Cunha, A.; Marques, A.; Silva, F. S.; Gasik, M.; Trindade, B.; Carvalho, O.; Bartolomeu, F.


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A. Cunha, A. Marques, F.S. Silva, M. Gasik, B. Trindade, O. Carvalho, F. Bartolomeu

A novel multi-material design concept to be applied in plastic injection moulds is proposed combining the mechanical resistance of the 420 stainless steel alloy and the high thermal conductivity of copper, in a single component, fabricated in just one event by means of homemade 3D Multi-Material Laser Powder Bed Fusion equipment. The processing strategy and the interface region between both materials are analysed and discussed both from a metallurgical and mechanical point of view. The results show a good metallurgical bonding between the two materials, with a diffusion zone of about 10 µm, capable of providing mechanical interlocking, i.e. entrapment of one material in the other, creating a physical link between them. Both materials have low porosity and the pores detected present a sub-micrometre size distribution. A few pores and cracks on both the top and cross-section surfaces were detected with some tens of micrometres in size, at the interface zone. The hardness of the 420 stainless steel and copper varied from 482 to 532 HV and 99 and 116 HV, respectively.

1. Introduction

The plastic injection moulding (PIM) industry is one of the fastest-growing industries in the world due to the massive use of plastic in daily life products [1–3]. However, although this industry has numerous advantages such as high dimensional and geometric precision, repeatability, and adaptability to a wide range of materials, the costs associated with the mould and the injection machine are high [4–6]. Thus, a reduction in the cycle time of the PIM, particularly the cooling time (~ 70 % of the cycle), has been a never-ending challenge since it has a direct influence on the production costs, productivity, and quality of the parts produced [3,7,8]. The use of conformal cooling channels and high thermal conductive inserts has been one of the main solutions to achieve this objective [9–11]. Additive manufacturing processes, particularly Laser Powder Bed Fusion (LPBF), have been used in the fabrication of tools with a high geometric complexity, challenging the traditional design guidelines for cooling systems in industrial heat transfer cases, namely in the plastic injection moulding industry [10,12–14]. This technology allows the development of several innovative designs for complex cooling systems in mould inserts without geometric limitations in terms of the mould’s cavity and core [11,15].

Steel alloys are frequently used for the fabrication of plastic injection moulds since they combine the most essential characteristics required from a mould, e.g. mechanical and corrosion resistance, hardness, wear resistance, resistance to fatigue, among others [5,16–18]. In particular, 420 stainless steel (420SS) is one of the steels most used for the production of moulds for plastic, due to its high strength, hardness and corrosion properties [19,20]. However, one of the main drawbacks is its low thermal conductivity (25 W/m K), which makes it difficult to extract heat after the injection cycle [21]. Copper and its alloys have been used in mould inserts to solve this problem [9]. Pure copper, although it has a high thermal conductivity (~ 400 W/m K), is a very soft and ductile material and therefore cannot be used in the production of the mould’s core and cavity. Therefore, alloying elements have been added to copper (Cu-Be, Cu-Co-Ni-Be, Cu-Ni-Si-Cr, and Cu-Al-Ni-Fe) [22,23] to improve its mechanical properties, although substantially lowering its thermal conductivity.

In recent years, the design of multi-materials capable of presenting a
set of properties, provided by their base constituents, has been a strategy adopted in additive manufacturing. This technology can be disruptive as, by using optimised processing parameters and high solididification rates, the diffusion process can be minimised, or even suppressed, avoiding the formation of fragile and undesirable intermetallic phases [24]. Different studies about multi-material solutions produced by additive manufacturing, namely 420SS-TiN [25], 420SS-Inconel 718 [26], 420SS-300 maraging steel [27], 300 maraging steel-copper (sheet) [28], H13 steel-copper [29], 316L stainless steel-C18400 alloy [30], have been reported in the literature. However, several authors have listed difficulties in combining steel and copper alloys, due to their different physical and chemical properties (metallurgical incompatibility). The solubility between Fe and Cu is quite low, and no intermetallic phases exist in the Fe-Cu phase diagram [28,31]. Moreover, the high laser reflectivity and low absorption of copper, as well as its high thermal conductivity, make sintering difficult [28,32]. On the other hand, the significant difference in the values of the thermal expansion coefficient and thermal conductivity between these materials leads to a large deformation and residual stresses in the joint, causing solidification cracks [28,33]. The present study explores the use of a 3D Multi-Material Laser Powder Bed Fusion (3DMLPBF) system to combine two distinct materials (420 stainless steel and copper) with unique and specific properties (high mechanical resistance and high thermal conductivity) in one new single multi-functional component to improve the heat extraction of a plastic injection mould. The design followed in this study differs from the works available in the literature since the construction of the part is in full 3D, in which both materials are deposited on the same layer so that there is a continuous bonding between them.

