

## Control of the Substrate Temperature during Plasma Spray Deposition

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Thermocouple temperature measurements with the Al<sub>2</sub>O<sub>3</sub> covered probe during vacuum plasma spray process were performed. The voltage of arc discharge during plasma spray was constant (31 V) and the arc discharge current varied from 420 A to 800 A. Temperature measurements were done for different distances between the probe and the nozzle of plasma gun (0.17 – 0.23 m). The gray body radiation model was used to describe the experimental results of temperature – time dependencies. The proposed model describes the kinetics of the sample heating in the plasma and the sample cooling after plasma spray had been turned off. The absorption factor of a probe surface was evaluated analyzing kinetics of the sample cooling.

*Keywords:* vacuum plasma spray; temperature; gray body.

### INTRODUCTION

Ceramic materials, due to their hardness and chemical inertness, have great potential for applications as ideal wear-resistant materials at high temperatures [1 – 2]. The plasma-spray method is most widely used to produce a considerable variety of oxide ceramic coatings, such as Cr<sub>2</sub>O<sub>3</sub>, Al<sub>2</sub>O<sub>3</sub>, ZrO<sub>2</sub> and TiO<sub>2</sub>, etc. Common features of plasma spraying are the high temperatures of melted particles and high deposition rates, up to 500 μm h<sup>-1</sup> [3]. The plasma spray process is based on the generation of a plasma jet consisting of argon or argon with admixtures, which are ionized by a high current arc discharge in a plasma torch. The powders to be sprayed are injected into the plasma where they are accelerated, melted and finally projected onto the substrate. Ceramic coatings are formed by solidification and flattening of molten or partially molten particles at impact on the substrate [4]. Such formed coatings have different applications: as a protective materials from hostile environments, a hard coatings of they can be used in energetic sector for producing of electrodes for solid oxide fuel cells (SOFC). By operating the spray process in a chamber with reduced pressure, by selecting different powders it could be deposited a sufficient porous anode or cathode layer and thin, completely dense layer as required for the electrolyte [4 – 6].

In plasma spray deposition process part of plasma energy is transferred to the substrate. In this way temperature of the substrate affected by plasma becomes one of the most important technological parameter – defining properties of the coatings. It is always important to control the temperature in order to prevent softening and possible distortion of finished element. On the other hand for a chosen powder to be sprayed on the substrate, the temperature field of the plasma flame is very important parameter as the in-flight particles take heat from the plasma flame [7 – 11].

In this paper investigations of temperature field in a vacuum plasma spray torch are presented. A small-mass (2.7 g) probe of Al<sub>2</sub>O<sub>3</sub> ceramic was chosen for the experiment. It is shown that gray body model may be used to describe the temperature-time dependences of small mass substrate affected by plasma torch.

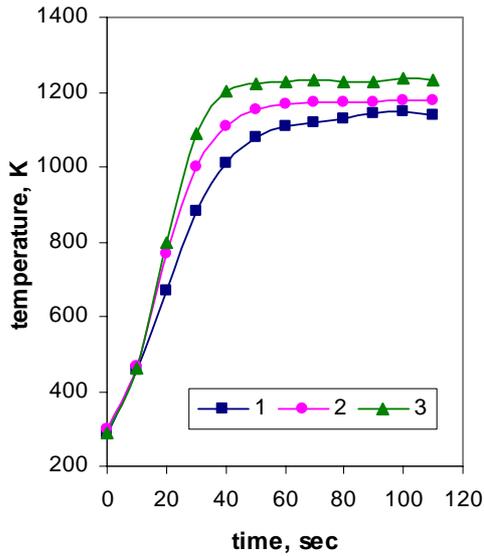
### EXPERIMENTAL

The employed experimental equipment consists of plasma gun (SG-100 Miller Thermal Inc.), vacuum chamber and system for chamber cooling [12]. Argon (Ar) was used as the working gas. This device was employed to produce yttria stabilized zirconia coatings and yttria stabilized ZrO<sub>2</sub> powder with 8 % mol of yttria was used in the plasma spray experiments. The temperature of a small-mass Al<sub>2</sub>O<sub>3</sub> probe in the deposition chamber was measured at the same plasma conditions as it is used to produce ceramic coatings by means of thermocouple and special electronic circuit. These temperature measurements were obtained at 10 s time intervals. The plasma spray process was executed at a constant voltage 31 V and the arc discharge current varied between 440 and 800 A.

