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17. IOWAITE IN SERPENTINITE MUDS AT SITES 778, 779, 780, AND 784: A POSSIBLE CAUSE FOR THE LOW CHLORINITY OF PORE WATERS[1]

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ABSTRACT

The <63-µm fractions of serpentinite muds from two seamounts on the Mariana and Izu-Bonin forearcs were analyzed for mineral composition by X-ray diffraction and for chemical composition by X-ray fluorescence. The silt fraction of the muds consists predominantly of chrysotile, brucite, and ample amorphous constituents. Chlorite and smectite are less abundant components.

Of special interest is the occurrence of iowaite, a brucite-like, Cl-bearing mineral with a layered structure. Iowaite was not found in the samples from the summit site of one of the seamounts drilled; however, it is scattered throughout the strata, composing the flanks of both seamounts investigated. No systematic change of the iowaite abundance with depth was observed.

The distribution of iowaite is confined to the surface of the flanks of the seamount. Based on the distribution on the mineral and its chemical composition, we suggest that the iowaite formed by oxidation of some of the ferrous iron in brucite contained in the serpentine mud as it contacted abyssal seawater during protrusion onto the seafloor. The resulting positive charge imparted to the brucite was compensated by the uptake of seawater chloride. Consequently, the formation of iowaite is restricted to the seafloor where oxygen and chloride are available for these reactions. The availability of oxygen is considered the limiting factor.

We conclude that iowaite formation cannot be a major cause for the low chlorinity of pore fluids inside the seamounts.

INTRODUCTION

One of the drilling objectives of Leg 125 was to obtain additional information on the composition of the pore fluids circulating through serpentinite seamounts on the Mariana and Izu-Bonin forearcs (Fryer et al., 1990). The pore fluids recovered from cores of Sites 778–780 and 784 (Fig 1.) are characterized by a strong decrease in chlorinity at shallow depths below seafloor (Fryer, Pearce, Stokking, et al., 1990; Mottl, this volume). Aboard the ship the interpretation of this dramatic decrease in chlorinity was that the fluids most likely were derived from dehydration of hydrous minerals at depth (Fryer, Pearce, Stokking, et al., 1990). The dehydration reactions generate relatively fresh water, which flows upward and decreases the chlorinity of the pore waters.

The chloride-bearing mineral iowaite was found by X-ray diffraction (XRD) analysis of the mineral content of serpentinite mud samples. The detection of iowaite prompted our interest in addressing the question of whether the formation of iowaite might be a possible alternative cause for the low chlorinity of the interstitial waters recovered from the serpentinite seamounts.

METHODS

Sample Preparation

The silt fraction (<63 µm) was separated from the bulk sample by sieve and sedimentation techniques, eliminating the sand- and gravel-sized fraction of unaltered parent rock and coarser serpentinite. Thus, only the "matrix" fraction of the serpentinite muds was analyzed.

Because the serpentinite muds strongly tended to flocculate, sedimentation methods to separate a finer fraction often failed. The silt fractions of such samples were obtained by using a <63-µm sieve. Careful desalting by dialysis for 24 hr did not stop flocculation completely; however, unlimited dialysis could not be run without removing water-soluble components from the samples; in particular, chloride. Thus, allowance was made for minor salt contents rather than risk the alteration of the chemical composition of the sample.

Most of the minerals were identified by XRD of oriented specimens. Nonoriented specimens were prepared by filling a rectangular sample holder of 1-cm^3 volume with the powdered samples.

X-ray Diffraction Analysis

X-ray diffraction was performed by a Siemens 500 automatic diffractometer using Ni-filtered CuKα–radiation. Scanning speed with routine runs was 1°/min. Air-dried, oriented samples were scanned from 1° to 40° 2θ Samples suspected of containing expandable clays were run again after glycolation and heat treatment. Nonoriented specimens were scanned from 2° to 70° 2θ. Slower scanning speeds over particular ranges were used when the identification of a particular mineral phase demanded higher resolution and precision.

Semiquantitative estimates of the abundances of the mineral species were based on the peak areas. The calculated abundances were revised so that the sum of all identified components totaled 100%. When applying this standard procedure to a quantitative estimate of minerals, we must make allowances for large margins of error. Because of the lack of suitable reference samples of known abundances and, above all, because of large differences in the degree of crystallographic order of the different minerals, an exact estimate with a precision of 1% cannot be obtained. This is true in particular for the minerals iowaite and chrysotile. Abundances of iowaite will probably be overestimated. On the other hand, abundances of serpentine minerals might be underestimated. Nevertheless, the calculated abundances are reported even in numbers below 10. This is done to show the differences in composition between the samples rather than to put major significance on the absolute values. If the sample preparation and machine settings are kept constant, differences in abundances among the analyzed samples can be evaluated with sufficient reliability.

Mineral Identification

Clay minerals were recognized in general by their basal reflections in oriented samples. The various nonclay minerals were detected by their prominent reflections unless the reflections interfered with those of other minerals present. Brucite was recognized by its peaks at

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[1] Fryer, P., Pearce, J. A., Stokking, L. B., et al., 1992. *Proc. ODP, Sci. Results,* 125: College Station, TX (Ocean Drilling Program).

