The Effect of Ultrasonic Treatment on the Adhesion of a Surface Composition Made of SME Materials at Various Stages of Its Formation

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***Abstract*– The article describes results of an experimental study of ultrasonic treatment as a part of a complex temperature-force effect on the adhesive properties of a surface composition made of materials with thermoelastic phase transformations at various stages of formation: at the stage of the base preparation; at the stage of the coated part finishing. The established patterns of modifying effects of ultrasonic treatment on the distribution of residual stresses at the surface-to-surface interface make it possible to predict the functional and mechanical properties of the layers that make up the surface composition.**

Keywords* – *residual stresses, Sachs method, adhesion, ultrasonic treatment, coatings, high-velocity oxygen-fuel spraying

I. INTRODUCTION

At present, various types of thermal spraying are being increasingly used to form coatings. The the most widely used one is the technology of high-velocity oxygen-fuel spraying (HVOF). This method allows obtaining coatings of the required thickness with high adhesive strength and low porosity at high productivity. HVOF also allows us to adjust the temperature of the gas jet; this expands the range of powder materials used for spraying. However, thermal spraying technologies, including HVOF, are used restrictedly to create coatings on details that work under conditions of increased loads, because of the adhesion strength that does not meet the requirements. Therefore, in order to expand the scope of use and the nomenclature of details subjected to HVOF, it is necessary to ensure reliable adhesion strength at the border “base – coating made of material with shape memory effect (SME material)”, as well as between layers. We also need to develop ways to increase this strength. The relevance and significance of this task is confirmed by the sharp increase in global publishing activity on the stated subject [1, 2].

Adhesive strength is a combination of two interaction types of the sprayed coating with the substrate: mechanical and chemical ones. The mechanical interaction is determined by the adhesion of the coating particles to the substrate due to the unevenness of the surface and is not the main factor in ensuring adhesion. The chemical interaction takes place due to the formation of common compounds between the elements of the sprayed material and the substrate, mutual penetration into the crystal lattice of the coating atoms and the substrate. The chemical component of adhesion is the main indicator. Therefore, many scientific works [3] study how to increase it in all types of coatings and films.

The peculiarity of thermal spraying methods lies in the fact that the detail, as well as the coating particles, is still heated and then cooled sharply due to the removal into the surrounding atmosphere. This causes residual stresses, which either drastically change the sign at the interface of the coating-substrate, or dramatically change the magnitude of the voltage, which is a negative factor for adhesion [4]. This is especially important for coatings made of SME materials, since during operation the coating can undergo both a temperature change, leading to a direct or reverse martensitic transformation, and to deformation due to the occurance of deformation martensite. These processes lead to a change in the structure of the coating material due to the shift of atoms, which, in turn, causes stresses at the interface of the coating-substrate, and with multiple phase transformations can cause delamination. This is characteristic of coatings with low adhesion and in cases when adhesion of the coating was provided only by the mechanical component of adhesion. All this explains the necessity to develop technologies that provide for obtaining coatings made of SME materials with high adhesive strength.

There are different methods to improve adhesion: deposition of intermediate layers with high interaction potential both with the substrate and with the coating; optimization of the substrate temperature; optimization of the granulometric composition, activation of characteristics and thermophysical properties of the sprayed material; optimization of HVOF modes including particles’ speed and combustible gas composition. All these methods depend on the requirements for coating, and on the operating conditions of the product. In order to increase the adhesion we can use acoustic stimulation, ultrasonic action, heat treatment, which initiates diffusion processes in the contact zone of the coating with the substrate. Earlier, we developed a technology for electromechanical pulsed processing, which ensures an increase in adhesion by creating reliable contact points at the interface “coating-substrate” [5]. The advantage of this technology is the possibility to optimize the magnitude and direction of technological residual stresses with regard to operational requirements (Pat. RF № 2625508). An analysis of existing technologies for preparing the surface of the substrate before coating has shown that in order to increase the adhesive and cohesive strength when forming the surface layers or compositions of SME materials, it is optimal to use ultrasonic treatment (UST) at all stages of coating production. This worl presents the results of a study of one of the ways to increase the adhesion strength of the base and the coating made of SME material formed by means of HVOF with subsequent UST [4–8].

II. Materials and Equipments

To assess the adhesion strength we used the pin method (State Standard 28844-90 "Gas-thermal, hardening and restoring coatings"). The tests were carried out on the machine Instron 8801.

