PERIÓDICO TCHÊ QUÍMICA
ARTIGO ORIGINAL

ESTADO NO DOMÍNIO DOS REVESTIMENTOS RESISTENTES AO CALOR PARA LIGAS E AÇOS DE NÍQUEL À PROVA DE CALOR

STATE IN THE FIELD OF HEAT-RESISTANT COATINGS FOR HEAT-PROOF NICKEL ALLOYS AND STEELS

СОСТОЯНИЕ ВОПРОСА В ОБЛАСТИ ЖАРОСТОЙКИХ ПОКРЫТИЙ ДЛЯ ЖАРОПРОЧНЫХ НИКЕЛЕВЫХ СПЛАВОВ И СТАЛЕЙ

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RESUMO

Atualmente, em muitos setores como, por exemplo, aviação, espacial (foguetes) e engenharia é dada atenção especial ao aumento das características táticas e técnicas dos produtos que estão sendo criados, aumentando sua confiabilidade e economia. A solução de problemas requer o uso de materiais que possam trabalhar em ambientes agressivos sob cargas pesadas, altas temperaturas, pressões e vibrações, o que determina a relevância do problema descrito no artigo. O objetivo do artigo foi realizar uma análise crítica de revestimentos desenvolvidos na Rússia com diversas características de composição, propriedades tecnológicas e operacionais, do ponto de vista da possibilidade de usá-los para fornecer um sistema confiável de proteção de material sob condições que causam corrosão por gás de alta temperatura e erosão da camada superficial. No trabalho foram considerados os principais métodos de proteção de materiais como pulverização térmica, pulverização a plasma, feixe de elétrons, pulverização por catodo de arco elétrico a plasma, deposição a vácuo, tratamento químico-térmico, revestimento por difusão à base de alumínio. Entre os métodos mais eficazes, destacaram-se o método de síntese autopropagável a alta temperatura, o método da química do plasma de alta energia e os métodos de deposição física no vácuo. Também foram destacados os tipos de revestimentos (de óxido, metálicos, intermetálicos, de esmalte de vidro, de vidro cristalino e vitrocerâmico) e suas características. Foi dada especial atenção aos revestimentos resistentes ao calor à base de esmaltes refratários uma vez que suas vantagens incluem baixo custo e a possibilidade de aplicá-los diretamente em peças sem *primers*, subpêlos, protetores. Foi identificada uma série de soluções técnicas que protegem as ligas da corrosão e erosão por gases de alta temperatura durante operações de longo prazo em produtos de aeronaves a altas temperaturas. Os resultados do artigo podem ser úteis para pesquisas posteriores, pois os desenvolvimentos individuais não são informativos do ponto de vista da solução de problemas específicos e exigem testes de bancada caros do resultado técnico declarado.

Palavras-chave: *resistência ao calor, revestimento, ligas de níquel, revestimentos vitrocerâmicos, tecnologias de aplicação.*

ABSTRACT

Currently, in many industries, special attention is paid to increasing the reliability and economy of the products. Solving problems requires the use of materials that can work in difficult conditions, which determines the relevance of the problem stated in the article. The purpose of the article was to conduct a critical analysis of coatings developed in Russia that are diverse in composition, technological and operational properties, from the standpoint of the possibility of using them to provide a reliable material protection system under conditions that cause high-temperature gas corrosion and surface layer erosion. The main methods of materials protection were considered in the work: thermal spraying, plasma spraying, electron beam, plasma electric arc cathode spraying, vacuum deposition, chemical-thermal treatment, diffusion coating based on aluminum. Among the more effective methods, the method of self-propagating high-temperature synthesis, the method of high-energy plasma chemistry, the methods of physical deposition in vacuum are noted. The types of coatings were also highlighted and their short characteristics were given. Particular attention was paid to heat-resistant coatings based on refractory enamels since their advantages include low cost and the possibility of applying them directly

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to parts. A number of technical solutions have been identified that protect the alloys from high-temperature gas corrosion and erosion under long-term operation in aircraft products at high temperatures. The results of the article may be useful for further research since individual developments are uninformative from the standpoint of solving specific problems and require expensive bench testing of the claimed technical result.

Keywords: *heat resistance, coating, nickel alloys, glass-ceramic coatings, application technologies.*

АННОТАЦИЯ

В настоящее время во многих отраслях особое внимание уделяется повышению надежности и экономичности продукции. Решение проблем требует использования материалов, способных работать в сложных условиях, что и определяет актуальность проблемы, изложенной в статье. Целью статьи является проведение критического анализа разработанных в России покрытий, отличающихся по составу, технологическим и эксплуатационным свойствам, с точки зрения возможности их использования для обеспечения надежной системы защиты материалов в условиях, вызывающих высокую температуру. газовая коррозия и эрозия поверхностного слоя. В работе рассмотрены основные методы защиты материалов: термическое напыление, плазменное напыление, электронный луч, плазменное электродуговое катодное напыление, вакуумное напыление, химико-термическая обработка, диффузионное покрытие на основе алюминия. Среди наиболее эффективных методов отмечены метод самораспространяющегося высокотемпературного синтеза, метод химии плазмы высоких энергий, методы физического осаждения в вакууме. Также были выделены типы покрытий и даны их краткие характеристики. Особое внимание было уделено термостойким покрытиям на основе тугоплавких эмалей, поскольку к их преимуществам относятся низкая стоимость и возможность нанесения их непосредственно на детали. Был определен ряд технических решений, которые защищают сплавы от высокотемпературной газовой коррозии и эрозии при длительной эксплуатации авиационных изделий при высоких температурах. Результаты статьи могут быть полезны для дальнейших исследований, поскольку отдельные разработки являются неинформативными с точки зрения решения конкретных задач и требуют дорогостоящего стендового тестирования заявленного технического результата.

