High-Rate Diamond Deposition by Combustion Flame Method Using Twin Acetylene/Oxygen Gas Welding Torch

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(Submitted June 8, 2009; in revised form August 28, 2009)

To develop a high-rate diamond deposition process using combustion flame method, diamond deposition equipment with twin acetylene/oxygen welding torch was manufactured, and diamond deposition by using this equipment was carried out. 304 Stainless steel plates and molybdenum plates were used as substrates. The diamond deposition was conducted under the following conditions: oxygen flow rate: 1.25 SLM, acetylene/oxygen flow ratio: 1.15, and diamond deposition temperature: around 1473 K. Consequently, diamonds could be deposited even on the stainless steel substrate, and diamond deposition rate was promoted by using twin torch equipment. Besides, the diamond/molybdenum hybrid coating using diamonds deposited by twin torch equipment have the same wear-resistant property as that using diamonds by the single torch equipment. From these results, this technique was thought to have high potential for high-rate diamond deposition in combustion flame method.

Keywords	APS	moly	bdenum	coa	atings,	chemical	vapor
-	depos	ition,	combust	ion	flame	method,	testing,
	wear						

1. Introduction

Recently, wear-resistant thermal spray coating has been widely used to elongate the lifetime of sliding parts of automobiles. Especially, molybdenum, with good wear resistance properties, has been widely used as an important material for many applications. However, since molybdenum has some problems, for example, lower wear resistance in comparison with practical ceramics and drastic decrease of wear resistance ability under a wet condition, thermal sprayed molybdenum coating needs to be improved to expand its utility. Although the infiltration of the liquid-phase PTFE (Ref 1) and the low viscosity glass into the coatings and the dispersion of the glass (Ref 2), ceramics, and/or diamond particles into them are available as the processes which improve the wear resistance by deposition molybdenum thermal sprayed coatings, more convenient methods were demanded because the above-mentioned processes have some problems such as longer infiltration time, low uniformity of the dispersion profile of the particles, and so on. Therefore, since diamond addition using atmospheric chemical vapor deposition (CVD) is thought to be effective because of uniform diamond deposition in open air by the process, combustion flame CVD diamond/ atmospheric thermal spray molybdenum hybrid coating has been conducted in our research group. Combustion flame method is a diamond deposition process using acetylene/oxygen welding torch (Ref 3). Since this process has some merits, for example, high deposition rate (approximately 40 µm/h), atmospheric process, and so on, it is hoped to be utilized as a low-cost diamond deposition process. However, the combustion flame method has some disadvantages such as thermal damage of the substrate during diamond deposition due to direct irradiation of the combustion flame and small diamond deposition area. As the protection process from the thermal damages of the substrate, techniques using substrate surface cooling (Ref 4) and insertion of the thermal stress buffer layer between substrate and top coating (Ref 5) were developed, and some effects could be confirmed. Besides, since it was proved that diamond could be deposited even on the 304 stainless steel substrate by combustion flame CVD in our recent work (Ref 6), the diamond/molybdenum hybrid coating could be fabricated by molybdenum coating after diamond deposition on the stainless steel substrate without taking account of peeling off and crack running of the

This article is an invited paper selected from presentations at the 3rd Asian Thermal Spray Conference (ATSC2008) and has been expanded from the original presentation. ATSC2008 was held at Nanyang Executive Centre, Singapore, November 6-7, 2008, and chaired by K.A. Khor.

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molybdenum coating during diamond deposition. Therefore, the diamond/molybdenum hybrid coating could be fabricated without using thermal stress buffer layerincluded substrate and taking account of crack running and peeling off of the molybdenum coating during diamond deposition. As for the solution for the small diamond deposition area, two methods, multiflame deposition (Ref 7) and traversing torch method (Ref 5) traversing substrate (or traversing welding torch) during operation, were proved to be available (Ref 8). However, these methods have some disadvantages. Although the multiflame deposition using some parallelly located single torches is thought to be excellent on high-rate diamond deposition, substrate is heavily damaged during diamond deposition due to large heat-affected zone. On the other hand, in the case of traversing substrate, if this method is applied, diamond deposition rate should be promoted to solve the newly generated problem that diamond deposition rate will decrease with increasing substrate traversing speed. Therefore, high radical density combustion flame generator is demanded for large-area high-rate diamond deposition.

