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Calcium-poor pyroxene reaction relations in calc-alkaline magmas

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Abstract

In several calc-alkaline intrusive complexes calcium-poor pyroxene, which is a major phase in the basic compositions, disappears from the more differentiated members of the suite. Several models have been used successfully to explain the disappearance of calcium-poor pyroxene from tholeiitic and potassic suites, but they cannot satisfactorily explain its disappearance from calc-alkaline magmas. The existence of a reaction relationship involving the disappearance of calcium-poor pyroxene and appearance of hornblende is preferred for calc-alkaline suites. An increase in water content with differentiation from $<0.5P_{\text{total}}$ to $>0.6P_{\text{total}}$ at approximately 4–5 kbar is required to explain the crystallization sequence of two well-documented calc-alkaline complexes—Garabal Hill and Guadalupe—in which the disappearance of calcium-poor pyroxene has been reported. There may be considerable variations in the water content of parental magmas to calc-alkaline suites, but they may be significantly undersaturated with water at the pressures of crystallization.

Introduction

The disappearance of calcium-poor pyroxenes from tholeiitic magmas, both intrusive and extrusive, has frequently been observed, and several models have been presented to explain the phenomenon (Poldervaart and Hess, 1951; Brown, 1957; Carmichael, 1960). The mineral may also cease to crystallize in certain calc-alkaline differentiated suites (Nockolds, 1941; Best, 1963), but the previous models cannot be satisfactorily applied to them. It is the intention here to discuss various ways by which calcium-poor pyroxene may cease to crystallize from calc-alkaline magmas.

The term calcium-poor pyroxenes is used to include orthopyroxenes, pigeonites, and inverted pigeonites, and calcic pyroxene covers the augite to hedenbergite series.

Mineral assemblages involving calcium-poor pyroxene disappearance

There are at least four different series of mineral assemblages which may show the disappearance of a calcium-poor pyroxene during magmatic differentiation. (Subsolidus recrystallization and late-stage igneous or secondary alteration to form bastite, actinolitic amphibole, *etc.* are excluded from this discussion.) The crystallization sequences are:

(1) The Skaergaard intrusion (Wager and Brown, 1967); (a) plagioclase–calcic pyroxene–calcium-poor pyroxene; (b) plagioclase–calcic pyroxene–calcium-poor pyroxene–olivine; (c) plagioclase–calcic pyroxene–olivine.
(2) pitchstone glasses from Iceland (Carmichael, 1960); (a) plagioclase–calcic pyroxene–calcium-poor pyroxene; (b) plagioclase–calcic pyroxene; (c) plagioclase–calcic pyroxene–olivine.
(3) the Garabal Hill complex (Nockolds, 1941); (a) plagioclase–calcic pyroxene–calcium-poor pyroxene–hornblende–biotite; (b) plagioclase–calcic pyroxene–hornblende–biotite; (c) plagioclase–calcic pyroxene–hornblende–biotite–quartz.
(4) the Guadalupe complex (Best, 1963); (a) plagioclase–calcic pyroxene–calcium-poor pyroxene–hornblende–biotite–quartz; (b) plagioclase–calcium-poor pyroxene–hornblende–biotite–quartz; (c) plagioclase–hornblende–biotite–quartz.

There may be other possible crystallization sequences in which calcium-poor pyroxene disappears, *e.g.* the Cuillin intrusion, Scotland (Wager and Brown, 1967; Fig. 224), the Baltimore gabbro, Maryland (Herz, 1951), Insch, Scotland (Read *et al.*, 1961); but there are less detailed mineralogical data available for these.

