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Density Measurements of Be Shells

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From: Bob Cook

Subject: Density measurements of Be shells

The purpose of this memo is to lay out the uncertainties associated with the measurement of density of Be ablaters by the weigh and volume method. I am counting on the readers to point out any faulty assumptions about the techniques or uncertainties associated with them. Based on the analysis presented below we should expect that 30 μm thick shells will have an uncertainty in the measured density of about 2% of the value, coming more or less equally from the mass and volume measurement. The uncertainty is roughly inversely proportional to the coating thickness, thus a 60 μm walled shell would result in a 1% uncertainty in the density.

Let me quickly go through the error analysis involved when making density measurements on Be shells. Clearly the density is

$$\rho = m / V \quad (1)$$

where m is the mass of the Be and V is its volume. Thus the uncertainty in the density, $\delta\rho / \rho$, is

$$\frac{\delta\rho}{\rho} = \frac{\delta m}{m} + \frac{\delta V}{V} \quad (2)$$

where $\delta m / m$ and $\delta V / V$ are the relative uncertainties in the mass and volume measurements.

The measurement of the mass is relatively straightforward. If the mandrel has been removed it is simply the mass of the shell, if the mandrel has *not* been removed (and this is generally the case) then one needs to know the mass of the plastic mandrel. In some cases we have known this (or at least think we do) because a relatively few mandrels were coated at one time, and their diameters, which are measured beforehand, are different enough so that after coating the shells can be associated with specific mandrels. However what is missing here is any mass loss of the mandrel during the initial stages of coating when the mandrels are exposed to relatively high temperatures and gaseous decomposition products may diffuse away. Thus in this case the mass we might use is an upper bound to the mass of the mandrel. Since we are subtracting this from the mass of the coated shell to get the Be mass, the result will be lower than the actual Be mass by the amount of the mandrel decomposition. We currently have no good estimate for the mass of the mandrel lost during the initial stages of coating. There is evidence, however, that changes are taking place, since we

have seen increases in diameter of the shells during coating (this might occur without mass loss of course). To get some idea of the magnitude of this "error" let's look at a typical shell. The mandrel weight is perhaps 180 μg . If this is coated with 30 μm of Be, the composite mass might be 850 μg , thus the mass of Be would be by difference 670 μg . But if 10 μg of mandrel were lost the correct mass would be 680 μg , or about 1.5% higher. Clearly this error increases with thinner coatings, and decreases with thicker coatings. And of course we have no reason to expect 1 μg , 10 μg , or more of loss. There may be some experiments that we can do to quantify this, and maybe some data already exists to put some bounds on it. The rate of Be deposition is about 1 $\mu\text{m}/\text{h}$, probably the loss stops if there is 10 μm of Be on the shell,¹ and the temperature reached by the shell is probably not more than 250 $^{\circ}\text{C}$. There probably is (or certainly could be) TGA mass loss data on bare mandrels heated to 250 $^{\circ}\text{C}$ that could be used to bound the effect.

The situation when individually uncharacterized mandrels are used is certainly worse than this, but under the best of circumstances not much worse. Evelyn characterizes the mass of a batch (as received from GA) by weighing a number of shells.² Typically mandrels within a batch vary in mass by about 10 μg . See Figure 1 for the characterization data on 6 batches. The standard deviations for the batches K, P, and Q were 2.9, 3.3, and 4.1 μg . Thus the uncertainty in the mass if a single batch is used is about 5-7 μg (half of the range of measured shells or about 2 standard deviations) in the mandrel mass if the batch average mass is used. Thus the effect on the Be mass is about 0.75% for a 30 μm coating.

