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On the Induction Plasma Deposition of Tungsten Metal

X.L. Jiang, R. Tiwari, F. Gitzhofer, and M.I. Boulos

The central particle injection and long residence time characteristics of induction plasma have given rise to the complete melting of tungsten particles injected into an Ar-H_2 plasma under soft vacuum conditions. The influences of process variables such as power level, chamber pressure, and spray distance on splat morphology, apparent density, and deposition efficiency have been studied. Dense tungsten deposits with no oxidation have been obtained. Scanning electron microscopy (SEM) micrographs reveal a well-flattened lamellar structure in deposits. Radiative cooling is observed to play a significant role in the plasma spraying of this refractory metal.

1. Introduction

THERMAL plasma technology is a well-established interdisciplinary science with many applications in materials processing. Plasma processing may be classified on the basis of the role played by the plasma source. For instance, the plasma may be used as a source of high-temperature energy as in plasma spray-forming and powder spheroidization. Plasma is also a source of chemically active species, involving both physical and chemical processes. There are two main types of plasma sources: (1) direct current (dc) plasma and (2) inductively coupled radio frequency (rf) plasma sources. The latter has been extensively used for spray-forming applications.[1-4]

The induction plasma spray process is characterized by a large volume and low velocity plasma discharge.[5] An important feature of the induction plasma process is the lack of electrodes and the ability for axial injection of the feedstock powder into the plasma discharge along with longer residence times, on the order of 10 to 25 ms compared to 0.5 to 1 ms for the dc plasma process. This allows for the melting of relatively large particles at high powder loading without loss of melting efficiency. Due to the absence of electrodes, the deposition environment may be either inert, reducing, or oxidizing, and there is no deposit contamination coming from electrode erosion.

Tungsten is a refractory metal with a high melting point (T_m = 3683 K) and is a candidate material for many defense and high-temperature structural applications. However, it has low oxidation resistance at high temperatures and is susceptible to oxygen embrittlement. These considerations make conventional refractory metal-processing techniques less attractive. Induction plasma spraying, with its ability to handle high powder feeding and large particle sizes under an inert or reducing environment, emerges as a promising technique for tungsten or other refractory metal processing.

In this research work, a two-part effort was undertaken to study the plasma deposition of tungsten metal. The viability of a spray deposition process depends on the quality of the deposit and the material utilization. Because deposit density is an important parameter influencing its structural integrity and mechanical properties, it was used to assess the influence of process variables on the deposition process. Also, the dependence of the deposition efficiency, a measure of the material utilization, on the various process variables was calculated. Considering that a plasma deposit may be described as an aggregate structure comprised of successively deposited splats, attention was given to the influence of the process variables on the shape of splats formed on the substrate. The process variables considered are the plasma power level, chamber pressure, and spray distance, which have a very distinct effect on the deposit properties. In the first part of this article, the influence of these variables on the splat formation, apparent density (which is a percentage ratio of the deposit density to the theoretical density of feedstock material), and the deposition efficiency have been examined. In the second part, free-standing tungsten metal deposits were obtained using the optimum processing conditions, and their microstructures were analyzed.

Key Words: induction plasma processing, processing conditions, splat morphology, tungsten

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2. Experimental Procedure

Plasma spraying was performed using a 50-kW, 3-MHz induction plasma unit (Fig. 1). It consisted of a high energy density induction plasma torch with a 50-mm ID ceramic tube operated with a mixture of argon-hydrogen as sheath gas and pure argon as the central and powder carrier gases. The spraying conditions are summarized in Table 1. The powder was axially injected into the center of the plasma discharge using a water-cooled stainless steel probe positioned at the top of the induction coil. The exit flange of the torch was connected to a water-cooled stainless steel deposit chamber that was connected to a vacuum system.

