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Influence of the Fusion State of Particles During Slat Formation on an Oxidized Substrate

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Achieving a good coating through thermal spray process involves understanding the mechanisms connected to the slat formation. The main objective of this paper is to analyze how the substrate is affected by the thickness of the oxide layer. The next thing to think about is how melting aluminum particles will react with an oxidized substrate. To this end, the fluid structure interaction technique (IFS), the finite element method and the volume of fluid method (VOF) are used to simulate the deposition of particles (molten or semi-molten) onto the part to be covered. The findings reveal that the percentage of the fusion state present in the particle acts directly on the morphology of the splat.
Keywords: structure fluid interaction (IFS), finite element, volume of fluid method (VOF), oxide layer, spreading, solidification.

1. Introduction

Thermal spray coating technology is extensively used in industries, such as aerospace, automotive, oil/gas industry and renewable Sector. The principle of this process involves the ejection of molten or semi molten droplet onto a previously prepared substrate. When the droplet contacts the substrate, the spreading process begins followed by the cooling and solidification of this droplet. Nevertheless, as the droplet impact the solid substrate, it exerts forces on it, leading to deformation and stress in the structure of the substrate. Improving the performance of coatings in thermal spraying requires understanding the dynamics of droplet impact and its interaction with the substrate.

There have been numerous researchers who have made significant contributions to the field of fluid-structure interaction (FSI).

Using numerical simulations and models to study the impact process of molten metal droplets on solid surface, with a specific focus on the interfacial thermal contact resistance. A three-dimensional time-dependent numerical model of free-surface flows and heat transfer was integrated with the analytical model. It was found that the existence of surface oxide or other impurities are likely to cause additional variations in contact resistance, and these changes cannot be anticipated solely based on surface roughness (Xue et al., 2007). The Galerkin finite element method and the VOF method in Ansys/Flotran code was used to simulate the behavior of partially molten aluminium particle during thermal spray process. The outcomes reveal that the partially molten particle does not totally flatten on the solid substrate (small contact area). Yet, at the impact point it exercises a high impact pressure which leads to better substrate/ splat contact which improves the adherence of the coating. And with the increase of the molten fraction of the particle the spreading increases (great contact area) (Zirari et al., 2010).

Another work was done by Franci et al. (2017), suggested a focus on simulating and analyzing the behavior of nuclear reactor cores under extreme conditions, such as accidents or meltdowns, using the Particle Finite Element Method (PFEM). Their study demonstrates the effectiveness of the suggested formulation to reproduce the structural failure mechanisms triggered by corium, which could manifest during a meltdown scenario. Although, Tahar et al. (2018) studied the impact and spreading of Nickel droplet on rough stainless-steel substrate with changing the temperature of the substrate. In their study they use the finite element method and the fluid volume method (VOF) to simulate a two-dimensional model of the droplet impact. Their results indicate that, a good spreading of the droplet is obtained for the round form for both cold and hot substrate, resulting in uniform solidified layer which is desirable in coating. Even so, the preheated substrate reduced splashing, it produces an oxide coating on its surface.

A novel 3D lattice Boltzmann method (LBM) was proposed for simulating a molten droplet impacting and solidifying on a cold smooth substrate surrounded by air. The entrapment of air within the molten droplet during impact creates a thermal resistance between the droplet and the substrate. It is demonstrated that the air on the solid surface can not be entirely ejected due to the no-slip velocity, and the air space between the droplet and the substrate is compressed by the falling droplet. To better understand how microgravity affects the deposition of droplets on vertical substrates and how it differs from the deposition on a horizontal substrate (Xiong & Cheng, 2018).

The study of the dynamics of particles impact onto a solid surface was conducted by Liang et. (2019), focusing mainly the behavior of the liquid when it forms a "splat" upon impact. Results show that increasing Weber number decreases the wave propagation slightly, but this effect becomes less significant at higher Weber number. Shena et al. (2019), developed a numerical model for examining the spreading and solidification dynamics of a Yttria-Stabilized Zirconia (YSZ) droplet with high temperature and high velocity, upon the impact on a preheated substrate, under the conditions typical of supersonic plasma thermal spraying processes.The results showed that, although the existence of a large thermal gradient between the droplet and the preheated substrate, the solidification is assumed to take place mostly after spreading. It was also found that, the higher the impacting velocity, the larger the maximum spread, and the lower thermal contact resistance which leads to more strong contact.

The behavior of metal droplets on vertical substrates was discussed by Huang et al. (2020), in the context of 3D printing technology under microgravity conditions. It was found that the asymmetrical spreading, retraction and solidification shape of the droplet originate from the interaction of gravity (the droplet tilts to gravity direction). And this undesired solidification shape would be effectively suppressed with larger impact inertia or slower solidification rate.

In order to investigate the effect of the impact dynamics during splat formation Mekhtiche et al. (2021) conducted a simulation of aluminum particles impinging at different impact velocities. The findings show that an impact velocity lower than the reference speed does not allow the formation of a lamella, whereas for a higher speed the lamella splashes partially or totally proportional to the speed of impact. In additionally, significant impact velocity could cause deformations that propagate along the interface of the lamella/substrate.

In another work of Tahar et al. (2022) conducted a numerical simulation of the successive impact of aluminium droplet on an oxidized substrate using the finite element method. It was found that, the thickness of the oxide layer increases with the substrate temperature. This oxide layer can act as a protective layer against corrosion, it prevents direct contact with the substrate and the corrosive substances in the environment, resulting in an increase in the durability of the coating.

For the purpose of better understanding the interaction between the particle and the substrate, an oxide-coated H13 tool steel substrate is suggested for the investigation of the effects of a partially melted and molten alumina particle. employing the technique of fluid- structure interaction.

