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Finite element analysis of thermal stress in Cu_2O coating synthesized on Cu substrate

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ABSTRACT

Purpose: The paper aims to find the magnitude and nature of thermal residual stresses that occur during cooling of a copper sample with a thermally synthesized oxide layer of Cu_2O.

Design/methodology/approach: Thermo-mechanical analysis was performed by the finite element method using Ansys Software. The results of thermal analysis were used to study the resulting stress-strain state of the thin film/coating system after cooling.

Findings: Based on the modeling results, the paper determined the most stress-strain areas of the sample with a coating, which are the free edges of the interfaces between the copper substrate and the Cu_2O oxide layer.

Research limitations/implications: The main limitations of the study are the use of certain simplifications in the condition setup, for instance, uniform cooling of the thin film/coating system, homogeneity and isotropy of substrate and thin film materials, invariance of their properties with temperature changes, etc.

Practical implications: The results obtained can be used to control the stress-strain state of the thin film/coating system and prevent deformations and destruction of thin-film structures during their production and operation of products with them.

Originality/value: The study of new promising methods for the formation of oxide nanostructures, for instance in a plasma environment, requires a sufficient theoretical basis in addressing the origin and development of stresses.

Keywords: Stress-strain state, Residual stress, Oxide layers, Thermo-mechanical modelling, Coefficient of thermal expansion

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METHODOLOGY OF RESEARCH, ANALYSIS AND MODELLING

1. Introduction

Advances in science in the field of thin-film structures are one of the drivers of development of many industries. Of particular note are the oxide nanostructured layers CuO/Cu_2O and nanotubes, which due to their properties have found application in the production of a new generation of lithium-ion batteries [1], semiconductors [2], solar panels

[3,4] and more. In addition, the direction of using nanomaterials as a reinforcing element in the production of nanocomposites with ceramic, metal and polymer matrices is promising [5,6]. Among the variety of methods for synthesizing these structures, the most common is thermal oxidation [7]. A more promising method in terms of intensification and control of the synthesis of oxide layers and nanotubes is the use of plasma [8].

One of the key parameters of the growth of oxide nanostructures is the process temperature, which at the same time causes thermal stresses [9]. The development of these stresses is due to the difference in thermo-mechanical properties of the substrate and film materials. The sum of thermal, external stresses, and stresses that occur in the film during synthesis results in the residual stresses acting in the film-substrate system. These residual stresses directly affect the properties of thin films [10,11]. Prevention of deformations and destruction of thin-film structures is associated with understanding the causes of the origin of residual stresses, their sign and propagation in the film-coating system.

Analytical models [12,13], and mechanical (destructive) and physical (non-destructive) methods [14] have been proposed to determine thermal stresses, but their use does not allow us to analyze the process of stress formation and create a scientific basis necessary for effective control of the stress-strain state of a coated product. Computational models are limited by a number of assumptions, and the issue of determining the localization of residual stresses remains open. With the development of computer technology, software products such as Abaqus, Nastran, Ansys, etc. have appeared. Currently, the use of the latter is a powerful tool for calculating residual stresses, which makes it possible to study their propagation in the volume of the studied bodies, determine critical areas and prevent possible destruction of thin films and coatings [15]. In addition, process modeling allows us to take into account not only one source of stress occurrence, but also to simultaneously study the influence of several factors.

As mentioned above, the difference in the thermo-mechanical properties of the film and substrate materials causes stresses. Thus, one of the important parameters that significantly affects the thermal residual stresses is the coefficient of thermal expansion (CTE) [12]. In the case of formation of oxide thin-film layers on a metal substrate, the difference in CTE values can reach ten or more times [16]. The process of synthesis of Cu_2O/CuO oxide layers on a copper substrate can result in system temperature reaching 700 °C upon completion of their formation [17]. In many cases, the process temperature is the main parameter which affects the intensification of thin films growth [18]. Further cooling of the substrate with the oxide layer to ambient temperature given that the CTE of the latter is significantly less than the CTE of the copper substrate leads to compressive stresses in the oxide layers.

The analysis of thermal residual stresses by the finite element method that occur in the coating-substrate system is presented in the works of many researchers.

