Influence of 3D-Printing Parameters on Bimetallic Products Manufacturing Process of Cu-Fe System

K S Osipovich¹, D A Gurianov¹ and A V Chumaevsky¹

¹Institute of Strength Physics and Materials Sciences, SB RAS, 2/4 Akademicheskii Ave., Tomsk, 634055, Russian Federation

E-mail: osipovich_k@ispms.ru

Abstract. The article is devoted to selection of optimal parameters for manufacturing defect-free high quality products. Bimetallic samples with different parameters of modes were printed by the method of electron beam wire-feed additive technology. Materials of stainless steel 304 and copper C11000 selected for printing have different physical properties. Based on macrostructural research, the paper presents various external defects, which were formed in different modes of production. The study showed the effect of external defects on the formation of internal defects in experimental samples. It was noted that in addition to the basic parameters of the manufacturing process, such as beam power and wire feed rate, geometric parameters such as electron beam sweep diameter and substrate thickness play a major role. The article presents macrostructural studies of bimetallic samples of stainless steel 304 and C11000 made using different modes of production.

1. Introduction
At present, the material science of the main industries is facing the task of developing new promising materials with minimum costs and for a short period of time. Polymetallic designs with various functionality can offer unique decisions of engineering problems in comparison with monometallic structures. This approach has been previously tested for the obtaining of various polymetallic structures, ranging from titanium [1, 2], CoCrMo [3], stainless steel [4] and composites based on Inconel [5, 6]. The polymetallic structures consist mainly of two or more materials and demonstrate improvement of such properties as hardness [3, 4], wear resistance [7, 8] and even reduction of elastic modulus [8] in comparison with monometallic parts. Numerous advantages of polymetallic materials in comparison with monometallic ones are determined by unique structural, thermal and mechanical properties of the final product. Due to the flexibility in design and production, optimized properties of the two metals and more complex functions, such materials are widely used in various fields of mechanical engineering [9]: for example, in applications in which the operating conditions of the parts change depending on the location, and therefore, the requirements for the material also change depending on the location [10]. For applications in extreme environmental conditions, e.g. aerospace [11] or nuclear power, parts are required that, for example, must operate at radically different temperatures in different parts zones [12]. The aerospace industry requires a large number of dissimilar metal compounds. Copper and steel compounds are widely used in rocket engine nozzles with high thrust. However, due to its different physical and chemical properties, the manufacture of dissimilar materials from copper and steel faces the following difficulties: the high thermal
conductivity of copper quickly dissipates heat from the melt bath, which leads to difficulties in achieving the melting temperature and rapid solidification of the melt bath, resulting in high porosity when printing pure copper. Low solubility between Cu and Fe [13], a significant difference in thermal expansion coefficient and thermal conductivity between copper and steel inevitably causes high residual stresses in the material, which can lead to cracks during solidification and cooling. Hydrogen is a highly soluble element in liquid copper, which can usually form pores in the joint area [14]. Therefore, it is difficult to obtain copper and steel joints with better characteristics by conventional welding methods. The most widespread methods of manufacturing of multi-material designs are welding and related processes. At welding of dissimilar metals often use intermediate layers which divide two dissimilar metals [14], or use methods with high energy density, such as laser welding, to prevent cracking or embrittlement [14]. These problems are usually caused by insufficient solubility, crystalline lattice mismatch, change in thermal expansion and formation of thermodynamically stable brittle intermetallic phases along the gradient transition region. Additive Manufacturing (AM) is ideally suited for the structural method of manufacturing functionally graded materials. The main additive methods currently available are based on the principle of exposure to a high-power energy source: a laser or an electron beam with a material. Due to the above-mentioned difficulties in traditional manufacturing methods, several studies on laser additive technology for obtaining copper and steel compounds have been conducted [15, 16]. Interface diffusion and good adhesion between copper and H13 were achieved due to hybrid welding with ultrasonic influence of these two powders [15]. Similar metallurgical diffusion results were also obtained in the polymetallic regions between UNS C18400 copper alloy and SLM 316 L stainless steel, while massive cracks and pores were observed at the phase interface and no data on bond strength are available [16]. In addition, direct laser deposition of metal from H13 steel on a copper substrate after stretching observed cracks in the bond areas or heat-affected zones (HAZ) [16]. The researchers have demonstrated the feasibility of laser additive production of poly metals between copper and steel, but could not identify the influence of laser parameters, metallurgical mechanism of communication and the evolution of the interfacial microstructure. At present, articles on the manufacture of poly metallic products by electron-beam additive technology using copper and steel wire only appear in the public domain. In the article by X. Shu et al. [17] investigated the formation of the structure of a poly metallic sample using SS 304 steel wires and T2 copper wire by EBF3 method. The analysis showed that thin cracks up to 40 m long are formed near the contact boundary of SS 304 steel and copper T2. In this paper we will study the nature of pore and crack formation using a similar method of creating a bimetallic sample using wire filaments - SS 304 and copper M1. The purpose of the article is to establish the influence of the main parameters of the bimetallic product working process on the external defect formation. It is also necessary to establish the relationship between external defects formed during the manufacturing process and internal defects in the type of pores and cracks.

