Earth and Planetary Science Letters 296 (2010) 319–328

Contents lists available at ScienceDirect

Earth and Planetary Science Letters

journal homepage: www.elsevier.com/locate/epsl

Evidence for a low-O_2 Archean atmosphere from nickel-rich chrome spinels in 3.24 Ga impact spherules, Barberton greenstone belt, South Africa

Alexandra E. Krull-Davatzes [a,b,*], Gary R. Byerly [c], Donald R. Lowe [b]

[a] Temple University, Department of Earth and Environmental Science, Temple University, Philadelphia, PA 19046, USA
[b] Stanford University, Department of Geological and Environmental Science, Stanford University, Stanford, CA 94305-2115, USA
[c] Louisiana State University, Department of Geology and Geophysics, Louisiana State University, Baton Rouge, LA 70803-4101, USA

ARTICLE INFO

Article history:
Received 12 January 2010
Received in revised form 13 May 2010
Accepted 17 May 2010
Available online 12 June 2010

Editor: R.W. Carlson

Keywords:
spherule
spinel
Barberton
Archean
oxygen
atmosphere

ABSTRACT

The composition of spinels in 3.24-billion-year-old Archean impact spherules in the S3 spherule bed in the Barberton greenstone belt, South Africa, provides important clues about the environments within which they formed, including the redox conditions of the impact plume and the oxygen levels in the early atmosphere. Despite pervasive diagenetic alteration of the impact spherules and nearly complete alteration of primary mineralogy, primary Ni-rich chrome spinel is preserved. The impact spinels are significantly more oxidized than detrital spinels of komatiitic origin that are also present in the spherule bed. The average Fe^{+3}/Fe_T (atoms) value in the Ni-rich impact spinels is 0.43 whereas the average Fe^{+3}/Fe_T of detrital spinels is 0.17. Fe^{3+}/Fe_T ratios of the impact spinels range from 0.26 to 0.69 (atoms) and suggest formation at oxygen fugacities below 10^{-4} bar based on comparisons with experimental results. Comparison of the S3 impact spinels with similar spinels from the K/T boundary layer, Eocene impact layer, and Late Pliocene impact layer also suggests much lower O_2 levels in the 3.24 Ga atmosphere. Oxidation of spinels present in condensed melt droplets was affected by the oxygen fugacity of the atmospheric component mixed with the ejected plume, and the broad range in values may reflect fO_2 heterogeneity both temporally and spatially within the impact-produced rock-vapor plume. Compositional and morphological variability of spinels in the spherules suggests temperature and cooling rate were heterogeneous through the plume.

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1. Introduction

1.1. Impact spherules

Impact spherules formed by the condensation of rock vapor in impact plumes have been identified throughout the geologic record. Criteria for identifying spherules as products of meteorite impacts include their anomalously high Ir contents (Montanari et al., 1983), non-terrestrial Cr-isotopic ratios (Shukolyukov and Lugmair, 1998), radiating quench textures (Simonson and Glass, 2004), centrally offset vesicles (Simonson and Glass, 2004), and the presence of Ni-rich chrome spinel (Smit and Kyte, 1984). Ni-rich spinel has been identified in spherules from the K/T boundary layer (Montanari et al., 1983), in upper Eocene spherules (Pierrard et al., 1998), in spherules from the Late Pliocene Eltanin impact (Margolis et al., 1991) and in Archean spherules from the Barberton greenstone belt (BGB), South Africa (Byerly and Lowe, 1994).

Studies of impact spinels within spherules from the BGB (Byerly and Lowe, 1994), K/T and Eocene impact suggest that they formed within melt droplets in high-temperature rock-vapor plumes shortly after the impact events (Kyte and Bohor, 1995; Kyte and Bostwick, 1995; Pierrard et al., 1998; Ebel and Grossman, 2005). In Phanerozoic impact deposits, spinels are highly oxidized, in contrast to volcanic or meteoritic chromites, which have very low ferric/ferrous ratios (Brearley and Jones, 1998; Kyte and Bohor, 1995; Kyte and Bostwick, 1995; Margolis et al., 1991; Pierrard et al., 1998). Kyte and Bohor (1995) identified Ni-rich mangnesiowüstite inclusions within magnesioferrite spinel in K/T spherules. Because mangnesiowüstite and magnesioferrite can crystallize together only within a very refractory ultramafic Mg-rich liquid, Kyte and Bohor (1995) concluded that Mg enrichment was due to fractionation of SiO_2 from MgO in a high-temperature vapor, and therefore that these spinel-bearing spherules originated from an impact-produced rock vapor.

