Synthetic fluid inclusions. XV. TEM investigation of plastic flow associated with reequilibration of fluid inclusions in natural quartz

Abstract The nature and abundance of dislocations in quartz surrounding fluid inclusions were studied to obtain a better understanding of processes associated with fluid inclusion reequilibration. Synthetic fluid inclusions containing 10 wt% NaCl aqueous solution were formed in three samples at 700 °C and 5 kbar. One of the samples was quenched along an isochore to serve as a reference sample. The other two samples were quenched along a P-T path that generated internal pressures in excess of the confining pressure. The two samples were held at the final reequilibration P-T conditions of 625 °C and 2 kbar for 30 and 180 days, respectively. Following the experiments, microstructures associated with fluid inclusions were examined with the TEM. Quartz in healed fractures in the reference sample that was quenched isochorically shows a moderate dislocation activity. Quartz adjacent to reequilibrated fluid inclusions in the other two samples, however, showed a marked increase in dislocation activity compared to the un-reequilibrated sample. Deformation of the inclusion walls occurred anisotropically by expansion of mobile dislocations in their slip systems. Dislocation expansion was controlled by glide in the rhombohedral planes {1 0 1 1} that was restricted to narrow zones (≤3 μm) in the immediate vicinity of the fluid inclusion walls outside of the healed fracture plane. These plastic zones were observed after both short term (30 days) and long term (180 days) experiments and are attributed to hydrolytic weakening of quartz around fluid inclusions owing to diffusion of water into the quartz matrix during the experiment. The close spatial association of submicroscopic water bubbles with dislocations, and the rarity of water bubbles in the reference sample, show clearly that in both the 30 and 180 day experiments reequilibration involves water loss from the fluid inclusions. Our results indicate that synthetic fluid inclusions in this study recover (chemically and volumetrically), even at relatively fast experimental loading rates, such that internal stresses never reach the point of brittle failure. The driving force for fluid inclusion deformation involves two related mechanisms: plastic deformation of hydrolytically weakened wet quartz in the healed fracture, and water leakage associated with preexisting and strain-induced dislocations.

Introduction

Larson et al. (1973) suggested that anomalously high homogenization temperatures of fluid inclusions in fluorite reported earlier were the result of inclusion volume increases generated during sample preparation and/or heating tests. These workers coined the term “stretching” to describe this behavior. Soon, many other papers were published which described inclusion reequilibration caused by laboratory heating at 1 atmosphere confining pressure (e.g., Leroy 1979; Swanenberg 1980; Pêcher 1981; Bodnar and Bethke 1984; Gratier and Jenatton 1984; Guilhaumou et al. 1987; Prezbindowski and Larese 1987; Bodnar et al. 1989; Wanamaker and Evans 1989) or during freezing (Lawler and Crawford 1983). Other studies described inclusion reequilibration at elevated confining pressure (e.g., Poland 1982; Pêcher and Boullier 1984; Boullier et al. 1989; Sterner and Bodnar 1989; Vityk and Bodnar 1995a, b). In the studies listed above, it was generally assumed that stretching involved only a change in the inclusion volume, with no loss of fluid from the inclusion. Other workers have investigated
compositional changes and/or mass loss during fluid inclusion reequilibration, both in the laboratory and in nature (e.g., Hall and Bodnar 1990; Bakker and Jansen 1990, 1991, 1993, 1994; Hall and Sterner 1993, 1995; Cordier et al. 1994; Kotelnikova 1994; Mavrogenes and Bodnar 1994; Barker 1995; Johnson and Hollister 1995; Sterner et al. 1995; Vityk et al. 1995). These workers used the term “leakage” to describe this behavior.

In nature (and in the laboratory), fluid inclusion reequilibration at high $T$ and low loading rates might include plasticity accommodated by glide and climb, with both being assisted by water diffusion in wet quartz. These processes involve both volume change and fluid loss, suggesting that inclusion stretching and leakage are related. Consider, for example, high $P$-$T$ reequilibration of fluid inclusions under conditions of tectonic decompression. Under these conditions, the inclusions might undergo a volume increase (stretching or decrepitation) related to internal overpressure. However, diffusion of water into the surrounding quartz is probably required to initiate and facilitate the process, as dry natural quartz is virtually undeformable (Griggs and Blacie 1964, 1965). That is, dry quartz can only respond to an applied stress by fracturing – it cannot deform plastically. However, once water diffuses into the surrounding quartz, the quartz becomes ductile as a result of hydrolytic weakening, and can undergo plastic deformation much more easily (e.g., Kerrich 1976; Wilkins and Barkas 1978; Boulleier et al. 1989; Kronenberg et al. 1986; Gerretsen et al. 1993; Cordier et al. 1994). The nucleation and propagation of mobile dislocations associated with plastic deformation extracts some water from fluid inclusions (pipe diffusion along the dislocation cores) and carries this water away from inclusions under the action of differential stress (e.g., Hollister 1990; Bakker and Jansen 1990, 1991; Cordier et al. 1994). Thus, stretching of inclusion walls by dislocation multiplication must involve some loss of water from the inclusion. Diffusion of water into the surrounding quartz, in turn, enhances inclusion plasticity which promotes fluid density changes associated with stretching.

Stretching and decrepitation (or leakage) have in the past been considered to involve two different deformational mechanisms which result in different changes to the original fluid inclusion. Figure 1 is an attempt to relate the various observations, environments and mechanisms associated with these two reequilibration processes. Thus, stretching (type of reequilibration) is associated with plastic deformation (mechanism) in a low strain environment. There is generally no detectable loss of fluid during stretching (observation), except for small amounts of hydrogen which may be lost by diffusion (mechanism; cf., Mavrogenes and Bodnar 1994). Decrepitation, on the other hand, involves the complete loss of water by advection along fractures resulting from brittle deformation in a high strain environment. Figure 1 suggests that there is a continuum between stretching and decrepitation, which represent the two end-members of reequilibration behavior. Thus, an inclusion which stretches by plastic deformation in a low strain environment with no loss of fluid may begin to lose fluid (leak) along dislocations and/or microfractures if the strain rate increases. This type of reequilibration is often referred to as partial decrepitation, and the fluid loss is often detectable through microthermometric analysis (cf., Hall and Sterner 1993; Audédat and Günther 1999). The same inclusion may eventually decrepitate with loss of all fluid by explosive brittle failure if the strain rate increases further.

In order to use results from naturally reequilibrated fluid inclusions to infer $P$-$T$ histories, it is necessary to understand the mechanisms of stretching and decrepitation of fluid inclusions, and how these mechanisms operate to affect inclusion density and composition. If the inclusions fail in a brittle manner, then one should observe dry fractures around the fluid inclusions. If, however, the crystal surrounding inclusions in quartz behaves plastically, then one should observe a high density of dislocations in glide and/or climb configuration around fluid inclusions, as well as precipitation of water in the form of tiny bubbles. Among the first workers to recognize the important role of dislocations in the reequilibration of fluid inclusions were Wilkins et al. (1981), Bodnar and Bethke (1984), Sterner and Bodnar (1986), Sterner et al. (1988), Boulleier et al. (1989), Bakker and Jansen (1990) and Hollister (1990). To understand better the mechanisms operating during inclusion deformation, we have conducted a detailed TEM study of the lattice defects around reequilibrated aqueous synthetic inclusions in natural Brazilian quartz,