Ключевые слова: *жаростойкость, покрытие, никелевые сплавы, стеклокерамические покрытия, технологии нанесения.*

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1. INTRODUCTION

Presently, in many industries (aviation, space, rocket, engineering), special attention is paid to increasing the tactical and technical characteristics of the products being created, increasing their reliability, resource, and economy. Solution of problems requires the use of materials capable of operating in various aggressive environments, under cyclic and alternating loads, high temperatures, pressures, vibrations, including when interacting with high-speed high-enthalpy flows of gases (air, fuel combustion products) (Tomarov and Shipkov, 2018).

High-temperature gas corrosion of nickel alloys and alloy steels is accompanied by the formation of scale on their surface, which is represented by phases of variable composition, as well as zones of inner oxidation under hardened layers (Burkov *et al.*, 2018). As a result, the de-alloying of alloys, especially those containing Nb, Mo, and W, takes place, and for steels, there is also decarburization. Changes in chemical composition, in turn, lead to deterioration in the mechanical properties of materials and their operational characteristics (Thomas *et al.*, 2018).

The problems are significantly aggravated by the using of alloys under the influence of high-speed flows of oxygen-containing gases (Astapov and Terentieva, 2014; Astapov and Terentieva, 2016; Terentieva and Astapov, 2018). As a result, their oxidation processes are significantly accelerated, which are accompanied by the destruction and delamination of formed oxide films and local plastic deformation of the surface layers. In supersonic and hypersonic flows, local gas corrosion and selective oxidation of individual alloy components are intensified, surface microrelief in the form of roughness, corrosion-erosion pits and caverns are more intensively developing, which, in turn, increases gas turbulence in boundary regions and erosive destruction of materials.

Protection of alloys from high-temperature gas corrosion and erosion using thin-layer heat-resistant coatings in many cases is the only possible way to realize their heat-proof characteristics and functional properties (Tomarov and Shipkov, 2018).

The requirements for protective coatings can be very diverse depending on the service time, which can range from a few seconds to thousands of hours, and on working conditions. Heat-resistant coatings, in addition to actually effective protection against oxidation and erosion, high physical and chemical compatibility with the base material, should have increased heat resistance and mechanical strength, and in addition, they should have low catalyticity and certain optical properties (radiation, reflection). The minimal catalytic activity on the surface at exothermic reactions of heterogeneous recombination of atoms and ions of high-speed flows reduces the chemical component of aerodynamic heating to the smallest value.

A high degree of blackness of the coating makes it possible to intensify the process of re-emission of the received heat into the surrounding space, which is important when organizing thermal protection systems for bodies of high-speed aircraft. While ensuring the thermal conditions of the flow paths of propulsion systems, the coating should have a high heat-reflecting ability in order to maximize heat dispersion through the material (Tomarov *et al.,* 2018).

Extensive literature, including inventions, articles, monographs (Astapov and Terentieva, 2014; Abraimov and Eliseev, 2001; Terentieva, 2008; Kablov *et al., 1999*; Gayamov *et al.,* 2014), is devoted to various types of heat-resistant coatings and methods for applying them to heat-proof structural materials, in particular, high-alloy chromium-nickel alloys and steels. It seems appropriate to make a critical analysis of coatings developed in Russia that are diverse in composition, technological and operational properties, from the standpoint of the possibility of using the accumulated theoretical knowledge and applied experience in relation to the tasks of providing a reliable protection system for the abovementioned structural materials under conditions that cause high-temperature gas corrosion and surface erosion layers (Astapov and Terentieva, 2016).

2. MATERIALS AND METHODS

2.1. Single-layer heat resistant coatings

Among the many single-layer heat-resistant coatings, oxide, metal, intermetallic and enamel coatings are widely used (mainly for protecting the blades of gas turbine engines (GTE) and for technological use) (Shao *et al.*, 2019). Purely *oxide coatings* based on refractory oxides (Al_2O_3, Cr_2O_3, Y_2O_3, La_2O_3, MgO, BeO, ZrO_2, HfO_2, ThO_2 or a combination thereof), applied, as a rule, by various methods of thermal spraying, are not very promising due to the high brittleness of oxides, a significant difference in the thermos-physical characteristics of the base materials and coatings, which lead to cracking of the protective layer during quick changes in temperature, thermal cycling and thermal shock, and also due to absence of reserve component, providing restoration of the coating at in case of random defects (Astapov and Terentieva, 2016). At the same time, refractory oxides are widely used as an integral part of complex – multilayer, composite heat-resistant coatings, especially if needed, in addition to protection against high-temperature gas corrosion, to provide thermal insulation properties of the surface layers of the protected parts.

