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GRAIN REFINEMENT OF ARC-MELTED BERYLLIUM-6 WT% COPPER

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Abstract

Beryllium doped with 6 weight % copper is the material of choice for fabrication of target capsules for the National Ignition Facility because of its combination of attractive neutronic, physical, and mechanical properties. The target capsules are very small (2 mm in diameter) and thin-walled (150 microns), and the material must be fine-grained and of low inclusion content. Arc-melted Be-Cu is being produced to eliminate the oxide content. Equal channel angular extrusion (ECAE) is being used to refine the as-cast grain structure. Be-Cu rods have been processed by ECAE at temperatures from 500 to 750°C in tooling with a 120° angle. Selected samples have been annealed for 1 hour at temperatures from 700 to 775°C. The ECAE processing creates a heavily deformed and finely subdivided structure, and the annealing can produce an equiaxed microstructure with a grain size of approximately 20 μm.

Keywords: beryllium, grain size, target capsules, texture

Introduction

Beryllium offers many advantages, including neutronic, hydrodymanic, and mechanical, as compared to polymers, for the production of capsules for the Inertial Confinement Fusion program. These include high density, high tensile strength, high thermal conductivity, and low opacity [1]. Be material should be strong enough to allow room-temperature handling of a filled capsule, which would result in significant savings over the cryogenic operation that a polymer capsule will require. Be doped with 0.9 at% Cu (6 wt%) has the necessary x-ray opacity. Current designs for the National Ignition Facility target call for a capsule 2 mm in diameter with walls 150 μm thick. In order to assure that the capsule walls respond uniformly, both to internal pressure from the deuterium-tritium fuel, and the exterior drive pressure, there must be numerous grains through the wall thickness, and little or no crystallographic or mechanical anisotropy in the Be material. Thus, a grain size of 10 μm, or even less, is desirable. The required surface finish for the capsule is approximately 30 μm RMS. This finish is difficult to achieve with conventional powder-metallurgy source Be materials because of their high oxide content (typically about 1 wt%), as oxide particle pullout limits the quality of the final polished surface.

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To avoid the problem of oxide particles, an ingot metallurgy approach is needed. Unfortunately, this typically results in a very large grain size because of the high heat capacity of Be. Special thermomechanical processing will be needed to refine the grain size to the sizes needed for the NIF capsules. Work is underway at Los Alamos National Laboratory to produce high-purity Be-Cu material, and to develop the processing procedures to refine the grain size to a suitable level. Arc-melting has been used to produce small quantities of the Be-0.9 at% Cu material, and severe plastic deformation has been used to break down the coarse as-cast structure to produce an equiaxed, greatly refined grain size.

Experimental Procedure

The starting materials for the arc-cast Be-Cu alloys were Brush-Wellman Be lump and Alfa Aesar 99.9999% Cu shot. The Cu shot was annealed in a 96% Ar-4% H mixture at 900°C to remove dissolved oxygen. The Be was arc-melted in small buttons weighing 10-15 g. The arc-melting chamber was evacuated to less than 5×10^{-5} torr, and then backfilled with ultra high-purity Ar gas that was passed through a purifying train to achieve impurity levels of less than 10 parts per billion. The arc melting was conducted with a low pressure of approximately 300 torr. After arc-melting the buttons were repeatedly etched to remove any slag using a mixture of nitric and hydrofluoric acids. The small Be buttons were cut into several (5-10) small chunks. The appropriate amount of Cu was weighed out and melted with one of these pieces. The remaining Be was added one chunk at a time. The final alloyed button was then etched to remove slag, cut into several parts, and remelted 6-10 times. The etching process for Be-Cu alloys is more complicated than that for pure Be because of the differing chemical activities of Be and Cu. The first etch is a mixture of nitric, sulfuric and hydrofluoric acids. This removes the adherent BeO slag. The button is then etched with dilute sulfuric acid to remove Be, but not Cu. The button is re-etched with the acid mixture to get rid of the Cu. The final etch is warm concentrated nitric acid which leaves the surface clean with a thin oxide layer. There is no rinsing between etchants.

