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Fabrication Capabilities for Spherical Foam Targets Used in ICF Experiments*

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Abstract—This paper reviews the processes developed at General Atomics (GA) in the past several years to fabricate a variety of spherical foam targets at various densities for the inertial confinement fusion (ICF) community. The two most common chemical systems used to produce spherical foam targets have been resorcinol-formaldehyde (R/F) aerogel and divinylbenzene (DVB). Spherical targets have been made in the form of shells and beads with diameters ranging from approximately 0.5 to 4.0 mm, and densities from 100 mg/cc to 250 mg/cc, with typical high yield of intact shells or beads of 90%–95%. Permeation barriers have been developed and deposited on both R/F and DVB shells. We have also made R/F foam shells with higher pore size (0.10–0.50 μm) in order to increase the cryo-fill fraction when these shells are cryogenically layered with D_2 . Another spherical target that is currently under development that will also be discussed is silica aerogels shells and beads. Other foam target materials currently under development, such metal doped R/F aerogel beads for extreme ultra violet (EUV) source experiments will also be discussed.

I. INTRODUCTION

This paper will discuss the variety of spherical foam targets we have developed over the past several years for a variety of ICF experiments, such as R/F, DVB, silica aerogels, as well as metal-doped R/F. Foam shells are needed for and have been used on current direct drive experiments on OMEGA at the Laboratory of Laser Energetics (LLE) and for future experiments on the National Ignition Facility (NIF). The shells needed for OMEGA experiments require diameters of ~ 800 – $900 \mu\text{m}$. These are scaled down versions of the targets that are required for high gain wetted foam direct drive ignition designs on NIF [1]. The required shell diameter for OMEGA experiments is ~ 800 – $900 \mu\text{m}$. The required wall thickness and foam density depend on the application. Cryogenic targets require a wall thickness of 50–100 μm with a foam density of 30–150 mg/cc. The room temperature foam shell surrogates, mimicking the D_2 or DT ice layer need to have a wall thickness of 80–120 μm and a foam density of 180–250 mg/cc. These targets are designed to reduce the need for difficult to field cryogenic shots with the foam serving to simulate the cryogenic fuel layer.

An important requirement for these foam shells is the wall uniformity. The wall uniformity specifications for these OMEGA targets are usually quoted in terms of nonconcentricity defined in Eq. (1), respectively. For OMEGA size wetted foam shells the NC must be $\leq 5\%$.

$$\text{NC} = \frac{\text{Offset of the Inner - Outer Wall}}{2 (\text{Avg. Wall Thickness})} \quad (1)$$

A pinhole free permeation barrier is needed for both the room temperature surrogates and for the cryogenic application in order to retain the gas and avoid evaporation of the ice layer, respectively.

Standard small pore ($< 100 \text{ nm}$) R/F formulation has been used for fabricating these shells due to their optical transparency. However, fabrication of large pore foam shells has also become necessary in order to increase the cryo-fill fraction when these shells are cryogenically layered with D_2 . These larger foam shells have been fabricated by developing formulations of R/F foam suitable for shell manufacturing and also by using DVB foam which characteristically has larger pores (approximately few microns). Silica aerogels have also been fabricated as direct drive designs also include this material for the foam shell.

As we will discuss below, the key factors in shell fabrication using any of these formulations are proper chemistry of the foam/aerogel system and proper agitation of the shells/beads during the curing process. Besides direct drive experiments, spherical foam targets have other applications such as x-ray drive symmetry, high energy physics experiments and Extreme Ultra Violet (EUV) source development. These targets often need metal doping. We have developed and will discuss the fabrication of metal-doped beads for EUV source experiments in this paper [2].

II. R/F FOAM SHELLS FOR LLE

R/F was chosen as the system of choice for foam shell fabrication because of its characteristically small pores ($\approx 0.10 \mu\text{m}$), making the foam transparent. This allows use of routine characterization, which is used for full density polymer shells.

The R/F foam/aerogel was first developed by Pekala, et al. [3]. The R/F foam is formed by a polycondensation reaction using a two-step (base/acid catalysis) gelation process and is described in [3–5]. Fabrication of R/F foam shells was developed by Lambert, et al. [4]. We have extended this work by transforming the process into a repeatable, reliable, high yield production process to fabricate 100 mg/cc R/F foam shells [5]. Two factors that improved the yield of shells were the gentle agitation of the shells during the curing process, and changes in the chemistry that slowed down the gelation time. This improved the yield of intact shells from 30–95%. Slowing down the gelation time also improved the non-concentricity of the shells by giving the shells time to center. By modifying the gelation time, ~ 30 – 50% of the shells meet the NC specification of $< 5\%$.

