

Fused silica capillary capsule as a hydrogen fugacity sensor in hydrothermal experiments below 400 °C

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Fused silica capillary capsule (FSCC; 0.38 mm OD, 0.1 mm ID and 1 to 2 mm long; [1]) has been used to measure hydrogen pressure (P_{H_2}) at temperatures (T_s) below 400 °C in (1) a cold-sealed pressure vessel equilibrated under P_{H_2} between 0.1 and 0.5 MPa; and (2) a gold capsule containing an oxygen buffer and H₂O under 100 MPa Ar external pressure. The P_{H_2} in the quenched FSCC was measured quantitatively at room T using Raman spectroscopy. The relationship between H₂ Raman peak height near 4156 cm⁻¹ and P_{H_2} , which was prepared in a high-pressure optical cell [2], was first established at room T for our Raman system (Horiba Revolution model). Based on this relationship and the Raman peak height of H₂ collected from the quenched FSCC, the P_{H_2} of the sample was then determined. Our results at 250 °C indicate that it requires 24 hours or less to reach osmotic equilibrium with about 2% P_{H_2} uncertainties for 0.5 MPa H₂ pressure, and that about 3 days are needed to obtain a steady state P_{H_2} reading for each oxygen buffers, including Ni-NiO, Co-CoO, Fe-Fe₃O₄, and WO₂-WO₃.

By using the isopiestic method to control the redox state of their samples in hydrothermal experiments at low T 's (≤ 400 °C), Timofeev et al. [3] identified a new uranium chloride species (UCl₄^o) and claimed that it is more stable under reducing than oxidized conditions. For each experiment, their sample solution was exposed to an oxygen buffer through the vapor phase without involving diffusion of H₂ through a membrane. Even though, this is an excellent experimental method, verification of the actual redox state of their sample will further support their conclusions, and the use a FSCC redox sensor, exposed directly to the vapor phase, will be ideal. Furthermore, the FSCC redox sensor has a great potential in hydrothermal experiments for the systems containing hydrocarbons and sulfur at T 's below 400 °C.

[1] Chou et al. (2008) *Geochim. Cosmochim. Acta*, **72**, 5217-5231. [2] Chou (2012) *EMU Notes in Mineral.*, **12**, 227-247. [3] Timofeev et al. (2018) *Nature Commun.* **9**:1469
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