Xi Zhang *et al.* studied the effect of the number of layers and their thickness of gradient coatings on the thermal residual stress distribution of the Ni/Yttria-stabilized Zirconia system (YSZ) using the Ansys package [19]. This paper showed the effect of using intermediate layers on the reductions of values of residual stresses, which reduces the probability of destruction of coatings.

The findings of the cooling process of the coated substrate and the effect of residual stresses on the destruction of the latter are given in [20]. Y. Kahraman established the relationship between the thickness of coatings and the value of stresses by the finite element method, and the findings show that the increase in thickness of boride layers leads to the decrease in thermal residual stresses. In addition to finding the value and sign of residual thermal stresses, computer simulation can prevent the destruction of thin-film structures that occurs when the stresses reach the limit values.

Q.M. Yu *et al.* studied the crack formation process of thermal barrier coatings using the Abaqus software product [21]. It was found that the destruction of coatings can be observed when the coating-substrate system is cooled at the interface of the thermally grown oxide (thermally grown oxide) and the intermediate coating layer, and when the system is heated, but the formation of cracks can be observed at the interface of the thermally obtained oxide and the main protective layer of the coating.

Computer simulation of residual stresses can be performed for both thick and thin coatings [22,23]. Finite-element modeling of thermal stresses in oxide layers obtained by ion-beam sputtering on substrates of different materials is presented in the paper of X. Tian *et al.* [22]. The obtained results were compared with the results of calculations using an analytical model and verified experimentally. The results of calculations correlate well with the results of modeling and are confirmed by experiments. Based on the results obtained, it is concluded that it is possible to use numerical methods to prevent the destruction of thin-film structures and optimize the spraying process.

Experimental verification of the obtained results of finite element analysis of thermal stresses in oxide layers was performed H.-C. Chen *et al.* [24]. According to the results of the conducted research, it was found that the error in the results of modeling and experiments amounted to 2...6%.

Experimental validation of numerical simulations performed using the Ansys package was performed by M. Elhoriny *et al.* [25]. Comparing the obtained results, it can be concluded that the finite element analysis of residual stresses is adequate and complies with experimental data.

The aim of the paper was to analize the thermal residual stress distribution in Cu_2O coating on Cu substrate to determine the most likely damage initiation location in the above mentioned system. To achieve this, a thermo-mechanical finite-element model was implemented to estimate thermal and stress fields developed in coating/substrate system. The results of thermal analysis was applied as input parameters for the subsequent stress-strain analysis. The research is the preliminary work for understanding the crack initiation mechanism and behaviour of plasma synthesized coatings under the total residual stresses acting in the film-substrate system.

2. Finite element modelling

2.1. Simulation procedure

Ansys Academic Software (Workbench 2021) was used to study the distribution of thermal residual stresses in a thin film-coating system. The algorithm for performing finite element analysis is shown in Figure 1.

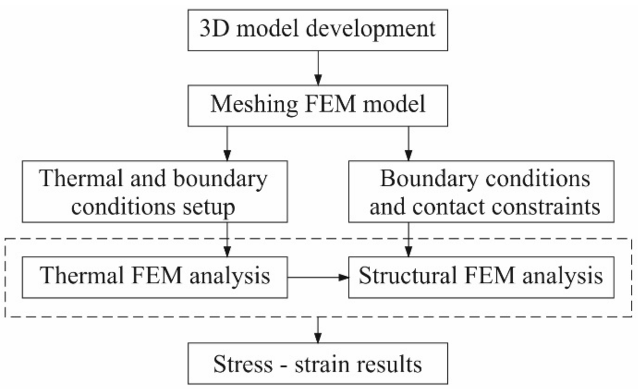

Fig. 1. Flowchart of the FEM procedure

2.2. Model geometry

At the first stage, a 3D model of Cu substrate with Cu_2O oxide layer was built using the SpaceClaim module integrated in Ansys software CAD. A schematic of 3D finite element model is shown in Figure 2. The model can also be imported from any other CAD software product. The substrate is a cylinder with a diameter of 14.0 mm and a height of 2.0 mm. An oxide layer with a thickness of 0.1 mm is evenly obtained on the upper end surface of the sample. The contact limit of the substrate with the coating was set as "Bonded". Since the model is symmetric, ¼ of the model was used to reduce the calculation time.

