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UKRI Engineering and Physical Sciences Research Council

EPSRC Programme Grant

Enabling Next Generation Additive Manufacturing

Final report

UNIVERSITY OF BIRMINGHAM

WARWICK
THE UNIVERSITY OF WARWICK

Contents

Image (cover): Dr Feiran Wang printing a graphene structure in a Dimatix printer.
Image (this page): Coloured electron microscopy image of clustered silver nanoparticles inkjet printed onto silicon dioxide and heated at 500°C.

Foreword

Professor Richard Hague
Principal Investigator

As we gather to mark the conclusion of this extraordinary six-year journey for our EPSRC Enabling Next Generation Additive Manufacturing (AM) Programme Grant (PG), I am very proud of the significant advances and contributions that we have made by meeting our original vision of driving disruptive change, rapid development, and adoption of next generation additive manufacturing.

Our PG, with its continual focus on advancing processes, developing novel materials and devices in additive manufacturing, has pushed the boundaries of scientific and engineering understanding, resulting in positive outcomes across various challenges. Beyond the significant scientific discoveries, I'd like to highlight the impact of our collective efforts on the broader AM landscape.

Beyond our academic achievements, one of the most tangible measures of our success lies in the extensive career progression of our researchers. Witnessing the growth, development, and achievements of each member of our team has been nothing short of inspiring. From opportunities to lead projects and co-supervise master's and PhD students, to securing their first grants and earning well-deserved promotions, our postdoctoral researchers have demonstrated exceptional dedication and expertise to keep up the momentum of the Programme throughout the various challenges faced. Of equal importance, I would like to highlight the contributions of our PhD students who, through PG-aligned projects, have provided significant input to our research.

Equally noteworthy is the emphasis we have placed on knowledge exchange and engagement with non-academic audiences. Through our outreach events, research inputs to the political arena, and collaborations with industrial partners, we are actively contributing to shaping the future of research strategy in additive manufacturing.

As we reflect on our accomplishments, it is crucial to acknowledge the collaborative spirit that has underpinned our journey. Each breakthrough, each milestone reached, has been the result of collective endeavour, fuelled by the diverse perspectives, expertise, and unwavering commitment of every individual involved.

Looking ahead, as we transition from this chapter to the next, we will carry forward the lessons learned, the collaborations established, and the aspirations built. Whilst our formal Programme may be ending, the impact of our work will endure, shaping the future of additive manufacturing and inspiring generations to come.

In closing I extend my sincere thanks to our funder, EPSRC, for their continuous support, and to our partner institutions – the Universities of Warwick and Birmingham and the National Physical Laboratory (NPL) – alongside a host of other academic and industrial partners.

Finally, I would like to give my heartfelt thanks to our Advisory Board for their guidance throughout this journey, and the support team for their professionalism.

As we come to the end of this chapter, I invite you to look out at our achievements and share my feelings of accomplishment and anticipation for the limitless possibilities that lie ahead.

Highlights

People

41 Members

12 Academics

3 Universities

25 Researchers

18 Aligned PhD research projects

Dissemination

131 Publications across all research challenges

24 Keynote speaker appearances

21 Committees and leadership activities

3 Book chapters

Engagement activities

24 STEM outreach activities

2 STEM toolkits developed

16 Media appearances

28k+ YouTube viewings

16 Undergraduate summer projects

Influencing policy

£47m+ Research grant leveraged

9 Engagement activities with policy makers

15 Advisory committees

1 Leading UK manufacturing spin out reform

Business partnership

£2m Industrial funding leveraged from the PG

13 Industrial partnerships

6 Expert panels hosted

Vision

Our Programme Grant focusses on understanding how to control the co-deposition (interfacing) of both functional and structural materials to significantly extend Additive Manufacturing's (AM)'s reach beyond the well understood geometric freedoms it currently enables. We do this by leveraging the application foci of 3D electronics, pharmaceuticals and healthcare devices and now, regenerative medicine.

This vision has been achieved through projects developed in line with four main research challenges detailed below. In the final years of the PG, a combination of the findings of each research challenge were combined to deliver the PG's overall vision.

Strategic research priorities

Research Challenge 01

Determination of the interface/ interphase evolution at microscale

Aim: To understand, through precision experiments, the spatio-temporal interface/interphase evolution between successively deposited droplets or voxels of multimaterials at the microscale. Specifically, the projects in this research challenge focussed on the fundamental understanding of the interfaces of materials produced by AM processes.

Research Challenge 02

Multifunctional AM computational modelling framework

Aim: To develop a multifunctional Additive Manufacture Computational Framework that will guide the manufacturing strategies to be employed to create functional objects; delivered across all the academic partners at the universities of Warwick, Birmingham and Nottingham. Having a framework that provides an understanding across the processes leads to greater design capability for multifunctional additive manufacturing. To accurately model and simulate multimaterial AM, from constituent materials to multifunctional components.

Research Challenge 03

Controlling connectivity and anisotropy for enhanced functionality

Aim: To control the connectivity of additively manufactured layers in order to enable the material's full functionalisation. In the first part of the Programme, there was a focus on processes and material library development for two-photon polymerisation (2PP), as well as advancements in both the 3D inkjetting and MetalJet processes. During the second part of the Programme, we focussed on the manufacturing processes themselves.

Research Challenge 04

Controlled co-deposition of multimaterials

Aim: To investigate and demonstrate strategies for the macroscale co-deposition of functional and structural materials via piezo-driven-inkjetting, high temperature metaljetting and functionalised multi-photon techniques, with a focus on multimaterial structures with potential applications in electronic devices and healthcare

This individual research-challenge approach was used to facilitate the delivery of Programme outcomes. However, the symbiotic relationship between the research challenges has meant that delivery of each one, but interconnected with each other, greatly enhanced the outcomes achieved.

Members

Investigators

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Principal Investigator and Joint RC1 Lead

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Co-Investigator and Joint RC1 Lead

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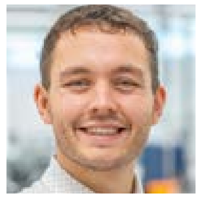

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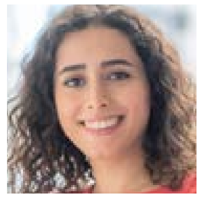

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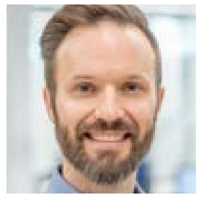

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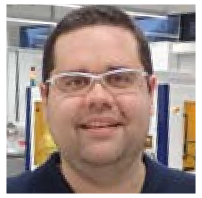

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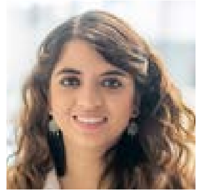

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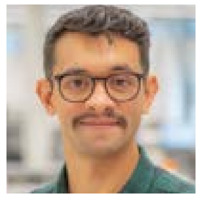

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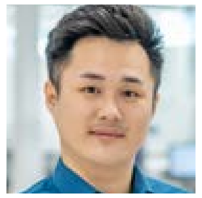

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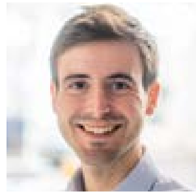

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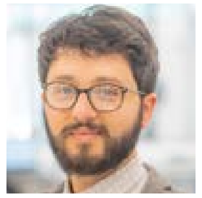

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Mirela Axinte
Programme Manager (inception), Faculty of Engineering, University of Nottingham

“ This has been an exciting programme, and it has been a pleasure to see the progress being made throughout the timeline, with the multi-disciplinary, multi-party teams working extremely effectively together within and across the four Research Challenges. On the one hand the programme has generated some real, fundamental understanding of the jetting of multimaterials and the mathematical modelling of this, whilst on the other hand we have seen new applications developed in both electronics and biopharma areas. It is satisfying to see these latter developments being taken forward into a new multi-year programme. ”

Dr William Barton, Chair of the Advisory Board

Image: 3D-printed pills will allow digital design of medication release. In this image, the insoluble core and barriers used to control the drug release of a 3D-printed pill.

Our final year at a glance

The past year has been one of significant advancement within the field of multimaterial additive manufacturing (AM). The long-term research undertaken within this Programme Grant is showing great promise, placing the UK at the forefront of next-generation AM research.

This PG enabled us to develop and consolidate expertise within analytical methods, processes, modelling and functional materials – the foundations for a device-led approach. These efforts pushed the boundaries of what is possible in multimaterial AM and confirm the Centre for Additive Manufacturing as an international leading group in research into next generation AM processes and materials.

Key advancements in the past year

Analytical methods

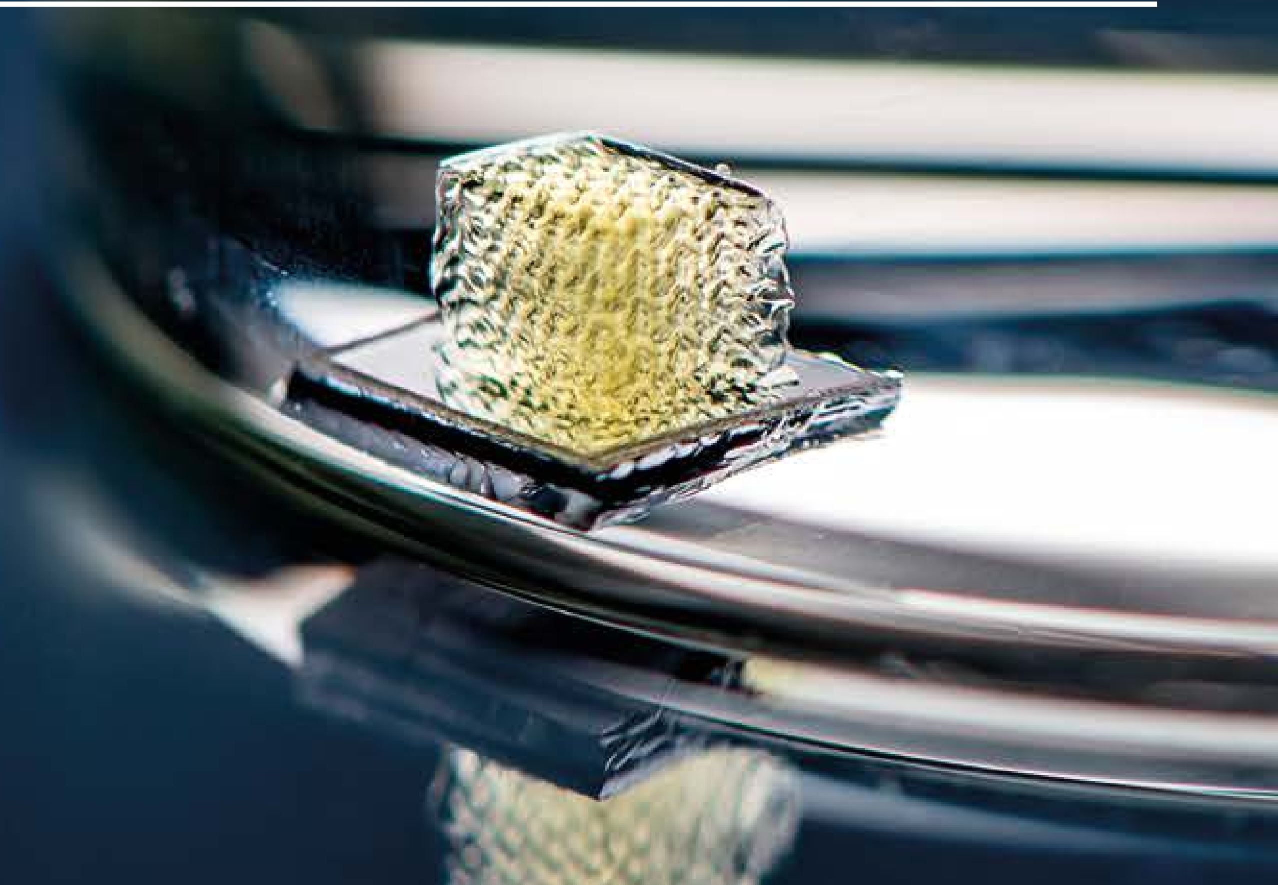
- Advanced interface analysis to drive strategies for novel ink formulations containing nanoscale materials such as graphene, hexagonal boron nitride and functionalised gold nanoparticles alongside perovskite nanocrystals and quantum dots.
- With the National Physical Laboratory (NPL) and concentrating on devices, we explored the use of the 3D orbiSIMS instrument to exploit its potential for multimaterial AM.

Functional materials and demonstrators

- The development of functional demonstrators, alongside the investigation of both conductive and dielectric materials for inkjet printing.
- Development of inkjet printing for perovskite nanocrystals and printing of photodetectors.
- Investigation of post-deposition functionalisation of low dimensional materials for inkjet-printing electronic and optoelectronic devices, with multiple vertically stacked 2D material layers (such as capacitive sensors and digital processors), and with graphene functionalised by optically active materials (photon sensors).

Processes

- We completed the installation of a new multimaterial jetting system based on Xaar 1003 printheads to enable printing of more viscous ink formulations and developed a low-cost multimaterial jetting platform based on Xaar 128 printheads to expedite functional material development and formulation.
- We completed the construction of our multi-head MetalJet platform for the co-deposition of two high temperature metallics in the same print. We also developed a new metaljetting material based on alloys (Cu-based and Al-based).
- We developed 2PP deposition of functional materials and ordered sub-micron arrays for applications in electronic and optoelectronic devices and we continue to explore opportunities for the post-deposition functionalisation of 2PP structures with nanomaterials.



With a continued eye on the future and capitalising on the advancements of this PG, we have achieved significant development of demonstrators, including:

- ongoing developments of printed electronics demonstrators, including graphene sensors, fluorescent sensors, perovskite LEDs
- ongoing developments in multimaterial metal printing for design freedom and applications operating in extreme environments
- advancements in technology for a clean energy future, for example, printed batteries and hydrogen storage

At the same time, our focus on developing the next generation of postdoctoral researchers into future leaders continues. From an academic point of view, three of our senior researchers have accepted academic positions, which opened an opportunity to continue the development of some of the PhD students from our centre. Six research associate positions were filled, and one has already progressed to research fellow. This opportunity was possible due to the flexibility of the PG mechanism and our current device-led approach in driving the research direction, giving us significant traction in work associated with our application areas of 3D electronics, clean energy and extreme environments.

During the summer we also offered an undergraduate placement programme, with a record number of applicants and five undergraduate students joining the team. The cohort successfully delivered projects in four different areas: MetalJet, hydrogen storage device, interface investigations using Raman spectroscopy, and battery development. In the same remit of developing future generations, we have expanded our participation in outreach events. In our last year, we participated in the prestigious Royal Society Summer Science Exhibition in London, and joined four relevant STEM events in the Midlands. These are positive examples as to how new talent can be attracted to continue developing leading research in STEM disciplines, very aligned to our ethos.

Delivering next generation additive manufacturing

1. Multimaterial interface analysis and control

Image: Multiple versions of 3D-printed finger stent exemplar on the SEM stubs.

To enable next generation additive manufacturing (AM), one of the key challenges tackled during this Programme Grant was related to the difficulty of inter- and intralayer coalescence/bonding for functional-structural and functional-functional materials. One of the primary challenges in multimaterial deposition is the potential difference in the co-deposited materials' physical state, chemistry and temperature at deposition or conversion. Through precision experiments and the development of new methodologies for ex situ materials analysis, we have been solving these challenges.

At earlier stages of the PG, we established a series of analytical strategies for ex situ materials interface analysis. These comprise sample preparation procedures suitable for each printing method and baseline material characterisation for a suite of core analytical techniques, such as time-of-flight secondary ion mass spectrometry (ToF-SIMS), focussed ion-beam scanning electron microscopy (FIB-SEM), Raman spectroscopy, X-ray photoelectron spectroscopy (XPS) and atomic force microscopy (AFM). Having these protocols in place enabled our researchers to focus on the fundamental understanding of the materials interfaces. In the following years, we have implemented and integrated these methods across the Programme, thereby informing the modelling and building optimisation strategies across different processes.

In the last years of the Programme, we have increased our collaboration via a series of projects with the National Physical Laboratory (NPL) and other internal groups, including, the Nanoscale and Microscale Research Centre (NMRC), as well as international collaborations with the University of Sao Paulo and CERN, the European Organization for Nuclear Research. Through these collaborations, we have been investigating inkjet-printed interfaces of functional materials using ToF-SIMs, FIB-SEM, and FIB-TEM to map the printed interfaces at the sub-micron scale.