In this study, to develop a high-rate diamond deposition process using combustion flame method by high radical density combustion flame creation, diamond deposition equipment with twin acetylene/oxygen welding torch was fabricated, and the diamond deposition using this equipment was carried out.

2. Experimental Procedure

Figure 1 shows the schematic diagram of the conventional (single torch type) combustion flame diamond deposition equipment. This equipment consists of acetylene/oxygen combustion flame welding torch, gas supply system including mass flow controller, and substrate holder. A sample was deposited on the stage of the substrate holder. Inlet pressure of C2H2 and O2 were 0.25 and 0.34 MPa, respectively. Mass flow of O2 was fixed at 1.25 SLM, while mass flow ratio of C_2H_2/O_2 was also fixed at 1.15. Deposition distance was fixed at 10 mm to make the acetylene feather of the combustion flame irradiate surface of the substrate. Deposition time was 20 min. As substrates, $12 \text{ mm} \times 10 \text{ mm} \times 1 \text{ mmt}$ Mo plate and $15 \text{ mm} \times 15 \text{ mm} \times 1 \text{ mmt} 304 \text{ stainless steel plate were}$ used. These substrates were polished by 400# waterresistant sand paper before diamond deposition. The deposition temperature during operation (T_d) was observed by a radiation thermometer. $T_{\rm d}$ was controlled by varying contact condition between the substrate and the cooling pipes located on the substrate surface. Therefore, the temperature was controlled by moving the positions of the cooling pipes located on the surface of the substrate without varying the conditions of combustion flame (such as quenching, deactivation of radicals, and so on).

Figure 2 shows twin torch-type combustion flame diamond deposition equipment. Both combustion flames



(a) Schematic diagram



(**b**) Photograph during operation

Fig. 1 Schematic diagram of the single welding torch combustion flame diamond deposition equipment

were generated on the same condition as the combustion flame in the case of the single torch-type equipment.

After diamond deposition, investigation of the microstructure and phase composition of the diamonddeposited substrate was carried out using optical microscope and x-ray diffraction. Table 1 shows the diamond deposition conditions. Substrate temperature was measured by infrared radiation thermometer. The thermometer measured the temperature of the point where the combustion flame contacts with the substrate.

For confirmation of the effects of diamond addition to the atmospheric plasma spray (APS) molybdenum coating on its wear-resistant property, wear-resistant test of the diamond/molybdenum hybrid coatings was conducted by using SUGA-ISO-3 wear testing equipment shown in Fig. 3, based on the condition shown in Table 2. Before the wear test, the diamond/molybdenum hybrid coatings were created by APS molybdenum coating deposition on the diamond-deposited molybdenum and 304 stainless steel substrates (Fig. 4). Table 3 shows conditions of the molybdenum APS coating. Thickness of the hybrid films was approximately 100 μ m.





(b)

Fig. 2 Schematic diagram and photograph during operation of the twin welding torch combustion flame diamond deposition equipment. (a) Schematic diagram and (b) photograph during operation

Table 1	Diamond	deposition	conditions

Working gas	C_2H_2/O_2
C_2H_2 flow rate	1.4 SLM
O_2 flow rate	1.25 SLM
Deposition distance	10 mm
Deposition time	5-20 min(a)5-15 min(b)
Deposition temperature	1473 K
Substrate	Molybdenum, 304 stainless steel

(a) In the case of the single welding torch equipment. (b) In the case of the twin welding torch equipment

3. Results and Discussion

3.1 Effects of Twin Welding Torch on Diamond Deposition Rate in the Case of Molybdenum Substrate

Figure 5 shows the optical micrographs of the surfaces of the substrates after 5 and 10 min combustion flame irradiation. As shown in this figure, in this case of single torch, it took over 5 min to create the diamond nuclei on the substrate and only small diamond particles (under 5 µm in diameter) could be deposited even after 10 min combustion flame irradiation. In the case of twin torch, nucleation occurred within 5 min and over 10 µm diameter diamond particles were deposited after 10 min deposition as shown in Fig. 6. From these results, twin torchtype equipment was proven to be effective for promotion of diamond deposition rate in combustion flame method. As the reason why the deposition rate was lower than twice of that in the case of the single torch, it was thought that cooling pipe of the substrate surface cooling system quenched and deactivated the radicals in the combustion flame. Figure 7 shows the appearance of the molybdenum substrate after 15 min diamond deposition using twin