2. Materials and methods

The samples were obtained on laboratory experimental setup for additive electron-beam production of metal products. C11000 copper wire with diameter of 1 mm and 304 stainless steel wire with diameter were chosen for 3D printing of samples. The wire was deposited onto a 3 mm thick rectangular substrate made of stainless steel 304. The production of the samples consisted of depositing layers of stainless steel wire on the substrate. When 10 layers were reached, the stainless steel wire feed was replaced by a copper wire feed to apply the next 10 layers. It should be noted that the production of samples in some modes was stopped due to the accompanying defect formation of negative factors for the process.

In this way 5 experimental samples were produced, varying the main process parameters given in Table 1:
Table 1. The wire-feed electron beam additive technology deposition process parameters.

| Mode | Material | Deposition speed \( v_c \), mm/min | Current \( I_c \), mA | Beam sweep, mm | Voltage \( U \), kV |
|------|----------|--------------------------------|---------------------|----------------|-----------------|
| 1    | Fe       | 250                             | 45 \( \div \) 34    |                | 5               |
|      | Cu       | 250                             | 35 \( \div \) 34    |                |                 |
| 2    | Fe       | 250                             | 45 \( \div \) 34    |                | 5               |
|      | Cu       | 400                             | 70 \( \div \) 68    |                |                 |
| 3    | Fe       | 250                             | 80 \( \div \) 48    |                | 3               |
|      | Cu       | 440                             | 44 \( \div \) 68    |                | 30              |
| 4    | Fe       | 250                             | 90 \( \div \) 48    |                | 3               |
|      | Cu       | 440                             | 45                  |                |                 |
| 5    | Fe       | 250                             | 90 \( \div \) 48    |                | 3               |
|      | Cu       | 600                             | 80 \( \div \) 45    |                |                 |

In the vacuum chamber on a stainless steel substrate mounted on a movable three-axis water-cooled table, an electronic beam formed a melt bath, for modes 1, 2 and 3 the electron beam sweep was 5 mm in diameter, for modes 4 and 5 – 3 mm. A steel wire was fed into molten pool and a 3D printing of the vertical wall was carried out by the method of material layer-by-layer application. At the beginning of each new layer the table was returned to its original position with height adjustment. Fed rate of steel wire was 250 mm/min and did not change for different modes as it is optimised proceeding from the literary analysis and earlier received steel samples. The same situation is observed for the beam current, the value of which for the formation of steel layers changes slightly \( \pm \) 10 mA. As for parameters such as feed rate and current for the production of copper layers, they vary depending on the quality of the pilot sample, i.e. the presence of external defects.

