# THE SPLAT FORMATION ISSUE IN THERMAL SPRAY PROCESSES

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#### ABSTRACT

Transition phenomenon in the flattening behavior of the thermally sprayed metallic particles has been recognized in our series of experimental works. Based on the results, a hypothesis has been proposed, namely, ultra-rapid cooled chill structure, preferentially formed at the bottom part of the splat, plays an essential role for the generation of the disk splat. Universality of this hypothesis beyond material difference was verified experimentally in our recent study, by using several kinds of ceramic materials with different thermal properties. To perform this, Al<sub>2</sub>O<sub>3</sub>, Y<sub>2</sub>O<sub>3</sub> and YSZ powder materials were plasma sprayed onto AISI304 stainless steel substrate, and a fractional change of the disk splat with a substrate temperature increase was investigated, followed by a precise observation of the cross-section microstructure of the splats. The results obtained showed that unique amorphous and chill structures were observed in Al<sub>2</sub>O<sub>3</sub> and Y<sub>2</sub>O<sub>3</sub> splat at the bottom, respectively, indicating that similar disk formation mechanism in metallic material may act in these materials. On the other hand, only normal columnar structure was recognized in YSZ splat. It was indicated that a rapid increase in viscosity may act on this material. Consequently, our hypothesis was verified partially, beyond the materials difference.

Keywords: thermal spray, splat, transition temperature, chill structure, viscosity.

### **1 INTRODUCTION**

Objective of our series of observations [1] on the flattening behavior of a thermally sprayed single splat is to establish the controllability or reliability of the thermal spray process. Since an individual particle is a fundamental element for the coating formation, the flattening behavior of thermally sprayed particles onto the flat substrate surface has been precisely investigated. The results showed that a transitional phenomenon was recognized in the flattening behavior both with the substrate temperature and ambient pressure changes. That is, the flattening of the splat changes transitionally from a distorted shape with splash to a disk-shaped splat without splash at certain critical temperature and pressure values, respectively. A transition temperature,  $T_t$  and transition pressure,  $P_t$  were defined and introduced by the author [2], respectively, for those critical conditions. Correspondingly, it was confirmed that the coating adhesive property on the blasted substrate surface changes transitionally on those critical temperature and pressure ranges, respectively. Based on the results, three-dimensional transition curvatures by combining both transition curves was proposed for each sprayed material, as a controlling principle for the practical usage in the thermal spray industries [3]. Namely, information given in the single splat has a meaningful contribution onto the practical usage of the thermal spraying.

On the other hand, based on the fluid dynamics, namely the splashing parameter or so-called Sommerfeld Number, K [4], restraint mechanism for the splashing have to be clarified from an academic viewpoint. In our series of observations on the metallic materials, it was indicated that ultra-rapid solidified chill structure preferentially formed at the bottom part of the splat plays an essential role for the disk splat formation [5]. In our recent study, to verify the universality of this hypothesis beyond material difference between metal and ceramic, three kinds of ceramic powder particles with different thermal conductivity,  $Al_2O_3$ ,  $Y_2O_3$  and YSZ, were thermally sprayed onto the metallic substrate. The effect of cooling or



solidification behavior at bottom part of the splat on the flattening was investigated precisely in the present study and our hypothesis was verified experimentally.

## 2 EXPERIMENTAL PROCEDURES

Commercially available metallic and ceramic powders with diameter of several tens micrometers were used as feedstock powders as shown in Table 1. AISI304 stainless steel plates with dimensions of  $25\text{mm} \times 25\text{mm} \times 6\text{mm}$  finally polished with 0.3  $\mu$ m Al<sub>2</sub>O<sub>3</sub> buff were used as substrates. Sprayed particles were collected on the substrate surface using both atmospheric and low-pressure plasma spray equipment. The splat shape was classified and the number of both disk splat and splash splat were counted on every trial. The fraction of the disk splat was defined as the ratio of the number of disk splat to the total splat. The cross-section microstructure of the splat was precisely observed by means of a scanning ion microscope, SIM and transmission electron microscope, TEM after cutting the splat by focused ion beam microscope, FIB.

# **3 RESULTS AND DISCUSSION**

### 3.1 Transition mechanism in metallic materials

It is known that the break-up phenomenon of the liquid film generated in the collision of the liquid particle onto the flat solid surface can be evaluated by the splashing parameter, K, in the fluid dynamics field [4]. K is defined as  $We^{0.5}Re^{0.25}$  and it is based on the in-flight kinetic information of the liquid particle. Here,  $We = \rho dv^2/\gamma$ , and  $Re = \rho dv/\eta$ : where  $\rho$  is density, d is diameter, v is velocity,  $\gamma$  is surface tension and  $\eta$  is viscosity, respectively. Here, K has a critical value Kc = 57.7 empirically decided, and if the K value of the particle exceeds Kc, the liquid film shows the break-up after the collision onto the solid surface. To verify the real K value of the regular thermally sprayed particles, in-flight measurement onto the sprayed particles was conducted by using the DPV-2000 system. By introducing the measured temperature into each physical constant (i.e., density, surface tension, and viscosity), the actual K value was calculated for several kinds of sprayed metallic materials. As a result, it was found that K values in each sprayed particle were remarkably larger compared to Kc, regardless of the sprayed material [3]. It means that the regular thermally sprayed particles have enough higher driving force for the splashing. The result indicates that the essential issue in the transition phenomenon lies in not why and how the splashing occurs on the cold substrate surface, but why and how the disk splat appears on the hot substrate surface or in the reduced pressure condition instead of the splash splat.

