



# In situ oxygen isotope compositions in olivines of different types of cosmic spherules: An assessment of relationships to chondritic particles

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## Abstract

Cosmic spherules collected from deep sea sediments of the Indian Ocean having different textures such as scoriaceous (4), relict-bearing (16), porphyritic (35) and barred olivine (2) were investigated for petrography, as well as high precision oxygen isotopic studies on olivine grains using secondary ion mass spectrometry (SIMS). The oxide FeO/MgO ratios of large olivines (>20 μm) in cosmic spherules have low values similar to those seen in the olivines of carbonaceous chondrite chondrules, rather than matching the compositions of matrix. The oxygen isotope compositions of olivines in cosmic spherules have a wide range of  $\delta^{18}\text{O}$ ,  $\delta^{17}\text{O}$  and  $\Delta^{17}\text{O}$  values as follows:  $-9$  to  $40\%$ ,  $-13$  to  $22\%$  and  $-11$  to  $6\%$ . Our results suggest that the oxygen isotope compositions of the scoriaceous, relict-bearing, porphyritic and barred spherules show provenance related to the carbonaceous (CM, CV, CO and CR) chondrites. The different types of spherules that has experienced varied atmospheric heating during entry has not significantly altered the  $\Delta^{17}\text{O}$  values. However, one of the relict-bearing spherules with a large relict grain has  $\Delta^{17}\text{O} = 5.7\%$ , suggesting that it is derived from  $^{16}\text{O}$ -poor material that is not recognized in the meteorite record. A majority of the spherules have  $\Delta^{17}\text{O}$  ranging from  $-4$  to  $-2\%$ , similar to values in chondrules from carbonaceous chondrites, signifying that chondrules of carbonaceous chondrites are the major contributors to the flux of micrometeorites, with an insignificant fraction derived from ordinary chondrites. Furthermore, barred spherule data shows that during atmospheric entry an increase in  $\sim 10\%$  of  $\delta^{18}\text{O}$  value surges  $\Delta^{17}\text{O}$  value by  $\sim 1\%$ .

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## 1. INTRODUCTION

The Earth is continuously bombarded by an extraterrestrial material flux of  $\sim 30,000$  tons per annum (Love and Brownlee, 1991). While more than 90% of this material gets evaporated in the atmosphere, a fraction of the colliding particles survive atmospheric entry heating to reach the

Earth's surface (Taylor et al., 1998; Yada et al., 2004; Plane, 2012; Prasad et al., 2013). The survivability of these particles that penetrate the Earth's atmosphere depends on various factors such as mass, density, velocity, zenith angle during entry and chemical compositions that are markedly different for different precursors (Love and Brownlee, 1991, 1993; Rudraswami et al., 2015a). The micrometeorites that have entered Earth's atmosphere at high zenith angle and low velocity are interesting because their petrologic and isotopic characteristics are largely preserved thereby providing constraints on the parent body they have originated from.

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However, those micrometeorite particles that enter at low zenith angle and high velocity experience large heating and vaporization during entry, thereby altering the chemical and isotopic compositions. Under such circumstances, relating the chemical composition of the micrometeorites/cosmic spherules to their origins is marred by uncertainties (Kurat et al., 1994; Beckerling and Bischoff, 1995; Brownlee et al., 1997; Greshake et al., 1998; Cordier et al., 2011; Rudraswami et al., 2012).

Oxygen isotope studies complemented by mineralogical compositions are vital to unravel the complexity involved in distinguishing the known components of micrometeorites and their parent bodies. For carbonaceous chondrites, the oxygen isotope compositions of the chondrules, CAIs and matrix fall close to the Carbonaceous Chondrite Anhydrous Minerals (CCAM) line with slope  $\sim 1$  (Clayton, 1993). Mixing of these different components with nebular gas, and parent-body processing after accretion, led to changes in  $\delta^{18}\text{O}$  and  $\Delta^{17}\text{O}$  values (Yurimoto et al., 2008). However, the chondrules in ordinary chondrites have a different trend than carbonaceous chondrites and fall above the CCAM line (Clayton et al., 1991; Kita et al., 2010). For cosmic spherules, interactions with the terrestrial atmosphere during atmospheric entry without substantial evaporative mass loss do not alter the  $\Delta^{17}\text{O}$ , but can change the  $\delta^{18}\text{O}$  value significantly (Engrand et al., 2005; Suavet et al., 2010; Rudraswami et al., 2015b). Despite the effects of atmospheric mixing, some cosmic spherules retain relict grains that preserve their primary oxygen isotope signatures, indicating that they did not crystallize from a melt during atmospheric entry (Engrand et al., 2005; Yada et al., 2005; Genge et al., 2008; Suavet et al., 2010; Rudraswami et al., 2015b).

