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Splash Splat to Disk Splat Transition Behavior in Plasma-Sprayed Metallic Materials

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A variety of metallic powder particles were thermally sprayed onto the mirror polished metallic substrate surface and the effect of both substrate temperature and ambient pressure on the flattening behavior of the particle was systematically investigated. In the flattening behavior of the sprayed particle onto the substrate surface, critical conditions were recognized both in the substrate temperature and ambient pressure. That is, the flattening behavior changed transitionally on that critical temperature and pressure range, respectively. A transition temperature, T_t, and transition pressure, P_t, were defined and introduced, respectively for those critical conditions. The fact that the dependence of both transition temperature and transition pressure on the sprayed particle material had similar tendency indicated that the wetting of the substrate by the molten particles seemed to be a domination in the flattening. Three-dimensional transition curvature by combining both transition temperature and transition pressure dependence was proposed as a practical and effective controlling principle of the thermal spray process.

Keywords thermal-sprayed particle, flattening behavior, substrate temperature, ambient pressure, three-dimensional curvature, transition temperature, transition pressure

1. Introduction

Thermal spraying has been established as a common technology to fabricate thick coatings of most industrial materials. It is, however, pointed out that the process controllability or reliability of thermal spraying is still insufficient. As a flattening behavior of an individual particle impinging onto the flat substrate surface can be recognized as a fundamental phenomenon of the coating formation in the thermal spray process, a clarification of the flattening behavior of the particle is essential to establish the process controlling. Most of the theoretical or numerical analyses ever conducted indicate that flattening degree of the splat is simply given as a function of the Reynolds number, Re (Ref 1). This means that both in-flight temperature and velocity of the particle are the dominating factors in the flattening of the sprayed particles, and thus for controlling the thermal spray process. In fact, conventional thermal spray technology has developed into two different ways up to today, that is, temperature dominated and velocity dominated processes. Plasma spraying and HVOF spraying can be regarded as representatives for both.

However, instead of these particle oriented factors, it has been pointed out in our previous results that the particle/substrate interface oriented factors, such as a substrate temperature and ambient pressure affect more significantly the flattening behavior of the sprayed particles (Ref 2-6). In particular, our experimental results have revealed that a splat shape of most materials onto the flat substrate surface undergoes a transition from a distorted shape with splash to a disk shape without splash over a narrow temperature range with an increase of the substrate temperature (Ref 2-3). The transition temperature, T_t, has been defined by the author as a critical in the substrate temperature. Since then, the drastic change in the splat pattern nearby the transition temperature has become of great concern in recent years. Based on the experimental, analytical, or numerical data, a unified explanation have been sought to describe the transition behavior. As the most possible domination, the role of solidification in the flattening particle (Ref 7), wetting at particle/substrate interface (Ref 2, 3), and desorption of adsorbates on the substrate surface (Ref 8) on the flattening have been investigated.

To establish the controlling principle for thermal spray process, the transition behavior in the flattening of thermally sprayed particles on the substrate surface as a function of the substrate temperature was investigated

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more precisely. Additionally, similar transition behavior as a function of ambient pressure was investigated in the present study.

2. Experimental Methods

Powder materials used are a variety of commercially available metallic materials, Ti, Ni, Cu, Fe, Cr, Kojundo Chemical Lab. Co., Ltd., Japan. These were made mostly by atomizing method. Size distributions of each powder given by the producing company were commonly in the range of 10-44 μm. Mirror polished AISI 304 stainless steel plates with 25 mm × 25 mm × 5 mm were used for the substrate. Both atmospheric plasma-spraying equipment, Sulzar Metco Co., Ltd., 9MB, Switzerland, and low-pressure plasma-spraying equipment, Plasmadyne, SG-100, USA, were used for the spraying and the spraying conditions are summarized in Table 1. To investigate the effect of substrate temperature on the flattening behavior of the particles, powder materials were sprayed onto the substrate kept at various elevated temperature conditions. Moreover, to investigate the effect of ambient pressure on the flattening behavior of the particles, powders were sprayed onto the substrate kept at various pressure conditions in LPPS. In LPPS experiments, substrate temperature was kept at room temperature by cooling by water. Spraying in LPPS was conducted at the designated pressure after once evacuated to the lowest pressure condition of the equipment. Both in-flight temperature and velocity of the sprayed particles were measured by using DPV-2000 system, Tecnar Ltee, St. Bruno, Quebec, Canada. Through the measurement, average values both of in-flight velocity and temperature were given by this equipment. Transition temperature, T_t, and transition pressure, P_t, for each powder material were measured. For the particle collection, both stainless steel plate with ϕ10 mm hole and graphite plate with ϕ20 mm hole were used. By falling graphite plate rapidly just in front of the fixed steel plate, sprayed particles passed through the two holes could be collected on the substrate surface. Details of the measuring methods can be found in our previous report (Ref 2). In the measuring, around 50 or more splats were collected and observed on the substrate on every trial in the experiments.