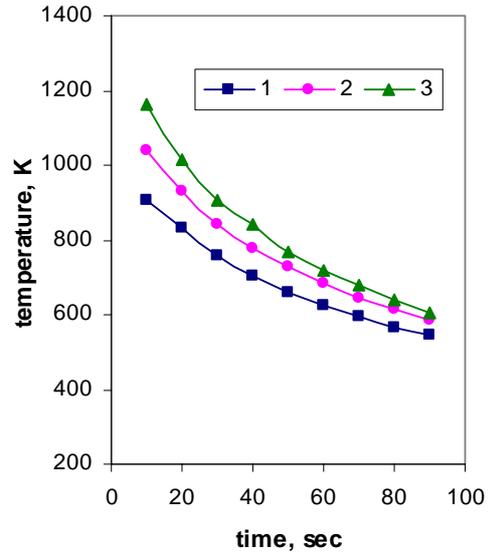
### RESULTS

Typical temperature - time plots measured for the fixed distance probe are shown in Fig. 1. The probe temperature was measured until steady state was reached. After reaching the steady state, plasma spray was stopped and probe temperature variation was measured keeping the probe in a vacuum chamber. Fig. 2 shows temperature - time dependences after the plasma spray process had been turned off. These curves are essential for the evaluation of the absorption factor of the probe surface. It was found that temperature - time dependencies were sensitive to the applied plasma power as well as distance between the probe and nozzle of the plasma gun. In Fig. 3 the temperature - time measurements for the different distances between the probe and plasma gun are presented. A steep growth of the probe temperature with the decreasing of distance was found.

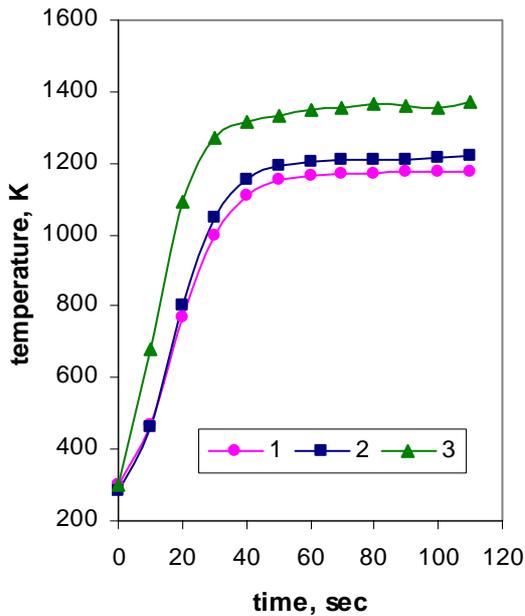
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**Fig. 1.** The temperature - time dependences (curves 1 – 3) of a probe ( $m = 2.7$  g,  $S = 864$  mm<sup>2</sup>) placed in a plasma spray torch measured for the constant voltage (31 V) and different arc discharge currents (curve 1, 440 A; curve 2, 620 A; curve 3, 800 A). The Ar gas flow ( $F = 34$  l/min) and distance ( $d = 0.23$  m) during plasma spray were constant



**Fig. 2.** The temperature - time dependences (curves 1 – 3) of a probe ( $m = 2.7$  g,  $S = 864$  mm<sup>2</sup>) after plasma torch had been turned off. The experiment conditions before plasma turning off were (curve 1, 440 A; curve 2, 620 A; curve 3, 800 A) the same as they are presented in Figure 1



**Fig. 3.** The temperature - time dependences (curves 1 – 3) of a probe ( $m = 2.7$  g,  $S = 864$  mm<sup>2</sup>) placed in a plasma torch measured for the constant arc discharge power (voltage  $U = 31$  V, current  $I = 620$  A) and different distances (curve 1, 0.23 m; curve 2, 0.20 m; curve 3, 0.17 m). The Ar gas flow ( $F = 34$  l/min) was fixed

