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Vladimir V. Bardushkin, Yulia I. Shilyaeva, Sergey A. Gavrilov, Maxim V. Silibin, Victor B. Yakovlev, Mikhail L. Zheludkevich and Natalia I. Popenko

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PREDICTIVE MODELING OF MECHANICAL PROPERTIES OF METAL FILLED ANODIC ALUMINUM OXIDE

VLADIMIR V. BARDUSHKIN, YULIA I. SHILYAEVA, SERGEY A. GAVRILOV, MAXIM V. SILIBIN, VICTOR B. YAKOVLEV, MIKHAIL L. ZHELUDKEVICH AND NATALIA I. POPENKO

Filling dielectric porous matrices, particularly anodic aluminum oxide, with metal confers a promising solution for nanocomposite creation. In this regard, the problem of estimating and predicting the physical and mechanical properties of such materials is of prime importance. The present work focuses on the numerical modeling of the effective and ultimate stress-strain (under compression) characteristics of nanocomposites based on anodic aluminum oxide with unidirectional filamentary pores filled with different metals (In, Sn, and Zn). The dependences of the tensor components of the effective elastic moduli, coefficients of elastic anisotropy (in different directions), and compression strength (along the nanowires) on the structure parameters and the concentration of nanowires are investigated.

1. Introduction

One of the possible ways to create nanocomposites is filling dielectric ordered porous matrices with metal. In this regard, porous anodic aluminum oxide (AAO) [Yao et al. 2008] is the most widely used candidate. AAO-based nanocomposites have unique characteristics and are promising for a wide range of possible applications for optical systems and sensors, as well as functional parts of solar and thermoelectric batteries [Poddubny et al. 2013]. The high thermal stability and the possibility of precise control of geometrical parameters and consequently size-dependent properties make AAO the material of choice in many cases. Produced by a two-step anodization by the method of [Masuda and Fukuda 1995], AAO has a high degree of ordering — representing an array of hexagonal cells with cylindrical pores. It allows the creation of nanocomposites with well controllable properties and is an ideal model structure for fundamental research. Moreover, AAO-based template synthesis is an alternative to expensive lithographic techniques.

The mechanical properties of nanocomposites become very important when considering practical applications. The basis of such analysis is the calculation of performance (effective) and ultimate stress-strain properties [Shermergor 1977; Pobedrya 1984; Khoroshun et al. 1989; Mori and Tanaka 1973; Kanaun and Levin 1993; Milton 2002; Buryachenko 2007; Böhm 2013; Bardushkin and Yakovlev 2011; Sychev and Bardushkin 2013]. Thus, the task of developing predictive methods for physical and mechanical properties of matrix composites like "AAO-nanowires" is very relevant.

The problem of predicting the effective elastic properties of heterogeneous media has been considered by many authors. A detailed review of studies on this subject can be found in [Buryachenko 2007; Böhm 2013; Bardushkin and Yakovlev 2011; Sychev and Bardushkin 2013].

Keywords: nanocomposite, modeling, anodic aluminum oxide, performance elastic characteristics, compressive strength.

Predicting the distribution of local (internal) stress and deformation fields is key for a correct analysis of the ultimate strength properties of heterogeneous media. The local elastic characteristics include operators (tensors) of stress/deformation concentration and volumetric density of deformation energy [Buryachenko 2007; Böhm 2013; Bardushkin and Yakovlev 2011; Sychev and Bardushkin 2013]. The distribution analysis of the above characteristics allows correct predictions of the behavior of materials under loads (especially extreme). It can provide recommendations on the selection of components taking into account the concentration of heterogeneous elements and features of the shape structure and inclusion orientation in the material matrix.

