Surface & Coatings Technology 383 (2020) 125246

Contents lists available at ScienceDirect

Surface & Coatings Technology

journal homepage: www.elsevier.com/locate/surfcoat

Structure and phase states modification of AL-11SI-2CU alloy processed by ion-plasma jet and pulsed electron beam

D. Zaguliaev[a], V. Gromov[a], Yu. Rubannikova[a,*], S. Konovalov[b], Yu. Ivanov[c], D. Romanov[a], A. Semin[a]

[a] Department of Natural Sciences, Siberian State Industrial University, Novokuznetsk 654007,Russia
[b] Department of Metals Technology and Aviation Materials, Samara National Research University, Samara 443086, Russia
[c] Laboratory of Plasma Emission Electronics, Institute of High Current Electronics of the Siberian Branch of the RAS, Tomsk 654052, Russia

ARTICLE INFO

Keywords:
Processing with ion-plasma jet
Titanium
Yttrium
Electron beams
Phase composition

ABSTRACT

In the research the investigations into structure, elemental and phase composition, defect substructure of Al-11Si-2Cu hypoeutectic alloy subjected to processing with ion-plasma jet consisting of titanium and yttrium oxide with subsequent irradiation by high intensity pulsed electron beam with energy density of beam of 25 J/cm^2 (two-stage processing) were carried out by methods of scanning and transmission electron diffraction microscopy, micro-X-ray spectral analysis. It has been found that structure of Al-11Si-2Cu alloy in cast state demonstrates a polyphase morphologically varied character of the material. Microcraters, solid aluminium-based compounds, intermetallide and eutectic compounds are present in it. It has been revealed that two-stage processing of Al-11Si-2Cu alloy results in cardinal transformation of samples' surface layer structure and it is manifested in dissolution of silicon inclusions and intermetallides of micron and submicro dimensions typical of Al-11Si-2Cu cast alloy. It has been detected that ion-plasma jet effect on alloy surface is accompanied by both alloying of surface layer by plasma elements and penetration of initial powder particles of yttrium oxide. Along with atoms of initial material (Al, Si, Cu, Ni, Fe) the surface layer is enriched by atoms of titanium, yttrium, oxygen. It has been shown that a multielemental polyphase layer ≈60–70 μm thick having a submicro-nanocrystalline structure is formed as a result of two-stage processing.

1. Introduction

Aluminium alloys are considered to be leading materials among non-ferrous metals by volumes of production and they are also widely employed in many branches of industry due to unique features of aluminium: a good combination of mechanical and technological properties, low density, high corrosion properties, distribution of raw material for its production in nature. It is known that 60% of aluminium castings being produced in the world are manufactured from aluminium silicon (Al—Si) alloys [1]. Distribution of Al—Si alloys is dictated first by requirements of different manufacturers because they are used as modern structural materials in automobile, aircraft and ship building industries, in different workpieces and joints of railway transport and oil industry as well as in building and other branches of industry [2,3].

Due to their technological and industrial importance aluminium alloys are widely investigated experimentally and theoretically. Nowadays many research teams try to improve physical and mechanical characteristics of alloys. Polyphase composites based on Al—Si system [4] are being created. Problems of plastic deformation effect on tribological characteristics of $AlSi_9Cu_3$ alloy are being studied [5]. In research [6], the effect of alloying elements and rate of crystallization on structure and mechanical properties of Al-11Si-2Cu eutectic alloy was determined. One of widespread methods of Al-alloys' hardening is surface layer modification to which such types of processing as plasma spraying of wear-resistant coatings [7–9], electron beam processing [10,11], laser irradiation [12–16] and magnetic treatment [17,18] belong. Main feature of methods mentioned is pulsed and local character of surface effect that is a significant economical advantage of their application compared with traditional methods of treatment as alloying [19,20] thermal treatment [21,22] pressure shaping.

