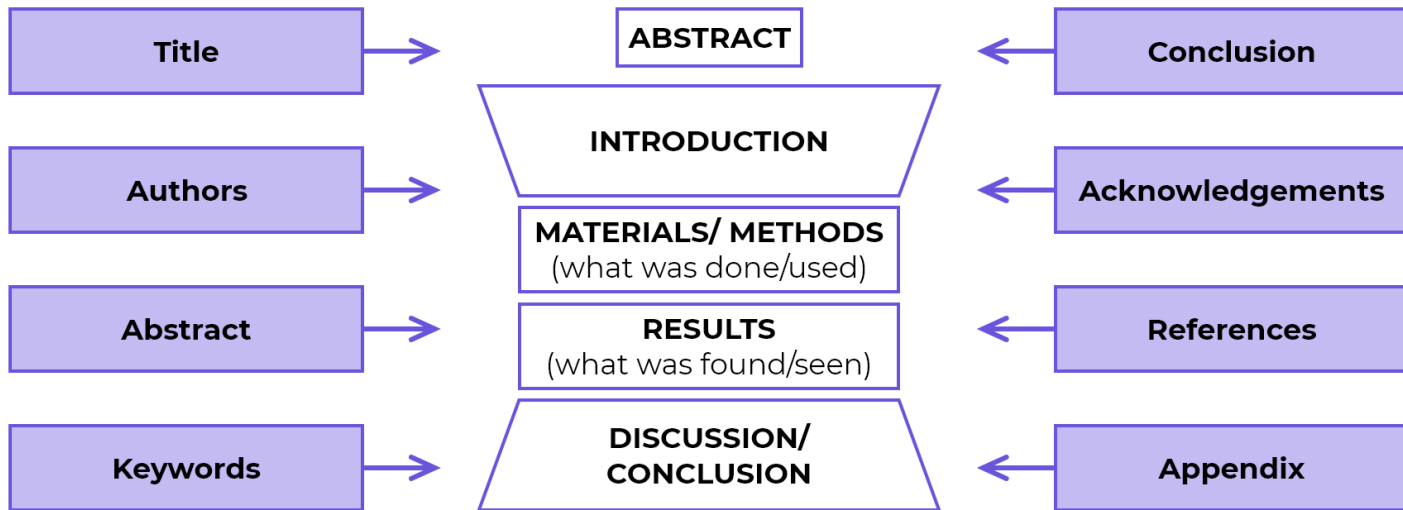


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Full Length Article

Towards digital metal additive manufacturing via high-temperature drop-on-demand jetting

Marco Simonelli^{a,*}, Nesma Aboulkhair^a, Mircea Rasa^b, Mark East^a, Chris Tuck^a, Ricky Wildman^a, Otto Salomons^b, Richard Hague^a

^a Centre for Additive Manufacturing, The University of Nottingham, UK

^b Océ – A Canon Company, the Netherlands

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ABSTRACT

Drop-on-demand jetting of metals offers a fully digital manufacturing approach to surpass the limitations of the current generation powder-based additive manufacturing technologies. However, research on this topic has been restricted mainly to near-net shaping of relatively low melting temperature metals. Here it is proposed a novel approach to jet molten metals at high-temperatures ($> 1000\text{ }^{\circ}\text{C}$) to enable the direct digital additive fabrication of micro- to macro-scale objects. The technique used in our research – “MetalJet” - is discussed by studying the ejection and the deposition of two example metals, tin and silver. The applicability of this new technology to additive manufacturing is evaluated through the study of the interface formed between the droplets and the substrate, the inter-droplets bonding, the microstructure and the geometrical fidelity of the printed objects. The research shows that the integrity of the samples (in terms of density as well as metallurgy) varies dramatically in the two investigated materials due to the different conditions that are required to melt the interface of the stacked droplets. Nevertheless the research shows that by a careful choice of the jetting strategy and sintering treatments 3D structures of various complexity can be formed. This research paves the way towards the next generation metal additive manufacturing where various printing resolutions and multi-material capabilities could be used to obtain functional components for applications in printed electronics, medicine and the automotive sectors.