Previously proposed models for calcium-poor pyroxene disappearance

Poldervaart and Hess (1951) suggested that calcium-poor pyroxene might disappear through a reaction relation with liquid to produce olivine and tridymite, a reaction demonstrated by the experimental data of Bowen and Schairer (1935; Fig. 5). In view of the low-temperature breakdown of iron-rich calcium-poor pyroxene to give these phases (Lindsley and Munoz, 1969; Smith, 1972) this seems plausible. A slightly different reaction mechanism is embodied in the phase diagram of Yoder *et al.* (1963; Fig. 16, point *B*) where only tridymite, and not olivine, is produced. However, Carmichael (1960) presented mineralogical data for pitchstones for which the calcium-poor pyroxene ceased to crystallize *before* iron-rich olivine appeared, and a silica polymorph was not involved. Hence, these two reaction relations cannot explain the disappearance of calcium-poor pyroxene in this particular example.

Brown (1957) presented an alternative model to explain the disappearance of calcium-poor pyroxene without necessitating the appearance of another phase. He suggested that there may be a reaction relationship of calcium-poor pyroxene and liquid giving a calcic pyroxene for relatively iron-rich (low-temperature) magmas, whereas for the more magnesium-rich (higher temperature) magmas the two pyroxenes coprecipitated. This model fits extremely well with the experimental data of Kushiro (1972a). The reasoning is explained in some detail as it is important in discussing calc-alkaline magmas.

Phase relations in the system forsterite–diopside–quartz at 1 atmosphere pressure (Kushiro, 1972a), and various natural pyroxene compositions are shown in Figure 1. The relation between the curve calcic pyroxene–pigeonite–liquid (*a*–*b*) and the composition of the minerals is of interest here. The composition of the most calcium-rich calcic pyroxene, which is in equilibrium with a sub-calcic pyroxene, analyzed by Brown (1957) is plotted on Figure 1 as Composition 1.[1] The extension of the phase-boundary curve involving two pyroxenes and liquid intersects the tie-line joining the coexisting pyroxene compositions. [The calcium-poor pyroxene composition is not shown, but plots close to the (Mg, Fe)SiO_3 composition.] Hence, the two pyroxenes *coprecipitate*. With continued crystallization the calcic pyroxene becomes less calcic. The calcic pyroxene with the lowest Ca content in equilibrium with a calcium-poor pyroxene from Brown (1957) is plotted on Figure 1 as Composition 2. Now, the extension of the phase boundary curve intersects the two pyroxene tie-line, not between them, but on an extension of the line further away from the calcium-poor pyroxene [using the position of Composition 2 determined by the method of Kushiro (1962) or Cawthorn and Collerson (1974)]. This geometrical situation demands that the calcium-poor pyroxene be in reaction relation with the liquid to produce calcic pyroxene. Using this diagram, it can be seen that if the calcic pyroxene becomes less calcic than C^1 a reaction relationship with the disappearance of calcium-poor pyroxene may be anticipated. Thus Brown's (1957) model appears to be explained by the

