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Effect of Interface Wetting on Flattening of Freely Fallen Metal Droplet onto Flat Substrate Surface

Masahiro Fukumoto, Eiji Nishioka, and Toshikazu Matsubara

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A free-falling experiment was conducted as a simulation of a thermal spray process. The flattening behavior of the freely fallen metal droplet impinged onto a flat substrate surface was investigated in a fundamental way. The substrates were kept at various temperatures, and the substrates were coated with gold by physical vapor deposition (PVD) and were prepared in order to investigate the effect of wetting at the splat-substrate interface on the flattening behavior of the droplet. A falling atmosphere was created with atmospheric pressure of nitrogen to prevent the oxidation of the melted droplet. Experiments under low-pressure conditions also were conducted. The different types of splat morphology were recognized in experiments conducted under a nitrogen atmosphere with atmospheric pressure. The splat morphology on a substrate at room temperature was of the splash type, whereas that on a substrate at high temperature was of the disk type. The microstructure observed on a cross-section of the splat obtained on the substrate at room temperature was an isotropic coarse grain, whereas that on the substrate at high temperature was a fine columnar grain. The grain size changed transitionally with increasing substrate temperature. The temperature of the transition on the gold-coated substrate was higher than that on the naked substrate. The microstructure of the cross-section of the splat obtained under low pressure was finely columnar even on the substrate at room temperature. The results indicate that the metal droplet wets better under the low-pressure condition than under the atmospheric pressure nitrogen condition and that wetting has a significant role in the flattening of the droplet.

Keywords freely fallen droplet, interface wetting, solidification, splat morphology, substrate temperature, thermal spraying, transition phenomenon

1. Introduction

Our previous studies on the flattening behavior of the particle thermally sprayed onto the flat substrate have shown that as the substrate temperature increases, the splat morphology changes from a splash type to a disk type.[1-3] Hence, the substrate temperature is one of the dominating factors in the flattening of a sprayed particle. It is, however, difficult to clarify the effect of other factors on the flattening and solidification behavior of the particle because they are hard to separate in a complex spraying process. In order to examine the transition phenomenon of the splat morphology, we conducted an investigation with a simulation of the thermal spraying. Free-falling experiments[4,5] have received more attention because the relating factors can be controlled easily.

In this study, the free-falling experiments, in which the metal droplet fell freely and impinged on the flat substrate, were conducted as a simulation and a simplification of the thermal spray process. The effect of wetting on the flattening behavior of the droplet was investigated to clarify the formation mechanism of the splash splat.

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2. Experimental

Figure 1 shows the experimental apparatus used in this study. Results of a previous study[1] have shown that the splat morphology depends on the spraying materials. Commercially available pure copper wires (optical density, 2 mm; >99.9% purity) were used to create the droplets. These wires were heated and melted by a radiofrequency heater (RWN-1-20, Nihon Koshuha Co, Ltd). Because the droplet starts falling when the equivalent volume of wire is completely melted by heating, the temperature of the droplet can be estimated by its melting point. The velocity of the droplet just before the collision with the substrate surface can be easily determined by adjusting the falling distance, that is, the height of the heating electrode from the substrate level. The mirror-polished AISI304 stainless steel substrates were heated and kept at various designated temperatures, and the temperature was measured by a K-type thermocouple. In order to change the material property of the substrate surface and to examine the

Nomenclature	
T_t	Transition temperature
T_s	Substrate temperature
L	Grain size
S	Solidification rate

