EBM FABRICATION AND CHARACTERIZATION OF HIGH PURITY NIOBIUM FOR SUPERCONDUCTOR APPLICATIONS

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Abstract

Superconducting radio frequency (SRF) cavities are used to accelerate charged particles to near the speed of light for elemental studies. Currently, SRF cavities are typically fabricated using different forming processes including deep-drawing and spinning to mechanically shape niobium into the desired geometry. This research presents the development of processing parameters for high purity niobium (powder size range of 25-125μm) using electron beam melting additive manufacturing technology. Fabrication parameters were improved to obtain dense parts in a time-efficient manner. A specific procedure was used to maintain powder purity, and powder chemistry was monitored at different stages of fabrication. In addition, a series of experiments were performed to obtain 99.9% dense parts and a maximum building height of ~85mm.

Introduction

The Electron Beam Melting (EBM) process is an additive manufacturing (AM) technology used for the rapid manufacturing of near net-shape metal components. There are many advantages of this process over other AM techniques. For example, since the process is performed in vacuum, it allows for the fabrication of high melting point metals and reactive materials such as nickel, cobalt and titanium alloys while eliminating the negative effects of impurities or oxidation. Also, EBM is considered a hot process because the electron beam is utilized prior to fabrication of each layer to preheat the powder metal bed resulting in the fabrication of parts that have minimal to no residual stresses [1]. The EBM process has been used mostly for medical and aerospace applications, although continued research is widening the spectrum of materials that can be processed as well as their applications.

The EBM process has been commercially available since 2000 by the Swedish company Arcam (Arcam AB, Sweden). This process is catalogued as a powder bed fusion AM technology and it uses a stream of electrons to sinter and melt selectively in a layer-by-layer fashion. Metal powder particles (30μm – 120μm in diameter) are used as the precursor material for fabrication. In the process, layers of metal powder with a thickness in the range from 50-120 μm are evenly spread by a raking mechanism. Then the electron beam is used to preheat the powder bed followed by the selective melting in specific regions through the bed using higher energy on the beam. The electron beam is accurately and rapidly steered to scan specific geometries with the use of electromagnetic lenses or coils positioned in the gun column. After processing of one layer, the build table is lowered in the Z direction by a thickness of one layer and the process repeats until the part is finished. The process is carried out in vacuum levels that are maintained at ~10^{-4} Torr or below. The main components in the Arcam EBM process (Figure 1) are the electron gun (1), electromagnetic coils (2), powder hoppers or containers from which powder is gravity fed (3), the raking arm (4) and the build stage (6). Normally, the process uses a build

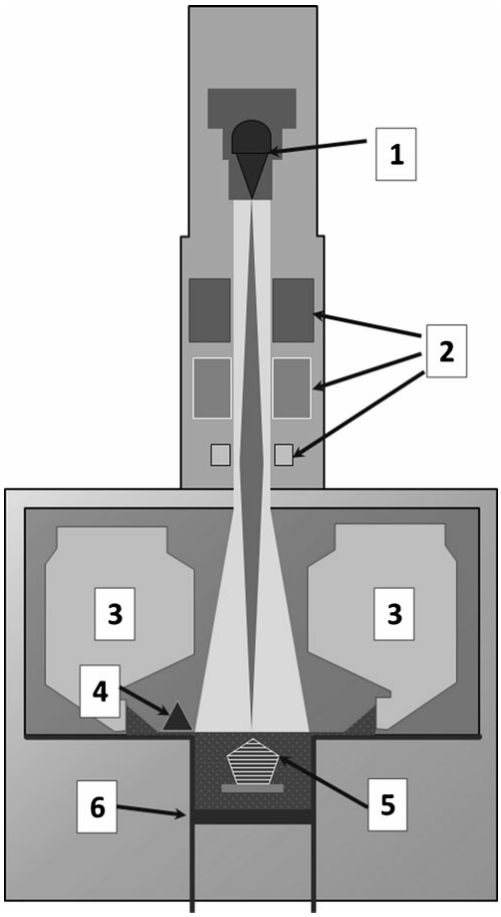

Figure 1 – Schematic of the Arcam electron beam melting system

substrate to commence fabrication and provide anchoring of the initial layers of the part being produced (5).

SRF Cavities

In the field of particle physics, superconducting radio frequency (SRF) devices are used to generate electric fields that accelerate elemental particles such as electrons to study their interaction upon collision. SRF cavities are key components in particle accelerators since they transfer the energy to the traveling particles, and their purity is critical as it directly affects the accelerating field that can be achieved during operation. Assemblies of multiple SRF cavities are normally employed in a particle physics laboratory setup. In the setup, voltage is applied intermittently to the cavity assembly while particles are injected from a source at a rate that is in phase with the voltage. As the voltage oscillates through the assembly, each cavity experiences a change in polarity that provides the energy kick and accelerates the particles.

