Influence of processing parameters and heat input level on the functional properties of atmospheric plasma sprayed Al$_2$O$_3$-3TiO$_2$ commercial coating

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Abstract. This paper concerns the combined influence of spraying parameters and heat flux level during atmospheric plasma spraying process of commercial alumina-titania coating. One of the main industrial application of alumina-titania are wear and abrasion resistant protective thermally sprayed coatings. Commercial high throughput plasma torch with cascaded construction is used as the spraying equipment. Three sets of spraying parameters were used for the basic evaluation of their influence. Additionally, three levels of heat input during spraying process were added to some of those sets of parameters. Those heat input levels are called “cold”, “hot” and “extra hot” depending on the relative amount of direct heat input and maximum achieved surface temperature. Comparison of surface hardness, microstructure and especially abrasion resistance is carried out. Results confirmed that for successful design of such ceramic coatings, appropriate and sufficient heat input during spraying process into the complex of layer-substrate is required.

1 Introduction

Atmospheric plasma spraying (APS) process, being one of thermal spraying technologies, is able to melt and subsequently deposit almost any material thanks to the temperature above 15 000 K in the plasma jet. Such high temperature is sufficient to deposit most materials, including refractory metals and ceramics and APS technology is commercially used mainly for deposition of ceramic coatings. Alumina-titania coatings were developed to produce wear resistant coatings, where titania addition to alumina significantly increases toughness of the coating compared to pure alumina. Al$_2$O$_3$ - 3 % TiO$_2$ commercial powder was used in this experiment as feedstock material, deposited by cascaded design plasma torch. Coatings sprayed on low carbon steel substrate underwent superficial Rockwell indentation, optical microscopy on polished cross section, adhesion strength testing and dry sand abrasion test. [1] [2] [3]

2 Spraying equipment, feedstock and substrate material

Atmospheric plasma spraying of all samples was done with cascaded plasma torch (SinplexPro by Oerlikon Metco) with argon - hydrogen mixture as plasma gas and 9 mm torch nozzle diameter. Samples were sprayed in department of thermal spraying of Research and Testing Institute Pilsen (VZÚ Plzeň s.r.o.).

Commercial powder Amperit 742 (fused) by Höganäs (Sweden) with nominal particle size distribution between 22 and 45 micrometers was chosen as the default material. Nominal chemical
composition is 97% of $\text{Al}_2\text{O}_3$ and 3% of $\text{TiO}_2$. Trace amount of other oxides is presented ($\text{SiO}_2$, $\text{Fe}_2\text{O}_3$). 
S235 construction steel served as the substrate material. Specimens with cross section of 25 mm x 5 mm and 75 mm in length were used for Dry sand rubber wheel test and specimens of same cross section and 50 mm length for metallography and indentation. In addition, cylindrical samples of 25 mm diameter and 6 mm height were used to evaluate adhesion strength of the coating according to ASTM C633.

3 Testing methods
Because good wear behavior and particles abrasion resistance are one of the commercially desired properties of alumina-titania coating, dry sand abrasion test was chosen in order to compare such resistance of specimens sprayed with different spraying parameters and heat input levels. Dry sand rubber wheel test according to ASTM G-65 were deployed on tree specimens of each parameter set. An output of this test was average material mass loss of tested specimens after five testing cycles. The ASTM testing standard however demands evaluation of volume loss of tested material, but due to the general difficulties of measuring the specific density of thermally sprayed coating (especially caused by closed porosity), mass loss was employed instead. It was assumed, that no relevant differences of density between samples of same material are presented and therefore mass loss can be compared with relevancy. Rockwell superficial indentation (HR 15N) were carried out on specimen from every set. Polished cross sections also underwent optical microscopy to compare microstructure, porosity and eventually presence of cracks developed in coating due to the possible overheating. Surface temperature was measured with laser pyrometer right after finishing every spraying cycle (at the start of the cooling cycle) on the coating surface. Pyrometer was calibrated to the emissivity of the coating material at ambient temperature.