Table 1 Plasma-spraying conditions both for APS and LPPS

Parameter	Value
Atmospheric plasma spray	
Spray distance, mm	200
Arc current, A	600
Operating gas flow rate, L/min—1st: N_2	70.8
Operating gas flow rate, L/min—2nd: H_2	4.72
Arc voltage, V	70
Powder carrier gas: Ar, L/min	5.0
Powder feed rate, g/min	6.0
Low-pressure plasma spray	
Chamber pressure, Torr	30-760
Spray distance, mm	200-300
Arc current, A	900
Operating gas flow rate, L/min—1st: Ar	50
Operating gas flow rate, L/min—2nd: He	12
Arc voltage, V	40
Powder carrier gas; Ar, L/min	5.0
Powder feed rate, g/min	6.0

3. Results and Discussion

3.1 Definition of Transition Temperature, T_t

To confirm the effect of practical process parameters on the particle's velocity and temperature, in-flight velocity and temperature were measured by DPV-2000 system. As a typical example, effect of both operating gas flow rate and spray distance of the normal plasma spraying of Ni particle on the velocity and temperature were measured. Results obtained are shown in Fig. 1. From the figure, it is clear that there are no significant changes in velocity and temperature with these process parameters. This means that in a reasonable variation range of each process parameter, both in-flight velocity and temperature do not change so significantly.

Instead of the particle oriented factors, the effect of particle/substrate interface oriented factors, namely, effect

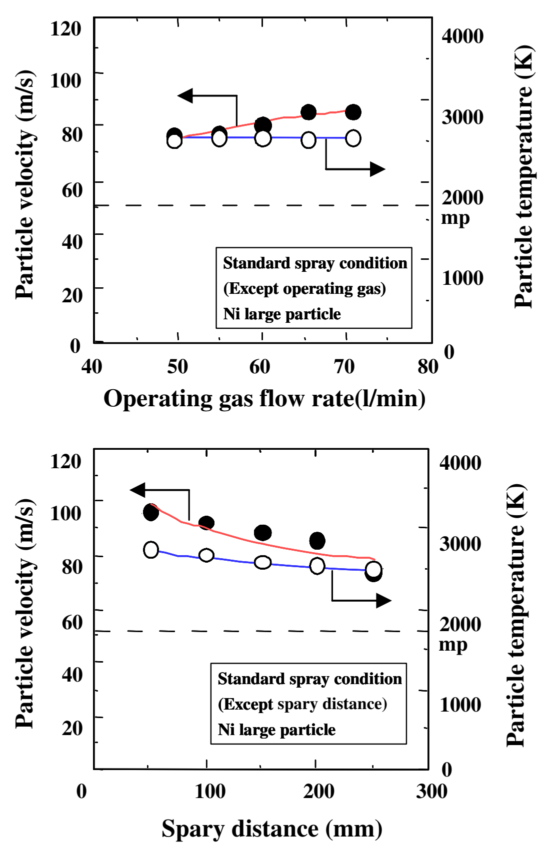

Fig. 1 Effect of spray parameters on particle in-flight conditions in atmospheric plasma spraying measured by DPV-2000 system

