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Formation of tungsten coatings by gas tunnel type plasma spraying

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Abstract

Tungsten is a material that has the highest melting point of 3422 °C among metals. Therefore, when deposited as a coating, it can protect the substrate surface from high heat flux. In this study, tungsten (W) sprayed coatings were formed on stainless steel substrates by gas tunnel type plasma spraying at a short spraying distance. The Vickers micro-hardness of the W coating was measured as a guide to its mechanical properties. The tungsten coating has a hardness of $Hv = 260-300$ along its cross section, which is a little lower than the hardness usually quoted for pure tungsten. Regarding the microstructure, the coating contains some pores. The results of X-ray diffraction and energy dispersive X-ray spectroscopy show that the coating consists of pure tungsten.

Keywords: Tungsten coatings; Gas tunnel type plasma spraying; Hardness test; XRD

1. Introduction

Tungsten is a special material, which has a high melting point, whose melting temperature is the highest among the metals ($T_m = 3422$ °C). It is therefore desirable for high temperature applications. Furthermore, 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.

As one example for actual use, tungsten is currently being used as the primary target material for generating X-rays using electron beams. Tungsten targets need to be rotated at high speed (~10,000 rpm) to achieve adequate cooling. The high weight of tungsten is one problem for the mechanics of such a high-speed rotating target. Using a coating would help solve the problem. Active research is ongoing to replace the W-target with a low weight silicon carbide (SiC) substrate coated with tungsten. Although chemical vapor deposition (CVD) is being used to deposit tungsten onto the SiC substrate, CVD is a slow and relatively costly and the resultant coating may contain contaminated species. Plasma spraying of tungsten is a more economical and convenient method for coating preparation. Another example application is the tungsten-plasma spraying coated SiC-fiber (foam) for high heat-resistance. It is helpful to enhance the SiC-fiber heat-resistance that tungsten has almost the same expansion coefficient as SiC, but higher melting point than SiC, whose melting point is 2500–2700 °C.

However, the high melting point of tungsten requires the plasma spray equipment to run at high power. High-power gas tunnel type plasma spraying has been developed [2] and is used to achieve efficient melting and coating deposition of tungsten. Compared with conventional plasma spray equipment, the high-power gas tunnel type plasma spraying can produce coatings with 20–30% higher hardness and density. By the way, several ceramic coatings deposited using the gas tunnel type plasma spraying have been investigated and reported previously [3–6]. For example, zirconia coatings produced at short spraying distances (L) have a surface layer with higher hardness than the inner layers, which indicates the graded functionality of the coatings [8,9]. The Vickers micro-

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hardness of the zirconia coatings was about $Hv=1200$ [7] at power of $P=33$ kW and spraying distance of $L=30$ mm. 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 [10].

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 TBC of the hot sections of gas turbine engines and the high temperature parts of detonation furnace. In order to enhance the quality of the TBC, plasma spraying has been contributed to combine the high heat capability of tungsten and the low thermal conductivity of ZrO_2 even for developing more heat resistant TBCs.

In this study, tungsten sprayed coatings were formed on stainless steel substrates using the gas tunnel type plasma spraying at a short spraying distance of 40 mm, in order to develop a high performance. The Vickers micro-hardness of the tungsten coatings was measured by the micro-hardness tester. Also, the microstructure of the coatings was viewed by an optical microscope and a scanning electron microscope (SEM). The crystalline structure of the coatings was investigated by X-ray diffraction (XRD) and the composition of the coatings was analyzed by energy dispersive X-ray spectroscopy (EDX). Variations of tungsten coatings in those properties were also examined with different coating thickness.

2. Experimental procedure

The used gas tunnel type plasma spray torch which has been developed at Osaka University [2,3] is shown schematically in Fig. 1. The experimental methods for production of the high hardness ceramic coatings by means of the gas tunnel type plasma spraying have been described in the previous publications [4–6]. For the current studies, a gas divertor nozzle diameter of 20 mm was chosen.