Heterostructures

The manufacturing of heterostructures requires the interfacing of dissimilar materials in controlled geometries. Arranging these printed materials into different architectures enables a range of device functionalities to be achieved, but the performance of these devices is highly dependent on the quality of interfaces between neighboring (adjacent) materials.

We developed methods to probe the interfaces of printed heterostructures using a combination of time-of-flight secondary ion mass spectroscopy (ToF-SIMS) and focussed ion beam scanning electron microscopy (FIB-SEM) and assessed how different printing parameters affect interface quality. By adjusting our ink formulations, printing strategies, and post-processing techniques, we demonstrated control over printed heterostructure interfaces, and thus control over the performance of printed heterostructure devices.

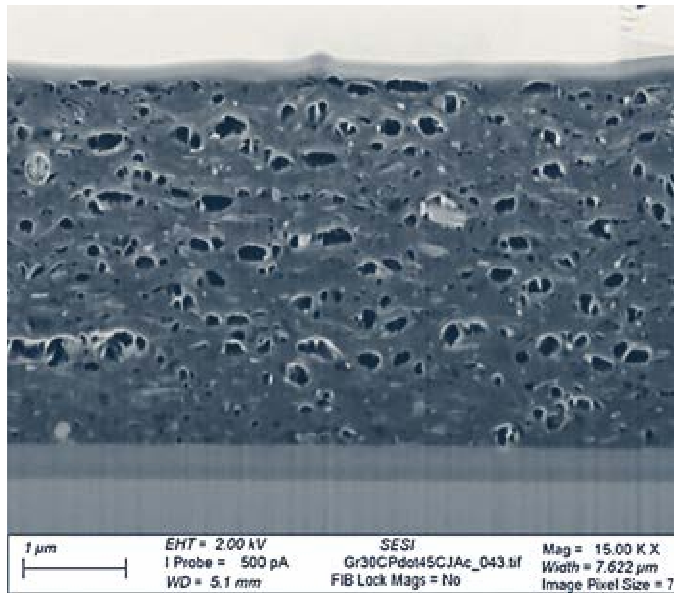


Image: FIB-SEM image of inkjet printed PEDOT:PSS/graphene heterostructure on Si/SiO2 substrate. The printing parameters were selected to control the pore sizes within the graphene layer.

Phase separation

The discovery of how phase separation influences drug release is an advancement in personalised medicine, particularly within the realm of 3D printing. This finding explains the creation of intricate microstructures in pharmaceuticals, which can be engineered to control the timing and quantity of drug release. Utilising phase separation in 3D printing brings a new strategy for the fabrication of drug delivery systems with tailored release profiles, essential for optimising therapeutic outcomes and minimising side effects. This technique is especially beneficial in developing controlled-release applications, where drugs can be designed to release active ingredients over specific durations, from hours to months. By integrating phase separation into 3D printed pharmaceuticals, researchers will be able to offer a new level of customisation in treatment, providing patients with more effective and personalised medication.

Related publications

Molecular formula prediction for chemical filtering of 3D OrbiSIMS datasets.
Analytical Chemistry, 2022.

Protein identification by 3D OrbiSIMS to facilitate in situ imaging and depth profiling.
Nature Communications, 2020.

In situ protein identification, mapping and depth profiling using 3D OrbiSIMS.
University of Nottingham data repository, 2020.

Chemical imaging of buried interfaces in organic–inorganic devices using focussed ion beam-time-of-flight-secondary-ion mass spectrometry.
ACS Applied Materials and Interfaces, 2019.

Delivery

2. Novel functional materials

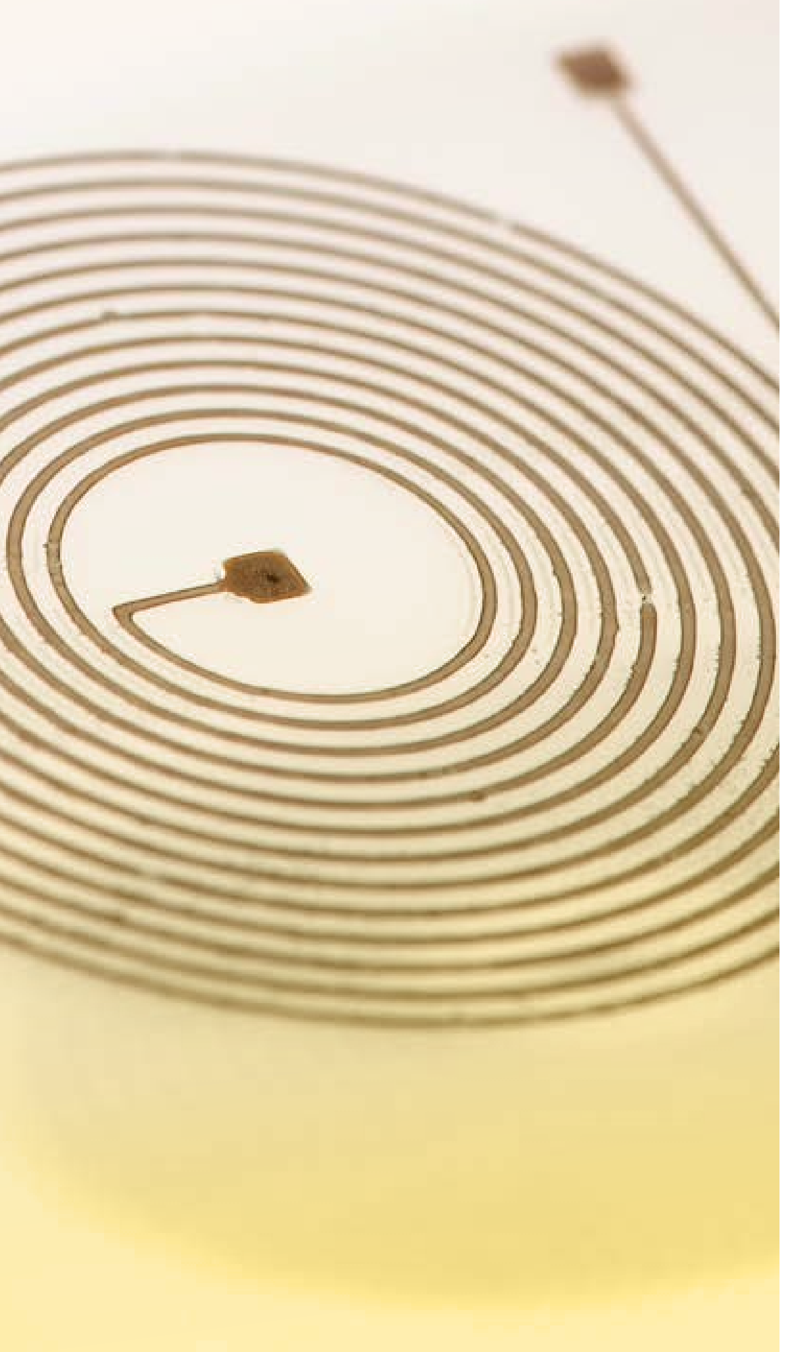


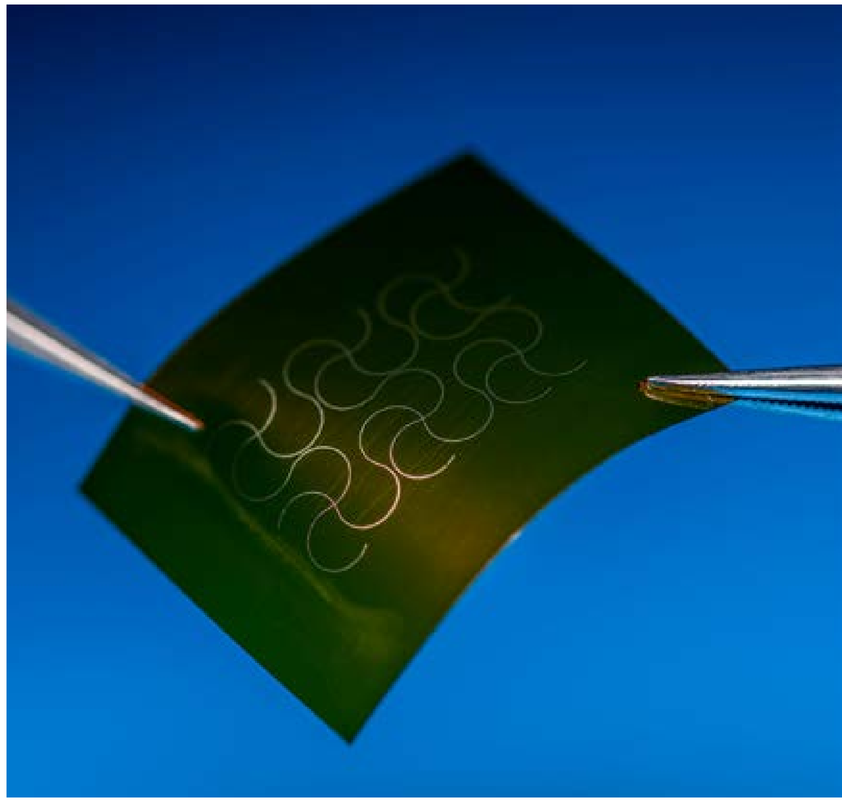
Image: A planar spiral inductor inkjet printed onto a flexible substrate using a gold nanoparticle ink formulation.

Polymers

Throughout the Programme Grant we have been researching advancements to polymers in additive manufacturing at all levels of development. At the fundamental level, we have researched the influence of monomer and polymer chemistry upon the ability to print polymeric and composite structures. This has involved the synthesis of new monomer species that are designed to impart specific surface effects when converted into polymer articles, for example, the ability to prevent the build-up of biofilms. This has also included the synthesis of new monomeric species derived from sustainable sources. Based on this promising initial work, we plan to increase the sustainability and circular economy footprint of AM by making this a key research area for the Centre for Additive Manufacturing (CfAM) going forward.

We have also investigated the use of pre-polymers with defined three-dimensional molecular structures (that is macromeric, comb, star and hyperbranched structures) in formulating AM inks and resins. Use of these polymers, when compared to the typical linear polymers used up to this point, revealed positive enhancements that these more complex structures impart upon the physical and material properties, molecular reactivity and formulation characteristics of the resultant AM inks and resins, as well as useful functional attributes in the final printed polymers. Thus, we generated a new understanding of how carefully selecting combinations of complex polymers and monomers can control functionality alongside inks and resin viscosity, reactivity and flow properties; all critical aspects defining which blends will be processable by AM techniques. This work also revealed the influence that the structure and concentration of these complex polymers had upon the level of cure, the material properties, print fidelity and surface characteristics of the final printed structures; all properties vital for an AM produced device to be fit-for-purpose for adoption in a specific end-use application.

Image: Silver nanoparticle used to print conductive (electrocardiogram) elements.



Metal nanoparticles

The metal nanoparticle work developed during this PG focussed on three main metals: silver, gold and copper. Each of the metal nanoparticle formulations present their own unique challenges and are at different stages of development. They can be used for a range of functional applications as described below.

Silver nanoparticles (AgNPs)

Silver nanoparticles are commonly used to print conductive elements, with the PG using them to print near-field communications (NFCs) for sensors and a variety of connective circuitry. We have also clearly demonstrated that stabilising additives in the ink cause a significant reduction in conductivity in the vertical, intra-layer build direction, and are working to develop ways to overcome this issue. This PG is also focussed on producing complex 3D architectures out of AgNPs for use within metamaterial and electronic devices, including geometries such as pillars and strut-based lattices.

Gold nanoparticles (AuNPs)

We have developed photocleavable, ligand-functionalised, gold nanoparticles for UV assisted sintering. This eliminated the need for high-temperature sintering which can be damaging for flexible polymer substrates. This breakthrough approach enables in-situ printing and UV-sintering for a continuous manufacturing process and multimaterial printing of complex conductive circuits. Optimised ink formulation combined with multimaterial co-deposition strategies for inkjetting have been allowing us to progress with the printing of multifunctional structures.

Copper nanoparticles (CuNPs)

CuNPs are highly attractive for deposition via inkjet printing due to their lower cost when compared to silver and gold. However, deposition of electrically conductive structures using CuNP inks has so far been challenging due to their tendency to oxidise during printing and post-processing. Initial studies indicate that the size of the CuNPs used and the composition of the ink need to be further optimised to enable their sintering into conductive inks. To facilitate this process we are exploring the addition of a small amount of AuNPs into the ink formulations. We have also explored the addition of small amounts of AuNPs into the CuNP inks to improve sintering to achieve high conductivity films.

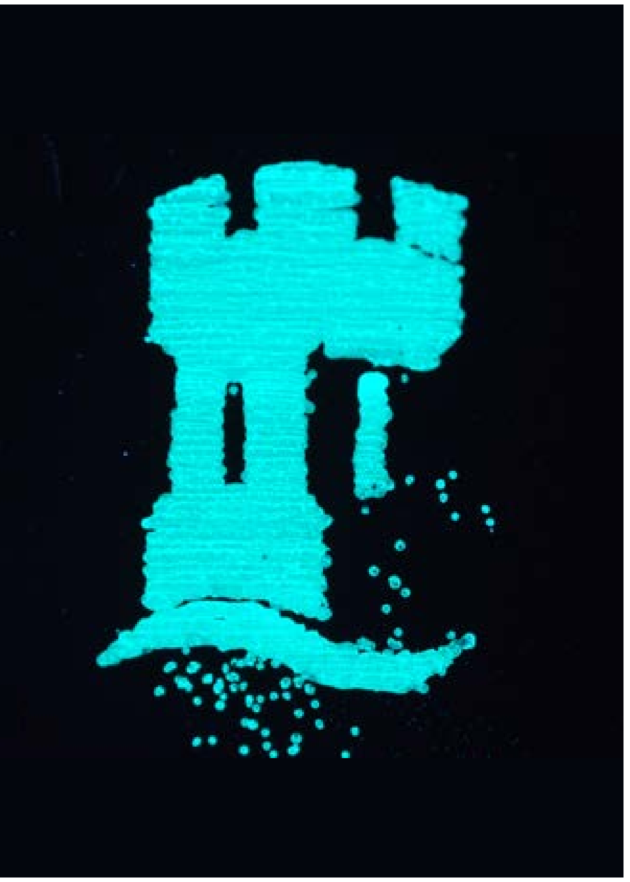


Image: University of Nottingham logo inkjet printed using all-inorganic perovskite nanocrystals with green photoluminescence.

Semiconductive nanoparticles

In recent years, semiconducting nanoparticles (NPs) have been used extensively in optoelectronic devices such as solar cells, photodetectors, and LEDs because they possess a variety of useful optical properties, such as tuneable band gap energies and high photoluminescent quantum yields. The deposition of semiconducting NPs via inkjet printing enables upscaled manufacturing of optoelectronic devices, controlled deposition of different NPs on a single chip/device and enables the fabrication of devices on flexible substrates.

A variety of semiconducting NP inks with bandgap energies tuneable by the nanocrystal size and composition across the ultraviolet, visible, and near-infrared (UV-Vis-NIR) range, including all-inorganic perovskite nanocrystals (NCs), PbS quantum dots (QDs), lanthanide-doped upconverting NPs, and graphene QDs. Printing of these NPs was demonstrated with both water-based inks and inks based on non-polar solvents, and deposition strategies were optimised to achieve thin printed films which maintained their optical properties after printing and post-processing.

The developed inks were used to photosensitise graphene field effect transistors to fabricate high performance photodetectors with tuneable responsivity range UV-Vis-NIR range. Fully printed photodetectors were also demonstrated on flexible substrates by inkjet printing graphene, AgNPs and semiconductor NPs as a conductive channel, electrical contacts and an optically active layer, respectively.

2D materials

Graphene inks, containing liquid exfoliated graphene flakes and ethyl cellulose dispersed in a mixture of solvents, were successfully inkjet-printed onto rigid and flexible substrates. Optimised post-deposition strategies have been developed, including thermal annealing and photonic annealing, as confirmed by high conductivity and signatures in micro-Raman spectroscopy. Alongside deposition strategy, formulation improvements of polymeric stabiliser have been achieved by using hyper-branched polymers to promote dispersion of graphene flakes. Ceaseum lead halide perovskite nanocrystals (PeNCs), lanthanide doped upconverting nanoparticles and PbS quantum dots have been formulated into inks suitable for inkjet printing, with promising optoelectronical applications due to their tunable optical properties, narrow emission bandwidth, and high photoluminescence quantum yield.

