Formation of chlor- and fluor-apatite in layered intrusions

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Abstract

Apatite in most igneous intrusions has a high Cl/F ratio. However, chlor-apatite has been reported in the lower portions of the Bushveld and Stillwater Complexes. This has been used as evidence supporting the early separation of a Cl-rich discrete hydrous fluid in these intrusions. Mineralogical evidence is presented here to demonstrate that the Bushveld Complex, at least, formed from a nearly anhydrous magma, and did not release a hydrous fluid before apatite began to crystallize. It is suggested that apatite in the earliest cumulates equilibrated with trapped interstitial liquid, which converted it from the typical F-rich composition of cumulus apatite to a Cl-rich composition. This is an analogous process to that in which cumulus mafic minerals may become more Fe-rich on cooling and reaction with interstitial liquid.

KEYWORDS: apatite, layered intrusions, Cl/F ratio, Bushveld Complex.

Introduction

APATITE is a cumulus phase in many layered basic intrusions, associated with the more differentiated rocks. In such cases, it is typically F-rich (e.g. Brown and Peckett, 1977). However, it is also present in very minor amounts even in some of the earliest cumulates (Boudreau *et al.*, 1986). The detailed study of apatite as an intercumulus phase in the Stillwater and Bushveld intrusions (Boudreau and McCallum, 1989; Boudreau and Kruger, 1990) focused on the halogen content, and showed that the ratio of Cl/F may vary widely both within single samples and in short vertical sections. The model proposed by these authors to explain the high Cl content of apatite presumes that a hydrous Cl-bearing fluid phase separated at an early stage of fractionation and preferentially removed Cl, thereby decreasing the Cl/F ratio in the magma and in apatite which subsequently formed. Fundamental to this model is the requirement that a fluid phase be liberated from the magma before apatite saturation.

Confirmation of this process is important because Cl-rich fluids may transport base metals and possibly also platinum-group elements (PGE) (Boudreau and McCallum, 1992). This paper briefly reviews information about apatite in layered intrusions, examines the evidence for fluid-saturated conditions, especially in the Bushveld Complex, and then proposes an alternative mechanism for variations in halogen content of apatite.

Apatite in layered intrusions

The abundance of apatite in layered intrusions is extremely variable. For example, up to 30% apatite has been reported in layers in the uppermost Bushveld Complex (Cawthorn and Walsh, 1988), while near the base of this intrusion chemical analyses indicate that some adcumulate rocks may contain as little as 0.01% apatite, and is normally overlooked during optical investigation. When a cumulus mineral, apatite displays euhedral hexagonal to elongated, prismatic outline often erratically distributed throughout the rock in clusters. Boudreau and Kruger (1990) reported that apatite only occurred in 30% of all thin sections studied from samples close to the Merensky Reef in the Bushveld Complex, and that it occurred in tiny pools of intercumulus grains of clinopyroxene, mica and quartz. Apatite is idiomorphic and up to 0.02 mm in size. It often occurred close to grain boundaries of cumulus phases.

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In the lower half of the Stillwater Intrusion, Boudreau and McCallum (1989) reported very similar observations for the intercumulus apatite. The mineral was never enclosed in cumulus grains and was anhedral when abutting such phases. The uppermost sequence of the Stillwater which presumably would have contained cumulus apatite is not exposed. Similar relationships are noted for the Munni Munni Intrusion (Hoatson and Keays, 1989; Boudreau, in press), where 0.01 mm grains of apatite occur with any of quartz, alkali feldspar, biotite and magnetite, interstitial to plagioclase and pyroxene.

In the Skaergaard Intrusion apatite becomes a cumulus phase close to the top of the intrusion, although is present as small, anhedral grains throughout the entire sequence (Brown and Peckett, 1977). Analyses of a great many whole rock samples from this intrusion by McBirney (1989) indicate that the P_2O_5 and hence apatite content varies considerably. The lowest values are reported, not near the base of the intrusion, but in the Middle Zone, suggesting that these are more adcumulate than the lowest rocks and contain as little as 0.1% to as much as 1% apatite. Even at a level below the appearance of cumulus apatite its abundance increases, as evidenced by increased P_2O_5 contents, indicating in excess of 1% apatite. Once it becomes a cumulus phase, typically 2% is present in most rocks. Examples of cumulus and intercumulus apatite are well documented by Wager and Brown (1968).