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As stated earlier, a pinhole free permeation barrier needed to be developed and deposited on the foam shells in order for the shells to hold a fill gas such as D_2 or DT. The permeation barrier that was chosen was glow discharge polymer (GDP) [5]. It took approximately 3-4 μm of GDP to create a pinhole free permeation barrier. The shells were tested for gas retention by filling them with argon (Ar), and analyzing the shells by X-ray fluorescence (XRF), by following the decay of Ar signal in the shells as a function of time. This has become a standard gas retention technique for the overcoated foam shells. Approximately 60-70% of the shells were gas retentive. Developing a production process and permeation barrier for 100 mg/cc R/F foam shells enabled the first cryogenic foam shots in the U.S., which produced a yield over clean (YOC) of 100% [6].

We have extended this work to develop a production process for higher density R/F foam targets (180-250 mg/cc). Again, the gelation time played a key factor in fabricating a high yield of intact shells with good wall uniformity. A gelation time that was too slow or too quick resulted in a low yield of intact R/F shells with poor surfaces and bad wall uniformity [Fig. 1(a)]. Because the gelation behavior of the R/F precursor played a factor in the fabrication of a high yield of intact shells, the gelation time and behavior of the R/F precursor solution was checked by measuring its viscosity at its curing temperature of 70°C over time by using a rheometer (Bohlin CVO50 from Bohlin Instruments). The results of the gelation studies of several high density R/F precursors from the rheometer show that a gelation time similar to that of the 100 mg/cc R/F foam shells resulted in high yield of intact shells without wrinkled surface finish which was another usual failure mode in fabrication of these targets [Fig. 1(b)]. The results of the rheometer studies on the high density R/F precursor are shown in Fig. 2.

Factors that helped produce shells with good wall uniformity was a slight density mismatch of the inner oil (O1) droplet with the R/F solution, as well as a gelation time that would give the shells time to center was also key in producing shells with good wall uniformity and NC. The densities of the R/F precursor solution were precisely measured on a densitometer (Anton Paar, DMA 4500). The densitometer is accurate to 0.05 mg/cm^3 and can measure densities at temperature range of 0 to 90°C. Approximately 20-40% of the shells analyzed meet the NC specification of $\leq 5\%$ depending on the density mismatch between the O1 and the R/F precursor solution (Fig. 3).

GDP was also used as the permeation barrier for these high-density R/F shells, and the gas retention of these shells are similar to the 100 mg/cc R/F shell. It required approximately 3-4 μm of GDP to create a pinhole free permeation barrier with the addition of a thin coating of aluminum ($\sim 1000\text{\AA}$) as a barrier against deuterium at OMEGA [7]. These pinhole free room temperature deuterium filled foam shells were shot at OMEGA for the first time in the U.S. These experiments will continue at LLE in the future.

III. SILICA AEROGEL SHELLS

Another alternative to the R/F shells for direct drive experiments on OMEGA is Silica aerogel shells. The silica aerogel synthesis begins with an alkoxide or inorganic precursor that is made to gel via a catalyst. Silica aerogel (SAG) shells and beads have been recently fabricated at GA.

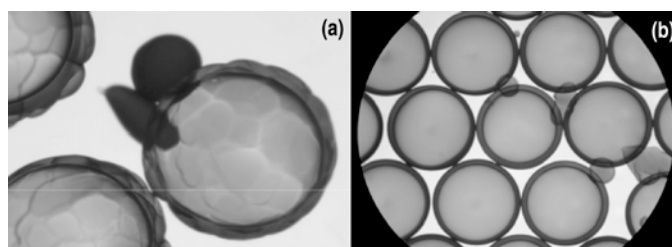


Figure 1. (a) 180 mg/cc R/F shells with improper gelation, this leads to shells with bad surfaces (soccer ball type shells). (b) 180 mg/cc R/F foam shells with proper gelation, which produces good surfaces and higher yield of intact shells.