The use of single-layer *intermetallic* and/or *ceramic* oxygen-free compounds (aluminides, silicides, borides, carbides, nitrides) and their compositions as protective coatings is also unpromising for practically the same reasons. Due to the significant difference in temperature coefficients of the linear expansion of refractory oxides, high-temperature intermetallic compounds, ceramics, and protected structural materials, the coatings are in tension state after application. During the cooling process, a partial relaxation of tensile stresses occurs, which is accompanied by cracking. Mono-coatings, with the exception of enamel, are used mainly as functional layers in complex – multilayer, composite systems for protecting heat-proof alloys (Shao *et al.,* 2019).

Single-layer *metal* coatings (mainly in Me-Cr-Al-Y system, where Me is Ni, Ni-Co) are still used to protect GTE blades up to 1050-1100 °C. Since the 80s of the XX century, they have replaced traditional diffusion aluminide coatings, in which, at high temperatures, the “resorption” of diffusion layers occurs and the aluminum content is unacceptably reduced. Most of multicomponent coatings of this system (Abraimov, 1993; Abraimov and Eliseev, 2001; Abraimov and Geykin, 2018; Kolomytsev, 1979; Terentieva, 2008) are formed by methods based on the processes of vacuum physical evaporation of materials with their subsequent condensation (vacuum-arc, electron-beam, magnetron), as well as using plasma spraying methods.

2.2. Multilayer, composite heat-resistant coatings

The works (Muboyadzhyan, 2011; Kablov

et al., 1999; Kablov *et al.,* 2007), demonstrate the original technological processes developed at FSUE "VIAM" for producing ion-plasma diffusion, condensed and condensation-diffusion coatings for protecting nickel blade alloys from high-temperature gas corrosion in the temperature range of 1000-1100 °C and 1100-1200 °C. For complex alloying of ion-plasma diffusion coatings, the deposition of condensed nickel-based sublayers is proposed. Alloying elements included in the nickel sublayer should modify the coating, and the sublayer becomes an obstacle to the dissolution of refractory alloy elements in the diffusion coating layer. In some cases, thin (~ 3–5 μm) anti-diffusion barrier layers of metal carbides are used, made, for example, by high-energy plasma chemistry (Muboyadzhyan *et al.,* 2010; Muboyadzhyan, 2010; Muboyadzhyan, 2011; Kablov *et al.,* 2007). It was mentioned that condensation-diffusion coatings have significantly higher protective properties compared to conventional condensed coatings. Nonetheless, they provide no information about their properties, except for the extreme temperature range of their protective ability (1000-1200 °C).

The authors of work (Poklad *et al.,* 2010) mention that the combined use of two or more technologies makes it possible to more successfully solve the problems of increasing the operability of heat-resistant coatings of Ni(Co)-Cr-Al systems up to 1200 °C. This is connected to the expansion of the usability of complex alloying with refractory elements (Ta, Re, W), effectively inhibiting the diffusion "resorption" of coatings, and micro additives (Y, La, Ce, Hf, Si), thus increasing their functional properties. The concentration limits of the content of the elements in high-temperature coatings of this system are provided.

Of interest is the information (Samoilenko, 2006) on the effect of various elements involved in mutual diffusion in the systems "nickel alloy – aluminide coatings (complex, conditioned, combined)" on the destabilization of chemical and phase compositions of coatings, their structure and, as a consequence, on the decrease of protective resource. The conclusions that were made confirm the need to suppress diffusion processes between a heat-resistant coating and a protected heat-proof alloy. For these purposes, coatings are alloyed with elements that, being in β- or γ′-phase, impede diffusion processes, or participate in formation of inner coating zone containing carbides and intermetallic compounds that impede mutual diffusion of the elements. A similar effect is made by diffusion barrier layers created in the surface layers of the protected alloys with a purpose to inhibit diffusion into the coating of refractory elements (Mo, Nb, V, Ta). The latter, entering the coating, reduce protective properties of the oxide film, lead to the loss of Al and, thereby, significantly reduce stability of the β phase and durability of the protective systems. This circumstance lets us conclude that stability of β-phase can be rather effectively controlled by targeted alloying and the creation of barrier layers, thereby solving the problem of increasing the mechanical properties of coatings while maintaining high resistance to gas corrosion and surface erosion.

2.3. Heat-resistant coatings

Attention to heat-resistant silicate enamels in aeronautical engineering is primarily conditioned by fact that, characterized by a simple formation technology and low cost, many of them can provide antioxidant protection and special surface properties (erosion resistance, anticatalyticity, emissivity) of elements of heat-loaded structures from heat-proof materials in a fairly wide temperature range. Another advantage of enamel coatings is the possibility of applying them directly to parts without any primers, undercoats, protectors.

Almost all enamels are applied by slip technology from pre-melted frits – oxide alloys synthesized at high temperatures (from 800-1000 °C for low-melting compositions, 1200-1400 °C – for medium-melting compositions and up to 1600-1650 °C – for refractory). The technological process of enameling is established (Vargin *et al.,* 1958; Solntsev and Tumanov, 1976; Solntsev, 1984; Appen, 1974; Appen, 1976) and includes:

- preparation of slip suspension (slip);
- surface preparation of protected material (degreasing, hydro-sandblasting, etching in a soda bath);
- applying of slip (with a spray gun, dipping, dousing or brush);
- drying of layer (air or oven at 50-100 °C);
- heat treatment (firing) with a purpose to reflow the enamel and firmly fix it on the surface of the product. In case of defects, the burnt enamel is removed (by sandblasting, etching in an alkaline bath), and after that, the enameling process is repeated.

