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Coexisting sodic and calcic amphiboles from high-pressure metamorphic belts and the stability of barroisitic amphibole*

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SUMMARY. Compositions of glaucophanes and actinolite-hornblende solid solutions occurring in chemically similar metabasaltic rocks from blueschist terranes in east-central Shikoku, W. California, Valtournanche (W. Alps), and W. Liguria are compared. Chemical contrasts among coexisting Na and Ca amphibole pairs, which reflect disparate *P*-*T* histories under the presumed attendance of local equilibrium, include: Na contents are rather high among barroisitic hornblendes from the western and Ligurian Alps, as well as among high-grade tectonic blocks from California; in contrast, actinolitic amphiboles from both lower-grade Franciscan tectonic blocks and *in situ* schists and the blueschists of Shikoku are impoverished in Na relative to blue-green hornblendes. Sodic amphiboles contain less than 0.5 Al^{iv} per formula unit, whereas Al^{vi} is very high; a situation reversed among calcic amphiboles. The Na + Ca contents of glaucophanes are strongly clustered around the sum of 2.0 (i.e. A site vacant) whereas calcic amphiboles have a wider range with the A site variably occupied. No solvus has been detected within either sodic or calcic amphiboles under blueschist facies conditions. For low-grade metabasaltic parageneses, a miscibility gap separates these two amphibole groups; at relatively high grade such compositions have sodic calcic amphiboles of barroisitic type; this may mean that glaucophane + hornblende assemblages are metastable, accounting for textural relations indicating that the sodic amphibole typically did not grow at the same time as the barroisite. Ti, Mn, and K appear to be concentrated in calcic amphibole compared to coexisting glaucophane, probably in the M_2, M_4, and A sites, respectively.

Contrasts in coexisting amphibole tie lines are thought to be a consequence of the fact that the parageneses of Shikoku and California reflect high *P* and very high *P* prograde *P*-*T* paths respectively, whereas those from Valtournache and W. Liguria show evidence of decompression recrystallization (or back-reaction) of high *P* (i.e. eclogitic) protoliths. Comparison of the inferred physical conditions operating during the production of these four contrasting paragenetic sequences allows the provisional assignment of a *P*-*T* stability region for barroisitic amphibole in metabasaltic rocks as: *P* 4-5 kb at *c.* 350°; *P* 5-7 kb at *c.* 450 °C.

MESOZOIC and younger blueschist terranes discontinuously border the Pacific Ocean and form portions of the Alpine-Tethyan-Himalayan orogen. Dense, H_2O-bearing metamorphic mineral assemblages developed in such belts require geothermal gradients about 5-15 °C/km (Ernst, 1973*a*; Newton and Fyfe, 1976). Such high-pressure, low-temperature regimes are associated with downgoing lithospheric slabs (Hasebe *et al.*, 1970; Oxburgh and Turcotte, 1970, 1971; Toksöz *et al.*, 1971, 1973; Griggs, 1972; Turcotte and Schubert, 1973). Hence glaucophane-schist belts are commonly regarded as products of subduction zone metamorphism and may delimit the ancient convergent plate boundaries (Dewey and Bird, 1970; Ernst, 1970, 1971*a*, 1975).

Although metasedimentary rocks predominate in many subduction zone complexes, metabasalts are widespread. The high-*P*, low-*T* recrystallization of the latter has given calcic, sodic calcic, and sodic amphibole-bearing assemblages. This study focuses on the compositions of these coexisting clinoamphiboles in metabasaltic units from four different blueschist terranes: east-central Shikoku, Japan; W. California, USA: Valtournanche, W. Alps; and W. Liguria, Italy. Prograde and retrograde metamorphic parageneses and the chemistries of the coexisting amphiboles in the different areas are examined and estimates made of the physical conditions attending the formation of sodic calcic amphiboles in metabasaltic rocks.

Paragenetic sequences and P-T *paths*

Prograde mineral assemblages in rocks of basaltic composition from SW. Japan, W. California, and the W. Alps (Valtournanche) have been summarized recently by Ernst (1977); the retrograde paragenesis in W. Liguria was also described.

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Through comparison with a large variety of internally and externally consistent phase-equilibrium studies and some oxygen isotopic and other geothermometers, as well as thermodynamic calculations, physical conditions attending the progressive and decompressive recrystallizations were derived. This synthesis, coupled with newer work (Ernst and Dal Piaz, 1978) allows the comparison of coexisting calcic and sodic amphiboles from the above four regions. The compositions of the host schists have been given in previous investigations and except for some of the Ligurian rocks, which are rather iron-rich, those from the other areas have chemically similar ranges.

