Surface temperature measurements of TiCuN-A7 system during pulsed electron-beam treatment

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Abstract. The paper deals with surface temperature measurements of the TiCuN-A7 system during the pulsed submillisecond e-beam treatment. The variable parameters include beam energy density, pulse duration, and coating thickness. The authors determine the temperature dependences, maximum temperature values, as well as the heating and cooling rates of the surface treated with a pulsed electron beam. They establish the optimal conditions for electron-beam treatment of the TiCuN-A7 system.

1. Introduction
Currently, the scope of combined surface modification methods is becoming increasingly expanded to embrace the state-of-the-art techniques in place to enable a wide range of properties, compositions and structures of the surface layer of a metal material [1]. One of the promising methods of interest to the researchers is the coating process to be followed by electron-beam treatment of a TiCuN-A7 system [1]. Subject to specific energy densities, coating compositions and substrate properties, the technology allows for the production of different surface alloys ~ 0.1–100 μm thick [2], or fusion of single-layer hard coatings into lighter substrates with a view to obtaining highly adhesive films [3], or formation of relatively thick stable coatings (> 6 μm) in multi-cycle deposition-irradiation conditions.

For all electron-beam TiCuN-A7 modifications, the main criterion for a modified layer to be generated is the temperature over the area subject to electron beam irradiation as well as the rate of its change and the rate of heating and cooling of a material. The temperature characteristics are primarily affected by the energy of an electron beam on the substrate (energy density per pulse), the properties of coating and substrate materials (thermal conductivity, melting point, etc.), the coating thickness, and the cooling rate of the specimens.

The study aims to measure the surface temperature of the TiCuN-A7 system irradiated with a pulsed submillisecond electron beam; to determine temperature dependences for different coating thicknesses, different beam energy densities in a pulse, and different pulse durations; to identify optimal electron-beam treatment conditions for the TiCuN-A7 system.

2. Materials and methods
TiCuN coatings were deposited and subsequently treated with a pulsed electron beam in a single vacuum cycle on an automated COMPLEX setup [4] developed and manufactured at the Institute of...
High Current Electronics SB RAS (Tomsk) and designed for electron-ion-plasma surface engineering of materials and products. Specimens were made of commercially pure A7 aluminum (Al – 99.7%; Fe – up to 0.16%; Si – up to 0.16%; Ti – up to 0.02%; Cu – up to 0.01%; Zn – up to 0.01%) and were electrochemically polished to the arithmetic roughness average $R_a = 0.1–0.2 \mu m$. The initial microhardness $HV_{0.5}$ was 0.3 GPa.

The specimens were coated via plasma-assisted arc deposition during the evaporation of sintered Ti-12% Cu cathode [5] in a nitrogen medium at a discharge current $I_d = 40$ A. The ion current density of a gas plasma constituted 30% of the total ion current density of a gas-metal plasma on the substrate. The growth rate of TiCuN coatings was $11 \mu m/h$. For stripping an oxide film off the surface to increase coating-to-substrate adhesion, it was preliminarily bombarded with titanium ions at low pressure (< 0.1 Pa) and high negative bias (~ 1 kV). Electron-ion-plasma treatment of Al substrates involved coating them with TiCuN 1 $\mu m$ thick and irradiating the TiCuN-A7 system with a pulsed electron beam during the 6-cycle operations until a relatively thick coating ($\geq 6 \mu m$) was formed in a single vacuum cycle. The EBT conditions were as follows: accelerating voltage $U_a = 10$ kV, energy density per pulse $E_s = 5$ and 10 J cm$^{-2}$, pulse repetition rate $f = 0.3$ s$^{-1}$, pulse duration $\tau = (100–400)$ $\mu s$, number of pulses $N = 3$. Such ranges were chosen so as to relieve the internal stresses accumulated in the relatively thick nitride coatings during their formation; to provide fusing of a hard coating into a low-melting substrate avoiding a significant change in the basic properties of a coating, its phase composition and structure after treatment.