2. Mathematical Formulation

Three main sorts of interactions occur when a particle is injected into a plasma: Plasma-Particle momentum, heat and mass transfer.

Consider a solid particle of radius R. The temperature distribution in the sphere can be described by the equation of continuity of the thermal energy flow, adapted to its spherical geometry (Vardelle et al, 1983):

$$\rho_p \, C_p \, \frac{\partial T}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \, K_p \, \frac{\partial T}{\partial r} \right) \quad (1)$$

Consider the following boundary conditions:

At the center of the particle (r = 0):

$$\frac{\partial T}{\partial r} = 0 \quad (2)$$

While, on the surface (r = R), heat transfer takes place by convection and radiation

(Abdellah El-Hadj & Ait-Messaoudene, 2005):

$$K_p \frac{\partial T}{\partial r} = h.(T_\infty - T_P) + \delta (T_\alpha^4 - T_P^4) \quad (3)$$

K_p, the thermal conductivity of the particle;

h, the convection coefficient is calculated from the Nusselt number (Bounder et al, 2003)

$$Nu = \frac{h\, d_p}{\overline{K_g}} = 2.0 + 0.514\, Re^{0.5} \quad (4)$$

Where $\overline{K_g}$ is the plasma conductivity evaluated as an integral average value as proposed by Ben ettouil et al. (2007):

$$\overline{K_g}(T) = \frac{1}{T - T_a} \int_{T_a}^{T} K_g(\theta)\, d\theta \quad (5)$$

The basic equation for the motion of a particle injected into a thermal projection system is a representation of the balance of the force acting on it. The equation governing the transfer of momentum between a spherical particle with diameter d_p and the gas mixture, considering the effect of the substrate's presence, can be expressed in the following form (Lugscheider, 2000; Carslaw, 1959):

$$\rho_P \frac{dv_P}{dt} = F_D + F_{th} \quad (6)$$

$$\frac{12\,\pi\,.\,\mu_\infty\,.\,v_\infty\,.\frac{d_p}{2}\,.\frac{C_S}{T_\infty}\,\overrightarrow{\mathrm{grad}}T}{\left(1 + 6\,C_m.\lambda/d_p\right)\left(1 + \frac{2\,k_\infty}{k_p} + 4\,C_t\,.\lambda/d_p\right)} \quad (7)$$

With F_{th} and F_D are the thermal force and the drag force respectively, given as follows: $\vec{F}_{th}$ =

$$\vec{F}_D = -\frac{1}{2}\, C_D\,.\pi\, \frac{d_P^2}{4}\,.\,\rho_\infty\,.|V_\infty - V_P|\,.(V_\infty - V_P) \quad (8)$$

The drag coefficient C_D is calculated and presented by the following equation:

$$C_D = \frac{24}{Re_p} + Re_p \quad (9)$$

The effects of stress and deformation are denoted as σe and εe, respectively.

Vector stress tensor:

$$T_\sigma = \begin{bmatrix} \sigma_e & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} \quad (10)$$

The deviations of effective and multi-axial stresses are matched, without taking into account hydro-static pressure.

$$D_\varepsilon = \begin{bmatrix} \sigma_\varepsilon^{-\frac{\sigma_\varepsilon}{3}} & 0 & 0 \\ 0 & \frac{\sigma_\varepsilon}{3} & 0 \\ 0 & 0 & -\frac{\sigma_\varepsilon}{3} \end{bmatrix} = \begin{bmatrix} \frac{2\sigma_1 - \sigma_2 - \sigma_3}{3} & 0 & 0 \\ 0 & \frac{2\sigma_1 - \sigma_2 - \sigma_3}{3} & 0 \\ 0 & 0 & -\frac{2\sigma_1 - \sigma_2 - \sigma_3}{3} \end{bmatrix} \quad (11)$$

By assimilating the second invariant of the respective deviators, the following is the effective stress expression written:

$$\sigma_e = \sqrt{\frac{1}{2}[(\sigma_1 - \sigma_2)^2 + (\sigma_2 - \sigma_3)^2 + (\sigma_3 - \sigma_1)^2]} \quad (12)$$

The effective strain has the following three principal components:

$$\varepsilon_1 = \frac{1}{E}\, \sigma_e = \varepsilon_e \quad (13)$$

$$\varepsilon_2 = \frac{1}{E}\, u\, \sigma_e = -u\, \varepsilon_e \quad (14)$$

$$\varepsilon_3 = \frac{1}{E}\, u\, \sigma_e = -u\, \varepsilon_e \quad (15)$$

Therefore, the effective deformation tensor is equal to:

$$T_\varepsilon = \begin{bmatrix} \varepsilon_e & 0 & 0 \\ 0 & -u\varepsilon_e & 0 \\ 0 & 0 & -u\varepsilon_e \end{bmatrix} \quad (16)$$

Repeating the procedure used for the effective stress, there is a match between the multi-axial and effective deviator:

By assimilating the second invariant of the respective deviators, the deformation expression is written:

$$\varepsilon_e = \frac{1}{(1+u)\sqrt{2}} \sqrt{(\varepsilon_1 - \varepsilon_2)^2 + (\varepsilon_2 - \varepsilon_3)^2 + (\varepsilon_3 - \varepsilon_1)^2} \quad (18)$$

3. Results and Discussion

This study focuses on simulating the impact of aluminum particles on a steel substrate.

Table 1 provides the thermo-physical parameters of the materials.