Frit compositions for enamel coatings used to protect steel and nickel alloys from high-temperature gas corrosion which are produced in

Russia, the USA, Japan, France, Germany, and other developed countries, as a rule, range within the following limits in terms of the content of the main components, mass. %: SiO_2 – 25÷85, BaO – 20÷50, B_2O_3 – 0÷20, Al_2O_3 – 0÷5, MgO – 0÷3, CaO – 0÷5 (Solntsev, 1984). To improve the adhesive properties of formed coatings, the so-called adhesion oxides — CoO, NiO, MoO_3 — are introduced into the frits in small amounts. With the purpose to increase the functional characteristics of coatings (chemical resistance, resistance to erosion, degree of blackness, heat-reflecting ability), fillers are included (Cr_2O_3, Al_2O_3, TiO_2, ZrO_2, CeO_2, $ZrSiO_4$, SiB_4, SiC) through the mixture for welding upon receipt of the frit or in the form of mill additives in the preparation of the slip. Depending on the chemical and physical-mineralogical composition, glass-enamel (glassy), glass-crystalline (glass-metal) and glass-ceramic coatings can be distinguished.

3. RESULTS AND DISCUSSION

3.1. Research on single-layer heat resistant coatings

The advantages and disadvantages of vacuum spreading methods are quite fully presented in studies (Kablov *et al.,* 1999; Muboyadzhyan, 2010), the authors of which, on the basis of comparative multi-parameter analysis, demonstrated that the most preferred method (in terms of the combined parameters of density, adhesion, structural perfection of coatings combined with simplicity and reliability of equipment, performance, and process accuracy) is the vacuum-arc evaporation of alloys. Another step forward was the original high-energy ion-plasma technology developed at FSUE "VIAM" and the creation of ion-plasma systems MAP-1M, MAP-2, MAP-3. A significant amount of developed thin-layer ion-plasma diffusion, condensed, and condensation-diffusion coatings for protecting turbine blades from high-temperature gas corrosion using this method is presented in literature (Muboyadzhyan *et al.,* 2010; Muboyadzhyan, 2011; Kablov *et al.,* 1999; Kablov *et al.,* 2007). The main chemical systems with these coatings, their abbreviations and applications are given in works (Muboyadzhyan, 2011; Kablov *et al.,* 1999; Kablov *et al.,* 2007).

The monographs (Abraimov, 1993; Abraimov and Eliseev, 2001; Abraimov and Geykin, 2018; Kolomytsev, 1979; Terentieva, 2008) describe the most significant theoretical and experimental data obtained by Russian and international researchers in the process of developing protective coatings mainly for nickel-blade alloys. Attention is directed to the technology for producing diffusion coatings, which, according to the authors, will remain the most common for a long time. These coatings include, primarily, the technologies of powder and slip alitization, chromoalitization, aluminosilication, characterized by simplicity and economy. While they are used as basic ones, a large number of new coating compositions have been created in which Al and Cr remain the main alloying elements. They are present in almost all high-temperature coatings since they provide the formation of oxide film with high protective properties. The compositions of many coatings include one or more microalloying elements (Hf, Zr, Y, La, Ce, Yb), designed to improve the adhesive ability of protective films. In order to increase the thermochemical stability, metal coatings are alloyed with refractory elements (Ta, Re, Ru, Nb, W), which inhibit the diffusion "resorption" of protective layers.

A brief review of Russian materials for making single-layer condensed coatings and developed abroad (grades, chemical composition) is given in (Terentieva, 2008). Condensed coatings of the same chemical systems as diffusion ones have better properties in general since the method opens up the possibility of producing coatings of a given composition with a constant or specified distribution of alloying elements in thickness.

The use of nickel, cobalt, iron, or combinations thereof, as well as a set of alloying elements, as a basis for coatings of the Me-Cr-Al-Y system, depends on the purpose of the coatings, the material of the parts to be protected, and their operational conditions. An analysis of literature in the field of development of heat-resistant coatings for the protection of parts made of heat-proof nickel alloys and steels shows that a positive solution to the problem is usually associated with an individual approach to both the choice of coating composition and method of its formation, even within the same method. Thus, it makes no sense to consider the countless number of developed compositions of heat-resistant coatings and methods for their application from the standpoint of applicability or improvement. But attention should be paid to practicability of choosing a basic system and concentration ratio of the components of protective coatings. Therefore, from analysis of general characteristic of the properties of electron-beam coatings of Me-Cr-Al-Y systems

(Me – Ni, Co, Fe and their combination) (Terentieva, 2008) it follows that coatings have the highest protective ability on blade nickel alloys (type ZhS_6U) Ni-Cr-Al-Y, and the maximum heat resistance and high ductility are found in Ni-Co-Cr-Al-Y coatings. Coatings of Fe-Cr-Al-Y system are, usually, unstable on nickel alloys and are not recommended for use. 37% Cr in coatings of the Co-Cr-Al system is seen as a threshold concentration at which reliable protection of nickel alloys from gas corrosion is ensured for a wide temperature range. Small additives Y, Hf, Si significantly improve protective ability of coatings (Ni, Co)-Cr-Al, but their number also has its limits, for example, in Co-Cr-Al-Y – no more than 0.2-0.3% for reasons of maintaining sufficient ductility (Terentieva, 2008).

