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DEPOSITION OF TITANIUM COATINGS ON THE AlN CERAMIC SUBSTRATES BY THE D-GUN SPRAYING METHOD

T. Chmielewski

Dr inz.

D. Golański

Dr inz.

W. Wysoczański

Dr inz.

Warsaw University of Technology, Warsaw, Poland

This paper presents the selected investigation results of the D-gun spraying method applied to the deposition of titanium coatings on the ceramic substrates. The process of D-gun thermal spraying has been characterized. The presented results cover the microstructure of deposited metallic coatings on the ceramic substrate, microhardness profile across the coatings and residual stress analysis. The technology requirements of the D-gun spraying method have been developed taking into account the service properties of deposited coatings.

titanium coating, D-gun spraying method, ceramic substrate

1. Introduction. Thermal spraying of metals onto ceramic substrates has the potential to become cheaper processing method comparing to common but expensive and complex techniques used for ceramic metallization. The proper application of the thermal spraying method can provide many savings when used in electro-technical, electronic, and electron industry. The metallization of advanced ceramic materials is often necessary in the advanced science and technology. But joining of ceramic to metals reveals several problems resulting from the differences in material properties.

The ceramic-to-metal joints are used nowadays in many areas:

— optoelectronics (multilayer ceramic-metal substrates, housing for semiconductor digital and neon devices, housing and other parts used in the liquid crystal technique),

— investigation of fuel cell (non-conducting fuel barriers made of βAl_2O_3 and austenitic steel, conducting fuel barriers made of ZrO_2 joined with nickel-chromium steels),

— laser techniques,

— semiconductor industry (housing for diodes and thyristors and microwave semiconducting elements),

— lamp industry (elements of microwave lamps, delay lines made of alumina ceramics and molybdenum),

— ultra high vacuum techniques (current passages, windows made of sapphire and copper alloys)

The metallic layers thermally sprayed on the ceramic substrates could also be used as an intermediate layers applied for brazing or friction welding of brittle materials.

2. Detonation spraying. There are several methods of thermal spraying used in surface engineering industry. The choice of a proper method depends on the requirements for surface materials, the possibility of their application for a given coating and substrate materials, the level of coating adhesion and the costs and availability of the spraying technique in a specified condition.

The detonation gun technique (HVOF D-Gun) is based on the controlled detonation of the gas mixture (mostly acetylene with oxygen) started by the electric ignition between the electrode sparks. The detonation of the gas generates the shock wave heating each particle of coating material and accelerates them in a special gun up to 1200 m/s [1]. Due to the high temperature of the heat source and high kinetic energy of transported particles the deposited coating is characterized by the even, well compressed and multilayered structure. The structure of the deposited surface layer may change depending on the kinetic energy of the particles [3].

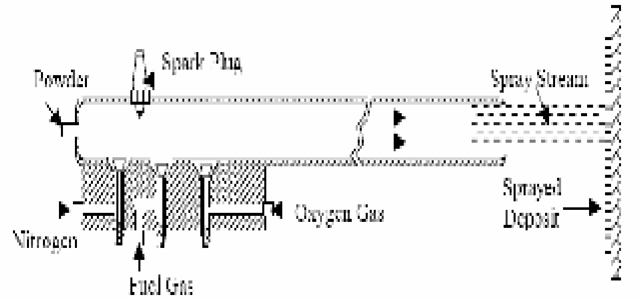

Fig. 1. The scheme of D-GUN thermal spraying method [2]

The scheme of the equipment used for the D-Gun spraying is shown in fig. 1. The coating layer is built up in a discreet manner e.g. each shot (detonation) make the heated particle to clutch to the substrate on a small area covering the diameter of around 20-25 mm and the thickness about 10 mm.

The detonation frequency is controlled in the range between 1 and 12 Hz. Other main processing parameters include the energy gas pressure, oxygen pressure, powder material fluidization gas pressure (nitrogen), spraying distance, length of detonation tube and the velocity of gun movement against deposited substrate.

The D-Gun spraying technique has been selected in this research because it allows generating high kinetic energy of transporting particles which makes the strong adhesion between coating and substrate reaching up to 80 MPa [2]. The discreet manner in which particles are transported in this method allows the coating/substrate system to heat up to much lower temperatures comparing to the other thermal spraying methods.

