



Chromium isotope heterogeneity in the mantle



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ABSTRACT

To better constrain the Cr isotopic composition of the silicate Earth and to investigate potential Cr isotopic fractionation during high temperature geological processes, we analyzed the Cr isotopic composition of different types of mantle xenoliths from diverse geologic settings: fertile to refractory off-craton spinel and garnet peridotites, pyroxenite veins, metasomatised spinel lherzolites and associated basalts from central Mongolia, spinel lherzolites and harzburgites from North China, as well as cratonic spinel and garnet peridotites from Siberia and southern Africa. The $\delta^{53}\text{Cr}_{\text{NIST 979}}$ values of the peridotites range from $-0.51 \pm 0.04\text{‰}$ (2SD) to $+0.75 \pm 0.05\text{‰}$ (2SD). The results show a slight negative correlation between $\delta^{53}\text{Cr}$ and Al_2O_3 and CaO contents for most mantle peridotites, which may imply Cr isotopic fractionation during partial melting of mantle peridotites. However, highly variable Cr isotopic compositions measured in Mongolian peridotites cannot be caused by partial melting alone. Instead, the wide range in Cr isotopic composition of these samples most likely reflects kinetic fractionation during melt percolation. Chemical diffusion during melt percolation resulted in light Cr isotopes preferably entering into the melt. Two spinel websterite veins from Mongolia have extremely light $\delta^{53}\text{Cr}$ values of $-1.36 \pm 0.04\text{‰}$ and $-0.77 \pm 0.06\text{‰}$, respectively, which are the most negative Cr isotopic compositions yet reported for mantle-derived rocks. These two websterite veins may represent crystallization products from the isotopically light melt that may also metasomatize some peridotites in the area. The $\delta^{53}\text{Cr}$ values of highly altered garnet peridotites from southern Africa vary from $-0.35 \pm 0.04\text{‰}$ (2SD) to $+0.12 \pm 0.04\text{‰}$ (2SD) and increase with increasing LOI (Loss on Ignition), reflecting a shift of $\delta^{53}\text{Cr}$ to more positive values by secondary alteration.

The Cr isotopic composition of the pristine, fertile upper mantle is estimated as $\delta^{53}\text{Cr} = -0.14 \pm 0.12\text{‰}$, after corrections for the effects of partial melting and metasomatism. This value is in line with that estimated for the BSE ($-0.12 \pm 0.10\text{‰}$) previously.

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1. Introduction

Chromium (Cr) is a compatible and moderately siderophile element that tends to be concentrated in mantle-derived ultramafic rocks as well as in the core. Chromium is also redox sensitive and is present in multiple valence states in different geological settings (e.g., +2, +3 in mantle-derived rocks and +6 in crocoites (Frost, 2004) and highly oxidizing surface environments), which makes the Cr isotope system a valuable tool for reconstructing the evolution of atmospheric oxygen and paleo-redox conditions of the oceans and atmosphere (Frei et al., 2009, 2011; Frei and Polat, 2013). Most previous studies of mass dependent Cr isotope frac-

tionation have focused on large kinetic isotopic fractionation that occurs at Earth's surface induced by redox reactions such as oxidation of Cr^{3+} to Cr^{6+} in response to the rise in the atmospheric oxygen level, or biotic or abiotic reduction of Cr^{6+} (Døssing et al., 2011; Ellis et al., 2002, 2004; Frei et al., 2009; Han et al., 2012; Johnson and Bullen, 2004; Kitchen et al., 2012).