2. Materials and methods

Multi-material 420SS-Cu parts with 2.5 × 4 × 1 mm were fabricated using homemade 3DMLPBF equipment developed at CMEMS (Center for MicroElectroMechanical Systems) of the University of Minho. 420SS (particle size in the range 15–45 µm) and 99.7 % purity Cu powders (15–45 µm) were purchased from Carpenter Additive (United Kingdom) and TLS Technik (Germany), respectively. The 3DMLPBF equipment is operationalised with a point-wisely high-design freedom software connected to an Nd: YAG laser. As illustrated in Fig. 1, it works with multiple powder layer deposition functions and vacuum cleaning procedures to avoid mixing the different powders. An argon protective atmosphere was used to avoid the formation of oxides resulting from the increase in temperature due to the interaction between the powder bed and the laser beam. The argon flow into the process chamber induces a dilution of the oxygen and impurities initially present in the atmosphere [34]. After some series of preliminary tests, a set of processing parameters was used for each material: laser power (P) of 10 and 12 W for the 420 stainless steel and Cu, respectively, scan speed (s) of 120 mm/min, hatch spacing (h) of 200 µm, layer thickness (t) of 30 µm, and laser spot size of 200 µm. A pulsed laser with a frequency of 10 Hz and duration of 0.3 ms was used.

The design consists of nine 420SS single scan tracks intercepted by two copper lines acting as heat conductors and was produced with a bidirectional scan strategy, without rotation between each layer (Fig. 2). The result was a 420SS-Cu multi-material with, approximately, 18 % of copper and 82 % of 420SS. A schematic representation of the multi-material part proposed is shown in Fig. 3. The part was built vertically upwards on a 420 stainless steel substrate, parallel to the z-direction.

The morphological and chemical analyses of 420SS and Cu regions performed individually and at their interface, were conducted by scanning electron microscopy (SEM) (Nano-SEM - FEI Nova 200) and electron dispersive spectroscopy (EDS) (EDAX - Pegasus X4M). Both secondary electron imaging (SEI) and backscattered electron imaging (BEI) modes were used for this analysis.

The Vickers hardness was evaluated with a micro-hardness tester (DuraScan of EMCO-TEST). Five indentations were made in three distinct zones of each material under applied loads of 10, 50, and 100 gf and a dwell time of 20 s. These values were selected based on previous works [27,29,30,35,36].

3. Results and discussion

The multi-material processing of metals, particularly in materials such as steel and copper alloys, is problematic due to their metallurgical incompatibility and the wide difference in physical properties [28,31]. Due to the unique characteristics of each of the materials, 420 stainless steel absorbs more energy from the laser when compared to copper, so the complete melting of the first material is significantly easier. Nevertheless, this can be compensated for by increasing the laser energy.
density to sinter copper. Decreasing the layer’s thickness, scanning speed, and/or hatch spacing, and/or increasing laser power can be decisive in this regard. This will compensate for the energy loss that is inherent to copper due to its high reflectivity and thermal conductivity [28–30]. Therefore, the processing parameters were optimised individually for each material, using a higher laser power for the sintering Cu (12 W) than for the 420SS (10 W). However, it is important to note that the previous studies were based on a 2D multi-material concept which means that only one of the materials is being solidified in each layer [28–30,37]. In this study, the two materials are solidified in the same layer (3D multi-material concept) and therefore the low thermal conductivity of the 420 stainless steel helps to retain the heat in the melt pool when the copper lines are being solidified, facilitating their sintering. The morphology of the successive tracks and their cross-section were evaluated (Fig. 4) to assess the possibility of severe keyhole formation or lack of fusion with the underlying substrate.