The surface temperature was measured on a SOLO vacuum electron-beam setup [6], developed and manufactured at the Institute of High Current Electronics SB RAS and used for surface modifications of various materials and products [7]. It is equipped with a plasma-cathode electron source based on a low-pressure pulsed arc discharge [8], and is capable of generating an electron beam with the pulse duration of 20–200 $\mu s$, the current of 20–300 A, the electron energy of 5–25 keV, the energy density of up to 100 J cm$^{-2}$ at a repetition frequency of 0.3–20 s$^{-1}$. The operating pressure range of the electron source is 0.01–0.05 Pa, which is controlled by a constant working gas (argon) inlet through the electron source. Being irradiated by a pulsed electron beam, the surface of metal materials is heated and cooled with a rate of up to $10^8$ K s$^{-1}$ and $10^6–10^7$ K s$^{-1}$, respectively, which leads to surface smoothing, pulsed quenching, and, hence, to significantly enhanced physical, chemical and service properties of materials and products as such.

The target surface temperature of coated aluminum specimens was measured using a Kleiber KGA 740–LO high-speed infrared pyrometer connected by a flexible waveguide to an LVO 25 lens suitable for adjusting the focus and distance to the irradiated surface. Figure 1 shows a temperature measurement circuit, where 1 – an electron source; 2–3 – magnetic coils; 4 – manipulator table; 5 – irradiated target; 6 – chromel-alumel thermocouple; 7 – Fluke 175 multimeter; 8 – LVO 25 lens; 9 – KV quartz glass; 10 – Kleiber KGA 740-LO high-speed infrared pyrometer; 11 – power and control units of an electron source; 12 – Tektronix TDS 101B oscilloscope. The temperature range of the pyrometer was 300–2300 °C, bandwidth – 2–2.2 $\mu m$, response time – 6 $\mu s$, and temperature difference – 1 °C. In order to reduce the heat loss during measurements, the specimens were fixed on the surface of a manipulator table with thin stainless steel strips.

The pyrometer lens was located inside a vacuum chamber with a specially designed manipulator, but was kept at atmospheric pressure. The temperature inside the vacuum chamber was collected with a quartz glass 1.5 cm thick transparent for a specified bandwidth. The pyrometer had an analog output of 0–10 V for signal transmission to an oscilloscope. To ensure calibration of pyrometer readings, a chromel-alumel thermocouple was spot welded on the back of a specimen to prevent the thermocouple from a direct electron beam impact. The voltage from the thermocouple was measured with a multimeter. The lens focus and the emitting surface area were visually controlled with a backward light beam (figure 1). Since the lens was fixed at an angle of $\approx 30^\circ$ to the irradiated surface, the emitting surface area represented an ellipse with dimensions $\approx 4\times8$ mm$^2$. The pyrometer has a potential to set an emissivity $\varepsilon$ by setting a required potentiometer value in the range 0.1–1. The procedure for calibration of pyrometer readings and estimation of surface emissivity $\varepsilon$ was effected as
follows. A thermally insulated specimen with a welded thermocouple was uniformly heated to 500 °C by a pulsed electron beam under pulse-frequency conditions (repetition frequency 8 s⁻¹, pulse energy density 0.5 J·cm⁻², pulse duration 50 μs, electron energy 10 keV). By varying the potentiometer value, the temperature 500 °C on the thermocouple corresponded to 1 V on the oscilloscope. The final potentiometer value indicated the emissivity for a given material and the state of its surface at the time of measurement. As the specimen was cooled after pulse-frequency electron-beam heating, several more temperature (thermocouple) – voltage (oscilloscope) matches were measured to plot a calibration curve to determine the surface temperature in specified irradiation modes. Each specimen was calibrated before and after measurements to detect emissivity deviations from the initial value. The target surface temperature in A7 Al specimens with TiCuN coatings was measured at the following parameters: accelerating voltage $U_a = 15$ kV, pulse energy density $E_S = 5$ and 10 J·cm⁻², pulse duration $\tau = 100$ and 200 μs.

![Figure 1. Schematic for measuring surface temperature during electron beam irradiation.](image)

3. Results and discussion

Measurements of the target temperature and electron beam parameters showed that the temperature-time relationship at the target surface is similar to the waveforms of electron beam current pulses that are rectangular and have a rise time of 30–40 μs and FWHM of 100–200 μs. The indicative electron beam waveform and time dependences of the surface temperature for Al with a 1-μm-thick TiCuN film at $E_S = 10$ J·cm⁻², $\tau = 200$ μs, are shown in figure 2. During the beam current rise to be $\approx 4 \times 10^6$ A·s⁻¹, the increase in the target temperature was $\approx 2 \times 10^7$ deg·s⁻¹.

