Internal structure of type I deep-sea spherules by X-ray computed microtomography

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Abstract—The internal structures of type I spherules (melted micrometeorites rich in iron) have been investigated using synchrotron-based computed microtomography. Variations from sphericity are small—the average ratio of the largest to the smallest semimajor axis is 1.07 ± 0.06. The X-ray tomographs reveal interior cavities, four spherules with metal cores with diameters ranging from 57 to 143 µm and, in two spherules, high attenuation features thought to be nuggets rich in platinum-group elements. Bulk densities range from 4.2 to 5.9 g/cm3 and average grain densities from 4.5 to 6.5 (g/cm3) with uncertainties of 10–15%. The average grain densities are those expected for materials containing mostly oxides of iron and nickel. The tomographic density measurements indicate an average void space of 5.8 ± 8%. The void spaces may be contraction features or the skeletons of bubbles that formed in the molten precursors during atmospheric passage.

INTRODUCTION

With the wider availability of intense X-ray sources and improved software for image reconstruction, the application of computed X-ray microtomography (CMT) to extraterrestrial materials has attracted increasing interest (e.g., Tsuchiyama et al. 1997, 2002, 2003; Kondo et al. 1997; Carlson and McCoy 1998; Koeberl et al. 2002; Hertz et al. 2003; Murray et al. 2003). Arnold et al. (1983) first demonstrated the potential of CMT for non-destructive mapping of extraterrestrial objects. Here we report on the application of CMT to the rare class of melted, iron-rich micrometeorites known as the type I spherules. These spherules were collected from mid-Pacific sea floor with the cosmic muck rake (Brownlee et al. 1979; Brownlee 1985).

Type I spherules range in diameter from ~0.01 mm to 0.50 mm (Czajkowski 1987) and consist mainly of iron, nickel, and terrestrial atmospheric oxygen. Their nearly spherical shapes indicate rapid cooling from a liquid phase. The key features observed in microscopic examination of polished sections are thought to reflect progressive oxidation of molten metal (e.g., Castaing and Fredriksson 1958; Kyte 1983; Czajkowski 1987; Brownlee et al. 1997). In particular, some spherules, frozen before oxidation was complete, retain an unoxidized core of metal, usually positioned off center. Other spherules consist nearly entirely of oxides, either because they lost a metal core in transit or because oxidation of iron and nickel was complete. Many of the spherules have monomineralic rims a few micrometers thick, which Engrand et al. (1998) have identified as magnetite and Kosakevitch and Disnar (1997) as hematite.

The interior oxides vary in composition and texture and, in polished section, often appear to be interlaced with cracks, which may or may not be polishing artifacts. Some type I spherules also contain micrometer-size nuggets enriched in Ir and other Pt-group elements (PGE) (Brownlee et al. 1984; Bonté et al. 1987).

We set out to see whether CMT might be a good way to map the structures of type I spherules, to determine their porosities, and to draw inferences from the structures about the formation process. Some specific questions of interest concerned the frequency of occurrence of metal cores and of
the micrometer-size nuggets mentioned above and the shape of any pore space. Feng et al. (1999) have given a preliminary account of this work.

**EXPERIMENTAL METHODS**

The type I spherules described came from the collection of Brownlee (Brownlee et al. 1979; Brownlee 1985), two from the KK1 and twenty-two from the KK2 collections. We weighed (±3 µg) and measured the diameter (±5 µm) of each type I spherule from the KK2 collection using a Cahn microbalance and an optical microscope, respectively. The spherule position chosen for the optical measurement was the one in which the particle happened to land on the microscope stage. This approach would likely favor orientations in which a longer axis lay parallel to the stage surface. We also tomographically measured three perpendicular diameters for each spherule (see below)—such measurements are more difficult with an optical microscope.

For tomography, we glued each spherule to the end of a glass or lucite rod with Duco cement. The rods were mounted on a stage that could be translated vertically and rotated with a precision of ±1 µm and of ±0.001°, respectively. X-rays transmitted by the spherules were detected with a YAG:Ce scintillator viewed through a magnifying lens by a position-sensitive CCD camera (active area: 3088 pixels wide; 2056 pixels high).

