

Plasma spray forming of tungsten coatings on copper electrodes^①

JIANG Xian-liang(蒋显亮)¹, F. Gitzhofer², M. I. Boulos²

(1. School of Materials Science and Engineering, Central South University, Changsha 410083, China;
2. Plasma Technology Research Center, University of Sherbrooke, Sherbrooke, J1K 2R1, Canada)

Abstract: Both direct current dc plasma and radio frequency induction plasma were used to deposit tungsten coatings on copper electrodes. Fine tungsten powder with mean particle size of 5 μm and coarse tungsten powder with particle size in the range from 45 μm to 75 μm were used as plasma spray feedstock. It is found that dc plasma is only applicable to spray the fine tungsten powder and induction plasma can be used to spray both the coarse powder and the fine powder. The tungsten coating deposited by the induction plasma spraying of the coarse powder is extremely dense. Such a coating with an interlocking structure and an integral interface with the copper substrate demonstrates high cohesion strength and adhesion strength.

Key words: plasma spraying; tungsten; particle size; interlocking structure; adhesion

CLC number: TG 156.8

Document code: A

1 INTRODUCTION

An essential requirement for Tokamak accelerator is that the injected fuel should entrain minimum impurities^[1]. To satisfy this requirement, the coating material on the copper electrode of Tokamak accelerator must have a high melting point, good electric conductivity, low absorption and release of gases. It is found that this goal can be achieved only through plasma spray forming of tungsten coatings on the copper electrode. Tungsten coatings^[2-5] were developed for nuclear fusion devices. For application of tungsten coatings in Tokamak accelerator, tungsten coatings should have the thickness of approximately 300 μm , density more than 97% of the theoretical, purity above 99. 9%, good toughness, high adhesion, smooth surface, and clean coating/substrate interface.

Direct current plasma has the characteristics of small plasma volume, high energy density, lateral injection of powders, high particle velocity, and short particle residence time. It has been used in industry for depositing anti-wear coating, corrosion-resistant coating, thermal barrier coating, etc. Radio frequency induction plasma has the characteristics of large plasma volume, low energy density, central injection of powders, low particle velocity, long particle residence time, and lack of electrode contamination^[6]. Induction plasma spraying, as a new technology, is being applied to the melting, spheroidization and deposition of the refractory materials such as W, Mo

and Al_2O_3 ^[7-10], as well as to the synthesis and processing of ceramic materials^[11-15].

In this study, both direct current plasma and radio frequency induction plasma are used to spray fine tungsten powder and coarse tungsten powder. Comparisons of the microstructure, adhesion strength and toughness of tungsten coatings formed by the two methods are presented.

2 EXPERIMENTAL

Both direct current plasma spraying and radio frequency induction plasma spraying were conducted in the spraying chamber connected to a vacuum system. The soft vacuum/ low pressure (10-30 kPa) of the spray chamber was used for preventing the oxidation of tungsten particles melted in plasma. Argon was used as primary plasma gas and hydrogen was used as secondary plasma gas. The substrate used is a hollow copper cylinder with approximately 20 mm in diameter and 400 mm in length. The hollow cylinder was mounted on a rotary shaft cooled by high pressure cooling water. Fine tungsten powder used as spray feedstock has the mean particle size of 5 μm and coarse tungsten powder has the particle size in the range from 45 to 75 μm .

During dc plasma spraying, tungsten coatings were generated by moving plasma gun horizontally on the rotary copper substrate. A sphere-shape chamber with a diameter of about 1 500 mm was used. Tungsten powder was laterally fed into the plasma via two

① Received date: 2004-03-27; Accepted date: 2004-06-10

Correspondence: JIANG Xianliang, Professor, PhD; Tel: +86-731-8876307; E-mail: xljiang@mail.csu.edu.cn

opposite powder injectors. The condition for the dc plasma spraying is given in Table 1. During radio frequency induction plasma spraying, tungsten coatings were generated by moving the copper substrate horizontally and rotating it simultaneously because plasma gun/torch was fixed to an oscillating unit of power supply system and could not be moved. In contrast to lateral injection of powders in dc plasma spraying, tungsten powders were axially fed into the center of the plasma with a water-cooled powder injector in induction plasma spraying. The condition for the induction plasma spraying is given in Table 2. It should be pointed out that spray distance was calculated from the tip of the water-cooled powder injector to the surface of the copper substrate.