4 Spraying parameters and heat input levels
Based on the former experience with deposition of both pure $\text{Al}_2\text{O}_3$ and $\text{Al}_2\text{O}_3$ based ceramic coatings and with utilization of results of papers concerning this topic [4, 5, 6], set of deposition parameters was designed. Main goal of spraying parameters optimization was to achieve crack-free, hard, wear resistant coating, especially with good abrasive resistance. As main variable process parameters, input torch power and argon flow rate were chosen. Those parameters have chief influence on the formation of plasma jet. Three levels of Critical Plasma Spraying Parameters (CPSP) [4, 5, 6] were deployed valuing 0.83, 1.01 and 1.17. Those three levels represent low, normal and high plasma conditions respectively. Constant spraying distance of 120 mm was used for all three sets of parameters. A fourth set of parameters was added, with the same spraying parameters and CPSP value as set No. 3 (1.17 CPSP) but with decreased spraying distance from 120 mm to 105 mm. This correction was done in order to deal with relatively short plasma jet of No. 3 set resulting from lowering argon flow from 60 l/min down to 50 l/min. For all variable parameters see Table 1. All other process parameters were constant during whole spraying, with 6 l/min of hydrogen flow rate as secondary plasma gas, 36 g/min of powder feed rate and argon also as a carrier gas. Torch linear speed was constant during spraying of all specimen.

| No. | CPSP | Power input $\text{kW}$ | Argon flow $\text{l/min}$ | Spraying distance $\text{mm}$ |
|-----|------|-------------------------|---------------------------|-------------------------------|
| No. 1 | 0.83 | 47                      | 60                        | 120                           |
| No. 2 | 1.01 | 57                      | 60                        | 120                           |
| No. 3 | 1.17 | 55                      | 50                        | 120                           |
| No. 4 | 1.17 | 55                      | 50                        | 105                           |
With process parameter sets chosen, all four sets were sprayed several times with different heat load levels to the complex sample – coating. To achieve different heat loads with same spraying parameters, different system of spraying passes and cooling cycles were utilized. Three heat load levels were named “cold”, “hot” and “extra hot”. For spraying of “cold” sets, temperature of the substrate-coating complex did not exceeded 150 °C. After every cooling cycle, spraying was resumed after coating surface temperature dropped below 30 °C (this “ambient” temperature cooling threshold was same for all heat levels). “Hot” sets were sprayed with cooling cycles employed after 3 continuous torch passes. Maximal surface temperature after 3 continuous torch passes varied from 240 °C (set No.1) up to 300 °C (sets No. 2 and No.4). “Extra hot” was the highest heat load level, with cooling cycle after 5 continuous torch passes. Temperatures of the coating surface of those “extra hot” samples went up to 340 °C in set No. 3 and even up to 360 °C for sets No. 2 and No. 4. Not all three heat load levels were used in each spraying parameter sets. Table 2 in the section 5 displays the distribution of heat load levels in individual spraying sets as well as the maximum temperatures during spraying mentioned above.

5 Results

Dry sand rubber wheel test was used as the principal test for the evaluation of the deposited coatings, with the search for the most abrasive resistant coating and thus revealing proper spraying parameters and also evaluation of the influence of heat load to the quality of the coating. Abrasive mass loss during individual cycles of the abrasive track test was plotted to a graph. Linear regression was employed to get the linear function of mass loss on the abrasive track length. Slope of this linear function was used to evaluate abrasive resistance of all coatings (results shown in Table 2), where lower the value of the slope, the lower mass loss during test and thus higher abrasive resistance of the specific coating. Rockwell superficial indentation results are also included in Table 2 along with average thickness per torch pass. Rockwell superficial indentation results shown quite positive correlation with results of abrasion resistance.