Table 1 Induction plasma spray parameters

Central gas flow rate	40 slpm Ar
Sheath gas flow rates	90 slpm Ar + 9 slpm H_2
Carrier gas flow rate	4 slpm Ar
Plate power	25 to 45 kW
Chamber pressure	200 to 400 torr
Spray distance	175 to 300 mm
Powder feed rate	12 to 40 g/min

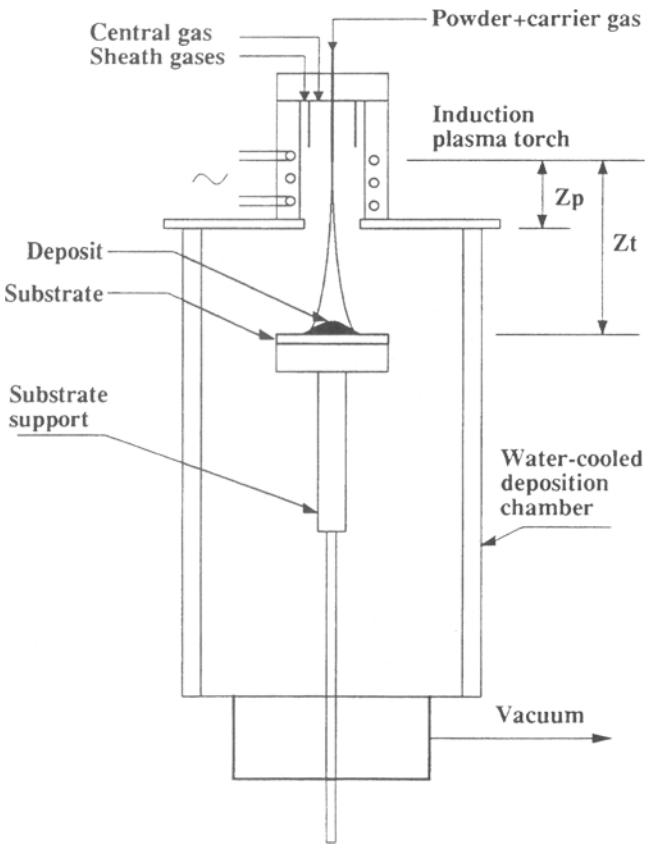

Fig. 1 Schematic of induction plasma spray setup. *Zp* = probe position; *Zt* = spray distance.

The feedstock tungsten powder, obtained from Sylvania, Chemicals/Metals, Towanda, PA, had a particle size distribution in the range of −75 +45 μm. Typical parameter values were 40 kW power level, 200 torr chamber pressure, 200 mm spray distance, and 20 g/min powder feed rate. Power levels of 25, 30, 35, 40, and 45 kW; chamber pressures of 200, 300, and 400 torr; and spray distances of 175, 200, 225, 250, 275, and 300 mm were used to study the effect of several process variables. The deposits were obtained on stationary graphite substrates that were presprayed with a boron nitride aerosol to enable removal of the deposits after spraying. Density measurements were carried out using water displacement methods. These were made for both the full deposit and for small samples cut from different sections of the deposit. Deposition efficiencies were also measured for each case. Optical and scanning electron microscopy along with X-ray diffraction studies were used for microstructural and phase composition characterization.

3. Processing/Microstructure Studies

The influence of the induction plasma process macrovariables, e.g., power level, chamber pressure, and spray distance, on the splat-flattening behavior and deposit formation is discussed below.

3.1 *Power Level*

The influence of the applied power level on the splat morphology is shown in Fig. 2. The chamber pressure was kept constant in this case at 200 torr, and the spray distance was 250 mm. At 40 kW, a greater degree of splat flattening is obtained com-

