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A revised empirical model to calculate the dynamic viscosity of H_2O–NaCl fluids at elevated temperatures and pressures ($\leq$1000 °C, $\leq$500 MPa, 0–100 wt % NaCl)

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ABSTRACT

Examination of viscosities for fluids in the system H_2O–NaCl predicted by the commonly-used model of Palliser and McKibbin has identified regions of pressure-temperature-salinity (*PTx*) space in which the model delivers values that are inconsistent with some experimental data and exhibits discontinuities and trends that are unexpected. Here, we describe a revised empirical model to calculate viscosity of H_2O–NaCl fluids that shows good correlation with experimental values and shows trends that are consistent with known or expected behavior outside of the region where experimental data are available. The model described here is valid over the temperature range from the H_2O solidus (~0 °C) to ~1000 °C, from ~0.1 MPa to $\leq$500 MPa, and for salinities from 0 to 100 wt % NaCl.

1. Introduction

The circulation of fluids in the Earth's crust plays an important role in the transport of chemical constituents (mass) and heat (energy), and is the major control on the formation of hydrothermal ore deposits [6]. In recent decades, our understanding of the underlying principles associated with fluid circulation in response to thermal perturbations in the crust has advanced significantly through application of numerical fluid flow models. These models incorporate the transport equations that describe conservation of mass, energy and momentum of the system, and require as input the physical and thermodynamic properties of the circulating fluid. Early numerical simulations incorporated the properties of pure H_2O to simulate the natural hydrothermal fluid. For example, the properties of H_2O were used to approximate the hydrothermal fluid in systems associated with shallow continental plutons [7,28–30], submarine hydrothermal systems [12,13,18,19], and sedimentary environments [8,14,47]. While most hydrothermal fluids are indeed H_2O-rich, most also contain varying amounts of salts such as NaCl, KCl, $CaCl_2$, $FeCl_2$, etc., or volatiles such as CO_2 and CH_4 [5,16,37,48].

While the one-component system H_2O is a reasonable approximation for the composition of many low-salinity aqueous hydrothermal fluids, the fact that H_2O is a one-component system results in a phase topology that differs significantly from that of multi-component fluids. An important difference in phase topology of the one component H_2O system, compared to multi-component fluid systems, is that in a one component system liquid and vapor only coexist along the liquid-vapor equilibrium curve that terminates at a critical end point (Fig. 1a; C.P.), beyond which only a single-phase (supercritical) fluid may exist. Additionally, at temperatures above the critical temperature in a one-component system such as H_2O, all physical and thermodynamic properties of the fluid vary smoothly and continuously with changing temperature and pressure, and no discontinuities in fluid properties occur. Over the past several decades, a large experimental database describing the Pressure-Volume-Temperature-Composition (*PVTx*) properties of aqueous electrolyte systems has been generated, especially for the system H_2O–NaCl [3,4,23,39]. A complete summary of available experimental data thorugh 2008 is available in Ref. [42]. The available experimental data have been used to develop algorithms to estimate the physical and thermodynamic properties of more complex fluids at high temperatures and pressures [2,10,11,32–34,41]. These various algorithms have subsequently been incorporated into numerical fluid flow models [25,46] that describe the circulation of hydrothermal fluids in the crust.

One of the basic physical properties required to model fluid flow

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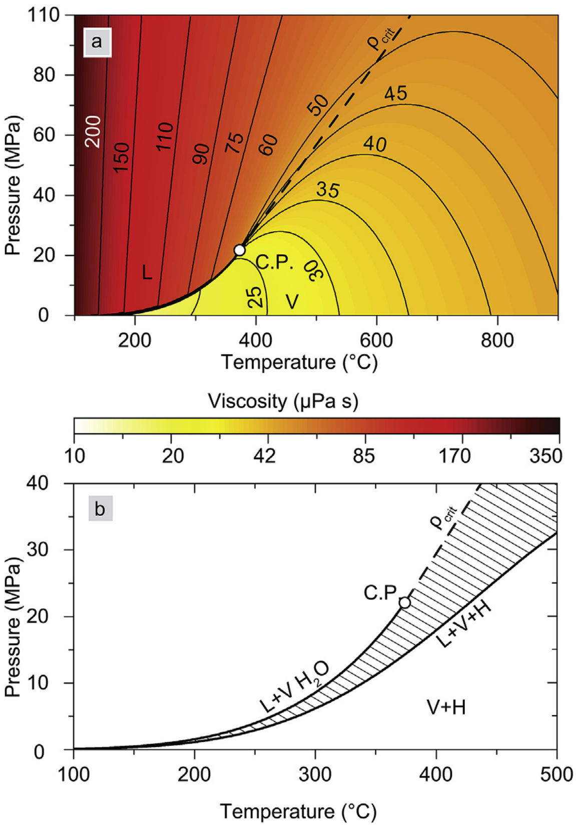