Glass

In the past year, a step change in AM glass production using silica nanoparticle-loaded photocurable resins has been achieved. Through digital light processing of silica-polymer resins followed by post-process debinding and sintering, highly transparent and crack-free microstructured quartz glass has been produced. Novel doping and surface decoration have also been demonstrated to alter the optical properties and induce additional functionality. This enables miniaturised and integrated systems for micro-optics and microfluidics applications, paving the way for integrated multifunctional components and benefiting sectors such as energy, pharmaceuticals, quantum technology, automotive and more.

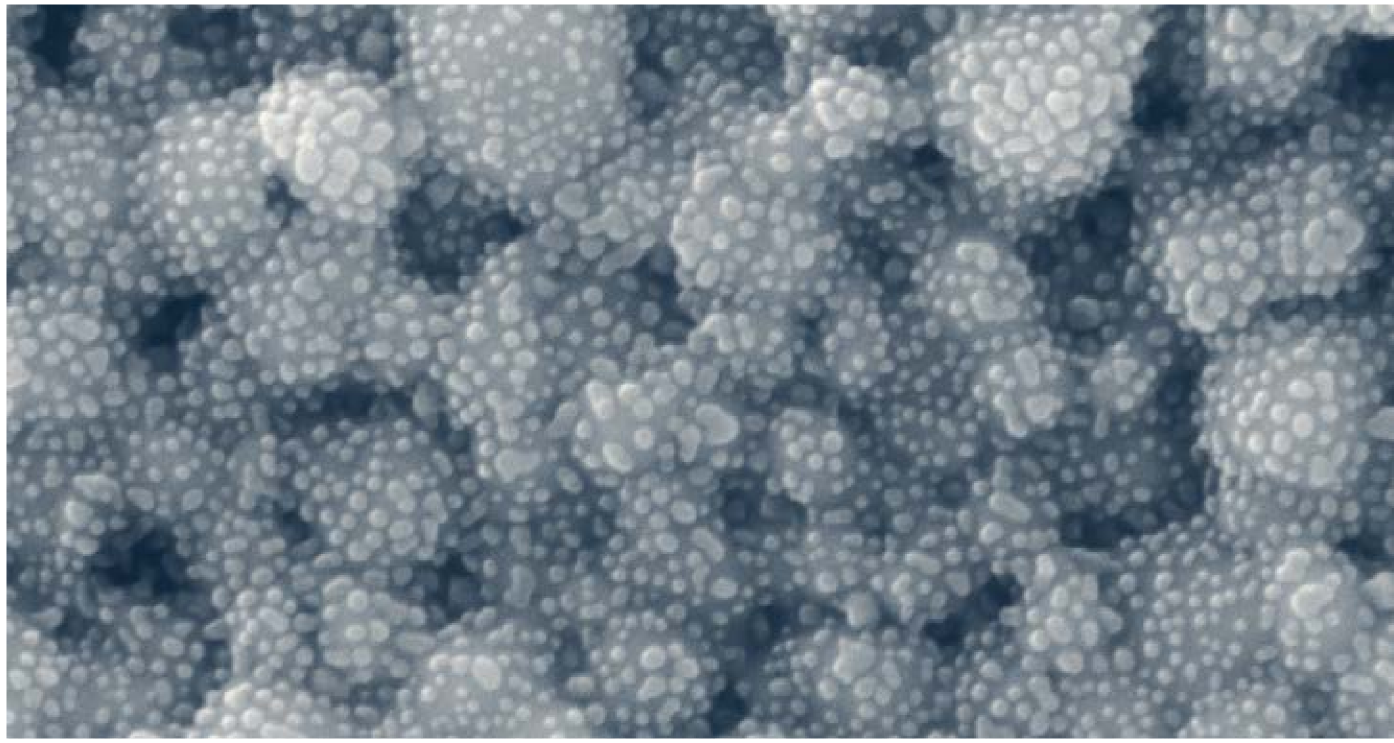


Image: SEM image of two-photon polymerisation printed silver nanoparticles and heated at 300 ° C.

Related publications

Functionalised gold nanoparticles with a cohesion enhancer for robust flexible electrodes. *ACS Applied Nano Materials, 2022.*

A spacer cation assisted nucleation and growth strategy enables efficient and high-luminance quasi-2D perovskite LEDs. *Advanced Functional Materiala, 2022.*

Magnetic and electric field dependent charge transfer in perovskite/graphene field effect transistors. *Advanced Electronic Materials, 2022.*

Residual polymer stabiliser causes anisotropic electrical conductivity during inkjet printing of metal nanoparticles. *Communications Materials, 2021.*

Light-induced stark effect and reversible photoluminescence quenching in inorganic perovskite nanocrystals. *Advanced Optical Materials, 2021.*

Core/shell metal halide perovskite nanocrystals for optoelectronic applications. *Advanced Functional Materials, 2021.*

Core/shell perovskite nanocrystals: synthesis of highly efficient and environmentally stable FAPbBr3/CsPbBr3 for LED applications. *Advanced Functional Materials, 2020.*

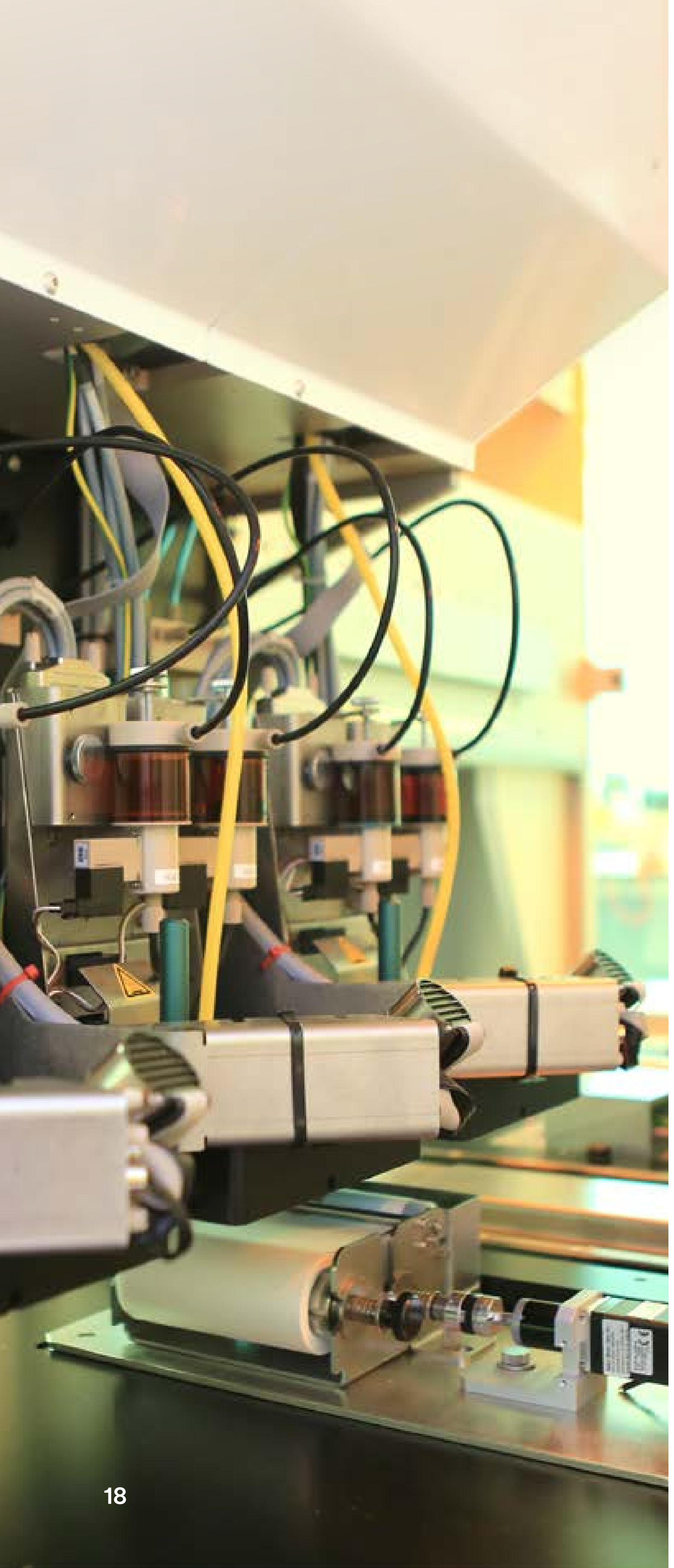
Design of highly stabilised nanocomposite inks based on biodegradable polymer-matrix and gold nanoparticles for Inkjet Printing. *Scientific reports, 2019.*

Atom chips with free-standing two-dimensional electron gases: advantages and challenges. *Journal of modern optics, 2018.*

3D-printed components for quantum devices. *Nature Scientific reports, 2018.*

Delivery

3. Process development



Advancements in MetalJet

Over the past six years under the Programme Grant, our focus has been to deeply understand the six main pillars of the metaljetting process: droplet formation, in-flight droplet cooling, droplet spreading and solidification, interface formation, residual stress development, and microstructure evolution. We have employed an integrated computational, analytical, and experimental approach to gain insights into these multiscale and multiphysics parameters.

The acquired expertise in process operation and our understanding of the underlying physics of the process has been transferred from the single-head system to a new multi-head system.

1. Through an experimental approach, combined with analytical analysis, we gained insights into the mechanism of microdroplet formation. This understanding facilitated the optimisation of the actuation waveform, enabling the printing of several materials with improved precision.
2. Computational and analytical modelling of molten drop cooling during flight, validated by experiments, facilitated the development of an effective printing strategy to maintain drops in their liquid state upon impact. Additionally, these models predicted the droplet temperature upon deposition onto the substrate, serving as an input for subsequent models.
3. Numerical and computational models were developed to precisely simulate the spreading and solidification of droplets upon impact on the substrate, ultimately enabling the prediction of the final morphology of droplets. This information facilitated the optimisation of printing parameters to achieve fully dense structures.
4. The established computational models were instrumental in elucidating the critical elements necessary to achieve sufficient bonding at interfaces, as well as identifying the various modes of bonding. This information was used to optimise process parameters, thereby facilitating the manufacturing of 3D structures with high structural integrity.
5. Advanced characterisation techniques, combined with computational analysis, shed light on the microstructure of MetalJet manufactured components, revealing the influence of thermal conditions on developing diverse microstructures. This provides exciting avenues for tailored microstructure design using this technology.
6. Through computational analysis, validated by experiments, the physical mechanism of residual stress generation during the process was revealed. Armed with this understanding, efforts were made to identify solutions to rectify these issues through virtual experiments, hence providing a basis for a more targeted experimental investigation.

Image: Inside view of Toucan, a bespoke inkjet based 3D printer, which is capable of co-print six different materials.

Printing functional components

We investigated the feasibility of directly printing molten metal droplets using the MetalJet platform for the production of customised 2D and 3D functional components. MetalJet presents a distinct advantage over other technologies for creating such functional objects due to its direct fabrication process routes of high-temperature metallics, eliminating the need for pre- and post-processing. This contrasts with systems using inks loaded with metal nanoparticles, which require post-processing steps to evaporate the ink, adversely affecting performance.

Crucially, MetalJet surpasses the limitations of other technologies in 2D structures, enabling the creation of 3D structures and expanding the spectrum of design options and their efficiency.

As a proof of concept, we conceptualised, fabricated, and evaluated a MetalJet-printed antenna. We have demonstrated that with the superior properties obtained, the technology offers significant opportunities in the realm of electronics and beyond.

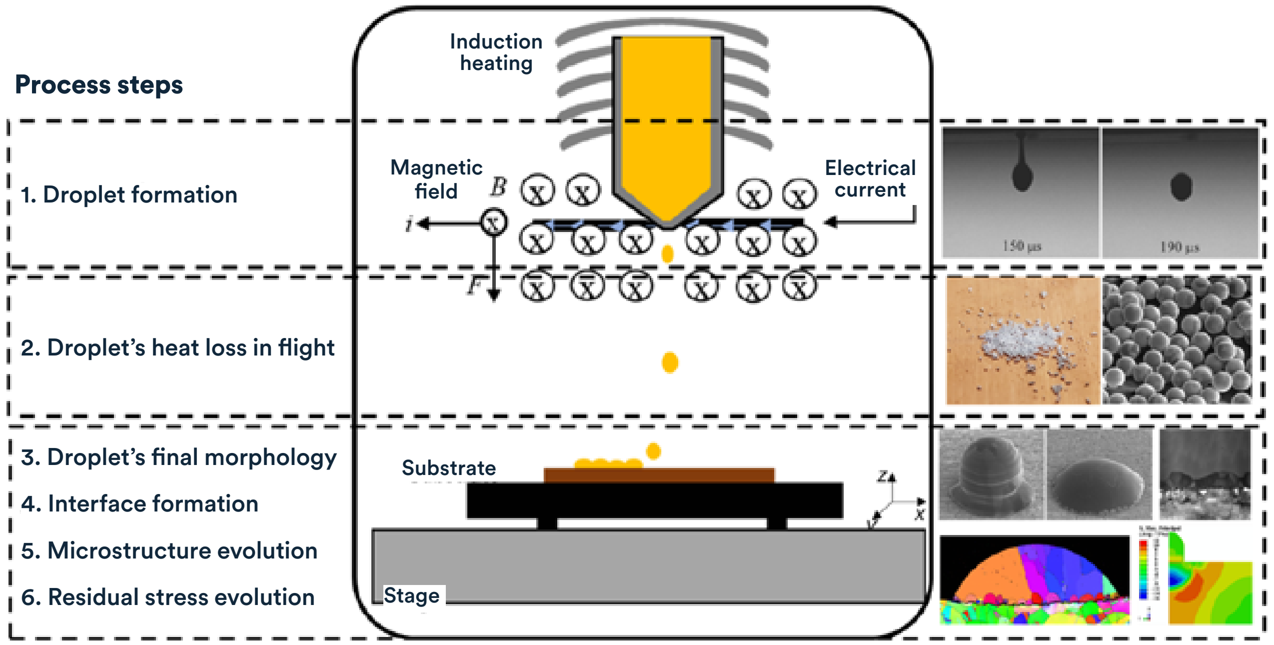


Figure: MetalJet – a multiphysics process.

