

Fabrication of high-thickness and low surface roughness metal parts by a hybrid electrochemical manufacturing process

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ABSTRACT

Electroforming on additively manufactured templates has the potential to become a manufacturing process for making complex metal parts in a flexible and economical way. However, the main challenge of electroforming is the high surface roughness when deposition thickness increases (thickness > 100 µm), due to the high current density on edges and co-deposition of impurities while electroforming. In this work, a hybrid electrochemical manufacturing process to fabricate high-precision metal parts is proposed combining electroplating and surface levelling to achieve thick deposits (up to 1 mm) with a low surface roughness. The presented work includes a comparative study of electroforming technologies to fabricate precision metal parts on additively manufactured templates. The potential of this methodology was demonstrated by forming a copper layer on additively manufactured ABS (acrylonitrile butadiene styrene) models. The proposed hybrid electroforming method results in a simple process design, as no specific chemistry (e.g. lack of additives) is needed. Another benefit of this indirect additive manufacturing process is the reduction of the carbon footprint compared to direct printing by usage of metal salts instead of refined metals.

1. Introduction

Additive Manufacturing (AM) has gained significant interest among academia and industry for many years, due to its unlimited freedom of design and its ability to produce personalised components on-demand with a highly efficient material use (Leach and Carmignato 2020; Tuncer and Bose 2020). A wide range of applications can benefit from this technique, ranging from prototyping to production of medium to large series (Duda and Raghavan 2016).

However, to produce net-shapes high-precision metal parts by AM technology is still very challenging due to unpredictable shrinkage, residual stresses and limited process stability and robustness, hampering its use as a real precision manufacturing method (Leach and Carmignato 2020; Witvrouw et al., 2018). In addition, post-processing is often required because of the remaining porosity and the relatively high roughness of the produced parts (Ra ranging from 3 µm to 25 µm depending on the deployed AM technology) (Tuncer and Bose 2020; Leach and Carmignato 2020).

Among alternative technologies to fabricate metal parts layer by layer rather than subtractively, electroforming is an interesting candidate. Currently, electroforming is used for the manufacturing of metallic

micro-parts using high-precision molds typically fabricated by LIGA or similar processes (MacGeough et al., 2001). From this established technology, the idea emerged to replace the expensive molds fabricated by clean-room technologies by more affordable methods, such as low-cost additive manufacturing methods. Recent advancements show that high-precision polymer models can indeed be manufactured using novel AM approaches (Ge et al., 2020) and even by more traditional AM methods for polymers, such as Fused Deposition Modeling (FDM) ensuring reasonable precision products (Msalleem et al., 2020).

Recently, the feasibility of this approach using thin depositions of metals by electroplating on 3D printed models was demonstrated by several authors. Cook et al., (2018) used 3D printed molds by digital light processing stereolithography to electroform by current pulse deposition traveling-wave tube circuits of typical thickness of 40 µm. Lazarus et al., (2019) studied the possibility of using electroplating to create conductive circuits on 3D printed parts. Kim et al., (2019) successfully achieved excellent coatings on horn antennas by electroplating of 3D printed parts with conductive filaments. Ghazali et al., (2018) metalized a 3D printed waveguide structure for the design of microwave passive components by first sputtering a seed layer followed by copper electroplating. Angel et al., (2018) tried to incorporate a non-conductive filament and a

