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Influence of Surface Character Change of Substrate Due to Heating on Flattening Behavior of Thermal Sprayed Particles

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The authors have confirmed that in the thermal spraying of practical powder materials, the splat shape changes with increasing substrate temperature to a circular disk shape from a fringe shape with splashing at a critical substrate temperature, T_t. The increase of the substrate temperature may accompany a kind of essential change on the substrate surface, because the effect is maintained until the substrate is cooled down to room temperature. However, the nature of the substrate surface change due to the heating has not been clearly understood yet. In this study, AISI 304 stainless steel was used as a substrate material, and the substrate was heated in an air atmosphere or laser treated as a pretreatment. Substrate surface topography in nanometer scale was analyzed precisely by atomic force microscope (AFM). The relationship between surface topography in nanometer scale and splat morphology was discussed. Moreover, to evaluate the effect of chemical composition of the substrate surface, gold was coated onto the substrate surface by physical vapor deposition (PVD) after the heat treatment. The effect of adsorbate/condensate on the substrate surface on the flattening behavior of thermal sprayed particles was also verified.

Keywords adsorbate/condensate, flattening behavior, laser treatment, sprayed particles, substrate temperature, surface topography, thermal spraying

1. Introduction

As a flattening behavior of individual thermally sprayed particle on a substrate surface is recognized to be a fundamental phenomenon of the coating formation, a clarification of the flattening behavior of an individual particle is essential for control of the thermal spray process. Theoretical analysis typically represented by Madejski (Ref 1) has indicated that both in-flight temperature and velocity of the particle dominate the flattening behavior of the sprayed particles. However, the authors have pointed out that the particle/substrate interface oriented factors, such as substrate temperature or atmospheric pressure, can have a greater effect on the flattening (Ref 2-6). In particular, the authors' experimental results have revealed that the splat shape of most metallic and ceramic materials on a flat substrate surface change remarkably from a splash splat to a disk shape with an increase of the substrate temperature (Ref 2, 3).

The transition temperature, T_t, at which the splat shape of the particle changes to a splash splat from the disk shape was introduced by the author (Ref 3). Moreover, it was verified that the adhesion strength of the coating changed with the increasing substrate temperature, and its dependence on the substrate temperature corresponded quite well to that of the splat shape. Thus, investigation of the flattening behavior of thermal sprayed particle on the substrate surface is really meaningful from a practical viewpoint as well. Up to today, unified explanations of the transition behavior have been proposed, based on experimental, analytical, or numerical data both in actual thermal spray process and simulative freely fallen metal droplet experiment. Possible dominant factors are the initial solidification (Ref 7), wetting at particle/substrate interface (Ref 2, 3), several surface conditions (Ref 8-10), and so on.

The effect of substrate surface topography change due to heat treatment on the transition in flattening behavior of thermal sprayed particle was mainly investigated in the current study. Especially, mean roughness R_a and skewness Sk both in nanometer scale were selected as the evaluating parameters for the roughness character of the substrate surface. Laser irradiation treatment was also used to change the surface topography of the substrate and the influence of surface change on the flattening behavior of thermal sprayed particle was investigated. Furthermore, the effect of surface character in both physical (surface roughness and adsorbates/condensates) and chemical (chemical composition change, namely, surface oxidation) aspect on the flattening behavior of thermal sprayed particles was investigated.

2. Experimental Procedures

2.1 Experimental Setting

Commercially available pure copper (Cu) powder with 75 μm or less in diameter was used for the spraying. Mirror-

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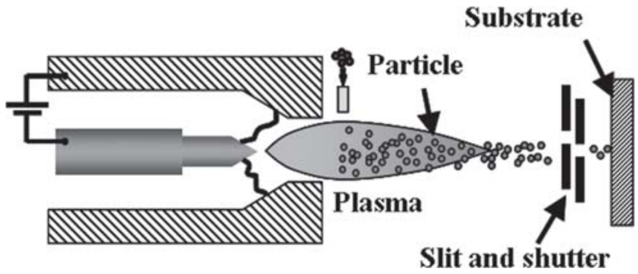