The concept of concentration tensors connecting the average values in the phase with average values in the whole body (or in a representative volume element) was introduced by Hill [1963]. The solution to the problem of strength concentration on the surface of an ellipsoidal heterogeneity in an anisotropic medium was obtained in [Laws 1977; Kunin and Sosnina 1973; Buryachenko and Lipanov 1986; Yakovlev and Nikitin 1997; Shermergor and Yakovlev 1998; Maslov 1987]. Concentration functionals and operators expressing stress and strain in a heterogeneous body by using those in a homogeneous body with efficient features were described in [Pobedrya and Gorbachev 1984]. In [Pobedrya and Gorbachev 1984; Gorbachev 1989], explicit analytical expressions for the concentration tensors in the case of a layered composite and in the case of a cylindrical hole in an infinite homogeneous isotropic medium were obtained. The concentration tensors in an n-dimensional elastic medium with an n-dimensional spherical inclusion were suggested in [Gorbachev and Mikhailov 1993]. By assuming homogeneity of the stress and strain fields (via the generalized singular approximation used to determine the effective properties of heterogeneous media [Shermergor 1977]), the expressions for calculating these operators were obtained in [Yakovlev 2000; Kolesnikov et al. 2005; Bardushkin and Yakovlev 2005; Bardushkin et al. 2013], in which the effect of composite microstructures on their local characteristics was investigated.

2. Formulation of the problem and model

The aim of this work is to solve the two main issues of predicting the elastic-strength properties of nanocomposites based on anodic aluminum oxide with filamentary pores filled with metal (In, Sn, and Zn), namely:

(1) predicting the operational (effective) elastic characteristics, and

(2) predicting the ultimate stress-strain (under compression) characteristics.

The problem of predicting the mechanical properties can be solved firstly by constructing a model taking into account the structure of the nanocomposites by introducing a dimensionless structural parameter associated with the concentration of heterogeneous elements; and secondly by performing the numerical modeling of elastic-strength characteristics.

2.1. ***Introduction of a dimensionless structural parameter.*** The following points should be resolved before creating a numerical model for the performance and ultimate stress-strain properties of matrix composites like "AAO-nanowires":

(a) a correlation between the distance between the nanowires and structural parameters that can be measured directly, and

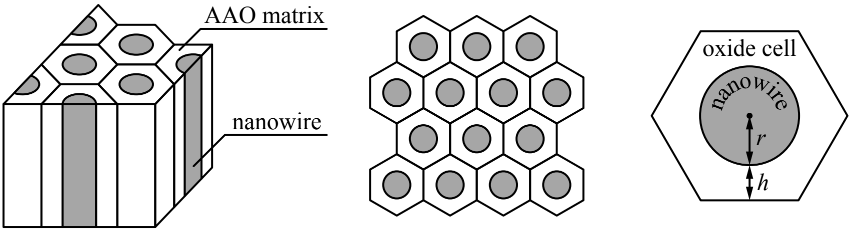

Figure 1. Schematic representation of the composite structure (left), several elementary volumes (middle), an elementary volume in the cross-sectional plane (right).

(b) the derivation of mathematical formulas convenient for numerical analysis without losing information about the structure of the composite [Bardushkin and Yakovlev 2011; 2005; Kolesnikov et al. 2005; Bardushkin et al. 2013; Shilyaeva et al. 2013b; 2013a; 2014].

The actual structure of composites must be taken into account when conducting simulations, as described earlier [Shilyaeva et al. 2014]. As it is known, AAO consists of densely packed hexagonal cells that are adjacent to each other with their sides. Therefore, we assume that in a considered uniaxially reinforced composite the components are isotropic and the position of nanowires in the template is random. However, the material is assumed to be statistically homogeneous as a whole. This assumption results in the existence of an average distance between wires that may be related to the loading of the metal in the composite matrix. An average volume element in the shape of a regular hexagonal prism with one cylindrical nanowire oriented along the z axis in the center can be considered. Some of these elementary volumes are schematically shown in Figure 1.

One can assume that each nanowire has an average radius r and the distance from the center of a regular hexagon to its side is $r+h$ (see Figure 1, right). The base area of the elementary cell is then $S = 2\sqrt{3}(r+h)^2$, and the cross sectional area of the wire is $S_w = \pi r^2$. Defining the concentration of wires as $v_w = S_w/S$, we have $v_w = \pi/(2\sqrt{3}(1+h/r)^2)$, $v_m = 1 - v_w$.

The index "w" here and below denotes the values related to the metallic wires, while "m" indicates those related to the matrix.