Investigations in the field of surface modification show that combination of different methods of its treatment has a greater effect on physico-mechanical properties and surface relief formation than a single-stage treatment. However, if we compare one method of treatment and different combination of methods then the most promising one is seen to be the modification of products' surface by ion-plasma jet

* Corresponding author.
E-mail address: rubannikova96@mail.ru (Y. Rubannikova).

https://doi.org/10.1016/j.surfcoat.2019.125246
Received 31 October 2019; Received in revised form 2 December 2019; Accepted 8 December 2019
Available online 11 December 2019
0257-8972/

[23] followed by surface irradiation with high intensity pulsed electron beam [24,25]. Processing with ion-plasma jet enables alloying to be carried out by both simple metals and complex compounds - carbides, oxides, borides etc., providing a high level of surface operational properties [26,27].

Subsequent processing by high intensity pulsed electron beams enables one to obtain a more homogeneous coating with improved physico-mechanical properties by means of melting of surface layers of a material and subsequent high velocity crystallization [28,29]. Moreover, the modification method makes it possible to control and regulate the quantity of energy supplied to surface being processed, to perform processing locally only in the immediate regions of fracture in the process of operation. The method possesses small coefficients of energy reflection, superhigh rates of heating and cooling of surface layer.

The purpose of the research is to analyze changes in structure, defect substructure, elemental and phase composition of surface layer of Al-11Si-2Cu hypoeutectic alloy subjected to two-stage processing including the effect on sample by ion-plasma jet followed by irradiation with high intensity pulsed electron beam.

2. Material and methods

Hypoeutectic alloy Al-11Si-2Cu was used as material under study. Chemical composition obtained according to results of X-ray spectral analysis of samples is shown in Fig. 1, a. The research samples were made in the form of plates and had dimensions 20x20x10 mm^3 (Fig. 1, b.).

Surface modification of Al-11Si-2Cu alloy samples was performed by two-stage processing. Fist, Al-11Si-2Cu alloy samples were subjected to ion-plasma jet processing consisting of Ti-Y_2O_3 system (titanium foil mass, m_{Ti} - 58.9 mg; mass of Y_2O_3 powder, $m_{Y_2O_3}$ - 58.9 mg; discharge voltage - U = 2.8 kV) and subsequent effect by high intensity pulsed electron beam (energy density of electron beam - 25 J/cm^2; energy of accelerated electron - 17 keV; length of electron beam pulse - 150 µs; number of pulses - 3). Electrodischarge unit EVU-60/10 was used for ion-plasma jet processing. Spraying was carried out in vacuum according to the following procedure: weighed sample of Y_2O_3 powder was placed on titanium foil clamped between two coaxial electrodes. Then, voltage was supplied to it through vacuum discharger and explosion took place under effect of electric current of great density. Products of explosion consisting of plasma component and including Ti and Y_2O_3 particles of different dispersion directed to sample along technological chamber and deposited on sample with melting through the surface layers of the material. Thus, a polyphase and multicomponent coating was formed on surface of the product being processed. The next stage in processing became the irradiation of resultant coating with high intensity pulsed electron beam. Irradiation was carried out at unit 'SOLO' [30]. After processing with ion-plasma jet the sample was placed in vacuum technological chamber of the unit and oriented to electron beam by plane modified with jet.

Studies of elemental and phase composition, state of defect substructure were carried out by methods of scanning electron microscopy (device Philips SEM-515), transmission electron diffraction microscopy (apparatus JEM-2100F) [31–33]. Foils manufactured by methods of ion thinning of plates cut out perpendicular to surface of irradiation were analyzed. Such location of foils enables one to perform the analysis of structure and elemental composition of the material depending on distance from modification surface. Elemental composition of material was studied by methods of micro-X-ray spectral analysis.

3. Results and discussion

Structure of Al-11Si-2Cu alloy in cast state obtained by methods of scanning electron microscopy demonstrates a multiphase, morphologically varied character of the material, typical images of structure of alloy under study are presented in Fig. 2. Inclusions of the second phases have different shapes. Dimensions of inclusions vary from units to tens of micrometers. Solid compounds based on aluminium (Fig. 2, region 1), intermetallide (Fig. 2, region 2) and eutectic (Fig. 2, region 3) compounds are observed in alloy structure and presence of microcraters (Fig. 2, region 4) is observed in some places.