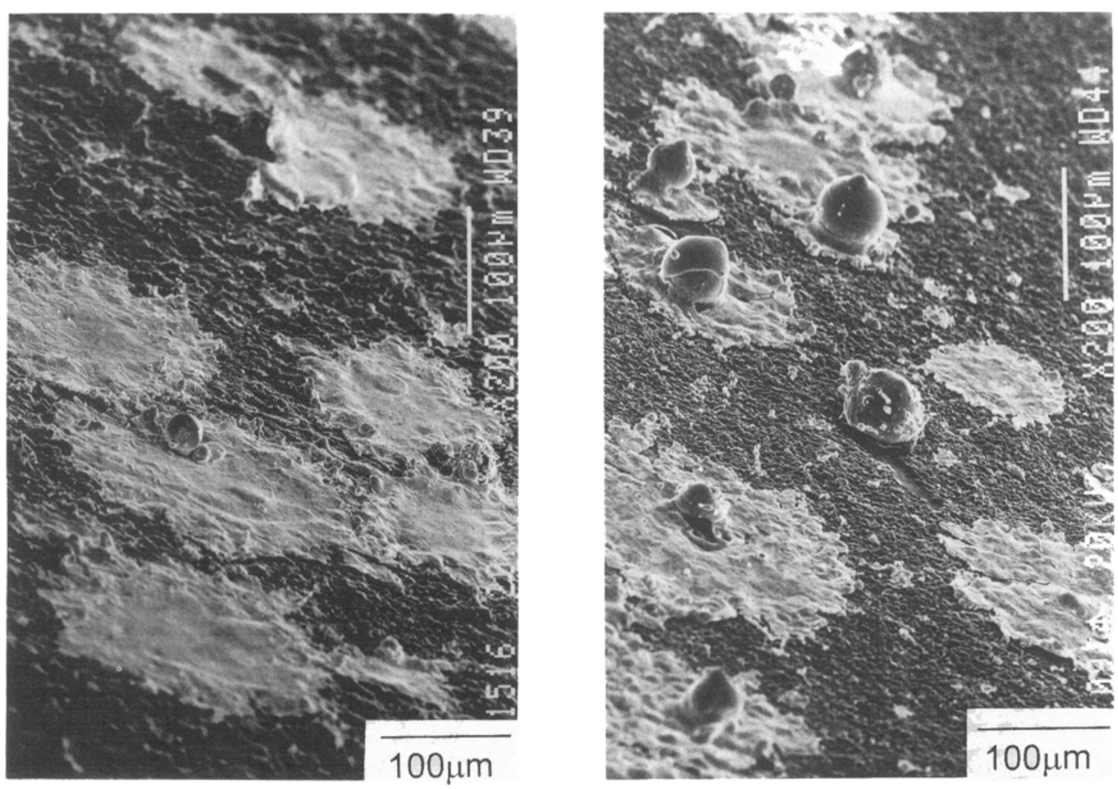

Fig. 2 Effect of power level on tungsten splat morphology at a chamber pressure of 200 torr and a spray distance of 250 mm. (a) 40 kW. (b) 30 kW.

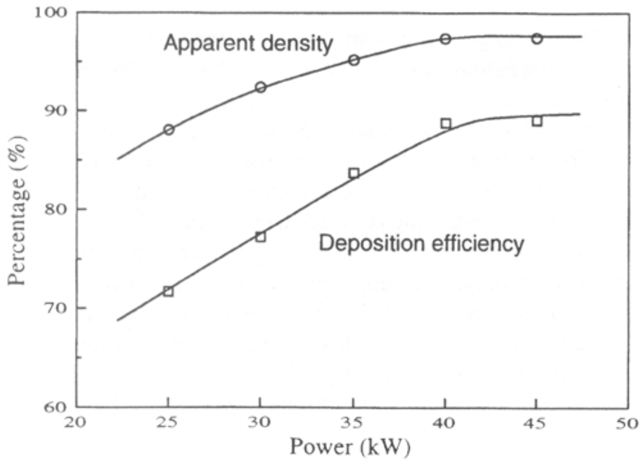

Fig. 3 Variation in deposit apparent density and deposition efficiency with plasma power level.

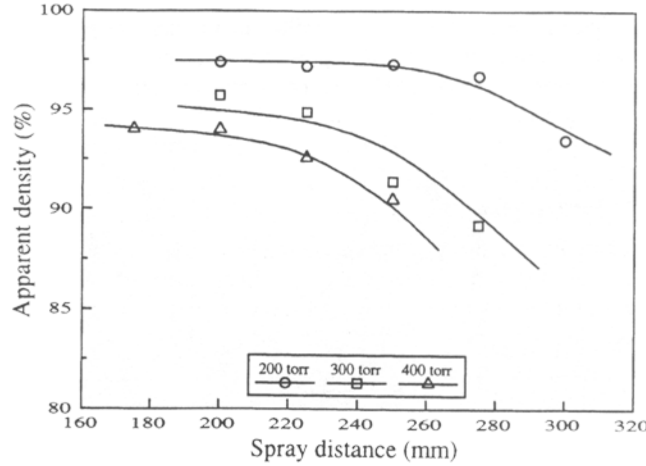

Fig. 5 Effect of chamber pressure and spray distance on the apparent density of tungsten at a plasma power of 40 kW.