Table 1 dc plasma spray condition for tungsten coatings

Plasma gas flow/ (L·min ⁻¹)	Plasma power/ kW	Powder feed rate/ (g·min ⁻¹)
40(Ar) + 14(H ₂)	45	50
Carrier gas flow/ (L·min ⁻¹)	Chamber pressure/ kPa	Spray distance/ mm
1(Ar)	13	80

Table 2 Induction plasma spray condition for tungsten coatings

Sheath plasma gas/ (L·min ⁻¹)	Central plasma gas/ (L·min ⁻¹)	Plasma power/ kW
90(Ar) + 9(H ₂)	40(Ar)	40
Powder feed rate/ (g·min ⁻¹)	Carrier gas flow/ (L·min ⁻¹)	Chamber pressure/ kPa
20	4(Ar)	26
		Spray distance/mm
		250

Tungsten coatings on the substrate were cooled to room temperature before being taken out from the spray chambers. Coating samples were cut by a low speed rotary diamond saw. After being ground and polished, the samples were examined under the JSM-840A scanning electron microscope equipped with an EDAX system. Density of the coating, chiseled from the copper substrate, was measured using the method of water displacement.

3 RESULTS AND DISCUSSION

3.1 Microstructure and density of tungsten coatings

Morphology of the fine tungsten powder is shown in Fig. 1. The powder consists of non-uniform individual crystals. There are some connections a-

mong individual crystalline particles. The mean particle size of the powder is 5 μm . The microstructure of the coating formed via the dc plasma spraying of the fine powder is shown in Fig. 2. Because of the high velocity ($> 300 \text{ m/s}$) of tungsten particles attained inside the dc plasma at low pressure, molten tungsten particles have sufficient momentum to be deformed when impacted on the substrate. As a result, the stacking of deformed particles is highly dense and almost no pore can be found in the coating. Also, the interface between the coating and the substrate is clean and integral. Density measurement of the coating indicates that the coating has the apparent density of 97%.

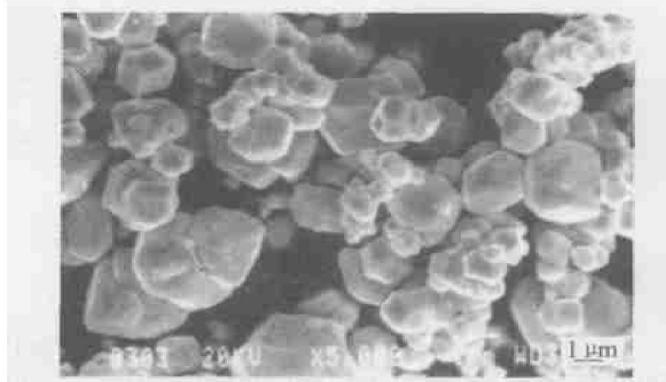


Fig. 1 Morphology of fine tungsten powder used for plasma spraying

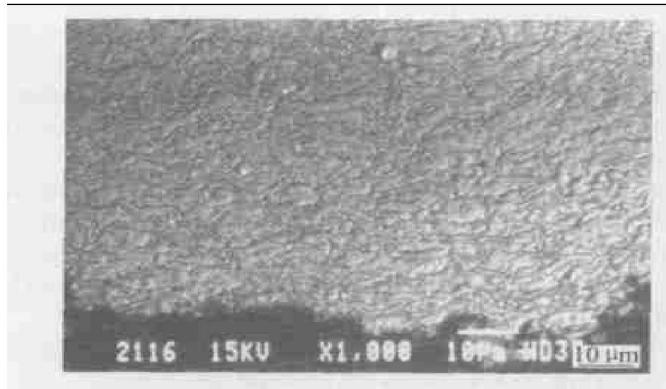


Fig. 2 Cross-sectional morphology of tungsten coating formed via dc plasma spraying of fine powder

