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Effect of Substrate Temperature on Deposition Behavior of Copper Particles on Substrate Surfaces in the Cold Spray Process

M. Fukumoto, H. Wada, K. Tanabe, M. Yamada, E. Yamaguchi, A. Niwa, M. Sugimoto, and M. Izawa

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The deposition behavior of sprayed individual metallic particles on the substrate surface in the cold spray process was fundamentally investigated. As a preliminary experiment, pure copper (Cu) particles were sprayed on mirror-polished stainless steel and aluminum (Al) alloy substrate surfaces. Process parameters that changed systematically were particle diameter, working gas, gas pressure, gas temperature, and substrate temperature, and the effect of these parameters on the flattening or adhesive behavior of an individual particle was precisely investigated. Deposition ratio on the substrate surface was also evaluated using these parameters. From the results obtained, it was quite noticeable that the higher substrate temperature brought about a higher deposition rate of Cu particles, even under the condition where particles were kept at room temperature. This tendency was promoted more effectively using helium instead of air or nitrogen as a working gas. Both higher velocity and temperature of the particles sprayed are the necessary conditions for the higher deposition ratio in the cold spraying. However, instead of particle heating, substrate heating may bring about the equivalent effect for particle deposition.

Keywords cold spray, deposition behavior, deposition ratio, gas pressure, gas temperature, particle size, particle velocity, substrate temperature

1. Introduction

Cold spraying has developed in this decade as a new technology for attaining high-quality coatings because of its lower heating of the powder materials (Ref 1-5). Mechanically softer materials have been effectively coated by the process, and coating properties that are superior compared with conventional coatings thermally sprayed have been indicated (Ref 6). However, the process is still in relatively new, and a fundamental aspect, the deposition mechanism, itself has not been well understood up to today. To establish the higher reliability or controllability of the cold spray process, fundamental research especially on the deposition mechanism becomes a key for the process development (Ref 7, 8). A typical characteristic of the process compared to the ordinal thermal spray process lies in that the structural component unit is a solid particle colliding on the substrate surface or prior deposited coating surface at extremely high velocity more than the so-called critical velocity. Thus, the adhesion mechanism in the cold spray process differs completely from the conventional thermal spraying; namely, it may be based on severe plastic deformation or shear instability in solid-solid plastic flow.

However, as a precise mechanism cold spraying has not yet been verified; fundamental trials to solve the problem are earnestly desired. The deposition behavior of sprayed individual metallic particles on substrate surface was fundamentally investigated in this study. Effects of both preheat treatment of particles on the deposition and postannealing of the coating on the coating property were investigated in the previous research (Ref 9, 10). However, the in situ improvement of deposition by typically increasing the substrate temperature has not yet been verified. Therefore, the effect of substrate temperature on particle deposition was precisely investigated in the present study.

2. Experimental

Cold spray equipment installations were self-designed and homemade. Basic capacities of the equipment were: spray nozzle had a 2.0 mm diam throat, a 130 mm long convergent divergent barrel nozzle, and a 4 mm diam exit;

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M. Fukumoto, H. Wada, K. Tanabe, and **M. Yamada**, Department of Production Systems Engineering, Toyohashi University of Technology, Toyohashi 441-8580, Japan; and **E. Yamaguchi**, **A. Niwa**, **M. Sugimoto**, and **M. Izawa**, Shintobrator, Ltd., Nagoya 450-0002, Japan. Contact e-mail: fukumoto@pse.tut.ac.jp.

heating temperature ranged from room temperature (RT) to 923 K, and gas pressure ranged up to 1 MPa. Powder materials used were commercially available pure copper with spherical shape and three mean diameters, namely, 5, 10.and 15 μm, respectively. Particle size distributions measured for each particle are shown in Fig. 1. Substrate materials used are AISI 304 stainless steel and aluminum (Al) alloy plates 25 by 25 by 5 mm. The particle-collecting surface was polished to a surface roughness (R_a) of 0.3 μm for each substrate plate, and they were preheated before powder collection up to a designated elevated temperature from 300 to 673 K. For the powder collection on substrate surface, substrate plates were traversed quickly at an angle of perpendicular to the cold spray torch axis to prevent temperature change of the substrate from its initial value. The deposition behavior of each particle was examined individually by scanning electron microscopy (SEM) observation of the substrate surface after deposition.