The characteristic parameter h/r defining the structure of the composite can thus be represented by the concentration of nanowires as

$$\frac{h}{r} = \sqrt{\frac{\pi}{2\sqrt{3}\cdot v_w}} - 1. \tag{1}$$

It is evident that the maximum theoretical value of the concentration of nanowires is observed when $h/r \to 0$, which corresponds to $v_w \to \pi/(2\sqrt{3}) \approx 0.9$. The minimum value of the concentration of wires is observed when $h/r \to \infty$; hence, $v_w \to 0$. This range of concentrations for nanowires corresponds to the boundaries of the applicability for the suggested approach of simulating such materials.

2.2. ***Elastic characteristics.*** The effective elastic characteristics of the considered composites can be determined by the fourth-rank tensor c^* ("$*$" here and below indicates that composite's effective characteristics are considered) connecting the average values of stresses $\langle\sigma_{ij}(\boldsymbol{r})\rangle$ and strains $\langle\epsilon_{kl}(\boldsymbol{r})\rangle$ in the material via

$$\langle\sigma_{ij}(\boldsymbol{r})\rangle = c^*_{ijkl}\langle\epsilon_{kl}(\boldsymbol{r})\rangle, \quad i, j, k, l = 1, 2, 3,$$

where $\boldsymbol{r}$ is the radius-vector of a random point in the medium. Angular brackets here and below define the procedure of ensemble averaging. For statistically homogeneous composites, i.e., when performing the hypothesis of ergodicity, it coincides with the averaging in volume [Shermergor 1977; Pobedrya 1984; Khoroshun et al. 1989; Mori and Tanaka 1973; Kanaun and Levin 1993; Milton 2002; Buryachenko 2007; Böhm 2013; Bardushkin and Yakovlev 2011; Sychev and Bardushkin 2013; Walter et al. 1993; Mitin et al. 2001].

The equations for the equilibrium of an elastic heterogeneous medium should be solved to conduct a correct analysis of elastic properties of composites which depend on the interaction of elements of heterogeneity, composition, shape, orientation, and concentration of components. The ratio for the numerical calculations of an effective elastic moduli tensor c^* is usually hard to obtain. Therefore, various approximations are used for its calculation. Within the framework of the generalized singular approximation of the theory of random fields [Shermergor 1977], only the singular component of Green's tensor of equations for the equilibrium is used. It depends only on the Dirac delta function. A homogeneous reference body whose material constants are included in the final expression for calculating c^* is also introduced. The physical meaning of the generalized singular approximation is the assumption of homogeneity of the stress and strain fields within the element of heterogeneity. In this case, the expression for calculating c^* is (indices omitted) [Shermergor 1977; Walter et al. 1993; Mitin et al. 2001]

$$c^* = \langle c(\boldsymbol{r})(I - g(\boldsymbol{r})c''(\boldsymbol{r}))^{-1}\rangle\langle(I - g(\boldsymbol{r})c''(\boldsymbol{r}))^{-1}\rangle^{-1}, \tag{2}$$

where I is the fourth-rank unit tensor; $c(\boldsymbol{r})$ is elastic modulus tensor; the double primes indicate the difference between the corresponding parameters of a heterogeneous medium and a homogeneous reference body, characteristics of which are denoted hereinafter by the superscript "ref": $c''(\boldsymbol{r}) = c(\boldsymbol{r}) - c^{\text{ref}}$; $g(\boldsymbol{r})$ is the integral of the singular component of the second derivative of Green's tensor of equations for the equilibrium, which is a fourth-rank tensor. Components g_{ijkl} of $g(\boldsymbol{r})$ tensor can be calculated upon knowing components a_{iklj} of the fourth-rank tensor A as

$$a_{iklj} = -\frac{1}{4\pi}\int n_k n_j t_{il}^{-1}\,\mathrm{d}\Omega, \tag{3}$$

and then symmetrization [Shermergor 1977; Walter et al. 1993; Mitin et al. 2001] is performed using pairs of i and $j$and k and l indices.