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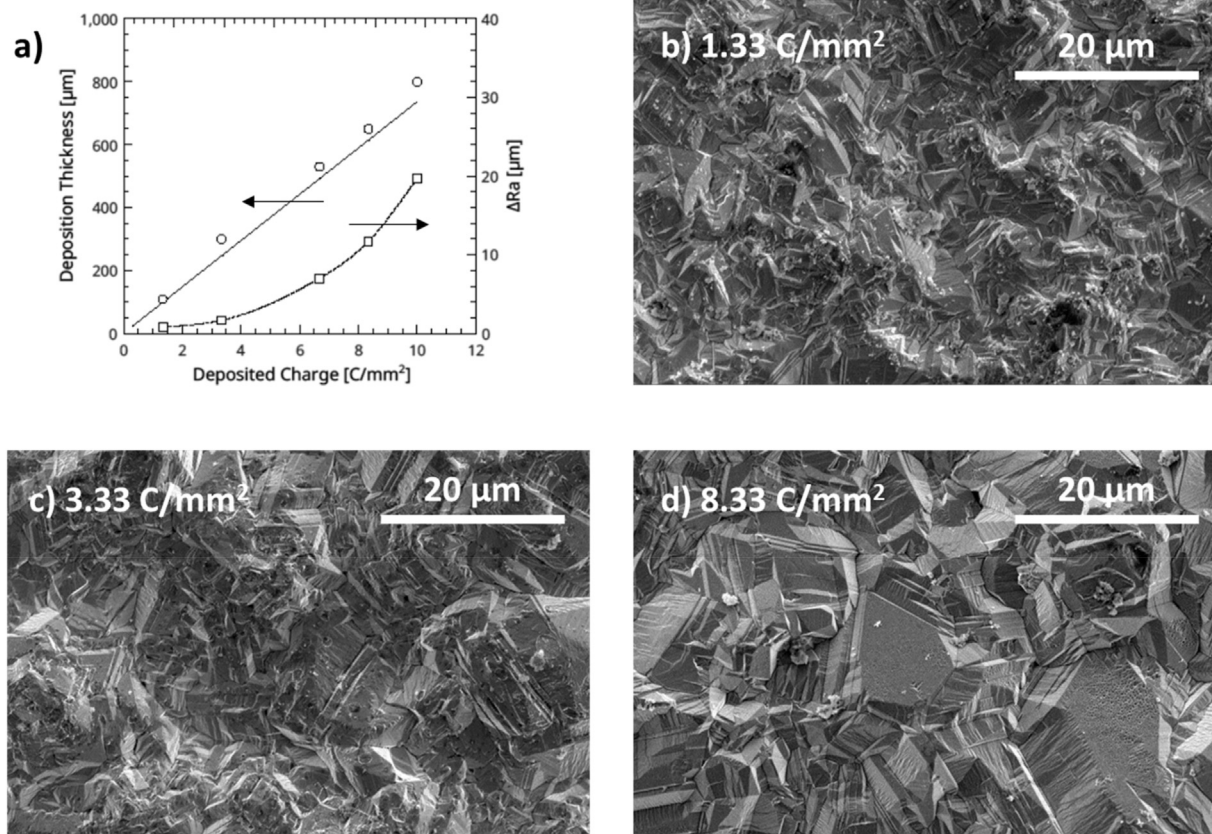


Fig. 1. a) The effect of varying the deposited charge on the thickness and surface roughness (Ra) of the deposits at a constant current density of 5 mA/cm² b) to d) SEM micrographs of the coatings obtained at a different quantity of electricity of 1.33, 3.33 and 8.33 C/mm² at a constant current density of 5 mA/Cm².

conductive filament to 3D print a part followed by a selectively electroplating onto the printed part to improve the electrical properties of the conductive filament. [Kunieda et al., \(1998\)](#), produced metal parts by selective electrodeposition using an electrolyte jet. The formed metal part was then mechanically lapped using an abrasive lapping machine to remove the irregularities on the deposits.

When deploying electroforming as an efficient manufacturing process for high-precision metal parts, the metal deposition step needs 1) to have a high degree of thickness control, and 2) provide a low surface roughness, in order to minimize subsequent post-processing steps. Electroplating is a suitable process when only thin-deposited layers (thickness up to 100 μm) are required. However, the main challenge of electroforming is the high surface roughness when deposition thickness increases (thickness > 100 μm), due to the high current density on edges and co-deposition of impurities while electroforming ([Jung et al., 2015](#)). Hence, surface roughness and dimensional accuracy are primary concerns in electroforming where a thick coating (higher than 100 μm up to typically a millimeter) is required ([Qu et al., 1997](#)). Both aspects are of high importance when using electroforming on 3D printed mandrels to form high-precision metal parts.

