



Experimental investigation of Fe³⁺-rich majoritic garnet and its effect on majorite geobarometer

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Abstract

Majoritic garnet [(Ca, Mg, Fe²⁺)₃(Fe³⁺, Al, Si)₂(SiO₄)₃] is one of the predominant and important constituents of upper mantle peridotite and ultra-deep subducted slabs. Majoritic substitution in garnet depends on pressure, and it has been used to estimate the formation pressure of natural majoritic garnet. Ferric iron (Fe³⁺) substitution occurs in natural majoritic garnets from mantle diamonds and shocked meteorites. However, available majorite geobarometers were developed without considering the effect of Fe³⁺ substitution in the structure. In this study, we systematically synthesized Fe³⁺-bearing majoritic garnets from 6.5 GPa to 15 GPa to evaluate the effect of Fe³⁺ on the majorite geobarometer. The Fe³⁺ contents of synthetic majoritic garnets were analyzed using the “Flank method” with the electron probe microanalyzer (EPMA). The results were compared with those based on the charge balance calculations. From the known synthesis pressures and measured Fe³⁺ contents, we developed a new majorite geobarometer for Fe³⁺-bearing majoritic garnets. Our results show that the existing majorite geobarometer, which does not take into account the Fe³⁺ substitution, could underestimate the formation pressure of majoritic garnets, especially for samples with a high majoritic component.

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1. INTRODUCTION

Aluminous garnet, (Ca, Mg, Fe²⁺, Mn)₃Al₂Si₃O₁₂, is an important constituent of mantle peridotite (~10 vol.%), and of lower crust and upper mantle eclogite (~50 vol.%) (Wood et al., 2013). The composition of aluminous garnets can generally be expressed in terms of four main end-member components: pyrope (Mg₃Al₂Si₃O₁₂), grossular (Ca₃Al₂Si₃O₁₂), almandine (Fe²⁺₃Al₂Si₃O₁₂), and spessartine (Mn₃Al₂Si₃O₁₂). Because the compositional variation of the aluminous garnet is sensitive to equilibrium temperature and pressure, geobarometers involving aluminous garnet-bearing equilibrium are often developed to esti-

mate temperature and pressure of metamorphic and igneous rocks (e.g. O'Neill and Wood, 1979; Bohlen et al., 1983). Iron has multiple valence states (e.g. Fe²⁺ and Fe³⁺). Different Fe³⁺/Fe²⁺ ratios may have a significant effect on the physical and chemical properties of iron-bearing minerals such as garnet (McCammon and Ross, 2003). Garnet could be the alternative host for Fe³⁺ as Fe³⁺-bearing spinel becomes unstable in the shallow mantle (>~50 km) (Robinson and Wood, 1998). The 6-fold-coordinated sites in garnet can incorporate significant amounts of Fe³⁺ (Luth et al., 1990; Woodland and O'Neill, 1993) and the Fe³⁺ contents can be used as an oxygen barometer for metamorphic rocks from subduction zones and garnet peridotites from the mantle (Gudmundsson and Wood, 1995; Canil and O'Neill 1996; Simakov, 2006; Stagno et al., 2013; Stagno et al., 2015). The geothermometers and geobarometers reveal a trend

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of an increasing skiagitic ($\text{Fe}_2\text{Fe}_2^{3+}\text{Si}_3\text{O}_{12}$) component in garnet with increasing temperature and pressure of equilibration, which is not coupled to a whole-rock increase (Canil and O'Neill, 1996; Woodland and Koch, 2003; Frost and McCammon, 2008). Meanwhile, the substitution of Fe^{3+} for Al is facilitated by the addition of Ca to the garnet, an example of the “reciprocal solution” effect (Wood and Nicholls, 1978).

In garnets formed in the Earth's lower upper mantle between 50–150 km, Si is exclusively in tetrahedral coordination. With increasing pressure (depth > 150 km) Si also partitions increasingly into the octahedral site, because of incorporation of pyroxene at high pressure (Ringwood and Major, 1971; Irifune and Ringwood, 1993). This implies that the Si content of garnet will increase at the expense of Al in the octahedral site and form majoritic garnet (i.e. Si will be hosted in both the tetrahedral and octahedral sites in garnet). Two key majoritic substitution mechanisms have been proposed to classify natural majoritic garnets (Schulz, 2003; Kiseeva et al., 2013); peridotitic substitution ($\text{Si}^{4+} + \text{M}^{2+} = 2\text{Al}^{3+}$) and eclogitic substitution ($\text{Na}^+ + \text{Si}^{4+} = \text{M}^{2+} + \text{Al}^{3+}$), where M^{2+} represents divalent Ca, Mg, and Fe^{2+} . In this study, we define the chemical compositions of garnet with Si (p.f.u.) > 3.00 as be majoritic garnet hereafter.

