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Particle melting, flattening, and stacking behaviors in induction plasma deposition of tungsten①

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[Abstract] Particle melting, flattening, and stacking behaviors during induction plasma deposition of refractory tungsten were studied for land-based turbine engine application. Scanning electron microscopy was used to observe the morphology of particles and splats as well as to examine the microstructure of tungsten deposit. Three kinds of pores were found in the deposit, i. e., large pores with $d > 10\ \mu m$, medium pores in the range of 1~ 10 μm, and small pores with $d < 1\ \mu m$. Both optimized plasma spray condition and use of spherical powder with a narrow particle size distribution are important in the elimination of large and medium pores and have significant influences on the formation of dense tungsten deposit. Highly dense tungsten deposit was obtained through complete melting, sufficiently flattening, and regularly stacking of tungsten particles.

[Key words] induction plasma; tungsten; deposition

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1 INTRODUTION

Thermal spray is one of the major techniques used for surface modification and coating deposition. Thermal sprayed coatings, particularly plasma sprayed coatings, are applied in anti-wear and erosion, against oxidation and corrosion, resisting heat, forming seals, and providing lubrication. Materials commonly used for these applications include metals, ceramics, polymers, and their composites. In plasma spraying, powders are injected into plasma through a powder-feeding unit. Particles melt, flatten, and stack, forming deposits or coatings on substrate.

Tungsten and molybdenum are candidate materials to be fabricated into turbine blades for land-based turbine engine application[1]. Tungsten coating is also used for nuclear fusion devices[2,3]. Properties of tungsten coating were measured[4,5]. Processes for molybdenum splat formation[6] and coating deposition[7] were also investigated.

Recently, induction plasma technology gains momentum to be industrialized. The advantageous characteristics of the radio frequency induction plasma include large plasma volume, long particle residence time, central injection of powders into plasma, and absence of electrodes[8]. All these characteristics favor the synthesis of ceramic powders[9, 10] and the formation of dense deposits and coatings from large size powders[11, 12].

2 EXPERIMENTAL

To form highly dense tungsten deposit via induction plasma spraying, particle melting behavior, particle flattening behavior, and splats stacking behavior were studied in three steps.

In the first step, fused and crushed tungsten powder was fed into the plasma. After plasma processing, the powder was collected. SEM was used to examine the change of particle morphologies. An experimental set-up for this purpose is illustrated in Fig. 1. Induction plasma was generated at the frequency of 3 MHz. Ar was used as central plasma gas with the flow rate of 40 L/min. A mixed gas of Ar+ H_2 was used as sheath gas with the flow rate of 90 L/min (Ar) + 9 L/min (H_2). The powder was axially fed into the plasma through a high pressure water-cooled injector with 2.5 mm in internal diameter. Most of the fed-in powder was collected at the bottom of the chamber that was connected to an evacuation system. The experimental condition used for the study of particle melting and spheroidization behavior is given in Table 1.

Table 1 Experimental condition for particle melting and spheroidization

Plasma power /kW	Chamber pressure /kPa	Particle size /μm	Powder feed rate /(g·min⁻¹)	Carrier gas flow /(L·min⁻¹)
30~ 40	47~ 87	45~ 75	20~ 80	4

In the second step, a substrate was put into the upper position of the chamber to receive particles and to form splats. A schematic of the experimental set-up for the study of particle flattening behavior is shown in Fig. 2. Two kinds of substrate were used.