One of the issues we wish to address here is whether exchange of  $\delta^{17,18}\text{O}$  between a micrometeorite and the atmosphere will preclude an understanding of its parent body. In an earlier study, we showed that most Mg-rich relict olivine grains of cosmic spherules have  $\Delta^{17}\text{O}$  ranging from  $-5$  to  $0\%$ , thereby suggesting that chondrules of carbonaceous chondrites could be the origin for the relict bearing cosmic spherules (Rudraswami et al., 2015b). In addition, some Mg-rich relict olivine grains are very  $^{16}\text{O}$ -rich, with  $\Delta^{17}\text{O}$  ranging from  $-21.9$  to  $-18.7\%$ , similar to oxygen isotopic compositions observed in calcium aluminium rich inclusions (CAIs), amoeboid olivine aggregates (AOAs), and some porphyritic chondrules from carbonaceous chondrites (Rudraswami et al., 2015b). Earlier studies were done on large sized relict bearing cosmic spherules largely dominated by forsteritic olivine. In the present study, we have performed high precision ion microprobe oxygen isotope analyses of olivines in 57 cosmic spherules having different textures (scoriaceous, relict-bearing, porphyritic and barred) that has experienced different heating (i.e. partially to large scale heating) during atmospheric entry. The objectives is to see the oxygen isotope variation in different type of cosmic spherules and compare with different chondrites and their components. This will benefit in recognizing and probably quantifying the range of contributors to the micrometeorite flux on the Earth. Secondly, the particle as it enter the stratosphere undergo oxygen isotopic modi-

fication leading to enrichment of  $\delta^{17,18}\text{O}$ . Distinguishing the variations in  $\delta^{17,18}\text{O}$  and  $\Delta^{17}\text{O}$  will be beneficial to identify the parent bodies of these particles.

## 2. SAMPLE SELECTION

Cosmic spherules chosen for the present investigation were collected from the deep-sea sediments of the Indian Ocean from a seafloor depth of  $\sim 5000$  m. A grab sampler of size  $50 \times 50$  cm (length  $\times$  breadth) has been used to sample the seafloor sediments: the sampler penetrates to a depth of  $\sim 15$  cm and has the capacity to pick up  $\sim 45$  kg of wet sediments (Rudraswami et al., 2011a, 2012; Prasad et al., 2013). The sediments were sieved using a  $200 \mu\text{m}$  mesh sized sieve and the  $>200 \mu\text{m}$  portions were dried and subjected to magnetic separation. Individual spherules were handpicked under a binocular microscope from these magnetic separates. A total of  $\sim 2500$  spherules were mounted in 12 sections which were carefully ground and polished to expose the internal textures. Fifty-seven cosmic spherules of different types, have experienced different stages of heating during entry and that have large olivine grain ( $>20 \mu\text{m}$ ), were selected for oxygen isotope studies from the total  $\sim 2500$  observed under scanning electron microscope (SEM). We focus on olivine oxygen isotope studies in different micrometeorites as a fairly robust indicator to understand precursor meteorite body, and different level of heating in different types of cosmic spherules will provide information on stratospheric oxygen isotope exchange during interaction. The selection criteria for spherules was to sample a representative selection of different petrographic textures and types such as scoriaceous (4), relict-bearing (16), porphyritic (35) and barred olivine (2).

## 3. ANALYTICAL TECHNIQUES

A JEOL JSM 5800LV SEM having an OXFORD INCA energy dispersive spectrometer detector (EDS,ISIS-300 at National Institute of Oceanography, Goa) was used to observe high resolution back scattered electron images (BSE) for classifying the spherules. The images were used to select the best spherules and identify phases for chemical and oxygen isotope analyses using electron microprobe and ion microprobe, respectively. A Cameca SX5 Electron Micro Probe Analyzer (EPMA at National Institute of Oceanography, Goa) equipped with four spectrometers was used to determine the elemental composition of different phases in the spherules, and the operating conditions are identical to those used in Rudraswami et al. (2015b). After SEM and EPMA analyses, we selected 57 cosmic spherules having olivines with grain sizes  $>20 \mu\text{m}$  for further oxygen three isotope studies based on morphological features and the grain sizes.

Oxygen three isotope analyses on cosmic spherules were performed using the Secondary Ion Mass Spectrometer (SIMS, Cameca IMS 1280 at University of Hawaii, USA). The  $\text{Cs}^+$  primary beam ion current  $\sim 1$  nA was rastered over  $7 \times 7 \mu\text{m}$  on the selected phase for measuring secondary ions. The mass resolving power ( $M/\Delta M$ ) used for  $^{16}\text{O}^-$  and  $^{18}\text{O}^-$  was  $\sim 2000$ . Higher mass resolving

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