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Original Research Paper

Increasing the amorphous yield of $\{(Fe_{0.6}Co_{0.4})_{0.75}B_{0.2}Si_{0.05}\}_{96}Nb_4$ powders by hot gas atomization

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ABSTRACT

The synthesis of metallic glasses requires high cooling rates leading to product size limitations of a few millimeters when using conventional casting techniques. One way to overcome these size limitations is powder metallurgy. Melt atomization and the subsequent powder processing can result in larger, amorphous components as long as no crystallization takes place during powder consolidation.

An iron-based glass-forming alloy $\{(Fe_{0.6}Co_{0.4})_{0.75}B_{0.2}Si_{0.05}\}_{96}Nb_4$ was formed through both ambient room and high temperature inert gas atomization at various melt flow rates (close-coupled atomization). The use of hot gas generally decreases the droplet size and hence leads to an increased cooling rate and amorphous fraction of the atomized powders.

Hot gas atomization results in a lower gas consumption, a smaller gas-to-melt mass flow ratio (GMR), smaller particles and a smaller geometric standard deviation.

Particles atomized in ambient temperature were fully amorphous up to a particle size fraction of 90 μm. Larger particle size fractions resulted in a higher crystalline fraction. According to the XRD and DSC analyses, hot gas atomization has only a very small influence on the cooling rate and the amorphous fraction. However, the amorphous yield is significantly increased using hot gas atomization.

1. Introduction

Metallic glasses exhibit excellent physical and chemical properties such as soft magnetic properties, good catalytic performance, high strength and good corrosion resistance [1,2]. However, the properties of these amorphous materials can only be obtained when the melt is rapidly cooled. Ongoing efforts to modify glass-forming alloys and the use of rapid solidification techniques such as water quenching and copper mold casting has led to the synthesis of bulk metallic glasses. These methods provide the required cooling rates to avoid crystallization, but the relatively low heat conduction of these alloys generally results in dimensional restrictions. This limits the critical casting thickness ranges from micrometers to a few centimeters [3,4]. A further challenge is the required high purity of the feedstock material that can also limit the commercialization of metallic glasses at a large scale.

The combined use of melt atomization and a subsequent powder metallurgy process is an alternative way to overcome size limitations. Gas atomization is a suitable process for the generation of amorphous powders [5,6], as it provides an appropriate range of cooling rates (10^3–10^5 K s^{-1}).

Today, there is a strong tendency to process metallic glasses for use in structural components. Different manufacturing techniques such as selective laser melting [7], warm hydrostatic extrusion [8], warm rolling [9], spark plasma sintering [10,11], severe plastic deformation [12], cold spraying [13,14], and thermoplastic forming [15–17] have been used. These techniques require primarily amorphous powder as a base material. Consequently, generating metallic glasses as glassy powders is essential for these technologies as it avoids dimensional restrictions as long as no crystallization takes place during subsequent powder processing.

The final properties of the consolidated material are influenced by particle size, particle shape, impurities, oxygen level and amorphous fraction. These properties are interconnected as the amorphous fraction also depends on the cooling rate and the cooling rate, in turn, depends on the particle size. For instance, an increased droplet size generally results in lower cooling rates as the specific surface of the melt droplets decreases [5]. The design

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of appropriate cooling strategies is therefore required to achieve an amorphous fraction of 100%.

One of the most attractive possibilities to alter the cooling rate is the use of hot gas to generate smaller particles [18,19]. Hot gas atomization can yield smaller particles as the gas velocity increases. The cooling rate, the amorphous fraction, and the amorphous yield hence increase due to the synthesis of smaller particles. An additional benefit is a reduction of gas consumption.

The applicability of hot gas atomization poses the question: does hot gas atomization decrease the cooling rate and the amorphous fraction even though smaller particles are being generated?

2. Experimental procedure

2.1. Sample synthesis

The iron-based glass-forming alloy $\{(Fe_{0.6}Co_{0.4})_{0.75}B_{0.2}Si_{0.05}\}_{96}Nb_4$ (at.%) was chosen due to its economical relevance that results from its unique mechanical properties, high availability, and relatively moderate feedstock costs. Iron-boron (81.43 wt% of Fe and 17.94 wt% of B), FeNb (31.40 wt% of Fe and 65.20 wt% of Nb), pure Fe (99.99 wt%), Co (99.0 wt%) and Si (99.9 wt%) were melted in an Al_2O_3 crucible under an argon atmosphere. The total weight of the feedstock material was 2000 g. The melt temperature of 1833 K was constant for 20 min before atomization started.

2.2. Atomization

The schematic layout of the gas-atomization facility is shown in Fig. 1a. The system consists of three main components: a vacuum vessel including the inductive melting system, a spray chamber with a total height of 4.80 m and the nozzle system (close-coupled atomization) as seen in Fig. 1b.