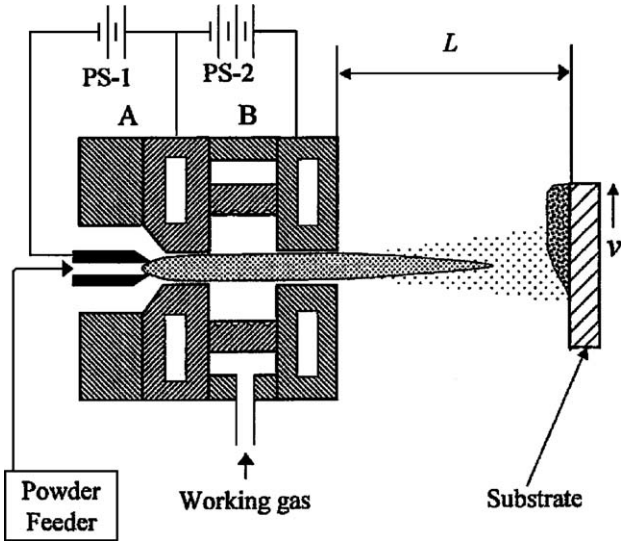

Fig. 1. Schematic of the gas tunnel type plasma spraying torch used for tungsten coating preparation. L: spraying distance.

Table 1
Overall experimental condition

Power input, P (kW)	13.5
Spraying time, t (s)	8–15
Working gas flow rate, Q (l/min)	170
Powder feed gas, Q_{feed} (l/min)	10
Spraying distance, L (mm)	40
Powder feed rate: w (g/min)	~16
Gas divertor nozzle diameter, d (mm)	20

The overall experimental conditions for the plasma spraying of tungsten are shown in Table 1. The input power to the plasma torch was about $P=13.5$ kW. The power input to the pilot plasma jet was supplied by the power supply PS-1 and was turned off after starting of the gas tunnel type plasma jet. A short spraying distance of $L=40$ mm was chosen for all tungsten plasma spraying deposition processes. Argon was used as the working gas and its flow rate was $Q=170$ l/min. The powder feed rate of tungsten was about $w=16$ g/min and the gas flow rate of carrier gas was 10 l/min. The substrate was not traversed during the spraying.

The chemical composition and particle size distribution of the used tungsten powder are also given in Table 2. The tungsten powder was 99.9% in purity and the average particle size was 12 μm. The used AISI304 stainless steel ($3\times50\times50$ mm^3) substrate was sand-blasted before spraying deposition.

The Vickers micro-hardness of the sprayed tungsten coating was measured at those cross-sectional regions in which no pore is contained. 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. But the Vickers micro-hardness distribution in the cross section of the tungsten coatings was obtained by measurement at 10 sites chosen with equal gap distance along the thickness direction.

The cross section of the tungsten coatings was observed for surface morphology at magnifications of 200 or 400 times with an optical microscope. The X-ray diffraction (XRD) with a $\theta-2\theta$ geometry conducted for the crystal

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

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

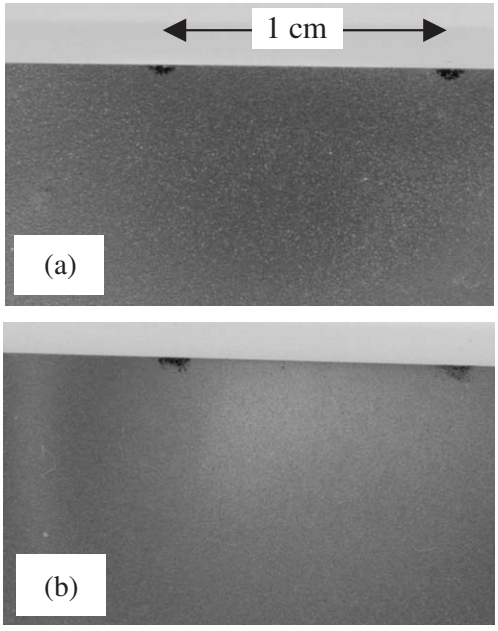

Fig. 2. Surface photographs of the coatings sprayed at $L=40$ mm with $P=13.5$ kW. (a) Thicker coating and (b) thinner coating.

structure of the coatings utilizes a Co target and tube voltage of 30 kV and current of 14 mA. The energy dispersive X-ray spectroscopy (EDX) used for composition analysis of the coatings is one affiliated to the used SEM.

3. Results and discussion

Fig. 2 shows the surface photographs of (a) a tungsten thicker (~180 μm) coating and (b) a thinner tungsten (~70 μm) coating. The photographs were taken from the central area of the deposition area, and the cutting line for coating cross-section observation is shown at their upper part. The color of the thicker sample (a) was grey coating. On the other hand, the sample (b) has a light and bright appearance. Plasma sprayed pure tungsten coatings are black in color. But generally yellow appears, which indicates the presence of tungsten oxides.

