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PLASMA COATINGS BASED ON SELF-FLUXING NiCrBSi ALLOY WITH IMPROVED WEAR RESISTANCE PROPERTIES

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The structure and properties of plasma coatings sprayed with a composite material based on a self-fluxing NiCrBSi alloy (PG-10N-01 alloy) modified with a composite material obtained by self-propagating high-temperature synthesis were studied. Titanium powders, carbon black, aluminum, iron oxide, PT-NA-01 thermosetting powder and PGOSA-0 refractory clay were used as the initial components of modified with a composite material. Mixing and mechanical activation of the initial powders was carried out in a BM-1 ball mill for 15 minutes at 130 rpm in a ratio of 1 to 40 of the mass of the charge to the mass of the falling bodies (steel balls with a diameter of 6 mm). Initiation of the self-propagating high-temperature synthesis was carried out using a special device by introducing a heated nichrome spiral. The process of coatings spraying was performed on the MPN-004 microplasma spraying unit at a current of 45 A, a voltage of 30 V with a distance of 100 mm on samples made of 65G steel with a thickness of 3 mm. Argon was used as a plasma-forming and shielding gas. In order to substantiate the feasibility of the self-propagating high-temperature synthesis, a part of the samples was sprayed with a self-fluxing alloy PG-10N-01 with the addition of a mechanical mixture of starting powders. It was established that as a result of plasma spraying of the PG-10N-01 alloy and the composite material of the modified with a composite material + PG-10N-01 composition, coatings with a dense and multiphase structure are formed. The microstructure of the PG-10N-01 alloy coating consists of a solid solution based on nickel (γ-Ni) with inclusions of nickel borides Ni_3B and chromium carbides Cr_3C_2. When adding modified with a composite material in a nickel-based solid solution, in addition to the phases indicated above, borides of titanium TiB_2, carbides of titanium TiC and silicon SiC were detected. Their presence leads to an increase in the microhardness of such coatings and their greater wear resistance under conditions of abrasive wear in comparison with the spraying coating of the PG-10H-01 alloy.

***Keywords**: self-propagating high-temperature synthesis (SHS process), composite material, spraying, plasma coating, structure, phase composition, microhardness, wear resistance.*

Relevance of the research topic

The plasma method of coatings spraying consists in the formation of the coating part of material particles heated and accelerated by a high-temperature plasma jet on the surface. When the particles collide with the surface of the base or sprayed material, their connection occurs. This method is recommended for spraying strengthening, protective or other types of coatings made of metal powders, carbides, oxides, nitrides, refractory compounds, as well as composite materials, the individual phases of which perform specified special functions [1]. Composite materials combine the properties of each original component, as their properties are formed due to either the addition of components (additivity) or their joint strengthening (synergism) [2].

One of the methods of obtaining composite materials is the self-propagating high-temperature synthesis (SHS process), the essence of which is the local initiation of exothermic reactions between the starting reagents. This makes it possible to generate a significant amount of heat for the propagation of the front of physicochemical transformations capable of forming materials of the predicted phase composition [3–6].

Analysis of recent studies and papers

The physical and mechanical properties of the Ti-Fe-C system composite, obtained using the SHS process, are considered in [7]. PTS-1 titanium Ti powder, carbon C, and Fe iron powder (SHKH15 steel) were used as starting materials. The latter was added from slurry grinding using the technology of the Lutsk National Technical University [8]. Cylindrical samples with a diameter of 30 mm and a height of 60 mm were pressed from the obtained powders. The SHS process was carried out in a laboratory reactor manufactured at the university.

As a result of chemical reactions, a synthesized sample (spike) was obtained. The Rockwell hardness of the synthesized spike was: 63 HRC at the center of the spike, 38 HRC at a radius of 0.5 mm, and 18 HRC at a radius of 0.9 mm. In addition, the authors determined the ultimate strength and ultimate deformation under static loading. It was established that the maximum stress at the moment of the spike failure is σ_{max}=106 MPa, and the relative deformation is e_{max}=0.038. Based on the research results, the authors recommend using the obtained composite as a construction material.

The method of obtaining carbon nanomaterials using the SHS process is considered in [9]. In its implementation, the initiation of the SHS process is carried out by pulsed heating of the local zone of the powder mixture with an electric discharge applied to a wire made of refractory metal, which is in surface contact with the mixture. The use of this method allows to increase the amount of powders of carbon nanomaterials and reduce the probability of an undesirable "explosive combustion" mode of the mixture, while maintaining the simplicity and stability of the initiation.