An Al_2O_3 crucible with a capacity of 1000 ml was placed into the vessel. Twenty minutes after the atomization temperature (1833 K) was kept constant, a stopper rod was pulled to start the atomization process (the feedstock material was melted 350 K above the liquidus temperature). Three different nozzle outlet diameters were used (1.5 mm; 2 mm; 3 mm) leading to different melt mass flow rates ranging from 81 to 313 kg h^{-1}. The gas temperature was heated with a heat exchanger to temperatures in the range of 293–600 K.

A novel convergent-divergent close-coupled atomizer with an annular slit of 0.8 mm (CD-CCA-0.8) was used [20]. The new nozzle configuration design allows the stable atomization at low pressures of 0.8 MPa as well as higher gas pressures.

The high velocity gas stream impinged on the melt stream which resulted in the disintegration of the melt into droplets. The droplets then solidify in the presence of an inert gas during their flight inside the spray chamber. Liquid Ar $\geq$ 99.996 with an oxygen impurity $O_2 \leq 4$ ppm was used. The powder yield was above 95% (the remaining five percent corresponded to residues of the melt in the crucible and flakes or fibres due to the collision of semi-solidified droplets with the chamber wall – flakes and fibres were subsequently removed during sieving).

Table 1 summarizes parameters and characteristics of the atomization process including the gas-to-melt-mass flow ratio (GMR), mass median particle diameter $d_{50,3}$, and geometric standard deviation σ. The geometric standard deviation was calculated from the cumulative mass distribution according to $\sigma = (d_{84.3}/d_{16.3})^{1/2}$.

A reference powder was synthesized with the Nanoval process [21,22] in order to calculate the amorphous fraction from differential scanning calorimetry (DSC) measurements. The feedstock was melted in an argon atmosphere and subsequently atomized in a nitrogen atmosphere at 1770 K (oxygen content < 200 ppm). The GMR was 3.0 (nitrogen (kg)/melt (kg)). The obtained reference powder had a particle mass median diameter of $d_{50,3}$ = 23.2 µm and a geometric standard deviation of 2.0.

2.3. Analyses

A laser diffraction instrument (Malvern Mastersizer 2000) with a wet dispersion unit was used to measure the mass median particle diameter $d_{50,3}$ and the geometric standard deviation σ (nine measurements were conducted and averaged).

XRD measurements were performed with a Bruker D8 equipped with a CuKα source. The powder samples were run from 10 – 80°

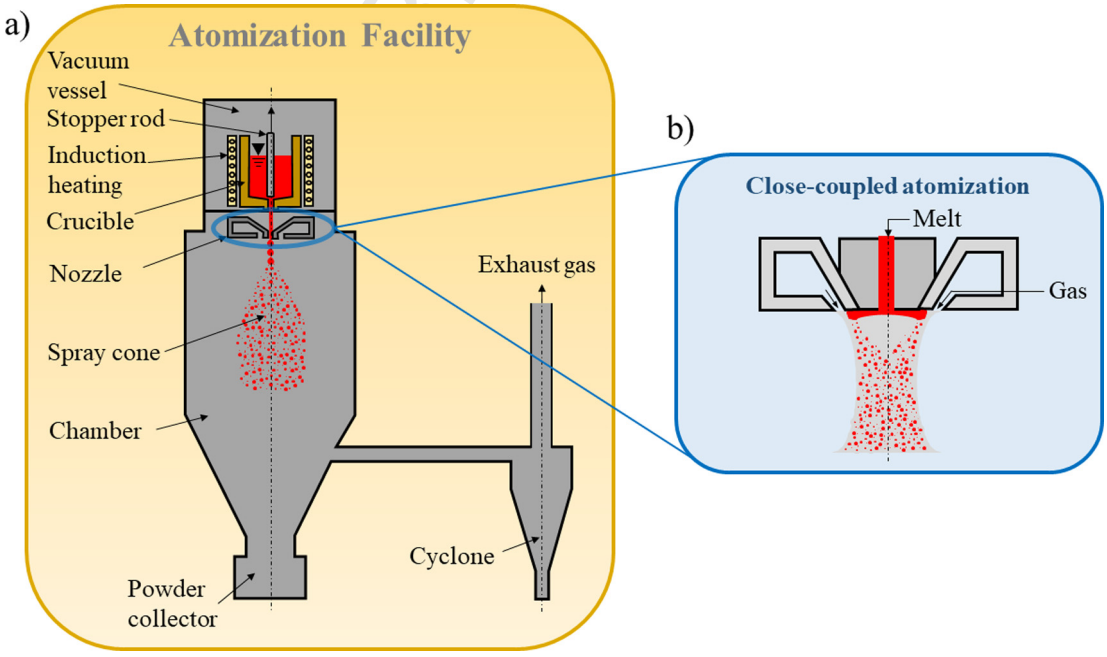

Fig. 1. Schematic illustration of the gas-atomization set-up (a) including the close-coupled atomizer (b).

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