In any metamorphic terrane, the effects of retrogression have been superimposed to some extent on the prograde paragenesis. Sometimes late recrystallization has been virtually lacking, as in W. California, or indistinct as in Shikoku. In contrast, the W. Alps, including parts of W. Liguria, have been substantially recrystallized after the high-*P* maximum and locally the early Alpine high-*P* assemblages have been completely obliterated.

Another factor is the relationship between individual hand specimens and the areal disposition of large segments of a metamorphic terrane that contains a range of mineral assemblages. For example, where belts of differing metamorphic facies (occurring in rocks of similar composition) are juxtaposed, the boundaries may be either isograds or tectonic contacts and it is essential that identification of the true nature of the contact be made. The paragenetic sequences displayed by a moderate number of individual rocks may not represent the *P–T* history of a metamorphic belt very accurately, for different parts of the terrane will each have been subjected to their own prograde and retrograde *P–T* paths. This subject has been dealt with by England and Richardson (1977).

Bearing in mind such provisos, generalized *P–T* paths inferred from the mineralogical sequences in the four areas are illustrated in fig. 1. Amphibole-bearing parageneses are now described.

East central Shikoku. In the Sanbagawa belt, progressive mineral assemblages in the eastern half of Shikoku grade from prehnite–pumpellyite facies through interlayered blueschists and greenschists to albite amphibolites containing lenses of eclogitic material (Iwasaki, 1963; Banno, 1964; Ernst, 1964; Ernst *et al.*, 1970; Kurata and Banno, 1974). Sodic amphibole occurs sporadically in the most feebly metamorphosed units, but is widespread in middle-grade mafic schists where it is associated with aluminous actinolite, albite, epidote, phengite, and chlorite. As garnet joins the assemblage in the higher-grade rocks, blue-green calcic amphibole increases at the expense of sodic amphibole, which ultimately disappears. The synchronous(?) prograde *P–T* path has been evaluated by Ernst and Seki (1967) and Ernst *et al.* (1970). Temperatures apparently ranged from about 200 to over 400 °C, and pressures from 4 to nearly 7 kb. Retrograde recrystallization is indistinct.

Western California. The progressive sequence of Franciscan *in situ* mineral assemblages in adjacent rocks ranges from feebly recrystallized zeolitic assemblages through successively more intensely metamorphosed pumpellyite-bearing rocks to lawsonite-, omphacite-, and epidote-bearing blueschists (Coleman and Lee, 1963; Blake *et al.*, 1969; Ernst, 1971*b*; Ernst *et al.*, 1970; Suppe, 1973; Platt, 1975). Even higher-grade tectonic blocks of eclogite, amphibolite, and blueschist are scattered in this terrane (Coleman *et al.*, 1965; Coleman and Lanphere, 1971; Ghent and Coleman, 1973). Sodic amphibole is virtually ubiquitous in metabasalts at grades higher than that characterized by pumpellyite; calcic amphibole is not common and tends to be actinolitic where both sodic and calcic

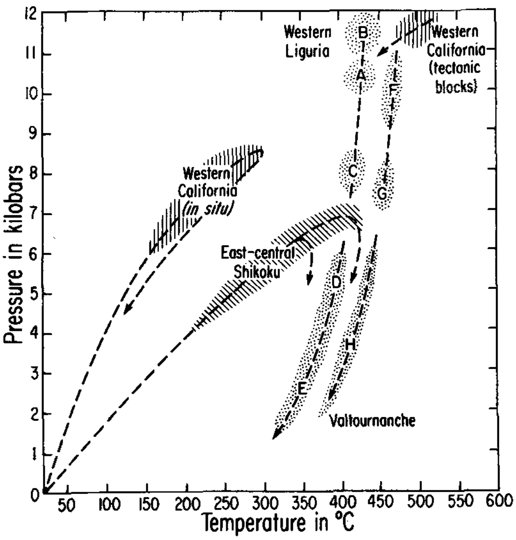

FIG. 1. *P–T* paths inferred from the observed mineral assemblages combined with available experimental and theoretical phase equilibria, and stable isotope geothermometers (Ernst, 1973*a*; Newton and Fyfe, 1976). For W. Liguria and Valtournanche, stages A, B, and F represent eclogitic recrystallization, C and G glaucophane production, and D, E, and H represent portions of a continuum in the conversion of pre-existing rocks to barroisitic amphibolites and actinolitic greenschists. Oxygen isotopic data indicate that Franciscan tectonic blocks equilibrated at temperatures considerably higher than those attending recrystallization of the *in situ* schists (Taylor and Coleman, 1968).