After manufacturing the vertical wall-shaped product from the received blanks with the help of electro-erosion machine DK7750 the samples in cross-section were cut out for microstructure study. Before the microstructure examination, the samples were grinded and polished on diamond paste. To reveal the copper grain structure, etching of polished sample surfaces in 10 ml \( \text{HCl} + 1 \text{ g FeCl}_3 + 20 \text{ ml H}_2\text{O} \) solution was performed. Metallographic examination of specimen structure was carried out using confocal microscope OLYMPUS LEXT OLS4100.

3. Results and discussions

To establish the dependence between the parameters of the manufacturing process and the defects of the product, the shape of the product was chosen as a standard vertical "wall". The distance from the feeders to the product being shaped and the wire feed angle were calibrated. Fluctuations in the wire feed angle lead to asymmetrical forming and deviations in the product growth from the specified vertical axis. It is known that the main parameters in the additive manufacturing process are the current of the electron beam \( I_e \), the linear formation speed of each layers \( V_w \) and the wire feed rate \( V_{wf} \).

It was found that the optimal value of the accelerating voltage of the electron beam \( U \) is 25 kV. Additional studies have shown that increasing the accelerating voltage to 30 kV significantly stabilizes the formation of long additive products. Varying the listed parameters, as well as the reamer diameter, five experimental samples were obtained (Figure 1). It can be clearly seen that each sample has an external defect. It is necessary to note, that selection of optimum parameters of the press is necessary for formation of the experimental sample with sharp boundary of two materials contact, quantity of each layers is 10.
The first defect that occurs to a greater or lesser extent in all samples is the bending of the substrate. A temperature gradient is typical for electron beam wire-fed additive technology. Since the substrate is fixed on a three-axis water-cooled table, its temperature at the initial moment is close to the table temperature and is 20 °C. At layer-by-layer deposition of the material with an electron beam, a melt bath is formed in which the supplied wire melts. At this moment the operating temperature tends to the melting point of the used material. In this way a thermally unstable state is appears. Thus, the lack of thermal stability leads to the presence of internal stresses, which remain and accumulate in the material, because when the metal solidified, it shrinks. To avoid this defect it is necessary to strive for a state close to thermal stability. To do this, it is necessary to reduce the ratio of reamer diameter to substrate thickness, which will prevent the accumulation of residual internal stresses. In modes 3, 4 and 5, a reamer with a diameter of 3 mm was used.

The following observed defects depend on the variation of the main parameters. For better visualization of the parameters presented in the method of experiment, the change in the value of energy input [19] was used (1)

\[ E = \frac{60 \times U \times I}{1000 \times \nu}, \]

where \( U \) and \( I \) are the voltage and current of the electron beam, V and A, respectively, \( \nu \) – printing speed, m/s. When processing the results of the calculated energy input values the dependence of the energy input value on the assumed number of layers was constructed (Figure 2).

Figure 1. General view of experimental bimetallic samples made on: a – 1 mode; b – 2 mode; c – 3 mode; d – 4 mode; e – 5 mode.

Figure 2. Change in energy input values as a function of layers in the manufacture of steel-copper bimetallic samples.
The energy input value is a characteristic of the thermal gradient. The lack of thermal stability mentioned above is due to sharp thermal gradients and low thermal conductivity of the material. This leads to a curved boundary between the first layer and the substrate (Figure 3). When the top layer expands, it is limited to the much colder bottom layer, causing elastic compressive deformation. However, at elevated temperatures, the yield strength of the top layer is reduced, allowing the top layer to be compressed plastically. The cooling of the plastically compressed top layer now leads to its shrinkage, causing a bending angle in relation to the electron beam. This leads to tensile stress in the direction of growing. It is important to note that this mechanism occurs in the solid phase (does not require melting of the material).

Figure 3. Macrostructure of experimental bimetallic samples made on: a – 1 mode; b – 2 mode; c – 3 mode; d – 4 mode; e – 5 mode.