Formation technology of the surface composition included several stages: preparation of the base using UST; mechanical activation of the deposited material, enhanced by ultrasonic action; application of a transition adhesive layer of a material having unlimited solubility with the base material and chemical affinity with the material of the functional layer. High-velocity oxygen-fuel spraying was carried out on a GLC-720 installation in protective atmosphere of argon with subsequent thermomechanical treatment combining surface plastic deformation (SPD) and UST. The temperature regime was chosen taking into account the possibility of beginning the process of thermal diffusion mass transfer and increasing the adhesion of the substrate-coating. Similarly, a functional layer made of SME material was applied at a temperature regime that provides the beginning of the process of thermal diffusion mass transfer to increase the interlayer adhesion of the coating-coating.

We took Steel 45 as a base when developing surface layers made of SME materials with increased requirements for adhesion. We used nickel with unlimited solubility with iron and chemical affinity with the material of the functional layer as a transition layer. We applied a three-component SME material Ti33Ni49Zr18 to form a functional layer. This material has a high-temperature memory effect: it obtains 20° C in a martensitic state. We also applied a four-component SME material Ti40Ni25Cu25Hf10 which has a low-temperature shape memory effect, and is at room temperature of 20° C in the martensitic state.

Preparation of the base included an ultrasonic treatment with a hardening spherical element, which allows increasing the dislocation density on the base surface. This increasese the number of vacant places in the structure of the base material for diffusional penetration of coating atoms. This technology increases the value of the chemical component of adhesion. Thus, ultrasonic treatment can be attributed to the mechanical activation of the substrate surface before coating. Fig. 1. shows installation for UST

After base surface activation, the samples were subjected to chemical treatment, including degreasing of the surface and etching with a mixture of hydrochloric and nitric acids. The preparation of the applied material consists in the mechanical activation (MA) of the material, which was carried out in the Hephaestus-2 AGO-2U ball mill. Coating was carried out on a modernized GLC-720 installation in an argon protective atmosphere.

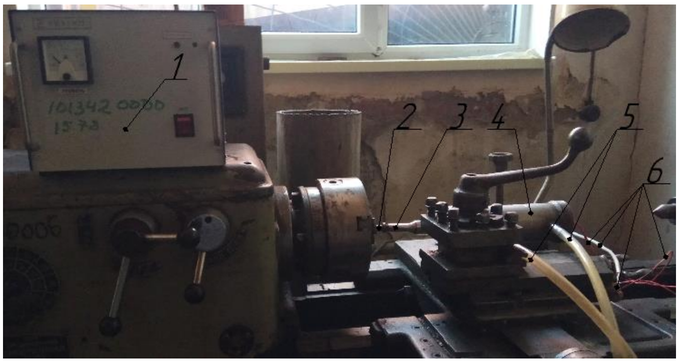


Fig. 1. Installation for ultrasonic treatment: 1 – ultrasonic generator; 2 – coated sample; 3 – waveguide with indenter; 4 – magnetostrictive transducer; 5 – water cooling system; 6 – wires

Studies of residual stresses were carried out on special cylindrical samples of Steel 45 with a height of 20 mm and diameters of 8–10 mm. Part of the samples were subjected to mechanical processing, after which they were coated (Ni – TiNiZr and Ni – TiNiCuHf). The second part of the samples was subjected to a full processing cycle using UST at the stages of base preparation and at the finishing stage after coating according to the earlier described technology. The formation of a composite coating includes: applying a transition adhesive layer on a high-velocity oxygen-fuel spraying device GLC-720 in argon protective atmosphere; the subsequent combined SPD and UST treatment under temperature, providing the beginning of the process of thermal diffusion mass transfer and an increase in the adhesion “substrate-coating”; deposition of a functional layer of SME material by high-velocity oxygen-fuel spraying in argon protective atmosphere, followed by a combined SPD and UST treatment under temperature effects/ This ensures the start of the process of thermal diffusion mass transfer and increases interlayer adhesion "coating-coating".

The thickness of the obtained coatings on samples is 0.1–1 mm, the thickness of the adhesive layer varied 0.05–0.1 mm, the resulting height of cylindrical specimens is 20 mm and diameters are 8.2–10 mm. When preparing, the face ends of the samples were ground and polished on a universal surface grinder with a horizontal spindle 3B722 and on an automatic grinding and polishing machine MR-300.