Table 1(Adapted from article of (Xue et al, 2007)

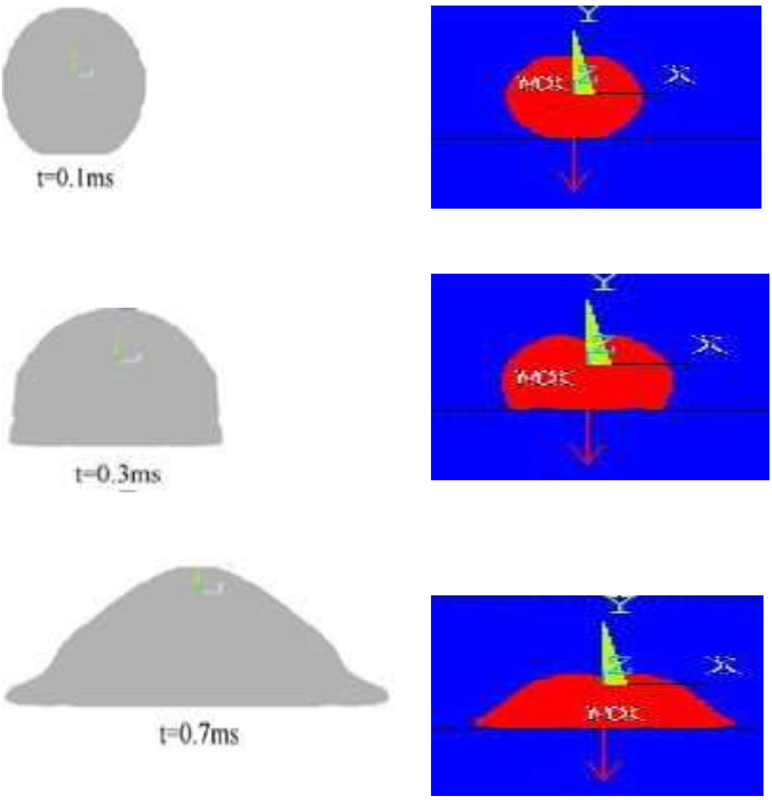
Properties of the materials used

Properties	Particle: Alumina	Substrate: Steel H13	Fe2O3 Oxide Layer	unity
Density	2570	7800	5240	(Kg/m³)
Viscosity (liquid)	°c 0 4.5 E-7			(m²/s)
	700 4 E-7			
Thermal conductivity	70	26	2.9	(W/m K)
Liquid specific heat	1000		670	(J/Kg K)

Solid thermal conductivity	°c 400 144 500 147 630 70			(W/m K)
Specific heat	°c 100 400 300 500 400 800 570 1050 580 1038	°c 20 460 200 502 500 550		(J/Kg K)

For identification purposes, the results were compared with those from simulations conducted by Xue et al. (2007), under the same conditions. Figure 1 illustrates the simulation results of the impact of a completely molten aluminum particle on a stainless-steel substrate. Figure 2 compares the spread factors in the present work with those of Xue et al. (2007).

The numerical results indicate that the droplet began to propagate at 0.1 ms. The spreading and shape of the drop are similar during the impact. The convex form gradually reduces its incline over time as it slides slowly across the substrate, eventually achieving complete spreading. At t = 7 ms, the droplet stabilizes and takes the final shape of the splat. A crucial detail to note is that the liquid adhered to the surface topography.



