Proceedings of the III Int. School and Symposium on Physics in Materials Science, Jaszowiec '98

# INFLUENCE OF PLASMA DYNAMICS ON MATERIAL SYNTHESIS PRODUCT OF IPD PROCESS

M. Rabiński $^{a*}$  and K. Zdunek $^{b}$ 

<sup>a</sup>Department of Plasma Physics and Technology
The Andrzej Sołtan Institute for Nuclear Studies
05-400 Otwock-Świerk, Poland
<sup>b</sup>Faculty of Materials Science, Warsaw University of Technology
Narbutta 85, 02-524 Warsaw, Poland

In the present work we discuss recent studies concerning plasma dynamics influence on the material synthesis product of the impulse plasma deposition process. Conditions favourable for evaluation of the Rayleigh–Taylor instability on the current sheet surface were found during the computational studies of plasma movement in the coaxial accelerator. Appearance of this phenomenon explains non-uniform phase composition and morphology of coatings. By modifying the design of the plasma accelerator, we succeeded in reducing substantially Rayleigh–Taylor instability and in obtaining  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> coatings instead of common  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>.

PACS numbers: 07.05.Tp, 52.30.-q, 52.35.Py, 52.75.Rx

#### 1. Introduction

In surface engineering plasma can be used as an efficient source of mass and energy for the synthesis and deposition of various materials as layers. During the impulse plasma deposition (IPD) process [1] plasma is generated within the working gas due to a high-voltage high-current discharge, ignited and accelerated within an interelectrode region of a coaxial accelerator [2, 3]. System of two coaxial metal electrodes (a cylinder and a rod, insulated from one another by a ceramic material), in which the plasma is accelerated by the Lorentz force, is the most efficient solution for surface engineering. At the outlet of the accelerator, plasma consists of the ionized atoms of the working gas and the eroded electrode material. In addition, these products may react chemically, including the phase nucleation. Schematic diagram of the complete apparatus for the IPD synthesis of coatings can be found in Fig. 1.

<sup>\*</sup>e-mail: rabinski@cx1.cyf.gov.pl

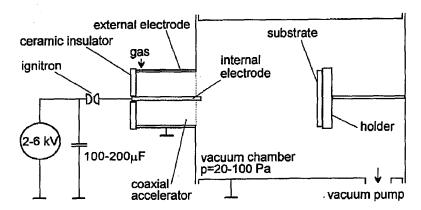


Fig. 1. Scheme of the impulse plasma coaxial accelerator.

A short lifetime ( $< 100~\mu s$ ) of the plasma and its high ionization are the characteristic features of this technique. Coatings made of diamond, titanium nitride, multicomponent metallic alloys and aluminum oxide, adhere well to the substrates, although no external source heats them during the plasma process. IPD has been implemented on the industrial scale at the Steel Works of Stalowa Wola, Poland, for depositing TiN coatings on cutting tools.

The study presents the results of physico-mathematical modelling of the plasma dynamics in the coaxial accelerator under the IPD process conditions. The modelling throws new light on the experimental results obtained earlier, suggesting that the previous interpretation should be modified. In this study we concentrated our attention on the influence of plasma dynamics on material synthesis product. Consequences of Rayleigh-Taylor instability on plasma configuration explain observations of a toroidal ring in the front of the central electrode. By modifying the design of the plasma accelerator we succeeded in reducing this instability. The experimental observations proved that the  $\gamma \to \alpha$  phase transformation takes place in aluminium oxide after change in electrode geometry.

### 2. Dynamics of impulse plasma

In our earlier study [3] we proposed the physical model of dynamic phenomena in coaxial accelerator used in surface engineering for IPD synthesis. According to our model, at the initial stage of the process, an axially symmetric electric current sheet forms on the insulator surface. Current flowing within this layer induces an angular magnetic field behind. The Lorentz force accelerates the current sheet in the axial direction, sweeping the gas over its surface and leaving vacuum behind it. Magnetic field behind the current-conducting plasma sheet functions as a magnetic piston. Because of plasma being continuously accelerated, a shock wave forms in front of it, preionizing the working gas. Under the action of the electric current flowing through gas, it is transformed into plasma. Paraboloidal shape of the magnetic piston induces plasma flow along the sheet to the outside, much like the snow plow (see Fig. 2).