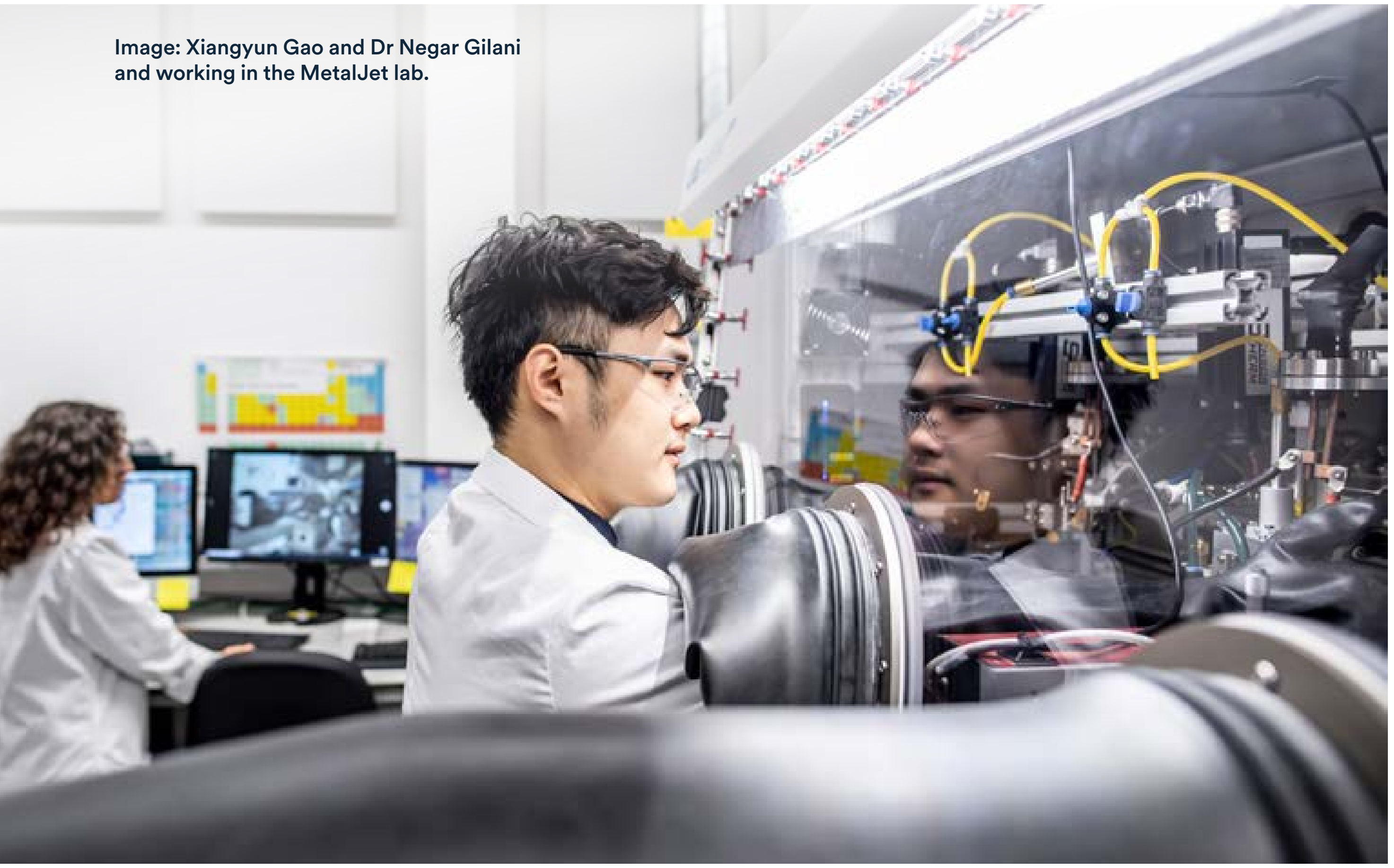


Image: Xiangyun Gao and Dr Negar Gilani and working in the MetalJet lab.

Developing the world's first multi-metaljetting platform

Our in-house multi-MetalJet platform is the first drop-on-demand multi-metaljetting technique that enables printing functional components from at least two different metals in one printing operation. This is accomplished by coordinating the deposition of dissimilar metallic materials on a substrate such that components with spatially varied compositions can be directly created within the build process. This provides a unique opportunity to 3D print components with diverse properties throughout, while eliminating the need for joining procedures. The voxel-by-voxel deposition of various metallic microdroplets offers unmatched control for fine-tuning the local properties of 3D parts and sets this technology apart from the existing multi-metal AM techniques.

Our expertise in process operation and our understanding of the underlying physics of the process have been transferred from the single-head system to the multi-head system, accelerating our progress. The system is currently fully operational, and research on the interaction between droplets of various materials at a microscale is ongoing.

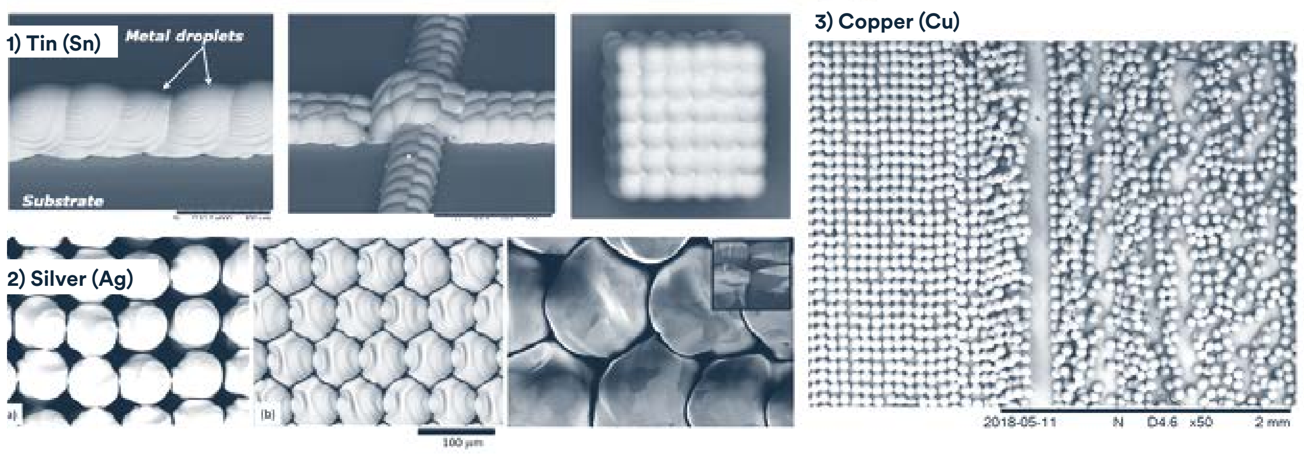


Image: Metal droplets in tin, silver and copper printed using MetalJet system.

Enabling printing a range of materials

Our research within the Programme Grant has led to significant advancements in the printability of both low- and high-melting-point metals. Tin and indium find widespread usage in electronic applications, with their low melting temperatures (232°C and 157°C) making them suitable for printing on polymer and flexible substrates. The resulting components demonstrate high structural integrity and electrical conductivity, comparable to the bulk material, thanks to the robust metallurgical inter-droplet bonding. Furthermore, copper and silver (1084°C and 962°C) are distinguished by their unparalleled electrical and thermal conductivities, placing them at the forefront of materials used in electronics and a wide array of other applications. The strategic selection of both low- and high-temperature metals was instrumental in enabling multimaterial printing. Key tasks included waveform optimisation, ejection temperature adjustments, and compatibility verification with the cartridges.

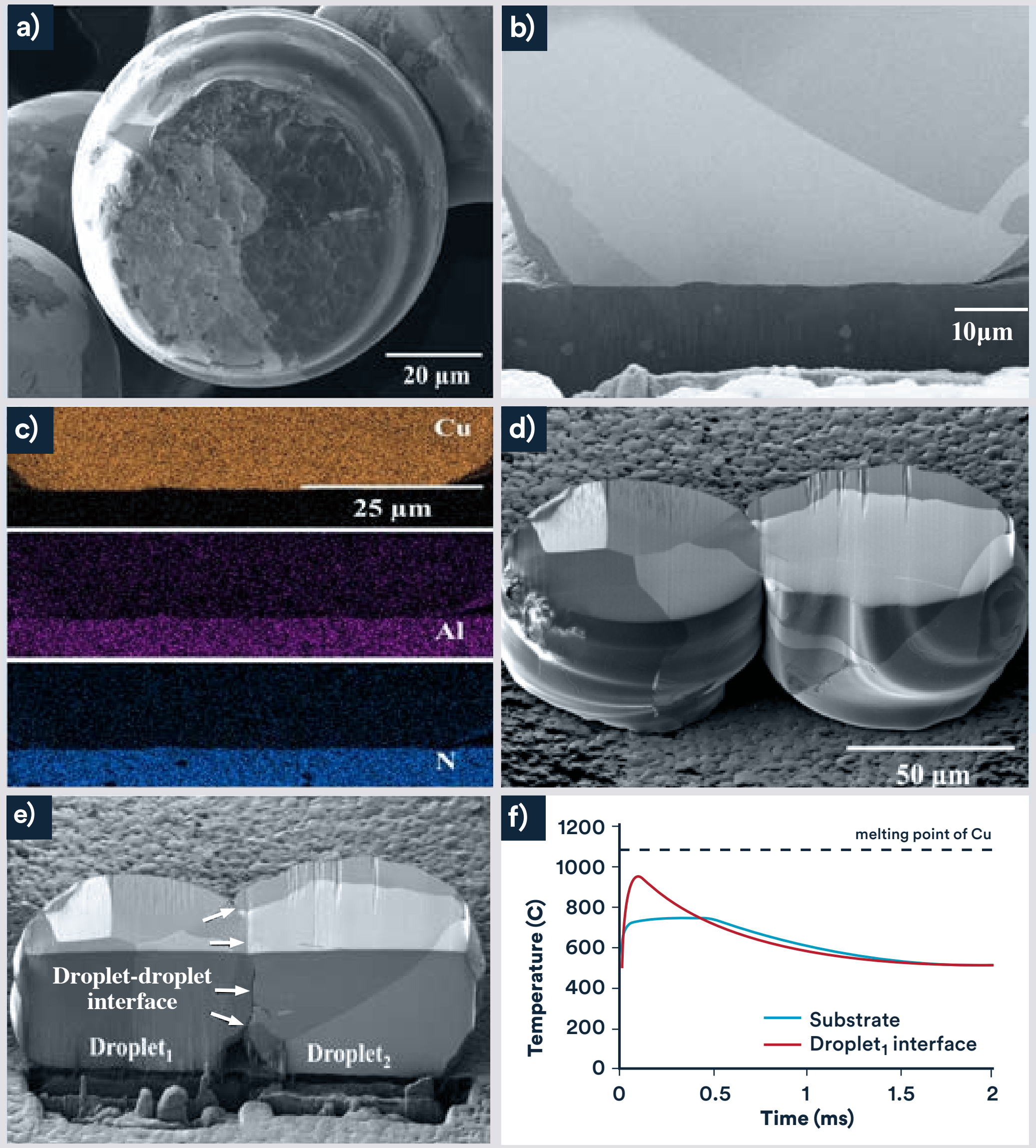


Figure:

a) Interface of an individual Cu droplet with AlN substrate captured by peeling and mounting droplets on a C tape.

b) Cross-section of Cu droplets with AlN substrate illustrating the lack of fusion bonding at the interface.

c) EDX analysis of the cross-section, showing no diffusion at the interface.

d) Cross-sectional view with the interface between the droplets and the grain structure.

e) Cross-section displaying inter-droplet and droplet-substrate interfaces.

f) Temperature evolution at the interface of previously deposited droplets during the deposition of new droplets, the temperature evolution of substrate during the deposition of a single droplet.

Advancements in

Two-photon polymerisation

Materials and formulations

Two-photon polymerisation (2PP) based additive manufacturing has emerged as a powerful technology to fabricate complex three-dimensional micro- and nanoscale architectures. We developed a comprehensive understanding of the effect of printing parameters on the functional properties of these structures, which allowed us to produce multi-material designs. We developed a number of resin formulations based on different monomers and initiators. We established three complementary strategies for integration of nanomaterials into 2PP produced structures: (a) in-situ formation of metal nanoparticles (MeNPs) through a single step photoreduction process, (b) integration of pre-formed NPs into 2PP resin and (c) site-selective MeNPs decoration of 3D 2PP structures. A number of nanoparticles were successfully used, including AgNPs, AuNPs, CuNPs and more recently nanodiamonds.

The link between the 2PP process and final material properties was established by a combination of advanced analytical techniques, such as Raman spectroscopy, mass spectrometry by ToF-SIMS and high resolution electron microscopy. This PG achieved controlled deposition of different materials and multimaterial structures with high precision, enabling rational design and manufacture of complex geometries, including structures with integrated nanomaterials, relevant for applications in photonics, metamaterials, plasmonics, catalysis, bio-sensing and drug delivery.

2PP modelling: towards full-structure simulations

The previously developed extension of a single-voxel model of two-photon polymerisation (2PP) allows allows the straightforward simulation of a few dozen or hundred voxels, hence it cannot model whole 2PP structures, which can consist of millions and even billions of voxels.

Due to the long-range nature of heat and mass transfer during the 2PP process, full-structure simulations are required. Through this project, we have confirmed the validity of significant coarse-graining of our model, which enables simulations of much larger structures. Such simulations reproduce non-uniform polymerisation, also observed experimentally, and have attracted significant interest from industry.

Image: Researcher using the 2PP Nanoscribe.

Advancements in

Inkjet

Throughout this Programme Grant, a range of theoretical modelling, experiments and computational modelling have been used to develop a flexible framework for multimaterial inkjet process optimisation. In parallel, we have been working on formulation optimisation and developing new techniques to improve accuracy, allowing an increased functionality and extending use in applications.

Image: Strain sensor inkjet-printed onto a stretchable surface.

Theory

Advancements in understanding the transport nature of inkjet-printed graphene network

We have further investigated the transport properties of inkjet-printed graphene devices in high magnetic fields and low temperatures, which enable observation of quantum phenomena, such as weak localisation and negative magnetoresistance. The intra-flake and inter-flake classical and quantum transport mechanisms are used to explain the quantum nature of carrier transport in inkjet-printed graphene, where the electrical conductivity can be controlled by the thickness of the graphene film.

Advancements in theoretical modelling and simulations in inkjet-printed graphene

We have worked on developing quantum tunnelling and thermally assisted hopping calculations to explain the temperature and gate voltage dependences of graphene networks in inkjet-printed devices. We have developed a universal model that can be used in the analysis of monolayer graphene devices under varying temperatures, connecting key transport parameters, and performed detailed modelling of the effect of temperature on the electrical properties. This model reproduces, explains, and unifies experimental mobility and conductivity data from a wide range of samples and provides *a priori* a way to predict all key transport parameters of graphene devices.

Formulation

Optimisation of formulations for Dimatix inkjet cartridges

A change in the architecture of Dimatix inkjet cartridges – a previous workhorse of our inkjet-development activity – midway through the Programme required re-optimisation of previously formulated inks in order to cope with more restrictive viscosity parameters. Most of the functional inks previously used, both commercially available and those at research-scale, had been developed and optimised for the previous generation of printhead, and were not compatible with the new printheads. As such, the PG took on the reoptimisation of the library of existing functional materials, whilst also developing new, functional inks for the inkjetting of functional devices.

These included reformulation of: low-dimensional nanoparticle inks for electronics (graphene, gold nanoparticles, conductive polymer PEDOT:PSS, silver nanoparticles), bio-polymers (GelMA blends, resilin-like peptides), and structural polymers (TPGDA, TCDMDA). New functional inks, include optically active nanoparticles (perovskites, lanthanide-doped UCNPs, graphene quantum dots, PbS quantum dots), and semiconductor polymers (pTPD, TPBi). These leading-edge materials are now primed for use in the next generation of printers and printed functional devices.

Print process

Boosting printing resolution: going "Off the Grid" to overcome design limits

During this PG, new techniques for inkjet printing were developed, enabling greater control and increasing options for positioning of the droplets enabling higher degree of freedom needed to produce intricate inkjet printed patterns. Classical printing techniques make a flawed assumption that all available positions of deposited droplets should be multiples of the desired droplet spacing, confining droplets to a grid of possible positions. This results in aliasing: the grid pattern cannot accurately reproduce curves or angles. During this PG, we overcame this limitation, using the Off the Grid technique, which consists of the individual droplet positions and desired spacing being encoded directly into the print design using the printer's maximum resolution. This significantly reduces the aliasing effect, achieving accurate printing of thin curved or angled lines, as well as high-resolution control over the gaps between adjacent printed shapes. Implementation on image-based printers demonstrated, without alterations to the printer software, promising rapid improvement to inkjet printing of detailed devices and structures.

