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Investigation of Cold Spray on Polymers by Single Particle Impact Experiments

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Abstract

Cold spray has been proved to be a viable method for metallization of polymers and polymer composites. It has been reported that the mechanism of cold spray on polymeric substrates is different from the conventional mechanism on metallic substrates (i.e. adiabatic shear instability). In this work, single particle impact experiments were performed on polymeric substrates as well as mild steel. The particle-substrate interactions on different substrates were analyzed. Based on the results, the mechanism of cold spray on polymeric substrates is discussed and compared to that on metallic substrates.

Introduction

Metallization of polymers has received increasing attention in a wide range of applications during the past few decades [1]. In the early 1990s, the evolution in microelectronics industry stimulated intensive research on this topic due to the great need for miniaturization and reduction of propagation delay in devices consisting of copper and polymers [2]. For such applications, metals were usually evaporated onto polymers[2]. More recently, polymer and polymeric composites have been increasingly used as structural materials, e.g. in the aerospace industry, due to their low density and high specific strength, but the low conductivity has limited their application [1, 3]. Hence, metallization of large-scale polymer and polymeric composites have been studied, and a number of coating technologies have been investigated [1, 4]. Among these technologies, cold spray has been demonstrated to be a feasible approach to metallize polymers and polymeric composites [3, 5-9]. In particular, cold spray features low processing temperatures, which will minimize the thermal damage to the polymeric substrates.

Despite the fact that successful coatings have been produced by cold spray onto various polymeric substrates, the deposition mechanism of metals on polymers is not thoroughly understood. Conventionally, when cold spray metals onto metallic substrates, the particles are considered to be bonded by a combination of mechanical interlocking and metallurgical bonding [10-15]. It is widely accepted that the bonding is achieved through the adiabatic shear instability mechanism [10, 12]. However, when it comes to the polymeric substrates, the adiabatic shear instability mechanism may not be applicable since the polymers possess largely different mechanical properties when compared to metals; indeed, such a mechanism has barely been observed in experiments [3, 16, 17]. Moreover, the polymers can be classified into two groups, thermoplastics and thermosets, and they have different deformation responses and different thermal processing behaviors. It has been reported that cold spray onto thermoplastics is more successful than thermosets [16-18].

Single particle impact tests have been used to investigate the particle/substrate interaction upon impact. Such a test enables a clear observation of the particle deformation, e.g. splat formation, in conventional cold spray. This work aims at studying the cold spray deposition behavior of metals on polymers by doing single particle impact tests. Copper particles were sprayed onto various polymeric substrates. The interaction between the copper particles and the polymeric substrates was observed and the deposition mechanism was discussed.

Experimental methods

The substrates used in this work were commercially available ABS (acrylonitrile butadiene styrene), PEEK (polyether ether ketone), PEI (polyethylenimine), as well as CFRP (supplied by Bombardier Aerospace, Montreal, QC, Canada), and 1020 mild steel. ABS, PEEK, and PEI are all thermoplastic polymers, but have different glass transition temperatures, T_g, as well as different mechanical properties, as listed in Table 1. The CFRP used in this work has a thermosetting matrix. Mild steel was used as a benchmark substrate. Prior to the cold spray experiments, the CFRP and mild steel sections were degreased with acetone, and the ABS, PEEK and PEI sections were degreased with ethyl alcohol.

Commercial purity copper powder (Plasma Giken, Japan) was used in this work. The average particle size is 18 µm, as measured with a laser scattering particle size analyser (LA-920; HORIBA, Japan). The particle size distribution and scanning electron microscope image of the feedstock powder are shown in Fig. 1. It can be seen that the copper particles are generally spherical.

