Tailoring sphere density for high pressure physical property measurements on liquids

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(Received 31 October 2000; accepted for publication 7 January 2001)

We present a new method of tailoring the density of a sphere for use as a probe in high pressure-temperature physical property experiments on liquids. The method consists of a composite sphere made of an inner, high density, metallic, spherical core and an exterior, low density, refractory, spherical shell or mantle. Micromechanical techniques are used to fabricate the composite sphere. We describe a relatively simple mechanical device that can grind hemispherical recesses as small as 200 μm in diameter in sapphire and as small as 500 μm in diameter in ruby hemispheres. Examples of composite spheres made with a Pt or WC core and Al_2O_3 shell used in metallic liquids pressurized to 16 GPa and 1900 K are shown. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1351838]

I. INTRODUCTION

Physical properties of materials under conditions of high pressure (P) and temperature (T) provide important insight into the composition, evolution, present state, and dynamic processes occurring within the deep interior of Earth and other planets. The determination of some physical properties such as liquid density and viscosity poses special challenges because of the limited methods that are currently available to measure them under the spatial confinement of pressurized environments. Yet, they are among the most fundamental properties of any liquid and provide anchors for our understanding of basic processes in the interior of planetary bodies.

One method that has been used repeatedly for many years to measure the density and viscosity of silicate melts and metallic alloy liquids at high static pressures is the moving sphere method.[1–3] For both properties, a sphere of a known density is placed in the sample and, because of its contrasting density with the density of the liquid sample at the experimental conditions, the sphere either rises or falls through the molten sample. In the case of density, the direction of motion of the sphere indicates a relative density of the sample with respect to the sphere and several experiments are used to bracket the density of the sample. In the case of viscosity, some timing mechanism is employed in order to derive a sphere velocity through the liquid. The velocity of the sphere in turn provides a viscosity, if the sphere and liquid sample densities are known, through the well-known Stokes equation. In both types of experiments, there are several constraints placed on the choice of sphere material. The sphere material must withstand the experimental P and T and not react with the sample. In addition, the sphere must be of appropriate density to allow accurate determination of its velocity, by some means, in the case of viscosity measurement, and to allow narrow bracketing in the case of density measurement. The material must also be shaped into a sphere for direct application of Stokesian theory. There are other constraints that are specific to the method applied, such as the electrical resistivity contrast between the sphere and sample when the electrodetection method of sphere velocity measurement is used[4] or the x-ray absorption contrast when using the radiography technique.[5] In this article, we report on a new method of a composite sphere, as well as its construction, that allows the composite density to be chosen by selective assignment of the core and mantle radii.

II. METHOD FOR GRINDING CONCAVE RECESSES IN SUBMILLIMETER DIAMETER HEMISPHERES

We have devised a simple mechanical method for grinding a concave recess in submillimeter sized hemispheres. In our application, we have used sapphire (Al_2O_3) and ruby (Al_2O_3+0.5%Cr^{3+}) as the material to be ground but as will become apparent, the technique is applicable to virtually any solid, cohesive material. The method utilizes a small milling machine and a shop-made, powered, vertical rotary spindle, shown in Fig. 1. Any small milling machine may be used provided the spindle can be tilted to an angle of ~30° from vertical. A suitably sized diamond burr (from any dental tool supplier) is used as the grinding tool and is mounted in the milling machine spindle using a ''ww'' style watchmaker's collet to maintain concentricity with the hemisphere. The reasons for grinding at an angle are (1) the surface to be ground is in contact with more diamond particles than if the burr were vertically oriented where no cutting action would result from the rotation of a single diamond particle on the center point; (2) the shape of the recess more closely approximates the radius of the burr tip as opposed to the conical shank of the burr; (3) concentricity of the recess and the hemisphere is maintained; and (4) the diameter of the recess can be easily regulated using finely controlled translation of the burr along the x and y axes of the milling machine.

The vertical rotary spindle, shown in an enlarged view in Fig. 2, consists of an aluminum body with a shaft riding in

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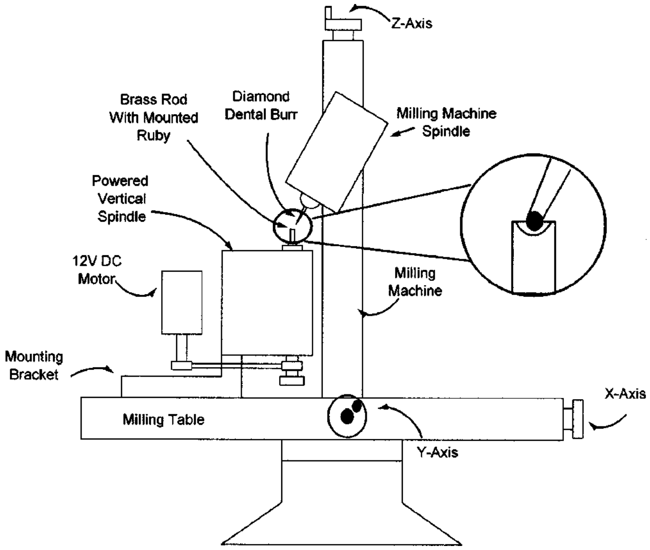

