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Textural character and chemistry of plagioclase and apatite in the Marathon Cu-PGE deposit, Ontario: Implications for mineralizing processes

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ABSTRACT

The Marathon deposit is hosted within the Two Duck Lake gabbro (TDLG) of the Mesoproterozoic Coldwell alkaline complex, and comprises three zones (Footwall Zone, Main Zone, and W Horizon), which have different textural, mineralogical, and geochemical characteristics. Plagioclase and apatite in the host gabbro units have complex textures and chemistry. Magmatic plagioclase is characterized by a strong positive Eu anomaly, and ΣREE increases from core to rim. Plagioclase with replacement rims occurs adjacent to granophyric patches in the vicinity of the footwall, suggesting alteration by fluids exsolved from the granophyric melts. Apatite both predated and postdated plagioclase. Apatite crystals from the Main and Footwall zones generally have high Cl contents, whereas apatite from the W Horizon has low Cl content. Rocks with higher grades of mineralization in the Main and Footwall zones generally contain apatite with higher Cl contents, whereas, in the W Horizon, higher grades correlate with lower Cl in apatite. In addition, zoning, recrystallization textures, and the presence of primary fluid inclusions are more common in apatite from the Main and Footwall zones. ΣREE in late apatite overlaps that of the core and rim of early apatite, suggesting that late apatite probably crystallized from a new influx of magma rather than from continued crystallization of a resident magma. The spatial variations in apatite Cl/F could be explained if compositionally distinct magma pulses were responsible for each mineralized zone, as most early apatite crystals are euhedral to subhedral, and magmatic. Intracontinental basaltic melts are, however, reported to be poor in Cl, suggesting local addition of Cl. Chalcopyrite in the Main Zone commonly replaces pyrrhotite and is intergrown with hydrous silicate minerals, suggesting Cu (re-)mobilization. Therefore, a zone-refining process in which volatiles, derived from footwall country-rock dehydration, migrated through the crystallizing gabbros and transported Cu to the Main Zone is an attractive model by which Cl could also be added to the system. The low Cl contents of apatite in the W Horizon can be explained if these fluids did not reach the W Horizon, or the W Horizon represents late-stage magma infiltration.

INTRODUCTION

Despite extensive study, aspects of the genesis of contact-style, low sulphide PGE-Cu (-Ni) deposits (e.g. Naldrett, 2004; Mungall, 2005; Zientek, 2012) remains a controversial subject, in particular the relative importance of magmatic and fluid (volatile)-mediated processes in Cu and PGE enrichment remains a point of debate. Many recent studies have discussed this issue, and have suggested that these deposits are magmatic ore deposits modified by syn- or post-magmatic fluid activity (Harris and Chaumba, 2001; Sharman-Harris et al., 2005; Holwell et al., 2007; Gál et al., 2011; Holwell et al., 2014).

The Marathon deposit, Ontario, Canada, is classified as a contact-type PGE-Cu deposit. It is hosted by the Two Duck Lake gabbro (TDLG) of the Mesoproterozoic (1108 ± 1 Ma) Coldwell alkaline complex. The process of platinum-group element (PGE) enrichment in the Marathon deposit is also still controversial: can the concentration mechanisms be dominantly attributed to PGE-rich immiscible sulphide liquids that separated from the host gabbroic magmas (Good and Crocket, 1994; Good, 2010; Ruthart et al., 2010) or were the volatile-rich fluids passing through the crystalline or nearly crystalline rocks also important (Watkinson and Ohnenstetter, 1992; Dahl et al.,

2001; Barrie et al., 2002). The Marathon deposit comprises three zones of mineralization with different textural, mineralogical, and geochemical characteristics: Footwall Zone, Main Zone, and W Horizon.

In order to improve our understanding of the magmatic and fluid-rock interaction processes that led to the formation of the Marathon deposit, including the thermal history of the rocks, and hence further develop models for contact-type low-sulphide deposits, we have studied the chemical and textural characteristics and evolution of plagioclase and apatite at Marathon, in the context of sulphide distribution in the various mineralized zones.