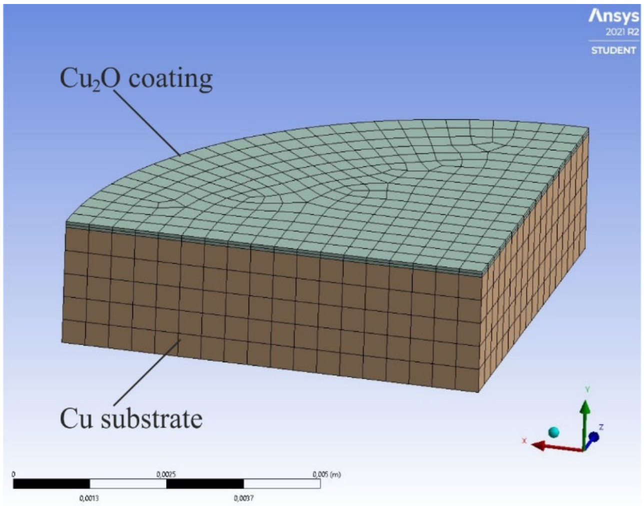

Fig. 2. Calculated 3D model of the sample with the oxide layer

2.3. Meshing

After creating the model, the next step is to build a finite element mesh. This is one of the most important stages of Finite Element Analysis, since it ensures the adequacy of calculation results.

The accuracy of finite element calculations depends on the correct choice of the type and size of finite elements. From the point of view of Finite Element Analysis, it is optimal to divide it into elements that have the shape of the simplest equilateral figures.

For the purpose of this paper, a rectangular shape of elements with element size of 0.4*0.4 mm was set. Looking ahead, it should be noted that the mesh size was chosen by gradually reducing the size with an analysis of the results obtained after all the process settings. Further reduction of the size does not affect the difference in results, but significantly increases the calculation time. An element order was set as "Quadratic". For a thin film with the thickness of 0.1 mm herein, 2 elements were set for the film thickness. The total number of elements of the model is 2525, and the model consisted of 13768 nodes.

2.4. Governing equations

The total strain induced in the material can be expressed as [26]:

$$\varepsilon_\Sigma = \varepsilon_{th} + \varepsilon_{el}, \quad (1)$$

where ε_{th} and ε_{el} are thermal and elastic strains respectively.

The thermal stress can be calculated as:

$$\varepsilon_{th} = \alpha(\Delta T), \quad (2)$$

where is the thermal expansion coefficient and is the temperature difference between T_2 and T_1.

The elastic stress can be calculated with Hooke's law [26]:

$$\sigma = D\varepsilon_{th}, \quad (3)$$

where D is the elastic matrix related to the elastic modulus, E, and Poisson's ratio, v, for the given material at given temperature.

The stress-strain state for isotropic material can be expressed by combining of Eq. (1) – (3):

$$\varepsilon_{xx} = \frac{1}{E}\left[\sigma_{xx} + v\left(\sigma_{yy} + \sigma_{zz}\right)\right] + \alpha(\Delta T), \quad (4)$$

$$\varepsilon_{yy} = \frac{1}{E}\left[\sigma_{yy} + v\left(\sigma_{xx} + \sigma_{zz}\right)\right] + \alpha(\Delta T), \quad (5)$$

$$\varepsilon_{zz} = \frac{1}{E}\left[\sigma_{zz} + v\left(\sigma_{yy} + \sigma_{xx}\right)\right] + \alpha(\Delta T), \quad (6)$$

where E and v are Young's modulus and Poisson's ratio, respectively.

2.5. Initial and boundary conditions

At the third stage, the initial and boundary conditions were set. For the calculated model, the regions of symmetry were given and the following materials were assigned: Cu for the substrate and Cu_2O for the thin film. The properties of materials in terms of layer thickness are unchanged, i.e. homogeneous and isotropic. The properties of the materials are shown in Table 1.

Table 1.
Properties of materials used in the study

Properties \ Layer	Cu substrate	Cu_2O oxide layer
CTE α, $\times10^{-6}$ 1/°C	16.5	0.9
Elastic modulus E, $\times10^{3}$ MPa	123.0	25.0
Poisson's ratio v	0.35	0.462

The initial temperature of the thin film coating system was 500°C, 600°C, and 700°C, which corresponds to the synthesis temperature of the oxide layers, and the system cools down to ambient temperature (T_2 = 25°C). Cooling occurs naturally, i.e. without artificial acceleration, while the convective heat transfer coefficient was set to 10 W/m^2*s. No heat exchange with the surface on which the coated sample lies is observed, and the conditions for sample fixation are absent. The cooling time of the system was set to 4000 seconds.