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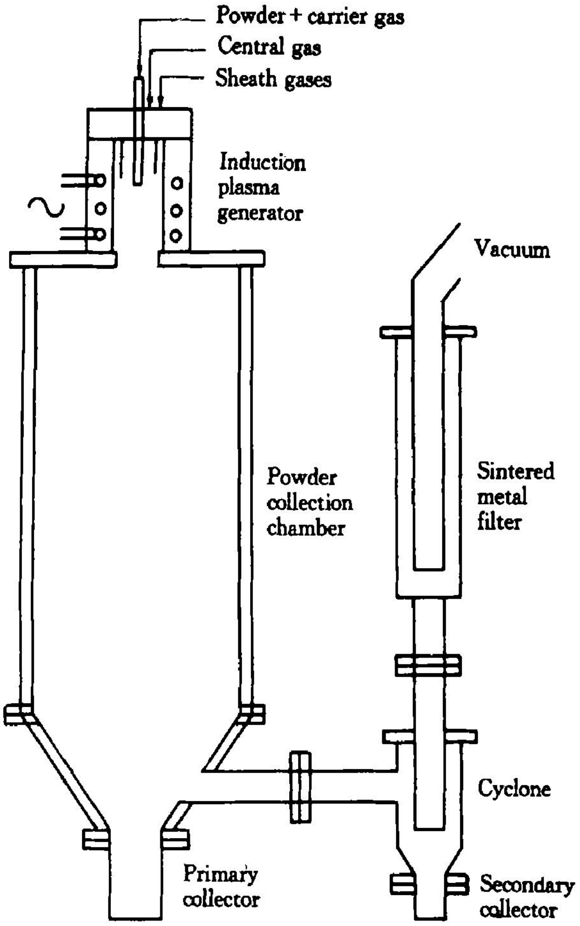

Fig. 1 Schematic of experimental set-up used for melting and spheroidization of tungsten powder

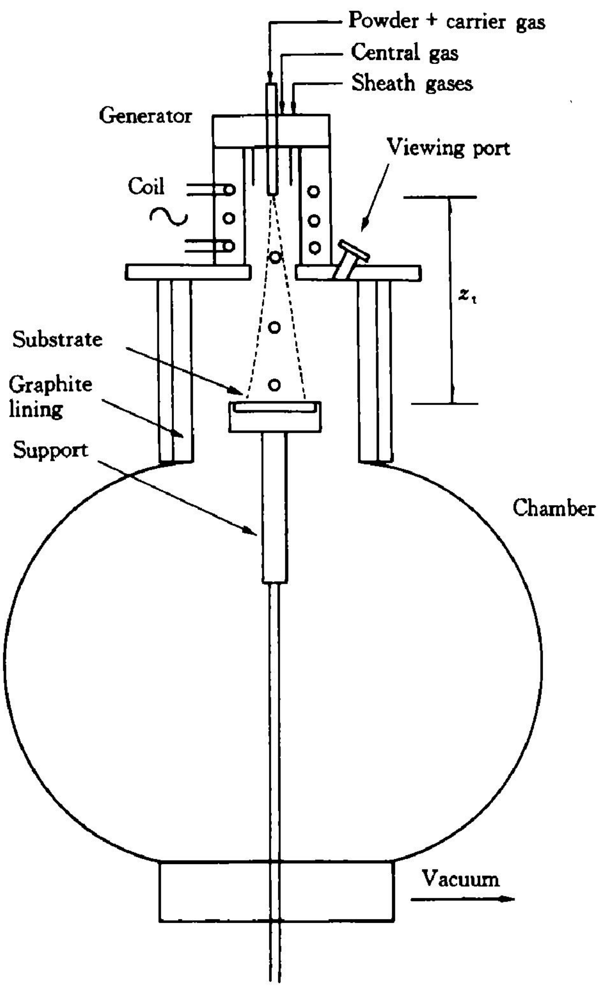

Fig. 2 Schematic of experimental set-up used for splat formation and deposition of tungsten in induction plasma

For spray distance of less than 300 mm, smooth alumina substrate was used. For spray distance of more than 300 mm, stainless steel substrate without grit blasting was used. A substrate support was moved up and down for adjustment of spray distance. The parameters used in the second step are shown in Table 2.

In the third step, the same experimental set-up as in the second step was used. Graphite substrate, 5 mm thick and 100 mm in diameter, was used in the third step for study of splats stacking behavior and for deposit formation. Before the deposition, BN aerosol was sprayed on the graphite substrate to facilitate removal of the deposit from the substrate. The process parameters studied in the third step are given in Table 3.

Tungsten deposit samples were cut by a diamond blade. SEM was used for examination of microstructure and pore size, X-ray diffraction for detection of any oxide, and water displacement method for measurement of deposit density.