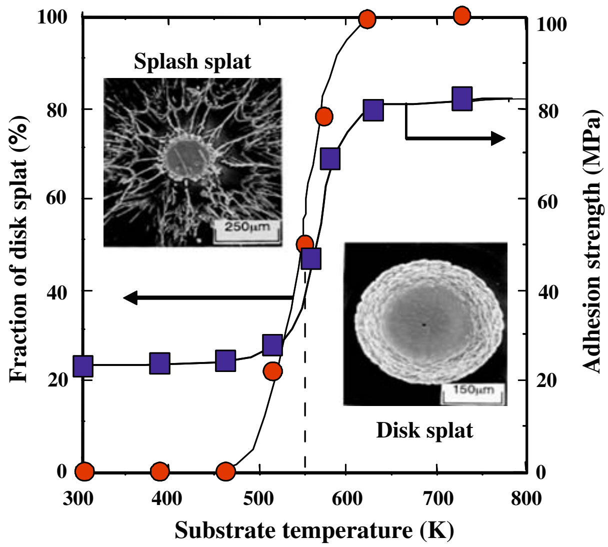

Fig. 2 Dependence of fraction of disk splat and coating adhesion strength on substrate temperature

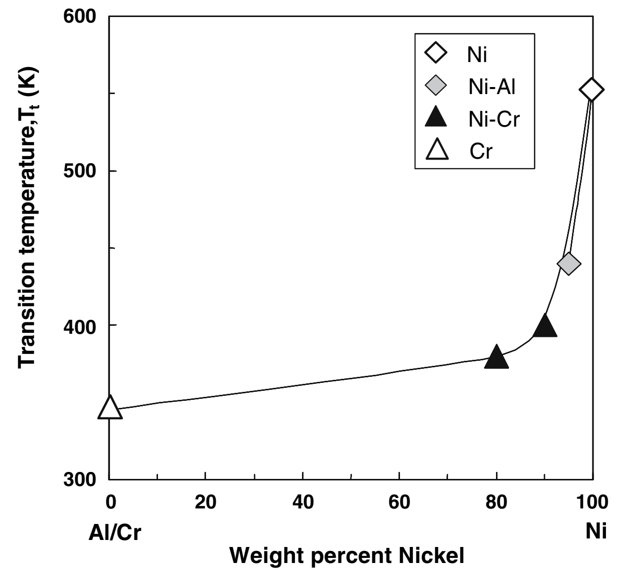

Fig. 3 Dependence of chemical composition on transition temperature both in Ni-Al and Ni-Cr systems. Substrate: AISI304 stainless steel

Table 2 Transition temperature for each powder material on AISI 304 stainless steel substrate

Powder material	Transition temperature, K
Ni	610
Mo	474
Cu	394
Cr	387
Cu-30Zn	505
Al_2O_3	318
TiO_2	350
YSZ	345

Substrate: AISI 304 stainless steel. Spraying device: APS

of substrate temperature on the flattening behavior of the sprayed particles were investigated (Ref 2-6). Especially, our experimental results have revealed that a splat shape of most metallic and ceramic material onto the flat substrate surface has a transitional changing tendency from a distorted shape with splash to a disk shape without splash at a narrow temperature range with an increase of the substrate temperature as shown in Fig. 2, which was a typical case of Ni particle on AISI304 substrate (Ref 2, 3). The transition temperature, T_t, at which the particle's splat pattern change to the form without splashing from the one with splashing was defined and introduced by the authors (Ref 2). Moreover, it was verified experimentally that the adhesion strength of the coating changed transitionally with the substrate temperature increasing, and its dependence on the substrate temperature corresponded quite well to that of the splat pattern as shown additionally in Fig. 2 (Ref 3). Similar results were given by other researchers (Ref 9, 10). Thus, the investigation of the flattening mechanism of the sprayed particle on the substrate surface is significantly meaningful for the practical usage of the thermal spray process.

3.2 Physical Meaning of Transition Temperature

The transition temperature, T_t, for each powder material onto stainless steel substrate was measured. To conduct this, the fraction change of the disk splat on the substrate surface with substrate temperature increasing was systematically measured. The measured T_t values are summarized in Table 2. Standard deviation of T_t is approximately 5-10 K. From the table, it is recognized that the T_t varies widely with particle material and oxide ceramic materials have relatively low T_t.

Here, the problem lies in "what is the physical meaning of transition temperature?" T_t values both of Ni-Al and Ni-Cr material systems are summarized in Fig. 3. From the figure, it is found that both Ni-Al and Ni-Cr materials have lower T_t values compare with pure Ni, namely, small amount of Al or Cr addition to Ni causes a significant decrease in T_t. As T_t is defined as a critical substrate temperature for an appearance of the coating with higher adhesion strength with the substrate temperature, it is quite beneficial for Ni-Al or Ni-Cr to be used as undercoating materials instead of pure Ni. In other words, higher coating property can be given by utilizing Ni-Al or Ni-Cr with less effort in preheating conditions of the steel substrate.