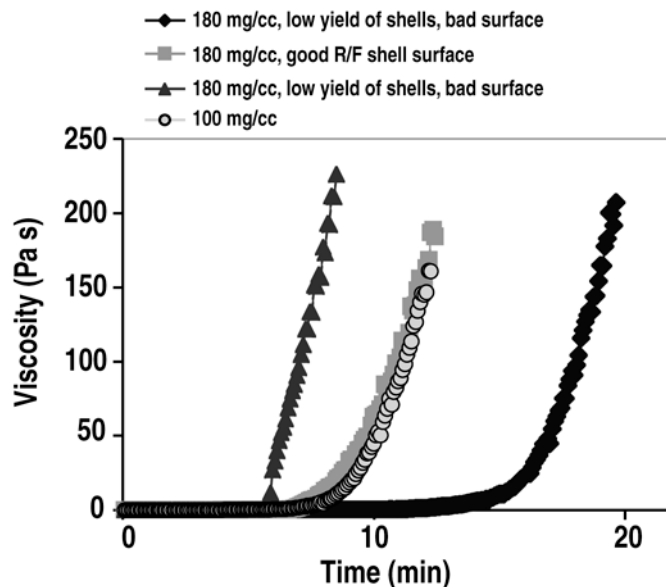


Figure 2. Viscosity readings of different high-density R/F precursors and the optimal one for low density shells at the curing temperature 70°C. As the precursor starts to gel, the viscosity increases. The R/F precursors that gel too fast or slow compared to the low density case resulted in a low yield of shells with poor surface and wall uniformity. The high-density R/F precursor that is similar to the 100 mg/cc low-density R/F precursor results in a high yield of shells with unwrinkled surfaces.

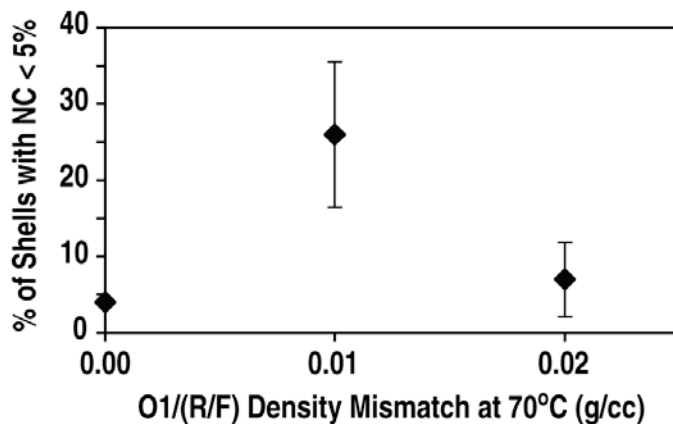


Figure 3. The yield of shells with the specification of NC $< 5\%$ is highest when the O1 is mismatched in density with the 180 mg/cc R/F solution at the curing temperature of 70°C by 0.01 g/cc.

Beside direct drive experiments, SAG beads can be used for x-ray drive symmetry experiments. A picture of these shells is shown in Fig. 4, the picture shows a batch of 100-120 mg/cc OMEGA size shells that are transparent [2]. Fig. 5 shows a picture of 100-120 mg/cc SAG beads, these are also transparent as in the case of other foam shells such as R/F, the SAG shells/beads were fabricated by using the triple orifice droplet generator. The droplet generator allows for precise control of the shell diameter and wall thickness. A surfactant and oil system that was compatible with the SAG precursor was needed in order to fabricate SAG Shells and beads. This creates a stable emulsion that did not agglomerate. A proper gelation time of the SAG solution allowed time for the shells to center properly in order for the shells to have good wall uniformity. The shells were characterized by visual characterization techniques for wall uniformity. Results show that 65% of the shells have a 4π NC of $<5\%$ (Fig. 6). Though the yield of shells that meet the wall uniformity specification is not 100%, it is a sufficient enough yield to provide targets for these experiments because each batch contains thousands of shells. With a stable emulsion and proper gelation time, SAG shells as well as beads were successfully fabricated. Details of fabrication of these shells are given in [2]. Future work will concentrate developing a gas permeation barrier for the SAG shells.

