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Origin of silica and fingerprinting of Australian sedimentary opals

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ABSTRACT

Opal is Australia's national gemstone with a significant fraction of the global supply mined from highly weathered Cretaceous sedimentary rocks within the Great Artesian Basin. Surprisingly, relatively little is known about the petrography and trace elemental composition of opal and its host rocks and consequently about the source of silica that underpins its formation. Using laser ablation-inductively coupled plasma mass spectrometry (LA-ICP-MS) of precious and common opal from key opal mining areas in the Great Artesian Basin coupled with multivariate analyses of 59 detectable elements in opal, we show that a mining region from which an opal originates can be constrained by using a combination of Hf, Ba, Zr and Gd with a high degree of confidence. Likewise, precious opal can be distinguished from common (non-precious opal) using a combination of Bi, Ta, Sn and Ca as these particular elements are especially low in concentration in precious opal. Although the opal from the Great Artesian Basin is sedimentary, the Ba content of opals from the eastern part of the basin suggests a volcanic origin. The most likely source of Ba and hence of silica for these opals are feldspars, now altered to kaolinite, sourced as volcanoclastic sediment from the Cretaceous Whitsunday Volcanic Province that marked the rifting and breakup of eastern Gondwana. The alteration of detrital feldspars to kaolinite and their replacement by void-filling opal confirms that weathering has played a critical role in the formation of Australian opal. The opal host rocks are severely weathered with a chemical index of alteration (CIA) up to 92. For the majority of opals studied, the silica is most likely derived locally from the opal host rocks, which impart a unique elemental signature on the opal at any particular locality. Mintabie opal, however, has very low Zr/Hf ratio, which is decoupled from its host rock, suggesting that the silica source is different from all the other opals, or that the silica fluid has experienced intense trace element fractionation, or both. The combination of analytical and statistical methods used here provides a powerful tool for a wide range of provenance studies, not just gemstones, where relationships between a large number of major and trace elements are difficult to unravel.

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1. Introduction

Geochemical fingerprinting is critical for the correct identification of the origin of precious gemstones and plays an important part in the prevention of trade in fake and illegal stones, the valuation process as well as in understanding the geological processes under which the gemstones formed. Major and trace elements are known to impart a distinctive chemical signature for many gemstones and their concentrations have been successfully used in studies of diamonds and sapphires (e.g., [Guillong and Günther, 2001](#); [Resano et al., 2003](#)) but never for opal. Opal consists of amorphous $\text{SiO}_2 \cdot n\text{H}_2\text{O}$ comprising silica spheres typically ranging from 150 to 400 nm in diameter ([Jones et al., 1964](#); [Sanders, 1964](#)), which in precious opal are of similar size and form an ordered network allowing light to diffract into a striking array of rainbow colours ([Sanders, 1964](#)) known as play-of-colour. Common opal, which is intimately associated with precious opal, has the same composition but is composed of randomly arranged silica spheres of variable sizes ([Sanders, 1964](#)) and is usually dull white, grey or brown in colour.

Opal also contains 1–9 wt.% of water ([Segnit et al., 1965](#)) and trace amounts of “impurities” such as Al, Fe and Ti ([Gaillou et al., 2008](#)) which are incorporated in the opal by substitution of tetravalent silicon species or as submicroscopic mineral grains during the opal formation process ([Erel et al., 2003](#)). Opal is found in sedimentary and volcanic environments ([Smallwood et al., 2008](#)) where it forms under low temperature ([Iler, 1979](#)) and is commonly referred to as ‘sedimentary opal’ and ‘volcanic opal’. Sedimentary opal is classified as opal-A and volcanic opal is classified as opal-CT based on X-ray diffraction analysis. Other major differences include the degree of crystallinity, water content and density with sedimentary opals being amorphous and containing 4–9% water compared to the slightly crystalline volcanic opals in which the water content is generally 9–18% ([Smallwood et al., 2008](#)). The formation of non-opalescent opal-A has also been documented from hot spring environments (e.g., [Akahane et al., 2004](#)). Virtually all opal mined in Australia is sedimentary opal-A ([Smallwood et al., 2008](#)) with Australia supplying over 95% of the world's precious opal, which to this day is mined privately by minor operators. The opal occurs almost exclusively within Cretaceous sedimentary rocks of the Great Artesian Basin and is mined from a small number of locations ([Fig. 1](#)). However, despite a long history of mining and a well-constrained geological