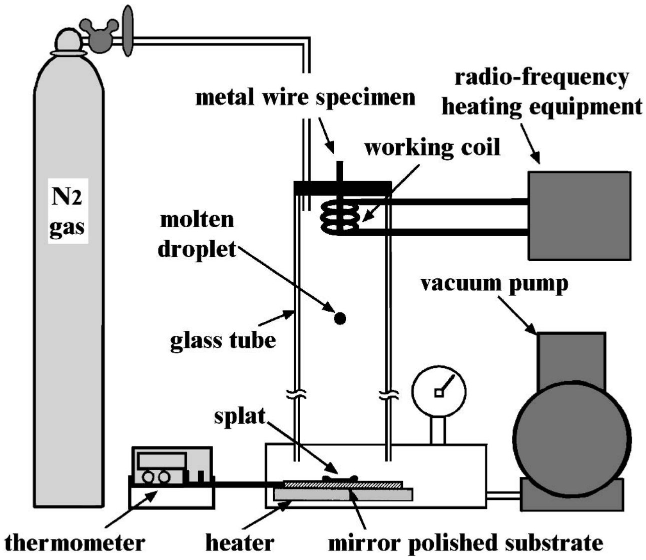


Fig. 1 Schematic drawing of the experimental setup

effect of wetting at various splat-substrate interface conditions, a thin gold layer was coated onto the substrates by physical vapor deposition (PVD). The flattening behavior on the coated substrate was compared with those on the naked substrate. The experiments were performed both in the atmospheric pressure nitrogen (107 kPa) and a low-pressure (1 kPa) condition. For a microstructure observation, the cross-section of the splat was etched with a mixture of 10 ml HNO_3 (1.4 N) and 70 ml methanol (96% pure).

3. Results and Discussion

3.1 Cross-Section Microstructures of the Splats

Figure 2 shows the representative splat morphology on the stainless steel substrate. The splat morphology on the substrate at room temperature was of the splash type, whereas that on the substrate at high temperature was the disk type. The substrate temperature at which the splat morphology changes transitionally from splash to disk was defined in our previous study[1] as the transition temperature (T_t).

In order to investigate the cooling behavior in the splat, the cross-section microstructures of the splats were observed and are shown in Fig. 3. The microstructure of the cross-section of the splat obtained on the substrate at room temperature (Fig. 3a) was an isotropic coarse grain, whereas that on the substrate at high temperature (Fig. 3b) was a fine columnar grain. The mean grain size of the splat obtained on the substrate at room temperature is obviously larger than that on the substrate at high temperature. In order to examine the relationship between the microstructure and the substrate temperature in more detail, the grain size was measured as the width of the column on the cross-section of the splat. The grain size was measured randomly for a total of 20 points on the cross-section of the splat, and the mean value was calculated from the data obtained. The results are shown in Fig. 4(a). As the substrate temperature increased, the grain size decreased transitionally, and the splat morphology changed correspondingly.

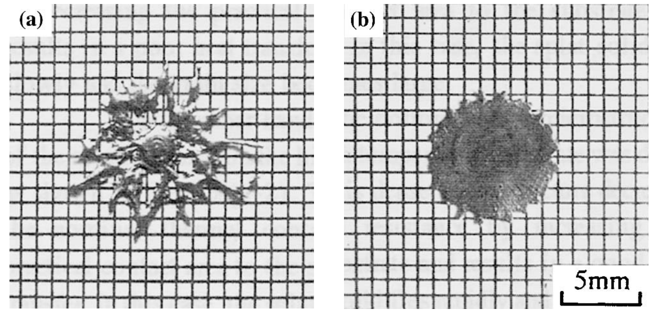


Fig. 2 Observation results of splat morphology. **(a)** T_s = 300 K; and **(b)** T_s = 600 K

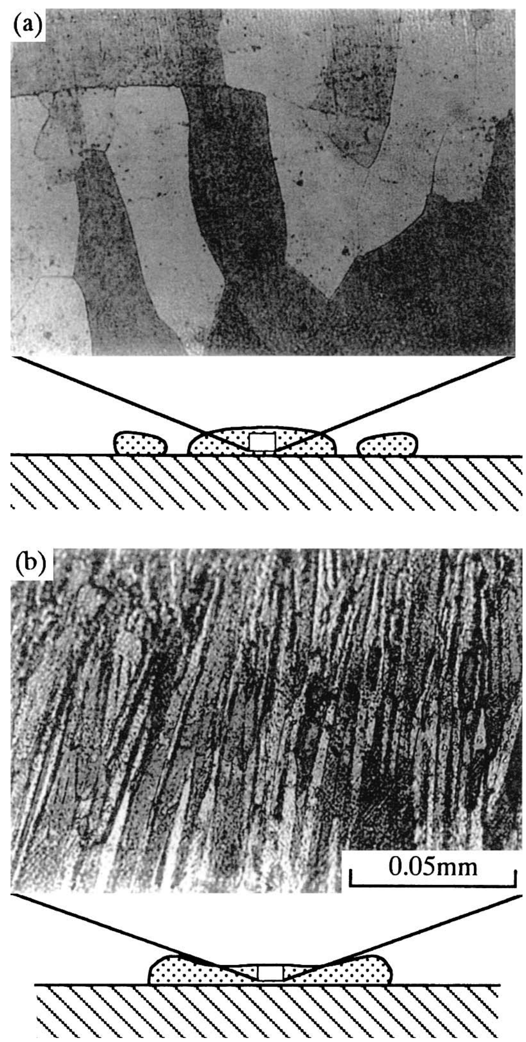


Fig. 3 Observation results of a cross-section of the microstructure of splats. **(a)** T_s = 300 K; and **(b)** T_s = 600 K