Major and trace element mineral chemistry, combined with petrography, have been used by a number of authors to elucidate magma-chamber processes, such as magma sources, the nature and degree of fractional crystallization, magma recharge, and fluid-rock interaction (Hall, 1987; Kruger, 1992; Best, 2003; Streck, 2008). Plagioclase chemistry has been long recognized as a good recorder of crystallization history and the chemical evolution of magmas due to the easily recognized compositional zoning (Ginibre et al., 2002; Humphreys et al., 2006; Ginibre et al., 2007; Humphreys, 2009; Gorokhova et al., 2013). Also, the distribution and composition of halogen-bearing minerals can be used to assess the possible role of volatile components during crystallization, and the potential for Cl-bearing fluids to act as metal transport agents (Boudreau and McCallum, 1989; Boudreau et al., 1986; Boudreau and Kruger, 1990; Willmore et al., 2002; Boudreau and Hoatson, 2004). The effects of the migration of Cl-rich fluids in the mobilization of base and precious metal sulphides in the Bushveld and Stillwater complexes have been extensively reported (Boudreau et al., 1986; Boudreau and McCallum, 1989; Boudreau and Kruger, 1990; Willmore et al., 2000). Therefore, the significance of hydrothermal fluids in the mineralizing mechanisms, either directly forming mineralization or causing considerable modification of it, cannot be ignored. In this study, petrographic analyses have been combined with detailed mineral chemical analyses to evaluate the petrogenetic evolution of the Marathon host rocks in the context of magmatic and fluid-rock interaction history as a framework for understanding the relationship of this evolution to the formation of the ore minerals.

RESULTS

Petrography

The TDLG is coarse-grained to pegmatitic, and comprises plagioclase with variable amounts of interstitial ophitic clinopyroxene, olivine, and magnetite. Apatite, biotite, and amphibole are common accessory minerals. Hydrothermal alteration is a common feature, especially in the Main and Footwall zones, and is dominated by replacement of primary igneous minerals by amphibole, chlorite, biotite, and serpentine. Epidote and carbonate are also present as alteration minerals in some samples.

Plagioclase is a ubiquitous silicate phase in the TDLG and occurs as cumulus euhedral to subhedral tabular prisms. The grain size varies typically from a few millimetres to 1 centimetre, but can be considerably coarser. Plagioclase crystals contain a wide variety of internal textures, including resorption surfaces, normal or reverse zoning, oscillatory zoning, and patchy zoning. Plagioclase crystals can be classified into four types based on the nature of internal zonation: (1) homogeneous crystals lacking zonation, (2) crystals showing normal or oscillatory zoning (Fig. 1a, b), (3) plagioclase crystals with resorption surfaces (Fig. 1c, d, e), and (4) crystals with replacement rims (Fig. 1f). Resorption textures are relatively common in plagioclase, and can contain one or two resorption surfaces (i.e. two or three stages of plagioclase growth; Fig. 1c, d).In some cases, in rocks that contain sulphides, the outermost resorption zone is characterized by a porous texture in which the pores are filled with sulphide (Fig. 1e). Finally, plagioclase crystals with replacement rims occur in the vicinity of quartz-feldspar granophyre patches (Fig. 1f).

Apatite is an accessory mineral and mostly occurs as interstitial euhedral to subhedral acicular and tabular prisms, and as anhedral crystals hosted by both altered and unaltered minerals. Although apatite is commonly hosted by plagioclase, it is also hosted by pyroxene, sulphide, magnetite, and hydrous minerals (Fig. 2). In different rocks, apatite either predated (pre-plagioclase apatite; Fig. 2a, b) or postdated plagioclase crystallization (post-plagioclase apatite; Fig. 2g, h). Apatite grains vary in size but are usually not more than a few millimetres. However, unusual oikocryst apatite, up to a few centimetres across, occurs in pegmatitic gabbro. The latter are more common in the W Horizon, and mostly postdate plagioclase crystallization. SEM-CL imaging shows that apatite crystals can exhibit growth (oscillatory) and replacement zoning. Zoning, re-crystallization textures (Fig. 3), and the presence of primary fluid inclusions, are more common in apatite crystals from Main and Footwall zone samples compared to those from the W Horizon.