Refractory powders of zirconium diboride ZrB_2 were also obtained using the SHS process [10]. Enriched borate ore (boron oxide content up to 40%), zirconium silicate $ZrSiO_4$, zirconium oxide ZrO_2, aluminum powder Al and magnesium Mg (to increase the combustion temperature), as well as hydrochloric acid HCl (its concentration is 37.5%) were used as initial components. Mixing and mechanical activation of the initial components of the charge was carried out in a "Pulverisette 5" planetary mill (Fritsch, Germany). The preparation of the charge was carried out taking into account the stoichiometric ratios of the initial components. A glass-carbon crucible, into which the charge was poured, was used to carry out the SHS process. Initiation of the SHS process was carried out in a high-pressure reactor in an argon medium Ar under a pressure of 10 atm by introducing a red-hot tungsten spiral. The obtained refractory submicron nano powders of zirconium diboride ZrB_2 (94.7%) and zirconium boride ZrB (3.3%) can be used as abrasive powders, ceramic and composite materials in the technologies of various coatings application.

The results of research on the production of chromium borides using the SHS process are shown in the paper [11]. As initial components of the charge, the authors used boron powder H_3BO_3 with a purity of 99.9%, chromium oxide Cr_2O_3 and aluminum Al. Mixing and mechanical activation of the charge was carried out in the "Pulverisette 5" planetary mill, after which cylindrical samples with a diameter of 20 mm and a height of 20 mm were pressed from the resulting mixture. In contrast to previous studies, the authors of this paper conducted the SHS process without a protective environment.

Using the X-ray phase analysis method, it was established that secondary reaction products are contained in the combustion products of the charge, namely chromium borides of the types CrB, CrB_2, Cr_2B, Cr_5B_3, CrB_6 and Cr_3B_4. Crystalline products, which are a mixture of chromium boride CrB_2 and aluminum oxide Al_2O_3, were found in the sample. These materials are recognized as promising from the point of view of industrial use and development of composites with a metal matrix.

Comprehensive scientific research on the development of composite powders based on the FeMoNiCrB amorphizing alloy with the addition of refractory compounds for gas-thermal coating technologies is given in [12]. Chemical compounds ZrB_2, (Ti,Cr)C and $FeTiO_3$ were used as refractory ones. The compounds ZrB_2 and $FeTiO_3$ were obtained by the method of mechanical doping, and the compound (Ti,Cr)C - by the SHS method.

Paper [13], which analyzed the structure and amorphizing properties of composite detonation coatings based on FeMoNiCrB+ZrB_2, FeMoNiCrB+(Ti,Cr)C, FeMoNiCrB+$FeTiO_3$ alloys, became a continuation of these studies. The coatings applied by detonation spraying had a dense lamellar multiphase structure. The microhardness of the coatings was: FeMoNiCrB – 4855±1023 MPa, FeMoNiCrB+ZrB_2 – 3830±570 MPa, FeMoNiCrB+(Ti,Cr)C – 4450±700 MPa, FeMoNiCrB+$FeTiO_3$ – 3750±620 MPa. Electrochemical tests of the coatings showed that their corrosion resistance depends on the pH solution. The resulting coatings with a thickness of 500 μm were tested in acidic solutions (3% NaCl and 5% NaOH). The wear resistance of FeMoNiCrB+ZrB_2, FeMoNiCrB+(Ti,Cr)C, FeMoNiCrB+$FeTiO_3$ detonation coatings under conditions of wear on unattached abrasive particles of SiO_2 in B_4C is by 2.6–3.3 times greater than that of 30KhGSA steel in SiO_2 environment and by 1.9–2.9 times in the B_4C environment. The highest wear resistance was achieved on the coating type FeMoNiCrB+(Ti,Cr)C with a microhardness of 4450±700 MPa.

Composite materials obtained on the basis of self-fluxing alloys of the NiCrBSi system (PG-10N-01, PG-10N-04, PG-12N-02, etc. powders) are used for various coating technologies [14–16]. The main coating

phases of such alloys are solid solution based on nickel (γ-Ni) and eutectic, which, as a rule, consists of γ-Ni and nickel boride Ni_3B. The addition of modifying and strengthening additives obtained by SHS to the NiCrBSi alloy makes it possible to control the phase composition of the composite material and obtain new compounds (phases) in the nickel matrix, which provide a higher level of physical and mechanical properties of coatings on their base.

One of the further directions of obtaining materials using the SHS process, which can be used as modifiers, is the expansion of the nomenclature of initial reagents, namely the addition of various types of mineral raw materials (clay, sandy materials), solid industrial and radioactive waste (smelting slag, metal shavings) [6]. Refractory clay, the basis of which is known to consist of oxides of silicon SiO_2 and aluminum Al_2O_3, was chosen as one of the starting materials [17].

