On the chemistry of pillow lavas and the origin of spilites

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Summary. Twenty-five new analyses of 10 spilitic pillows from the British Isles offer evidence of considerable variability in distribution of components, notably Si, Na, Mg.Fe^{2+}, Ca, Fe^{3+}, and water. Selvedges of pillows have compositions strongly contrasted with but essentially complementary to those of cores. Redistribution of components is believed to have occurred during devitrification. Geosynclinal spilites are regarded as basalts which have suffered such redistribution under conditions of diagenesis or low-grade metamorphism.

MOST pillowy bodies, both ancient and modern, display textural contrast between margin or selvedge and core. In pillows of recent age the selvedges are typically glass-rich and surround more crystalline cores. Textures (divergent, arborescent, intersertal, etc.) within such pillows indicate rapid chilling. Calcic plagioclase, with high- or transitional-optics, and pyroxene are the most common crystalline phases. Few chemical data concerning these recent basaltic pillows are available. The Pliocene example from Poggio Raffo, Sicily, studied by Vuagnat (1959) is taken as representative (table I, nos. 1, 2). Here the compositions of core and glassy selvedge are closely similar. The selvedge is clearly chilled basaltic liquid.

Older pillows, especially those deeply buried in geosynclinal sequences, are as a rule not glassy. Divergent or arborescent arrangements of phenocrysts and microlites are common but the place of glass is taken by microcrystalline aggregates of minerals such as chlorite, epidote, or prehnite. The typical feldspar is now albite (usually An_{0-8} with low-temperature optics). Pyroxene, where present, is apparently metastable. Pillows of this type are not found among recent volcanic products.

Chemical similarity between selvedge and core, noted in the modern glassy pillow, is not typical of the older bodies. Slavik (1928) pointed out contrasts in pre-Cambrian material from Bohemia. More recently, Vuagnat has provided many chemical data on non-glassy pillows from the Alps, Britain, and elsewhere. In these, the selvedges are notably poorer in Si and Na and richer in Fe and Mg than the cores. Another

TABLE 1 (See Key opposite)

	1	2	3	4	5	6	7	8	9	10	11	12	13
SiO_2	50·87	52·74	51·82	32·08	50·48	35·12	49·71	34·50	49·06	37·94	38·27	52·31	33·63
Al_2O_3	12·79	13·16	14·29	16·36	14·72	14·75	15·34	13·91	15·64	14·48	10·62	10·41	14·36
Fe_2O_3	3·52	3·09	2·51	5·76	2·59	3·52	3·54	7·17	2·90	3·06	4·63	9·71	7·01
FeO	6·53	7·36	6·32	13·34	4·20	13·00	9·15	15·66	7·43	16·52	6·61	6·99	15·81
MgO	7·49	7·13	4·93	14·29	3·91	13·71	5·15	9·99	6·67	12·77	4·09	4·23	8·86
CaO	9·66	9·48	8·79	3·89	8·40	3·63	6·18	4·66	7·33	4·14	14·06	3·64	3·67
Na_2O	2·90	2·76	4·82	0·56	6·38	1·45	4·37	1·58	4·59	0·31	4·04	3·88	1·13
K_2O	0·12	0·17	0·18	0·72	0·35	0·46	0·02	0·12	0·26	0·24	0·06	0·04	0·00
H_2O+	3·00	2·31	2·29	9·01	2·10	8·36	2·22	7·71	3·71	7·64	1·77	1·67	7·58
H_2O-	1·32	0·04	0·62	1·30	0·80	3·02	0·26	1·01	0·28	0·46	0·08	0·05	0·43
TiO_2	1·59	1·87	1·39	2·44	2·36	2·70	1·43	3·37	2·00	2·20	2·86	2·39	4·44
MnO	0·10	0·13	0·14	0·22	0·09	0·15	0·22	0·28	0·18	0·22	0·29	0·15	0·31
P_2O_5	0·21	0·00	0·02	0·01	0·52	0·36	0·28	0·28	0·28	0·26	0·45	0·55	0·11
CO_2			1·82	0·05	3·28	0·31	2·14	0·00	0·32	0·41	11·96	3·94	2·64
Total	100·10	100·24	99·94	100·03	100·18	100·54	100·01	100·24	100·65	100·65	99·79	99·96	99·98
S.G.			2·82	2·90	2·78	2·86	2·88	3·01	3·01	3·30	2·86	2·90	3·06