Fig. 1 Atmospheric dc plasma spray apparatus

polished AISI 304 stainless steel plates and pure gold plates with 10 by 10 by 5 mm were used as the substrate material. The plates were polished with 0.3 μm alumina (Al_2O_3). In addition to the heating by regular heating equipment (hot plate), laser irradiation treatment was introduced. Laser power density was varied from 25.83 to 90.4 W/mm^2. Traverse speed of the laser was 0.4 mm/s commonly for all experiments. Oxygen or argon was used as assist gas for laser treatment. A couple of substrates were laser treated simultaneously under the same condition, and one was used for the spraying and another was used for the measurement by atomic force microscope (AFM). Moreover, to investigate the effect of elapsed time on the flattening behavior of the sprayed particle, substrate materials after heat treatment were stored for appropriate periods in an atmosphere, namely, at room temperature not in high humidity, but in a dry condition. As the substrate material used in the study is stainless steel, it is estimated that no significant oxidation occurred by holding under this situation. Hence, surface roughness of the substrate after keeping in an atmosphere was not remeasured in the experiment. To eliminate the effect of chemical composition of the substrate surface on the flattening behavior of the particle, gold was coated onto the substrate by normal physical vapor deposition (PVD) method both as polished and after heated conditions. Heat treated and laser treated samples were allowed to cool to room temperature prior to plasma spraying.

Atmospheric direct-current (dc) plasma spray equipment (Sulzer Metco 9MB, Sulzer Metco, Winterthur, Switzerland) was used for the spraying. The schematic diagram of atmospheric plasma spray (APS) apparatus is shown in Fig. 1. The sprayed particles that passed through the shutter were collected onto the substrate as the splat. As the substrates were completely shielded from plasma both by slit and shutter, all substrates were at room temperature when plasma spraying was initiated commonly in this study. The spraying experiments were conducted immediately after the heat treatment to the substrate. Surface roughness of the substrate in nanometer scale was measured before the spraying experiments by AFM on an area of 1 μm^2.

2.2 Evaluation of Substrate Surface

In this study, two surface roughness parameters were used to evaluate the surface roughness character, that is, mean roughness, R_a, and skewness, Sk. The definitions of these two parameters are, respectively:

$$R_a = \frac{1}{l}\int_0^l |z-m|\,dz \quad \text{(Eq 1)}$$

$$Sk = \frac{1}{\sigma^3}\int_{-\infty}^{\infty} (z-m)^3 \phi(z)\,dz \quad \text{(Eq 2)}$$

where z is surface height, m is mean value of the surface height, and l is sampling length; where $\phi(z)$ is distributing function of the surface heights, and Sk is the parameter to evaluate distribution of the surface heights (Ref 11, 12). Nanometer scale surface topography of the substrate may affect friction coefficient between sprayed particle and substrate surface, flattening speed, and thermal conduction. Effect of substrate surface topography on the flattening of the sprayed particle may be evaluated by the measurement for these parameters given in the AFM analysis.

3. Results and Discussion

3.1 Relation between Substrate Roughness Parameter and Splat Morphology

Typical AFM observation results on both as-polished and once-heated substrate surface are shown in Fig. 2. As compared with as-polished substrate, projections were observed on once-heated substrate, as shown in Fig. 2(b). Mean surface roughness R_a value of once-heated substrate became bigger than that of the as-polished substrate. Correspondingly, the splat shape on the laser treated surface changed to the disk type also, as shown in Fig. 2(b). However, as the splash-type splat was observed on the substrate as polished, which was polished by 1 μm polishing media and had the mean surface roughness R_a of almost 3.0 nm, it became clear that the evaluation with R_a was not enough. Thus, instead of R_a, skewness, Sk, was introduced. From the measurement result, it became clear the Sk value of once-heated substrate became bigger and changed from negative to positive, as shown in Fig. 2(b), and the splat morphology changed to disk shape from splash shape, correspondingly.