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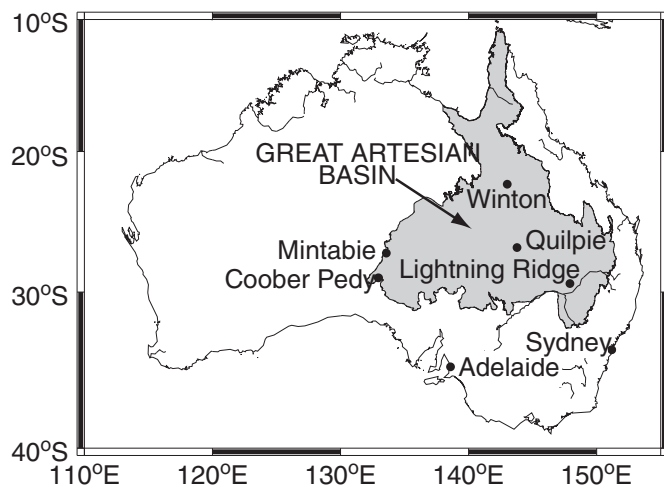


Fig. 1. Present-day extent of the Great Artesian Basin and major opal mining townships.

history of the basin, the formation of sedimentary opal and its uniqueness to the Australian continent are poorly understood (e.g., Merdith et al., 2013). Although the opal has been studied petrographically (Gallacher, 2001), very little is known about the petrology of its host rocks. Likewise the elemental composition of Australian sedimentary opals is poorly documented with only three studies based on inductively coupled plasma mass spectrometry (ICP-MS) conducted to date (Gallacher, 2001; Brown et al., 2004; Gaillou et al., 2008). Here, we have used laser ablation ICP-MS (LA-ICP-MS) on precious and common opal from key opal mining areas in order to constrain the possible sources of silica involved in opal genesis (a critical aspect of opal formation) and to assess whether any elements could be used to determine the provenance of opal with respect to a particular mining area. This approach had proven extremely useful for the fingerprinting of sapphires and diamonds (e.g., Resano et al., 2003) but until now has never been applied to opals.

2. Opal and its host rocks

The Cretaceous sediments, which comprise the host rocks for the opal in the Great Artesian Basin, were deposited from ca. 125 Ma to 95 Ma when an epicontinental sea flooded central Australia (Frakes et al., 1987; Campbell and Haig, 1999) as the Australian plate was moving eastward over a west-dipping subduction zone (Fig. 2) (Matthews et al., 2011). A silicic-dominated large igneous province comprising the Whitsunday volcanics, developed along the eastern Australian margin ca. 132–95 Ma (Fig. 2) marking the onset of continental break-up in eastern Gondwana (Bryan et al., 2000). Much of the volcanogenic material is now preserved within marine and fluvial Cretaceous sedimentary rocks in the Great Artesian Basin and is intimately associated with opal (Fig. 3). The basin experienced a major period of uplift between 100 and 65 Ma with erosion removing a package of sedimentary rock up to 3 km thick (e.g., Raza et al., 2009). Intense weathering resulted in extensive silicification within the Tertiary regolith (Thiry et al., 2006). In Winton and Quilpie (Fig. 1) the opal occurs within the late Albian to Cenomanian fluvial, volcanoclastic Winton Formation (Fig. 3) (Exon and Senior, 1976), which is red–brown ironstone, dominated by goethite and kaolinite mineralogy with minor quartz and illite (Table 1); in Lightning Ridge within late Albian kaolinite-rich (Table 1) Finch Claystone deposited in a flood plain environment; in Coober Pedy within the early Cretaceous kaolinite-rich (Table 1), marine Bulldog Shale (Fig. 3) (Exon and Senior, 1976); in Mintabie within Ordovician fluviodeltatic sandstones of the Mintabie Beds (Fig. 3) (Barnes et al., 1992), the only field that has produced opal from Paleozoic rocks. The mineralogy of the Bulldog Shale and Finch Claystone is broadly similar

