



Mineralogy and diagenesis of 3.24 Ga meteorite impact spherules

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ABSTRACT

Spherules in the S3 bed of the Barberton greenstone belt, South Africa are distal fallout formed within an impact plume after a large impact event at 3.24 Ga. Since that time, diagenesis and lower greenschist grade metamorphism of the spherules has changed the mineralogy, though shape and texture are largely preserved. Alteration of the S3 bed has resulted in spherules composed of quartz, phyllosilicates, Ti- and Fe-oxides, and some Ni-rich chromites. Initially, glassy spherules were palagonitized and silica cementation of the spherules began during low-temperature interaction with seawater. Further alteration by Si- and K-rich fluids resulted in a mineralogical assemblage of quartz, feldspar, and clays. Crystalline minerals were replaced by dissolution-precipitation processes, preserving relict textures. Further silica cementation resulted in complete lithification of the bed. Most of this alteration occurred at the seafloor and during shallow burial. With continued burial, amorphous silica recrystallized to microcrystalline quartz. Later recrystallization of clays to micas occurred during regional metamorphism at peak temperatures of 300–320 °C. Late-stage shearing and mineralization preferentially affected the northern region of the belt. Samples from different sections record highly variable local conditions. Water depth, the amount of pre-depositional transport, location within the belt, and proximity to igneous dikes all affect the diagenesis of the S3 spherules. Silica and barite concentrations are lower, and carbonate concentrations are higher in the deep-water depositional environments. Element mobility during diagenetic and lower greenschist grade metamorphism can be inferred based on studies of multiple sections throughout the BGB. The most immobile elements are Al, Zr, Ti, Sc and the high field strength elements and present element ratios can be used to infer original composition of the spherules. The large ion lithophile elements are highly mobile, as are the light rare earth elements (REEs), which are particularly susceptible to mobilization during carbonate diagenesis and phosphate authigenesis. Of the REEs, Ce and Eu show the largest variability, suggesting significant mobilization during diagenesis and low-grade metamorphism. Consistent Cr/Ir ratios, particularly in the high concentrations suggest limited mobility, and segregation of platinum group elements into Ni-rich chromite-bearing spherules. Sulfide mineralization has not affected the Ir concentration in the S3 layer.

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1. Introduction

The earliest physical evidence of meteorite impacts on Earth in the geologic record is contained in layers of spherules in the Barberton greenstone belt (BGB), South Africa (Lowe and Byerly, 1986b, 2010) that range in age from 3.47 to 3.23 Ga (Lowe et al., 1989; Lowe and Byerly, 2010). Lowe and Byerly (1986b) present evidence that the beds are composed in large part of sand-sized spherical particles, termed spherules, which are thought to have formed by the condensation of impact vapor plumes ejected from

the sites of large impacts. Mapping of the regional extent and stratigraphic relationships, petrographic analysis, and geochemical studies, including iridium (Ir) measurements and Cr-isotopic measurements (Byerly and Lowe, 1994; Kyte et al., 1992, 2003; Lowe et al., 1989; Shukolyukov et al., 2000) provide strong evidence that these Archean spherule beds contain non-terrestrial components and were formed as a result of meteorite impacts.

One of the primary problems in interpreting the origin and implications of the BGB spherule beds is the pervasive alteration that has obliterated all but the most hardy components of virtually all rocks in the belt (Condie et al., 1977; Duchac and Hanor, 1987; Hofmann, 2005; Lahaye et al., 1995; Lecuyer et al., 1994; Lowe, 1999; Reimer, 1972). Like other beds in the BGB, the spherule beds have experienced silica and potash metasomatic alteration (Duchac and Hanor, 1987; Hanor and Duchac, 1990; Hofmann, 2005; Lowe et al., 1983, 2003). However, ratios of original immobile elements

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(Al, Ti, P, Cr, Nb, Y, Zr) and rare earth elements have been shown to be preserved in many highly altered BGB komatiites and sedimentary units (Hanor and Duchac, 1990; Hofmann, 2005; Reimer, 1972). This paper outlines the nature of diagenetic alteration and metasomatism that has affected the S3 spherules through the use of textural observations, immobile element ratios, and comparison to modern spherules. By comparing sections from different sedimentological settings that have been exposed to variable diagenetic histories, this work provides a necessary framework for further investigations of the original mineralogy of the spherules. It also provides a better understanding of local and regional diagenesis and metamorphism of the BGB.

2. Geologic setting

2.1. The Barberton greenstone belt, South Africa

The Barberton greenstone belt (BGB) includes three major lithostratigraphic units (Figs. 1 and 2) deposited between about 3.5 and 3.2 Ga. At the base is the Onverwacht Group, which consists of about 10 km of mainly mafic and ultramafic volcanic rock, at least one major felsic igneous and volcanoclastic unit, and thin interlayered chert beds (Fig. 2) (Lowe and Byerly, 1999b; Viljoen and Viljoen, 1969a,b,c). The overlying Fig Tree Group is composed of greywacke, shale, chert, and felsic volcanoclastic rocks (Fig. 2) (Lowe and Byerly, 1999a; Condie et al., 1970). The top of the sequence is the Moodies Group, which consists of up to 3000 m of feldspathic and quartzose sandstone, conglomerate, some siltstone and minor shale (Fig. 2) (Anhaeusser, 1976; Eriksson, 1982; Heubeck and Lowe, 1999).