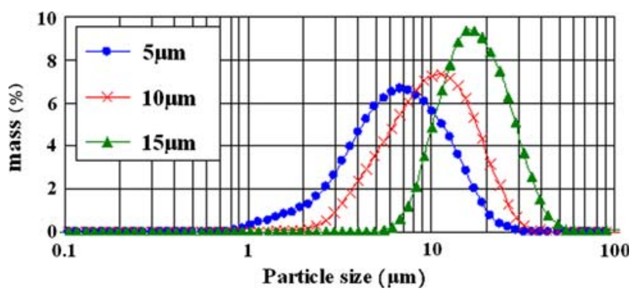

Fig. 1 Particle size distribution of Cu powders

Table 1 Cold spray conditions

Powder	Cu: 5, 10, 15 μm
Substrate	AISI 304, A6063
Substrate temperature	RT
Working gas	Air
Gas pressure	0.4-1.0 MPa
Gas temperature	523-923 K

3. Results and Discussion

3.1 Effect of Process Parameters on Deposition Behavior of Particles on RT Substrate Using Air as Working Gas

Under the various combinations of gas pressure and gas temperature, three kinds of copper (Cu) particles were collected on the substrate surface, which was kept at room temperature. Cold spray conditions used are shown in Table 1. In this experiment, compressed air was used as a working gas. First, the effect of gas pressure on the particles deposition behavior was investigated. The morphologies of collected particles for both 5 and 10 μm mean diameter on AISI 304 substrate are shown in Fig. 2. The figure shows that the collected copper particles had a hemispherical shape on the flat AISI 304 substrate surface, regardless of the mean particle size. On the other hand, lots of craters were recognized on the substrate surface. In this case, softer copper particles collided on the harder AISI 304 substrate. The increase in hardness due to the plastic deformation at extremely higher strain rate in copper particles on impact may induce the plastic deformation of harder AISI 304 substrate material. In the case of 10 μm particles as shown in Fig. 2(d) to (f), particles

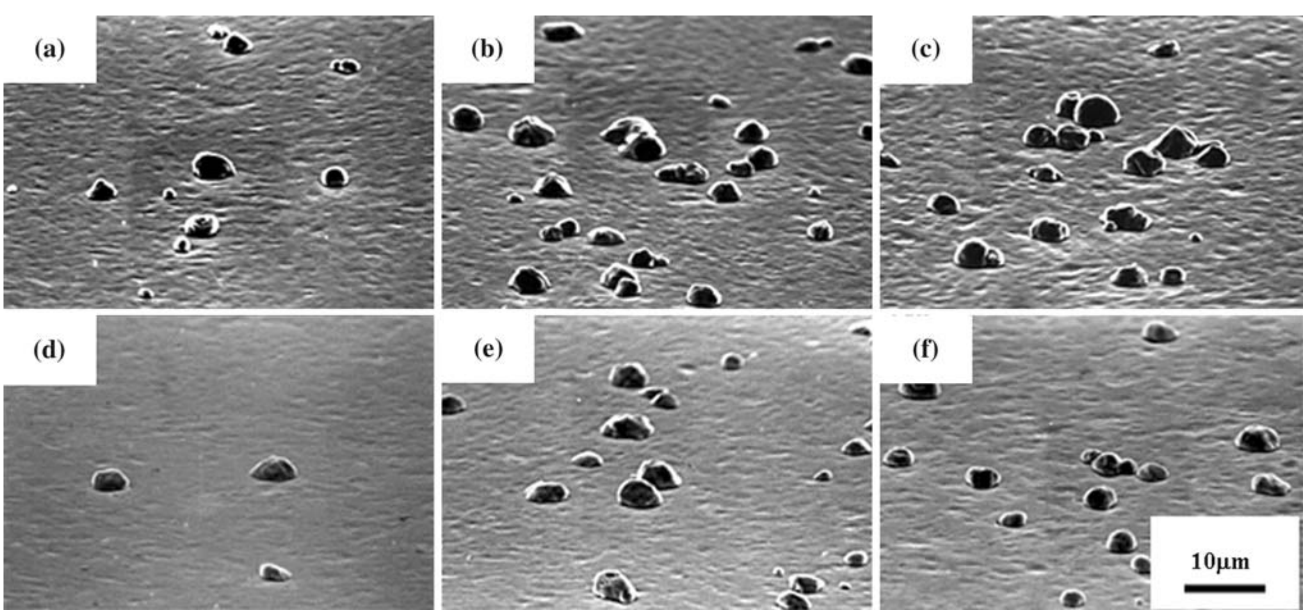

Fig. 2 Morphologies of collected particles (Gas temperature: 523 K, AISI 304 substrate). (a) Particle size: 5 μm, gas pressure: 0.4 MPa. (b) 5 μm, 0.5 MPa. (c) 5 μm, 0.6 MPa. (d) 10 μm, 0.4 MPa. (e) 10 μm, 0.5 MPa. (f) 10 μm, 0.6 MPa

larger than 5 μm could not be observed on the surface. That is, adhered particles on the substrate had an almost similar size distribution as in the case of particles with 5 μm mean size, as shown in Fig. 2(a) to (c). The result indicates that under the spraying conditions in this experiment, moderate maximum particle size may exist for the deposition. Comparison of Fig. 2(a) to (c) found that the particle deposition ratio increased remarkably with the increase of gas pressure for 5 μm particles. A similar tendency was observed in the results of 10 μm particles. This may be attributed to an increase of particle velocity caused by the increase of gas pressure.

