Water in opals : new insights from thermal analysis

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Opal is a variety of natural hydrous amorphous silica that may contain significant amount of water (up to 21% wt). Although the speciation of water can be identified in part using vibrational spectroscopy, its location and configuration in the microstructure remain poorly understood. Thermal analyses (Differential Scanning Calorimetry, DSC; Thermogravimetric Analysis, TGA) were carried out on a wide sampling of well characterized opals (30, both opal-A and opal-CT [2]). Using DSC (from -85°C to 30°C), the exo- and endothermic peak area (during freezing and melting respectively) can be used to calculate the amount of water that may crystallize ("free water", as opposed to bonded water)[3]. Combined with TGA, which measures the total water content, we determine the percentage of crystallizable water compared to the total water content. In addition, DSC analysis can be used to estimate the pore size and size distribution of the pores containing this water.

DSC analysis reveals that our samples contain 0.3%wt to 4,4 %wt of crystallizable water. Our opals having a water content ranges from 3%wt to 18%wt (measured by TGA), we determine that 5% to 50% of this water is crystallizable, suggesting that the remaining water is either trapped in silica cages, adsorbed or bonded in some way. Moreover, the peaks observed display a wide range of shape and width, implying that opals have multiple pore size distribution (i.e discrete size distribution involving narrow peaks).

Thermal analyses indicate that the amount of crystallizable water varies from sample to sample. This suggest that molecular water is included in diverse types of pores. The mobility of water likely plays a role in the stability of opal, resulting in cracking and/or whitening. This investigation brings new evidences to the understanding of the mobility of water and its control by the microstructure, and subsequently, the opal stability.

[2] Chauviré *et al.* (2017) *Eur. J. Mineral.* **29**. [3] Thomas *et al.* (2013) *J. Therm. Anal. Calorim.* **113**, 1255-1260.

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