Research of the intermetallics formation mechanism during the synthesis of functionally graded layered steel-aluminum compositions

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Abstract. The features of the formation and growth of intermetallic compounds in the production of functionally graded layered steel-aluminum compositions by the arc surfacing process have been revealed. The influence of the intermediate aluminum layer formed by the arc surfacing process on the diffusion zone formation mechanism has been shown.

1. Introduction
The friction unit elements (bearing shell, blocks of sliding bearings and others) responsible for the operating mode and resource of machines and mechanisms are mainly bimetallic structures [1-3]. The bimetallic structure consists of working antifriction layer and low-carbon steel substrate which restricts plastic deformation. However, the level of performance properties of existing antifriction materials from non-ferrous metal alloys (Al, Cu, Pb, Sn, etc.) has reached maximum values, and the potential for further improvement due to alloying or special processing has exhausted itself, as evidenced by the analysis of plain bearings emergency failure [4, 5]. Therefore the use for the working layer of new particle-reinforced composite materials (CM) based on aluminum is a promising and effective solution. The CM have both high performance properties and low specific weight, and also the possibility of multiple recovery by surface treatment [6, 7]. The use of fundamentally new functionally graded layered compositions with CM working layers will not only improve the level of performance properties, but also will reduce the friction units cost. However, the main obstacle limiting the wide using of such compositions is associated with the low adhesive strength between the coating and the substrate due to the formation of an iron-aluminum system intermetallic brittle layer at the interface [8]. Therefore, the study of the intermetallic layer formation mechanisms is important to overcome the obstacle when developing technologies for applying aluminum matrix composite coatings to steel substrates and their successful operation which has been the purpose of this work.

2. Materials and methods
Composite coatings were deposited on plates of size 150x200x2 mm made of high-quality low-carbon steel 20 (0.17-0.24 wt.% C; 0.17-0.37 wt.% Si; 0.35-0.65 wt.% Mn; ≤0.25 wt.% Cr; Fe - the rest according to GOST 1050), by an argon arc surfacing process, characterized by universality and efficiency. Preliminarily, an intermediate layer of A5 grade aluminum (0.2-0.35 wt.% Fe; 0.1-0.25 wt.% Si, ≤0.015 wt.% Cu; Al - the rest according to GOST 7871) had been formed on the steel substrate surface. According to previous studies the intermediate layer allows to limit interfacial interaction between the substrate material and the CM matrix [8]. The use of pure aluminum for the intermediate layer made it possible to take into account the influence on the formation mechanism and diffusion zone characteristics of alloying elements from only the filler metal composition.

A promising liquid phase method of arc surfacing - the process of “Cold Metal Transfer” (CMT), was used to obtain the aluminum intermediate layer. CMT process is characterized by minimal heat input and allows limiting the growth of intermetallic compounds. CMT technology was carried out on specialized welding equipment TransPuls Synergic 2700, Fronius using a A5 grade welding wire with a diameter of 1.2 mm. The CMT technological parameters (pulse current - 150 A; base current - 40 A; voltage - 18 V; surfacing speed – 0.5 m/min) ensured the formation of an intermediate layer up to 2 mm thick and were selected according to [9]. Previously the steel plate’s surface was treated by hot dip galvanizing in order to improve its wetting with aluminum melt.

The presence or absence of high-strength, refractory reinforcing particles does not affect the formation and growth mechanisms of intermetallic compounds, and first of all determines the CM characteristics level. Therefore, more cheaper materials similar in chemical composition to CM matrix alloys in the form of OK Tigrod 4047 rods (≤0.6 wt.% Fe; 11-13 wt.% Si; ≤0.15 wt.% Mn; ≤0.05 wt.% Cu; Al - the rest according to AWS A5.10) with a diameter of 2.4 mm were used. The process of argon arc surfacing of the matrix alloy was carried out according to the scheme shown on Figure 1. Technological parameters of the process, the main of which are welding current (I) - 150-160 A; arc voltage (U) - 18-20 V; surfacing speed (V) - 11-13 m/h, were selected according to [7], and provided full remelting of the intermediate layer, i.e. contact of the melt with a solid substrate.