amphiboles occur. The higher-grade amphibolites and eclogites exhibit local replacement of hornblende (and omphacite) by glaucophane, and these amphibole pairs may not be equilibrium assemblages. The tectonic blocks seem to have formed at about 500 °C (Taylor and Coleman, 1968) and over 11 kb. In contrast, the prograde $P-T$ paths derived from the *in situ* schists have much lower temperatures (Ernst *et al.*, 1970, Chapter XIII) about 150-300 °C and 5-8+ kb. Except for the transformation of aragonite to calcite, the effects of retrogression are virtually non-existent in Franciscan rocks of the California Coast Ranges.

Valtournanche. The early Alpine high-P paragenesis in the W. Pennine terrane has been described and summarized by many workers: (Niggli, 1960, 1970; Niggli *et al.*, 1973; Bearth, 1962, 1966; Bocquet, 1971; Ernst, 1971*a*, 1973*b*; Dal Piaz *et al.*, 1972; Dal Piaz, 1974*a*, *b*; Frey *et al.*, 1974). Eclogitic rocks represent the culmination of this sequence, and at Valtournanche have been retrograded to a series of blueschists, barroisitic amphibolites, and greenschists (Dal Piaz *et al.*, in press; Dal Piaz and Ernst, 1978). In contrast to the previous two areas discussed, the amphibole-bearing assemblages reflect reactions that probably took place during decompressive recrystallization. Glaucophane replaces omphacite in the initial stages of this back-reaction, but later is joined by barroisitic amphibole, epidote, then albite, paragonite, phengite, and chlorite. Actinolite only appears after the sodic amphibole has ceased to be stable in mafic schists. The $P-T$ attending eclogitic recrystallization is estimated at 470±50 °C and 10±2 kb, replacement by glaucophane at 450±50 °C and 7+2 kb, and greenschist conditions at 400±50 °C and 3±2 kb, with barroisitic amphibole formed between the last two $P-T$ conditions (Ernst and Dal Piaz, 1978).

Western Liguria. Similar to Valtournanche, the metagabbroic rocks of the Gruppo di Voltri display eclogitic mineral assemblages characteristic of the prograde metamorphic peak variably recrystallized (Messiga and Piccardo, 1974; Chiesa *et al.*, 1975; Cortesogno *et al.*, 1975*a*, *b*, 1977; Ernst, 1976). Early retrogression replaced omphacite by sodic amphibole, followed by the entrance of barroisitic amphibole, then the gradual generation of minor albite, epidote, and white mica plus chlorite. As calcic amphibole and plagioclase increase sodic amphibole diminishes and disappears by the stage at which the Ca-amphibole has become aluminous actinolite. The $P-T$ path during inferred unloading was deduced by Ernst (1976) and is very similar to that postulated for the Valtournanche paragenesis: eclogitic stages A+B 430±50 °C, *c.* 10 kb; glaucophanic stage C, *c.* 400 °C, 8 kb; and barroisitic to greenschist stages D+E, 300-75 °C, 2-5 kb.

Compositions of coexisting sodic and calcic amphiboles

Most studies of element distribution between coexisting minerals assume that the minerals crystallized together in chemical equilibrium. Textural relations may provide unequivocal evidence that minerals crystallized at different times but the lack of such features cannot be employed to demonstrate equilibrium. The coexisting sodic and calcic amphiboles studied chiefly form separate, spatially associated grains, sometimes intergrown, but a few exhibit possible replacement textures. The only chemically zoned samples detected were a few from Valtournanche and W. Liguria and only the rim compositions were employed in the study of coexisting pairs. The times of growth of sodic and calcic amphiboles seem to have overlapped extensively in all four areas and so it is supposed, but not proved, that they are stable assemblages.

