Cation ordering in synthetic and natural Ni–Mg olivine

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Abstract

The crystal structures of natural liebenbergite—(Ni\(_{1.55}\)Co\(_{0.09}\)Fe\(_{0.45}\)Mg\(_{0.34}\))SiO\(_4\)—from Barberton, South Africa, and of synthetic liebenbergite—(Ni\(_{1.16}\)Mg\(_{0.84}\))SiO\(_4\)—synthesized at 500°C, have been studied in order to determine the intracrystalline Ni–Mg distribution. The natural liebenbergite is fully ordered, with M1 occupied only by Ni, whereas the synthetic sample is only partially ordered with \(K_D = [\text{Mg}(\text{M2}) \cdot \text{Ni}(\text{M1})]/[\text{Ni}(\text{M2}) \cdot \text{Mg}(\text{M1})] = 9.9(4)\), \(\Delta G_{\text{M1}}^\circ = -3.5\) kcal/mole. Comparison with the results of Rajamani et al. (1975), who found \(K_D = 9.2(2)\) and \(\Delta G_{\text{M1}}^\circ = -6.9\) kcal/mole for a sample synthesized at 1280°C, implies that the 500°C synthetic sample in the present study crystallized metastably in a disordered or partially ordered state and ordering proceeded slowly.

Although site size effects are small in Fe–Mg, Ni–Mg, and Co–Mg olivines, the crystal field stabilization energy is important in determining the observed cation distribution in Ni–Mg and Co–Mg olivines. Electronegativity or covalency effects are known only qualitatively, but there is a preference of less electronegative ions (Mg, Ca) for M2.

Ordering of Ni into the M1 site of olivine should appreciably affect Ni partitioning between olivine and melt, and activity-composition relations have been examined assuming ideal solution behavior. Deviations from Raoult's law increase with increasing order, but variations in activity coefficients are less than 10 percent below 10 mole% Ni.

Introduction

Although nickel is a common and important minor constituent in olivines, liebenbergite, the nickel-rich olivine, has been found only in the unusual Bon Accord deposit in Barberton, South Africa. The material at Bon Accord originally filled interstices between trevorite grains in the assemblage trevorite–nickel serpentinite–nickel ludwigite–bunsenite–violarite–millerite–gaspeite–nimitite but is now almost completely replaced by secondary nickel serpentinite. Only small irregular crystals remain in the serpentinite matrix (de Waal and Calk, 1973). The liebenbergite appears to have formed at about 730°C and less than 2 kbar during thermal metamorphism, possibly of a nickel-rich meteorite (de Waal, 1978). I have obtained crystals of liebenbergite through the courtesy of Dr. de Waal.

The natural occurrence of liebenbergite presents us with the unique opportunity to examine a mineral that previously had been studied in synthetic form only (e.g. Rajamani et al., 1975). The natural sample equilibrated over a long period at a fairly low temperature (730°C), so it should possess an equilibrium distribution of cations. Typical synthesis experiments are of short duration at high temperature. For example, the Ni–Mg olivine examined by Rajamani et al. (1975) was crystallized at 1280°C and cooled in several days yielding a zoned crystal. The authors questioned whether the observed \(K_D\), \([\text{Mg}(\text{M2}) \cdot \text{Ni}(\text{M1})]/[\text{Ni}(\text{M2}) \cdot \text{Mg}(\text{M1})]\), represented an equilibrium distribution of cations.

During a study of hydrous nickel–magnesium silicates (Brindley et al., 1979), euhedral crystals of liebenbergite were synthesized by reacting natural pimelite (nickel–magnesium talc) at 500°C and 2.1 kbar for 4 weeks. These crystals should differ significantly from the products of short-term reactions at temperatures in excess of 1200°C.

Numerous studies have focused on the nature and causes of intracrystalline cation distributions in natural and synthetic olivines (see Rajamani et al., 1975, for a summary). In an attempt to understand order-
### APPENDIX A

**OBSERVED AND CALCULATED STRUCTURE FACTORS FOR Ni-Mg OLIVINE (Synthetic)**

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