ALUMINUM SURFACE COATING OF COPPER USING HIGH-CURRENT ELECTRON BEAM

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High-current electron beam irradiation has been applied for surface coating of copper with aluminum in ablative mode at the TEMP-A accelerator with energy of 350 keV, pulse length of 5 µs, and fluence 10…200 J/cm^2. The aluminum-rich surface layer with average thickness around 25 µm, microhardness of 6.7 GPa and elasticity modulus of 122 GPa was formed on the copper template.

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INTRODUCTION

The high-current electron beams (HCEB) are less known as a technique for modification and testing of materials compared to the laser sources due to technological complexity of the e-beam facilities, radiation protection, etc. Nevertheless, there were numerous investigations on irradiation of solids by HCEB [1, 2] (and references cited therein). Intense e-beam exposure stimulates proceeding of simultaneous processes such as fast heating, melting, evaporation, ejection of plasma and neutral vapor from the surface, generation of deformations and high dynamic stresses in the solid medium, etc [3 - 7]. In fact, the variety of these processes, called ablative processes, is defined by the electron energy, specific beam power, impulse duration, and depend on the material characteristics. The induced fast heating and cooling rates, high stresses lead to significant changes in structural-phase state of the subsurface layers, modification of the structure-dependent properties of different materials as described in [4, 6, 7]. In this paper, we only deal with the microsecond pulses [6] of e-beams, so, duration of exposure is far bigger than the characteristic electron-phonon relaxation time for the target medium. We operate with e-beams in nearly relativistic range (~0.35 MeV) in ablation mode, that enables deeper penetration of electrons into the bulk of material and provokes explosive melting with liquid splashing. It should be noted, that the low-energy e-beams have relatively shallow impact on the material surface, that is why they are usually replaced by the laser sources for such purpose.

Microsecond e-beam pulses are the effective technique for generating the directed vapor and liquid flaws as well as the bulk material fusing, thus creating new opportunities for the HCEB-assisted coating technologies. The existing ones can be classified into two major types according to distance between the materials for deposition and the processing surface: (i) remote and (ii) nearby-placed (i.e. precoating before the following e-beam treatment). Another way to divide the methods based on the characteristics of physical processes during the HCEB-impact: (i) rapid melting and solidification, (ii) surface alloying of coatings into the bulk, (iii) surface fusing of coatings to the bulk [1]. The recent experimental studies in the same vein are presented by Weisenburger et al. in [1]. They have shown practical perspectiveness of the surface alloying and surface fusing methods for coating of the cladding tubes to prevent corrosion. According to [1], coatings previously deposited or foils placed on the material's surface (T91 ferritic and 316L austenitic steels) are melted together by the impinging 120-keV electron beam with an energy density of 30…50 J/cm^2, and those efficiently reduce oxidation and corrosion rates in liquid lead alloys.

Some efforts have been made on the experimental investigation of the HCEB modification of material's bulk [4, 6], gas-plasma cloud generation [3], and the rapid solidification coating [7] by our team. The radiation-thermal strengthening after HCEB exposure was observed in [4]. In other work [6], aluminum, titanium as well as stainless steel were deposited on the cold copper templates to gain better understanding about remote surface coating using HCEBs. The targets for deposition were placed at some distance apart sample surfaces, which, in their turn, were not subjected to e-beam irradiation. It was noticed, that coatings, obtained using such method, are nonuniform and rough due to hydrodynamic instability of melted metal during its transport to template, but their hardness increased compared to the similar coatings got using conventional methods.

However, both the literature and our previous research represent none efforts about surface coating of activated template, when the combined effect of material deposition from target and activation of template's surface is achieved simultaneously by one e-beam shot. Thus, the main purpose of this study is to investigate the general specifics of such coating under exposure by the microsecond HCEB at the TEMP-A pulsed electron beam accelerator facility [6]. As the candidate materials for such research, we have chosen the previously studied [6] couple: aluminum for the deposition target and cooper for the template. Both of them were subjected to the HCEB impact, but with different fluences. The strategy was to irradiate the aluminum plate in the intense ablative mode to generate damage products enough for deposition on the copper foil, which was thermally activated by the tangent impact of the e-beam, impinging at low angle to the copper surface (Fig. 1) significantly decreasing fluence. The most important is to find experimentally advantages provided by the proposed method in contrast to the remote surface coating method on cold template, studied previously [6].