These examples illustrate the differences in apatite mode between intrusions which contain dominantly adcumulate rocks (Bushveld) and those with orthocumulate rocks (Skaergaard).

Cl/F in apatite

The Cl/F ratios in apatite from several intrusions are summarized in Fig. 1. Typically there is a wide scatter of compositions even within single samples, although individual grains are usually homogeneous (Boudreau and McCallum, 1989; Boudreau and Kruger, 1990). Apatite in the lower portion of ultramafic cumulates and in the J-M Reef, where platinum-group element (PGE) mineralization

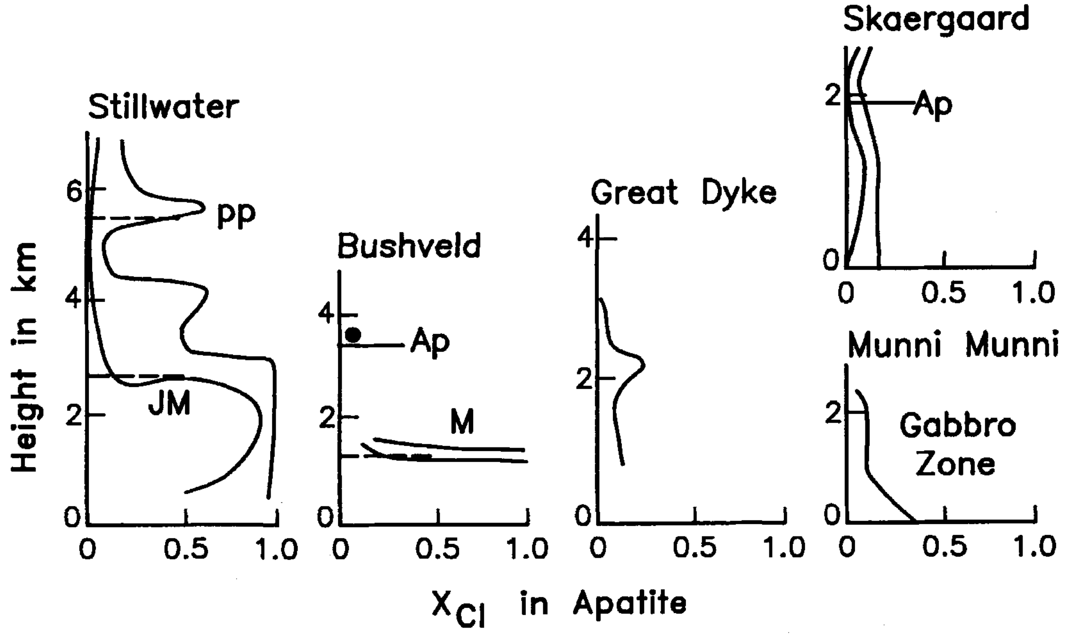

Fig. 1. Plot of the range of X_{Cl} (Cl/Cl + F + OH) values in apatite as a function of height in various intrusions. Data are taken from Brown and Peckett (1977), Boudreau *et al.* (1986), Boudreau and Kruger (1990), Love *et al.* (1992), Boudreau (in press). JM and PP in the Stillwater Complex refer to the Johns-Manville and Picket Pin PGE-mineralized horizons; and M in the Bushveld Complex refers to the Merensky Reef. For the Munni Munni Intrusion only analyses from above the mineralized zone are included. For the Bushveld Complex and the Skaergaard Intrusion the appearance of cumulus apatite is indicated by Ap.

occurs, of the Stillwater Intrusion is extremely high in Cl, and decreases irregularly upwards. Fewer data are available for the Bushveld Complex. Close to the level of the Merensky Reef with its PGE-mineralization the apatite is Cl-rich. Cumulus apatite, near the top of the intrusion, is F-rich. In contrast, throughout the Skaergaard Intrusion the apatite is poor in Cl, decreasing slightly upwards (Brown and Peckett, 1977). An unusual gold-bearing horizon, which is also significantly enriched in palladium occurs within the Skaergaard Intrusion (Bird *et al.*, 1991). While no analyses of apatite are available from this horizon, an analysis from less than 100 m above this level was reported by Brown and Peckett (1977) and was poor in Cl. In the Munni Munni Intrusion, which has a zone of very significant PGE enrichment, dominated by palladium (Hoatson and Keays, 1989), the concentration of Cl in apatite is also fairly low, and decreases upwards. The Great Dyke also has a PGE-mineralized horizon (at a height of 2 km in Fig. 1), and the Cl content of apatite is low. Hence, PGE enrichment does not necessarily appear to be associated with chlor-apatite.