Prior to the final melting and casting of the Be-Cu buttons, a Zr button was arc-melted for at least 60 s to getter the arc-melter atmosphere. The Be-Cu buttons were arc-melted on a two-piece Cu hearth with a cylindrical mold cavity approximately 5 mm in diameter and 40 mm deep. When the button was ready for casting, a vacuum assist was used to overcome the surface tension of the melt and quickly draw the molten metal down into the mold cavity. The hearth was then removed from the arc melter, and the 2 pieces were separated to allow removal of the Be-Cu ingot. The hot-top and the bottom of the arc-melted Be-Cu rods were removed with a slow-speed diamond saw to leave a rod approximately 5 mm in diameter by 30 mm long.

Preliminary experiments had shown that commercially available powder-metallurgy-source S200F could be processed by ECAE at 425°C [2]. However, the arc-melted material would have a vastly greater initial grain size than the approximately 30-μm-grain-size S200F material, and would also contain 6 wt% Cu as an alloying addition, which would strengthen the alloy [3]; both factors suggested higher temperatures would be needed for ECAE of the Be-Cu.

To prevent any exposure of Be to the laboratory atmosphere, and also to prevent any contact between the Be and the ECAE tooling, the Be was enclosed in a can made of commercial purity nickel (Ni200) [2]. The can was 45 mm long (1.77 in.) and 9.5 mm in diameter (0.374 in.), with walls 2.25 mm thick (0.088 in.). A Ni200 plug 5 mm in diameter by 7.44 mm in length (0.197 by 0.293 in.) was inserted in the end of the can, and the can was sealed by electron-beam (EB) welding. The can and plug were in the annealed condition.

The 120°-angle die was fabricated from H13 tool steel, heat-treated to R_C 52, with an inlet channel 9.53 mm (0.375 in.) in diameter. The outer corner radius at the intersection of the inlet and outlet channels was 4.78 mm (0.188 in.), or half the inlet channel diameter. The outlet channel was slightly smaller in diameter (9.35 mm [0.368 in.]). The outlet channel had a land that extended 5.1 mm (0.20 in.) from the inner corner; the outer channel then expanded to 9.80 mm (0.386 in.) in diameter to reduce the area over which friction acted.

The billet and tooling were lubricated with a nickel-containing high-temperature anti-seize compound (NIKAL Pure Nickel Anti-Seize Lubricant, Jet-Lube, Houston, TX). A short plug approximately 19 mm long (0.75 in.) of Ni200 was inserted in the die inlet channel at the tail of the billet. The Ni plug at the billet tail allowed the billet to be pushed through the die corner and beyond the outlet channel land without interruption. The Ni plug remained in the die corner, and was expelled by the next extrusion. The billets were extruded at a ram speed of 6.35 mm/s (0.25 in./s). Route B_C (90° rotation of the billet in the same direction) was used, with the welded end of the billet being inserted first into the die for all passes. The billets were preheated in air to the extrusion temperature for 1 h prior to the first pass; the extrusion temperature ranged from 500 to 750°C in 50° increments. After each pass, the billet was cooled in water, cleaned, and carefully examined to verify that the can and weld were intact. After each extrusion, a small bulge of material was noted on the top surface of the billet immediately behind the billet nose; this bulge was removed by abrasive grinding. For each subsequent pass, the billet was preheated for 0.5 h. The ECAE tooling was preheated and maintained at approximately 500°C throughout the processing by a removable split-box furnace mounted on a stand. The temperature of the tooling was monitored with a thermocouple that was placed inside the inlet channel of the tooling.

After the 4 passes, the billet was divided approximately in half transversely with a diamond saw. Slices approximately 2 mm thick were encapsulated in quartz tubing in an Ar atmosphere, with a Ti getter, and then heat treated for 1 h at 700, 725, 750, or 775°C. The slices were then mounted and polished to show the effect of the annealing heat treatment on the microstructure.