1. MATERIALS AND METHODS

A plate of technically pure Al (99.5 %wt.) and a copper template (Cu 99.9 %wt.) were irradiated at the TEMP-A pulsed e-beam accelerator [6], located in the NSC Kharkiv Institute of Physics and Technology. The beam's parameters were the following: the current of 2 kA, electron energy ~ 0.35 MeV, impulse duration τ_p ~ 5 µs. One-impulse irradiation was conducted under the pressure of about 10^{-5} Torr. The thickness of the Al-plate and copper template were 2 mm and 200 µm respectively. The Gaussian-shaped solid cross-sectional beam had a diameter ~ 40 mm and a full-width half-maximum (FWHM) of about 10 mm. The complex target was irradiated normally to the collector surface that means the copper template was subjected to sliding irradiation with the energy fluence in the range of 10 to 40 J/cm^2, and the Al-plate was irradiated normally in order to provide the maximal incident energy density around 200 J/cm^2. The main part of the Al-plate (Fig. 1) was fixed on the accelerator collector perpendicularly to the incident e-beam, and its other bended part served as a holder for the Cu-template, which was initially inclined at β ~ 40° to the e-beam axis with one free side directing towards the collector (see Fig. 1). The bended part of Al-plate was inclined at α ~ 30° to the e-beam axis. Such construction enables thermal activation of the copper template, condensation of the dense plasma cloud and liquid splash on its surface, as well as prevents it from destruction.

The specimens for metallography, fractography and hardness measurements were taken in the epicenter zone and in the periphery zone of the e-beam impact (Fig. 2). The sectioning of special parts was performed by means of shears. The fractionation was done by manual tensile bending rupture at the room temperature.

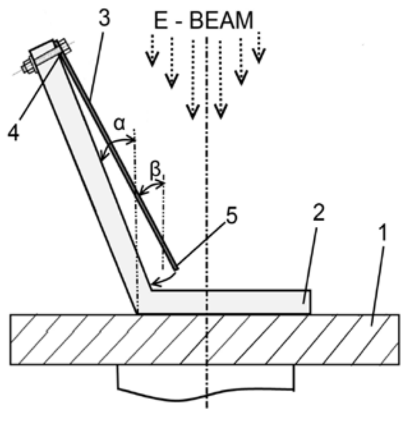

Fig. 1. Schematic view of the prepared target: 1 – collector; 2 – Al-plate; 3 – Cu-template; 4 – strong fixation of template to Al-plate; 5 – free side of template

Preliminary visual and morphological analysis of the irradiated copper foil and its cross-fractures was performed using the light microscope Bresser BioLux NV. Fractographic and energy-dispersive X-ray spectroscopic analyzes of the cross-fractures were conducted using the scanning electron microscope JEOL JSM-840. Then, the cross-sections were prepared for metallographic and hardness analyzes. The specimens were polished using a micron diamond powder W0-1. To reveal the hardness H^{50} properties of the copper foil after modification, we used the PMT-3 microhardness testing machine equipped with Berkovich trihedral diamond pyramid with an applied load of 50 kgf. Estimates of the elastic modulus E and nanohardness H of the modified copper template were obtained using the continuous stiffness method on MSSI's Agilent Nano Indenter G200 by Berkovich indenter to the indentation depth up to 500 nm. The hardness was calculated using the Oliver-Pharr analysis method. After hardness measurements had been performed, metallographic analyses were performed. To detect the microstructure of the material, the chemical etching was carried out using the Kroll's reagent (2 ml HF, 6 ml HNO_3, and 92 ml H_2O) at the room temperature; etching time of 45 s. The average grain sizes were measured by chord intercept method.

We conduct numerical finite element (FE) modeling (with finite difference (FD) discretization of time operators) of the thermal and stress evolution in the samples to find the corresponding fluences and optimize the modification process. It was performed in 64-bit Ubuntu 15.04 LTS using a C++-like language in the free-source software FreeFem++-3.36. We applied the thermoelastic ablation numerical model, which was previously described in [4]. The energy deposition profiles and the scattering factors was calculated in Casino 3.2v.

2. RESULTS AND DISCUSSION

Fig. 2 shows the copper template with the formed Al-rich coating after the HCEB irradiation. The e-beam heating was intense enough to melt the surface of the copper template and deflect its free side from angle β to α. Some amount of copper was ejected into vacuum and condensed onto the aluminum target.