In the case of low thickness electroplating (up to typically 100 μm) several methods exist to address the issue of growing surface roughness with increased deposit thickness.

The most common approach is the use of additives in the plating bath to improve the surface quality and properties of the deposits ([Zhu et al., 2008](#); [Cook et al., 2018](#)). However, the additives need to be optimised and developed specifically to each deposition bath and it is not straight forward to monitor the additive concentration during a long electroforming period, making process control challenging.

Another approach is to use mechanical polishing with hard particles

to diminish the irregularities on the deposit surface, such as pinholes and nodules, by polishing the part surface during the electrodeposition process ([Zhu et al., 2008](#); [Cook et al., 2018](#)). Although improved surface finish can be obtained, such mechanical approaches are less appropriate for high-precision parts as they alter the dimensional accuracy of the produced parts. It was also reported that the use of galvanostatic pulse-reverse electrodeposition results in an improved surface finish of the deposits compared with surface finish obtained by direct current deposition ([Ghazali et al., 2018](#); [Jung et al., 2015](#)). Although all these approaches are very successful for creating high-quality depositions up to typically 100 μm, their applications for higher thickness deposits remain to be explored.

In this paper, a hybrid electrochemical manufacturing process to achieve high-precision metal parts is proposed combining electroplating and surface levelling to achieve thick deposits (up to 1 mm) with low surface roughness. The presented work includes a comparative study of electroforming technologies to fabricate precision metal parts on additively manufactured templates. Copper was selected for the hybrid electroforming-surface levelling process since it is cost-effective and well-established as a plating metal. In addition, among metals, copper has an excellent electrical conductivity, which is favorable in applications requiring high electrical conductivity, such as electronic devices.

The proposed hybrid electroforming method results in a much simpler process design, as there is no need to develop specific additives. An additional advantage of forming indirectly metal parts, compared to direct printing, is the usage of metal salts (here: copper sulphate) instead of pure metal (here: copper), which reduces the carbon footprint of the designed process by eliminating refining process steps from metal salts to solid metals ([Elsentriecy et al., 2015](#); [Pitt and Wadsworth 1981](#); [Giurco and Petrie 2007](#)).