On the top surface, successive passes of the laser can be detected, for each of the materials. The average overlap for both materials was approximately 12 %, while between the two materials it was 18 %, ensuring a well-defined and dense interface. In the cross-section and particularly in the last sintered layer, it is possible to see that the width of the melt pool is close to twice the depth, which characterises stable melt pools [38,39]. Fig. 5 shows SEM images of the polished top and cross-section surfaces of the 3D multi-material 420SS-Cu parts produced.

The results show that the top and cross-section surfaces of the 420SS are almost free of defects. Only some irregular and circular-shaped pores, with a sub-micrometre size, are perceptible that might be originated from gas trapped during the sintering process [40–42]. Furthermore, Zhang et al. [41] have reported that for the bidirectional scan strategy, at the beginning and end of a scan track, the laser power is unstable and scan speed is gradually reduced which reduces a relatively higher laser energy and the formation of defects. Two copper lines of approximately 200 µm can be seen on the top surface, with no significant number of pores along their length. On the cross-section image, it is possible to observe a good definition of the copper lines throughout the 420 stainless steels.

The copper-related areas were melted and consolidated after the 420 stainless steel powder and therefore the consolidation of the interface region is done with the parameters associated with the most conductive material (copper). It is important to mention that when the copper powder is melted close to the surface of 420SS, the adjacent surface of the steel alloy also undergoes melting and mixes with the copper deposited. Wits et al. [43] stated that stretching the diffusion zone is advantageous as it creates a gradual transition between the two materials. This makes it possible to lower the level of residual stresses between materials due to the gradual change in physical, metallurgical, and mechanical properties, which improves the component’s performance. However, a compromise between this gradual effect between two distinct materials and the final properties of the part must be considered, since the aim is to have well-densified components with good thermal and mechanical properties.

It is also possible to observe some pores and cracks at the interface zone on both the top and cross-section surfaces in Fig. 5, with a few tens of micrometres in size. The pores can indicate one of two causes: (i) there was not enough overlap to create a bond between the 420SS and copper or (ii) due to the difference in physical properties between the two materials, the processing parameters used did not allow an effective melting. Liu et al. [30] have pointed out two main reasons for the existence of cracks at the interface between 316 L stainless steel and a copper alloy C18400, which are the incompatibility of physical properties and the infiltration of copper in the austenitic grain boundaries of the steel. The significant difference in the values of the thermal expansion coefficient and thermal conductivity between 420SS and copper leads to large deformation and residual stresses in the joint. Therefore, long cracks at the interface are caused by the warp effect, since residual stress generated from larger thermal gradients during solidification, tends to bend the consolidated layers towards the laser beam [28,30,43]. Furthermore, the tendency to form cracks is associated with the amount of copper in the melt pool. If there is a lower amount of copper, a
A dilute solution with iron is formed, and no significant stresses and consequent cracks occur in the fusion zone. On the other hand, the greater the amount of copper is, the greater the tendency to form cracks in the steel will be, due to the thermal incompatibility between the two materials (thermal conductivity, thermal expansion coefficient, ...) [30]. Moreover, the diffusion of copper resulted in the embrittlement of the austenitic grain boundaries, which can promote microcracks induced by thermal stresses [44].

The EDS results (Fig. 6) show a narrow interface region of ~10 µm wide (position 60–70 µm) between the steel (iron) and copper. Liu et al. [30] have stated that the cooling rates associated with the LPBF process are very high, the melt pool undergoes supercooling, leading the heat removal rate to exceed the heat of the fusion releasing rate. This supercooling is further amplified by the high thermal conductivity associated with copper and its alloys.

According to the Fe-Cu phase diagram, the elements Fe and Cu are immiscible, and no intermediate phases exist. However, since these elements are miscible in the liquid state, the two liquids undergo diffusion within the melt pool during the sintering of the interface. Because the cooling rates associated with the LPBF process are very high [30], the diffusion of these elements is minimised [30,43] and supersaturated Fe-Cu solid solutions may be formed at room temperature. The formation of Cu-Fe supersaturated solid solutions has been claimed concerning other far from equilibrium processing techniques such as mechanical alloying [45].

It is well known that in the zones relating to each material individually there is always some weight percentage residual of the other constituent due to the placement of different powders in the same layer and incomplete aspiration between the deposition of each material.