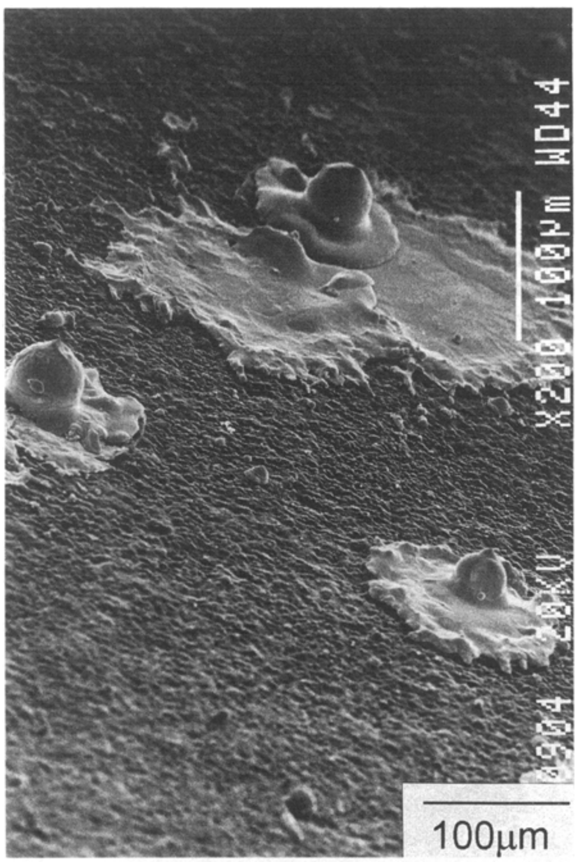

Fig. 4 Tungsten splats obtained at a plasma power of 40 kW, chamber pressure of 400 torr, and spray distance of 250 mm.

pared to that at 30 kW. This is due to the high temperature and hence the decreased viscosity of the molten particles prior to their impact on the substrate. This greater splat flattening, at higher power levels, gives rise to higher deposit densities, as shown in Fig. 3. A greater flowability of the splat at higher power levels minimizes the interlamellar porosity, thereby yielding higher deposit densities. The deposition efficiency is also enhanced at higher power levels.

The applied power level influences the plasma discharge volume and enthalpy. An increase in the applied power level causes a greater degree of ionization of the plasma gas, and thus, a higher plasma specific enthalpy and discharge volume is obtained. The higher enthalpy results in a greater degree of particle melting. Greater particle melting also results from the longer residence time of the particles in the plasma due to the increased discharge volume. This lowers the particle viscosity and leads to denser deposits.

3.2 *Chamber Pressure*

The chamber pressure is an important variable during low-pressure induction plasma spraying because it influences particle velocity and temperature. Bronet and Boulos,[6] using laser doppler anemometry, observed that a decrease in chamber pressure from 400 to 150 torr led to an increase in the particle velocity from 20 to 50 m/s. At lower chamber pressures, the particle velocities are enhanced due to the higher plasma velocity for the same mass flow rate of the powder carrier gas, central, and sheath gases. This effect is more pronounced in the case of dc plasma spraying. Additionally, Takikawa et al.[7] have reported a rapid decrease in particle temperature with an increase in chamber pressure. Particle cooling is enhanced because of the reduced velocity. In the case of plasma deposition of refractory metals, the radiative heat loss, which is proportional to the fourth power of the particle temperature, is a significant factor. Thus, it may be expected that, during plasma deposition of tungsten metal, the splat flowability and deposit densities are very sensitive to the chamber pressure. Figure 4 shows insufficient splat flattening at a chamber pressure of 400 torr, which can be attributed to the reduced particle velocity and consequently the increased heat loss by radiation. Operation at 200 torr is observed to be the optimum chamber pressure for splat formation and deposition. It should be mentioned that much lower chamber pressures, on the other hand, can lead to instability of the plasma and insufficient heat transfer to large feedstock particles due to the Knudsen effect.[8]

Figures 5 and 6 illustrate the influence of chamber pressure on the apparent density and deposition efficiency at different spray distances, respectively. At lower chamber pressures, the higher particle temperature and particle velocities combine to yield higher apparent densities and deposition efficiencies.