Sulphide minerals occur as disseminations throughout the gabbro or as massive to semi-massive accumulations of chalcopyrite, pyrrhotite, bornite, minor pentlandite, and accessory pyrite and cubanite. The Footwall Zone contains massive to semi-massive pyrrhotite with minor chalcopyrite, pentlandite, and pyrite. In the Main Zone, interstitial disseminated sulphides comprise chalcopyrite, pyrrhotite ± pentlandite, and cubanite. In some
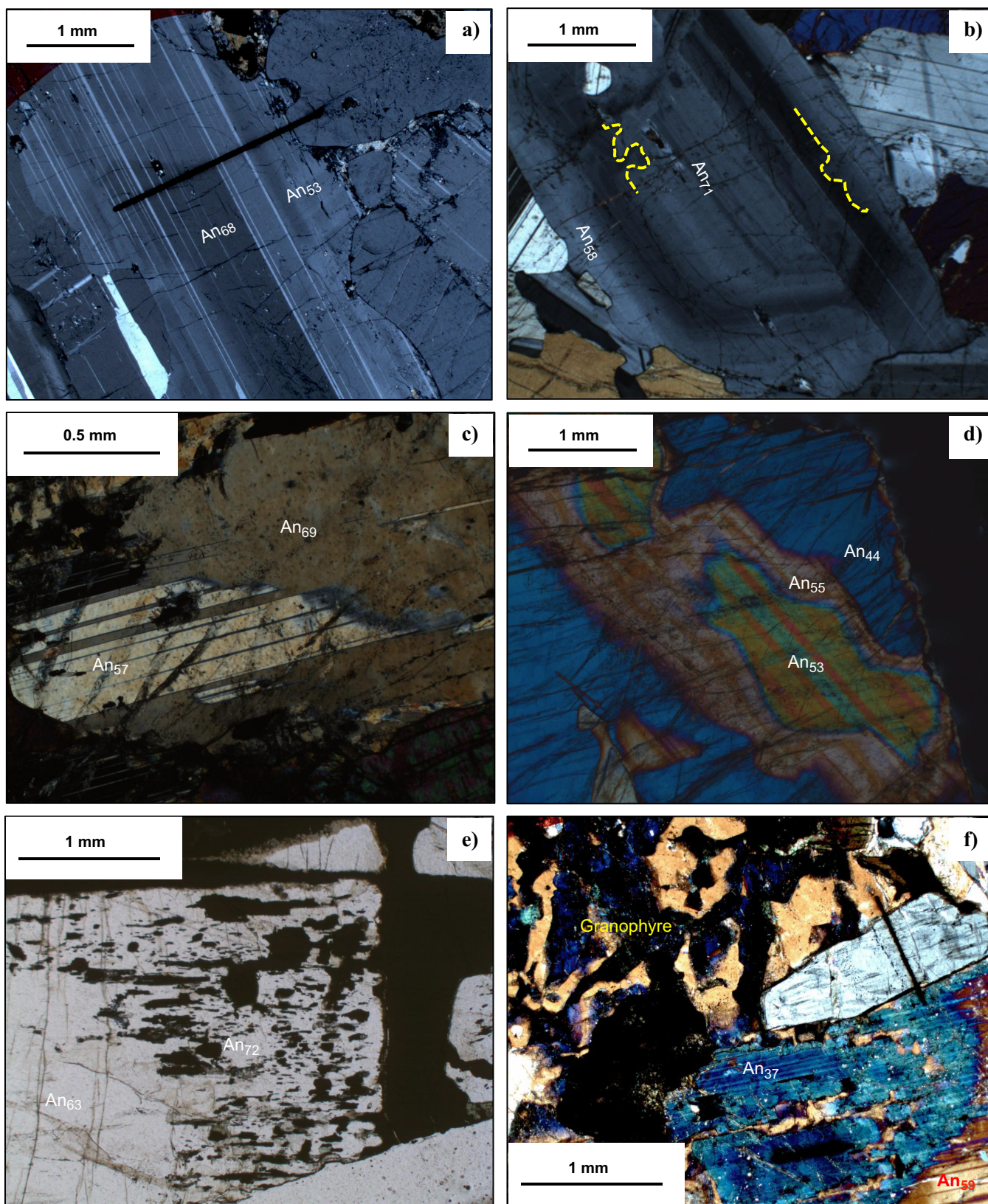
Figure 1. Photomicrographs representing the variation in textural characteristics of plagioclase: **(a)** normal zoning; **(b)** oscillatory zoning (highlighted by a yellow dotted line); **(c)** early crystallized plagioclase resorbed and overgrown by more calcic plagioclase; **(d)** two stages of resorption: early