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Comparison between metallic and ceramic splats: Influence of viscosity and kinetic energy on the particle flattening

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ABSTRACT

Plasma-sprayed coating properties are strongly linked to the real contacts between layered splats, defects created by splashing of melted particles and inclusion of unmolten ones, and pores and cracks formed during coating generation. In this paper, the impact, flattening and solidification of millimetre- and micrometre-sized ceramic (alumina) and metallic (Ni5Al) drops or droplets onto a smooth 304 L stainless steel (Ra = 0.06 μm) substrate have been studied to understand the impact phenomena and respond to the following questions:

- Which dimensionless numbers must be considered and how do they compare at both scales?
- How do experiments developed to compare droplets (plasma-sprayed) and drops (free-falling) differ?
- Are the flattening behaviours comparable at both scales?

It has been shown that for droplets (40 μm in diameter) the thickness of the liquid flow boundary layer between flattening droplets and substrate plays a more important role than with drops. It must also be pointed out that with the metal drops or droplets, solidification at their surface can occur, thus modifying the matter flattening. The flattening process depends strongly upon the substrate temperature and the modification of its wettability. Finally, it has been shown that the flattening velocity is strongly linked to the amounts of adsorbates and condensates existing on the substrate, according to heat treatments prior to spraying.

1. Introduction

Plasma-sprayed coating properties depend strongly upon:

- The real contacts between layered splats: typical contacts vary between about 10–20% of splat surfaces for poor ones and 60% or over for good ones [1].
- Splashing of melted particles upon impact that could drastically affect coating properties [2,3].
- Pores [4] and cracks [5] formed during coating generation.

According to the importance of splat real contacts on coating properties, many works have been devoted to this topic since the last two decades:

1.1. Splat interface

- Only a few studies of the direct interaction between splat and substrate and their interface at the microscopic level have been performed, as the preparation and observation of cross-sections at a high spatial resolution are relatively difficult and require specific instrumentation [6–17]. They showed that the shape and real contact between splat and polished substrate and the occurrence of inter-diffusion depend very strongly on:
- The substrate surface chemistry: on the one hand, the oxide or oxyhydroxyle layer formed during substrate preheating or boiling treatment; on the other hand, desorption or not of adsorbates and condensates during preheating.
- The wettability of the flattening droplet onto the substrate that, for example, can be improved by the diffusion of elements such as Mg and Cr towards the substrate surface for NiCr on stainless steel [10].
- The localised inter-diffusion between splat and substrate with possible localised melting of the substrate.

1.2. Modeling of a single particle flattening

Even for the impact of a single, fully melted particle onto a smooth surface, the modeling is very complex. Results depend on: impacting particle parameters (temperature, velocity, impact angle, oxidation stage that can be controlled by diffusion and/or convection phenomena as well as by the quantity of oxygen entrained in the plasma jet along the particle trajectory) and substrate parameters (material, initial surface roughness, oxide composition, thickness and roughness depending on substrate preheating conditions, presence of adsorbates and condensates, possible preferential diffusion of substrate elements towards its surface, especially during the impact of the molten droplet). As

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summarised in the three reviews published about the splat formation [17–19], two models have been considered: analytical and numerical.

- Analytical methods are based on dimensionless numbers characterising the droplet flattening and cooling. However, the weak point of these models is the thermal contact resistance R_{th} between the flattening droplet and substrate. This key parameter controls the heat transfer and, therefore, the cooling and solidification of the splat under formation (as shown, for example, by the 1D model of Dhiman et al. [20]).
- Numerical simulations of the fluid flow and heat transfer during the droplet impact result in very sophisticated models [18,19]. However, the thermal contact resistance between flattening particle substrate is again the key parameter, the difficulty being that R_{th} varies with time along the radius of the flattening droplet [21]. Thus, instead of a constant value of R_{th}, as assumed by most models, a variable one should be considered, but the best model assumes two R_{th} values: one for the central core of the flattening particle, and the other, with a higher R_{th} value, for its periphery [22]. At last, all models developed assume that nucleation occurs at the melting temperature (T_m), which is not the case — nucleation taking place at a temperature below T_m. Nucleation can be easily considered in 1D transient models [23] but unfortunately not in 3D transient ones.

