Last developments in diagnostics to follow splats formation during plasma spraying

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Abstract. Plasma sprayed coatings adhesion-cohesion, as well as thermo-mechanical properties depend strongly upon the sprayed particle flattening and solidification, taking place in a few $\mu$s and the corresponding splat formation. Sophisticated fast pyrometers (response times of less than 100ns) have been developed for studying the flattening particle temperature evolution when impacting on smooth surfaces. However the signal interpretation is far to be easy without following simultaneously the evolution of the particle shape. As the flattening time is in the $\mu$s range and the acquisition frequency of the fastest camera is only one image/$\mu$s, it is not possible to follow the flattening particle shape evolution. That is why, different particles considered to have the same impact parameters, were photographed at different flattening times separated by a few hundreds of nanoseconds. In parallel measurements on millimetre-sized drops (metals and ceramics), which flattening time is in the ms range, have been developed because fast cameras (~ 5000 images/s) can be used to follow the flattening phenomena. In this paper are presented the experimental devices developed and impacts of alumina particles in the 40$\mu$m and 5mm ranges, but with Weber numbers as close as possible, are compared. Measurements at both scales demonstrate the importance of the substrate preheating control (time and temperature) to achieve disk shaped splats and explain the phenomena induced.

1. Introduction

In thermal spray the coatings are build up by successive impacts of particle, which form splats after flattening and solidification. So coating properties depend mainly of the adhesion between splats and between splat and substrate. The link between the first splats layer and the substrate plays the main role on the coating adhesion. To improve the contact between splat and substrate, some authors studied the splat obtained and particularly its shape. If this technique gives an idea of some trends, it is difficult with these results to conclude on the influence on splat generation of the different in flight parameters. Indeed when the substrate surface temperature is increased over a given temperature, called transition temperature, splats are disk shaped. In fact it exists a temperature for which the proportion of circular and fragmented particles is identical. However it is mandatory to associate the in-flight particle parameters to splats formation to draw sound conclusions on the substrate temperature effects, which is not yet really possible. So to better understand the involved phenomena, many works have been devoted to the study of a single splat formation; see the reviews of Fauchais and al [1, 2]. They have emphasized the difficulty of the study: impact of particles in a molten state...
with sizes between 20 and 60 µm with velocities ranging between 50 and 600 m/s, resulting in flattening and solidification times in the µs time range. If sophisticated pyrometers have been developed with response times of about 100ns, the interpretation of their signals is far to be straight forward without the visualization of the flattening phenomenon. In spite of the development of fast cameras, it is still impossible to follow this flattening for the same micrometer sized particle. That is why, since the mid nineties, many works have been devoted to the flattening of millimeter sized metal drops [1, 2] occurring in the millisecond time range instead of the microsecond one. However most results were obtained with free falling drops (velocities below 3m/s) resulting in Weber numbers much lower than those obtained in plasma spraying conditions. The comparison of phenomena at both scales (mm and µm sized) is thus rather difficult. If for tin drops, using a centrifugal droplet set-up, impact velocities up to 30 m/s have been achieved [3], this material is unfortunately not sprayed by thermal processes.

The main difficulty is to achieve imaging at the micrometer scale. To our knowledge only one author has used a very fast camera (1 frames/µs) for following the flattening of a single zirconia particle impacting on a cold substrate [4]. Some authors have also studied the impact of metal and ceramic particles on glass or inconel substrates at different times of the flattening process but for different particles [5-6].

In this paper are presented first the experimental devices developed and the comparison of the impacts of alumina particles in the 40 µm and 5 mm ranges, but with Weber numbers as close as possible. Measurements at both scales demonstrate the importance of the substrate preheating control (time and temperature) to achieve disk shaped splats and explain the phenomena induced. At last some examples of the different impacts are presented.

2. Experimental setup

Experimentations are carried out with two experimental techniques for studying impact and flattening phenomena:

- The first one is a modified free-falling set-up to study millimeter-sized drops with low or high Weber numbers.
- The second one uses a plasma spraying set-up with micrometer-sized drops.