The aim of the paper: research on the structure and properties of plasma coatings based on a self-fluxing alloy of the NiCrBSi system, modified with a composite material obtained using the SHS process.

Research materials and methodology

For spraying, a composite material obtained on the basis of a self-fluxing alloy of the NiCrBSi system (PG-10N-01 alloy) modified with a composite material (MCM) obtained by self-propagating high-temperature synthesis was used as the initial material. The composite material was obtained in two stages. At the first stage, powders of titanium Ti of PTM-1 grade, technical carbon C of the P-803 brand and refractory clay grade PGOSA-0 were used to obtain MCM. In order to enhance the thermal effect of the reaction, aluminum Al in the form of PAP-1 powder, iron oxide Fe_2O_3 and thermo-reactive powder PT-NA-01 were added to the initial charge. The particle size of the initial powders did not exceed 100 μm. The ratio of powders was equimolar, so that during the further passage of the SHS process, the synthesis of carbides of titanium TiC and silicon SiC of stoichiometric composition took place.

Mixing and mechanical activation of the charge was carried out in a BM-1 ball mill for 15 min at 130 rpm and a ratio of 1:40 of the mass of the charge to the mass of the falling bodies (steel balls with a diameter of 6 mm). After mechanical activation, the maximum particle size of the charge did not exceed 40 μm [17]. 10% of "Metylan" glue was added to the processed charge, after which a cylindrical sample with a diameter of 16 mm and a height of 20 mm was pressed. The sample was dried for 72 hours. Initiation of the SHS process of the sample was carried out with a heated nichrome spiral with a diameter of 0.8 mm (Fig. 1) using a specially developed device [18]. The SHS process was carried out in an argon medium with pure Ar 98%.

At the second stage, the MCM obtained in the form of a spike was crushed to a powder-like state, after which an amount of 10% to 30% of MCM was added to the matrix material – self-fluxing PG-10N-01 alloy and mechanical activation was carried out for 15 minutes. Some of the samples were sprayed with the PG-10N-01 alloy with the addition of a mechanical mixture (MM) of the initial components of the charge Ti-C-Al-SiO_2-Al_2O_3-Fe_2O_3-PT-NA-01. The amount of MM, as well as the amount of MCM, was from 10% to 30%, respectively. The choice of such concentrations was made based on recommendations for the production of wear-resistant composite materials [12] and the results of previously performed research.

a b c

Fig. 1. The spike obtaining using the SHS process:
a – preliminary heating of the sample; b – SHS process; c – obtained spike

The application of plasma coatings was carried out using the MPN-004 microplasma spraying unit at the E. O. Paton Electric Welding Institute of the National Academy of Sciences of Ukraine. The design of the MP-04 microplasmatron has an external anode, which provides the possibility of introducing the sprayed material into the high-temperature region of the microplasma jet, which is the arc gap in front of the anode spot [19–21]. Spraying process parameters are: electric current I=45 A, voltage U=30 V, consumption of plasma-forming gas Q_{pl}=70 l/h, consumption of protective gas Q_{pg}=280 l/h, spraying distance L=100 mm. Spraying was carried out on samples made of 65G steel with a thickness of 3 mm. Previously, the samples were subjected to jet-abrasive treatment and ultrasonic cleaning was carried out on the surface with isopropyl alcohol for 15 minutes.

The microstructure and phase composition of the coatings were studied using a Neophot-32 optical microscope and a Tescan Mira 3LMU scanning electron microscope with an Oxford X-max energy dispersive spectrometer. A PMT-3 microhardness tester with a load of 0.1 kg was used to measure the microhardness of sprayed coatings.

The Kh4-B test machine was used to study the wear resistance of the coatings. The relative wear resistance of sprayed coatings containing MCM and MM was compared with the amount of wear of the coating sprayed with matrix material PG-10N-01. The amount of wear of the sprayed coating was determined by the weight method using VLR-200 analytical scales.