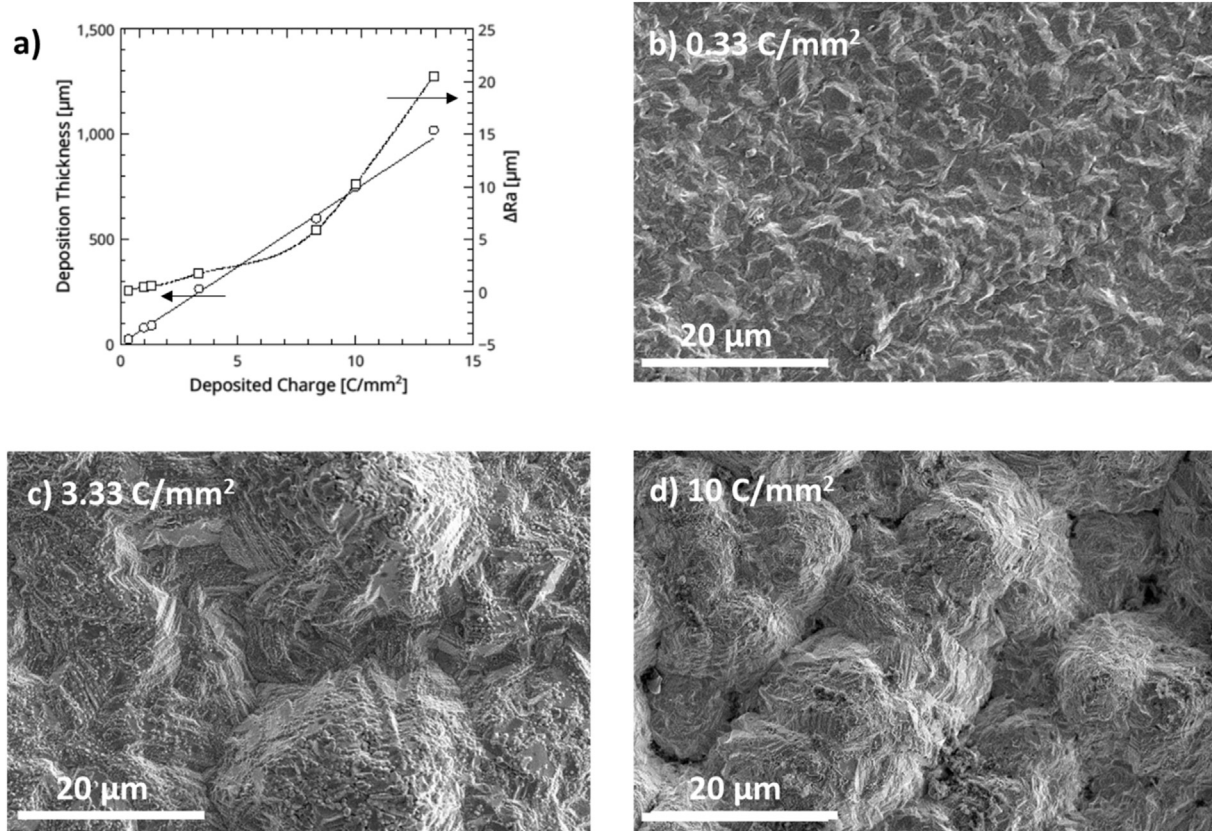


Fig. 2. a) The effect of the deposited charge on the thickness and surface roughness (Ra) of the deposits for a constant potential of -150mV vs. Cu b) to d) SEM micrographs of the coatings obtained at different quantity of electricity of 0.33, 3.33 and 10 C/mm^2 at a potential of -150 mV .

2. Material and methods

2.1. Electroplating of copper substrates

Copper electroplating was performed in a solution containing 1 M H_2SO_4 (98.08%, J.T.Baker) and 0.5 M $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ (98%, Sigma-Aldrich) at room temperature (23°C). The working electrode (WE) was a 110-annealed copper plate (McMaster-Carr) of $10 \times 50\text{ mm}^2$ and 0.8 mm thickness with an exposed area of $10 \times 15\text{ mm}^2$.

The counter electrode (CE) was a cylindrical copper sheet surrounding the working electrode to ensure a similar distance between the WE and CE (about 4 cm). A 1 mm diameter copper wire was deployed as pseudo-reference electrode (RE) located near the working electrode. Experiments were conducted using a SRS EC301, Stanford Research Systems, potentiostat/galvanostat.

The as-deposited layers were characterized by SEM (Hitachi, S-3400N), and surface roughness R_a (arithmetical mean deviation) of the electrodeposits were measured by a 3D confocal laser microscope (Olympus OLS4000).

2.2. 3D printing of ABS templates and subsequent electroforming

Acrylonitrile butadiene styrene (ABS) templates were manufactured by an Ultimaker 2+ fused filament fabrication printer using ABS filaments followed by copper electroless plating (adapted from (Glenn O and Hajdu, 1990; Qi et al., 2020)) to make the exposed surfaces conductive to enable the electrodeposition afterwards. The ABS template was subsequently dissolved in acetone to obtain the final electroformed part.

3. Results

In a first step, three electro-forming strategies on copper substrates

were compared: galvanostatic, potentiostatic, and the proposed hybrid electroforming-surface levelling process. For all methods, the thickness Δt , the increase in roughness ΔR_a (the difference between roughness of deposit and roughness of the initial substrate) and morphology of the obtained deposits in function of deposited charge were investigated. The deposit thickness, ranging up to $800\text{ }\mu\text{m}$, was compared to the calculated thickness according to Faraday's law (Eq. (1)) for a given deposition charge Q (assuming 100% of the charge was used to form the deposit):

$$\Delta t = \frac{QA}{2\rho FS} \quad (1)$$

Where A is the atomic weight of copper (63.55 g/mol), F is the Faraday constant ($96\,485\text{ C/mol}$), S is the surface area of the sample (1.5 cm^2), and ρ is the copper density (8.96 g/cm^3).

