Simulation of multilayer coating growth in an industrial magnetron sputtering system

Simulacija rasti večplastnih prevlek v industrijski napravi za magnetronsko napprševanje

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Abstract: Layered coatings are mainly prepared by physical vapor deposition such as magnetron sputtering. In industrial deposition systems layered coatings (e.g. multilayer or nanolayered coatings) are produced by the rotation of the substrates along the spatially separated targets. In order to assure uniform deposition on all parts of the substrate with complex geometry (e.g. tools), two- or three-fold rotation is typically applied. Such rotation is similar to the planetary rotation. A consequence of the planetary rotation are layered coatings whose structure depends on the type of the rotation. In this paper we describe a model of the sputter deposition in the deposition systems with the planetary rotation. Such model helps us understand the influence of the rotation on the layer structure of the coatings. Results of the model for different types of the substrate rotation are presented. In addition, we prepared TiAlN/CrN nanolayered coatings in an industrial magnetron sputtering system and compared their layered structures with the calculated ones. The comparison confirms the accuracy of the developed model.

Izvleček: Večplastne prevleke pripravljamo s fizikalnimi postopki nanašanja iz parne faze (PVD), kot je magnetronsko naprševanje. V industrijskih napravah večplastne prevleke pripravimo z vrtenjem podlag okoli prostorsko ločenih tarč. Podlage imajo v splošnem kompleksno geometrijo (npr. orodja), zato se morajo vrteti okrog dveh ali treh osi, pri čemer je vrtenje podobno planetarnemu vrtenju. Tako zagotovimo enakomeren nanos prevleke na vse dele

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orodja. Rezultat planetarnega vrtenja so različne večplastne prevleke, katerih struktura je odvisna od načina vrtenja. V članku opisujemo model nanašanja večplastnih prevlek, ki smo ga razvili za magnetronsko naprševanje v industrijskih napravah s planetarnim vrtenjem. Model nam pomaga razumeti vpliv različnih parametrov na večplastno strukturo prevlek. V članku predstavljamo rezultate modela za različne vrste vrtenja. Za preverjanje natančnosti modela smo v industrijski napravi CC800/9 (CemeCon) pripravili nanoplastne prevleke TiAlN/CrN in njihove večplastne strukture primerjali z izračunanimi strukturami. Rezultati potrjujejo točnost modela.

Key words: modeling, layered structures, PVD, magnetron sputtering, TEM

Ključne besede: modeliranje, večplastne strukture, PVD, magnetronsko naprševanje, TEM

INTRODUCTION

Hard coatings are thin films, which are deposited on the tools and components in order to improve hardness, friction, wear and corrosion reistance of the surface. In this way the lifetime of the tools is prolonged, therefore the productivity is enhanced. Moreover, the use of hard coatings reduces the consumption of lubricants and often enables machining of new materials. Hard coatings are commonly prepared by physical vapor deposition (PVD), which offers an easy way of depositing coatings in a form of a single layer or multilayers. Layered structures are prepared by alternately depositing two or more different materials. They are composed of a few or up to several hundred layers. The thickness of the individual layers can vary from a few atomic layers up to micrometers, the structures can be periodic or aperiodic. When the thickness of the individual layers is in the nanometer range, the term nanolayered coatings is used.

Unique property of the nanolayered coatings is an extremely high hardness, which is much higher than the hardness of individual layers[1, 2]. In 1987, HELMERSSON et al.[3] published a paper in which they reported on drastic enhancement of hardness in TiN/VN nanolayered coatings. They showed that the hardness of the coating exceeded 50 GPa for the thickness of layers ≈2–4 nm, which is much more than the hardness of a single layer TiN (≈22 GPa) and VN (≈16 GPa) coatings. High hardness was interpreted as a consequence of numerous interfaces between the individual layers and the small thickness of the layers[4]. Interfaces obstruct the movement of dislocations, while a few nanometers thick layers reduce the formation of new dislocations. Consequently, hardness of nanolayered coating can be higher than the hardness of the second hardest material, the cubic BN.