![Figure 1. Scheme of the argon arc surfacing process. The numbers indicate: 1 – deposited layer; 2 - intermediate layer; 3 - substrate; 4 – filler rod.](image)

The structure of the diffusion zone located between the substrate and deposited coatings was studied using optical and electron microscopy. Leika DMILM light microscope equipped with a digital camera, as well as a Helios NanoLab 660 scanning electron microscope equipped with an attachment for X-ray microanalysis were used.

Microhardness (Vickers hardness - HV) of the samples was measured according GOST 9450 with using a DuraScan 70 equipment to assess the diffusion zone size, as well as to obtain quantitative data on the characteristics of the intermetallic compounds that make up it.

The characterization of the functionally graded layered compositions properties was carried out by assessing the adhesive strength of the deposited coatings according to the results of peeling and shear tests according to [10]. These test schemes were chosen due to their wide using in industry for bimetallic structures of friction units and composite bimetallic materials manufacturing. The selected
schemes were carried out using a tensile testing machine 2054 R-5. The load was applied stepwise until the samples were destroyed.

3. Results and discussion

During CMT process, the continuous intermetallic layer with a thickness of 4 to 10 μm (an average value of 8.2 μm) is formed at the interface between the steel substrate and the pure aluminum intermediate layer (Figure 2a). It should be noted that the longitudinal cracks dividing the intermetallic layer into two regions are observed along the entire length of the interface. The region located near the deposited layer has an average thickness of 3.6 μm and according to the X-ray microanalysis data consists of intermetallic compounds based on aluminum FeAl₃ and Fe₂Al₅ (Figure 2b). These phases have maximum hardness values among intermetallic compounds of the Fe-Al binary system [11].

![Figure 2. Microstructure (a) and chemical elements distribution (b) in the intermetallic layer between the steel substrate and intermediate layer of A5 grade aluminum.](image)

The formation of the FeAl₃ intermetallic phase occurs upon contact of the aluminum melt with solid steel due to the dissolution of iron in aluminum [8, 11, 12]. Subsequently, the Fe₂Al₅ compound is formed on the steel side as a result of chemical reaction:

\[ \text{FeAl₃} + \text{Fe} = \text{Fe₂Al₅} \]  

Iron-based intermetallic compounds (Fe₂Al₅, Fe₂Al₂, Fe₃Al) are located in the region adjacent to the steel substrate. These intermetallics are characterized by the shape of tongue-like outgrowths in the direction of steel (Figure 2b). They form a continuous layer of variable thickness, the average value of which is 4.6 μm.

Different values of the average thickness of the intermetallic layer regions formed by compounds based on aluminum and iron, respectively 3.6 and 4.6 μm, are probably associated with a longer duration of the diffusion process of aluminum into iron compared with the opposite direction of diffusion of iron into aluminum. The unevenness of the intermetallic layer thickness is caused by the anisotropy of the intermetallic phase Fe₂Al₅ diffusion properties. The Fe₂Al₅ phase is characterized by the ability to pass aluminum atoms in the direction perpendicular to the interface. The maximum values of the intermetallic layer thickness on the steel substrate side are observed under the conditions of the Fe₂Al₅ crystal growth along the normal to the surface (Figure 3). The crystal growth deviation from the normal direction leads to a decrease in the growth rate until it is completely stopped.

The formation of intermetallic compounds is accompanied by a significant increase in the final phase volume, which reacts with volumes of steel and aluminum. The new intermetallic compounds layers have an effect on previously formed ones. Significant stresses arising as a result of such an action can lead to the appearance of longitudinal cracks in the intermetallic layer. It explains their presence in the obtained samples, and is consistent with the results of [11].

The thickness of the 4047 aluminum alloy coatings deposited by an argon arc welding process with full remelting of the intermediate layer is up to 5 mm. The main feature in this case is the interaction of the silicon-containing aluminum melt not with the surface of the steel substrate, but with the
intermetallic layer existing at the steel-aluminum interface formed during the intermediate layer deposition.

