cell Al occur. Assuming that Al is replaced by  $Fe^{3+}$ , we may conclude that  $Fe^{2+} = (Al + Fe_{total}) = 1.5$ . According to analyses presented in Table 3, this value equals 0.0 to 0.16 atoms/unit cell; the degree of iron oxidation is as high as  $Fe^{3+}:Fe_{total} = 0.88$  to 1.0. This high value is confirm-

TABLE 2  
Microprobe Analyses of Smectite

Oxides	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
SiO <sub>2</sub>	55.8	53.28	54.20	56.5	49.13	50.8	50.9	43.49	31.39	51.63	49.66	39.02	29.7	45.02	42.16	45.44	52.51
TiO <sub>2</sub>	0.0	0.0	0.0	0.28	5.86	0.0	0.0	0.88	2.25	0.06	0.09	0.0	0.05	4.6	0.41	0.02	0.0
Al <sub>2</sub> O <sub>3</sub>	32.0	26.30	23.51	15.2	14.76	6.0	6.7	4.93	3.99	4.27	3.73	5.57	3.73	5.53	6.13	6.36	0.03
FeO	1.3	3.02	4.34	—	8.28	15.7	12.9	—	—	—	—	—	—	—	—	—	—
Fe <sub>2</sub> O <sub>3</sub>	—	—	—	15.94	—	—	—	27.62	35.99	13.06	15.41	28.70	39.50	19.08	7.92	17.81	10.18
MnO	0.08	0.06	0.0	0.0	0.03	0.0	0.0	0.01	0.08	0.04	0.04	0.0	0.03	0.0	0.0	0.0	0.01
MgO	3.9	5.38	3.54	3.3	3.24	15.7	18.3	10.64	9.59	20.6	15.67	10.32	6.12	11.15	14.8	16.14	24.64
CaO	0.47	0.48	0.50	1.0	4.04	1.5	1.8	1.80	2.22	0.82	0.82	1.24	0.95	1.38	2.14	1.93	0.55
Na <sub>2</sub> O	0.35	0.08	0.09	0.0	3.6	0.0	0.0	0.0	0.03	0.10	0.09	0.05	0.0	0.23	0.09	0.0	0.06
K <sub>2</sub> O	0.95	0.59	1.77	0.28	1.20	0.0	0.0	2.36	0.79	0.14	0.13	1.60	3.30	4.6	0.08	0.07	0.12
V <sub>2</sub> O <sub>5</sub>	—	—	—	—	—	—	—	0.13	0.24	—	—	—	—	—	—	—	—
Total	94.85	89.19	87.96	95.28	90.14	89.7	90.6	90.75	85.13	90.2	85.15	85.35	81.8	86.23	83.01	87.06	87.7
Si	9.48	3.56	3.71	3.80	3.52	3.72	3.64	3.45	2.88	3.71	3.78	3.11	2.97	3.31	3.30	3.37	3.75
Al	0.52	0.44	0.29	0.20	0.48	0.28	0.36	0.46	0.43	0.29	0.22	0.52	0.44	0.48	0.57	0.56	—
Fe	—	—	—	—	—	—	—	0.09	0.69	—	—	0.37	0.59	0.21	0.13	0.07	0.25
Al	1.82	1.63	1.59	1.00	0.76	0.24	0.21	—	—	0.07	0.11	—	—	—	—	—	—
Fe	0.07	0.17	0.25	0.81	0.49	0.96	0.77	1.67	1.95	0.75	0.88	1.35	2.58	0.85	0.92	0.92	0.30
Mg	0.36	0.54	0.36	0.33	0.34	0.71	1.95	1.26	1.31	2.20	1.77	1.23	0.91	1.22	1.72	1.78	2.62
Mn	—	—	—	—	—	—	—	—	0.01	—	—	—	—	—	—	—	—
Ti	—	—	—	0.02	0.32	—	—	0.05	0.16	—	—	—	—	0.26	0.02	—	—
Ca	0.03	0.04	0.04	0.08	0.31	0.12	0.14	0.15	0.43	0.06	0.06	0.11	0.10	0.11	0.18	0.15	0.04
Na	0.04	0.01	0.02	0.0	0.50	—	—	—	0.01	0.02	0.02	0.01	—	0.04	0.02	—	0.01
K	0.08	0.05	0.15	0.02	0.11	—	—	0.23	0.10	0.01	0.01	0.17	0.42	0.43	0.01	—	0.01
V	—	—	—	—	—	—	—	—	0.02	—	—	—	—	—	—	—	—