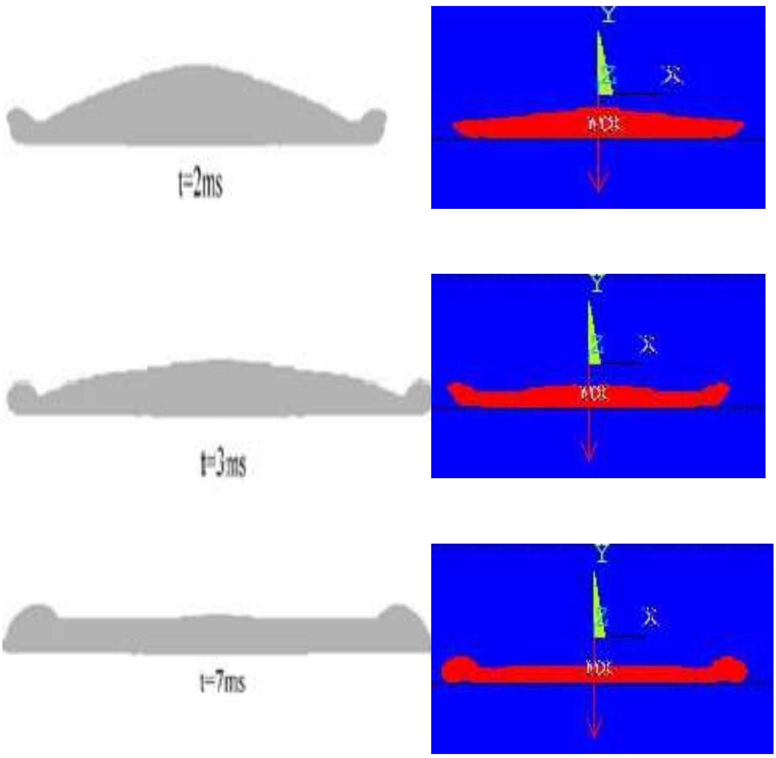


Figure 1 Impact of an aluminum particle on a steel substrate

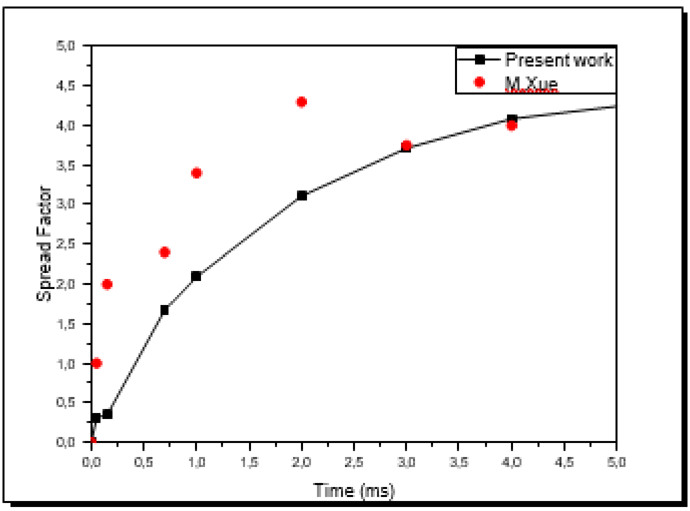


Figure 2 Comparison between spreading factors under the same conditions

The simulation of the impact of a completely molten aluminum particle with a diameter of 3.92 mm, initially at a temperature of 630°C and a velocity of 3 m/s, on a steel substrate initially at 200°C was studied. The formation of an oxide layer (Fe2O3) was taken into account (Figure 3). The oxide layer can lead to an increase in contact resistance.

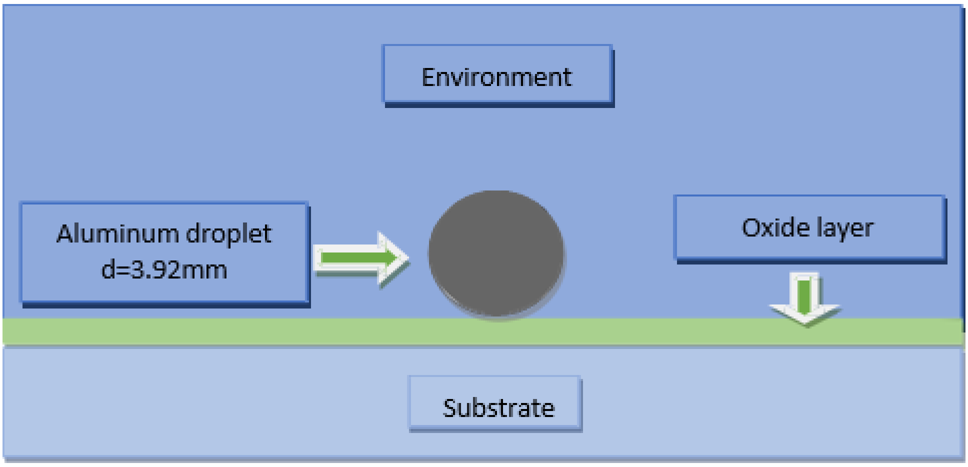


Figure 3 The impact of a particle on the substrate with the presence of an oxidation layer

Figure 4 represents a comparison between the spread factors for four cases of oxide layer thickness (0 mm, 0.1 mm, 0.3 mm, and 0.5 mm). The numerical results exhibit a high degree of similarity for all the cases. At t = 0.1 ms, the droplet spreads quickly, and by t = 7 ms, the droplet reaches maximum spreading and forms the lamella.

Table 2 indicates the temperature distribution during the impact of a droplet onto a steel substrate in the presence of an oxide layer for all the cases (0.1 mm, 0.3 mm, and 0.5 mm). The results show that the oxide layer can act as a protective barrier, reducing heat transfer between the particle and the substrate. Additionally, there is a delay in cooling due to the oxide layer, extending the duration of higher temperatures within the aluminum particle. The shape of the droplet is similar during the impact for all four cases, and the "fingers" situated at the edges of the formed lamella solidify at a later stage, depositing onto the rough surfaces of the substrate.

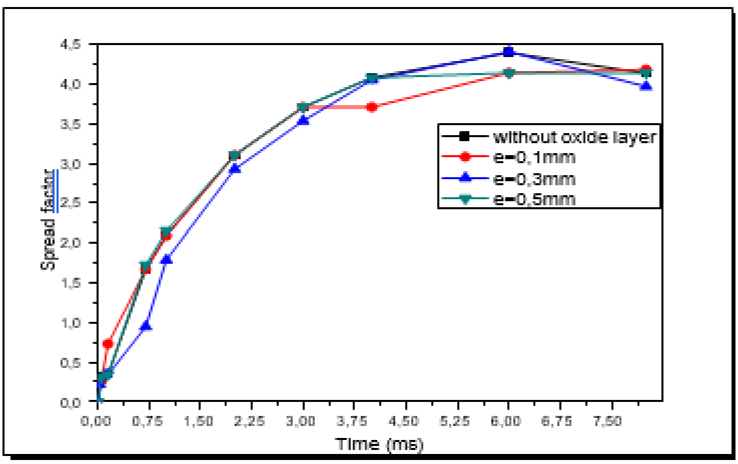


Figure 4 Comparison between the spread factors for the four cases (Without oxide layer, 0.1mm, 0.3mm and 0.5mm)

Table 2 Spatial representation of the cooling of the particle during the formation of the Lamellae

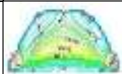
	Without oxide layer	0.1mm	0.3mm	0.5mm
0.05				
0.15				
0.7				
3				
8				

Figure 5 presents a comparison of the temperature profiles over time for the four cases at the impact. As expected, at the initial moments following the contact of the droplet with the substrate, the temperature of the substrate increases rapidly due to the high initial temperature of the molten aluminum droplet. As the droplet partially spreads, the contact surface area between the substrate and the droplet increases, facilitating more efficient heat transfer. Consequently, the substrate's temperature stabilizes and remains nearly constant during the subsequent spreading phase.