In (3), $\mathrm{d}\Omega = \sin\theta\,\mathrm{d}\theta\,\mathrm{d}\phi$ is an element of the solid angle in a spherical system of coordinates; t_{il}^{-1} are the elements of the reverse matrix T where elements $t_{il} = c_{iklj}^{\text{ref}} n_k n_j$; n_k and n_j $(k, j = 1, 2, 3)$ are components of a vector of an external normal to the inclusion's surface. For ellipsoidal inclusions with principal semiaxes l_1, l_2, and l_3, the components of the normal vector are determined by the relationship

$$n_1 = \frac{1}{l_1}\sin\theta\cos\phi, \quad n_2 = \frac{1}{l_2}\sin\theta\sin\phi, \quad n_3 = \frac{1}{l_3}\cos\theta.$$

The relation (2), as shown in [Bardushkin and Yakovlev 2011], can be used to calculate the effective characteristics of a statistically homogeneous matrix composite with ellipsoidal inclusions oriented relative to each other.

As it was mentioned, under the condition of ergodicity it is possible to use volume averaging for each component of the composite [Shermergor 1977; Pobedrya 1984; Khoroshun et al. 1989; Mori and Tanaka 1973; Kanaun and Levin 1993; Milton 2002; Buryachenko 2007; Böhm 2013; Bardushkin and Yakovlev

2011; Sychev and Bardushkin 2013]. Then, the averaging operation over the entire material volume for some random variable $a(\boldsymbol{r})$ is reduced to summing

$$\langle a(\boldsymbol{r})\rangle = \sum_s v_s \langle a_s(\boldsymbol{r})\rangle,$$

where v_s is the volumetric concentration of the s-type component and $a_s(\boldsymbol{r})$ is random variable $\sum_s v_s = 1$ corresponding to the specified component. In particular, for a two-component composite containing isotropic inclusions and the matrix, the procedure of averaging is reduced to summing

$$\langle a(\boldsymbol{r})\rangle = v_w a_w + v_m a_m. \tag{4}$$

When considering inclusions in the form of nanowires with principal semiaxes $l_1 = l_2 = r$ and $l_3 \to \infty$ for the normal vector components, the obtained ratios are

$$n_1 = \frac{1}{r}\sin\theta\cos\phi, \quad n_2 = \frac{1}{r}\sin\theta\sin\phi, \quad n_3 \to 0.$$

The elastic characteristics of the matrix can be taken as the parameters of the reference body [Khoroshun et al. 1989]. Then in (2) $c''(\boldsymbol{r}) = c(\boldsymbol{r}) - c_m$, and $c''(\boldsymbol{r}) = c_w - c_m$ in calculations for the nanowires and $c''(\boldsymbol{r}) = 0$ for the matrix. Considering (4), (2) will take the form for calculating the effective elastic properties of composites as

$$c^* = (v_w c_w (I - g_w(c_w - c_m))^{-1} + v_m c_m) \times (v_w (I - g_w(c_w - c_m))^{-1} + v_m I)^{-1}. \tag{5}$$

In (5), c_w and c_m are the elastic moduli tensors for the wires and matrix, respectively; g_w is a tensor $g(\boldsymbol{r})$ (for the wires) with the components calculated by (3).

2.3. ***Ultimate strength characteristics.*** When solving the problem of predicting ultimate strength properties of composites under compression, the situation associated with the fast fracture of materials is considered. The solution is based on the method of predicting the ultimate strength properties of matrix composites under compression, which is based on the use of the stress concentration operator (fourth-rank tensor). This operator binds the local values of the stress tensor $\sigma_{ij}(\boldsymbol{r})$ with the average external stress of material $\langle\sigma_{kl}(\boldsymbol{r})\rangle$ [Buryachenko 2007; Böhm 2013; Bardushkin and Yakovlev 2011; Sychev and Bardushkin 2013]

$$\sigma_{ij}(\boldsymbol{r}) = K^{\sigma}_{ijkl}(\boldsymbol{r})\langle\sigma_{kl}(\boldsymbol{r})\rangle, \quad i, j, k, l = 1, 2, 3. \tag{6}$$

The kind and magnitude of the stress $\sigma_{ij}(\boldsymbol{r})$ occurring inside the heterogeneous element of any type can be determined knowing the nature of the external impact $\langle\sigma_{kl}(\boldsymbol{r})\rangle$ based on the definition (6) for $K^{\sigma}(\boldsymbol{r})$. It should be emphasized that the emerging local (internal) stresses either in the matrix or in the wires will differ in appearance and size from the applied impact $\langle\sigma_{kl}(\boldsymbol{r})\rangle$ [Bardushkin and Yakovlev 2011].