We determined the residual stresses by the Sachs method in the following sequence: measurement of the outer diameter and length of the sample with further drilling and boring of the hole with measurement of circumferential and axial deformation at the outer radius. Drilling was carried out on a 1E61M engine lathe. Dimensioning was carried out using the ACCURA coordinate measuring machine. It is designed to measure the geometrical dimensions samples with complex shape, deviations of the shape and location of the surfaces shown in Fig. 2

Removal of coating material layers on a cylindrical sample from the hole was carried out by chemical etching using HCl (2 ml) + HF (1 ml) + HNO_3 (1 ml) + H2SO4 (1 ml) solution with tracking changes in axial and circumferential deformations on the outer radius. The calculation of residual stresses (circumferential, radial and axial) was carried out in the software package MathCad Prime 4.0 according to equations (1–3) [9, 10]:

$$\sigma_r(r) = \frac{E}{1-\mu^2} \cdot \frac{R_2^2 - r^2}{2r^2}\left[\varepsilon_{\theta 2}(r) + \mu\varepsilon_{z2}(r)\right] \quad (1)$$

$$\sigma_z(r) = \frac{E}{1-\mu^2}\left[\frac{R_2^2 - r^2}{2r^2}\left(\frac{d\varepsilon_{z2}}{dr}(r) + \mu\frac{d\varepsilon_{\theta 2}}{dr}(r)\right) - \varepsilon_{z2}(r) - \mu\varepsilon_{\theta 2}(r)\right] \quad (2)$$

$$\sigma_\theta(r) = \frac{E}{1-\mu^2}\left[\frac{R_2^2 - r^2}{2r^2}\left(\frac{d\varepsilon_{\theta 2}}{dr}(r) + \mu\frac{d\varepsilon_{z2}}{dr}(r)\right) - \frac{R_2^2 + r^2}{2r^2}\left(\varepsilon_{\theta 2}(r) + \mu\varepsilon_{z2}(r)\right)\right] \quad (3)$$

where: σ_r (r) is the radial residual stress; σ_z (r) is the axial residual stress; σ_θ (r) is the axial residual stress; r is the radius of the drilled or pickled hole; R2 is the outer diameter of the cylindrical sample; ε_θ2 (r) is the deformation in the circumferential direction; ε_z2 (r) is the deformation in the circumferential direction; E is the modulus of elasticity of the material; μ is the Poisson's ratio of the material.

Fig. 2. ACCURA coordinate measuring machine

III. Analysis of Simulation Results and Experimental Data.

The use of UST at the preparation stage of the substrate surface leads to the work-hardening of the surface due to intense plastic deformation, which, in turn, leads to an increase in the intergranular space in the surface structure of the substrate. This increases the intergranular diffusion of coating atoms with higher energy into the base material. As a result of intensive plastic deformation, the dislocation density inside the the base material grain itself increases and new dislocation structures are generated due to their generation by Frank-Read sources. An increase in the dislocations density leads to an increase in vacancies for the penetration of atoms, which contributes to diffusion (intragranular one). One of the conditions for intense diffusion is the similarity of the atomic radii of the base material and the diffusing element, as well as the similarity of the characteristic crystal lattice types. This similarity, in turn, determines the solubility of the elements in each other at diffusion saturation. Thus, the use of nickel, which has unlimited solubility in iron, during the UST of the substrate at the preparation stage, reduces the required activation energy for the diffusion process, improves the chemical component of the adhesive strength of the coating substrate. In addition, as a result of UV energy absorption by the base atoms, they are excited and, consequently the diffusion of the base atoms into the coating material is observed during spraying. Thus, a mutual diffusion process takes place between the elements of the coating and the substrate at the interface of the coating-substrate. Therefore, UST at the preparation stage of the surface substrate to spraying can be attributed to the activation the of products' surface. Apparently, this is why, at the praparation stage of the base surface before spraying, we observed a higher adhesion value of the specimens, whose surface was subjected to UST. We carried out the assessment of the adhesion strength of the "coating-substrate" of specimens subjected to and not subjected to UST. The scatter of adhesion strength values of specimens subjected to UST is ± 5 %. The scatter of the values of adhesive strength of samples without UST is ± 20 %. Fig. 3 shows a dependency graph of adhesion versus coating thickness for samples with UST before spraying and samples without UST before spraying SME of coatings where nickel was used as an adhesive layer.