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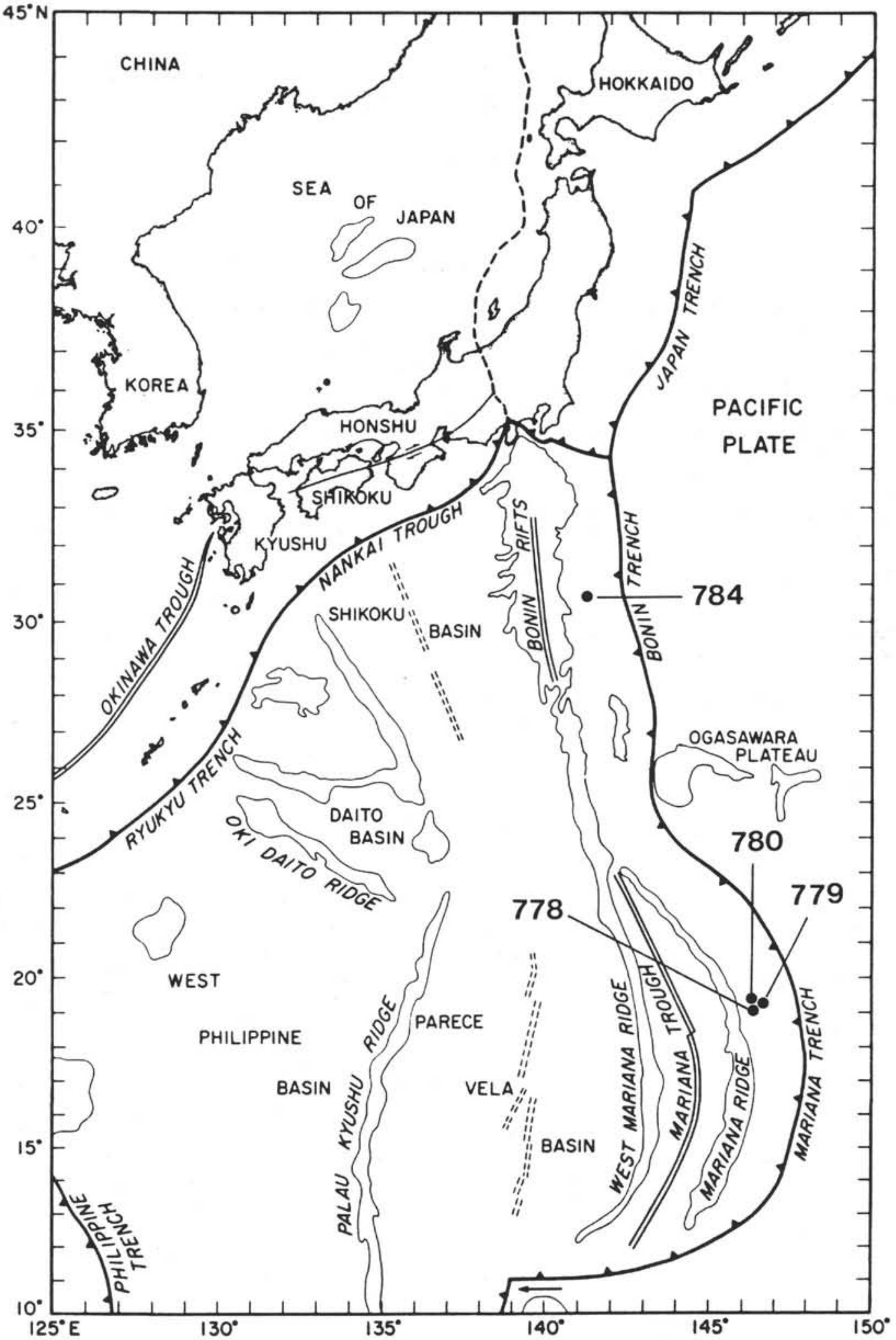

Figure 1. Location of Site 786 in the Izu-Bonin forearc. Adapted from Fryer, Pearce, Stokking, et al. (1990).

4.77Å and 1.79Å. Chrysotile was identified by its strong basal reflections at 7.31Å and 3.65Å and by its polytype-specific reflections at 2.20Å and 2.09Å. It was also recognized by its fibrous habit in scanning electron micrographs (Pl. 1, Fig. 1). Antigorite was not found and lizardite occurred only in traces.

Iowaite

In certain oriented samples, distinct reflections were observed at 8.045Å and 3.985Å (Figs. 2 and 3). These peaks were assigned to a "magnesium-iron-hydroxychloride" which was discovered by Kohls and Rodda (1967) in a Precambrian serpentinite from Sioux County, Iowa. At the location of discovery it was associated with chrysotile, dolomite, brucite, calcite, magnesite, and pyrite, all of which postdated the serpentinization of the rock.

Kohls and Rodda (1967) assigned the peak at 8.109Å (I = 100%) to the indication 003, and the peak at 4.042Å (I = 40%) to the indication 006. The next peak in line of the integral series was 009 at 2.697Å with the very weak intensity of <1. The first and second peak of the mineral under investigation in this study agrees well with the data of Kohls and Rodda (1967). The third peak at 2.697Å could not be detected with certainty, probably because of its weak intensity. The basal spacing of iowaite is 24.33Å. The complete integral series of basal reflections was readily recognized and is proof of the layered structure of the mineral. The mineral was described as displaying a greasy luster, bluish in color, and having a soapy feel. All of these properties agree well with the mineral found in some serpentine samples of Leg 125. Scanning electron micrographs show the platy, hexagonal structure of iowaite (Pl. 1, Fig. 2). Chlorite was recognized by its weak 001 reflection at 14.30Å, which did not shift upon glycolation, and its much stronger 002 peak at 7.15Å. Smectite was recognized by its basal reflection at 12.62Å, which shifted to 17Å after glycolation. Chlorite-smectite (corrensite) was identified by its reflections at 28.90Å and 14.20Å and by its swelling properties upon glycolation.

The chemical composition of iowaite derived from the analysis by Kohls and Rodda (1967):

$$4Mg(OH)_2FeOCl \cdot nH_2O \ (n = 1\text{–}4).$$

An improved structural formula was given by Allmann and Donnay (1969) on the basis of their chemical analysis:

$$[Mg_4Fe(OH)_{10}]^+[Cl(H_2O)_{x-1}]^- \ x = 4,$$

with the actual composition expressed as:

$$[Mg_{0.775}Fe^{II}_{0.022}Fe^{III}_{0.203}(OH)_2]^{0.203+}[Cl_{0.203}(H_2O)_{0.6}]^{0.203-}$$

According to this structural formula the mineral is composed of two subunits. The first consists of four brucite layers associated with one layer in which a ferric iron (Fe^{3+}) ion is coordinated by two hydroxyls, yielding a positive charge. The second subunit consists of one Cl-ion coordinated by three H_2O molecules, resulting in one negative charge by which the positive charge of the first subunit is neutralized. Kohls and Rodda (1967) found that the water is given off gradually as the temperature rises to 280° C, and the structure begins to collapse at 315° C.