Electron-beam treatment of coated specimens strongly relies on the coating thickness, since the heat conduction from the TiCuN-Al system is influenced not only by the dimensions and thermophysical parameters of the substrate, but also by the characteristics of the coating itself. Provided that commercially pure aluminum with no TiCuN coating (figure 6 in [9]) is exposed to a pulsed electron beam, the temperature-time relationship grows to a maximum of $\approx 700$ °C and falls after the pulse ceases, at the following parameters: $E_S = 8$ J·cm⁻², $\tau = 100$ μs. The heating rate in the target area was $\approx 5 \times 10^7$ deg·s⁻¹, while the cooling rate was $\approx 2 \times 10^6$ deg·s⁻¹.
For a TiCuN coated specimen ($h_{\text{TiCuN}} = 1 \, \mu m$) (figure 3), an increase in the surface temperature during the beam current rise time, varies in the range $(0.9–3.0) \times 10^7 \, \text{deg} \cdot \text{s}^{-1}$ and depends on the heating rate, i.e. beam power density. The cooling rate calculated during the beam fall time is within $(1.1–2.0) \times 10^6 \, \text{deg} \cdot \text{s}^{-1}$. During the e-beam pulse, an increase in the target temperature also occurs at $E_S = 5 \, \text{J} \cdot \text{cm}^{-2}$ by $\approx (50–90) \, {^\circ C}$, at $E_S = 10 \, \text{J} \cdot \text{cm}^{-2}$ by $\approx (100–115) \, {^\circ C}$. With an increase in the pulse duration from 100 to 200 $\mu$s, the rate of temperature increase during the pulse slows down by an order of magnitude from $2 \times 10^6$ to $5 \times 10^5 \, \text{deg} \cdot \text{s}^{-1}$. Obviously, when the pulse duration increases at the same beam energy density, the maximum reported temperature decreases, when the energy density with the same pulse duration grows, it increases (figure 3). The maximum heating temperature $(789 \, {^\circ C})$ for a TiCuN film of 1 $\mu m$ thickness is measured at $E_S = 10 \, \text{J} \cdot \text{cm}^{-2}$ and $\tau = 100 \, \mu s$. Remarkably, this appears to be the only mode for a melting point ($T_m = 660 \, {^\circ C}$) to be achieved for Al with a TiCuN film 1 $\mu m$ thick. During the pulse, the target surface temperature is no lower than $450 \, {^\circ C}$, regardless of a treatment mode.

Given that the coating thickness in the TiCuN-Al system increases to 3 $\mu m$ (figure 4a), the temperature increase during the beam current rise, the cooling rate of the target surface, and the temperature increase during the pulse all remain the same as for the thickness 1 $\mu m$. For all e-beam parameters, the melting point of Al is reached, the maximum temperature depending on the irradiation parameters lies in the range 682–944 $\, {^\circ C}$, the temperature during the pulse does not fall below $600 \, {^\circ C}$. The maximum temperature 944 $\, {^\circ C}$, as in the first case, is measured for $E_S = 10 \, \text{J} \cdot \text{cm}^{-2}$ and $\tau = 100 \, \mu s$.

A further increase in the coating thickness up to 6 $\mu m$ does not provoke a change in the experimental temperature dependences (figure 4b). However, the temperature rate during the pulse for $\tau = 100 \, \mu s$ increases by a factor of 1.5–2 due to an increase in the coating thickness that has a thermal conductivity $\approx 10$ times lower [10] than an Al substrate (220 $\, \text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$). For all pulse durations and energy densities, the Al substrate is melted; the maximum temperatures for different beam parameters lie in the range 683–990 $\, {^\circ C}$. The maximum temperature for a coating thickness 6 $\mu m$ (990 $\, {^\circ C}$) is measured, as in the first two cases, for the beam parameters $E_S = 10 \, \text{J} \cdot \text{cm}^{-2}$ and $\tau = 100 \, \mu s$.