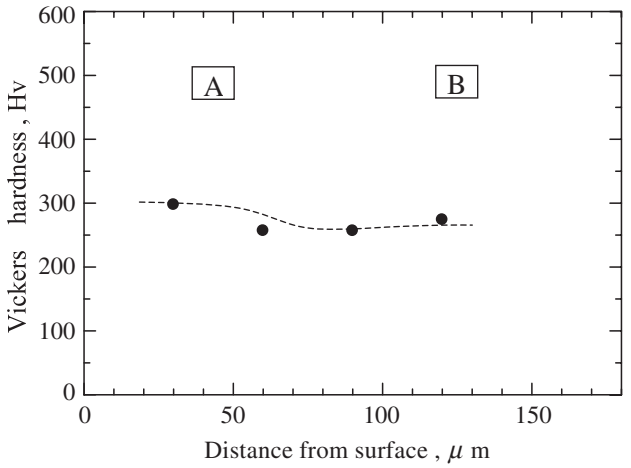

Fig. 4. Distribution of Vickers micro-hardness along the cross section of the thicker coating. (A) Surface region and (B) bond interface.

The cross-section images of tungsten coatings were taken by an optical microscope. Fig. 3(a) and (b) show the cross-sectional images of the same tungsten coating samples as in Fig. 2, but the microscope spans only a scope of 1 cm in the center of deposition area.

Inside the thicker (~180 μm) coating, the surface side [region A] was re-melted and dense tungsten layer was formed. In the meanwhile, the cross section near the substrate [region B] shows that the particles were deposited separately and were condensed together during

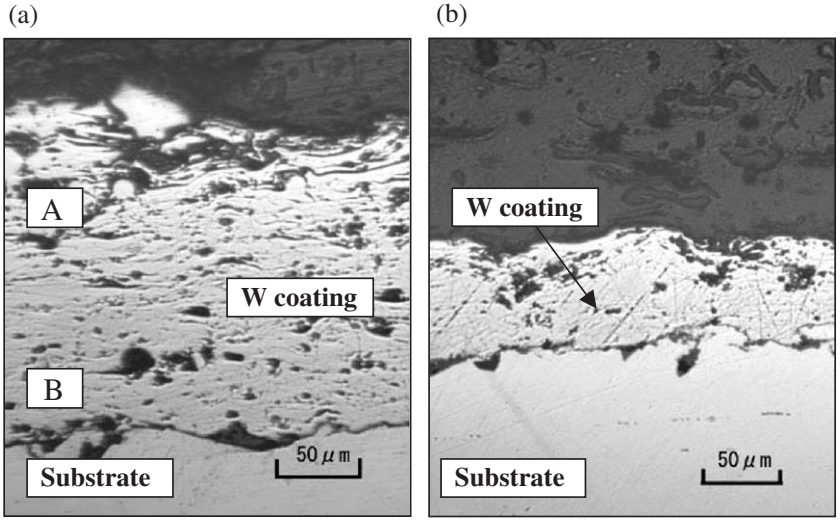

Fig. 3. Cross-sectional optical microscope images of coatings sprayed at $L=40$ mm with $P=13.5$ kW. (a) Thicker coating and (b) thinner coating. (A) Surface region and (B) bond interface.

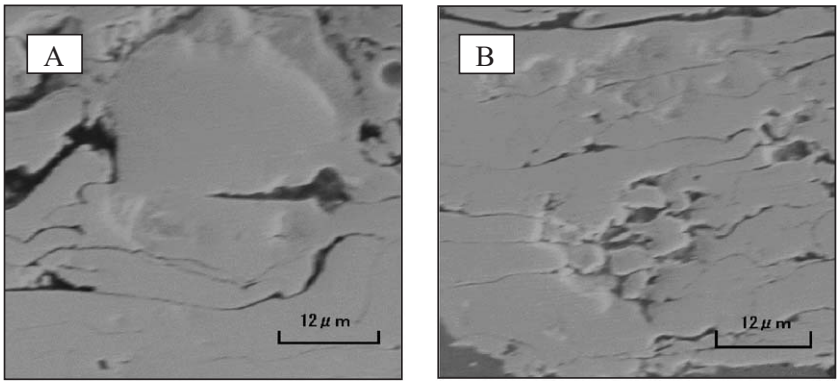

Fig. 5. Cross-sectional optical microscope images from 2 regions of the thicker coating. (A) Surface region and (B) bond interface.

the initial stage of deposition. It is the coating heat transfer features changing gradually along with the undergoing deposition, which results in graded changes in the morphology and density.

