

# Formation of the surface alloys by high-intensity pulsed electron beam irradiation of the coating/substrate system

Yu F Ivanov<sup>1,2</sup>, E A Petrikova<sup>1,2</sup>, A D Teresov<sup>1,2</sup>, O V Krygina<sup>1,2</sup>, M E Rygina<sup>3</sup>

<sup>1</sup> Institute of High Current Electronics SB RAS, Tomsk, 634055, Russia

<sup>2</sup> National Research Tomsk State University, Tomsk, 634050, Russia

<sup>3</sup> National Research Tomsk Polytechnic University, Tomsk, 634050, Russia

E-mail: yufi55@mail.ru

**Abstract.** The results of the analysis of the structure and properties of the surface layer of aluminum A7 subjected to alloying by the intense pulsed electron beam melting of the film / substrate system. Fold increase in strength and tribological properties of the modified surface layer due to the formation of submicro - nanoscale multiphase structure have been revealed.

## 1. Introduction

The possibility of synthesis the nanoscale metastable phases with unique physicochemical and performance characteristics in the surface layer of treated material is the main advantage of the approach based on the formation of surface alloys. This approach implemented in a number of methods: formation of coatings by electroexplosive method, welding powders using CW laser and compression plasma flows, high dose ion implantation, diffusion saturation in plasma of gas discharges and many others [1-3]. Along with the obvious advantages, these methods have and disadvantages.

For example, electroexplosive method and welding deposition method leads to the formation of inhomogeneous microstructure, the large grain size and high porosity, due to the high dimensional heterogeneity of the original powder coating and the relatively small rates of the melt quenching [3, 4].

The aim of this work is the developing of a method of alloying the surface layer of metals and alloys based on formation and irradiation of the film/substrate system by high-intensity pulsed electron beam submillisecond exposure time.

## 2. Materials and methods of study

Aluminum A7 was used as a material for study [5]. Formation and liquid-phase mixing of the film / substrate (A7) system was carried out in a single vacuum cycle by a pulsed electron beam on the "SOLO" setup (IHCE SB RAS) [6]. Silumin of a grade AK12 (Al-12%Si) was used as electron beam sputtering target (disk with diameter of 30 mm, a thickness of 6 mm) [7]. Aluminum substrate (plates 15\*15\*5 mm), located at the distance  $\approx 50$  mm from the target is attached to a flat plate installed on the shaft of the step motor (the last one located parallel to the plane of the manipulator) to allow rotation of the sample between the sputtering and mixing processes. At the initial moment, the modifiable surface of aluminum substrate was faced to the target and it located at an acute angle to the surface of the table. The target, fixed on a water-cooled table, irradiated by a pulsed electron beam with the following parameters: the energy density of 40 J/cm<sup>2</sup>, pulse duration of 100  $\mu$ s, the pulse



frequency 1 Hz. Under these conditions, the deposition rate  $\approx 5$  nm/pulse. The coating thickness varied from 500 to 1000 nm, and determined by the number of irradiation pulses. After sputtering the plate with the sample, using a step motor was located parallel to the plane of the table and moved to the area of influence of the electron beam. Changing the mode of operation of the electron-beam setup "SOLO" (electron energy of 16 keV; the pulse frequency  $0.3 \text{ s}^{-1}$ ; the pulse duration of electron beam of 50  $\mu\text{s}$  and 150  $\mu\text{s}$ ; the energy density of the electron beam (10 ... 25)  $\text{j/cm}^2$ ; the number of pulses 1, 3, 5), was conducted liquid phase mixing of the film/substrate (A7) system to form the surface alloy. All processes have been carried out in a vacuum chamber at a pressure of the working gas (Ar)  $\approx 3,5 \cdot 10^{-2}$  Pa. Thus, the formation and liquid phase mixing of the film / substrate system have been performed in a single vacuum cycle. Examinations of the structure of the surface alloy was carried out by optical (device Microvisor metallographic  $\mu\text{Vizo}$  - MET-221) and scanning electron (device LEO EVO 50) microscopy. The elemental composition of the surface layer was studied using the methods of electron microprobe analysis (device LEO EVO 50). Estimation of elastoplastic properties and hardness of the surface alloy were conducted on a dynamic ultramicrohardness tester (nano indenter) Shimadzu DUH-211S and micro hardness tester PMT-3. Studies of wear resistance were carried out on a tribometer «CSEM Tribometer High Temperature S / N 07-142»; as a counterbody were used a ball of a hard alloyVK8. The measurements were carried out in a «rotation of the sample at a fixed counterface» scheme. Measurements of the profile of friction groove of the surface of the samples with the numerical determination of the depth of the friction groove and the area of its cross-section have been conducted at the end of the test using 3D-profilometer MICRO MEASURE 3D station French company STIL.