Two of the spherules were tomographed at the National Synchrotron Light Source (NSLS) of Brookhaven National Laboratory (BNL) with a white beam of X-rays from a bending magnet. Sheets of Al (1.6 cm thick) and of Mo (0.1 mm thick) were used to attenuate the low-energy portions of the X-ray distribution and, thus, remove effects caused by attenuation of the low-energy component of the beam in the sample. The X-ray beam, 3 mm wide and 0.5 mm high, had a roughly Gaussian intensity profile in the vertical direction that varied by <10% over the central 300 µm. The size of one volume element or voxel was measured by moving a straight edge by a known distance in the X-ray beam. For these measurements, one voxel corresponds to 3.60 µm³ when binning and optical magnification are taken into account.

The X-ray energy spectrum shown in Fig. 1 was calculated using the theoretical X-ray energy distribution for the NSLS bending magnet (Thomson and Vaughan 2001) and the X-ray attenuation coefficients of the attenuators.

We obtained tomographic data for 22 other spherules by using a monoenergetic beam with a photon energy of 25 keV so that no attenuator foils were required. For these measurements, one voxel corresponds to 2.27 µm³ when binning and optical magnification are taken into account.

The data for the tomographic volumes were obtained by making 1000 exposures as the sample was rotated in 0.18° steps between 0° and 180°. Data were corrected for the binning and optical magnification are taken into account.

Table 1. Calculated approximate X-ray energy distribution of photons used in the white beam experiments. The calculations include the effects of 6350 µm Al and 298 µm Zr attenuators placed in the beam path. These attenuators differ from those used for tomography, but not by enough to affect conclusions drawn below.

![Fig. 1. Calculated approximate X-ray energy distribution of photons used in the white beam experiments.](image-url)
keyboard, the software allows us to change dynamically viewing parameters such as the color table (the colors chosen to represent different attenuation coefficients) and opacity scheme. The user can control the viewing content by adding or removing isosurfaces, cutting planes, color scales, histograms, and other displays. Other features include stereo visualization and animation. To enhance the data analysis, we added various user defined filters and routines.

To check the results of the tomography, we embedded spherule KK1-98-6 in epoxy and examined successively polished sections prepared at depths of 8, 13, 16, 22, 27, 28, and 31 µm. For compositional analysis of the section at a depth of 31 µm, we used a scanning electron microscope in quantitative, energy-dispersive mode with polished metal standards and ZAF corrections (Newbury et al. 1986).

RESULTS

Volumes

Figure 2 and Table 1 show the semi-major axes (a, b, and c) that were determined tomographically and plotted against the radius (r_{optical}) measured once with an optical microscope. In most cases, the tomographic radii agree with the microscopic measurements to within 10%, and in all cases to within 16%. The ratio of the largest to the smallest tomographic radius varies from 1.00 for spherule 14 to 1.20 for spherule 23, averaging 1.07 ± 0.06. Both these observations indicate relatively small variations from sphericity. We have calculated a volume for each spherule from the formula for the volume of an ellipsoid, \( V = \frac{4}{3} \pi abc \) (Table 1). This approach may overestimate the true volumes of objects with large irregularities on their surfaces (e.g., spherule 27).

Bulk Densities

We calculate bulk density (Table 1) by dividing the spherule mass by the volume, calculated above. Void spaces make the calculated bulk densities lower than the densities of the constituent grains (see Grain Densities section). Nonetheless, the average bulk density for our spherules, 5.0 ± 0.5 g/cm³, is similar to the density expected for objects consisting mainly of wüstite (5.70 g/cm³), hematite (5.24 g/cm³), and magnetite (5.14 g/cm³).

The bulk densities of the spherules could provide some information about the maximum interior temperatures of the spherules at the time of freezing, and hence, set lower limits on maximum temperatures. In particular, the exterior shell presumably froze around a continuous, molten interior. If spherules today preserve the size of that shell, the bulk densities should give the densities at the time of spherule formation. If density were known as a function of temperature (which is not the case), then we could infer the average interior temperature of the melt.