The microstructure of the tungsten coating made from the fine powder using the induction plasma spraying is illustrated in Fig. 3. This coating is less dense than the coating formed by dc plasma spray. Pores and unmelted particles can be found in the coating. There are two reasons for explaining the more pores and unmelted particles found in the coating formed via induction plasma spraying. Firstly, induction plasma has lower particle velocity ($< 100 \text{ m/s}$) than dc plasma and the kinetic energy of the small particles with the lower velocity is insufficient to generate a high impact force. Secondly, the spray dis-

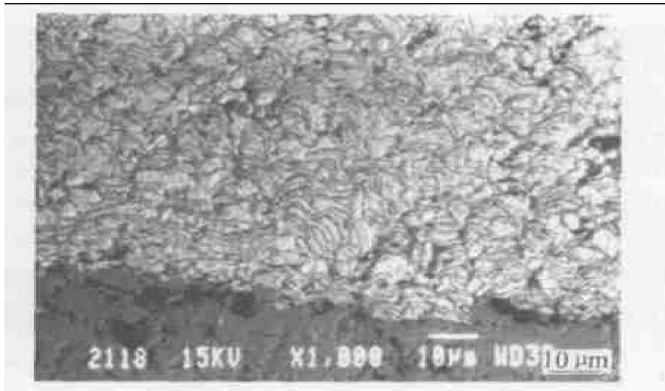


Fig. 3 Cross-sectional morphology of tungsten coating formed via induction plasma spraying of fine powder

tance (250 mm) used in the induction plasma spray is longer than the one (80 mm) used in the dc plasma spray and thus some of the molten tungsten particles have been solidified before arriving at the substrate.

The morphology of the coarse tungsten powder is shown in Fig. 4. This powder, consists of spheric particles, has been processed by induction plasma and thus the tungsten particles were spheroidized. The powder used for plasma spray forming of the coating has the particle size in the range from 45 to 75 μm . When the coarse tungsten powder was used in the dc plasma spraying, however, tungsten coating can not be generated. Only a few particles were melted and attached on the substrate. A dominant portion of the injected particles were not melted and bounced away from the substrate. This phenomenon is due to the very limited particle residence time ($< 10 \text{ ms}$) in dc plasma spraying, especially operated at low pressure.

When the coarse tungsten powder was used in the induction plasma spraying, some advantages have been demonstrated over dc plasma spraying. Fig. 5 shows the SEM micrograph of the cross section of a typical tungsten coating formed via induction plasma spraying of the coarse powder. It is seen that the coating, consists of large interlocking laminar layers,

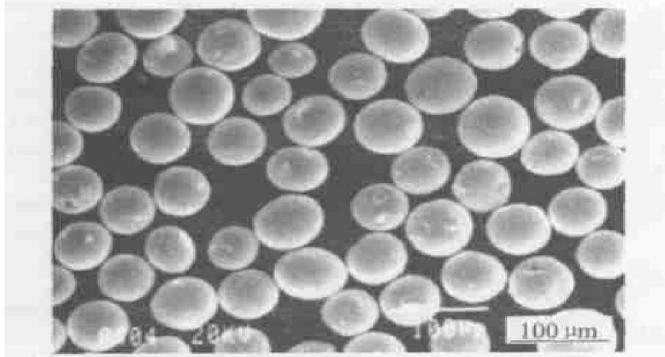


Fig. 4 Morphology of coarse tungsten powder used for plasma spraying

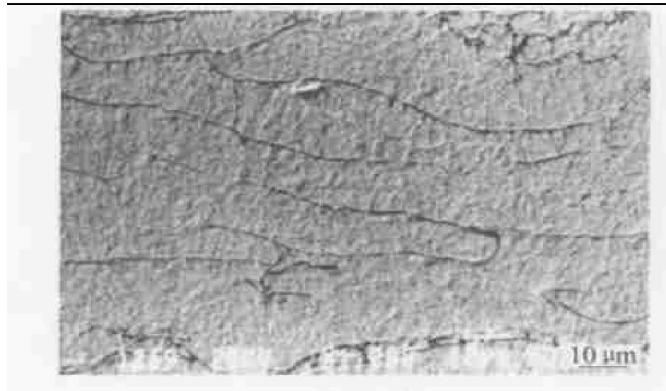


Fig. 5 Cross-sectional morphology of tungsten coating formed via induction plasma spraying of coarse powder