### 3. Experimental results and their discussion

There is rather multiplicity of the methods, which allow changing purposefully the properties of a detail at modification of structural and phase state of its surface layer [1-3]. One of the methods is the formation of surface alloys. That based on the use of the concentrated fluxes of energy (CFE) for irradiation of the film/substrate system. In the present work as it was above, the alloying of aluminum surface layer as carried out in a uniform vacuum cycle, using an intensive pulse electron beam as CFE. The results of the change of friction coefficient ( $\mu$ ), wear rate ( $V$ ) of A7 aluminum in an initial state and after a surface alloying are presented in the table. The estimates of wear rate were carried out by equation (1):

$$V = \frac{2\pi RA}{FL}, \quad (1)$$

where R – the radius of a track of wear [mm], A – the area of cross-section of wear track [ $\text{mm}^2$ ], F – the normal load [N], L – the distance [m] passed by a ball.

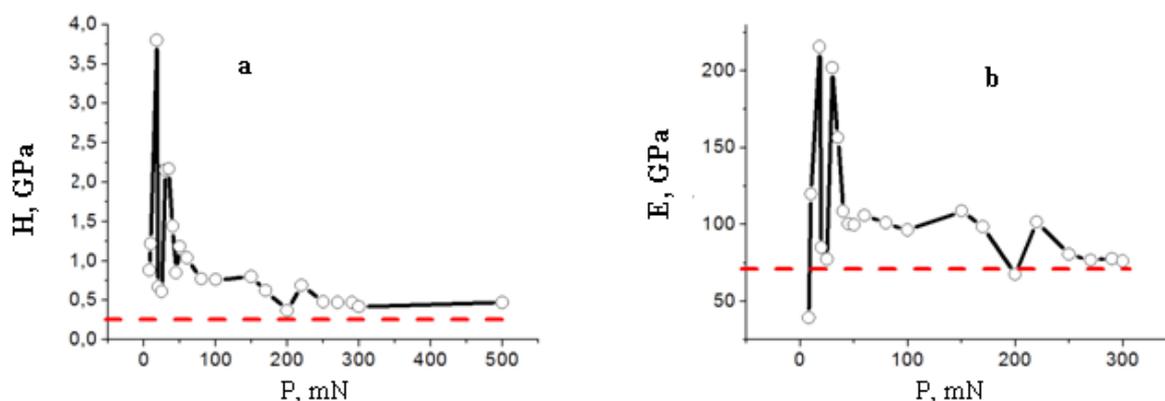
The analysis of the data presented in the table 1 showed that the best result is observed at deposition of a film with thickness of 1  $\mu\text{m}$  and the subsequent irradiation of film/substrate system by intensive pulse electron beam at the following parameters: 20  $\text{J/cm}^2$ , 150  $\mu\text{s}$ , 5 pulses,  $0,3 \text{ s}^{-1}$ .