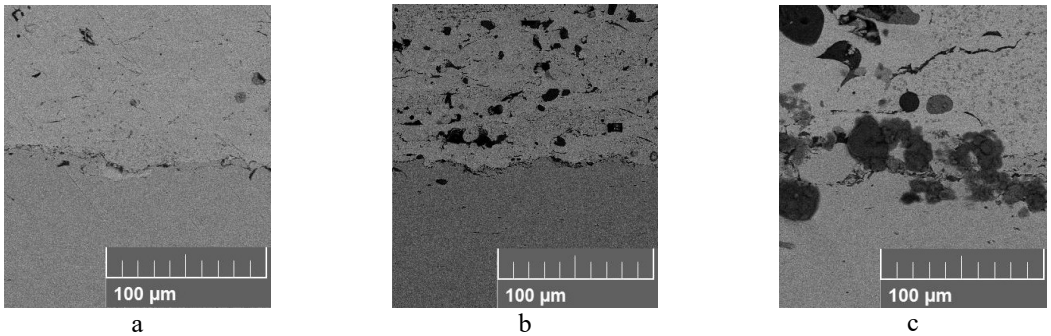
The main research material

When spraying both the PG-10N-01 alloy and the composite material with a composition of 10% MCM + 90% PG-10N-01, dense, uniform in thickness coatings are formed (Fig. 2). The microstructure of the coating 10% MCM + 90% PG-10N-01 consists of the matrix material of the self-fluxing alloy PG-10N-01, in which inclusions of different sizes are located (Fig. 2, b), and with an increase in the content of MCM in the composite material, their number increases [22]. Coating with a composition of 10% MC + 90% PG-10N-01 is the matrix material of the PG-10N-01 alloy, which contains inclusions of different sizes and pores (Fig. 2, c).

Using scanning electron microscopy with energy dispersive analysis, it was established that the basis of the sprayed coating PG-10N-01 is solid solution of nickel (γ-Ni) with inclusions of nickel borides Ni_3B and chromium carbides Cr_3C_2 (Fig. 3, a). Coatings sprayed with a composite material with a composition of 10% MCM + 90% PG-10N-01, in addition to the solid solution based on nickel γ-Ni and the inclusions indicated above, contain the phases of titanium carbides TiC and silicon SiC, as well as titanium diborides TiB_2 (Fig. 3, b). Microstructure of 10% MM + 90% PG-10N-01 coating consists of a solid solution based on γ-Ni nickel and Ni_3B–Cr_3C_2 inclusions, however, unlike the previous sample, it contains phases of silicon oxides SiO_2, titanium oxides TiO_2 and TiB_2 diborides (Fig. 3, c).

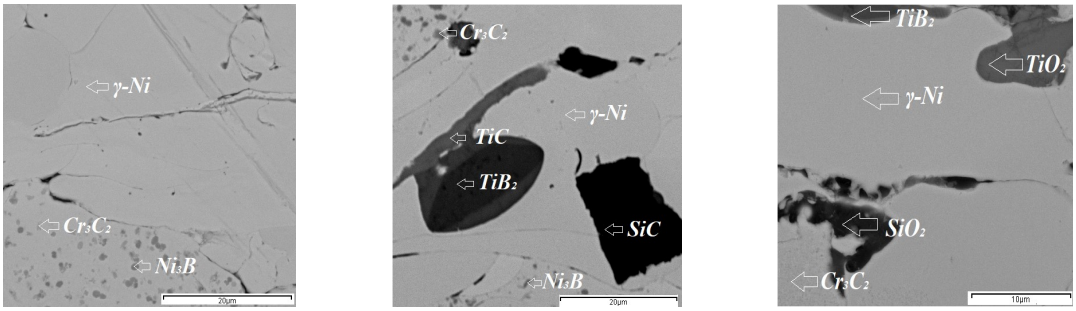
The distribution of elements in the sprayed coating was determined by the method of scanning electron microscopy with energy dispersive analysis along the scanning line (Fig. 4).

The microhardness of coatings, measured in the direction from the surface of the sprayed coating to the surface of the base, is shown in Fig. 5.