Widespread occurrences of sodic and calcic amphibole pairs in low-grade metabasalts of various ages and settings demonstrate the existence of a miscibility gap within this system. No clear evidence of such a gap within the sodic amphiboles is known, for although Black (1970) reported a glaucophane+riebeckite-arfvedsonite pair from New Caledonia, it was not possible to resolve the question of an equilibrium versus non-equilibrium association. Accordingly, based on the widespread occurrence of single sodic amphibole-bearing assemblages and the lack of exsolution lamellae, this group is regarded as showing virtually complete solid solution among the four end-members glaucophane, ferroglaucophane, magnesioriebeckite, and riebeckite (Miyashiro, 1957). The situation is ambiguous but appears to be different for calcic

TABLE I. *Literature sources of amphibole analyses*

East-central Shikoku: Iwasaki (1963, Tables 13, 14); Ernst (1964, Table 2); Banno (1964, Table 6); Klein (1969, Tables 4, 5); Ernst *et al.* (1970, Tables 26, 28-30).
West-central California: Lee *et al.* (1966, Table 2); Coleman and Papike (1968, Table 1); Himmelberg and Papike (1969, Table 1); Klein (1969, Tables 3, 4); Ernst *et al.* (1970, Tables 11-17, 27, 31, 32); Burns and Greaves (1971, Table 1); Hermes (1973, Table 5).
Valtournanche: Ernst and Dal Piaz (1978, Tables 3, 4).
Western Liguria: Cimmino *et al.* (1974, Table 2); Ernst (1976, Tables 3, 4).

amphiboles (e.g. Sampson and Fawcett, 1977). At moderate to high pressures, compositions evidently range continuously from tremolite-actinolite to aluminous, alkalic hornblendes (Ernst, 1972; Graham, 1974) but under some lower-pressure regimes the pair actinolite + hornblende exists—suggesting the presence of a solvus (Ernst, 1968, pp. 27–32; Klein, 1969; Cooper and Lovering, 1970; Cooper, 1972; Brady, 1974; Misch and Rice, 1975; Tagiri, 1977; Allen and Goldie, 1978). In contrast, Grapes and Graham (1978) conclude that most actinolite-hornblende pairs result from disequilibrium recrystallization, especially the abrupt appearance or disappearance of phases participating in amphibole-generating or amphibole-consuming reactions. If such a solvus does exist, evidently it is disfavoured by increased pressures, and the blueschist parageneses considered here have only a single calcic amphibole in addition to a sodic amphibole.

Compositions of amphibole pairs. Table I lists the literature sources for the clinoamphibole compositions. Gravimetric analyses show that most sodic amphiboles contain similar amounts of Fe^{2+} and Fe^{3+}, whereas most calcic amphiboles have substantially higher Fe^{2+}/Fe^{3+}. Accordingly, microprobe analyses were computed assuming that 50% of the iron in sodic amphiboles is Fe^{2+} and that 100% of the iron in calcic amphiboles is Fe^{2+}. The proportions of the cations are not influenced by assumptions regarding oxidation state of the iron; but Al^{iv}, Al^{vi}, Si, $\Sigma(M_1+M_2+M_3)$ cations, ΣM_4 cations, and A-site occupancy, etc. are influenced especially for Fe-rich, Al-poor specimens. Both 'wet' and microprobe analyses of minerals are dealt with similarly, for all the data are probe-derived except for three Californian tectonic blocks: 50-CZ-60 (Lee *et al.*, 1966); GRS (Ernst *et al.*, 1970, Tables 27 and 32, for glaucophane major elements only).

Other methods have been employed to assign amphibole Fe^{3+}/Fe^{2+} ratios from microprobe analyses (e.g. Stout, 1972; Brady, 1974; Papike *et al.*, 1974) but are not clearly superior. Calculation of thirty-six Valtournanche amphiboles using the method of Papike *et al.* (1974) gave values for eleven of the analyses that were unsatisfactory as regards site-occupancy totals and so the empirical assignment of $Fe^{3+} = Fe^{2+}$ in sodic amphiboles and $Fe^{3+} = 0$ calcic amphiboles is used.

From figs. 2 and 3 the following features are evident:

The Valtournanche and W. Ligurian compositions are remarkably similar, and contrast with the coexisting calcic and sodic amphiboles from Shikoku and the *in situ* schists of W. California. Specifically, the Na and Al^{iv} in calcic amphiboles coexisting with glaucophanes in the Ligurian and W. Alps are higher than analogous pairs from Shikoku and California. The Franciscan tectonic blocks have a great range of Na, Ca, Al^{iv}, and Al^{vi} with some of these exotic samples similar to those from the Ligurian and W. Alps, whereas others show compositions indistinguishable from the *in situ* Franciscan schists.

In general, sodic amphiboles contain less than 0.5 Al^{iv} per formula unit, and $Al^{vi} \gg Al^{iv}$. Among calcic amphiboles, however, $Al^{iv} \geqslant Al^{vi}$, and the Al^{iv} ranges from 0.0–0.3 in actinolites, and up to 1.7 and over in hornblendes.