For printing with steel wire, in this case the selection of parameters was based on already known data [19]. The combination of low value of energy input (225 kJ/m) for printing with a steel wire and wire feed coefficient (0.9), realized in mode 2, allows form a product. However, even in this case, the heat input is not sufficient for the complete melting of the filament, which leads to the appearance of areas with the not melted steel wire (Figure 1 (a)). A higher energy input of 500 kJ/m allows the filament to melt completely regardless of the feed factor (0.9 or 1.3) [19]. But at the same time there is an excessive melting of previously formed layers. Thus the melting of material of formed product can be partially compensated by increased volume of filament with higher value of its feed factor. The combination of a moderate value of energy input (324 kJ/m) with a low value of filament supply coefficient (0.9), realized in mode 2, allows to form a stable product with a satisfactory appearance. When using a high filament feed factor (1.3) it is necessary to increase the energy input value to 400 kJ/m [19]. As follows from comparison of the geometrical sizes (and appearance) of products (Figure 1 (a), (b)), formed on modes 1 and 2, at the last increased value of energy input on 100 kJ/m has allowed completely to melt supplied filament without melting of earlier formed layers.

For copper wire printing, it is not possible to use the previously obtained modes when deposited to the steel substrate. This is due to the fact that already additive steel layers will play the role of the substrate. When using the values of energy input for printing with copper wire close to the values when printing with a steel wire it is difficult to produce a quality product. The energy input value (228 kJ/m) is high for printing with copper wire regardless of the feed factor (0.9 or 1.3). It leads to full melting of the feed material and its subsequent spreading on already deposited layers.
section of the sample (Figure 3 (a)) there is an uneven distribution of copper material, forming an overlap on additive steel. At carrying out of parameters optimization for printing by a copper wire on already put steel layers one more important aspect was considered. Namely, minimization of energy input. As, insufficient energy input does not allow to melt filament material in full, and excessive energy input leads to melting of a product. Incorrect selection of basic parameters leads to overheating of the material as it is removed from the substrate. This increases the thickness of the product, which is undesirable. Therefore, the average energy input for copper wire printing is 168 kJ/m. At this value, copper spillage is minimized. When printing with copper wire in modes 3 and 4, a rapid decrease in the energy input value from 180 kJ/m to 90 kJ/m results in an uneven melting of copper filament. This manifests itself in needle-like whole wire residues on the outside of the experimental sample (Figure 1 (c), (d)). Uneven melting from sharp spikes of thermal investment leads to pores in the copper area of the bimetallic piece. As it is known [20], defects by the type of pores are the place of preferential nucleation of fatigue cracks. Smooth change of heat investment values to minimum values, as in mode 5 (Figure 2), leads to the formation of a high-quality defect-free vertical "wall". In addition, it is shown that the geometric parameters of the print (for example, the diameter of the beam reamer) has no less influence on the process of product formation. If the diameter is larger, the angle of wire feed is oscillating, which leads to a deviation of the product growth from the given axis and asymmetrical formation (Figure 3 (b)). This defect is difficult to estimate when analyzing an experimental sample in longitudinal direction.

In the samples shown in the Figure 3 (c-e), the electron beam sweep diameter was 3 mm and the product formation is symmetrical. In addition, the boundaries of the melt bath can be observed, especially on the first layers. With an increase in the number of product layers, the boundaries of the melt bath are already difficult to distinguish, it is associated with thermal cycling in the growth of the product. The structure of additive steel is dendritic, the structure of copper is grain. Epitaxial growth is observed in the direction of growing experimental samples. But, in the areas formed due to the melting of copper, there were some signs of new grains originating due to the surface grain nucleation mechanism [21]. The melt pool surface can be undercooled thermally to produce surface nucleation by reduction or removal of the heat input as form of heat radiation. The boundary between the first layer of copper and the last layer of steel is similar to the mechanical mixture of these materials. The material was distributed as steel particles on the first copper layer shown in Figure 1 and 3. There is a significant distribution of iron in the first copper layer. At deposition of the second and subsequent layers, the impact with subsequent melting of steel layers, the volume fraction of steel in the copper layers decreased. The boundary zone is present in all samples and its size is within 300 – 700 µm. It should be noted that this area is free of visible defects because of the metallurgical bonding between SS 304 and C11000.