Nanolayered coatings are mainly prepared by magnetron sputtering or cathodic arc evaporation[5]. In laboratory deposition systems, nanolayered coatings are usually formed by sequential switching between two target sources[3], whereas in industrial deposition systems, nanolayered coatings are formed when the substrates rotate along spatially separated targets[6]. In the industrial deposition systems, the substrates have to rotate around two, three or even four axes in order to insure uniform coating on all parts of the substrates with complicated geometry such as tools. Rotation around different axes causes periodic and aperiodic layer structures. The layer structure depends on the number of rotational axes, revolution time around the individual axes, initial position of the substrates and on the target arrangement. The nanolayered coatings prepared in the same batch therefore have different layer structures.

The objective of our work was to develop a model of a sputtering process in an industrial deposition system with planetary rotation and to calculate the layer structure of the coatings for different parameters of the deposition. This is important because the parameters, such as planetary rotation, cause considerable variations in the thickness of individual layers and thus can influence the mechanical properties of nanolayered coatings. In this paper we are presenting the model and the results of the model for different types of the rotation. In addition, we prepared samples for transmission electron microscope and compared calculated layer structures with deposited TiAlN/CrN nanolayered coatings.

Industrial physical vapour deposition system

Nanolayered coatings are usually prepared by magnetron sputtering. A schematic top view of the industrial magnetron sputtering system CC800/9 from company CemeCon is shown in Figure 1. The deposition system has four planar magnetron sources with dimensions 500 mm × 88 mm. The sources are arranged in the corners of the rectangle. The substrates can be positioned at different heights. The turntable has the possibility of a 3-fold planetary rotation; the first axis of rotation is in the centre of the turntable while the substrate towers rotate around the second axis, which is positioned 137 mm away from the first axis. The substrate towers rotate around the first and the second axis. The rotation around the third axis is not continuous but is achieved by a switch fixed on the rod. For every rotation of the substrate tower around the second axis, the switch turns the sample for a specific angle. The distance from the second axis to the third axis is 58 mm. The revolution time of the turntable can be adjusted from 38 s to 97 s, while the revolution time of the substrate towers is determined by the gear ratio between the turntable and the substrate tower; this ratio is 100/37.

For the experiments TiAlN/CrN nanolayered coatings were prepared by three types of the rotation; 1-, 2- and 3-fold. Coatings were deposited on D2 tool steel, hard metal and silicon substrates. Prior to deposition the samples were ground and polished, ultrasonically cleaned and ion-etched in deposition system. Substrate temperature during the deposition was ≈450 °C, power on the Cr targets was 4.5 kW and on the TiAl targets 9.5 kW. A mixture of nitrogen, argon and krypton gases was used with flow rates of (70, 150, 100) mL/min, respectively. Total gas pressure during deposition was 0.6 Pa and a DC bias of –100 V was applied to the substrates. One rotational cycle of the turntable was 97 s while the deposition time was 125 min.

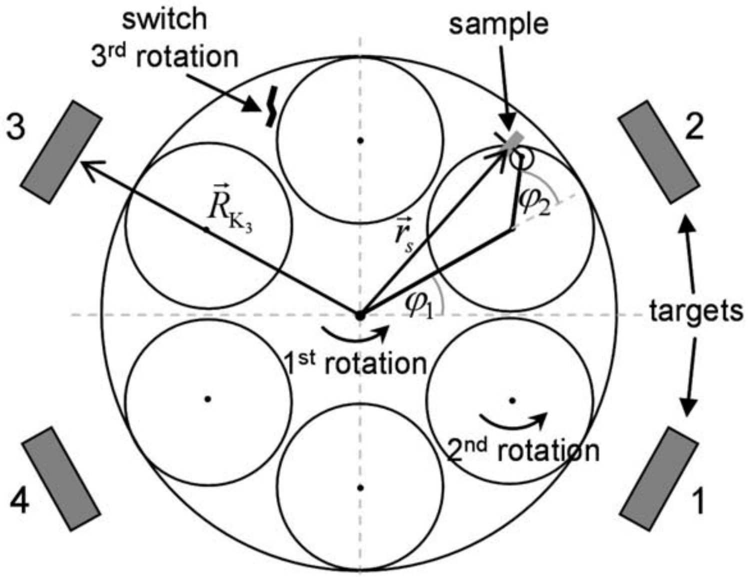

Figure 1. Schematic top view of the CC800/9 (CemeCon)