## INTERPRETATION OF THE DATA

As a result of the interaction between the arc plasma and the probe (or object to be coated) the temperature of the probe increases during plasma spray deposition process. Using the principle of conservation of energy for the thermally isolated probe (heat losses due to thermal

conductivity are minimized) placed in a plasma torch one can write [7]:

$$-mcdT - \varepsilon\sigma T^4 Sdt + K_e j_e E_e Sdt + K_i j_i E_i Sdt + K_n j_n E_n Sdt = 0 \quad (1)$$

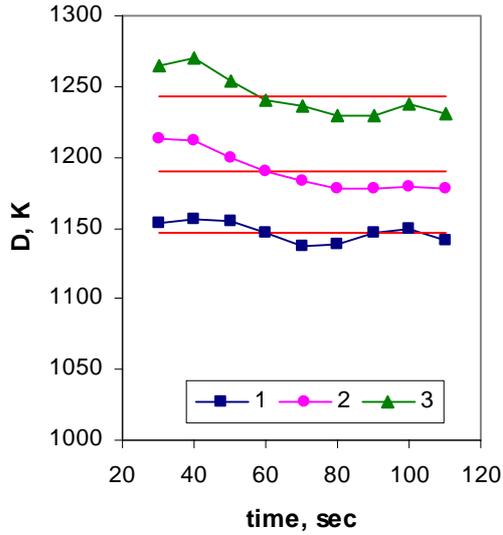
where  $m$  is the probe mass,  $c$  is the specific heat capacity,  $T$  is the object temperature,  $\varepsilon$  is the absorbtion factor (for gray body  $\varepsilon < 1$ ),  $\sigma$  is the Stefan constant,  $S$  is the area of the probe surface,  $t$  is the time,  $j$  (m<sup>-2</sup>s<sup>-1</sup>) is the flux density of the particles,  $E$  is the energy of the incident particle,  $K$  is the coefficient of proportionality between kinetic energy transferred to the heat, and the indexes  $e$ ,  $i$  and  $n$  correspond to the fluxes of electrons, ions and neutral hot gas atoms respectively.

Turning off the plasma makes the fluxes of electrons, ions and hot neutral atoms to become equal to zero and the temperature of the object decreases owing to thermal radiation. In this case a solution of Eqn. (1) describing the temperature - time dependence is

$$T^{-3} = \frac{3S\varepsilon\sigma}{mc}t + T_{01}^{-3} \quad (2)$$

where  $T_{01}$  is the steady state temperature (corresponding the moment when the plasma spray had been turned off).

The experimental curves (Fig. 2) plotted as  $T^{-3} = f(t)$  reveals linear behavior at high temperature (over 800 K). Analysis of the temperature vs. time allows one to estimate the absorbtion factor  $\varepsilon$  of a gray body, because the value  $\frac{3S\varepsilon\sigma}{mc} = b$  ( $b$  is the slope of the lines) during the experiment was constant. In this way the absorbtion factor of the Al<sub>2</sub>O<sub>3</sub> ceramic sample surface was found to be  $\varepsilon = 0.73$ .



**Fig. 4.** The calculated parameter  $D$ -time dependences. The experimental data used for the parameter  $D$  calculation was obtained (see Fig. 1) for the constant voltage (31 V) and different arc discharge current (curve 1, 440 A; curve 2, 620 A; curve 3, 800 A ). Distance between the probe and the nozzle of plasma gun was fixed  $d = 0.23$  m

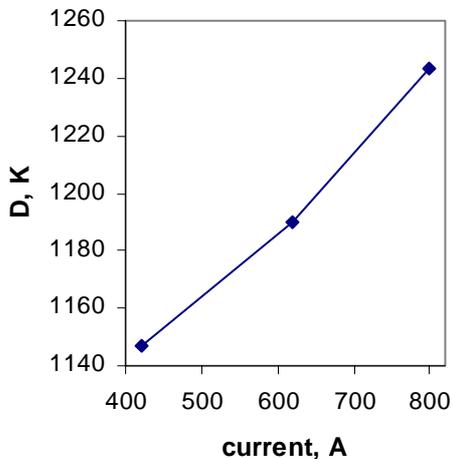
Solution of the Eqn (1) in general is

$$\frac{1}{4D^3} \left[ \ln\left(\frac{D+T}{D-T}\right) - \ln\left(\frac{D+T_{02}}{D-T_{02}}\right) \right] + \frac{1}{2D^3} \left[ \tan^{-1}\left(\frac{T}{D}\right) - \tan^{-1}\left(\frac{T_{02}}{D}\right) \right] = \frac{S\varepsilon\sigma}{mc} t \quad (3)$$