4. Conclusion
The work revealed that in the manufacture of bimetallic samples using dissimilar materials, it is necessary to minimize the values of energy input. With the minimum optimal ratio of energy input values to the filament supply coefficient, it is possible to form experimental samples without under melted and remelted areas of material. Optimization of the production parameters of the part will improve the thermal conditions of the process and get rid of the above defects. This, in turn, contributes to the absence of defects in the type of pores. In the present work the presence of pores was connected with the presence of unmelted wire, thus it was established the influence of external defects on the formation of internal defects. In addition, it was found that geometrical parameters, along with energy input, are an important characteristic of the main process parameters. Control and optimization of the above mentioned parameters allows achieve higher accuracy of product formation and surface quality.
5. References
[1] Lima D D, Mantri S A, Mikler C V, Contieri R, Yannetta C J, Campo K N, Lopes E S, Styles M J, Borkar T, Caram R, Banerjee R 2017 Mater. Des. 130 8-15
[2] Hofmann D C, Roberts S, Otis R, Kolodziejcska J, Dillon R P, Suh J, Shapiro A A, Liu Z-K, Borgonia J-P 2015 Sci. Rep. 4 5357
[3] Sahasrabudhe H, Bose S and Bandyopadhyay A 2018 Acta. Biomater 66 118–128
[4] Gualtieri T and Bandyopadhyay A. 2018 Mater. Des. 139 419–428
[5] Hong, C Gu D, Dai D, Gasser A, Weisheit A, Kelbassa I, Zhong M, Poprawe R 2013 Opt. Laser Technol. 54 98-109
[6] Carroll B E, Otis R A, Borgonia J P, Suh J O, Dillon R P, Shapiro A A, Hofmann D C, Liu Z K, Beese A M 2016 Acta Mater. 108 46-54
[7] Balla V K, Xue W, Bose S, Bandyopadhyay A 2008 Acta Biomater 4 697-706
[8] Das M, Bhattacharya K, Dittrick S A, Mandal C, Balla V K, Sampath Kumar T S, Bandyopadhyay A, Manna I 2014 J. Mech. Behav. Biomed. Mater 29 259-271
[9] Raj S V, Ghosn L J 2003 Coating Development for GRCop-84 Liners for Reusable Launch Vehicles Aided by Modeling Studies Research and Technology NASA/TM-2004-212729 35–37
[10] Mortensen A and Suresh S 1995 Proc. Int. Mat. Rev. 40 (6) 239-265
[11] Suresh S, Mortensen 1998 IOM Com. Ltd (London)
[12] Shakil M, Ahmad M, Tariq N H, Hasan B A, Akhter J I, Ahmed E, Mehmood M, Choudhry M A, Iqbal M 2014 Vacuum 110 121-126
[13] Yao C, Xu B, Zhang X, Huang J , Fu J, Wu Y 2009 Opt. Lasers Eng. 47 807-814
[14] Sun Z and Karppi R 1996 J. Mat. Proc. Technol. 59 (3) 257-267
[15] Al-Jamal O M, Hinduja S, Li L 2008 CIRP Annals 57 (1) 239-242
[16] Liu Z H, Zhang D Q, Sing S L, Chua C K, Loh L E 2014 Mater Charact 94 116-125
[17] Imran M K, Masood S H, Brandt M, Bhattacharya S, Mazumder J 2011 Sci. Eng. A 528 3342-3349
[18] Shu X, Chen G, Liu J, Zhang B, Feng J 2017 Materials Letters 213 374-377
[19] Tarasov S Yu, Filippov A V, Savchenko N L, Fortuna S V, Rubtsov V E, Kolubaev E A, Psakhie S G 2018 The Int. J. of Advanced Manufacturing Technology 99 2353–2363
[20] Solberg K and Berto F 2019 Int. J. of Fatigue 122 35-45
[21] Karimia P, Sadeghia E, Åkerfeldt P, Ålgårdhac J, Anderssona J 2018 Materials & Design 160 427-441

Acknowledgments
The work was performed according to the Government research assignment for ISPMS SB RAS, project No. III.23.2.11.