Do fluid inclusions in high-grade metamorphic terranes preserve peak metamorphic density during retrograde decompression?

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ABSTRACT

It is often assumed that fluid inclusions in quartz do not maintain their density if they experience internal pressures 2 kbar in excess of confining pressure. However, results of a long-term (180 d) laboratory experiment simulating a nearly isothermal decompression P-T path showed that two of 160 inclusions recorded the original density. These two inclusions thus maintained about 2 kbar internal overpressure for the duration of the experiment without modification. Comparison with results reported in the literature reveals that a small percentage of fluid inclusions in quartz from high-grade metamorphic rocks maintain peak densities during retrograde uplift. These inclusions represent outliers in homogenization temperature histograms and are often ignored during data interpretation.

INTRODUCTION

The use of fluid inclusions to reconstruct P-T histories of mineral formation has generated many questions concerning the interpretation of inclusions from metamorphic environments. One question often asked is, if fluid inclusions are trapped at peak P-T conditions in highgrade metamorphic rocks, will those inclusions maintain their original density during retrograde nonisochoric cooling? The bases for this and related questions are results of earlier experimental studies, including those by Sterner and Bodnar (1989). In that study, synthetic fluid inclusions in natural quartz were reequilibrated along a decompressional path that generated up to 1.5-1.7 kbar internal overpressure in the inclusions. The results indicated that none of the inclusions maintained the original density; all inclusions showed some density decrease and approached the final, reequilibration density. These data were interpreted by Sterner and Bodnar (1989, p. 251) to indicate that "a P-T relation inferred from inclusion microthermometry alone provides neither a maximum nor a minimum estimate of the P-T conditions attending a particular stage of metamorphism."

In this paper we present results of two laboratory reequilibration experiments using H_2O -rich synthetic fluid inclusions in natural quartz. These are referred to as the short-term experiment (7 d at final reequilibration conditions) and the long-term experiment (180 d at final reequilibration conditions). The homogenization-temperature histograms for these inclusions are compared with data reported in the literature for synthetic and natural inclusions. Although the results presented here support the general conclusions of Sterner and Bodnar (1989), they also suggest that rigorous application of their results may lead to an incorrect interpretation of inclusion data.

RESULTS

Experiments described in this paper were conducted in four steps. First, 10 wt% NaCl-H₂O inclusions were trapped in three quartz samples in three separate pressure vessels at 5 kbar and 700 °C (STEP 1, Fig. 1), using techniques described by Bodnar and Sterner (1987). After 7 d one sample was quenched isochorically, removed from the pressure vessel, and studied on a heating-freezing stage to determine the microthermometric characteristics of original inclusions that have not been reequilibrated. The remaining two samples were cooled in 1 kbar and 25 °C increments every 7 d (STEPS 2, 3, and 4, Fig. 1) from the original conditions to final reequilibration conditions of 2 kbar and 625 °C. The final conditions (STEP 4, Fig. 1) resulted in a cumulative overpressure of 2.1 kbar relative to the original isochore. At the final conditions the two samples were held for 7 and 180 d, respectively. Note that after formation the reequilibrated samples were not quenched to room temperature until the completion of the experiment.

Fluid inclusions originally formed at 700 °C and 5 kbar for 7 d homogenize at 282–285 °C with a mean value of 284 °C (Fig. 2A). Histograms showing homogenization temperatures (T_h) of 160 randomly selected fluid inclusions from the two samples subjected to decompression are shown in Figure 2B and 2C. Also shown are the T_h corresponding to the original isochore (284 °C) and the T_h (380 °C) representing the final reequilibration conditions. All the fluid inclusions in both the original and reequilibrated samples have ice-melting temperatures of -6.7 ± 0.1 °C, which corresponds to 10.0 ± 0.1 wt% NaCl. Therefore, the inclusion composition did not change during the reequilibration experiments.

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Following the short-term (7 d) decompression experi-



Fig. 1. *P*-*T* path simulating nearly isothermal decompression during reequilibration of 10 wt% NaCl-H₂O synthetic inclusions in natural quartz samples. Isochores project from the conditions of reequilibration (solid circles) to the liquid-vapor curve of 10 wt% NaCl-H₂O (open circles). The isochores for 10 wt% NaCl-H₂O solution were calculated using the equation of state of Bodnar and Vityk (1994).

ment, eight fluid inclusions maintained their original density, as evidenced by the homogenization temperature (284 °C) (Fig. 2B). About 100 inclusions (more than 60% of inclusions measured) showed T_h values within \pm 15 °C of the original T_h , suggesting that most of the inclusions were slightly modified during the experiment. Note that about 20 of these fluid inclusions exhibit T_h values lower (up to 16 °C) than the original T_h . A satisfactory explanation for this behavior is not available. However, we do not believe that the difference is a result of experimental error, as this would require temperature errors of approximately 25 °C or pressure errors of about 400 bar, both of which are much larger than errors typical for such experiments (approximately equal to \pm 1% of total range for both P and T).