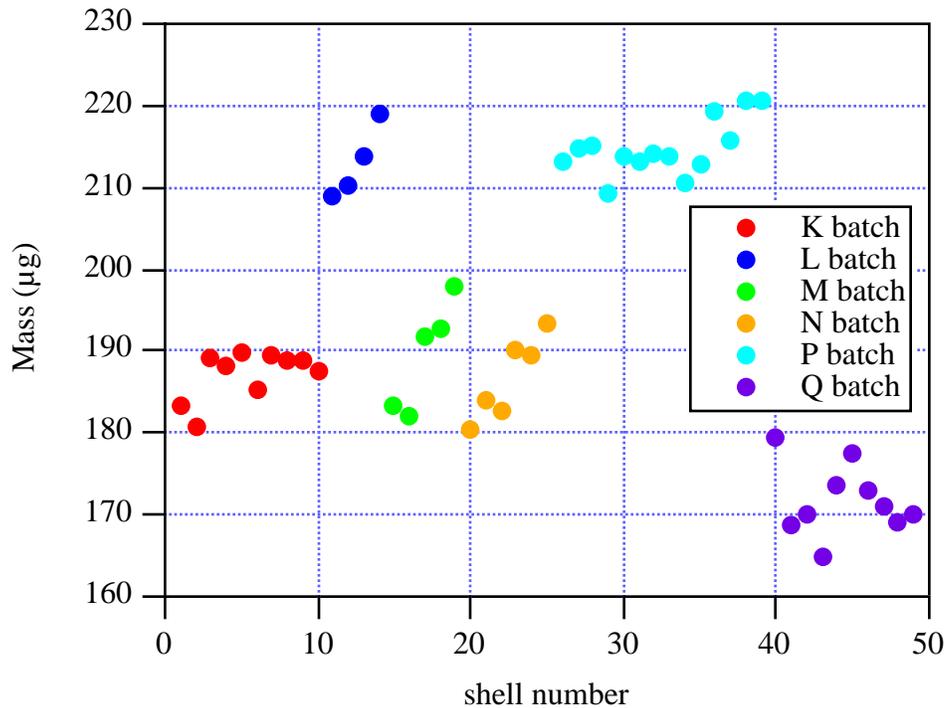


Figure 1. Distribution of masses within a batch.

¹ Some data from stove Letts showed that Be shell with 10 μm of coating and *no* drilled hole lost mass when heated in a furnace to 500 $^{\circ}\text{C}$. Those with 20 μm of coating did not.

² Of course other batch characteristics such as diameter, out of round and wall thickness are also measured.

One last small correction that should be made in the mass measurement is that there is a buoyancy correction for the masses measured for Be coated shells because they are evacuated. The mass of the equivalent volume of air is given by

$$m_{\text{buoy.}} = \frac{4}{3}\pi r_i^3 \times \frac{n \cdot M_{\text{air}}}{V} = \frac{4}{3}\pi r_i^3 \times \frac{P \cdot M_{\text{air}}}{RT} \quad (3)$$

where r_i is the inner radius (~ 0.1 cm), and M_{air} is the average molecular weight of air (~ 29 g/mol). At 1 atm and 25 °C this amounts to about 5 μg , the mass that should be added to the mass measured for the Be shell.

Thus the situation for mass is not as good as we thought, at least if the mandrel is still within. It looks like the $\delta m / m$ term in eq 2 may be approaching 1% for thin (30 μm) shells based on average mandrel mass if the mandrels used were all from the same batch. However in the "Christmas" run both batches P and Q were used, and these differed in average mass by nearly 43 μg , thus the mandrel mass in a given shell is really unknown. Using the average of the two batches insures that one will be off by about 20 μg , some high and some low. This represents a 3% relative error in the mass measurement if the shell were 30 μm thick, however since they are ~ 100 μm thick the uncertainty this brings to the mass measurement is still about 1%. The diameters of the P and Q batches were about the same, so this doesn't allow for any sorting. I suppose one could still do sorting as follows. For each shell determine the density with the mandrel mass equal to the average (193 μg) plus and minus the 21 μg "error". Looking at the results one might be able to determine whether the "plus" or "minus" (and thus the batch) was correct. But this is probably not worth the time, and the "Christmas" run was graded, which is another problem (though not serious as shown below).

Though not very useful for quick turn around, it might be worth doing some densities on capsules that have had their mandrels burned out. For these the mass measurement is good to about 0.2 μg , which for a 30 μm thick coating is 0.03%. One doesn't need to worry about the mass loss of the hole, a 6- μm -diameter hole through a 30- μm wall represents less than 10^{-3} % of the mass.

The situation for the volume measurement is as follows. Currently the best way to make this measurement is to measure the outer diameter, d , of the capsule by RACI and then to break the capsule open and measure the wall thickness, w , by SEM. In this way the Be capsule volume is given by

$$V = \left(\frac{4}{3}\right)\pi \left(\left(\frac{d}{2}\right)^3 - \left(\frac{d-2w}{2}\right)^3 \right) = \frac{1}{3}\pi w \left(3d^3 - 6dw + 4w^2 \right). \quad (4)$$