Running in parallel these two studies enable comparing particle flattening and cooling on smooth 304L substrates, preheated or not, at spatial and time scales differing by almost three orders of magnitude.

2.1. Millimeter system

To produce liquid ceramic drops, an alumina rod is introduced and melted in an electrical arc furnace (fig 1). A suspended drop is formed. When the gravity force overcomes that of the surface tension, the drop falls. This set-up is disposed into an argon controlled atmosphere chamber. Thanks to the use of a jack, the substrate can be moved up and the relative impact velocity can reach up to 10 m/s [7].

A detector located at the chamber output, generates a TTL pulse when one single drop crosses its measuring volume. This pulse is then directed to the measuring system composed of a fast camera (Photron 4000 image/s) targeting the substrate in two configurations, first tangentially in order to follow the matter ejection during the drop flattening and second with an angle of 45° to follow the whole flattening. Of course, for the first position, in order to better determine the location of ejections, the camera isn’t absolutely parallel to the substrate but a small angle (2°) exists, allowing seeing the opposite edge of the splat.
Finally, during the movement of the substrate, a fast “on board” two-color pyrometer fixed on a jack controls the drop temperature at impact. The main difficulty is due to the abrupt deceleration of the substrate, which permits to study the flattening only during the first 14 ms.

![Figure 1. Drop generator for millimetre sized particle](image1)

### 2.2. Micrometer system

Particles are plasma sprayed using a direct current (D.C.) plasma torch (PTF4 type) with a 6 mm internal diameter anode-nozzle and running with a mixture of argon-hydrogen. The arc current is 650A, the argon flow rate 33 L/min and the hydrogen volume percentage 25%. An alumina powder with particle sizes between 40 and 50 µm is used in this study. These spray conditions result in fully melted particles with velocities at impact around 200 m/s. Splats are collected on a smooth (Ra=0.06 µm) 304L substrate fixed 110 mm downstream of the nozzle exit (figure 2).

![Figure 2. Drop generator for micrometer sized particle](image2)

Three shields are used to select particle trajectories close to the plasma axis:
- A moving water cooled shield (hole of 5 mm) fixed 70 mm downstream of the torch nozzle exit
- Another one (hole of 1.5 mm) fixed 10 mm behind the first shield.
- A last one with a hole of 600 µm fixed 10 mm upstream of the substrate.
2.2.1. In flight velocity measurement system

The in-flight velocity characterization is made by a two points measuring optical detector, the signal treatment permitting to filter the raw signal coming from PM, and generating a TTL pulse on the falling edge of each smooth signal (see figure 3). These two signals, thanks to specific hardware, called velocimeter and running under LabView, permit to measure the time between both TTL pulses. After the velocity calculation, the hardware calculates at which time the particle will impact the substrate. Finally a pulse to start the different measurement systems at a given time (mainly at the impact) is generated. The relative random error on velocity is about 10% and 5% on the acquisition triggering.

![Figure 3. Example of in-flight velocity measurement for an alumina particle 40 µm in diameter with an impact velocity of 140 m/s. The triggering acquisition is the reference time for all measurement systems (pyrometer and cameras)](image)

2.2.2. Particle temperature in flight and during the flattening

A fast (50ns) pyrometer follows the particle temperature during its flattening and cooling. The two wavelengths of the pyrometer are 690 µm and 710 µm. A smoothing is applied on the two original signals (for two wavelengths), but in order to avoid a modification of the profile curves, there are three smoothing areas: - the first area, noted 1 in figure 4, corresponds to the particle signal during its flight just before the impact, - the second area, noted 2, corresponds to the particle flattening on the substrate, - finally, the last area, noted 3, corresponds to the splat cooling and / or the splat diameter reduction. After that, the hardware calculates the dichromatic temperature. When the signal begins to decrease the temperature is calculated thanks to monochromatic function.