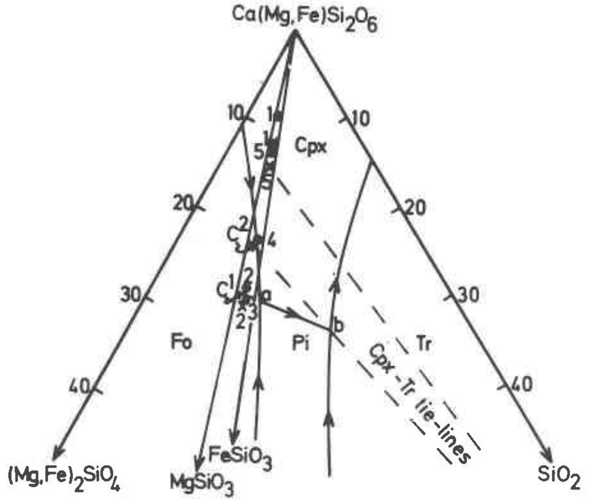

FIG. 1. Phase relations in part of the system $(Mg,Fe)_2SiO_4$–$Ca(Mg,Fe)Si_2O_6$–SiO_2 (in weight percent) with phase boundaries for the iron-free system at 1 atmosphere from Kushiro (1972a). Calcic pyroxene compositions: 1—analysis 1 from Brown (1957), 2—analysis 7 from Brown (1957), 3—least calcic mineral from Insch complex, Scotland (Wager and Brown, 1967), 4—mineral composition where calcium-poor pyroxene disappears from Insch complex, Scotland (Wager and Brown, 1967), 5—analysis 153 from Best and Mercy (1967). Solid circles are compositions determined from their Ca:Mg:Fe^{2+} ratio; crosses are compositions determined by methods of Kushiro (1962) and Cawthorn and Collerson (1974). C^1 and C^2 are calcic pyroxene compositions for which reaction relationship of calcium-poor pyroxene is possible (see text). Abbreviations: Fo-forsterite; Pi-pigeonite; Cpx-calcic pyroxene; Tr-tridymite.

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[1] There are two alternative methods for plotting natural pyroxene compositions in Fig. 1. From the Ca, Mg, and Fe^{2+} content of the mineral, the weight percent wollastonite, enstatite, and ferrosilite may be calculated. Alternatively, allowance may be made for Ti, Al, Fe^{3+}, Na as pyroxene end-member compositions by using the methods of Kushiro (1962) or Cawthorn and Collerson (1974) and the weight proportions of wollastonite, enstatite, and ferrosilite found. These latter two calculations give almost identical compositions in Fig. 1, but are slightly different from that obtained by the first method.

experimental phase diagram, provided there is no significant shift in the orientation of the phase boundary (*a–b*) with changing Mg/Fe ratio.

There is a second potential reaction relationship which may be anticipated from inspection of Figure 1. This occurs at the pseudoinvariant point (*b* in Fig. 1) where calcic pyroxene–calcium-poor pyroxene–tridymite–liquid coexist, and is a more precise location of this point inferred by Yoder *et al.* (1963; Fig. 16). If the composition of the calcic pyroxene in this equilibrium is less calcic than C^2 in Figure 1, this point will be a peritectic, and with further crystallization the liquid will migrate along the curve involving calcic pyroxene and tridymite. However, if the calcic pyroxene plotted closer to diopside than C^2, this point would become a eutectic, and calcium-poor pyroxene would not be in reaction relationship.

A possible example of a calcium-poor pyroxene reaction at a point analogous to *b* in Figure 1 is the Insch complex in Scotland. The data reported by Wager and Brown (1967; Fig. 236) show calcium depletion in calcic pyroxenes to a point very close to C^1 (Composition 3 in Fig. 1) and a subsequent enrichment in calcium, while still in equilibrium with a calcium-poor pyroxene. The calcium-poor pyroxene reacts out only when the calcic pyroxene has a composition close to C^2 (Composition 4 in Fig. 1). For compositions of calcic pyroxene between 3 and 4, coprecipitation of two pyroxenes may be expected while the liquid changes along *a–b*; but a reaction relationship would occur as soon as the liquid reached point *b*. However, the Insch complex has syenitic residual liquids, and quartz is a very minor phase, hence the use of Figure 1 may be questioned.

The syenites from Insch contain an appreciable amount of alkali feldspar and biotite (Sadashivaiah, 1954), and so the study by Luth (1967) on potassium-rich melts may be more appropriate. He demonstrated the plausibility of two reactions by which calcium-poor pyroxene may disappear:

enstatite + orthoclase + liquid = phlogopite + quartz

enstatite + orthoclase + vapor = phlogopite + liquid.

By equating orthoclase and phlogopite in the experimental study with alkali feldspar and biotite respectively for natural material, these reactions may be relevant to the crystallization sequence of the Insch mass.

It therefore seems that for tholeiitic and potassic liquids the disappearance of calcium-poor pyroxene has been adequately explained.