	14	15	16	17	18	19	20	21	22	23	24	25	26	27
SiO_2	52·29	53·66	52·65	56·16	54·46	49·54	38·36	51·00	36·12	53·84	46·57	40·85	52·98	36·62
Al_2O_3	16·77	17·98	18·01	15·33	15·00	16·39	17·05	17·82	20·29	18·81	17·06	12·80	14·89	2·46
Fe_2O_3	4·02	3·91	5·46	4·26	5·00	3·93	7·49	1·73	9·15	4·01	6·87	1·10	13·31	49·86
FeO	5·03	4·64	3·31	5·04	5·51	8·57	11·62	6·28	5·81	6·18	8·48	4·28	1·73	1·58
MgO	5·83	5·57	4·61	1·75	1·81	3·87	6·08	5·72	5·08	2·38	3·39	1·86	5·54	3·08
CaO	3·57	3·15	2·75	4·23	4·60	3·90	9·10	4·80	16·44	2·17	5·01	13·80	0·44	0·17
Na_2O	5·57	3·95	2·01	6·70	6·34	5·50	0·92	5·64	0·48	6·70	5·08	4·58	6·56	0·53
K_2O	0·54	1·21	4·14	0·68	1·28	1·30	0·86	0·28	0·15	0·78	0·49	0·96	0·00	1·76
H_2O+	4·48	4·72	5·22	1·88	1·69	3·14	5·13	3·62	4·06	1·62	3·09	1·96	2·64	2·05
H_2O-	0·36	0·98	1·58	0·03	tr.	0·09	0·18	0·38	0·40	0·19	0·13	0·13	0·03	0·00
TiO_2	0·36	0·29	0·27	2·88	3·45	3·02	2·88	1·90	2·10	1·74	3·01	15·78	1·46	1·49
MnO	0·13	0·13	0·16	0·09	0·07	0·14	0·30	0·11	0·12	0·17	0·12	0·07	0·09	0·03
P_2O_5	0·04	0·02	0·00	1·22	1·20	0·92	0·30	0·33	0·22	0·14	0·59	1·47	0·28	0·30
CO_2	1·22	0·00	0·00	0·22	0·08	0·06	0·24	0·40	0·32	1·44	0·00	0·33	0·00	0·00
Total	100·21	100·21	100·17	100·47	100·49	100·37	100·51	100·01	100·74	100·17	99·89	99·97	99·95	99·93
S.G.	2·71	2·70	2·72	2·83	2·87	2·88	3·06	2·97	3·31	2·87	2·91	3·01	2·73	3·94

type of variation involving marginal enrichment in Ca has been recognized in Devonian pillows from Nundle, N.S.W. (Vallance, 1960). The present study extends sampling of British pillow lavas and examines chemical variations in these basic bodies with fairly regular textural heterogeneity.

The new analyses listed in table I (nos. 3–27) are considered to be representative of 10 pillows selected for study. Core and margin were analysed in each case; bodies containing intermediate zones visible macroscopically were sampled more extensively. The material ranges in age from pre-Cambrian to Devonian. Three samples are from the Mona Complex–Porth yr Halen, Llanddwyn Island, and Cerrigceinwen Church, Anglesey (Greenly, 1919), and Porth Dinlläen, Lleyn Peninsula, Carnarvonshire (Matley, 1928). The pillows from An Aird, Tayvallich (Peach *et al.*, 1911), and Rudha Cuillin, Loch Awe, Argyllshire (Bailey, 1913) belong to the Dalradian epidiorite suite. Ordovician pillows are

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Key to the Table

1, 2. Interior (1) and vitreous margin (2) of basaltic pillow (diam. 2 m), Poggio Raffo, Sicily (Vuagnat, 1959).