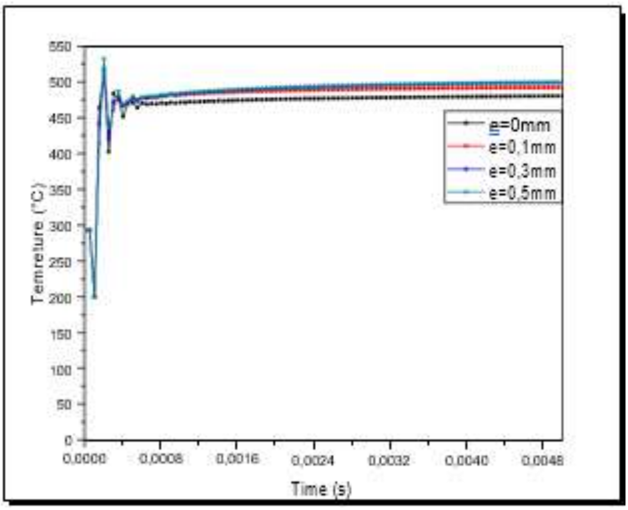
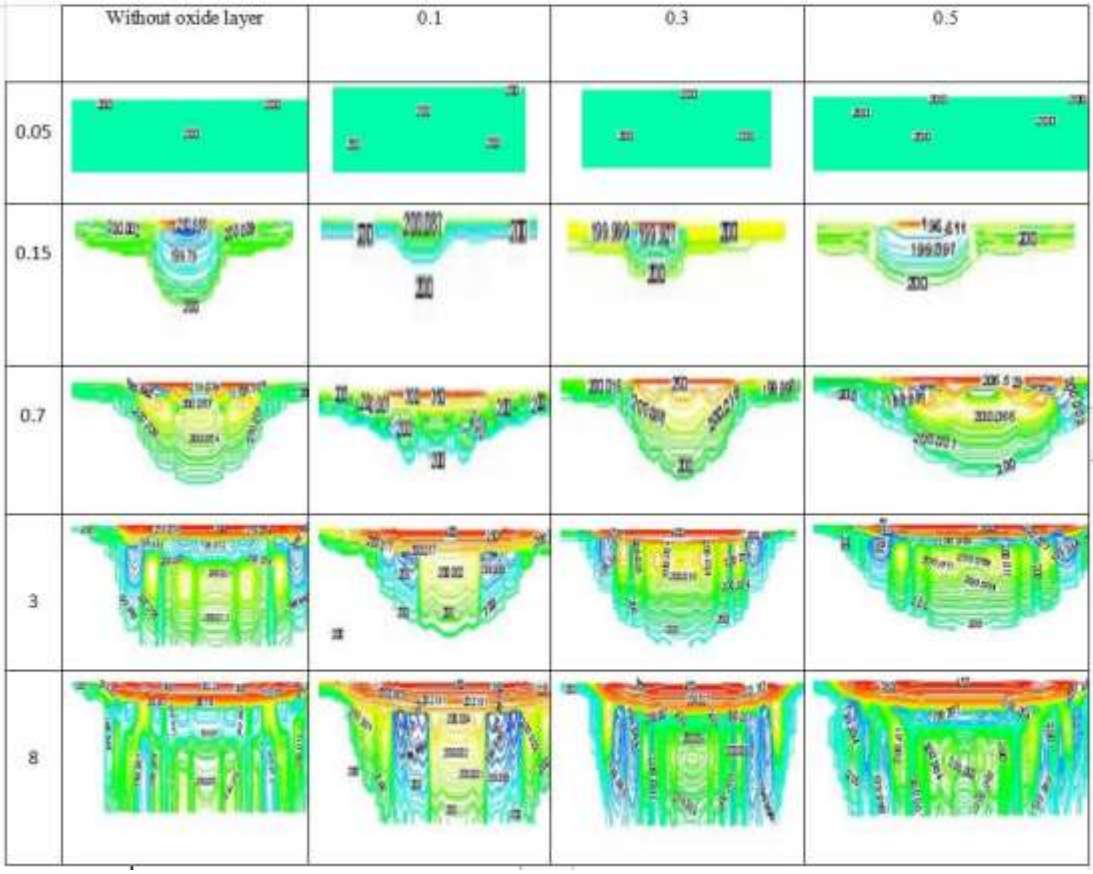
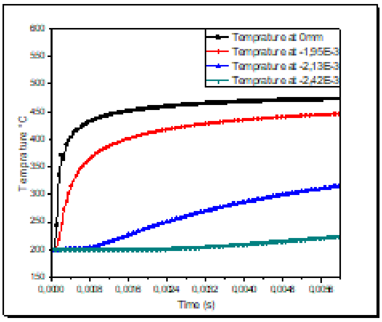


Figure 5 Comparison of temperature profiles over time for four cases at the point of impact

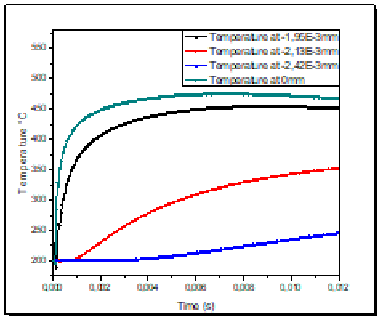
Table 3 Temperature distribution upon impact of an aluminum droplet on a preheated and pre- oxidized substrate



For positions below the substrate surface, the temperature increases much more slowly, as demonstrated in Figure 6. The presence of the oxide layer increases the associated contact resistance, which promotes radial thermal diffusion within the lamella (Tahar et al, 2022). Consequently, the oxide layer influences the dissipation of heat in the substrate by acting as a thermal barrier. This barrier restricts direct heat transfer vertically into the substrate, causing more heat to be conducted laterally within the spreading lamella. This results in a slower temperature rise in the substrate beneath the impact site and alters the overall thermal profile of the system.