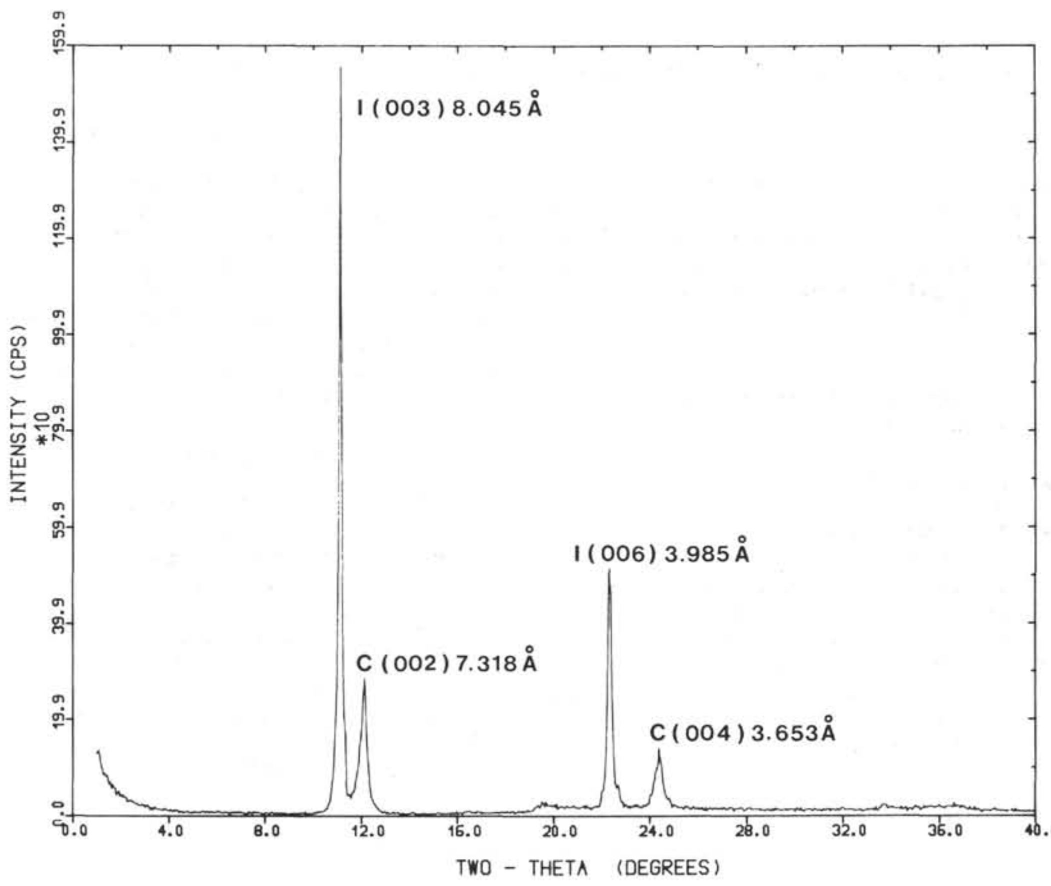

Figure 2. XRD pattern of chrysotile and iowaite (oriented, air-dried), Sample 125-784A-36R-1, 43–45 cm.

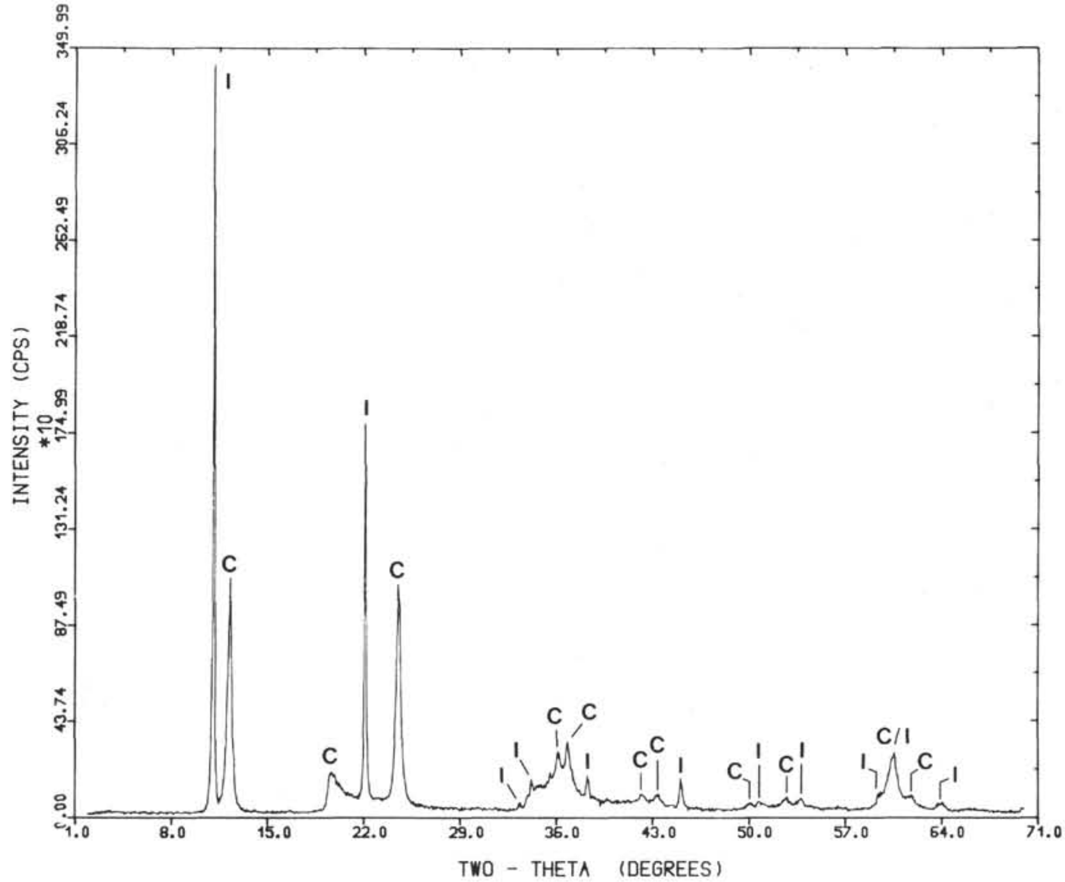

Figure 3. XRD pattern of chrysotile and iowaite (nonoriented, air-dried), Sample 125-784A-36R-1, 43–45 cm.

Carbonates

Most of the aragonite exceeded 63 µm in size and was separated from the silt fraction used for mineral analyses. Traces that were present in the analyzed fraction were identified by reflections at 3.40Å and 1.97Å, but were not considered in the quantitative evaluation. Aragonite is visible in the sand fraction as needles up to 5 mm long.

Magnetite

Magnetite was identified by its distinguishing reflections at 2.53Å and 1.48Å, as well as by its magnetic properties.

X-ray Amorphous Constituents

Amorphous matter, present in most samples, was detected as a hump in the base line of the XRD patterns between 20° and 30° (2θ). We attempted a rough estimate of the abundances of the amorphous fractions by measuring the height of the hump. Because of the high Ni-content determined by trace element analyses, the amorphous fractions probably contain predominantly garnierite, commonly associated with chrysotile. Silica and alkali-earth-hydroxides are presumed to be additional amorphous constituents of the "garnierite" fraction. Clay minerals in abundances below the limit of detection might also be included (Figs. 2 and 3).

X-ray Fluorescence Analyses

The compositions of both the major elements and the trace elements of selected samples were analyzed by X-ray fluorescence (XRF) using a Siemens wavelength dispersive, sequential X-ray fluorescence spectrometer, Model SRS 300. Major element compositions were determined with melt tablets, produced by mixing the sample with 9 parts of lithium tetraborate (Spectromelt A 1000). Trace element concentrations were determined using pressed powder tablets. Loss on ignition was determined in the temperature range between 105° C and 1050° C.

Thermogravimetric Analyses

Thermogravimetric (TGA) analyses were conducted using a DuPont Model 1090 thermoanalytic system, equipped with chromel/alumel thermoelements and a thermoanalytic balance, in order to study the decomposition of iowaite by raising the temperature. Only samples containing iowaite and chrysotile exclusively were analyzed by TGA. These minerals are easily distinguished by their different decomposition and conversion temperatures.