High purity niobium is the material of choice for fabrication of SRF cavities since the element has the highest critical temperature of all pure substances (9.2K or -263.95ºC) and it becomes superconducting when cooled below this point. Subtractive (i.e. machining) manufacturing is rarely used in fabrication of SRF cavities because of niobium's upscale price.

Therefore, three forming methods (Figure 2) are currently used in the fabrication of cavities which are deep drawing, spinning and hydroforming [2]. Deep drawn cavities are formed by stamping a thin (~3mm) niobium sheet into a set of dies. The spinning operation is performed similarly by starting with a niobium disc of similar thickness that is bolted from the center and rotated. Then an adjustable mandrel is moved to apply pressure and form the shape of the cavity. Both techniques mentioned before have the limitation that only half-cavities with uniform thickness can be produced. Post-processing steps to form the full cavity are required, which include the welding of the half-cavities from the equator; a process that introduces variability in thickness and possible contamination of the precursor purified niobium. The third process employed is hydroforming where a single niobium tube is attached in between a set of dies and hydraulic pressure is applied at the interior of the tube to form the cavity profile. This process has the advantage of producing multi-cavity designs but further development is necessary to achieve uniform thickness of the resulting parts [2].

Issues with Current SRF Cavities

During experimental operation of cavity assemblies, there are many factors that affect their performance. From the standpoint of the mechanical stability of the parts, the development of the so called Lorentz forces cause the mechanical deformation of cavities. These forces appear as an outward pressure at the equator and an inward pressure at the iris in the cavity profile eventually causing its deformation (Figure 3). These local deformations cause the detuning in the frequency of the cavity assembly ultimately reducing the accelerating field that

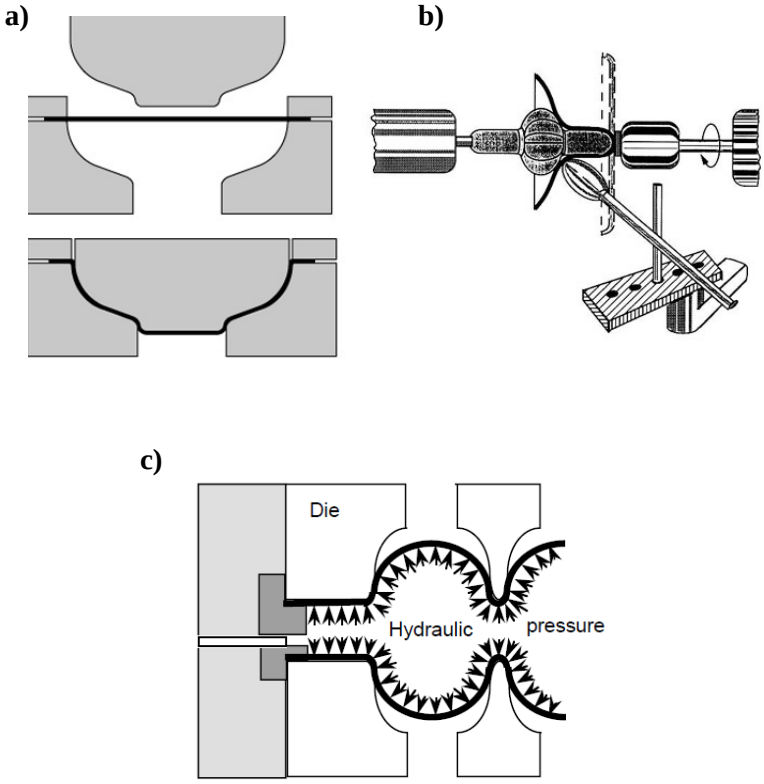

Figure 2 – Traditional fabrication methods for SRF cavities. a) deep-drawing, b) spinning and c) hydroforming.