1. Introduction

Mirroring the Drop-on-Demand (DoD) benefits of conventional polymeric 2D and 3D ink-jetting, DoD jetting of fully melted liquid metallics conceptually gives a promising approach for the production of both 2D and 3D constructs. The attractiveness of DoD liquid metal “3D printing” lies within its simple workflow, as structures can be created through the digitally-driven, on-demand deposition of discrete volumes of metals, as opposed to the complex handling of metallic powders intrinsic to all other conventional powder-bed fusion, binder-jetting or powder-feed-based Additive Manufacturing (AM) techniques [1]. Additionally, DoD jetting-based techniques are inherently scalable and give the newly-offered potential to shape functional parts made of dissimilar materials [2,3], that is in stark contrast to conventional single-material metallic AM systems. However, to date, both research and industrial implementation of metallic DoD jetting has been confined to relatively low-temperature ($< 700\text{ }^{\circ}\text{C}$) alloys in large droplets ($> 200\text{ }\mu\text{m}$, typically $> 500\text{ }\mu\text{m}$), leading to near-net structures

requiring post-process machining rather than net-shapes that can be used directly [4–11]. Key to DoD liquid metal 3D printing is the generation and control of a stable stream of metallic droplets. Various jetting techniques have been presented in the literature. Ejection through piezoelectric actuation, perhaps the most common form of actuation, is limited by the operating temperature of the piezoelectric crystal and therefore has only been used for printing particulate-loaded carrier inks, where the carrier ink is evaporated post deposition and the particulates (which are $\sim 1\text{ }\mu\text{m}$ in size) are then furnace processed to sinter them together [12,13], low melting point metals (tin, gallium, indium and their alloys) and other eutectics [4,5,14,15]. The use of plunger rods to shield the piezoelectric crystals from the molten metal has also been proposed to extend the number of printable metals by piezoelectric actuation resulting in the successful jetting of metals with relatively higher melting point such as aluminum alloys [16]. By having no moving parts in direct contact with the molten metal but inert gas, pneumatic actuation has been shown to overcome the temperature limitation of the piezoelectric ejection [7,17]. Pneumatic actuation is

2. Materials and methods

2.1. Materials

The printing materials were Sn and Ag grade 5 N supplied by ESPI Metals in the form of shots and rods, respectively. The substrates used for printing were Cu plates (99.9%) supplied by Goodfellow, UK except for the case of 1) the tracks printed for electrical resistivity measurement, where Cu plates were coated by a commercial Cu ink solution (Promethean Particles Ltd, UK) and 2) the Ag samples printed for sintering, where Al_2O_3 substrates were used (CSC Ceramics, UK). For sintering treatments, Al_2O_3 substrates were used as Ag and Cu form an eutectic alloy at approx. 780 °C so any sintering treatments above this temperature would cause a collapse and dissolution of the entire structure. In addition Al_2O_3 was also chosen as it generally accepted that ionocovalent structures (where oxygen ions form a hexagonal close-packed structure with aluminium ions filling two-thirds of the octahedral interstices) might facilitate diffusive bonding with metals that possess great affinity to oxygen.

2.2. Jetting process

Fig. 1(a) shows the working principle of the MetalJet technology used in this research [22].

Under an argon protective atmosphere, desired metals (Sn or Ag in the present study) are melted in distinct cartridges by induction heating. Each cartridge's ejecting chamber is surrounded by permanent

remaining tail retracts into the nozzle ready for a new jetting sequence. As the meniscus in the nozzle varies depending on the jetting temperature, wettability and impurities, tuning the actuation pulse is essential at the beginning of each printing experiment. During this initial phase, the amplitude of the current pulse was typically varied between 20 and 120A until the meniscus head of the liquid metal reached the tip of the nozzle with a typical velocity of 1 m/s. Meniscus with lower inertia protrude outside of the orifice but never detach from the nozzle. Adjusting the pulse duration, it was then possible to control the volume of jet (and therefore that of the droplet) as well as the length of its tail. The jetting pulse shape was thus optimized to control the break off of the jet tail and to allow molten metal to detach without secondary satellites. As the jet snaps off, it quickly shapes into a droplet by surface tension with the remaining tail being retracted into the orifice in the equilibrium position by capillary action. At this stage of the research, the parameters vary for each jetting and printing experiment. Research is currently ongoing [33] to establish the exact physics of the jetting process (particularly at high temperatures above 1000 °C) and establish general jetting protocols. The jetting process (droplet formation, droplet speed, radius and trajectory) was monitored by a JetXpert drop-watcher, which enabled droplet stroboscopic backlighting synchronous to the firing of the print-head.