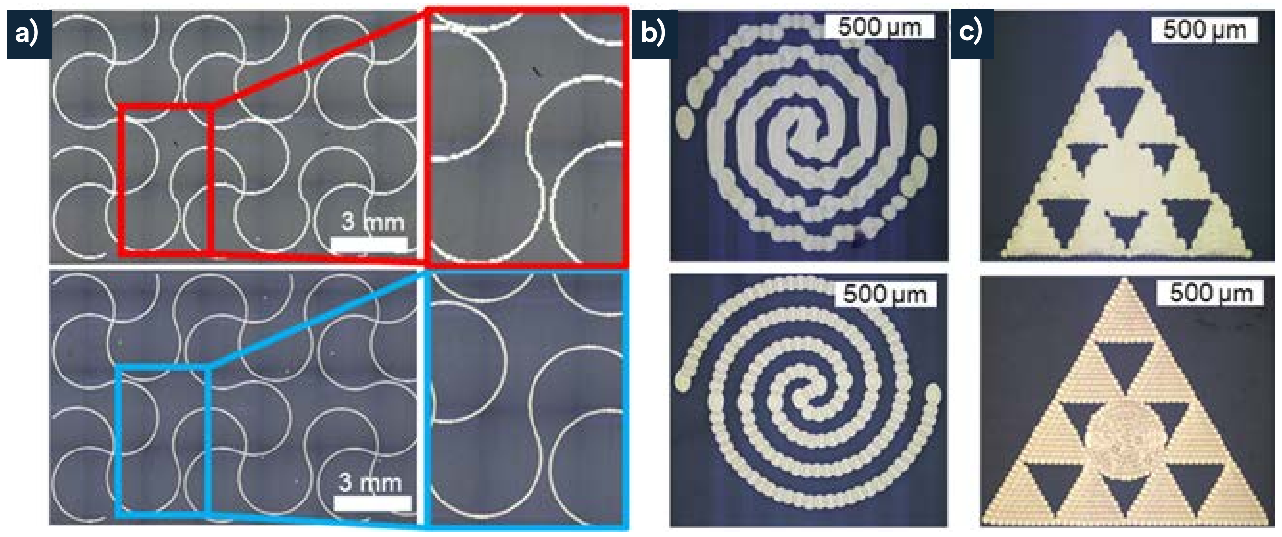


Figure: Optical microscopy images of printed (a) electrocardiogram (ECG) pad traces, (b) an Archimedean spiral antenna, and (c) an X-band Sierpinski triangle antenna. In all cases, samples are printed using AgNP inks (AgCite®) using traditional (top) and OtG (bottom) strategies. The OtG triangular sections in (c) are printed with Sn = 60 µm, and the circular section with Sn = 53 µm.

Inkjet electronic circuits

A major sector in the industrial-level inkjet printing for functional materials has come in electronic circuitry. Silver nanoparticles, when sintered, form the conductive structures, and an insulating polymer – usually a UV-curable polyacrylate – forms the bulk. These circuits are often used for rapid prototyping or creating 3D circuit boards that can fit into oddly shaped spaces. It is also possible to embed components such as RFID tags, antennas, or even electromagnets to create a wide array of unique circuitry that is cumbersome to create through conventional means. Currently we are working on embedding circuitry into 3D printed materials and connecting a variety of printed sensors into fully printed devices, including the ability to wirelessly send results via NFC.

Image: Electronic circuits inkjet printed in a flexible substrate.

Ordered arrays towards metamaterials

Metamaterials have an ordered structure which impacts their properties beyond their chemical composition. Within this Programme Grant, we have been using inkjet printing to create selective reflectarray antennas, miniaturised antennas, and isotropic dielectrics with silver and polymeric materials, easily adjusting spacing and symmetries for a wide range of useful structures.

High-viscosity inkjet upgrades

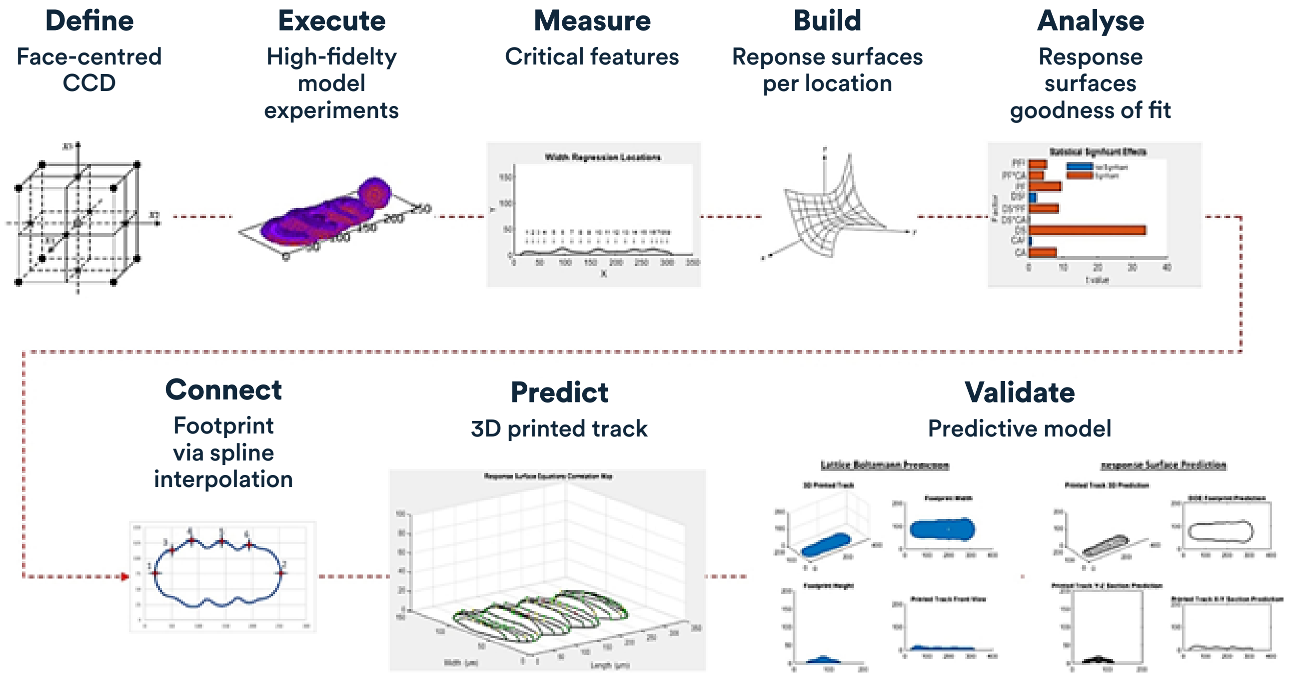
Throughout this grant, new opportunities to expand capabilities in multimaterial inkjet have been presented. In addition to the high-resolution inkjet opportunities mentioned elsewhere, upgrades to our LP-50 printing system dramatically expanded its capabilities, allowing use of XAAR-128 printheads. These new printheads are robustly built in a small footprint, making them easy to maintain and operate, while doubling the number of independent heads on the printer, allowing us to perform multimaterial printing with up to four co-printed materials. They enable rapid printing of large areas with our inks, including dielectric polymers, and electrically conductive or functional inks. Additionally, modifications made to the printheads allow high viscosity resins to be jetted; for the first time, commercial resins intended for SLA printing have been successfully jetted without modification, enabling inkjet to access a large, well-developed materials library.

Alongside these modified systems, we have also obtained a Notion Systems large format inkjet printer, one of two in the world. This uses industrial-scale XAAR 1003 printheads, with 1000 active nozzles per printhead. This enables us to level-up our research going forward, enabling our advancements in inkjet materials, formulations, and techniques from this grant to continue research towards industrial scale-up.

Modelling

Reactive inkjet process optimisation advancements

Figure: Surrogate modelling framework to improve the inkjet-printed track morphology created by the sequential deposition of microdroplets on non-porous substrates.



The development of optimisation models for reactive inkjet processes includes the creation of surrogate modelling and an inverse problem optimisation framework.

The surrogate model focusses on improving the inkjet-printed track morphology created by the sequential deposition of microdroplets on non-porous substrates. The model captures transient effects observed in experiments and builds track morphology in seconds, enabling efficient optimisation of printing and wetting parameters. The simplicity of the proposed technique makes it a promising tool for model-driven inkjet printing process optimisation, including real time process control, and paves the way for better quality devices in the printed electronics industry.

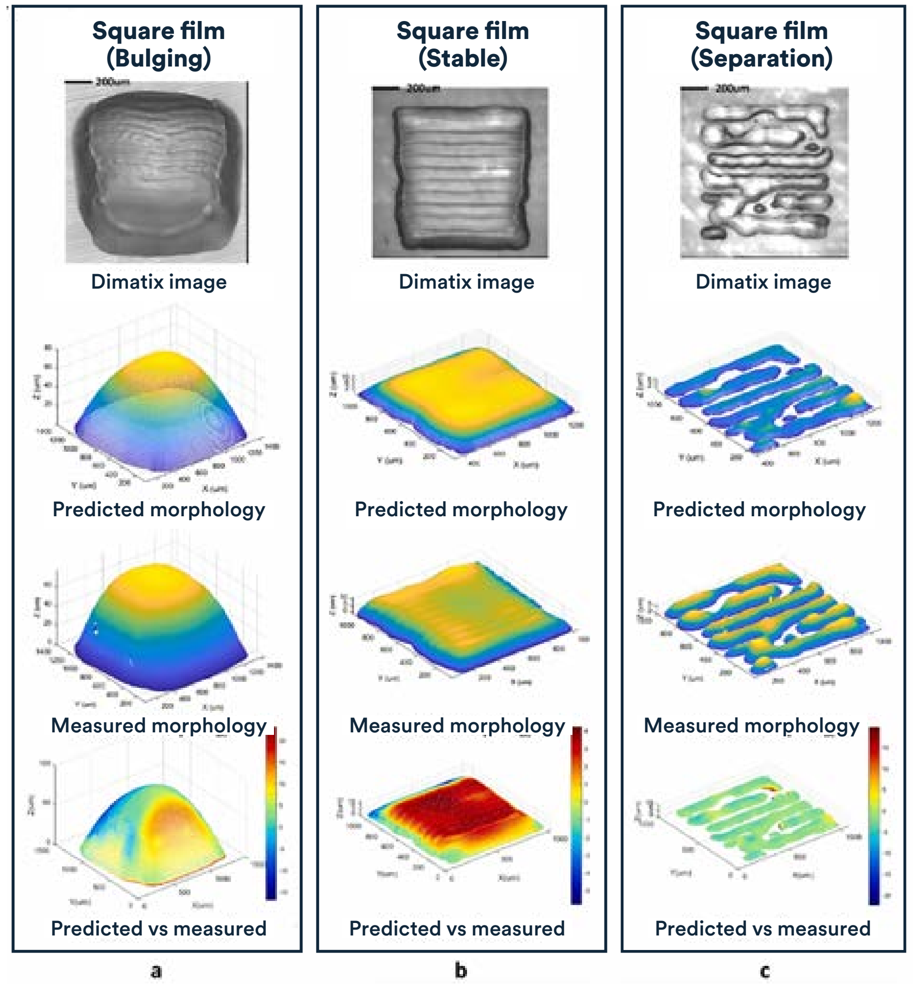


Figure: Reconstructed film, measured morphology, and predicted shape deviations from target to assess the accuracy of the SFS approach.

The inverse problem framework facilitates prediction of the morphology of printed features by utilising a Shape from Shading (SFS) photometric technique coupled with the rapid optimisation of process parameters to enhance the quality of the product. The framework incorporates the critical printing parameters including drop spacing, printing frequency and standoff distance such that we can predict the morphology of complex patterns with an error percentage of less than 10 % in seconds. This framework is highly versatile since it can be utilised in conjunction with computer vision algorithms for accurate in-situ inspection of printed features. This framework can also be utilised to find the optimal printing parameters for freeform patterns, in a fraction of the time needed for experimentation or explicit modelling methods, enabling the consideration of a wide range of different materials and substrates to improve the quality of printed parts.

Computational fluid dynamics (CFD) modelling framework

The development of a CFD framework is among the key outcomes achieved during this PG. This work focussed on the underlying mechanisms of reactive inkjet printing through baseline experiments and theoretical understanding, including the integrated modelling of the core physical and chemical phenomena, such as dynamic wetting, droplet coalescence and photochemical polymer curing.

As part of this project, we developed a framework comprised of seven sub models: (1) fluid dynamics of droplet impact, (2) droplet coalescence and film formation, (3/4) single/multi-layer UV dosing and attenuation; (5) liquid film rheology under UV dosing and attenuation; (6) droplet impact on a partially cured film and substrate using a three-phase model; and, (7) phase change and liquid-solid (solidified liquid) interaction using the three-phase model. This model will increase understanding and enable process optimisation in the jetting of photo-curable polymers.

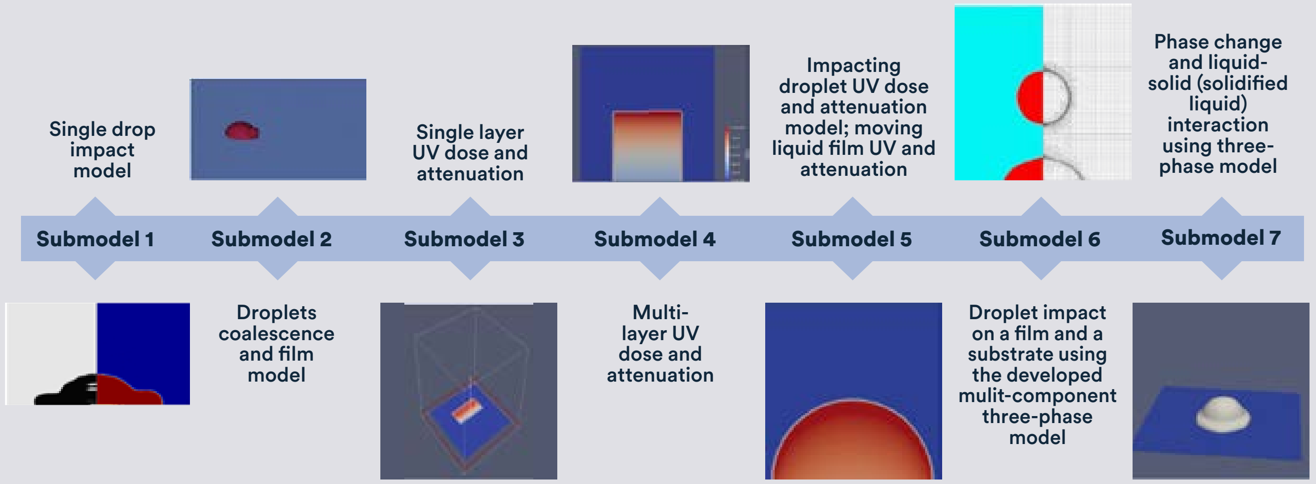


Image: Alasdair Bulloch and Adam Whitbread working on the AconityMIDI+ system.

Advancements in Laser powder bed fusion (LPBF)

Advancements in LPBF have centred around new capabilities to understand, control and manipulate the produced metallic materials, their microstructure and properties. These innovations have been possible due to the recent acquisition through EPSRC Strategic Equipment funding (Grant reference: EP/V029010/1) of an AconityMIDI+, an open multi-laser (2x400W, 1x1kW) LPBF system.

The custom-specification system offers unparalleled flexibility in the processing of both single and multimaterial structures, providing researchers with the ability to conduct cutting-edge research into LPBF. Research into multimaterial LPBF has been made possible through the acquisition of a dual material selective powder deposition recoater developed by Aerosint. This state-of-the-art recoating unit allows for the simultaneous and selective deposition of two separate powders onto the build area.

Advances during this PG will underpin future research on the development of robust bi-metallic parts and customisable functional materials produced through LPBF.