![Scheme of the intermetallic compounds growth in the normal direction to the interface.](image1)

**Figure 3.** Scheme of the intermetallic compounds growth in the normal direction to the interface.

The significant changes occur at the interface between the obtained samples compared with the initial state. As a result two characteristic regions can be distinguished in them (Figure 4a):

- region I, resulting from the complete remelting of the aluminum intermediate layer;
- region II subjected to thermal influence during argon arc welding.

The intermetallic layer formed in region I, limited by the dimensions of the weld pool in contact with the solid steel, is continuous and uneven in thickness (Figure 4b). Its thickness values are in the range from 7 to 22 μm (average 18 μm). In region II, formed as a result of heating above the temperatures of intensive growth of the Fe-Al system intermetallic compounds and characterized by a length of up to 400 μm from the deposited metal, the thickness of the intermetallic layer is from 6 to 14 μm with an average value of 12 μm (Figure 4c).

![Scheme of the obtained samples](image2)

**Figure 4.** Scheme of the obtained samples (a), the microstructure and composition of the intermetallic layer in regions I (b) and II (c). The numbers indicate: 1 – deposited layer from 4047 aluminium alloy; 2 – intermediate layer of A5 grade aluminum; 3 – substrate from steel 20.
The complete remelting of the intermediate layer during surfacing leads to the partial dissolution of the Fe-Al binary system intermetallic layer formed during the aluminum intermediate layer deposition, as well as the development of silicon diffusion into it, as evidenced by the X-ray microanalysis results (Fig. 4, b). The maximum silicon concentration is achieved in the region of the diffusion zone located near the deposited aluminum alloy 4047 coating, and consisting of the Al7.4Fe1.8Si intermetallic phase. The silicon amount in the diffusion layer composition decreases with further movement in the steel substrate direction. It indicates the appearance of the Fe(Al, Si)3 compound. The formation of such ternary phases of the Fe–Al–Si system occurs due to the substitution of aluminum by silicon and occupation by it of structural vacancies in the initial Fe–Al binary intermetallic compounds (Fe2Al3, FeAl3) [13-15]. It is important to note that the result of this is a decrease in the rate of aluminum and iron diffusion [15-17].

Full remelting of the Fe-Al binary system continuous intermetallic compounds layer located at the steel-aluminum interface is practically excluded during surfacing. The temperature of the weld pool which prevents direct exposure to a high-temperature electric arc does not exceed 1173 K according to [18]. This temperature is significantly lower than the intermetallic compounds melting temperature (Table 1).

**Table 1.** The melting temperature of the Fe-Al system intermetallic compounds of various stoichiometric composition [19]

| Intermetallic compound | Intermetallic compound composition (in parts) | Melting temperature, K |
|------------------------|---------------------------------------------|------------------------|
| FeAl                   | Fe1/2Al1/2                                   | 1583                   |
| FeAl2                  | Fe1/3Al2/3                                   | 1365                   |
| FeAl3                  | Fe1/4Al3/4                                   | 1430                   |
| Fe2Al5                 | Fe2/7Al5/7                                   | 1444                   |
| Fe3Al                  | Fe3/4Al1/4                                   | 1825                   |

Thus, the intermetallic layer formation mechanism during surfacing of coatings with full remelting of the intermediate aluminium layer obtained by the CMT process can be represented as follows (Figure 5):

1) at the first stage, the aluminum-silicon melt is wetted and spreads over the continuous intermetallic layer surface (Figure 5a). At the same time, the surface layer of intermetallic compounds having the FeAl3 composition dissolves. It subsequently provides the possibility of physical contact of the melt with a layer of Fe2Al5 intermetallic compounds located near steel substrate (Figure 5b).

2) the second stage is characterized by the development of the diffusion process of silicon into the crystal lattice of Fe2Al5 intermetallic compounds which occurs by the vacancy mechanism. The silicon penetration depth achieved at this stage is almost comparable with the entire thickness of the intermetallic layer. As a result triple intermetallic compounds - θ-Fe(Al, Si)3 with a melting point of 1373 K are formed (Figure 5c).