The effect of gas temperature on particle deposition behavior was investigated. The results are shown in Fig. 3. By comparing the particle deposition behavior at the same pressure conditions, for example between Fig. 3(a) and (d), it can be said that the particle deposition ratio remarkably increased with the increase in gas temperature. Particle velocity was measured using particle imaging velocimetry (PIV) measurement equipment to find the relation between particle velocity and both gas pressure and gas temperature. The results obtained are shown in Fig. 4 and 5. Here, the standard deviation in each measurement was around 20 to 30% of each mean value. For simplicity, the expression for standard deviation was omitted from all figures in this study. Figure 4 shows that the particle velocity increased with gas pressure increasing in each gas heating condition. However, no significant increase in particle velocity with increase in gas temperature is recognized. This tendency can be similarly confirmed in the measurement results shown in Fig. 5. Figure 5 reveals that a remarkable decrease in particle velocity can be observed with an increase in particle mean diameter. The results shown in both Fig. 4 and 5 indicate that the particle velocity strongly depends not on the gas temperature, but on the gas pressure, and particle velocity is quite sensitive to its mean size. By using the findings obtained from Fig. 4 and 5, the experimental results shown in Fig. 2 and 3 can be explained as follows. That is, the particle deposition ratio can be increased remarkably mainly by the increase in gas pressure, and this may be attributed to the increase in particle velocity caused by the increase in gas pressure. However, an increase in gas temperature can also bring about a higher deposition ratio, as shown in Fig. 3. In this case, it is considered that an increase in the temperature of the particles or the

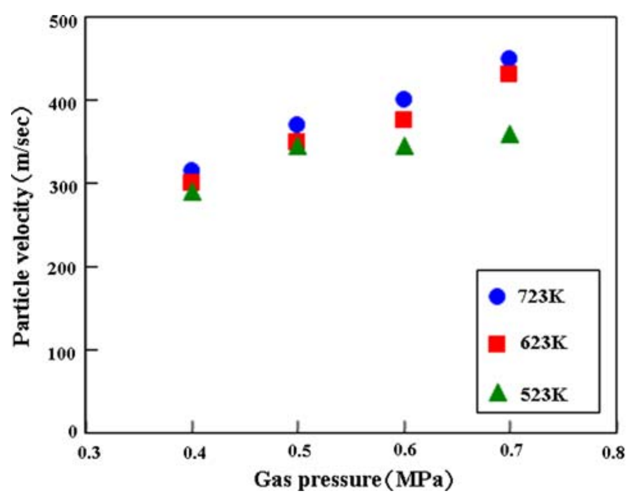

Fig. 4 Relation between particle velocity and both gas pressure and gas temperature (particle size: 5 μm)

