Mechanical properties of multilayer coatings TiAlN

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Abstract. This study aims to examine multilayer TiAlN coatings. The mechanical properties of the coatings were obtained based on micromechanics methods and finite element modeling of a representative volume of a layered medium in ANSYS. Our findings are in line with the data on coating nanoindentation. Besides, we have succeeded in finding the dependence of the mechanical properties of multilayer coatings on various Al/Ti ratios.

1. Introduction
In recent years there has been considerable interest in vacuum ion-plasma wear-resistant coatings that may have either a monolayer structure or a sequence of thin plane-parallel layers [1–6]. Each layer has a thickness of up to hundreds of nanometers, or a heterogeneous 3D-type dispersed multiphase structure [1]. Research and directional modeling of the mechanical properties of multilayer coatings are crucial issues when designing such materials since they depend on a variety of factors [2–9]. For instance, as noted by [7, 8] the properties of films much depend on several technological parameters of the deposition on a substrate, e.g., the energy of deposited atoms, the substrate roughness, etc. In this case, the mechanical properties of coatings can be obtained based on the approaches of micromechanics [10–12] and molecular dynamics modeling [7]. Mathematical modeling of deposition and growth of thin films using supercomputer modeling is given in [7]. Besides, Grigoriev et al. [7] calculated mechanical stresses that arise during the growth of a silicon dioxide film on a substrate and are caused by the difference in the physical and chemical properties of the substrate and the film, as well as by the deposition conditions. Tillmann et al. [8] experimentally investigated the residual stresses in both coating and substrate depending on the deposition method (DCMS/HiPIMS). It should be mentioned that residual stresses in the coating and substrate can affect the coating adhesion and tribological characteristics. Much work on the tribological properties of media with a microstructure has been carried out based on contact problems in quasi-static and dynamic formulations considering friction in the contact area [13–16]. Experimental studies of tribological characteristics are given in works [17–18].

Within the framework of this study, we calculated the mechanical properties of layered coatings depending on the atomic ratio of their coating components. Our approach is based on a differential scheme of the self-consistency method and finite element modeling of the representative volume of a layered medium in the ANSYS software package. Then, the results of numerical simulation of the mechanical properties of multilayer coatings were compared with data on coating nanoindentation.

2. Mechanical properties of multilayer coatings
The study focuses on the TiAlN layered medium with alternating TiN, AlN layers. The layered coating is a transversely isotropic medium, whose transtropy axis is perpendicular to the plane of the
layers. The tensor of elastic constants of a layered medium $C^{\text{eff}}$ was determined based on a differential scheme of the self-consistency method for oblate spheroidal inclusions with a volume concentration $\phi$ in a transversely isotropic medium as follows:

$$
\frac{dC^{\text{eff}}(\phi)}{d\phi} = \frac{1}{1-\phi} \left( C_H - C^{\text{eff}}(\phi) \right) \left( E + S(\phi)(C^{\text{eff}}(\phi))^{-1} \left( C_H - C^{\text{eff}}(\phi) \right) \right)^{-1}, C^{\text{eff}}(0) = C_I,
$$

where $S$ is the Eshelby tensor for the transtropic medium, $E$ is the unit tensor, $C_I$ and $C_H$ are the tensors of the elastic constants of main material and of inclusions, $\phi = V_I / (V_I + V_H)$. It is worth noting that equation (1) allows calculating the effective elastic moduli of a layered medium, modeling the layers as infinitely thin disc-shaped plate inclusions.

Thin lamellar disc-shaped inclusions look like spheroids, whose semiaxes are $a_1 = a_2 = a$, $a_3 = \zeta a$, moreover $\zeta << 1$. In this case, the Eshelby tensor components are expressed by double integrals that have no singularities in the integral domain. The equation (1) -based numerical experiments on calculating the effective elastic moduli of a layered medium were done in the Matlab software package. To validate the data obtained by equation (1)-based calculations, the program was initially tested on special cases for the Eshelby tensor components for an isotropic medium with spherical and thin disc-shaped inclusions. The Eshelby tensor components were obtained in explicit expression without any quadratures [19].

The second method for calculating the effective elastic constants of a layered coating is a finite element modeling of a representative volume with thin layers in ANSYS. In this case, the Ti/Al atomic ratio was satisfied by the thickness of the layers, as well as by their number. Determining five independent elastic constants implies the solution of the corresponding boundary value problems:

$$
\nabla \cdot \sigma = 0, \ \sigma = C^{\text{eff}} : \varepsilon, [u] = 0, n \cdot [\sigma] = 0,
$$

where $\sigma$, $\varepsilon$ are tensors of the second rank of stresses and strains, $C^{\text{eff}}$ is the elastic moduli tensor of the fourth rank. Tensors are written in the non-index form. The brackets denote a jump off the variable enclosed in them, across the layer interface. Therefore, the definition of five independent elastic constants is reduced to solving five boundary value problems, assuming separate components $\varepsilon_i \neq 0$ in equation (2). It should be mentioned that, as in the case of the first approach, upon reaching a certain layer thickness (number of layers), the elastic constants tensor components basically do not change. As an increase in the number of layers results in a decrease in their thickness, we had to finish the numerical experiment when the number of layers was equal to 30.