3.1. Galvanostatic mode

Electroforming in galvanostatic mode is a promising approach for electroforming on additively manufactured parts. According to equation (1), thickness of the fabricated part can be controlled by electroforming time (at a constant current, the charge (Q) is directly proportional to the deposition time). A constant current density of 5 mA/cm^2 was used to keep the deposition surface roughness low (Kazuo Kondo et al., 2014).

Fig. 1a shows the deposition thickness Δt and increase of roughness ΔR_a in function of deposition charge at a constant current of 5 mA/cm^2 . It can be seen from Fig. 1a that by increasing the deposited charge, both thickness and surface roughness of the deposit increase.

Fig. 1b-d shows the SEM micrographs of the deposits for various charges. It is observed that as the deposit charge increases, defects and multifaceted structures (V-shape grains) increase, resulting in a significant increase in roughness, as confirmed by Fig. 1a.

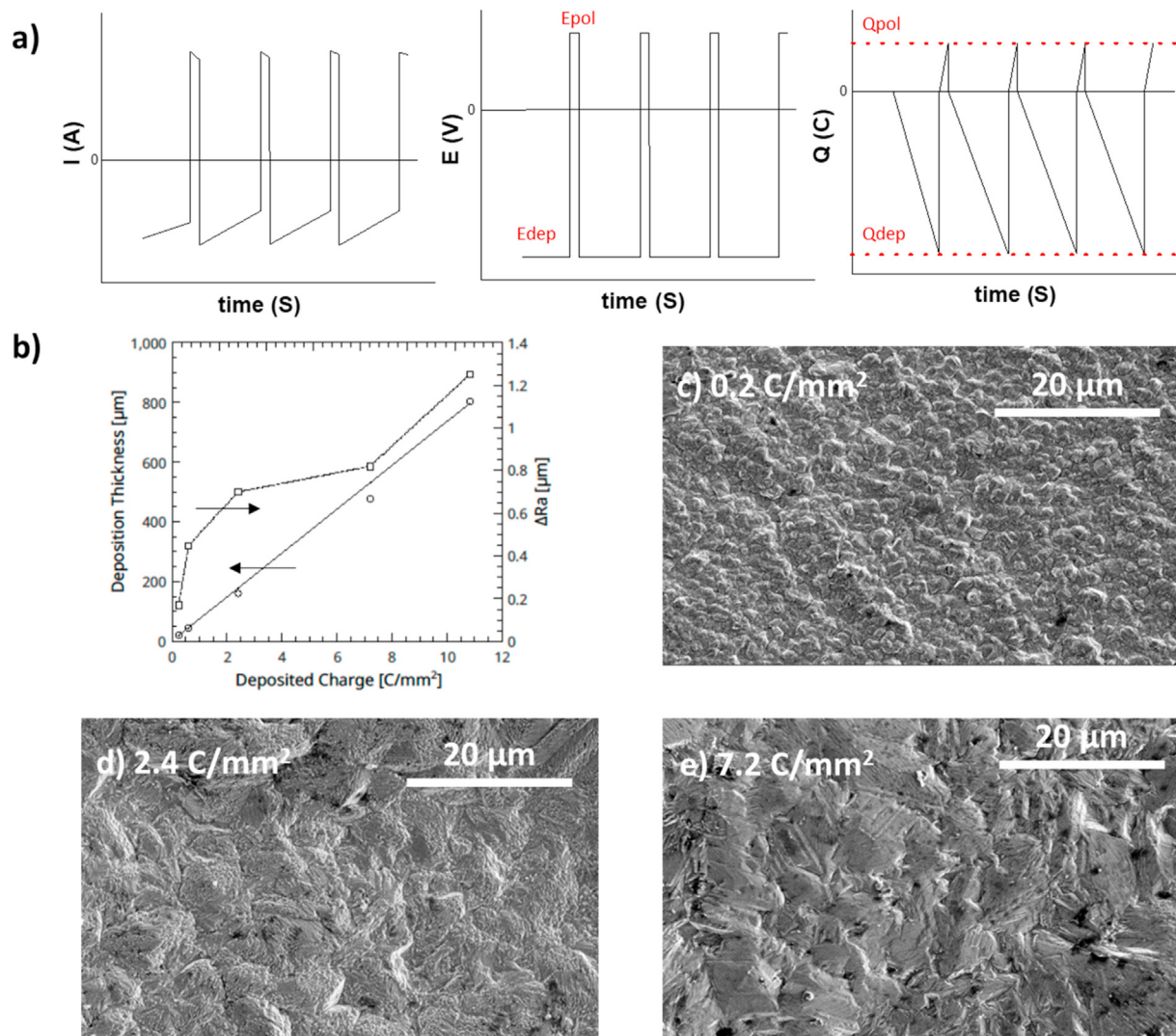


Fig. 3. a) Current, potential and charge curves associated with the pulsed process ($E_{dep} = -150 \text{ mV vs Cu}$, $E_{pol} = +100 \text{ mV vs Cu}$, Q_{dep} is the charge required to form a thickness of $1 \mu\text{m}$ and Q_{pol} is a defined percentage of Q_{dep}) b) The effect of varying the deposited charge on the thickness and surface roughness in pulse deposition mode where 10% of the deposition charge is used to determine the polishing charge c) to e) SEM micrographs of the pulse plated coatings obtained at the different total quantity of electricity of 0.2, 2.4 and $7.2 \text{ C}/\text{mm}^2$.

3.2. Potentiostatic mode

In potentiostatic mode, a constant (cathodic) potential of -150 mV vs. Cu was used. This potential was chosen to keep hydrogen evolution low to avoid negatively affecting the surface roughness obtained in the deposition process (Zhu et al., 2008).