Calcium-poor pyroxene disappearance from calc-alkaline magmas

The crystallization sequence of the Guadalupe complex was compared with that of the Skaergaard complex by Best (1963; Fig. 6). A similar diagram is presented here on Figure 2 for the data compiled by Nockolds (1941) on the Garabal Hill complex in Scotland. Olivine does not reappear in either of these crystallization sequences, and quartz is not a major phase until *after* the disappearance of calcium-poor pyroxene (Fig. 2; and Best, 1963, Fig. 6). Hence the models of Poldervaart and Hess (1951) and Bowen and Schairer (1935) cannot apply here. The alternative model of Brown (1957) is unsatisfactory for the Guadalupe complex, as Best (1963) showed that the calcic pyroxene disappeared *before* the calcium-poor pyroxene. Alkali feldspar does not appear in the Garabal Hill sequence until *after* calcium-poor pyroxene has reacted out, and so neither of the two reactions of Luth (1967) involving alkali feldspar and calcium-poor pyroxene can have taken place. Hence, none of the previously postulated models, which explain the calcium-poor pyroxene reaction relationship in tholeiitic and potassic melts, appear to explain this phenomenon in calc-alkaline magmas.

Several analyses of calcic pyroxenes from the Guadalupe complex were presented by Best and Mercy (1967). These minerals do not show any significant decrease in Ca-content during crystallization as displayed by the Skaergaard pyroxenes. Therefore, the composition of the calcic pyroxene when calcium-poor pyroxene disappears shows very limited solubility towards $(Mg,Fe)SiO_3$. As the Skaergaard pyrox-

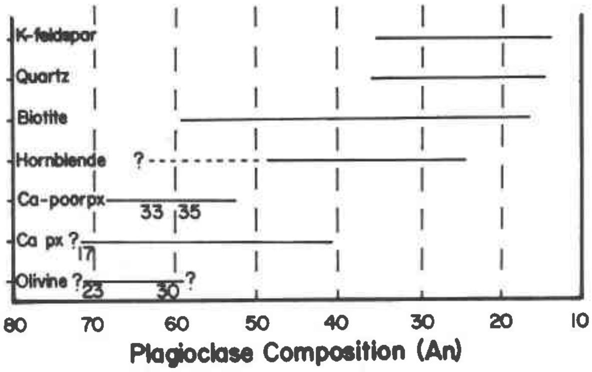

FIG. 2. Mineralogical variation in the Garabal Hill complex (Nockolds, 1941) for comparison with Skaergaard and Guadalupe intrusions in Best (1963; Fig. 6). Numbers indicate $Fe^{2+}/(Fe^{2+} + Mg)$ ratio for olivine and pyroxenes. Secondary hornblende after calcic pyroxene is not unambiguously distinguishable from igneous hornblende, and hence its range is uncertain.

enes crystallized at approximately 1050–1100°C (Brown, 1957) whilst those from Guadalupe probably crystallized at about 950°C (Best and Mercy, 1967), this decreased solubility is exactly as predicted by Davis and Boyd (1966) from the shape of the pyroxene solvus.

The least calcium-rich calcic pyroxene in equilibrium with calcium-poor pyroxene from Guadalupe is plotted on Figures 1 and 3 as Composition 5. It is distinctly more calcic than Composition C^2, 'and hence a reaction relationship is not possible (in terms of the 1 atmosphere pressure phase relations). With increasing water pressure the position of the invariant point involving 2 pyroxenes–quartz–liquid will shift. However the only way it could produce a reaction relationship situation would be for it to plot in the stippled region of Figure 3, within the tie-triangle SiO_2–$Ca(Mg,Fe)Si_2O_6$–calcic pyroxene, Composition 5. This would imply that during crystallization calc-alkaline magmas become extremely rich in normative diopside relative to hypersthene (to plot in the stippled area of Fig. 3). Chayes (1969) showed exactly the reverse trend in normative compositions, in that calc-alkaline magmas had a much lower normative diopside to hypersthene ratio than tholeiitic suites, and that frequently calc-alkaline magmas were corundum-normative. This would indicate that the phase boundary curves which control calc-alkaline magma crystallization move in the opposite direction from that required to produce a calcium-poor pyroxene reaction relationship.

