# Preliminary results of experimental re-equilibration studies of natural H<sub>2</sub>O-CO<sub>2</sub>-NaCl-bearing fluid inclusions in quartz

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### INTRODUCTION

Experimental diffusion studies with fluid inclusions are performed under hydrothermal conditions to obtain diffusion rates of water related species through the quartz crystal. Synthetic fluid inclusions are synthesised under well defined experimental conditions und re-equilibrated with an external fluid at elevated temperatures and pressures (see Doppler et al., this volume). In addition to synthetic inclusions natural, negativecrystal shaped inclusions (Fig. 1A) in quartz veins in gneisses from the Alpeiner Scharte (Olperer, Tauern Window, Austria) are used for reequilibration studies. Before re-equilibration, those inclusions were analysed accurately by optical investigation (estimation of volume fractions, size and shape of the inclusions), microthermometry and Raman spectroscopy to characterize their bulk fluid properties.

## NATURAL FLUID INCLUSIONS

The fluid in natural inclusions belongs to the ternary NaCl-CO<sub>2</sub>-H<sub>2</sub>O system. Solid phases such as mica, most probably muscovite, and an unknown hairy-like phase are found in some inclusions. The perfect shape of the fluid inclusions and the perfectly spherical vapour bubble do not allow an optical observation of separated CO<sub>2</sub> liquid and vapour phase, i.e. the CO<sub>2</sub> liquid phase is hidden in the dark rim of the vapour bubble. Therefore, the density of the vapour phase  $(CO_2)$ was determined after the method described by Fall et al. (2011) by measuring the distance between the main Raman peaks of CO<sub>2</sub> at about 1285 cm<sup>-1</sup> and 1388 cm<sup>-1</sup> at 40 °C. Salinity calculations were performed with the computer program Q2 (Bakker, 1997). From the obtained data, the fluid composition is calculated to about 74 mass% H<sub>2</sub>O, 19 mass% CO<sub>2</sub> and 7 mass% NaCl. The total molar volume of the inclusions is about 25

cm<sup>3</sup>/mol. According to these fluid properties the specific isochore of fluid trapping was calculated by using the program ISOC (Bakker, 2003). Re-equilibration experiments are performed along this specific isochore to beware of pressure difference and of inclusion stretching and/or decripitation.



Fig. 1. A) Natural NaCl-CO<sub>2</sub>- $H_2O$  inclusions before re-equilibration. B) Fluid inclusions after re-equilibration with pure H2O.

#### **RE-EQUILIBRATION EXPERIMENTS**

First experiments were carried out at 600 °C and about 400 MPa with 1) pure water and 2)  $D_2O$  as external fluid.  $D_2O$  does not play a role in

natural fluid systems, but it is used as tracer for diffusion studies because it has similar properties as  $H_2O$ , and it is easily identified by Raman spectroscopy by its specific vibrational modes between 2200 and 2800 cm<sup>-1</sup>.

Differences in the chemical potential (concentration of species) are known as the controlling parameters of diffusion. In theory,  $CO_2$  and NaCl outward diffusion through the quartz crystal should occur in both experiments. As the  $CO_2$  molecule and also the atomic radii of Na<sup>+</sup> and Cl<sup>-</sup> (dissolved in the aqueous solution) are relatively large compared to atomic distances in the crystal lattice of quartz, no efficient transport is assumed within 19 days of re-equilibration. In addition Na<sup>+</sup> and Cl<sup>-</sup> occur as charged ions, which also decelerate the transport out of the inclusions due to adsorption processes.

The difference in water fugacity at experimental conditions for the pure water reequilibration is chosen to be very little (about 217 MPa in the external fluid and 182 MPa in the fluid inclusions; calculated with the software LONER AP and LONER HKG, Bakker, 2003) and therefore no significant fluid flow is assumed. Preliminary results illustrate minor changes in fluid composition. Nevertheless, inclusions shape may modify during re-equilibration as it is shown in Figure 1.

Fluid inclusions which were re-equilibrated with  $D_2O$  show modifications in fluid composition. Minor amounts of  $D_2O$  are clearly identified by Raman spectroscopy (see Fig. 2). The amount of  $D_2O$  transported into the inclusion varies. The intensity of variation is mainly caused by the position of the inclusion in the quartz sample (3dimensional distance from the quartz surface) and the total volume of the inclusions.

#### OUTLINE

Further experiments with different compositions of the external fluid at different experimental conditions (temperature and pressure) will be carried out to obtain new data of diffusion rates in natural quartz-fluid systems in order to create new diffusion models and compare them with already established ones.



Fig. 2. A) Raman spectrum of  $H_2O$  measured in a fluid inclusion before re-equilibration. B) Raman spectrum of  $D_2O-H_2O$  mixture after 19 days of re-equilibration.

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