The total Na + Ca contents of sodic amphiboles are strongly clustered near 2.0 (i.e. A site nearly empty) whereas in the calcic amphiboles, although the A position is essentially unoccupied in actinolite, it is progressively filled as the calcic amphibole grades towards barroisitic compositions.

A solvus between hornblende and actinolite is not evident based on the proportions of Al^{iv}, Al^{vi}, Na, and Ca in agreement with previous studies of medium- and high-pressure belts.

Where compositional zoning has been measured, as in a few samples from Valtournanche and W. Liguria, among calcic amphiboles there is a regular change from aluminous and sodic (i.e. barroisitic) cores towards Al-poor, calcic (i.e. actinolitic) rims; in contrast, Valtournanche sodic amphiboles display only an indistinct increase in Al + Na towards their rims.

Generally, a compositional gap separates the calcic and sodic amphiboles. In the Shikoku samples, blue and green amphiboles occur together at lower grades but at higher grades a single, sodic calcic amphibole is present that plots at intermediate positions on the Na–Ca and Al^{iv}–Al^{vi} diagrams, apparently bridging the miscibility gap in the Shikoku schists. Therefore if the solvus between sodic and calcic amphiboles has closed under conditions approaching 400 °C and 7 kb, the observed glaucophane + barroisitic amphibole assemblages from W. Liguria and Valtournanche, as well as the Californian high-grade tectonic blocks, probably represent non-equilibrium pairs. In support of this hypothesis is the observation that generally the sodic amphibole either grew earlier or later than the barroisitic amphibole.

Among the Franciscan tectonic blocks, at least two populations of samples appear to be present. The group of glaucophane + Na- and Al-rich actinolite pairs displays compositions and tie lines similar to the lower grade *in situ* schists from California. The second group is more diverse, but consists of hornblende + glaucophane associations and reflects a paragenesis like that described for the Gruppo di Voltri and Valtournanche rocks in