However, the thinner (~70 μm) coating in Fig. 3(b) shows a more uniform highly dense or re-melted structure. The spraying time (~15 s) for the thin layer was longer than that for the thicker coating. It is therefore believed that sufficient plasma torch heating occurred during the thinner layer deposition for re-melting to occur. For the both coatings, the number of pores is substantially lower than that of zirconia coatings deposited under comparable conditions.

The Vickers micro-hardness cross-sectional distribution was measured from the coating surface to the substrate-coating interface. The result for the thicker coating was shown in Fig. 4. The hardness distribution was gradually decreased from the surface to the interface. Near the surface [region A], the Vickers micro-hardness was around $Hv=300$, while near the substrate [region B], it dropped to about $Hv=270$. It is hinted that the distribution of hardness is in coincidence with the variation of the coating structure. Fig. 5(A) and (B) show the SEM photographs of the cross section of both [region A] and [region B]. The structure of [region A] was much denser than that of [region B], in which the powders were deposited separately.

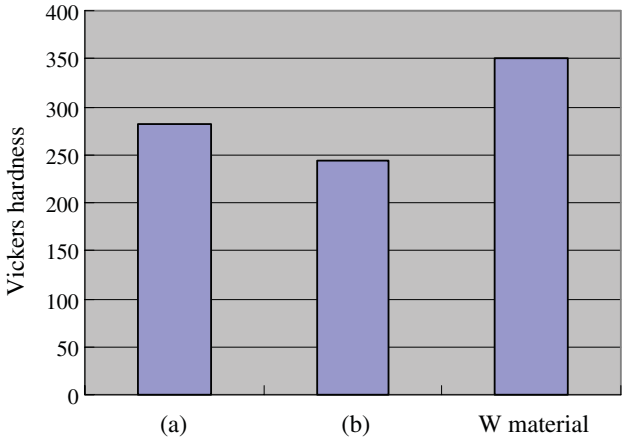

Fig. 6. Comparison of Vickers micro-hardness at different coating thickness. (a) Thicker coating and (b) thinner coating.

The thicker coating (a) and thinner coating (b) were compared in Fig. 3 on Vickers micro-hardness along cross section, which was measured for 10 sites scattered over the whole cross section of each coating. An average value along each cross section was used as the overall Vickers micro-hardness and the results were shown in Fig. 6. The Vickers micro-hardness of the thicker and thinner coatings were $Hv=280$ and $Hv=250$, respectively, which means the average value for plasma sprayed tungsten coating was comparable to each other. However, the tungsten coating Vickers micro-hardness was lower than that of pure tungsten, which is about $Hv=350$, probably because of the pores in the coatings.

XRD measurements of the coatings are shown in Fig. 7, which reveals several strong tungsten peaks and indicates the presence of pure tungsten phase. These XRD spectra obtained for the tungsten coatings contained only metallic tungsten phase. No tungsten oxide was observed. The absence of tungsten oxide peaks shows that only minimal oxidation occurred during the deposition processes.

The composition in the W coating structure analyzed by EDX on the cross sections of the tungsten coatings is shown in Fig. 8. There exist only tungsten metal peaks in the spectra and no oxygen was recognized, which shows again that tungsten oxide was not formed sufficiently.

Generally, to avoid oxidation, tungsten or any other refractory metals require 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