In general, the relation between the grain size, L, and a solidification rate, S, is represented as L^2S = constant.[6] In this case, the grain size, that is, the width of the column, seems to be a representative value for its microstructure. By substituting the measurement results of the grain size into the relation, it was

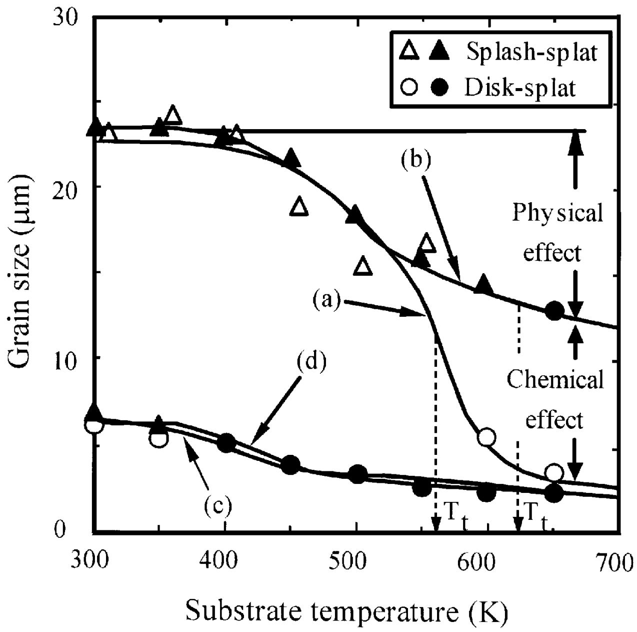


Fig. 4 Grain size in a cross-section of the splat obtained in different ambient pressures: **(a)** on stainless steel in atmospheric pressure; **(b)** on gold-coated stainless steel in atmospheric pressure; **(c)** on stainless steel under low pressure; and **(d)** on gold-coated stainless steel under low-pressure

found that the solidification rate inside of the splat obtained on the substrate at room temperature was considerably lower than that on the substrate at high temperature. According to Newton's law of cooling, the larger the temperature difference between the droplet and the substrate is, the more rapidly the splat is cooled and the smaller the grain size. The measurement result, however, showed an opposite tendency to Newton's law of cooling.

3.2 Bottom Surface Microstructures of the Splats

Bianchi et al.[7] showed that the microstructure of the splat depends on a thermal contact resistance at the splat-substrate interface. The bottom surface microstructure of the splat, which affects thermal contact resistance, was observed and is shown in Fig. 5(a). The bottom surface microstructure of the splat was observed by peeling off the splat from the substrate surface by using appropriate adhesives. The bottom surface microstructure of the splat obtained on the substrate at room temperature was porous, as is shown in Fig. 5(a-1). This porous microstructure increases the nominal thermal contact resistance by decreasing the practical contact area with the substrate and results in a poor heat transfer. In this case, the splat solidifies slowly and the grain becomes large.

On the other hand, the bottom surface microstructure on the substrate at high temperature was dense, as shown in Fig. 5(a-2). This dense microstructure decreases the nominal thermal contact resistance by increasing the practical contact area with the substrate and results in a good heat transfer. In this case, the splat solidifies rapidly and the grain becomes fine. It was found that the bottom surface microstructure, in other words, a contact condition at splat-substrate interface, was affected by the substrate temperature.