The upper portions of the Munni Munni, Stillwater and Great Dyke are not preserved or exposed, and so the composition of apatite when it became a cumulus phase is not known. However, in the three intrusions, Skaergaard (Brown and Peckett, 1977), Kiglapait (Huntington, 1979) and Bushveld (Boudreau and Kruger, 1990) which do have a preserved zone of cumulus apatite, its composition is F-rich (Fig. 1). If such a composition reflects selective loss of Cl in a fluid phase as proposed by Boudreau and McCallum (1992), then the inference is that all such intrusions reached vapour-saturation long before apatite became a cumulus phase. Hence, all such intrusions must have had a high initial water content.

Water content of the Bushveld magma

Boudreau (1992) stated that the Bushveld magma became saturated with water after about 25% crystallization, liberating a water-dominated fluid. It was estimated that the magma contained 5% H_2O at this stage. This requires that the parental magma contained about 4% H_2O, an unusually high water content for a basic to ultrabasic magma. Several lines of evidence suggest that this is implausible.

The parental magma to the lower portion of the Bushveld Complex has been inferred from sill compositions (Cawthorn *et al.*, 1981) on which anhydrous melting experiments confirmed the observed crystallization sequence (Cawthorn and Davies, 1983). If the magma had contained as much as 4% H_2O then the crystallization sequence would have been very different, specifically resulting in olivine being the sole liquidus mineral for a far greater temperature interval. This would have disagreed with the observed orthopyroxene dominance in the Lower and Critical Zones of the Bushveld Complex (Cameron, 1978).

The pyroxenes in the Bushveld Complex provide qualitative constraints on their temperature of crystallization. Their extensive exsolution indicates crystallization at high temperature. Further, the composition at which orthopyroxene is replaced by pigeonite in the crystallization sequence, as determined by Atkins (1969), is consistent with the results of studies by Lindsley (1983) under anhydrous conditions. The presence of pigeonite and the extent of exsolution in pyroxenes of the Bushveld Complex contrasts with the pyroxenes in the Guadalupe Complex which did form under hydrous conditions (Best, 1963). In the Guadalupe Complex mutual solid solution is less extensive and primary orthopyroxene is present throughout the entire differentiation sequence. The Bushveld pyroxenes also contrast with those from the Fongen-Hyllingen complex (Wilson *et al.*, 1981), which formed under elevated water pressure. There the extent of mutual solubility, and hence subsequent exsolution, is much lower than in the Bushveld Complex, and the crystallization of pigeonite only commenced at extremely iron-rich compositions. Thus the pyroxenes of the Bushveld Complex have all the characteristics of phases formed under anhydrous conditions.

Finally, Spulber and Rutherford (1983) showed that typical basaltic magma would crystallize hornblende at higher temperatures than plagioclase at pressures of 2 kbar under water-dominated conditions. Hornblende only becomes a minor phase at the extreme top (about 200 m below the roof) of the Upper Zone (Molyneux, 1974), whereas apatite became a cumulus phase 600 to 1000 m below the roof (Molyneux, 1974; Cawthorn and Walsh, 1988).

These three lines of evidence suggest that the primary Bushveld magma was not close to water-saturation, and that the estimate of several per cent water by Boudreau (1992) is excessive.

Origin of chlor-apatite

Where apatite is a cumulus phase, as in the Skaergaard, Bushveld and Kiglapait Intrusions the mineral is F-rich, and hence the Cl-rich compositions in the lower portions of the Bushveld and Stillwater intrusions are anomalous. Two mechanisms which explain why intercumulus apatite from far below the level of apatite saturation is Cl-rich can be envisaged, and may be distinguishable when more data are available, especially for the Bushveld Complex.