**Table 1.** Wear rate and the friction coefficient of the modified surface layer of aluminium A7<sup>a</sup>.

| $E_s, \text{J/cm}^2$ | Thickness of the film, $\mu\text{m}$ | $V, 10^{-6}, \text{mm}^3/(\text{N}\cdot\text{m})$ | $V_0/V$ | $\langle\mu\rangle$ | $\langle\mu_0\rangle/\langle\mu\rangle$ |
|----------------------|--------------------------------------|---|---------|---------------------|---|
| 0,0 (initial A7)     | 0,0                                  | 7590  |         | 0.48                |   |
| 15                   | 0,5                                  | 6080  | 1,3     | 0.39                | 1,5                                     |
| 15                   | 1,0                                  | 6570  | 1,4     | 0.40                | 1,2                                     |
| 20                   | 0,5                                  | 5470  | 1,4     | 0.35                | 1,4                                     |
| 20                   | 1,0                                  | 4790  | 1,6     | 0.31                | 1,6                                     |

<sup>a</sup> The mode of treatment of the film / substrate system by electron beam: 150  $\mu\text{s}$ , 5 pulses.,  $0,3 \text{ s}^{-1}$ ;  $E_s$  - energy density of the electron beam;  $V_0$  and  $\mu_0$  - wear rate and friction coefficient of aluminum A7 in the initial state (before modification).

Formation of surface alloy is followed by change of strength characteristics of material. In Figure 1 the results of hardness change for the alloyed aluminum surface layer are presented. It is clear that the hardness and the Young's modulus of surface layer smoothly decreases from the values which is repeatedly exceeding strength characteristics of initial material to the values corresponding to A7 alloy. Therefore, the modified layer smoothly passes into the main volume of a sample. The boundary, which can contain concentrators of tension, is absent.



**Figure 1.** Dependence of hardness ( $H$ ) and Young's modulus ( $E$ ) on normal load (nanoindentation method) of aluminium surface layer modified by silumin deposition (thickness is  $1\ \mu\text{m}$ ) and the subsequent treatment by an electron beam on the mode of  $20\ \text{J}/\text{cm}^2$ ,  $150\ \mu\text{s}$ , 5 pulses,  $0,3\ \text{s}^{-1}$ . The dash-dotted lines designated the characteristics of initial aluminium A7 alloy.

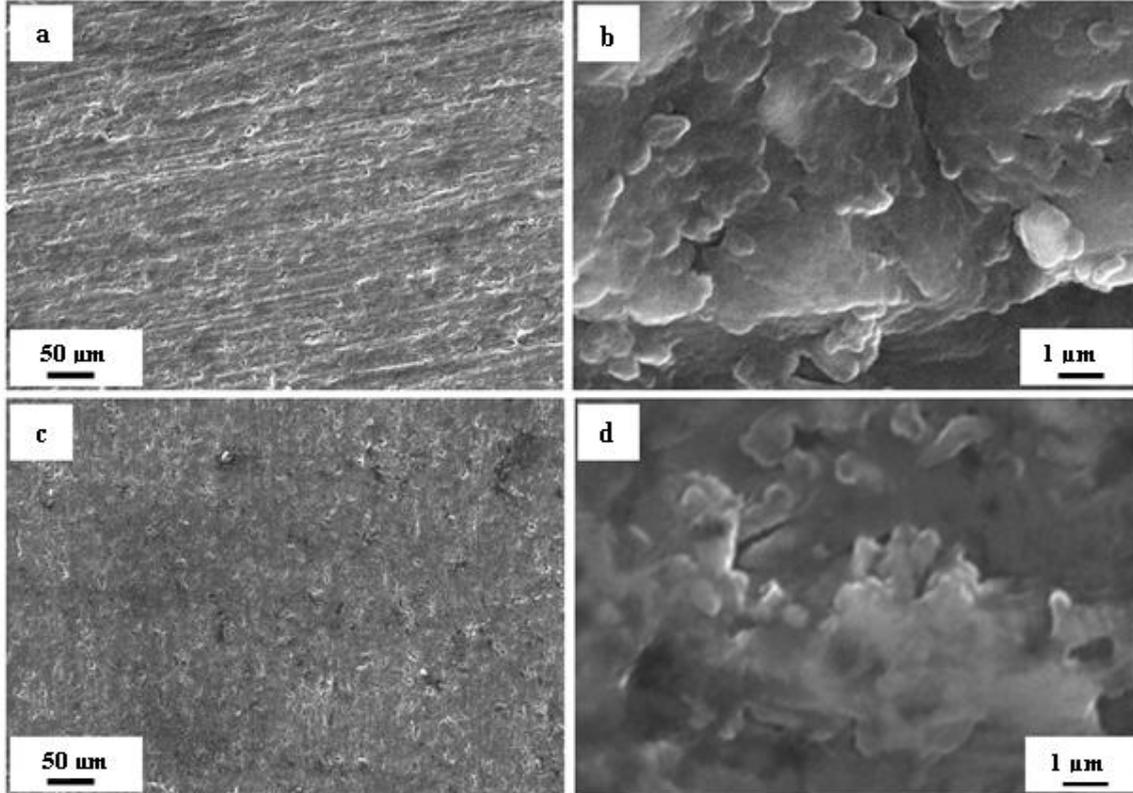
Fold increase in strength and tribological properties of the surface layer of aluminum indicates a significant transformation of elemental, phase and structural state of the material. Figure 2 shows electron microscopic images of the surface structure formed by irradiation of the film/substrate system by a pulsed electron beam. Clearly shows that in the surface layer is formed submicrocrystalline structure, crystallite sizes vary from  $0.3\ \mu\text{m}$  to  $0.6\ \mu\text{m}$ .