The idea of using the relation (6), providing a means to link macroscopic stresses with microscopic stresses within the microstructure of the material, in order to predict the ultimate strength characteristics was described in [Fritsch et al. 2009; 2010; 2013; Pichler and Hellmich 2011]. The solution to the problem of predicting the strength characteristics for the matrix composites (within the framework of the generalized singular approximation of random field theory [Shermergor 1977]) is given in [Kolesnikov et al. 2014].

The matrix plays a fundamental role in the composites by making the material monolithic and redistributing the mechanical stresses between all elements of heterogeneity. The matrix breakdown leads to full failure of the material. Therefore, it is believed [Kolesnikov et al. 2014; Bardushkin et al. 2015] that stress applied to the composite (for example, compression in a certain direction) becomes destructive only when the internal stress in the matrix exceeds its ultimate strength. Here the magnitude of the internal stress in the matrix under external impact is compared to the known value of the ultimate strength of the matrix set experimentally or taken from the respective references. The value of the ultimate strength of the matrix should correspond to the external stress (for example, compression performed in the same direction as that for the considered composite) applied to a homogeneous body consisting only of the matrix material.

The equations for the equilibrium of an elastic heterogeneous medium must be solved for the correct analysis of the local stress concentration in the composite that allows the consideration of the interaction of elements of heterogeneity, composition and structure of the material, and concentration of wires. However, in general (as well as when calculating the effective elastic properties c^*), it is impossible to derive the relation for numerical calculations of the stress concentration operator $K^\sigma(\boldsymbol{r})$. Therefore, we use the generalized singular approximation of the theory of random fields for its evaluation (see Section 2.2). In this case, the expression for $K^\sigma(\boldsymbol{r})$ is (indices omitted) [Bardushkin and Yakovlev 2011; Sychev and Bardushkin 2013]

$$K^\sigma(\boldsymbol{r}) = c(\boldsymbol{r})(I - g(\boldsymbol{r})c''(\boldsymbol{r}))^{-1} \times \langle c(\boldsymbol{r})(I - g(\boldsymbol{r})c''(\boldsymbol{r}))^{-1}\rangle^{-1}. \tag{7}$$

The analysis of (7) shows that when evaluating the local stress-strain state of a heterogeneous medium using a stress concentration operator, the information about an external mechanical impact is excluded, because $K^\sigma(\boldsymbol{r})$ depends only on the material parameters of the medium and the composite structure. As before (see Section 2.2), taking the elastic characteristics of the matrix as the parameters for the reference body and considering (4), (7) will take the form

$$K_m^\sigma = c_m(v_w c_w(I - g(c_w - c_m))^{-1} + v_m c_m)^{-1}. \tag{8}$$

3. Numerical calculations

3.1. ***Numerical modeling of the elastic properties.*** For unidirectional matrix composites with isotropic components, like metal nanowires of In, Sn, and Zn in the AAO matrix, the model calculations for the effective elastic moduli c^* were carried out. Information about elastic characteristics of the composite components is known from the literature [Grigor'ev and Meilikhov 1991; Xia et al. 2004; Gu et al. 2004], and is given in Table 1.

Material	Young's modulus E (GPa)	Poisson's ratio ν
Indium	10.5	0.46
Tin	48	0.33
Zinc	115	0.325
Aluminum oxide	140	0.32

Table 1. Elastic characteristics of the composite components.

