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SHIL’KO Sergey V., Ph. D. in Eng., Assoc. Prof.
Head of the Laboratory[1]
E-mail: shilko_mpri@mail.ru

PETROKOVETS Ekaterina M.
Researcher[1]
E-mail: katya_petro@mail.ru

ZHANG Qiang, D. Sc. in Eng., Prof.
Professor[2]
E-mail: zhang_tsiang@hit.edu.cn

[1]V.A. Belyi Metal-Polymer Research Institute of the NAS of Belarus, Gomel, Republic of Belarus
[2]Harbin Institute of Technology, Harbin, People’s Republic of China

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STRUCTURAL DESIGN AND TAILORING OF COMPOSITES TO OBTAIN NEAR ZERO COEFFICIENT OF LINEAR THERMAL EXPANSION

The paper discusses the realization of high dimensional stability of products, including minimization of thermal strains by the use of additives having negative coefficient of linear thermal expansion, as well as auxetics having negative Poisson’s ratio. Mechanisms of change of thermal stress state of the porous, dispersion-reinforced and layered materials are established during heating and cooling, depending on elasticity moduli, the size of inclusions, a parity of rigidity of the matrix and the filler as well as boundary conditions. The results of structural design make it possible to improve dimensional stability and reduce residual stresses of microelectronics elements, precise equipment for aerospace engineering and measuring instruments.

Keywords: *measuring instrument, microelectronics elements, functional materials, aluminum alloys, auxetics, zirconium tungstate, thermal expansion, residual stresses, finite element analysis*

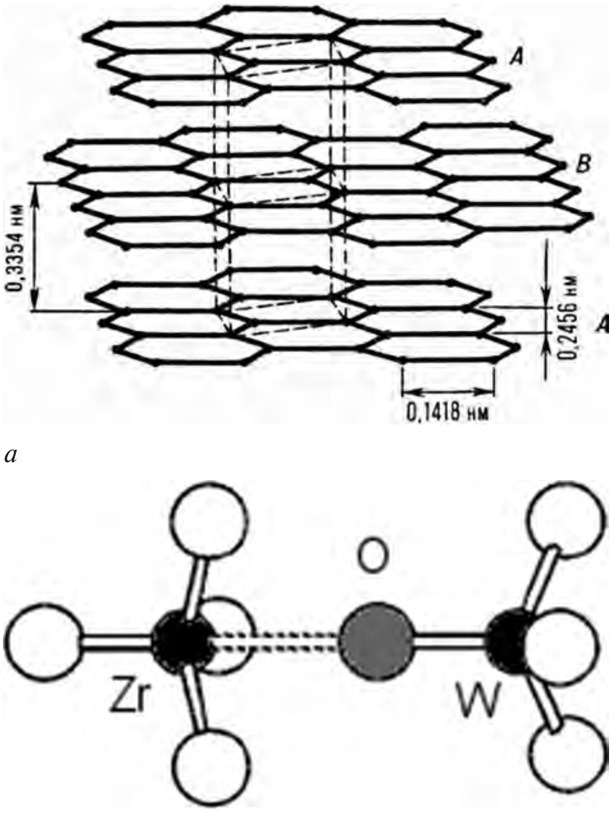
Introduction. It is known [1] that high dimensional stability is assumed as the important requirement to measuring instrument, microelectronics elements as well as the constructions for aerospace engineering and so on. Insufficient rigidity of the structures causes unwanted deformations (contact, bending, tension, compression, and their combinations) under the action of mechanical loads. Obviously, materials with high elastic moduli under tension and shear should be used to reduce strains mentioned above.

Apart from that, thermal strains may cause notable dimensional changes with temperature fluctuations. To minimize this factor, materials are required that have a low (near zero, ideally zero) coefficient of linear thermal expansion (CTE) in a given temperature range.

In both cases, the idea of an improvement of dimensional stability of products by using materials with peculiar (anomalous) physical and mechanical properties, described, for example, in [2], is a very attractive. The possibilities of computer design of composites in terms of managing thermal deformations and stresses are discussed below.

First of all, we assume the “direct” method of thermal strains minimization by mixing of materials with positive and negative CTE [3–8]. The second way of tailoring thermostable composites with zero thermal expansion coefficient is based on the high resistance of auxetics (materials with negative Poisson’s ratio) to form change [9–12]. In this regard, we studied the thermal deformations of auxetic composites with different reinforcement schemes and the ratio of the Poisson’s ratio of the matrix and the filler. In the strength aspect, the concentration of thermal stresses in the inhomogeneity zone (cavity or inclusion) is evaluated.