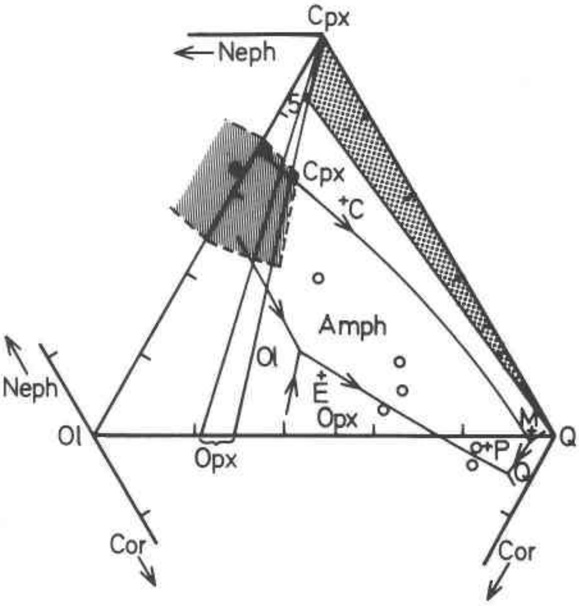

FIG. 3. Projection from plagioclase into the system olivine–calcic pyroxene (or corundum)–quartz (or nepheline) in weight percent, showing phase relations under 3–5 kbar water pressure. Location of amphibole primary phase field inferred from experimental data on Composition C (from Cawthorn *et al.*, 1973, sample 198) and E (from Eggler and Burnham, 1973) both at 5 kbar pressure, P (from Piwinskii and Wyllie, 1970, sample 678) and M (from McDowell and Wyllie, 1971, sample 26272) both at 3 kbar pressure. Stippled area—liquid composition required for reaction relation of calcium-poor pyroxene to give calcic pyroxene (Composition 5, same as Fig. 1). Open circles—analyses of uncontaminated fresh rock samples from Nockolds (1941). Solid circles—amphibole analyses from Best and Mercy (1967). Ruled area—amphibole solid solution limits in igneous rocks from Cawthorn (1976).

Crystallization conditions of calc-alkaline magmas

To explain why calcium-poor pyroxenes disappear from calc-alkaline suites during differentiation it is necessary to determine the physical conditions of crystallization. Nockolds (1941) and Best and Weiss (1964) reported the mineral assemblage andalusite–sillimanite–cordierite–biotite in the contact-metamorphic aureoles around the Garabal Hill and Guadalupe complexes. Using the temperature–pressure grid of Hess (1969), this assemblage may be stable under 4–5 kbar pressure. Yoder and Tilley (1962) and Holloway and Burnham (1972) showed that at pressures greater than 2 kbar, amphibole crystallized before plagioclase from basic magmas for $fH_2O > 0.6P_{total}$. In both the Guadalupe and Garabal Hill complexes plagioclase begins to crystallize before hornblende (Best, 1963; Nockolds, 1941), and if these rocks formed at 4–5 kbar, the water activity must have been fairly low. There are other arguments against high water fugacity in the magma during the early stages of crystallization. Under 5 kbar water pressure the assemblage olivine–calcic pyroxene–plagioclase is not stable (Cawthorn, 1976) but is replaced by hornblende-bearing equilibria. Yet, this gabbroic assemblage is observed in many calc-alkaline bodies. At high water pressure calcium-poor pyroxenes may not crystallize from olivine-normative magmas (Kushiro, 1972b). Some of the rocks from Guadalupe and Garabal Hill are olivine-normative and yet contain calcium-poor pyroxene. If these rocks are assumed to represent liquids rather than cumulates their compositions again suggest fairly low water pressure.