3 RESULTS AND DISCUSSION

3.1 Particle melting and spheroidization behavior

Physical properties of tungsten metal are given in Table 4. SEM micrograph of starting tungsten powder is shown in Fig. 3(a). Most particles of the powder were in the range of 45 ~ 75 μm. Fig. 3(b) is the

Table 2 Experimental condition for particle melting and splat formation

Plasma power / kW	Chamber pressure / kPa	Spray distance / mm	Particle size / μm	Powder feed rate / (g·min^{-1})	Carrier gas flow rate / (L·min^{-1})
30~ 40	27~ 53	200~ 350	53~ 63	20	4

Table 3 Experimental condition for splats stacking and deposit formation

Plasma power / kW	Chamber pressure / kPa	Spray distance / mm	Particle size / μm	Powder feed rate / (g·min^{-1})	Carrier gas flow rate / (L·min^{-1})	Deposition time / min
25~ 45	27~ 53	175~ 320	45~ 75	12~ 40	2~ 8	2~ 40

Table 4 Physical properties of tungsten metal

Theoretical density $/(g\cdot cm^{-3})$	Thermal conductivity $/(W\cdot m\cdot K^{-1})$	Specific heat $/(J\cdot kg\cdot K^{-1})$	Melting point /℃	Latent heat of fusion $/(kJ\cdot kg^{-1})$
19.3	180	134	3410	192

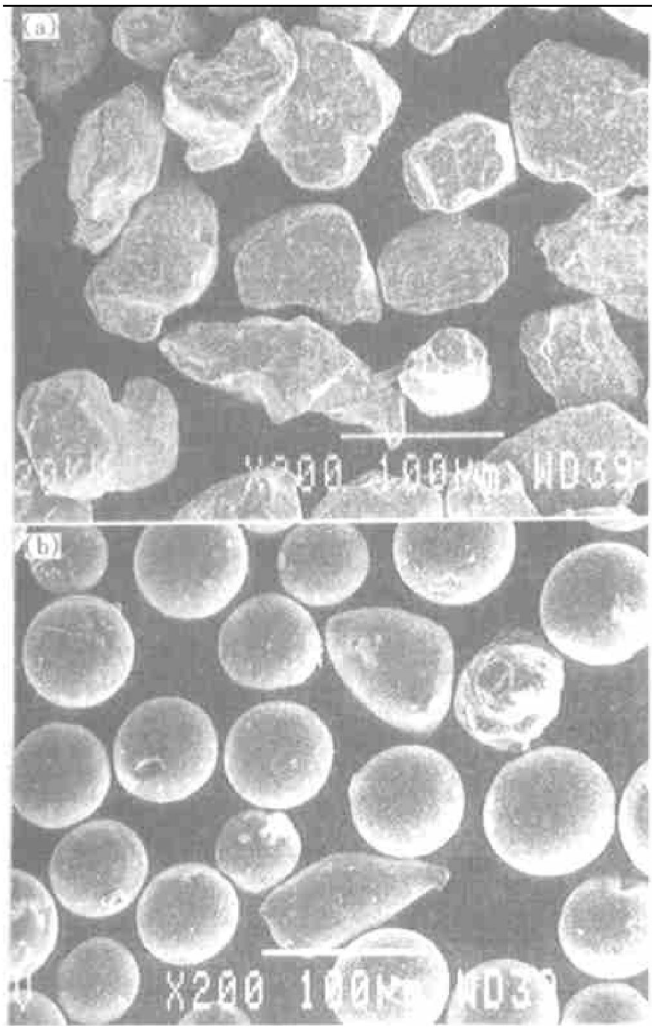

Fig. 3 SEM micrographs of tungsten powder
(a) —Starting state; (b) —After plasma processing

SEM micrograph of the powder collected at the chamber bottom, after being processed under the condition of plasma power of 40 kW, chamber pressure of 67 kPa, and powder feed rate of 80 g/min. Almost all particles of the plasma-processed powder were melted and spheroidized. Analysis of particle size distribution indicated that there was a slight increase of mean particle size because a fraction of the plasma-processed powder was extracted to the cyclone where small particle size powder was collected. SEM examination of the cross-sections of the plasma-processed powder revealed that most of the particles were dense, as shown in Fig. 4.