Fig. 2a shows the deposition thickness and increase of roughness ΔR_a in function of the total charge (obtained by integrating the current signal) at a potential of -150 mV vs. Cu . A good agreement between equation (1) and the measurements can be observed, showing that the thickness of the deposit can be accurately controlled in the potentiostatic mode.

Note that up to a charge of about $8 \text{ C}/\text{mm}^2$, there is a slight increase in ΔR_a , which is followed by a remarkable rise. SEM micrographs (Fig. 2 b to d) provide further insights about the reason for this increase. As the formation of larger copper grains is more energy favorable than small ones, the formation of larger copper grains with clear boundaries can be observed. As the coating thickness increases, more surface defects have been observed. In fact, small initial defects serve as preferential nucleation sites for subsequent deposits, leading to an increase in the roughness progressively.

3.3. Hybrid electroforming-surface levelling

Both galvanostatic and potentiostatic deposition modes allowed for the control of deposition thickness but resulted in increased surface roughness as the deposit thickness grows. To address this issue, a hybrid approach was investigated combining electroplating and surface levelling using pulsed signals. As electroplating under constant potential provided more promising results than under constant current, surface levelling will be combined with potentiostatic electrodeposition.

In this approach, first, a (cathodic) pulse of -150 mV vs. Cu is applied. The pulse is applied until a deposit charge corresponding to one-micron deposit thickness is obtained (a charge of $41 \text{ mC}/\text{mm}^2$, according to Equation (1)). In the present work, this charge will be referred as the deposition charge Q_{dep} . This pulse is followed by a second (anodic) pulse at $+100 \text{ mV vs. Cu}$. At this potential, surface levelling of the deposit takes place. The duration of this pulse is chosen to be 10% of the deposition charge passing through the electrochemical circuit (i.e., $4.1 \text{ mC}/\text{mm}^2$). This charge will be referred as the polishing charge Q_{pol} (Fig. 3a).

