Study of Bonding Mechanisms in Cold Spray of Metal-to-Polymer through a Numerical Approach

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Abstract. Cold spray (CS) is a low-temperature process that can be used for the metallization of temperature-sensitive materials, such as polymers or polymer matrix composites, so coupling the lightweight of polymers with the wear resistance, physical properties and hardness of metals. The study of the cold spray of metal particles applied to polymers is still in its early stage and the deposition mechanisms underlying the process are not thoroughly understood yet. Moreover, numerical studies of cold spray of metal-to-polymer are almost completely absent in literature. Therefore, aiming to fulfill this gap of knowledge, the scope of this work is to develop a numerical FE model capable of predicting the impact and the adhesion of a micron size metallic particle onto a polymeric substrate. The results from the model were compared with the experimental outcomes found in literature to establish the effectiveness of the model that was used as a powerful tool to better understand the bonding mechanisms and all the related phenomena ruling the CS process of metal-to-polymer.

Introduction

Cold Gas Dynamic Spray, or generally referred as cold spray, is a relatively new additive coating technique developed a few decades ago in the Soviet Union by professor Papyrin [1]. It is framed in the realm of thermal spray processes for the deposition of both metallic and/or non-metallic particles on a target surface. The powders, in form of micron size particles (10-100 µm in diameter [2]), are dragged by a pressured carrier gas, typically air, nitrogen, helium, and accelerated at high velocities through a converging-diverging de Laval nozzle [3]. When the particles exit from the nozzle and impact with the target surface, conversion of kinetic energy to plastic deformation occurs, the solid particles and the substrate deform and bond together [4].

Contrary to traditional thermal spray techniques, CS minimizes the effects of oxidation, melting, evaporation and other common problems experienced with other spray processes [5]. In addition, cold spray is characterized by low toxic gas/smoke emissions, so it is in line with the European program to direct production technologies to sustainable and environmentally friendly processes [6].

All these features make cold spray a valid technique for the surface metallization of materials sensitive to high temperature, such as polymers or polymer matrix composites (PMCs), so improving the functionality of these products [7]. Several applications of polymers and PMCs are still excluded to date because of their poor electrical and thermal properties, reduced wear resistance and lightning strike protection. The surface metallization can expand their use making them furtherly attractive in industrial fields [8,9].

In the last decades, the study of metal-to-metal deposition through cold spray was extensively carried out, both experimental and numerical activities were developed to analyze the process in its wholeness [10,11]. On the contrary, the metallization of polymer-based materials is still in its early stages and the deposition mechanisms of metal-to-polymers are not thoroughly understood yet [12–14].

The experimental results found in literature show that the mechanical interlocking mechanism plays a key role for the adhesion of metallic particles on polymers, the adiabatic shear instability phenomenon underlying the CS processes of metal-to-metal was proved to be ineffective for bonding due to the different nature of the involved materials [15]. However, the experimental tests required to study in detail the particles deformation and the bonding mechanisms in CS processes are cost and time consumptions [16]; moreover, the study of the impact of particles of such small size and time scales could be very difficult, if not impossible, experimentally. In this scenario, the numerical models can bring significant insights into the study of CS process and can provide helpful tools to overcome the abovementioned experimental limitations [17,18].

In the case of metal depositions on PMCs, there are very few numerical works in literature, meaning the complexity to model the bonding under CS conditions. For instance, Chen et al. [8] developed a 2D-axisymmetric finite element (FE) model to investigate the impact deformation of Cu spherical particle on PEEK substrate. It was found from this study the key role of material jetting that surrounds the particle and promotes the bonding at higher propelling gas pressure. Tsai et al. [19] carried out similar simulations by focusing attention on material modelling and its effects on FE capability to predict the experimental results. Heydari Astaraee et al. [20] developed a 3D-FE model able to describe the behavior of both the particle and the substrate during impact, also proving that there exists a threshold value velocity (or critical velocity) above it an effective anchorage of the particle with the surface can be observed.