To confirm the effect of surface topography change on the flattening behavior artificially, laser irradiation treatment onto the substrate surface was carried out. Sprayed particles were collected on the laser treated substrates, and the effect of surface character change by laser treatment on the flattening behavior of thermal sprayed particles was investigated. The result of AFM analysis on the laser treated substrate surface and splat morphology are shown in Fig. 3. Figure 3(b) shows the typical surface topography image for laser treated substrate with a power density of 90.4 W/mm^2 after polishing with 0.3 μm Al_2O_3. It can be seen from the figure the R_a value of laser treated substrate became larger than that of the as-polished substrate. Moreover, the Sk value changed from negative to positive as expected. Correspondingly, splat morphology on the laser treated substrate was disk type. Change of the fraction of disk splat due to laser treatment was measured more systematically. To perform this, change of fraction of disk splat with increasing laser power was investigated. Figure 4 shows the relationship between laser power density and fraction of disk splat. From the figure, it became clear that fraction of disk splat almost linearly increased with increasing the laser power density. To confirm the effect of substrate laser treatment on the roughness change, these substrate surfaces were analyzed by AFM. Figure 5 shows the relationship between laser-power density and surface roughness parameters, both of mean surface roughness, R_a, and skewness, Sk.

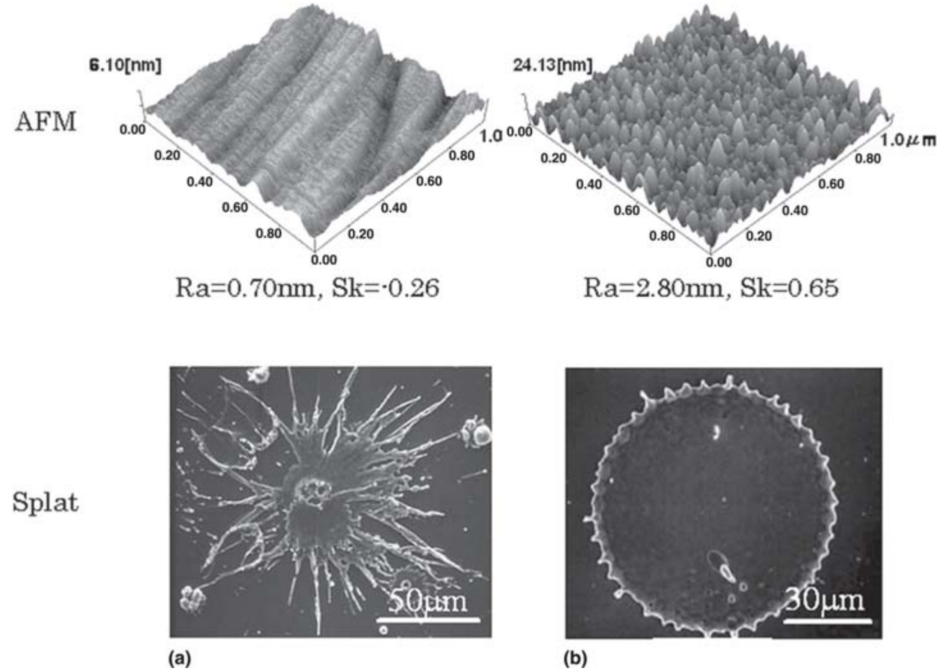

Fig. 2 AFM profiles and splat morphologies on AISI 304 substrate: (a) as polished; (b) once heated to 673 K