Structural and functional analysis of materials with zero and negative CTE. A review of the literature on materials that demonstrate zero and negative linear thermal expansion coefficients [3–8] indicates the presence of a number of inorganic compounds possessing the indicated “anomalous” property. These include graphite with $\alpha = –1.22\cdot10^{-6}$ K^{-1} along principal crystallographic axis (Figure 1 *a*) and some minerals: silicon oxides SiO_2 with $\alpha = –4.2\cdot10^{-6}$ K^{-1} and complex oxides of lithium, aluminum, silicon $LiAlSiO_4$ with $\alpha = –4,0\cdot10^{-6}$ K^{-1}; aluminum and phosphorus $AlPO_4$ with $\alpha = –11.7\cdot10^{-6}$ K^{-1}; lead and titanium $PbTiO_3$ with $\alpha = –3.5\cdot10^{-6}$ K^{-1}; thallium and vanadium ($TaOVO_4$ with $\alpha = –4.4\cdot10^{-6}$ K^{-1}; scandium and tungsten $Sc_2W_3O_{12}$ with $\alpha = –2.2\cdot10^{-6}$ K^{-1}; lithium and tungsten LiW_3O_{12} with $\alpha = –6.8\cdot10^{-6}$ K^{-1}; zirconium and tungsten α-ZrW_2O_8 with $\alpha = –8.7\cdot10^{-6}$ K^{-1}; β-ZrW_2O_8 with $\alpha = –4.9\cdot10^{-6}$ K^{-1} and



a

b

Figure 1 — Structural elements of linear thermal expansion graphite (*a*) and compounds ZrW_2O_8 (*b*)

γ-ZrW_2O_8 with $\alpha = -1.0 \cdot 10^{-6}$ K^{-1} (see Figure 1 *b*). Abnormal thermal deformations of these substances are due to the specific configuration of chemical bonds, in which the atoms in the process of heating are displaced in the opposite direction (compared to most substances) as shown in Figure 1.

Direct tailoring of experimental samples with zero and negative CTE. A rather restricted list of the above-mentioned non-structural materials (minerals) makes it relevant to search for and obtain materials in the class of metals, polymers, etc. This is possible by using the methods of composite materials science. For example, the aluminum matrix composites with near-zero coefficient of thermal expansion can be obtained by adding negative thermal expansion phases into the aluminum alloys. Zirconium tungstate, ZrW_2O_8, is such a peculiar material, exhibiting isotropic and NTE properties from 0.3 K to its decomposition temperature of 1050 K. ZrW_2O_8 has three phases: the room stable α-phase, a high-temperature stable β-phase, and high-pressure stable γ-phase. The α-phase ($\alpha = -8.7 \cdot 10^{-6}$ K^{-1}) undergoes a reversible transformation at 155 °C to β-phase ($\alpha = -4.9 \cdot 10^{-6}$ K^{-1}). When subjected to pressures above 0.2 GPa, the cubic α-phase transforms to the orthorhombic high-pressure γ-phase with a dramatically higher CTE value, increasing from -8.7 to $-1.0 \cdot 10^{-6}$ K^{-1}. Upon heating to 120 °C, the γ-ZrW_2O_8 will be reconverted to α-ZrW_2O_8. Therefore, ZrW_2O_8 is an ideal filler material to produce light-weight aluminum matrix composites with near-zero coefficient of thermal expansion.

In this work, a 70 vol.% ZrW_2O_8/Al–Si composite was fabricated by a pressure infiltration process. The matrix was an Al–Si alloy for its low CTE and excellent casting fluidity. A steel mold was filled with ZrW_2O_8 particles, and a vertical pressure was used to press these ZrW_2O_8 particles into a cylindrical preform according to the given volume fraction. The preform was then preheated; while at the same time, the Al–Si allow was heated in a graphite crucible. After the Al–Si alloy was molten, it was poured into the steel mold. A ram was then driven vertically downwards and a mechanical pressure was applied immediately, causing downwards infiltration of the molten Al–Si alloy into the ZrW_2O_8 preform. After the Al–Si alloy solidified completely, the ZrW_2O_8/Al–Si composite was obtained. Since the ZrW_2O_8 will decompose into ZrO_2 and WO_3 at temperature higher than 777 °C, the infiltration temperature should be controlled carefully to avoid the decomposition of ZrW_2O_8.