3, 4. Core (3) and selvedge (4) of pillow (diam. 12 cm), Mullion Island, Cornwall (Nat. Grid, SW 661174). Spec. 15401. Anal: W. H. Herdsman.

5, 6. Core (5) and selvedge (6) of pillow (diam. 15 cm), Port Vad, Ayrshire (Nat. Grid, NX 093869). Spec. 21416. Anal: Avery and Anderson.

7, 8. Core (7) and selvedge (8) of pillow (diam. 20 cm), Strumble Head, Pembrokeshire (Nat. Grid, SM 897414). Spec. 20659. Anal: W. H. Herdsman.

9, 10. Core (9) and selvedge (10) of pillow (diam. 20 cm), Porth Dinlläen, Carnarvonshire (Nat. Grid, SH 276420). Spec. 20626. Anal: Avery and Anderson.

11, 12, 13. Inner core (11), 16 cm from margin, outer (main) core (12), 10 cm from margin, and selvedge (13) of pillow (diam. 35 cm), Chipley, Devon (Nat. Grid, SX 807721). Spec. 15306. Anal: W. H. Herdsman.

14, 15, 16. Core (14), 20 cm from margin, outer core (15), 4 cm from margin, and margin (16) of pillow (diam. 40 cm), small quarry near bridle-path, Cummer, ½ mile NW. of Finny, Co. Mayo. Specs. 18081, 18086. Anal: W. H. Herdsman.

17, 18, 19, 20. Core (17), 30 cm from margin, (18), 20 cm from margin, (19), 10 cm from margin, and selvedge (20) of pillow, An Aird, Tayvallich, Argyllshire (Nat. Grid, NR 703838). Specs. 20934, 20933, 20935, 20936. Anal: Avery and Anderson.

21, 22. Core (21) and selvedge (22) of pillow (diam. 12 cm), Porth yr Halen, Llandwyn Island, Anglesey (Nat. Grid, SH 391630). Spec. 20600. Anal: Avery and Anderson.

23, 24, 25. Core (23), 17 cm from margin, outer core (24), 10 cm from margin, and selvedge (25) of pillow (diam. 40 cm), Rudha Cuillin, Loch Awe, Argyllshire (Nat. Grid. NM 918054). Specs. 15095, 15096. Anals: W. H. Herdsman (23, 24) and Avery and Anderson.

26, 27. Core (26) and selvedge (27) of pillow (diam. 25 cm), W. of Cerrigceinwen Church, Anglesey (Nat. Grid, SH 423737). Spec. 20592. Anal: W. H. Herdsman.

represented from Mullion Island, Cornwall (Flett, 1946), Strumble Head, Pembrokeshire (Thomas and Thomas, 1956), Port Vad, north of Ballantrae, Ayrshire (Peach *et al.*, 1899), and near Finny, Co. Mayo (Gardiner and Reynolds, 1912). The youngest pillow is from Chipley, Devon (Middleton, 1960). Local geological details are given in the references cited. To this list of analysed British pillows must be added those from Newborough, Anglesey (Vuagnat, 1949*a*), Tayvallich (Vuagnat, 1949*b*), and Llanellwedd,[1] Radnor (Nicholls, 1959).