crystallized plagioclase was first resorbed and overgrown by more calcic plagioclase and then a second stage is represented by resorption followed by sodic overgrowth; **(e)** resorption texture with sulphide blebs in the outermost zone (overgrowth); **(f)** replacement rim of more sodic plagioclase replacing a more calcic crystal.
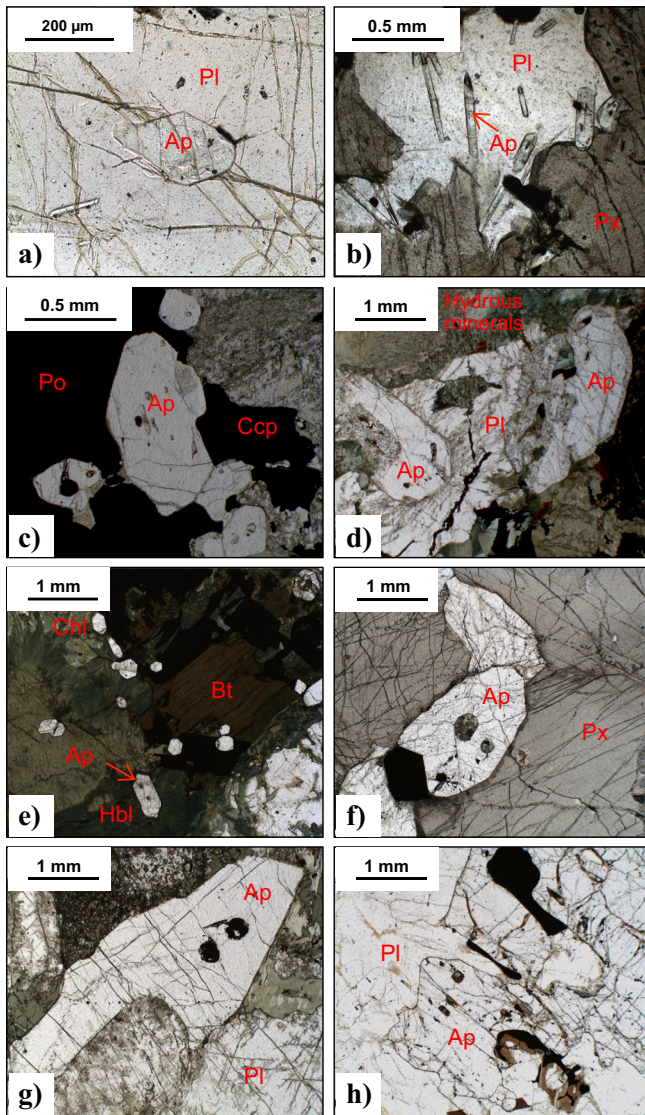
Figure 2. Photomicrographs representing the variation in textural characteristics and host assemblages of apatite from the three mineralized zones: **(a and b)** Main Zone; **(c and d)** Footwall Zone; **(e and f)** W Horizon; and **(g and h)** apatite that postdates plagioclase crystallization. Ap = apatite; Bt = biotite; Ccp = chalcopyrite; Chl = chlorite; Hbl = hornblende; Pl = plagioclase; Po = pyrrhotite; Px = pyroxene.

