Influence of Particles Velocity and Temperature on the Properties of Thermally Sprayed Coatings

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Abstract
The knowledge about the influence of thermal spray process parameters on the final coating properties is a crucial part in coatings design [1-10]. Nowadays, many sophisticated on-line particle diagnostic systems are available on market. Based on different physical methods such systems allow for the measurement of particle speed, temperature, and size distributions as a function of time and place. Particle temperatures and velocities during the moment of impact on the substrate are believed to be the main factors influencing splashing and spreading of the droplets, beside the substrate roughness and substrate temperature itself. This paper presents some results received using DPV-2000 diagnostics system and shows the impact on final coating microstructure.

Introduction
To get a better understanding of the basics of thermal spray processes and coating formation, many particle measurement devices came on the market during the last decades, like among them Laser Doppler Anemometers (Figure 1) for particle velocity measurements [11-17], LaserStrobe [18], or high speed (quotient) pyrometers for temperature measurements [19-24] and combined systems like the DPV-2000 [25], the CCD camera based devices [26-31] such as the Accuraspray [32, 33], the PlumeSpector [32], the SprayWatch system [34, 35], the IPP-2010 [36], and the ThermaViz system [37].

Laser- and DPV- based systems extract local information out of a few hundred µm³, which have to be averaged (i.e. scanned) for getting representative particle distributions in the
plume. CCD camera based devices offer the advantage of a larger image field (dm²), but lower local resolution.

In this study the optimization and reliability of atmospherically plasma sprayed (APS) coatings, especially highly porous thermal barrier coatings (TBC), was investigated using the DPV-2000 particle monitoring equipment. This study aims to elucidate the correlation between the basic plasma spraying parameters (like electrical current, plasma gas (composition and amount), stand-off distance between plasma generator outlet and substrate, etc.) and the deduced physical properties like particle temperatures and velocities and the final mechanical and thermal properties of coatings.

Experimental

Thermal Spraying
A commercially available 8 wt% Yttria stabilized ZrO₂ powder (YSZ) was used for this study. The particle size fraction was -136 +30 µm. The spraying tests were carried out using a plasma spray facility type Medicat M60 with a modified F4 torch working at atmospheric pressure conditions. Different plasma gas compositions and flow rates (Ar, H₂) were used. The electrical input power as well as the nozzle diameter and stand-off distance between nozzle exit and samples were varied and the resulting particle speed and temperatures were monitored (Table 1).

Table 1: Plasma parameter variations.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Range</th>
</tr>
</thead>
<tbody>
<tr>
<td>Plasma Gas (Argon)</td>
<td>22 ... 40 l/min</td>
</tr>
<tr>
<td>Additional Gas (Hydrogen)</td>
<td>0 ... 9 l/min</td>
</tr>
<tr>
<td>Plasma Current</td>
<td>600 ... 800 A</td>
</tr>
<tr>
<td>Electrical Input Power (EIP):</td>
<td>20 ... 40 kW</td>
</tr>
<tr>
<td>Stand-off Distance</td>
<td>75 ... 200 mm</td>
</tr>
<tr>
<td>Nozzle Diameter</td>
<td>6 ... 8 mm</td>
</tr>
<tr>
<td>Robot Speed</td>
<td>200 ... 400 mm/s</td>
</tr>
<tr>
<td>Powder Feed Rate</td>
<td>10 ... 40 g/min</td>
</tr>
</tbody>
</table>

The parameter field covered a wide range of particle temperatures (1800 to 2900 °C) and particle speed (50 to 300 m/s). For most of the parameter settings, samples were sprayed on different substrate materials like stainless steel and Hastelloy and afterwards metallographically inspected.

Particle Monitoring Device
For spray parameter optimization, the portable in-flight particle diagnostics system DPV-2000 of TECNAR Automation Ltd. (Canada) was used. The equipment consists of an optical sensing device based on a patented technology developed by the National Research Council of Canada.

The system uses infrared pyrometry along with a dual slit optical device in order to perform real time in-flight particle temperature, velocity and diameter measurements (Figure 2). Because of the slit mask a particle passing in front of the sensor will generate a two-peaks
signal (Figure 3). Out of the known optical geometry (magnification factor), the speed of the particles can be calculated.