The maximum T_h value (358 °C) for inclusions in the short-term experiment approximates closely the T_h of the isochore representing the final reequilibration conditions (Fig. 2B). These could be inclusions that have completely reequilibrated to the new *P*-*T* conditions, or they could be new inclusions formed at the reequilibration conditions. We are unable to distinguish between these two possibilities owing to the experimental procedure used, in which both the original and reequilibration fluid have the same composition.

After the 180 d decompression experiment, one inclusion with the original density ($T_h = 285$ °C) was found in a microfracture near the central portion of the sample. This inclusion maintained its original density, even after being subjected to 2 kbar of internal overpressure for 180 d (Fig. 2C). A second inclusion showed a T_h value within 4 °C of the original T_h . The inclusion that showed no change in T_h measured 30 × 15 µm and was regularly shaped (shape factor $\approx 6-7$ according to classification of Bodnar et al., 1989). The inclusion that showed a 4 °C change in T_h was 3 × 7 µm and also regularly shaped (shape factor 7-8). The total range in size of the 160 inclusions was from about 1 × 3 µm to 13 × 53 µm, and the two inclusions that maintained their densities are in the middle (30 × 15) to lower end (3 × 7) of this range. The shapes of both of these inclusions are classified as regular.

The inclusion with T_h (372 °C) closest to the final reequilibration T_h (380 °C) was also the largest inclusion measured (13 × 53 µm). This inclusion occurred near the edge of the quartz chip. This could be an inclusion that reequilibrated to the final *P*-*T* conditions, as Bodnar et al. (1989) and others have shown that the largest inclusions decrepitate or stretch more easily than smaller inclusions. However, it could also be a new inclusion formed at the final reequilibration *P*-*T* condition during continued fracture healing.

Most of the T_h values for the 180 d experiment lie between 308 °C (1.6 kbar of internal pressure) and 349 °C (0.7 kbar of internal pressure), with three frequency peaks and a mean value at 320 °C (Fig. 2C). The variable T_h of these inclusions suggests variable degrees of reequilibration. Peaks in the histogram possibly represent opening of interconnected portions of fractures during the reequilibration process and then rehealing to produce numerous inclusions of the same intermediate density. Some inclusions may also have formed during periods of continued fracture opening and healing during decompression.

DISCUSSION

In the 7 d decompression experiment most inclusions did not change density significantly. Our observations differ from results of Sterner and Bodnar (1989), in which no inclusions maintained the original density. In this earlier experimental study the differential pressure was applied almost instantaneously, which may have resulted in shock-induced fracturing or stretching of fluid inclusions during rapid depressurization. Also, preserved inclusions might have been overlooked in that earlier study because data were reported for only 19–38 inclusions in each sample.

Our experimental data indicate that aqueous inclusions may preserve the original density, even if the inclusions experience differences between internal and confining pressures of up to 2 kbar for as long as 180 d. This experiment was conducted at pressures (≤ 5 kbar) and temperatures (≤ 700 °C) appropriate for amphibolite and granulite metamorphism. The internal overpressure (≤ 2 kbar) is similar to overpressures commonly reported for fluid inclusions from high-grade metamorphic terranes. However, fluid inclusions in these rocks are commonly CO₂-rich, and it is well known that reequilibration behavior depends on inclusion composition. Thus, interpretation of CO₂ inclusions on the basis of reequilibration

behavior of aqueous inclusions in our experiments may not be justified. However, Hall and Wheeler (1992) showed that fluid inclusions with appreciable quantities of CO₂ are less susceptible to reequilibration than aqueous inclusions, and that high-salinity inclusions may reequilibrate more easily than those containing low-salinity fluid. These workers also suggested that preservation of CO₂-rich inclusions may be favored over aqueous inclusions in high P-T environments. A similar conclusion was also reached by Vityk et al. (1994), who showed that at elevated P and T, H_2O -rich inclusions decrepitate at conditions of about 2 kbar of internal underpressure, whereas methane-rich inclusions remain intact up to about 4 kbar of internal underpressure. These results suggest that inclusions used in our experiments should reequilibrate much more easily (i.e., at a lower internal pressure or shorter period of time) in comparison with the CO₂rich inclusions in natural samples, all else being equal.

