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| Citation | Transactions of JWRI. 38(1) p.57-p.62 |
| Issue Date | 2009-09 |
| oaire:version | VoR |
| URL | https://doi.org/10.18910/10535 |
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# Microstructure and Hardness of Tungsten Coating for High Heat Resistant Material Produced by Means of Gas Tunnel Type Plasma Spraying†

**KOBAYASHI Akira * and PURIC Jagos ****

**Abstract**

*Tungsten (W) is the metal which has the highest melting point of 3422 °C, therefore when deposited as a coating it can protect the substrate surface from high heat flux. The W composite produced by coating methods will be a superior candidate for aerospace materials which require high heat resistance* (such as TBC)*. Thermal spraying method is one of potential candidates to produce W composites. In this study,* pure *W coatings were produced on stainless steel substrates by gas tunnel type plasma spraying at a short spraying distance (40-50mm). Regarding the microstructure, the W coating contained some pores. The Vickers micro-hardness of the W coating was measured as the mechanical property. The W coating had a hardness of Hv=260-320 along its cross section, which is a little lower than the hardness for bulk tungsten. The results of X ray Diffraction showed that the coating consists of pure tungsten, without oxidation.*

**KEY WORDS:** (Tungsten), (Sprayed coatings), (Fusion materials), (Gas tunnel type plasma spraying), (Microstructure) (Vickers Hardness), (XRD)

## 1. Introduction

Tungsten (W) is a special material, which has a high melting point, which is the highest among the metals ($T_m$ =3422°C). It is therefore desirable as a refractory material for high temperature applications in advanced industrial fields. Tungsten has the lowest sputtering yields among metals and is therefore being developed for fusion reactor applications such as the first wall [1], which requires minimum sputtering of foreign elements into the plasma.

For actual use as a tungsten target of an X-ray machine, using a coating would help to solve the problem of heavy weight of W. Although chemical vapor deposition (CVD) is being used to deposit tungsten onto a SiC substrate, CVD is a slow and relatively costly method, and the resultant coating may contain contaminated species.

Plasma spraying of tungsten is a more economical and convenient method for coating preparation. For example, it is helpful in enhancing the SiC- fiber heat-resistance that tungsten has almost the same expansion coefficient as SiC, but a higher melting point than SiC, whose melting point is 2500-2700°C. This material will be a good material for space propulsion. Therefore W composites will be superior candidates for high heat resistant materials for the aerospace industry as well as thermal fusion fields.

However, the high melting point of tungsten requires the plasma spray equipment to run at high power. High-power gas tunnel type plasma jet has been developed and its advanced properties were described in previous work [2-4]. It can be used to achieve efficient melting and coating deposition of tungsten, and it has a large possibility to widen the application fields [5,6]. Compared with conventional plasma spray equipment, the high-power gas tunnel type plasma spraying [7] can produce coatings with 20-30% higher hardness and density. Several ceramic coatings deposited using the gas tunnel type plasma spraying have been investigated and reported previously [8-11]. For example, the Vickers micro-hardness of a zirconia coatings was about *Hv*=1200 [12] at a power of *P*=33 kW and spraying distance of *L*=30 mm.

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Transactions of JWRI is published by Joining and Welding Research Institute, Osaka University, Ibaraki, Osaka 567-0047, Japan

Zirconia coatings produced at short spraying distances (*L*), also have a surface layer with higher hardness than the inner layers, which indicates the graded functionality of the coatings [13-14]. The mechanical properties and the bonding strength between the zirconia coatings and substrates have also shown that use of the gas tunnel type plasma spraying results in superior coatings [15, 16]

In this study, tungsten sprayed coatings were formed on the stainless steel substrates using the gas tunnel type plasma spraying at a short spraying distance, in order to develop a high performance material for thermal fusion and aero space components. The Vickers micro-hardness of the tungsten coatings was measured by a micro hardness tester. Also the microstructure of the coatings was examined by an Optical Microscope (OM) and a Scanning Electron Microscope (SEM). The crystalline structure of the coatings was investigated by X-ray Diffraction (XRD). Variations in those properties were also examined with different coating conditions.

## 2. Experimental Setup and Procedure

### 2.1 Preparation of W coatings

The gas tunnel type plasma spray torch developed by the author [7-8] is shown schematically in **Fig. 1**. The experimental methods for production of high hardness ceramic coatings by means of the gas tunnel type plasma spraying have been described in the previous publications [9-11]. For the current studies, a gas divertor nozzle diameter of 20mm was chosen. The overall experimental conditions for the plasma spraying of tungsten are shown in **Table 1**.