The next step is to set the analysis task. In this case, 2 separate tasks – Thermal and Mechanical – are related to each other. After the settings, the paper suggests a condition that the results of calculations of the thermal problem are used in Mechanical analysis.

3. Results and discussions

The modeling helped to obtain the results of the distribution and magnitude of thermal stresses that occur during cooling of a copper cylindrical sample with a synthesized Cu_2O oxide layer at one of its edges.

Figure 3 shows a temperature trend for the thin film/coating system during a set cooling time. It can be seen that significant cooling from 700°C to 200°C occurs during approximately the first 500 seconds, and then it monotonically falls to the ambient temperature (T_2 = 25°C). Since the Cu_2O oxide layer is thin relative to the substrate ($t_c << t_s$), it can be seen that the temperature of the Cu_2O layer corresponds to the temperature of the substrate during cooling.

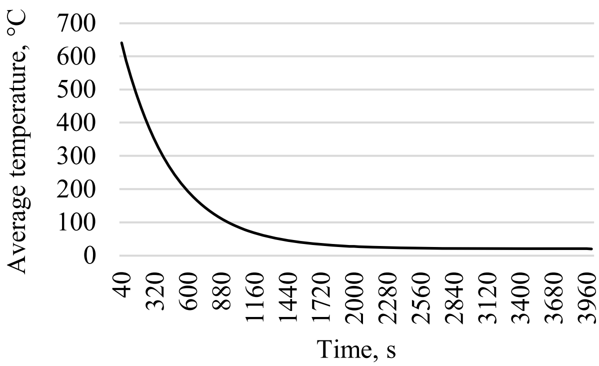

Fig. 3. Cooling graph of Cu_2O/Cu system

Figures 4 and 5 show the results of modeling the cooling of a thin film/coating system synthesized at different temperatures: tangential, radial, and maximum and minimum Von-Mises effective stresses of Cu_2O/Cu system.