To the authors' best of knowledge, none of these few works studied the adhesion mechanisms between the metal particle and the polymer substrate through a detailed numerical model capable to simulate and capture the real anchorage of the particle. Therefore, based on these premises, in this work, an original FE model able to predict the bonding and the rebounding phenomena of a single particle in cold spray-based methods was proposed. The scope is to study in more details the deposition mechanisms and the complex adhesion/rebound phenomena of the particles, which govern the performances of the cold spray process of metal-to-polymer. For this purpose, a 2D-axisymmetric FE model of a copper particle impacting onto a PEEK substrate was developed. The model was validated through the literature results in terms of critical particle velocity and deformation regime. The effects of the particle impact velocity and the particle/substrate temperature were analyzed. For the first time, a cohesive behavior option was also implemented into the model to simulate the bonding between the impacting particle and the substrate under specific working conditions.

Numerical Methodology

In this section, the authors propose the FE modelling of the impact of a single copper particle on a PEEK substrate by using a commercially available FEA program; the following subsections describe the geometrical properties and the materials modelling. Particular attention was devoted at models describing the interaction between the particle and the target surface for the adhesion modelling. Note that this combination of materials was chosen due to relatively high availability of experimental data found in literature [21,22].

Geometrical properties. Both the particle and the substrate were modelled in a Lagrangian reference frame that is widely used in literature to model high-velocity impact with reduced computational costs [23–25]. A spherical particle impacting normally on a substrate at least 5 times larger than the particle was simulated by using a 2D-axisymmetric model and solving an explicit dynamic analysis including adiabatic heating effects. This size ratio between the particle and the substrate was defined taking into account that the elastic waves reflecting from the boundaries did not reach the interface during impact.

The particle diameter d_p was selected to be 18 μm. The computational domain was opportunely partitioned into several regions aiming to ensure a refined mesh with the gradual change of mesh density at the particle-substrate interface. Both the particle and the substrate were discretized by using the 4-node reduced integration elements (CAX4R). The nominal meshing size for the particle was set to $d_p/40$. The interface region in the substrate has the same meshing size of the particle in order to

ensure the solution precision. The use of the mesh size described above is justified by a sensitivity analysis on the mesh size dependency which is not reported here for the sake of brevity. However, the comparison of the maximum values of temperature and Von Mises equivalent stress and the computational costs were considered for the convergence verification. Both the bottom and the right side of the target surface were fixed in all degrees of freedom, while proper symmetry constraints were applied along the symmetric axis.

The velocity of the particle and the temperatures of the substrate and particle (assumed to be at room temperature prior to impact unless specified in the text) were imposed as initial conditions. The effects of gravity and air resistance were neglected.

Friction and plasticity-induced heating were integrated into the model. The tangential behavior was defined by a friction coefficient assuming the surface-to-surface penalty contact algorithm. A friction coefficient value of 0.35 was assigned to the whole model including the interface between the copper particle and PEEK substrate [26].