Table 1. *Some properties of the three thermoplastic materials used in this work, data summarized from [19].*

Property	ABS	PEEK	PEI
T_g (°C)	105	145	215
Hardness (M)	30-50	55-100	109-112
Tensile Strength (MPa)	30-60	70-100	90-100
ASTM D256 Impact (J/m)	100-350	80-85	50-60

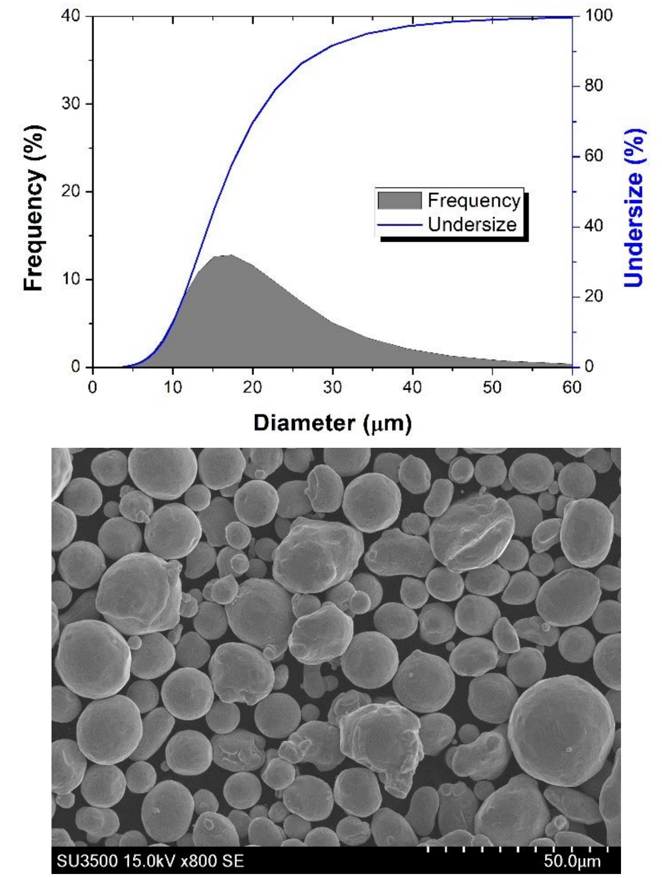

Figure 1: Particle size distribution (top) and scanning electron microscope image (bottom) of the feedstock powder.

Single particle impact cold spray experiments were carried out at the McGill-NRC cold spray facility at National Research Council Canada, Boucherville using a commercially available Plasma Giken system (PSC-800, Plasma Giken, Japan). Nitrogen was selected as the carrier gas, and the gas pressure was fixed at 2 MPa. A nozzle stand-off distance of 40 mm was used. Before cold spray, the commercially available ©Kinetic Spray Solutions (KSS) software (Kinetic Spray Solutions, Germany) was used to simulate the process and calculate the particle temperatures (details of the simulation reported in [14, 20]). Based on the simulation results, two gas temperatures were selected for each polymer substrate, with one leading to a particle temperature (for mean particle size) above T_g when reaching the substrate and the other below. For CFRP (cannot reflow when heated), gas temperatures of 425°C and 250°C were used, and 425°C was chosen for steel.

To achieve single particle impact and avoid successive impingement, a high gun travel speed of 1000 mm·s^{-1} and low powder feed setting, no higher than 0.2 RPM, were used. At such high gun travel speed, the thermal effect, i.e. heating, would be different from the previous experiments [16] in which successful coatings were obtained at much lower gun travel speed (25 mm·s^{-1}). Considering the thermal effect may be important for polymeric substrates, a preheating pass was conducted just before the single impact tests to minimize the difference in heating. Namely, the gun was operated at the set condition but without powder feeding and moved across the substrate at 25 mm·s^{-1}, followed immediately by the single impact spray as a return pass at 1000 mm·s^{-1} with powder feeding.

After the cold spray experiments, the samples were characterized with a Hitachi SU 3500 SEM in variable pressure mode (due to the low conductivity of the polymers).