3.2 Particle solidification and splat formation behavior

Tungsten splats with sufficient flattening on substrate will not be formed when plasma spray condition is not optimized. During the experiment of particle

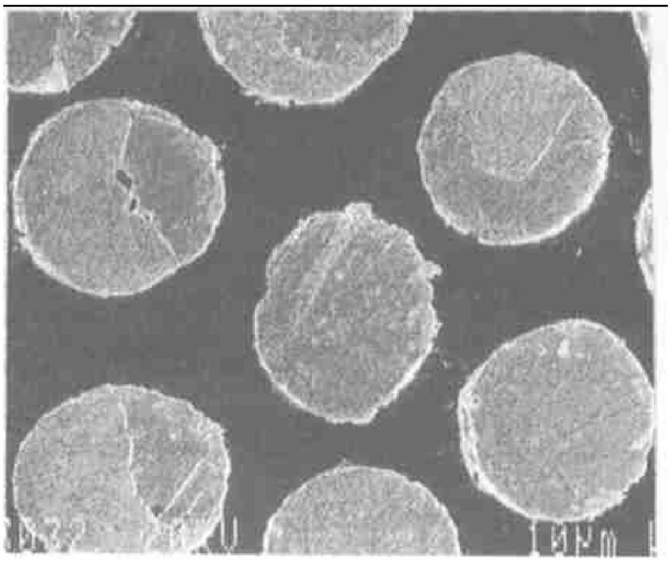

Fig. 4 SEM micrograph of cross sections of tungsten particles with plasma processing

solidification and splat formation, unmelted/solidified particles are bounced away from substrate. Stationery substrate was used in this experiment.

There are a variety of tungsten splats that are formed on the substrate during the induction plasma spraying, but they can be categorized into three groups. The first group refers to the splats with sufficient flattening on the substrate before the solidification of molten particles begins. A typical splat in this group is illustrated in Fig. 5(a). It was believed that the molten particle reached much higher temperature than tungsten melting point in the plasma. When it impinged on the substrate, the particle had time to spread sufficiently before its temperature reached 3410 ℃. The ratio of diameter to thickness (d/t) of such a splat was as high as 15.

The splat shown in Fig. 5(b) belongs to the second group. It was seen from this SEM micrograph that only surface layer of the molten particle was spread but its center was not deformed when the particle impacted with the substrate. Such a flattening behavior revealed that the temperature of the particle was just above its melting point. It did not have enough time to flatten before its solidification begins.

The splats formed on the substrate without obvious flattening represent the third group, as shown in Fig. 5(c). The molten particle was almost solidified at the moment it arrived at the substrate. This phenomenon happened when large spray distance was used.

It was found that a few principal parameters had great effects on the patterns of tungsten splats formed