is very dense and almost no pore is found even though the density measurement is only 97%. X-ray diffraction analysis of the coating did not reveal any oxidation of tungsten particles and any involvement of impurities. The high density of the coating results from the two facts. One is that inside induction plasma the tungsten particles have much longer residence time due to the combination of low particle velocity and large spray distance. Thus, the induction plasma even generated with the power of only 40 kW can melt almost all the large tungsten particles. Another fact is that the impact force of a large particle is about 100 times higher because the mass of the large particle (45–75 μm) is 1 000 times higher than the mass of a small particle (5 μm) although the particle velocity is a few times lower.

It was surprised to find that few large solidified particles were completely surrounded by the molten tungsten particles without the formation of any pore nearby, as shown in Fig. 6. Such a phenomenon implies the sufficiently low viscosity of molten tungsten particles obtained and the high impact force resulted from the large mass of individual particles. It can be said that the coating has a high cohesion strength. The good toughness of the coating with interlocking structure was confirmed by the fact that no crack was found in the coating after the coating was knocked with a high impact force. Although there is large difference in the coefficients of thermal expansion between tungsten ($4.5 \times 10^{-6} \text{K}^{-1}$) and copper ($16.6 \times 10^{-6} \text{K}^{-1}$), the phenomena of coating cracking and spallation never happened during the induction plasma spraying of the coarse tungsten powder on the copper substrate.

3.2 Interface between tungsten coating and copper substrate

The interface between tungsten coating and copper substrate is important for application of the coating to Tokamak accelerator because a clean and com-

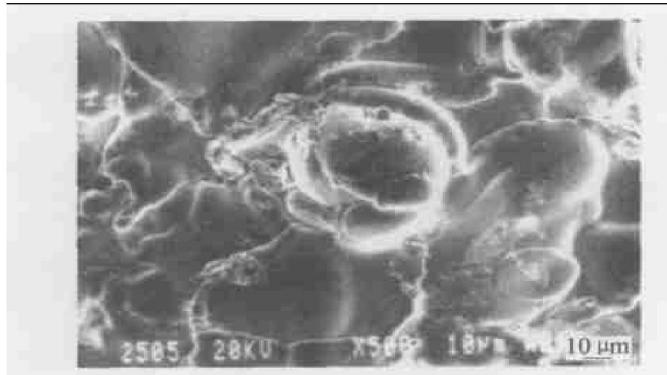


Fig. 6 Surface morphology of tungsten coating formed via induction plasma spraying of coarse powder

plete interface is required for heat release and electric conduction. Tungsten coating formed on the copper substrate by dc plasma spraying of the fine powder has been shown in Fig. 2. Although no obvious pore is found at the interface, the bonding between the substrate and the coating is not strong. When surface polishing was conducted, the tungsten coating was occasionally spalled from the substrate due to large residual stress in the coating and weak bonding at the interface.

Fig. 7 shows the morphology of the interface between the copper substrate and the tungsten coating formed via induction plasma spraying of the coarse powder. The large lamellae transformed from molten tungsten particles were completely adapted to the rough surface of the copper substrate. When surface polishing was performed, the coating was never spalled from the substrate. The coating was separated from the substrate only when a chisel was used. Energy dispersive analysis of X-ray (EDAX) revealed that tungsten residue was attached on the side of copper substrate and copper residue was found on the side of tungsten coating. Such a tight contact between the coating and the substrate at the interface not only provides great adhesion of the tungsten coating to the substrate but also gives rise to low electric resistance.