aggregates, chalcopyrite shares equilibrium boundaries with pyrrhotite. Much of the chalcopyrite in the Main Zone, however, replaced pyrrhotite and is intergrown with hydrous silicate minerals. In both the Footwall and Main zones, sulphides occur as replacing pyroxene lamellae in altered samples. Sulphides in the W Horizon (the highest stratigraphic zone) are characterized by disseminated chalcopyrite, bornite ± pyrrhotite, pentlandite, cubanite, and pyrite. Bornite occurs as irregular patches in chalcopyrite but is mostly represented by flame-like exsolution within chalcopyrite.

Mineral Chemistry
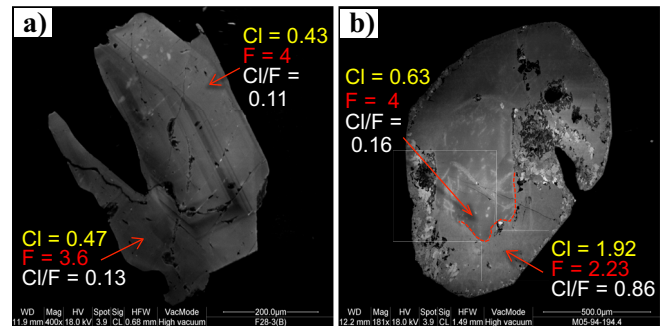
The anorthite content of plagioclase in the TDLG