Several distinct patterns of chemical variations are evident within these pillows. In each case Na is more abundant in the core than in the selvedge which also, as a rule, carries less Si and more H_2O. Notable variety exists, however, in the distribution of $Mg.Fe^{2+}$, Ca, and Fe^{3+}. Apparent marginal enrichment in $Mg.Fe^{2+}$ with decrease in Ca is evident in the samples from Mullion Island, Port Vad, Strumble Head, Porth Dinlläen, Tayvallich, and Newborough (cf. Slavik and Vuagnat). The pillows from Porth yr Halen and Rudha Cuillin, like that from Nundle and the margined sac from Llanellwedd have Ca-rich selvedges. Both Ca and $Mg.Fe^{2+}$ are concentrated in the An Aird selvedge and at the margin of the 'apparently homogeneous sac' of Nicholls (1959). A tendency to marginal depletion of Ca and $Mg.Fe^{2+}$ is seen in the Finny material. The Cerrigceinwen selvedge is considerably enriched in Fe^{3+}, a component concentrated to a much smaller extent in margins of other pillows. Distribution of the other components estimated is somewhat irregular. The weight percentage Al is roughly uniform throughout most of the pillows. K and Ti are often more abundant in selvedges, Ti especially so in the Ca-rich margin at Rudha Cuillin. CO_2 tends to be more abundant in the cores.

In all the pillows the only sodic phase is albite, generally occupying the textural positions of phenocrysts and microlites. The present distribution of Na is thus related to original feldspar sites. Selvedges, like those from Mullion Island, Porth Dinlläen, and Porth yr Halen, lacking phenocrysts and with few microlites are distinctly Na-poor. In contrast the Finny pillow has little real selvedge and is feldspathic to the margin. Nicholls's homogeneous sac appears similar. The Rudha Cuillin pillow has a true selvedge zone but this carries numerous regular albite crystals taken to be relict phenocrysts. Si also tends to be richest in those parts containing most relics of primary crystalline material. There

[1] The analysed bodies from this locality are called sacs by Nicholls who claims they should be distinguished from pillows. Having examined these rocks in the field and in thin section, the writer is still unsure why the distinction is necessary.

is, in fact, evidence of a fairly simple relation between Na and Si (fig. 1). The apparent exceptions (Chipley and Finny) have free quartz accumulated in the outer cores whereas the inner cores are richer in carbonates. K shows little tendency to be fixed in original feldspar sites but occurs (most commonly as mica, sometimes as K-feldspar) in authigenic growths and fine veinlets.

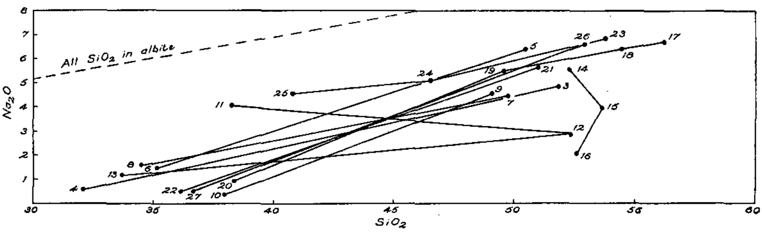

FIG. 1. Relations between Na_2O and SiO_2 (wt. %) in the analysed British pillows listed in table I. The dashed line represents an ideal case in which all Si (and Na) is fixed in albite.

Those pillows marginally enriched in $Mg.Fe^{2+}$ have selvedges composed largely of aggregates of microcrystalline chlorite (e.g. Mullion Island and Porth Dinlläen). Calcic selvedges at Porth yr Halen and Nundle contain abundant crystals and granules of epidote set in microcrystalline chlorite. Prehnite is also common in some calcic selvedges at Nundle. Nicholls has identified pumpellyite as an important marginal phase at Llanellwedd. It occurs, too, as a cavity-filling in the Finny pillow lava. Earthy or granular sphene is an accessory in most selvedges; at Rudha Cuillin it becomes an essential constituent. The Cerrigceinwen selvedge, of course, contains much haematite.

Following a uniformitarian view for the formation of these ancient pillows, it may be expected that the microcrystalline selvedges represent chilled material, once probably glassy. Indeed, in several areas studied, there is good evidence for replacement of glass. Some pillowy rocks at Nundle (e.g. 22970)[1] have fine chlorite aggregates with clear relics of perlitic crackling. Euhedral epidote crystals have grown in the aggregates and across the pillow margin into the matrix. Here both chlorite and epidote surely replace glass. Battey (discussion in Nicholls, 1959) refers to a pillow selvedge showing perlitic cracking but now crystalline. The selvedge from Mullion Island (15401) consists of fine chloritic

[1] Numbered specimens are in the collections of the University of Sydney.

aggregates with concentric banding and small crescentic patches of brown sphene within the bands. The whole gives the impression of being a crystallized colloid. Sub-parallel thin patches of sphene in banded chlorite occur in the Porth Dinlläen selvedge (20626) but the crescentic character, noted above, is not present. The other British pillows studied have less characteristic selvedges, but in all except that from Finny there is much more abundant microcrystalline material at the margins than in the cores.