Attenuation Coefficients

Figure 3 shows as frequency plots the attenuation coefficients determined for the 22 type I spherules tomographed with monochromatic X-rays. The attenuation coefficients range from -0.010 pixel^{-1} to ~0.030 pixel^{-1} with uncertainties of ±5% (see above). Negative values generally occur for voxels close to the spherule periphery and are thought to reflect X-ray scattering within the spherules. Most of the frequency distributions have two main peaks: one peak centered on values near zero, which correspond physically to void space either within or exterior to the spherules; and a second peak comprising attenuation coefficients ranging upward from 0.005 pixel^{-1}. These values indicate the presence of matter. Attenuation coefficients between 0.007 pixel^{-1} and 0.010 pixel^{-1} bracket the secondary peaks for most spherules. Closer inspection reveals that the secondary peak position correlates weakly with its width (Fig. 4). A few frequency distributions have a distinct third peak.

Grain Densities

We use the term grain density to mean the spherule mass divided by the volume of solid material. To obtain the volume
of solid material, $V_{\text{solid}}$ (Table 1, column 9), we counted the total number of voxels with X-ray coefficients indicating the presence of matter. The volumes determined in this way have uncertainties of about 10%. The grain densities range from 4.5 g/cm$^3$ to 6.5 g/cm$^3$ (Table 1, column 6), have uncertainties of 5–10%, and average to 5.3 ± 0.6 g/cm$^3$. All but two of the spherules have grain densities that lie within one sigma of the density of wöstité or magnetite. For both outliers, KK298-A4 and -A22, the densities are about 1.2 standard deviations lower than that of magnetite.

**Fractional Void Space (Porosity)**

Conventional petrographic observations cannot readily distinguish holes indigenous to a polished section from those produced by plucking. Non-destructive CMT does not create voids and makes the identification of holes larger than ~5 μm easy because of the strongly contrasting density of the surrounding solid material. Our tomographic measurements and previous measurements (Tsuchiyama et al. 1997) reveal void spaces ranging from small, enclosed regions to larger unbounded regions that connect to the outer surface of the spherules.

We calculate the fraction of void space from the relation $1 - V_{\text{solid}}/V$ (Table 1). On average, the void spaces calculated by this relation account for 5.3% of the total volume. Murray et al. (2003) and Tsuchiyama et al. (2003) have recently reported the presence of void space in chondrules. The porosity of chondrules, about 3% (Tsuchiyama et al. 2003), appears to be similar to, although perhaps somewhat smaller than, that of the type I spherules.

**Oxides**

The X-ray attenuation coefficients for the major phases known to be present in type I spherules (metal, wöstité, and magnetite/hematite) can be estimated from published X-ray attenuation coefficients of the relevant materials (Berger et al. 1999), which are shown in Fig. 5, from the linear thickness of the absorber (assumed constant), and from the atomic composition of the material. The values calculated for hematite/magnetite are about one fourth of those for wöstité and about half those for metallic iron. From analogous data for Pt (Berger et al. 1999; not plotted), we estimate that the values for hematite/magnetite are about 10% less than those for wöstité and about half those for metallic iron. From analogous data for Pt (Berger et al. 1999; not plotted), we estimate that the values for hematite/magnetite are about 10% less than those for wöstité and about half those for metallic iron. From analogous data for Pt (Berger et al. 1999; not plotted), we estimate that the values for hematite/magnetite are about 10% less than those for wöstité and about half those for metallic iron.
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spherule KK1-98-6 at a depth of 31 µm. We have enhanced the contrast to emphasize the differences among the various phases. The bright feature (upper right), seen in Figs. 6a and 6b, is a PGE-rich nugget discussed below. We identify the more abundant and darker phase as magnetite and the lighter phase as wüstite. From the scale of Fig. 6, we see that a spatial resolution of 1 µm or less is needed to resolve the regions containing the two minerals. Although the tomography does not resolve the various oxides, their occurrence in different proportions almost certainly influences the positions and widths of the secondary peaks of Fig. 3.