Fig. 1. (a) Viscosity of pure H_2O in the *PT* range 0–100 MPa and 100–900 °C calculated using the [17] model. C.P. is the critical point (373.946 °C, 22.064 MPa), L refers to the liquid phase (density greater than the critical density), V refers to the vapor phase (density less than the critical density), and ρ_{crit} indicates the critical isochore. (b) Pressure-Temperature plot showing the liquid-vapor coexistence curve of pure water (L+V H_2O) that terminates at the critical point (C.P.) and the critical isochore for H_2O (dashed line labeled ρ_{crit}). Also shown is the vapor + liquid + halite equilibrium curve (L+V+H) for H_2O–NaCl, and the field in which an H_2O–NaCl vapor phase and halite are in equilibrium (V+H). The hatched region defined by pressures less than the H_2O liquid-vapor curve and pressure along the critical isochore, and pressures greater than the pressure on the L+V+H curve identifies the region of *PT* space in which the Palliser & McKibbin model may predict viscosities that are too low owing to the fact that in this *PT* region H_2O is a vapor (density less than the critical density) whereas many H_2O–NaCl compositions are liquid in this *PT* region.

is the fluid viscosity. In most early models, the viscosity of pure H_2O was used to approximate the viscosity of the natural fluid. In the liquid region, the viscosity of pure H_2O decreases smoothly and continuously with increasing temperature at constant pressure above the critical pressure (22.064 MPa), up to approximately the *PT* conditions along the critical isochore (Fig. 1a). With increasing temperature above the critical isochore, the viscosity shows a reversal and increases with increasing temperature at *PT* conditions to the low-pressure side of the critical isochore, i.e., in the vapor field (Fig. 1a). While the viscosity of pure H_2O provides a reasonable approximation for low salinity fluids and at *PT* conditions that are in the liquid field for pure H_2O (i.e., at pressure greater than the pressure on the liquid-vapor curve, or greater than the pressure on the critical isochore for a given temperature), the viscosity of pure H_2O is a poor analog for the viscosity of more saline fluids, especially at *PT* conditions in the vicinity of the pure H_2O liquid-vapor curve and at *PT* conditions that would be in the vapor field for pure H_2O.

The dynamic viscosity (η – Pa·s) is included in the expression for conservation of momentum associated with fluid flow. The conservation of momentum can be defined as [31]:

$$\vec{v} = -\frac{k\rho}{\eta}(\nabla P - \rho\vec{g}) \tag{1}$$

where $\vec{v}$ is a vector quantity describing mass flow rate per unit area ($kg \cdot m^{-2} \cdot s^{-1}$), k is the permeability (m^2), ρ is the fluid density ($kg \cdot m^{-3}$), η is the dynamic viscosity (Pa·s), ∇P is the pressure gradient and $\vec{g}$ is the standard acceleration due to gravity ($9.81\ m\ s^{-2}$).

Numerical models describing the viscosity for pure H_2O were developed within the International Association for the Properties of Steam (IAPS), which in 1989 became the International Association for the Properties of Water and Steam (IAPWS). The viscosity of H_2O established in 1964 was initially presented as a set of skeleton tables, together with recommended interpolation equations by [22]. Later numerical models for the viscosity of H_2O included revisions to improve the accuracy of predictions [20] and to better estimate the asymptotic divergent behavior of the viscosity near the critical point of water [45] (see Fig. 1a). In the latest release of the IAPWS formulation 2008, the viscosity of pure H_2O is based on [17] and replaces the IAPS formulation 1985 for the Viscosity of Ordinary Water Substance issued in 2003 [9]. The IAPWS 2008 formulation is based on the density of H_2O calculated by the IAPWS-95 model of water and steam properties developed by [44]. The viscosity of pure H_2O in the IAPWS formulation 2008 has estimated uncertainties between 1 and 7%, and is limited in its *PT* range of applicability by the solidus of H_2O at low temperature (~0.0 °C), an upper temperature limit of 900 °C and an upper pressure limit varying from 1000 MPa at lower temperatures to 300 MPa at higher temperatures. The viscosity of H_2O predicted by the IAPWS formulation 2008 is shown on Fig. 1a.

The most commonly used model to calculate the viscosity of H_2O–NaCl at elevated *PT* conditions is that of Palliser & McKibbin [34]. During our earlier studies to investigate fluid flow in submarine hydrothermal systems [40,49], we observed unexpected behavior that could be traced back to viscosities predicted by the [34] model. This motivated us to investigate further, leading to the

Table 1
Summary of experimental data for the viscosity of H_2O and H_2O–NaCl fluids.

Salinity (wt % NaCl)	Temperature (°C)	Pressure (bars)	Method	Uncertainty (%)	Number of data points	Reference
0.03–27.5	0–154	1–389.8	*n.p.*	*n.p.*	1161	[55]
0–6.8	0–300	100–1500	CAP	2	110	[38]
0–24.7	10–40	1–311.6	OSD	0.3	166	[53]
1–20	20–350	2–300	CAP	0.3	186	[35]
2.7–24	18–154	1–311.6	OSD	0.3	189	[52]
0.6–6.5	5–95	1	Ub	0.02	60	[54]
5.9–26.1	24–201.5	1–321	OSD	0.2	381	[21]
0.1–26	20	1	*n.p.*	*n.p.*	23	[50]
100	800–975	1	OSC	2.4	40	[51]

OSD: oscillating disc; Ub: Ubbelohde; CAP: capillary; n.p: not provided.