Nanolayered coatings were prepared in cross-section for the transmission electron microscopy. The samples were first cut into small pieces, glued face-to-face, fixed into brass disk holders, mechanically polished to ≈100 µm, thinned to 20 µm by dimpling and ion milled to electron transparency. Investigations were carried out on field-emission electron-source high-resolution transmission electron microscope JEOL 2010F operated at 200 keV.

Modeling of multilayer growth

The layer structure of the coating is obtained by calculating the deposition rate from a magnetron source on the surface of a rotating substrate. The deposition rate from a particular target depends on the distance from the target, the orientation between the target and the substrate and on the angular distribution of a particle flux from the target. The particle flux was modeled by two point sources where each source has a cosine angular distribution while the intensity falls with square of the distance. Similar model was introduced by Rother et al. [7–9]. In order to give a realistic description of the deposition process a shading of the particle flux by the batching material was also considered. The model assumes the following:

1. The deposition rate on the surface of the sample depends on the particular position and orientation of the substrate. The position and the orientation of the substrate (e.g. the trajectory) are defined by the planetary rotation.
2. The planetary rotation is described by the equations

$$\boldsymbol{r}_{\mathrm{s}}(t)=\left(\sum_{i=1}^{N}\left(R_{\mathrm{i}}\cos\left(\sum_{j=1}^{i}\left(\frac{2\pi}{t_{\mathrm{j0}}}t+\varphi_{\mathrm{j}}\right)\right)\right),\; R_{\mathrm{i}}\sin\left(\sum_{j=1}^{i}\left(\frac{2\pi}{t_{\mathrm{j0}}}t+\varphi_{\mathrm{j}}\right)\right),h_{\mathrm{s}}\right) \tag{1}$$

$$\boldsymbol{n}_{\mathrm{s}}(t)=\left(\sum_{j=1}^{N}\cos\left(\frac{2\pi}{t_{\mathrm{j0}}}t+\varphi_{\mathrm{j}}\right),\; \sum_{j=1}^{N}\sin\left(\frac{2\pi}{t_{\mathrm{j0}}}t+\varphi_{\mathrm{j}}\right),0\right) \tag{2}$$

where $\boldsymbol{r}_{\mathrm{s}}$ is a vector from the center of the turntable to the substrate, $\boldsymbol{n}_{\mathrm{s}}$ is orientation of the substrate, N is the number of the rotational axes, R_{i} are the radii around the individual axes, t_{j0} are time periods of one rotational cycle around the axis j, φ_{j} are initial angles around the individual axes and h_{s} is the height of the substrate (cf. Figure 1). The rotation around the third axis is not continuous. It is achieved by the switch, which turns the substrate for a certain angle when the substrate makes one cycle around the second axis. This is also taken into account in equations (1) and (2).

3. The magnetron targets are considered as two point sources representing a racetrack. A particle flux j_p is modeled by the cosine angular distribution (Figure 2)

$$j_p = \frac{A}{r^2}(\cos\vartheta)^n \tag{3}$$

where r is the distance from the source to the substrate, ϑ is the angle between the sources' normal and the direction of the sputtered particles, A is the flux intensity, and n is the lateral particle distribution coefficient.