The strategy of combining electrodeposition followed by surface levelling offers two benefits. First, during the polishing pulse, defects and

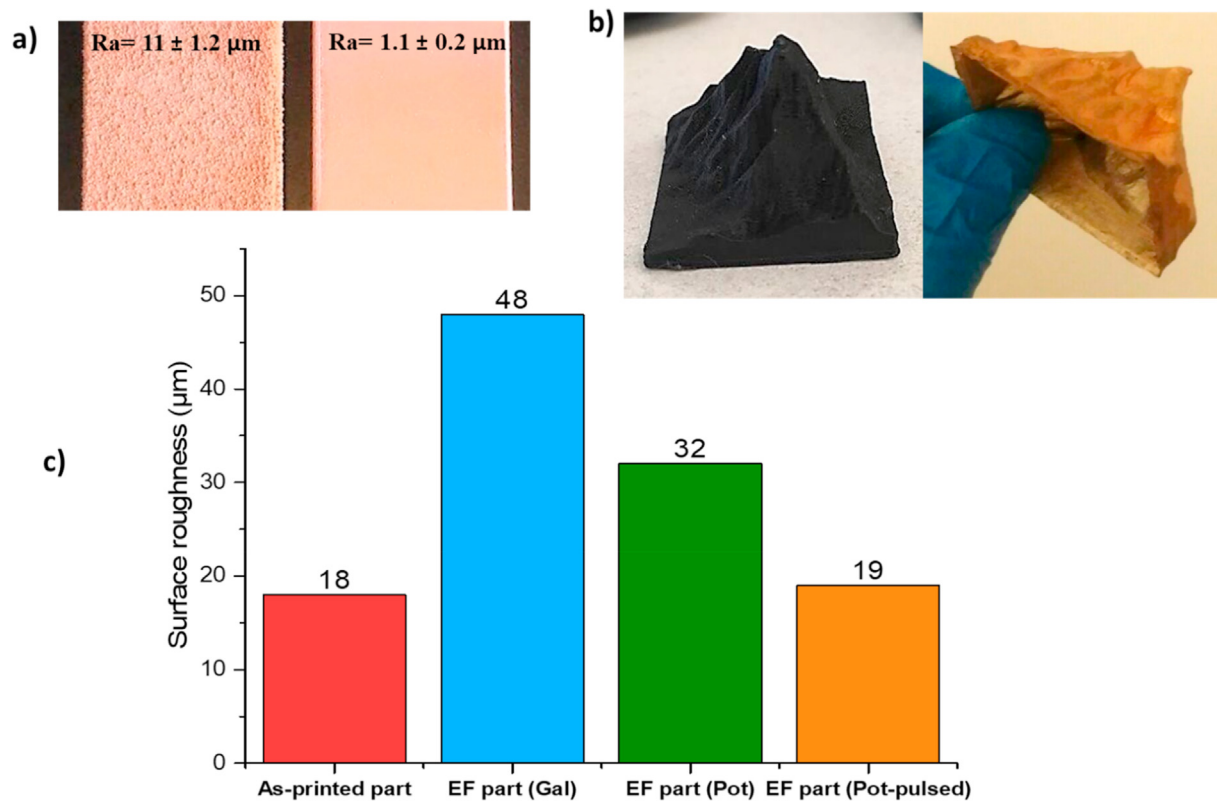


Fig. 4. a) Copper deposits obtained by potentiostatic and pulse-potentiostatic plating modes from left to right, for the same deposition charge of 10 C/mm² (thickness of 770 μm) b) Copper electroformed AM part (view from the inside showing a hollow part) c) Surface roughness comparison of electroformed parts (EF) created by different electroplating modes (Gal: galvanostatic, Pot: potentiostatic, Pot-pulsed: potentiostatic pulsed plating).

asperities are eliminated, avoiding their propagation as the coating is growing. Second, the coating thickness can be controlled by controlling the number of pulses, as each pulse is controlled to last for a given charge, rather than a given time. Indeed, keeping the pulse duration constant (as in traditional pulse electroplating) will not result in layers of constant thickness. The electrochemical state of the surface is changing as the part grows thicker and consequently the current density will not remain identical for fixed over-potentials. Beside the surface state, the deposition bath state will in general change too, again resulting in different currents for fixed potentials. Working at constant charge rather than constant time eliminates both variations in the process.

Fig. 3b shows the thickness of the deposit in the function of the deposition charge using a 10% polishing charge (note that in this case, the polishing charge has to be taken into account in equation (1), since material removal takes place during the polishing). An excellent agreement with Equation (1) can be seen, showing that a very accurate deposition thickness control can be achieved. Similar results were achieved in the case of 1% and 30% polishing charges.