Recently, several studies suggested that the Cr isotopic system has the potential to trace large-scale planetary processes at high temperatures. Moynier et al. (2011) found that carbonaceous, ordinary and enstatite chondrites have $\delta^{53}\text{Cr}$ of -0.2‰ to $\sim -0.4\text{‰}$, which is isotopically lighter than that of the bulk silicate Earth (BSE) ($\delta^{53}\text{Cr} = -0.12 \pm 0.10\text{‰}$, 2SD) (Schoenberg et al., 2008). Thus, Moynier et al. (2011) suggested that there could be significant Cr isotopic fractionation during core segregation, and that light Cr isotopes may have partitioned into the core. A light Cr iso-

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topic composition for one chondrite ($\sim -0.30\%$) was also reported by Schiller et al. (2014). In contrast, Qin et al. (2015), Bonnand et al. (2016a) and Schoenberg et al. (2016) observed that carbonaceous, ordinary and enstatite chondrites have BSE-like stable Cr isotopic compositions (-0.22% to -0.02%). So far, only one study has been done to constrain the Cr isotopic composition of the deep Earth (Schoenberg et al., 2008). In that study, no Cr isotopic fractionation during magmatic processes was identified. Recent studies by Qin et al. (2015) and Bonnand et al. (2016b), however, both documented that magmatic processes can fractionate Cr isotopic composition. Thus, further constraints on Cr isotopic behavior during partial melting and magma differentiation, as well as during other high-temperature processes, are necessary to better constrain the Cr isotopic composition of the BSE before firm conclusions can be made about whether the BSE and chondrites have the same, or different, isotopic compositions. Moreover, both Farkaš et al. (2013) and Shen et al. (2015) found that mantle-derived chromites were isotopically heavier than chromite-bearing and chromite-free mantle peridotites. These observations further imply the potential Cr isotopic fractionation during chromite crystallization from mafic and ultra-mafic melts.

In addition, because Cr is a first-group transition element, the properties (e.g., valence and ionic radius at mantle oxygen fugacity) of Cr ions (Cr^{2+} and Cr^{3+}) are similar to those of Fe ions (Fe^{2+} and Fe^{3+}) to some extent. Previous studies have documented significant Fe isotopic fractionation during high temperature processes, such as partial melting and fractional crystallization (e.g. Schoenberg and Blanckenburg, 2006; Weyer and Ionov, 2007; Williams et al., 2005; Zambardi et al., 2013; Zhu et al., 2002). Schoenberg et al. (2008) concluded that Cr isotopic fractionation during partial melting may be negligible because they found similar isotopic compositions of mantle xenoliths, ultramafic rocks, cumulates and basalts from different locations in their study. The petrogenetic relationships among their samples, however, were not clear. Thus, systematic investigations of Cr isotope behavior during partial melting and magma differentiation are necessary before Cr isotopic composition be applied to constrain high-temperature processes.

To address these questions, we present a systematic Cr isotopic study using a high-precision method on both whole rocks and individual minerals of well-studied mantle xenoliths from different geologic settings. The samples were selected to represent, first, fertile mantle and residues of different degrees of melt extraction from fertile mantle and, second, products of melt-rock reactions and metasomatism in the mantle. The goal of this study is to assess the effects of partial melting and late-stage and secondary processes (metasomatism, alteration) on Cr isotopic fractionation. In addition, since Cr is a compatible element and the mantle is the most important Cr reservoir of the bulk silicate Earth, studying mantle xenoliths provides important constraints on the Cr isotopic inventory of the bulk silicate Earth.

2. Samples

Mantle xenoliths

Forty-five mantle xenoliths from seven locations are investigated here, including garnet-spinel lherzolites, spinel lherzolites, spinel harzburgites, garnet peridotites, a clinopyroxene-bearing garnet orthopyroxenite and spinel websterites.

Mongolia: Mantle xenoliths from Tariat, central Mongolia, are well known for their high proportions of fertile (CaO and Al_2O_3 contents near 4 wt%) peridotites, which are otherwise rare worldwide (Ionov, 2007; Ionov and Hofmann, 2007; Press et al., 1986). Although a majority of the Tariat samples in this study are fertile, the sample suite includes a compositional range of spinel peridotites that reflects the removal of variable amounts of melts