The crystallite size of the surface layer depends on the thickness of the deposited film and on the electron beam energy density reaches the minimum sizes when irradiated by an electron beam with an energy density of  $20\ \text{J}/\text{cm}^2$  (Figure 2, c, d).

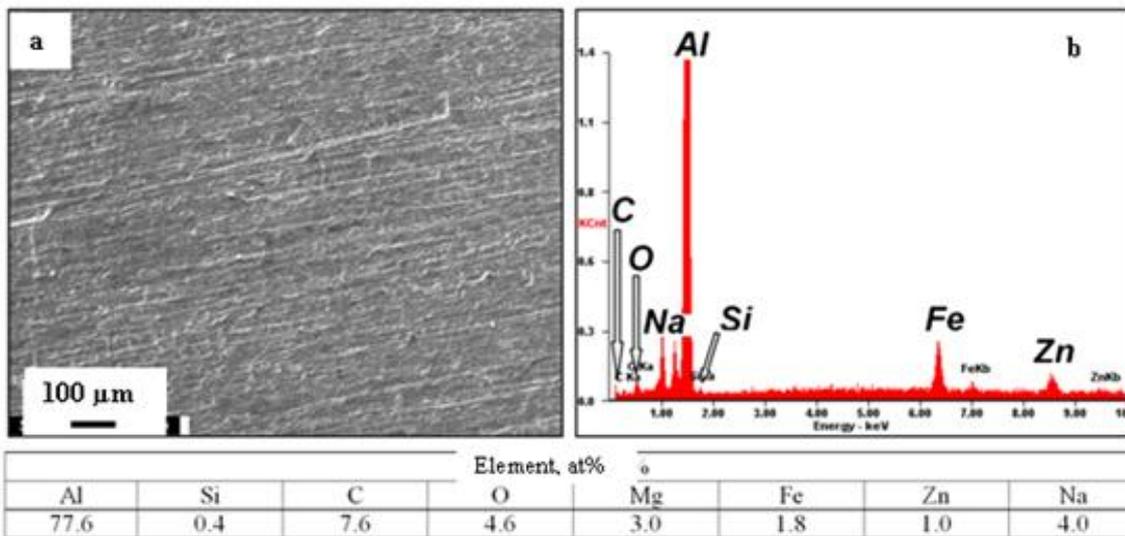
The elemental composition of the modified layer was determined by electron microprobe analysis. Typical images of the energy spectrum obtained with this method for studying of the elemental composition is shown in Figure 3, b. The results showed that the surface layer is characterized by complex elemental composition, the main element of which, except aluminum, is oxygen and carbon. The concentration of silicon in a surface layer of the substrate repeatedly (in 25-30 times) lower than in the deposited sample. Consequently, the hardening of the modified layer and the improving of its tribological properties are due to the formation of oxycarbide phases based on aluminum.