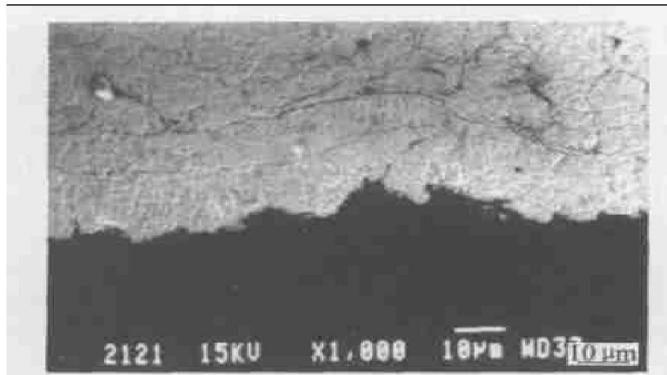


Fig. 7 Morphology of interface between copper substrate and tungsten coating formed via induction plasma spraying of coarse powder

4 CONCLUSIONS

Plasma spraying can be used to form tungsten coatings on the copper electrodes. Highly dense tungsten coatings can be obtained when dc plasma is used to spray fine powder or induction plasma is used to spray coarse powder. Tungsten coating formed by induction plasma spraying of coarse powder consists of interlocking laminar structure. With such an interlocking laminar structure, the tungsten coating has high cohesion and adhesion strength. Induction plasma spraying of coarse tungsten powder at low pressure is preferred to deposit tungsten coatings satisfying the requirement of Tokamak accelerator.

REFERENCES

- [1] Raman R, Thomas J C, Hwang D Q, et al. Design of the compact toroid fueler for center fuelling tokamak de varennes[J]. Fusion Technology, 1993, 24: 239 - 250.
- [2] Mallener W, Hohenauer W, Stoever D. Tungsten coatings for nuclear fusion devices[A]. 9th National Thermal Spray Conf[C]. USA: Cincinnati, 1996. 1 - 6.
- [3] Cavasin A, Brzezinski T, Grenier S, et al. W and B₄C coatings for nuclear fusion reactors[A]. International Thermal Spray Conf[C]. France: Nice, 1998. 957 - 962.
- [4] Varacalle D J, Lundberg L B, Miller B G, et al. Air plasma spray of tungsten coatings[A]. International Thermal Spray Conf[C]. Japan: Kobe, 1995. 377 - 382.
- [5] Valdes M U, Saint-Jacques R G, Moreau C. Thermal shock resistance of plasma sprayed tungsten coatings[A]. United Thermal Spray Conf[C]. USA: Indianapolis, 1997. 55 - 58.
- [6] Boulos M I. The inductively coupled radio frequency plasma[J]. Journal of High Temperature Materials and Processes, 1997, 1: 17 - 39.
- [7] Jiang X L, Tiwari R, Gitzhofer F, et al. On the induction plasma deposition of tungsten metal[J]. Journal of Thermal Spray Technology, 1993, 2: 265 - 270.
- [8] Jiang X L, Boulos M I. Particle melting, flattening, and stacking behaviors in the induction plasma deposition of tungsten[J]. Trans Nonferrous Met Soc China, 2001, 11: 811 - 816.
- [9] Fan X B, Gitzhofer F, Boulos M I. Statistical design of experiments for the spheroidization of powdered alumina by induction plasma processing[J]. Journal of Thermal Spray Technology, 1998, 7: 247 - 253.
- [10] Jiang X L, Boulos M I. Radio frequency induction plasma spraying of molybdenum[J]. Plasma Science & Technology, 2003, 5: 1895 - 1900.
- [11] Ishigaki T, Jurewicz J, Tanaka J, et al. Compositional modification of titanium carbide powders by induction plasma treatment[J]. Journal of Materials Science, 1995, 30: 883 - 890.
- [12] Jiang X L, Boulos M I. Induction plasma reactive deposition of tungsten carbide from tungsten metal

powder[J]. *Acta Metallurgica Sinica*, 2001, 14: 352 – 358.

[13] Jiang X L, Boulos M I. Effect of process parameters on the induction plasma reactive deposition of tungsten carbide from tungsten metal powder[J]. *Trans Nonferrous Met Soc China*, 2001, 11: 639 – 643.

[14] Jiang X L, Boulos M I. Synthesis of titanium carbide by induction plasma reactive spray[J]. *Trans Nonferrous Met Soc China*, 2004, 14(1) : 15 – 19.

[15] Gitzhofer F, Boulos M I, Heberlein J. Integrated fabrication processes for solid-oxide fuel cells using thermal plasma spray technology[J]. *MRS Bulletin*, 2000, 25: 38 – 42.

(Edited by LONG Huai-zhong)