3) the third stage is associated with the formation on the site of the dissolved compound of the composition FeAl3 of the intermetallic phase τ5-Al7.4Fe1.8Si, which has a melting point of 1123 K (Fig. 5, c). At elevated temperatures (above 893 K), this compound is formed by a monovariant peritectic reaction [13, 14]:

\[ L + \theta \rightarrow \tau_5 \]  \hspace{1cm} (2)

The τ5 phase crystallizes simultaneously with θ-Fe(Al, Si)3 intermetallic compounds according the reaction. At temperatures less than 893 K the formation of the τ5 phase occurs in accordance with the quasi peritectic reaction [12]:

\[ L + \theta \rightarrow \tau_5 + (Al) \]  \hspace{1cm} (3)

It is important to note that the formation of a continuous intermetallic layer of the τ5-Al7.4Fe1.8Si compound at the coating-substrate interface prevents the further growth of the θ-Fe(Al, Si)3 phase.
4) the fourth stage is accompanied by an increase in the size of the τ₅-Al₇Fe₁₈Si intermetallic compounds located near the deposited coating and crystallization of the aluminum-silicon melt (Figure 5d).

Figure 5. Scheme of the intermetallic layer formation during surfacing of an aluminum-silicon coating on steel with full remelting of the intermediate aluminum layer obtained by the CMT process. ● Atoms of Al and Si melt; ○ Atoms of Fe.

The phase composition of the intermetallic layer located in region II is characterized by the similar structure to that formed when the intermediate layer of grade A5 aluminum is produced by the CMT process (compare Fig. 2, b and Fig. 5, b). The main differences are related to its thickness, as well as the presence of numerous cracks in it. The heating of the intermetallic layer located in region II during arc surfacing to temperatures exceeding the onset of intensive growth of intermetallic compounds leads to increasing of up to 50% (from 8 to 12 μm) of its average thickness compared to the initial state. The intermetallic layer located near the intermediate aluminum layer is characterized by many cracks. Their appearance is associated with high hardness and brittleness of the phases based on aluminum (FeAl₃ and Fe₂Al₅). The increase in the average thickness of the aluminum-based intermetallides interlayer reaches 66% (average thickness: 3.6 μm in the initial state, and 6 μm after arc surfacing). The other part of the intermetallic layer located near the steel substrate has a comparable average thickness (the maximum increase was 30% of the initial value: from 4.6 to 6 μm) and consists of iron-based Fe-Al binary intermetallic compounds (Fe₂Al₃, FeAl, Fe₃Al). The compounds formation and growth are associated with the diffusion process of aluminum into steel during arc surfacing process. These results confirm the earlier conclusion that in the solid phase, the diffusion process of iron into aluminum through the intermetallic layer has a higher speed compared to diffusion of aluminum into iron.

Tests for adhesive strength were carried out in order to assess the mechanical characteristics of the samples. The obtained values of adhesive strength did not exceed 15 and 25 MPa when tested for shear and peeling, respectively. Thus, both increase in the diffusion rate as a result of thermal action...
during the arc surfacing of an aluminum coating on steel with complete remelting of the intermediate layer of pure aluminum obtained preliminarily by the CMT process combined and uneven distribution of intermetallic compounds of various stoichiometric composition, lead to the thickness of the intermetallic layer increasing by up to 50% in comparison with the initial samples, as well as significant cracking mainly from the deposited layer side, whereby the adhesive strength values correspond to 25 MPa.

4. Conclusion
The mechanism for the formation and growth of intermetallic compounds upon contact of an aluminum-silicon melt with intermetallic compounds of the Al-Fe system during the process of arc surfacing of functionally graded layered steel-aluminum compositions with working coatings made of aluminum based CM is proposed.

It is shown that the CM matrix melt contacts with the Fe-Al system intermetallic layer located at the interface during arc surfacing on steel with a pre-deposited intermediate aluminum layer. The intermetallic layer partial dissolves during the contact with CM matrix melt. The degree of its dissolution depends on temperature and time the existence of a weld pool. The dissolution rate of the initial intermetallic compounds is lower than the formation rate of new ternary Fe-Al-Si compounds. It leads to an increase from 8.2 to 18 μm in the intermetallic layer average thickness in the produced functionally graded layered steel-aluminum compositions, which can be recommended for use in friction units whose adhesive strength is 25 MPa.

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