From a practical point of view, it can be pointed out that T_t has an indicative role as an optimum preheating condition of the substrate. Up to now, we have recognized the usefulness of the substrate preheating empirically in practical thermal spray process. However, we have no answer to "what is an optimum preheating condition for a given particle/substrate combination?" or "what is the physical meaning of the preheating itself?" If there exists an optimum preheating condition for a given particle/substrate combination, T_t may correspond to this and give the useful information.

3.3 Essential Point in Transition Problem

From an academic viewpoint, the initial problem to be solved is ''why and how the splashing occurs on the cold substrate surface?'' The possible reasons can be the rapid solidification on the bottom surface of the splat, adsorbates on the substrate surface, poor wettability and so on. Computer simulations on the flattening behavior of the sprayed particle have been conducted by several researchers (Ref 11). Fard et al. indicated that the unstable situation in liquid flow occur at the spreading periphery of the particle after the collision. And their simulation revealed that the rapid solidification on the bottom surface of the splat occurred correspondingly. The preferential solidification at the bottom part of the splat may affect the flowing of the melted part located on the solidified layer, namely, induce the rapid expansion of the liquid as a thin film. Thus, their simulation result has indicated that the rapid solidification on the bottom surface can be a trigger for the splashing. We have also pointed out that the initial solidified layer affects the splashing of the particle in the case of metal particles (Ref 2, 4).

To clarify this, alumina splats were collected on the substrate whose surface was partially Au coated. T_t is 420 K and 370 K for Au coated and naked stainless steel substrate, respectively. The substrate temperature was 400 K, at which the splash splat was observed on Au-coated substrate and the disk splat was observed on naked substrate (Ref 6). The splat morphology was observed, which impinged onto the boundary between Au coated and naked substrate surface. The typical splat morphology is shown in Fig. 4. It was clear that the splat was composed of both characters, that is, the splash splat appeared on the Au-coated surface and the disk splat on the naked surface. The result is quite natural. However, it is notable that on the Au-coated surface, the splash splat was formed without any initial solidification part of the splat. Similar experimental results has been presented by other researchers (Ref 12, 13). This fact indicates that the initial solidification is not always a necessary condition for the splashing. Thus, we have estimated that the wetting affects more strongly to the transition.

Another possible explanation for the transition phenomenon can be given by the desorption of the adsorbates on the substrate surface due to the heating. The effect of adsorbates/condensates on the substrate surface on the flattening behavior of the thermal-sprayed particles has been precisely investigated by Jiang et al. (Ref 8). The transition phenomenon from splash splat to the disk splat with substrate temperature increasing can be explained reasonably by this hypothesis. Namely, on the hot substrate surface, better contact between droplet and substrate can be attained by the desorption of adsorbates. On the cold substrate surface, on the other hand, adsorbates become a source of interface porous microstructure and also assist the radial flow from central to the periphery on the bottom surface of the splash splat. Moreover, in a certain case, adsorbates can induce the extremely rapid expanding or flowing of the splat due to its explosion by the rapid heating.

In general, however, it is well known that the break-up phenomenon of the liquid film generated by the collision of the particle to the solid surface can be evaluated by the splashing parameter, K, in the fluid dynamics field (Ref 14). 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, $We = \rho d v^2/\gamma$, and $Re = \rho d v/\eta$ and ρ: density, d: diameter, v: velocity, γ: viscosity, η: surface tension, respectively. K has a critical value, $K_c = 57.7$, and if the K value of the particle exceeds K_c, the liquid film show the break-up after the collision onto the solid surface. To investigate the real K value, measurement on the in-flight information of the regular thermal-sprayed particles was conducted by using DPV-2000 system. By introducing the measured temperature into each physical constant, that is, density, surface tension, and viscosity, the real K value was obtained as shown in Fig. 5 for a few kinds of sprayed metallic materials. K_c value is indicated in the figure for comparison. As obviously recognized in the figure, K values obtained were remarkably larger compared to K_c regardless of the sprayed material. More details on K value of the sprayed particles were investigated recently (Ref 15) and almost similar tendency have been confirmed. Namely, it is confirmed that the regular sprayed particle has enough driving force for the splashing. The results mentioned above indicates quite important fact that the essential point of this transition problem lies in not ''why and how the splashing occurs