3. The residual stress analysis in metallic coatings deposited onto ceramic substrate. The state of residual stresses together with the adhesion level of the coating is very important and affects the service properties of deposited layers. The distribution of residual stresses have the important effect on the coating adhesion because the unfavorable stress state may lead to the coating delamination or cracking during production and service life. The source of the residual stresses in coating/substrate system results from the difference in thermal, physical and mechanical properties of applied materials within a given temperature gradient.

During the deposition process the coating material is heated up to a certain temperature (depending on the process conditions) which decreases after the process is completed. Upon cooling of the coating an uneven shrinkage develops within coating and substrate which leads to formation of thermal residual stresses at room temperature. As a result the compressive or tensile stresses may exist in coating/substrate system with the level exceeding the strength of materials which may lead to the coating self-destruction.

In the selection process of compatible coating and substrate materials we should take into account their thermal, physical and mechanical properties (e.g. thermal conductivity, coefficient of thermal expansion, the Young's modulus) that mostly affect the residual stress state.

The residual stresses generated during the first stage of thermal spraying can be roughly estimated by the following formula applied within the elastic range [2]:

$$\sigma_q = \alpha_c \left(T_m - T_c\right) E_c \,, \qquad (1)$$

where: a_c – coefficient of thermal expansion of a coating, T_m – melting temperature of a coating, T_c – temperature of a substrate, E_c – Young Modulus of a coating,

The presented equation is valid when heated material particles hit the substrate material whose temperature is lower that the coating temperature. We may suppose that an initial preheating of the substrate material could help to reduce the stress level in this system.

Within the next stage of this process, during cooling of deposited layer the residual stresses are generated resulting from the mismatch of material properties.

We may calculate these stresses by a simplified equation [4]:

$$\sigma_c = \frac{\left[E_c\left(T_f - T_r\right)\left(\alpha_c - \alpha_s\right)\right]}{\left[1 + 2\left(\frac{E_c t_c}{E_s t_s}\right)\right]} \,, \qquad (2)$$

where: T_f – temperature of deposited layer, T_r – ambient temperature, a – coefficient of thermal expansion of a coating (c) and substrate (s), E – Young Modulus of a coating (c) and substrate (s), t – thickness of a coating (c) and substrate (s). The final stress level in the coating/substrate system will be the sum of the stresses created in the spraying phase and stresses generated during cooling of the coating to the room temperature. The stress level also depends on the mutual thickness relation between coating and substrate. The formula presented above are used as a simplified solution of residual stresses generated in a plate model composed of a uniform coating and substrate materials and is valid for isotropic material within the elastic range. It is also to note that the substrate temperature depends on the coating thickness according to the Fourier law [6]:

$$T_s = T_f - \frac{q t_c}{kA} \,, \qquad (3)$$

where: T_s – temperature of a substrate, T_f – temperature of a deposited coating, q – the velocity of heat transfer, t_c – thickness of a coating, k – thermal conductivity of a substrate, A–– substrate area.

The more advanced and precise methods of stress analysis in such systems are based mostly on the finite element method. This technique has been performed to study thermal residual stresses in a numerical model of Ti coating deposited on AlN ceramic. The substrate material consisted of AlN disk shape samples with 100 mm diameter and 2 mm height.

The finite element numerical analysis has been preformed by using the commercial code LUSAS FEA. The problem has been solved as a nonlinear, thermal, elastic-plastic with the isotropic von Mises yield criteria. It was assumed that coating and substrate are cooled down together loaded with different temperature gradients (DT=1000°C for coating and 250°C for substrate).

The calculation results show that the tensile residual stresses s_x (radial) are formed in the whole coating while the

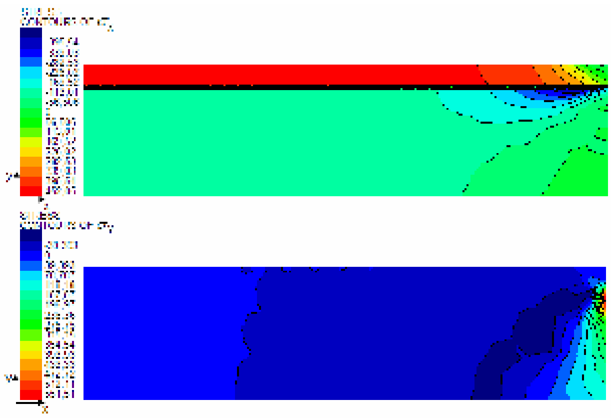

Fig. 2. Distribution of radial (s_x) and axial (s_y) stresses in Ti/AlN model calculated by FEM (Ti thickness=0.2 mm)