a) 0.1mm



b) 0.3mm

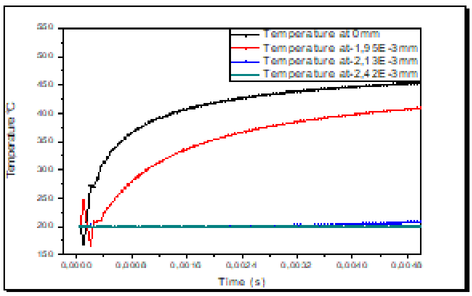


Figure 6 Temperature profile for different positions below the substrate

The variation of the pressure at the impact for the four cases is presented in the graph below (Figure 7). The results show that all cases produce high pressure at the initial impact with the substrate. However, the increase in pressure is greater in the third and fourth cases compared to the first two. This higher pressure is sufficient for achieving strong adhesion between the droplet and the substrate.

From a physical perspective, the higher pressure enhances the mechanical interlocking between the lamella and the substrate surface. Additionally, the increased pressure reduces the thermal contact resistance, which facilitates better heat transfer across the interface. This improved heat transfer can lead to a stronger metallurgical bond as the molten aluminum can more effectively wet and adhere to the steel substrate. Thus, the pressure increase in the later cases contributes to both stronger adhesion and more efficient thermal conduction.

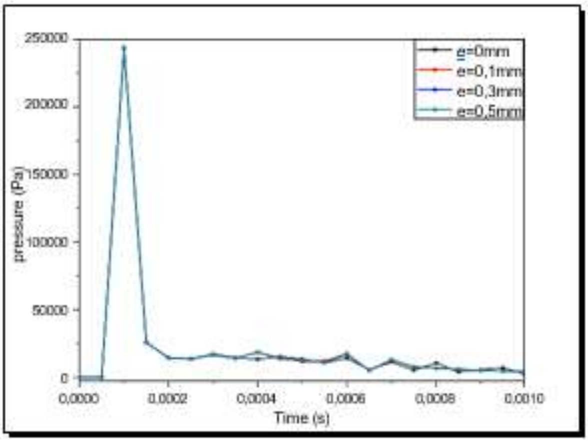


Figure 7 The variation of the pressure at the impact for the four cases

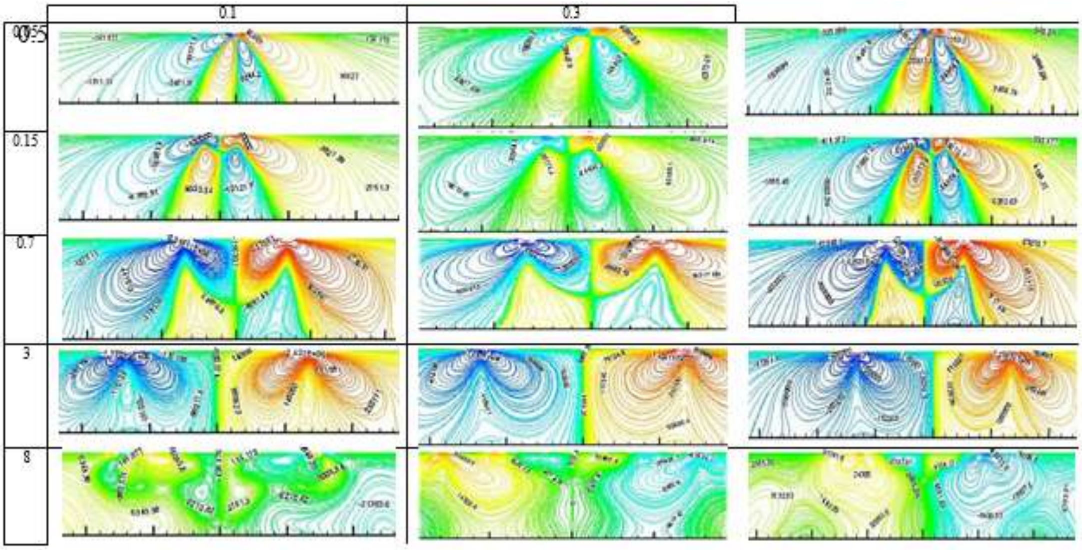
To better understand the interaction between the particle and the substrate, the concentration of stresses within the substrate and the associated deformations are studied. Stress concentration is a phenomenon where the contact area between the particle and the substrate experiences higher stress than the average stress, leading to deformation of the substrate.

Initially, the level of stress is high at the moment of impact and then relaxes during the spreading phase (Table 4). It is noteworthy that the presence of an oxide layer can influence the stress distribution. A thicker oxide layer causes higher stress due to the increased contact resistance and mechanical interaction.

Figure 8 illustrates the deformation at the point of impact for the three cases of the oxide layer thickness. According to the results, the oxide layer makes the substrate more susceptible to deformation upon impact. This increased deformation is due to the oxide layer's effect on the stress distribution, as it can act as a brittle layer that concentrates stress. Over time, this deformation is minimized as the spreading of the droplet progresses and the stresses are redistributed.

Physically, the presence of the oxide layer alters the mechanical properties of the contact interface. The oxide layer, being harder and more brittle than the substrate, can lead to localized stress concentrations and increased deformation upon impact. This effect is more pronounced with thicker oxide layers, which further impede the even distribution of stress and heat, leading to greater deformation initially. However, as the droplet spreads and the stresses are redistributed, the overall deformation tends to decrease.