If the selvedges were chilled to glass it may be taken, by analogy with modern pillows, that the chemical components were once roughly uniform throughout. This is clearly not the case with the pillows now. The selvedges are chemically unlike normal igneous rocks while the cores would qualify for the name spilite in its commonly used sense. Intersertal patches of microcrystalline material, texturally similar to selvedge aggregates, exist in the cores of many pillows. These patches, too, may once have been glass. In view of the compositional changes in once-glassy material, and presumably connected with devitrification, it is reasonable to expect that neither selvedges nor cores of altered pillows now have the composition of chilled magma. The greater the amount of original glass the greater is the likelihood of extensive departure from original composition. It is surely significant that pillows with limited selvedges (and hence less rapidly chilled?) show fewer signs of variation in the distribution of components (cf. the Finny pillow).

The high H_2O^+ contents of most pillows suggest mineral adjustments took place in aqueous environments. These could have occurred during cooling or after consolidation. Where cooling was rapid the opportunity for purely magmatic adjustment was doubtless limited. However, changes initiated early may be completed after consolidation. In areas where altered pillowy flows are associated with sediments containing igneous detritus the latter tends to be mineralogically similar to the materials of the flows. Growth of authigenic albite, chlorite, epidote, prehnite, or pumpellyite in these sediments indicates response to the same physical conditions as affected the lavas. In such régimes post-consolidational alteration of pillows is to be expected. With the metamorphic environments at Tayvallich and Rudha Cuillin this is hardly surprising but common adjustment of sediments and flows is also evident in terrains not regarded as typically metamorphic. The successions at Strumble Head, Port Vad, and Finny, for example, offer many signs of adjustment to low temperature, hydrous conditions which should probably be considered as diagenetic. In all of the cases studied adjustment must have been accomplished above the stability range of zeolites. The present mineral assemblages apparently range from those included in Coombs's (1960) prehnite–pumpellyite metagreywacke facies to others belonging to low-grade greenschist facies.

Experimental work on the solubility of crystalline basalt in H_2O and H_2O+CO_2 at low to moderate temperatures and pressures (Iiyama, 1961) offers a clue in elucidating the chemical distribution patterns in pillows. Of the components studied by Iiyama (Si, Al, Mg, Ca, Na), Si and Na are notably soluble in both neutral and acid water. The solubility of Ca is increased considerably in the presence of CO_2. Al and Mg are relatively insoluble over most of the range of conditions investigated. Applying these results to partly glassy rocks it may be predicted that the glass will be particularly reactive. Crystallization of glass in the presence of excess H_2O at low to moderate temperatures and pressures will involve not only reorganization of the glass structure but doubtless also solution of various components. Si, Na, and Ca will tend to pass into solution from crystallizing glass as they do from crystalline phases. These three components are widely and variably distributed in altered pillows. They may be fixed elsewhere within the bodies or even lost to the surroundings. If, as seems likely in the cases studied, low-albite replaces original calcic feldspar rather than forming authigenic growths in new sites, extra Si will be fixed with Na in feldspar-rich zones. By analogy with the experimental evidence, relations between Ca and $Mg.Fe^{2+}$ appear to be controlled in part, at least, by pH. Pillows with Ca-poor selvedges tend to have cores with higher Ca than those with Ca-rich margins (fig. 2). However, carbonates mainly in veins, patches and vesicles are more abundant in cores associated with Ca-poor selvedges. It seems apparent that redistribution of Ca was aided by the presence of CO_2. Though proportions of Ca and $Mg.Fe^{2+}$ vary widely relations between Mg and Fe^{2+} are usually less variable. Small increases or decreases in Mg/Fe^{2+} from core to selvedge are apparent but these cannot be systematized in terms of present compositions. It may be significant that pillows showing decrease in Mg/Fe^{2+} are carbonate-poor. Although it seems evident that Ca may migrate in solution, the not uncommon occurrence of chlorite in cavities suggests that $Mg.Fe^{2+}$-rich selvedges are not merely residual accumulations dependent on removal of Ca. Nevertheless $Mg.Fe^{2+}$ is probably less migratory than Ca. The Ca locally removed can be fixed in cores but is perhaps mainly lost to the pillow. Veins and patches of Ca-minerals and even material like the 'chert' (SiO_2 33·57; Al_2O_3 21·49; CaO 20·46) interstitial to pillows in Borneo (Reinhard and Wenk, 1951) probably represent Ca from pillows.