3.3 Effects of the Material Combination and Substrate Temperature on the Flattening Behavior of the Droplet

It is obvious that the contact property at the interface strongly depends on the wetting of the liquid metal to the solid substrate. The flattening phenomenon of the liquid droplet on the substrate is regarded not as a static wetting but as a dynamic wetting. Dynamic wetting is determined by two kinds of characters. One results from a material combination between the splat and the substrate, and another results from the temperature field of the substrate itself. Here, the former is defined as the chemical character of wetting, and the latter is defined as the physical character. Each character of wetting in the flattening behavior of the droplet was investigated. If the stainless steel substrate is heated, iron and chromium oxides are formed preferentially on the top surface of the substrate. Therefore, the wetting condition on the naked stainless steel substrate involves the changes of both characters simultaneously. On the other hand, only the physical character will change in the wetting condition on the gold-coated substrate, because the gold is stable and will not form any oxides, even at high temperatures. The splat obtained on the gold-coated substrate represents the physical character of wetting. The chemical character of wetting can be estimated by comparing the results of the splats obtained on the naked substrate and of those obtained on the gold-coated substrate at the same substrate temperatures.

First, the physical character of wetting was examined. The grain size of the splat obtained on the gold-coated substrate was observed and is shown in Fig. 4(b). From a comparison of the grain size on the substrate at room temperature with that on some high-temperature substrate, one can see that the grain size decreases gradually with increasing substrate temperature. This change seems to be caused by the promotion of the physical wetting. The bottom surface microstructure of the splat obtained on the gold-coated substrate is shown in Fig. 5(b). The microstructure of the splat obtained on the substrate at room temperature was porous, as is shown in Fig. 5(b-1). This microstructure is almost similar to that obtained on the naked substrate. The microstructure of the splat obtained on the substrate at high temperature was also porous, as is shown in Fig. 5(b-2).

Next, the chemical character of wetting was examined. From the comparison between the grain size of the splat obtained on the naked substrate and that obtained on the gold-coated substrate, the difference in grain size increases remarkably with increasing substrate temperature, as is shown in Fig. 4(a) and (b). The transition temperature on the gold-coated substrate is slightly higher than that on the naked substrate. This fact indicates that the molten copper contains some oxygen and that the gold coating on the substrate makes the wetting poor. Moreover, further promotion of wetting was caused by the chemical character of the naked substrate. Therefore, the drastic change of the grain size on the naked substrate with the increasing substrate temperature seems to be due to the promotion of wetting both in chemical and physical ways.

3.4 Effect of Ambient Pressure on the Flattening Behavior of the Droplet

The flattening phenomenon of the liquid droplet on the substrate is regarded as dynamic wetting. Therefore, it is believed

