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New insights on opal destabilization from thermal, optical and spectroscopy analysis.

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Opal is a gemstone that may suffer of instability: with time, some specimens may crack (development of fractures) and/or whiten (decrease of transparency), when many others remain unchanged with time (Aguilar-Reyes, 2004; Rondeau et al., 2011). Destabilization process strongly affects the marketability. The stability of a given specimen is barely predictable, making opal a suspicious gemstone. Scientific literature on opal stability is restricted to its description and concludes to a link between destabilization and the release of water and/or a change in its speciation (Pearson, 1985; Paris et al., 2007). To investigate which parameters primarily control instability, a comprehensive set of opals from various origins and structures has been selected. Susceptibility to cracking and/or whitening has been checked based on heating experiments.

Thermogravimetric analyses show that most samples do not lose any weight until $200-300^{\circ}$ C, followed by a sudden release of water. In these samples, cracking usually occurs between $200-300^{\circ}$ C, simultaneously with the water loss. By contrast, other samples either lose gradually their water up to 1000° C, or dehydrates totally before 150° C.

Before cracking, opal is isotropic under crossed polars but shows, after cracking, anisotropic strain-related bands following the cracks network. In addition, Raman bands are characterized by slight shift in the Raman modes, consistent with strain in the material (Tallant et al., 1988; Malchow et al., 2015). Hence, Raman spectroscopy and optical microscopy both demonstrate that cracking induces strain in opal.

Water-filled pore volumes were measured using thermoporosimetry, based on differential scanning calorimetry. We observed that analyzed opal contains water only in nanometric pores (from 1 to 20 nm).

We propose that cracking may develop with desiccation of silica constituting opal. Actually, the decrease of volume during dehydration induces strain in silica. An assumption to explain whitening was that empty larges pores enhance light scattering and subsequently decrease the transparency. However, samples whiten even though filled pores are too small to be involved in whitening by light scattering. Further investigations on changes in opal during destabilization are required to better constrain the parameters involved.

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