As shown in Fig. 3b, the deposit surface roughness is significantly lower than that of the two previous modes. Only a surface roughness (Ra) increase of about 1 μm is observed over the full deposition process of about 900 μm thickness. This is one order of magnitude better than under constant current or potential deposition. For a 1% polishing charge, higher surface roughness are obtained (similar to the two other deposition modes). Using a 30% polishing charge, a further reduction of surface roughness increase can be obtained (about 75% reduction) but with at the cost of a significantly longer process time.

Fig. 3c–e shows the deposit morphology and confirm the significantly smoother surface finish compared to the other two deposition modes for the same deposition thicknesses. Grain growth occurs and no sharp edges are present due to the elimination of defects during the anodic pulse.

3.4. Electroforming on a 3D printed ABS model

Fig. 4a exhibits the notable difference in the surface quality of the deposits obtained by potentiostatic mode with an applied potential of -150 mV (left) and pulsed deposition with -150 mV as the cathodic and $+100 \text{ mV}$ as anodic potentials (right) for the same total deposited charge of 10 C/mm² (corresponding to a deposition thickness of 770 μm). Surface roughness (R_a) of the potentiostatic and pulsed deposition coatings were measured to be $11 \pm 1.2 \mu\text{m}$ and $1.1 \pm 0.2 \mu\text{m}$, respectively.

Fig. 4b shows an example of a copper part manufactured by electroforming on a 3D printed ABS template (mountain geometry with intrinsic irregular surface). The printed template surface was made conductive by electroless copper deposition. After copper electroforming by pulse plating, the printed model was dissolved in acetone. The thickness uniformity of the fabricated part ($765 \pm 10 \mu\text{m}$) was confirmed by measurements with a point-micrometer (Mitutoyo) on various locations. Surface roughness (R_a) of the 3D plated part was also measured at different locations over the entire mountain and the average roughness has been reported to be $19 \pm 0.4 \mu\text{m}$. It has been noted that the initial surface roughness of the ABS printed part was about $18 \pm 0.8 \mu\text{m}$, suggesting that the surface roughness of the 3D part has only increased less than 1 μm after the pulse-electroforming.

As shown in Fig. 4c, the hybrid electroforming-surface levelling process resulted in very similar surface roughness as the initial printed template (an increase of about one micron in surface roughness), confirming that the pulse-plating electrodeposition only adds very few surface defects. Applying galvanostatic and potentiostatic electrodeposition on the same substrate resulted in surface roughness increase of about 30 μm and 14 μm , respectively.

4. Conclusion

In this present study, a hybrid electroforming-surface levelling process was proposed for the fabrication of high-precision metal parts. The results showed that this hybrid manufacturing process allowed for the formation of thick deposits (up to about 1 mm) with a low surface roughness (less than 1.2 μm). A comparative study was conducted on the copper plates evaluating galvanostatic, potentiostatic and the developed hybrid electroforming-surface levelling deposition processes on achievable thickness, surface finish (roughness) and morphology. It was observed that both thickness and surface roughness of the deposits increase as the deposit charge increases for three different electroforming methods. However, in a case of hybrid electroforming process, surface roughness (Ra) of the deposit increased only 1 μm after achieving 900 μm thickness due to the surface leveling effect during the anodic pulses. Furthermore, surface morphology of the hybrid electroformed part confirmed a smoother surface finish as compared to the other two methods.

The potential of the hybrid methodology was demonstrated by forming free-standing copper parts on additively manufactured ABS models, showing only a slight increase in surface roughness (about 1 μm) as compared to a rise of 30 and 14 μm with respectively the galvanostatic and potentiostatic methods.

The proposed hybrid electroforming method resulted in simple process design, as no specific chemistry (e.g. lack of additives) is needed. Another benefit of this indirect additive manufacturing process is the reduction of the carbon footprint compared to direct printing by usage of metal salts instead of pure metals eliminating refining process steps from metal salt to solid metal.

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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