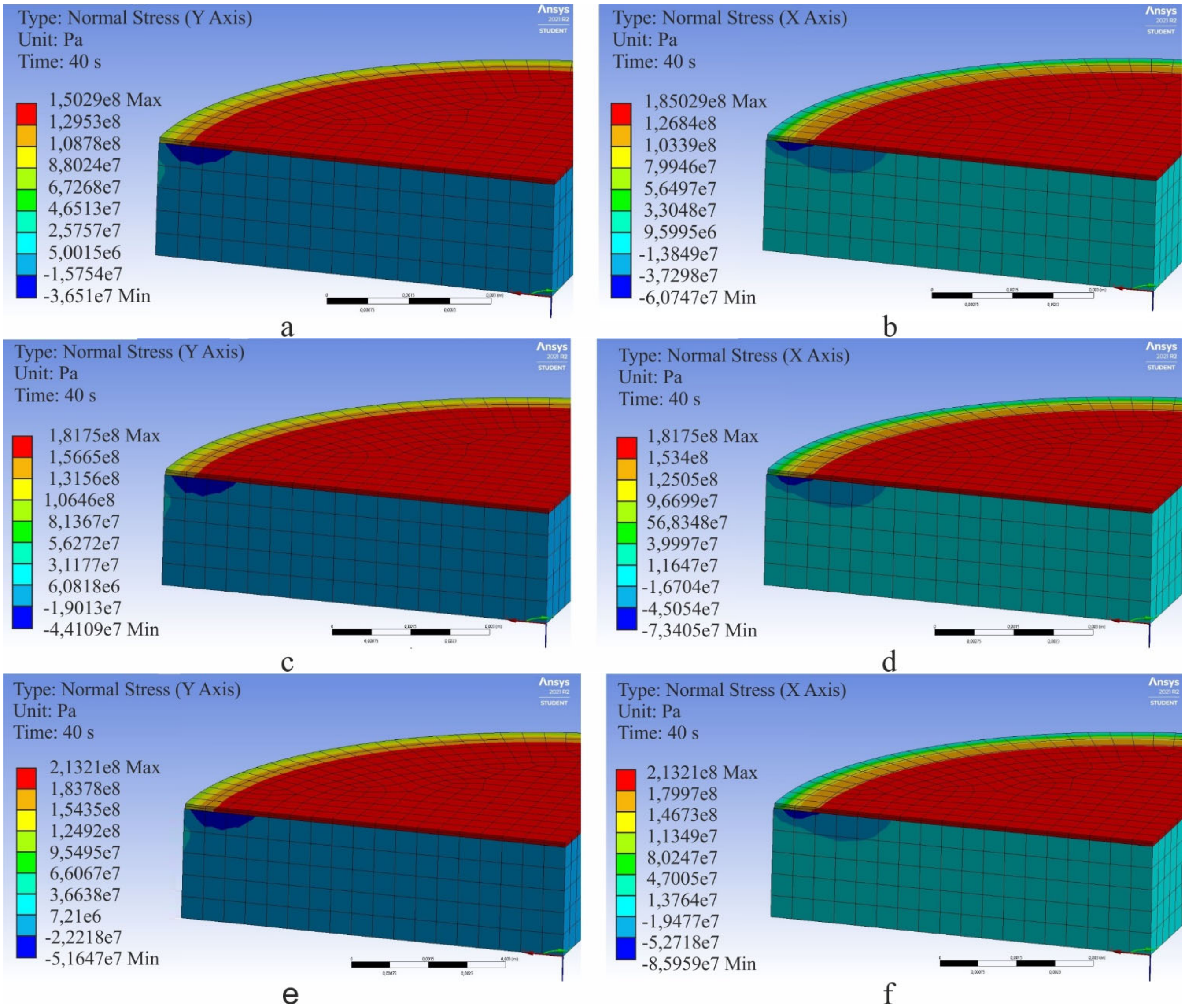

Fig. 4. Simulation results at different synthesized temperatures: tangential and radial stresses at 500°C (a, b), 600°C (c, d), and 700°C (e, f), respectively

The analysis of the results of the distribution of temperature fields in the oxide layer and the substrate during cooling of this system shows that the largest temperature gradient occurs between these bodies due to the difference in coefficients of thermal expansion, which leads to thermal stresses. The above figures show that compressive residual stresses occur in the oxide layer, while tensile stresses occur in the metal substrate. These results are expected, since the coefficient of thermal expansion of the Cu_2O layer is significantly smaller than the coefficient of thermal expansion of the copper substrate ($\alpha_c \ll \alpha_s$), which is also confirmed by the results of other studies [12,13].

It can be seen, that with increasing of deposition temperature from 500°C to 700°C, the residual stress also increased. There was found that the maximum values of tangential and radial compressive residual stresses in Cu_2O obtained at 700°C are the same – 213.2 MPa. The minimum compressive stress in Cu_2O (150.3 MPa) was obtained at 500°C in tangential direction. The increase in tensile residual stress of Cu substrate (from 37.3 MPa to 86 MPa) with increasing of deposition temperature from 500°C to 700°C was found. It should be added that maximum value of tensile stress in Cu substrate was obtained in radial direction at 700°C, while 51.6 MPa was found in tangential direction.