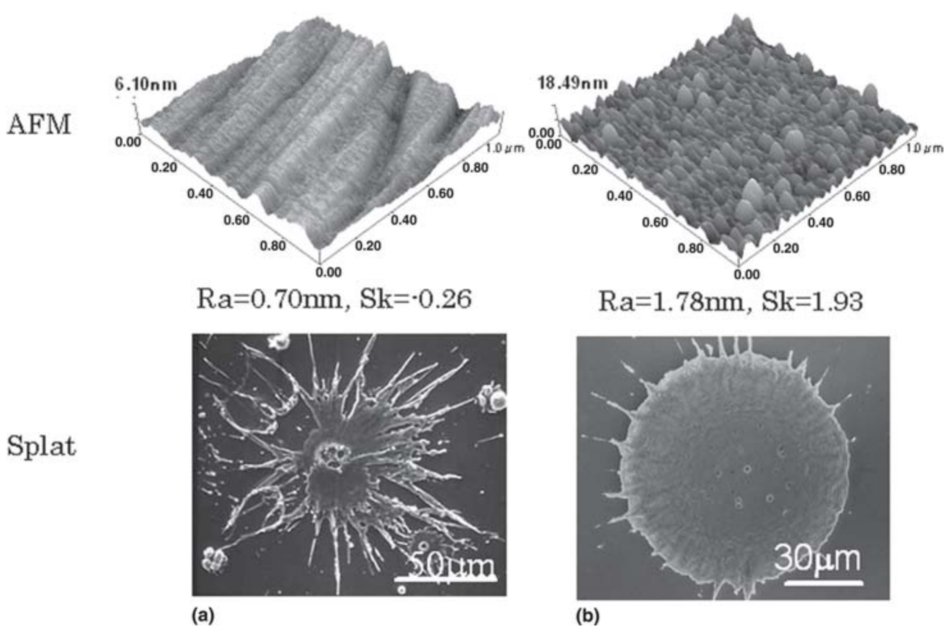

Fig. 3 AFM profiles and splat morphologies on AISI 304 substrate: (a) as polished; (b) laser treated

From the result, it became clear the R_a value stayed almost the same as that of the polished substrate surface up to 50 W/mm^2; however, it remarkably increased with laser power density over 50 W/mm^2. On the other hand, the *Sk* value increased with laser power density over 50 W/mm^2, and it changed from negative to positive, correspondingly. From the results mentioned previously, it was indicated that substrate surface topography in nanometer scale might be a possible dominating factor in the flattening of thermal sprayed particles.

3.2 Effect of Chemical Factor and Adsorbate on Flattening Behavior of Thermal Sprayed Particles

Meanwhile, the effect of both chemical factor and adsorbate/condensate on the flattening was investigated. To do this, the fraction of disk splat was investigated on the substrate once heated and kept in an air atmosphere at room temperature for a long time. In other words, the effect of elapsed time after heating

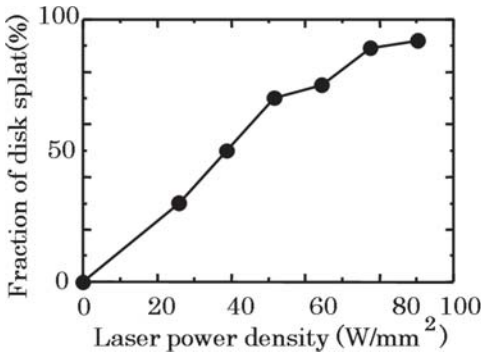

Fig. 4 Relationship between laser power density and fraction of disk splat

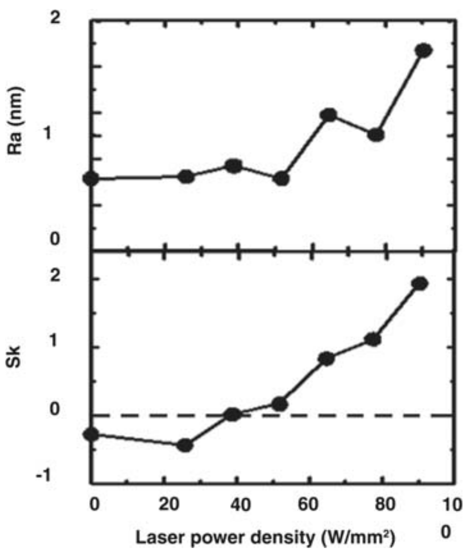

Fig. 5 Surface character change of the laser treated substrate

on the splat morphology change was investigated. Elapsed time was 24, 48, 72, 96, and 120 h. The result is shown in Fig. 6. As shown in Fig. 6, 24 h after heating, a higher fraction of disk splat was maintained. When 50 h or more had passed since the substrate was heated, however, the fraction of disk splat began to decrease. Finally, the fraction of disk splat decreased to less than 50% over 72 h and more. Measurement results both on R_a and *Sk* of once-heated substrate kept in an air atmosphere at room temperature for a long time revealed that the values were almost the same as those of the substrate just after cool down from heating, regardless of the elapsed time. Thus, some other factors, for example, change in the condition of adsorbates/condensates on the substrate surface, may have an effect.