The first peak observed starts at 270° C and reaches its maximum at 293° C. At the latter temperature the chloride of the iowaite is given off (Fig. 4). Zeolitic water has been gradually released. The second peak appears at 315° C and displays its maximum at 340° C, probably

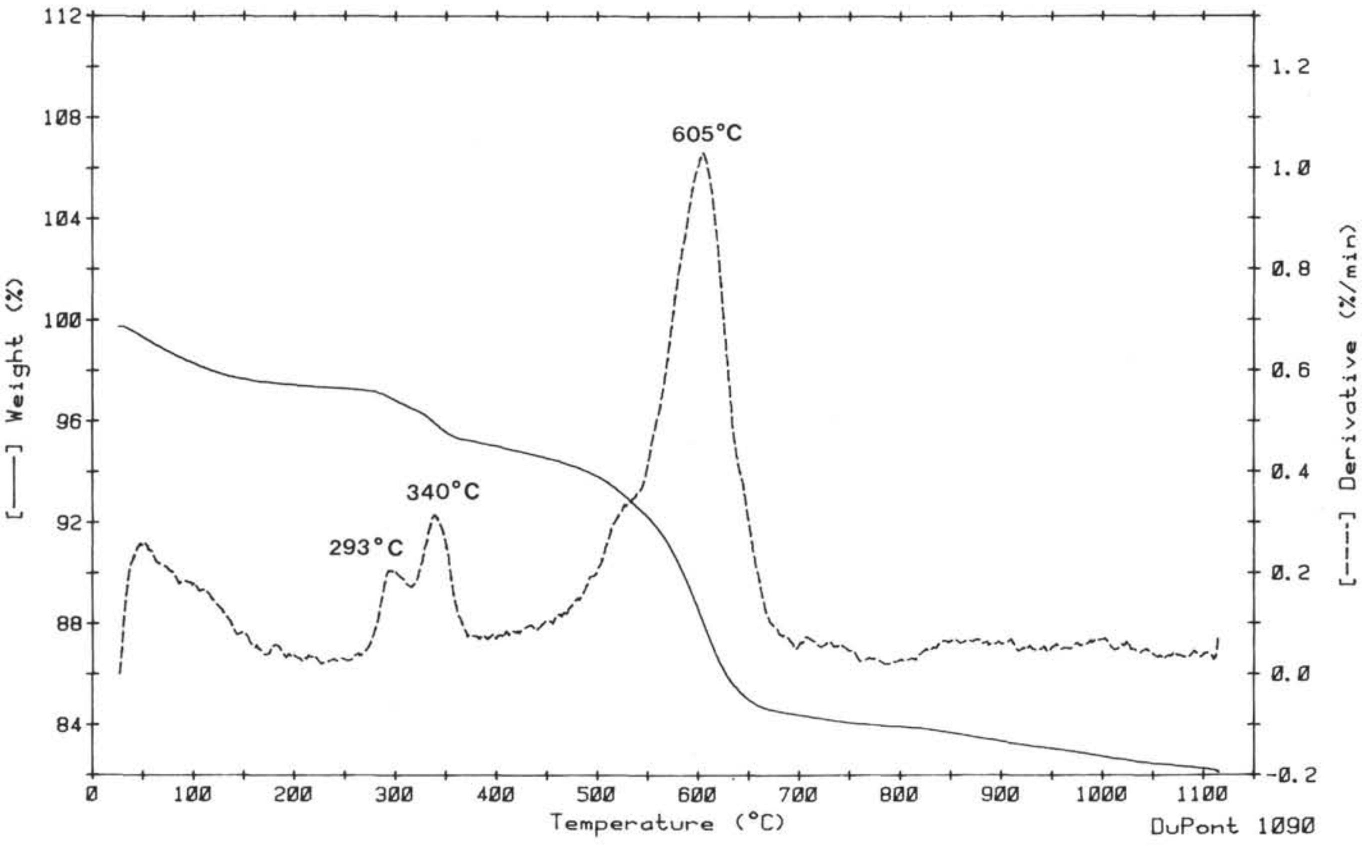

Figure 4. TGA diagram showing the decomposition of iowaite and chrysotile, Sample 125-784A-36R-1, 43–45 cm.

caused by the emission of water from the brucite layers, leading to the complete decomposition of iowaite. The third peak appears at 500°C and reaches its maximum at 605° C, when the water of the chrysotile is released. The peaks assigned to the iowaite agree well with the differential thermoanalytic (DTA) data given by Kohls and Rodda.

RESULTS

Mineralogical Composition

The results of the mineralogical analyses of the Sites 778, 779, 780, and 784 are shown in Figures 5 through 8.

Chrysotile is the prevalent mineral in nearly all investigated samples. Lizardite occurs only in minor amounts and was not given further consideration. Iowaite occurs in many samples from Sites 778, 779, and 784, but was not found in samples from Site 780. Brucite is present in considerable amounts at Site 780. At all other sites it is a minor component and often absent. Apparently brucite is more abundant where iowaite is absent. We suspect garnierite is present in all samples in moderate abundances. Aragonite occurs only in samples from Conical Seamount. Magnetite occurs in the coarse fractions of a few samples. Chlorite was found in some samples, but in small amounts. Smectite was found in Sample 125-778A-11R-1, 56–58 cm, and corrensite in Samples 125-778A-13R-1, 83–85 cm, and 125-779A-36R-1, 80–82 cm.

Chemical Composition

The results of the major and trace element composition of the analyzed samples are listed in Tables 1 and 2. The chemical composition of the serpentine matrix is uniform. The major element composition is characterized by high concentrations of SiO_2, MgO, Fe_2O_3, and a low concentration of Al_2O_3. The SiO_2/MgO ratio is near one, as is characteristic for serpentines. In samples containing only serpentine minerals the ratio is >1; in samples containing iowaite the ratio is <1. Because of the high water content of both serpentine and iowaite, the loss on ignition is high. The maximum values of 17.26% are found in samples containing iowaite. The trace element composition is marked by exceptionally high concentrations of Ni and Cr. Co, V, Zr, and Zn are also relatively abundant. The concentrations of Rb, Th, and Y are below the detection limit of 5 ppm. The concentrations of Sr result from the presence of aragonite, where Ca is partly substituted by Sr.

The locations of iowaite occurrences in the analyzed samples are given in Table 3.

DISCUSSION

The most interesting finding of our investigation is the occurrence of the rare mineral iowaite. According to all the evidence found, iowaite is thought to have formed from brucite by uptake of chloride. This conclusion is consistent with the structural similarities of iowaite and brucite. Moreover, the abundances of iowaite in the serpentinite muds of the seamounts in the Mariana and Izu-Bonin forearcs correlate approximately negatively with the abundances of brucite. Brucite [$Mg(OH)_2$] generally contains some ferrous iron as a substitute for Mg^{2+} in the layer lattice. As cited by Deer et al. (1962), the FeO content of brucite can reach a maximum of 10%. The authors suggest that the greater the contribution of fayalite, the greater the Fe content.