Followers of the school of Kolomytseva P.T. still continue to develop and improve heat-resistant protective coatings obtained by chemical-thermal treatment by powder mixtures and diffusion saturation in circulating gas medium (Abraimov, 1993; Abraimov and Eliseev, 2001; Abraimov and Geykin, 2018). The patents (Arzamasov *et al*., 2006; Eliseev *et al.,* 2007) offer methods for the formation of thin-layer (30–40 μm) of chromoaluminide coatings on the ZhS_6U alloy in a circulating halide medium using different sources of diffusing elements and different technological methods for chromoalification.

Various techniques are included in technological cycle of producing aluminide coatings (Eliseev *et al.,* 2003a; Eliseev *et al.*, 2003b; Eliseev *et al.,* 2003c; Eliseev *et al.,* 2007): changing the chemical composition of successively applied layers, using pre-melted ingots of special alloys, from which transition coating layers are formed by various spraying methods. At the same time, it is mentioned that, due to the shortcomings of the methods of electron beam, plasma and electric arc cathode sputtering from ingots (high porosity of the resulting coatings and the unevenness of their thickness, especially on complex parts), diffusion coatings based on aluminum are preferable, in particular: gas, or slip, or powder alitization, chromoalation, aluminosilicon.

In (Levashov *et al.,* 2009), an effective method was proposed for increasing the life of blades made of ZhS_6U alloy by applying electro spark alloying coatings using a KhTN-61 alloy made by self-propagating high-temperature synthesis to form a coating from consumable electrode. The phase composition of the coatings after one minute of treatment according to the given optimal regime is: cobalt-based solid solution and double carbides (Nb, Ti) C and (Mo, Ti) C. The presented test results show that this method of coating formation allows the creation of high-quality multifunctional antifriction alloys on the ZhS_6U alloy (decrease in the friction coefficient by 5 times), wear-resistant (increase in wear resistance by more than 10 times), including layers with increased hardness (at thickness of 40 μm $H_μ$ = 5.2 GPa). Despite the effectiveness of the considered method, a significant increase in the wear resistance, hardness, and antifriction of the resulting coatings, it remains doubtful that with this phase composition it is possible to provide protection against high-temperature gas corrosion even at moderately high temperatures.

The significant disadvantages of most diffusion saturation methods include prolonged exposure to high temperatures (1050–1230 °C) (Abraimov and Eliseev, 2001) during the formation of coatings of optimal thicknesses (80–100 μm), which cannot but adversely affect the structure stability of the protected nickel alloys, and, therefore, its properties. Moreover, it is very problematic to use these methods on "delicate" thin-walled structural elements of the flow paths of propulsion systems, which, as a rule, have complex geometry of surfaces to be protected and sufficiently large overall dimensions.

Enamel coatings are discussed in another section of this article. The developers of almost all single-layer coatings, including the above-mentioned coating, in their subsequent publications, state that two-, three-stage, mainly multilayer, complex, combined composite heat-resistant coatings have the best protective properties. They are obtained in various ways, within the same method – condensation (using electron beam, cathode, plasma, laser, and other technologies), diffusion (powder, slip, gas technologies), and by a combination of different methods and specific technics.

3.2. Specifics of multilayer, composite heat-resistant coatings

The work (Kablov *et al.,* 2007) shows the advantages and possibilities of using high-energy ion-plasma technology for applying protective and hardening multicomponent coatings based on industrial application experience for this technology. The features of ion-plasma multicomponent condensed coatings of Ni(Co, Ni-Co)-Cr-Al-Y systems developed at the VIAM Federal State Unitary Enterprise are highly dispersed structure in the initial state (~ 0.1 μm), high adhesion (> 100 MPa), high deposition accuracy and relatively low cost. Depending on the composition, they can provide at a minimum thickness of about 50 µm long protection blade alloys from hot gas corrosion up to temperatures of 800-1100 °C. Coatings are successfully used both as independent protection since they have a minimal effect on the mechanical characteristics of the base material, and as a part of sublayers for air-diffusion and heat-protective coatings. For example, a recent report (Smirnov and Budinovsky, 2017) about a significant increase in service characteristics of the currently used heat-resistant condensation-diffusion coatings of composition Ni-Cr-Al-Ta-Re-Y-Hf + Al-Ni-Y to protect the widely used heat-proof blade alloy ZhS_{32} by creating an internal nitride barrier layer by ion-plasma technology at MAP-2 installation. A similar positive effect was obtained by the authors of works (Gayamov *et al.,* 2014) when creating heat-resistant ion-plasma coatings for Al-Ni-Cr-Y system with a sublayer formed from the heat-resistant alloy of the Ni-Cr-Al-Ta-Hf-Re-Y system due to the formation of a thin barrier layer containing tantalum carbide particles.