To confirm the effect of surface topography on the flattening of thermal sprayed particle more precisely, gold (Au) was coated both on as-polished and once-heated substrates surface to make the chemical composition on the surface exactly similar. The results both of splat morphology and AFM analysis are shown in Fig. 7. From the comparison with Fig. 2, it was recognized that both R_a and *Sk* values of Au-coated substrates showed almost similar to those of the uncoated substrates. The projections were observed typically on the Au-coated once-heated substrate, and

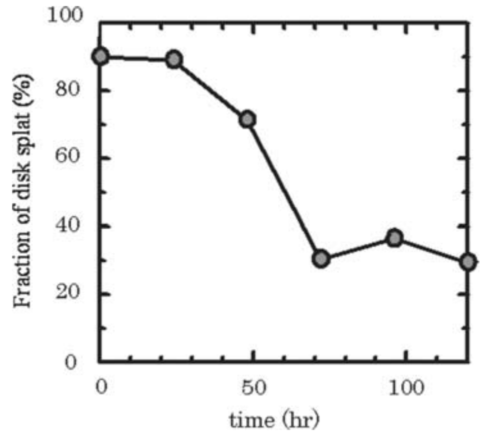

Fig. 6 Relationship between elapsed time after heating to 723 K and fraction of disk splat

the *Sk* value was positive. However, it was found that the splat morphologies on both Au-coated substrates were similar and not disk type but a splashlike shape. The result indicates that a physical factor such as a surface topography is not always the dominant factor in the flattening of thermal sprayed particles.

To confirm this finding, pure gold Au substrate was used and the flattening behavior of the thermal sprayed particle on the Au substrate was observed. As shown in Fig. 8, the R_a value of once-heated substrate changed to be rather bigger than that of the substrate in the as-polished condition, while the *Sk* value of both substrates was almost the same and indicated as negative. However, it was found that the splat morphology on once-heated Au substrate was disk type. As Au is not oxidized so easily by heating to several hundred degrees Celsius even in air, both as-polished and once-heated substrates should have a pure Au surface commonly. This means that both chemical and physical factors do not affect the flattening of the thermal sprayed particle.

The common fact through all observation results mentioned previously is that the disk-type splat appeared only on the substrate once heated in air and within a short period after the heating. Namely, elapsed time in a scale of hour may bring about some kind of change on the substrate surface condition. By summarizing the results mentioned previously, it is suggested that one of the most possible dominating factors in the flattening behavior may be the rest factor, that is, adsorbate/condensate on the surface. However, as the existence of adsorbates on the substrate surface cannot be detected directly in most practical case, the role of the factor has to be investigated more precisely in future study.

4. Conclusions

Laser treatment was used, and the influence of surface change by laser treatment on flattening behavior of thermal sprayed particles was investigated. Furthermore, the effect of surface character in both physical and chemical aspects on flattening behavior of thermal sprayed particles was verified. Results obtained in this study can be summarized as:

- Mean surface roughness, R_a, on the laser treated surface ranged transitionally increased with laser power density in a high-power region, correspondingly the *Sk* value changed