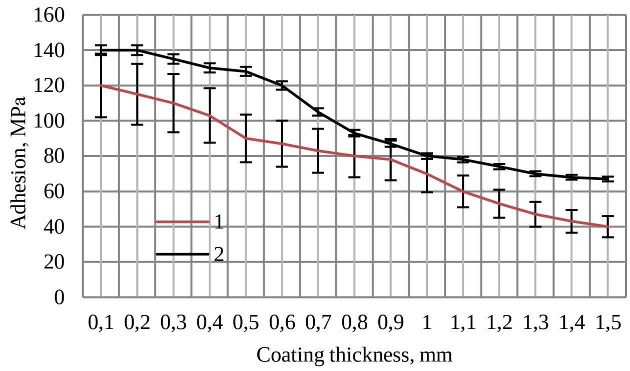
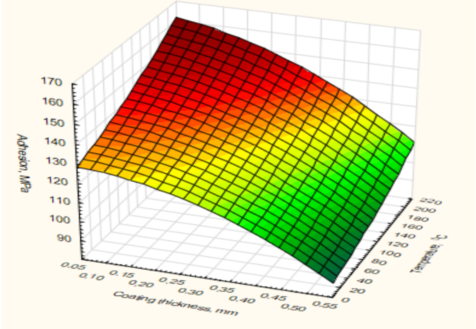
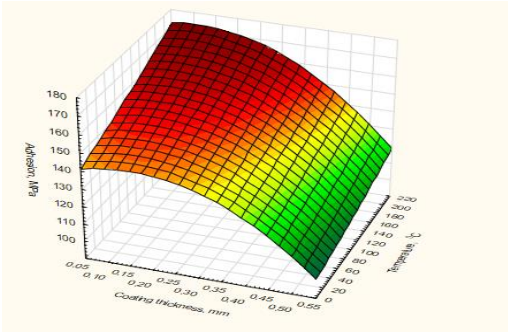


Fig. 3. The dependence of adhesion on the coating thickness (SME material Ni): 1- for samples without UST before spraying; 2- for samples with RCD before spraying

The use of UST after the deposition of the coating at elevated temperature allows activation of the diffusion process. At the same time, it is recommended to carry out SPD immediately before UST, since deformation allows reducing the porosity both in the thickness of the coating and at the interface of the coating-substrate. SPD helps to increase the adhesion density of the coating to the substrate. It is necessary in order to start the process of thermal diffusion mass transfer, and it also affects its intensity. The best result was obtained with two-stage use of UST: at the preparation stage of the substrate surface before spraying and at the finishing stage after spraying at elevated temperature. This temperature depends on the chemical composition of the sprayed material and the temperature of its phase transformations. It must be noted that SPD was carried out directly before the UST. Fig. 4 shows the dependences of adhesion strength on the coating thickness and the on heating temperature at various stages of the formation of coatings made of SME materials.



a



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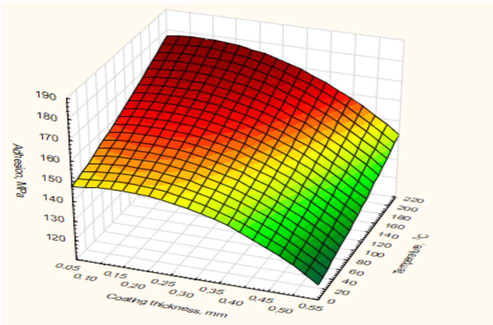