Hydrodynamic and thermodynamic models of impact plume composition also indicate that glassy spherules and their included spinels can form by condensation from an impact vapor (Ebel and Grossman, 2005). These models suggest that spinels crystallize within liquid droplets that quench into glass spherules at temperatures greater than 1600 °C (Ebel and Grossman, 2005; Gayraud et al., 1996). A range of spherule compositions may be produced that reflects the vapor conditions when the spherules quenched (Ebel and Grossman, 2005), as well as the composition of target materials, composition of

* Corresponding author. Temple University, Department of Earth and Environmental Science, Temple University, Philadelphia, PA 19046, USA.
E-mail addresses: alix@temple.edu (A.E. Krull-Davatzes), gbyerly@geol.lsu.edu (G.R. Byerly).

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doi:10.1016/j.epsl.2010.05.014

the projectile, and mixing and fractionation processes in the plume. There is some question as to whether the spinels crystallize in a condensed melt or through direct condensation from vapor (Ebel and Grossman, 2005; Gayraud et al., 1996; Kyte and Bohor, 1995; Robin et al., 1992; Siret and Robin, 2003), but crystallization within a condensed melt is most consistent with the distribution of spinels within spherules and thermodynamic models (Ebel and Grossman, 2005). Further work by de Niem et al. (2008) indicate that spinels form in the earliest condensates of the impact plume when the liquid-to-gas ratio is less than 50%.

In this paper, we report analyses of Ni-rich spinels from a 3.24 Ga impact-produced spherule bed in the Barberton greenstone belt, South Africa. Because these spinels formed within an impact plume that was interacting with the early atmosphere, they preserve a unique record of oxygen levels in the Archean. All Phanerozoic impact spinels display high ferric/ferrous ratios resulting from crystallization in an O_2-rich atmosphere (Gayraud et al., 1996; Margolis et al., 1991; Pierrard et al., 1998; Robin et al., 1992). Comparison of the Archean impact spinel compositions with that of Phanerozoic impact spinels and volcanic chrome spinel, combined with published experimental results, provides a powerful means of estimating oxygen levels in the Archean atmosphere.

1.2. Geologic setting

Rocks of the Barberton greenstone belt have been divided into three major lithostratigraphic units deposited between 3.55 and 3.22 Ga: the Onverwacht, Fig Tree and Moodies Groups (Lowe and Byerly, 1999; Viljoen and Viljoen, 1969; Visser, 1956) (Fig. 1). The sedimentary rocks in this sequence are remarkably well preserved over large areas. Bedforms, textures, and grain shapes down to <10 μm can still be seen in most units. However, primary silicate mineralogy has been severely altered by metasomatism. Microcrystalline quartz is the most common replacement mineral, and silicification appears to have occurred soon after deposition through low-temperature interaction with sea water (Lowe and Byerly, 1999) or penecontemporaneous hydrothermal fluids (Duchac and Hanor, 1987). Aluminous silicate minerals have been altered to fine-grained phyllosilicates, predominantly sericite, chlorite, Cr-sericite, and Cr-phengite. Only the most resistant minerals are preserved, including chrome spinels and detrital zircons.