When some ferrous iron in brucite is oxidized to the ferric state by the oxygen in seawater, one positive charge per oxidized Fe^{2+} in the brucite elementary cell is left unsatisfied. This positive charge would readily be compensated by a Cl-ion from seawater if the Cl^- is introduced as an interlayer anion between the brucite layers. Iowaite is formed by this uptake of chloride. Accordingly, the formation of

Table 1. Major element concentrations of selected samples from Sites 778, 779, 780, and 784.

Sample (interval in cm)	Depth (mbsf)	Major elements (%)											
		SiO_2	MgO	Na_2O	Fe_2O_3	MnO	TiO_2	P_2O_5	CaO	K_2O	Al_2O_3	LOI	Total
125-778A-1R-1, 57–60	0.58	38.53	37.42	0.00	7.27	0.10	0.06	0.03	1.06	0.13	0.48	14.70	99.78
125-779A-2R-3, 88–90	4.00	35.07	33.36	0.72	6.45	0.08	0.02	0.02	8.78	0.16	0.00	14.00	99.13
125-780C-1R-1, 65–69	0.67	38.92	35.96	0.35	7.91	0.12	0.10	0.03	1.18	0.15	0.86	13.50	99.08
125-784A-													
34R-2, 106–108	312.26	39.97	36.48	0.12	8.96	0.31	0.03	0.01	0.31	0.03	1.00	13.25	100.47
35R-1, 98–100	320.28	40.00	36.55	0.02	6.92	0.21	0.02	0.01	1.32	0.02	1.07	14.10	100.24
35R-2, 39–41	321.20	41.65	37.32	0.07	7.70	0.12	0.02	0.01	0.56	0.15	0.00	12.30	99.72
36R-1, 43–45	329.72	36.48	38.84	0.05	7.62	0.10	0.01	0.02	0.06	0.13	0.00	16.50	99.81
36R-1, 64–66	329.93	35.01	38.22	0.00	8.21	0.10	0.03	0.00	0.27	0.01	1.21	17.26	100.32
39R-1, 60–62	358.50	39.91	38.39	0.00	7.14	0.07	0.02	0.00	0.24	0.01	0.00	13.46	100.24
40R-1, 48–50	367.98	40.21	37.80	0.00	6.33	0.09	0.02	0.00	0.28	0.01	1.04	13.85	99.63
41R-1, 48–50	377.78	38.66	38.19	0.00	7.13	0.11	0.02	0.01	0.41	0.01	1.23	14.67	100.44
42R-1, 71–73	387.61	37.60	38.40	0.02	7.35	0.10	0.02	0.01	0.48	0.01	1.16	15.15	100.30
44R-1, 76–78	407.06	39.15	37.55	0.07	6.80	0.09	0.03	0.01	0.87	0.01	1.33	14.62	100.53
45R-1, 20–22	415.26	39.27	38.03	0.07	6.68	0.09	0.03	0.01	0.84	0.01	1.26	14.11	100.40

Note: LOI = loss on ignition.

Table 2. Trace element concentrations of selected samples from Sites 778, 779, 780, and 784.

Sample (interval in cm)	Depth (mbsf)	Trace elements (ppm)							
		Ni	Cr	Co	V	Cu	Zr	Sr	Zn
125-778A-1R-1, 57–60	0.58	1514	1086	89	52	9	20	131	44
125-779A-2R-3, 88–90	4.00	1587	876	70	46	8	86	1196	22
125-780C-1R-1, 65–69	0.67	1455	921	90	55	6	15	22	24
125-784A-									
34R-2, 106–108	312.26	2768	650	112	29	18	84	137	56
35R-1, 98–100	320.28	2650	664	93	29	8	31	140	45
35R-2, 39–41	321.20	1738	1498	102	46	<5	16	9	26
36R-1, 43–45	329.72	1621	1571	98	46	<5	14	<5	28
36R-1, 64–66	329.93	2235	1309	127	30	10	26	108	77
39R-1, 60–62	358.50	2702	580	187	30	5	20	129	41
40R-1, 48–50	367.98	2232	886	138	29	<5	23	115	41
41R-1, 48–50	377.78	2349	2002	122	31	12	16	121	37
42R-1, 71–73	387.61	2108	1600	106	31	12	14	106	37
44R-1, 76–78	407.06	1932	1133	104	31	19	15	104	34
45R-1, 20–22	415.26	1903	1960	95	29	8	14	99	34

Table 3. The occurrence of iowaite in the analyzed samples from seamounts on the Mariana and Izu-Bonin forearcs.

Seamount	Site	Position	Iowaite occurrence
Conical	778	Halfway up the southern flank	In most samples between 0 and 100 mbsf
	779	Halfway up the south-eastern flank	In two of the samples analyzed from 0 to 300 mbsf
	780	Near summit	In none of the 13 samples taken from the depth interval from 0 to 10 mbsf, and in none of the 11 samples from the depth interval from 10 to 160 mbsf
Torishima Forearc	784	Western flank	In four of 17 samples analyzed from 0 to 420 mbsf

Note: Fryer and Mottl (this volume) found iowaite in a subsample from Site 780 (summit of Conical Seamount) at 1.0 mbsf.

