Fluid immiscibility at metamorphic conditions: experimental evidence of fluid inclusions

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The important role of fluids in geological processes is undoubtedly proven by the presence of fluids in abundant rock types, from sedimentary, metamorphic, igneous to extra-terrestrial rock. Metamorphic reactions, melting behaviour, alterations, weathering, ore generation are just a few of those processes that are affected or triggered by the presence of fluid phases. One aspect of fluid research is the knowledge of fluid immiscibility, even at higher temperatures and pressures. Immiscibility of low-density and high-density fluid phases may cause fractionation of economically relevant metals that are dissolved in these fluid phases. Temperature-pressure-composition-density properties of fluid-immiscibility fields are only roughly defined in literature, and lack any thorough thermodynamical modelling. Whereas unary fluid systems (e.g. pure H_2O) have well defined fluid immiscibility conditions (liquid-vapour curve), already binary fluid systems are lacking sufficient experimental data and thermodynamic models. One exception: the most-studied fluid system is H_2O-NaCl, and its fluid immiscibility conditions are well known. An important question that arises: can immiscible fluids be preserved in fluid inclusions? The present work presents a first attempt to investigate synthetic fluid inclusions in quartz that were trapped in the presence of heterogeneous fluids, i.e. immiscible liquid-rich and vapour-rich H_2O-NaCl fluids. The capability of doing so is a prerequisite for a correct interpretation of naturally occurring fluid inclusion assemblages. The experimental conditions (T, P) are selected to obtain equal amounts of 2.41 mass% NaCl vapour-rich fluid (58.51 cm^3/mol) and 41.40 mass% NaCl liquid-rich fluid (29.34 cm^3/mol), that is obtained from on original 30.07 mass% NaCl solution (loading conditions). Both fluids can be trapped simultaneously in small cracks within quartz crystal. The first results illustrate that these fluids are not preserved within newly formed synthetic fluid inclusions. Inclusions with relatively large vapour bubbles contain the original 30 mass% NaCl solution, which was trapped before the immiscibility field was reached in the experiment. The liquid-rich inclusions reveal properties that resulted from the mechanically mixing of little salt crystals with the liquid phase. In conclusion, narrow cracks in quartz crystal may heal instantaneously, within one hour in an experimental heating run from room temperature to 600 ˚C. At the final experimental conditions only traces of the liquid-rich fluid are trapped in the remaining cracks, which may be coated with small NaCl crystals, that formed during the unmixing process. The vapour-rich fluid is not trapped, due to its wetting properties, which prevent entering of small bubbles into narrow cracks. These cracks are completely wetted with the liquid-rich phase. There is no reason to assume that the crack healing process in the experiment is different from natural processes. These results put some restrictions on recognizing heterogeneous entrapment processes in natural rock.