A mixture of different sized ZrW_2O_8 particles was used to increase their volume fraction (70 %) and fine ZrW_2O_8 particles occupied the interstitial positions around coarse particles efficiently. No particles clustering was observed. A high pressure applied during fabrication and solidification process ensured a complete infiltration of molten Al–Si alloy into ZrW_2O_8 preform, and then the composite was dense and homogeneous.

In the as-fabricated ZrW_2O_8/Al–Si composite, a mixture of γ-ZrW_2O_8 and α-ZrW_2O_8 was found, which is deleterious to the low thermal expansion property of the composite. Annealling treatment was used to reduce the residual stress, causing the conversion of γ-ZrW_2O_8 to α-ZrW_2O_8. This is demonstrated by the relative linear length change with the variation of temperature for the ZrW_2O_8/Al–Si composites. The as-annealed composite exhibits much lower linear length change when compared with as-fabricated composite. Therefore, the mean CTE of the as-annealed ZrW_2O_8/Al–Si composite in the temperature range –40~60 °C is only $1.35 \cdot 10^{-6}$ K^{-1}, exhibiting a near-zero thermal expansion property by the combination of Al–Si alloy and NTE ZrW_2O_8 phase.

Structural design of materials with near zero CTE. Apart from direct mixing with positive and negative CTE of components, the authors consider the possibility of obtaining thermal stable materials by introducing auxetic phase with negative Poisson's ratio into the base structural material. Thus, it is hypothesized that it is possible to compensate for thermal displacements by using the Poisson effect in the region of negative values ν. For a purposeful search for the structure of this material, it is advisable to use a micromechanical approach, which is to simulate a piece of material in the form of a set of identical structural elements (periodicity cells).

Using the finite element method (FEM), micromechanical models were built, covering some important composite structures as shown in Figure 2.

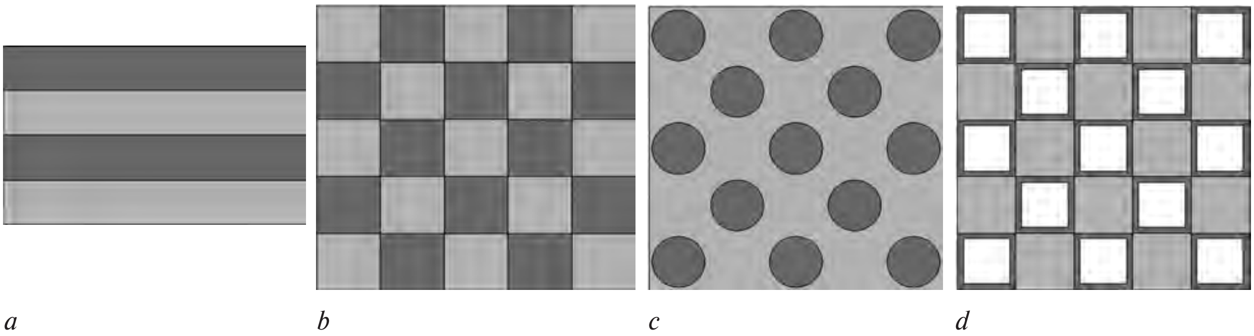


Figure 2 — Model of composite structures with auxetic phase: layered (*a*); porous (*b*); continual with disk (*c*) and rectangular (*d*) inclusions

In all the performed test calculations, the coefficient of thermal expansion $\alpha = 10^{-5}$, typical of polymers, was specified. The calculation of thermal deformations was performed in the environment of the ANSYS software under the assumption of a plane-deformed state. Displacement along the *x* and *y* axes was used as output parameters. A fragment consisting of 5·5 cells was used as a representative volume. Young's modulus of material in all cells had a constant value.

Analysis of thermal linear expansion for a layered system (see Figure 2 *a*) showed that introducing the component with $\nu = -0.5$ makes it possible one to reduce the displacement of u_x by 1.22 times, u_y by 1.44 times as compared to the component with $\nu = 0.48$ (practically incompressible rubber type material). A more noticeable effect is provided by the alternation of layers (a decrease in the named component of displacement by 1.90 and 1.36 times, respectively). The most significant effect of reducing CTE (3.62 and 5.60, respectively) is achieved when using the auxetic phase with the minimum theoretically possible value for an isotropic medium $\nu = -1$.

