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Investigation on spheroidization of refractory tungsten powders by laminar DC plasma torch

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Abstract

The spherical tungsten powders were prepared by a low-power DC laminar plasma torch. At a discharge current of 50 A, the tungsten powder fed downstream cannot be spheroidized by using Ar discharge, while a small number of spherical particles can be observed with upstream feeding. When N₂ is added to the working gas, the input power is increased to maintain the same discharge current, which leads to the increase of plasma jet length, thereby expanding the effective high temperature region. Optical emission spectroscopy measurements show that the electron excitation temperature of N₂-Ar mixed discharge is higher than that of argon discharge. In addition, compared with argon plasma, N₂-Ar mixed gas plasma has larger thermal conductivity due to the molecular dissociation. Therefore, the heat is more efficiently transferred from the plasma to the tungsten particles. The improvement of plasma jet and heat transfer characteristics by introducing nitrogen into the discharge contributes to the increase of spheroidization rate. EDS analysis shows that the oxygen content in the tungsten particles decreases after spheroidization, which is caused by the evaporation of tungsten oxide during plasma treatment.

1. Introduction

There is a growing demand for particles with spherical shape on powder injection molding, 3D printing, thermal spraying, etc. [1–3]. Particles with spherical shape have low friction coefficient, high flowability and stacking density in the powder processing technologies, which have an important impact on the quality of the manufactured objects [4–6].

Tungsten is one of the widely used refractory metals in manufacturing [7, 8]. Spherical tungsten particles cannot be prepared by traditional methods due to its high melting point. In recent years, tungsten spheroidization by using thermal plasma has attracted great interest [9, 10]. Thermal plasmas are characterized by high temperatures, controllable temperature fields, and flow states, exhibiting the unique advantages in spheroidization of refractory metals involving the one-step treatment [11, 12]. When injected into the thermal plasma, the tungsten powders in the effective high temperature region (melting point or above) are melted rapidly by absorbing great amount of heat, and form spherical droplets under the action of surface tension. After leaving the high temperature region, these droplets rapidly cool and solidify to form spherical particles.

Thermal plasma can be generated by different excitation modes. Radio frequency inductively coupled plasmas (ICP) have been successfully applied to tungsten spheroidization [13–17]. Li et al prepared fine-grained spherical tungsten powders with narrow particle size distribution by ICP torch [16]. He et al simulated velocity field in plasma to improve the spheroidization rate of small particles [17]. Compared with the ICP discharge, the direct current (DC) thermal plasma has higher power conversion efficiency. DC thermal plasma can operate in laminar flow state under appropriate conditions. When a laminar DC plasma torch is used for tungsten spheroidization, the injected particles have a longer residence time in the plasma [18]. Pershin et al have obtained spherical WC powder particles by using a DC plasma torch over 30 kW [19]. At present, there are limited experimental investigations on tungsten spheroidization by using DC plasma, especially at low-power levels.
High-power plasma torches possess large effective high temperature regions, which provides a more favorable environment for powder spheroidization. The plasma spheroidization process is complex, affected not only by the plasma temperature field, but also by the gas flow rate, gas composition, etc. However, the effects of these processing parameters are difficult to distinguish in high-power mode. When the DC plasma operates at low-power level, the interaction time between the injected tungsten powders and the plasma is shortened due to the small volume of the plasma, which is beneficial to study the key factors affecting spheroidization. More work needs to be done in order to promote our fundamental understanding of plasma spheroidization process.

In the present work, a low-power DC arc plasma torch in laminar flow state was employed to spheroidize tungsten particles. The spheroidization experiments were carried out in two ways of upstream and downstream feeding by using N$_2$/Ar gas discharge. The plasma features, the surface morphology and particle size distribution of tungsten particles were investigated, and the plasma spheroidization process was discussed.

2. Experimental details

The experimental set-up is schematically described in figure 1, including a DC arc plasma torch, powder feeding equipment and optical emission spectroscopy.

The DC plasma torch consisted of four discharge electrodes: cathode, initiating arc electrode, the intermediate electrode, and anode. Argon or a mixture of argon and nitrogen was introduced as working gas from the discharge gas inlet. By controlling the gas flow rate and input power, a laminar DC plasma was generated. This was a compact plasma torch operating at low-power levels. During the plasma spheroidization experiments, the torch current was kept at 50 A. Since the voltage at 50 A current discharge changed with the composition of the working gas, the corresponding input power varied in the low power range of 1.0–3.5 kW under different working gas compositions, see the section of Results and discussion for details.