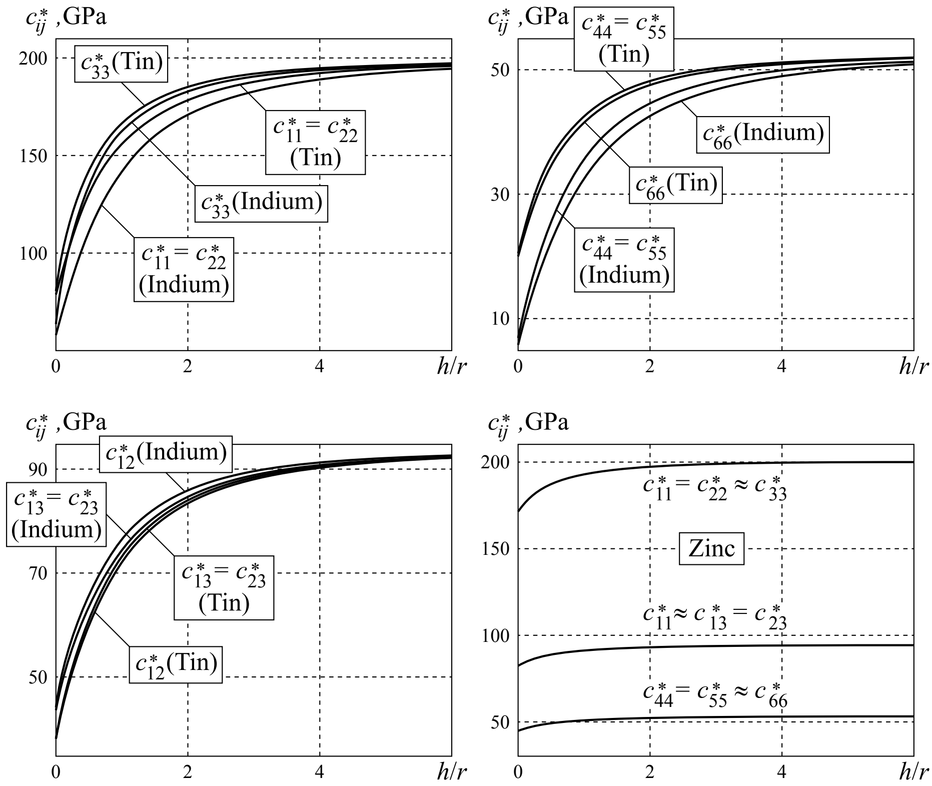

Figure 2. Effective elastic moduli as a function of h/r for model composites.

Considering wires as ellipsoids of rotation with semiaxes $l_1 = l_2 = r = 1$ and $l_3 \to \infty$, calculations of the tensor components c^* depending on the structural parameter h/r were carried out using (5).

In the calculations, we used tensors written in the matrix form. The nonzero elements c_{ij} $(i, j = 1, \dots, 6)$ of the symmetrical matrix of the elastic moduli tensor c for an isotropic material can be expressed through its Young's modulus E and Poisson's ratio ν [Shermergor 1977]:

$$c_{11} = c_{22} = c_{33} = \frac{E(1-\nu)}{(1+\nu)(1-2\nu)};$$
$$c_{44} = c_{55} = c_{66} = \frac{E}{2(1+\nu)};$$
$$c_{12} = c_{13} = c_{23} = \frac{E\nu}{(1+\nu)(1-2\nu)}.$$

The results of the numerical modeling of the nonzero elements c^*_{ij} of the matrix of the effective elastic moduli tensor c^* depending on the structural parameter h/r are given in Figure 2.

The model calculations of the elastic anisotropy coefficients A_x and A_z (along x and z axes respectively) were also carried out: $A_x = (c^*_{11} - c^*_{12})/(2c^*_{44})$, $A_z = (c^*_{33} - c^*_{23})/(2c^*_{66})$. The values of the anisotropy coefficient A_y (along the y axis) are similar to the values of the anisotropy coefficient A_x.