Results and Discussion

The arc-melted rods were generally clean and shiny, with occasional surface pits and defects. Transverse and longitudinal sections through the rods showed that the as-cast grains were very large, as Fig. 1 shows, frequently extending half-way across the billet; i.e. up to 2.5 mm in length, and 200 μm in diameter. Few inclusions were observed in the material.

Fig. 1. The as-cast Be-Cu microstructure (as-polished; polarized light).

The Ni cans were readily sealed by EB welding. The welds showed no indications of flaws or other defects. The canned billets were extruded for 4 passes at the 6 temperatures from

500 to 750°C. At the lowest temperature, fluctuations in the load-displacement record were present, suggesting that perhaps the internal Be-Cu rod had failed. However, in all cases the Ni can remained intact, with no indications of failure of the weld or the can wall.

When the canned billets were cut in half, it was obvious that the Be-Cu material had failed during the extrusions at 500 and 550°C; it is not known on which pass failure occurred. However, the Be-Cu material appeared sound for the samples extruded from 600 to 750°C. Slices from these 4 billets were annealed for 1 h at temperatures of 700, 725, 750, or 775°C. These temperatures were chosen based on previous experience with similar Be-Cu material that had been extruded and annealed over a similar temperature range [4].

The microstructures of the as-extruded material showed that the as-cast structure had been extensively sub-divided by the ECAE process (Fig. 2). In some cases, traces of the prior as-cast structure could still be discerned. The deformed structure had many features that suggested twinning played a significant role in the deformation at the elevated extrusion temperatures. Rounded and equiaxed regions of new grains were observed with increasing extrusion temperatures, suggesting that dynamic recrystallization had occurred during the extrusion. However, the new grains were not distributed homogeneously, and did not fully replace the as-cast structure, even in the 750°C extrusion.