3. Results and discussion

We carried out a numerical experiment for TiAlN layered composition with the following properties: $E_{\text{TIN}} = 259$ (GPa), $\nu_{\text{TIN}} = 0.25$ [9], $E_{\text{AlN}} = 273$ (GPa), $\nu_{\text{AlN}} = 0.21$ [6, 9], $\zeta = 0.001$. Table 1 summarizes the data on $E^*_i$, $E^*_i$, $E^*_i$, $E^*_i$ values of the effective Young’s modules for a transversely isotropic medium, depending on Al/Ti mass. % calculated by the differential scheme of the self-consistency method ($E^*_i$) and finite element modeling in the ANSYS software package ($E^*_i$). In the numerical experiment, we chose TiN as the main material and AlN as the inclusions. The relative margins of error of Young's modulus were quantified by $\varepsilon_i = \frac{[E^*_i - E^*_i]}{E_{\text{AlN}}} \cdot 100\%$, $i = 1, 3$ and did not exceed 8%. It should be noted that in this case, the layered medium, whose layers possess the aforementioned mechanical properties, behaves essentially as isotropic.

Then, we compared our findings with the data obtained by nanoindentation [1]. In the TiAlN system coatings, the Al/Ti ratio in at.% was approximately the same at the level of $25/25 = 1$ (values scatter was $1.0 – 1.12$), which in mass.% corresponds to $25/45 = 0.56$ (values scatter was $0.51 – 0.63$) [1], which corresponds to TiN – 0.641, AlN – 0.359 in the TiAlN system.
Table 1. Effective Young's modulus of multilayer TiAlN composition.

| ϕ      | $E_1^M$ (GPa) | $E_1^A$ (GPa) | $E_3^M$ (GPa) | $E_3^A$ (GPa) | $\varepsilon_1$ (%) | $\varepsilon_3$ (%) |
|--------|---------------|---------------|---------------|---------------|--------------------|--------------------|
| 0.1    | 259.0649      | 278.0600      | 259.0676      | 277.1733      | 6.95               | 6.99               |
| 0.2    | 259.2721      | 278.3150      | 259.2834      | 278.0839      | 6.98               | 7.25               |
| 0.3    | 259.6150      | 280.6477      | 259.6402      | 279.0003      | 7.70               | 7.47               |
| 0.359  | 259.8818      | 280.8577      | 259.9173      | 279.5542      | 7.68               | 7.58               |
| 0.4    | 260.0833      | 281.0655      | 260.1265      | 279.9225      | 7.68               | 7.64               |
| 0.5    | 260.6627      | 281.1509      | 260.7270      | 280.8534      | 7.5                | 7.77               |
| 0.6    | 261.3365      | 281.2728      | 261.4233      | 281.7901      | 7.3                | 7.86               |

The Young's modulus values for a layered transversely isotropic medium in a plane perpendicular to the isotropy plane are presented in table 1. The nanoindentation test results (reduced modulus of elasticity) are detailed in table 1 in [1].

Hence, relative margins of error were in the range of 9.3 – 13.1% and 19 – 24% for coatings with 150-200 (nm) thick layers for two types of substrates and coatings with 30-60 (nm) thick layers, respectively. A large scatter of experimental data and, consequently, relative margins of error can be explained by the coating technology applied [6, 7, 9].

The mechanical properties of films substantially depend on many technological parameters of the process of their deposition on a substrate [2–9]. We carried out a numerical experiment for $E_{TiN} = 600$ (GPa), $\nu_{TiN} = 0.25$ [5] at the same values of Young's modulus and Poisson's ratio AlN. Table 2 presents the Young's modulus of the TiAIN layered system, which were calculated based on equation (1), where TiN is the main material and AlN is the material of inclusions.

Table 2. Effective Young's modulus of multilayer TiAlN composition.

| ϕ      | $E_1^M$ (GPa) | $E_3^M$ (GPa) |
|--------|---------------|---------------|
| 0.1    | 598.5322      | 596.9264      |
| 0.2    | 593.8454      | 587.3060      |
| 0.3    | 586.0755      | 571.9955      |
| 0.4    | 575.4469      | 552.2622      |
| 0.5    | 562.2625      | 529.5499      |
| 0.6    | 546.8883      | 505.2619      |

In this case, the degree of anisotropy of the medium under consideration is greater than in the case of the mechanical characteristics of TiN, AlN, on the basis of which the data in table 1 were obtained.
4. Conclusions
In this work, two approaches are considered for determining the mechanical properties of multilayer coatings of the TiAlN system depending on various Ti/Al ratios. The first of them is a differential scheme of the self-consistency method, and the second is a finite element modeling of a representative volume of a layered medium. The data obtained as a result of the numerical experiment were compared with the available results of nanoindenting of multilayer TiAlN coatings. The relative margin of error (less than 8%) between the two numerical-analytical approaches demonstrates the efficiency of the proposed methods for calculating the mechanical properties of multilayer coatings.

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