3.3 *Spray Distance*

During induction plasma spraying, the spray distance (*Zt*) is referred to as the distance between the tip of the powder feeding probe and the substrate surface, as shown in Fig. 1. The molten particles of tungsten metal are cooling as they approach the substrate past a certain point, and therefore, the particles impact the substrate with increased viscosity. The combination of higher particle temperature at reduced spray distance and larger velocity at lower chamber pressure gives rise to the explosive splats, as shown in Fig. 7(a), whereas at increased spray distance, a low degree of splat flattening is obtained due to the low particle temperature, as shown in Fig. 7(b). The reduced splat flattening leads to a decrease in the apparent density, which is illustrated in Fig. 5. In this study, the radiative heat loss is a principal mechanism that accounts for particle cooling. Therefore, a drastic decrease in the apparent density of the tungsten metal deposit occurs with increasing spray distance. A similar effect on the deposition efficiency is observed in Fig. 6.

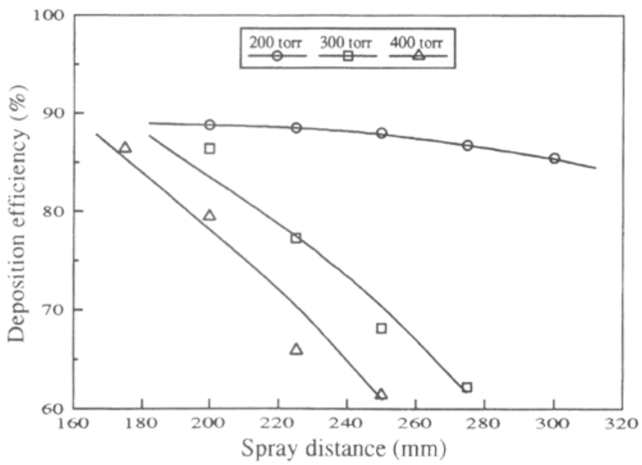

Fig. 6 Effect of chamber pressure and spray distance on the deposition efficiency of tungsten at a plasma power of 40 kW.

4. Microstructure Characterization

Free-standing tungsten metal deposits were obtained on fixed substrates using the optimum power level of 40 kW, chamber pressure of 200 torr, and spray distance of 200 mm for the particle size range and tungsten particle morphology used. The tungsten deposits have a cone-like structure. The axial and radial distribution of deposit density is shown in Fig. 8. The apparent density of the entire sample before cutting into pieces was measured to be 97.8 ± 0.2%. The deposit density is higher near the center of the substrate (98.8%) and decreases axially to 98% at the top of the deposit. The radial variation is greater because of the lower particle temperature and velocity near the periphery of the deposit. Also, it was observed that an increase