The material replacing glass in most pillows is typically aluminous. Among calcic phases epidote, prehnite, pumpellyite, and hydrogrossular

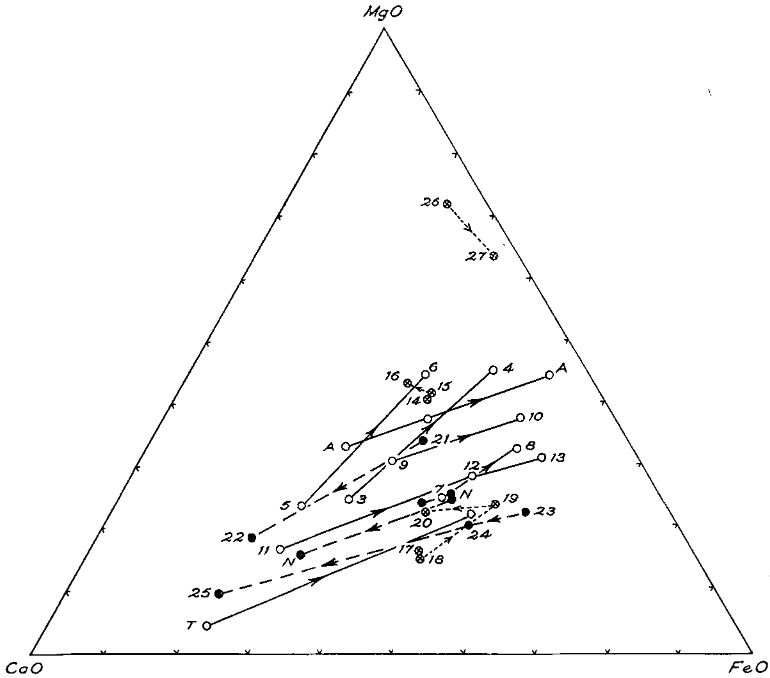

FIG. 2. Variations in MgO, FeO, and CaO (wt. %) in analysed spilitic pillows. Arrows point from core to selvedge. For convenience, pillows showing similar styles of variation are indicated by similar conventional symbols. The numbered points refer to data in table I; *A*, *N*, *T* represent analysed pillows from Anglesey (Vuagnat, 1949*a*) Nundle (Vallance, 1960), and Tayvallich (Vuagnat, 1949*b*), respectively.

(Scott, 1951) are recorded. The chlorites, optically similar to those described by Battey (1956), are doubtless aluminous. Glass becoming progressively organized in the presence of H_2O and losing Si and Na appears to have crystallized largely as hydrous Al-bearing silicates. Removal of Ca must favour growth of chlorite. Retention and accumulation of Ca leads to development of hydrous CaAl-silicates in addition to chlorite. Low-Al selvedges are found only in those unusual cases where the environment has favoured accumulation of non-aluminous phases such as sphene or haematite. Formation of the latter clearly requires oxidizing conditions; the reasons for Ti-enrichment are less apparent.

