

FABRICATION OF LARGE DIAMETER ALUMINO-SILICATE K⁺ SOURCES*

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Abstract

Alumino-silicate sources that are used for Heavy Ion Fusion (HIF) drivers can produce high current beams, and have a long lifetime, typically many months of normal operation. The usual ion source diameter is about 10 cm with a curved emitting surface. Recently we achieved reliable fabrication of such large diameter sources with high quality alumino-silicate coating on tungsten substrates. The effects of substrate porosity, powder size distribution, coating technique, particle packing density, drying, and heat firing temperature have been investigated.

INTRODUCTION

Alumino-silicate K⁺ sources have been used in HIF experiments for many years [1,2]. These sources have a typical current density of $\sim 10\text{-}15\text{mA}/\text{cm}^2$, for 5-10 μs . but much higher current density has been observed (using smaller size sources). In comparison to ionizers, alumino-silicate sources have a considerably long experimental run time, on the order of months, and lower neutral emission levels. Recently we have improved our fabrication techniques and are able to reliably produce large diameter ion sources with high quality emitter surface without defects. This note provides a detailed description of the procedures employed in the fabrication process. The variables in the processing steps affecting surface quality are substrate porosity, powder size distribution, coating technique on large area concave surfaces, drying, and heat firing temperature.

SUBSTRATE PREPARATION

Alumino-silicate K⁺ ion sources are made by melting a layer of synthetic alumino-silicate onto a tungsten substrate, the process is similar to working with ceramics [3]. Since the ion source will be operated at temperatures above 1050 degree C and the melting process is around 1550 degree C, the substrate is usually made of a refractory metal such as tungsten. 80% dense porous tungsten is suitable for ion source application, anything of lighter density is usually too fragile.

There is another very important reason to use porous tungsten substrates. In comparison to a solid substrate, the absorption of molten ceramic material and a rough surface provides the basis for mechanical gripping of alumino-silicate during the melting stage. The HCX source has a dimension of 10cm diameter with a concave surface of

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20.32cm spherical radius. The emitting concave surface area is 82.4cm^2 ($\pm 1\text{-mil}$ tolerance). The tungsten substrates supplied by the vendor have pores closed by the finishing machining process so the first step is to reopen the emitting surface pores by electrolysis etching.

Tungsten etching is accomplished by a DC electro-chemical process. A 5 molar KOH solution is produced by mixing 2 liters of high purity deionized water and 500g of potassium hydroxide (KOH). A tungsten substrate is attached to a positive electrode with the region to be etched exposed and facing the negative electrode. The negative electrode has the shape of a convex spherical cap with a smaller spherical radius of 19.05cm, to fit the concentric contour of the substrate etching area and retain uniform electric field line distance across the entire etched surface at a separation of $\sim 1.2\text{cm}$. In etching the 10cm diameter source, a 20A DC current with $\sim 2.5\text{V}$ applied across the electrodes for 40 minutes will etch away $\sim 8\text{-mil}$ of tungsten material and is enough to re-open the pores and produce a uniform surface free of any remaining machining marks. SEM photos (Fig. 1) show that the surface pore average size is typically $\sim 15\mu\text{m}$.

After electro-chemical etching, residual KOH electrolytic solution must be removed from the substrate. Typical practice is to soak and dilute the tungsten substrate in deionized water for a few days until the pH value reaches ~ 6.0 or higher. Drying can be done in a vacuum oven at a temperature of ~ 100 degree C to remove all remaining water vapor. Finally, the substrate is clean fired at 1750 degree C in a vacuum furnace at a pressure of $\sim 10^{-6}$ Torr.

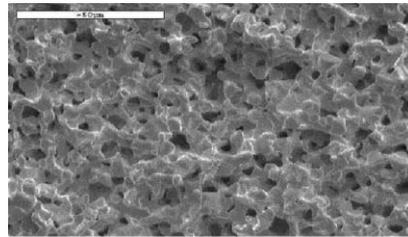


Figure 1: SEM photo of etched, porous, 80% dense tungsten surface (50 μm scale) with $\sim 15\mu\text{m}$ pore size.

ALUMINO-SILICATE PREPARATION

This procedure describes the preparation of synthesizing potassium-aluminosilicate, as it is heated to ~ 1450 degree C to produce the Leucite crystalline phase, $\text{K}_2\text{OAl}_2\text{O}_3 \cdot 4\text{SiO}_2$ [4]. It begins with thoroughly mixing raw high purity chemicals followed by a two-step reaction process in an atmospheric air furnace.