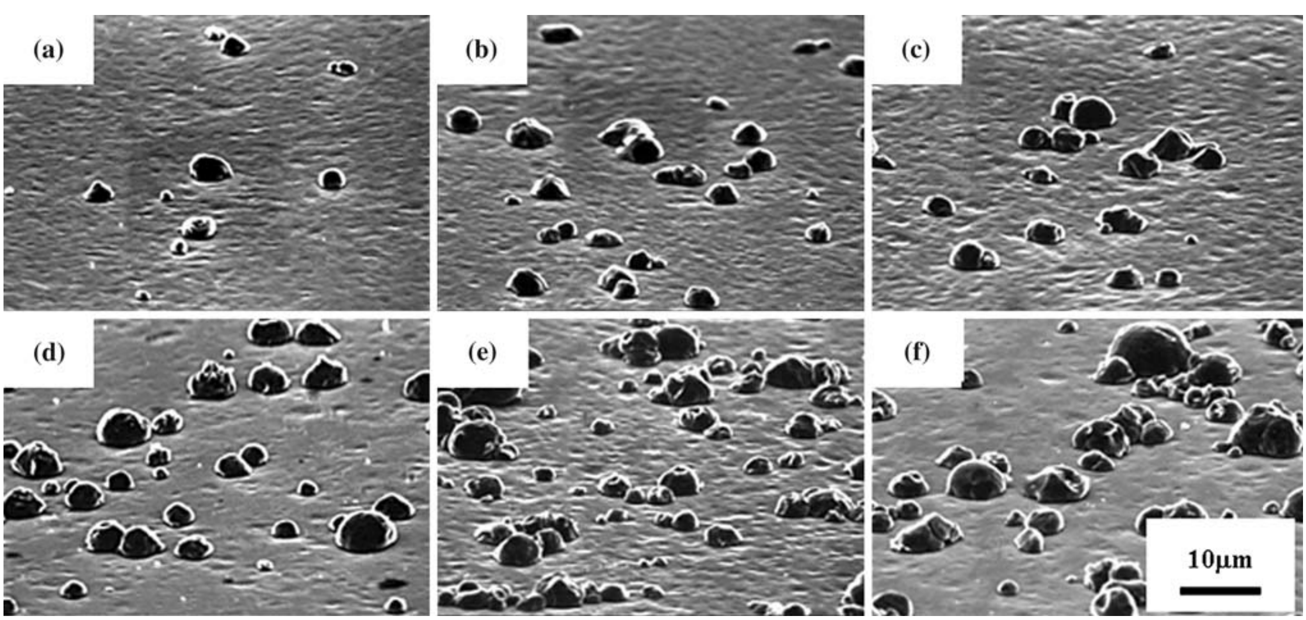

Fig. 3 Morphologies of collected particles (particle size: 5 μm, AISI 304 substrate). (a) Gas pressure: 0.4 MPa, gas temperature: 523 K. (b) 0.5 MPa, 523 K. (c) 0.6 MPa, 523 K. (d) 0.4 MPa, 923 K. (e) 0.5 MPa, 923 K. (f) 0.6 MPa, 923 K

substrate due to an increase in gas temperature might bring about the higher deposition ratio. The effect of substrate temperature on particle deposition is discussed in detail in the next section.

Particle deposition behavior on the surface of different substrate materials was observed. The results obtained for both AISI 304 and A6063 substrates are shown in Fig. 6. Compared to the AISI 304 substrate, extreme roughening was recognized on the A6063 substrate surface. This is simply attributed to the lower hardness of A6063 substrate compared to AISI 304 substrate. From the observation results on the cross-section microstructure for both substrates, it was revealed that the adhered copper particles deformed themselves on the harder AISI 304 substrate while both particles and substrate deformed each other on the softer A6063 substrate surface, which was almost the same result as reported in Ref 11. However, as already pointed out in Fig. 2, rebounding copper particles left many craters on the harder AISI 304 substrate surface. This indicates that a certain energy balance may exist between adhesion and rebounding of the sprayed particles on the substrate surface. More details on this phenomenon should be investigated in future study.

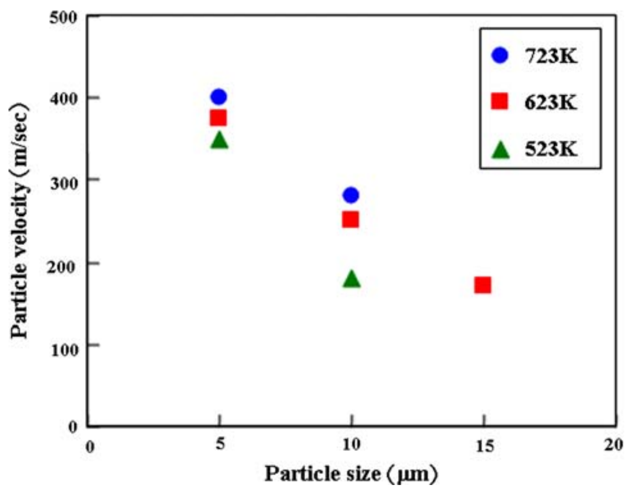

Fig. 5 Relation between particle velocity and both particle size and gas temperature (gas pressure: 0.6 MPa)

3.2 Effect of Substrate Temperature on Deposition Behavior of Copper Particles on Substrate Using Helium as Working Gas

As the significant effect of gas temperature on the deposition ratio was indicated in the experimental results in Fig. 3, the effect of substrate temperature on the deposition behavior of particles on the substrate surface was also investigated. The cold spray conditions used are shown in Table 2. Helium was used as a working gas in

Table 2 Cold spray conditions

Powder	Cu: 5, 15 μm
Substrate	AISI 304
Substrate temperature	RT-673 K
Working gas	He
Gas pressure	0.2-1.0 MPa
Gas temperature	RT, 673 K