Similar calculations were carried out for a composite material consisting of square-shaped cells (see Figure 2 *b*) with alternating auxetic and non-auxetic cells. In particular, for $\nu = -0.5$, the predicted reduction in CTE was 1.22 and 1.28, respectively. When varying Poisson's ratio in the interval of admissible values from 0.5 to –1, it was established that thermal displacements close to zero are achieved when $\nu = -1$.

The calculations for a porous material formed by the alternation of square-shaped hollow cells (see Figure 2 *c*) also showed the possibility of reducing the CTE in the region of positive CTE values of this system.

Composite materials with auxetic phase in the form of disk inclusions of equal radius (see Figure 2 *d*) demonstrate a decrease in the CTE of 1.34 and 1.41 times and a matrix of 1.81 and 2.45 times, respectively. Thus, a comparison of the characteristics of thermal expansion for various modeling structures showed that the most noticeable decrease in CTE is achieved using a homogeneous auxetic structure. This is most noticeable with the minimum value of the Poisson's ratio $\nu = -1$. The effect of the structure is smoothed with increasing Poisson's ratio and becomes insignificant when Poisson's ratio exceeds 0.3.

As a technical applications of auxetics, two coupled thermoelastic problems are considered below.

Example 1. Let assume the heating of composite structure (semiconductor laser) in the form of a heterogeneous body consisting of 3 nested regions (Figure 3), the physical and mechanical characteristics of which are listed in the Table 1. Initial data includes absorption coefficient *adep* = 1.8, sample width $y_0 = 0.6$, input stream $I_0 = 1$, $Qrodp = adep \cdot I_0 \cdot \exp[-adep \times x] \cdot \exp[-(y/y_0)^2]$ and temperature of the remote heat sink $Tb = 0$.

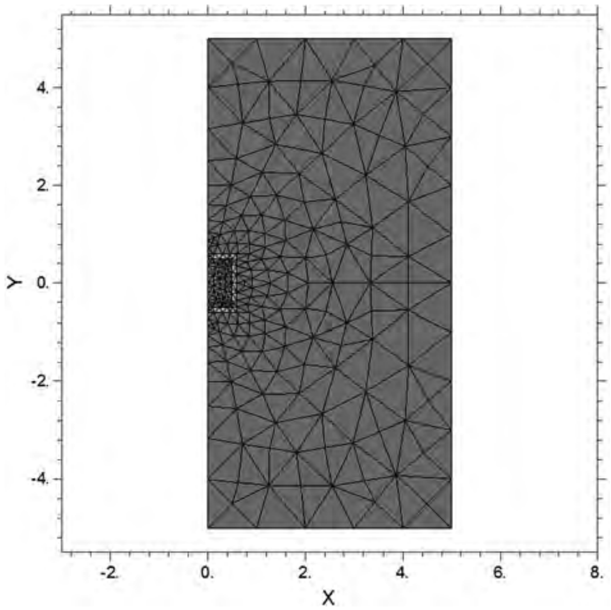


Figure 3 — FEM discretization of composite structure

Table 1 — Initial data for the calculation of the thermal stress state

Characteristics	1	2	3
Coefficient of thermal conductivity *k*	0.083	0.083	0.0098
Heat source *Q*	0	0	*Qrodp*
Young's modulus *E*	117e3	60e3	282e3
Poisson's ratio ν	0.4	varies	0.28
Linear expansion coefficient α	10e-6	16e-6	710e-6

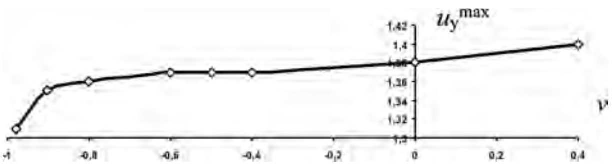


Figure 4 — Dependence of maximal displacement u_y on the Poisson's ratio v

The FlexPDE software for solving partial differential equations is used as a means of numerical analysis of the thermoelastic problem. The investigated two-dimensional region is approximated by triangular finite elements of small size, as compared with the solution region (see Figure 3). An approximate solution is sought as an expansion over the basis of a finite-dimensional space of dimension N.