The input power to the plasma torch was about *P*=20 kW, and was supplied by the power supply PS-2. The power input to the pilot plasma jet was turned off after starting the gas tunnel type plasma jet. A short spraying distance of *L*= 40-50 mm was chosen for all tungsten plasma spraying deposition processes. The SUS304 stainless steel (3x50x50mm) substrate used was sand-blasted before spraying deposition. Argon was used as the working gas, and its flow rate was *Q*=170, 180 l/min. The W powder was fed to the plasma in an axial direction from the plasma gun cathode, the feed rate of tungsten was *w*=16, 24 g/min and the gas flow rate of carrier gas was 10 l/min. The substrate was traversed at 12 times or 32 times during the spraying.

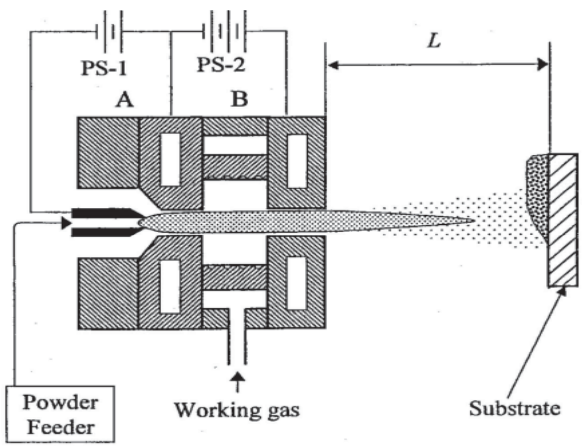


**Fig.1** Gas tunnel type plasma spraying used in this study (L=spraying distance).

**Table 1** Spraying Conditions.

| | |
|---|---|
| Arc current | 350A, 400 A |
| Voltage | 50V |
| Spraying distance | 40 -50 mm |
| Working gas Ar flow rate | 170, 180 l/min |
| Feed gas flow rate | 10 l/min |
| Powder feed rate | 16-24 g/min |
| Traverse number | 12, 32times |
| Spraying time | 24, 48s |

**Table 2** The chemical composition and particle size of the tungsten powder used.

| | |
|---|---|
| Material: | Tungsten (W) |
| Melting point: | $T_m$ = 3422°C |
| Purity: | 99.9% |
| Particle size: | 12 μm (average) |

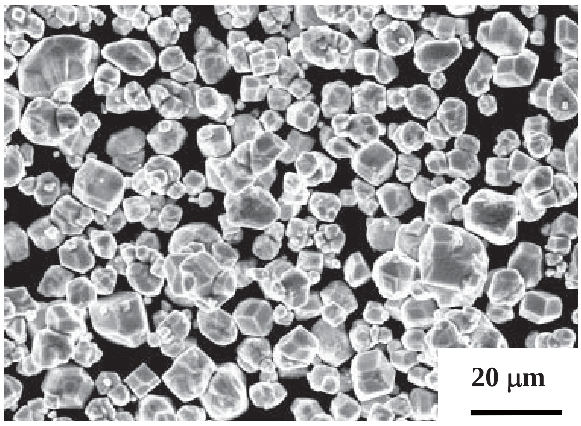


**Fig. 2** SEM micrographs of W powder (average size: 12 μm).

The chemical composition and particle size of the tungsten powder used are also given in **Table 2**. The tungsten powder was 99.9% in purity and the average particle size was 12 μm.. **Figure 2** shows the microphotographs of tungsten powder by SEM. The profile of the powder was not spherical but of angular type. The result of XRD measurement for the powder showed that the powder was pure tungsten.

### 2.2 Characterization of the W coatings

The cross section of the tungsten coatings was observed by optical microscope (OM) at magnifications of 200 or 400 times. The average thickness of the sprayed coatings was decided by the cross section. Also the porosity on the cross section of the metal glass coating was measured by the Image Processing method by using the results of optical micrographs of the coating cross section. The surface morphology of the W powder and the W coating cross-section was also examined by a scanning electron microscope (SEM) to observe clearly the structure.

X-ray diffraction (XRD) was conducted for measuring the crystal structure of the W powder and W coatings utilizing a Cu target (CuKα radiation source) and tube voltage of 30 kV and current of 14 mA.