Related publications

MetalJet

Drop-on-demand metal jetting of pure copper: On the interaction of molten metal with ceramic and metallic substrates. *Materials and Design, 2024.*

Additive manufacturing processes for metals. *Book chapter, 2023.*

Quality analysis of additively manufactured metals simulation approaches, processes, and microstructure properties. *Additive manufacturing processes for metals (book chapter), 2023.*

Rolling and sliding modes of nanodroplet spreading: molecular simulations and a continuum approach. *Physical Review Letters, 2023.*

Decreasing contact angles at accelerating three-phase moving contact lines. *Journal of Fluid Mechanics, 2022.*

From impact to solidification in Drop-on-Demand metal additive manufacturing using MetalJet. *Additive Manufacturing, 2022.*

The onset of solidification: From interface formation to the Stefan regime. *Journal of Chemical Physics, 2022.*

Insights into Drop-on-Demand metal additive manufacturing through an integrated experimental and computational study. *Additive Manufacturing, 2021.*

Solidification and dynamic wetting: A unified modeling framework. *Physics of Fluids, 2021.*

Towards digital metal additive manufacturing via high temperature Drop-on-Demand jetting. *Additive Manufacturing, 2019.*

Reactive material jetting of polyimide insulators for complex circuit board design. *Additive Manufacturing, 2019.*

Two-photon

Strategies for integrating metal nanoparticles with two-photon polymerization process: toward high resolution functional additive manufacturing. *Advanced functional materials, 2023.*

The influence of printing parameters on multimaterial two-photon polymerisation based micro additive manufacturing. *Additive Manufacturing, 2022.*

Inkjet

Off the Grid: A new strategy for material-jet 3D printing with enhanced sub-droplet resolution. *Additive Manufacturing Letters, 2024.*

Quantum nature of charge transport in inkjet-printed graphene revealed in high magnetic fields up to 60T. *Nano Micro Small, 2024.*

Stable large area Drop-on-Demand deposition of a conductive polymer ink for 3D-printed electronics, enabled by bio-renewable co-solvents. *Additive Manufacturing, 2023.*

Formulation of functional materials for inkjet printing: A pathway towards fully 3D printed electronics. *Materials Today Electronics, 2023.*

A surrogate modelling strategy to improve the surface morphology quality of inkjet printing applications. *Journal of Manufacturing Processes, 2023.*

Material jetting high quality components via an inverse problem framework. *Additive Manufacturing, 2023.*

Modelling the influence of UV curing strategies for optimisation of inkjet-based 3D printing. *Materials and Design, 2021.*

Inter-flake quantum transport of electrons and holes in inkjet-printed graphene devices. *Advanced Functional Materials, 2020.*

Delivery

4. Applications

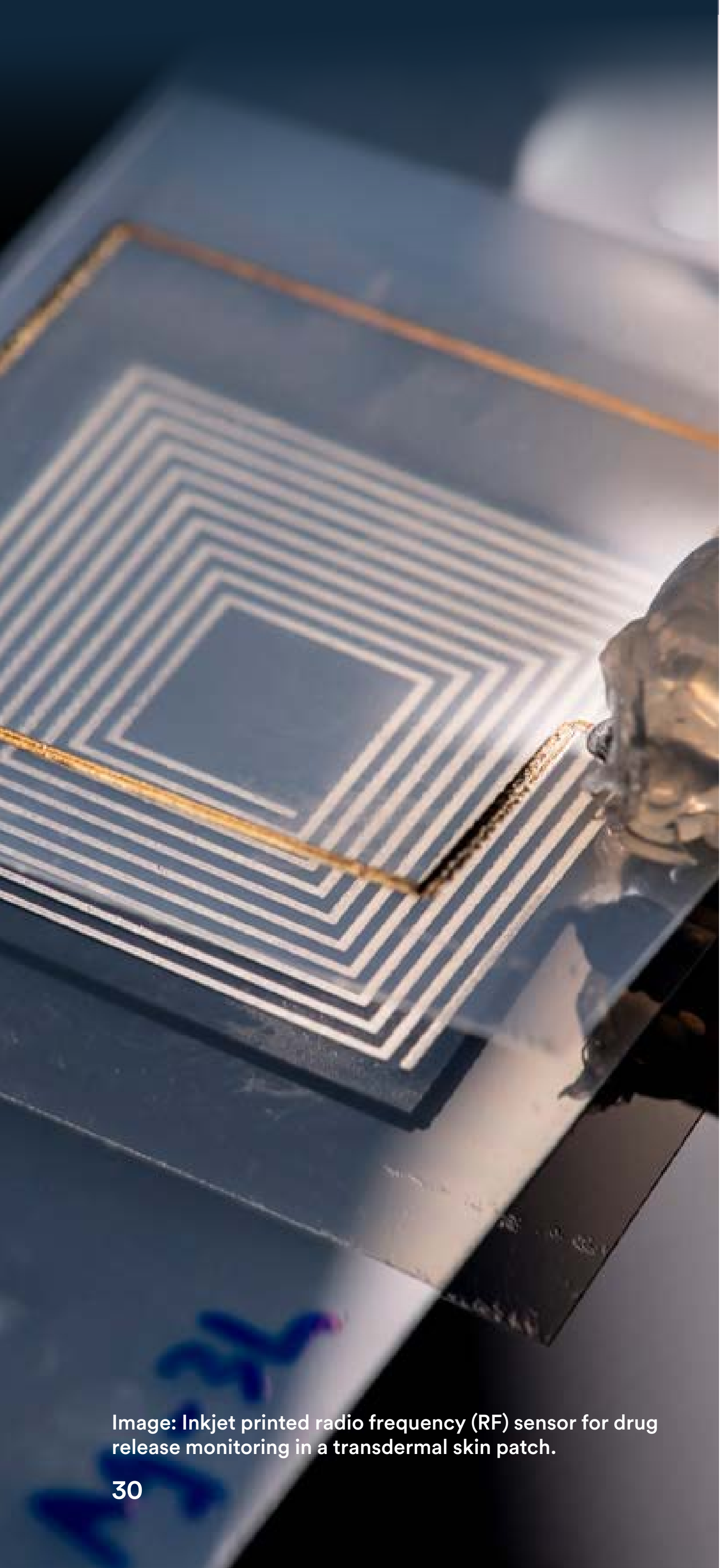


Image: Inkjet printed radio frequency (RF) sensor for drug release monitoring in a transdermal skin patch.

Much of this Programme Grant (PG) has focussed on the development of techniques, materials formulation and understanding via modelling and characterisation. Nonetheless, within the latter half of the Programme we have made significant progress in the development of demonstrators, that is, exemplars have been produced combining the latest advancements used as a proof-of-concept. Earlier demonstrators produced were focussed on healthcare devices and, at the second stage, functional electronic devices and bimetallic parts were also achieved.

Healthcare

The multimaterial additive manufacturing techniques developed in this project represents a radically different approach to creating customised medical devices tailored to individual patient needs. Despite its potential, the adoption of this technology in healthcare is challenged by a lack of in-depth research and understanding in materials, fabrication, and design. Material compatibility with the printing process is crucial, as the absence of well-defined material properties can hinder the production and validation of design toolsets.

To address this, the PG focussed on developing processes and new materials specifically designed for multimaterial printing, aiming to harness their potential in various healthcare applications. This effort is pivotal in advancing the technology's practicality and ensuring its successful integration into medical device customisation, offering promising prospects for personalised treatment and care.

Among the projects developed are included: batch production of customised pills using a six printheads inkjet system; printing cells using a Computed Axial Lithography (CAL) system, experimental replication of shape-like functional units of liver (liver lobules) and bone (osteons) and development of a new biocompatible hydrogel formulation for fabricating flexible wound fillings to deliver analgesics and antiseptics.

Currently, healthcare applications projects have been transferred to the recently awarded PG: *Dialling Up Performance For On Demand Manufacturing*, to continue their development from conception to product idealisation.

Image: 3D inkjet printing of a compacted pill made from a novel reactive prodrug ink that contains four different regions, each with distinct releasing behaviours.

Low dimensional graphene-based optoelectronics

At the early stages of the PG, we successfully developed deposition and post-process strategies for inkjet printing low dimensional graphene-based devices, including a fully printed field effect transistor and electrodes for an InSe photodetector, which both featured a quantum transport behaviour. Electrical characterisation has demonstrated the devices' functionality, which has also been analysed and confirmed by a theoretical model. Device performance is defined by the quality of intermixing, which was confirmed by ToF-SIMS compositional analysis.

These advancements in the inkjet deposition of graphene enable us to produce devices such as transistors, multifunctional sensors, and photodetectors.

A field effect transistor, based on multimaterial stacks of low-dimensional materials, was fabricated by inkjet printing a multi-layer graphene channel with a boron nitride gate and a silver electrode on top. The electrical measurements demonstrated a characteristic gate effect on the electrical conduction. A fully inkjet-printed solar blind UV detector and antennae were also fabricated using graphene ink.

In the past year, a scalable fully inkjet-printed multifunctional sensor matrix which integrated humidity, thermal, and pressure sensors was fabricated using two layers of graphene arrays separated by a dielectric Tripropylene Glycol Diacrylated (TPGDA) layer. The integration of all three sensing capabilities into a single device structure required that each sensor provide output responses to a specific stimulus without being interfered by other stimuli. Additional functions can be achieved by tailoring the sensor geometry, such as monitoring drug release, gas sensing, among others.

We are currently working on the development of inkjet-printed graphene sensors for extreme environments, including high temperature, high pressure and heavy metals detection.

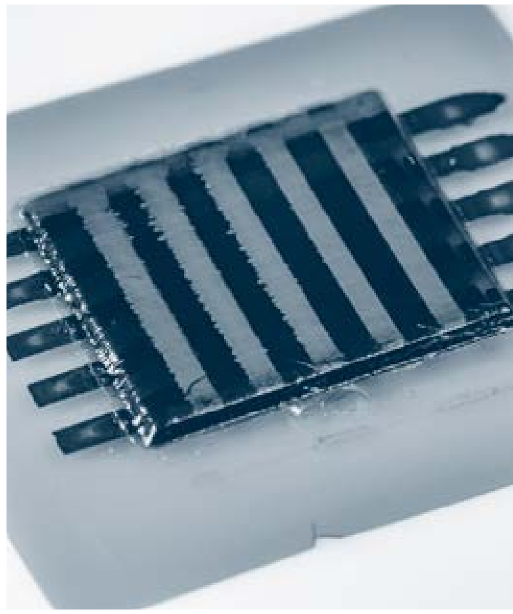
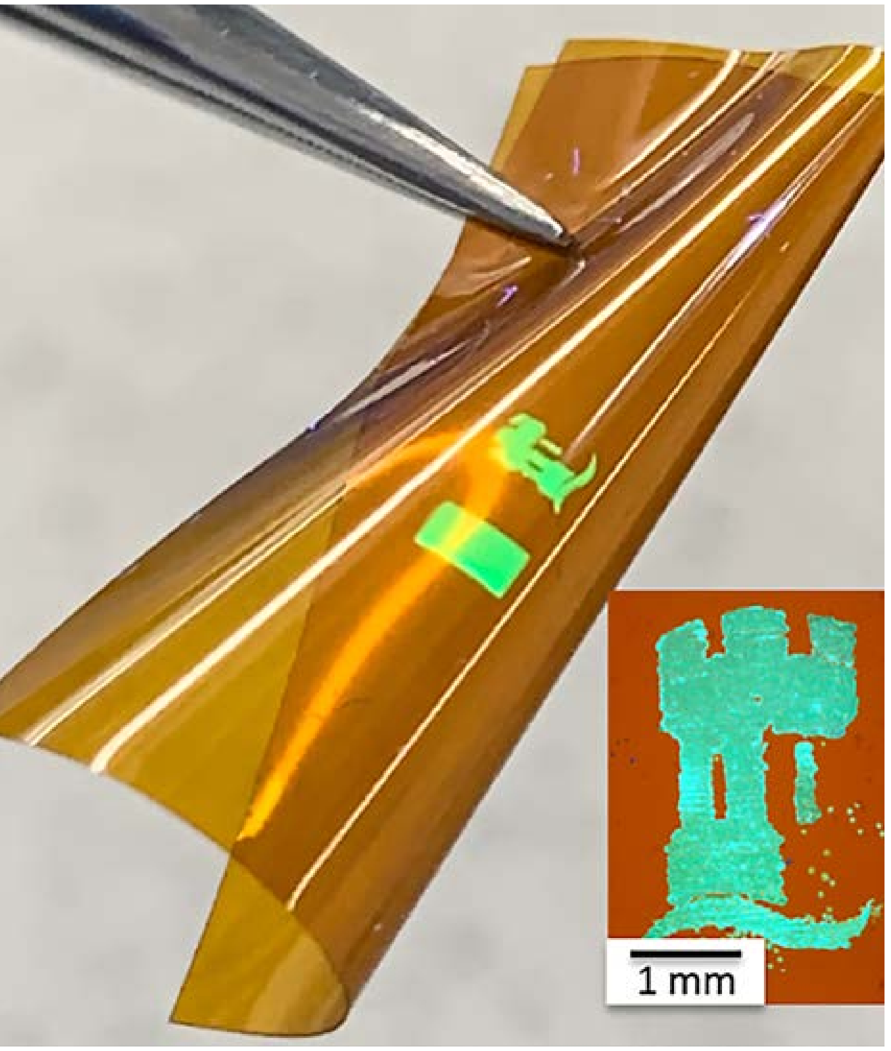


Image: Multifunctional inkjet-printed graphene sensor consisting of graphene layers and TPGDA dialectric medium.

Ongoing research:

A glance at the future of devices in additive manufacturing (AM)



Perovskite LED

Fully printed LED devices have exciting potential in due to the complex and customisable pattens enabled by inkjet printing. However, they have so far proven challenging to fabricate. This is because LED devices require the deposition of several different material layers into heterostructure. Several materials required for the LED are yet to be developed via inkjet printing, each with a uniform thickness of ~ 50 nm.

Towards the goal of fabricating a fully printed perovskite LED, a number of inks were developed, including $CsPbBr_3$ perovskite NCs for the emissive layer, the organic materials poly-TPD, TPBI, and PEDOT:PSS for the charge transport layers, and AuNPs and graphene for the electrodes. All layers of the device were deposited via inkjet deposition on flexible substrates and electrical properties suggest the formation of a *p-n* junction were recorded, which shows that our fabrication technique for this device may be possible with further optimisation.

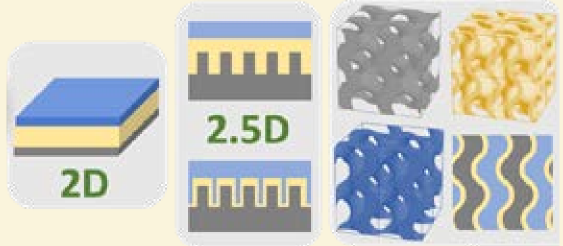
Image: Optical image of printed CsPbBr3 perovskite nanocrystals in the shape of the university logo on flexible transparent Kapton substrate. The NCs display bright green fluorescence under UV (λ = 365 nm) illumination.

Hydrogen storage device

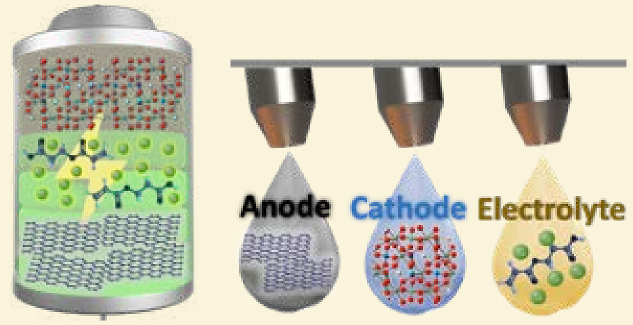
Metal Hydride-based Thermal Energy Stores (MH-TES) can provide highly efficient and promising closed systems for space and water heating. In collaboration with the Advanced Materials Group at the University of Nottingham and the Hydrogen Materials Group at University of Birmingham, Titanium-Vanadium-Magnesium alloy-based nanoparticles have been synthesised by supercritical water hydrothermal method and formulated into inks suitable for inkjet printing. Using multimaterial inkjet printing of advanced designs with interlaced graphene layers to enhance the thermal conductivity, a prototype TES module has been fabricated and tested for hydrogen storage properties, paving the way for a greater range of practical heating applications with significantly higher energy density and no energy degradation in time, lower volume requirements, and good adaptability.

Additive manufacturing of sodium-ion batteries (NIBs)

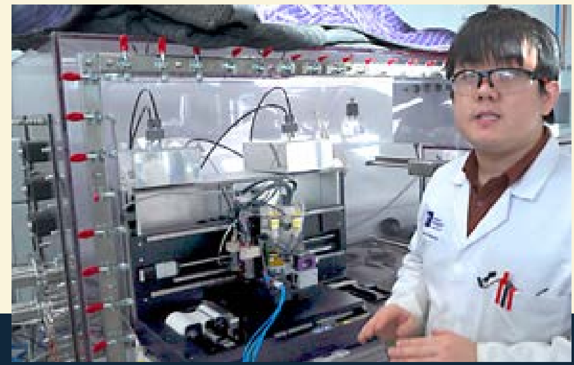
Emerging sodium-ion batteries (NIBs) show promise in complementing lithium-ion battery technology and diversifying the battery market.



Conventional production routes have drawbacks due to limited control to fabricate the solid electrolytes with desired shape and size, impeding their maximum performance.



Additive Manufacturing has the potential to be used for solid state NIBs with high energy density and faster charging, by delivering design freedom with desired thickness, shape, precise control, topological optimisation of complex structure and composition.



Utilising our cutting-edge LP50/Xaar multimaterial inkjet printing system, NIBs can potentially be fabricated which are far smaller, lighter, safer and less expensive to produce than those currently available.

All key materials for a working NIB, including those for the anode, cathode and electrolyte, have been successfully formulated with active material concentration for low-viscosity inkjet printing, achieving a stable printing.