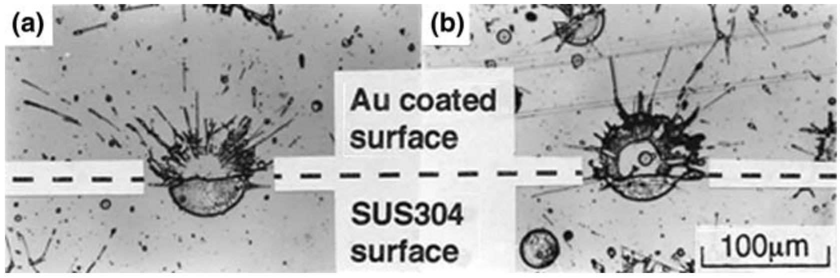

Fig. 4 Al_2O_3 splat morphologies onto AISI 304 substrate at 400 K

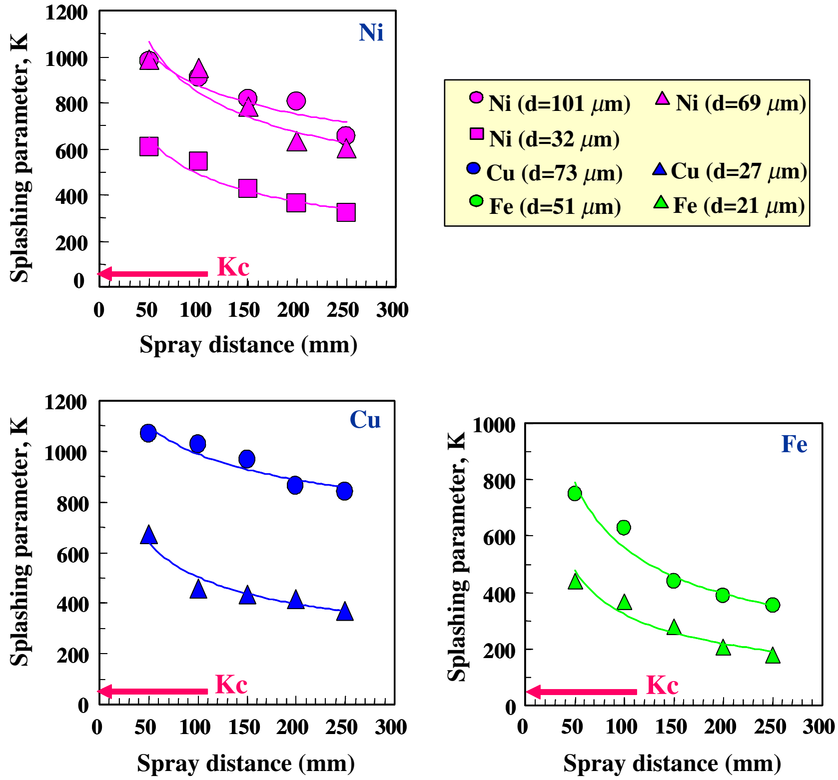

Fig. 5 Measured splashing parameter, K values for regular sprayed particles

on the cold substrate surface?'' but ''why and how the disk splat appears on the hot substrate surface instead of the splash splat?'' This indicates that we need to find a mechanism which induces the occurrence of disk splat while inhibiting the splashing.

3.4 Definition of Transition Pressure, P_t, and three-dimensional Transition Curvature in Thermal Spray Process

Instead of the substrate temperature change, effect of desorption of adsorbates on the flattening behavior was systematically investigated in the low-pressure plasma spraying by reducing the ambient pressure. The substrate temperature was kept at room temperature. The typical splat morphologies of metallic particles onto stainless steel substrate surface were shown in Fig. 6. From the results, it is found that the disk splat appears easily by reducing the ambient pressure while the substrate temperature is kept constant at room temperature. Experiments also revealed that the transition behavior from the splash splat to the disk one was recognized in most of the metallic materials by reducing the ambient pressure as shown in Fig. 7.