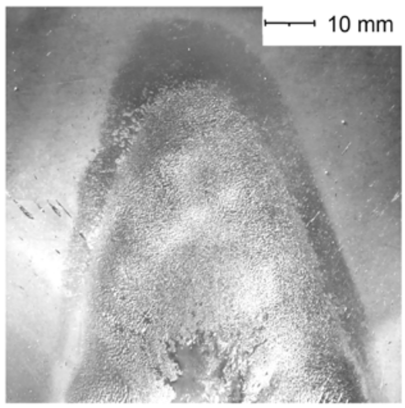

Fig. 2. Surface of Cu-template after the HCEB irradiation

The Al-target was exposed to the aforementioned high fluence, which provoked damage with explosive melting accompanied by liquid splash and generation of gas-liquid and plasma cloud. These ablative products also interacted with the ejected material from the template itself. Almost 30% of ablated aluminum condensed on the thermally activated copper template. It was easy to notice, that the coating was formed during fast solidification of the droplet-gas cloud, preceded by some mixing with melt on the template. In fact, such construction of the complex target is simple and reliable method to detect expansion of the gas-plasma torch in contrast to the conventional ones [3]. The obtained coating directly replicates the density of the ejected ablative products, preserving the shape of torch.

Noteworthy, when the complex target was extracted from the vacuum chamber, the copper template was stranded. While the coating was deposited at an elevated temperature and cooled down rapidly to normal temperature, the thermal expansion mismatch between the Al-rich coating and the copper template results in compressive residual stress.

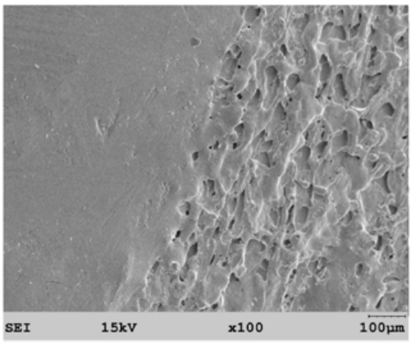

Fig. 3. Surface of coating on the boundary between smooth epicenter and wavy periphery zones

The obtained coating consists of the epicenter zone, where the dense core of the cloud condensed, and the periphery zone. These two characteristic zones are clearly distinguished by the surface roughness (see Figs. 2, 3). The first one has relatively smooth surface in the micrometer scale with few dimples and ridges of (1…5) µm deep. On the “as-fabricated” surface, using optical microscopy and SEM, we noticed a needle-like alignment of surface morphology at low magnification, and dendrite-like character was found at higher magnification. The coating on the periphery is very wavy and not solid. It has deep dimples, ridges and voids. SEM analysis showed, that it consists mainly of deformed droplets. Such complicated nonuniform surface was created due to high kinetic energy of the deposited vapor and melt, which had not been accumulated by the copper melt layer on the template. Moreover, the total density of material flux and surface tension were not enough to shrink the gaps between the droplets during fast cooling through thermal conduction to the bulk. Inasmuch as the roughness of the periphery zone surface approximately equals its thickness, so, such coating cannot be considered as functional.

Then, the fracture surface morphology was investigated (Fig. 4). The nonirradiated copper bulk has long coarse fatigue striations, which were found perpendicular to applied load (e.g. parallel to the surface). They appeared due to tensile bending rupture process. We observed ductile character of fracture at higher magnification along with limited macroscale brittle. Periodic shear lips were also found. We noticed the transitional layer in the sample between the unmodified copper material and the Al-enriched coating. Formation of this layer was caused by the tangent e-beam irradiation and further thermal conduction from the hotter coating. It was melted and then resolidified with coating upon it. This heat-affected layer is a mediate between the coating and substrate. Its thickness is around 30 µm. Its rupture has relatively flat surface and characterized by brittle fracture. Several big cracks directing from the coating to the bulk were observed (see Fig. 4). Their origins begin on the boundary with the coating. Such cracks could be formed when the stranded template was manually expanded for sample preparation, whether it was provoked by the residual stresses.

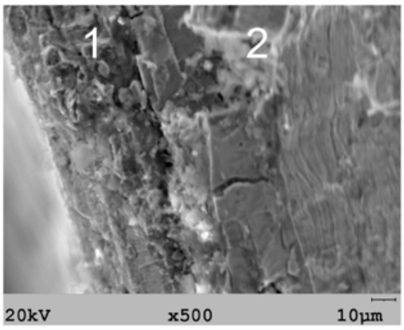

Fig. 4. Fractogram of aluminum coating on the copper template: 1 – coating; 2 – copper template

The formed coating (see Figs. 4, 5) is characterized by the rough fracture surface that revealed ductile-brittle fracture mechanism. It has short intergranular branched cracks in preferable vertical direction. We also found a net of parallel cracks inside the coating, especially in the epicenter zone. Worth to mention, they did not cause any significant delamination of the coating from the template. The parallel cracks are considered to be caused by relaxation of compression residual stresses. Interestingly, we noticed some fine debris – particles with linear size of (1…5) µm, and several 5-µm deformed facets.