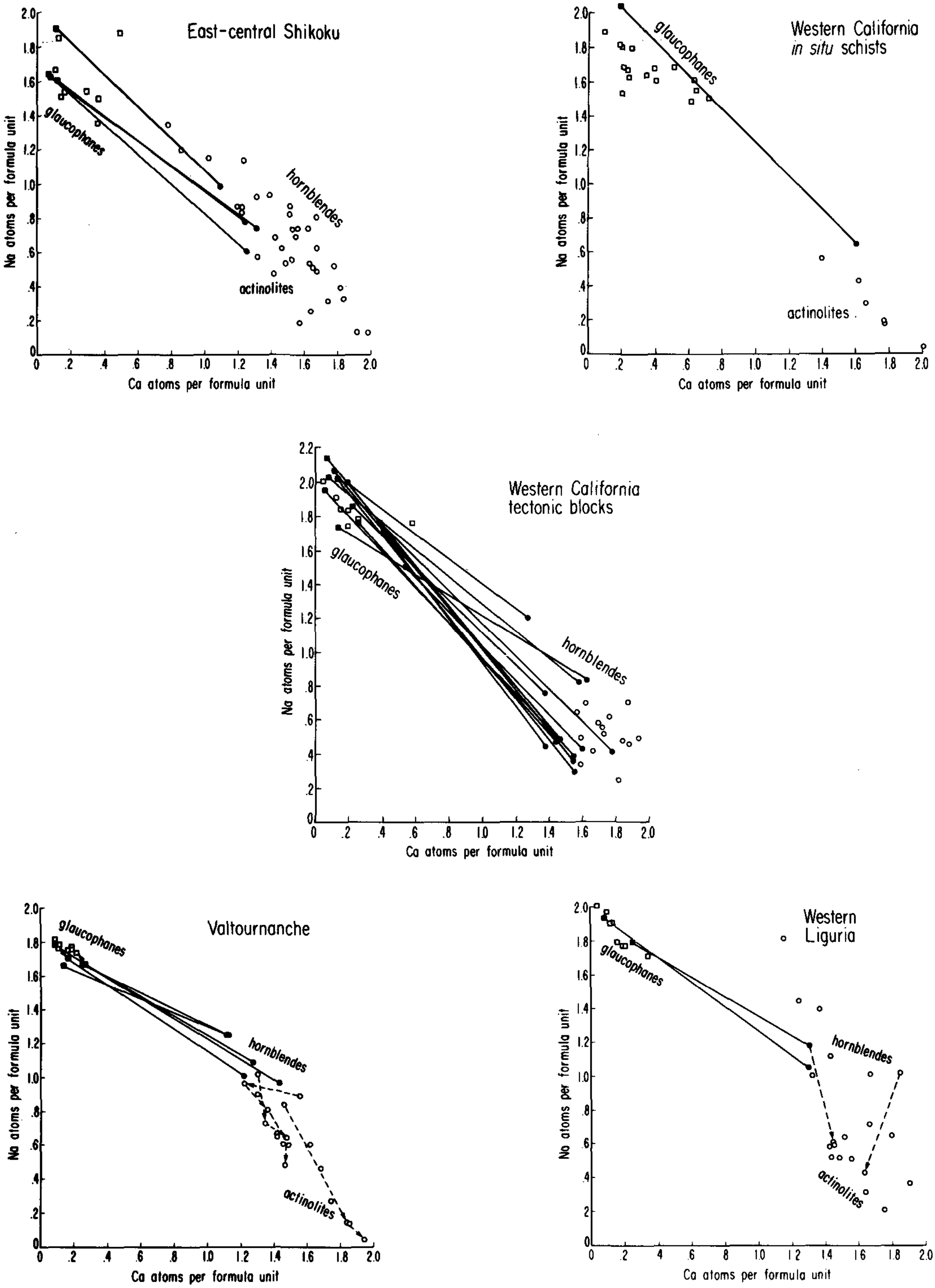

FIG. 2. Na and Ca contents of analysed amphiboles, in the formula unit based on forty-six anionic charges exclusive of the oxygen bonded to hydrogen. Solid symbols represent coexisting amphibole pairs.