Results and discussion

Simulation and gas temperature selection

The simulation results from the ©KSS software are shown in Fig. 2. The software's fluid mechanics module enables the calculation of velocities and temperatures of the gas and particle at the set process conditions (for a given nozzle and defined powder characteristics). The temperatures presented in Fig. 2 are those for the mean particle size at 40 mm from the nozzle exit (when reaching the substrate in experiments). With the calculated particle velocity and temperature, the software may further calculate the deposition efficiency (DE) by comparing the particle velocity to the critical velocity, v_{crit}, at that temperature. It can be seen from Fig. 2 that the particle temperature generally increases linearly with gas preheating temperature. On the other hand, DE increases dramatically below 350°C, from below 50% at 250°C to 89% at 350°C, but shows only a slightly increasing trend at temperatures higher than 350°C. The simulation results suggest that when spraying copper at a gas pressure of 2 MPa, successful coatings can be obtained in the gas temperature range of 250 to 550°C, although that DE varies significantly. However, it should be noted that the critical velocity concept was developed on metallic substrates by assuming that the particle and substrate materials are identical [21], the results will be different for polymeric substrates.

Nevertheless, it was assumed in this work that 250°C to 550°C is within the conventional deposition window. It was proposed in our previous paper that cold spray onto the polymeric substrate is a two-step process, the development of the first layer and the following buildup; each step has its own window and the overall window is where the two windows overlap [16]. Therefore, to develop a non-monolayer coating, in practice, the process parameters need to be selected within the buildup window (i.e. the conventional spray window); if those parameters also fall into the first window, a thick coating can be achieved. However, it is relatively difficult to practically determine the window for developing the first layer, partially due to the largely different properties of different polymers (the properties change with temperature). It was expected in this work that the single impact tests may offer insights into the particle/polymer interaction. For thermoplastics, their properties show a relatively sudden change at T_g, thus it is worthwhile comparing the impact behavior at particle temperatures above and below T_g. For PEI, PEEK and ABS, their $T_g s$ are presented in Fig. 2 by the dashed lines (top to bottom in sequence)., Gas temperatures of 550°C and 425°C were chosen for PEI, 425°C and 350°C for PEEK, and 400°C and 250°C for ABS. For CFRP, which has a thermosetting matrix, 425°C and 250°C were used, which resulted in 108°C difference in particle temperature according to the simulation.

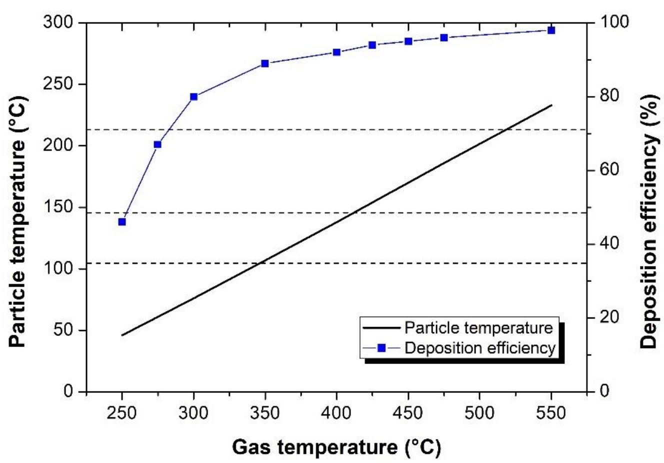

Figure 2: DE and particle temperature results as a function of gas preheating temperature, calculated by the ©KSS software.

SEM characterization

Figure 3 shows the SEM image of a copper particle cold sprayed onto the steel substrate at 425°C. The splat formation is well captured. It can be seen that the particle successfully adhered to the substrate, showing obviously flattening and significant amount of plastic deformation especially at its rim. This indicates that the adiabatic shear instability is activated and is in accordance with the simulation results (i.e. particle velocity higher than critical velocity). However, such plastic deformation and flattening were not observed on the polymeric substrates, even when spraying under the same condition.