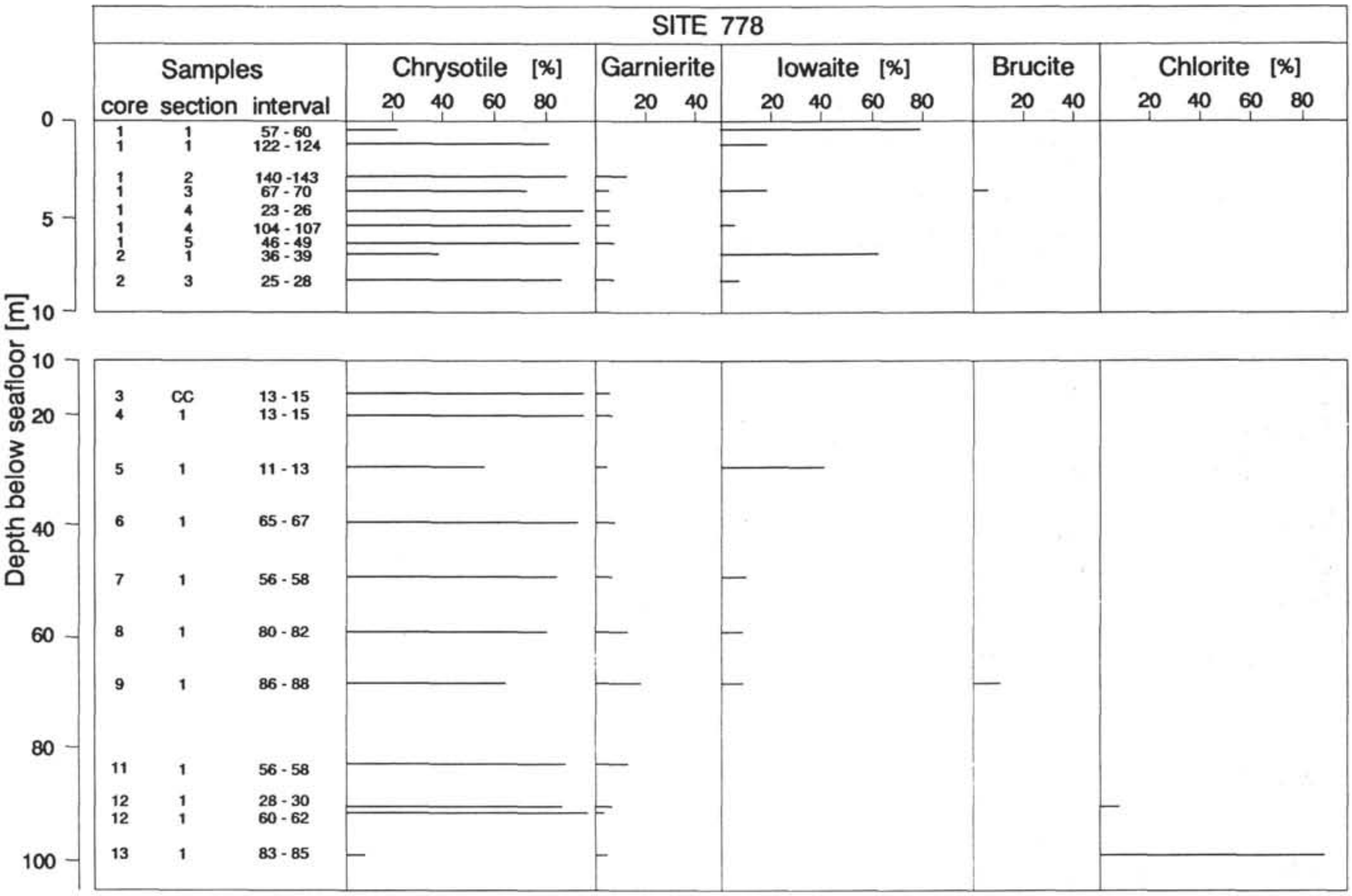

Figure 5. Mineral composition of serpentinite muds, Site 778.

iowaite depends on two processes: (1) some Fe^{2+} of the brucite has to be oxidized to Fe^{3+}, and (2) Cl^- has to be supplied to balance the layer charge.

Because of the abundance of methane in the serpentine muds, reducing conditions should prevail inside the seamounts. No oxygen is contained in the pore fluids. Thus, the oxygen for the oxidation of the ferrous iron of brucite is supplied by seawater. According to Mantyla and Reid (1983) the O_2 concentration of the abyssal seawater on the Mariana forearc is near 100 mg/L. The chloride that must be incorporated with brucite to form iowaite could be supplied by seawater when muds protrude onto the seafloor. If this deduction is true, iowaite is formed on the seafloor and therefore could be found on the flanks of the seamounts where the brucite-bearing serpentine mud protruded and mixed with seawater as it flowed down the flanks. Our samples of the muds on top of the seamount lack iowaite. Fryer and Mottl (this volume) found iowaite in one subsample from Site 780 (near the summit of Conical Seamount) at 1.0 mbsf. The abundance of the iowaite in this sample was not specified. We suspect that this iowaite formed by reacting with seawater chloride that migrated down to 1.0 mbsf by diffusion, possibly at a time when the rate of protrusion of serpentinite muds was low. Generally, however, the adventive, pressure-induced flow of seawater down into the diapir must be considered unlikely. The fluid vents observed at the seafloor (Fryer et al. 1987) prove that the pressure gradient is directed upward similar to the thermal gradient or the flow of compaction fluids.

The conclusion that seawater does not seem to penetrate deeply into diapiric seamounts is in accordance with Haggerty's (1987) interpretations, deduced from the chemical and isotopic composition of aragonite found in serpentinites from Mariana forearc seamounts. Haggerty concluded that fluids other than normal seawater migrate through the sediments. According to Haggerty's hypothesis, iowaite could form only in oxidizing environments. Therefore, iowaite cannot form unless the muds have contact with seawater. The iowaite found today in strata below the seafloor is supposed to have been formed earlier (i.e., before the iowaite-bearing strata were covered by younger flows). The oxidizing potential on the seafloor is considered the limiting factor in iowaite formation. Other factors might be the duration of exposure on the seafloor, grain size, or mixture with seawater.

Iowaite is sometimes associated with brucite, but brucite is usually absent where iowaite is the prevailing mineral phase. Where the accompanying brucite is missing, it might be totally transformed to iowaite. Where brucite is present, the oxygen supply might not have been sufficient to completely transform to iowaite.

It is possible that some chloride is included in the serpentinite muds in the course of the serpentinization process (Rucklidge, 1972). However, as long as only chloride, but no oxygen, is added to the brucite-bearing serpentine muds, iowaite cannot be formed.

CONCLUSIONS

Iowaite, a rare, brucite-like, chloride-bearing mineral was found irregularly distributed in the serpentinite mud flows of two seamounts of the Mariana and Izu-Bonin forearcs. The mineral probably formed from brucite produced along with serpentinites at greater depths.

The oxidation of ferrous iron to ferric iron in brucite, accompanied by the uptake of Cl^- to balance the charge, forms iowaite. The essential oxygen is thought to be supplied by seawater. Accordingly, iowaite is formed after the brucite-bearing, serpentine muds have protruded onto the seafloor. The oxidation of the ferrous iron generates a layer