Investigations into elemental composition of different regions of Al-11Si-2Cu alloy in cast state were carried out by methods of X-ray spectral analysis (Fig. 3). Results presented in the figure testify to inhomogeneous distribution (in volume) of chemical elements that form compounds being different in dimensions, contrast, morphology and elemental composition.

When analyzing the graph shown in Fig. 3b, it can be noted that compounds (regions 1,3,6,7 in Fig. 3, a) possess the increased content of nickel and copper that is indicative of their belonging to intermetallide phase. In regions 2 and 5 the increased content of silicon testifying to their belonging to eutectic phase is observed. Region 4 consisting of solid aluminium-based compounds possesses the chemical composition close to that of initial material.

Characteristic electron microscopic images of surface structure of Al-11Si-2Cu alloy processed by ion-plasma jet followed by irradiation with high intensity pulsed electron beam are shown in Fig. 4. As a result of two-stage processing the relief surface containing a large number of microcraters (Fig. 4, a, microcraters are designated by white arrows) and inclusions of rounded shape (Fig. 4, a, particles are designated by black arrows) is formed. Surface layer being formed is divided into regions whose dimensions are less than 1 µm (Fig. 4, b). A two-stage processing of Al-11Si-2Cu alloy results in cardinal transformation of structure of samples' surface layer. First of all, it is manifested in dissolution of silicon inclusions and intermetallides of micron and submicron dimensions typical of Al-11Si-2Cu cast alloy.

Results of investigations into elemental composition of modified surface layer of Al-11Si-2Cu alloy depending on distance to two-stage processing surface are depicted in Fig. 5. When analyzing the results it can be noted that thickness of the layer alloyed by titanium, oxygen and

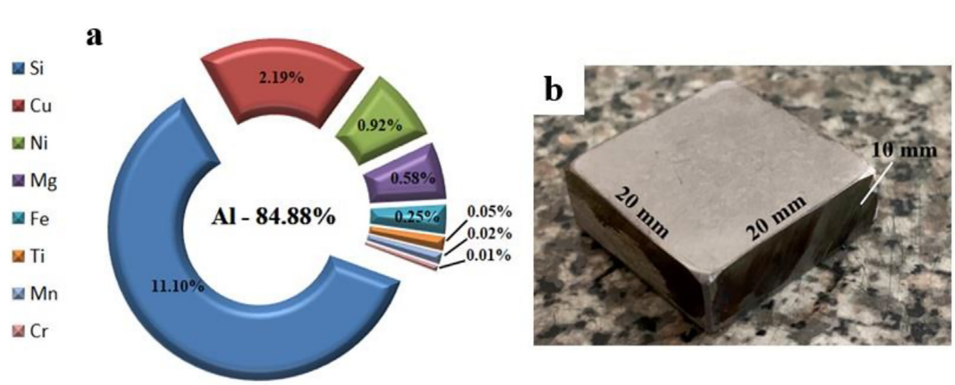

Fig. 1. Results of X-ray spectral analysis of Al-11Si-2Cu alloy (a), geometrical dimensions of sample (b).

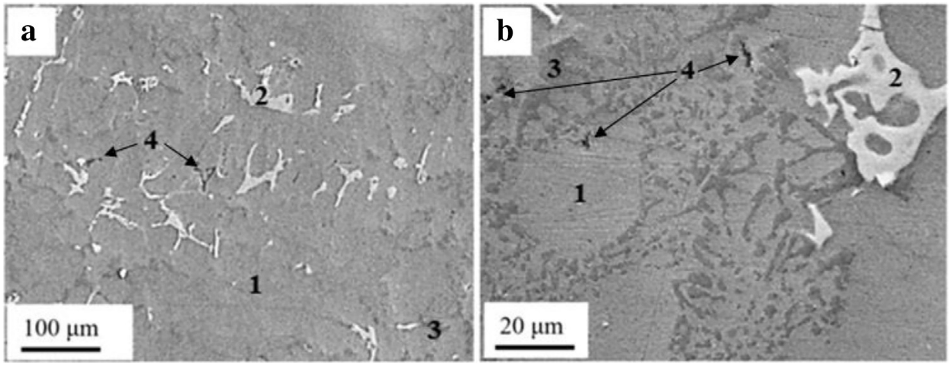

Fig. 2. Structure of Al-11Si-2Cu alloy in cast state.