FIG. 1. Schematic drawing of the sphere grinder. The inset in the circle at the right shows an expanded view of the hemisphere mounted in a brass rod with the diamond burr tip grinding a hemispherical recess.

bearings which in turn is driven by a small 12 V dc motor via pulleys and a round polycord belt. The shaft is hollow with a tapered top to accept a ww style watchmaker's collet and drawbar. A brass rod of ~3 mm diameter is faced and then drilled in a small lathe exactly on center in one end with a diameter matching that of the hemisphere and to a depth equaling one half diameter of the hemisphere to be ground. The ruby hemisphere is then attached to the rod by a strong glue (e.g., any cyanoacrylate based adhesive) so that the flat surface of the hemisphere is flush with the face of the brass rod. When chucked in the collet, the spindle is then rotated counterclockwise at a speed of ~150–200 rpm using a 12 V dc variable power supply. The diamond dental burr is mounted into the milling machine spindle using a ww collet and is lined up on the center of the ruby hemisphere, with the aid of a microscope, using the *x*- and *y*-axes handwheels of the milling machine. The *z*-axis handwheel is then used to make contact with the ruby with the milling machine running

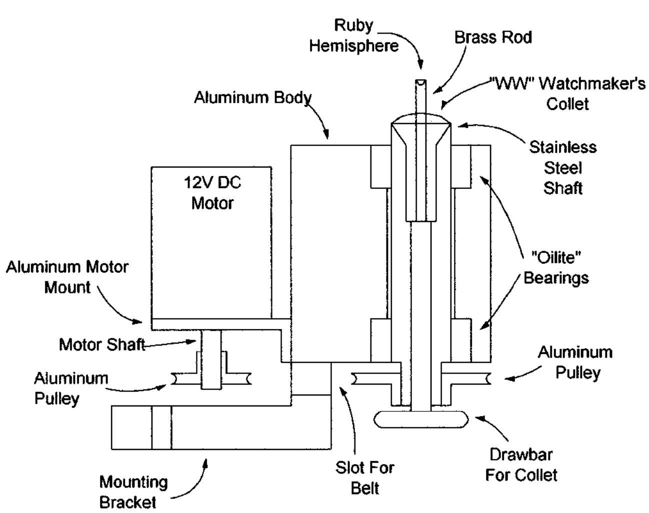

FIG. 2. Schematic drawing detailing the components of the powered vertical spindle.

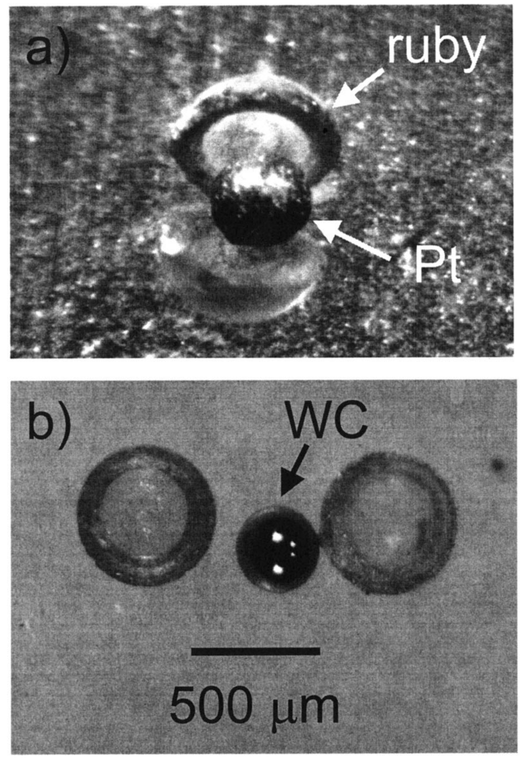

FIG. 3. Examples of two sets of ruby hemispheres with recesses, ground to accept inner spheres made of (a) Pt and (b) tungsten carbide, shown prior to assembly of the composite sphere.

clockwise at the fastest speed of our Sherline milling machine, 2800 rpm. Water is used as the grinding lubricant. The *z*-axis handwheel is then advanced 0.0005 in. at a time until the desired depth is reached. The ruby hemisphere, with recess complete, can be removed by adhesive-specific debonder or acetone.

Examples of ruby hemispheres with ground recesses are shown in Fig. 3. The reproducibility of this grinding technique allows two hemispheres with very closely matching recesses to accept a core made of Pt or WC. The core sphere is made using an air driven grinder of the Bond type.[6] The two mantle hemispheres are held together using a thin layer of high temperature Al_2O_3-based cement placed on the opposing rims of the hemispheres.