Table 4 Spatial representation of XY stress for the three cases



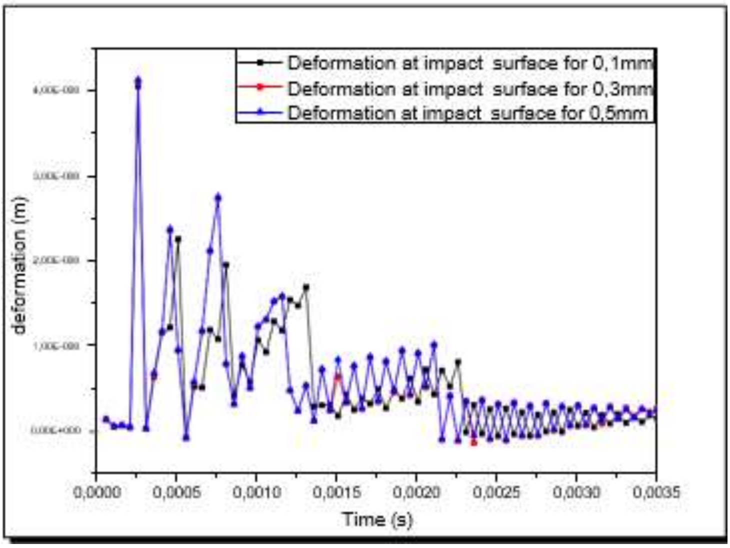


Figure 8 The variation of the deformation at the point of impact

The current model simulates the impact and deposition of a partially molten aluminum particle in four cases (0%, 25%, 50%, 75% molten) on a pre-oxidized substrate at 200°C. The temperature distribution during the impact of the particle is represented in Table 5.

Partially molten particles with a higher percentage of molten material have a higher temperature, which promotes splashing, as shown in the table below. Consequently, the formed lamella breaks up at its edges during the spreading process, and the final splat morphology changes from a disk shape to a splash.

In contrast, unmolten particles or those with a lower percentage of molten material solidify quickly. Due to the internal temperature distribution, these particles have a more significant temperature difference from their melting point, leading to faster solidification. Consequently, the ultimate shape of the lamella is influenced by the fusion state of the particle.

From a physical perspective, the degree of melting within the particle affects its thermal and mechanical behavior upon impact. Higher molten percentages mean that the particle retains more heat, which reduces the viscosity and enhances the fluidity of the material. This increased fluidity leads to more extensive spreading and splashing, as the molten material can flow more freely and is less resistant to deformation.

On the other hand, particles with lower molten percentages have a greater proportion of solid material, which restricts the flow and increases the viscosity. The solid regions within the particle act as rigid inclusions, promoting quicker solidification as the heat is conducted away. This rapid solidification prevents extensive spreading, resulting in a more stable, disk-like lamella.

Therefore, the fusion state of the particle significantly influences the final morphology of the splat, with higher molten percentages leading to splashing and lower molten percentages resulting in more controlled, disk-shaped deposits.

Table 5 Spatial representation of solidification the particle during the formation of the lamella

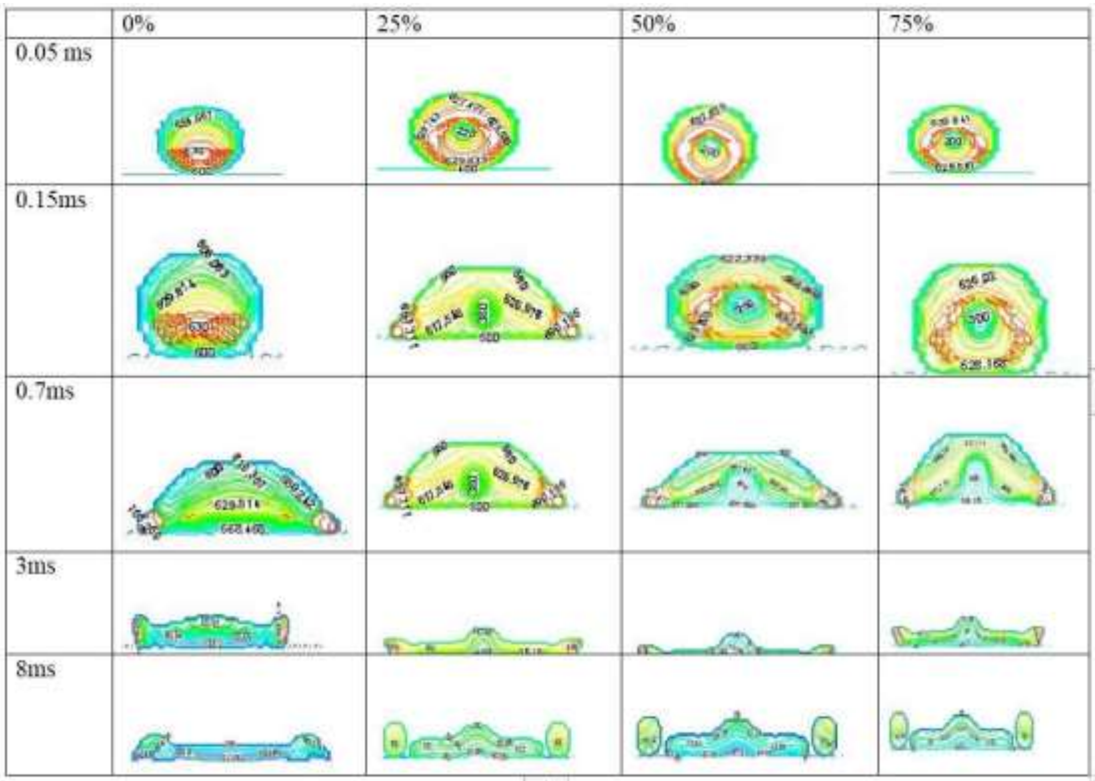


Figure 9 presents a comparison of the spread factors between the four cases. The lower the percentage of the fusion state of the particles in the droplet, the less the spreading occurs. This low spread factor is due to the presence of unmolten metal, which prevents the particle from sliding across the substrate surface .