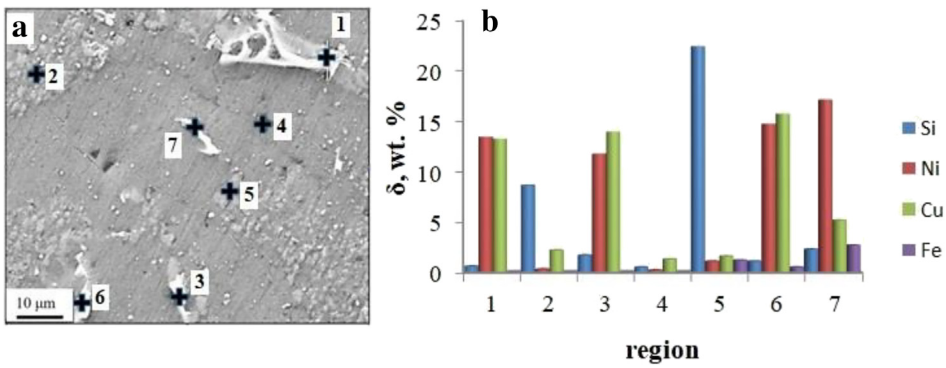

Fig. 3. Electron microscopic image of Al-11Si-2Cu alloy structure. a - regions wherein micro-X-ray spectral analysis of material's elemental composition was performed; b - results of micro-X-ray spectral analysis of sample's surface region.

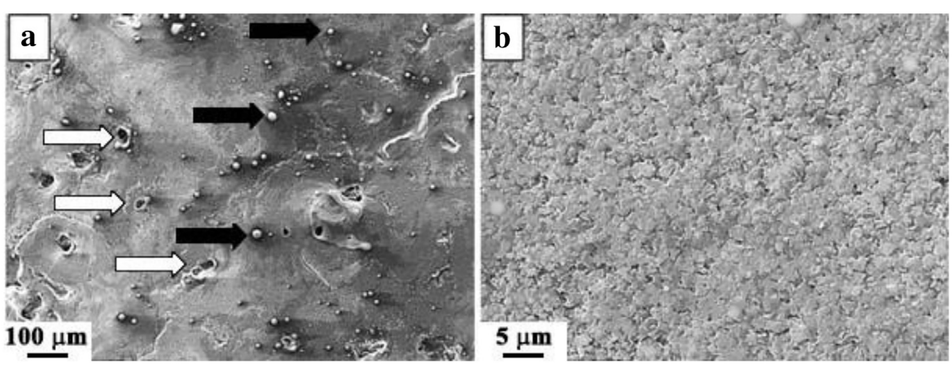

Fig. 4. Structure of Al-11Si-2Cu alloy surface subjected to ion-plasma spraying and subsequent irradiation by high intensity pulsed electron beam. In (a) microcraters are designated by white arrows, inclusions of rounded shape – by black arrows.

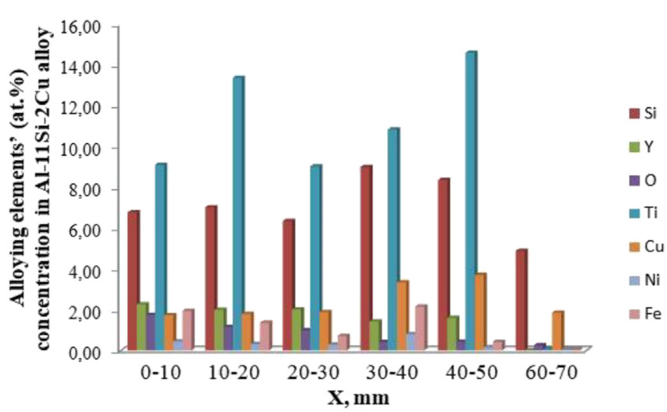

Fig. 5. Dependence of alloying elements' (at.%) concentration of Al-11Si-2Cu alloy on distance from surface of the modification.

yttrium amounts to 60–70 µm. When moving away from surface of modification the concentration of the elements was negligible. Elemental composition of modified layer depends on distance to surface of processing. As the distance from surface of processing increases the concentration of titanium, oxygen and yttrium atoms decreases the most significantly.