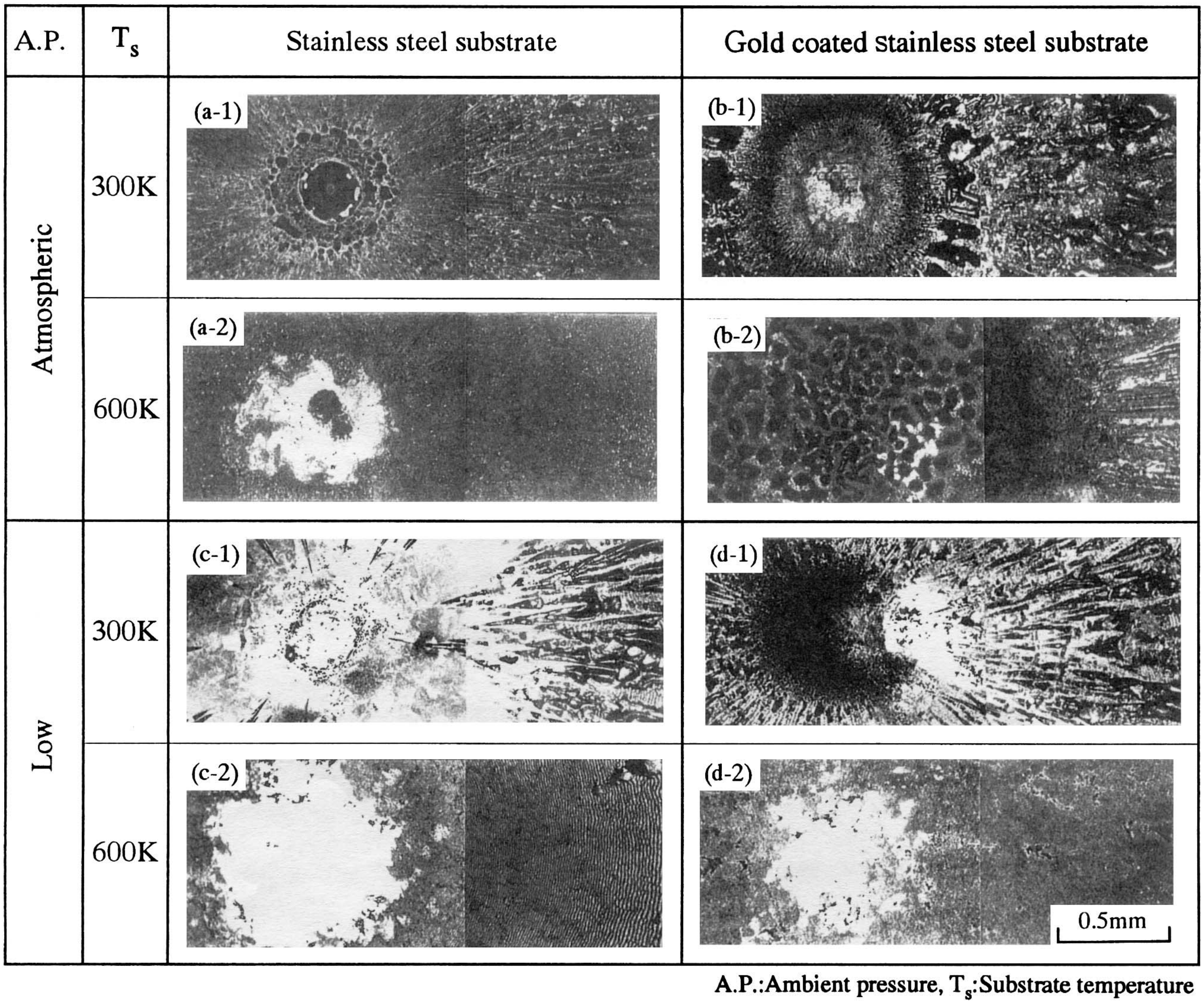


Fig. 5 Observation results of the bottom surface microstructure of splats

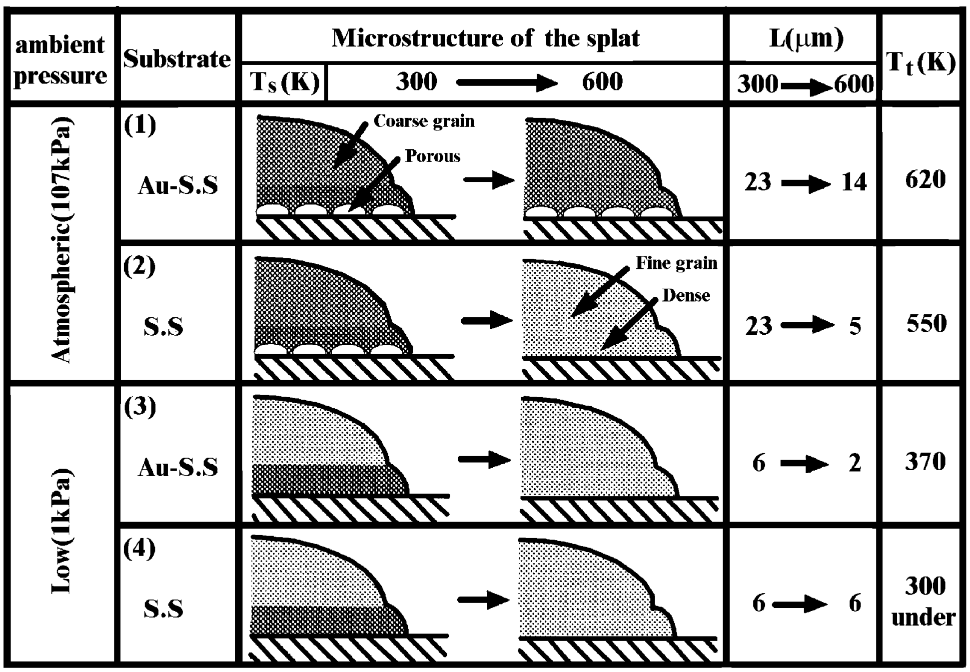
that wetting can be affected physically by the existence of a surrounding gas at the interface. That is, as a possible reason for the formation of the porous microstructure in the bottom surface of the splat, a physical intake of an ambient gas to the splat-substrate interface during the flattening is expected. The effect of the intake of the ambient gas on the flattening behavior of the droplet was investigated. In order to examine the intake behavior of the ambient gas, the splats were collected under the low-pressure condition. Under the low-pressure condition, the amount of the ambient gas is small. The grain size in the cross-section microstructures of the splats obtained under the various substrates temperatures were measured and are shown in Fig. 4(c) and (d).