The Arbitrary Lagrangian-Eulerian (ALE) method with adaptive meshing was used to overcome the unrealistic excessive distortion of elements at high impact velocities. In particular, an "adaptive meshing frequency" equal to 5 and a "remeshing sweeps per increment" equal to 7 were chosen in this activity. The schematization and the details of the FE model developed are shown in Fig. 1.
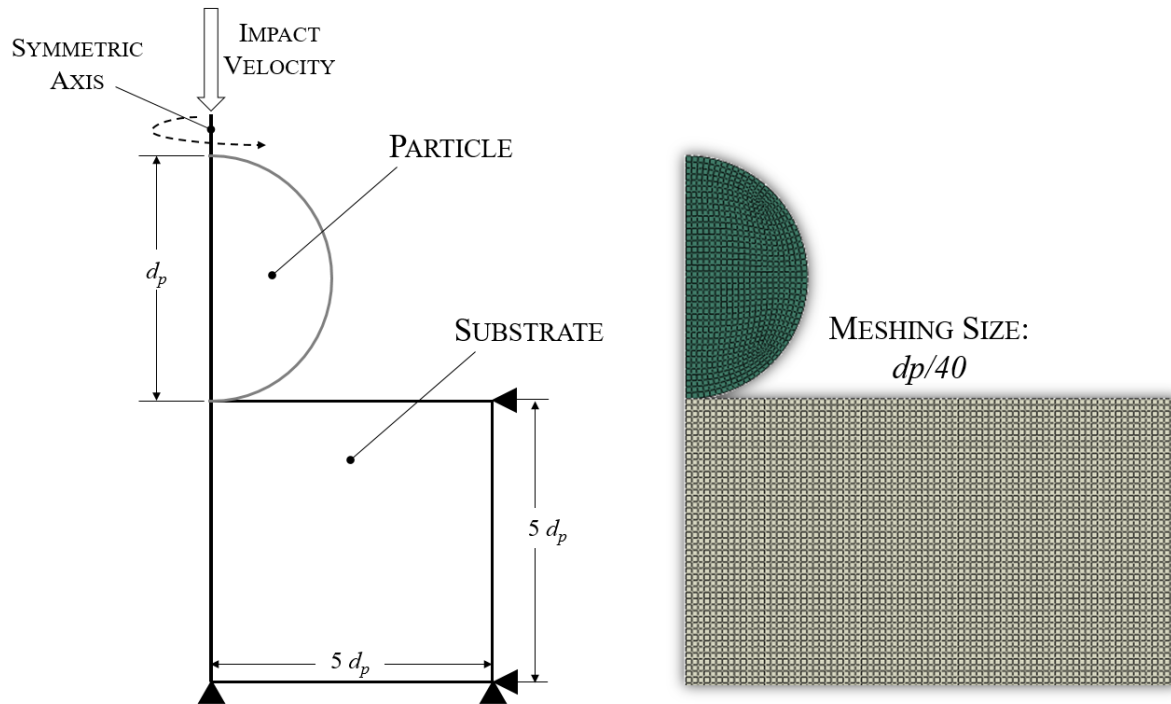

Fig. 1. Schematization and details of 2D-FE model.

Material Model. The Johnson-Cook (JC) plasticity model, which considers effects of large strains, ultra-high strain rates and thermal softening, seems to be very suitable to solve numerically engineering impact problems for its simple multiplication form [27]. This material model combination was used for both the particle and the substrate in this research activity. The stresses were expressed according to the Von Mises plasticity model. The flow stress (σ) of material is expressed as follows (Eq. 1):

$$\sigma = (A + B \cdot \varepsilon^n) \cdot \left(1 + C \cdot ln\frac{\dot{\varepsilon}}{\dot{\varepsilon_0}}\right) \cdot [1 - (T^*)^m] \tag{1}$$

where ε is the equivalent plastic strain, $\dot{\varepsilon}$ is the actual strain rate, $\dot{\varepsilon_0}$ is the reference strain rate, normally taken as 1 s^{-1} and T* is the homologous temperature defined as the following:

$$T^* = \begin{cases} 0, & for \ T < T_R \\ (T - T_R)/(T_m - T_R), & for \ T_R \leq T \leq T_m \\ 1, & for \ T > T_m \end{cases} \tag{2}$$

In Eq. 2, T is the current temperature, T_R is the reference or transition temperature and T_m is the melting point of the material. The five material constants are $A, B, n, C,$ and m. A represents the quasi-static yield strength of the material, B and n define the strain hardening behaviour, and C and m define the strain rate hardening and thermal softening behaviours, respectively. Material properties of the copper particle and the PEEK substrate used in the model are provided in Table 1 [28,29]. Young's modulus of PEEK increases vs. plastic strain and strain rate, while it decreases vs. temperature [30]. However, for the sake of simplicity, a single value was used in these analyses. Moreover, note that the glass transition temperature (T_g) of PEEK is 143 °C.

Table 1. Material parameters for copper and PEEK.

Material parameter	Copper	PEEK
Density, [kg/m^3]	8960	1300
Young's modulus, [GPa]	124	3.5
Poisson ratio	0.34	0.4
Thermal conductivity, [W/m °C]	386	0.25
Heat capacity, [J/kg·°C]	383	2180
Melting temperature, [°C]	1083	341
A, [MPa]	90	132
B, [MPa]	292	10
n	0.31	1.2
C	0.025	0.034
Reference strain rate, $\dot{\varepsilon}_0$, [s^{-1}]	1.0	0.001
Thermal exponent, m	1.09	0.634
Inelastic heat fraction	0.9	0.9
Reference temperature, T_R, [°C]	25	143

Although the Johnson-Cook model was not fully able to capture the viscoplasticity of PEEK, especially at high temperatures, it was the only available immediate approximation regarding the complexities of characterizing the viscoplastic behavior of PEEK at extremely high strain rates and temperatures encountered in CS.