Phase composition of the modified layer was analyzed by the methods of transmission electron diffraction microscopy using dark-field images and the technique of microelectron diffraction patterns' interpretation [31–33]. Fig. 6a shows electron microscopic image of the layer.

Selected area electron diffraction (SAED) contains a large number of reflections of different intensities (Fig. 6, b). Interpretation of SAED permitted the detection of reflections of the following phases: silicon, α-titanium, SiY, SiTi, Cu_2YSi_2. Reflections belonging to silicon crystal lattice form the diffraction rings (Fig. 6, reflection [111] Si), that is indicative of small particles' dimensions of the phase (10–20 nm)

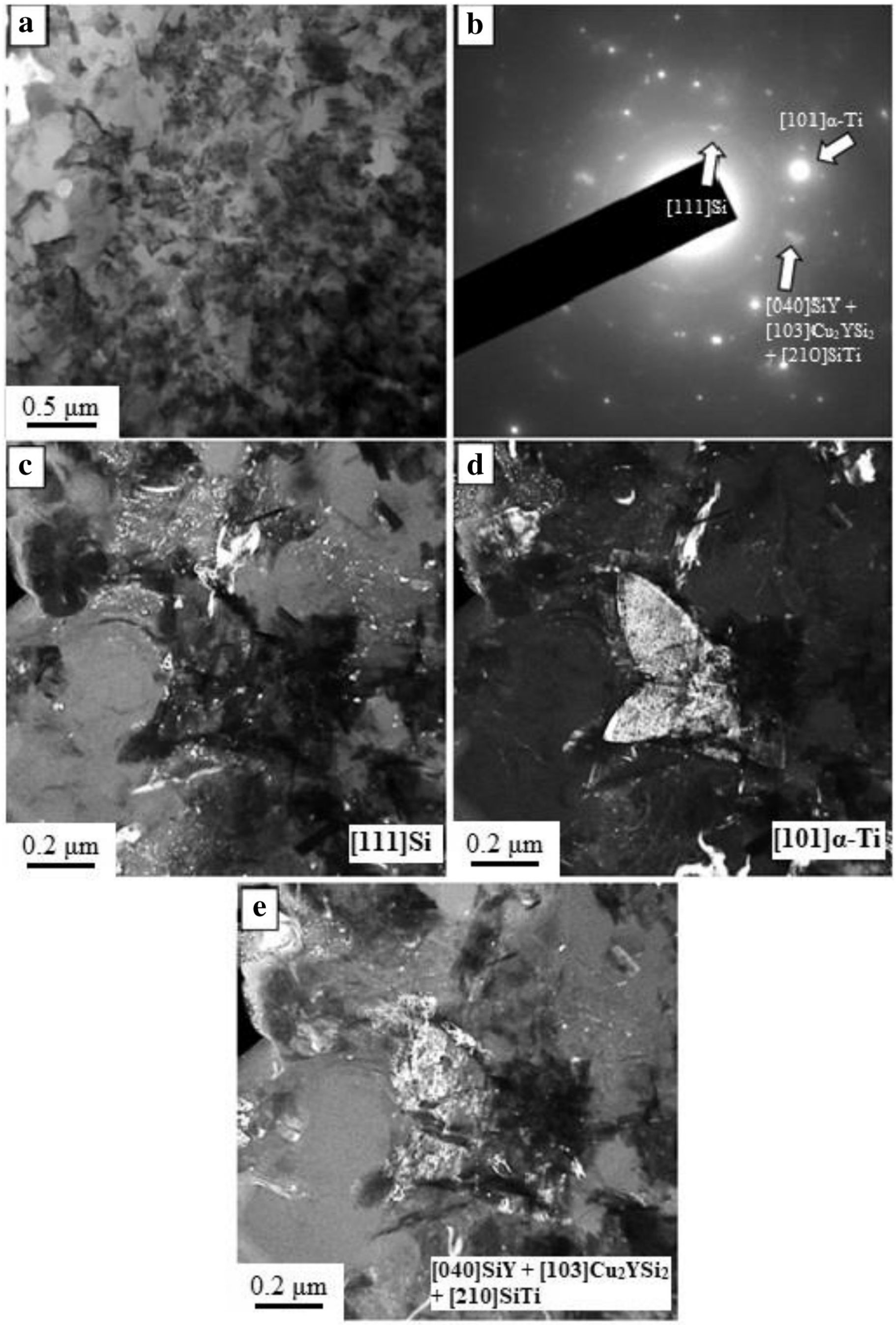

Fig. 6. Electron microscopic image of surface layer structure of Al-11Si-2Cu alloy subjected to two-stage processing; a - bright field; b - selected area electron diffraction (arrows designate reflections of obtaining the dark fields); c–e - dark fields obtained in reflections of [111] Si (c), [101] α-Ti (d), [040] SiY + [103] Cu_2YSi_2 + [210] SiTi (e).

(Fig. 6, c). The most intensive reflection of SAED (Fig. 6, b) corresponds to [101] α-Ti. Dark-field image (Fig. 6, d) of foil obtained in the reflection testifies to the fact that particles with faceting are formed by α-titanium. The most complex for interpretation is dark-field image obtained in closely located reflections of [040] SiY + [103] Cu_2YSi_2 + [210] SiTi. Analysis of microelectron diffraction pattern enables us to suggest that the reflections belong to the phases of SiY, Cu_2YSi_2 and SiTi that form the shell of α-titanium particle judging from the image presented in Fig. 6, e.

4. Conclusion