Worthy of attention are the works (Kuznetsov *et al.,* 2007; Kuznetsov *et al.,* 2011; Lesnikov *et al.,* 2012) devoted to the creation of a coating system for protection against high-temperature gas corrosion of internal cavity and external path surface of single-crystal high pressure turbine blades (HPT) of modern gas turbine engines made of carbon-free heat-proof alloys with a high content of refractory elements (Re, Ru, Ta). A particularity of these blades is the "openwork" design with a complex system of internal cavities and perforations with a size of ~ 0.5–1.0 mm. The temperature of the outer and inner surfaces of the HPT blades varies around 250-300 °C, and the temperature of the outer path surface 1150-1250 °C. The authors believe that the solution of such complex problems is possible only by combining various methods and technologies of coating formation (gas circulation, thermal diffusion saturation, high-energy ion-plasma technology, reactive deposition during magnetron evaporation). They introduce the developed diffusion-condensation coating, formed by sequential alitization, then applying the chromium-aluminum layer by the gas circulation method, then the main layer, consisting of β-phase (Ni, Co) Al with a high Al content and alloyed 1 at.% Cr ion-plasma technology. It is stated that at present, gas circulation coatings are the most effective and, in fact, the only ones for protecting the internal cavity and perforations of cooled HPT blades. To protect the external tract surface, an ion-plasma coating of the Al-Ni-Cr-Y system has been developed. The temperature-time resource of the HPT blades protected by this complex coating is estimated as – 1150 °C, 1000 h.

The research results presented in work (Kachalin and Mednikov, 2013) show the possibility of creating heat-resistant nanostructured coatings on the details of the gas paths of aviation and space technology using physical methods of deposition in vacuum, in particular, the magnetron method in forming such coatings. It is stated that recently heat-resistant and erosion-resistant nanostructured coatings have been developed that not only protect chrome-nickel alloys from high-temperature gas corrosion at temperatures above 1000 °C but also have resistance to shock dynamic effects. It is stated that to ensure the long-term performance of the blades at t > 1000 °C, combined diffusion-conditioned multilayer coatings (based on the Ni-Cr-Al-Si-O system) containing a barrier sublayer of Ni-Cr-Al-Si nano-layer structure have been developed, inhibiting the diffusion of atoms at the boundary with the protected alloy and, thus, stabilizing the main phase of NiAl for a sufficiently long period. The addition of oxygen during the magnetron sputtering of Ni-Cr-Al-Si (or Cr-Al-Si) material helps to obtain heat-resistant protective coatings based on oxides of the mentioned systems. The structure of the coatings is a combination of microlayers with sizes from 0.5 to 3–4 µm; each of them consists of nano-layers with sizes from 20 to 40 nm. The static heat resistance of structural alloys (for example, EI652, EP693) with these coatings in the air of a chamber laboratory furnace at t = 1050 ° C, 50 hours increases by no less than 4-5 times. The test results of samples of EP693 alloy with the same coating for drop-impact erosion (droplet diameter of 800 µm, collision velocity of 250 m/s) demonstrated that no ablation occurs in the first 500-600 min, i.e. the duration of the incubation period of the ablation of the mass of the alloy protected by the coating increases by 4-6 times.

Thus, the results of numerous studies have shown that the main trends in improving heat-resistant coatings are: 1. transition from single-layer to two- and multilayer coatings, which, along with the heat resistance, make it easier to provide the required set of properties (anti-erosion, wear-resistance, tribological, special); 2. creation of diffusion barriers, since the main mechanism for the exhaustion of protective properties of coatings at high temperatures is diffusion process in the systems with "substrate-coating" and "coating-environment".

At the same time, a large reserve remains in the use of layers of heat-resistant ceramic compounds in the coating structure. There are also promising, but still weak, attempts to use nanotechnology in the development of coatings, the development of which currently goes in two ways: the creation of nanostructured coatings (with the size of individual grains in the range of 1–100 nm in three directions) and nano-layer structures with thickness of each layer within nanometer range.

3.3. Heat-resistant coatings based on refractory enamels

A wide range of enamel resource coatings has been developed to ensure the performance of materials in question at temperatures of 900-1100 °C for a long time and 1200 °C for a short time, including operation in high-speed aggressive gas flows. Among these developments, the majority belongs to heat-resistant coatings for effective protection of parts and assemblies of gas turbine engines and turbo-pump units (Solntsev *et al.,* 2001; Solntsev *et al.,* 2002; Solntsev *et al.,* 2004b; Solntsev *et al.,* 2006; Solntsev *et al.,* 2008; Solovieva *et al.,* 2009; Kablov *et al.,* 2016); some technical solutions aimed at increasing the reliability of the structural elements of liquid rocket engines (for manned and cargo spacecraft, strategic ballistic missiles, space stations) (Solntsev, 2009; Prilepsky *et al.,* 1993); there are practically no developments in the field of protection of heat-loaded parts of gliders of hypersonic aircraft and their propulsion systems (Astapov *et al.,* 2019a). The latter, first of all, is caused by temperature-time factors, significantly limiting the possibility of applying traditional structural materials in so-called hot structures. They are usually made of more heat-proof materials – alloys based on refractory metals (Nb, Mo, W), graphites, carbon composites (Astapov and Rabinskiy, 2017; Yurishcheva *et al.,* 2018; Astapov *et al.,* 2019e; Astapov and Terentieva, 2014; Astapov and Terentieva, 2016; Terentieva and Astapov, 2018) and high-temperature ceramics (Astapov *et al.,* 2019b; Astapov *et al.,* 2019c; Astapov *et al.,* 2019d). Though, the problem of ensuring the short-term performance of steels and nickel alloys at temperatures of 1250-1350 °C under the influence of high-speed flows (air, fuel combustion products) remains extremely important.