Obviously, that sputtering of the target by a pulsed electron beam is accompanied by modification of a material surface layer. Figure 4 shows the electron microscopic images of the surface structure of AK12 silumin before irradiation (Figure 4a, b) and after treatment with a pulsed electron beam with the parameters of  $40\ \text{J}/\text{cm}^2$ ,  $100\ \mu\text{s}$ , 100 pulses,  $1\ \text{s}^{-1}$  (Figure 4, c, d). Initially, the structure of the polished surface of the target is characterized by the presence of coarse inclusions of plate and globular forms of silicon (Figure 4, a, b). A contrast of silicon inclusions is slightly different from the contrast of the aluminum substrate when capturing an image of the surface structure of silumin in back-scattered electrons, as aluminum and silicon has similar atomic masses. Inclusions with acicular and skeletal shape in addition to silicon are detected (Figure 4, a). Comparatively bright contrast of these inclusions indicates that they are enriched with the heavier elements relative silicon and

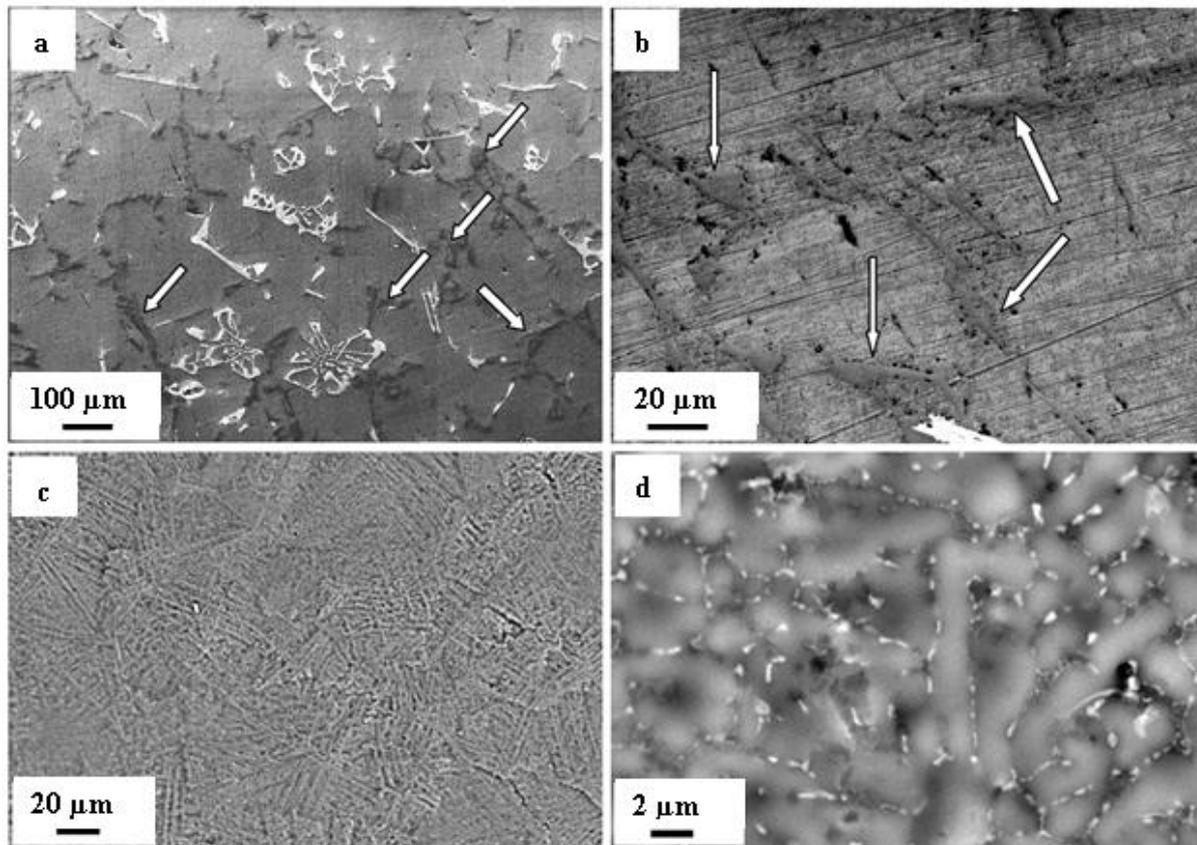
aluminum. Indeed, electron microprobe analysis revealed the presence in such inclusions of atoms of iron, copper and manganese in addition to aluminum and silicon. In [8, 9] are shown that the inclusion of such forms may be phases of  $Al_5SiFe$  ( $\beta$ -phase) and  $Al_{15}(FeMn)_3Si_2$  ( $\alpha$ -phase).