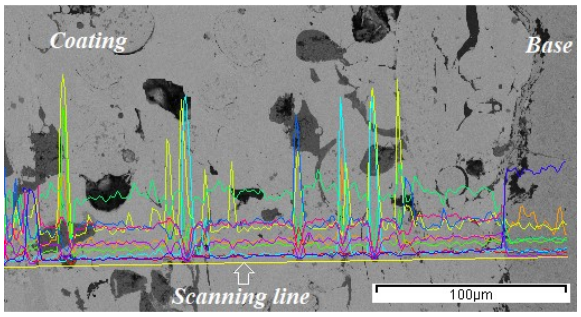
a b c

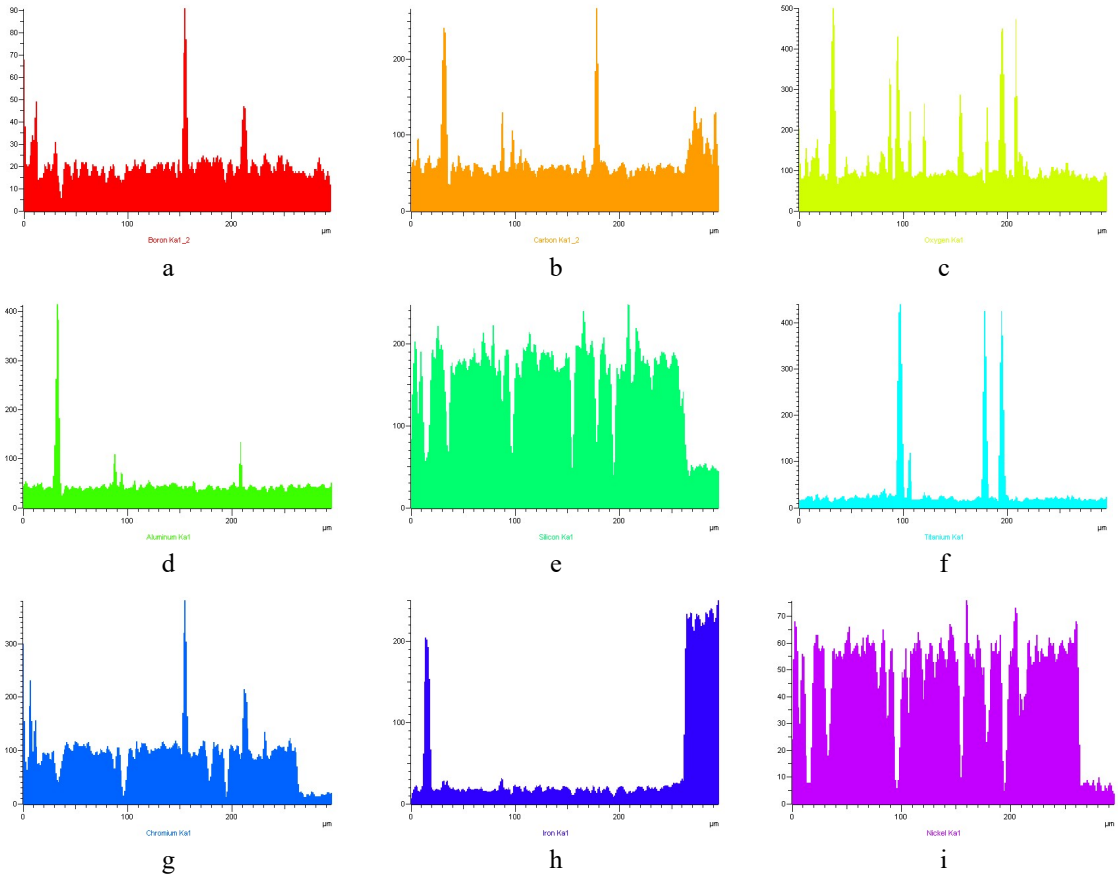
Fig. 2. Microstructure of sprayed coatings:
a – PG-10N-01; b – 10% MCM + 90% PG-10N-01; c – 10% MM + 90% PG-10N-01



a b c

Fig. 3. Microstructure and phase composition of sprayed coatings:
a – PG-10N-01; b – 10% MCM + 90% PG-10N-01; c – 10% MM + 90% PG-10N-01





a b c

d e f

g h i

Fig. 4. Distribution of elements of plasma coating with a composition of 10% MCM + 90% PG-10N-01:
a – B; b – C; c – O; d – Al; e – Si; f – Ti; g – Cr; h – Fe; i – Ni

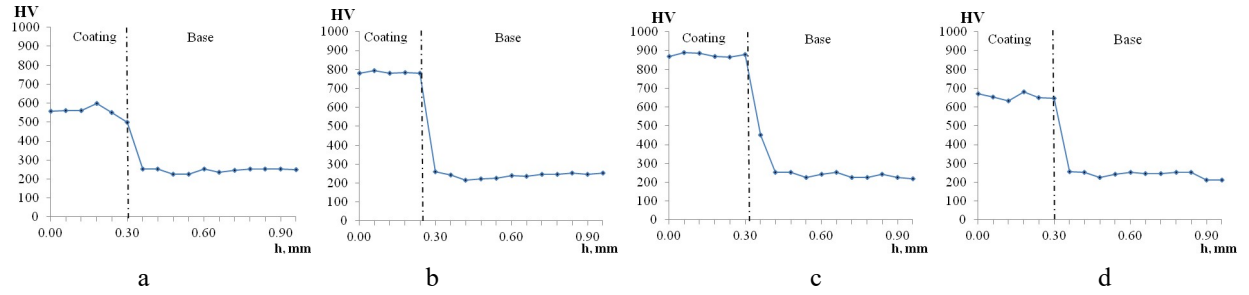


Fig. 5. Change in microhardness (HV) of sprayed coatings in the direction from the surface of the coating to the surface of the base:
a – PG-10N-01; b – 10% MCM + 90% PG-10N-01; c– 30% MCM + 70% PG-10N-01; d – 10% MM + 90% PG-10N-01