For all the polymeric substrates, the copper particles found at the top surfaces did not show significant amounts of plastic deformation. Figure 4 presents the copper particles cold sprayed at 550°C and 425°C on PEI substrates. The former led to a particle temperature of ~235°C and the latter ~155°C. It can be seen that both particles penetrated the substrates and remained adhered. There are interaction zones in the substrate around the particle and clear signs of substrate deformation in both samples. The deformation should be caused by a combination of thermal softening (e.g. melting) and deformation under force (e.g. plastic deformation). There is no significant difference in the two samples, even though one has particle temperature above T_g and the other below. It should be pointed out that the copper particles in Fig. 4 look different from those in Fig. 1, and this may be attributed to that the SEM images of the polymeric substrates were in BSE mode (SE for feedstock powder), and it may also be caused by thermal etching.

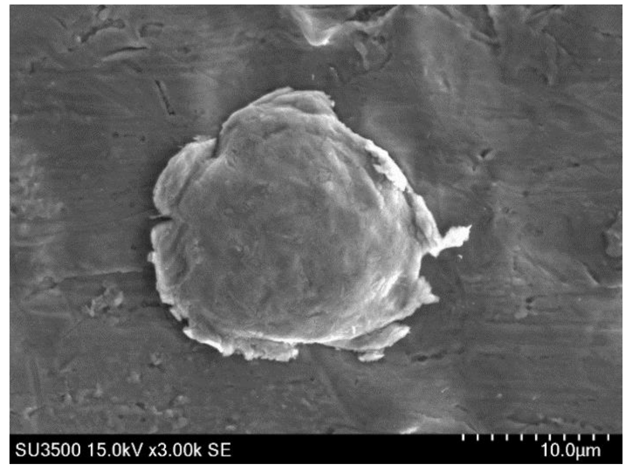

Figure 3: SEM image showing a copper particle cold sprayed onto the steel substrate at 425°C.

For PEEK cold sprayed at 425°C and 350°C, the results are shown in Fig. 5. It can be seen that numerous copper particles adhered to the substrates, with a substrate 'interaction' zone around each particle. However, there significantly fewer particles remained adhered at the lower temperature, leaving more craters in the sample. It should be mentioned that during the SEM characterization of the 350°C sample, some copper particles were swept away from the substrate by the electron beam, signifying weak adhesion to the substrate. This is understandable considering that: a) the particle temperature, 107°C, was well below T_g, and b) PEEK is a semi-crystalline thermoplastic in which the crystalline part does not gradually soften with temperature increase [22].

The SEM images showing the ABS substrates after single impact tests are presented in Fig. 6. When cold spraying at 400°C, a copper particle was found penetrated deeply into the substrate. This may be attributed to the weak resistance to heat of ABS. It is inferred that cold spray of copper at temperatures higher than 400°C can cause severe substrate erosion. Indeed, severe erosion was observed in the previous work when cold spraying copper onto ABS at 425°C [16]. At a lower temperature of 250°C, particle adherence and the associated interaction zones were observed. Since ABS possesses weak resistance to heat but high impact resistance, future work on cold spraying of metals onto ABS at relatively low temperature but high pressure is recommended.