IV. LARGE PORE FOAM SHELLS

A direct extension of the work on R/F foams shells for direct drive experiments was fabrication of large pore foam shells in order to increase the cryo-fill fraction when these shells were cryogenically layered with D_2 . Optical clarity of the D_2 ice layer was severely reduced when the standard R/F foam shells were cryogenically layered [8]. It was theorized that because the pore size of standard R/F foam is <100 nm, it can lead to under-filling of the D_2 . This would cause voids or micro-ice structures that scatter light, reducing the clarity of the ice layer. If larger pore foams were used, it could increase the cryo fill fraction, which could lead to a clearer ice layer. We investigated two ways to produce large pore foam shells. First, was increasing the pore size of the R/F foam, and secondly, to use a foam system that characteristically has large pores such as DVB.

A. Large Pore R/F Shells

Changing the pore size of the R/F foam by changing the processing conditions had been successfully demonstrated previously [9,10]. The traditional technique involved reduction of the base catalyst concentration in the R/F formulation. The base catalyst (carbonate) allows the reaction of the resorcinol (R) and the formaldehyde (F). Using this technique has a dramatic effect on the pore size (Fig. 7). We used that technique, as well as a using a new process involving the changing the acid concentration in the formulation [11]. The acid in the R/F formulation is the curing catalyst which helps the aggregation of the R/F clusters to form a foam network. Increasing the acid concentration in the R/F formulation successfully increased the pore size of the R/F (Fig. 7). These large pore R/F foams were also measured by nitrogen absorption and the results show that the surface area increases as pore diameter decreases due to the increase in volume occupied by larger sized pores, this is a good indication that the pore size of the foam increased (Fig. 8). Details of the characterization of the pore sizes obtained can be found in [11].

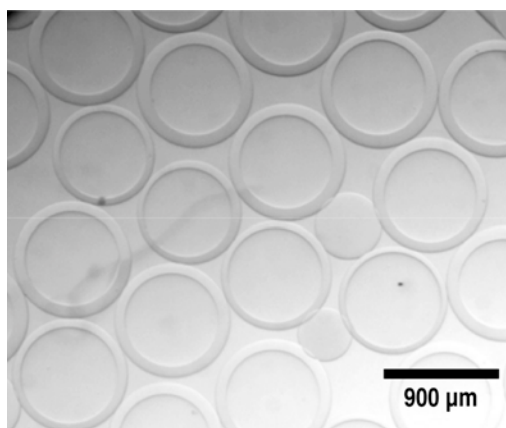


Figure 4. These are ~ 900 μm silica aerogel shells (~ 100 - 120 mg/cc) produced by the microencapsulation process using a droplet generator.

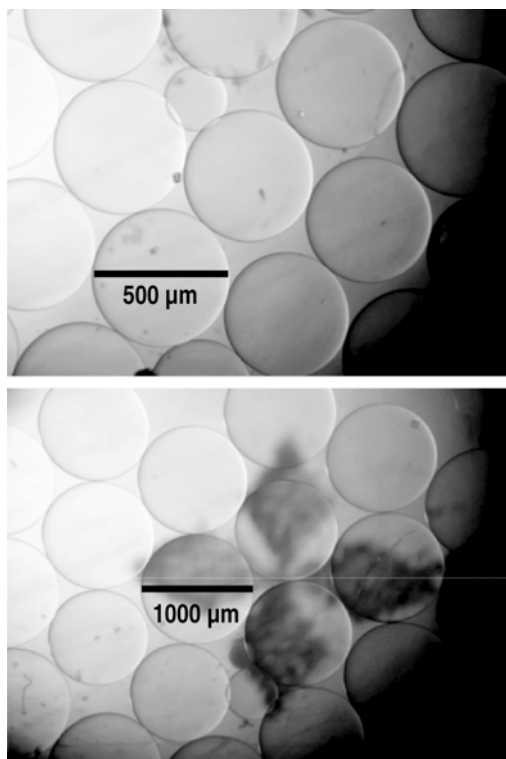


Figure 5. These are silica aerogel beads (~ 100 - 120 mg/cc) produced by the microencapsulation process using a droplet generator.

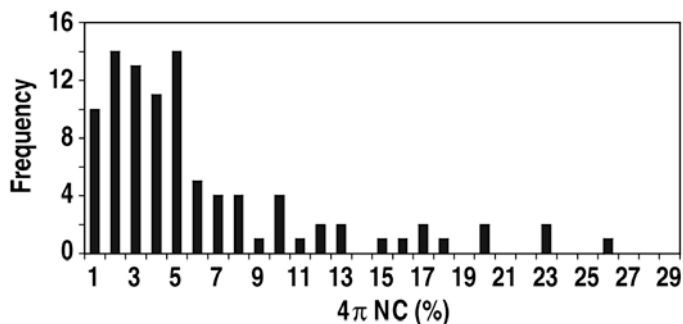


Figure 6. Histogram of the 4π NC of OMEGA sized (800 - 1000 μm) Silica aerogel shells. Approximately 65% of the shells produced meet the target specification on NC (4π NC $< 5\%$).