Known enamel coating (Solntsev *et al.,* 2001), designed to protect high-temperature nickel alloys from destruction from high-temperature gas corrosion in a high-speed gas stream during operation. The coating contains SiO_2, B_2O_3, Al_2O_3, BaO, CaO, MgO, TiO_2, Cr_2O_3 as well as mineral complex compound based on SiO_2. The coating is formed using slip-firing technology, heat treatment is done at a temperature of 1100-1200 °C for 2-5 minutes. The authors declare a significant increase in properties for heat-proof nickel alloys protected by this kind of coating at working temperatures of 1100 °C and higher, namely: heat resistance more than 10 times at 1100 °C, heat resistance 4 times at $1100 \leftrightarrow 20$ °C and 9 times at $1200 \leftrightarrow 20$ °C, the expansion of effective softening range of more than 150 °C. Unfortunately, it is not indicated under what conditions all the above characteristics of the properties are obtained. Apparently, it was tested in thermal furnaces under conditions of natural air convection, since there is no information about the influence of high-speed flows of hot gases on the structure and properties of coatings.

Later, the same authors patented a different composition (Solntsev *et al*., 2002) of an effective glass-enamel coating for protection of heat-proof alloys from destruction under conditions that cause gas corrosion. The advantages include a decrease in temperature of coating formation to room temperature, an increase in continuity (by 6-10%), heat resistance (by ~ 1.5-2 times) and heat resistance at temperatures exceeding 1000 °C (more than 10 times), which is confirmed by experimental data. Since the coating is formed at room temperature, it can be used not only as a resource but also as a repair tool with the possibility of application in field (airfield, polygon) conditions.

Glass-crystal coatings, in comparison with glass-enamel ones, have higher protection properties (Solntsev, 2009). Coatings of this group are characterized by a high degree of cohesion of silicon-oxygen framework, and, as a consequence, have enhanced characteristics of heat resistance, temperature resistance, and thermodynamic stability in aggressive environments containing abrasive particles.

In work (Solntsev, 2010), it is reported that for a wide range of parts made of heat-proof alloys (combustion chambers, afterburners, heat pipes, stabilizers), a series of glass-crystal coatings (EVK-103, EVK-103M, EVK-112, EVK-75, EVK-127), stable in high-speed gas flows at temperatures of 900-1000 °C for a long time and 1200 °C for a short time. These coatings are characterized by strong adhesion to the surface of alloys, bulk micro-crystallization, gas density, strong chemical bonding, high heat and heat resistance characteristics. Publication (Solntsev *et al.,* 2014), presents a glass-crystal coating of EVK-104M with system SiO_2-BaO-B_2O_3-Al_2O_3. The coating is operational for a long time ineffective protection of parts from high-temperature gas corrosion at higher extreme temperatures (1050 °C for a long time) and, like the previous ones, up to 1200 °C for a short time (when temperature peaks). The use of coating can significantly reduce the rate of oxidation of alloys (on VZh159 alloy – by 5-8 times).

In work (Denisova, 2018), new high-temperature glass-ceramic coatings based on the refractory frits of the BaO-Al_2O_3-SiO_2 system and silicon tetraboride SiB_4 are presented. It was declared that in terms of a combination of operational properties and advantages in application technology, they surpass domestic and foreign counterparts. Additive SiB_4 brings the temperature of formation of coatings closer to the temperatures of operation. The results of experimental verification indicate that they can provide effective protection of nickel alloys and heat resistant steels against high-temperature gas corrosion and ignition up to 1250 °C for more than 100 high performance of glass-ceramic coatings in strong oxidizing environments is associated with amorphous structure of their matrices, optimization of the ratio of refractory ceramic particles and glass-forming components, and the presence of bulk micro crystallization.

In works (Solntsev, 2009; Solntsev, 2014), the possibilities of using theoretical experience obtained at the "VIAM" Federal State Unitary Enterprise on example of development of reaction-curable thermoregulating erosion-resistant coatings for reusable thermal protection of materials (quartz tiles) of the Buran reusable spacecraft were demonstrated. Using silicon tetraboride (SiB_4) and silicon hexaboride (SiB_6) in the development of temperature-controlled firing “black” coatings formed as a result of chemical reactions between atmospheric oxygen, borides and high-silica matrix glass, provided the necessary complex of functional properties at operational temperatures up to 1250 °C: high degree of blackness ($\varepsilon \geq 0.9$), low catalyticity of the surface (constant of velocity of heterogeneous recombination of atoms $K_w \sim 1$ m/s), very high characteristics of heat resistance, thermostability and thermoelasticity.

Promising are some concepts based on the use of refractory oxides and more complex synthetic compositions of oxygen-free ceramics, and oxides having a common structure-forming component. As such components, Si, B, and other elements can be used. When protecting thin-walled parts of complex configuration, arises a problem of lowering the temperature of coating formation while maintaining the basic operational properties. In work (Solntsev *et al.,* 2004a), to solve the problem it was proposed to use the reaction curing effect when chemically active components (borides and fusible borosilicate glasses) are introduced into silicate systems. The latter can actively interact with components of the silicate system with the formation of liquid borosilicate phase, which bonds the particles of the refractory phase and promotes the relaxation of thermoelastic stresses. Such coatings are formed over a wide temperature range of 1120-1160 °C. The effectiveness of their protective action is not worse than that of glass-crystal coatings. In (Solntsev *et al.,* 2004a), a comparative analysis of the averaged properties of glass enamel, glass crystalline, refractory, and reaction-cured coatings was carried out. The presented data indicate that, according to the combined criteria of formation temperature, heat resistance (1000-1100 °C ↔ 20 °C mode) and time to failure in a gas stream at 1000, 1100, 1200 °C, refractory coatings are in the lead, and the properties of glass-crystalline and reaction-cured coatings are almost the same.