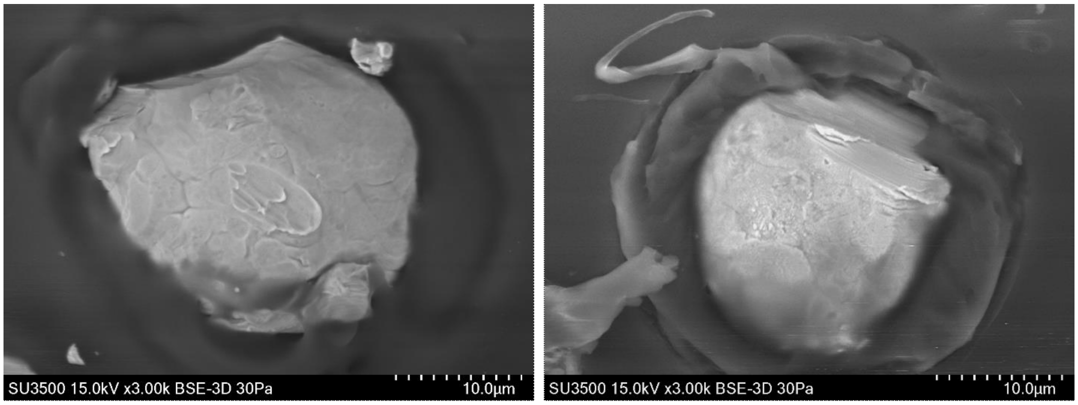

Figure 4: SEM images showing copper particles cold sprayed onto PEI at 550°C (left) and 425°C (right).

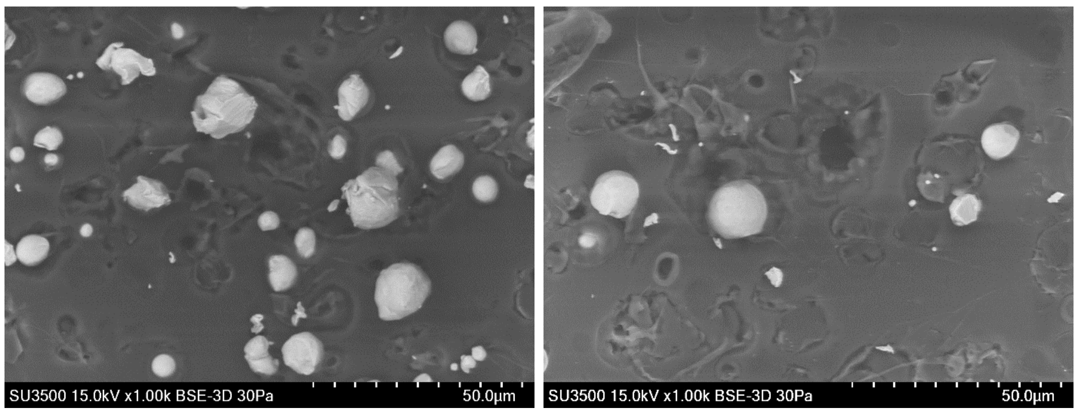

Figure 5: SEM images showing copper particles cold sprayed onto PEEK at 425°C (left) and 350°C (right).

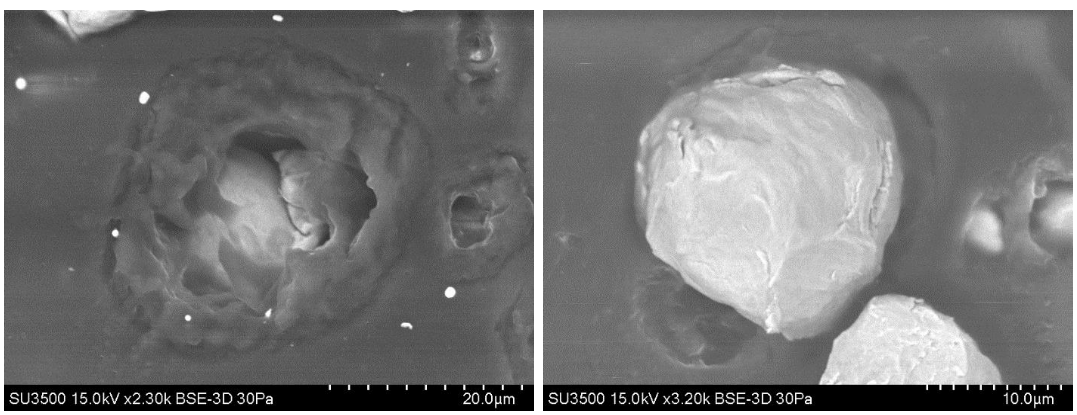

Figure 6: SEM images showing copper particles cold sprayed onto ABS at 400°C (left) and 250°C (right).