The cohesive model. In all the simulations dealing with the impact of a metal particle on a polymeric substrate existing in literature for CS, the particle after impact leaves the substrate with a velocity related to its kinetic rebounding energy [8,19,20]. Aiming to overcome this limitation and model the bonding between the particle and the substrate, the surface-based cohesive behavior option was implemented into the model. This allows for the identification of the window of impact velocities and temperatures at which the particle remains firmly anchored to the substrate. The cohesive behavior function provides a simplified way to model cohesive sticky interactions between surfaces already or coming into contact.

The surface-based cohesive behaviour is a linear elastic traction-separation model with damage initiation criteria and damage evolution laws. The failure occurs if the normal stress at the interface is greater than a critical tensile stress (σ_{lim}). The damage evolution law describes the rate at which the cohesive stiffness (k_{hh}) is degraded once the above mentioned initiation criterion is reached. It is

defined on the energy that is dissipated as a result of the linear damage process, also called the fracture energy (G_c). The numerical tests were carried out by setting both the critical tensile stress and the separation δ across the interface based on the values found in literature, and varying k_{hh}, in order to calibrate the model in terms of bonding velocities compared with the experimental literature [31]. A simple sketch of the traction-separation response is shown in Fig. 2.

Summarizing, in this work, the particle after impact does not necessarily detach from the substrate, but can adhere, exactly as it happens in experimental tests of copper deposition on PEEK.
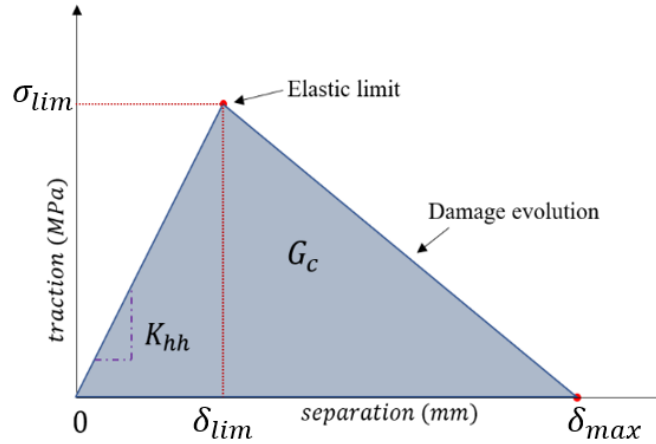

Fig. 2. Traction-separation cohesive behavior from linear elastic response to complete failure.

Results and Discussion

It is known that the kinetic energy of the particle reduces during the impact onto the target surface due to dissipation phenomena involving mainly the plastic deformation [22]. When the particle velocity goes to zero and the deformation no longer occurs, the particle recovers the elastic strain obtaining a reverse velocity, here referred as "rebound velocity", V_r. The ratio between V_R and the initial velocity, V_I, is an index of the residual energy possessed by the particle to detach from the substrate, referred below as C_R. In Fig. 3, it is reported the trend of C_R vs. V_I calculated through the model.
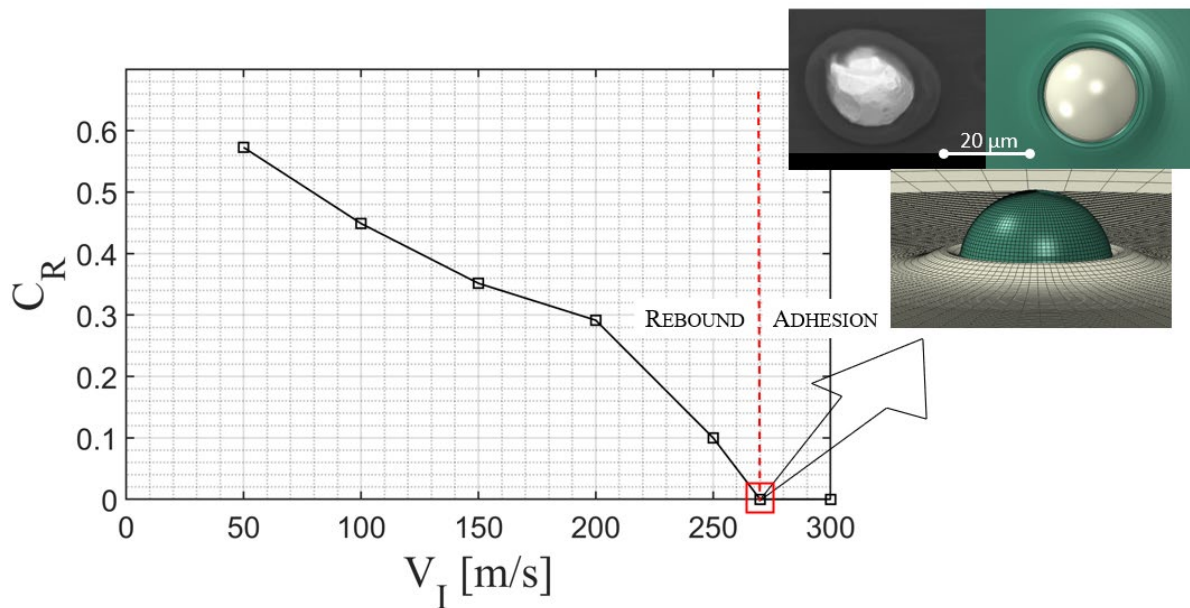