The transition pressure, P_t, as a critical pressure at which the fraction of disk splat exceeds 50% with reducing the ambient pressure was defined and introduced by the author. P_t values were given experimentally by measuring the fraction of disk splat on the substrate kept at room temperature with reducing the chamber pressure. As the plasma length is easily changed by the chamber pressure, the effect of spraying distance on the fraction of disk splat appearance under three kinds of chamber pressures was investigated. From the results, it was found that the fraction of disk splat did not change so much regardless of the spraying distance and in any chamber pressures. This indicates that the in-flight conditions, that is, both velocity and temperature of regularly sprayed particles does not change so much even in low-pressure spraying, and thus in-flight conditions do not strongly affect the flattening. The reduction in ambient pressure, reduced the vapor pressure of the adsorbates. This probably accelerated vaporization/desorption of the adsorbates from the surface. Desorption of the adsorbates improved splat-substrate wetting during flattening, resulting in disk-like splats. Hence, the fact mentioned above indicates that the desorption of adsorbates can affect independently the flattening behavior as a transition from splash to the disk. Transition pressure distribution for each sprayed material was summarized in Fig. 8. It is quite noticeable that material order corresponds well to Periodic table, that is, Ti, Cr, Ni, and Cu. As the physical or chemical meaning of this material order is not clarified well at this moment, this has to be investigated in the future research.

In the figure, dependence of the transition temperature on the sprayed material was also shown for the comparison purpose. From the figure, it is recognized that the

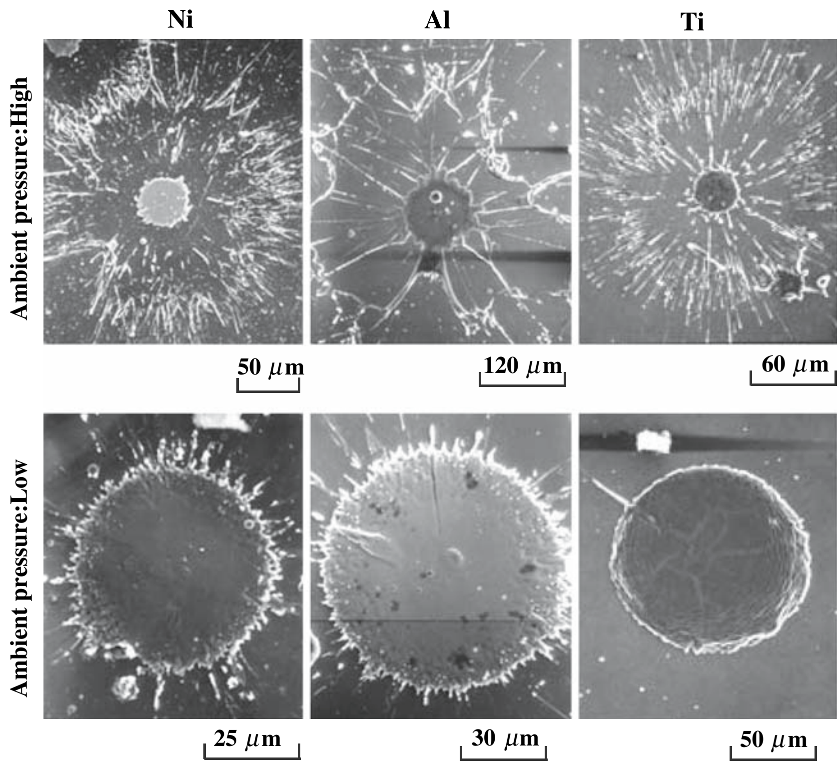

Fig. 6 Splat morphologies of metallic particles on AISI304 steel substrate at different atmospheric pressures