1.3. Experimental study of a single particle flattening

The first measurements were those devoted to the evolution of the particle temperature during the flattening thanks to sophisticated two-colour pyrometers with response times in the 50 ns range [18,19]. However, the interpretation of the cooling curves was very difficult without images showing the evolution of the flattening droplet. Unfortunately, flattening occurs in times of the order of one microsecond, while the fastest camera available allows an interval between each frame of 1 μs and exposure time of each frame of 500 ns [24]. Solutions have been found by photographing one particle during its flattening in a given time range [25–27]. Thus, varying the time range for different particles, assumed to be identical on impact (same diameter, temperature and velocity), it becomes possible to obtain few photographs (fewer than ten) of the flattening at different times. Such images with the evolution of the particle thermal emission and observation of the resulting splat allow a much better understanding of the phenomena taking place. As such experiments are extremely difficult and time-consuming, in parallel many experiments have been performed on millimetre-sized particles [18,19]. In this case, flattening time is in the millisecond range, with fast cameras (5000 images/s) allowing following the whole flattening of a single particle. Unfortunately, almost all experiments, except for those with tin droplets (not really a sprayed material), have been performed with free-falling drops with velocities below 3 m/s. Few experiments have also been devoted to the measurement of the thermal contact resistance during millimetre-sized drop impacts thanks to a thermocouple measuring at the centre of the substrate and the temperature evolution during flattening [28–30]. Experiments with millimetre- and micrometre-sized particles show the existence of a so-called transition temperature, T_t. When the substrate is preheated over T_t, splats are mostly disk-shaped [18,19] while they are extensively fingered or fragmented below it. This has been attributed to desorption of adsorbates and condensates or a better wettability of flattening droplets/substrate when preheating the substrate or reducing the pressure of the surrounding atmosphere. Correspondingly, all models and comparisons with flattening drop or droplet temperature evolution show that over T_t, thermal contact resistances are in the range of 10^{-7}–10^{-8} m^2 K W^{-1}, while below T_t they are superior to 10^{-6} m^2 K W^{-1}.

To conclude, in spite of all efforts devoted, during the last decade, to splat formation on smooth substrates, many works are still necessary for a better understanding of the different phenomena involved. Moreover, many results are based on measurements performed with millimetre-sized particles for which it has not been clearly established that flattening behaviours at the micrometre and millimetre scales are similar. (See, for example, the recent papers of Yang et al. [31].)

The goal of this paper is to address this point. For that a stainless steel substrate has been chosen: 304 L stainless steel, due to its relatively thin oxide layer (a few tens of nanometres) and its good resistance to oxidation. For the sprayed material, either plasma-sprayed (particles with a mean size of about 40 μm) or free-falling (about 5 mm in diameter), an alloy NiAl (5 wt%) and an oxide (Al_2O_3) have been chosen. Table 1 summarises the main properties of both sprayed materials. In the following, melted micrometre-sized particles which are plasma-sprayed will be called droplets, while free-falling millimetre-sized particles will be called drops.

The following points will be successively presented:

- Which dimensionless numbers must be considered and how do they compare at both scales?
- Experiments have been developed to compare droplets (plasma-sprayed) and drops (free-falling).
- Are the flattening behaviours comparable at both scales?

2. Comparison of dimensionless numbers at millimetre and micrometre scales

Many works have been devoted to the impact of water, ethanol, ink… drops and droplets on a solid surface for watering, ink printers [32–35] [36,37]… Upon flattening of sprayed droplets or free-falling drops, temperature effects must also be considered. Their viscosity and surface tension vary as they cool down during flattening; furthermore, and of course, when the nucleation temperature is reached, solidification occurs and its propagation from the bottom of the flattening drop or droplet to its top drastically modifies the liquid flow. Moreover, the very hot drops and droplets interfere with the substrate through the evaporation of condensates and adsorbates, the diffusion of species from substrate to flattening particles or reverse (if temperatures are sufficiently high), and local melting. However, it must be kept in mind that heat transfer from flattening drops or droplets to substrate depends strongly on the transient contact between them and substrate, characterised by R_{th} along the flattening drop or droplet radius. Unfortunately, the local R_{th} is impossible to measure.

The two dimensionless parameters that are considered are the Reynolds number, Re, and the Weber number, We, representing (respectively):

- Re, which is the ratio of inertial force to the viscous one:

$$Re = \frac{\rho_p v_p d_p}{\mu_p}$$

where ρ_P, d_p, and v_p are the specific mass, diameter and velocity of the drop or droplet on impact, while μ_p is its viscosity.

- We is the ratio of the particle inertia to its surface tension:

$$We = \frac{\rho_p {v_p}^2 d_p}{\sigma_p}$$

where σ_p is the surface tension of the drop or droplet.

Table 1
Properties of sprayed materials [41–43].

	Al_2O_3	NiAl
Viscosity (mPa s)	60 (2050 °C)	5 (1450 °C)
	40 (2400 °C)	4 (1600 °C)
Surface tension(mN m^{-1})	652 (2050 °C)	1780 (1450 °C)
	600 (2400 °C)	1740 (1600 °C)