Devitrification products in these pillows have compositions unlike those of palagonitic material recognized as alteration products of basic glass in some recent volcanic associations. Altered selvedges of the types discussed here cannot be considered simply as basalt glass + water (cf. Peacock's, 1930, view of palagonite). If spilitic pillows ever passed through a palagonite stage all evidence of it is now obliterated.

The origin of spilites

Spilites have been variously regarded as derivatives from originally sodic (spilitic) magma by primary crystallization or late-magmatic deuteric activity or from water-rich basaltic magma by so-called hydromagmatic crystallization. Some consider the sodic character is due to acquisition of Na from sea-water. Others hold spilites to be merely altered basalts. The numerous hypotheses have been reviewed recently elsewhere (Vallance, 1960). Concerning the genesis of these rocks it is important to recognize that spilitic mineral assemblages, adjusted to low temperature, hydrous conditions, imply no unique set of causes. Rocks from widely different environments have been called spilites. Many, but by no means all, have developed in submarine associations. Some apparently formed during, others after magmatic consolidation. Intrusive spilites or albite dolerites, especially those showing evidence of more extensive mineral adjustment within the bodies than at the margins, are probably more often related to magmatic consolidation than the rapidly-chilled flows quickly buried in geosynclinal piles.

The absence of representatives of originally sodic spilitic magma among recent volcanic products argues against the existence of such material. Balsillie's (1932) labradorite-bearing 'tachylite' at Stockenray Bay, Ayrshire, has been regarded as comagmatic with (in fact, a chilled equivalent of) the Arenig spilites (cf. Bailey and McCallien, 1960). Other samples from this locality are devitrified but contain calcic feldspar and analcime (e.g. 21386). The occurrence of these analcime-bearing basalts obviously out of equilibrium with the associated spilites casts doubt on the assumed comagmatic relation. Identification of source material of spilites will probably have to be made by more indirect means than searching for undevitrified quenching products in spilitic terrains. Again, the hydromagmatic theory receives little support from the hydrothermal experiments of Yoder and Tilley (1962). It is certainly difficult to reconcile this theory with the numerous occurrences of augite with albite in spilites (e.g. Mullion Island, Ballantrae, Porth Dinlläen, Nundle) or with the records of basalts containing fresh pyroxene dredged from the deep ocean floors.

From the earlier discussion it is evident that albite-rich cores of altered pillows are intimately associated with material of common origin but contrasted and complementary composition. Similar contrasts are observable in massive spilites but these are less completely documented. In massive bodies sampling poses greater problems than in more regularly variable pillows. Surely it cannot be fortuitous that chemical variety among rocks described as spilites in the literature is almost as great as in altered pillows (cf. ninety-two analyses in Vallance, 1960).

The pillows considered here probably behaved as open systems with their environments during alteration. Identification of original magmatic material based on calculated bulk compositions of pillows is thus open to much uncertainty. Quantitative estimates of gains and losses are hence also of doubtful value. Nevertheless, it is suggested that in certain cases pillows may be regarded approximately as closed to all components except water. Thus the analysed pillow (23055) from Nundle (Vallance, 1960) is compact and fits closely against its neighbours with no signs of veining or filling of original interspaces. The bulk composition of this pillow is that of basalt with extra water. None of the British pillows studied satisfies these requirements; all have probably exchanged material with their surroundings.

In view of the environmental diversity among rocks called spilites the probability of all having formed in the same manner cannot be high. However, it is suggested that geosynclinal spilitic flows are most reasonably interpreted as rapidly-chilled basalts which have undergone diagenetic or low-grade metamorphic adjustments involving redistribution of components. The various redistribution patterns recognized appear to be determined not only by original composition and excess water but also by local factors such as pH and oxidation-reduction potential. Na, fixed largely in existing feldspar sites, could have come mainly if not entirely from the igneous bodies themselves. The most obvious extraneous source of Na, if such were required, would be provided by base exchange with sea-water during cooling (Yoder and Tilley, 1962) or from sea-water trapped in adjacent sediments after magmatic consolidation.

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