Fig. 3 Schematic diagram of the wear testing equipment

Table 2Wear-resistant test conditions

Load	10 N
Stroke	30 mm
Sliding speed	2400 mm/min
Sliding time	1250 min
Opponent material	#2000 SiC sand paper





welding torch equipment. Although the size of the largest diamond particle (approximately 10 μ m) was almost the same as that in the case of 10 min, the particle density was dramatically increased in comparison with that in the case of 10 min. Compared to the case of the single torch, since it took 20 min to deposit diamonds with the same size and particle density, it was proved that diamond deposition rate was promoted by using twin welding torch equipment. Figure 8 shows x-ray diffraction pattern of the molybdenum substrate after 15 min diamond deposition using twin welding torch equipment. As shown in this figure, not only the peaks of diamond but also that of molybdenum carbide (Mo₂C) was observed. From this result and the fact that the interatomic distance of diamond is close to that of

Table 3Spray parameters for the deposition of the APSmolybdenum coating

Ar
40 SLM
50 V
600 A
80 mm
304 stainless steel

 Mo_2C compared to that of Mo, it was thought that the creation of Mo_2C enables the deposition of diamonds on molybdenum substrate.

3.2 Effects of Twin Welding Torch on Diamond Deposition Rate in the Case of Stainless Steel Substrate

Figure 9 shows the appearances and the surface optical micrograph of the 304 stainless steel substrate after 20 min diamond deposition using single welding torch equipment. Although 304 stainless steel had low melting point in comparison with molybdenum, it was proved that diamonds could be deposited on the stainless steel substrate without melt down of the substrate by proper layout of the cooling pipes located on the substrate surface. Figure 10 shows the appearances and the surface optical micrograph of the 304 stainless steel substrate after 15 min diamond deposition using twin welding torch equipment. Even in the case of twin welding torch equipment, though the heat flux from the combustion flame to the substrate was approximately twice higher than that in the case of the single welding torch equipment, meltdown did not occur during diamond deposition. Besides, in the case of the molybdenum substrate, it took 15 min to create diamonds



Fig. 5 Optical micrograph of the substrate surface after diamond deposition in the case of the single welding torch equipment. (a) After 5 min deposition and (b) after 10 min deposition



Fig. 6 Optical micrograph of the substrate surface after diamond deposition in the case of the twin welding torch equipment. (a) After 5 min deposition and (b) after 10 min deposition



Fig. 9 Appearance and surface optical micrograph of the diamond deposited sample in the case of the single welding torch equipment. (a) Appearance and (b) surface optical micrograph

torch equipment also in the case of stainless steel substrate. Figure 11 shows x-ray diffraction pattern of the 304 stainless steel substrate after 15 min diamond deposition using twin welding torch equipment. Also in this case, not only the peaks of diamond but also that of iron carbide (Fe₃C) was observed. According to the literature on diamond deposition on 304 stainless steel substrate by combustion flame method (Ref 9), the Fe₃C layer was concluded to play a roll of buffer layer for hetero-epitaxial diamond growth. In this case, the same phenomenon was thought to occur during diamond deposition.

3.3 Wear Resistance of the Diamond/Molybdenum Hybrid Coating

Figure 12 shows the results of the wear-resistant test of the APS molybdenum coating and diamond/molybdenum hybrid coatings deposited on molybdenum and 304 stainless steel substrates by the single welding torch equipment and the twin welding torch equipment. In our previous study (Ref 4-6), it was proven that wear-resistant property was promoted by addition of diamond particles. Also in this study, almost the same wear resistance, which was



Fig. 7 Appearance and surface optical micrograph of the Mo substrate after 15 min diamond deposition in the case of the twin welding torch equipment. (a) Appearance and (b) surface optical micrograph