4. The surface of the substrate is coated only if it is in a direct view of the target otherwise the deposition rate is zero. Shaded areas are defined by the dot products of the following vectors (cf. Figure 3):

$$\frac{\boldsymbol{r}_\mathrm{s}(t)-\boldsymbol{R}_\mathrm{h}(t)}{|\boldsymbol{r}_\mathrm{s}(t)-\boldsymbol{R}_\mathrm{h}(t)|}\cdot\frac{\boldsymbol{R}_{\mathrm{K}_i}(t)-\boldsymbol{R}_\mathrm{h}(t)}{|\boldsymbol{R}_{\mathrm{K}_i}(t)-\boldsymbol{R}_\mathrm{h}(t)|} < \cos(90°) \tag{4}$$

$$\frac{\boldsymbol{r}_\mathrm{s}(t)}{|\boldsymbol{r}_\mathrm{s}(t)|}\cdot\frac{\boldsymbol{R}_{\mathrm{K}_i}(t)}{|\boldsymbol{R}_{\mathrm{K}_i}(t)|} < \cos(75°) \tag{5}$$

where $\boldsymbol{R}_\mathrm{h}$ is the vector from the center of the turntable to the center of the substrate tower and $\boldsymbol{R}_{\mathrm{K}_i}$ is the vector from the center of the turntable to the sputtering source i. The shading originates from two contributions. The substrates, which are in the shade of its own substrate tower, are described by the relation (4). The substrates, which are in the shade of other substrate towers, are described by the relation (5). The vector $\boldsymbol{R}_{\mathrm{K}_i}$ is fixed and the vectors $\boldsymbol{r}_\mathrm{s}(t)$ and $\boldsymbol{R}_\mathrm{h}(t)$ change with the time.