**Figure 2.** Electron microscopic image of the surface structure that is formed by irradiation of the film/substrate system by intense pulsed electron beam with the parameters: a, b -  $15 J/cm^2$ ,  $150 \mu s$ , 5 pls.,  $0.3 s^{-1}$ ; c, d -  $20 J/cm^2$ ,  $150 \mu s$ , 5 pulses,  $0.3 s^{-1}$ ; a, b - film thickness of  $0.5 \mu m$ ; c, d -  $1 \mu m$ .



**Figure 3.** An electron microscopic image of the surface (a) formed upon irradiation of film/substrate system electron by pulsed electron beam; b – the energy spectrum obtained from the plot shown in (a); table elemental composition specified the area shown in (a). The thickness of the deposited film is  $1 \mu m$ , electron beam irradiation mode:  $20 J/cm^2$ ,  $150 \mu s$ , 5 pulses,  $0.3 s^{-1}$ .



**Figure 4.** The structure of the silumin target surface in the initial state (a, b) and after irradiation by pulsed electron beam ( $40 \text{ J/cm}^2$ ,  $100 \mu\text{s}$ , 100 pulses;  $1 \text{ s}^{-1}$ ) (c, d). The arrows in (a, b) are shown the silicon particles.

After sputtering of target by pulsed electron beam the surface layer structure is changing radically. Firstly, there is the formation of a dendritic-cellular structure with high speed crystallization (Figure 4, c, d). The cell dimensions are vary from 1 to 3  $\mu\text{m}$ . Inclusions of the second phase with size of 100 ... 300 nm are located at the cell boundaries. The electron microscopic images of the structure obtained in back-scattered electrons are shown that the nano-sized particles have a relatively more bright contrast, indicating the content of elements with high atomic weight in them, along with aluminum and silicon. Indeed, electron microprobe analysis of these particles revealed atoms of iron and copper in addition to aluminum and silicon. Secondly, the element and, obviously, the phase composition of the surface layer are changes: it is determined by methods of microprobe analysis that the average volume surface layer concentration of silicon is much lower ( $\approx 7 \text{ at.}\%$ ) relative to the initial state ( $\approx 13 \text{ at.}\%$ ). Therefore, the sputtering of target (AK12 silumin) is accompanied by a change not only the structure (formed submicro- nanoscale homogeneous structure with high speed crystallization), but also the elemental composition of the surface layer (the concentration of silicon reduces substantially in  $\approx 2$  times).

#### 4. Conclusion

A liquid phase mixing and formation of film/substrate system by intense pulsed electron beam in a single vacuum cycle is carried out. The mode of deposition ( $40 \text{ J/cm}^2$ ,  $100 \mu\text{s}$ ,  $1 \text{ s}^{-1}$ , 1000 pulses) and irradiation ( $20 \text{ J/cm}^2$ ,  $150 \mu\text{s}$ , 5 pulses,  $0.3 \text{ s}^{-1}$ ) of the film/substrate system by intense pulsed electron beam, allowing to multiple increase in hardness and wear resistance of the surface layer of A7

aluminum is identified. It has been established that the increase of the strength and tribological properties of the modified surface layer caused by formation of submicron-nanoscale multiphase structure.

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