Admittedly, TiCuN coatings treated at a beam energy density of $E_S = 10 \, \text{J} \cdot \text{cm}^{-2}$ changes the characteristic golden color to silver, which is starkly illustrated for a pulse duration of $\tau = 100 \, \mu s$. This signifies the TiCuN destruction and justifies the low properties compared to the coatings treated at $E_S = 5 \, \text{J} \cdot \text{cm}^{-2}$. The destruction can be caused by the thermal heating $\geq 850 \, {^\circ C}$ [11]. Thus, the optimization of electron-ion-plasma conditions for the TiCuN-Al system requires, first of all,
addressing the thickness and thermal properties of the coating. In the selected range of TiCuN parameters and thicknesses, it makes sense to conduct EBT at $E_S = 5 \text{ J cm}^{-2}$, $\tau = 200 \mu\text{s}$ for coating thicknesses up to $6 \mu\text{m}$; $E_S = 5 \text{ J cm}^{-2}$, $\tau = 100 \mu\text{s} - 1 \mu\text{m}$; $E_S = 10 \text{ J cm}^{-2}$, $\tau = 200 \mu\text{s} - 1 \mu\text{m}$. At $E_S = 10 \text{ J cm}^{-2}$, $\tau = 100 \mu\text{s}$, e-beam treated coatings with a thickness of $1 \mu\text{m}$ or more can be destructed and, hence, lose the desired properties.

Figure 4. Experimental surface temperature for TiCuN-coated Al ($h_{\text{TiCuN}} = 3 \mu\text{m}$, (a) and $h_{\text{TiCuN}} = 6 \mu\text{m}$, (b)) versus time at $E_S = 5 \text{ J cm}^{-2}$, $\tau = 100 \mu\text{s}$ (1); $E_S = 10 \text{ J cm}^{-2}$, $\tau = 100 \mu\text{s}$ (2); $E_S = 5 \text{ J cm}^{-2}$, $\tau = 200 \mu\text{s}$ (3); and $E_S = 10 \text{ J cm}^{-2}$, $\tau = 200 \mu\text{s}$ (4).

4. Conclusion
Thus, surface temperature measurements in TiCuN-coated A7 aluminum alloy treated by a pulsed submillisecond electron beam using a high-speed infrared pyrometer show that the surface temperature depends on coating thickness, beam energy density in a pulse, and pulse duration. An increase in the coating thickness affects the target surface temperature due to a decreased thermal conductivity of the TiCuN-Al system. The maximum temperature (990 °C) is measured for a coating thickness of 6 $\mu\text{m}$ at electron beam parameters: $E_S = 10 \text{ J cm}^{-2}$ and $\tau = 100 \mu\text{s}$.

The heating rate of the target specimens during the beam current rise time is $(0.9–3.0)\times10^7 \text{ deg s}^{-1}$ depending on the heating intensity (power density) and coating thickness; cooling rate during the fall time lies in the range $(1.1–2.8)\times10^6 \text{ deg s}^{-1}$; the rate of temperature increase during the pulse is $(1.0–50)\times10^6 \text{ deg s}^{-1}$. During the pulse, irrespective of the mode, the surface temperature is no lower than 450 °C for a coating thickness of 1 $\mu\text{m}$, 600 °C – for 3 $\mu\text{m}$, and 610 °C – for 6 $\mu\text{m}$. The maximum temperature in the range of the selected parameters with a coating thickness of 1 $\mu\text{m}$ is 790 °C, 3 $\mu\text{m}$ – 944 °C, and 6 $\mu\text{m}$ – 990 °C.

Each coating thickness requires such e-beam treatment when the target surface temperature does not exceed $\approx 800$ °C that is commonly followed by the destruction of the nitride coating. Electron beam treatment is reasonable at the following parameters: $E_S = 5 \text{ J cm}^{-2}$, $\tau = 200 \mu\text{s}$ with coating thicknesses up to $6 \mu\text{m}$; $E_S = 5 \text{ J cm}^{-2}$, $\tau = 100 \mu\text{s} - 1 \mu\text{m}$; $E_S = 10 \text{ J cm}^{-2}$, $\tau = 200 \mu\text{s} - 1 \mu\text{m}$. At $E_S = 10 \text{ J cm}^{-2}$, $\tau = 100 \mu\text{s}$, e-beam treated coatings with a thickness of $1 \mu\text{m}$ or more can be destructed and, hence, lose the service properties.

Acknowledgements
This work was supported by the Russian Science Foundation (grant No. 19-19-00183).

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