

^{57}Fe Mössbauer spectroscopy of mineral assemblages in mantle spinel lherzolites from Cenozoic alkali basalt, eastern China: Petrological applications

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ABSTRACT

Mineral assemblages in spinel lherzolite xenoliths from the Cenozoic alkali basalt of eastern China were analyzed by ^{57}Fe Mössbauer spectroscopy for $\text{Fe}^{3+}/\Sigma\text{Fe}$ and distribution of $\text{Fe}^{2+}/\text{Fe}^{3+}$ in non-equivalent crystal sites. Orthopyroxene, clinopyroxene and spinel have 0.08–0.13, 0.19–0.31 and 0.13–0.23 $\text{Fe}^{3+}/\Sigma\text{Fe}$, respectively. The cation-exchange equilibrium temperatures, Fe^{3+} partition coefficient and oxygen fugacity of the upper mantle peridotites were calculated. The equilibrium temperatures are between 1103 K and 1405 K consistent with those reported elsewhere. The oxygen fugacities of spinel lherzolites calculated with olivine–orthopyroxene–spinel ($\Delta\log f_{\text{O}_2}$ from -1.1 to 0) and clinopyroxene–olivine–orthopyroxene oxybarometers ($\Delta\log f_{\text{O}_2}$ from -2.0 to 0.7) are consistent with previously reported upper mantle values.

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1. Introduction

The nature of upper mantle mineralogy and geochemistry is a fundamental question for the structure and evolution of Earth. Our understanding of this issue is mainly based on peridotite xenoliths brought to the surface by magma. Oxygen fugacity is an important issue for the understanding of the heterogeneity of upper mantle and mantle–crust interactions (Arculus, 1985). As iron is the most abundant transition metal with multivalent states in the mantle mineral assemblage, $\text{Fe}^{3+}/\Sigma\text{Fe}$ values in the coexisting minerals decisively buffers mantle oxygen fugacity (Woodland et al., 2006). With high abundance of iron and negligible Fe^{3+} content, olivine is a substantial reservoir of Fe^{2+} in the mantle (Canil and O'Neill, 1996; Nakamura and Schmalzried, 1983; O'Neill et al., 1993; Woodland and Angel, 2000). Therefore, the distribution of Fe^{3+} between orthopyroxene, clinopyroxene and spinel determines the redox state of the lherzolite xenoliths which also represents the oxygen fugacity of the upper mantle.

Although there are several previous Mössbauer spectroscopic studies on the distribution of Fe^{3+} between the co-existing phases in mantle peridotite from different locations, the whole data set is still small. A suite of mantle xenoliths' data on British Columbia were reported by Canil et al. (1990) and Luth and Canil (1993). Data of Fe^{3+} distribution in minerals of spinel peridotite xenoliths mostly from the western United States was reported by Dyar et al. (1989, 1992) and McGuire et al. (1991). A significant set of data on a broad range of samples were reported by Canil and O'Neill (1996).

Despite the importance of quantitative estimates of mantle oxygen fugacity based on Mössbauer analysis, such data are very limited in eastern China with a vast distribution of Cenozoic alkali basalts.

Cenozoic basalts are widespread in eastern China and formed in three major stages: Paleogene, Neogene and Quaternary (Chi, 1988). Generally, these basalts erupted along major continental rift zones or as flood basalts unrelated to those continental rift zones; they are also exposed at, or near, continental plate boundaries between the Eurasian and Indian plates (Zhou et al., 1988). Xenoliths that can be commonly found in these Cenozoic basalts include spinel- and garnet-peridotites and augite high-pressure megacrysts. The latter mainly occur in Neogene and Quaternary alkali basalts. Liuhe-Nanlianshan, Panshishan and Nüshan are three well studied areas for mantle xenoliths in eastern China. Previous studies have presented elemental and isotopic geochemical and petrological results (Chen et al., 1994a, 1994b; Li et al., 1999; Lu et al., 2012; Yu et al., 2005a, 2005b). In this study, ^{57}Fe Mössbauer spectroscopy was used to measure the hyperfine parameters, the distribution of Fe^{3+} and Fe^{2+} interstitially and intrastitially in the peridotite assemblages to provide a data set from China for mantle oxygen fugacity and accurate cation-exchange equilibrium temperature of the upper mantle. These data are useful in the calculation of the regional mantle redox state and expand the worldwide data set of mantle oxygen fugacity.