2.3. Tuning of the jetting process

To assess the typical printing accuracy of the process, the droplet jetting stability at an ejection rate of 500 Hz was assessed by

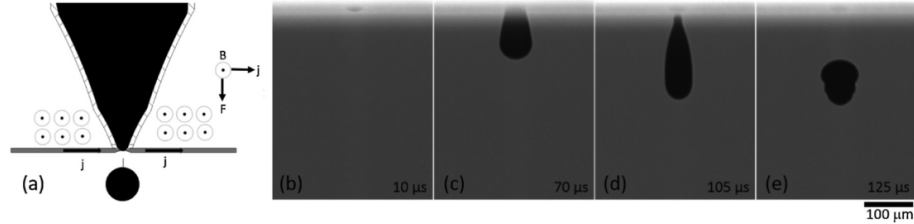


Fig. 1. The MetalJet ejection mechanism. (a) Jetting by magnetohydrodynamics (MHD): the directions of the current density j and the Lorentz force, F , acting on the metal are indicated in the diagram by black arrows. The direction of the perpendicular magnetic field B (pointing out of the screen) is indicated by the concentric circles. As an example, snapshots of the formation of Ag droplets are shown after (b) $10\ \mu\text{s}$; (c) $70\ \mu\text{s}$; (d) $105\ \mu\text{s}$ and (e), $125\ \mu\text{s}$ from the start of the MHD actuation.

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Table 1
Sn and Ag jetting stability: droplet speed, radius and trajectory.

		Sn	Ag
Average droplet speed v_j	[m/s]	1.72	1.88
Speed deviation σ_{v_j}	[m/s]	0.01	0.04
Average trajectory	[°deg]	90.3	89.9
Angle deviation ϑ	[°deg]	0.13	0.2
Average radius r	[μm]	39	38

and L are the cross-sectional area and length of the track, respectively. Two-layer tracks with a length of 40 mm were printed on a Al_2O_3 plate coated with un-sintered Cu ink to ease the detachment of the printed structure prior to measurement. The dimensions of the tracks were calculated by microscopy analysis. The resistance of the tracks was measured using a Hameg LCR high precision meter (HM 8018). The reported values are an average of the electrical resistivity of five different tracks.

quency of 500 Hz and $d = 1$ mm, the positional error caused by the variation in the droplet speed is $\delta_v \sim 0.1 \mu\text{m}$ (Sn) and $\delta_s \sim 0.5 \mu\text{m}$ (Ag). The combined total positional accuracy uncertainty is below $4 \mu\text{m}$ for both metals. The presented equipment is a research platform that is not optimized for printing speed or cost. Nevertheless, by considering the current capabilities a realistic deposition rate up to $2 \text{ cm}^3/\text{hour}$ per print head can be achieved. However, the deposition rate clearly scales with the volume of the jetted droplets, the building block of our 3D parts. With the necessary changes in the nozzle geometry, it would be straightforward to jet larger droplets and increase the deposition rate dramatically, as per target requirements.

2.4. Printing setup

Droplets were sequentially jetted from stationary print heads onto a substrate mounted on a movable stage (PRO225LM and ANT130-L Aerotech stages for the x-, z- and y-axes, respectively) with a nozzle to substrate distance of 1 mm. A LabVIEW program was used to read the stack of bitmaps corresponding to the printing structure and coordinate the printing process. First, each bitmap was scanned line by line to inform the x- and y-stages on the desired path to follow - the x- and y-axes substrate movements were controlled by simply converting the black pixels of the desired bitmaps into coordinates so that the droplet pitch matched the bitmap resolution. The program then synchronizes the jetting of each droplet with the position of the substrate: droplets are jetted on the basis of the pixel value and substrate speed is directly derived from the desired jetting frequency and droplet pitch. At the end of each layer, the substrate is lowered (z-axis) by the dictated thickness of the deposit before printing the following layer/bitmap.

3. Results and discussion

3.1. Additive manufacture of 3D structures in Sn

To demonstrate the printing capabilities associated with the MetalJet approach various structures in Sn were fabricated to allow the study of how objects are made one droplet at the time (Fig. 2).