Cl and F in parental magmas. It is now well established that the mineralized horizons of the Stillwater and the Bushveld intrusions represent the boundaries between two very different magma compositions (Kruger and Marsh, 1982; Irvine *et al.*, 1983). In both cases it is possible that the first magma was rich in Cl relative to F. In this event, apatite from all the samples below the mineralized zones would be Cl-rich, and those above would be F-rich. The Bushveld magma had a very high initial $^{87}Sr/^{86}Sr$ ratio, which has been attributed to crustal contamination (Cawthorn *et al.*, 1981; Barnes, 1989). Partial melting of crustal rocks, enriched in Cl relative to basic magma (Faure, 1992) could have led to elevated Cl abundances. In contrast, the Great Dyke has an initial $^{87}Sr/^{86}Sr$ ratio typical of mantle (Hamilton, 1977). Thus, high Cl content may reflect crustal contamination rather than fluid activity.

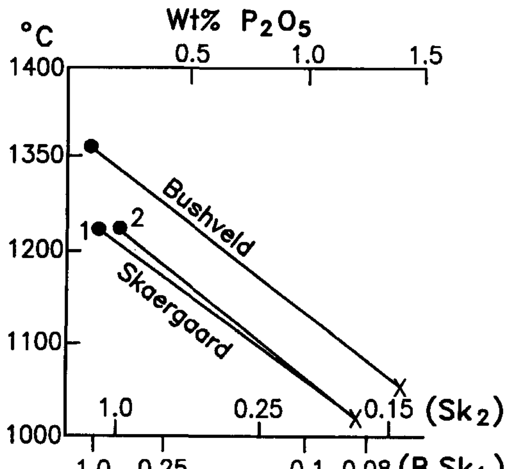

Fig. 2. Plot showing degree of fractionation required for apatite saturation in the Bushveld Complex and Skaergaard Intrusion. The parental magmas are shown by solid circles, that to the Bushveld Complex being taken from Cawthorn *et al.* (1981) and two estimates for the Skaergaard Intrusion from Wager and Brown (1968) and Hunter and Sparks (1987). Liquidus temperatures are taken from Cawthorn and Davies (1983) and Biggar (1974). The temperature and P_2O_5 contents at apatite saturation are taken from Green and Watson (1982). The fraction of liquid remaining (F) at apatite saturation is shown.

The data in Fig. 1 suggest that there may be a gradual decrease in the ratio of Cl/F in apatite upwards in the Skaergaard, Great Dyke, the gabbro zone of Munni Munni and somewhat more irregularly also for the Stillwater intrusions. These are the only intrusions for which an adequate database exists. These trends suggest that variations in Cl/F in apatite are not solely due to multiple intrusion of magmas with different Cl/F ratios as suggested in the previous paragraph.

Degree of fractionation at apatite-saturation. It is here suggested that the proportion of residual liquid in the rock at the time that intercumulus apatite formed is crucial to the prediction of its composition. The initial composition of the parent magma and the P_2O_5 content at apatite-saturation are required to demonstrate this effect. The P_2O_5 content of the parental magmas to the Skaergaard and Bushveld intrusions are shown in Fig. 2, based on analyses by Wager and Brown (1968) and Hunter and Sparks (1987) for Skaergaard, and Cawthorn *et al.* (1981) for the Bushveld Complex. Two different initial parent compositions given for Skaergaard reflect the possible range of values. Liquidus temperatures are taken from the experimental studies of Biggar (1974) and Cawthorn and Davies (1983). The saturation of apatite in the Skaergaard magma has been determined experimentally by Green and Watson (1982), and from variation diagrams using lava compositions (Hunter and Sparks, 1987). Both approaches yielded a value close to 1.2% P_2O_5. The fractionation trend for P_2O_5 is shown as a function of temperature and fraction of liquid remaining (F) on Fig. 2; the numbers below the axis refer to a Bushveld and Skaergaard parent magma with 0.1% P_2O_5. The numbers above the axis relate to an initial P_2O_5 content of 0.2% for Skaergaard (Hunter and Sparks, 1987). As the anorthite content of the plagioclase at apatite saturation in the Bushveld Complex (An_{48}) is higher than for Skaergaard (An_{39}) (Molyneux, 1974; Wager and Brown, 1968), a higher temperature is inferred for apatite saturation in the Bushveld Complex. This requires a greater P_2O_5 content, which demands greater degrees of fractionation. The fraction of liquid remaining at apatite saturation can be seen to vary from 0.2 to 0.05 depending on the intrusion, the parent magma composition, and the temperature of apatite saturation.