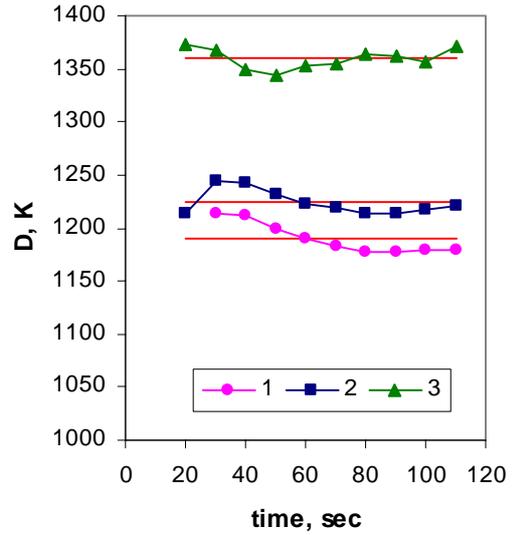
where

$$D = \left( \frac{1}{S\varepsilon\sigma} (K_e j_e E_e + K_i j_i E_i + K_n j_n E_n) \right)^{1/4} \quad (4)$$

$T_{02}$  is the probe temperature at the time  $t = 0$ . The physical meaning of parameter  $D$  may be understood from the definition (4) of this value; it corresponds to the maximum achievable temperature of the object for a fixed arc discharge power.

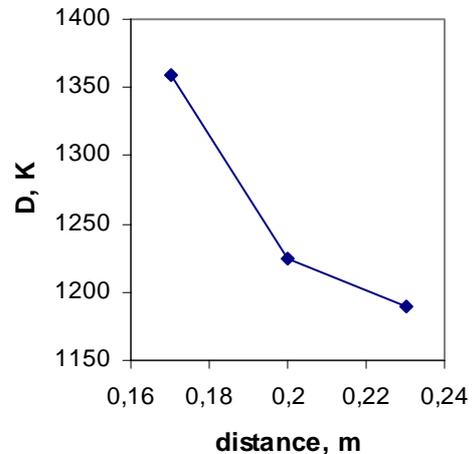


**Fig. 6.** Dependence of parameter  $D$  on the arc discharge current



**Fig. 5.** The calculated parameter  $D$ -time dependences. The experimental data used for the parameter  $D$  calculation was obtained for the constant arc discharge power (voltage  $U = 31$  V, current  $I = 620$  A) and different distances (curve 1, 0.23 m; curve 2, 0.20 m; curve 3, 0.17 m )

The values  $D$  were calculated using Eqn. (3) and experimental data (curves presented in Fig. 1 and Fig. 3). Fig. 4 and Fig. 5 shows the dependence of the parameter  $D$  versus time as well as its average value (dashed line). It should be noted that the average values of parameter  $D$  are close to the steady state probe temperature during plasma spray process. This means that parameter  $D$  does not depend on the time and may be used to describe real temperature conditions of the small-mass object or the temperature field of plasma spray equipment. Besides according to Eqn. (4) the parameter  $D$  does not depend on the mass of the object and it is defined only by the flux transferred by high energy particles – electrons, ions, neutrals, the area and the surface absorption factor of irradiated object.



**Fig. 7.** Dependence of parameter  $D$  on the distance between the probe and the nozzle of plasma gun

In Fig. 6 and Fig. 7 the parameter  $D$  average value dependences on the arc discharge power and distance (between the probe and the nozzle of plasma gun) are presented. The curves are plotted to guide the eye and one can see that the parameter  $D$  increases with increasing the arc discharge power and decreasing distance from the nozzle of plasma gun respectively.