When measuring the microhardness, it was found that the average microhardness of the sprayed coating of the composite material with a composition of 10% MCM + 90% PG-10N-01 is 780 HV, coating with 20% MCM + 80% PG-10N-01 is 835 HV and coating with 30% MCM + 70% PG-10N-01 is 880 HV, which exceeds the average microhardness of the sprayed coating of the PG-10N-01, which is equal to 555 HV. The stable nature of the microhardness distribution in the sprayed coatings of the composite material indicates the uniform distribution of TiC and SiC carbides and TiB_2 diborides in the matrix material (Fig. 5, b and c). The average microhardness of the sprayed coating with a composition of 10% MM + 90% PG-10N-01 is 650 HV (Fig. 5, d), a coating with 20% MM + 80% PG-10N-01 is 680 HV, and a coating with 30% MM + 70% PG-10N-01 – 720 HV. Such coatings have a less uniform distribution of microhardness over the coating thickness and have lower microhardness compared to coatings containing MCM (Fig. 6).

The results of studies of wear resistance of sprayed coatings are shown in Fig. 7.

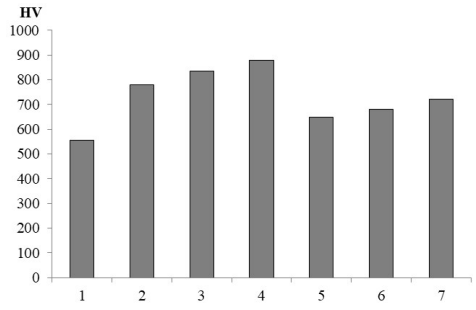


Fig. 6. Average microhardness (HV) of deposited coatings (measured in the direction from the surface of the deposited coating to the surface of the base):
1 – PG-10N-01;
2 – 10% MCM + 90% PG-10N-01;
3 – 20% MCM + 80% PG-10N-01;
4 – 30% MCM + 70% PG-10N-01;
5 – 10% MM + 90% PG-10N-01;
6 – 20% MM + 80% PG-10N-01;
7 – 30% MM+ 70% PG-10N-01

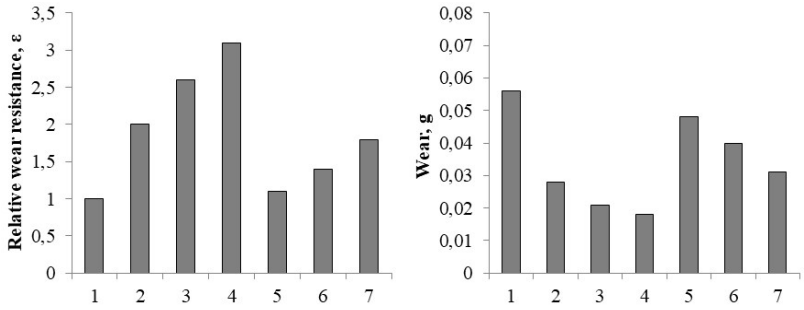
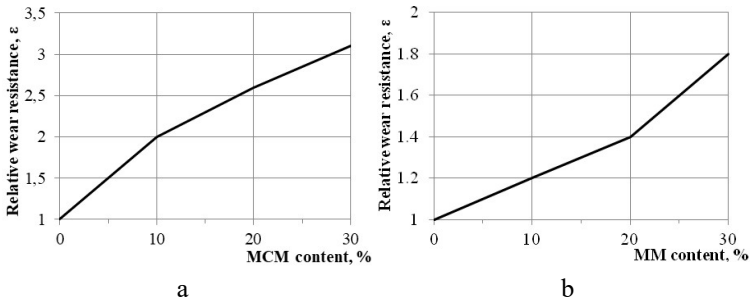


Fig. 7. Wear resistance of sprayed coatings in the process of abrasive wear:
a – relative wear resistance; b – wear of sprayed coatings;
1 – PG-10N-01;
2 – 10% MCM + 90% PG-10N-01;
3 – 20% MCM + 80% PG-10N-01;
4 – 30% MCM + 70% PG-10N-01;
5 – 10% MM + 90% PG-10N-01;
6 – 20% MM + 80% PG-10N-01;
7 – 30% MM+ 70% PG-10N-01