![Figure 2: Principle of DVP 2000 system with sensing head and optical beam splitting for quotient pyrometry (F, F1) and velocity measurements (Ref. Tecnar).](image1)

![Figure 3: Signal (time versus intensity) generated from a particle passing in front of the dual slit for speed measurements (Ref. Tecnar).](image2)

The signal is further split in two different wavelengths (F, F1) and the temperature of this particle is analyzed by quotient pyrometry (PD-1, PD-2). Assuming spherical particles, the amplitude varies proportionally to the square of the particle's diameter, which leads to the indication of the diameter distribution. However, because of signal filtering, a better estimate of the diameter is obtained from the time integral of the complete signal normalized for (i.e. multiplied by) velocity.

**Table 2: Technical specifications (according to the manufacturer):**

<table>
<thead>
<tr>
<th>Specification</th>
<th>Details</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diameter measurement range</td>
<td>1 to 300 µm (depending on photo mask used)</td>
</tr>
<tr>
<td>Velocity measurement range</td>
<td>10 to 400 and 400 to 1200 m/s (depending on photo mask used)</td>
</tr>
<tr>
<td>Temperature measurement range</td>
<td>1'100 to 4'500 °C (depending on photo mask used and emissivity of the powder)</td>
</tr>
<tr>
<td>Analysis rate</td>
<td>up to 800 particles per second</td>
</tr>
<tr>
<td>Calibration module for</td>
<td>Temperatures between 1800 ... 2500 °C and speed between 40 ... 90 m/s</td>
</tr>
</tbody>
</table>

**Metallographic Investigations**

The sprayed coatings on Hastelloy and stainless steel substrates were prepared according to the DVS guidelines [38]. A previous study [1] showed the influence of the embedding media on the apparent porosity. For the quantitative results of porosity, image analysis by optical microscopy was performed on least 5 different locations of each sample.
Three-Point Bending Tests
For investigating the mechanical coating properties, three point bending tests using Dynamic-Mechanical Analysis (DMA) were performed on a PerkinElmer DMA 7e device (PerkinElmer Inc., Norwalk, Connecticut, USA). The maximum applicable force was 8.5 N and an increment of 500 mN/min was applied until the sample broke. The geometry of the free standing coatings were approximately 1.8 x 11 x D mm, where D was the coating thickness ranging from approx. 0.3 to 0.6 mm. The elastic modulus was extracted from the slope of the stress-strain curve measured at room temperature.

Results / Discussion
Influence of Spray Parameters on Particle Speed and Temperature
For different positions from the nozzle exit (75 ... 250 mm), particle velocities and temperatures were measured using the DPV 2000 and afterwards samples were sprayed on Hastelloy and stainless steel substrates using the same conditions. As an example Figure 4 shows the typical particle speed and temperature distributions in the cross-section (horizontal- and vertical-axe) of the plasma jet at 100mm from the nozzle exit ensuring homogeneous and symmetrical particle loading.

The experiments were done for different plasma gas compositions and amounts, different plasma current levels and finally nozzle diameters. For two specific nozzle diameters any stable process parameters (including extreme and critical settings) are experienced regardless of the resulting coating formation (efficiency, adhesion, etc.). Figure 5 shows the distinct regimes of the cumulative results of the following variations with stable plasma conditions (current: 600 ...800 A, plasma gases Ar: 24 ...37 l/min, H2: 3 ...7 l/min and stand-off distances: 75 ...200 mm) for two different nozzle diameters, namely 6 and 8 mm.
There are distinct v-T-regimes for the two nozzle types. The main influence caused by the different nozzle diameters can be seen for the particle speed, whereas the mean temperatures of the particles do not differ much. The slope between temperature and speed is steeper for the 8 mm nozzle compared to the 6 mm nozzle.

Influence of Particle Velocity and Temperature on Microstructural Features:
The structural differences of the coatings sprayed at vastly different parameters were examined using image analysis and mechanical testing. Figure 6 shows the dependencies of particle temperature and speed in relation to the total porosity measured by image analysis. It could be seen that for decreasing particle speed and temperature the porosity increased and the microstructures showed less cracks, compared to the dense coatings sprayed at “hot” and “fast” conditions. In Figure 6, one can also see that the pronounced trend of porosity increase with the lowering of particle in-flight parameters vanishes when the mean particle temperature drops below the melting point (ca. 2600 °C for YSZ). The explanation could be that the temperature of particles, which actually form the coating, stays nearly unchanged, whereas the deposit rate dramatically decreases due to bouncing-back effect.
Figure 6: Comparison of differently sprayed TBC coatings and their total porosity (mean value measured at 5 different places) compared to corresponding particle temperature and velocity.

Micrographs from previously resin-infiltrated cross sections reveal entirely different microstructures for the parameter range depicted in Figure 6. Two candidates from the “extreme” positions (i.e. (1) “hot/fast” and (2) “cold/slow”) are shown in Figure 7 and Figure 8.