From a physical perspective, the degree of fusion within the particle directly affects its ability to spread upon impact. Particles with a higher percentage of molten material have lower viscosity, allowing them to spread more easily across the substrate. In contrast, particles with a lower percentage of molten material contain more solid regions, which increase the overall viscosity and impede the flow of the droplet.

The unmolten portions of the particle act as rigid structures that resist deformation and spreading. These solid regions hinder the droplet's ability to flatten and spread out, resulting in a lower spread factor. Additionally, the increased viscosity of the partially solidified droplet reduces its momentum transfer across the substrate, further limiting the extent of spreading.

Therefore, the fusion state of the particle plays a crucial role in determining the spread factor, with lower molten percentages leading to restricted spreading due to the presence of unmolten metal, which acts as a barrier to the flow and spreading of the droplet.

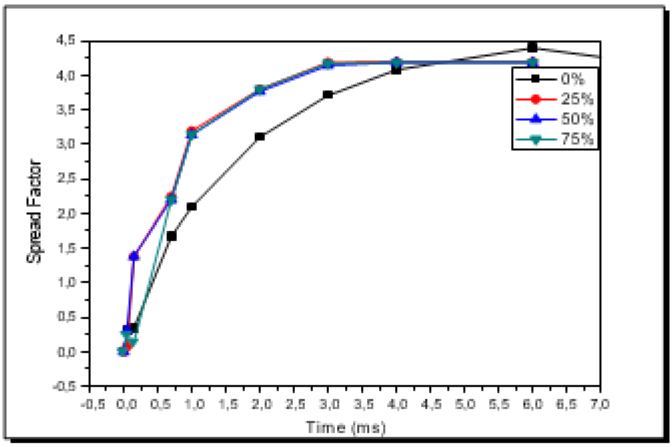


Figure 9 Comparison of the spread factor between the four cases

A comparison of the pressure at impact for unmolten and partially melted particles is shown in the graph in Figure 10. Upon impact, the pressure increases for all cases, which enhances adhesion. However, the pressure is significantly higher for the 75% molten case. Thus, the melting state of the particle results in higher impact pressure. This pressure decreases during the spreading of the particle.

From a physical perspective, a higher pressure at impact reduces contact resistance, leading to stronger adhesion of the deposit. The increased pressure facilitates better mechanical interlocking between the droplet and the substrate, improving the overall bond strength.

Additionally, lower contact resistance due to higher impact pressure minimizes radial thermal diffusion, ensuring efficient heat transfer toward the substrate. This enhanced heat transfer promotes rapid solidification and stronger metallurgical bonding between the droplet and the substrate.

In summary, the fusion state of the particle significantly influences the impact pressure, with higher molten percentages resulting in greater pressures. This higher pressure reduces contact resistance, promotes efficient heat transfer, and ultimately leads to stronger adhesion of the deposited material.

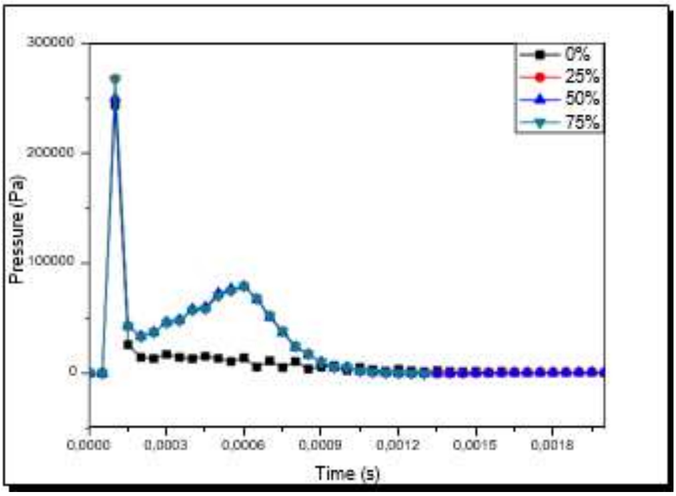


Figure 10 Comparison of the pressure at the impact for the four cases

4. Conclusion

This numerical study investigates the impact and solidification behavior of aluminum droplets on a steel substrate, considering the presence of an oxide layer and various degrees of particle fusion. The governing equations of the model are solved using the finite element method combined with the volume of fluid method. The model's predictions align well with both previous numerical simulations and experimental findings. The key findings of this study are as follows:

The spread of the particle conforms to the substrate's underlying topography, influencing the final shape and contact area of the deposited lamella.

The oxide layer on the substrate increases thermal resistance, impeding heat transfer between the molten fluid and the steel substrate. This results in delayed solidification of the deposited lamella.

The presence of the oxide layer renders the substrate more susceptible to stresses and deformations due to localized stress concentrations at the interface.

Higher impact pressures are observed, particularly with more molten particles, which enhances the adhesion between the droplet and the substrate. This effect is crucial for ensuring a strong metallurgical bond.

Unmolten droplets solidify rapidly upon impact compared to partly molten droplets. Furthermore, a higher percentage of particle fusion results in greater spreading of the droplet upon impact, influenced by viscosity and fluidity dynamics.

In summary, this study underscores the complex interplay between particle fusion state, oxide layer effects, thermal dynamics, and mechanical interactions during droplet impact and solidification on a substrate. These insights are critical for optimizing deposition processes and understanding the formation of splats in various industrial applications involving molten metal coatings.

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