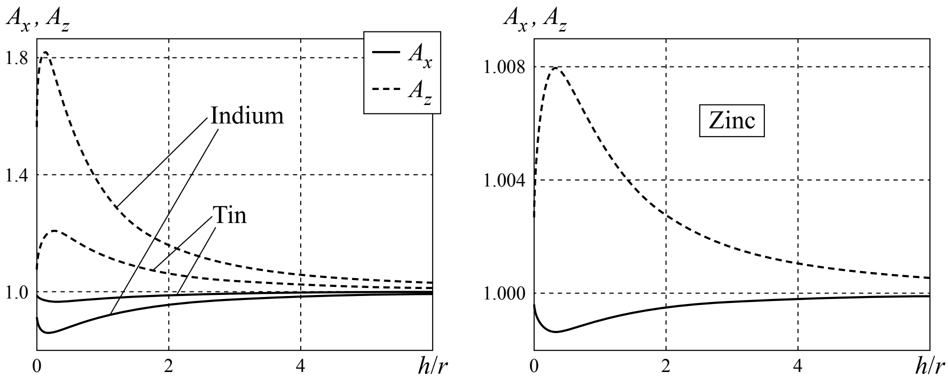

Figure 3. Anisotropy coefficients as a function of h/r for model composites.

Figure 3 shows the results of the numerical modeling of the anisotropy coefficients A_x and A_z depending on the structural parameter h/r.

Note that in Figures 2 and 3, the resulting curves for the composite with Zn nanowires are shown separately. This is because the symmetry of the effective elastic properties of the material is close to isotropic (see the elastic moduli of the components presented in Table 1). Thus, for the effective elastic moduli the relations can be expressed as $c^*_{11} = c^*_{22} \approx c^*_{33}$, $c^*_{44} = c^*_{55} \approx c^*_{66}$, $c^*_{12} \approx c^*_{13} = c^*_{23}$, $c^*_{66} = (c^*_{11} - c^*_{12})/2$, and are applied [Shermergor 1977], and the anisotropy coefficients A_x and A_z for this composite are slightly different from 1.

3.2. ***Numerical modeling of the ultimate stress properties.*** This subsection discusses failure of composites when exposed to compressive stress directed along the z axis (i.e., along the wires). This situation is most frequently encountered in practice.

The analysis of the dependence of compressive strength limits on the composition and concentration of components was carried out for model composites.

The computational procedure used to determine the ultimate compressive characteristics for model composites was arranged in the work as follows. It was assumed that the external action $\langle \sigma_{kl}(\boldsymbol{r}) \rangle$ (MPa) was given in the laboratory coordinate system $Oxyz$ by a (3×3) matrix with the only nonzero element $\sigma_{33} = B$. First, any value of the structural parameter h/r was recorded for the model composite. Then the operator K^σ_m was calculated by formula (8). Then a certain positive value B was assumed. Then, elements σ_{ij} $(i, j = 1, 2, 3)$ of the matrix of the stress tensor (in AAO) were calculated based on the definition (6) of the stress concentration operator. After that the comparison of the values of the computed element σ_{33} with the value of the limit of compressive stress for aluminum oxide equal to $\sigma_m = 4000$ MPa [Grigor'ev and Meilikhov 1991, p. 63] was made. If $\sigma_{33} < \sigma_m$, then value B was increased by 1 MPa and the calculation of the stress tensor matrix elements σ_{ij} in AAO was repeated again. The computational procedure in each case was stopped as soon as the condition $\sigma_{33} \geq \sigma_m$ was met, and the last value of B was taken as the compressive strength limit of the whole composite in the direction parallel to the wires. Then a new value of the structural parameter h/r was recorded and the calculation of the compressive strength limit for the model composite was repeated again.

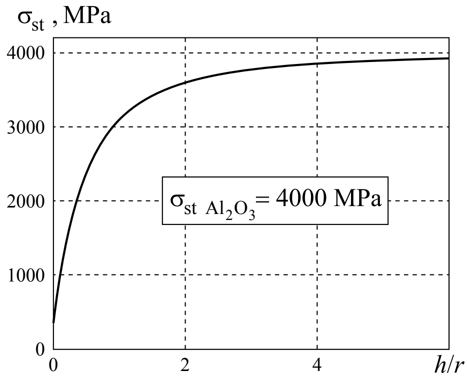

Figure 4. Compressive strength as a function of h/r for model composites.

Figure 4 shows the results of the numerical modeling of compressive strength limits σ_{st} for the above described load mode of model composites depending on the structural parameter.

4. Conclusions

The performed model studies allow us to draw the following conclusions.