Fig. 3. Trend of C_R vs. V_I of a copper particle (d_p=18 μm) impacting onto a PEEK substrate at a predefined temperature field of 155 °C (a little above T_g).

It can be seen from the figure that C_R decreases with the increasing of the impact velocity and it goes to zero when the impact velocity is set to 270 m/s, in agreement with the experimental results found in literature [21,22]. That means the FE model proposed, for the first time in literature, is capable of predicting the adhesion velocity for CS of metal-to-polymer, with the particle that numerically bond with the target surface. It can be also seen that the particle retains its spherical shape, and remains anchored to the substrate, with an obvious substrate interaction zone around the particle. The interaction zone is similar to a wave, which is why it is called "wave effect" [21]. Based on these observations, it can be asserted that the FE model seems to produce sufficiently converged results and it can be effectively used to investigate more in details on the bonding mechanisms in CS process.

The effects of the particle temperature on C_R coefficient plotted for different impact velocities are reported in Fig. 4.
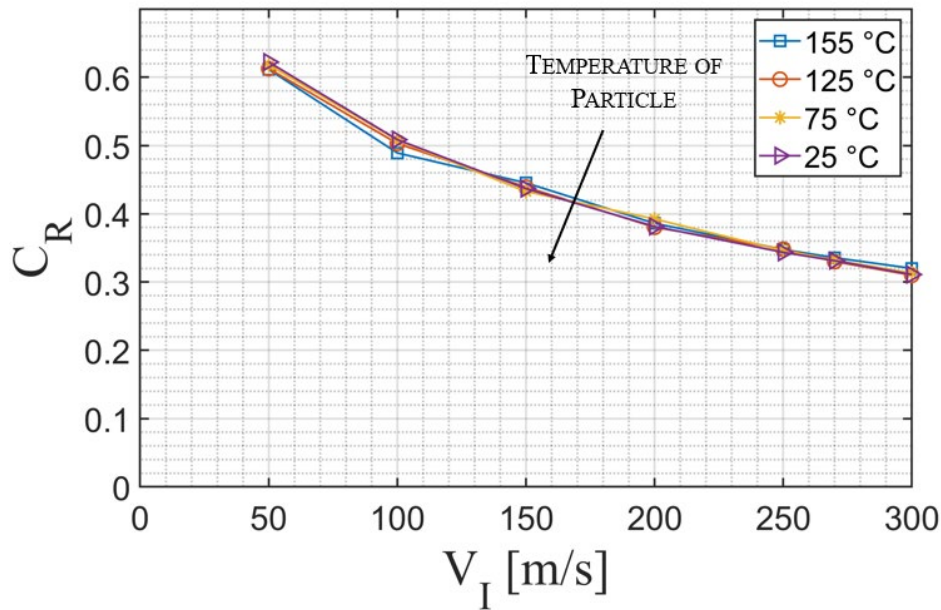

Fig. 4. Trend of C_R vs. V_I of a copper particle (d_p=18 μm) impacting onto a PEEK substrate by varying the temperature of the particle (substrate at room temperature).