The high-pressure majoritic garnet is also expected to incorporate Fe^{3+} in the octahedral site at mantle conditions. Fe^{3+} -rich majoritic garnets ($\text{Fe}^{3+}/\Sigma\text{Fe}$ ratio ranging from 0.03 to 0.95) have been synthesized at high pressure and high temperature using the multi-anvil press, under different oxygen fugacity conditions, both with and without water (McCammon and Ross, 2003; McCammon et al., 2004). Mössbauer spectroscopic measurements indicated that the Fe^{3+} contents in the synthetic majoritic garnet increase both with the total Fe content and increasing oxygen fugacity (McCammon et al., 2004). The $\text{Fe}^{3+}/\Sigma\text{Fe}$ ratio in the (Mg, Fe)(Si, Al) O_3 majorite is not significantly different from that in the Al-free majorite under the same redox conditions (McCammon and Ross, 2003).

Because majoritic garnet is an ultrahigh-pressure phase, its recovery from mantle-derived rocks is very rare. However, majoritic garnet inclusions are widely reported in mantle diamonds from different locations, including South Africa (Moore and Gurney, 1985; Haggerty and Sautter, 1990), Guinea (Hutchison, 1997; Stachel et al., 2001), Brazil (Hutchison, 1997), Russia (Sobolev et al., 2004), and China (Wang et al., 2000), and are classified as either peridotitic or eclogitic suites. However, most majoritic garnets are inferred to have a crustal origin according to the trace element and isotopic features (Stachel, 2001; Harte, 2010; Kiseeva et al., 2013; Ickert et al., 2015). The occurrence of majoritic garnet in shocked meteorites has also been documented. In fact, the first majorite was discovered in shock veins in the Coorara L6 chondritic meteorite (Smith and Mason, 1970). More majoritic garnets were subsequently reported in different meteorites, including further L6 chondrites (Mao et al., 1982; Chen et al., 2004; Ohtani et al., 2004; Tschauner et al., 2014) and H chondrites (Semenenko and Golovko, 1994; Tomioka and Kimura, 2003).

Collerson et al. (2010) summarized the chemical compositions of majoritic garnets from mantle diamonds and shocked meteorites, and calculated the formula of majoritic garnet assuming all Fe is present as Fe^{2+} on the basis of 12 oxygen atoms. In this study, we examined the chemical compositions of the reported natural majoritic garnets and re-calculated the formulas of majoritic garnets according to the charge balance method (Table S1). The charge balance calculations show that majoritic garnets from mantle diamonds and shocked meteorites could contain significant Fe^{3+} contents ($\text{Fe}^{3+}/\Sigma\text{Fe}$ up to 0.94 in Table S1). Notably, we recently reported Fe^{3+} -rich majoritic garnets ($\text{Fe}^{3+}/\Sigma\text{Fe} > \sim 0.81$) from the deep upper mantle (Xu et al., 2017). These observations motivated us to investigate the role of Fe^{3+} in the majoritic garnet structure, especially the effect of the Fe^{3+} content on the majorite geobarometer.

Majoritic substitution in garnet has been used to estimate the formation pressure of natural majoritic garnets. Because the Si^{4+} excesses and ($\text{Al}^{3+} + \text{Cr}^{3+}$) deficits in the majoritic garnet are a function of pressure based on high-pressure experiments (e.g., Irifune, 1987), pressure estimates for the natural samples are often obtained from this simple relation (e.g., Stachel, 2001; Banas et al., 2007; Scambelluri et al., 2008). Collerson et al. (2010) refined pressure determinations based on majoritic garnet compositions synthesized from a variety of protolith compositions including peridotite-pyrolite, kimberlite, komatiite, garnet pyroxenite, and MORB, over a wide pressure and temperature range. For samples with a high Na_2O content, Bobrov et al. (2008) proposed a Na content-dependent majoritic geobarometer based on experimentally determined phase relations on the diopside-jadeite-hedenbergite join up to 24 GPa. Wijbrans et al. (2016) experimentally investigated the stability of Cr-bearing majoritic garnet from 6 to 14.5 GPa and derived a new majorite geobarometer for peridotitic and eclogitic compositions by taking into account of the effect of Cr on the majorite substitution. Recently, Beyer and Frost (2017) formulated a new majorite barometer based on an empirical parameterization of the two major majoritic substitutions using their new experimental data between 6 and 16 GPa combined with previous published experimental data in a wide pressure range. They did consider the Fe^{3+} substitution in garnets in their geobarometer, but the limited range in the Fe^{3+} content ($\text{Fe}^{3+}/\Sigma\text{Fe} < 0.2$) in their synthetic majoritic garnets hindered the discussion on the effect of Fe^{3+} content on majorite geobarometer. The goal of this study is to perform systematic experiments over a large Fe^{3+} content range to investigate the effect of Fe^{3+} content on the majorite geobarometer.

Ferric iron in the majoritic garnet is often ignored because of technical limitations in determining the Fe^{3+} content in the natural samples. At present, the technique most widely used for $\text{Fe}^{3+}/\Sigma\text{Fe}$ determination is Mössbauer spectroscopy. The technique usually requires large sample volumes, especially for samples with a natural abundance of ^{57}Fe . The development of the “milliprobe” has improved the capability to probe micrometer-size area (McCammon et al., 1991, 2001). Nonetheless, a spatial resolution of >50 μm is still too large for Fe^{3+} measurements

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