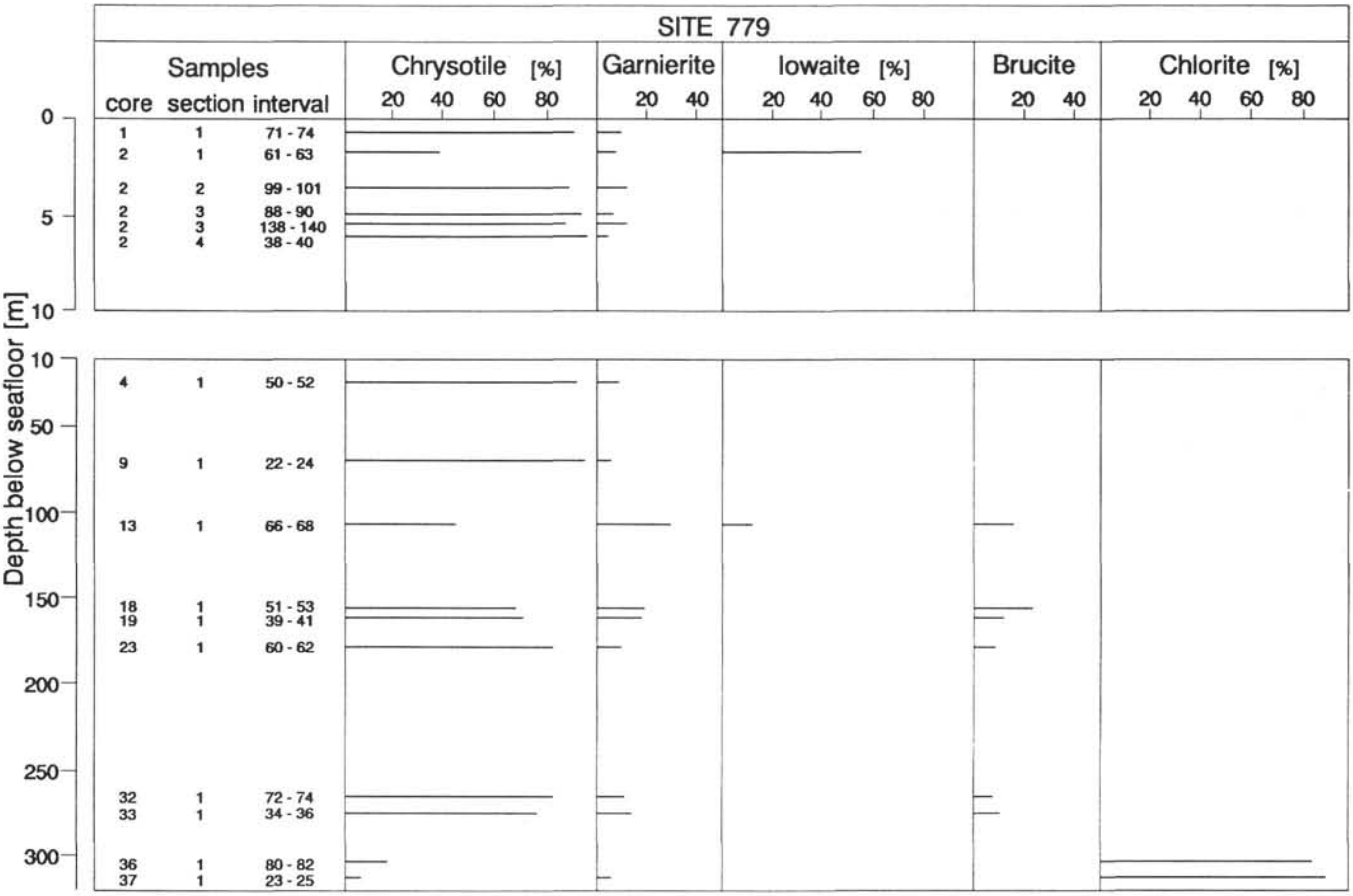

Figure 6. Mineral composition of serpentinite muds, Site 779.

charge, which is compensated by chloride, taken up from the seawater. The oxidation of ferrous iron and the abundance of brucite are considered to be the limiting factors in iowaite formation.

The distribution of the analyzed iowaite seems to be independent of depth. Therefore, it is unlikely that iowaite is formed diagenetically (i.e., under considerable overburden). The independence of depth further shows that the temperatures in the sampled depth intervals do not affect the iowaite stability. In accordance with the data obtained in this study, we conclude that the formation of iowaite cannot be interpreted to be a major cause for the low chlorinity of the fluids from the investigated seamounts.

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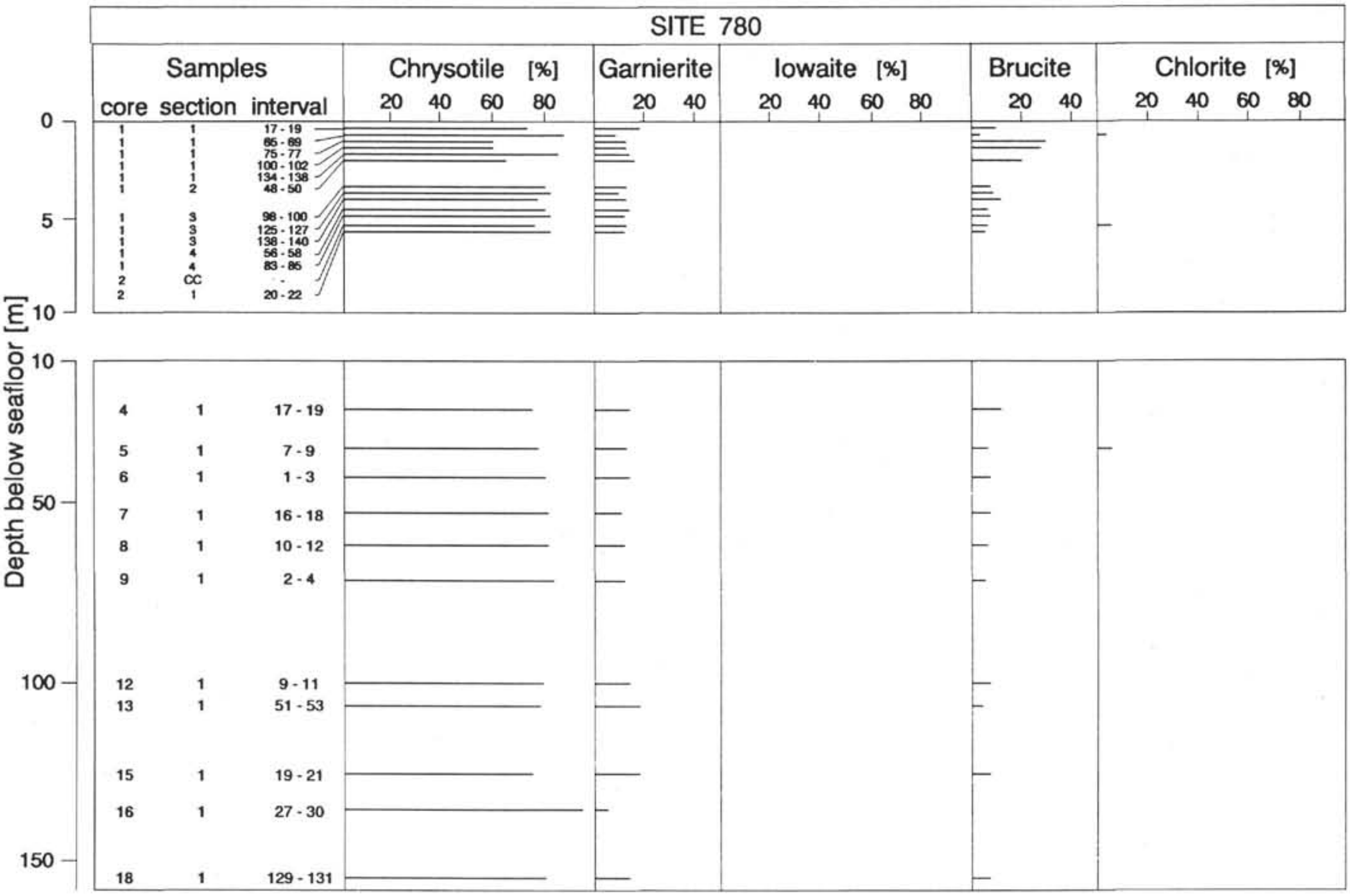

Figure 7. Mineral composition of serpentinite muds, Site 780.