Figure 3. SEM-cathodoluminescence images of apatite crystals with EDS analyses of different zones: **(a)** growth zoning in a crystal from the Main Zone with no variation in Cl/F ratios between zones; **(b)** outer replacement zone with a higher Cl/F ratio than in the relict core (from the Footwall Zone); boundary marked by the dotted line.
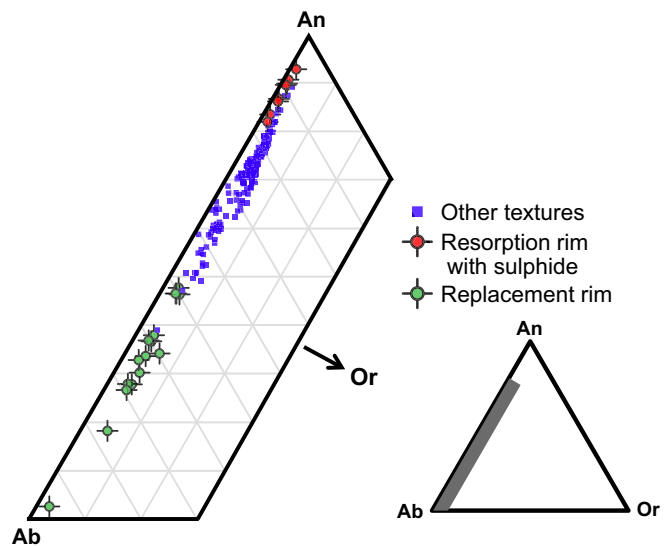

Figure 4. Part of a ternary diagram (grey field in the An-Ab-Or ternary diagram) showing the variations in An content of plagioclase with different textures.

varies widely, from An_{15} to An_{74}. The anorthite content of the plagioclase varies by 4 mol% between different oscillatory zones; however, the general trend is a decrease in An content toward the rim. The later overgrowth phases, after resorption, generally have higher An contents (An_{52-74}) than the earlier stages of crystallization, whereas replacement rims in the vicinity of the granophyric patches have lower An contents (An_{33-50}) compared to the magmatic plagioclase that was replaced (An_{60-75}) (Fig. 4).

Trace element analyses of plagioclase indicate that only the rare earth elements, S, Ni, Cu, Ti, and Pb exhibit significant differences among the three zones, and that other analyzed elements show either no variation or the variation is less than the precision of the analytical method. Magmatic plagioclase with normal or oscillatory zoning is characterized by a distinct positive Eu anomaly, and mostly shows an increase in ΣREE towards the rims of crystals. The change in the
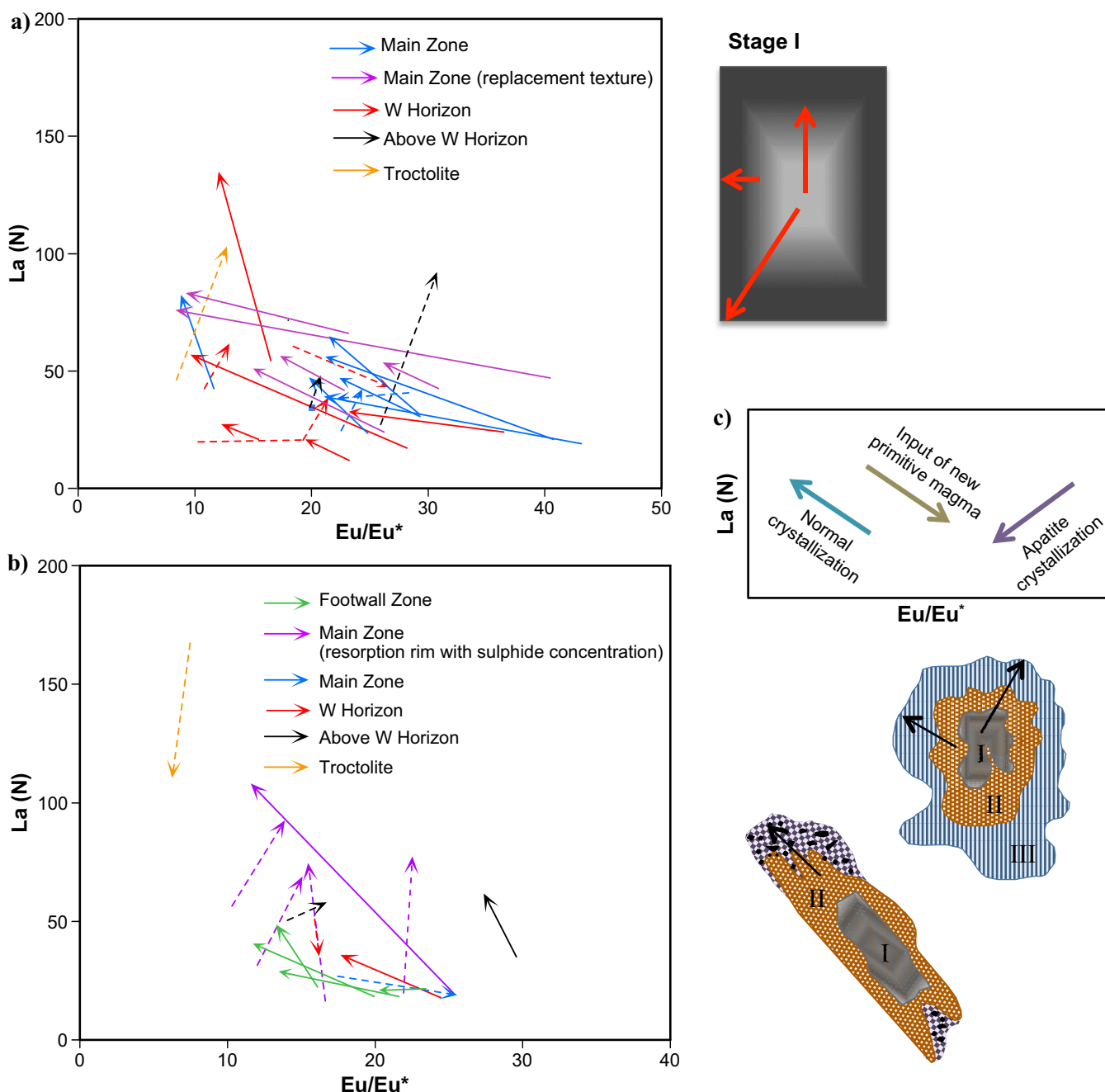
Figure 5. Trace element variation in plagioclase for different textures showing trends from early to late crystallization: **(a)** plagioclase crystallization showing normal or oscillatory zoning without any resorption; **(b)** plagioclase crystals showing overgrowth after resorption; **(c)** sketch showing expected trends of REE for early to late phases of plagioclase based on different scenarios.