|  | Cu              | Al <sub>2</sub> O <sub>3</sub> | Y <sub>2</sub> O <sub>3</sub>  | YSZ                           |
|--|-----------------|--------------------------------|--------------------------------|-------------------------------|
| Heat conductivity<br>[W•m <sup>-1</sup> •K <sup>-1</sup> ] | <b>401</b> : RT | <b>35</b> : RT<br>5.0: 2000K   | <b>14.5</b> : RT<br>2.2: 1600K | <b>3.0</b> : RT<br>1.5: 2000K |
| Melting point [K]  | 1358            | 2345                           | 2647                           | 2715                          |
| Specific heat<br>[kJ/kg•K]                                 | 0.386           | 0.8                            | 0.466                          | 0.46                          |
| Specific gravity   | 8.9             | 3.9                            | 5.01                           | 6.0                           |

Table 1: Metallic and ceramic powder materials used.





Figure 1: Cross-section microstructures of Cu splats obtained onto AISI304 stainless steel substrate at (a) room temperature substrate under atmospheric pressure, (b) heated substrate at 773K under atmospheric pressure.

Since the temperature difference of the substrate may cause the difference of the cooling rate, solidification behavior inside of the splat, solidification microstructure on the bottom surface of the splat was precisely observed to verify the transition mechanism in the metallic materials. On the bottom surface of the splat, after peeled off from the substrate surface, obtained at heated substrate or reduced pressure conditions indicated that ring shaped dense zone with fine microstructure was observed near the peripheral region. This fine microstructure near peripheral region indicates that rapid solidification might be occurred at this region. To confirm the solidification rate inside of the splat directly and to elucidate the effect of rapid solidification on the flattening, cross section microstructures of the splats were precisely observed by SIM. The results were shown in Fig. 1.

In the splash-typed splat collected on the room temperature substrate under atmospheric pressure (Fig. 1(a)), coarser columnar grains perpendicular to the substrate surface are observed in whole part of the splat. On the other hand, in the disk-typed splat collected on the 773K heated substrate, finer columnar grains inside of the splat are observed in whole part of the splat (Fig. 1(b)). Finer structure indicates that faster solidification occurred in the disk splat given on the heated substrate. However, since it is pointed out that the columnar solidification structure itself is generally formed after finishing the flowing of the liquid, it is found that the solidification itself inside of the splat never affect the flattening of the splat. Hence, difference in the mean cooling rate, and solidification behavior inside of the splat cannot be a reason for the difference in the splat shape.

More precise observation was conducted on the cross-section microstructures of the splats, and it was found that a layer composed with equiaxial fine grain, so-called chill crystal structure which usually caused by ultra-rapid cooling, indicated as blue rectangular shown in Fig. 1(b), was observed at the bottom part of the splat obtained on the heated substrate surface. The essential point is that chill crystal layer appeared on the heated substrate is much thicker than that on the unheated substrate. Moreover, the thickness of the chill structure layer becomes thicker close to the periphery region of the splat compared with the central region. This indicates that ultra-rapid cooling and resultant ultra-rapid increase of the viscosity and/or solidification occurred near the peripheral region preferentially and this ultra-rapid material change might act as the restraint for the splashing. This is actually a new finding in the relating research field.

### 3.2 Transition mechanism in ceramic materials

Universality of the new finding, namely ultra-rapid solidified chill structure in the splat has a restraint role for the splashing, obtained in the metallic materials was verified in the ceramic materials, since the similar transition behavior is recognized also in the ceramic splats. The change of the splat shape and fraction of disk splat with substrate temperature increase for each sprayed ceramic material was summarized in Fig. 2, including the data on Cu particle. It was found from the figure that the ceramic material with higher thermal conductivity has a tendency having a lower transition temperature. The result shows that the ceramic material having a higher heat conductivity tends to show a transitional behavior easier, indicating that a certain solidification property of the material may affect the flattening. To verify this tendency, the cross-section microstructures of the disk splats for each ceramic material collected on the heated substrate surface was observed. The results were summarized in Fig. 3.

In Al<sub>2</sub>O<sub>3</sub> splat shown in Fig. 3(a), having the highest heat conductivity, an amorphous layer, mostly half thickness of the splat, was clearly observed at bottom part of the splat entirely whole part in the radius direction. In case of  $Y_2O_3$  shown in Fig. 3(b), on the other hand, having a middle value of the heat conductivity, chill crystal layer similar to the metallic materials at bottom part of the splat was clearly observed entirely whole part in the radius direction. In these two materials, ultra-rapid solidification at bottom part of the splat may cause the disk splat, namely, the result supports the above hypothesis given in the metallic materials. However, in ZrO<sub>2</sub> splat shown in Fig. 3(c), only the columnar structure similar to the metallic material was observed without any typical ultra-rapid solidified microstructure at entirely bottom part of the splat, even in the disk splat. In this case, solidification cannot be



Figure 2: Splat shape and fraction change with substrate temperature increase in each material.





Figure 3: Cross-section microstructures of ceramic splats obtained onto heated AISI304 stainless steel substrate.

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