The authors of the patent (Prilepsky *et al.,* 1993) developed a glass-ceramic coating with a favorable set of properties for protection of alloy steel products working in high-temperature gas flow conditions, with high surface blackness values ($\varepsilon = 0.78$-0.84), and heat resistance (600-950 h at 1000 °C) and temperature resistance (920 ↔ 20 °C 500-1000 heat exchange). The protective capacity of the coating is provided by its composition, which, in addition to oxides SiO_2, TiO_2, Al_2O_3, Cr_2O_3, CaO, BaO, MnO, CoO, MoO_3, contains oxygen-free compounds SiC and SiB_4. The latter are introduced in order to increase the service life and emissivity. The thin-layer coating (40–60 μm) is applied in the form of an aqueous slip layer, which is then briefly burned at temperatures of 1130-1200 °C. Also, the coating has a favorable set of technological properties – opacity, wetting ability, continuity, strong adhesion to the base. In conditions of interaction with high-speed gas flows, the coating loses its working capacity at temperatures above 1200 °C. This is because of the transition of coating to a viscous-fluid state, in which the resistance to mechanical entrainment (erosion) is significantly weakened, which leads either to formation of a wavy (different thickness) surface with local defects, or to partial or complete runoff of the coating, or to its blowing off, i.e. to expose the substrate.

The authors of this invention were able to solve these problems (Astapov *et al.,* 2019a) by applying a coating layer 60–70 μm thick over the specified coating, containing, in addition to the previously considered components, an additional mill additives Al_2O_3 and Cr_2O_3. In the process of formation, takes place the surface crystallization of the coating and the formation of highly dispersed crystals of barium aluminosilicate with the composition $BaAl_2Si_2O_8$ ranging in size from 0.5 to 1.5–2 μm (less often up to 3–4 μm), concentrated exclusively in the surface layer with a thickness of 3–5 μm. Their refractoriness (melting point 1760 °C), high thermodynamic stability and location contribute to an additional increase in surface resistance to erosion entrainment, which positively affects the allowable working temperature of the coating. During high-temperature operation of coated products, coagulation of $BaAl_2Si_2O_8$ crystals and gradual spreading of crystallization deep into the coating layer are registered. The partial dissolution of Al_2O_3 in the barium silicate glass phase increases the viscosity of the matrix by creating a unified glass-forming framework. As a result, there is an increase in temperature resistance and resistance to erosion of the coating, expanding the limits of its short-term performance on the protected alloys – up to 1350 °C.

In summary: analyzing the presented data, it should be noted that glass enamels, refractory, glass crystalline, and reaction-cured coatings belong to the class of high-resource coatings, characterized by a combination of high physicochemical, technological, and corrosion-resistant properties. Coatings can protect structural elements from heat-proof nickel alloys and steels from high-temperature exposure to aggressive media at temperatures up to 1200-1250 °C (for a short time up to 1350 °C). Nonetheless, there is no information on the effectiveness of the protective action and mechanisms of operability of these coatings in high-speed gas flows (with rare exceptions (Astapov *et al.,* 2019a), or they are very scarce and uninformative (Solntsev *et al.,* 2004a).

4. CONCLUSIONS

A critical analysis of many years of research by conducted by Russian scientists in the field of creating single-layer and multi-layer heat-resistant coatings for heat-proof nickel alloys and steel for protection against high-temperature gas corrosion, including operation under conditions of interaction with high-velocity high-enthalpy flows of oxygen-containing gases, was carried out. Depending on the chemical and structural-phase composition, the following types of coatings were distinguished – oxide, metallic, intermetallic, glass-enamel, glass-crystal and glass-ceramic. Single-layer coatings cannot provide some of the numerous requirements for the surface of structural materials. Thus, in recent years, the main attention has been paid to the development of the architecture of multilayer, composite protective coatings. So, single-layer coatings are mainly used as intermediate layers in the "substrate – heat-resistant coating" system, where they perform barrier-compensation functions.

A significant place in this review is given to coatings based on refractory enamels. Now, a wide range of enamel resource coatings has been developed to ensure the performance of materials in question for temperatures of 900-1100 °C for a long time and 1200 °C for a short time, including operation in high-speed aggressive gas flows. Among these developments, the vast majority belongs to heat-resistant coatings for the effective protection of parts and assemblies of gas turbine engines and turbopump units; some technical solutions are aimed at increasing the reliability of the structural elements of liquid rocket engines; there are practically no developments in the field of protecting heat-loaded parts of gliders of hypersonic aircraft and their propulsion systems. The latter, primarily, is caused by temperature-time factors, significantly limiting the possibility of using traditional structural materials in high-temperature structures.

A number of technical solutions have been found; they protect the alloys from high-temperature gas corrosion and erosion during long-term operation in aircraft products at temperatures up to 1200-1250 °C with the possibility of short-term temperature peaks up to 1300-1350 °C. Some developments seem to be effective, however, they are uninformative from the standpoint of solving specific problems and require expensive testing of the claimed technical result in conditions that mimic operational conditions as applied to a particular alloy and specific features of structural elements made of the material.

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