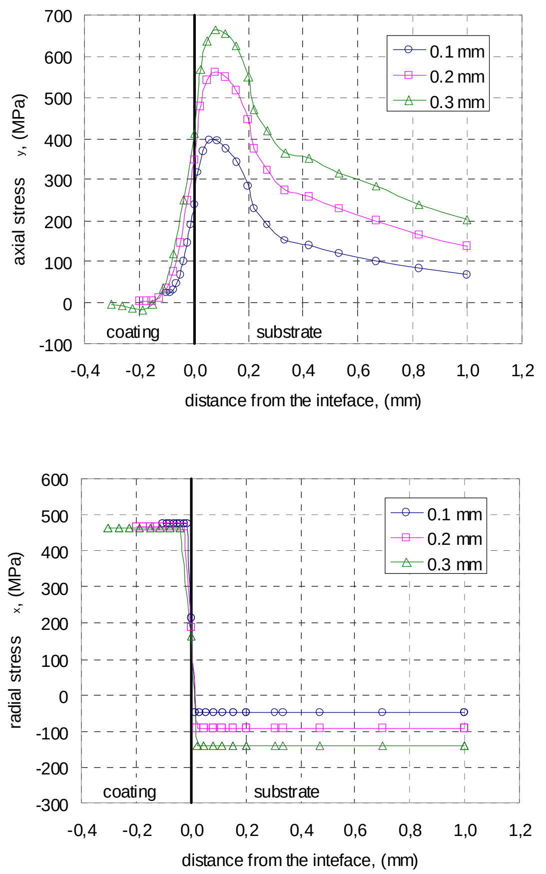

Fig. 3. Axial (s_y) and radial (s_y) stress profiles across the coating thickness calculated in the Ti/AlN system for three coating thicknesses (left- along line 1, right - along line 2 in fig. 2)

tensile axial stresses s_y are concentrated in a small region laying in the substrate material near the model edge towards the interface with Ti layer (fig. 2).

The changes in the residual stress profile can be seen on the model edge. In this region the axial stresses are dominating and they have very similar distribution to the maximal principal stresses. Also, the tensile stress concentration can be visible within the ceramic part (about 0.1 mm from the interface line) as well as within the coating material near the bonding line with ceramic substrate (fig. 3).

The axial stress level is increasing with the increase of the coating thickness and may result in coating delamination starting at the edge of the model. Additionally, high tensile stresses in the ceramic part can also be the source of cracking initiation in this area. From fig. 3 we may see that the maximal axial s_y stress has increased almost 60% with the increase of the coating thickness from 0.1 to 0.3 mm both in the AlN substrate and in the Ti coating near the interface.

From the fig. 3 it is seen that the axial stresses are diminishing in areas far from the coating/substrate interface line. The plastic strains that developed in the metallic part helped to partially reduce the stress level in this area. The magnitude of the stress redistribution depends on the yield stress of the applied coating material.

4. The microstructure of deposited coatings. Fig. 4 presents the microstructure of titanium coating sprayed onto the AlN substrate. It is seen that the structure of deposited layer is uniform and consists of multilayered squeezed and tightly packed particles of coating material. The coating shows good adhesion to the substrate filling each visible surface micro roughness. The measured thickness of the deposited coating shown in fig. 4 varied in the range from 185 up to 200 mm.

It is also possible to obtain smaller thicknesses of deposited titanium coatings as shown in fig. 5 where the layer thickness ranged between 55 and 65 mm. We may also see the enlarged area of the interface between titanium and aluminum nitride.

Also, fig. 5 presents the results of AlN-Ti interface analysis observed by the scanning microscope (SEM) in the cross-section perpendicular to the surface. The linear element distribution profiles indicate that there was a mutual penetration of AlN and Ti which was partially confirmed by a good adhesion of titanium to ceramic substrate.