The powder feeding equipment sent powders from the barrel into the mixing chamber through a motor-driven screw. The mass flow and velocity of powders entering the plasma can be independently controlled by adjusting the motor speed and carrier gas flow rate. Ar was used as the carrier gas. The carrier gas containing tungsten powders in the mixing chamber was injected into the plasma from the upstream feed inlet installed in the middle of DC plasma torch, or the downstream feed inlet 10 mm away from the torch nozzle.

A thorlabs CCS 200/M spectrometer was employed to investigate the plasma feature. The optical emission from the plasma was focused by a quartz lens, and then transferred to the optical fiber head. The origin of the rectangular coordinate system was set at the center of torch nozzle, and the Z axis was parallel to the direction of the plasma jet. The optical fiber head with lens was placed on a two-dimensional mobile platform on one side of the plasma jet, and the optical axis was along the X direction. Through the movement of the platform, the optical emission spectroscopy (OES) measurements with a 1 mm spatial resolution on the Y-Z plane were achieved. The samples were characterized by GeminiSEM 500 scanning electron microscopy (SEM) with energy dispersive spectrometer (EDS) and a Microtrac Sync particle analyzer for particle size distribution.
3. Results and discussion

The tungsten powder was treated by using Ar discharge. The discharge gas flow rate was set to 7 sLm, and the input power was 1.2 kW. The mass flow rate of tungsten powders and Ar carrier gas flow rate were kept at 3 g min$^{-1}$ and 2 sLm, respectively.

Figure 2 shows the SEM images of the tungsten powders before and after plasma spheroidization. The raw tungsten powders in the as-received condition exhibit polygonal shape with irregular edges in figure 2(a). Figures 2(b) and (c) display the SEM images of treated tungsten powders fed from the downstream and upstream feeding inlets, respectively. As shown in figure 2(b), the tungsten particles after plasma treatment with downstream feeding cannot be spheroidized, but the particle edge becomes blunt. When the tungsten powders are injected into plasma through upstream feeding, one can observe a few spherical particles in figure 2(c).

The characteristic spectra of argon discharge plasma at different positions of Z-axis are given in figure 3(a). Argon atomic emission lines are observed near the plasma torch nozzle and in the downstream region, while the first positive system (FPS) of N$_2$ molecules also appears at $Z = 15$ mm, which originates from the ambient air entrained into plasma due to the momentum transfer between the plasma jet and air [20].

Figure 3(b) shows the electron excitation temperature distribution on the Z-axis, and the inset gives electron excitation temperature distribution in Y-Z plane. The electron excitation temperature distribution decreases rapidly with increasing Z. The inset in figure 3(b) shows the temperature spatial distribution of the plasma jet, indicating that the electron excitation temperature gradually decreases from the inner layer to the outer layer. The upstream feed inlet is located in middle of the torch, and
the temperature in this region should be higher than that of the plasma jet. Unfortunately, it is impossible to obtain OES measurement of plasma in the torch.

It is difficult to achieve plasma spheroidization of tungsten with low-power Ar discharge. A mixed N$_2$-Ar discharge was used to treat tungsten powders with current of 50 A, where the powders were fed from the upstream inlet. The discharge gas flow rate and the carrier gas flow rate were maintained at 7sLm and 2sLm, respectively. Figure 4 shows the SEM images of surface morphology of tungsten particles after plasma spheroidization at different N$_2$/N$_2$ + Ar flow ratios. It is found that the addition of nitrogen to the discharge gas has a significant influence on spheroidization.

In contrast to the pure Ar discharge treatment with only a few spherical tungsten particles in figure 2, a large number of spherical tungsten particles of different sizes are observed in figure 4(a) where the N$_2$/N$_2$ + Ar ratio is 25%. At higher N$_2$/N$_2$ + Ar ratios, the spheroidization rate increases to around 70% in figures 4(b)–(c).