Hard carbon is a promising anode candidate for NIBs, due to its high capacity, sustainability, wide availability, and stable physicochemical properties. In collaboration with Professor Magda Titirici's Sustainable Energy Materials group at Imperial College, we have formulated two inkjet-compatible anode materials for NIBs: one was formulated by dispersing hydrothermal carbonised and subsequent pyrolysed disordered graphite with carboxymethyl cellulose binder solution, and the other was water-dispersed and ethyl cellulose encapsulated graphene oxide with in-situ UV-reduction.

Sodium vanadium phosphate (NVP) was selected as the main cathode material for its three-dimensional diffusivity of Na ions with high structural and thermal stability. Superconductive carbon black as an additive to promote easy charge transport to the NVP. The ink formulation was produced by combining the NVP and the superconductive carbon black with polyvinyludene fluoride binder and N-methyl pyrrolidone solvent. An in-house synthesised UV curable solid electrolyte ink has also been developed by mixing sodium hydrogen carbonate and carboxyethyl acrylate with PEGDA crosslinkers.

Our next steps include carrying out extensive materials and electrochemical characterisation of the inkjet-printed electrodes and electrolytes to study the correlation of structural features with electrochemical performance and assembling anode-electrolyte half cells and cathode-electrolyte half cells and test galvanostatic discharge and charge cycles for fabrication strategy optimisations. On a following stage, we will focus on fabricating fully inkjet-printed NIB fullcell systems with different geometry designs and produce different 3D geometries to compare design and performance.

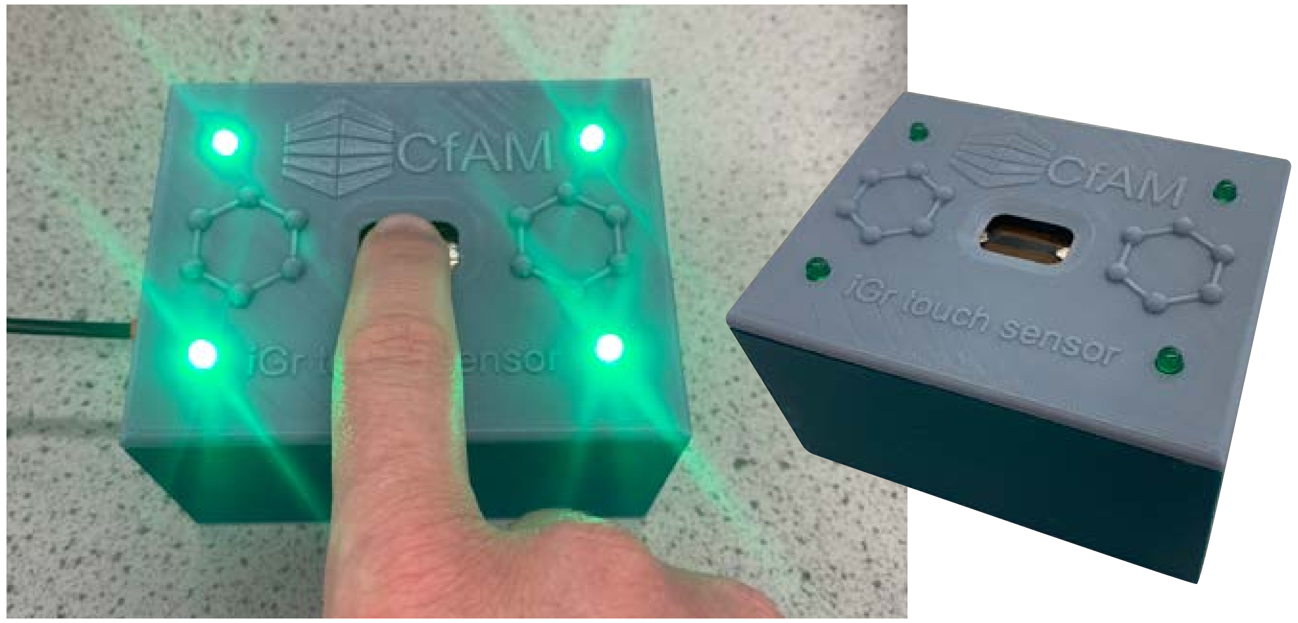


Image: Inkjet printed graphene (iGr) touch sensor displayed in 'on' (left) and 'off' (right) conditions. Graphene was printed onto a Kapton film, with a protective monomer capping layer, and subsequently housed in an FDM produced packaging. The signal produced in response to touch from a human finger was used as a trigger to send power to the set of LEDs.

Printed electronics demonstrators:

Graphene sensor

We have been exploring the use of inkjet-printed graphene as a multifunctional sensing material. When printed in a 2D form, graphene layers are highly responsive to external stimuli such as temperature, pressure and humidity. In this instance, we demonstrate a printed graphene device, which shows a distinct change in resistance when subjected to touch from human skin. The signal received from this contact is used to trigger an LED response.

Graphene-based sensors could also revolutionise sensor manufacturing for extreme environments, enabling next generation of wearable devices as well as manufacturing of sensors onto existing components. We are currently further developing and demonstrating additive manufacturing of multifunctional graphene-based sensors for temperature, pressure and chemical sensing with additional opportunities for photon detection, gas, humidity and magnetic field sensing.

These devices can be used to detect and monitor the environmental factors, potential failures and hazards with improved efficiency.

Photodetector sensor

We are currently working on a device which will showcase the bright fluorescence and sensing capabilities of the printed materials, with potential applications in wearable healthcare devices.

The device, a wearable 'laser warning sign', will be printed on flexible and transparent PEN, with some parts fluorescing red under ultraviolet (UV) light exposure and other parts fluorescing green under near-infrared (NIR) light exposure. Both UV and NIR are invisible to the human eye and potentially dangerous.

To achieve this, we are exploring printing strategies using the $CsPb(Br/I)_3$ perovskite nanocrystal (NC), which displays red fluorescence under UV illumination, and lanthanide-doped up converting nanoparticle (UCNP), which displays green fluorescence under UV light. Both $CsPb(Br/I)_3$ NCs and UCNPs based inks were developed in the PG.

Image: 3D printed bimetallic part using LPBF and 316L stainless steel to provide the structural strength and the CuSn10 acting as the cooling element.

Multimaterial metal printing demonstrator

Differences in the thermal and mechanical properties of the metals being combined can lead to defects at the interface such as porosity, cracking, and delamination. As such, understanding the interface which develops is critical for the development of a robust bimetallic part.

We are currently using laser powder bed fusion (LPBF) to create bimetallic parts. The bimetallic In718 – GRCop-42 samples, are produced using the AconityMIDI+ machine along with the Aerosint dual material recoater. These are observed under a microscope and analytical techniques such as EDX, EBSD and XRD are used to characterise the interface, and understand the effects of different process strategies.

Adding the capability to seamlessly change materials within a single part to the design freedom offered by LPBF could have a significant impact in industry. For instance, assemblies could be printed as a single structure, reducing lead time and the number of processing steps. Alternatively, greater design opportunities are available for existing parts.

Furthermore, selectively distributing the highly conductive properties of the copper alloy GRCop-42 within an Inconel 718 structure (which has exceptional strength and corrosion resistance at high temperatures) is particularly useful for applications operating in extreme environments. Examples include heat exchangers and the combustion chamber of rocket engines.

Related publications

Healthcare

Wireless electrical–molecular quantum signalling for cancer cell apoptosis. *Nature nanotechnology, 2024.*

Quantum leap against cancer. *Nature Chemical Biology, 2023.*

Personalised medicine: manufacturing oral solid dosage forms through additive manufacturing. *Additive manufacturing: materials, functionalities and applications, 2022.*

Bespoke 3D-printed polydrug implants created via microstructural control of oligomers (correction). *ACS Applied Materials and Interfaces, 2022.*

Ink-jet 3D printing as a strategy for developing bespoke non-eluting biofilm resistant medical devices. *Biomaterials, 2022.*

Customisable tablet printing: the development of multimaterial hot melt inkjet 3D printing to produce complex and personalised dosage forms. *Phamaceutics, 2021.*

Bespoke 3D-printed polydrug implants created via microstructural control of oligomers. *ACS Applied Materials and Interfaces, 2021.*

A multiscale optimisation method for bone growth scaffolds based on triply periodic minimal surfaces. *Biomechanics and Modeling in Mechanobiology, 2021.*

A reactive prodrug ink formulation strategy for inkjet 3D printing of controlled release dosage forms and implants. *Advanced Therapeutics, 2020.*

The next pharmaceutical path: determining technology evolution in drug delivery products fabricated with additive manufacturing. *Foresight and STI Governance, 2020.*

Optoelectronics

Surface functionalised graphene for UV-visible photon detection. *NATO STO Journal, 2023.*

Graphene FETs with high and low mobilities have universal temperature-dependent properties. *Nanotechnology, 2023.*

Additively manufactured 3D micro-bioelectrodes for enhanced bioelectrocatalytic operation. *ACS Applied Materials and Interfaces, 2023.*

Photosensitisation of inkjet printed graphene with stable all-inorganic perovskite nanocrystals. *Nanoscale, 2022.*

Magnetic and electric field dependent charge transfer in perovskite/graphene field effect transistors. *Advanced electronic Materials, 2022.*

Surface functionalisation and electroless plating of 3D-printed microstructures. *ECS Meeting abstracts, 2022.*

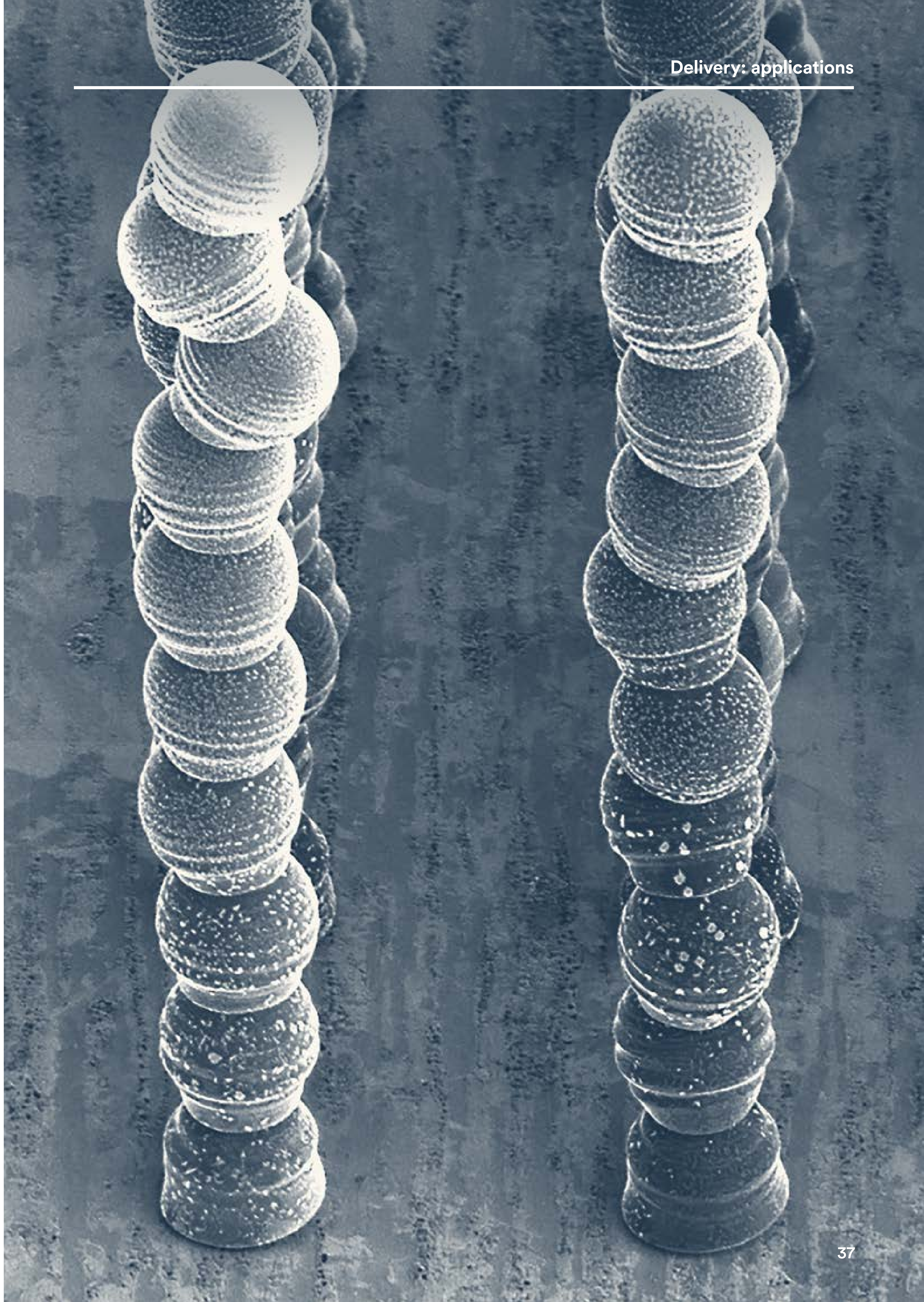
Impact of dielectric substrates on chipless RFID tag performance. *Cambridge University Press, 2022.*

Universal mobility characteristics of graphene originating from charge scattering by ionised impurities. *Nature Communications Physics, 2021.*

Inter-flake quantum transport of electrons and holes in inkjet-printed graphene devices. *Advanced Functional Materials, 2020.*

Tunnel spectroscopy of localised electronic states in hexagonal boron nitride. *Nature Communications Physics, 2018.*

Image: FIB-SEM image of pillars in silver (Ag), printed through the MetalJet platform.



Career progression

During the six years of this Programme Grant, we have had the opportunity to support students and researchers in progressing their careers in both academia and industry. On the following pages are some of the key academic achievements of our researchers and students.

"I began my journey with PG as a PhD student and transitioned to an academic role as the Programme concludes. The PG allowed me to collaborate with and learn from esteemed experts across various disciplines, accelerating my professional development. I'm grateful for this opportunity."

Dr Negar Gilani, University of Nottingham

PhD aligned projects

This programme has provided an important contribution to developing the next generation of researchers in additive manufacturing for the UK and beyond. We have been supporting their development through the progression of PhD students working on PG-aligned projects, providing academic guidance from PG's academic and researcher supervisors, alongside access to the state-of-the-art research equipment. As a result, these PhD students produced valuable inputs to the Programme's progress.

Current PG-aligned PhD projects

Name	Start date	University	PhD title
Nur Rofiqoh Eviana Putri	December 2019	Nottingham	Additive manufacture of vascularised bioactive scaffolds for bone tissue engineering
Oliver Nelson-Dummett	October 2020	Nottingham	3D multimaterial inkjet printing of electrically active materials
Kevin Bandeira	December 2020	Nottingham	Predictive modelling and design optimisation of erodible drug delivery systems
Xiangyun GAO	April 2021	Nottingham	Multimaterial metaljetting: developing a colour metal printer
Nathan Coombs	October 2021	Warwick	Computational modelling of 3D inkjet printing: PEDOT residue patterns
Alasdair Bulloch	October 2021	Nottingham	Design of metal structures of custom composition using additive manufacturing
James Caruana	October 2022	Nottingham	Synthesis of novel high performing polymers for the additive manufacture of bacterial biofilm resistant surfaces
Charles Heaton	October 2022	Nottingham	Inkjet deposition of low dimensional materials for flexible healthcare devices
Ryan McMullen	October 2023	Nottingham	Progressing flow chemistry through the additive manufacturing of novel functionalised systems
Ashley Murray	October 2023	Nottingham	Formulating functional materials for additive manufacturing of flexible electronics

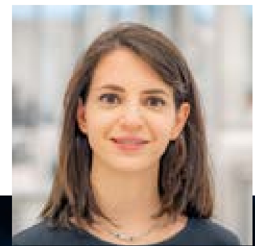
PG-aligned PhD projects completed during this PG

Congratulations!