Fig. 2(a) shows a lateral view of a single Sn droplet deposited on a Cu substrate. Upon contact with the substrate, the liquid Sn spreads driven by the dynamic pressure of the impact and then solidifies bottom up. The oscillations of the melt due to the balancing of inertial and capillary forces can be observed in the form of surface ripples that have frozen as the solidification front sweeps the entire droplet [24,25]. As the metal is deposited gently (Weber number, $2.6 < We < 4.6$) and in a superheated state, its flow is essentially unaffected by the viscosity (Ohnesorge number, $Oh < 0.1$) [26]. Due to the metallic compatibility of Cu and Sn, the droplet realizes a strong bonding with the substrate. Fig. 2(b–c) shows the interface formed by the Sn droplet on the substrate. The TEM investigation indicates that the liquid Sn reacts with the substrate material to form an interface consisting of two intermetallics ($\epsilon\text{-Cu}_3\text{Sn}$ and $\eta\text{-Cu}_6\text{Sn}_5$), as predicted by the Sn-Cu phase diagram. The layered interface suggests that $\epsilon\text{-Cu}_3\text{Sn}$ forms in the first stage of the solidification when Sn is in direct contact with Cu; $\eta\text{-Cu}_6\text{Sn}_5$, appearing with the typical scallop morphology [27], would instead grow in the latter stages of the solidification at the expense of $\epsilon\text{-Cu}_3\text{Sn}$ and liquid Sn. The chemical reaction at the interface promotes a strong adhesion of the droplets to the substrate. Continuous tracks made of conjoined droplets were then realised (Fig. 2(d)), where impinging droplets partially re-melt and create a metallurgical bond with

3.3. Macroscopic geometric fidelity to the original digital model

The dimensions of the corresponding printed specimens are reported in Table 2.

The results show a tolerance for the x- and y- dimensions of the parts comprised within 0.3% while it was observed a relatively larger deviation from the CAD z-height with the real average layer thickness

4. Conclusions

In summary, this research presents the MetalJet technology as a novel digital fabrication technique to form structures that extend in the macroscale with a lateral resolution of $80\text{ }\mu\text{m}$ (approximately double the accuracy achievable with laser powder-bed AM techniques) in low-

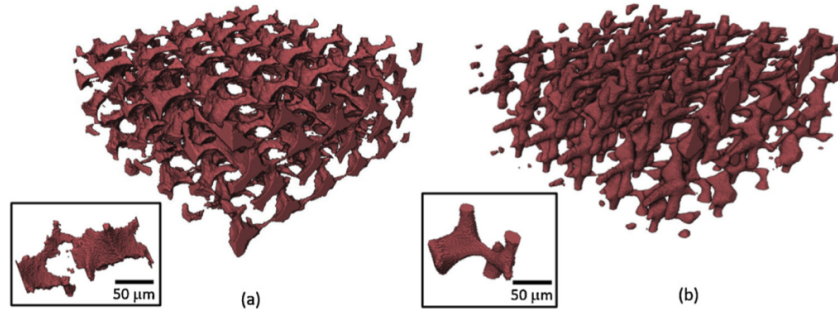


Fig. 7. Reconstructed porosity (in red) from X-Ray CT in (a) as-built and (b) sintered Ag sample. The sintering treatment causes a change of the morphology of the voids as shown in the image insets (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.).

Table 3

Surface roughness parameters Rt and Ra of the Sn and Ag cuboid structure (ISO 4287).

	xy-plane (Sn)	xz-plane (Sn)	xy-plane (Ag)	xz-plane (Ag)
Ra [μm]	1.60	0.39	5.98	n/a
Rt [μm]	7.98	3.43	30.21	n/a

and (relative) high-melting temperature metals. This research has shown the importance of the substrate choice to realize different levels of adhesion and sintering treatments. The temperature range in which metals are miscible, for example, can be used as a guideline to assess whether droplets will form a strong adhesion or a fragile interface. The strength and density of the printed objects can be directly correlated to the droplet bonding. This is a function of the temperature of the droplet interface, and as such liquid metal printing of solid structures in high melting point metals poses significant technological challenges. Nevertheless the DoD approach presented in this study allows the creation of precise “green” structures via deposition strategies that allow high packing density and promote sintering during post-process heat treatments. Various structures were realized by simply coordinating the droplet ejection to the movements of a X,Y,Z stage: by extension, additional axis of movement, for example a rotational axis

perpendicular to the droplet trajectory, could enable the production of filigree structures, such as winding coils, helices and mesh tubular structures for electronics and medical applications. Additionally, with further development of the hardware aimed at increasing the temperature of the droplets at landing/substrate and as more jetting materials will be developed we expect to extend the capabilities of the MetalJet fabrication towards the printing of a variety of objects made of single and dissimilar materials to fulfil the needs of the next generation additive manufacturing.

Declaration of Competing Interest

The authors declare no conflict of interest.