Figure 7: Coating microstructure sprayed with high particle velocity ($V_{\text{max}} = 307 \text{ m/s}$) and high temperature ($T_{\text{max}} = 2925 \text{ °C}$) showing an overall porosity of about 26 % and vertical cracks with a crack density of 2.7 cracks/mm in length.

The sample sprayed with high velocity ($V_{\text{max}} = 307 \text{ m/s}$) and high temperature ($T_{\text{max}} = 2925 \text{ °C}$) shows less porosity (approximately 26 %), but a high level of vertical cracks with a density of 2.7 cracks/mm in length (Figure 7) compared to the coating sprayed with low velocity ($V_{\text{max}} = 115 \text{ m/s}$) and low temperature ($T_{\text{max}} = 2520 \text{ °C}$) showing approximately 44 % porosity, but no visible cracks (Figure 8).
Figure 8: Coating microstructure sprayed with low particle velocity \( (V_{\text{max}} = 115 \, \text{m/s}) \) and low temperature \( (T_{\text{max}} = 2520 \, ^\circ\text{C}) \) showing higher porosity level (about 44 \%) than Figure 7, but no cracks.

The higher the particle speed and temperatures are, the higher the degree of melting and spreading will be, which resulted in a higher coating density. However, due to larger residual stresses, the coatings tended to crack, as already known from literature \[4, 39, 40\]. In contrast, the lower the temperature and the particle speed are, the more porous the coatings have been until no more adhesion and “grit blasting with cold particles” took place.

The gun electric input power (EIP), argon and hydrogen flow rates varied for each nozzle diameter and corresponding changes in the coating porosity were analyzed. As far as power is concerned, results of the experiments followed the well known trend of general increase in particle speed and temperature with the power and decrease of the coating porosity \[41-48\]. However, variations of the total gas flow rate and of relative content of the gas components when keeping the input power approximately constant have not revealed any clear trends in the particle temperatures. Nor there was any clear correlation between those parameters and the coating microstructure. Generally, increase in the flow rate of one or both gas components led to higher particles velocities while the temperature could decrease due to the reduction of particle dwell time and specific plasma enthalpy.

All statements are related to coatings in the „as sprayed“ state only. However, creating tendencies concerning the sensitivity to cracking may be partially transferred to the behavior under thermal cycling.

**Mechanical Analysis by Three-Point Bending Tests**

The results from the three-point bending tests showed for all coatings the typical brittle behavior and a total bending strain of about 0.3 \%. The pseudo-plastic behavior may be attributed to the crack propagation through the porous coatings. Figure 9 represents a typical
stress-strain curve of a three-point bending measurement for a TCB coating sprayed with reference conditions and with higher hydrogen content.

![Stress-Strain Curve](image)

**Figure 9**: Stress-Strain curve of two samples sprayed with “reference” and “higher hydrogen” parameters at 100 mm from the nozzle exit (porosity in both cases approximately 20%).

Out of the DMA data from Figure 9, the tangent E-Modulus was determined from the stress-strain slope and is shown as function of strain in Figure 10.

![Elastic Modulus](image)

**Figure 10**: Tangent E-Modulus of the TBC samples as a function of strain at “reference” and “higher hydrogen” parameters leading to the same porosity (about 20%), but approx. 65 % higher E-Modulus.

It can be seen that for coatings sprayed with different hydrogen content, the resulting porosity remained unchanged at about 20 %, whereas the mechanical testing showed large differences with about 65 % higher E-Modulus.
Conclusions

Particle monitoring devices can help finding optimized spraying parameters and even be used for self regulating feed-back systems. Depending on the measurement system used, local information as out of laser and DPV measurements (a few hundred µm³) has to be averaged (i.e. scanned) for getting representative particle distributions in the plume. CCD camera based measurements offer the advantage of a larger image field (dm³), but lower local resolution.

Using particle monitoring devices, changes in plasma conditions can be found, which strongly affect the final coating microstructure. From the experiments, the following consequences can be drawn:

- Strong influence of nozzle diameter on v-, T-regimes;
- For a specific nozzle diameter a discrete and restricted field of „in-flight parameter“ can be technically/physically realized under stable process conditions. The field and the parameter trend within this field is characteristic for the nozzle type.
- Variations of plasma gas flow rates could have ambiguous influence on the coating properties and can be hardly used for the purpose of coating quality control;
- Same level of porosity can show different E-Modulus;
- Three point bending tests can reveal mechanical coating data rather than porosity level can show from microscopy data.

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