



## Magnesium and oxygen isotopes in Roberts Victor eclogites



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### ABSTRACT

Magnesium and oxygen are critical elements in the solid Earth and hydrosphere. A better understanding of the combined behavior of Mg and O isotopes will refine their use as a tracer of geochemical processes and Earth evolution. In this study, the Mg-isotope compositions of garnet and omphacite separated from well-characterized xenolithic eclogites from the Roberts Victor kimberlite pipe (South Africa) have been measured by solution multi-collector ICP-MS. The reconstructed whole-rock  $\delta^{26}\text{Mg}$  values of Type I (metasomatized) eclogites range from  $-0.61\%$  to  $-0.20\%$  (Type IA) and from  $-0.60\%$  to  $-0.30\%$  (Type IB) (mean  $-0.43\% \pm 0.12\%$ ), while  $\delta^{26}\text{Mg}$  of Type IIA (fresh, least metasomatized) eclogites ranges from  $-1.09\%$  to  $-0.17\%$  (mean  $-0.69\% \pm 0.41\%$ ); a Type IIB (fresh, least metasomatized) has  $\delta^{26}\text{Mg}$  of  $-0.37\%$ . Oxygen-isotope compositions of garnet were analyzed in situ by SIMS (CAMECA 1280) and cross-checked by laser fluorination. Garnets have  $\delta^{18}\text{O}$  of  $6.53\%$  to  $9.08\%$  in Type IA,  $6.14\%$  to  $6.65\%$  in Type IB, and  $2.34\%$  to  $2.91\%$  in Type IIB. The variation of  $\delta^{26}\text{Mg}$  and  $\delta^{18}\text{O}$  in Type IA and IB eclogites is consistent with the previously proposed model for the evolution of these samples, based on major and trace elements and radiogenic isotopes. In this model, the protoliths (Type II eclogites) were metasomatized by carbonatitic to kimberlitic melts/fluids to produce first Type IA eclogites and then Type IB. Metasomatism has changed the O-isotope compositions, but the Mg-isotope compositions of Type IA are mainly controlled by the protoliths; those of Type IB eclogites reflect mixing between the protoliths and the kimberlitic melt/fluid. The combination of a large range of  $\delta^{26}\text{Mg}$  and low  $\delta^{18}\text{O}$  in Type II eclogites cannot be explained easily by seawater alteration of oceanic crust, interaction of carbonate/silicate sediments with oceanic crust, or partial melting of mafic rocks.

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### 1. Introduction

Magnesium is a major element in the solid Earth and an important element in the hydrosphere. It has three stable isotopes ( $^{24}\text{Mg}$ ,  $^{25}\text{Mg}$ , and  $^{26}\text{Mg}$ ) with large relative mass differences. Magnesium may therefore experience detectable isotopic fractionation between different phases under different geological conditions, providing information that can be further used to define geological environments and processes. The Mg-isotope results are normally reported in the  $\delta$  notation, where  $\delta^X\text{Mg} = [({}^X\text{Mg}/{}^{24}\text{Mg})_{\text{sample}} / ({}^X\text{Mg}/{}^{24}\text{Mg})_{\text{DSM3}} - 1] \times 1000$ ; X is 26 or 25. The widely used standard DSM3 was prepared using pure Mg provided by Dead Sea Magnesium Ltd., dissolved in  $\text{HNO}_3$  (Galy et al., 2003). The Mg-isotope compositions of major reservoirs have been investigated, and mean  $\delta^{26}\text{Mg}$  values have been defined for the mantle

( $-0.25\% \pm 0.10\%$ , 2SD,  $n = 325$ ), silicate crust ( $-0.23\% \pm 0.20\%$ , 2SD,  $n = 50$ ), carbonates ( $-5.57\%$  to  $0.75\%$ , average  $-2.14\% \pm 2.64\%$ , 2SD,  $n = 398$ ), river water ( $-1.05\% \pm 0.66\%$ , 2SD,  $n = 149$ ), and sea water ( $-0.83\% \pm 0.09\%$ , 2SD,  $n = 72$ ) (e.g., Tipper et al., 2006; Foster et al., 2010; Liu et al., 2010; Teng et al., 2010a; Ling et al., 2011; An and Huang, 2014; Geske et al., 2015; and references therein).