It can be seen from the figure that the curves tend to overlap meaning that the temperature of the particle does not affect significantly the impact conditions between the metal and the polymer. In particular, it can be seen that the bonding does not occur and the particle is able to detach from the substrate under the investigated impact conditions. This is due to the different nature of the materials involved that do not promote the adiabatic shear instability and the metallurgical bonding [15].

Contrary to what was observed in Fig. 4, it can be seen from Fig. 5 that the temperature of the substrate seems to have a great influence on C_R characteristic ratio. It can be seen that the higher the temperature of the substrate, the lower the velocity for which the adhesion can take place. The reason is that with the increase of the temperature of the substrate (as close as possible to the glass transition temperature of PEEK), the particle tends to penetrate deeply within the polymeric material promoting the mechanical interlocking [32]. In fact, T_g marks a transition of the substrate from a brittle state to a state where it has a rubber-like behavior. Therefore, once the glass transition temperature is exceeded, the substrate softens more easily, and deforms more to accommodate the particle.

This result suggests that for the deposition of copper on PEEK, it should be helpful heating the polymer substrate to a temperature equal to or higher than the glass transition temperature, in order to have adhesion of the particles to the substrate even at low velocities.
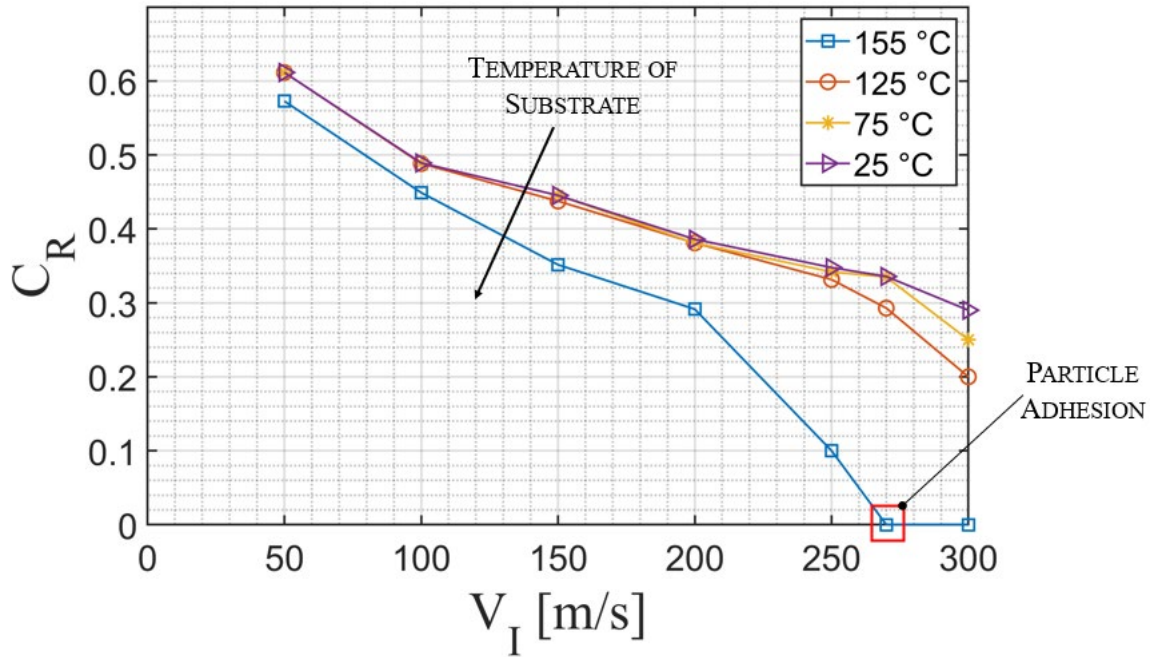
Fig. 5. Trend of C_R vs. V_I of a copper particle (d_p=18 μm) impacting onto a PEEK substrate by varying the substrate temperature (particle temperature set to 155 °C).