The investigations into structure, elemental and phase composition, defect substructure state of Al-11Si-2Cu alloy samples processed by ion-plasma jet and high intensity pulsed electron beam have been performed by the methods of scanning and transmission electron diffraction microscopy, micro-X-ray spectral analysis. The structure of Al-11Si-2Cu alloy in cast state demonstrates a polyphase morphologically varied character of the material wherein microcraters, solid aluminium-based compounds, intermetallide and eutectic compounds are present. As a result of two-stage processing the relief surface containing a large number of craters and rounded-shape inclusions is formed. The surface layer being formed is divided into regions whose dimensions are less than 1 µm. A two-stage processing results in structural transformation of samples' surface layer being manifested by dissolution of silicon inclusions and intermetallides of microns and submicron dimensions typical of Al-11Si-2Cu cast alloy. By means of the transmission electron diffraction microscopy and using the technique of microelectron diffraction pattern interpretation it has been possible to reveal the reflections of the following phases: silicon, α-titanium, SiY, SiTi, Cu_2YSi_2. It has been shown that the polyphase multielemental layer ≈60–70 μm thick having a submicro-nanocrystalline structure is formed as a result of two-stage processing.

Author contributions section

Dmitrii Zaguliaev: Validation, Writing-Review & Editing, Project administration. Victor Gromov: Supervision, Conceptualization. Yulia Rubannikova: Investigation, Writing-Original draft. Sergey Koovalov: Conceptualization, Formal analysis. Yurii Ivanov: Conceptualization. Denis Romanov: Methodology. Alexander Semin: Visualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

The research was supported by Russian Research Foundation grant (project № 19-79-10059).