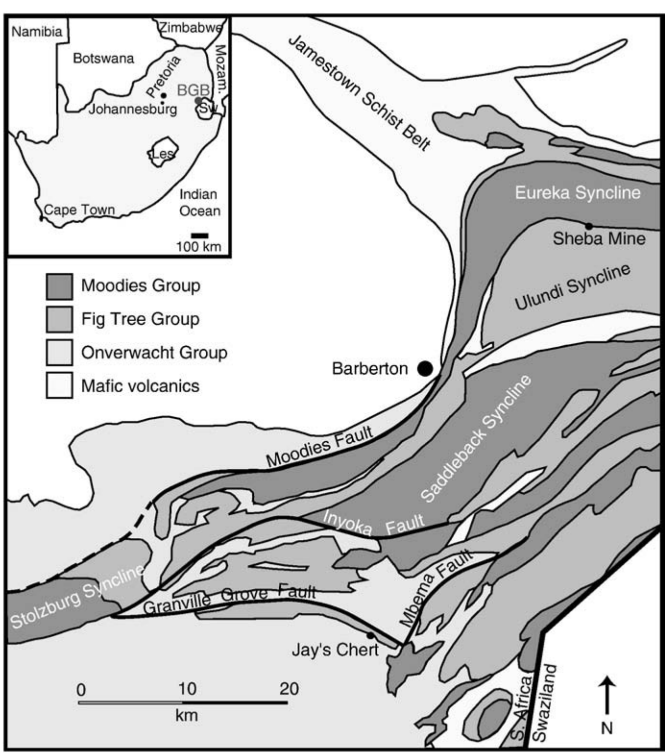

Fig. 1. Map of the Barberton greenstone belt, showing the two sample localities studied indicated with black dots and the location name. Insert map in upper left corner shows the location of the Barberton greenstone belt in a map of South Africa. Sw: Swaziland; Les: Lesotho.

Four impact-related spherule beds have been described from the Onverwacht and Fig Tree Groups (Lowe et al., 1989). The oldest (S1) occurs in the upper part of the Hooggenoeg Formation of the Onverwacht Group and is estimated to be 3470 ± 3 Ma (Byerly et al., 2002). S2 is located at the base of the Fig Tree Group and is estimated to be about 3258 ± 3 Ma (Byerly et al., 1996). S3 marks the base of the Fig Tree Group in the northern part of the belt and is approximately 3243 ± 4 Ma (Kroener et al., 1996). All dating is by single zircon U–Pb techniques on nearby tuffs or, in the case of S1, by zircons within the spherule bed. S4 is known from only one location where it occurs stratigraphically 6 m above S3 (Lowe et al., 1989).

Mapping, petrographic analysis, and geochemical studies of the spherule beds provide strong evidence that these beds are of impact origin: (1) Many spherules display quenched texture crystal pseudomorphs similar to those observed in Phanerozoic spherules (Lowe et al., 1989); (2) Beds S2 and S3 were deposited throughout the belt over a wide range of depositional environments. In shallow-water environments the spherules were reworked by waves and currents and mixed with locally derived detritus. In deep-water settings they appear to have been fallout deposits and contain very little admixed detrital material (Lowe et al., 1989); (3) The spherule bed samples are enriched in iridium up to ~900 ppb (Lowe et al., 1989); (4) Primary spinels are chemically distinct from the surrounding volcanic spinels and are similar to the low-Ti, high-Ni spinels in K/T spherule deposits (Byerly and Lowe, 1994); (5) The Cr-isotopic compositions of the spherule beds are extraterrestrial, and S2, S3, and S4 layers all display Cr-isotope values most consistent with carbonaceous chondrites (Kyte et al., 2003). Cr-isotope analyses have provided the most definitive evidence of the non-terrestrial origin of the spherules. Impact spinels contain abundant Cr (up to 56 wt.% Cr_2O_3) and are the primary carriers of chromium in the spherules. However, even where spinel is absent or has been destroyed, high levels of Cr showing extraterrestrial isotopic signatures are preserved in other mineral phases resulting from alteration of the original mineralogy, primarily Cr-phengite.

Nichromite ($Fe(Cr,Ni)_2O_4$) has been identified only in S3 (Byerly and Lowe, 1994). The spinel grains in this bed are significantly enriched in nickel relative to terrestrial spinel, a feature common in impact-produced spinels (Robin et al., 1992). Most S3 spinels have been classified as chalcophile, enriched in Ni, Co, and V, with minor amounts of lithophile (Al, Ti, Mg) spinel and manganese- and zinc-bearing spinel. Glikson (2005, 2007) reported a platinum-group element (PGE) nanonugget in a spinel from S3.

Two sections of S3 were sampled for this study: one at Jay's Chert (SAF-380) and a second at the Sheba Mine (SAF-381) (Fig. 1). Coordinates for these localities are given in Lowe et al. (2003). Nearly 5% of the spherules from both of these sections contain spinel. Samples from the Jay's Chert section have been highly silicified, and original textures and morphology are well preserved. Abundant erosional detritus has been mixed with spherules in S3 at this location. The sediments are inferred to have been deposited in a fan-delta environment and may have been affected by large impact-generated tsunamis (Lowe et al., 2003). In contrast, the spherule bed at Sheba Mine contains nearly 100% spherules in a 30-cm-thick bed, with a small amount of admixed detritus. S3 at this locality lacks current structures and appears to represent a fall deposit in deep water, with no reworking by tsunamis (Krull-Davatzes et al., 2006). Spherules in the Sheba Mine section have been structurally flattened and cemented by both carbonate and silica.

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