The considered composites possess hexagonal symmetry for the effective elastic properties [Shermergor 1977], since for nonzero elements c^*_{ij} in the symmetric matrix tensor c^* the following relations were performed: $c^*_{11} = c^*_{22}$, $c^*_{44} = c^*_{55}$, $c^*_{13} = c^*_{23}$, $c^*_{66} = (c^*_{11} - c^*_{12})/2$ (Figure 2).

When $0 < h/r < 6$, the dependence of the nonzero c^*_{ij} values on the average distance between the wires is essentially nonlinear. When $h/r > 6$ (i.e., when $\nu_w \to 0$), c^*_{ij} stabilizes around values equal to the isotropic elastic moduli of AAO, for the elements c_{ij} of the matrix elastic moduli tensor of which the following relations are valid: $c_{11} = 200.34$; $c_{44} = 53.03$; $c_{12} = 94.28$. Indeed, when $h/r > 6$, the following equations for c^*_{ij} start executing: $c^*_{11} = c^*_{22} = c^*_{33}$, $c^*_{44} = c^*_{55} = c^*_{66}$, $c^*_{12} = c^*_{13} = c^*_{23}$ (Figure 2). Moreover, $A_x \to 1$, $A_z \to 1$ (Figure 3). The strongest variation of anisotropy occurs along z axis. The values of anisotropy parameters in the composites show the strongest deviations from 1 when changing the structural parameter h/r in the range of 0 to 2.

In the considered composites, the dependence of σ_{st} on the parameter h/r has a steady and nonlinear character; moreover at $0.3 < h/r < 3$ this nonlinearity manifests itself most significantly.

Numerical modeling showed that the values σ_{st} are the same for composites with In, Sn, and Zn wires. This result is explained by the known stress properties of the aluminum oxide and the direction of the applied compression load. This follows from the simulation results of changing the values of the stress concentration tensor components K^{σ}_{m} (in matrix), described in [Bardushkin et al. 2013], since these values were similar for composites with In, Sn, and Zn wires.

At average distances between the wires $h/r > 6$ there occurs a stabilization of σ_{st} values. With the increase of h/r there is an increase in σ_{st} values of the ultimate stress limit for the composites up to the values $\sigma_m = 4000$ MPa for ultimate strength of the AAO matrix.

At average distances between the wires $h/r < 1$, the ultimate stress limit value σ_{st} is most sensitive to changes in its volume fraction.

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VLADIMIR V. BARDUSHKIN: bardushkin@mail.ru
Department of Intellectual Technical Systems, National Research University of Electronic Technology, Bld. 1, Shokin Square, Zelenograd, Moscow, 124498, Russia

YULIA I. SHILYAEVA: shyliaeva@gmail.com
Department of Intellectual Technical Systems, National Research University of Electronic Technology, Bld. 1, Shokin Square, Zelenograd, Moscow, 124498, Russia

SERGEY A. GAVRILOV: pcfme@miee.ru
Department of Intellectual Technical Systems, National Research University of Electronic Technology, Bld. 1, Shokin Square, Zelenograd, Moscow, 124498, Russia

MAXIM V. SILIBIN: sil_m@mail.ru
Department of Intellectual Technical Systems, National Research University of Electronic Technology, Bld. 1, Shokin Square, Zelenograd, Moscow, 124498, Russia

VICTOR B. YAKOVLEV: yakovlev@miee.ru
Department of Intellectual Technical Systems, National Research University of Electronic Technology, Bld. 1, Shokin Square, Zelenograd, Moscow, 124498, Russia

MIKHAIL L. ZHELUDKEVICH: mzheludkevich@ua.pt
Institute of Materials Research, Helmholtz–Zentrum Geesthacht, Zentrumfür Material-und Küstenforschung, Max–Planck–Straße 1, D-21502 Geesthacht, Germany

and

Department of Materials and Ceramic Engineering/CICECO, University of Aveiro, Portugal

NATALIA I. POPENKO: n.popenko@mail.ru
Department of Intellectual Technical Systems, National Research University of Electronic Technology, Bld. 1, Shokin Square, Zelenograd, Moscow, 124498, Russia

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