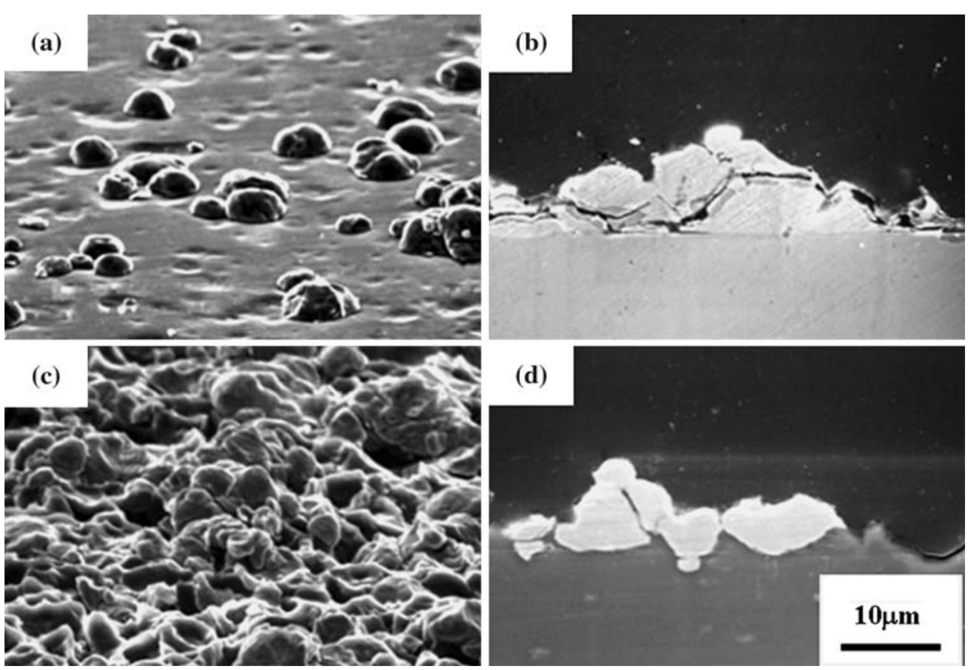

Fig. 6 Morphologies of collected particles (gas pressure: 0.7 MPa, gas temperature: 723 K, particle size: 5 μm). (a) AISI 304 substrate surface. (b) Cross section of particles on AISI 304 substrate. (c) A6063 substrate surface. (d) Cross section of particles on A6063 substrate

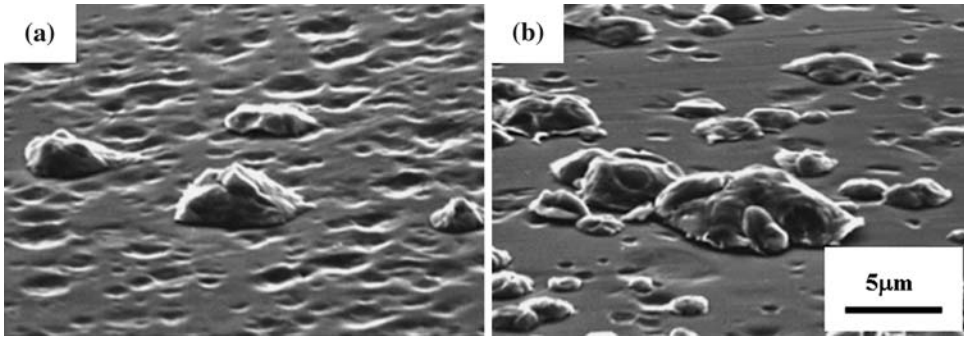

Fig. 7 Morphologies of collected particles (gas temperature: RT, substrate temperature: RT, particle size: 5 μm). (a) Gas pressure: 0.3 MPa. (b) 0.7 MPa

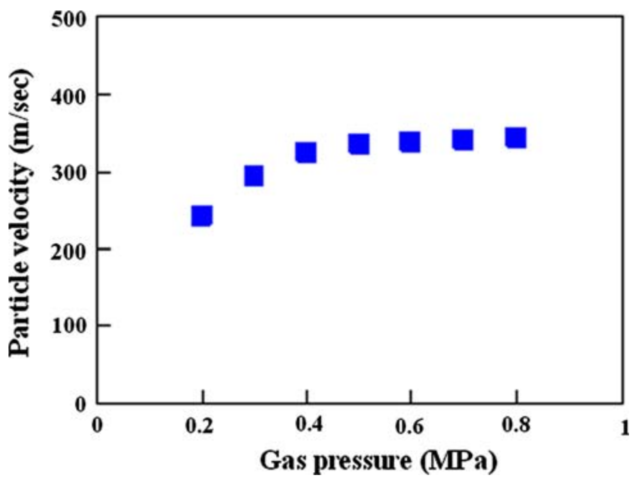

Fig. 8 Relation between particle velocity and gas pressure (gas temperature: RT, substrate temperature: RT, particle size: 5 μm)