Available theoretical and experimental work indicates that temperature (T) is generally the single most important parameter controlling equilibrium isotopic fractionation between phases (Eiler, 2001). Magnesium isotopes show large fractionations in low-T processes, such as surface chemical weathering, carbonate deposition, and clay mineral formation (e.g., Teng et al., 2010b; Wombacher et al., 2011; Huang et al., 2012c; Kasemann et al., 2014; Liu et al., 2014; Wimpenny et al., 2014a). Analyses of most mantle peridotites and oceanic basalts show little variation in  $\delta^{26}\text{Mg}$  ( $-0.25\% \pm 0.10\%$ ), within the uncertainty of current analytical methods, during mantle partial melting, mantle metasomatism, and magma-differentiation processes (Teng et al., 2007; Handler et al., 2009; Bourdon et al., 2010; Liu et al., 2010; Teng et al., 2010a). However, even at high T, some peridotites that have

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experienced mantle metasomatism have Mg-isotope compositions distinguishable from the nominal mantle value (Pearson et al., 2006; Huang et al., 2011; Xiao et al., 2013), indicating that the mantle is not completely homogeneous in terms of Mg isotopes and that it contains components with “non-mantle”  $\delta^{26}\text{Mg}$  values.

Eclogites, a minor (<1 vol.%) but important component of the lithospheric mantle (Schulze, 1989), have mafic rather than ultramafic compositions, in contrast to peridotites, the dominant component of the mantle (e.g., Jacob, 2004; Griffin and O'Reilly, 2007; Griffin et al., 2009; Gréau et al., 2011; Huang et al., 2012a). They have chemical compositions broadly similar to oceanic basalts collected at Earth's surface but have equilibrated at high T and pressure (P) (~1050 °C, 5–6 GPa) (e.g., Jacob, 2004; Gréau et al., 2011; Huang et al., 2012a; Huang et al., 2014a).

The mantle eclogites can be entrained and transported to Earth's surface by kimberlite/lamproite magmas as “xenolithic eclogites.” Their nature has long been controversial. Two end-member hypotheses for their origins have been proposed based on petrography and traditional geochemical studies (e.g., Hatton and Gurney, 1987; Barth et al., 2001; Jacob, 2004; Schmickler et al., 2004; Aulbach et al., 2007; Griffin and O'Reilly, 2007; Huang et al., 2012a; Smart et al., 2014): (1) metamorphosed oceanic basalts/gabbros subducted into the Earth mantle (subduction model) and (2) frozen lenses and veins of deep-seated magmas that crystallized and equilibrated at depth (magmatic model). Several studies also have documented that at least some eclogites have experienced a complex compositional evolution as a result of mantle metasomatism (Jacob et al., 2009; Smart et al., 2009; Huang et al., 2012a; Gréau et al., 2013; Huang et al., 2014a).

Magnesium isotopes are potentially a “robust” tool to trace the origin of xenolithic eclogites as they do not fractionate significantly during metamorphism or partial melting, and they apparently are not even easily modified during some types of mantle metasomatism (e.g., Teng et al., 2010a; Pogge von Strandmann et al., 2011; Wang et al., 2014b). The variation, if any, of their  $\delta^{26}\text{Mg}$  values therefore cannot be explained easily by these processes but must be related to other processes. For example, if carbonates are involved in the formation and evolution of eclogites, a large variation in  $\delta^{26}\text{Mg}$  would be expected (Wang et al., 2012, 2014a, 2015).

Xenolithic eclogites from the Roberts Victor kimberlite pipe, Kaapvaal Craton, South Africa, have been studied for decades (MacGregor and Carter, 1970; Garlick et al., 1971; Hatton, 1978; Ongley et al., 1987; McCandless and Gurney, 1989; Jacob and Jagoutz, 1991; Jacob et al., 2005; Gonzaga et al., 2010; Riches et al., 2016). More recently, a suite of samples collected by CCFS/GEMOC, Macquarie University has been extensively studied (Gréau et al., 2011; Huang et al., 2012a,b; Gréau et al., 2013; Huang et al., 2014a,b). The origin of Roberts Victor eclogites has been highly debated. They were originally suggested to be mantle magmatic products (Hatton, 1978). As more data on their petrology, trace elements, radiogenic isotopes, especially O isotopes became available, they were interpreted to be subducted metamorphosed oceanic rocks; the eclogites with low  $\delta^{18}\text{O}$  should originate from seawater-altered gabbros and the samples with high  $\delta^{18}\text{O}$  values from fresh or altered MORBs (MacGregor and Manton, 1986; Ongley et al., 1987). More recently, it has been found that most of the rocks have been heavily metasomatized and information on their protoliths has been obscured or obliterated (Gréau et al., 2011; Huang et al., 2012a). The evidence for the subduction model therefore is questionable.