Under the low-pressure condition, the grain size is remarkably smaller than that in the atmospheric pressure nitrogen condition and does not show a considerable change in size, regardless of the substrate temperature. The bottom surface microstructures of the splats under the low-pressure condition also were observed and are shown in Fig. 5(c) and (d). From these figures, the bottom surface microstructures are quite dense, regardless of the substrate temperature, and under the low-pressure condition, the microstructure of the bottom surface on the gold-coated substrate was almost similar to that on the naked substrate. This means that intimate contact between the splat and the substrate can be established under the low-pressure condition. Moreover, these results indicate that the ambient gas has a physical restraining effect on wetting by introducing the pores at the interface, and the ambient pressure seems to affect the cooling behavior of the splat through the promotion of wetting.

Consequently, the effect of the ambient pressure on wetting is remarkably stronger than the effects of the material combination and/or the substrate temperature change.

3.5 Relationship Between Wetting and Flattening of the Droplet

By accumulating the essential aspects of our research in a series of articles,[1-4] the relationship between wetting and the



S.S.: Stainless steel, T_s: Substrate temperature, L: Grain size, T_t: Transition temperature

Fig. 6 Relationship between wettability and the microstructure of the splat

flattening of the droplet has been summarized. Figure 6 shows a summary of the relationships among the ambient pressure, the surface material on the substrate, the microstructure in the splat, the grain size, and the transition temperature. In the case of the splat on the gold-coated substrate in atmospheric pressure nitrogen, as is shown in Fig. 6(1), the bottom part of the splat solidifies rapidly after impacting onto the substrate, and the porous microstructure is formed. This microstructure decreases the ability of the heat to transfer to the substrate. Then, the inside of the splat solidifies slowly, and the viscosity, which depends a great deal on the droplet temperature, does not increase so much.[4] Therefore, the droplet flattens rapidly at low viscosity. Rayleigh-Taylor instability[8] describes the unstable phenomenon occurring at the boundary between two fluids with different densities. The more rapidly the droplet flattens, the shorter the wavelength of the splat periphery is and the more complex the splat shape is. According to this instability, in this case the splash splat forms. The flattening behavior of the droplet on the substrate at room temperature in atmospheric pressure nitrogen, as is shown in Fig. 6(2), is almost similar to that mentioned above.

On the other hand, in the case of the splats both on the substrate at high temperature in atmospheric pressure nitrogen condition, as is shown in Fig. 6(2), and under the low-pressure conditions, as is shown in Fig. 6(3) and (4), the contact condition is good due to good wettability, and the formation of the porous microstructure is suppressed. Consequently, the inside of the splat solidifies quickly. In these cases, the viscosity of the droplet increases and most of the kinetic energy transfers to the viscous energy efficiently. So, the droplet flattens at lower velocity, and the formation of the disk splat results.

From the facts described above, we may conclude that a strong relationship between wetting and the cooling behavior of the splat has been recognized and that wetting has a significant role in the flattening of the droplet.

4. Conclusions

- The mean grain size of the splat obtained on the substrate at room temperature was larger than that on the substrate at high temperature. The solidification rate inside of the splat on the substrate at high temperature was higher than that on the substrate at room temperature.
- The contact condition at the splat/substrate interface is affected by the substrate temperature.
- The drastic change in the grain size on the naked substrate with the substrate temperature increasing seems to be due to the promotion of wetting in both chemical and physical ways.
- The intimate contact between the splat and the substrate can be established under the low-pressure condition, and the ambient gas has a physical restraining effect on wetting by introducing the pore at the interface.
- The effect of the ambient pressure on wetting is stronger than the effects of the material combination and/or the substrate temperature change.
- The strong relationship between wetting and the cooling behavior of the splat has been recognized, as has the fact that wetting has a significant role in the flattening of the droplet.

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