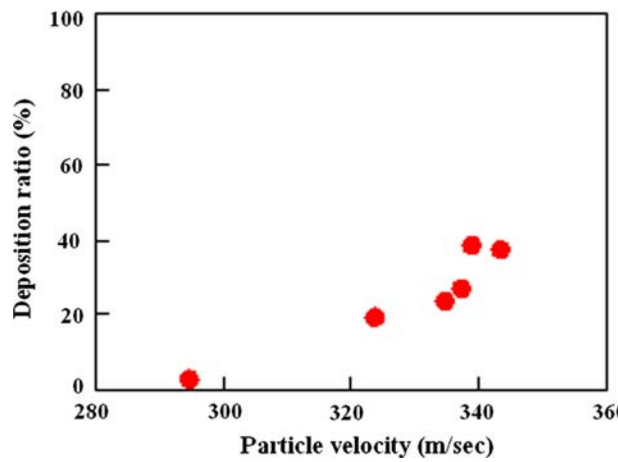

Fig. 9 Relation between deposition ratio and particle velocity (gas temperature: RT, substrate temperature: RT, particle size: 5 μm)

this experiment. As a preliminary experiment, particles were sprayed under with gas temperature at RT to confirm the significant effect of helium gas itself as a working gas. The substrate temperature was kept at RT in this experiment. The effect of gas pressure on the deposition was examined, and the results obtained are shown in Fig. 7. A comparison of Fig. 7(a) and (b) reveals that the deposition remarkably improved with the increase in gas pressure. Moreover, larger particles adhered to the substrate at higher gas pressure. The mean particle velocity change with the increase in gas pressure was measured using a DPV-2000 system (TECNAR Automation Ltd., St.-Bruno, Quebec, Canada). The measured results are shown in Fig. 8. The mean particle velocity gradually increased with increase in gas pressure; however, it almost saturated to 350 m/s over 0.5 MPa range. As the gas detachment from a nozzle wall and a corresponding saturation in particle velocity in a gas pressure range more than 0.5 MPa has been observed in our simulation, this saturation in particle velocity may be caused by the change in gas flow with the increase of gas pressure. Here, the deposition ratio was defined as a ratio of adhered particle number to the total number of adhered particles and rebounding particles (Ref 12). With data from the investigation of the deposition ratio for each gas pressure condition, the relation between deposition ratio and particles velocity is summarized in Fig. 9. The results indicate that the critical velocity in this experimental condition, that is, without heating of working gas and substrate, was found to be around 300 m/s, and an almost linear relation between particle velocity and deposition ratio was recognized in the velocity range over the critical velocity. The results indicate that the particle velocity can be a possible domination of the particle deposition in cold spray process.

The effect of gas heating on the deposition behavior of cold sprayed particles was investigated. The results obtained are shown in Fig. 10 for both 5 and 15 μm particles. The substrate temperature was RT. Most of the particles adhered onto the substrate surface under the gas heating condition typically shown in Fig. 10(b) and (d), while many rebounding traces were recognized on the substrate under the condition without gas heating as shown in Fig. 10(a) and (c). Moreover, large-size particles around 15 μm were observed on the substrate in the case of initial powder of 15 μm in the gas-heated condition as shown in Fig. 10(d). Thus, by using heated helium as a