In the plasma spheroidization process, the heating behavior of particles with different sizes is different. Figure 5 presents particle size distribution of tungsten powders before and after spheroidization. The as-received powders range from 10 $\mu$m to 200 $\mu$m with an average size of 58 $\mu$m. In general, particles with small size tend to evaporate easily due to their small heat capacity. After spheroidization by Ar discharging plasma in figure 5(b), the $D_{50}$ of treated powders is almost the same as that of as-received powder, while the particles below 20 $\mu$m disappear, and the $D_{90}$ is much larger than that of as-received powders. It is indicated that the evaporation and coalescence of small particles take place during plasma treatment. With the introduction of N$_2$ into the discharge, the average size increases to around 60 $\mu$m in figures 5(c)–(e) due to enhanced evaporation and coalescence effects.
A particle injected into a thermal plasma will usually undergo the following processes [21]: (1) heating of the solid phase from an initial temperature to the melting point of the material, (2) melting of the solid phase at constant melting temperature, (3) heating of the liquid phase from the melting point to temperatures at which evaporation takes place (approximately equal to the boiling temperature of the particle), and (4) evaporation of the liquid phase. The tungsten particles are melted in process 2. The tungsten in the liquid phase shrinks into spherical droplets under the action of the surface tension. At higher temperatures, there appears a strong tungsten evaporation.

The spheroidization process of powders in plasma is complicated. First, the plasma temperature field is not uniform, and the powders can only undergo phase transformation in the effective high temperature region (melting point or above). Second, a certain amount of tungsten powders needs to stay in the effective high temperature region for a period of time to absorb the latent heat of phase transition. The energy required for the phase transition for a certain amount of tungsten powder is determined, but the time required is affected by many factors. For simplicity, this time is mainly related to the heat transfer characteristics between the plasma and the particles, where the temperature and thermal conductivity of the plasma are important parameters.

In the pure argon plasma process shown in figure 2, due to the short plasma jet at 50 A, the tungsten powder cannot absorb enough heat to melt with downstream feeding. After the plasma treatment with the upstream feeding, a small number of spherical particles appears, which is thought to be due to the particles passing through a larger effective high temperature region.

Since the total ionization scattering cross section of nitrogen is smaller than that of argon [22], nitrogen discharge requires higher voltage than argon discharge under constant current condition. Figure 6 shows the input power as a function of nitrogen content in the working gas. As the nitrogen ratio increases, the input power is linearly increased to maintain the same discharge current, which leads to the increase of plasma jet length, thus expanding the effective high temperature region. On the other hand, OES measurement results showed that the electron excitation temperature increased from approximately 5000 K to 8000 K when nitrogen was added to the working gas. The thermal conductivity of nitrogen plasma around 7000 K is mainly derived from the reaction thermal conductivity due to the contribution of the dissociation reaction of nitrogen molecules [23]. Compared with argon, the dissociation reaction of diatomic molecules makes nitrogen plasma have higher thermal conductivity. In the argon-nitrogen mixed plasma, the higher thermal conductivity and temperature promote the heat transfer from the plasma to the particles. Improvements of plasma jet and heat transfer characteristics contribute to an increase in the spheroidization rate at high N$_2$/(N$_2$ + Ar) ratios.

The tungsten powders were characterized by energy dispersive spectroscopy. Figure 7 shows the distribution of tungsten element and oxygen element with corresponding characteristic spectra for tungsten powders. As can be seen, the oxygen content of the raw particle surface is higher and the spheroidized particle surfaces show reduced oxidation. During the spheroidization process, tungsten oxide evaporates from the tungsten particles due to its low boiling point, resulting in a significant reduction in the oxygen content of the spheroidized powders.
4. Conclusion

In summary, the spheroidization of refractory metal tungsten powders has been carried out by a low-power DC arc plasma torch in laminar flow state. Optical emission spectroscopy was used to investigate the plasma features. The morphology and the element contents of tungsten particles were characterized by SEM with EDS. It is difficult to achieve plasma spheroidization of tungsten powders with downstream feeding by using Ar discharge at the discharge current of 50 A, while a small amount of spheroidized particles can be obtained with the upstream feeding. With introducing N₂ into the discharge, the enhanced input power leads to an increase in the length of plasma jet, thereby expanding the effective high temperature region while maintaining the same discharge current. Meanwhile, it is found that the electron excitation temperature of N₂-Ar mixed gas discharge is higher than that of argon gas discharge. Due to the contribution of the dissociation of diatomic molecules, N₂-Ar mixed plasma has larger thermal conductivity compared with Ar plasma. As a result, the heat is more effectively transferred from the plasma to tungsten particles with introducing nitrogen in discharge. These are contributed to the improved spheroidization at high N₂/(N₂ + Ar) ratios. EDS measurement indicates decreases of oxygen content after spheroidization. It is believed that tungsten oxide evaporates from the tungsten particles due to its low boiling point in spheroidization process, resulting in a lower oxygen content in the spheroidized particles.

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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