(Ionov and Hofmann, 2007; Ionov, 2007). Samples analyzed in this study were collected from the Shavaryn-Tsaram and Haer basaltic eruption centers. They include fifteen spinel peridotites, one garnet-spinel peridotite, two spinel websterites (4230-15 vein and Sh11-12 vein) and one garnet orthopyroxenite (4399-24 vein) from Shavaryn (Ionov et al., 1998), and six spinel peridotites from Haer. Mantle xenoliths from Shavaryn can be subdivided into: (a) fertile or nearly fertile spinel lherzolites, (b) moderately refractory spinel lherzolites and (c) highly refractory spinel harzburgites that may have undergone metasomatism, as well as (d) spinel and garnet-spinel lherzolites that have Al_2O_3 and CaO contents and modal pyroxenes higher than primitive mantle due to the addition of or reaction with melts during or after partial melting (Ionov, 2007; Ionov and Hofmann, 2007; Ionov et al., 1994). Some of the spinel lherzolites from Haer also show indications of metasomatism, e.g. elevated FeO contents (Ionov, 2007; Table 1). All of the samples are fresh, with minimal signs of alteration. Besides the lithospheric mantle rocks, two samples of basalts (Sh11-1 and Cl11-4) hosting the mantle xenoliths from Shavaryn were also analyzed. A full description of these samples is in preparation and will be published separately.

Siberian craton: Peridotite xenoliths from particularly fresh facies of the Udachnaya-East kimberlite, central Siberia are well-known for their large size, freshness and homogeneity (Ionov et al., 2010). A set of five well-studied Udachnaya peridotites (Ionov et al., 2010, 2015a; Doucet et al., 2012, 2013, 2015) was analyzed that includes two low-opx spinel harzburgites, one garnet lherzolite and two garnet harzburgites that have experienced various degrees of partial melting and metasomatism. In addition, two spinel-rich peridotites from the Obnazhennaya kimberlite in the northeastern Siberian craton were analyzed to explore the effects of high Cr_2O_3 in these rocks (1.1–1.4% vs. 0.3–0.4% in normal peridotites) on Cr isotope compositions. One sample (Obn22-13) is a spinel lherzolite with relatively low Al_2O_3 and high Mg\# (0.917) and is a moderately refractory melting residue. The other one, dunite Obn60-13 with very low Al_2O_3 and CaO represents a refractory mantle source that interacted with evolved melts as suggested by its relatively low Mg\# of 0.888 (Ionov et al., 2015b).

Hannuoba: Four spinel lherzolites and one harzburgite are from Damaping (DMP) in the Hannuoba basalt field. This xenolith locality is in the central part of the North China Craton, which preserves crustal remnants as old as 3.8 Ga (Gao et al., 2002; Liu et al., 1992). However, the ancient, highly refractory mantle lithosphere beneath the craton is believed to have been replaced with generally more fertile materials (e.g. Gao et al., 2002). Hannuoba mantle xenoliths vary from relatively fertile spinel lherzolite to refractory harzburgite. Their major element compositions indicate variable degrees (0–25%) of melt extraction from a primitive mantle source (Rudnick et al., 2004). The Sr–Nd systematics and widely varying REE patterns of these xenoliths suggest that they might be influenced by an evolved metasomatic melt (Rudnick et al., 2004; Tang et al., 2013; Zhang et al., 2009). Detailed information on these samples can be found in Rudnick et al. (2004).

Southern Africa: Similar to the peridotite xenoliths from the Siberian craton, the Kaapvaal garnet peridotites were brought to the surface by kimberlite magmas (Lazarov et al., 2012a, 2012b; Simon et al., 2003, 2007). Our samples come from the Kimberley and Letlhakane kimberlites; they are composed of low-T garnet peridotites, spinel lherzolite and harzburgite which are characterized by low Al_2O_3 and high Mg\# (Table 2). Mantle xenoliths from Kimberley in the western part of the Kaapvaal Craton are mostly lherzolites and harzburgites, with rare dunites, wehrlites and websterites (Bell et al., 2005; Griffin et al., 1999; Simon et al., 2007), affected by multi-stage metasomatism by a variety of metasomatic agents (Grégoire et al., 2003; Janney et al., 2010; Lazarov et al., 2012b; Simon et al., 2007). Letlhakane contains a

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