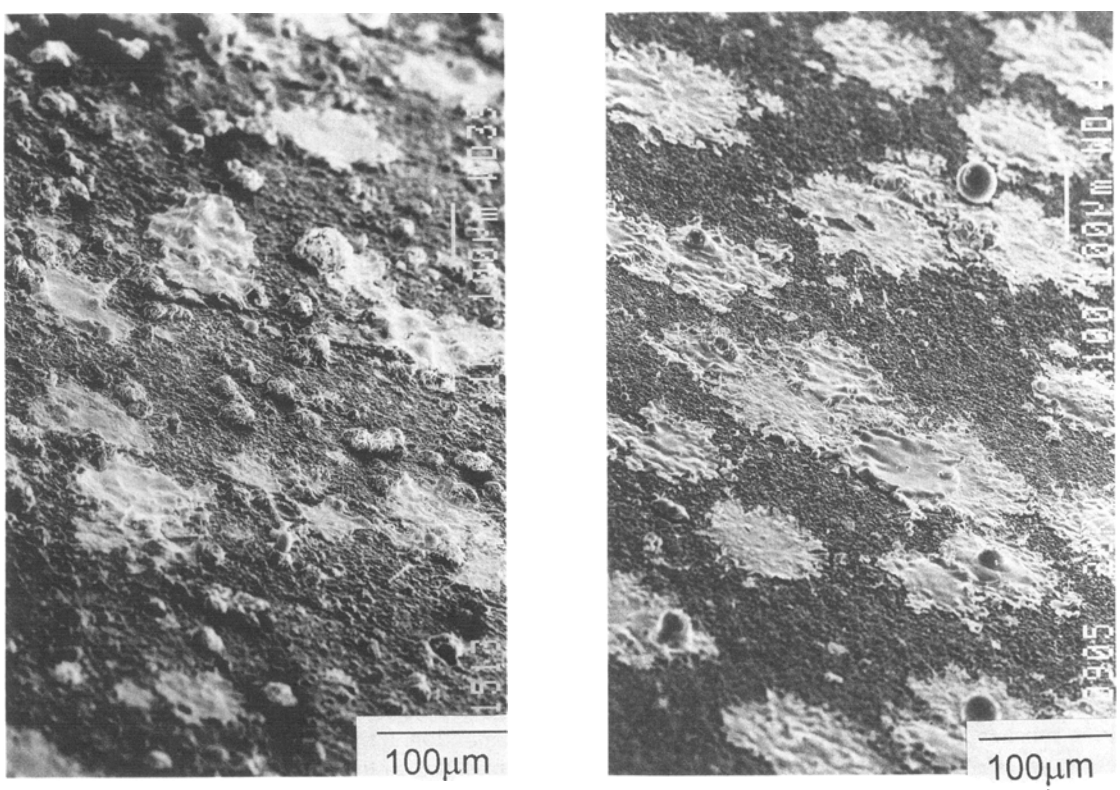

Fig. 7 Effect of spray distance on tungsten splat morphology at a plasma power of 40 kW and a chamber pressure of 200 torr. (a) *Zt* = 200 mm. (b) *Zt* = 300 mm.

in the powder feed rate over the range under study did not exhibit a significant decrease in the deposit density, as illustrated in Fig. 9.

The fracture surface of tungsten deposits reveals the lamellar and columnar structure shown in Fig. 10(a). Figure 10(b) is an electron micrograph showing the surface morphology of the as-sprayed deposit. A uniform overlapping of the splats is obtained, which could account for the low porosity levels obtained in these deposits. X-ray diffraction analysis (Fig. 11) shows the lack of oxidation of the tungsten metal during the deposition process. This is due to the inert low-pressure environment in the deposition chamber.

5. Conclusions

Induction plasma spraying has been shown to be a viable spray-forming technique for refractory metal processing. The inherent features of the induction plasma spray process, i.e., longer residence time due to lower plasma velocities and larger plasma volume and melting ability of axial injection of the feedstock powder, yield complete particle melting along with good splat flattening on the substrate and, consequently, a dense free-standing tungsten metal deposit of more than 97%. The applied power level, spray distance, and chamber pressure influence splat flattening and thereby the deposit density. Radiative heat

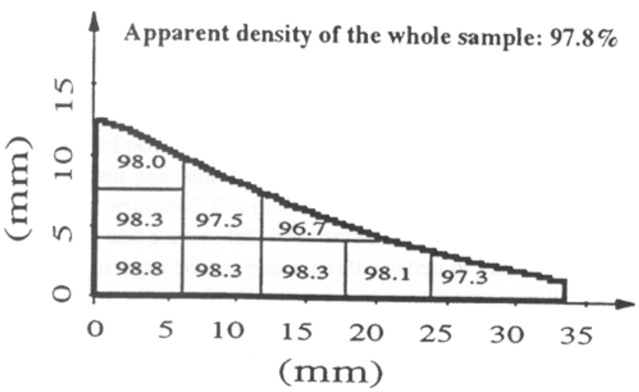

Fig. 8 Apparent density distribution of a tungsten deposit obtained at a plasma power of 40 kW, chamber pressure of 200 torr, and spray distance of 200 mm.