Note: 1 to 3 = montmorillonite-beidellyite (1 = Sample 417A-24-2, 81-84 cm, colorless smectite from a vesicle associated with calcite and zeolite; 2 = Sample 417A-30-6, 9 cm, pale green smectite from a plagioclase phenocryst; 3 = the same sample as No. 2, colorless smectite from a veinlet); 4 = Sample 417D-30-4, 25-27 cm, green beidellyite-nonttronite from a vesicle; 5 = Sample 417D-48-6, 85-87 cm, from a phenocryst of olivine(?); 6 = Sample 417D-52-4, 13 cm, from a vesicle; 7 = Sample 417D-59-3, 101-106 cm, from an olivine phenocryst; 8 to 11 = nonttronite from Hole 417D (8 and 9 = Sample 417D-60-2, 128-130 cm, intense yellow smectite from a vesicle associated with celadonite and Fe-hydroxides; 10 and 11 = 417D-69-1, 141-143 cm, dark brown and light brown, respectively, from altered glass in a groundmass of basalt); 12 to 17 = nonttronite from Hole 417A (12 to 14 = Sample 418A-69-5, 58-61 cm, bright brown, in the case of 12 and 13, and intensely green, e.g., 14, from a marginal part of the pillow; 15 and 16 = Sample 418A-79-1, 112-114 cm, brownish green smectite from a vesicle filled with calcite; 17 = Sample 418A-85-1, 32-34 cm, brown smectite from glass in basaltic groundmass).

TABLE 3  
Microprobe Analyses of Celadonite

Oxides	1	2	3	4	5	6	7	8	9	10	11
SiO <sub>2</sub>	51.02	50.68	50.56	35.8	46.5	53.3	49.06	51.7	50.37	46.56	53.8
TiO <sub>2</sub>	0.11	0.02	0.04	0.5	0.00	0.0	0.04	0.0	0.02	0.15	0.0
Al <sub>2</sub> O <sub>3</sub>	5.31	8.13	3.92	5.1	3.6	2.6	3.59	3.91	5.18	5.53	1.8
Fe <sub>2</sub> O <sub>3</sub>	27.02	25.63	25.00	33.86	28.44	23.34	25.69	26.17	24.74	24.11	27.92
MnO	0.04	0.01	0.0	0.0	0.0	0.0	0.01	0.0	0.01	0.03	0.0
MgO	4.43	3.72	3.95	7.3	6.2	5.1	5.06	5.14	5.59	5.55	3.9
CaO	0.43	0.35	0.38	0.6	0.6	0.45	1.22	0.99	1.62	1.65	0.4
Na <sub>2</sub> O	0.29	0.01	0.14	0.0	0.0	0.0	0.0	0.0	0.05	0.14	0.0
K <sub>2</sub> O	6.80	6.40	6.1	4.0	6.3	7.4	5.82	6.09	4.09	4.84	3
V <sub>2</sub> O <sub>5</sub>	—	—	—	—	—	—	—	—	0.02	0.02	—
Total	95.44	94.95	89.09	85.8	90.5	91.25	89.46	92.96	90.71	87.62	90.82
Si	3.64	3.60	3.70	2.94	3.51	3.90	3.69	3.73	3.68	3.63	3.93
Al	0.36	0.40	0.30	0.49	0.32	0.10	0.31	0.27	0.32	0.37	0.07
Fe	—	—	—	0.57	0.17	—	—	—	—	—	—
Al	0.09	0.28	0.04	—	—	0.12	0.01	0.06	0.13	0.14	0.09
Fe	1.47	1.38	1.32	1.53	1.46	1.29	1.46	1.42	1.36	1.43	1.53
Mn	—	—	—	—	—	—	—	—	—	—	—
Mg	0.47	0.39	0.78	0.89	0.70	0.56	0.57	0.55	0.61	0.65	0.42
Ti	—	—	—	0.12	—	—	—	—	—	0.01	—
Ca	0.03	0.03	0.03	0.05	0.05	0.03	0.09	0.08	0.13	0.14	0.03
Na	0.04	—	0.02	—	—	—	—	—	0.01	0.02	—
K	0.62	0.58	0.58	0.41	0.61	0.69	0.56	0.56	0.38	0.48	0.28

Note: 1 and 2 = Sample 417A-30-6, 9 cm (1 = bright green from a vesicle; 2 = green from a plagioclase phenocryst associated with pale green smectite, see Table 2, No. 2); 3 to 11 = Hole 417D samples (3 = Sample 48-6, 85-87 cm, green from a vesicle; 4 to 6 = Sample 49-2, 81-83 cm, from the altered glass in groundmass of basalts; 4 and 6 are green bands intercalated with colorless represented by 5); 7 to 10 = Sample 60-2, 128 cm (7 and 8 = yellowish green mica from a vesicle, where 7 = margin, 8 = inner part; 9 = reddish brown mica after glass in a marginal part of the pillow; 10 = deep yellow micaceous substance replacing glass and intercalated with the reddish brown substance of No. 9; 11 = Sample 52-4, 13 cm, green mica from an olivine phenocryst).