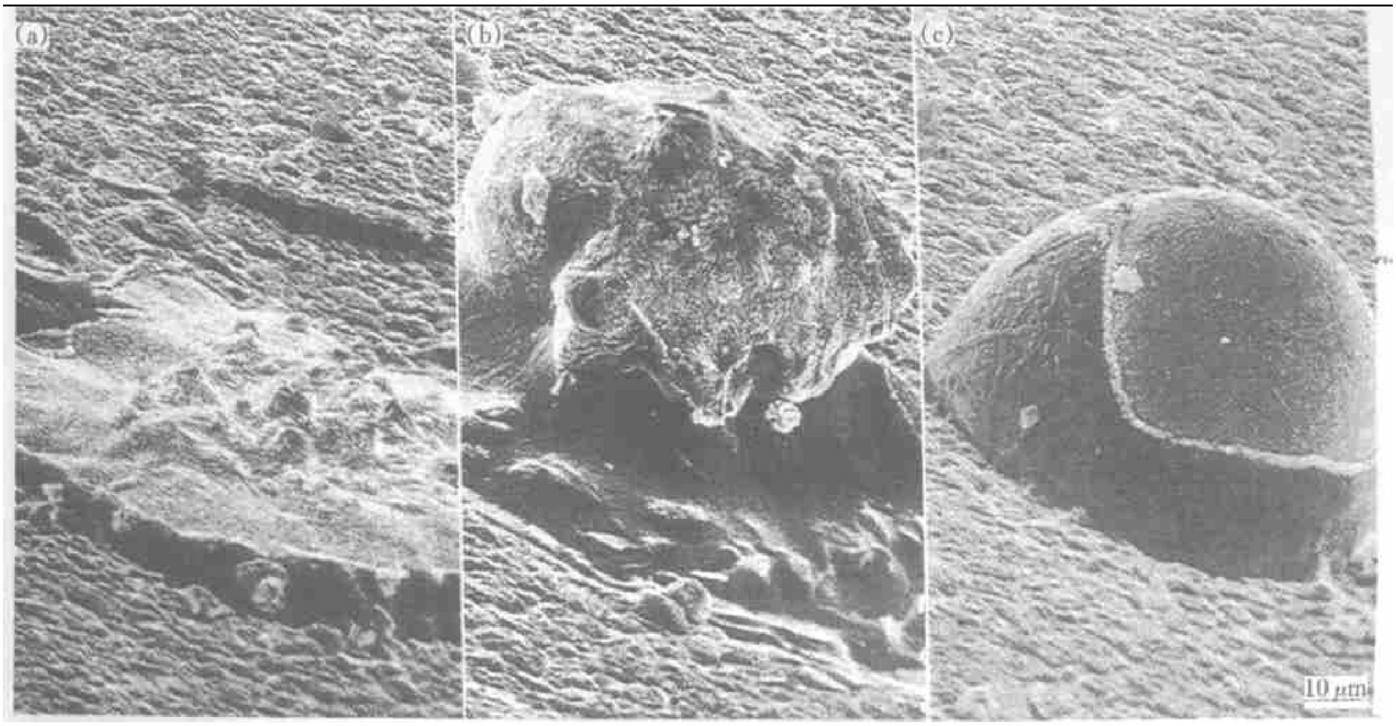

Fig. 5 SEM micrographs of representative tungsten splats
(a) —With sufficient flattening on substrate; (b) —With partial flattening on substrate; (c) —Without flattening on substrate

during the induction plasma deposition. For example, molten particles did not flatten very well on the substrate when chamber pressure was high, partially because the particles did not have enough kinetic energy. When substrate temperature was very low, molten particles solidified too quickly to flatten sufficiently on the substrate. Also, molten particles were not spread completely when substrate was very rough. Tungsten splats with a high ratio of diameter to thickness were obtained when major plasma spray parameters were optimized, as shown in Fig. 6. These splats were formed under the condition of plasma power of 40 kW, chamber pressure of 27 kPa, and spray distance of 250 mm.

3.3 Splats stacking and deposit formation behavior

Dense tungsten deposits will not be obtained when the splats are not stacked regularly. Experimental result on the induction plasma deposition of tungsten demonstrates that highly dense tungsten deposits can not be obtained only when plasma spray parameters are optimized but without use of sprayable tungsten powder. Under the viscous flow of induction plasma, the fused and crushed tungsten powders can not be uniformly distributed on substrate, which is required for the formation of dense deposits. However, the uniformity of the particle distribution on the substrate has been greatly improved after spherical tungsten powder with a narrow particle size distribution was used. Microstructures at the cross section and top surface of a representative tungsten deposit are shown Fig. 7. This deposit was made under the following condition: plasma power of 40 kW, chamber pressure of 27 kPa, spray distance of 200 mm, particle size of 45~ 75 μm, spherical powder feed rate of 20 g/min, and powder carrier gas flow rate of 4 L/min. Splats with sufficient flattening were found on the top surface of the deposit. Tight stacking of the splats made the deposit highly dense. Density measurement showed that there was no more than 2% porosity in the deposit.

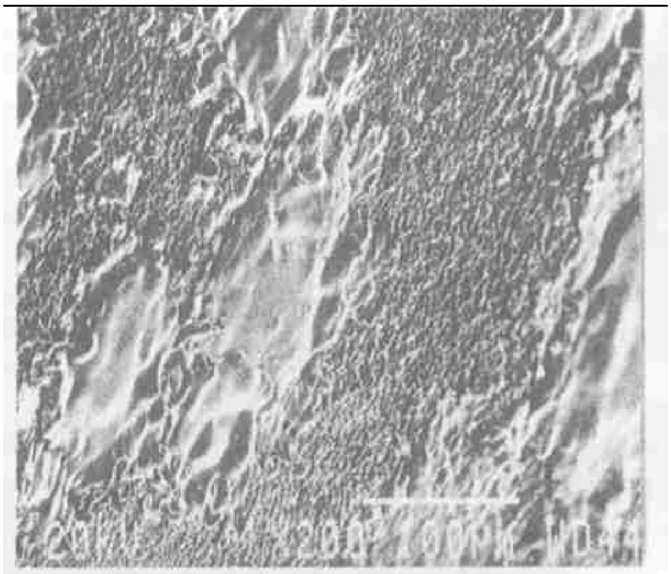

Fig. 6 SEM micrograph of tungsten splats with sufficient flattening on substrate under typical plasma spray condition