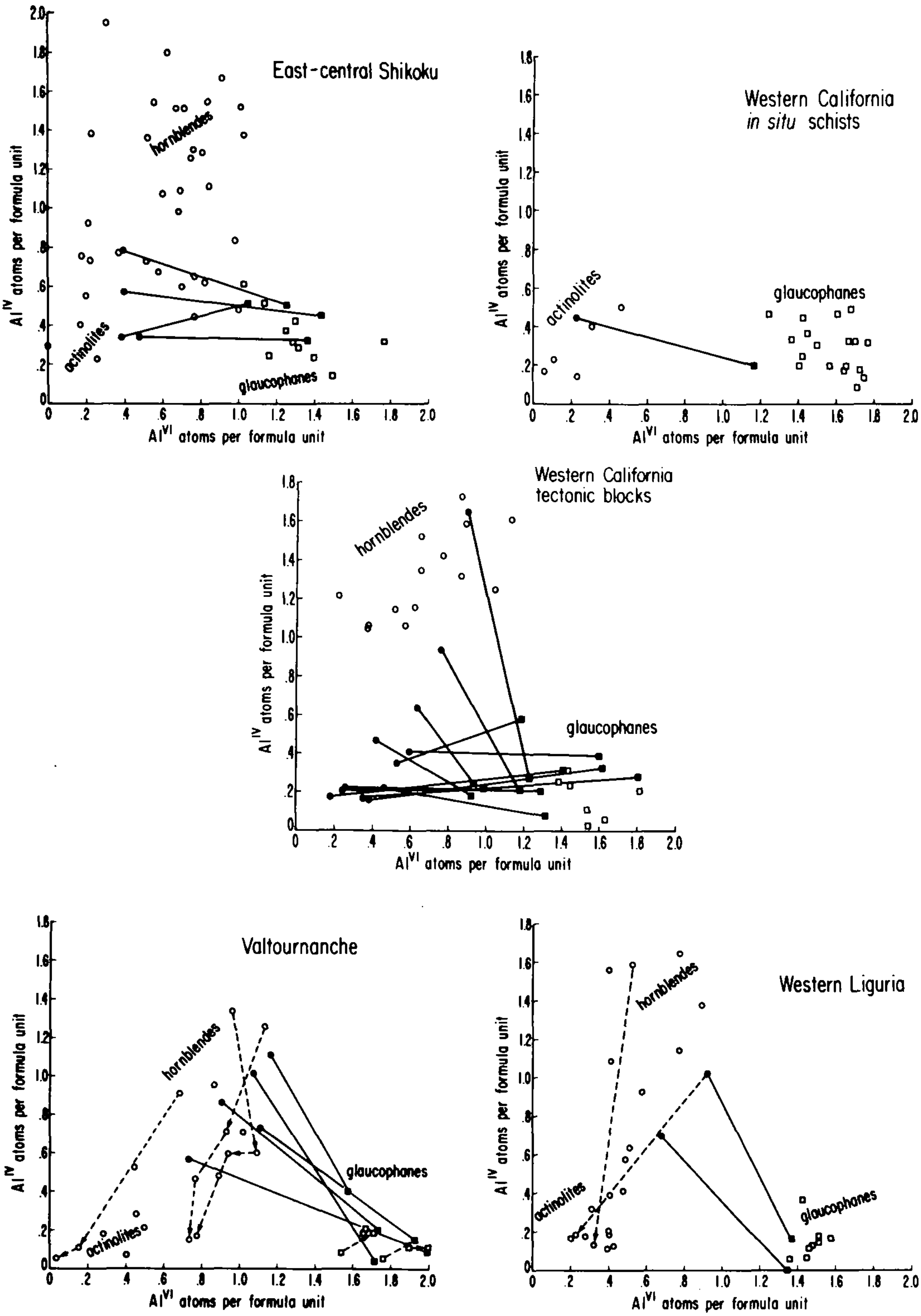

FIG. 3. Al^{iv} and Al^{vi} of analysed amphiboles in the formula unit. Solid symbols represent coexisting amphibole pairs.

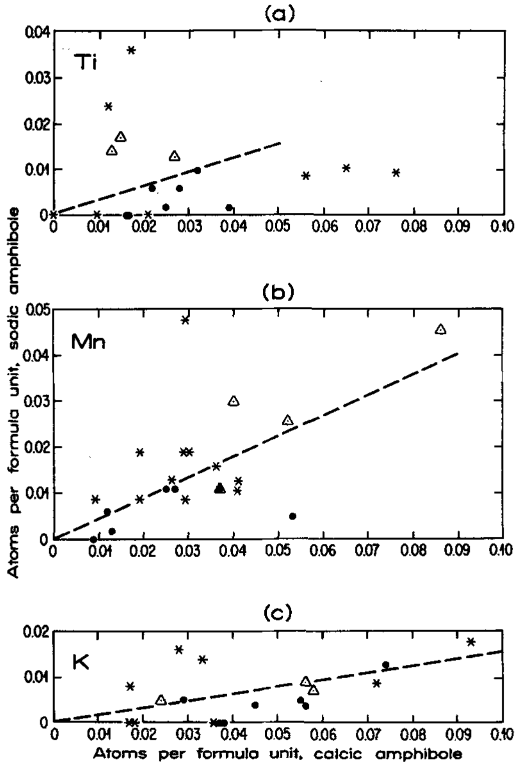

FIG. 4. Partitioning of (*a*) Ti, (*b*) Mn, and (*c*) K per formula unit for coexisting analysed amphibole pairs from the investigated terranes. Triangles = east-central Shikoku; stars = tectonic blocks of W. California; filled circles = Valtournanche and W. Liguria.

which earlier, higher-grade assemblages have been subjected to retrogression.

Numerous studies document the correlation between the Fe/Mg ratio of the host rock and that of the contained amphiboles (e.g. Coleman and Papike, 1968; Miller, 1977). Fractionation of Fe^{2+} and Mg between sodic and calcic amphiboles is modest, and values depend critically on the method of evaluating Fe^{2+}/Fe^{3+} ratios. A rough correlation appears to exist between Na in the calcic amphibole M_4 site as a function of Al^{iv} content, and pressure of formation (Brown, 1977) but because Al^{iv} values critically depend on the accuracy of the SiO_2 determinations and the oxidation states of the iron, some of the scatter evident in fig. 3 is predictable.

The minor element Ti resides chiefly in M_2 and is concentrated in actinolite and hornblende relative to glaucophane (fig. 4*a*). This selectivity seems to be more extreme for analysed pairs from W. Liguria and Valtournanche than for schists from Shikoku and the Franciscan tectonic blocks. However, the two Californian sodic amphiboles that exhibit high Ti content are the gravimetric analyses 50-CZ-60 and GRS. Such 'wet' analyses may reflect the presence of minute rutile or sphene inclusions in the minerals analysed, a view supported by the glaucophane from G-215B, which contains 0.60 wt% TiO_2 by 'wet' analysis, but only 0.11 wt% TiO_2 by microprobe analysis (cf. Ernst *et al.*, 1970, Tables 27, 32). Both Mn, which apparently occurs in M_4, and K, confined to A, are markedly enriched in calcic amphibole relative to coexisting glaucophane (fig. 4*b*, *c*). Black (1973) demonstrated an analogous concentration of Ti, Mn, and K in calcic amphiboles relative to sodic amphiboles from New Caledonia.