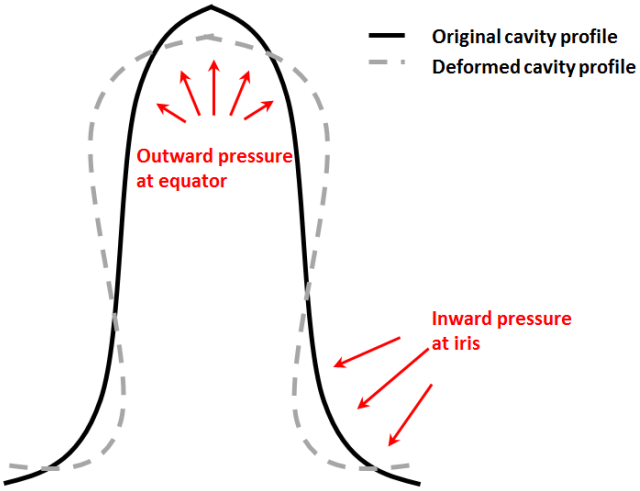

Figure 3 – Lorentz forces acting in the wall of an SRF cavity causing its deformation.

can be achieved. Welding of support structures and rings is usually performed as corrective measures to prevent detuning; however, this is done at the expense of introducing more complexity and variability to the components. The use of different joining techniques such as tungsten inert gas welding (TIG), metal inert gas welding (MIG), or electron beam welding (EBW) also increases the risk for contamination at the surface of cavities.

Other issues with detrimental factors to the electronic properties of niobium cavities are field emission and thermal breakdown (quenching), both problems related to the presence of impurities and defects in the cavity wall. In field emission, electrons are emitted from sites with impurities and their trajectories bent by the magnetic field causes them to strike the walls of other cavities and failure occurs by localized heat. In the case of quenching, small surface defects such as roughness or interstitial impurities cause unstable power dissipation leading to breakdown of the accelerating field [3].

EBM-AM of SRF Cavities

In this work, EBM was explored as an alternative fabrication process given the limitations of current methods to produce SRF niobium cavities. EBM provides the ability to fabricate nearly fully dense components with complex designs. This is an important advantage for the fabrication of SRF cavities as it can be used to manufacture designs that exhibit non-uniform thickness and/or with integrated stiffening support structures to resist the deforming forces and frequency detuning that develops during experimentation (Figure 4). The technology also offers the potential to produce cavities and other components in a monolithic form greatly reducing the post-processing by welding and hence maintaining the purity of the niobium once fabricated in the final shape.

Experimental Methods

The focus of this work was to optimize the processing parameters for EBM fabrication of dense niobium components using precursor powder with particle sizes in the range from 25 to 125µm. Also, part of the work focused on the implementation of a procedure for handling the niobium powder to minimize the exposure to environmental impurities and humidity (described

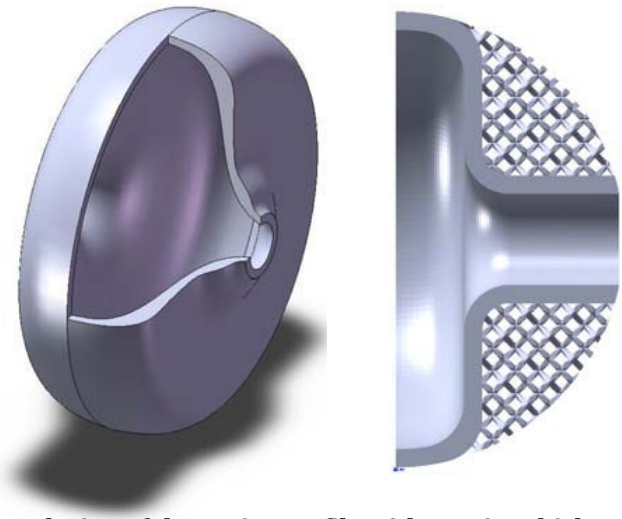

Figure 4 – CAD rendering of the cavity profile with varying thickness and integrated support structures to the wall of a cavity

in the powder handling section). The chemistry of niobium was traced from the wrought stock used, after its fabrication into wire, after the plasma atomization of the wire, and after EBM-fabrication.

The approach followed to improve the density of EBM niobium implemented the use of three sets of parameter combinations. Each experiment consisted of the fabrication of nine separate blocks or coupons with dimensions of 10x10x10mm forming a grid or matrix (Figure 5). The processing parameters from the melt explored were beam speed (mm/s), average current (mA) and focus offset (mA) of the electron beam. Across the rows of the grid, the speed and average current of the beam were varied while the focus offset was varied column wise. A total of three experiments were performed in which the fabrication parameters that provided the highest density were used for the next iteration.