magnitude of the Eu anomaly from core to rim of oscillatory zoned plagioclase crystals, however, is different in different crystals. For example, in most plagioclase crystals from the Main and Footwall zones, the magnitude of this anomaly decreases from the earlier to the later crystallized phase, whereas in some crystals from the W Horizon; in gabbro above W Horizon the opposite is observed (Fig. 5a). These different trends in REE character can also be found in plagioclase crystals with overgrowths that postdate resorption, where the Eu anomaly can increase or decrease from an early crystal that has been resorbed to one of the overgrowth phases or from an early overgrowth to a later one (Fig. 5b). The resorption rims characterized by sulphide-filled pores have higher An content than the crystal cores and have higher S, Ni, Cu, and Pb and lower Ti than all other types of plagioclase.

The energy-dispersive X-ray spectrometry (ed.) and wavelength-dispersive X-ray spectrometry (WDS) analyses indicate that the F and Cl contents in apatite
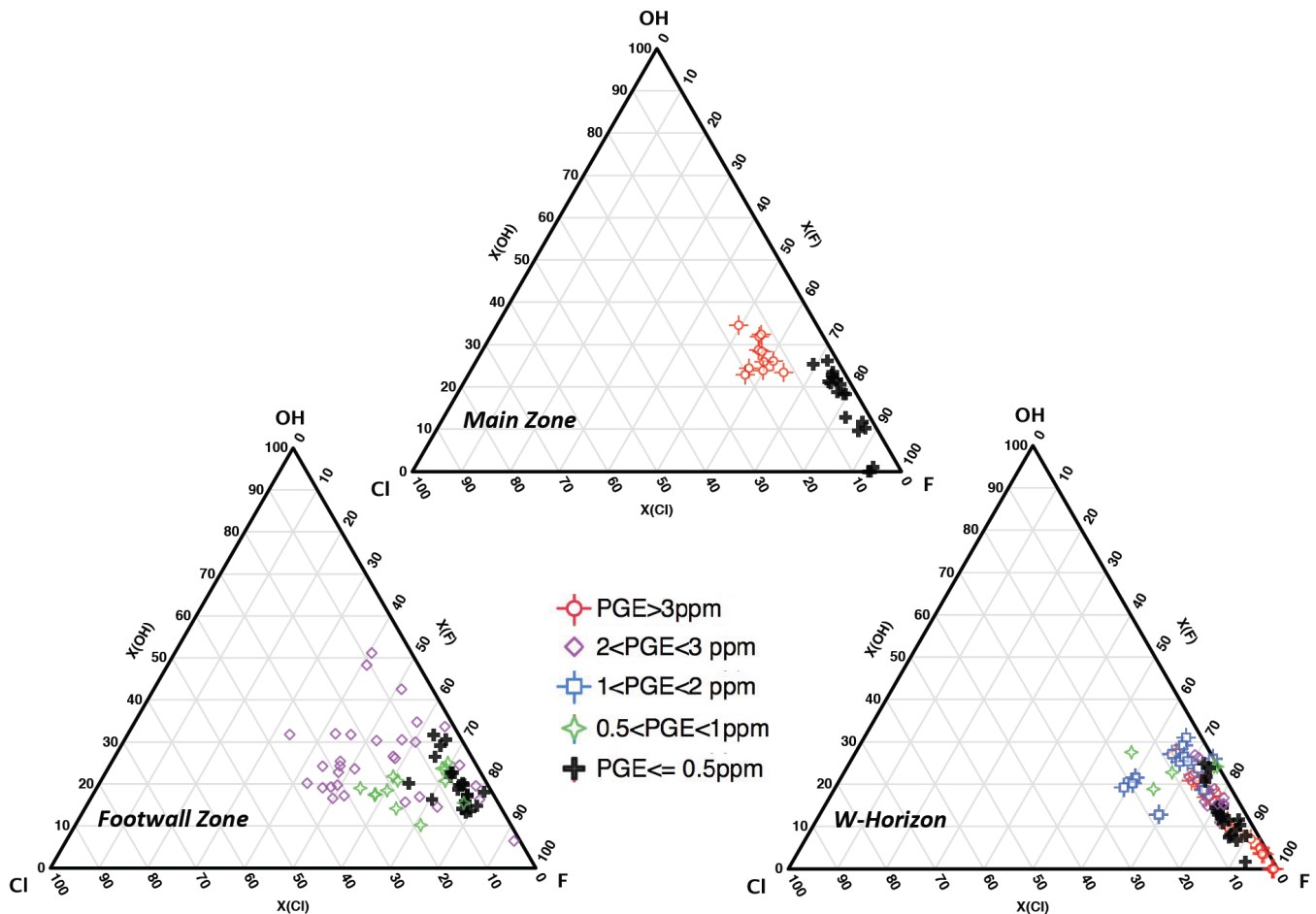
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Figure 6. Ternary diagrams showing F-Cl variations in apatite from the different mineralized zones and their relationship to the grade of mineralization.

are variable (Fig. 6). There is a systematic variation of halogen contents in apatite with stratigraphic level and grade of mineralization. Apatite crystals from mineralized Main and Footwall zone samples generally have high Cl contents, whereas apatite from the W Horizon has low Cl contents, and generally have fluorapatite end-member compositions. Apatite crystals from the Footwall Zone have the highest Cl/F values, whereas those from the W Horizon show the lowest Cl/F ratios. Rocks with higher grades of mineralization in the Main and Footwall zones generally contain apatite with higher Cl contents, whereas, in the W Horizon, higher grades correlate with lower Cl in apatite.

Cores of apatite crystals have lower ΣREE contents and a negative Eu anomaly, whereas the rims have higher REE contents and a stronger Eu anomaly. Both cores and rims of apatite crystals exhibit similar positive Ce anomalies. The range of REE compositions for post-plagioclase apatite spans the entire range of compositions for cores and rims of pre-plagioclase apatite. Replacement rims of apatite crystals, however, show lower ΣREE, and higher Cl contents compared to early apatite, and no significant Ce anomaly.

DISCUSSION

Although some crystals of plagioclase in the TDLG show normal zoning, it is common to find evidence of disequilibrium and resorption, indicating repeated input of new magma into the TDLG system, which was out of equilibrium with pre-existing crystals (Fig. 1c, d, e). Magma dynamics, including magma mingling, as recorded by plagioclase crystals, has been discussed in numerous studies (Browne et al., 2006; Ginibre et al., 2007; Gál et al., 2013; Thy et al., 2013). The expected trend from normal crystallization would be an increase in La and a decrease in the magnitude of the Eu anomaly as crystals grow (Fig. 5c). At Marathon, plagioclase crystals that exhibit normal or oscillatory zoning without any evidence of resorption show different chemical trends from core to rim (Fig. 5a). Most of the plagioclase crystals from the Main Zone and Footwall Zone follow this trend, however, some crystals from within and above the W Horizon and from the troctolite represent different trends: the magnitude of the Eu anomaly increases from early to late crystallization. This trend can be explained by changing magma chemistry (through influx of new magma) or crystallization of