2. Materials and method

Thirteen peridotite xenolith samples were collected from Liuhe-Nanlianshan of Anhui Province, Panshishan and Nüshan of Jiangsu Province of Eastern China (Fig. 1). The petrologic and the

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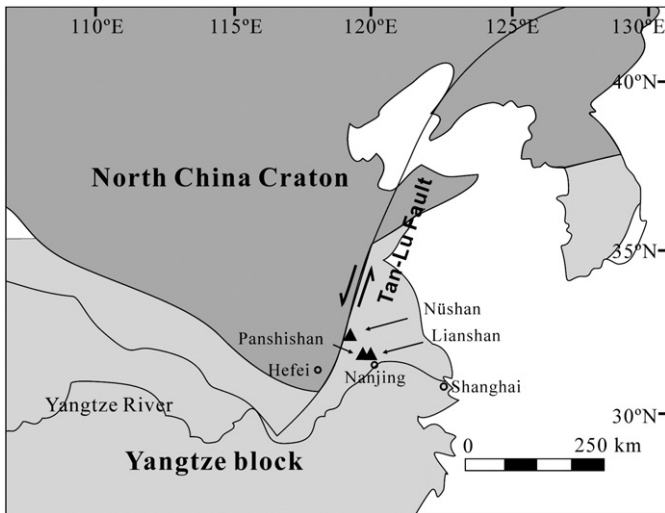


Fig. 1. Sketch tectonic map of eastern China showing peridotite xenolith localities.

geochemical background of these samples were reported in Yu et al. (2005a) and Yang et al. (2008). The samples are hosted by alkali basalts and are all spinel-facies peridotites, dominantly coarse grained spinel lherzolites with rare spinel harzburgites. Single crystals of optical purity from peridotite assemblages were picked under a binocular microscope. The selected minerals were immersed in dilute HCl acid for 2 h to remove the surface impurities before the analysis of chemical compositions on a JEOL733-JXA8800M microprobe operated at 25 kV accelerating voltage and 25 nA current at Nanjing University.

For Mössbauer spectroscopic measurements, the selected mineral crystals were immersed in anhydrous alcohol for grinding to 200 mesh in an agate mortar. The ^{57}Fe Mössbauer spectra were collected on OXFORD-MS-500 Mössbauer spectrometer with a 25 mCi $^{57}\text{Co}/\text{Pb}$ source at the University of Science and Technology of China. The finely ground minerals were mounted in an acrylic holder with relative thicknesses between 3 and <10 mg Fe/cm^2 depending on the nature of the sample and the condition of the radioactive source. Spectra were collected at room temperature in transmission mode with a constant acceleration. The velocity scale was calibrated relative to 25- μm $\alpha\text{-Fe}$ at room temperature. Lorentzian doublets were used and the recoilless fractions of iron in octahedral and tetrahedral crystallographic sites were considered equal (Gorski and Scherer, 2010) for fitting the areas of sub-spectra.

3. Results and discussion

3.1. Chemical composition

The major element compositions of olivine, orthopyroxene, clinopyroxene and spinel are listed in Table 1. The composition of olivine is homogeneous with forsterite component (X_{fo}) ranging from 0.88 to 0.91; orthopyroxene and clinopyroxene have X_{Mg} values of 0.90–0.92 and 0.90–0.94 respectively which are typical of mantle-derived spinel lherzolite. The Fe and Mg distributions among these three minerals are positively correlated showing thermodynamic equilibrium in the peridotite assemblages (Woodland et al., 2006).

The $\text{Cr}/(\text{Cr} + \text{Al} + \text{Fe}^{3+})$ in spinel, is negatively correlated with $\text{Mg}/(\text{Mg} + \text{Fe}^{2+})$ and Al contents of the co-existing clinopyroxene (Fig. 2). Such relations can be attributable to variable degrees of melt extraction experienced by the individual samples (Dick and Fisher, 1984; Woodland et al., 2006). Samples that have experienced higher degrees of partial melting have higher $\text{Cr}/(\text{Cr} + \text{Al} + \text{Fe}^{3+})$ and

lower $\text{Mg}/(\text{Mg} + \text{Fe}^{2+})$ in spinel and lower Al concentration in clinopyroxene (Fig. 2).