After fabrication of the 9 coupons with different parameters, the density of the parts was measured using a weight balance (1) with a density measuring kit including a plastic stand (2),

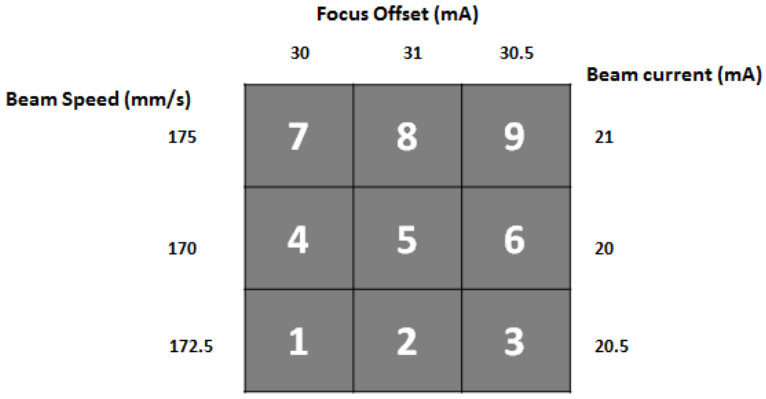

Figure 5 – Schematic of fabrication parameter space explored during experiments to improve the density of EBM niobium.

one bracket (3), one weight basket (4), and a nalgene beaker (5) to hold distilled water, as depicted by Figure 6. Measurements were done according to ASTM Standard B311-13 by the volume displacement method [4]. Weight measurements were made by weighing the parts in air and then in distilled water. The apparent density was calculated with the formula Density = (A x E)/(A - F). In this formula, A was the mass of the test specimen in air, F was the mass of the test specimen in water and E was the density of water at the working temperature. The percent relative density (%RD) for parts was calculated by dividing the obtained density values over the density for wrought reactor grade niobium of 8.57g/cm^3.

Since the surface roughness obtained from as-fabricated EBM parts is not optimized for SRF applications, a final experiment was done to measure the density of machined EBM niobium using the same setup described before. The edges or contours for one EBM-fabricated bar with dimensions of 12 x 12 x 85 mm were machined using a computer numerical control

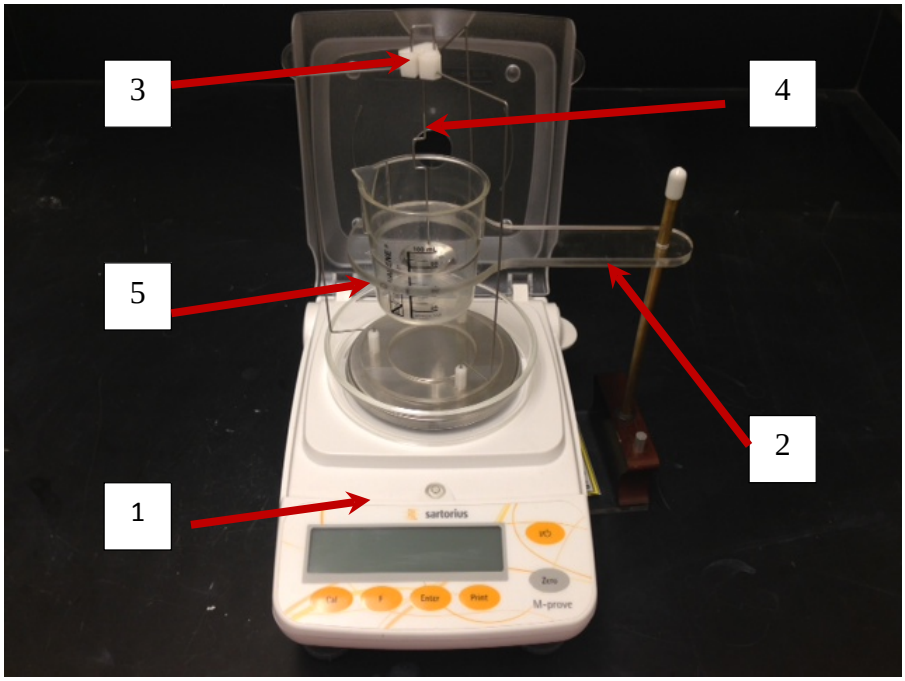

Figure 6 –Weight balance and density measurement kit used for EBM niobium parts.

(CNC) machine. Then, the part was cut into five equal sections and density measurements were taken for the parts. The values obtained provided a better indication for the density of the niobium when fabricated by the EBM process. Reported values in this work were the average from three measurements for each part.

Powder Handling

To maintain the purity of niobium, handling of the powder material was performed using a glovebox with a controlled N_2 gas atmosphere. The unit (Figure 7a) injected N_2 gas during operation maintaining the humidity at or below 5%. Inside the glovebox, a mechanical sifter (Figure 7b) was designed, fabricated (Figure 7c) and used to speed up the process to sieve the

powder for recycling purposes. The sifting process was carried out in ~20 minutes and the powder was poured manually into the hoppers of the A2 system.