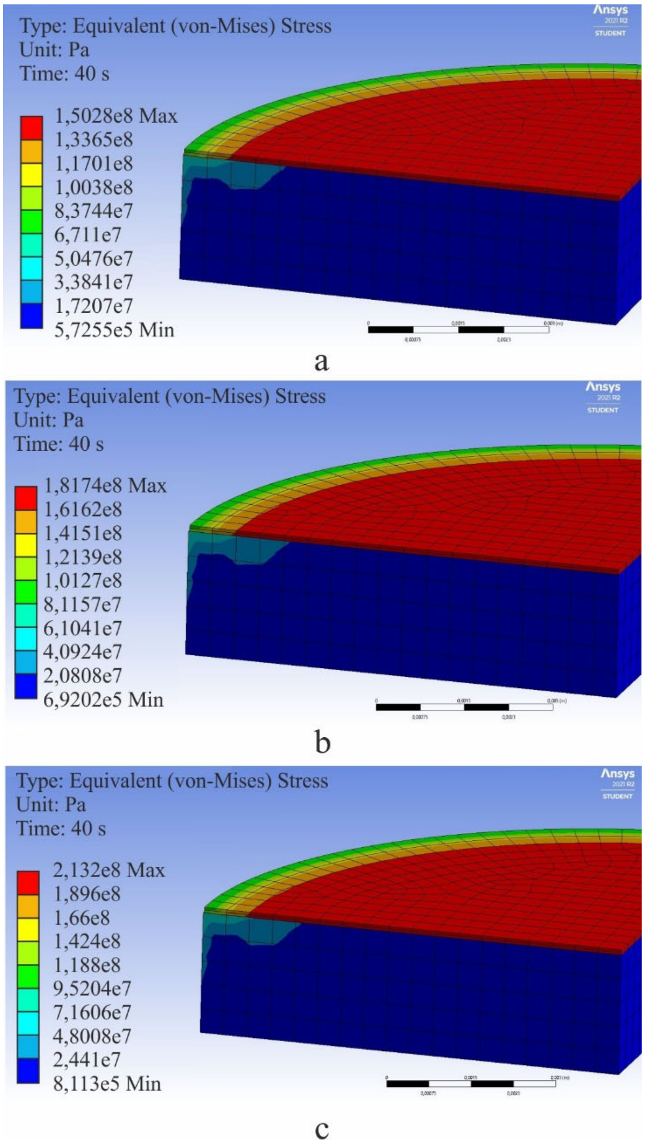

Fig. 5. Simulation results at different synthesized temperatures: maximum and minimum Von-Mises effective stresses at 500°C (a), 600°C (b), and 700°C (c)

Analyzing the obtained results it can be concluded that more intensive cooling of the coated sample in the edge zones and larger deformations lead to stresses that differ significantly in magnitude and distribution compared to the central part of the model. Therefore, the highest stress values can be observed in these edge zones.

Based on the modeling results, it is possible to identify areas where cracks are likely to occur, namely at the interface between the thin film and the substrate in the edge areas of the sample. In addition, it should be noted that these areas largely depend on the shape of the thin film sample (for instance, rectangular plate, cylinder, etc.), if the same materials of the thin film/coating system are considered. Similar results on the distribution of thermal stresses in coatings and thin films were obtained in the papers of other researchers [26,27], which confirms the reliability of the results.

It is difficult to conclude whether the maximum value of thermal stress is enough to initiate cracks or other forms of coating failure due to thermal stress is one of many components in total residual stress developed in coating/substrate system (e.g. intrinsic stresses induced by the lattice mismatch). Futher investigation of total residual stress in Cu_2O/Cu system should be carried out.

To verify the accuracy of the FE model used in this study, the residual thermal stress simulation was carried out using the same sample materials and process temperatures as in Ref. [7]. Baranov *et. al.* reported that any coating failure were obtained.

4. Conclusions

1) The study involved the modeling of thermal stresses in the Cu_2O oxide layer obtained on a copper substrate resulting from their cooling after the completion of the oxide layer synthesis process from the operating temperature of the process (500°C, 600°C, and 700°C) to the ambient temperature (25°C). With the increase of process temperature, the thermal residual stress in coating and substrate increase.
2) For thin-film structures with a clear separation boundary with the substrate, the thermal residual stresses depend on the physical and mechanical properties of the film and substrate materials (linear expansion coefficient, modulus of elasticity, Poisson's ratio), and the heating temperature of the system during the synthesis of the oxide layer.
3) When a 2.0 mm thick copper substrate with a 0.1 mm thick Cu_2O oxide layer is cooled from 700°C to 25°C, compressive thermal stresses occur in the latter ($\alpha_c << \alpha_s$) and reach maximum value of 213.2 MPa. The maximum value of tensile residual stress in Cu substrate was found 86 MPa.
4) The edge zones at the interface between the oxide layer and the substrate are those that receive the greatest deformations and stresses as a result of system cooling, which are most likely the places of crack formation and the initiation of destruction.
5) Futher investigation of total residual stress in Cu_2O/Cu system should be carried out due to other types of stresses are appeared during coating growth. After that the conclusion about coating failure and possible ways of its prevention can be done.

6) Since significant compressive stresses occur at the interface between the thin film and the substrate, the question arises whether temperature stresses can cause the thin layer of copper oxide to detach from the copper substrate. Therefore, the next step of research is to determine the stability of the thin film in order to prevent the destruction of thin-film structures.

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