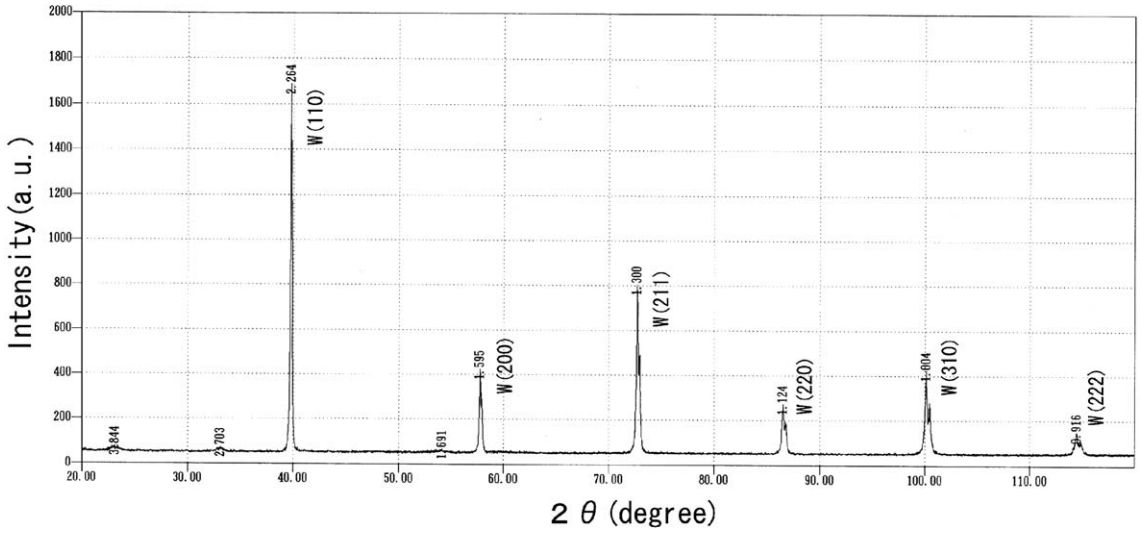

Fig. 7. X-ray diffraction spectrum of the tungsten coating surface.

inert gas, thus to allow it to cool quickly enough to avoid oxidation.

4. Conclusion

Pure tungsten coatings were sprayed onto stainless steel substrates using the gas tunnel type plasma spraying method. The following results were obtained.

(1) Thinner (~70 μm) and thicker (~180 μm) tungsten coatings could be coated onto stainless substrates at short spraying distances of less than 40 mm, when plasma torch was powered with $P=14$ kW.

(2) The prepared tungsten coatings have a Vickers micro-hardness of $Hv=260-300$. The Vickers micro-hardness of the thicker coating decreased from $Hv=300$ to 260 from the surface to the interface, which was a little lower than the hardness of pure tungsten of $Hv=380$.

(3) Regarding the microstructure of the coatings, a small number of pores were detected by optical microscope and SEM. The thinner deposition, the less porosity, which might be due to substantial re-melting during the deposition.

(4) The results from XRD and EDX show that the coatings consist of pure metal tungsten. The high-power plasma torch used must have supplied sufficient argon gas flow to keep the newly deposited

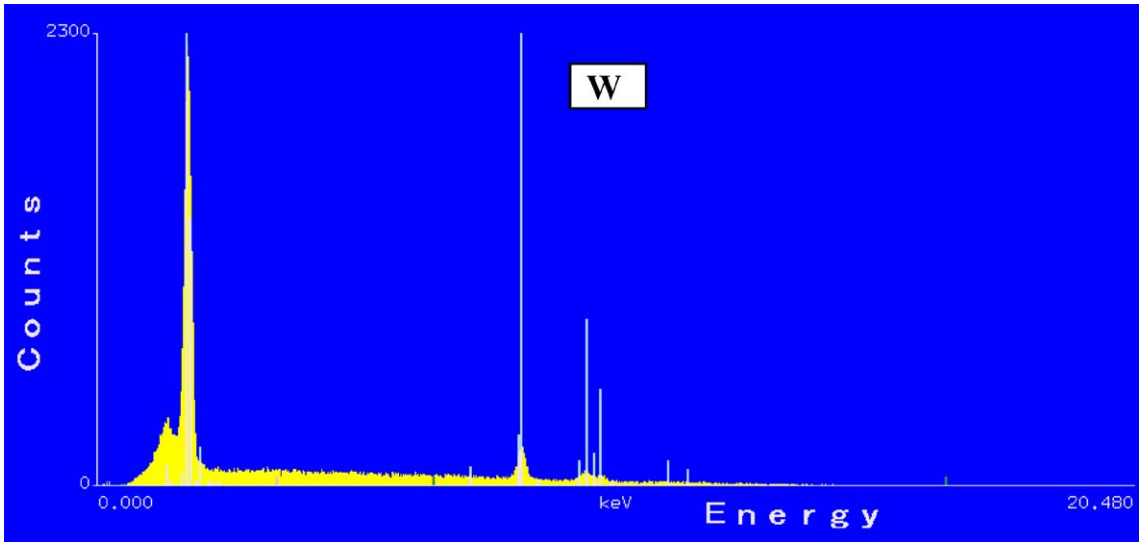

Fig. 8. EDX spectrum of the tungsten coatings.

tungsten under a shroud of inert gas, thus to prevent it from oxidation.

Acknowledgement

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