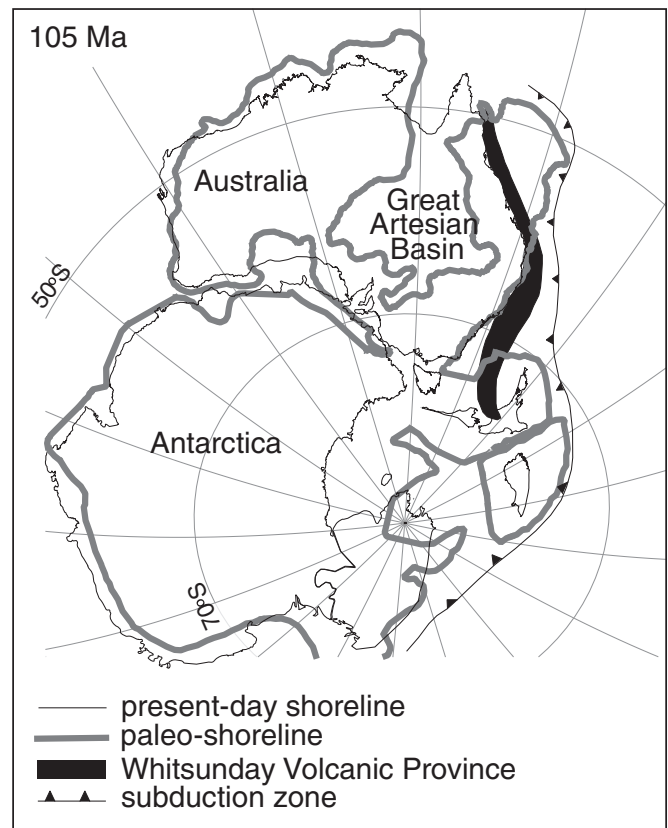


Fig. 2. Configuration of the Australian plate at 105 Ma showing the position of the paleo-shorelines (Golonka, 2007) and silicic large igneous province cluster comprising the Whitsunday Volcanic Province (Bryan and Ernst, 2008).

in that it is dominated by kaolinite, followed by quartz with smectite and illite comprising a relatively minor component of the clay fraction (Table 1). All opal host rocks contain a significant amorphous fraction (up to 63%; Table 1), which is attributed to opaline silica. The opal frequently occurs as cm to dm-thick bands or pockets at the interface between shale and sandstone (Barnes et al., 1992) filling primary and secondary pore spaces and fractures (Fig. 4) and is usually found down to 30 m below the surface. In our samples, the opal shows a variety of colours ranging from blue to green to red for precious opal and white to grey and brown for non-precious or common opal. Australian sedimentary opal has never been dated radiometrically due to its low U content (Gaillou et al., 2008) so its age can only be estimated using the age of the host rocks (Exon and Senior, 1976; Thiry et al., 2006), associated minerals (Newberry, 2004), formation of silcrete (Senior and Mabbutt, 1979; Thiry et al., 2006) and carbon inside cracks in opal (Dowell et al., 2002). Thus the age of the opal varies from 130 Ma to 1 kyr B.P., which could reflect a poor selection of material used in dating or alternatively, multiple phases of opal genesis.

3. Methodology

3.1. X-ray diffraction analysis

Three key samples, one from each major opal-hosting stratigraphic horizon, were analysed by X-ray diffraction (XRD) in order to determine the composition of the fine fraction (Table 1). Based on visual examination of hand specimens as well as polished sections, the host rock from Coober Pedy is similar to the host rock from Mintabie in terms of the fine fraction, while the Winton host rock is similar to the host rock in Quilpie. The Lightning Ridge host rock is slightly different in appearance from host rocks in South Australian and Queensland opal mining

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