Ventilation of the machine was also performed with N_2 gas by retrofitting a tank into the solenoid valve of the A2 machine. Once the system reached ambient temperature conditions, the N_2 gas was used to ventilate the chamber, delaying the adsorption of humidity into the niobium powder which could make it prone to an increase in interstitial impurities once fabricated by EBM.

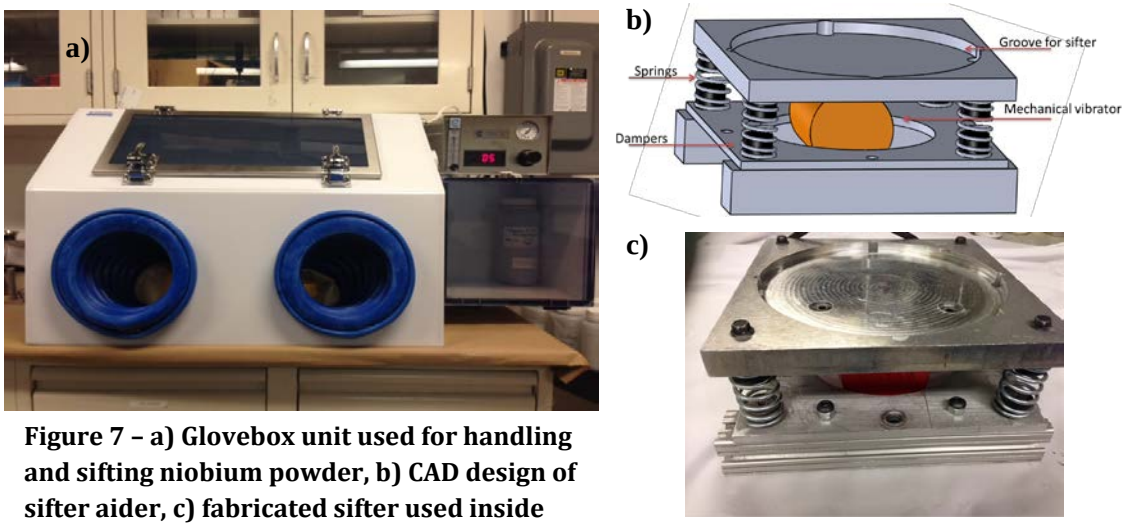

Figure 7 – a) Glovebox unit used for handling and sifting niobium powder, b) CAD design of sifter aider, c) fabricated sifter used inside glovebox.

Monitoring of Material Purity

The purity of niobium was traced using chemical analysis of the wrought bar, wire, atomized powder, and EBM-fabricated niobium. The chemical analysis on several specimens were sent to Element Materials Technology (Element, USA), where element tracing was performed by inductively coupled plasma mass spectrometry (ICP), to detect light elements such as hydrogen (H), oxygen (O) and nitrogen (N), and LECO combustion for carbon (C). The purity of EBM-fabricated niobium was also investigated with the use of X-ray diffraction (XRD).

An RGA-100 residual gas analyzer (SRS, USA) was also installed to evaluate the quality of the vacuum and thus the presence of impurities at the interior of the chamber (Figure 8). The RGA permitted monitoring of several gas species during fabrication using EBM. Use of the RGA is important since some of those gases, which include H, N, O, He, water vapor (H_2O), and carbon monoxide (CO_2) can become sources for the formation of interstitial impurities causing detriment of the electronic properties of niobium in SRF applications.

Figure 8 – RGA unit installed on the Arcam A2 system.

Results

Apparent and Percent Relative Densities

The bar charts depicted in Figures 9 through 12 show the results obtained for apparent and relative densities for the experiments performed. In experiment 1, the highest apparent and percent relative densities were those of part number 1 measured at 8.48g/cm^3 (98.9 %RD).

In experiment 2, the processing parameter space explored was near those of part number 1 in experiment 1. Improved density values were obtained for parts 4, 5, and 6 with %RD values above 99%. The highest density was that of part number 5 with measured values of 8.53g/cm^3 (99.5%RD).