The chemicals, all in powder form, are 99.995% Potassium Carbonate (K_2CO_3), 99.9% Silicon Oxide (SiO_2), and 99.8% Aluminum Oxide (Al_2O_3). The respective stoichiometric ratio by weight is 1.0:1.739:0.738. Since potassium carbonate is hygroscopic, the mixing process begins in a controlled low humidity environment, such as, flowing dry nitrogen gas through a glove box and keeping the humidity below 5%. After mixing by weight, the mixture is tape-sealed in a jar before removing from glove box to trap in the dry nitrogen gas.

The next step is to use a ball mill process to thoroughly blend the chemicals for several hours. Afterward the mixture is transferred to a platinum crucible. Chemical reactions occur when the mixture is heated in an air furnace for 4 hours at 1100 degree C. The reaction product expands and froths as CO_2 gas is released, and then shrinks to form a solid mass when cooled.

The comminution processes that follow after cooling consist of crushing and milling to further disperse the mixture and to reduce the average particle size of the material [5]. The solid mass is carefully removed from the crucible and the dry material ground with mortar and pestle to a fine powder, ~ 200 -mesh ($75\mu m$) size, before placing in a planetary ball mill for one hour minimum reducing it to a finer powder size as a final mixing step. The entire mixture is again transferred to the platinum crucible for re-heating in an air furnace for 2 hours at 1450 degree C, completing the reaction, releasing any remaining CO_2 , producing the Leucite crystalline phase, and cools to a solid mass. After removal from the crucible, the material is ground once more to produce the final particle-size powder distribution.

The particle-size powder distribution was found to be an important factor in minimizing defects, such as, warping, distortion, and cracks on the alumino-silicate layer. Two principal particle sizes for the coating process are separated using wire sieves. Fine particle-size 400-mesh ($<37\mu m$) powder is chosen as an initial pre-coat layer on porous tungsten substrates to penetrate, and lodge into the $\sim 15\mu m$ diameter pores of the tungsten substrate. The main coating layer consists of a coarser particle-size distribution between 200-mesh ($75\mu m$) and 270-mesh ($53\mu m$) powder.

COATING TECHNIQUE

A thin layer of ceramic may be formed by pouring aqueous slurry onto a surface and then moving a blade over the material to produce a thin film [5]. This coating method was used to apply a uniform adhering finish of 15-mil potassium alumino-silicate to cover large area concave and planar porous tungsten substrates.

The coating process occurs in two steps. Starting with 400-mesh powder, an aqueous slurry suspension is produced by mixing with high purity de-ionized water. Droplets of the 400-mesh aqueous mixture are then distributed over the entire surface, effectively imbedding small particles of fine 400-mesh alumino-silicate powder

into the pores of the tungsten substrate as a pre-coat layer. After a 12-hour drying period, the top layer of 400-mesh dry powder is gently brushed off and discarded.

Second step is to apply the main 15-mil coating layer, consisting of a coarser particle-size distribution between 200 and 270-mesh potassium alumino-silicate powder, and mixing with de-ionized water to form a thicker slurry.

Difficulties can arise in applying this main coating to large diameter and non-planar surfaces. A blade-scraping tool was designed to match the spherical radius of the concave shaped substrate with a smooth knife-edge down the center, and a 30-degree taper from the horizontal on both sides (Fig. 2).

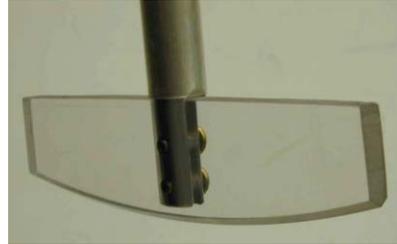


Figure 2: Blade-scraping tool, knife-edge has a 30-degree taper from the horizontal on both sides.

The main coating process begins by pouring alumino-silicate slurry to cover the emitting surface of the tungsten substrate. Because porous tungsten absorbs water readily, it is necessary to frequently apply de-ionized water droplets to keep the mixture near aqueous in viscosity. The scraping tool action is initiated by rotating slowly at the base of the support rod and applying de-ionized water droplets and adding slurry as needed to cover depleted areas. Alignment tolerance is held to ~ 1 -mil in a milling machine. The wet coated material gradually increases in viscosity as it dries and is nearly ready for the final lifting of the knife-edge tool off the surface. An ~ 1 -mil high thin track is typically left by the knife edge tool during final lift off. This is usually not a concern because the track will disappear when the layer of alumino-silicate melts during the heat firing process. With accurate digital vertical position control provided by a milling machine, it is possible to reset the scraping tool each time after removing excess slurry material, and then continue to form the layer until achieving uniform thickness. By slowly rotating the scraping tool and allowing the slurry to increase in viscosity before removing, uniform 15-mil coatings have routinely been achieved. After the coating process has been completed, the drying process should start immediately.