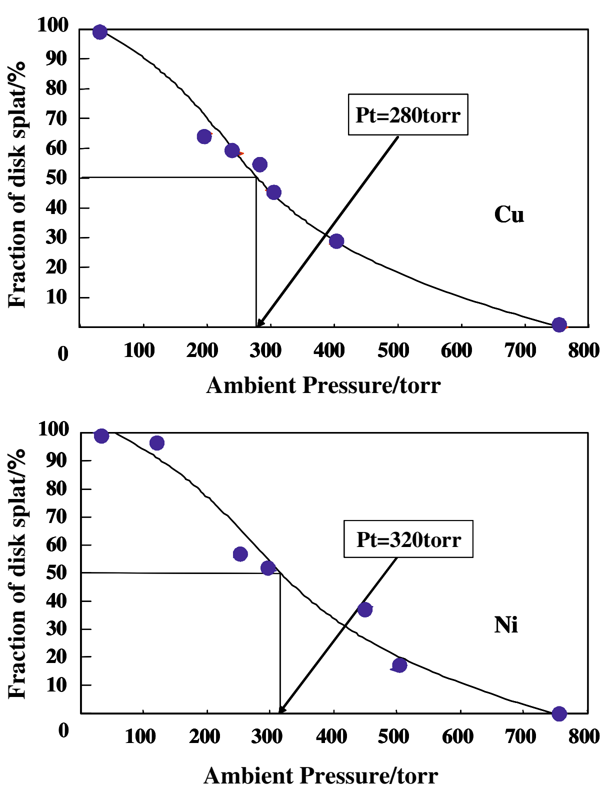

Fig. 7 Dependence of fraction of disk splat on ambient pressure and definition of Transition pressure, P_t

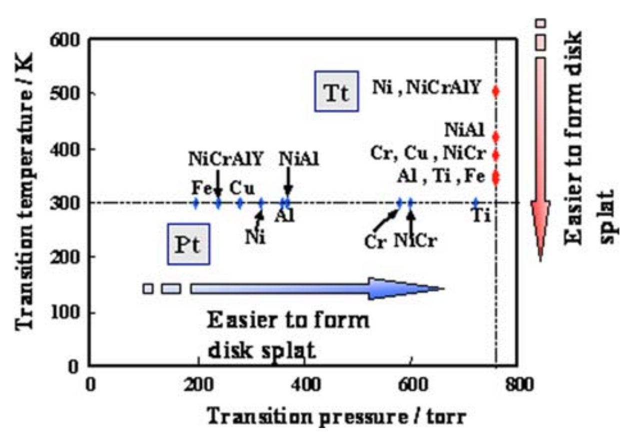

Fig. 8 Transition in thermal-sprayed particle with both substrate temperature and ambient pressure

dependence of T_t and P_t on the particle material has quite similar tendency. The fact indicates that equivalent effect in the flattening of splat behavior both by increasing the substrate temperature and reducing ambient pressure can be given from a certain essential common domination. Observation on the bottom surface microstructure change of the splat both by changing the ambient pressure and substrate temperature was carried out, respectively. The result was shown in Fig. 9. From the figure, it is clear that the similar tendency in microstructure change from the

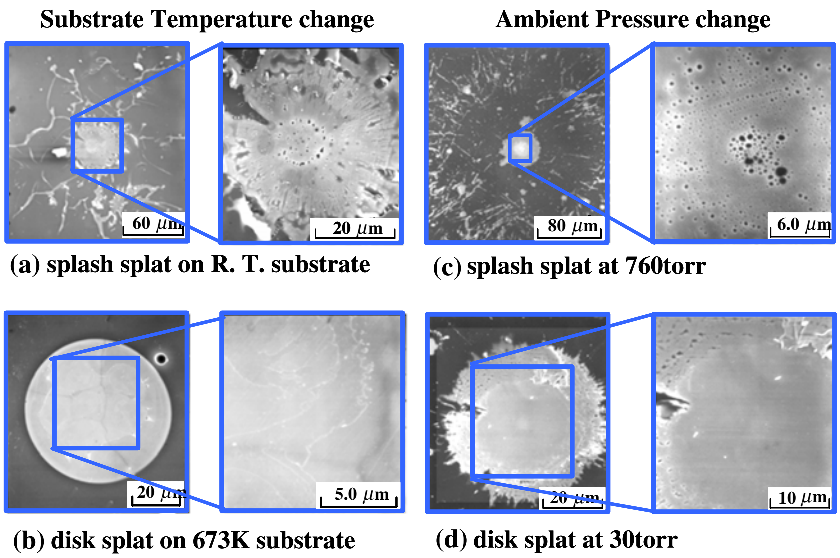

Fig. 9 Three-dimensional transition curvature of flattening behavior in thermal spray process

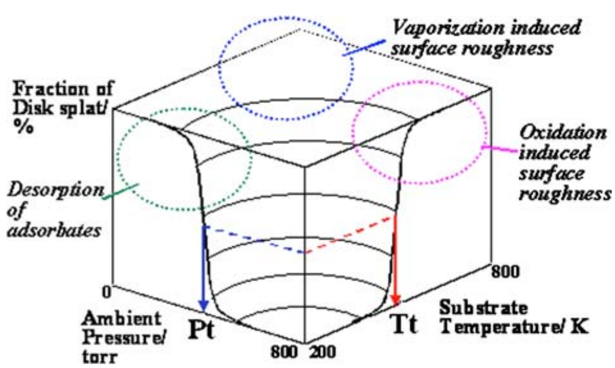

Fig. 10 Three dimensional transition map of flattening behavior in the thermal spray process

porous to the dense was observed in both cases. From the facts mentioned above, it can be concluded that wetting of the substrate by the molten particles seemed to be the essential domination for the flattening behavior of the thermal-sprayed particles.