TABLE 4  
Chemical Analyses of Palagonites and Glass

Oxides	1	2	3	4	5	6	7	8
SiO <sub>2</sub>	51.84	46.5	53.0	52.0	50.76	45.08	45.99	46.76
TiO <sub>2</sub>	0.96	1.6	1.9	1.9	0.32	0.0	0.0	0.0
Al <sub>2</sub> O <sub>3</sub>	13.31	13.7	14.2	15.0	8.12	5.78	6.05	5.75
FeO	11.12	10.9	12.0	11.9	17.78	15.96	15.35	15.66
MnO	0.17	0.16	0.0	0.0	0.0	0.0	0.03	0.0
CaO	10.55	12.3	2.4	2.5	2.18	1.36	1.31	1.06
Na <sub>2</sub> O	2.05	2.4	0.0	0.0	0.02	0.07	0.11	0.14
K <sub>2</sub> O	0.29	0.07	0.53	0.54	2.94	0.04	0.07	0.07
V <sub>2</sub> O <sub>5</sub>	0.0	0.0	0.0	0.0	0.04	0.0	0.0	0.0
Total	100.99	94.03	89.39	88.94	91.5	85.00	86.18	87.06

Note: 1 and 2 = unaltered glass from marginal parts of pillows (1 = Sample 417A-30-5, 67-73 cm; 2 = Sample 417D-30-5, 67-73 cm); 3 to 8 = palagonite from Hole 417D (3 and 4 = the same sample as No. 2; 5 = Sample 60-2, 128 cm; 6 to 8 = Sample 63-5, 22-26 cm).

ed by the high degree of iron oxidation in the rocks under study (see Table 2). Therefore, we may conclude that investigated hydromicas are very rich in the ferric iron that occupies not only octahedral positions, but sometime tetrahedral ones. The value of the  $Fe^{2+}:(Mg + Fe^{2+})$  ratio is very low (0 to 0.3). The Mg content is high, thus explaining why the value of octahedral cations in formulas is  $>2.0$ . In some cases, a portion of the Mg enters swelling interlayers, but this is obviously not the only cause; at least part of the Mg caused a surplus of octahedral cations.

Among cations in interlayers (except K), Ca is always present, with Na being less abundant. In the samples under study, vanadium (V) was checked. Because these samples are characterized by high Ca and may be mixed-layer phases, V may be fixed in the interlayers. This is plausible because some of the smectite are V-rich. Our analyses of celadonites and smectites coincide with data of J. R. Cann (personal communication) for "protoceladonite" ( $SiO_2 = 50$ ,  $Al_2O_3 = 1.5$ ,  $FeO = 26$  with total iron calculated as  $FeO, MgO = 5$ ,  $CaO = 0.7$ , and  $K_2O = 7$  wt. %) and "saponite" ( $SiO_2 = 44$ ,  $Al_2O_3 = 6$ ,  $FeO = 11$ ,  $MgO = 17$ ,  $CaO = 1$ ,  $Na_2O = 0.5$ , and  $K_2O = 0.5$ ). Palagonites investigated in basalts from Holes 417A, 417D, and 418A are intermediate products of smectites replacing basaltic glass. Palagonite is a brown isotropic material with low refractive indexes. Chemically, it is not homogeneous, varying within the same thin section in  $MgO$ ,  $Al_2O_3$ , and  $SiO_2$  content (Table 4, Nos. 3, 4). Palagonites differ distinctly from the glass with extremely low  $CaO$ , lower  $Na_2O$ , high  $H_2O$ , some higher  $Al_2O_3$ , and commonly high  $MgO$  and total Fe (Table 4, Nos. 5-8). Such strong alteration results in a structural reorganization of the glass; thus, X-ray diffractograms sometimes show only signs of layered silicates. Electronographic analyses indicate the

presence of crystal nuclei that are unordered along the C-axis layer structure. The next stage of alteration is growth of smectite crystals and an ordering of their structure. Chemically, a loss of  $CaO$  and  $TiO_2$ , and a gain of total Fe,  $MgO$ , and (sometimes)  $K_2O$  takes place (compare Tables 2 and 4). At this stage, smectites of different types are formed, i.e., rich in Fe or Mg. Therefore, palagonite is clearly not a stable mineral phase in the process of basalt alteration, but is a metastable intermediate product formed at the initial stage of smectite formation.

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