A wide range of depositional environments is represented by sedimentary rocks in the belt, including evaporitic, fluvial, and marine deposits. In general, north of the Inyoka Fault (Fig. 1), there is evidence that sediments in the Fig Tree Group were deposited in deep water (below wave base) whereas the southern facies represent a range of shallow water settings (Lowe and Byerly, 1999b).

Post depositional shortening through the development of fold and thrust belts along the northwestern and southeastern margins of the BGB occurred between 3240 and 3226 Ma (Lowe et al., 1999; de Wit et al., 2011). This phase, termed D₂ by Lowe (1999), therefore occurred shortly after the deposition of the spherules, concurrent with Moodies Group deposition (de Wit et al., 2011). Three other major stages of deformation have occurred since deposition of the spherules, including extension and uplift during deposition of the Moodies (D₃) and two additional stages of shortening after deposition of the Moodies (D₄ and D₅) that resulted in the tight folding and near vertical dips that are seen today (Lowe et al., 1999). Most of the major regional deformation of the BGB occurred during these later stages (Lowe et al., 1999; Ramsay, 1963).

2.2. S3 spherules

This paper focuses on the diagenesis and metamorphism of a single spherule bed, S3, selected largely because it is the spherule bed that is most widely distributed throughout the BGB. S3 marks the base of the Fig Tree Group in northern sections and is present up to 100 m above the base of the Group in the southern part of the BGB. A tuff immediately underlying S3 has been dated by single zircon U–Pb techniques at 3243 ± 4 Ma (Kröner et al., 1991). S2, located at the base of the Fig Tree Group in many southern sections is older, underlying a 3259 Ma tuff, showing that the Fig Tree-Onverwacht contact is diachronous across the belt.

There is abundant evidence that most rocks in most of the Barberton greenstone belt have been exposed to maximum

post-depositional temperatures of 300–320 °C (Tice et al., 2004; Xie et al., 1997) and that only localized regions have been subjected to higher temperatures, usually due to contact metamorphism around plutons or adjacent to widespread younger mafic dikes. The southernmost parts of the greenstone belt and greenstone remnants engulfed in surrounding plutons, including the Schapenburg greenstone remnant, both of which represent deeper crustal levels than the central parts of the belt, have also undergone significantly higher grades of regional metamorphism (Lecuyer et al., 1994; Diener et al., 2006). The rocks that are the focus of this study, however, lie within the central part of the BGB and have experienced only sub- or lower greenschist grade metamorphism.

Five main types of spherules are contained within the S3 spherule bed (Krull-Davatzes et al., 2006): (1) microcrystalline quartz spherules, (2) patchy mixed microcrystalline quartz and phyllosilicate spherules, (3) phyllosilicate spherules, (4) layered spherules, including spherules with (a) quartz-phyllosilicate layers, and (b) phyllosilicate-phyllosilicate layers, and (5) barred textured spherules (Fig. 3). All of the five types of spherules are present in every section of S3 discussed in this paper and collectively represent the altered remnants of a diverse population of impact-produced particles. Due to the pervasive silicification and K-metasomatism in the Barberton greenstone belt (Duchac and Hanor, 1987) quartz, phyllosilicates, and Fe- and Ti-oxides are the most common minerals in the spherules at present.

All of these spherule types were produced by the impact of a carbonaceous chondrite (Kyte et al., 2003) into basaltic and ultramafic rocks (Byerly and Lowe, 1994). Ni-rich chromite is the only known primary mineral that remains in the spherules. Published geochemical data for the Ni-rich chromites are available in Krull-Davatzes et al. (2010) and Byerly and Lowe (1994). This paper presents petrographic and geochemical analyses of the major diagenetic minerals in the S3 spherule bed.

3. Samples and procedures

Samples of S3 were collected from Jay's Chert (SAF 380, SA 306), Barite Valley (SAF 206), Maid-of-the-Mist (SA 528), Loop Road (SAF-295), Princeton Tunnel at Agnes Mine (SA-315), Florence and Devonian Mine (SA-294), and from two sections of Sheba Mine, Sheba 1 (SAF 381, SA 323), previously referred to by Wagoner and Wiegand (1986) as the Zwartkoppie section of Sheba Mine, and Sheba 2, previously called the Hospital Bar section of Sheba Mine (SA 336) (Fig. 1). Samples were collected through the complete thickness of the spherule bed where it is fully exposed, and from units above and below the spherule bed, where exposed. Petrographic thin sections were prepared from all sample sites. Heavy minerals were separated, mounted and studied using the petrographic microscope and the Scanning Electron Microscope (SEM). Small-grained titanium oxides and carbonaceous matter were identified from polished uncoated thin-sections using Laser Raman spectroscopy. Spectra were collected using a Kaiser Hololab D5000 Raman microscope with a 785 nm diode laser with a spot size of 1 μm.

Major, trace and rare earth element (REE) compositions of the spherule layers were determined by X-ray Fluorescence (XRF) and Inductively Coupled Plasma Mass Spectroscopy (ICP-MS) performed by the Washington State University GeoAnalytical Laboratory. The analyzed samples consisted of concentrated spherules containing little or no detrital matter and from a full thickness of the spherule bed from the Loop Road (SAF-295) the Barite Valley (SAF-206) and the Sheba 1, Sheba Mine (SAF-381) sections. These analyses therefore provide the most accurate representation of the overall spherule bed geochemistry. Spherule beds with abundant admixed detrital material were not analyzed for bulk chemistry.

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