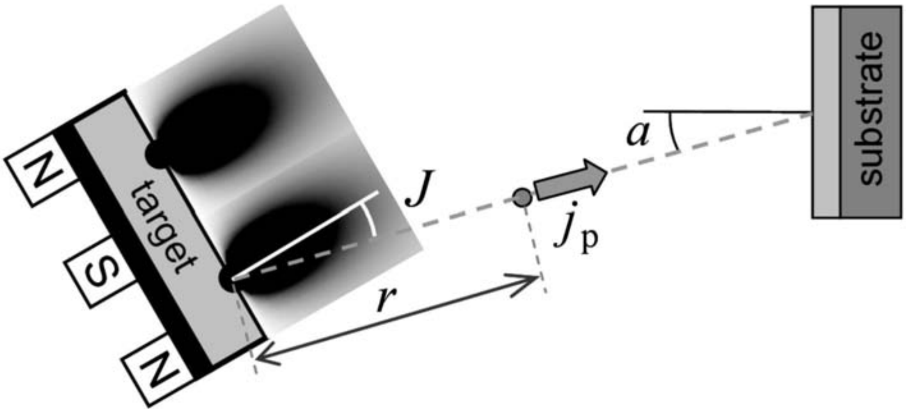

Figure 2. Particle flux from the magnetron target

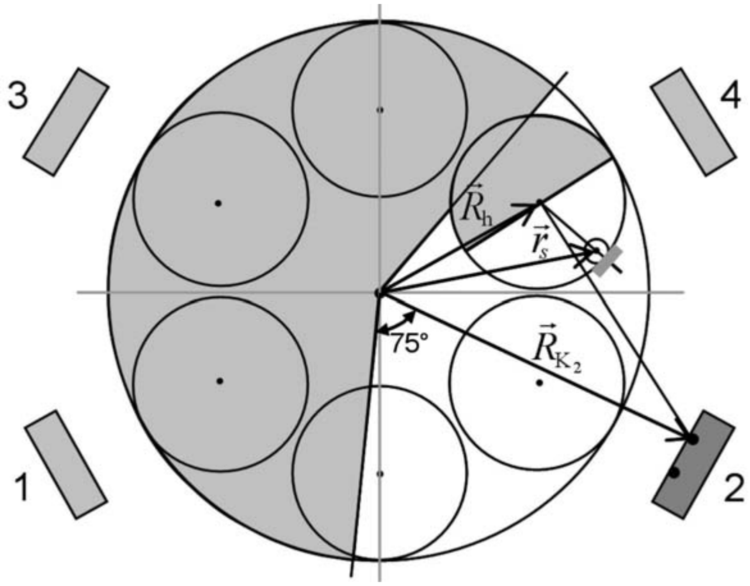

Figure 3. Shaded area (grey) of the particle flux coming from target 2

The deposition rate on the substrate changes due to rotation. When the substrate is close to the target and is fa-cing its direction, the deposition rate is high. If the substrate is far away from the target, the deposition rate is low, or zero if it is facing away from the target. The rotation is defined by equations (1) to (3). The distance between the target i and the substrate $|\boldsymbol{R}_{K_i} - \boldsymbol{r}_s(t)|$ changes with the rotation. The particle flux from all the targets at the position of the substrate is

$$j_p = \sum_{i=1}^{N} \frac{A_i}{(\boldsymbol{R}_{K_i} - \boldsymbol{r}_s)^2} (\cos\vartheta_i)^n \tag{6}$$

where ϑ_i is the angle between the target i normal and the direction of the particle flux to the substrate, A_i is the flux intensity from the target i, and n determines the shape of the angular distribution.

The deposition rate (v) on the surface of the substrate is proportional to the particle flux from all targets (j_{pi}) and it depends on the particle flux angle of incidence α_i (see Figure 2)

$$v = \sum_{i=1}^{N} j_{pi} \cos\alpha_i \tag{7}$$

In the simulation, it is considered that the growth of the coating is only possible if –90° <α_i < 90°. In these positions, the surface of the substrate faces the target, for all other angels the surface is shaded and $v = 0$.

The equation (7) describes the variations of the deposition rate in dependence of the time. Integration of the deposition rate with respect to the time gives the thickness (h) of the individual layers

$$h = \int_0^t v(t)dt \tag{8}$$

In the final step of the simulation, the calculated thicknesses of individual layers are graphically represented in a form of a layered structure. An example of the calculated deposition rate and layer structure is shown on Figure 4a and Figure 4b, respectively. Calculations were made for 3-fold rotation with two targets of equal material on one side of the deposition chamber and two targets of other material on the other side.

RESULTS AND DISCUSSION

The model described above was used to analyze the influence of different parameters on the layered structures. The layer structure depends on the initial position of the sample, type of rotation, configuration of targets and on geometrical parameters of the deposition system. Here we will discuss only the influence of 1-, 2- and 3-fold rotation on the layer structures. In order to prove the accuracy of the model we have also compared the TiAlN/CrN nanolayered coatings prepared by all types of rotation to the calculated layer structures.

Figure 5 shows the calculated deposition rate and layer structures for 1-, 2- and 3-fold rotation. Calculations were made for the CC800/9 deposi-

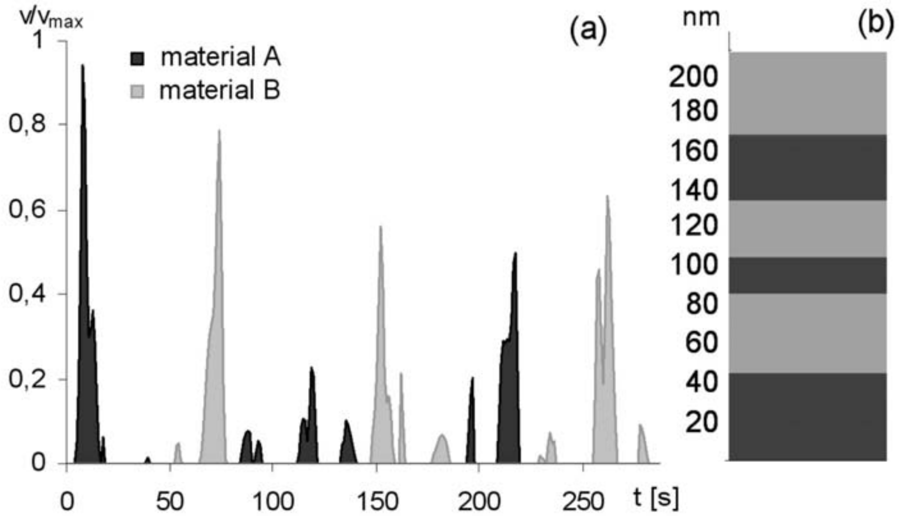

Figure 4. (a) The deposition rate as function of time and (b) the layered structure. Calculation was made for 3-fold rotation

