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Chemical Analysis Of Beryllium Shells

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Subject: Chemical Analysis of Beryllium shells

Summary:

There is a need to understand the level of high-Z impurities in Beryllium shells prepared by sputter coating. The Ignition Point Design Requirements[1] state the following: "Except for allowed ingredients, as listed in the ablator composition entries, the ablator material in all layers shall contain sufficiently low impurity levels that the sum over all impurities of atom fraction$*Z^2$ shall be less than or equal to 0.2." This is a tight specification that requires careful materials analysis. Early in the first quarter of FY06, we undertook a study of Be shell impurities via ICP-MS[2] and determined that the impurity levels in the sputtered shells are very close to the specification.

Experimental:

To measure trace impurities in Be shells, we followed the standard protocol for ICP-MS. First the Be shells were pyrolized to remove the mandrel and the remaining pieces (7 mg total amount) were dissolved in high purity nitric acid. This solution was then exposed to an Ar plasma to atomize and ionize the elements in the sample. The ions were then passed through a series of apertures (cones) and into a high vacuum mass analyzer. The isotopes are identified by their mass-to-charge ratio (m/e). The results are tabulated on a graph of counts as a function of mass number. The intensity of a specific peak is proportional to the amount of that element in the original sample. The instrument is calibrated for mass dependent sensitivity using concentration standards. A calibration curve is created using the data from these standards. The slope of the linear curve has units of counts per second (CPS)/concentration unit (usually ng/g or ppb). The samples are then run and a signal intensity (CPS) is obtained for one or more isotopes of the element of interest. This CPS is then converted into ng/g using the slope determined for the standards. Typical accuracies for this technique are in the several % range.

The Be sputter targets themselves, S-65 Structural Grade Be Block from Brush-Wellman, have impurity content that is close to the specification. The levels of impurities in Be coupons (Brush Wellman PF-60) are shown in Table 1, accompanied by the calculations for the Z^2 weighted sum of atom fraction. Note that the analyses values are only 1 significant figure, the

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[1] Steve Haan, Ignition Point Design Requirements, Number 1, Rev 0, WBS I.4.1.1

[2] ICP-MS: Inductively-Coupled Plasma Mass Spectrometry. Analysis done in the C&MS Department.

calculations based on these include more for clarity, but the final sum is only good to about 1 significant figure. The Brush Wellman analysis for the S-65 sputter targets is more limited, but the levels listed in Table 1 are about the same (0.08 instead of 0.06 wt % for Fe the most important variant). The sum of 0.160 is dominated by individual measurements for Fe and Ni, at 0.066 and 0.024 respectively. The results of the ICP-MS analysis for the Be shells themselves are shown in Table 2. The weighted sum was much higher, 0.554 in this case due to an unexpected amount of Cu in the sputtered samples. The weighted sum drops to 0.117 when the Cu is removed. This value does not change appreciable (only to about 0.12) if a small amount of Cu is taken into account assuming that the level of Cu should only be what it was in the analyzed coupon (Table 1).

Table 1. Impurities in a Brush Wellman Be coupon (PF-60) based on the Brush Wellman analysis that accompanied the samples. Also shown is the roll-up of these impurities as required to evaluate their level relative to the impurity specification. Conversion from the reported wt% to atom% is accomplished by multiplication by (At. W. Be)/(At. W. element).

Element	Z	At. W.	wt%	atom%	Z^2*at%/100
Fe	26	55.85	0.06	0.0097	0.066
Ni	28	58.71	0.02	0.0031	0.024
Si	14	28.09	0.02	0.0064	0.013
Al	13	26.98	0.02	0.0067	0.011
Cr	24	52.00	0.01	0.0017	0.010
Ca	20	40.08	0.01	0.0022	0.009
Pb	82	207.19	0.002	0.0001	0.006
C	6	12.01	0.02	0.015	0.005
Cu	29	63.54	0.003	0.0004	0.004
Others					<0.004
					Sum = 0.160

Table 2. ICP-MS analysis of Be shells produced in B298. The sum is shown both with and without Cu.

Element	Z	atom%	Z^2*at%/100
Cu	29	0.0520	0.437
Fe	26	0.0110	0.074
Ni	28	0.0021	0.0165
Al	13	0.0045	0.008
Mg	12	0.0043	0.006
Ti	22	0.0009	0.004
Cr	24	0.0005	0.003
Mn	25	0.0004	0.002
Mo, Co, V, Zn, Sc, Pd		<0.0001	<0.002
			Sum = 0.554
			Sum w/o Cu = 0.117

The higher level of Cu, at 0.052 atom % is surprising. Clearly there is residual Cu in the system that is transferred to the shells even when the Cu sputter guns are not being used for a given run. What is reassuring with these results is that there are no other surprises, for example an several fold increase in Fe relative to the sputter targets. What is in the targets seems to appear in the shells at about the same concentration.

The elements analyzed and quantified with the ICP-MS technique were specified prior to the experiment so the proper calibration could be performed. However, even if an element isn't specified beforehand, it is still possible to detect it. There was some indication in the ICP-MS results that there might be small amounts of U and W in the shells, most likely in the 10^{-3} atom % range. If we recalculate the weighted sum of atom fractions using 0.001 atom % as an estimate for each, then the sum without Cu is 0.256. This is slightly higher than the specifications allow. A change to a higher grade of Be (i.e. optical grade) would not necessarily lower the weighted sum of the high-Z impurities. The different categories of Be metal are graded primarily according to their BeO content. In fact, for the optical grade some of the Brush-Wellman specifications for Fe and other metallic impurities are actually higher in the optical grade than they are in the "lower" grades such as the S-65 material.

How we deal with these results depends upon how important this level of contamination is. Our guess is that the high Cu level is due to contamination either from the Cu gun directly or due to previously sputtered Cu in the chamber. If the latter it will be a very hard (impossible?) problem to fix. Even without the Cu problem we are just meeting the specification (to one significant figure). It is unlikely that we can get significantly purer Be targets (but we will inquire). Based on our results we think that the specification needs to be re-evaluated. Can the capsule in the nominally Cu free regions tolerate a 0.05 atom % Cu level? Can the sum of the contributions of other elements (atom fraction$*Z^2$) be as much as 0.4? Based upon these answers we will determine a path forward. In the mean time we plan to submit a second sample for analysis to include other elements (U, W, …) that were seen in this sample but not carefully analyzed for.

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