



## Invited review

# Supercritical fluids at subduction zones: Evidence, formation condition, and physicochemical properties



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## ABSTRACT

There is a general consensus that at subduction zones, mass transfer from the subducted slab to the overlying mantle wedge is mediated by a hydrous mobile phase. However, it is under intense debate whether this phase is an aqueous fluid, hydrous silicate melt, or supercritical fluid with intermediate composition (H<sub>2</sub>O concentration in the range of 30 wt%–70 wt%). Supercritical fluids, with fluid-like viscosity and melt-like wetting and element-carrying capability, are an ideal agent for chemical transport at subduction zones.

After clarifying the phase relations of silicate-H<sub>2</sub>O systems and the definition of supercritical fluids, this contribution reviews existing evidence for the presence of supercritical fluids at subduction zones, mostly from experimental investigation of phase relations of mineral-H<sub>2</sub>O and rock-H<sub>2</sub>O systems and the atomic structure and physicochemical properties of supercritical fluids. H<sub>2</sub>O-rich multi-phase solid inclusions from continental subduction zones, once their bulk water contents can be confirmed, may provide a direct record of supercritical fluids. Experimental results generally indicate that supercritical fluids can derive from the slab at 160 km depth, but there is still significant discrepancy between different studies on H<sub>2</sub>O-saturated solidus, fluid-melt critical curve, and the second critical end point (SCEP). Some novel experimental methods are proposed to resolve the controversies over the formation condition of supercritical fluids.

The special physicochemical properties of supercritical fluids arise fundamentally from their intermediate composition and intermediate degree of polymerization that are distinctive from aqueous fluids or silicate melts. Probing the structure and properties of supercritical fluids, which are required for understanding of their petrological and chemical behavior and geophysical characteristics, relies mainly on in situ spectroscopy, but first-principles molecular dynamics is becoming a potentially powerful alternative approach.

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## Contents

|   |    |
|---|----|
| 1. Introduction . . . . .   | 63 |
| 2. What is a supercritical fluid? . . . . .                               | 63 |
| 2.1. One-component system: H <sub>2</sub> O . . . . .                     | 63 |
| 2.2. Binary mineral-H <sub>2</sub> O system . . . . .                     | 63 |
| 2.3. Rock-H <sub>2</sub> O system . . . . .                               | 64 |
| 2.4. Common misconceptions about “critical” and “supercritical” . . . . . | 65 |
| 2.5. The definition of supercritical fluids . . . . .                     | 65 |
| 3. Evidence for supercritical fluids. . . . .                             | 65 |
| 3.1. Mineral solubility in water. . . . .                                 | 65 |
| 3.2. Water solubility in silicate melts . . . . .                         | 66 |
| 3.3. In situ observation of critical behavior . . . . .                   | 66 |
| 3.4. Termination of wet solidus . . . . .                                 | 66 |
| 3.5. Polymerization of fluid structure . . . . .                          | 66 |
| 3.6. Properties of supercritical fluids . . . . .                         | 67 |

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|--|----|
| 3.7. Records from continental subduction zones . . . . .           | 67 |
| 4. The unresolved issues . . . . .                                 | 67 |
| 4.1. Lack of robust record from fluid inclusions . . . . .         | 67 |
| 4.2. Controversies over the formation condition . . . . .          | 68 |
| 4.3. Inadequate knowledge about structure and properties . . . . . | 69 |
| 5. Concluding remarks . . . . .                                    | 69 |
| Acknowledgements . . . . .   | 70 |
| References . . . . .   | 70 |

## 1. Introduction

Subduction zones beneath convergent boundaries are characterized by a highly dynamic tectonic environment, where intensive crust-mantle interaction gives rise to earthquakes, magmatism, the destruction of oceanic crust, and eventually to the formation of new continental crust (Stern, 2002). The “subduction factory” involves a variety of physical and chemical changes that impact profoundly on the evolution of the Earth, global recycling of elements, and the genesis of ore deposits (Tatsumi and Kogiso, 2002; Hacker et al., 2003; Tatsumi and Stern, 2006; Sun et al., 2014). The trace element pattern and the hydrous nature of arc magmas, the output, are inferred to be inherited from the subducted slab, the input of the “subduction factory” (Bebout, 2014). Geochemical, geophysical and experimental studies reach the consensus that mass transfer from the subducted slab to the overlying mantle wedge is mediated by a hydrous mobile phase (Scambelluri and Philippot, 2001; Poli and Schmidt, 2002; Manning, 2004; Kessel et al., 2005a, 2005b; Hermann et al., 2006; Pommier, 2014; Frezzotti and Ferrando, 2015; Kawamoto et al., 2015). However, considerable controversy has arisen over the exact nature of that fluid (Manning, 2004; Mibe et al., 2011; Kawamoto et al., 2012; Schmidt and Poli, 2014).