III. TESTS OF COMPOSITE SPHERE AT HIGH PRESSURES AND TEMPERATURES

Several tests of assembled composite spheres pressurized and heated to high temperatures within a Fe–S matrix have been carried out. Examples of Pt core in sapphire spheres are shown in Figs. 4(a) and 4(b). Separate tests of pressurization in multianvil presses in octahedral cells to 3 GPa at room temperature [Fig. 4(a)] and to 16 GPa followed by heating to 1900 K [Fig. 4(b)] show the well-maintained spherical form of the spheres following recovery and sectioning. In two of the three spheres shown, the outer shell is spherical and contains the inner spherical core of Pt. In the sphere on the right side of Fig. 4(b), the depth of grinding is not sufficient to see the core. Electron microprobe analysis

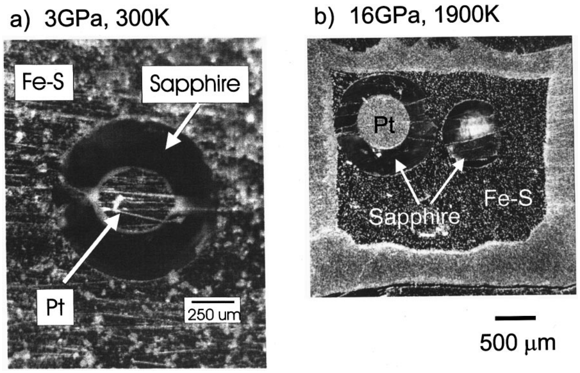

FIG. 4. Examples of recovered and sectioned composite spheres after exposure to high pressure and high temperature conditions. In both cases, the spheres are encased in a mixture of iron and sulfur. The cross-sectional views show the result of compression only in (a) and the result of compression and heating (to above iron–sulfur melting temperatures) in (b). The center of the sphere on the right in (b) is located in a deeper plane than the plane of the section and does not show the inner metallic sphere.

on recovered samples shows the spheres remain sealed by the lack of any contamination of the core, mantle, and surrounding sample.

We have also tested the composite sphere design under pressure on a synchrotron beam line with the intention of imaging, by radiography, the high absorption core sphere material in a falling sphere viscosity experiment. Figure 5 shows an example of a 280 μm Pt core sphere within a 500 μm ruby mantle as it falls through a liquid of composition Fe–10 wt %S. It is clear that despite the poor contrast of the ruby mantle, shown only by a faint halo around the dark Pt core, the ruby mantle remained intact.

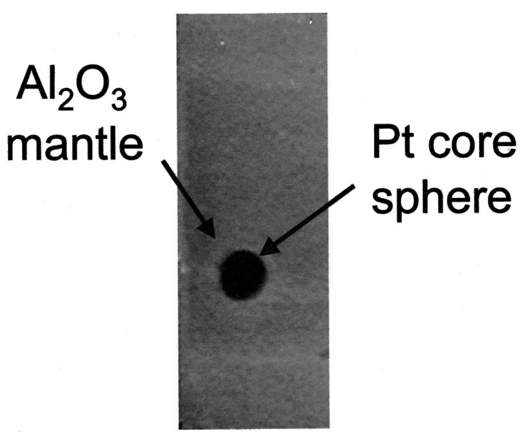

FIG. 5. Example of a 280 μm diam Pt sphere core within a 500 μm diameter ruby mantle as seen through a high pressure pyrophyllite cubic cell with synchrotron radiation. The ruby mantle is barely distinguishable as a halo in the image. The material surrounding the composite sphere is molten Fe–10 wt %S. The image was taken at pressure and temperature conditions of 2.6 GPa and ~1200 °C.

IV. DISCUSSION

Our method to produce spheres with densities that can be tailored also has the advantage that the outer shell material provides a means by which to insulate the inner core material from reaction with the matrix material. This is a critical consideration in high pressure viscosity and density experimentation that is sometimes overlooked. In one radiography experiment with an improperly closed composite sphere, the sudden rupture of the ruby mantle exposed the Pt to the very soluble Fe–S melt and the ~300 μm Pt core sphere diffused into the melt within 1–2 s.

The micromechanical device to drill hemispherical recesses into hemispheres as small as 500 μm described here has been used on more than 150 hemisphere grindings. The stability of composite spheres produced by this method, tested in several high pressure and temperature experiments with follow-up chemical analyses on postrun samples, shows that a properly cemented composite sphere system remains closed under extreme pressure and temperature conditions. This method provides a way of exploring a greater range of liquid viscosities and liquid compositions at high pressures and temperatures than is currently available.

ACKNOWLEDGMENTS

One of the authors (R.A.S.) gratefully acknowledges the Alexander von Humboldt Foundation of Germany for a fellowship and the members of the Bayerisches Geoinstitut for their generous hospitality during the early stages of this work. He thanks Hubert Schulze for his technical contributions to the the early stages of this work. Part of the work was supported by an NSERC grant to that author. Part of this work was performed at GeoSoilEnviroCARS (GSECARS), Sector 13, Advanced Photon Source, Argonne National Laboratory. GSECARS is supported by the National Science Foundation, Earth Sciences, Department of Energy, Geosciences, the W. M. Keck Foundation and the U.S. Department of Agriculture. Use of the Advanced Photon Source was supported by the U.S. Department of Energy, Basic Energy Sciences, Office of Energy Research, under Contract No. W-31-109-Eng-38. The authors thank T. Uchida of GSECARS for his help in obtaining the synchrotron images and Adam Jones for help with the figures.

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