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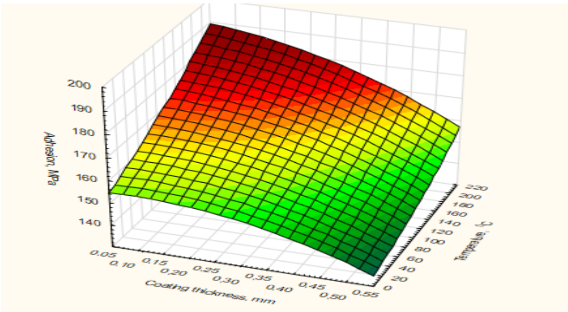
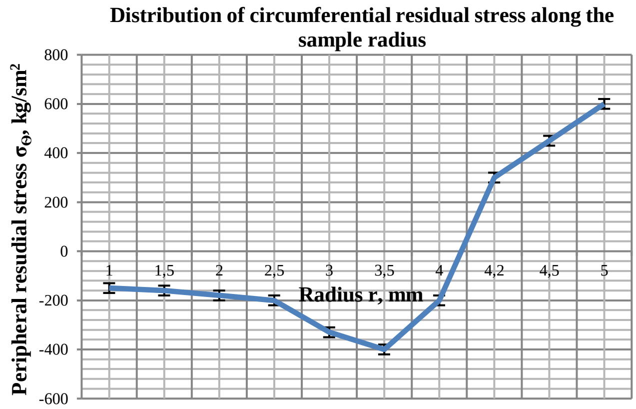
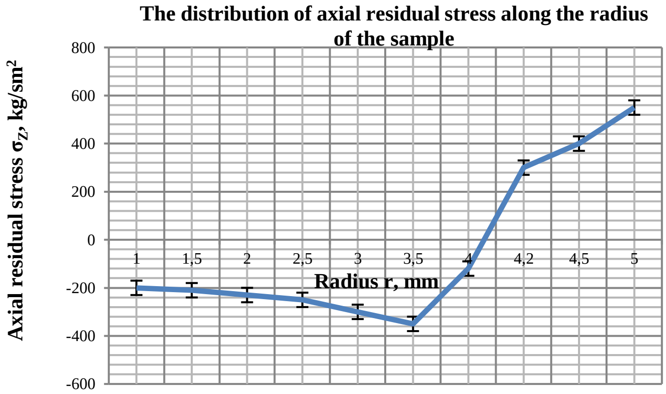


d

Fig. 4. Dependence of adhesion on the coating thickness at various stages of forming the surface composition “Ni-SME material”: a – without UST before spraying; b – UST before spraying; c – UST before spraying of Ni-TiNiCuHf coating and at the stage of finishing treatment; d – UST before spraying and at the stage of finishing operation, Ni-TiNiZr coating

Analysis of the results shown in Fig. 4 shows that when the coating thickness is more than 0.5 mm, the adhesion of the coating to the substrate for samples with UST after spraying coincides with the values of the adhesion of the coating to substrate for samples obtained without a UST after deposition. Thus, surface treatment of the coated part after spraying using UST and temperature effects are applicable for coatings whose thickness does not exceed 0.5 mm. Maenwhile in addition to increasing adhesion, there is an increase in coating density, a decrease in porosity and, as a result, increase of cohesive strength, which is an important factor.





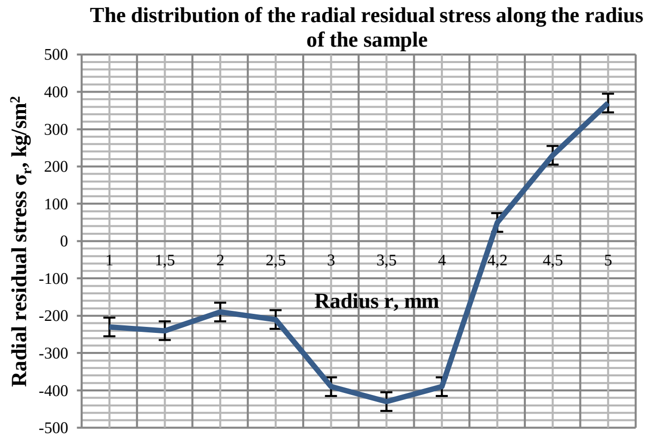
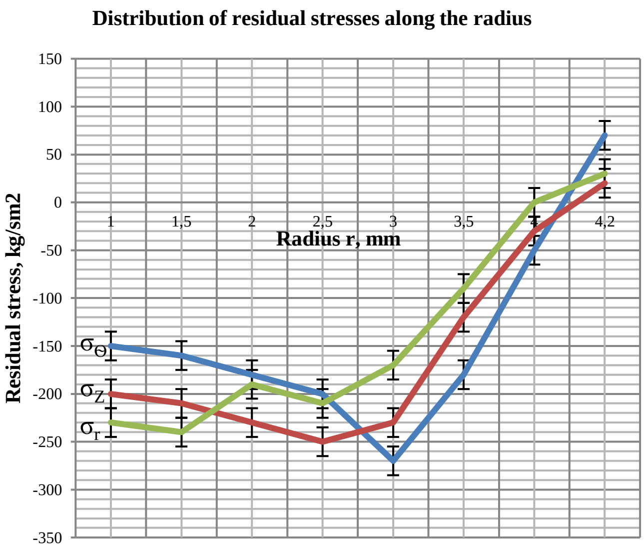


Fig. 5. Residual stress distribution over sample radius.