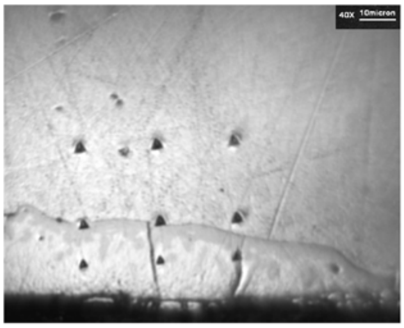

Fig. 5. Aluminum coating on the copper template

Metallographic data revealed the specifics of the coating’s microstructure. In the epicenter zone, its thickness is up to 30, and 20 µm in the periphery. It consists of the large grains with parallel size of (20…40) µm and vertical size of (6…12) µm. Among the boundaries of the large grains, there were found the abovementioned fine debris (their part ratio is around 15%). After deep etching of the samples, a few hidden round voids with size of 5…10 µm were observed in the coating. EDX analysis of the element composition showed, that in the epicenter zone it is around 25%Cu/75%Al (±5%, %wt.), and in the periphery it varies in the range of 25%Cu to 90%Cu.

We tested the hardness and elastic modulus for the dense coating at the epicenter zone. It has microhardness H^{50} ~ 6.7 GPa, that equals to its average nanohardness H measured experimentally (see Fig. 5), and

Young's modulus E ~ 122 GPa. The transition mediate has H ~ 1.6 GPa and E ~124 GPa, and the nonirradiated copper has 1.4, and 127 GPa, respectively. Thus, the physical and mechanical properties of the obtained coating is significantly higher, than in case of the remote surface coating on cold copper template described in [6], when the microhardness of nonuniform aluminum coating does not exceed H^{50}_{max} ~ 5.49 GPa.

Finally, these results motivate the use of the HCEB-assisted coating method on activated template as it provides good integrity of coating and adhesion to substrate. It is proposed, homogenization for elimination of surface roughness and voids can also be achieved through post-treatment with the light HCEB exposure. However, in-depth study about the melt dynamics along with the intense e-beam damage of polycrystalline materials is needed to obtain better practical results.

CONCLUSIONS

The Al-grade and the Cu-template have been irradiated simultaneously by the single HCEB impulse to create the Al-rich coating on the template. The copper template was exposed to the sliding impact, which guaranteed its thermal activation and light surface melting for further condensation on it of aluminum deposited from the nearby placed target. The condensation of the dense Al-cloud core resulted in formation of the compacted Al-Cu layer with smooth surface and good adhesion to the bulk material. This coating has hardness around 6.7 GPa and Young's modulus of 122 GPa. The obtained results have demonstrated, that the proposed coating method with microsecond HCEBs is a promising technique for improvement and protecting of the surface-sensitive physical and mechanical properties.

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ПОВЕРХНОСТНОЕ НАНЕСЕНИЕ АЛЮМИНИЯ НА МЕДЬ С ПРИМЕНЕНИЕМ СИЛЬНОТОЧНОГО ЭЛЕКТРОННОГО ПУЧКА

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Облучение сильноточным электронным пучком применено для поверхностного нанесения алюминия на медь в абляционном режиме на ускорителе ТЕМП-А с энергией 350 кэВ, длительностью импульса 5 мкс и флюенсом 10…200 Дж/см^2. На медной подложке сформировался поверхностный слой, обогащённый алюминием, со средней толщиной 25 мкм, микротвёрдостью 6,7 ГПа и модулем упругости 122 ГПа.

ПОВЕРХНЕВЕ НАНЕСЕННЯ АЛЮМІНІЮ НА МІДЬ З ВИКОРИСТАННЯМ СИЛЬНОСТРУМОВОГО ЕЛЕКТРОННОГО ПУЧКА

С.Є. Донець, В.Ф. Клепіков, В.В. Литвиненко, Ю.Ф. Лонін, А.Г. Пономарьов, Р.І. Старовойтов, О.А. Старцев, В.Т. Уваров

Опромінення сильнострумовим електронним пучком застосовано для поверхневого нанесення алюмінію на мідь в абляційному режимі на прискорювачі ТЕМП-А з енергією 350 кеВ, тривалістю імпульсу 5 мкс, і флюенсом 10…200 Дж/см^2. На мідній підкладці сформувався поверхневий шар, збагачений алюмінієм, з середньою товщиною 25 мкм, мікротвердістю 6,7 ГПа і модулем пружності 122 ГПа.