When analyzing the results of measurements of the coatings wear resistance, it was established that the sprayed coatings of the composite material have a higher abrasive wear resistance compared to the coatings of the self-fluxing alloy PG-10N-01. This is due to the fact that in the structure of the sprayed coating, along with the γ-Ni solid solution and Ni_3B–Cr_3C_2 inclusions, there are particles of TiC titanium carbides, SiC silicon carbides, and also TiB_2 titanium diborides. When the MCM content in the composite material increases, the amount

of these carbides and borides increases. Accordingly, the wear resistance of the coating in the process of abrasive wear increases (Fig. 8, a). When adding a mechanical mixture of the initial components of the charge in the amount of 10% to the PG-10N-01 alloy, the wear resistance of the sprayed coating is 1.2 times higher, the coating with 20% MM + 80% PG-10N-01 is 1.4 times higher, and the coating with 30% MM + 70% PG-10N-01 is 1.8 times higher compared to the wear resistance of the coating of the PG-10N-01 alloy (Fig. 8, b). This is explained by the fact that the structure of the matrix material contains phases of diborides TiB_2 and titanium oxides TiO_2, as well as silicon oxides SiO_2, which strengthen it.

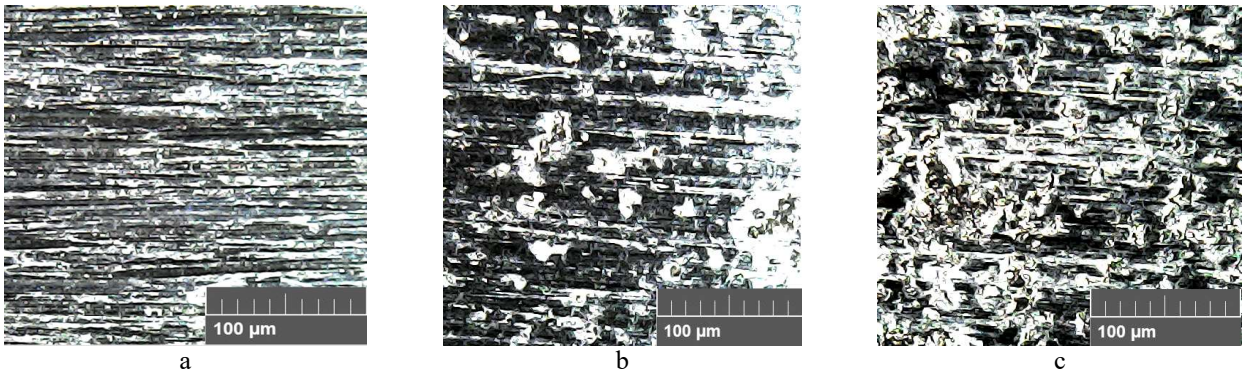


a b

Fig. 8. Relative wear resistance of sprayed coatings depending on the content of the additive to the PG-10N-01 alloy:
a – MCM; b – MM

The morphology of the friction surfaces is shown in Fig. 9. The friction surface sprayed with the PG-10N-01 alloy has lines up to 14 μm deep (Fig. 9, a), the friction surface of the composite material with 10% MCM + 90% PG-10N-01 has lines up to 7 μm deep (Fig. 9, b). When the MCM content increases in the composite material, the depth of the lines decreases [22]. The friction surface sprayed with a material with a composition of 10% MM + 90% PG-10N-01 has lines up to 12 μm deep.

The analysis of the obtained results shows that the coating based on the self-fluxing alloy PG-10N-01, modified with a composite material obtained by self-propagating high-temperature synthesis, has a structure of a solid solution based on nickel (γ-Ni) and Ni_3B-Cr_3C_2 inclusions, as well as strengthening phases of titanium carbides TiC, silicon SiC and diborides TiB_2. Introduced TiC, SiC, TiB_2 strengthen the nickel-based solid solution, which increases the microhardness and wear resistance of the coating.



a b c

Fig. 9. Morphology of friction surfaces:
a – PG-10N-01; b – 10% MCM + 90% PG-10N-01; c – 10% MM + 90% PG-10N-01

Conclusions

A composite material was developed based on the self-fluxing alloy PG-10N-01, modified by the composite material obtained by self-propagating high-temperature synthesis. Powders of titanium, carbon black, iron oxide, aluminum, thermosetting powder PT-NA-01, and non-refractory clay PGOSA-0 were used to obtain MCM using the SHS process.