The Vickers micro-hardness of the sprayed tungsten coating was measured at those cross sectional regions in which no pores existed. The hardness test loading weight was 100 g and the loading time was 25 s. The Vickers micro-hardness was calculated as a mean value of 10 measurements.

## 3. Results and Discussion

### 3.1 Formation of tungsten coating

The surface photograph of the tungsten coating sprayed at *L*=40mm with *P*=25 kW, taken by SEM, is shown in **Fig.3** (powder feed rate is16g/min, spraying time is 24s). This photograph shows that tungsten powder was sufficiently molten. The size of each splat is more than 20 μm in this case, and there were nano-size particles at some points on the coating surface. This means that there are some possibilities for controllability of spraying condition in order to make nano-size surfaces of W coatings in the future. Plasma sprayed pure tungsten coatings are black in color. But if yellow appears, this indicates the presence of tungsten oxides. The color of this coating sample was grey, which means that no oxidation existed in the coating.

**Figure 4** shows the cross sectional image of the same W coating which was taken by an optical microscope. The spraying time for this thin coating (~40μm) was 24s. The number of pores is substantially lower than that of zirconia coatings deposited under similar conditions, and the adhesion between W

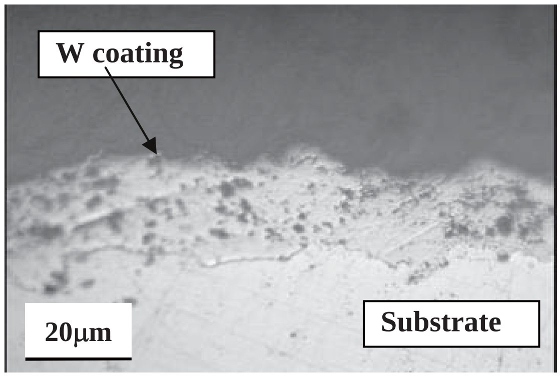


**Fig.4** Cross-sectional optical microscope image of W coating sprayed at *L*=40mm with *P* = 25 kW.

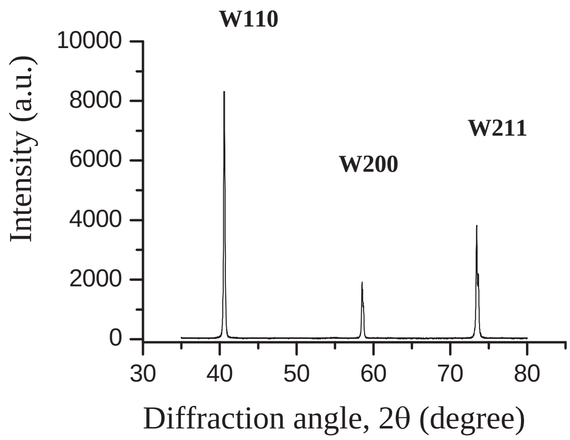


**Fig.5** XRD pattern on the surface of the W coating sprayed at *L*=40mm with *P*=25 kW.

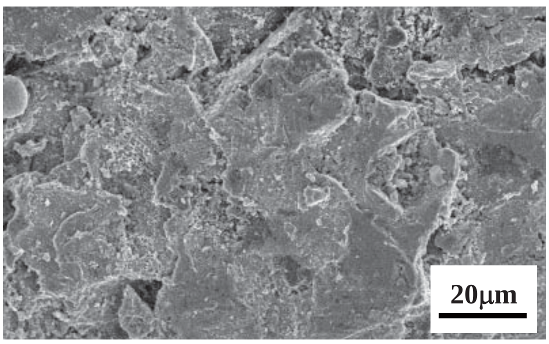


**Fig.3** Surface photograph of the coating sprayed at *L*=40mm with *P*=25 kW, taken by SEM.

coating and the substrate seems to be fine. This shows that the particles were deposited separately and were condensed together during the initial stage of deposition. It is therefore believed that sufficient plasma torch heating occurred during the thinner layer deposition for re-melting to occur.

**Figure 5** shows the XRD pattern of the surface of the W coating which is shown in Fig.4. There exist only tungsten peaks in this pattern. No tungsten oxide was observed. The absence of tungsten oxide peaks shows that only minimal oxidation occurred during the deposition processes.