Acknowledgement

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Appendix A

In-flight droplet cooling and re-melting of the previously deposited material

The metal droplets are jetted superheated above their liquidus temperatures and therefore are subject to rapid convective and radiative cooling during the in-flight phase. As per the small Biot number, the heat conduction in the interior of the droplet can be neglected. Assuming no significant heating by radiation from the nozzle of the cartridge, the droplet cooling during the time of flight can be expressed in the form of the Eq. (A.1):

$$\rho c \frac{4}{3} \pi r^3 \frac{dT}{dt} = -4\pi r^2 [h(T - T_{amb}) + \varepsilon \sigma (T^4 - T_{amb}^4)], \quad T(t=0) = T_{drop} \quad (\text{A.1})$$

where ρ is the density of the liquid metal, c is the specific heat capacity of the liquid droplet material, r is the droplet radius, T_{drop} is the droplet temperature at ejection, T_{amb} is the argon gas temperature, h is the heat transfer coefficient, ε and σ are the emissivity and Stefan-Boltzmann constant, respectively. As in the liquid phase there is only a minor variation with temperature of the specific heat c and density ρ it is assumed, for sake of simplicity, constant c and ρ during the cooling of the droplets (Table A1). The convective heat transfer coefficient h was calculated using the relationship $h = \left(\frac{Nu \times k}{D} \right)$, where Nu is the Nusselt number (the gas velocity is assumed to be equal to that the average droplet velocity), k is the thermal conductivity of Ar at the temperature in proximity to the print heads (measured by placing a thermocouple in proximity of the nozzle), D is the average diameter of the droplets. As the process occurs in inert atmosphere, it is assumed a constant value of the emissivity ε associated to the surface of non-oxidised droplets in the liquid state (Table A1).

A finite difference method was used to numerically solve Eq. (A.1), with a step size of 0.0001. With the typical time-of-flight of 1 ms and the jetting temperature used in these experiments the temperature of the Sn and Ag droplet just before impact varies between $595 < T < 615$ °C and $1015 < T < 1025$ °C, respectively.

The re-melting of the previously deposited material and therefore the occurrence of metallurgical bonding is modelled as a two-phase one-dimensional Stefan melting problem [31].

This model considers only the first few instants after the droplet impact ($t < 10\mu s$) and is valid when the characteristic length scale for the thermal conduction in the droplet ($\sim \sqrt{\alpha t}$) is much inferior than the droplet diameter as is the case in our research. This allows to conceptualize the problem

where α_l , α_s , k_l , k_s are the thermal diffusivity and the thermal conductivity of the liquid and solid phase (Sn and Ag respectively), and $S(t)$ is the moving solidification front. Applying the following boundary conditions:

$$T_l(x, t) = T_s(x, t) = T_m \text{ (constant) at } x = S(t), t > 0$$

$$T_s(x, t) = T_{sub} \text{ (constant) for } x = -\infty, t > 0$$

$$T_l(x, t) = T_{drop} \text{ (constant) for } x = +\infty, t > 0$$

The Neumann's analytical solution for the solidification front is given by Eq. (A.5):

$$S(t) = [k_s \frac{(T_m - T_{sub})e^{-\lambda^2}}{\sqrt{\alpha_s} [1 + \text{erf}(\lambda)]} + k_l \frac{(T_m - T_d)e^{-\frac{\alpha_s \lambda^2}{\alpha_l}}}{\text{erfc}(\lambda \sqrt{\frac{\alpha_s}{\alpha_l}})}] \frac{2\sqrt{t}}{\rho L \sqrt{\pi}} \quad (\text{A.5})$$

while the temperature profiles and distributions in the liquid and solid phase are:

$$T_l(x, t) = T_{drop} - (T_{drop} - T_m) \frac{\text{erf}(\frac{x}{2\sqrt{\alpha_l t}})}{\text{erf}(\lambda)} \quad (\text{A.6})$$

$$T_s(x, t) = T_{sub} + (T_m - T_{sub}) \frac{\text{erfc}(\frac{x}{2\sqrt{\alpha_s t}})}{\text{erfc}(\lambda \sqrt{\frac{\alpha_l}{\alpha_s}})} \quad (\text{A.7})$$

where λ relates to the initial conditions of the problem and is derived iteratively from the solution of following transcendental Eq. (A.8):

$$\frac{St_l}{e^{\lambda^2} \text{erf}(\lambda)} - \frac{St_s \sqrt{\alpha_s}}{\sqrt{\alpha_l} e^{\frac{\alpha_l \lambda^2}{\alpha_s}} \text{erfc}(\lambda \sqrt{\frac{\alpha_l}{\alpha_s}})} = \lambda \sqrt{\pi} \quad (\text{A.8})$$

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Summary

- Learned the overall structure of the paper
- Analysed the real-world application of structural components
- Prepared for in-depth IMRD component analysis