Fig. 8 XRD pattern of the Mo substrate surface after 15 min diamond deposition in the case of the twin welding torch equipment

with the same size and particle density as those by 20 min deposition using the single welding torch equipment. From this result, it was thought to be effective for promotion of diamond deposition rate to use the twin welding



Fig. 10 Appearance and surface optical micrograph of the diamond-deposited sample in the case of the twin welding torch equipment. (a) Appearance and (b) surface optical micrograph



Fig. 11 XRD pattern of the 304 stainless substrate surface after 15 min diamond deposition in the case of the twin welding torch equipment

1.5 times higher than that of the APS molybdenum coating, was indicated in any hybrid coatings. From these results, as for the wear resistance, deterioration of the diamond quality due to high-rate deposition using twin welding torch equipment was not confirmed.



Fig. 12 Results of the wear resistant test. #1—APS molybdenum coating and #2-5—diamond/molybdenum coating: #2: molybdenum substrate, single torch; #3: molybdenum substrate, twin torch; #4: stainless steel substrate, single torch; and #5: stainless steel substrate, twin torch

4. Conclusions

To develop a high-rate diamond deposition process using high radical density combustion flame, diamond deposition equipment using twin acetylene/oxygen welding torch was manufactured, and diamond deposition by using this equipment was carried out. Consequently, following results were obtained.

- 1. In the case of the Mo substrate, by using twin welding torch equipment, diamond deposition rate was promoted.
- 2. Even in the case of the 304 stainless steel substrate, diamonds could be deposited without melt down of the substrate by proper layout of the cooling pipe located on the substrate. Besides, by using twin welding torch equipment, as the case of the Mo substrate, promotion of the diamond deposition rate was confirmed.
- 3. Diamond/Mo hybrid coatings has high wear resistance in comparison with APS molybdenum coating even in the case of the 304 stainless steel substrate and twin welding torch use. Therefore, twin welding torch equipment is thought to be useful in high-rate diamond deposition process instead of the conventional single torch-type equipment.

From these results, this technique is found to have high potential for high-rate diamond deposition process.

References

- B.R. Marple and J. Voyer, Improved Wear Performance by the Incorporation of Solid Lubricants During Thermal Spraying, *Proceedings of ITSC2000*, 2000, p 909-918
- D.T. Gawne, Z. Qiu, T. Zhang, Y. Bao, and K. Zhang, Abrasive Wear Resistance of Plasma-Sprayed Glass-Composite Coatings, *Proceedings of ITSC2000*, 2000, p 977-981
- Y. Hirose, S. Amanuma, and K. Komaki, The Synthesis of High-Quality Diamond in Combustion Flames, *J. Appl. Phys.*, 1990, 68, p 6401-6405

- Y. Ando, S. Tobe, and H. Tahara, Creation of Diamond/ Molybdenum Composite Coating in Open Air, *Mater. Sci. Forum*, 2007, **534-536**, p 1097-1100
- Y. Ando, S. Tobe, T. Saito, J. Sakurai, H. Tahara, and T. Yoshikawa, Enlargement of the Diamond Deposition Area in Combustion Flame Method by Traversing Substrate, *Thin Solid Films*, 2004, **457**(1), p 217-223
- Y. Ando, S. Tobe, and H. Tahara, Diamond Deposition on Mo with Thermal Stress Buffer Layer Coated Mild Steel Substrate by Combustion Flame CVD, *Vacuum*, 2008, 83, p 102-106
- Y. Tzeng, R. Phillips, C. Cutshaw, and T. Srivinyunon, Multiple Flame Deposition of Diamond Films, *Appl. Phys. Lett.*, 1991, 58(23), p 2645-2647
- Y. Ando, H. Suzuki, K. Kobayashi, T. Mantani, S. Tobe, H. Tahara, and A. Kobayashi, Study on Diamond Deposition on APS Mo/Fe Coating and Stainless Steel by Combustion Flame CVD, Front. Appl. Plasma Technol., 2008, 1, p 69-76
- S. Kumar and M. Malhotra, Growth of Polycrystalline Diamond Films on Stainless Steel Without External Barrier Layers Using Oxy-Acetylene Flame, *Diam. Relat. Mater.*, 1998, 7(7), p 1043-1047