In this paper, we present the Mg-isotope compositions of Roberts Victor eclogites to examine the differences between types, to understand how mantle metasomatism affects Mg-isotope behaviour, to test models for their origin, and more generally to evaluate the homogeneity of Mg isotopes in the mantle and the fractionation of Mg isotopes at high T. We also present in situ O isotope data for the garnets to compare the behaviour of Mg and O isotopes during geological processes.

## 2. Samples

Roberts Victor eclogites were transported to the Earth's surface by Group II kimberlites erupted at  $128 \pm 15$  Ma (Smith et al., 1985). Most samples studied in this paper are large, ranging from fist-sized to  $24 \times 22$  cm, and ovoid in shape. However, some (RV07-30, 31, 33, 34) are only thumb-sized. All of the samples mainly consist of garnet (gnt) and clinopyroxene (cpx) and contain <5 vol.% of secondary minerals. They are different in this respect from orogenic eclogites, which typically have more complex mineral assemblages and may contain a number of secondary minerals (e.g., paragonite, phengite, epidote, zoisite). In terms of mineral compositions and microstructures, the Roberts Victor samples are classified into Type I and II eclogites (MacGregor and Carter, 1970; McCandless and Gurney, 1986; Schulze et al., 2000). Huang et al. (2012a) further divided Type I bimineralic eclogites into Type IA and IB, and Type II into Type IIA and IIB.

The systematic differences between Type I (IA + IB) and Type II (IIA + IIB) eclogites (Gréau et al., 2011; Huang et al., 2012a, 2014a,b) are summarized in Table 1.

## 3. Analytical methods

### 3.1. Magnesium-isotope analyses

Clean mineral separates were handpicked under a binocular microscope after the hand specimen was processed by SelFrag (electrostatic disaggregation). They were then given three ultrasonic rinses with MQ water (18.2 M $\Omega$ cm) before 5–18 mg were weighed into Savillex beakers. After the samples were fully digested, the solution was dried at 160 °C and then dissolved in 2 N HNO<sub>3</sub> in preparation for the column chemistry. Magnesium purification was achieved by cation exchange chromatography with Bio-Rad AG50W-X12 (200–400 mesh) resin. The sample solution with ~20  $\mu\text{g}$  Mg in 1 ml HNO<sub>3</sub> was loaded into the column, and Mg was purified and collected mainly using HNO<sub>3</sub>, except that HNO<sub>3</sub> with HF was used to elute Al, Ti, and Fe quickly and completely after the sample loading. Each sample was passed through the column twice to produce a pure Mg solution. The details of the chemical purification procedure are given by An et al. (2014).

The “pure” Mg solution and 1 ml solution collected both before and after the Mg cut were analyzed by ICP-MS to check that the column chemistry had efficiently removed matrix elements, which can cause analytical artifacts (Galy et al., 2001; Handler et al., 2009), and to assure the Mg yield was close to 100% because Mg isotopes fractionate significantly during the column separation (Chang et al., 2003; Teng et al., 2007). All samples in this study gave matrix/Mg < 0.05, and the yield is ~100%.

Magnesium-isotope values were obtained using a Neptune MC-ICP-MS and the sample-standard bracketing method in both the Institute of Geology and Geophysics, Chinese Academy of Sciences (IGGCAS), and the University of Science and Technology of China (USTC). The Mg solution (~2 ppm Mg in 2% HNO<sub>3</sub>) was introduced into the “wet” plasma using a quartz dual cyclonic-spray chamber and an ESI 50  $\mu\text{L}$  min<sup>-1</sup> PFA Teflon nebulizer. The solution was analyzed in medium-resolution mode with <sup>24</sup>Mg, <sup>25</sup>Mg, and <sup>26</sup>Mg detected in L3, C, and H3 Faraday cups simultaneously.

The precision and the accuracy of the chemical and analytical methods used for this work have been discussed by An et al. (2014). The in-run precision of <sup>26</sup>Mg/<sup>24</sup>Mg for a block of 60-cycles analyses with an integration time of 2.097 s per cycle is <0.02‰ (2SD). The internal precision of samples measured  $\geq 3$  times is <0.05‰ (2SD). The long-term external precision was evaluated by analysis of international Mg solution standards and rock standards and is generally better than 0.07‰ for  $\delta^{26}\text{Mg}$ . All the standards (CAM-1, IGGMg, SRM980, BHVO-2, BCR-2) have  $\delta^{26}\text{Mg}$  values identical to the published data (Table 2; An and Huang, 2014).

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