References

[1] I. Polmear, D. StJohn, J.F. Nie, M. Qian, Light Alloys: Metallurgy of the Light Metals, fifth ed., Butterworth-Heinemann, Boston, 2017.
[2] G.K. Sigworth, The modification of Ai-Si casting alloys: important practical and theoretical aspects, Int. J. Met. 2 (2008) 19–40, https://doi.org/10.1007/BF03355425.
[3] J. Gilbert Kaufman, E.L. Rooy, Aluminum Alloy Castings: Properties, Processes, and Applications, ASM International, 2004.
[4] O. Carvalho, M. Buciumeanu, S. Madeira, D. Soares, F.S. Silva, G. Miranda, Mechanisms governing the mechanical behavior of an AlSi–CNTs–SiCp hybrid composite, Compos. Part B 90 (2016) 443–449, https://doi.org/10.1016/j.compositesb.2016.01.032.
[5] D. Linsler, F. Schröckert, M. Scherge, Influence of subsurface plastic deformation on the running-in behavior of a hypoeutectic AlSi alloy, Tribol. Int. 100 (2016) 224–230, https://doi.org/10.1016/j.triboint.2016.01.033.
[6] K. Hasan, A. Aynur, Effect of alloying elements and growth rates on microstructure and mechanical properties in the directionally solidified Al–Si–X alloys, J. Alloys Compd. 694 (2017) 145–154, https://doi.org/10.1016/j.jallcom.2016.09.199.
[7] W. Qiang, R. Xing, W. Qing-Juan, B. Yu, D. Zhong-Ze, N. Wen-Juan, W. Wei, W. Kuai-She, G. Yuan, Bonding and wear behaviors of supersonic plasma sprayed Fe-based coatings on Al-Si alloy substrate, Surf. Coat. Technol. 367 (2019) 288–301, https://doi.org/10.1016/j.surfcoat.2019.04.003.
[8] J. Min-Seok, W.M. Sung, S. Jongsoo, S. Myungmo, H. Young, Adhesion of NCF to oxidized Si wafers after oxygen plasma treatment, Microelectron. Reliab. 78 (2017) 220–226, https://doi.org/10.1016/j.microrel.2017.09.001.
[9] O.F. Farag, Comparison of the effect of plasma treatment and gamma ray irradiation on PS-Cu nanocomposite films surface, Results Phys. 9 (2018) 91–99, https://doi.org/10.1016/j.rinp.2018.02.031.
[10] W. Deqiang, W. Xiaobing, W. Rong, C. Hongyang, Surface modification of 5CrMnMo steel with continuous scanning electron beam process, Vacuum 149 (2018) 118–123, https://doi.org/10.1016/j.vacuum.2017.12.032.
[11] T. Xuewei, Y. Zhengjun, Z. Shasha, L. Jun, L. Jing, Investigation on microstructure, mechanical and tribological properties of in-situ (TiB + TiC)/Ti composite during the electron beam surface melting, Surf. Coat. Technol. 337 (2018) 418–425, https://doi.org/10.1016/j.surfcoat.2018.01.054.
[12] K. Nan, M.E.L. Mansori, A new insight on induced-tribological behaviour of hypereutectic Al-Si alloys manufactured by selective laser melting, Tribol. Int. (2019), https://doi.org/10.1016/j.triboint.2019.04.035.
[13] S. Mahanty, Surface modification of Al–Si alloy by excimer laser pulse processing, Mater. Chem. Phys. 173 (2016) 192–199, https://doi.org/10.1016/j.matchemphys.2016.02.001.
[14] A. Viswanathan, D. Sastikumar, H. Kumar, A.K. Nath, Laser processed TiC–Al13 Fe4 composite layer formation on Al-Si alloy, Opt. Lasers Eng. 50 (2012) 1321–1329, https://doi.org/10.1016/j.optlaseng.2012.02.013.
[15] V. Oliveira, S.P. Sharma, M.F.S.F. de Moura, R.D.F. Moreira, R. Vilar, Surface treatment of CFRP composites using femtosecond laser radiation, Opt. Lasers Eng. 94 (2017) 37–43, https://doi.org/10.1016/j.optlaseng.2017.02.011.
[16] J.I. Ahuir-Torres, M.A. Arenas, W. Perrie, J. de Damborenea, Influence of laser parameters in surface texturing of Ti_6Al_4V and AA2024-T3 alloys, Opt. Lasers Eng. 103 (2018) 100–109, https://doi.org/10.1016/j.optlaseng.2017.12.004.