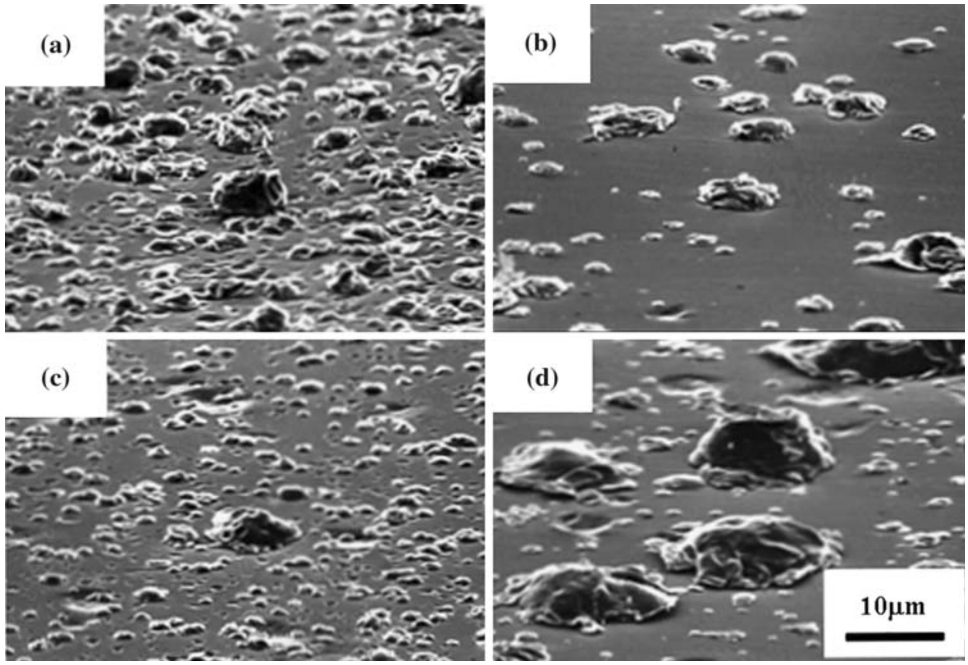

Fig. 10 Morphologies of collected particles (gas pressure: 0.8 MPa, substrate temperature: RT). (a) Particle size: 5 μm, gas temperature: RT. (b) 5 μm, 673 K. (c) 15 μm, RT. (d) 15 μm, 673 K

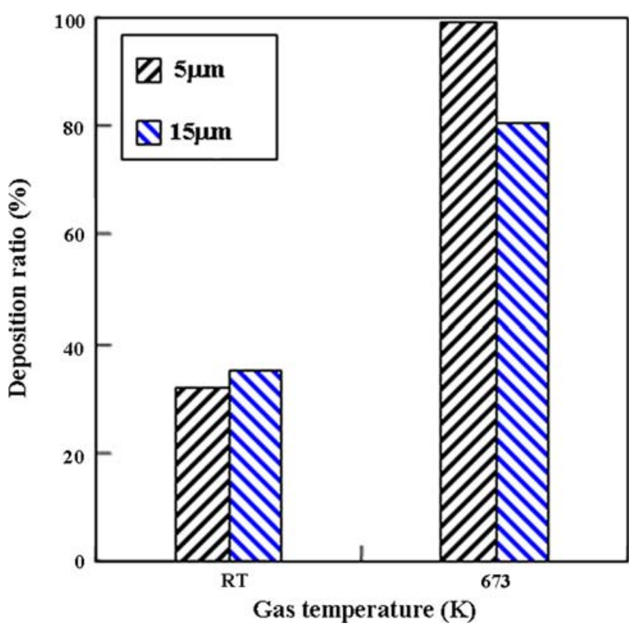

Fig. 11 Relation between deposition ratio and both gas temperature and particle size (gas pressure: 0.8 MPa, substrate temperature: RT)

working gas, higher deposition ability can be anticipated. The quantitative deposition ratio in Fig. 10 was measured, and results are shown in Fig. 11. The remarkable improvement in deposition ratio can be caused by the increase of working gas temperature, indicating that particle temperature can be another possible domination for the particle deposition.

However, it is inferred that the gas heating affects not only particle temperature but also substrate surface or particle/substrate interface temperature simultaneously in the normal situation of cold spraying. For example, it is usually observed that the deposition efficiency gradually increases with the increase in spraying time. Therefore, the effect of substrate temperature on the deposition has to be evaluated independently from the effect of particles heating. The effect of substrate temperature on the deposition behavior was investigated, and the results are shown in Fig. 12. To perform this experiment, gas temperature was kept exactly at RT to eliminate the influence of particle heating completely. It is clear from Fig. 12 that the deposition ability increases, and the number of craters decreases remarkably with the increase in substrate temperature. By considering that the particle temperature is below room temperature, it is clear that the substrate temperature plays a significant role in particle deposition in cold spraying. For further clarification, the quantitative value for deposition ratio with increase in the substrate temperature was measured. The results are shown in Fig. 13. It can be seen in Fig. 13 that the deposition ratio can be improved significantly with the substrate temperature increasing even under conditions without any heating of the working gas, in other words, without any heating of the particles. Moreover, a higher deposition ratio was given by increasing the gas pressure from 0.3 to 0.5 MPa, and it reached to almost 80%. The results show that substrate heating is quite effective for the formation of the first layer with a higher deposition ratio in the cold spray process. The finding may be quite beneficial from practical viewpoint, while