Adhesion and cohesive strength during the formation of HVOF coatings is determined by the temperature of the particles at the moment of contact with the substrate and the kinetic energy depending on the speed of movement of the particles, which are determined by the parameters of the technological process. The main parameters of the HVOF technological process include: distance and spraying angle, oxygen consumption, propane, flow rate and velocity of carrier gas, argon pressure in the chamber, burner speed, part rotation speed, powder utilization rate. For HVOF of multicomponent powder compositions of SME materials, the optimized parameters of the technological process were: propane consumption 60–85 l/min, oxygen – 120–160 l/min; flow rate of carrier gas (argon) – 40–50 l/min; spraying distance D – 100–250 mm; spraying angle – 70–90°; burner speed – 1–1.5 m/min; rotational speed of the coated part – 800–1000 min^{-1}; the residual pressure of argon in the chamber – 0.7–0.8 Pa, powder utilization rate E – 30–70 % [5].



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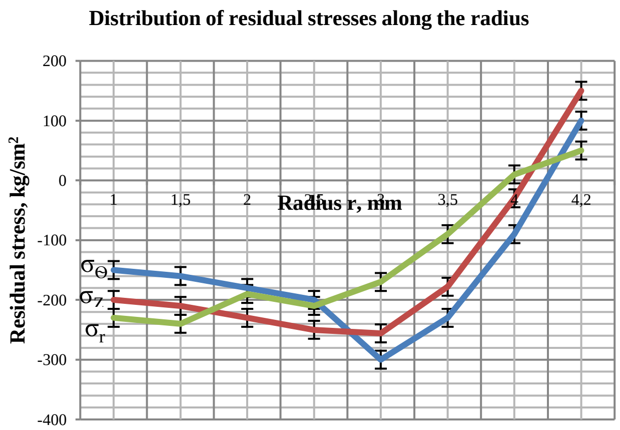

b

Fig. 6. Distribution of residual stresses along the sample radius (σ0 is the circumferential residual stress; σZ is the axial residual stress; σr is the radial residual stress) for the coating: a – Ni-TiNiZr; b – Ni-TiNiCuHf

As a result of spraying, the surface is heated, both due to the impact of the jet and due to the fact that the penetrating powder particles have a temperature of (0.9–1) Tpl. The main heat removal occurs in the direction of the part, so the temperature over the section of the part varies depending on the distance from the surface. All this leads to different rates of cooling of the part and the formation of non-equilibrium structures over the cross section of the coated part. This causes residual stresses, residual stresses cause cracking, delamination of the coating and changes in the shape of the products and distortion. Fig. 5 shows the distribution of residual stresses along the radius from the center of the sample.

As we can see from Fig. 5, at the interface between the substrate and the coating, there is a jump in the residual stresses and a change in the stresses sign from compressive to tensile one. This may be the reason for the delamination of the coatings. It is especially noticeable when applying a coating on thin-walled structures, which is accompanied by its warping, sometimes cracking and peeling of the coating.

Fig. 6 shows distribution of residual stresses on coated samples after performing an UST as a finishing operation at a temperature of 200 °C.

As can be seen from Fig. 6 after the UST of the sample with a coating at a temperature of 200 °C, the residual stresses decrease over the entire cross section of the coating. At the same time, there is a noticeable decrease in residual stresses in the base material directly along the border (border layer) and, as a result, the magnitude of the residual stress gradient at the coating – base interface decreases.

IV. Conclusion

The study results of the ultrasonic processing effect on the adhesion of a surface composition made of SME materials at various stages of its formation suggest that the use of UST at the preparation stage causes an increase in the dislocation density and the area of the grain boundary on the surface and that it is one of the mechanical activation types. The result of such exposure at the preparation stage of the base is an increase of the adhesion chemical component, as the energy required for the diffusion process decreases. The use of UST with temperature effects at the stage of finishing of the coated parts also allows providing the process of thermal diffusion mass transfer. This increases adhesion due to the chemical component of adhesion. As a result of this treatment, it was possible to reduce the magnitude of the jump in residual stresses at the boundary of the substrate-coating. This reduces the risk of coating delamination and allows obtaining a stable result without a large scatter of the values, which is characteristic for the coating deposition without a UST.

Acknowledgment

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