tion system with two Cr targets on one side of the vacuum chamber and two TiAl targets on the opposite side. The deposition rates and the layered structures differ between three types of rotation quite considerably. In the case of a **1-fold rotation** (Figure 5a), the deposition rate and layered structure is periodic. The substrates travel on identical trajectory for each rotation of the turntable, thus, the deposition rate and the layer structure are periodic. In each rotation of the turntable, the substrate is equally exposed to the TiAl and the Cr targets, thus, the individual layers have the same thickness (≈100 nm).

The deposition rate and layered structure for **2-fold rotation** is shown on Figure 5b. Additional rotation around the second axis influences the periodicity of the layer structure. The periodicity of the layer structures prepared by 2-fold rotation depends on the gear ratio between the turntable and the substrate tower. In the CC800/9 deposition system, the gear ratio is 100 : 37, which means that the substrate returns into an identical position only after 37 rotations of the turntable. Therefore, the layer structure for the 2-fold rotation repeats after $2 \cdot 37 = 74$ deposited layers (in each rotational cycle, 2 layers are deposited). Although the periodicity is quite large the thickness of the individual layers is approximately the same because in each rotational cycle of the turntable the sample is almost equally exposed to the targets; hence, the thickness of the individual layers varies only slightly.

In the case of a **3-fold rotation**, the periodicity of the layer structure is the most complex (cf. Figure 5c) due to the non-continuous rotation of the switch which turns the sample for a certain angle for each rotation around the second axis. Rotation around the third axis affects only the orientation of the substrate and less its position. The reason is the small radius of rotation around the third axis (e.g. 5 mm for drills) compared to the radii of rotation around the first (137 mm) and the second axis (58 mm). Thus, 3-fold rotation is essentially 2-fold rotation superimposed on a non-continuous third rotation, which only changes the orientation of the substrate. The periodicity of the layer structure produced by 3-fold rotation depends both on the gear ratio and the switch angle. In principle, the planetary rotation always produces periodic layer structures if the periodicity is observed on a large scale. However, on the scale of the deposition time (≈1 h), the layer structures can be periodic or aperiodic. The layer structure is periodic if the substrate returns into an identical position after a particular number of the turntable rotations. If this does not happen during the deposition time, the layer structure of the coating is aperiodic. In practice, layered structures prepared by 3-fold rotation are usually aperiodic.

From the Figure 5 it can be seen that the total thickness of the coating strongly depends on the type of rotation. In the case of 1-fold rotation, the total thickness for 5 rotations of the turntable is ≈900 nm, for 2-fold rotation ≈400 nm

