Experimental constraints on the dihedral angle between olivine and multicomponent aqueous fluids in the upper mantle conditions

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Generation and migration of aqueous fluid are crucial to understand the subduction zone dynamics including arc volcanism, seismic activities, and element recycling. In fluid-bearing rocks, the dihedral angle, θ, formed by two intersecting walls of a pore at a junction with two solid grains, is one of the key parameters to determine the distribution and connectivity of fluid phases that strongly affect the physical properties of rocks such as permeability, elasticity, and electrical conductivity. Theoretically, the dihedral angle of 60° is a threshold value to control whether an interconnected network could be formed or not in an isotropic system. Dihedral angles are dependent on temperature and pressure, but also on the composition of fluid. Fluid released from a descending slab is often assumed to be composed of pure H_2O. However, studies of fluid inclusions in the mantle xenolith (Kawamoto et al., 2013), upwelling slab-derived fluid in fore-arc arc regions (Morikawa et al., 2016), and melt inclusion from primitive arc basalts (Wallace, 2005; Keppler, 2017) suggested that CO_2 and NaCl could be the important constituents of aqueous fluid in subduction zones. The effect of H_2O-CO_2-NaCl fluid on dihedral angle is, however, still poorly constrained. We conducted the annealing experiments in the systems of olivine-H_2O-CO_2-NaCl at 1.0-4.0 GPa and 800–1100°C for 72–192 hours by using piston cylinder apparatus at Tohoku University and Bayerisches Geoinstitut, Bayreuth University. Measurements of dihedral angles were made from 200~220 secondary electron or backscattered images taken by FE-SEM at magnifications above×6,000 ~×150,000, depending on the size of grains and pores. Because our research is still in progress, here we present only the results obtained at 1 GPa and 1000°C. At this P-T condition, the olivine-fluid dihedral angles in the H_2O, H_2O-NaCl (~27.5 wt% NaCl), CO_2-rich fluid ((COOH)$_2$),H_2O-CO_2 (H_2O:CO_2 = 2:1 in mol) and H_2O-CO_2-NaCl systems were 64.1°, 56.1°, 78.2°, 62.7° and 58.2°, respectively. Since Watson and Brenan (1987) also investigated the olivine-fluid dihedral angle in the H_2O, H_2O-NaCl (~27.5wt% NaCl), and CO_2 system at the same P-T conditions, we can compare our results with theirs. In the H_2O system, the dihedral angle of 64.1° is in accordance with their study—65°. The dihedral angles of 56.1°in the H_2O-NaCl system is slightly smaller than that reported in the previous study (60°). The dihedral angle in the (COOH)$_2$ system (78.2°) is clearly lower that the pure CO_2 system (90°) in which $Ag_2C_2O_4$ was used as a starting material. NaCl in aqueous fluid decreased the olivine-fluid dihedral angle and CO_2 also slightly decreased the angle in the H_2O-CO_2 system although the angle in the CO_2-rich system is much larger than that in the H_2O system. In the H_2O-CO_2-NaCl system, the dihedral angle fell between the values in the H_2O-NaCl and H_2O-CO_2 systems. One of the key factors to understand these results can be the olivine solubility in the aqueous fluid. Yoshino et al., (2007) demonstrated that olivine-fluid dihedral angle decreased with increasing pressure, which was caused by the increase of olivine solubility. Recent experimental studies indicated that the solubility of olivine can be high in both H_2O-NaCl and H_2O-CO_2 systems compared to that in the H_2O system (Wykes et al., 2011; Tiraboschi et al., 2018), which could explain the low dihedral angles in both systems. According to this relationship, we expect that in the H_2O-CO_2-NaCl system CO_2 limited the olivine solubility in H_2O-NaCl fluid. The results of this research could provide indispensable information on the interpretation of seismic tomography and electric conductivity imaging and also forward geodynamic modeling in subduction zones.

Keywords: dihedral angle, multicomponent fluid, interconnected network, fluid migration, subduction zone