**3.2 Effect of spraying distance on the W coating property**

*3.2.1 Microstructure of tungsten coating*

In the case of different spraying distances of 45, 50 mm at fixed plasma current of 350 A, thick (more than 100 μm) W coatings were obtained as shown in **Fig.6**. **Fig. 6 (a)** shows the SEM photograph of cross section of the W coating spraying at *L*=45mm, and **Fig. 6 (b)** is W coating spraying at *L*=50mm.The powder feed rate was in both cases 24 g/min. The spraying time (48 s) for these thick W coating was longer than that for the thinner coating in Fig.3.

In the case of shorter spraying distance of *L*=45mm, a thicker (~120 μm) coating was obtained as shown in Fig.6 (a). On the other hand, as the spraying distance was larger (*L*=50mm), the coating thickness became a little thinner to 80-100μm and some crack was observed somewhere.

It shows a more uniform highly dense or re-melted structure. For both the coatings, the number of pores is substantially lower than that of zirconia coatings deposited under comparable conditions. The bonding strength of the coating seemed to be good.

*3.2.2 Crystal structure of tungsten coating*

XRD measurement of the W coating produced at 350A is shown in **Fig.7**, which reveals several strong tungsten peaks and indicates only the presence of pure tungsten phase. These XRD pattern obtained for the tungsten coatings at different spraying distance of 45, 50mm, contained only the metallic tungsten phase.

But there are some small impurity peaks near the Diffraction angle of $2\theta$=20 − 40 degree in this condition, which may be some problem for the quality of W coatings.

*3.2.3 Vickers hardness of tungsten coating*

The Vickers micro-hardness of the W coatings produced at I=350A was measured at different

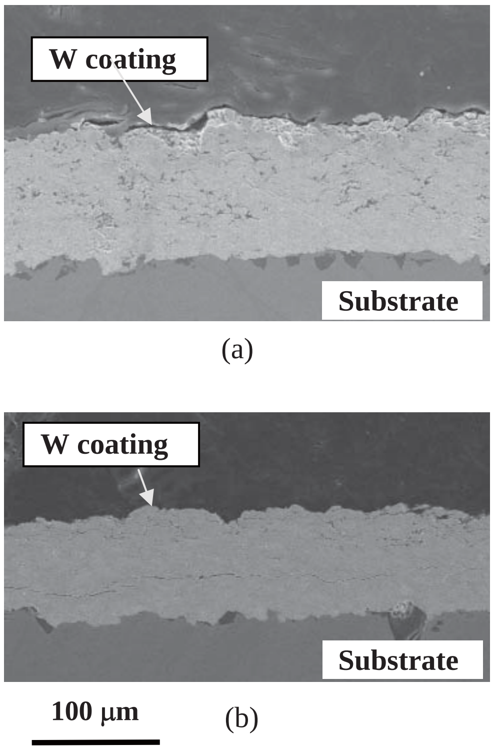


**Fig.6** SEM micrographs of the cross-section of W coating sprayed at plasma current of 350 A during the spray time of 48 sec, in the case of different spraying distance: (a) 45 mm, (b) 50 mm.

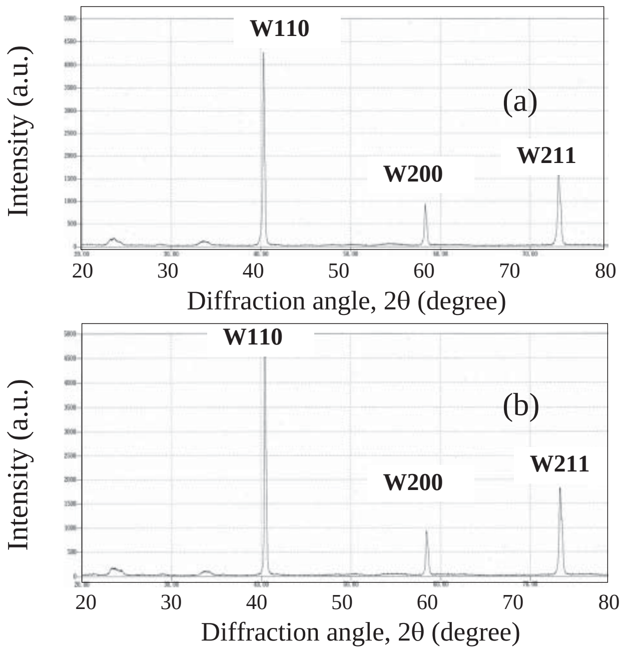


**Fig.7** XRD patterns on the surface of the W coatings sprayed at different distances for plasma current 350 A: (a) *L*=45mm and (b) *L*=50mm.

