**The size distribution of exsolution lamellae in iron-free clinopyroxene**

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**ABSTRACT**

The size distribution of pigeonite and diopside exsolution lamellae on “001,” obtained at temperatures of 1100, 1200, and 1300 °C and annealing times between 2 and 4320 h, was studied by transmission electron microscopy. A total of 5192 pigeonite and 5286 diopside lamellae was studied. At all three temperatures, the size distributions of pigeonite and diopside lamellae are smaller during exsolution compared to the subsequent coarsening process. The final size distributions are time invariant, indicating that a steady-stage distribution is reached. The theory of Ardell (1972a), which assumes volume diffusion as rate-limiting process and takes into account the non-zero volume fraction of the precipitates, describes the experimental size distributions quite well and also leads to the observed exponent of three in the rate law.

**Keywords:** exsolution, clinopyroxene, size distribution, exsolution lamella, electron microscopy, experimental petrology, kinetics

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**INTRODUCTION**

The wavelength of “001” exsolution lamellae [i.e., lamellae with composition planes approximately parallel to (001)] has been often used to constrain cooling rates of terrestrial and extraterrestrial rocks (e.g., Takeda et al. 1975; Grove 1982; Watanabe et al. 1985; Brizi and Mellini 1992; Weinbruch and Müller 1995; McCallum and O’Brien 1996; Weinbruch et al. 2001; Ferraris et al. 2002). The cooling rate estimates are based on coarsening of the exsolution lamellae observed during isothermal annealing (McCallister 1978; Nord and McCallister 1979; Weinbruch et al. 2003), or continuous cooling experiments (Grove 1982; Fukuda et al. 1987; Weinbruch et al. 2001).

In a recent study by Weinbruch et al. (2003), the exsolution mechanism and the coarsening kinetics in Fe-free clinopyroxene were investigated in detail. From the long-term isothermal annealing experiments (up to 4320 h), an exponent of approximately three was obtained in the rate law for coarsening (Weinbruch et al. 2003), indicating volume diffusion within both phases as rate-limiting process (e.g., Doherty 1983; Joesten 1991; Gleiter 1996). The grain-size distributions for coarsening of single-phase materials and for multiphase materials have been derived theoretically (see the excellent reviews by Martin and Doherty 1976; Ardell 1988; Joesten 1991; and references therein). However, the experimental database is scanty for geomaterials (e.g., Eberl et al. 1990, 1998; Joesten 1991).

In the present study, we investigate the development of the size distribution of pigeonite and diopside “001” exsolution lamellae during coarsening and compare the results with predictions of various theoretical models. This paper complements our previous publication (Weinbruch et al. 2003) where the exsolution mechanism and coarsening kinetics were described.

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**EXPERIMENTAL METHODS**

Isothermal annealing experiments were carried out with synthetic Fe-free clinopyroxene of composition En34.6Di65.4 at temperatures of 1100 °C (10 min to 4320 h), 1200 °C (1 h to 720 h), and 1300 °C (1 h to 360 h). The width of pigeonite and diopside “001” exsolution lamellae was determined by transmission electron microscopy (TEM). All experimental details can be found in our previous paper (Weinbruch et al. 2003).

**RESULTS**

The width of 30 to 480 pigeonite and diopside “001” exsolution lamellae was determined on each sample leading to a total of 5192 lamellae investigated for pigeonite, and 5286 lamellae for diopside. The exact locations, where the width of the individual lamella was measured, was chosen randomly to avoid any bias in the data. The development of the microstructure during coarsening at a temperature of 1200 °C is illustrated in Figure 1 (additional TEM images of exsolution lamellae can be found in Weinbruch et al. 2003). The arithmetic mean and the standard deviation for the pigeonite and diopside lamellae width as function of temperature and annealing time are listed in Table 1. The ratio of the diopside/pigeonite lamellae width (Table 1) increases with increasing annealing temperature, and the following mean values were obtained: 1.63 for 1100 °C, 1.95 for 1200 °C, and 2.28 for 1300 °C. From these ratios, the volume fraction of the two phases can be determined (1100 °C: 62.0 vol% Di and 38.0 vol% Pig; 1200 °C: 66.1 vol% Di and 33.9 vol% Pig; 1300 °C: 69.8 vol% Di and 30.2 vol% Pig).

The size distributions of exsolution lamellae encountered at a temperature of 1100 °C are displayed in Figures 2 (pigeonite) and 3 (diopside) as function of annealing time. Shown is the reduced lamellae width ω', which is obtained by normalizing (separately for each temperature and annealing time) the measured lamellae width l to the mean value \( \bar{l} \) (ω' = l/\( \bar{l} \)). The size distribution of the reduced lamellae width is defined that \( f'(\omega')d\omega' \) is the probability, normalized to unity, that a given lamella has a reduced

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