Proportion of trapped liquid in the rock at apatite saturation. The proportion of interstitial liquid remaining within the cumulate rocks when apatite forms can now be determined using these figures. This is shown in Fig. 3. The vertical axis shows the height in the intrusion up to the level where apatite becomes a cumulus phase. Immediately below this level there will be a packing of cumulus crystals with interstitial liquid which will be virtually apatite-saturated. Two models are shown here. In Fig. 3A it is assumed that there is 30% interstitial space, which might be appropriate for the Skaergaard Intrusion (Wager and Brown, 1968). Here apatite will begin to crystallize while there is 30% of interstitial liquid present. At progressively greater depths, where the trapped liquid is not apatite-saturated, an increasing degree of crystallization will have to occur within the interstitial space before it does become saturated with apatite. At the lowest levels exposed in the Skaergaard the weight fraction of liquid remaining when apatite saturation is reached will be 0.1 to 0.2 of the volume of interstitial liquid originally trapped as the intercumulus component (Fig. 2), or 3 to 6% of the total rock. (The upper figure of 6% is used in Fig. 3A.).

In the second model, the Bushveld Complex is more nearly an adcumulate and so there is less interstitial liquid. A figure of 10% for the total proportion of interstitial liquid is used here. This estimate is based on an evaluation of the incompatible trace element concentrations in the rocks of the Main and Upper Zones of the Bushveld Complex (Cawthorn and McCarthy, 1985, Cawthorn *et al.*, 1991). If the fraction of liquid remaining (F) at apatite saturation is only 0.08 and if there is only 10% interstitial liquid initially then the liquid will only occupy 0.8% of rock near the base of the intrusion when apatite crystallizes, as shown in Fig. 3B. The proportion of liquid remaining when apatite crystallized will be one of the most important factors controlling whether that liquid is truly trapped or can migrate and communicate with the overlying liquid.

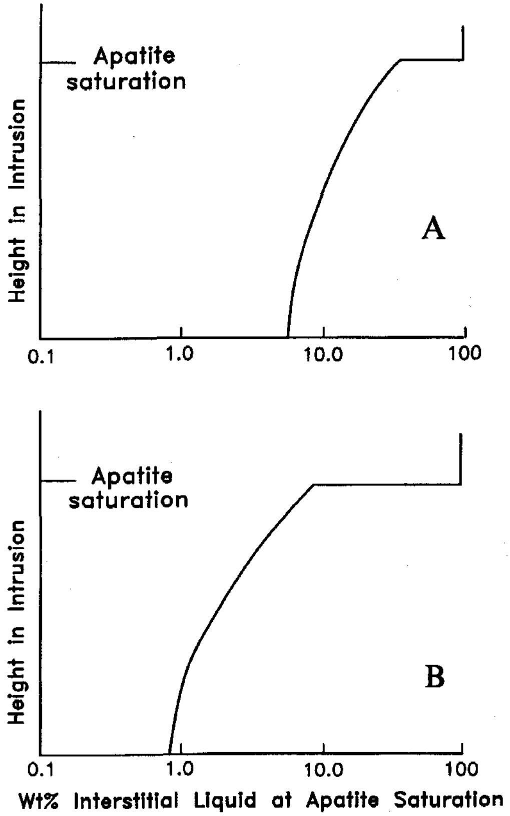

Fig. 3. Estimates of the proportion of interstitial liquid in rock at the point of apatite saturation in the Skaergaard Intrusion (Fig. 3A) and the Bushveld Complex (Fig. 3B). The lines denoted apatite saturation indicate where apatite becomes a cumulus phase.