Fig. 7 shows the Vickers microhardness values from different regions (periphery – zone 1, centre – zone 2, and periphery – zone 3) of each material in the part. The evaluation of different zones of the sample allowed us to evaluate the uniformity of the part, as well as the influence of the scan strategy. Furthermore, as the materials under study are quite different in terms of mechanical properties, three different loads were used (10, 50, and 100 gf).

The hardness values obtained in the 420SS region (area (a) in Fig. 7) varied from 482.0 to 532.8 HV, while in the Cu region (line (1) in Fig. 7) it varied from 99.3 to 115.7 HV. The values obtained were dependent on the applied load (the higher the load the lower the hardness) due to the indentation load/size effect (ISE) [46]. On the other hand, the hardness values obtained in different zones of the same material did not vary significantly, which proves the isotropy of the part along its length. Table 1 compares the hardness values obtained in this work and the ones for the same materials processed by different techniques. Concerning the 420SS, the values obtained are higher than the ones reported for the same material in the annealed condition (247 HV) [42] or processed by metal injection moulding (MIM) (490 HV) [47]. The cooling rates of these two processes are low, leading to equilibrium structures (or close to) with relatively high grain sizes and low hardness. However, they are consistent with the values reported in the literature for as-built LPBF
Fig. 4. SEM images of the surface morphology before polishing: (a) top surface, and (b) cross-section surfaces.

Fig. 5. SEM images of the multi-material 420SS-Cu produced by LPBF.
420SS parts (500–550 HV) [27,35,36] or wrought + quenched and tempered (567 HV) ones [48]. The same is true for copper, i.e., the values obtained are higher than the ones reported for the annealed condition (57 HV) [49] or LPBF parts (65 HV) [29] but consistent with those claimed by Yu et al. [49] for wrought pure copper (107 HV). According to the authors, this value corresponded to the copper in its factory state whilst the value of 57 HV was measured after annealing in a vacuum furnace at 600 °C for 4 h, followed by cooling to room temperature in the furnace. The different hardness values measured can be explained due to the different grain sizes and the number of dislocations present in the two samples. In another work on LPBF multi-material parts consisting of 316L stainless steel and C18400 copper alloy, Liu et al. [30] reported quite a low hardness value of 74 HV for the C18400 copper alloy, which was explained by the significant amount of porosity due to inadequate melting. The high hardness values obtained for copper in the present work are the result of the low porosity of the samples produced. Moreover, although a protective atmosphere of argon was used during the manufacturing process of the samples in this study, the
hypothesis that there was some incorporation of oxygen, which may have also contributed to these high hardness values, cannot be ruled out. It is known that an increase in oxygen in the copper lattice causes an increase in the strength with a corresponding decrease in the plasticity [50].

Finally, it should be noted that the results obtained in this work suggest a good metallurgical and mechanical behaviour of the 420SS-Cu parts. Further studies focused on a deeper microstructural, mechanical, corrosion, tribological as well as thermal characterisation are being carried out with the aim of understanding the potential of this solution in-service conditions of a plastic injection mould.

4. Conclusions

For the first time, 3D multi-material consisting of 420 stainless steel and copper was produced using 3D Multi-Material Laser Powder Bed Fusion equipment. The morphological analysis showed a low number of pores or cracks in both the individual materials and their interface. The existing pores in the 420 stainless steel and copper materials are sub-micrometre in size. At the interface zone, a few pores and cracks on both the top and cross-section surfaces were also detected, with some tens of micrometres in size. This region is well-defined with a thin diffusion zone of about 10 μm. Therefore, these results, together with the low porosity content, revealed a good metallurgical bonding at the interface as well as the mechanical interlocking between the two materials. Vickers microhardness tests indicated that the hardness values varied from 482 to 523 HV and 99 to 116 HV for the 420 stainless steel and copper, respectively.

CRediT authorship contribution statement

Angela Cunha: Conceptualization, Methodology, Validation, Investigation, Writing – original draft. Ana Marques: Methodology, Investigation. Filipe Samuel Silva: Validation, Supervision. Michael Gasik: Writing – review & editing. Bruno Trindade: Investigation, Validation, Writing – review & editing. Oscar Carvalho: Methodology, Investigation, Supervision, Writing – review & editing Flávio Bartolomeu: Methodology, Writing – review & editing.

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.

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