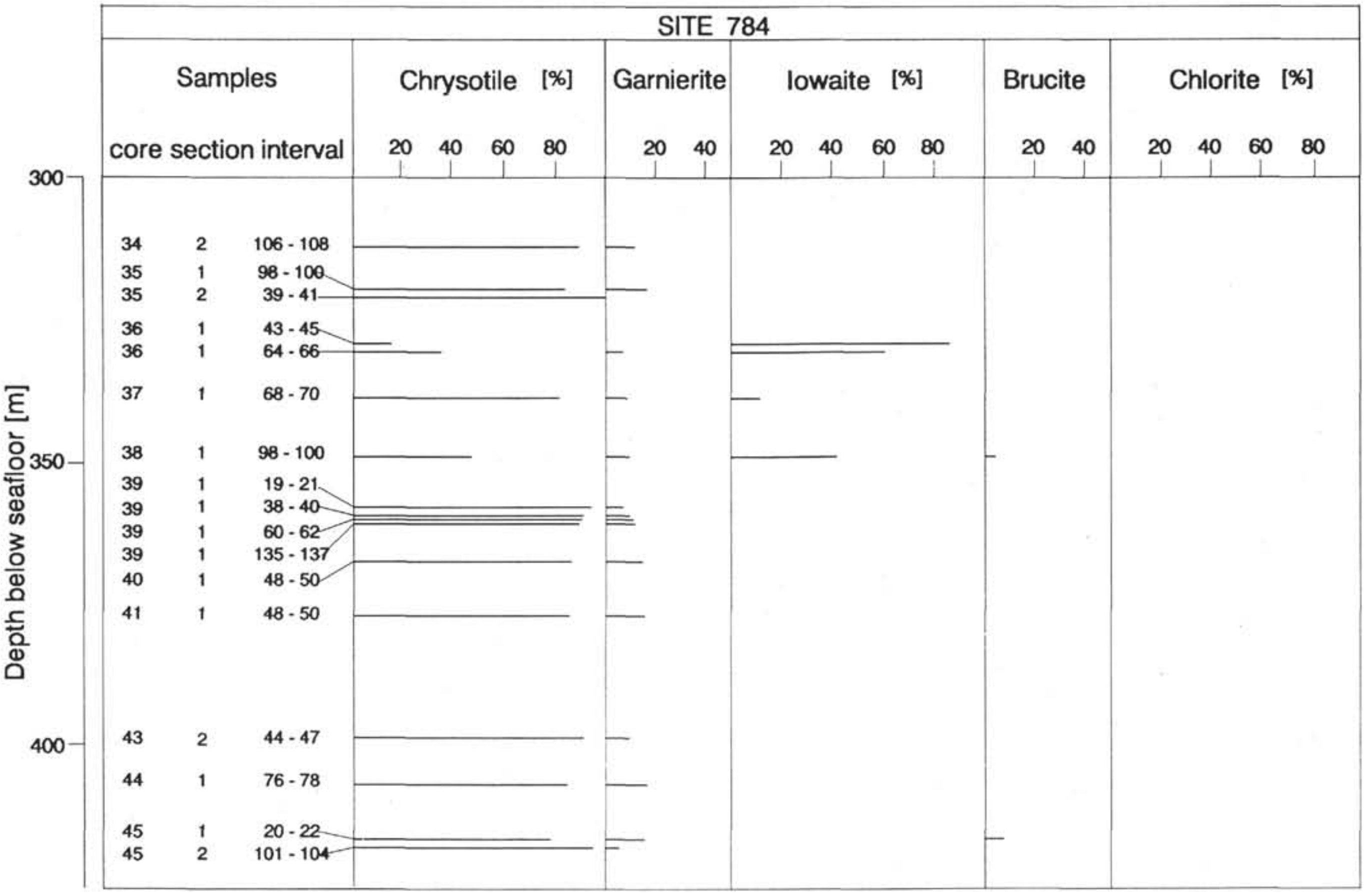

Figure 8. Mineral composition of serpentinite muds, Site 784.

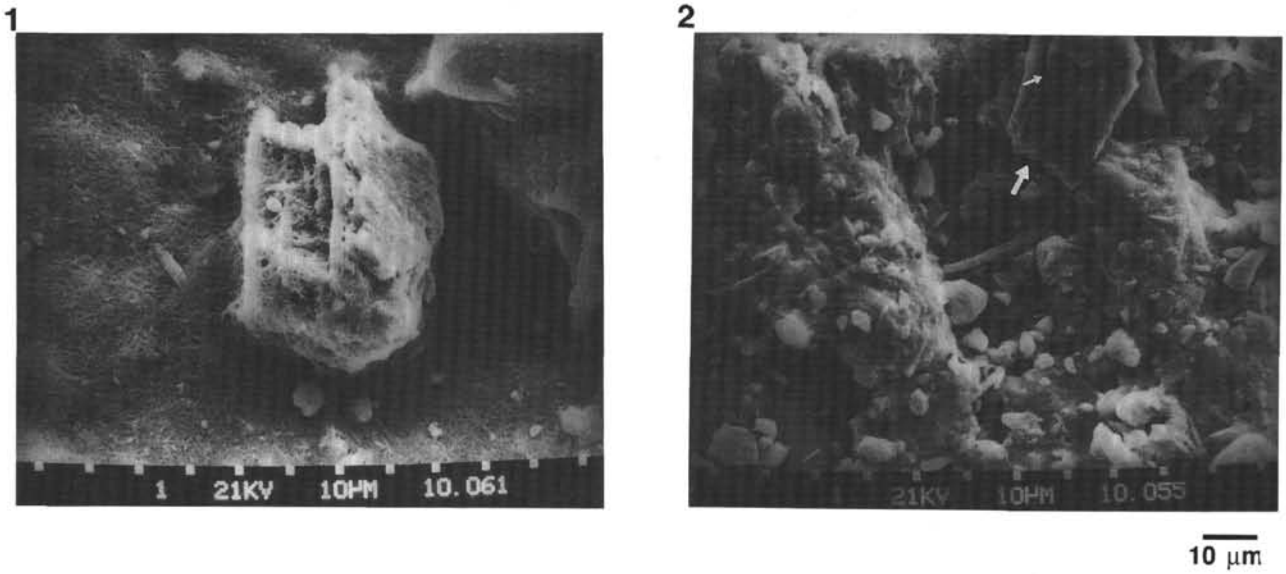

Plate 1. Scanning electron micrographs of serpentinite muds. **1.** Relict structure of an olivine crystal, overgrown by chrysotile fibers, Sample 125-780B-1R-1, 65–67 cm. **2.** Plate structure (arrow) and hexagonal symmetry (small arrow) of iowaite, Sample 125-784A-36R-1, 43–45 cm.