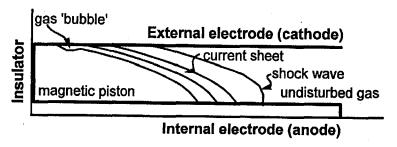


Fig. 2. Schematic pattern of the discharge region.

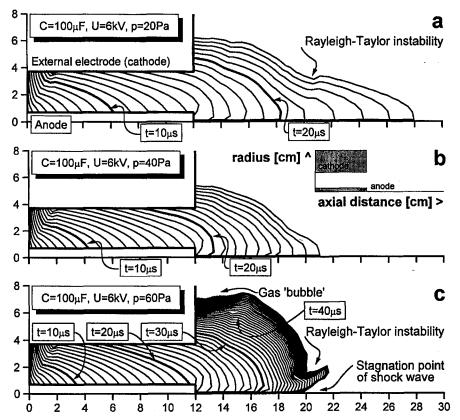


Fig. 3. Time evolution of the current sheet in the IPD accelerator (for different parameters of discharge). Current sheet position plotted with 1  $\mu$ s interval.

After the discharge reaches the end of the internal electrode, we have the phase of an arc discharge active along the system axis, in which the plasma is pinched, forming what is known as the "plasma focus".

Obviously neither the current sheet nor magnetic piston edge is an infinitely thin surface. The detailed structure of region between shock and magnetic piston can only be described by the solution of a complete magnetohydrodynamic model. The simplified two-dimensional "snow plow" model used assumes that all the mass swept up is compressed into infinitely thin layer immediately behind the shock. Thus, the magnetic piston edge, the current sheet, and the shock form the same interface. The plasma discharge region is reduced into an infinitely thin layer.

Figure 3 shows the dynamics of the electric current sheet, which spreads out within both the interelectrode space and behind the front face of the plasma accelerator electrodes. The shape of current sheet was obtained using the "snow plow" computer code [3] at various parameters of the plasma generation process. One can observe in the diagrams the characteristic features of the plasma dynamics:

- a) A toroidal reservoir of gas ("bubble") near the external electrode of the plasma accelerator. This structure is the result of the working gas sweeping by the moving layer.
- b) The Rayleigh-Taylor instability forming behind the front face of the electrodes, as a result of which the plasma front is composed of the two zones: one that spreads out along the system axis and, the other, above it, having the form of a torus of dense plasma. The toroidal region is continuously supplied with the gas swept away by the current layer, which enhances the instability.
- c) For a low energy of the electric discharge and a high gas pressure, additional dense plasmoid may form along the electrode axis at the stagnation point of the proceeding shock wave.

Thanks to the computer simulation, we could not only identify the characteristic features of the plasma dynamics but also find that it depends essentially on the plasma generation conditions.

## 3. Influence of Rayleigh-Taylor instability on IPD coatings

The described above structure of the plasma behind the electrode front faces is very important for the quality of the IPD coatings produced. Earlier studies on the products of the impulse plasma synthesis [2] and also the spectral examinations of the plasma have suggested that each individual plasma jet (a plasmoid) consists of the two fragments: one concentrated near the system axis, in which the plasma is isothermal, and the external portion which is highly unbalanced. Conditions prevailing at the boundary between these two parts are considered to be the most advantageous for the IPD synthesis of materials. Combining the results of computer simulation with the assumed plasmoid structure permits to anticipate the geometry of the deposition process. Also, the coating dependence on the parameters of plasma generation could be presumed. This however requires further research aimed at correlating the simulation with the experimental observations.

Investigations into this subject so far performed suggest that geometry of electrodes may lead during the plasma generation to the Rayleigh-Taylor instability on the surface of the current layer. Presence of this instability explains experimental observations of a toroidal ring in the front of the central electrode. On the other hand, configuration of plasma in the front of the electrode system is very important for the quality of the IPD coatings. It was found previously that

coatings obtained have nonuniform phase composition and morphology when the regions on the accelerator axis and away from it are compared. For example, the content of the solid solution Co(Cr, Al) increases with axial distance while the Cr and (CrCo) phases mostly occur on the axis [4]; in the regions close to axis coating of diamond is dominated mostly by graphite [5]; during TiN deposition one can find Ti and Ti<sub>2</sub>N mostly in the centre [6].