[17] K.N. Vdovin, G.A. Dubsky, V.B. Deev, L.G. Egorova, A.A. Nefediev, E.S. Prusov, Influence of a magnetic field on structure formation during the crystallization and physicomechanical properties of aluminum alloys, Russ. J. Non-Ferrous Met. 60 (3) (2019) 247–252, https://doi.org/10.3103/S1067821219030155.
[18] V.B. Deev, E.S. Prusov, A.I. Kutsenko, Theoretical and experimental evaluation of the effectiveness of aluminum melt treatment by physical methods, Metallurgia Italiana 110 (2) (2018) 16–24.
[19] S. Valkov, P. Petrov, R. Lazarova, R. Bezdushnyi, D. Dechev, Formation and characterization of Al–Ti–Nb alloys by electron-beam surface alloying, Appl. Surf. Sci. 389 (2018) 768–774, https://doi.org/10.1016/j.apsusc.2016.07.170.
[20] X. Han, Z. Conglin, L. Peng, C. Jie, J. Yunxue, G. Qingfeng, Surface alloying of aluminum with molybdenum by high-current pulsed electron beam, Nucl. Instrum. Methods Phys. Res., Sect. B 416 (2018) 9–15, https://doi.org/10.1016/j.nimb.2017.11.028.
[21] E. Czekaj, J. Zych, Z. Kwak, A. Garbacz-Klempka, Quality index of the AlSi7Mg0.3 aluminium casting alloy depending on the heat treatment parameters, Arch. Foundry Eng. 16 (2016) 25–28, https://doi.org/10.1515/afe-2016-0043.
[22] K. Gawdzińska, K. Bryll, D. Nagolska, Influence of heat treatment on abrasive wear resistance of silumin matrix composite castings, Arch. Metall. Mater. 61 (2016) 177–182, https://doi.org/10.1515/amm-2016-0031.
[23] A.Y. Bagautdinov, E.A. Budovskikh, Y. Ivanov, V.E. Gromov, Physical Fundamentals of Electroexplosion Alloying of Metals and Alloys, Pub. center SibSIU, Novokuznetsk, 2007.
[24] V.E. Gromov, Y. Ivanov, S.E. Vorobiev, S.V. Konovalov, Fatigue of Steels Modified by High Intensity Electron Beams, Cambridge, 2015.
[25] Y. Ivanov, V.E. Gromov, S.V. Konovalov, S. Chen, Formation of structure and properties of silumin on electron-beam processing, Fundam. Probl. Mod. Mater. Sci. 4 (2018) 506–512, https://doi.org/10.25712/ASTU.1811-1416.2018.04.009.
[26] D.V. Zagulyaev, V.E. Gromov, Y. Ivanov, E.A. Petrikova, A.D. Teresov, S.V. Konovalov, A.P. Semin, Study of the surface relief, structure and phase composition of the silumin composite layer obtained by the method of electron explosion alloying by Al-Y_2O_3 system, IOP Conf. Series: J. Phys. Conf. Series 1115 (2018) 032021, , https://doi.org/10.1088/1742-6596/1115/3/032021.
[27] S. Konovalov, V. Gromov, Yu. Ivanov, Multilayer structure of Al-Si alloy after electro-explosion alloying with yttrium oxide powder, Mater. Res. Express 5 (2018) 116520, , https://doi.org/10.1088/2053-1591/aadd29.
[28] W. Rong, C. Hongyang, H. Jiayue, J. Hongfei, Effect of the continuous electron beam process treatment in the surface modification of T10 steel, Nucl. Instrum. Methods Phys. Res., Sect. B 436 (2018) 29–34, https://doi.org/10.1016/j.nimb.2018.09.004.
[29] Y. Hangyu, C. Yuyong, W. Xiaopeng, K. Fantao, Effect of beam current on microstructure, phase, grain characteristic and mechanical properties of Ti-47Al-2Cr-2Nb alloy fabricated by selective electron beam melting, J. Alloys Compd. 750 (2018) 617–625, https://doi.org/10.1016/j.jallcom.2018.03.343.
[30] V.V. Uglov, A.K. Kuleshov, E.A. Soldatenko, N.N. Koval, Y. Ivanov, A.D. Teresov, Structure, phase composition and mechanical properties of hard alloy treated by intense pulsed electron beams, Surf. Coat. Technol. 206 (2012) 2972–2976, https://doi.org/10.1016/j.surfcoat.2011.12.032.
[31] C.S.S.R. Kumar (Ed.), Transmission Electron Microscopy Characterization of Nanomaterials, Springer, New York, 2014.
[32] C. Barry Carter, B. David, Transmission Electron Microscopy, Springer International Publishing, Berlin, 2016.
[33] R.F. Egerton, Physical Principles of Electron Microscopy, Springer, Berlin, 2016.