3.2. Mössbauer spectra of the co-existing minerals

The Mössbauer spectroscopic results of the co-existing minerals are listed in Table 2. Typical spectroscopic profiles of individual minerals are shown in Fig. 3.

3.2.1. Olivine

Olivine from all sites is Mg-rich forsterite in which the isomorphous cations occupy two non-equivalent oxygen coordinated octahedral sites M1 and M2 (Fig. 3A). Based on Stanek et al. (1986) and Dyar et al. (1989), doublets with quadrupole splitting (QS) from 2.76 to 2.92 mm/s and isomer shift (IS) from 1.05 to 1.15 mm/s were assigned to Fe^{2+} in the M1 site while doublets with QS from 2.94 to 3.15 mm/s and IS from 1.08 to 1.17 mm/s were assigned to Fe^{2+} in the M2 site (Table 2). No Fe^{3+} doublet could be fitted in any olivine samples, indicating undetectable Fe^{3+} in the structures.

3.2.2. Orthopyroxene

Orthopyroxene has two non-equivalent oxygen coordinated octahedral positions (M1 and M2). The orthopyroxene spectra were fitted with three quadrupole doublets for Fe^{2+} and one doublet for Fe^{3+} (Fig. 3B). Similar to Woodland et al. (2006) and Nikitina et al. (2010), one doublet with QS from 2.37 to 2.68 mm/s and IS from 1.13 to 1.19 mm/s was assigned to Fe^{2+} in the M1 site while two doublets with QS from 1.82 to 2.22 mm/s and IS from 1.07 to 1.17 mm/s were assigned to Fe^{2+} in the M2 site. The broadened line widths of M2 were fitted as two doublets M2a and M2b according to the next-nearest-neighbor (NNN) effect (Seifert, 1983). Doublets with QS from 0.39 to 0.84 mm/s and IS from 0.07 to 0.44 mm/s were assigned to Fe^{3+} in M1 site (Table 2). Dollase and Gustafson (1982) suggested that Fe^{3+} in tetrahedral position should have an average zero IS value and a larger QS (about 1.5 mm/s). In most samples of this study, Fe^{3+} occupies M1 site with the exception of LHLS-1 (IS = 0.07 mm/s and QS = 0.39 mm/s) which might suggest the occupation of tetrahedral site by some Fe^{3+} .

3.2.3. Clinopyroxene

Because of the complex composition and possible NNN effects of clinopyroxene, the fitted spectra were model-dependent. We fitted the spectra with two Fe^{2+} doublets and one Fe^{3+} doublet (Fig. 3C) as in previous studies (Canil and O'Neill, 1996; Luth and Canil, 1993; McCammon et al., 1998; Sobolev et al., 1999; Woodland et al., 2006). Doublets with QS from 0.48 to 0.88 mm/s and IS from 0.16 to 0.43 mm/s were assigned to Fe^{3+} in M1; and doublets with QS from 2.41 to 2.95 mm/s and IS from 1.09 to 1.25 mm/s were assigned to Fe^{2+} in the M1 position while doublets with QS from 2.00 to 2.14 mm/s and IS from 1.09 to 1.16 mm/s were assigned to Fe^{2+} in M2 sites (Table 2).

3.2.4. Spinel

In total four doublets were fitted including three Fe^{2+} doublets and one Fe^{3+} doublet for all spinels (Fig. 3D). On the basis of previous studies (Wood and Virgo, 1989; Woodland et al., 2006), doublets with QS from 0.68 to 0.89 mm/s and IS from 0.18 to 0.48 mm/s were assigned to Fe^{3+} in octahedral sites; and doublets with QS from 0.85 to 1.96 mm/s and IS from 0.80 to 1.17 mm/s were assigned to Fe^{2+} in tetrahedral sites (Table 2). Three doublets rather than a single doublet of Fe^{2+} were fitted to represent the split spectrum of tetrahedral Fe^{2+} which otherwise would show broadened line widths. The NNN effect stems from the distinct radii of trivalent cations (Fe^{3+} , Cr^{3+} , and Al^{3+}) in octahedral site and thus result in symmetric variances in the tetrahedral sites (Nikitina et al., 2010).

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