**Metallic Cores**

Metallic cores attenuate 25 keV X-rays more effectively than the various iron oxides (Fig. 5). Interestingly, separate peaks at the higher attenuation coefficients expected for metal appear in only four cases (Fig. 3): KK298A-10, -12, -27, and perhaps -7. Figures 7a and 7b show two visualizations of the core in spherule 10 (core diameter ~90 µm). They were created by projecting our 3D data set onto a 2D viewing plane. In particular, we used a “ray casting” technique, where the color and opacity (transparency) of each pixel in the 2D viewing plane is determined by adding color and opacity contributions along a ray passing through a volume that includes a voxel at the location of the pixel itself. In constructing Fig. 7a, we first removed voxels exterior to the spherule by setting a threshold attenuation coefficient of 0.002. We used the gray color scale shown at the bottom of Fig. 7a; opacity increases from 0 for attenuation 0.002 to 1 for attenuation 0.014. Superimposed on the gray scale is a frequency plot, which differs in appearance from that of Fig. 3 because the vertical scale is linear rather than logarithmic. The core is located at the spherule surface and has what

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Fig. 3. Measured frequency distributions of attenuation coefficients for 22 type I spherules tomographed with 25 keV X-rays. The values of the attenuation coefficients are indicative of the elemental composition of the individual voxels. The full names of the spherules can be obtained by prefixing KK298A- to all spherule numbers given in the legend.
appears to be a spherical dimple on its own surface, suggesting either a collapse on contraction or the ejection of a spherical bead. To show more clearly where the void space "begins," we have also visualized a semi-transparent isosurface at attenuation 0.005. In the second visualization, Fig. 7b, we have rotated the spherule clockwise by 90°, retaining only the metallic core and an equatorial slice taken perpendicular to an axis that transects the core. This equatorial slice has considerable, irregular void space and is typical in appearance.

Visualizations of spherules 7, 12, and 27 reveal that their cores (or more strictly, the regions with the highest attenuation coefficients) are spherical, are located near the surface, and have approximate diameters of 57 µm, 68 µm, and 143 µm, respectively. Even the largest core (that of spherule 27) occupies less than 10% of the total volume.

Most of the spherules that were tomographed do not contain large (>10 µm in diameter) metallic cores. The inferred scarcity of metallic cores is somewhat surprising but is consistent with the results of Xue et al. (1995) and Bonté et al. (1987), who studied other objects from the KK collection.

**Nuggets Rich in Pt-Group Elements**

Type I spherules may contain tiny nuggets rich in Pt-group metals. We expect the attenuation coefficients of nuggets to be considerably higher than those of lower Z, Fe-Ni cores. The tomographs obtained with white radiation revealed small features close to the surfaces of spherules...
line of sight; the images on the right show the voxels thought to be empty. A depression at the top of spherule 8 (see left hand image) continues downward into the interior in filmy, partly connected sheets (like spinach, perhaps) forming a roughly conical region overall (see right-hand image).

Spherule 12 has a well-defined metallic core, which we have outlined in white. The porous region in spherule 12 is roughly spherical with a few small but no large connections to the surface. This object may have cooled rapidly and uniformly over its entire surface. For spherule 4, the internal pore space
looks like a pancake that connects to the surface along a short segment of one edge.

Figures 9a and 9b show quasi-3D visualizations of spherule 10. One “surface” of the void space follows the contours of the metallic core, cupping it. The rest of the void space appears more or less spherical, i.e., to have cylindrical symmetry overall.

In most of the spherules, the centers of mass are displaced from the centers of the void spaces. In general, we see no clear relation between the positions or forms of the void spaces and those of the metallic cores, although there may be a weak tendency for the void spaces to lie close to the cores.