It was found that the use of material with pronounced auxetic properties $v < -0.85$ leads to a noticeable decrease in the maximum displacements u_y (Figure 4). Also, the dependence of temperature stresses on the Poisson's ratio is of interest, which is characterized by minimum values in the range of negative values of $-0.6 < v < -0.4$ for the component σ_x. It has been established that the use of a material with a negative Poisson's ratio leads to a significant change in the distribution of the components of thermal displacements and stresses in the adhesive joint of the composite layers.

Example 2. Analysis of the thermal stress state of the electrical contact. A common element in electronics is contact made from an electrically conductive material, usually a metal, adhesively bonded to a dielectric (glass, ceramic, polymer) substrate. The calculations show that as a result of contact heating during the installation of electronic components due to thermal expansion of the material, displacements and significant stresses occur in the zone of contact between the contact and the substrate (Table 2). To study the stress-strain state of the contact, an axisymmetric model was constructed in the form of an elastically deformable disk, adhesive bonded to a non-deformable substrate. The following initial data were set: Young's modulus $E = 100$ GPa, linear thermal expansion coefficient $\alpha = 10^{-6}$ K^{-1}, $\Delta T = 373$ K, geometrical parameters of disk: thickness $h = 0.2$ mm, diameter $d = 1$ mm.

An analysis of the results of finite element modeling shows that making a contact from an auxetic material with the Poisson's ratio $v = -1$, all other things being equal, makes it possible to reduce the maximum equivalent stresses by at least 40 %, and their distribution becomes more uniform. An important practical result is also a decrease in maximum thermal displacements. The maximum displacement u_x (Figure 5) decreases

Table 2 — Dependence of the stress-strain state parameters on Poisson's ratio

v	σ_{eqv}, MPa	u_x, mm	u_y, mm
–0.98	126	0.00361	0.00182
0.0	162	0.125	0.0862
0.3	180	0.152	0.128
0.48	204	0.168	0.164

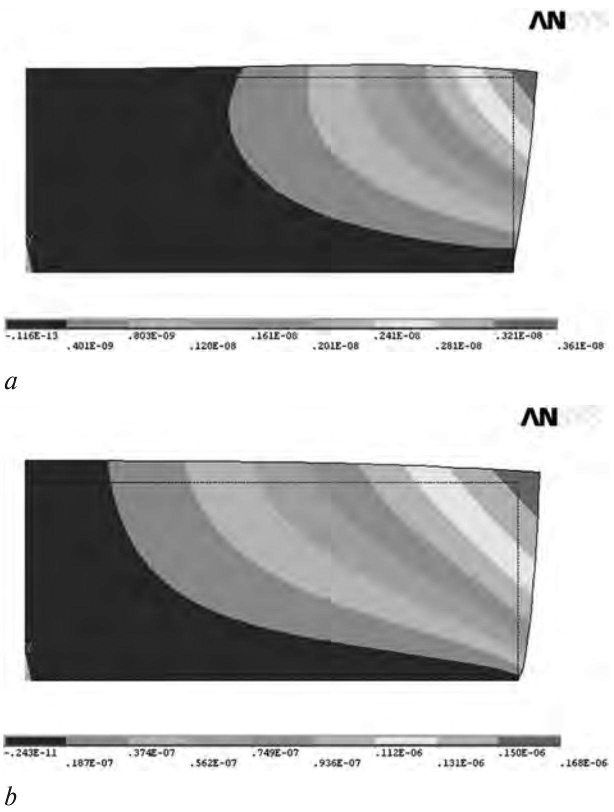


Figure 5 — Displacements along axis x: $v = -0.98$ (a) and $v = 0.48$ (b)

