Stability of magnesite in the presence of hydrous fluids up to 12 GPa: Insights into subduction zone processes and carbon cycling in the Earth’s mantle

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ABSTRACT

Understanding the stability of magnesite in the presence of a hydrous fluid in the Earth’s upper mantle is crucial for modeling the carbon budget and cycle in the deep Earth.

This study elucidates the behavior of magnesite in the presence of hydrous fluids. We examined the brucite-magnesite [Mg(OH)2-MgCO3] system between 1 and 12 GPa by using synchrotron in situ energy-dispersive X-ray diffraction experiments combined with textural observations from quenched experiments employing the falling sphere method. By subjecting magnesite to varying pressure-temperature conditions with controlled fluid proportion, we determined the stability limits of magnesite in the presence of a fluid and periclase.

The observed liquidus provides insights into the fate of magnesite-bearing rocks in subduction zones. Our findings show that magnesite remains stable under typical subduction zone gradients even when infiltrated by hydrous fluids released from dehydration reactions during subduction. We conclude that magnesite can be subducted down to and beyond sub-arc depths. Consequently, our results have important implications for the carbon budget of the Earth’s mantle and its role in regulating atmospheric CO2 levels over geological timescales.

Keywords: Deep carbon cycle, brucite dehydration, magnesite melting, EDXRD; Physics and Chemistry of Earth’s Deep Mantle and Core

INTRODUCTION

The fates of carbon and hydrogen in the mantle are intimately linked to each other. For instance, the release of CO2 from carbonate lithologies in the slabs during subduction is enhanced by dehydration reactions because water acts as a catalyst and solvent in decarbonation and dissolution reactions (summarized in Galvez and Pubellier 2019). With increasing water activity, the solidus of carbonate-bearing rocks is drastically reduced, promoting the formation of carbonate melts (Wyllie and Tuttle 1960; Poli 2015; Poli et al. 2009). Thus, infiltration of aqueous fluids (e.g., released in dehydration reactions) into slab sediments can produce a carbonate-bearing fluid and/or melt. The extent to which these processes operate is controversial (Kelemen and Manning 2015; Orcutt et al. 2019). Due to the breakdown of dolomite with increasing pressure, magnesite (MgCO3) is the most dominant carbonate phase in the CO2-peridotite system above ~4 GPa (Falloon and Green 1989; Dasgupta and Hirschmann 2006; Dasgupta et al. 2004; Shen et al. 2018) and plays an important role in the cycle of oxidized carbon into the deep Earth. However, to date, the stability of magnesite in the presence of a H2O-bearing fluid is still poorly constrained.

Carbonates in the upper mantle are nominally anhydrous, meaning their crystal structure does not incorporate H2O and their stability has been studied intensively (see Shatskiy et al. 2015 and references therein). In the (dry) MgO-CO2 system, magnesite dissociates up to 2.3 GPa and 1550 °C, where magnesite begins to melt incongruently forming periclase and carbonate liquid (Huang and Wyllie 1976; Irving and Wyllie 1975). Earlier studies reported congruent melting of magnesite at pressures ≥2.7 GPa (Huang and Wyllie 1976; Shatskiy et al. 2016; Katsura and Ito 1990). Sieber et al. (2022) recently revised the melting reaction, demonstrating incongruent melting of magnesite in the anhydrous system at 6 and 9 GPa.

A better investigation of the stability of magnesite in the presence of a free fluid phase is needed because only a marginally lower liquidus in the water-bearing system (1700–1750 °C) compared to anhydrous conditions (1750–1800 °C) was obtained from experiments on hydromagnesite at 6 GPa employing the falling sphere technique in ex situ experiments (Müller et al. 2017). Compared to a silicate-bearing system, such a small reduction of the liquidus temperature by H2O is surprising (e.g., Green et al. 2014). Interpretation of the equilibrium assemblage from quenched experiments based on crystal sizes and textural features is problematic due to the reactivity of the quenched vapors and solids (Walter et al. 1962). For instance, quenched periclase (MgO) reacts with a H2O-fluid at high pressure to brucite [Mg(OH)2] (Della Roy and Roy 1957) and with a CO2-H2O-fluid to magnesite, brucite, and/or nesquehonite (Walter et al. 1962). Therefore, in situ X-ray diffraction investigations of the melting conditions of magnesite in the presence of an aqueous fluid are strongly needed because, until now, melting conditions and phase equilibria are only available from quenched experiments.

We investigate the stability of magnesite in the presence of a hydrous fluid under mantle conditions between 1 and 12