Lamellae in the radial direction and columnar grains in the axial direction are presented in Fig. 7(a). Typical lamellae had the diameter of approximately 150 μm and the thickness of 10 μm. The

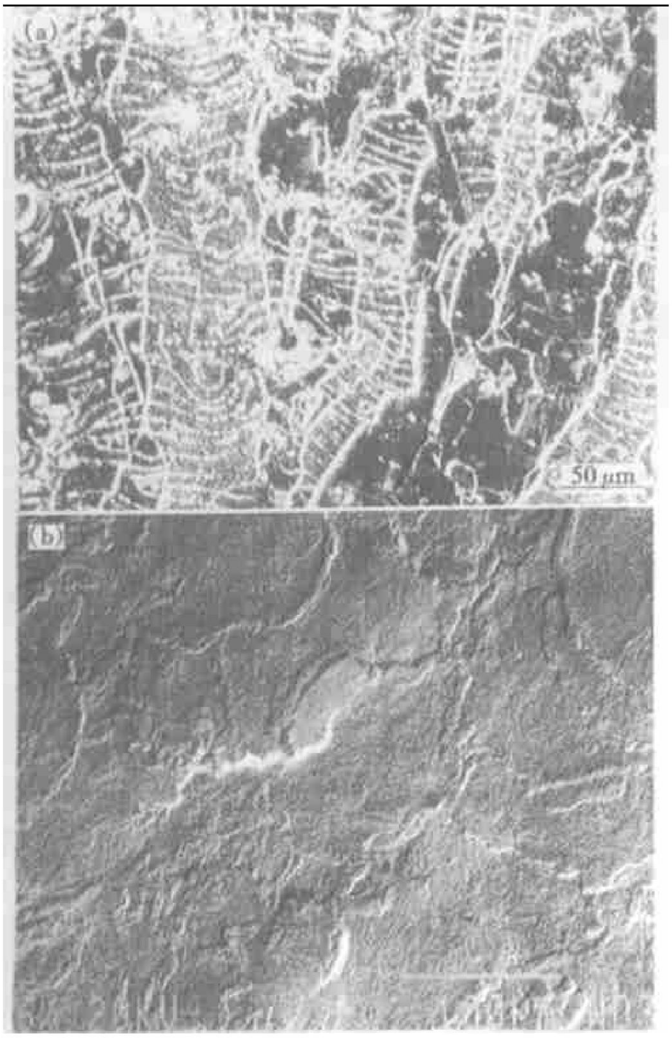

Fig. 7 Micrographs of representative tungsten deposit made from spheroidized powder under optimized plasma spray condition
(a) —Cross section (OM); (b) —Top surface (SEM)

large columnar grains had an average diameter of 80 μm. Length of the columnar grains was as high as 1 mm. Obviously, great grain growth occurred in the deposit on the substrate. Temperature measurement indicated that substrate temperature reached up to 1 800 ℃ after deposition for about 10 min. At such a high temperature, grain growth was inevitable. No obvious crack was found in the tungsten deposit. X-ray diffraction analysis did not reveal any oxidation of tungsten.

3.4 Large, medium, and small pores in tungsten deposit

Three kinds of pores in the tungsten deposit are identified, namely, large pores, medium pores, and small pores. Characteristics and formation mechanism of each kind of pores are discussed as follows.

Large pores found in the tungsten deposit are in the range of 10~ 50 μm, as shown in Fig. 8(a). The large pores resulted from the stacking of unmelted particles or particles solidified in-flight before reaching the substrate, accounting for up to 10% in volume. This kind of pores have been minimized when plasma spray parameters were optimized.