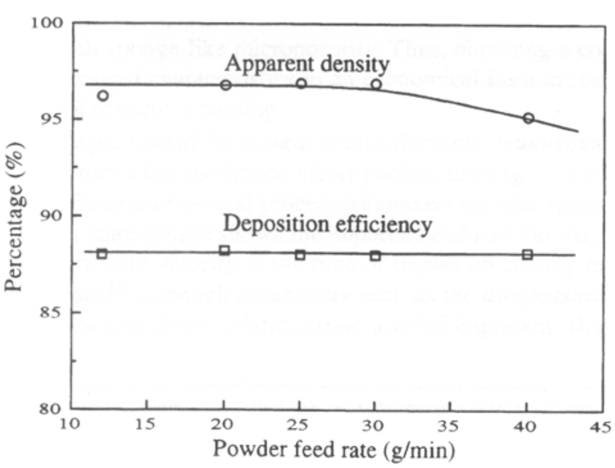

Fig. 9 Effect of powder feed rate on tungsten deposit density and deposition efficiency.

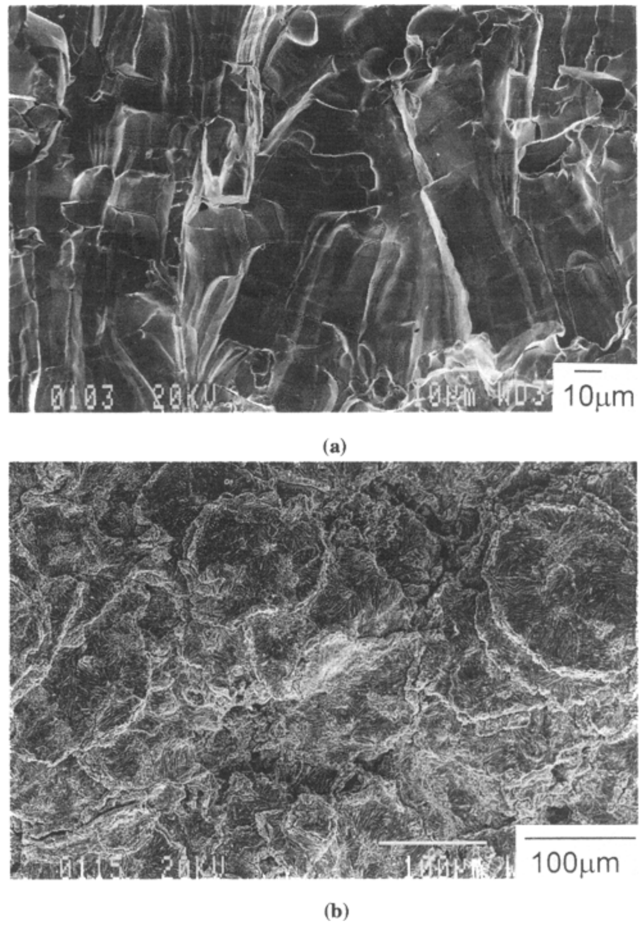

Fig. 10 SEM micrographs of a tungsten deposit obtained under the same condition as Fig. 8. (a) Fracture surface. (b) Top surface of the deposit.

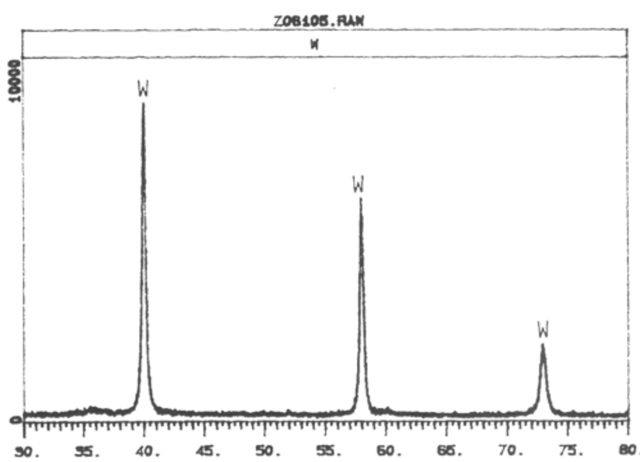

Fig. 11 X-ray diffraction pattern of a tungsten deposit obtained under the same conditions as Fig. 8.

loss is a significant phenomenon influencing the deposit density and deposition efficiency. The use of an inert low-pressure deposition environment leads to minimal deposit oxidation and increased deposit density.

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