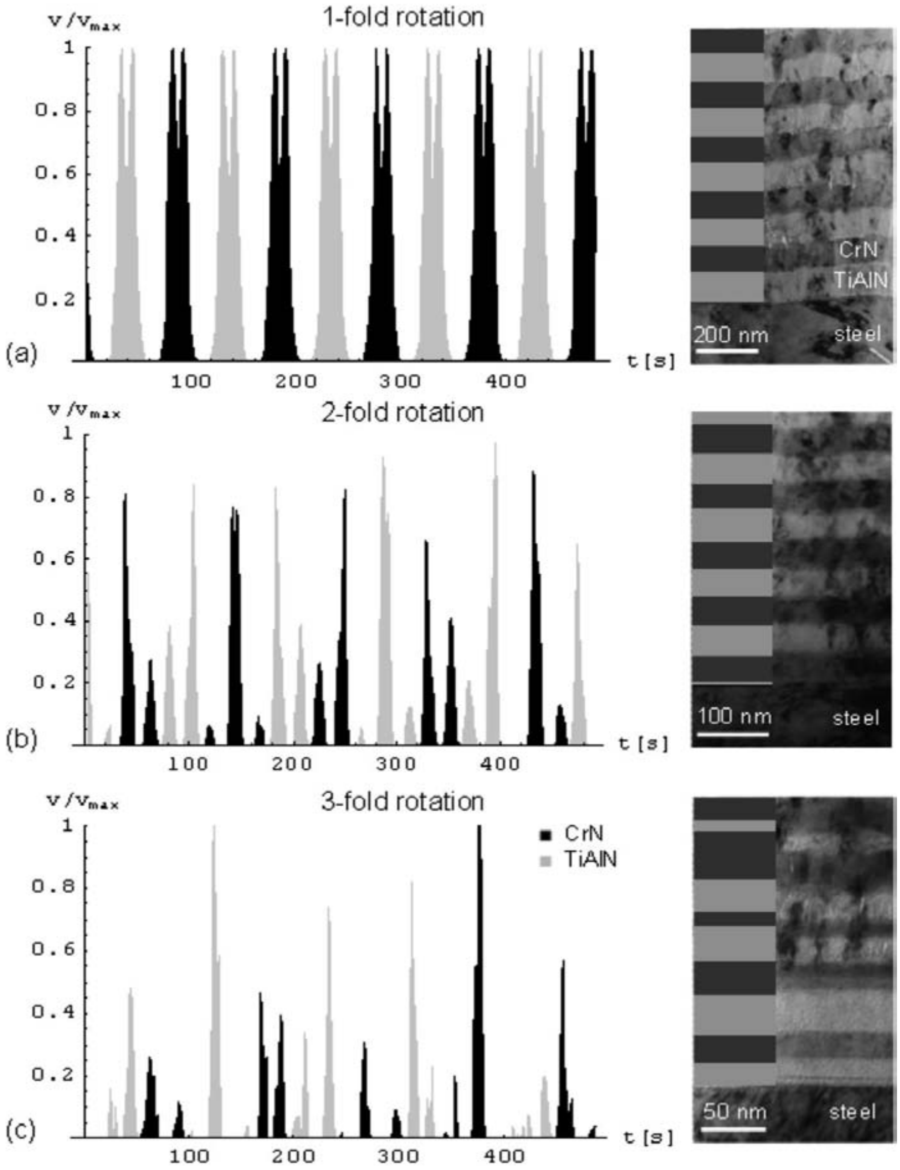

Figure 5. The deposition rate and the layer structure for (a) 1-, (b) 2-, and (c) 3-fold rotation. Calculated layered structures are compared to the TEM micrographs of the deposited TiAlN/CrN nanolayered coatings[10]

and for 3-fold rotation ≈220 nm. The corresponding average deposition rates are 1.8 nm/s, 0.8 nm/s, and 0.45 nm/s for the 1-, 2- and 3-fold rotation, respectively. Hence, the rotation around an additional axis considerably lowers the average deposition rate. Thus, if the coating in the same batch is deposited on the substrates with different types of the rotation, then the total thickness of the coatings will be considerably different.

On the right side of the Figure 5, the calculated layer structures are compared to the deposited TiAlN/CrN nanolayered coatings. The coatings were prepared by the three types of the rotation; 1-, 2- and 3-fold. The bright layers correspond to TiAlN and the dark layers to CrN. Comparison between the calculated and the deposited layer structures shows good overlap between the structures in the case of 1- and 2-fold rotations. In the case of the 3-fold rotation the agreement is less accurate although still satisfactory. Nevertheless, the discrepancy is mainly caused by the experimental difficulties during the deposition.