![Figure 4. Position of the different smoothing area on a typical thermal signal coming from pyrometer](image)
2.2.3. **Particle flattening by imaging technique**

Imaging techniques allow following the particle flattening. On each camera are fixed a macro lens with a focal distance of 180 mm and two converter (X2) lenses. The main advantage of these lenses compared to a long distance microscope (type Questar) is the more important collected light. One camera is disposed orthogonally to the substrate and other one tangentially to it. The main problem is due to the movements of high temperature gas produced by the plasma torch, which modify the refractive index and thus the focal point of both cameras. It is then necessary to use some cooling air system disposed in front of the camera in order to maintain a constant temperature of the optical path to keep a refractive index as constant as possible.

By adding a variable time delay to the calculated impact time, it is then possible to observe the flattening of micrometer sized particles at different times but for different particles (of course assuming they have the same impact parameters).

2.2.4. **Heating and cooling of substrate**

A monochromatic pyrometer (Ircon 5 μm, 10 ms of response time) controls the substrate temperature during the preheating stage, an automatic system permitting to keep a given temperature. In order to study the effect of the substrate oxidation it is necessary to freeze its oxidation state. For that the heating system is shifted to a cooling one, which permits to come back to room temperature in less than 2 minutes (see figure 5). If the cooling system is not used the substrate comes back to room temperature after 20 minutes.

![Figure 5. Typical signal of heating and cooling for a stainless steel substrate.](image)

\[ T_{\text{set point}} = 250^\circ \text{C} \]
\[ t_{\text{preheating}} = 10 \text{ mn} \]

2.2.5. **Typical procedure to study micrometer sized particle impact.**

With this system, the total time of an experiment, has to be very short. Indeed when the plasma torch is running too long, all optical adjustments are modified due to the hot temperatures, which induce the expansion of the metallic parts. So the spray system is controlled by a homemade hardware, following and controlling all successive steps to obtain one single splat. The main step consists in closing very fast (about 5 ms), the powder feeder and the mobile shield when one particle crosses the velocity measuring area, to avoid several particles on the measurement area. The total time to obtain a single splat onto the substrate is about 10 s.

2.3. **Substrate**

For both particle sizes, substrates are made of stainless steel (304L) and mirror polished by using SIC paper 4000. They are fixed on copper supports that can be heated up to 250 °C with a heating rate of 0.5 °C/s by two small resistances (each one with a power level of 150 W).

To illustrate the features of such systems, two typical examples of results are presented in the next paragraphs.
3. Millimeter sized particle results: Examples of alumina splat cooling during the first ten milliseconds for an impact with a Weber number close to that obtained with the micrometer sized particle plasma sprayed (We=2500)

Table 1 shows images of 5 mm alumina drops impacting onto the mirror-polished stainless steel substrate at two different temperatures.

On a cold surface (room temperature), the particle spreads out more, and also cools down faster. This cooling down is such that, according to the pyrometer measurements, the central part surface is solidified when the piston arrives at the end of its race (i.e. 14 ms after the impact). Splashing is due to the rupture of the thin film expanded around the central part of the splat and the presence of a matter accumulation at splat edges. After the break of the thin film, there is no splat evolution and the splat cooling is slower.

On a preheated surface, there is no thin film expanded around the splat centre. So it can be supposed that flattening is impeded by the splat solidification at substrate surface and the splashing observed is due to liquid droplets jetting out at its top surface still liquid. At the splat edges it can be observed some matter recoil towards the splat centre. Finally a disk shaped splat is obtained.