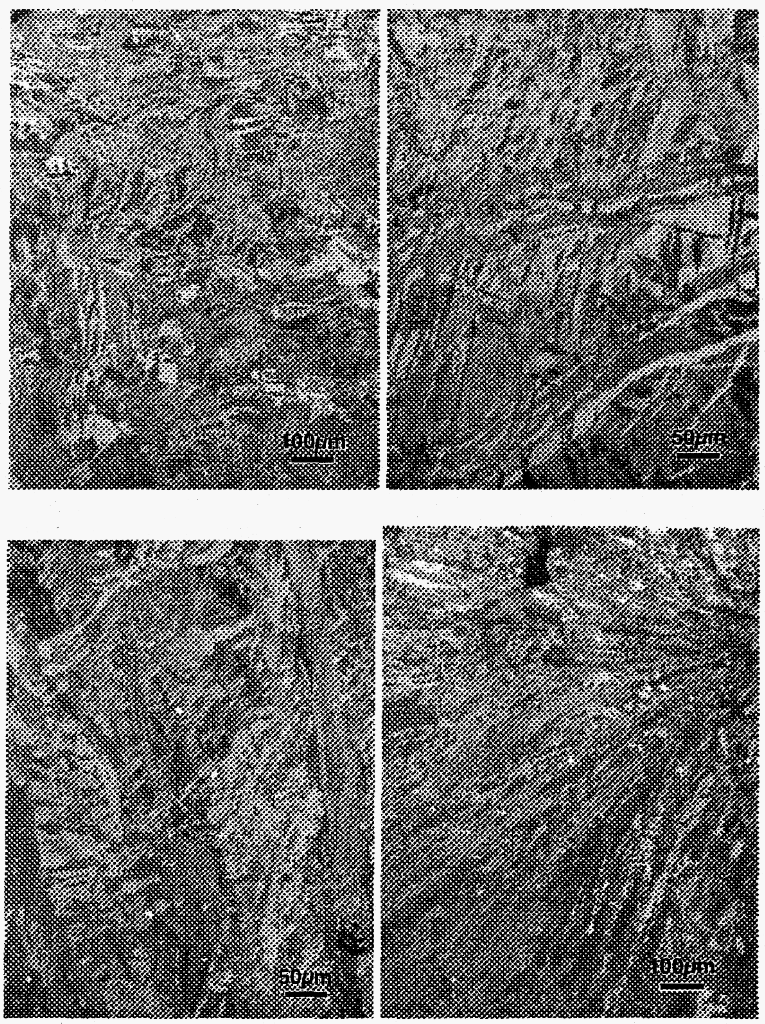

Fig. 2. The microstructures of the Be-Cu material after 4 passes of equal channel angular extrusion, 120° die, route B_C, at different temperatures (as polished, polarized light): a. 600°C; b. 650°C; c. 700°C; d. 750°C.

The 1-hour-annealings resulted in nucleation and growth of new, equiaxed grains. The process of nucleation of new grains is illustrated in Fig. 3, for the case of material processed at 750°C. At the lowest annealing temperature of 700°C, the deformed structure is still present, along with a few recrystallized grains. Approximately 25% of the structure is recrystallized at 725°C, 80% at 750°C, and at 775°C the structure is fully recrystallized. In the fully recrystallized structure the grains are equiaxed, and the grain size is approximately 20 μm. Texture measurements will be performed to determine if there is any texture to the recrystallized material.

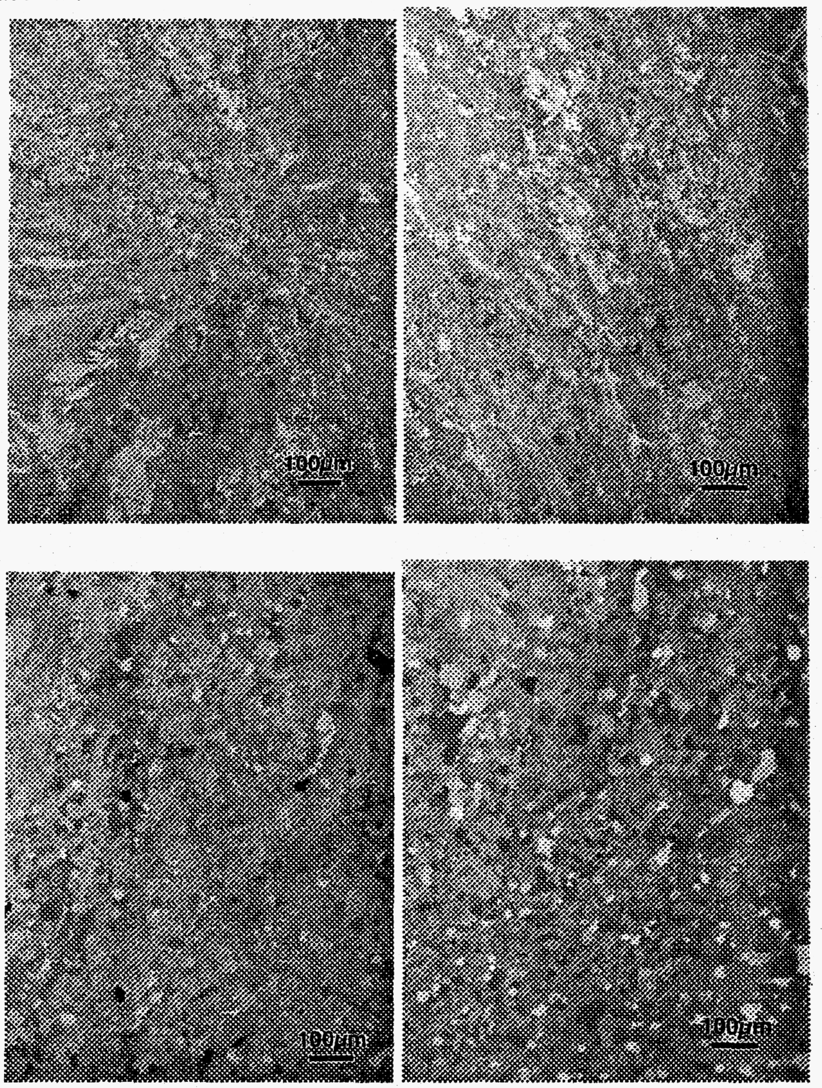

Fig. 3. Microstructures of the Be-Cu material after 4 passes of extrusion at 750°C, followed by annealing for 1 h at different temperatures (as polished, polarized light): a. 700°C; b. 725°C; c. 750°C, d. 775°C.