Examples of microstructure images are shown below (Figure 1 and Figure 2b). Cracks at the interpass interfaces and some minor vertical cracks were observed in all extra hot heat load level sets (Figure 1b and Figure 2b). [7]

**Table 2.** Results of abrasive testing, hardness indentation, deposition efficiency (thickness per pass) and with maximum surface temperature during spraying with different heat loads. Colored values in the abrasive mass loss line slope goes with red representing the worst results, over tones of orange and yellowish (average results) to the green values representing the best results.

| Set number | Plasma jet conditions (CPSP) | Heat input level | Maximum surface temperature °C | Thickness per pass µm/pass | Abrasive mass loss line slope (lower the better) | Superficial hardness (HR 15N) |
|------------|------------------------------|------------------|--------------------------------|---------------------------|-----------------------------------------------|-----------------------------|
| No. 1      | Low (0.83)                   | Cold             | 150                            | 32                        | 0.330                                         | 85.8 ± 0.9                  |
|            |                              | Hot              | 240                            | 46                        | 0.277                                         | 87.5 ± 0.4                  |
| No. 2      | Normal (1.01)                | Hot              | 240                            | 45                        | 0.163                                         | 88.3 ± 2.1                  |
|            |                              | Extra hot        | 360                            | 61                        | 0.118                                         | 89.4 ± 1.6                  |
| No. 3      | High (1.17)                  | Cold             | 150                            | 40                        | 0.380                                         | 86.7 ± 0.4                  |
|            |                              | Hot              | 240                            | 47                        | 0.194                                         | 91.0 ± 1.4                  |
|            |                              | Extra hot        | 340                            | 48                        | 0.183                                         | 91.3 ± 0.7                  |
| No. 4      | High (1.17)                  | Hot              | 300                            | 38                        | 0.150                                         | 90.2 ± 0.5                  |
|            |                              | Extra hot        | 340                            | 47                        | 0.089                                         | 90.9 ± 0.8                  |


Figure 1. Set No. 2 samples microstructure images (100x); a) hot and b) extra hot

Figure 2. Set No. 4 samples microstructure images (200x); a) hot and b) extra hot

6 Discussion
Results of abrasive resistance test shown that there are two ways to increase abrasive resistance and thus presumably overall wear resistance of alumina-titania coatings. First way is increasing CPSP parameter of plasma jet above value of 1. The simplest way to increase CPSP is to increase input electrical current and thus increase power of the torch. A decrease in a primary argon flow also leads to increase in CPSP, but at a cost of somewhat reduction in plasma jet length and velocity, which may lead to inferior performance of plasma jet during spraying (see differences between spraying sets No. 2 and No. 3, where a decrease in argon flow had to be utilized in order to achieve high CPSP value). To balance plasma jet velocity reduction caused by lowering plasma gas flow, a decrease in spraying distance (i.e. distance between torch nozzle and substrate) may be employed. Effect of this can be observed in comparison between spraying sets No. 3 and No. 4., which were sprayed with exactly same parameters, except lowering spraying distance from 120 mm to 105 mm.
Second way to increase wear resistance of alumina-titania coating is to increase heat flux during spraying. This can be done simply by optimizing spraying passes and cooling cycling. Results proved that significant increase in heat input during cooling cycles leads to increasing abrasive resistance. This is probably caused by more intensive phase transformation (from unstable phases (mostly $\gamma$-Al$_2$O$_3$) to
stable α-Al₂O₃ phase, that leads to improvement in mechanical properties) due to the higher temperature of the coating during spraying [8] [9]. Important fact is that both way of increasing wear resistance of alumina-based coating can be combined - increasing CPSP of plasma jet and optimizing spraying cycle to achieve higher temperatures leads to highly wear resistant ceramic coating. On the other hand, careful approach must be taken while increasing heat load and thus temperature of the coating during spraying, since subsequent increase in quenching stresses and thermal expansion mismatch between coating and substrate material may lead to development of cracks and also significant reduction in coating adhesion strength.

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