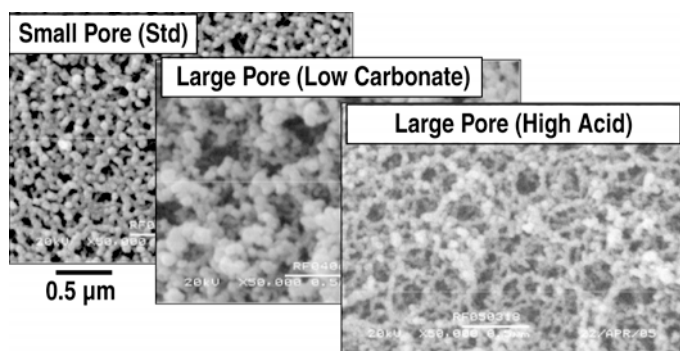


Figure 7. SEM images of the different R/F formulations. Lowering the carbonate, or increasing the acid concentration in the R/F formulation can change pore size.

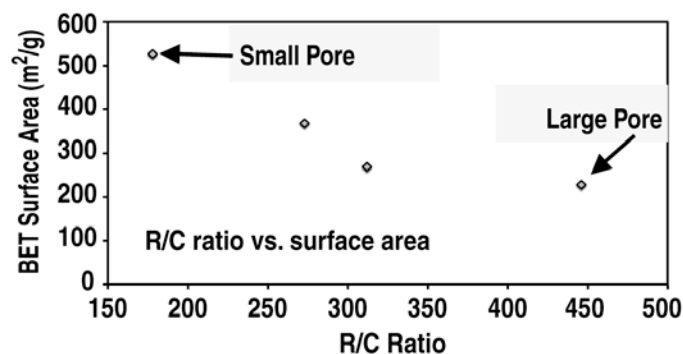


Figure 8. Nitrogen absorption data of the Large pore and standard (small) pore R/F foams. This shows that the surface area increases as pore diameter decreases due to the increase in volume occupied by larger sized pores.

The next challenge was to extend this work to shell fabrication. Both techniques described above lead to rapid gelation of the R/F solution. Rapid gelation of the R/F solution leads to three adverse effects to the fabrication of large pore R/F foam shells. The first effect was that rapid gelation of the R/F solution led to a low yield of intact shells and poor NC. In order to produce shells, the base catalysis time and the curing temperature were both decreased. These modifications slowed down the gelation time, giving the shells time to center, which resulted in a high yield of intact shells with good wall uniformity and NC [11]. Large pore R/F foam shells were successfully fabricated using both pore increasing techniques. The second problem with this formulation was reduction of density of final dry foam shells (~ 70 mg/cc) compared to the intended formulation (~ 100 mg/cc) due to inability of the all the constituents reacting [11]. Finally, these shells required over $10 \mu\text{m}$ of GDP coating in order to fully seal the foam due to the very large pore size. Despite these issues with the shells fabricated with the traditional technique, the shells were sent to LLE and they were cryogenically layered and better visibility of the ice layer in the shells was reported [8].

These issues were the motivation for using the acid modification technique in addition to the traditional base modification method. The shells produced by using the acid modification were more transparent and could be characterized by optical characterization techniques. They also required a thinner layer of GDP ($4\text{--}5 \mu\text{m}$) to create a pinhole free permeation barrier, unlike the shells made with the carbonate modification, which required a

GDP coating of $10 \mu\text{m}$. The acid modified large pore R/F shells lead to a final dry shell density, which was consistent with the intended calculated value. Large pore R/F shells fabricated with the acid modification still need to be cryogenically layered to see if it can create a more transparent ice layer.

B. DVB Foam Shells

DVB foam is another alternative to produce large pore foam shells. DVB shells have also been developed and previously fabricated [12]. This traditional way to fabricate DVB foam shells creates shells that crack and implode during the fabrication process; this leads to a low yield of intact shells. We improved the yield of this process using a dual initiator system which led to more robust shells which can survive the fabrication process [13]. The DVB shells that were produced met the NC specification of $<5\%$ and had a density of 100 mg/cc .