The results of this initial series of trials are very promising. Small rods of high-purity Be-Cu material have been made by an arc-melting process. The coarse as-cast Be-Cu material can be successfully processed by ECAE at temperatures from 600 to 750°C. With appropriate heat treatment, equiaxed recrystallized grains with a nominal diameter of about 20 μm can be produced. It should be possible to perform a second series of extrusions and anneals to achieve an even finer grain size. The first cycle of processing should allow subsequent extrusions to be made at significantly lower temperatures, since the grain size will have already been refined to the 20-μm range. Thus, temperatures as low as 400°C may be considered for the second round of processing. The lower extrusion temperature will reduce the amount of recovery that occurs during processing, so that the amount of stored energy will be greater than for the higher temperatures in the present work. This should allow recrystallization at lower temperatures such as 600°C, since the driving force for recrystallization will be greater. The number of nucleation sites should increase, thus nucleating more grains. The growth rate of the newly formed recrystallized grains will be slower at the lower annealing temperatures. The result should be a finer recrystallized grain size.

The present results suggest that the present annealed microstructure is fairly stable up to about 750°C, which is in agreement with the previous limited experience for similar Be-Cu material [4]. The stability of the microstructure has important implications for the bonding of the machined hemispheres that will form the final capsule. Should the microstructure prove to be stable at temperatures as high as 850°C, these temperatures can be used for the bonding cycle. A further refinement of the grain size may have an effect on the temperatures over which the microstructure is stable, as finer grains will be more susceptible to grain growth at elevated temperatures. The effects of time and temperature on grain growth in the Be-Cu material will need to be studied extensively to provide information for design of the bonding process. Although the mechanical properties of the fine-grained Be-Cu are expected to be more than adequate to allow the filled capsule to be held at room temperature, these will need to be measured.

Conclusions

High-purity Be-0.9 at% Cu (6 wt%) has been produced by arc melting. The Be-Cu was sealed in Ni cans, and processed by equal channel angular extrusion for 4 passes at temperatures from 500 to 750°C with 120° tooling, with a continuous 90° rotation between each pass (route B_C). Successful extrusions were possible at temperatures of 600°C and higher. The coarse, as-cast grains were extensively subdivided by the ECAE processing. Dynamic recrystallization of portions of the microstructure occurred at extrusion temperatures of 700 and 750°C. Annealing for 1 h was conducted at temperatures from 700 to 775°C. At 700°C, there was little change in the microstructure. As the annealing temperature increased, new equiaxed grains formed in the deformed material. At 775°C, the microstructure was fully recrystallized, with an equiaxed grain size of approximately 20 μm. Additional work will be conducted to further refine the grain size, and to measure the texture and the mechanical properties of the fine-grained material.

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References

1. D. C. Wilson et al., "The development and advantages of beryllium capsules for the National Ignition Facility", Physics of Plasmas, 1998, Vol. 5, No. 5, 1953.
2. R. D. Field et al., "Equal-Channel Angular Extrusion of Beryllium", Metall. Mater. Trans., 2002, Vol. 33A, 965.
3. F. Aldinger, "Flow and Fracture of Single Crystals", in *Beryllium Science and Technology, Vol. 1,* D. Webster and G. J. London, eds., Plenum Press, New York, 1979, 71.
4. J. C. Cooley and D. J. Thoma, personal communication, Los Alamos National Laboratory, 2000.