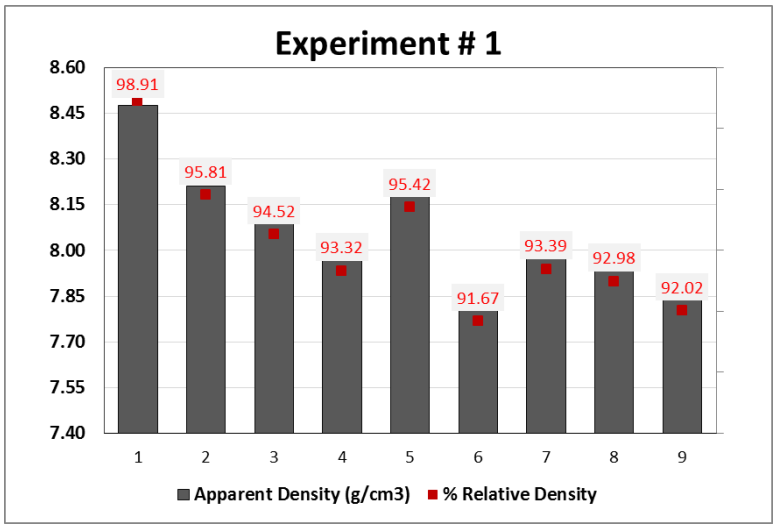

Figure 9 – Results for density measurements in experiment 1.

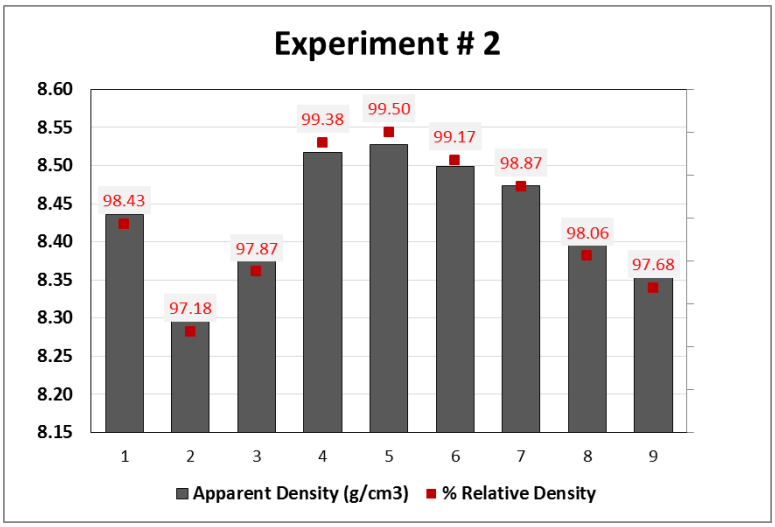

Figure 10 – Results for density measurements in experiment 2.

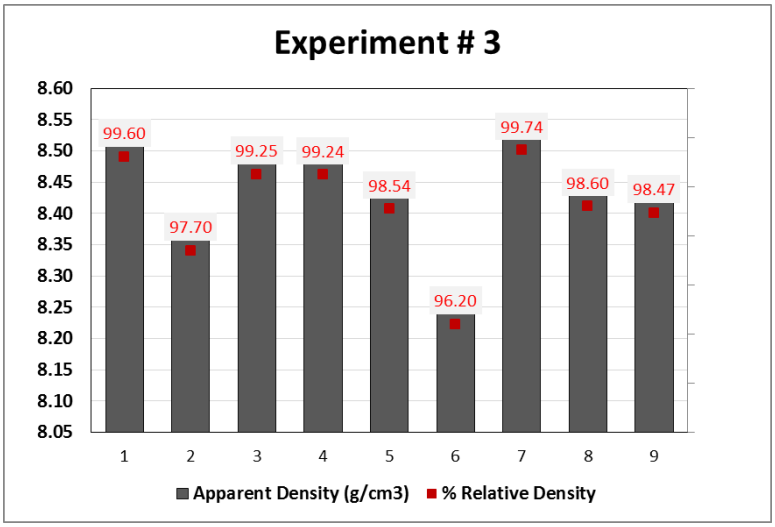

Figure 11 – Results for density measurements for experiment 3.