Within the forearc depth range (e.g., <80 km), the slab-derived fluid is found to be generally “dilute”, with solute concentration typically <15 wt%, as inferred from fluid/melt inclusions (Gao and Klemd, 2001; Kawamoto et al., 2013; Ribeiro et al., 2015), experimental investigation (Schneider and Egger, 1986; Brenan et al., 1995; Adam et al., 2014), and thermodynamic modeling (Manning, 1998, 2013; Dolejs and Manning, 2010). Even down to subarc depths (e.g., 80–130 km), the alkali aluminosilicate-dominated solute load could still lie below 30 wt% (Mibe et al., 2002; Kessel et al., 2005b; Dvir et al., 2011; Frezzotti and Ferrando, 2015). Such a fluid is normally referred to as aqueous fluid, aqueous solution, or vapor (Hermann et al., 2006; Zheng and Hermann, 2014; Green et al., 2014).

On the other hand, at hot subduction zones with young oceanic plates subducting, the sediments and basaltic crust atop the slab may undergo melting under water-saturated conditions and generate adakitic magma (Defant and Drummond, 1990; Prouteau et al., 2001; Mibe et al., 2011). The produced melts are inescapably hydrous since water is a highly incompatible component, but the concentration of silicate components is generally higher than 70 wt%, in contrast with “dilute” aqueous fluids.

Besides the above two scenarios, under the temperature and pressure conditions corresponding to deep subduction zones (i.e., subarc to backarc depths), the miscibility gap between aqueous fluid and silicate melt diminishes and may even close completely, allowing for the formation of a special fluid with intermediate composition – the concentrations of H<sub>2</sub>O and the silicate-dominated solute both fall into the range of 30 wt% to 70 wt%. In general, such a transitional fluid has to be formed at *P-T* conditions above the second critical end point of the given silicate-H<sub>2</sub>O system and is, therefore, often referred to as supercritical fluid (Manning, 2004; Hack et al., 2007; Sanchez-Valle, 2013).

In this contribution, we will first clarify the concept of supercritical fluids, and then review existing evidence for their possible presence in subduction zone setting. Most importantly, we will point out future directions that may settle the controversies over the presence and

formation condition of supercritical fluids and advance more precise understanding of their atomic structure and physicochemical properties.

## 2. What is a supercritical fluid?

The popular term “supercritical fluid” has been criticized by some authors (e.g., Liebscher and Heinrich, 2007) for causing a great deal of confusion in geosciences. Here we choose to keep this term for the lack of a better alternative. A detailed account of the phase relations involving fluids, melts and minerals is available in Manning (2004) and Hack et al. (2007). Below, we will use a few representative phase diagrams to illustrate what a supercritical fluid really means in simple and complex systems and to clear out some common misconceptions about supercritical fluid.

### 2.1. One-component system: H<sub>2</sub>O

For a one-component system exemplified by pure H<sub>2</sub>O, the definition of a supercritical fluid is unambiguous. Along the boiling curve, the density contrast between coexisting liquid water and vapor steam becomes less sharp toward higher *P-T* and eventually disappears at the critical point ( $T_c = 647$  K and  $P_c = 22$  MPa) terminating the boiling curve (Fig. 1a). Beyond the critical point there is no phase boundary between water and steam, giving rise to a supercritical fluid. Supercritical H<sub>2</sub>O bears some special physicochemical properties, such as low viscosity (like steam) and strong dissolving capability (like water). This phenomenon arises fundamentally from the fact that the density or intermolecular distance of supercritical H<sub>2</sub>O is intermediate between the commonly perceived “compact” water and “tenuous” steam.

It is also crucial to distinguish two different processes – dissolution and critical phenomenon – that can both lead to the homogenization of two fluid phases. Consider isochoric heating of H<sub>2</sub>O (Fig. 1b). If the bulk fluid density is above 0.322 g/cm<sup>3</sup>, the critical density  $\rho_c$ , the steam phase will gradually dissolve into water; below 0.322 g/cm<sup>3</sup>, water will dissolve into steam. But when the bulk density is close to  $\rho_c$ , neither water nor steam will be consumed significantly. Instead, the physical boundary between the two phases will become progressively indistinct in response to reduced density contrast. Across the critical point, the two phases of subequal amount will homogenize into a supercritical fluid by erasing the phase boundary completely and produce a critical phenomenon.

### 2.2. Binary mineral-H<sub>2</sub>O system

The situation is more complicated for silicate-H<sub>2</sub>O as a binary or multicomponent system. Here the concerned two mobile phases are an aqueous fluid and a hydrous silicate melt, which are different not only in density but also in composition. Instead of being fixed at a single *P-T*, critical point becomes univariant. For the albite-H<sub>2</sub>O binary system (Fig. 2a), for each given pressure there is a corresponding critical point (maximum of the fluid-melt solvus) at a specific temperature and a specific intermediate composition. Above the critical temperature, the miscibility gap between aqueous fluid and hydrous melt is closed.

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