The accuracy of the model used for calculating the layer structure is influenced by different factors. The simulation is only an approximation of the deposition process. In the model, the angular particle flux from the target was approximated by the cosine law. Different distributions were also tested (e.g. a cosine square distribution); however, this did not influence the layer structure considerably. A more important source of error in the model is a contribution of ionized particles to the deposition of the coating (ionization of the target material is ≈10 %). In the simulation, only particles (atoms) which travel in a straight line were considered. Ions whose path is determined by the electrical field (bias on the substrates) are neglected. The scattering of the particles and resputtering effects are also not considered in the model. However, these effects probably have only a minor influence on the accuracy of the model.

The most important source of error is probably the switch. The switch does not always turn the sample for the same angle. However, the simulations have shown that already a small change in the switch angle (e.g. 5°) produces considerable variations in the layer structure. Such stochastic rotational causes an increase of the error with every rotational cycle. This is seen, for example, in Figure 5c, where after a few deposited layers the agreement between the calculated and the deposited layer structure is lost.

Despite the approximations used in the model and the experimental difficulties it can be concluded that the model quite accurately describes the deposition process of layered structures. Therefore, we believe that this model can be used to explain various layer structures which are obtained by different parameters of the deposition. Thus, such simulations would be a benefit to the engineers who design industrial deposition systems.

CONCLUSIONS

A model of a sputtering process in an industrial magnetron sputtering system with the planetary rotation was developed in order to understand the influence of the rotation on the layered structures. Layered structures prepared by 1-, 2- or 3-fold rotation were analyzed by the model. The results of the model show that the periodicity of the deposition rate and consequently of the layer structure significantly depends on the type of rotation as well as on the other parameters. To verify the accuracy of the model we prepared TiAlN/CrN nanolayered coatings by the three types of rotation in the industrial magnetron sputtering system CC800/9 from CemeCon. The results show good agreement between the prepared and calculated layer structures. Thus, we can conclude that the model correctly describes the deposition process and therefore it could be used to predict the layer structures for different parameters of the process.

Acknowledgments

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REFERENCES

[1] M. STUEBER, H. HOLLECK, H. LEISTE, K. SEEMANN, S. ULRICH, C. ZIEBERT (2009): Concepts for the design of advanced nanoscale PVD multilayer protective thin films. *J. Alloys Compd.*, *483*, pp. 321–333.

[2] A. CAVALEIRO, J. T. M. DE HOSSON (2006): Nanostructured Coatings. (*Springer Science, New York*).

[3] U. HELMERSSON, S. TODOROVA, S. A. BARNETT, J. E. SUNDGREN, L. C. MARKERT, J. E. GREENE (1987): Growth of single-crystal TiN/VN strained-layer superlattices with extremely high mechanical hardness. *Journal of Applied Physics*, *62*, pp. 481–484.

[4] P. C. YASHAR, W. D. SPROUL (1999): Nanometer scale multilayered hard coatings. *Vacuum*, *55*, pp. 179–190.

[5] D. MATTOX (1998): Handbook of Physical Vapor Deposition Processing. (*Noyes Publications, Westwood*).

[6] W.-D. MUNZ (2003): Large-Scale Manufacturing of Nanoscale Multilayered Hard Coatings Deposited by Cathodic Arc/Unbalanced Magnetron Sputtering. *MRS Bulletin*, *28*, pp. 173.

[7] B. Rother (1994): *Surf. Coat. Technol.*, *64*, pp. 155.
[8] B. Rother, H. A. Jehn, H. M. Gabriel (1996): *Surf. Coat. Technol.*, 86–87, pp. 207.
[9] B. Rother, G. Ebersbach, H. M. Gabriel (1999): *Surf. Coat. Technol.*, *116–119*, pp. 694.
[10] M. Panjan, T. Peterman, M. Cekada, P. Panjan (2009): Simulation of a multilayer structure in coatings prepared by magnetron sputtering. *Surf. Coat. Technol.*, *204*, pp. 850–853.