Table 1. Flattening of a millimeter sized alumina drop onto a mirror polished stainless steel substrate held at two different temperatures

| Time (ms) | 0  | 1  | 2  | 3  | 4  | 5  | 10 |
|-----------|----|----|----|----|----|----|----|
| $T_{\text{substrate}}= 20^\circ\text{C}$ | ![Image](image1.png) | ![Image](image2.png) | ![Image](image3.png) | ![Image](image4.png) | ![Image](image5.png) | ![Image](image6.png) | ![Image](image7.png) |
| $T_{\text{particle}}= 2100^\circ\text{C}$ | ![Image](image8.png) | ![Image](image9.png) | ![Image](image10.png) | ![Image](image11.png) | ![Image](image12.png) | ![Image](image13.png) | ![Image](image14.png) |
| $D_{\text{particle}}=5 \text{ mm}$ | ![Image](image15.png) | ![Image](image16.png) | ![Image](image17.png) | ![Image](image18.png) | ![Image](image19.png) | ![Image](image20.png) | ![Image](image21.png) |
| $V_{\text{impact}}=10\text{ m/s}$ | ![Image](image22.png) | ![Image](image23.png) | ![Image](image24.png) | ![Image](image25.png) | ![Image](image26.png) | ![Image](image27.png) | ![Image](image28.png) |

In order to understand better the differences between both cases, the splat surface temperature is presented in figure 6. It can be seen first that the temperature of the drop is only of 2100°C at the impact, due to properties of the drop generator. Indeed the fall of the drop is only controlled by the one hand its surface tension and on the other the gravity force. The drop is kept during a long time into the electrical arc and its temperature is near its melting one. The second point that must be underlined is the temperature decreases during the droplet flattening followed by the increase of the temperature up to the melting point. This corresponds to the under-cooling phenomenon.

When both cases are compared, the under-cooling phenomenon seems to last longer on a hot substrate. This effect may be due to the good contact with the substrate, which pumps more energy of the splat inducing slower temperature increase. However an other explanation can be that the splat on a cold surface is much more expanded with more numerous nucleation sites, the thermal energy produced by the nucleation being then more important.

Another point is that the under-cooling phenomenon permits to the splat on a hot substrate to reorganize its edges in order to minimize the surface tension strength and obtain a disk.
4. Micrometer sized drop results: Examples of alumina particle impact at the micrometer size for different preheating conditions.

In figure 7 is presented the impact of an alumina particle on a cold (20°C) stainless steel. In the figure 7 (a) and (b) are presented respectively the views from the camera focusing orthogonally to the substrate and from that focusing tangentially. The exposure time of the first camera is 10 µs, the acquisition starting at the impact. For the second camera, the exposure times are sequenced (open 0.5 µs, close 0.5 µs; \( t_{\text{total}} = 10 \mu s \)) and the acquisition is starting 1 µs before impact in order to validate the velocity and have an estimation of the particle diameter. Thanks to the camera exposure time, it is possible to observe the all flattening history.

![Figure 6. Superposition of the splat shape and splat surface temperature time evolutions for a millimetre sized alumina drop impacting with a velocity of 10 m/s onto a mirror polished stainless steel substrate held at two different temperatures](image)

![Figure 7. Flattening of a micrometer sized alumina particle 40 µm in diameter with an impact velocity of 280 m/s onto a mirror polished stainless steel substrate held at room temperature (20°C). Views presented into two directions relatively to the substrate (a) orthogonal (b) tangential.](image)

First it can be noted that the maximum flattening of the drop is about 10 times larger than the initial particle diameter. The second point is that near the impact center, some ejections are present with a
starting location circle shaped. This circle corresponds roughly to the diameter of the impacting particle. It is thus clear that impact splashing occurs and induces some matter ejection at impact. Finally between the splat center and its edges a film seems to exist with matter accumulation at splat edges. Peripheral ejections seem to be generated at different locations and not on the whole periphery of the splat.

When ejections at the impact are compared with peripheral ones, in figure 7 (a) it can be seen that the first ejections have radial directions. But the second ejections have random directions. In figure 7 (b), it can be seen that there are many ejection angles, forming an ejection cloud where it is not possible to discriminate the different types of ejections.