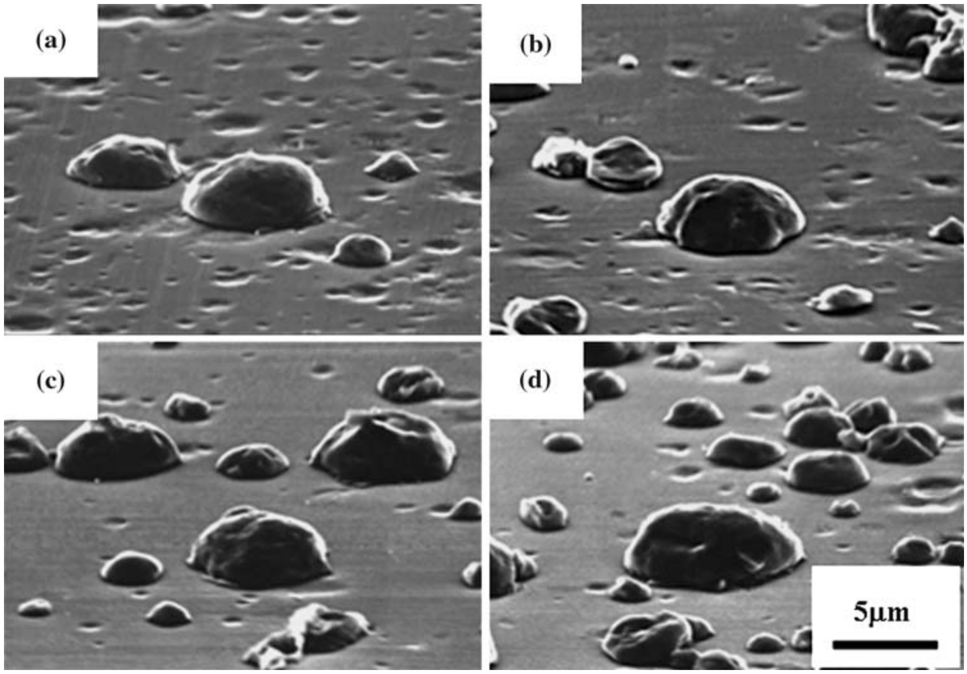

Fig. 12 Morphologies of collected particles (gas pressure: 0.3 MPa, gas temperature: RT, particle size: 5 μm). (a) Substrate temperature: 373 K. (b) 473 K. (c) 573 K. (d) 673 K

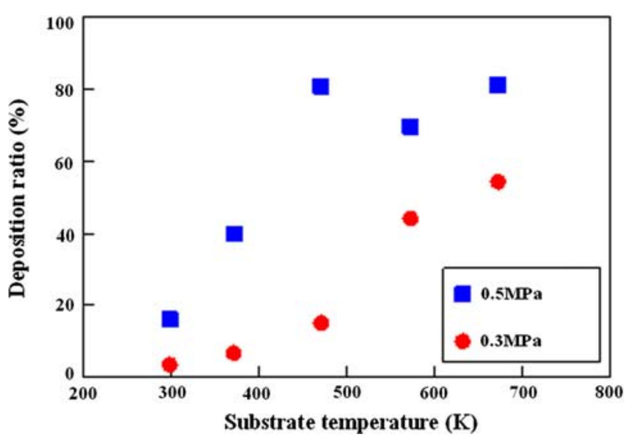

Fig. 13 Relation between deposition ratio and both substrate temperature and gas pressure (gas temperature: RT, particle size: 5 μm)

physical meaning of the higher deposition ability in this case has to be clarified in future study.

4. Summary

The relation between main process parameters and deposition behavior of cold spray process was fundamentally investigated in the present study. The results obtained are summarized as:

- The deposited copper particles showed hemispherical shape on the flat AISI 304 substrate surface regardless of the mean particle size. On the other hand, lots of craters were recognized on the substrate surface. Hardness increase caused by plastic deformation in extremely high strain rate in copper particles at impact may induce the plastic deformation of harder AISI 304 substrate material.
- The velocity of cold sprayed particles strongly depends not on the gas temperature but on the gas pressure, and particle velocity is quite sensitive to the particle mean size.
- Particle deposition ratio can be increased remarkably mainly by increasing gas pressure, and it may be attributed to the increase in particle velocity due to the increase in gas pressure.
- In the case of helium as a working gas, the particle deposition ratio can be improved significantly with substrate temperature increasing even under conditions without any heating of the working gas. The result indicates that substrate heating is quite effective for the formation of the first layer with a higher deposition ratio in the cold spray process.

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