Thus the relative uncertainty in the volume is

$$\frac{\delta V}{V} = \frac{\delta w}{w} + \frac{6d\delta d - 6d\delta w - 6w\delta d + 8w\delta w}{3d^2 - 6dw + 4w^2} = \frac{\delta w}{w} + \frac{6(d-w)\delta d - 2(3d-4w)\delta w}{3d^2 - 6dw + 4w^2} \quad (5)$$

or since typically $d \gg w$

$$\frac{\delta V}{V} \cong \frac{\delta w}{w} + 2 \frac{\delta d}{d} . \quad (6)$$

With the SEM we can make the wall thickness measurement to perhaps 0.2 to 0.3 μm . Wall thickness uniformity should not really be an issue since by process the wall ought to be very uniform (assumes no sticking, etc). So several measurements of wall thickness probably say more about the uncertainty of the measurement than differences in the wall thickness. We think that the SEM picture allows measurement of the wall thickness to say 0.3 μm , or for a 30 μm wall about 1%. Several measurements averaged would decrease this number somewhat. The uncertainty in the diameter is a different matter. The shells are typically out of round by a few μm s. This is say 2 μm out of 2000 μm or about 0.1%. Evelyn does 3 orthogonal views, ellipse fits to each, and the out-of-round is the difference between the largest major axis and the smallest minor axis. These shells are pretty round!! The average diameter is taken as the average value from these 6 axes. The error introduced in using this average value and a spherical model to determine the volume is certainly tiny. As an example, in 2-D a circle with diameter of 2000 μm has an area of 3141593 μm^2 . An ellipse with major and minor axes of 2005 and 1995 μm (a 2-D out of round of 10 μm) has an area of 3141573 μm^2 , a 0.0006% difference from the value one gets by using the average diameter and the equation for a simple circle. Further Evelyn's diameter measuring method involves first using a 2 mm silicon nitride ball bearing to calibrate the RACI exposure. The ball bearing is probably good to 0.1 μm , the calibration is good to 0.15 μm ; thus we can expect the measurement of each axes to be good to 0.3 μm , or $\delta d / d$ is about 0.015%. Even if the uncertainty was 1.0 μm , the relative uncertainty would still be only 0.05%, much less than the uncertainty in the wall measurement.

Thus it appears that the uncertainty in the volume is controlled by the wall thickness measurement, and is the same order as that associated with the mass, if shells from the same batch are coated. The mass measurement (of capsules with mandrel) under the best circumstances (i.e. no loss of mandrel mass due to decomposition) is uncertain to about 1%. The volume measurement is also uncertain to about 1% due to the wall thickness measurement. Thus a measured density value of 1.70 is uncertain to about 0.04 g/cm^3 , or assuming a "full" density of 1.85 (pure Be) this means that the sample is $91.9 \pm 2.2\%$ of full density, or $8.1 \pm 2.2\%$ porosity. This is not too bad precision, and probably an acceptable diagnostic. Both of these estimates are for 30 μm thick shells, in both cases the uncertainty drops linearly with the shell thickness (i.e. for a 60 μm wall the uncertainties are about 0.5% each).

Not included in this analysis are the possibilities of determinant or constant errors. Does the SEM routinely over or under measure the wall thickness, does RACI give the right value, which I guess means is the standard silicon nitride ball actually 2 mm? And is there mass loss of the mandrel on coating?

Lastly lets look at what "full" density for Cu doped samples actually is. The specific gravity of pure Be is 1.848 and for pure Cu its 8.96 (both at 20 °C).³ Since specific gravity is the ratio of the density of the material to the density of water, and the density of water at 20 °C is 0.9982 g/cm³,⁴ the densities of Be and Cu are 1.845 and 8.94 g/cm³. There are two ways we might compute the density of Cu-doped Be. The first is to assume that Cu atoms simply replace Be atoms, and the basic crystal structure remain the same. With this assumption the density of the composite is given by

$$\text{density Cu-doped Be} = 1.845 \times \left(\text{Atom fraction Be} + \frac{63.546}{9.012} \times \text{Atom fraction Cu} \right) \quad (7)$$

Note that at an atom fraction of Cu equal to 1 (pure Cu) the resulting density is 13.01 g/cm³, clearly wrong - the crystal structure of pure Cu is not the same as Be and the Cu atom is a bit bigger. But at low atom fractions of Cu this is not a bad approximation.

The other approach one could take is a simple mixing rule, which might be expressed as

$$\text{density Cu-doped Be} = 1.845 \times \text{Atom fraction Be} + 8.94 \times \text{Atom fraction Cu} \quad (8)$$

In Figure 2 I plot the two expressions for low atom fractions of Cu. The difference between the two is pretty small at the atom % Cu we are interested in - 1.884 versus 1.870 at 0.35 atom % Cu, 1.923 versus 1.895 at 0.70 atom % Cu. My guess is that the atom substitution method (eq 7, higher numbers) is more accurate.