Finally, in figure 8 (a) and (b), the resulting splat, at the end of flattening and cooling processes, is presented. It seems that the particle is completely destroyed at impact; the shape of the resulting splat corresponds to a circle around the impact center with a radius of about 200 µm (see figure 8 (a)). From the picture of the camera 1, it can be seen that splashing takes place at splat edges at about a distance of 200 µm from the splat center. So the matter circled around the impact center corresponds to the maximum flattening of the particle and probably to the peripheral ejection area. In the figure 8 (b), a zoom view of the splat center is presenting and it can be seen that it exists some areas without matter, which start from the impact center where there is very few matter. The diameter of the matter remaining corresponds roughly to the initial particle diameter.

Figure 8. Resulting splat view after flattening and cooling for a particle 40 µm in diameter with an impact velocity of 280 m/s onto a stainless steel substrate held at room temperature (20°C), (a) global view (b) Zoom view of the impact location.

On a hot surface (heating 10 min at 250°C) no splashing is observed (see figure 9 (a) and (b)) and the resulting splats are disk shaped with very few thin matter fingers, a thin matter ring being also present at the peripheral (see figure 9 (c)).

Figure 9. Flattening of a micrometer sized alumina particle 40 µm in diameter with an impact velocity of 290 m/s onto a mirror polished stainless steel substrate held at high temperature (250°C) during 10 min. Views presented into two directions relative to the substrate (a) orthogonal (b) tangential. (c) Resulting splat.
In order to understand the difference between these two impacts figure 10 (a) and (b) present the thermal signals from pyrometers and the corresponding calculated temperature in the case respectively of a cold and a hot substrate.

In both cases, during the impact, the particle temperature increases. It is due to the thermal gradient existing inside it. After this temperature increase, the cooling is much more faster onto a hot substrate. The flattening on a cold surface is very fast because the maximum flattening, in spite of the larger diameter, is reached before that obtained on a hot substrate. Indeed in 1.2 µs, the particle on a cold substrate reaches a radius of 200 µm, corresponding to the flattening speed of about of 170 m/s. On the hot substrate, the flattening speed is only to 50 m/s.

![Figure 10](image.png)

**Figure 10.** Thermal signals of the pyrometer generated by the flattening particle surface with the corresponding temperature for two substrate temperatures (a) 20°C (b) 250°C, 10 min. These signals correspond to the particles presented previously.

5. Conclusion

Plasma sprayed coating properties depend strongly on contacts between layered splats. This study is devoted to the understanding of the formation of a single splat on smooth stainless steel substrate according to particle parameters at impact, desorption or not of adsorbates and condensates at substrate surface. As it is not possible to follow the flattening of micrometer sized particle plasma sprayed (occurring in the microsecond time range), measurements have been performed in parallel with millimeter sized ones. The first experimental set-up developed generates millimeter sized (5mm) ceramic or metal drops with a Weber number close to that obtained when plasma spraying particles of the same material in the tens of micrometer size. It permits to follow the particle flattening and cooling during the first 14ms. The second set-up uses a direct current plasma torch with micrometer sized particles that gives global information about the impact of micrometer sized particles onto substrates held at different temperatures. Thanks to a substrate cooling system starting as soon as the splat is formed, it is possible to freeze the oxidation of the substrate and splat surfaces after the particle impact and to study the effect of the substrate surface chemistry on the splat shape. Correlating results obtained with fast (100ns response time) bi-chromatic pyrometer and the imaging techniques it becomes possible to observe the whole flattening history. Results permit to better understand the effect of the substrate surface modifications (oxidation state and corresponding roughness, as well as desorption of adsorbates and condensates) onto the splat shape. This system allows also following the matter ejection at impact and during flattening and solidification. Thus it becomes possible to improve the splat deposition onto the substrate by optimizing its preheating. When results obtained with both systems are compared correlations can be found on the flattening of particles at both scales. It is also possible to compare experimental results with flattening models at both scales and, especially, to better understand the flattening process for micrometer-sized particles and after for approaching nanometer scale. Indeed only a model can be used for observe the flattening of nanometer sized particle, due to the particle size and the very short distance between the torch and substrate.
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