GDP could not be used as the permeation barrier because it could not close the large pores of the DVB foam at thicknesses of a few μm s. Polyvinylphenol (PVP) has been used as an overcoat on other large pore foams in the past and is deposited on the shell by a polycondensation reaction [5,14]. One issue in using the PVP is that there is a 20% diameter shrinkage when $5 \mu\text{m}$ or more is coated on the foam. This could lead to densification of the foam shell, which is not desired. When $<3 \mu\text{m}$ of PVP is coated there is little to minimal shrinkage of the diameter, but it was found that a $3 \mu\text{m}$ coating could not provide a pinhole free permeation barrier. Therefore, the permeation barrier that was used was a dual coatings of PVP and GDP (Fig. 9) [13]. The DVB shell was coated with $2\text{--}3 \mu\text{m}$ of PVP, this helped fill most of the larger pores with minimal shrinkage, the rest of the pores were filled with a $3\text{--}4 \mu\text{m}$ coating of GDP. The shells with the dual coatings were tested for gas retention using XRF and $\sim 70\%$ of the shells had a pinhole free permeation barrier and were gas retentive.

V. METAL DOPED BEADS

A number of metal doped and metallic aerogels are currently under development in our laboratory including tantalum oxide and tin oxide [2]. Besides metal oxide aerogels, 100 mg/cc R/F foam has been loaded with Sn by suspending Sn nanoparticles. While most of these samples have been made in planar format, we have extended this to fabrication of beads by microencapsulation of the Sn loaded 100 mg/cc R/F solution. The Sn appears to be dispersed uniformly [Fig. 10(b) SEM picture with nice dispersion] if proper dispersion of the particulates is achieved by using aggressive ultrasonication (Fig. 10). Beads have been successfully made in diameter range of $300\text{--}600 \mu\text{m}$ doped with $0.1\text{--}1.0\%$ of full density Sn for EUV source development experiments [2].

VI. CONCLUSION

Foam shells were requested, developed and fabricated for various types of ICF experiments. Spherical foam targets in the form of shells and beads were fabricated for direct drive experiments on OMEGA with various types of foam/aerogel systems including R/F, DVB, and silica aerogels at densities of $100\text{--}250 \text{ mg/cc}$. Proper gelation time has been found the key in producing high yields of intact shells with good wall uniformity.

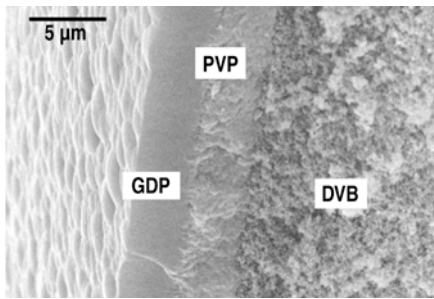


Figure 9. Cross section of a 100 mg/cc DVB foam shell (~900 μm) that is overcoated with PVP/GDP. ~70% of the OMEGA sized (800-1000 μm) shells coated this dual overcoat produced a pinhole free permeation barrier

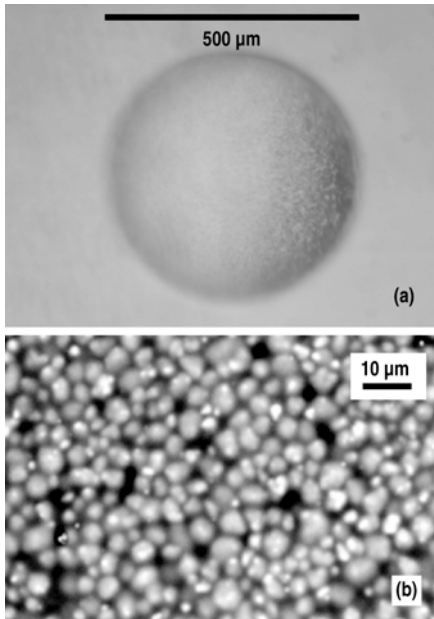


Figure 10. (a) 100 mg/cc R/F foam bead, doped with 1% full-density Sn fabricated by the microencapsulation process. (b) Backscatter SEM image of this Sn doped R/F foam bead. The SnO_2 particles (which appear as bright areas) in the R/F foam appear to be evenly dispersed, with the SnO_2 particles being ~5 μm .

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