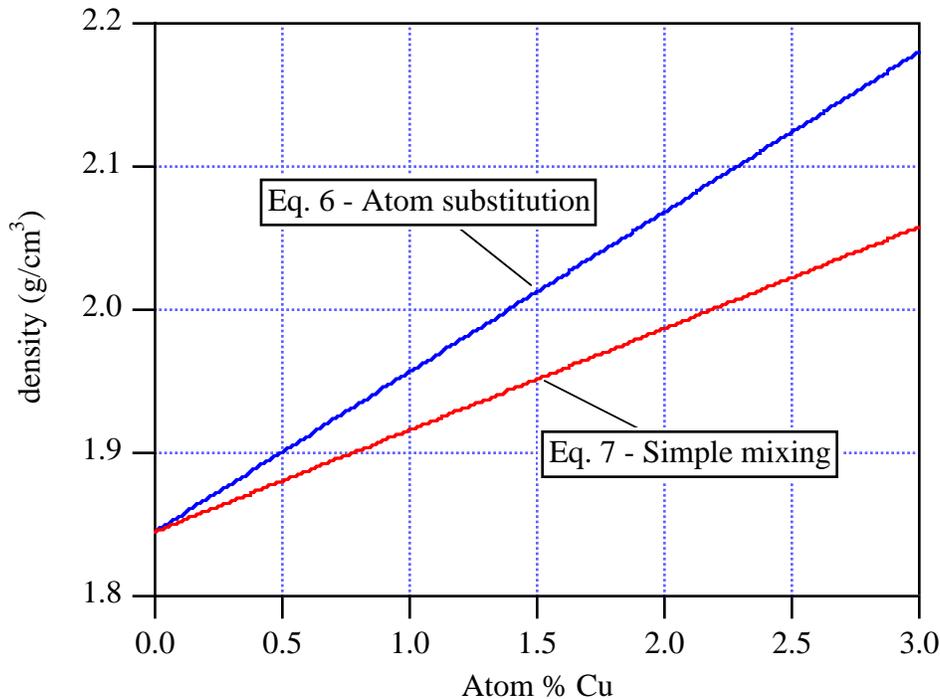


Figure 2. Computed densities of Cu-doped Be

³ *Handbook of Chemistry and Physics, 78th ed*, pgs 4-5 and 4-9.

⁴ *ibid.*, pg 6-5.

I guess the last thing to outline is what we do with graded capsules. At best by these methods we can only get a composite density. But we can calculate an expected "full" target density if we know the individual layer thicknesses. As an example lets consider a capsule that has an OD of 2210 μm . We then need to determine the individual layer thicknesses - and make sure that these add up to the total thickness of the deposited layer. This might not be such a problem given a polished cross-section. So lets assume for the sake of this example that starting from the outside we have 94 μm of pure Be, then 16 μm of 0.35 atom % Cu, followed by 55 μm of 0.70 atom % Cu, then 6 μm of 0.35 atom % Cu, and finally 6 μm of pure Be. This makes the total wall 177 μm thick.⁵ Lets number the layers starting from the outside as 1 through 5. The volume of the individual layers can be calculated as

$$V_i = \frac{4}{3}\pi \left(\left(R - \sum_{i=1}^i t_{i-1} \right)^3 - \left(R - \sum_{i=1}^{i-1} t_i \right)^3 \right) \quad (9)$$

where R is the outer radius of the capsule (2210/2 μm) and t_i is the thickness of the i th layer, t_0 being equal to 0. Given this notation the composite density of the shell in terms of the densities of the layers (ρ_i) is

$$\rho_{\text{composite shell}} = \frac{\sum_{i=1}^5 \rho_i V_i}{\sum_{i=1}^5 V_i} \quad (10)$$

Note that the units used for volume don't matter. For the example given above the composite density comes out to be 1.871 g/cm^3 . The result is not very sensitive to the measurement of layer thickness, if we take the Cu-doped layers as each being 1 μm thicker and decrease the outer pure Be layer by 3 μm , the composite density only changes by less than 0.001 g/cm^3 . It is critical however, to get the total layer thickness right, as it was for uniformly doped shells.

⁵ This is the original 300 eV design, see S. W. Haan, et al., "Update on the NIF Indirect Drive Ignition Target Fabrication Specifications," *Fusion Sci. and Technol.* **45**, 69 (2004).