The early crystallization of hypersthene gabbro builds up the fugacity of water to the point where hornblende crystallizes. Holloway (1973) showed that the thermal stability of pargasite increases dramatically for a small change in fH_2O, the critical fH_2O at 5 kbar being 0.5–$0.6P_{total}$. Hence, with a small degree of crystallization there will be a rapid change in the phase relations from those essentially anhydrous to those essentially hydrous. The phase relations under 5 kbar water pressure are shown in Figure 3. The extent of the amphibole stability field is determined by reference to the experimental studies of McDowell and Wyllie (1971) and Cawthorn *et al.* (1973) for the calcic pyroxene–amphibole curve, of Eggler and Burnham (1973) for the calcium-poor pyroxene–amphibole curve, and of Piwinskii and Wyllie (1970) for the amphibole–quartz curve. Typical calc-alkaline compositions from Garabal Hill now project within the amphibole stability field, and there is a rapid change in crystallization from 2 pyroxenes—plagioclase to amphibole–plagioclase. Thus the virtually simultaneous disappearance of both calcic and calcium-poor pyroxene, as observed in the Guadalupe and Garabal Hill complexes, is predicted. Therefore, the disappearance of calcium-poor pyroxene is due to increasing water fugacity, and is rather different from the usual kind of reaction relationship which may be explained by changes in the major element chemistry of the liquid. The trend of these calc-alkaline compositions can be seen in Figure 3 to be directly out from typical amphibole compositions, and hence the crystallization of amphibole may explain the chemical variation in these suites.

The stage during crystallization at which the reaction relation occurs may be largely dependent upon the initial water fugacity. According to the data of Best (1963; Fig. 6), calcium-poor pyroxene disappears when the feldspar composition is An_{23}, whereas for the Garabal Hill complex pyroxene disappears when the composition is An_{63}. Assuming the pressures of crystallization for these two suites to be similar (see above), this may indicate that the Guadalupe magma contained less water than that at Garabal Hill and underwent more extensive crystallization before the fugacity of water became sufficient to stabilize amphibole. Herz (1951) has presented an example of an essentially tholeiitic trend, which only in the very end-stages of differentiation crystallizes amphibole, loses calcium-poor pyroxene, and causes the well-known depletion in iron typical of calc-alkaline trends. This may represent the crystallization sequence of a magma with lower water contents than most calc-alkaline liquids, but higher water contents than most tholeiites.

Summary

The disappearance of calcium-poor pyroxene from two calc-alkaline intrusive complexes (Garabal Hill and Guadalupe) with differentiation cannot be explained by the models proposed for the reaction relation of calcium-poor pyroxene in tholeiitic or potassic magmas (Brown, 1957; Luth, 1967). Calcic pyroxene disappears before calcium-poor pyroxene in the Guadalupe complex (Best, 1963); thus invalidating the application of Brown's (1957) model for tholeiitic magmas, and alkali feldspar does not appear until after calcium-poor pyroxene has disappeared, invalidating Luth's (1967) model for potassic magmas. The mineralogy of the contact metamorphic rocks suggests that these complexes crystallized under approximately 4–5 kbar pressure. At constantly high fH_2O calcium-poor pyroxene would not crystallize from olivine-normative magmas (Kushiro, 1972b). At $fH_2O > 2$ kbar plagioclase would not crystallize before amphibole from gabbroic magmas (Holloway and Burnham, 1972). At high fH_2O the assemblage olivine-calcic pyroxene–plagioclase should not be stable (Cawthorn, 1976). All these predictions are contrary to the observations on the two complexes. The composition of coexisting pyroxenes is such that calcium-poor pyroxene could only react out if the liquid became extremely diopside-normative (Fig. 3), which is contrary to the calc-alkaline trends which usually have low normative diopside, or even normative corundum.

One way of explaining these observations is to suggest that the basic magmas crystallized under fairly low water fugacity which built up during fractionation until amphibole became stable (at $fH_2O = 0.6P_{\text{total}}$), replacing both calcic and calcium-poor pyroxene. The subsequent crystallization of amphibole may produce the calc-alkaline liquid evolution trends.

This interpretation does not conflict with any of the ideas presented by Nockolds (1941) or Best (1963) and Best and Mercy (1967)—it merely makes use of more recent experimental data to predict more closely the probable crystallization conditions of some calc-alkaline intrusive complexes and the reasons for the disappearance of calcium-poor pyroxenes.

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