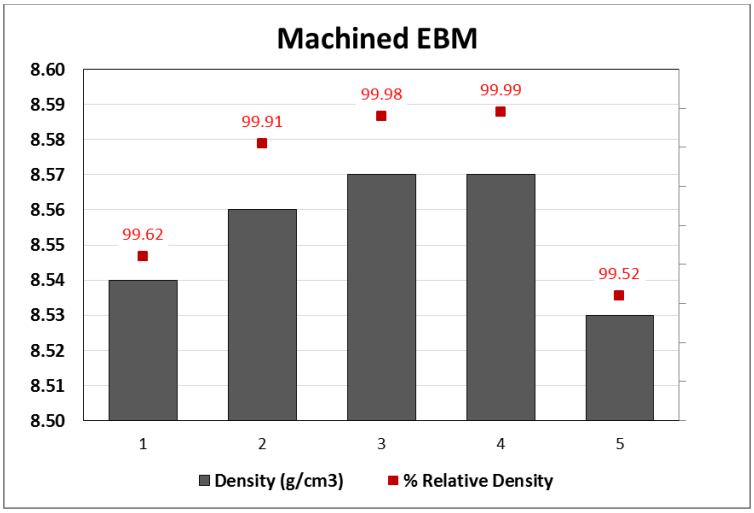

Figure 12 – Measured apparent and percent relative densities for EBM part with machined edges.

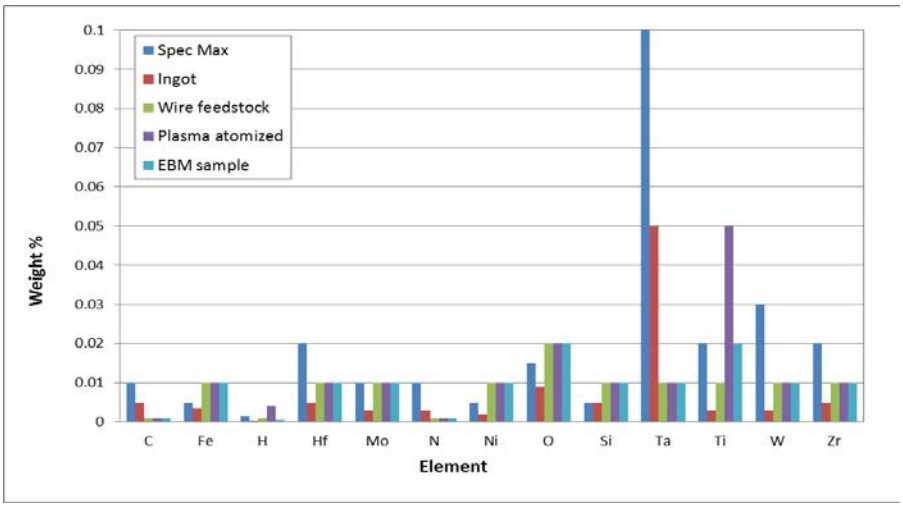

Figure 13 – Chemical analysis history of niobium

Results for experiment 3 show a further improvement in the density values. The densest part was number 7 with values of 8.548g/cm^3 (99.74% RD). The fabrication parameters for this specimen are a focus offset of 31 mA, an average current of 18.5 mA, and a beam speed of 172 mm/s for the melt themes. Since the fabrication parameters for this part yielded the best density in parts, further fabrication of parts was carried out with these parameters. As previously mentioned, the surface roughness is not optimal for SRF applications. The last experiment carried out consisted on machining the contours of a square bar measuring ~85mm and sectioning it into five equal parts. Density values obtained for all five sections averaged 8.55g/cm^3 and three nearly fully dense parts were measured at 99.9%RD (Figure 12).

Chemical Analysis

From the results of the chemical analysis it was deduced that the EBM process did not cause contamination of the niobium. By contrast, the atomization process to produce the powder from the niobium wire feedstock showed changes to the chemistry of the material, mostly as an increase in the titanium (Ti) content. Other elements that also showed levels above the standard