DISCUSSION

Structural Systematics

Many authors have discussed mechanisms for the formation of deep-sea spherules (e.g., Love and Brownlee 1991; Yada et al. 1996). Most of them agree that melting and varying degrees of oxidation occurred during deceleration in the Earth’s atmosphere. Isotopic evidence indicates that substantial evaporative losses from the melt took place at the same time (Clayton et al. 1986; Davis and Brownlee 1993; Engrand et al. 1998; Herzog et al. 1999). The spatial distribution of oxides, metallic cores, and nuggets seen in the
spherules follows naturally from the tendency during deceleration of higher density fluids to move forward in a lower density matrix. Thus, Ni-rich cores often occupy eccentric locations tangential to what was presumably the leading surface. If we extend this argument to the even higher density nuggets, which are residues left after the nearly complete oxidation of metal cores, then we would expect to find them near the leading surface as well. By the same token, lower density material and voids that formed during deceleration should tend to collect toward the rear. The observed spatial distributions of the void spaces are not always consistent with this simple picture.

**Oxides**

During spherule formation, as temperature increases at fixed oxygen fugacity, the most stable form of the iron changes from FeO to Fe$_3$O$_4$ to Fe$_2$O$_3$ (Yada et al. 1996). The trend is the same as oxygen fugacity increases at fixed temperature. We imagine that as the temperature of an incoming spherule rises and reaction with oxygen proceeds, a wüsite (FeO) melt forms first. On further heating and as the spherule encounters more oxygen, the ratio of oxygen to iron in the melt increases and magnetite forms. Moments later, when the spherule approaches the end of its trajectory and starts to cool, freezing begins at the surface with either magnetite (Levin et al. 1974) or possibly hematite (Kosakevitch and Disnar 1997), depending on the degree of oxidation. The exact proportions of the various oxides will depend on the availability of oxygen and kinetic considerations. Figure 5 implies that the average attenuation coefficient of the solid matter present in the spherules should decrease as the degree of oxidation increases. Thus, although we cannot resolve the various oxides of iron spatially, we can regard the spectral shapes of Fig. 3 as defining a kind of oxidation sequence, with the least oxidized members represented by KK298A-10 or -27 and the most oxidized by spherule KK298A-3. The spectra of spherules 10 and 27 have structure and considerable breadth at higher values of the attenuation coefficients. In contrast, the peak near 0.007 in spectrum 3, which is thought to comprise the voxels filled by mass, is relatively sharp.

**Void Space**

The void spaces could be: 1) contraction features created as the spherules froze, first at the surface and then inward (Tsuchiyama et al. 1997); 2) ghosts of bubbles that once contained gases that vaporized from the source material (Tsuchiyama et al. 1997; Wang et al. 1994); or 3) corrosion features if the spherules retained any metal.

Some shrinkage porosity would seem inevitable in type I spherules. Related effects have plagued the metal casting industry for centuries and are well known to metallurgists (e.g., Reed-Hill and Abbaschian 1992). Shrinkage occurs in three stages: as liquid freezes; as liquid remaining in the interior cools; and finally as solid cools. Metallurgists refer to conical defects due to shrinkage of cylindrical castings as pipes. Pipes form when liquid metal charges cool rapidly from one end. The void space in spherule 8 has roughly cylindrical symmetry and might have formed in this way. Engrand (personal communication) has observed that fresh-looking...
crystals of magnetite often border the void space, and that the texture of the assemblage seems consistent with direct crystallization.

Shrinkage is not the only way to create void space. The exsolution of gas can create bubbles. Wang et al. (1994) described a laboratory investigation of how iron and oxygen evaporate from a wüstite melt. To account for certain isotopic measurements, Wang and colleagues suggested that oxygen collects in bubbles, which may have been trapped when the spherule froze. Hydrogen, nitrogen, and H$_2$O are other possibilities for bubble-making gases.

The pore spaces assume various shapes, but most of them have roughly cylindrical rather than spherical symmetry. One possible explanation is that the molten spherules were oriented with residual cores to the front and that they spun or precessed about an axis parallel to the trajectory. The spin would tend to drive the mass to the exterior and any voids toward the center, more or less as observed. This scenario implies, however, that a significant portion of the void space formed while the spherules were still molten.