Most $T_{\rm h}$ histograms reported in the literature for inclusions in quartz from high-grade metamorphic terranes show a characteristic pattern manifested as a broad multimodal range in $T_{\rm h}$ with a characteristic gap between the minimum $T_{\rm h}$ and the main body of $T_{\rm h}$ values as shown in Figure 2D. As in our experiment, inclusions with minimum $T_{\rm h}$ values (highest density) are the rare exception rather than the rule (only about 1-2% of all inclusions measured). For example, several secondary CO₂ inclusions in quartz segregations in enderbites from Tromoy granulites homogenize at $-27 \,^{\circ}C$ (Fig. 2D). The isochore for this $T_{\rm h}$ passes through peak (granulite) P-T conditions, although the majority of the inclusion isochores for these samples do not pass through peak conditions. Fluid inclusions in these samples all appear to be of similar age, even though their densities are highly variable.

Homogenization-temperature histograms similar to those described here are common in the geologic literature. It is obvious that significantly different P-T histories would be inferred, depending on whether the worker used the maximum, minimum, mean or mode to interpret the data. Moreover, the outliers are often ignored. Each of these temperatures has significance, although a complete

Fig. 2. Histograms of homogenization temperatures for synthetic fluid inclusions in quartz after a 7 d period at formation conditions and isochoric cooling (A), 7 d at final reequilibration conditions (**B**), and 180 d at final reequilibration conditions (**C**). For comparison, a histogram of homogenization temperature (modified from van den Kerkhof et al., 1994) for CO₂ inclusions from Tromoy granulites, Proterozoic Bamble sector of southern Norway, is shown (**D**). Calculated internal pressures of fluid inclusions, corresponding to the measured $T_{\rm h}$, are shown along the top of each histogram. For inclusions from Tromoy granulites the internal pressures were estimated from a retrograde *P*-*T* fluid path and isochores corresponding to measured $T_{\rm h}$ of -27, +2, and +31 °C at 300 °C (see Fig. 6 in van den Kerkhof et al., 1994).



understanding of the relationship between histogram characteristics and P-T history is lacking.

CONCLUDING REMARKS

We realize that we can never experimentally simulate slow, natural loading rates. However, in this study we attempted to minimize the introduction of shock-induced inclusion modification. By changing the confining pressure in our experiments in 0.7 kbar increments over several weeks' duration, we found that a large percentage of aqueous synthetic inclusions in quartz maintained 2 kbar of internal overpressure for 7 d without changing volume. These results differ from previous experimental studies involving instantaneous loading at conditions of high internal overpressure, in which inclusions maintaining their original density were not reported.

Our experimental data further indicate that a small percentage of aqueous synthetic inclusions preserve their original density, even when subjected to internal overpressures of about 2 kbar for as long as 180 d. On the basis of these results and in comparison with naturally modified inclusions, we suggest that fluid inclusion data from high-grade metamorphic rocks may be interpreted to indicate that some of the inclusions maintained peak metamorphic densities.

We recognize that for some high-grade metamorphic terranes none of the inclusion isochores passes through peak metamorphic conditions. This behavior is presumably related to the very high internal overpressures that occurred after inclusion entrapment. This might also reflect an insufficient number of T_h measurements to identify the high-density inclusions or limited sample selection. Note that our experimental results, as well as data from natural samples, indicate that only about 1–2% of the inclusions record original densities.

An important extension of the work reported here is to develop criteria for recognizing inclusions most likely to maintain peak densities in natural samples. In our study, the inclusions that maintained the original density do not appear to be significantly different from the others. One did, however, occur in the interior of the sample. Whether this is significant or simply coincidental is unknown. Similarly, the inclusion that most closely approximated the final conditions was found near the edge of the sample. Careful reading of the literature provided little or no information concerning the distribution and textural characteristics of the highest density inclusions in natural samples. It is clear, however, that detailed petrographic studies and reexamination of samples that record peak densities are needed. Perhaps more of the P-T history of the rocks is recorded by fluid inclusions than we realize; we just have not yet learned to read the rock language.

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