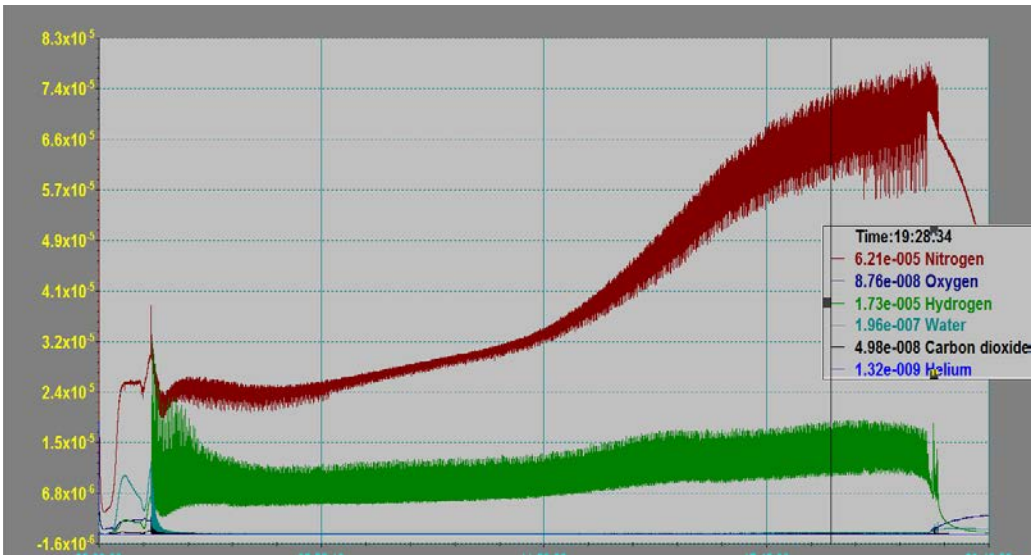

Figure 14 - Typical RGA plot observed during fabrication with niobium.

(ASTM B393) [5], such as nickel (Ni) and iron (Fe), and those prone to form interstitial impurities such as hydrogen (H) and oxygen (O) (Figure 13).

Residual Gas Analysis

The monitoring of different gas species during fabrication was done using the RGA. The plot of the partial pressures versus time shows they were maintained below the required limit ~10^{-5} Torr (Figure 14).

XRD Analysis

The XRD patterns obtained (Figure 15) for powder (a) and the fabricated EBM samples (b) also show a clean niobium spectrum with the preferred orientation planes. A bcc structure

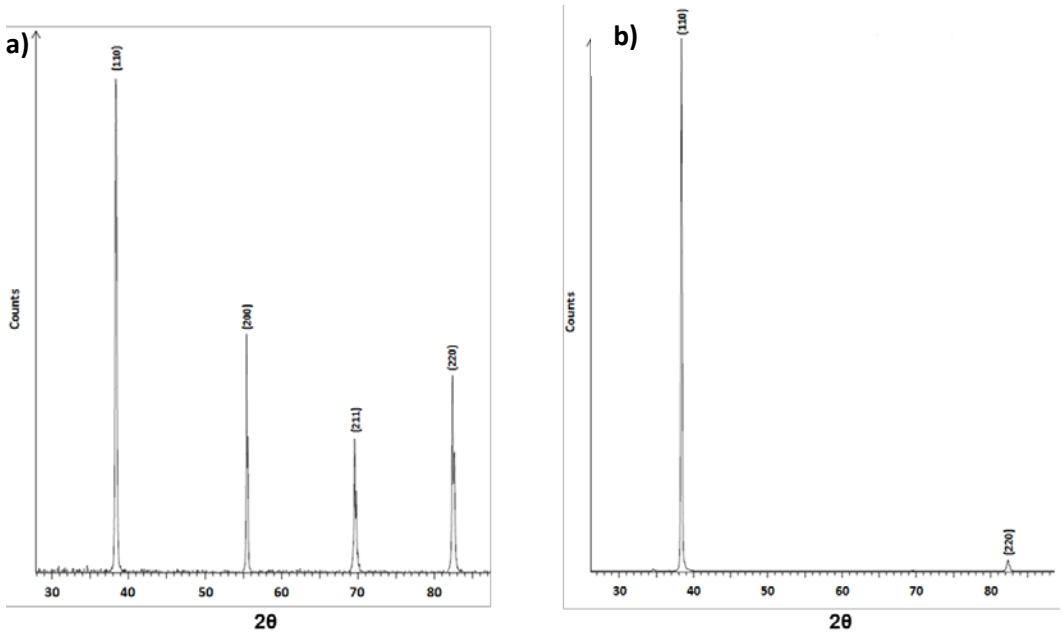

Figure 15 – XRD spectra for a) powder and b) EBM niobium specimens

with lattice parameter of a= 3.3Å is representative for both the powder and the fabricated specimen. The fabricated specimen analyzed belongs to a plane parallel to the fabrication direction (z-axis) and XRD analysis shows a preferential directionality in the (110) orientation.

Conclusions

In this work, a design of experiments was implemented to improve the density of niobium parts fabricated by the EBM process. Three parameters (beam speed, beam current and focus offset) were varied in each experiment in an iterative manner until the percent relative density, was improved to values over 99.9%. The work also reported on the monitoring of the purity of niobium at different stages in the production of niobium which are: a) niobium ingot, b) wire from stock material, c) powder state after plasma atomization, and d) after fabrication using the EBM process. Evidence that the material maintained the purity required by SRF applications after processing by EBM was obtained by the use of the RGA and the XRD analysis. Also, the use of chemical analysis for light elements such as H, N and O, and by LECO combustion to

trace carbon (C) evidenced the purity of the precursor powder material before and after use in the EBM process.

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