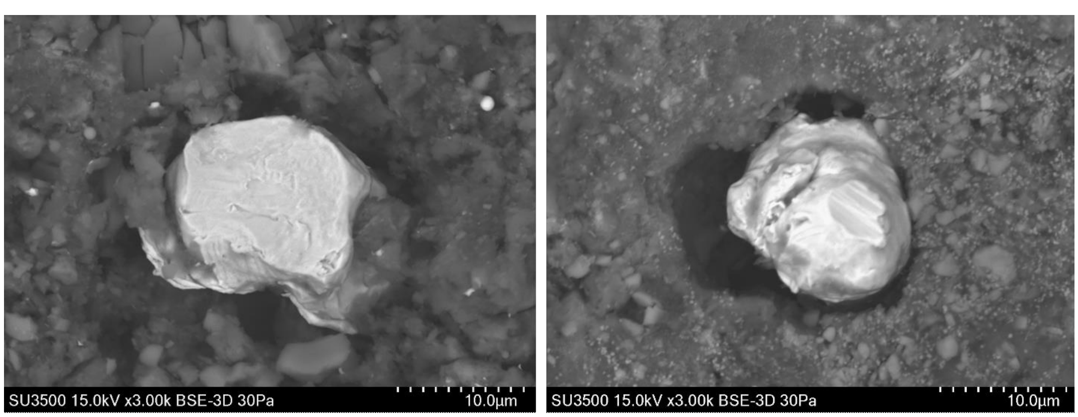

Figure 7: SEM images showing copper particles cold sprayed onto CFRP at 425°C (left) and 250°C (right).

Unlike the thermoplastics, the CFRP used in this work has a thermosetting matrix. The CFRP samples after single impact tests at 425°C and 250°C are shown in Fig. 7. It can be seen that at both temperatures, cavities were generated adjacent to the copper particle in the substrates after impact. The cavity formation should result from the brittleness of the CFRP substrate. Because of the cavity formation, the particles were not securely anchored, as can be seen from Fig. 7; it is reasonable to believe that if there were successive impacts, the particle would have been removed and enlargement of the cavity (i.e. erosion) would have occurred. The results signify that erosion of the substrate, instead of coating deposition, can occur during cold spraying of copper on CFRP at similar conditions. This is confirmed by the experimental results in [3, 16].

In general, it can be seen that the copper particles did not experience significant amounts of plastic deformation on the polymeric substrates. This indicates that the conventional adiabatic shear instability mechanism is inapplicable when developing the first metallic layer on polymeric substrates. The particles penetrated the polymers and generally remained spherical. Good particle/substrate interlocking is achieved when the substrate shows good capability of thermal deformation. This may result from the local softening, which has been observed by various researchers [16-18]. Only when good interlocking is achieved (i.e. first layer forms), can a metallic coating can be possibly deposited on the polymer after. In addition, the polymer's resistance to heat is another important factor. For polymers with weak resistance to heat, the gas temperature should be carefully selected and other process parameters should also be optimized to avoid excessive thermal effect.

Conclusions

Single particle impact tests were performed by cold spraying copper onto various polymeric substrates. The gas preheating temperatures were chosen from the conventional deposition window calculated by software simulation. Results show that the copper particle did not experience significant plastic deformation upon impact with the polymeric substrates, which are very different from mild steel. In general, the particle/substrate interlocking was achieved on all three thermoplastic polymers: the particle penetrated the substrates with an interaction zone around it. The deformation in the substrates is caused by a combination of thermal softening and deformation under force. On the other hand, cavities were formed in the CFRP substrate at the impact spots, which is a sigh of erosion, and the particles were not firmly interlocking with the substrate.

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