5. Conclusions. The applied D-Gun technique has shown the potential to become one of the unique methods for a successful thermal spraying of metallic materials on the ceramic substrates due to relatively small level of heat inputted into the substrate. The deposited coating particles are receiving high kinetic energy helping them to reach good adhesion to

Fig. 4. The microstructure of titanium coating deposited onto the AlN substrate by the D-gun technique (ґ100)

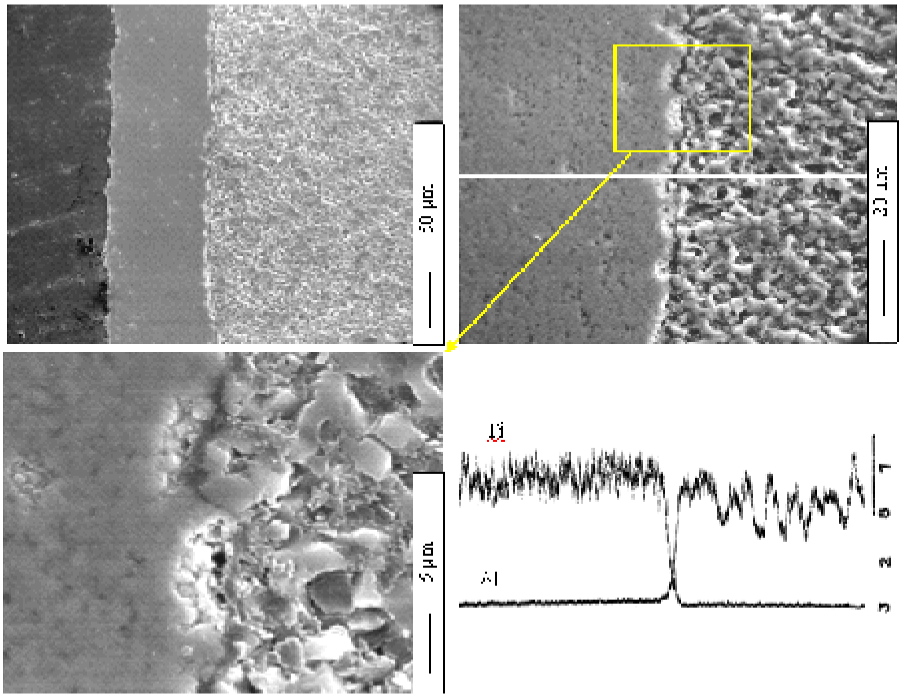

Fig. 5. The microstructure of titanium coating deposited onto the AlN substrate by the D-gun technique with linear distribution of Ti and Al elements

the substrate. To increase the service life of the coating we shall put a special attention to ensure low level of residual stresses in the coating/substrate system.

The selection of the coating and substrate materials should be performed in accordance with the main criteria put on the mismatch of material properties (e.g. thermal conductivity, thermal expansion coefficient, Young modulus) which should be kept at minimum. Also, low yield stress of metallic coating material may help to minimize the residual stresses through the stress redistribution. Other techniques which could possibly reduce the residual stresses in the thermally sprayed coating/substrate system may utilize the substrate preheating, which would also allow the increase of the free energy of this system [7] and reduce the activation energy required for the thermal spray processing. The thickness of sprayed coatings could be another important factor affecting the strength of the coating and should be kept at low value.

By utilizing the above aspects in the design of materials for thermal spraying it could be possible to perform the metallization of advanced ceramic materials. This approach would allow depositing coatings that may be applied in the surface modification of ceramic components or play the role of intermediate layers for joining ceramics to metals or metal matrix composites.

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Т. Хмілєвскі, Д. Голаньскі, В. Височанський

Нанесення титанових покриттів на керамічну основу методом розпилення

Варшавський технологічний університет, м. Варшава, Польща

У статті наведені результати дослідження застосування D-gun методу розпилювання для нанесення титанових покриттів на керамічну основу. Охарактеризований процес D-gun термічного розпилювання. Наведені результати охоплюють мікроструктуру нанесених металічних покриттів на керамічну основу, мікротвердість профілю вглиб покриття і аналіз залишкових напружень. Технологічні можливості D-gun методу розпилювання для нанесення покриттів розвинені з врахуванням службових характеристик покриттів.