The dependence of fraction of disk splat both on the substrate temperature and ambient pressure was summarized schematically in Fig. 10, as a three-dimensional transition curvature. By selecting the optimum operating conditions in the combination of both factors in the thermal spraying, we may be able to control the coating microstructure, and thus, any other properties, such as porosity, density, thermal conductivity, of the coating. To confirm this hypothesis, three-dimensional transition curvature for several kinds of powder materials were measured based on the experimental data, as shown in Fig. 11. As indicated in the results, each material had a

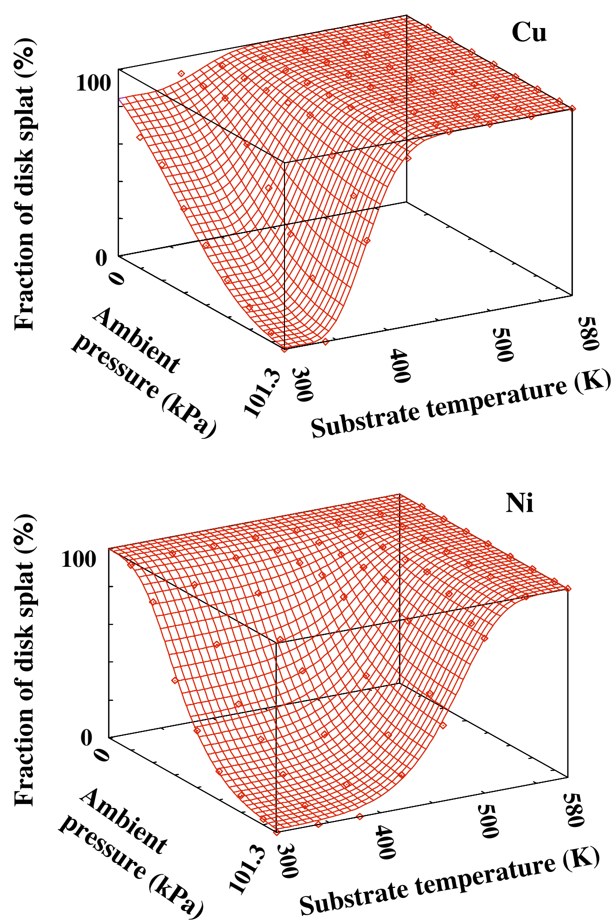

Fig. 11 Experimental results on three-dimensional transition curvature for each material

three-dimensional transition curvature actually. In these days, the thermal spray process is usually controlled by measuring the in-flight velocity and temperature of the particles and by feeding these data back into the input power of the equipment. Addition to this normal controlling method, the process can be controlled more effectively by introducing both T_t and P_t as for the practical and effective controlling principle of the thermal spraying.

4. Summary

The results obtained in this study are summarized as follows:

(1) From the measurement results in K value, it was found that the regular sprayed particle has enough driving force for the splashing. This indicates that the essential point of the transition problem lies in not why and how the splashing occurs on the cold substrate surface, but why and how the disk splat appears on hot substrate surface instead of the splash splat.

(2) The transition behavior from the splash splat to the disk one was recognized in most of the metallic materials by reducing the ambient pressure. The fact indicates that the desorption of adsorbates can affect independently the flattening behavior as a transition from splash to the disk.

(3) It was recognized that the dependence of T_t and P_t on the particle material has quite similar tendency. The fact indicates that both substrate temperature and ambient pressure may have an equivalent effect on the transition.

(4) By selecting the optimum operating conditions both in substrate temperature and ambient pressure in thermal spraying, we can control the coating microstructure and any other properties of the coating. Thus, the control of thermal spray process can be attained by introducing both T_t and P_t as for the practical and effective controlling principle of the thermal spray process.

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