Stability field of barroisitic amphibole in metabasaltic rocks

The compositions of the coexisting amphiboles, their mutual relationships, and the inferred *P-T* paths of the samples allow the delineation of a *P-T* stability field for barroisitic amphibole in mafic schists (fig. 5). It should be emphasized that the textural and compositional relations indicate that conversions of barroisitic to actinolitic amphibole and vice versa are completely gradational and so the low-pressure boundary of the barroisitic amphibole field is ill-defined, and continuous.

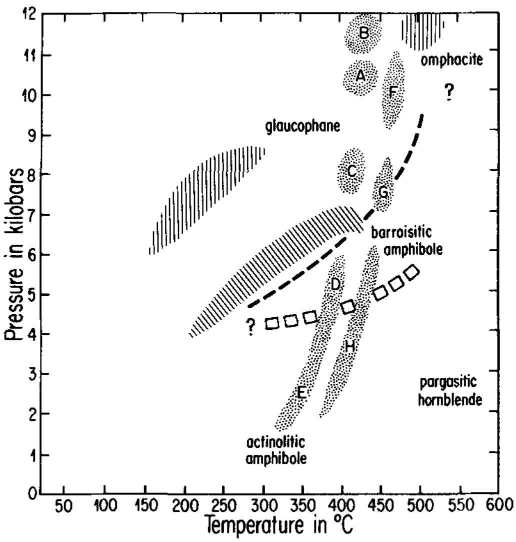

FIG. 5. Stability ranges of sodic and calcic clinoamphiboles in mafic schists as deduced from the information summarized in fig. 1. The designation of barroisitic amphibole and actinolite fields calls attention to chemical contrasts in a smoothly varying continuum of single calcic amphibole phase compositions (hence the use of the broad, open dashed line indicating the gradational nature of this zone 'boundary').

Phase equilibria between glaucophane and barroisitic amphibole are equivocal. The mutual association and the replacement of one by the other are features rather suggestive of the existence of a solvus, yet the fact that some higher-grade barroisitic amphiboles are roughly (but not exactly) intermediate in composition between lower-grade glaucophane and actinolite pairs argues for the possible high-temperature closure of a solvus. In any case, it is clear that blue-green hornblendes occupy a P-T field of stability intermediate between the higher-pressure glaucophanes and the lower-pressure actinolites, as shown in fig. 5. Provisional values for the barroisitic amphibole field are 4-5 kb at *c.* 350 °C and 5-7 kb at *c.* 450 °C.

Maresch (1977) has demonstrated that glaucophane is apparently stable only above 4 kb and temperatures $\leqslant$ 350 °C, but sodic amphiboles can range up to *c.* 550 °C at 10 kb. Hence, depending on the activity of H_2O, eclogitic assemblages evidently replace blueschist ones at such elevated pressures and temperatures. The P-T range for barroisitic amphibole at high temperatures is not known; however, hydrothermal experiments on oceanic tholeiite by Spear (1976) indicate that increased P and T favour the incorporation of substantial amounts of Na, Al^{iv}, and Al^{vi} in calcic, rather pargasitic, amphibole. At low temperatures—say, less than 300 °C—stable coexistence of glaucophane with actinolite in nature evidently signals the termination of the barroisitic amphibole field in metabasalts.

Concluding remarks

Comparable results to those described above have been obtained for coexisting clinoamphibole pairs from the glaucophane schists of the S. Urals, New Caledonia, and the Tauern Fenster of the E. Alps, but the data available to the author are limited (Chesnokov, 1959; Dobretsov *et al.*, 1971; Black, 1973; Miller, 1977; Raith *et al.*, 1977). As more information accumulates from blueschist terranes it may prove possible to employ the Ti contents, the Na/Ca or Na/K and, or, the Al^{iv}/Al^{vi} contrasts between coexisting glaucophanes and actinolite-barroisitic-amphibole solid solutions as indicators of the P-T conditions.

In any case, it is clear from the observed metabasaltic parageneses and assigned P-T paths that, for temperatures of 300-500 °C, barroisitic amphiboles have formed under a range of intermediate pressures, between those attending relatively high-pressure glaucophane schists and low pressures characteristic of greenschist recrystallization.

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