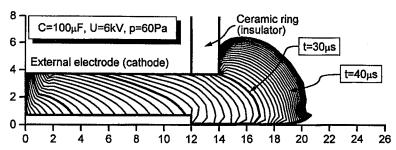


Fig. 4. Time evolution of the current sheet in the IPD accelerator with ceramic insulator at the external electrode front face (discharge parameters the same as in Fig. 3c).

By modifying the design of the plasma accelerator, we succeeded in reducing substantially the Rayleigh-Taylor instability and also in limiting the erosion zone of the internal electrode to the plane of its free end. The ceramic ring installed at the front face of the external electrode changes the geometry of plasma accelerator. This restricts the "climbing" of the electric current sheet upon the metallic wall of the vacuum chamber and modifies plasma dynamics. Figure 4 shows the computer simulation of the plasma motion for this situation. Comparing with the diagram of Fig. 3c, this shown in Fig. 4 suggests that the presence of the ceramic insulator reduces the tendency for the Rayleigh-Taylor instability to occur in the region behind the electrode front faces. As a result, the gas outflows undisturbed along the surface of the magnetic piston towards the external electrode, by that reducing the plasma energy dissipation. Since in the region preceding the zone of electric discharge there are no instabilities, the proceeding shock wave becomes stronger. Thus, the impulse heating is more intensive at the substrate surface on which the coating is condensed. This effect may be experimentally confirmed by the observation that, on the substrate not heated from any external source, the phase transformation of the Al<sub>2</sub>O<sub>3</sub> coating material from the metastable  $\gamma$  phase into the stable  $\alpha$  phase occurs only when the ceramic ring has been installed at the accelerator outlet [7].

#### 4. Conclusions

The mathematical model, adopted in the present study, allows effective modelling of plasma dynamics in the IPD accelerator. The computer simulation allows to anticipate the spatial and time distributions of the electric current layer dependent on the parameters of the plasma generation process. Numerical model also helps to interpret the experimental results. The modelling throws new light on the

experimental results obtained earlier, suggesting that the previous interpretation should be modified.

The correlation between plasma dynamics and material synthesis product of the IPD process has been explained. By proper modification of plasma motion using the ceramic insulator placed in the front face of the external electrode, we succeeded for the first time in reducing substantially the Rayleigh-Taylor instability and in obtaining  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> coatings instead of common  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>. Moreover, the proposed explanation of this phenomenon modifies the earlier interpretation of the ceramic ring influence upon the phase composition of the aluminium oxide coating produced. Till now, the ring was considered to function as a stationary heat source, whereas the results obtained with the proposed model suggest that this insert influences on plasma dynamics. The presence of ceramic ring reduces the instabilities on the current sheet and by that intensifies the effect of impulse heating of the substrate surface by the plasma itself.

### Acknowledgment

Some part of this study was supported by the Committee for Scientific Research under the contract no. 7T08C 050 12.

### References

- [1] M. Sokołowski, A. Sokołowska, J. Cryst. Growth 57, 185 (1982).
- [2] A. Rusek, K. Zdunek, Vacuum 39, 55 (1989).
- [3] M. Rabiński, K. Zdunek, Vacuum 48, 715 (1997).
- [4] K. Zdunek, J. Mater. Sci. 26, 4433 (1991).
- [5] A. Michalski, J. Mater. Sci. Lett. 3, 505 (1984).
- [6] A. Sokołowska, A. Olszyna, A. Michalski, K. Zdunek, Surf. Coat. Technol. 47, 144 (1991).
- [7] M. Rabiński, K. Zdunek, in: Sixth Int. Conf. on Plasma Surface Engineering, paper WBP30, Garmisch-Partenkirchen 1998, submitted to Surf. Coat. Technol.