other minerals, such as apatite, which could have affected REE concentrations in the magma. However, crystallization of apatite would cause REE depletion in the magma and later-crystallized plagioclase would be expected to show lower La, which is in contrast with what was observed. Therefore, it is more likely that these trends represent changing magma chemistry through the input of new magma. If the new magma was relatively primitive (the same as the original magma), we would expect the later phase to contain lower La because the REE partition coefficients in plagioclase decrease with increasing An content. The new magmas, therefore, probably came from a chamber undergoing fractional crystallization at depth. This model is supported by the observation that many crystals show evidence of resorption and that late-stage, post-resorption overgrowths have a higher An content as well as different REE trends.

The higher An and base metal contents of plagioclase crystals that contain sulphide blebs in post-resorption overgrowths suggests that these resorption rims are related to the mineralization, and crystallized from a sulphide-bearing magma or fluids. This could be attributed to the influx of metal-rich magma, or the interaction of the magma with metal-rich fluid. Both models could explain the higher An content of these resorptions rims than in the earlier stages of crystallization. One possible source of metal-rich fluids is the late-stage granophyric melts that were present in the system. Plagioclase replacement rims that occur in the vicinity of quartz-feldspar granophyric patches (Fig. 1f), however, have low An contents (An$_{33-50}$; Fig. 4c), consistent with alteration by fluids exsolved from the granophyric melts, which are therefore unlikely to have formed the sulphide-associated Ca-rich overgrowths (Fig. 4c).

The observed spatial variations in Cl/F in apatite can be explained by compositionally distinct magma pulses being responsible for the formation of the different mineralized zones, as most apatite crystals are euhedral to subhedral, formed early in the paragenetic sequence, and are magmatic. Boudreau et al. (1997) showed that magmas formed by simple extensional decompression melting are relatively volatile-poor and apatite that crystallizes from such magmas would be fluorine-rich. Given that the Marathon deposit is related to magmas formed in an intracontinental rift, we can therefore expect that the TDL magmas were relatively dry, and that apatite crystallizing from such a melt would be fluorapatite. This is supported by the lower Cl/F ratio of apatite from the TDLG compared to those from the Stillwater and Bushveld complexes (Boudreau et al., 1997).

Much of the chalcopyrite in the Main Zone has replaced pyrrhotite and is intergrown with hydrous silicate minerals, which suggests that Cu was introduced into the system by fluids. This observation can be explained by a process in which fluids flux through the Footwall Zone, transporting Cu to the Main Zone during its upward migration. Therefore, a zone-refining process, in which volatiles that were derived from footwall country-rock dehydration migrated up through the crystallizing gabbro, is an attractive alternative by which Cl could be added to the system. Zoning and recrystallization textures of Cl-rich apatite also indicate that Cl was not incorporated at the time of apatite crystallization, but rather Cl addition occurred at some point after the crystallization of apatite started. The low Cl contents of apatite in the W horizon can be explained if these fluids did not migrate up as far as the W Horizon. Alternatively, the W Horizon represents a late stage of magma intrusion, after the Cl-rich fluids had fluxed through the lower portions of the TDLG. Thus, the alteration of the primary igneous minerals, the re-crystallization textures, the high Cl/F exhibited by apatite within the Footwall and Main zones, and higher Cl and transition metal contents of the apatite replacement rims can be attributed to the interaction of a fluid phase at suprasolidus and/or subsolidus conditions.

IMPLICATIONS FOR EXPLORATION

A multistage-dissolution upgrading model has been widely accepted for the genesis of magmatic PGE-Cu deposits, and has been proposed for the formation of the Marathon deposit (Good, 2010). In this model, new, S-under-saturated magma pulses through magma conduits, resulting in dissolution of pre-existing sulphide but not PGE (Kerr and Leitch, 2005), and produces very high PGE/Cu and PGE/S ratios. The textures and chemistry of plagioclase and apatite indicate multiple influxes of magma into the Marathon gabbros, which would support this model. Therefore, the textural and chemical features described in this study can be used to indicate that these processes have occurred and thus could be used as an indicator of fertility of gabbro in exploration.

ACKNOWLEDGEMENTS

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