DRYING

The rate of drying and the particle packing density are essential factors of the forming process [6]. If the rate of drying is too fast, shrinkage of the ceramic slurry will cause coating defects to occur. A ceramic part dried too rapidly forms a hard surface film, which traps the moisture and leads to tearing at the surface. Hydrostatic forces, imparted to the mixture by water, are removed through the drying process. During the wet forming

process, the distribution of particle shapes and sizes is known as particle packing density. A higher particle packing density will effectively reduce shrinkage. Shrinkage will occur during drying as the liquid between the particles is eliminated and the interparticle separation distance decreases. Forming the ceramic material at a lower liquid content and increasing the mean particle size will reduce drying shrinkage and thus reduce coating defects. Shrinkage defects occurred using smaller particle size 400-mesh alumino-silicate powder for the main 15-mil coating layer, while shrinkage defects did not occur using increased particle size 200-mesh powder with proper drying techniques.

A basic method requiring limited effort is to control the drying rate by regulating humidity. For this task, a humidifier chamber was built by flowing air through a de-ionized water-filled flask to create water vapor flowing in parallel with dry air into a manifold and combining into a chamber. The air velocity is kept low for slower drying rates. Humidity is monitored at the output tube of the chamber with a humidity sensor. Immediately after wet coating, substrates are placed in the humidifier chamber and slow dried over a period of several days starting with >90% humidity, gradually lowering the humidity and ending with <10% humidity. A completed drying process is essential before heat firing in a vacuum furnace.

HEAT FIRING

Heat firing is the final process step that produces a melted layer of alumino-silicate that mechanically bonds to the tungsten substrate. Since bonding is mechanical, substrate porosity and surface roughness is essential to provide the basis for mechanical gripping. Also, there is thermal expansion mismatch to consider, metals usually have a higher thermal expansion coefficient than ceramics. Poor coating thermal shock resistance can lead to cracking and chipping at the surface. An effective way to enhance thermal shock resistance is to apply as thin a coating as possible with a high ceramic to metal bond. Defects can occur, such as craters and blowholes, from contamination of dust or metal flakes within the material vaporizing during heat firing.

A dry coated substrate is centrally placed in a vacuum furnace to reduce inherent temperature gradients. During the heat firing process, the vacuum furnace pressure is maintained in the 10^{-5} - 10^{-6} Torr range. Potassium alumino-silicate starts to melt at 1550 degree C. Typically, the heat firing process consist of bringing the temperature up to ~1565 degree C and holding for 45 minutes, with a heat-up and cool-down rate of 10 degree C per minute. Temperature control is achieved with thermocouples up until ~1500 degree C, then two optical pyrometers are utilized to achieve accurate temperature control near the melting point. The potassium alumino-silicate surface emissivity, equal to ~0.91, was calibrated by focusing on a blackbody in the vacuum furnace and then compared with a coated substrate at the same temperature, 1570 degree C, in the heat zone region.

Utilizing two optical pyrometers improved the accuracy of the temperature measurements to +/- 5 degrees C. The resultant phase structure is dependent upon the maximum heat firing temperature and the aluminosilicate coating thickness. Observed phase transformations, for uniform 15-mil coatings, range from glassy vitreous structure, ~1555 deg C, dull snowy bubble structure, ~1565 deg C, to crystalline needle-like structure, >1570 deg C. After heat firing, alumino-silicate coated layers have typically been reduced by ~30% in thickness due to shrinkage.

The maximum heat firing temperature must be approached slowly to avoid overfiring temperatures. According to SEM analysis, overfiring causes crystal growth and depletion of potassium. The glassy vitreous structure has desirable characteristics of lower defects and high current emission properties. Absorption depth of molten ceramic material into porous tungsten substrates has been detected to depths of ~25-mil to ~50-mil.

CONCLUSIONS

Previous attempts at fabricating ion sources have resulted in coatings displaying various defects, such as, shrinkage cracking, blistering, and craters (Fig. 3). After applying these coating techniques, the surface quality has significantly improved to produce smooth, uniform, and fully coated large diameter emitting surfaces with virtually a single-phase structure. As with most techniques, optimization of these processes is far from complete. For instance, the drying rate and the heat firing duration have not been fully optimized. The effect on surface quality will be a determining factor.



Figure 3: Comparison of significant coating technique improvements fabricating 10cm diameter, 0.4mm K⁺ alumino-silicate coatings on porous tungsten substrates.

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