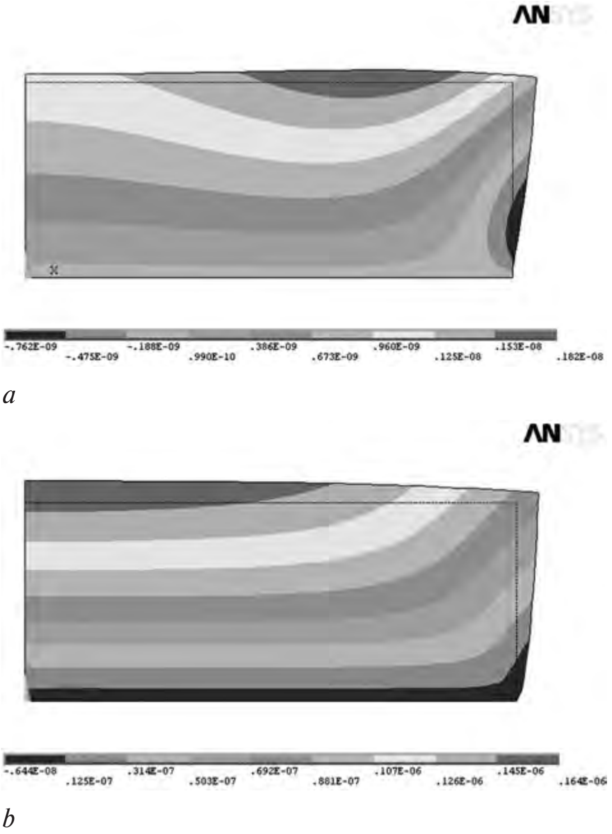


Figure 6 — Displacements along axis y: $v = -0.98$ (a) and $v = 0.48$ (b)

by more than 40 times (the zone of their localization moves from the center to the edge of the contact), and the similar component u_y (Figure 6) decreases by more than an order of magnitude in comparison with the value $v = 0.3$ for most materials (metals) used to make electrical contacts for the smallest value of the Poisson's ratio $v = -1$. Thus, the use of contacts from auxetic materials in electronic products has obvious technical advantages.

Conclusions. We assume the "direct" method of thermal strains minimization by mixing of materials with positive and negative CTE. The as-annealed composite exhibits much lower linear length change when compared with as-fabricated composite. For example, the mean CTE of the as-annealed ZrW_2O_8/Al–Si composite in the temperature range –40~60 °C is only $1.35 \cdot 10^{-6}$ K^{-1}, exhibiting a near-zero thermal expansion property by the combination of Al–Si alloy and NTE ZrW_2O_8 phase.

It is shown that the use of auxetic components, allowing for a wide range of regulation of bulk compressibility and shear rigidity, is a way to create thermostable composites for the manufacture of structural elements of measurement tools. Thermoelastic analysis of the heating of the composite structure showed that the use of components with pronounced auxetic properties ($v < -0.85$) leads to a noticeable decrease in the maximum displacements u_y. The temperature stresses are characterized by minimum values in the interval of negative values $-0.6 < v < -0.4$.

The numerical solution of the coupled thermoelastic problem of heating a composite structure of a semiconductor laser showed that the use of materials with pronounced auxetic properties ($v < -0.85$) leads to a noticeable decrease in thermal displacements u_y. Temperature stresses are characterized by minimal values in the range of negative values Poisson's ratio $-0.6 < v < -0.4$.

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С.В. ШИЛЬКО, канд. техн. наук, доц.
заведующий лабораторией механики композитов и биоматериалов[1]
E-mail: shilko_mpri@mail.ru

Е.М. ПЕТРОКОВЕЦ
научный сотрудник[1]
E-mail: katya_petro@mail.ru

Цян ЧЖАН, д-р техн. наук, проф.
профессор[2]
E-mail: zhang_tsiang@hit.edu.cn

[1]Институт механики металлополимерных систем им. В.А. Белого НАН Беларуси, г. Гомель, Республика Беларусь
[2]Харбинский политехнический университет, г. Харбин, КНР

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ФОРМИРОВАНИЕ СТРУКТУРЫ И РЕГУЛИРОВАНИЕ СВОЙСТВ КОМПОЗИТОВ ДЛЯ ПОЛУЧЕНИЯ БЛИЗКОГО К НУЛЮ КОЭФФИЦИЕНТА ЛИНЕЙНОГО ТЕПЛОВОГО РАСШИРЕНИЯ

Обсуждается проблема получения высокой формостабильности изделий, в том числе минимизация термических деформаций за счет использования компонентов с отрицательным коэффициентом линейного теплового расширения, а также ауксетиков с отрицательным коэффициентом Пуассона. Установлены механизмы изменения термонапряженного состояния пористых, дисперсно-армированных и слоистых материалов при нагреве и охлаждении в зависимости от модулей упругости, размера включений, соотношения жесткости матрицы и наполнителя, а также граничных условий. Результаты структурного дизайна позволяют повысить размерную стабильность и уменьшить остаточные напряжения элементов микроэлектроники, точного оборудования для аэрокосмической техники и измерительных приборов.

Ключевые слова: *измерительный прибор, элементы микроэлектроники, функциональные материалы, алюминиевые сплавы, ауксетики, вольфрамат циркония, тепловое расширение, остаточные напряжения, анализ методом конечных элементов*

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