Finally, the results reported in Fig. 6 show that by increasing the temperature of the substrate beyond the limit of the glass transition temperature, the particle penetrates deeper and deeper into the substrate, promoting erosion and degradation, without obtaining any improvement for adhesion; in fact, it can be also seen from the figure that the particle bonding area tends to decrease with the increase of the temperature of the substrate above T_g. Note that the model is unable to return results for temperatures higher than 240 °C, due to the large deformations involved.
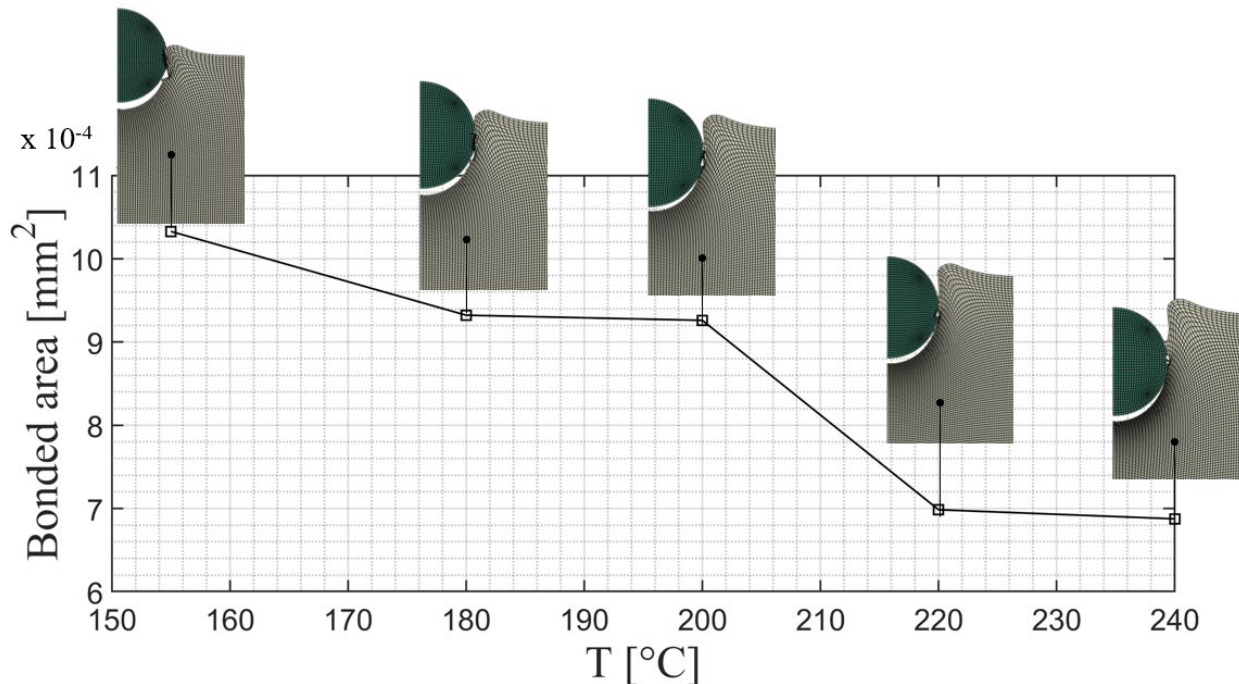

Fig. 6. Particle bonded area vs. substrate temperature above T_g (143 °C). Particle impact velocity equal to 270 m/s.

Conclusions

The aim of this work was to study the deposition mechanisms and the complex adhesion/rebound phenomena of the particles ruling the performances of the cold spray process of metal-to-polymer. For this purpose, an original FE model of a copper particle impacting onto a PEEK substrate was developed. Based on the results presented and discussed in the previous sections, the following considerations can be drawn:

- The numerical model developed was proved to be able to predict, for the first time in literature, the anchorage of the copper particle onto PEEK substrate under given experimental conditions.
- The temperature of the particle does not affect significantly the adhesion conditions due to the different nature of the involved materials.
- Heating the polymer substrate to a temperature equal to or higher than the glass transition temperature can be useful for deposition.
- Finally, increasing the temperature of the substrate beyond the limit of the glass transition temperature can promote erosion and degradation, without obtaining any improvement for adhesion.

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