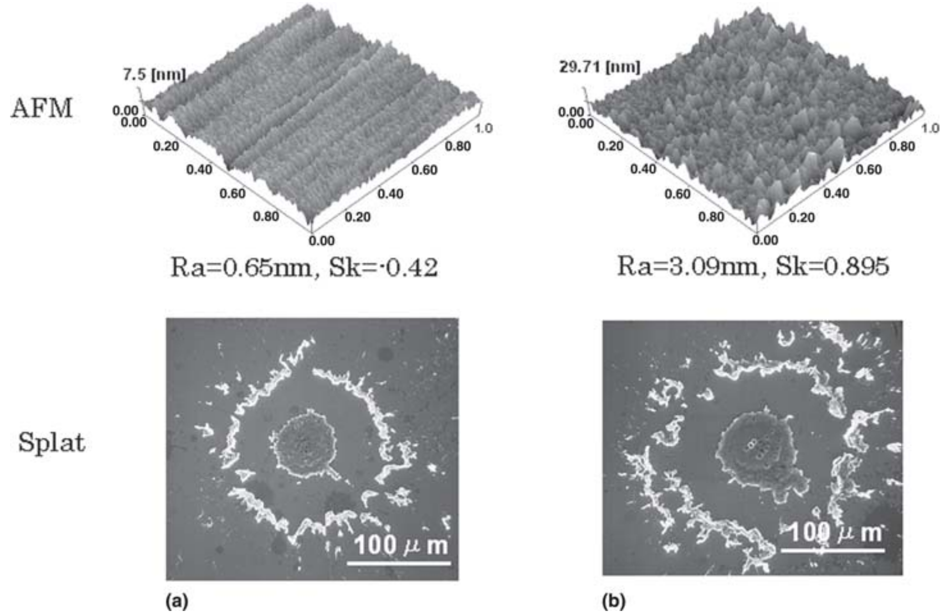

Fig. 7 AFM profiles and splat morphologies on Au-coated substrate: (a) as polished; (b) once heated

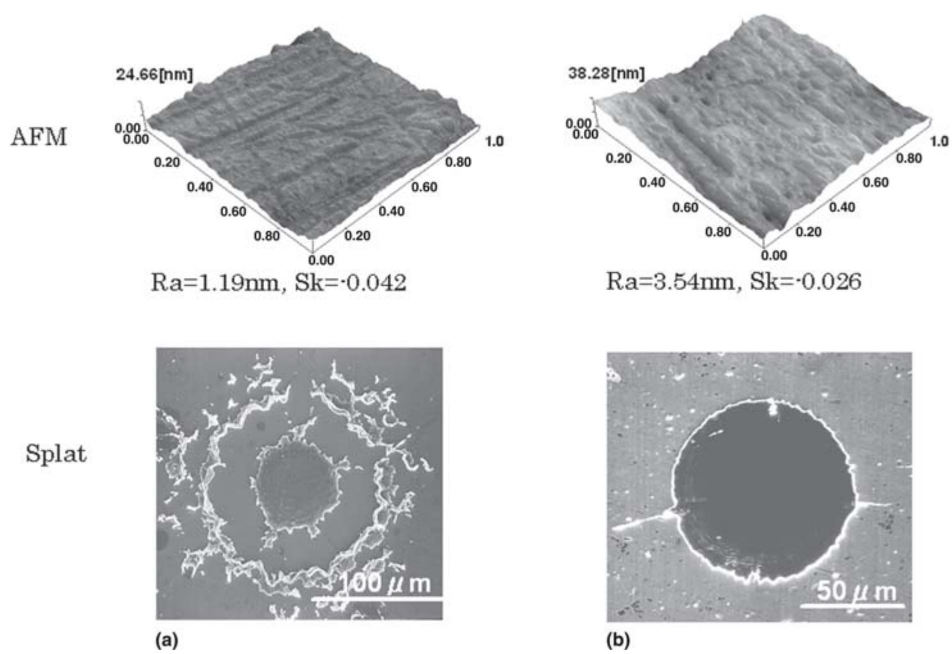

Fig. 8 AFM profiles and splat morphologies on Au substrate: (a) as polished; (b) once heated to 723 K

from negative to positive. Moreover, fraction of disk splat increased correspondingly with increasing of laser power density.

- The decreasing tendency in a fraction of disk splat was recognized on the substrate kept in an air atmosphere at room temperature for a long period. The result indicates that change in the adsorption condition on the surface may cause this change.
- Splash splat was observed on the Au-coated substrate even in the case of once heated in air. The result implies that physical factor as surface roughness character is not always the essential domination on the flattening behavior of thermal sprayed particle.
- It was found that the splat morphology on as-polished Au substrate was splash type, while that on once-heated Au substrate was disk type. The fact indicates that the chemical factor is not always the dominant factor in the flattening of thermal sprayed particle.

- It is likely that the disk-type splat was recognized only on the substrate once heated in air and the heating was effective only in a short period after heating.
- It is suggested that one of the most possible dominating factors in the flattening behavior may be the adsorbate/condensate on the surface. However, as the existence of adsorbates on the substrate surface cannot be detected directly in most practical case, the role of the factor has to be investigated more precisely in the future study.

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