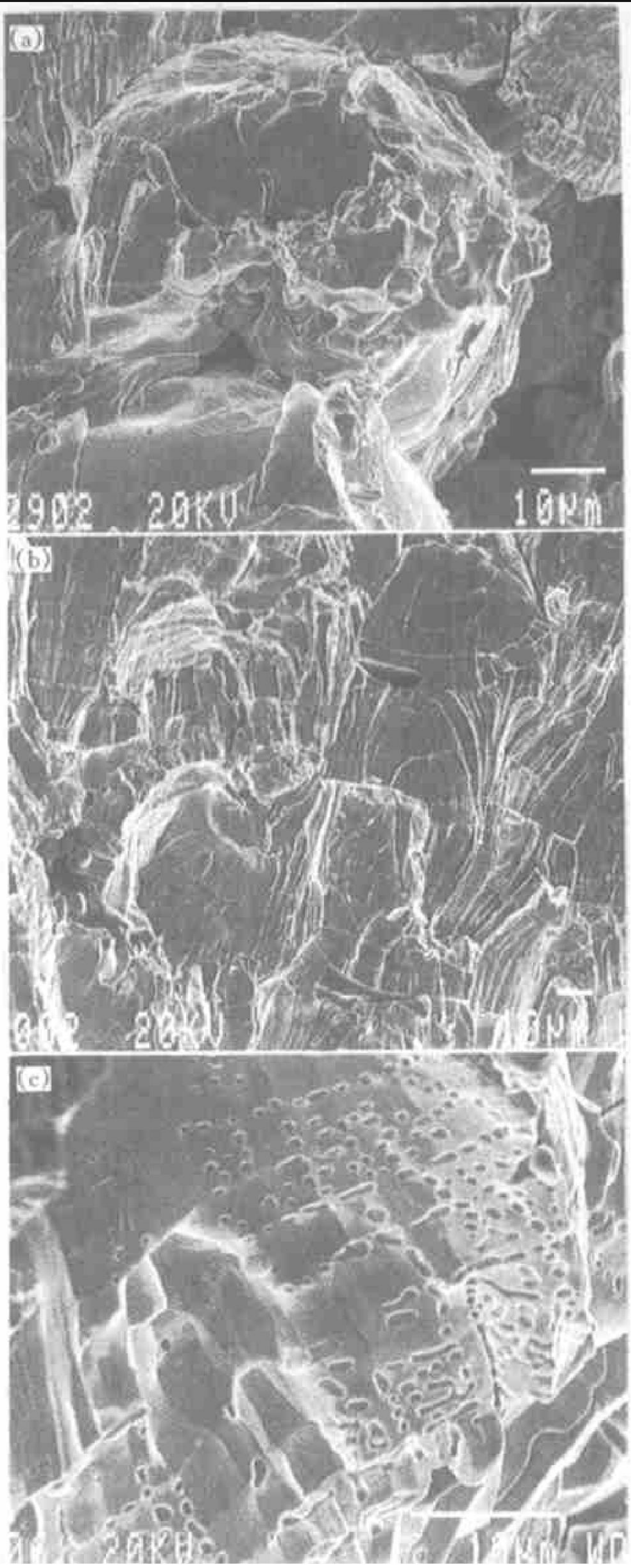

Fig. 8 SEM micrographs of fracture surfaces of tungsten deposits
(a) —Large pores; (b) —Medium pores; (c) —Small pores

Medium pores are referred to as the pores in the range of 1~ 10 μm, as shown in Fig. 8(b). This kind of pores was found at the inter-lamellae that were formed by either irregular stacking of splats or volume contraction when the tungsten deposit cooled. The medium pores have been minimized when an optimized plasma spray condition and spherical solid powder with a narrow particle size distribution were used.

Small pores are defined as those with the size of less than 1 μm. As shown in Fig. 8(c), a large number of small pores were found at the fracture surface of a tungsten deposit. Involvement of plasma gases into the deposit was the main cause to the formation of the small pores. This kind of pores, that accounting for up to 1% in volume, was impossible to be eliminated.

4 CONCLUSION

Highly dense tungsten deposit made from large size powder was obtained through complete melting of particles in the plasma, sufficient flattening of molten particles, and regular stacking of splats on the substrate, when plasma spray condition was optimized and spherical tungsten powder with a narrow particle size distribution was used.

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