Using optical and electron microscopy methods, it was established that the structure of sprayed coatings with a composite material consists of a solid solution based on nickel (γ-Ni) with inclusions of Ni_3B-Cr_3C_2 and contains solid strengthening phases in the form of carbides of titanium TiC and silicon SiC, as well as diborides of titanium TiB_2. When the MCM content in the composite material increases, the amount of TiC, SiC, and TiB_2 increases, which increases the microhardness of the coating and increases its wear resistance.

The microhardness of the sprayed coating of the composite material with a composition of 10% MCM + 90% PG-10N-01 is 780 HV, with 20% MCM + 80% PG-10N-01 it is 835 HV, with 30% MCM + 70% PG-10N-01 it is 880 HV, which exceeds the microhardness of the coating of self-fluxing alloy PG-10N-01, which is equal to 555 HV.

In the process of wear on fixed abrasive particles, the wear resistance of the coating with 10% MCM + 90% PG-10N-01 is 2 times higher, with 20% MCM + 80% PG-10N-01 it is 2.6 times higher, and with 30% MCM + 70% PG-10N-01 it is 3.1 times higher compared to the wear resistance of the PG-10N-01 alloy coating.

The microstructure of coatings sprayed with the addition of a mechanical mixture of the initial components of the charge consists of a solid solution based on nickel (γ-Ni) with Ni_3B-Cr_3C_2 inclusions, as well as silicon oxides SiO_2, TiO_2 oxides and titanium diborides TiB_2. The average microhardness of the sprayed coating with a composition of 10% MM + 90% PG-10N-01 is equal to 650 HV, coating with 20% MM + 80% PG-10N-01 – 680 HV and coating with 30% MM + 70% PG-10N-01 – 720 HV. Wear resistance of coating with 10% MM + 90% PG-10N-01 is 1.2 times higher, for coating with 20% MM + 80% PG-10N-01 it is 1.4 times higher, for coating with 30% MM + 70% PG-10N-01 – 1.8 times higher compared to the wear resistance of the PG-10N-01 coating.

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Плазмові покриття на основі самофлюсівного сплаву NiCrBSi з покращеними зносостійкими властивостями

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Досліджено структуру й властивості плазмових покриттів, напилених композиційним матеріалом на основі самофлюсівного сплаву NiCrBSi (сплаву марки ПГ-10Н-01), модифікованого композиційним матеріалом, одержаним самопоширюваним високотемпературним синтезом. Як вихідні компоненти модифікуючого композиційного матеріалу використані порошки титану, технічного вуглецю, алюмінію, оксиду заліза, термореагуючого порошку марки ПТ-НА-01 і вогнетривкої глини марки ПГОСА-0. Змішування й механічну активацію вихідних порошків проведено у кульовому млині КМ-1 протягом 15 хв при 130 об/хв у співвідношенні 1 до 40 маси шихти до

маси падаючих тіл (сталевих куль діаметром 6 мм). Ініціювання самопоширюваного високотемпературного синтезу здійснено з використанням спеціального пристрою шляхом підведення розжареної ніхромової спіралі. Процес напилення покриттів виконано на установці мікроплазмового напилення МПН-004 при струмі 45 А, напрузі 30 В з дистанцією 100 мм на зразки зі сталі 65Г товщиною 3 мм. Як плазмоутворюючий та захисний газ використано аргон. Для обґрунтування доцільності проведення самопоширюваного високотемпературного синтезу частину зразків напилено самофлюсівним сплавом ПГ-10Н-01 з додаванням механічної суміші вихідних порошків. Встановлено, що в результаті плазмового напилення сплаву ПГ-10Н-01 та композиційного матеріалу складу модифікуючий композиційний матеріал + ПГ-10Н-01 формуються покриття зі щільною й багатофазною структурою. Мікроструктура покриття сплаву ПГ-10Н-01 складається з твердого розчину на основі нікелю (γ-Ni) з включеннями боридів нікелю Ni_3B та карбідів хрому Cr_3C_2. При додаванні модифікуючого композиційного матеріалу у твердому розчині на основі нікелю, крім вказаних вище фаз, виявлені бориди титану TiB_2, карбіди титану TiC і кремнію SiC, наявність яких призводить до підвищення мікротвердості таких покриттів та їх більшої зносостійкості в умовах абразивного зношування у порівнянні з напиленим покриттям сплаву ПГ-10Н-01.

***Ключові слова**: самопоширюваний високотемпературний синтез (СВС-процес), композиційний матеріал, напилення, плазмове покриття, структура, фазовий склад, мікротвердість, зносостійкість.*

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