Migration versus trapping of interstitial liquid. McKenzie (1984, 1987) showed that at porosities of 1%, liquid could still migrate along grain boundaries through the mantle. Using the models of Higgins (1991) which incorporated a time-scale for cooling of igneous bodies in the crust, it can be inferred that, at low liquid contents, interstitial liquid would be effectively trapped rather than migrate. Hence, it is likely that at low porosity the interstitial apatite-saturated liquid would be incapable of communication with the supernatant liquid in the magma chamber. Given the huge vertical thickness of the Bushveld Complex and the extremely small residual porosity at apatite saturation (Fig. 3B), this might apply to the Bushveld Complex. In contrast, the smaller vertical distance and higher porosity in the Skaergaard intrusion at apatite saturation (Fig. 3A) may permit easier migration of interstitial liquid to the crystal–liquid interface. This distinction has major implications for the composition of the apatite.

Chemical effect of reaction with trapped liquid. Cumulus apatite ought to be F-rich, as F partitions strongly into apatite relative to Cl (Mathez, 1989). Thus, the low Cl/F ratio of apatite from the Skaergaard intrusion can be regarded as being that of the original phase unmodified by subsequent recrystallization. Where intercumulus apatite crystallized low down in a sequence with a thick pile of crystal

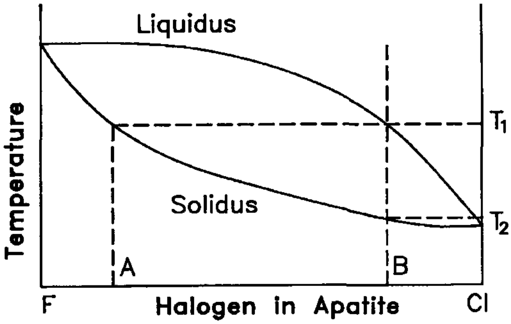

Fig. 4. Schematic phase diagram for variable proportions of Cl and F in apatite, demonstrating the range of Cl/F ratios obtainable in apatite depending upon the extent of equilibrium crystallization.

mush above, as in the Bushveld Complex, it did so from totally trapped liquid. Continued reaction with the liquid occurred. In the extreme case, the Cl/F ratio of the apatite became the same as that of the crystallizing liquid. This is illustrated in Fig. 4 which shows qualitatively that apatite crystallizing from a liquid with a Cl/F ratio given by B at temperature T_1 would have a Cl/F ratio A. However, equilibrium crystallization of this liquid will ultimately produce apatite at temperature T_2 with a Cl/F ratio of B. Isolation of apatite from the liquid at different times and temperatures during this cooling interval would produce the wide range of Cl/F ratios in apatite observed even within single samples (Boudreau and McCallum, 1989).

This process is analogous to the well-known trend of increasing Fe-enrichment in mafic minerals as they reequilibrate with residual liquid, referred to as the trapped liquid shift by Barnes (1986).

The Gabbro Zone of the Munni Munni Intrusion shows the transition from moderate Cl/F ratio in the apatite close to the horizon of mineralization to lower Cl/F ratios higher in the stratigraphy. This may represent a change from apatite crystallization from an essentially trapped intercumulus liquid which reequilibrated with apatite, to a situation where there was communication of the liquid with the supernatant magma. The exact height where cumulus apatite would have appeared in the Munni Munni Intrusion is not known, as the upper facies are not exposed. However, the gradual decrease in Cl/F ratio upwards to low values suggests that at the highest exposed level either there was a great deal of interstitial liquid or the magma was close to apatite saturation.

Conclusions

Chlor-apatite is characteristic of the lower portions of the Bushveld and Stillwater intrusions. It is not invariably associated with PGE mineralization, as the Great Dyke and Munni Munni intrusions contain PGE-mineralized horizons with Cl-poor apatite.

The model whereby the presence of fluor-apatite, is taken to indicate loss of Cl to a vapour, implies that not only are the Bushveld and Stillwater magmas water-saturated, but also Munni Munni, Great Dyke, Skaergaard and Kiglapait intrusions. In fact, in all intrusions where fluor-apatite is present, this model demands the magma reached water-saturation significantly before it reached apatite saturation. Independent evidence for this needs to be found before such a significant inference can be accepted. In fact, several lines of evidence refute this, at least for the Bushveld Complex.

The alternative model presented here demonstrates that reequilibration of apatite with trapped liquid could be adequate. In this model, the upward decrease in the Cl/F ratio of apatite, as seen for the Munni Munni intrusion, is to be expected as the system changes upwards in the intrusion from one where small volumes of liquid are effectively trapped to one where higher porosity permits liquid communication with the main body of magma.

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