It was found a large deviation of the values  $D$  at low temperatures (below 800 K). In order to avoid these large errors some of the values  $D$  corresponding to 10, 20 sec time intervals were eliminated (see Fig. 4, Fig. 5). This shows that experimental data of the temperature measurement, or predictions of the substrate temperature using Eqn. (3) must be considered with great care because the parameter  $D$  is very sensitive to the above – mentioned experimental conditions.

## CONCLUSIONS

It was demonstrated that the gray body radiation model at high temperatures may be used to describe heating of thermally isolated small-mass object during plasma spray process. It was found that temperature - time dependencies were sensitive to the applied plasma power as well as distance between the probe and nozzle of the plasma gun. A steep growth of the probe temperature with the decreasing of distance was found. It was shown that the highest possible probe temperature (or the parameter  $D$  entering into the temperature - time equation) depends on the energy flux transferred by electrons, ions and neutrals. It was shown the possibility to evaluate the absorption factor of a gray body analyzing temperature versus time curves of the sample cooling process after plasma turning off.

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## REFERENCES

1. **Ouyang, J. H., Sasaki, S., Umeda, K.** Low - Pressure Plasma – Sprayed  $ZrO_2 - CaF_2$  Composite Coating for High Temperature Tribological Applications *Surface and Coatings Technology* 137 2001: pp. 21 – 30.

2. **Khor, K. A., Gu, Y. W.** Thermal Properties of Plasma – Sprayed Functionally Graded Thermal Barrier Coatings *Thin Solid Films* 372 2000: pp. 104 – 113.
3. **Will, J., Mitterdorfer, A., Kleinlogel, C., Perednis, D., Gauckler, L. J.** Fabrication of Thin Electrolytes for Second-Generation Solid Oxide Fuel Cells *Solid State Ionics* 131 2000: pp. 79 – 96.
4. **Schiller, G., Henne, R. H., Lang, M., Ruckdaschel, R., Schaper, S.** Development of Vacuum Plasma Sprayed Thin – Film SOFC for Reduced Operating Temperature *DLR, Germany* 2000: pp. 7 – 12.
5. **Rambert, S., McEvoy, A. J., Barthel, K.** Composite Ceramic Fuel Cell Fabricated by Vacuum Plasma Spraying *Journal of European Ceramic Society* 19 1999: pp. 921 – 923.
6. **Singhal, S. C.** Advances in Solid Oxide Fuel Cell Technology *Solid State Ionics* 135 2000: pp. 305 – 313.
7. **Tamulevičius, S., Babilius, K., Matiukas, A.** Temperature Conditions During Arc Discharge Plasma Deposition of Titanium Nitride *Surface and Coatings Technology* 71 1995: pp. 250 – 253.
8. **Kulkarni, A., Vaidya, A., Goland, A., Sampath, S., Hreman, H.** Processing Effects on Porosity-Property Correlations in Plasma Sprayed Yttria-Stabilized Zirconia Coatings *Materials and Engineering A* 359 2003: pp. 100 – 111.
9. **Friis, M., Persson, C., Wigren, J.** Influence of Particle In-Flight Characteristics on the Microstructure of Atmospheric Plasma Sprayed Yttria Stabilized  $ZrO_2$  *Surface and Coatings Technology* 141 2001: pp. 115 – 127.
10. **Li Chang-Jiu, Sun Bo.** Effects of Spray Parameters on the Microstructure and Property of  $Al_2O_3$  Coatings Sprayed by a Low Power Plasma Torch with a Novel Hollow Cathode *Thin Solid Films* 450 2004: pp. 282 – 289.
11. **Jin, Z.-H.** Effect of Thermal Property Gradients on the Edge Cracking in a Functionally Graded Coating *Surface and Coatings Technology* 179 2004: pp. 210 – 214.
12. **Dargis, R., Tamulevičius, S., Slapikas, K.** Investigation of Plasma Properties During Low – Pressure Plasma Spraying *Medžiagotyra (Materials Science)* 2 (9) 1999: pp. 66 – 68.