**Nuggets**

Both KK1-98-2 and -6 lack metal cores but contain high-attenuation nuggets located near the surface. Based on this and other observations (Bonté et al. 1987; Brownlee et al. 1984), it seems likely that nuggets must occur widely in type I spherules that have undergone complete oxidation of Fe. The elements more resistant to oxidation, which persist as metal, may have moved toward the front of the spherule by virtue of their high density.

Were the nuggets molten? Brownlee et al. (1984) and Bonté et al. (1987) describe nuggets as spherical based on their circular appearance in polished sections; spherical shapes imply a melt. Our SEM image of the nugget in KK1-98-6 deviates from circularity by about 10%. Deviations of this magnitude, however, do not readily comply with the 3D figures of nuggets shown in the tomographs of Feng et al. (1999). Artifacts related to the high attenuation values could affect the shapes of the nuggets inferred from the tomographs.

On the other side, the general assumption of sphericity may merit closer examination. The scarcity of apparently non-spherical nuggets in published work could reflect selection effects or observational difficulties. For example, because of its thinness, the boomerang-shaped nugget in KK1-98-2 would have been difficult to find in any petrographic section not cut at precisely the right angle. Second, the data of Feng et al. (1999) show substantial variations in attenuation close to the nugget. With peripheral, matrix-rich regions (i.e., voxels containing both oxides and PGE) removed, the objects appear nearly spherical. Finally, we note that the PGE nuggets may freeze out of solution while under the stress of deceleration and might therefore take on non-spherical shapes. Non-spherical, PGE-rich objects have been seen in a few type-I spherules. Bonté et al. (1987) observed irregularly shaped, “disequilibrium” nuggets in magnetite and remarked specifically on their “unexpected geometrical habit.” Their relevance here is doubtful, however, as they occur in magnetite interior to the spherule, rather than at the edge.

**CONCLUSIONS**

With synchrotron CMT one can distinguish and map non-destructively internal porosity, metal cores, and PGE-rich nuggets in type-I spherules. In four of 24 spherules tomographed, we find evidence for metallic cores with diameters ranging from 57 µm to 143 µm, all eccentrically positioned and grazing the surface, just as seen in images of polished sections. Most of the spherules do not seem to have metallic cores, either because they were lost or because oxidation was complete.

Most spherules have some internal pore space. The pore space varies in shape from conical to spherical to ellipsoidal and, in degree of continuity, from small lacy, branching channels to larger holes. The pore space often connects to the surface, but it is not clear that it always does so. We believe that the pore spaces formed mainly as the spherules cooled and contracted. In some cases, exsolution of dissolved or reacted gas may also have produced porosity. Less probably, undersea corrosion could have occurred where metal remained and the contraction features afforded a point of entry. To check this point, one might search void peripheries for ions common in seawater.

Micrometer-scale spatial variations in the X-ray attenuation properties of two PGE-rich nuggets indicate compositional gradients, which may mirror (on a smaller scale) the systematic spatial variations in Ni reported by Kosakevitch and Disnar (1997). The elements more resistant to oxidation, which persist as metal, may concentrate toward the front of the spherule by virtue of their high density.

X-ray tomography of type I spherules captures the features mapped with optical microscopy and SEM. As the spatial resolution of the tomography improves, the 3D information it supplies should lead to a better understanding of the dynamic state of the type I spherule melts in the moments before they froze. Questions for the future include the absolute abundance of nuggets rich in platinum group elements, a matter that bears directly on the composition of the extraterrestrial precursors of the spherules; the fine structure of any rims, which may indicate total degree of oxidation (Engrand, personal communication); and the fine structure of the void space, which could help decide whether the spherules formed by processes that released gases.

Other potential targets for tomography among extraterrestrial materials include the siliceous micrometeorites. Genge and Grady (1998) argue for
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horizontal reduction of oxidized Fe to metal in silicaceous micrometeorites during passage through the Earth’s atmosphere. X-ray tomography should make it possible to test this assertion rapidly and non-destructively.

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