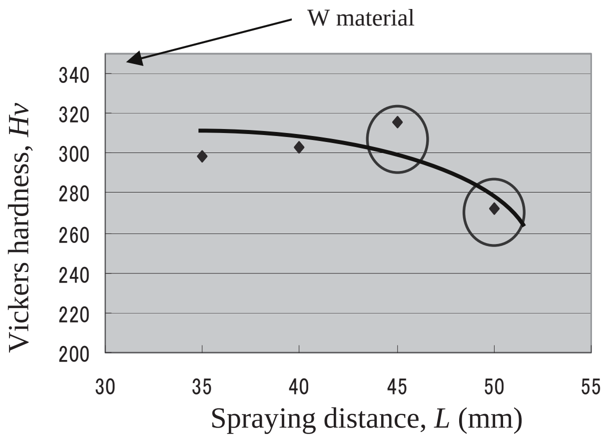


**Fig.8** Dependence of Vickers hardness of W coatings on the spraying distance for plasma current of 350 A.

spraying distance. The result for the effect of the spraying distance on the Vickers micro-hardness of W coatings was shown in **Fig.8.**

The Vickers micro-hardness of coatings were around *Hv*=260-320, and decreased with increase in the spraying distance. This corresponds to the dependence of coating thickness on the spraying distance. (120μm for 45mm, 100μm for 50mm) The thicker *Hv* = 300-320, and thinner *Hv* = 260-280 respectively, which represents the average value for plasma sprayed tungsten coating.

However, the tungsten coating Vickers micro-hardness was lower than that of pure bulk tungsten, which is about *Hv* = 350, probably because of the pores in the coatings.

### 3.3 Discussion about W coatings

The difference of hardness is in coincidence with the variation of the coating structure, namely the density and porosity of the coating. Also, the influence of thickness of the W coating on the Vickers micro-hardness was related to the coating structure. Inside the thick (~100μm) W coating, it was re-melted and a dense tungsten layer was formed. It is the coating heat transfer features, changing gradually along with the undergoing deposition, which result in graded changes in the morphology and density. Here, the lower Vickers micro-hardness than that of pure W, which is about *Hv* = 350, will probably be derived because of the pores in the coatings.

Generally, to avoid oxidation, tungsten or any other refractory metals requires spraying under controlled atmospheric conditions, such as an Argon back-filled chamber or use of vacuum plasma spray chambers. It is a significant finding that the high power plasma torch used here must have supplied sufficient argon gas flow to keep the newly deposited tungsten under a shroud of inert gas, thus allowing it to cool quickly enough to avoid oxidation. A few small oxide peaks shows that only minimal oxidation occurred during this deposition processes.

Ceramic coatings such as zirconia coatings are used for high temperature protection of metallic structures because of their high temperature resistance. Zirconia coating has been used as thermal barrier coating (TBC) of the hot sections of gas turbine engines and the high temperature parts of detonation furnaces. In order to enhance the quality of the TBC, plasma spraying has been contributing to combine the high heat capability of W and the low thermal conductivity of $ZrO_2$ even for developing more heat resistant TBCs.

## 4. Conclusions

The pure tungsten (W) coatings were produced on stainless steel substrates using the gas tunnel type plasma spraying. The following results were obtained.

(1) Thick (~120 μm) W coatings with uniform and dense structure could be coated onto stainless steel substrates at short spraying distances (40-50mm), when the plasma torch was operating at *I* =350A.
(2) Regarding the microstructure of the W coatings, a small number of pores were detected by SEM. The thicker the deposition, the less porosity, which might be due to substantial re-melting during the deposition.
(3) The W coating had a Vickers micro-hardness of *Hv*=260-320 along its cross section, which is a little lower than the hardness for bulk tungsten of *Hv* = 350. The hardness of the thicker W coating decreased from *Hv*=300 to 260 with increase in the spraying distance.
(4) The results of XRD method shows that the W coating consists of high purity metal tungsten, with very low oxidation. This plasma torch used must have supplied sufficient argon gas flow to keep the newly deposited W under a shroud of inert gas, thus to prevent it from oxidation.

**Acknowledgements**

The author would like to thank Mr. Makoto Kita for his help for the experiments and other coworkers for their valuable and helpful discussions.

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