Name	Viva date	PhD title
Dr Eric Lehder	September 2022	Optimising the geometry of a fracture healing assembly that includes a cell seeded scaffold and a stiffness graded auxetic fixation plate
Dr Jonathan Gosling	November 2022	Electronic properties of graphene-based electronics
Dr Negar Gilani	December 2022	3D inkjet printing of multimaterial pulsatile drug delivery implants
Dr Eva Kingwood	January 2023	3D inkjet printing of multimaterial pulsatile drug delivery implants
Dr Maria Ines Evangelista Barreiros	May 2023	Formulation strategies for the 3D extrusion printing of tablets containing a poorly soluble drug
Dr Kristian Plender	May 2023	Novel approaches to the long-term release of biomacromolecules
Dr Jonathan Austin	November 2023	Additive manufacturing based on hybrid low dimensional 0D/2D heterostructures
Dr Tien Thuy Quach	January 2024	Novel micro/nano scale characterisation of interfaces in multimaterial additive manufacturing
Dr Joseph Sefton	March 2024	Production and use of oligomers in additive manufacturing

Transition into research associate/ research fellow

The work carried out during some PhDs projects set the foundations for further appointments as Research Fellows, and the PG was instrumental in generating these valuable opportunities to advance their research careers. We are extremely proud of the achievements of so many of our students and their continued dedication to excellent research within the group in the past two years.



Dr Anna Lion

Dr Jonathan Gosling

Dr Jonathan Austin, featured right

Dr Negar Gilani, featured above left

"During the PG, I started and completed my PhD, and began my new role as a research fellow. The PG allowed me to work in a highly collaborative team with expertise across disciplines, accelerating my development as an independent researcher."

Dr Jonathan Austin, University of Nottingham

Postdoctoral research associate academic activities

Beyond the support provided for developing their research and attending relevant events in the field, our researchers were encouraged to continue developing their careers. They were provided with opportunities to co-supervise MSc and Nottingham Summer Engineering Research Programme (NSERP) students, and mentored through grant submissions and pursuing academic positions.

Grants

EPSRC Impact Acceleration Accounts	Lead	Award
Proof-of-concept: Development of gold conductive inks for material	Dr Jisun Im	£33,728
Ultrafast volumetric printing of wound fillings for immediate stabilisation of soft tissues	Dr Laura Ruiz-Cantu	£64,324
Enhancing osseo integration of additively manufactured implants by optimising surface architecture	Dr Laura Ruiz-Cantu	£6,939
Accelerated commercialisation of the Nottingham reactive 3D printing process	Dr Yinfeng He	£24,868
Scaling up and optimisation of the printing process for a patented powder-based 3D reactive inkjet technology	Dr Yinfeng He	£62,277

UNICAS-Researcher Academy (RA) Grant	Lead	Award
Direct 3D printing of high-resolution and high-aspect ratio electronics	Dr Negar Gilani	£14,600
Additively manufactured surface-enhanced Raman spectroscopy substrates: a novel 3D sensing platform for on-site, real-time food quality analysis, 2023	Dr Jisun Im	£8,940
Curbing antibiotic resistance: Repurposing systemic antibiotics for localised delivery via microneedles in chronic wound management, 2023	Dr Jisun Im	£2,910
Investigation of the potential cross-contamination in multifunctional additive manufacturing, 2022	Dr Tien Thuy Quach	£7,351
Tea for two-photon polymerisation: functional microdevice fabrication using a novel 3D printing approach for safe and controlled drug delivery, 2019	Dr Elizabeth Clark	£12,133

"The PG has been a wonderful journey for me to carry out interdisciplinary research across physics, materials and manufacturing, not only for the rational scientific discoveries, but also for the joyful experience of working in a collaborative and passionate team!"

Dr Feiran Wang, Research Fellow, University of Nottingham

Organisation	Title	Lead	Award
UKRI Research England Grant (UoN-ODA)	Knowledge exchange for multimaterial additive manufacturing, 2024	Dr Tien Thuy Quach	£20,000
C-Dice Energy Storage Sandpit	Developing next-generation metal hydride-based thermal energy stores – Decarbonising heat in buildings, 2022	Dr Feiran Wang	£29,971
Royal Society of Chemistry	Broadening horizons in the Chemical Sciences Programme, 2022	Dr Tien Thuy Quach	
EPSRC Doctoral Prize	Quantum mechanical approaches in the development of healthcare devices, 2022	Dr Jonathan Gosling	
Redistributed Manufacturing in Healthcare – RIHN	Ultrafast 3D printed personalised wound fillings for immediate stabilisation and regeneration of soft tissues, 2021	Dr Laura Ruiz-Cantu and Dr Yinfeng He	£80,405
iCURE (Innovate UK)	3D reactive jetting technology	Dr Yinfeng He	£15,000
Wellcome Trust	Development of a fatigue detection device to prevent musculoskeletal injuries in football	Dr Laura Ruiz-Cantu	£14,900
Wellcome Trust	Mechano-chemical integration at the osteochondral interface	Dr Laura Ruiz-Cantu	£14,900
São Paulo Research Foundation FAPESP (Brazil)	3D Printing of Micro-Patterned Gaseous Detectors: A route for sensor adaptability and optimisation	Dr Gustavo Ferraz Trindade	£40,000

Master's student projects

Being involved in academic activities and supervision is an important role for researchers' experience and career development. During the whole Programme Grant, researchers co-supervised on average eight to ten Master's student projects per year.

Academic positions

During the Programme Grant, we had the honour to witness the successful transition of some of the research fellows into academic positions. Congratulations to all and all the best for a fruitful continued research journey.

Dr Yinfeng He

Transitional Assistant Professor in Additive Manufacturing.

University of Nottingham (2018-present).

Currently on three-year secondment to University of Nottingham Ningbo Campus.

Dr Laura Ruiz-Cantu

Transitional Assistant Professor in Additive Manufacturing and Biofabrication.

University of Nottingham (2018-2022).

Dr Jisun Im

Assistant Professor in Materials Engineering, School of Engineering.

University of Warwick (2022-present).

Dr Negar Gilani

Assistant Professor of Solid Mechanics.

University of Nottingham (2023-present).

“ My time with CfAM working on this PG has given me the opportunity to establish my career, performing leading research in a world-class research group and facility. Thanks to this opportunity I’m now an Assistant Professor, and I can build on my research during the PG to build my independent research programme and give back to the group. ”

Dr Geoffrey Rivers

Assistant Professor of Functional Materials and Additive Manufacturing.

University of Nottingham (2023-present).

Nottingham Summer Engineering Research Programme students (NSERP) – a step towards the next generation of doctoral candidates

Summer placements were another important activity supported by this Programme Grant. This activity provided students with real-life researcher experience whilst allowing researchers to fully scope, lead and deliver projects aligned to their PG research.

Research title	Name
2023	
Studying the wettability and chemical interaction between molten metal and ceramics	**Afrid Hassan**
Additive manufacturing (3D printing) of multimaterial hierarchical structures for hydrogen storage	**Seyedeh Arefeh Jalali**
Additive manufacturing (3D printing): Towards additively manufactured sodium (Na) ion battery	**Shaheer Khan and Tingwei Deng**
Additively manufactured surface enhanced Raman scattering sensor platform (3D printing)	**Dongshuo Han**
2022	
Rheometry of polymeric materials in 3D printing	**Joseph Green**
2021	
Fabrication of energy autonomous electronic skin	**Xiaohang Cai**
3D printing of implants for cartilage repair	**Carolina Borrelli**
Additive manufacturing of polymeric materials	**Zachary Glassbrook**
Inkjet printing of inverse opal meshes composed of conductive polymers for biocompatible stretchable electrodes	**Noof Al Lawati**
2019	
Improving carbon fibre composite manufacturing processes by characterisation of composite preforms stabilised by inkjet printing methods	**Guy Lawrence**
Formulation of graphene containing ink for 3D printing and electronic device fabrication	**Eoin O’Connor**
3D printed flexible electronic devices: optimisation of the in-line sintering process for 3D inkjet-printed silver nanoparticles	**Adam Balogh**
Exploring routes to overcome functional anisotropy in two-photon lithography based 3D micro/nano fabrication	**Sylvia Tan**
Development of a new manufacturing protocol for the production of a carbon fibre composite: integration of inkjet in carbon fibre composites manufacturing	**Varinder Panesar**
Optimisation of the printing parameters of water-soluble polymers for the development of micro vascular channels	**Joni Wildman**

Knowledge exchange

This Programme Grant (PG) witnessed extensive knowledge exchange activities, evident in broad participation in conferences, outreach events, public lectures and research visits. However, the advent of the global pandemic in 2020 abruptly halted in-person activities, resulting in a shift to predominantly online engagement with fewer opportunities available.

During 2021, academic and industrial activity started to see a slow return. At this point, we were reaching the PG's mid point and needed to accelerate the full return of our engagement activities.

Maintaining our firm commitment to knowledge exchange activities, we developed a two-year deliverable plan comprising of the key areas: policy engagement, media appearances, outreach and STEM engagement, industrial collaborations and academic activities. This strategic framework enabled a swift recovery, culminating in a surge of activity throughout 2023 and early 2024. The scope and breadth of activities not only rebounded to prepandemic levels, but they surpassed them, fostering robust engagement across all sectors. This confirms our strong commitment to deliver to our stakeholders.

^ Policy engagement

A significant highlight of our policy engagement activity was hosting a visit from Nottingham South's Member of Parliament, Lilian Greenwood. Currently Shadow Minister for Arts, Heritage and Civil Society, Lilian came accompanied by local councillor, Pavlos Kotsonis. During their visit, they attended a tour of the Centre for Additive Manufacturing (CfAM) lab facilities and learned how the centre is developing additive manufacturing technology for some of the UK's leading companies, such as Astra Zeneca, Rolls Royce and Canon.

During this Programme Grant we have strengthened our research leadership position and have influenced policy through several engagement activities with policymakers, including workshops, rapid advice, contribution to national consultations, as well as scientific advisory board roles assumed by the team of academics.

< Public good

A key moment in our journey was our contribution to fight against the Covid-19 pandemic, when we designed, produced and delivered 5,000 face shields to Nottingham's NHS. The face shields successfully passed the BSI tests and were CE approved for use as part of PPE for healthcare workers' protection in hospitals.

< Outreach activities

As part of the PG's delivery plan and our drive to promote STEM disciplines, we developed targeted materials to school aged kids.

Since our first year, the team has participated in several dynamic and impactful STEM outreach activities, including Pint of Science, an annual science festival that aims to communicate contemporary scientific developments to the public; Family Discovery Day, a free event for families at University Park, Nottingham; Faculty of Engineering Christmas Lecture at the University of Nottingham; Nottingham Festival of Science and Curiosity; Real Science in Schools Symposium; and Science in the Park.

Over the past year, in steadfast dedication to our commitment to STEM outreach, we have delivered an array of engaging activities, highlighted by our participation in the prestigious Royal Society Summer Exhibition and several attendances to local STEM events.

We have developed a specialised educational toolkit for kids, a card game for teenagers, and engaging storytelling around the different materials, processes, and devices. This was complemented by a cohesive visual identity tailored to a younger audience and a curated selection of 3d printed pieces of a range of different shapes, sizes and levels of intricate detailing. These efforts ensure meaningful engagement with younger audiences in the realms of science and technology and encourage a passion for science across diverse communities.

In the media >

In recent years, the PG has seen a sharp increase in media presence, with numerous appearances across various platforms including podcasts, magazines, newspapers, and online media outlets. Some examples of outlets include:

- TCT Additive Insight, podcast and print edition article, February 2024
- The Observer
- Professional Engineering magazine
- Mail Online, tier 1 coverage
- 3DPOD podcast interview

Through these engagements, we have effectively amplified our message, reaching audiences far and wide to highlight the significance of our work. These appearances have driven awareness and understanding of the importance and potential of additive manufacturing within broader communities.

< Industrial activities

Maintaining our focus on to fostering successful industrial collaborations, our engagement with business and industrial partners has yielded significant results over the years.

From collaborations with partners and supporting organisations, including AstraZeneca, the National Physics Laboratory, Lawrence Livermore National Laboratory, Canon OCE and Texas Instrument, to newly developed strategic partnerships with BAE Systems, AWE, Qinetiq and ITP Aero.

These partnerships have driven innovation to joint initiatives, whilst solving industry challenges and leveraging our expertise to create mutual value. Notable highlights include the successful execution of projects covering novel printing of functional materials.

Academic activities >

Over the years, members of the PG have had recurrent and continued opportunities to disseminate our cutting-edge research to an academic audience, including several invites to act as guest speakers, keynote speakers, plenary speakers, technical committee members and technical networks.

During this time, PG researchers and academics attended a number of international conferences, delivering presentations and poster sessions. Some of the most relevant additive manufacturing annual conferences, including the Solid Freeform Formulation Symposium, Additive International and the Materials Research Society Meeting and Exhibit, had prominent participation from PG academics and researchers.

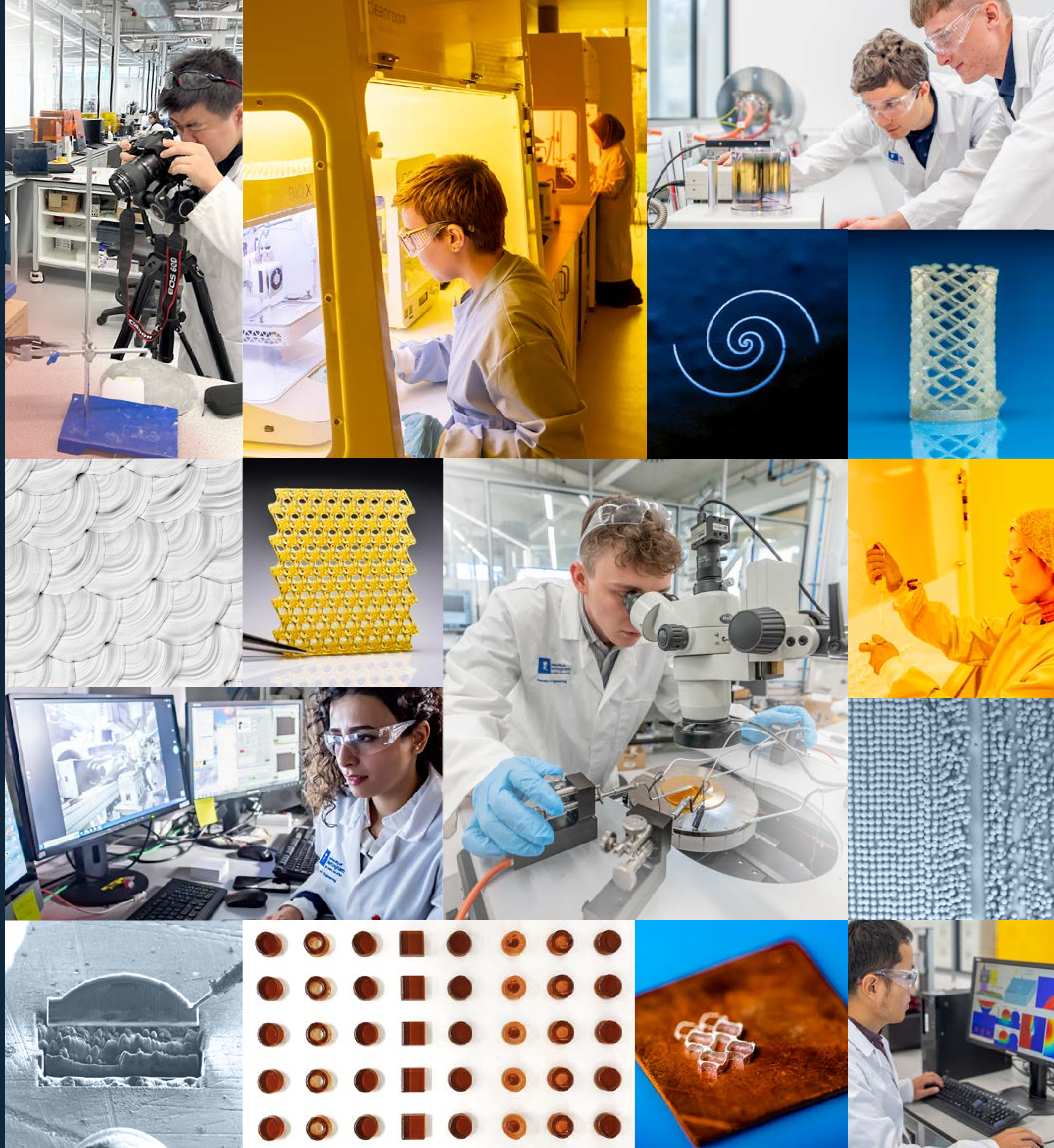
For a full list of conference attendance, please visit: nottingham.ac.uk/research/groups/cfam/major-epsrc-funding

The future

As we conclude our Programme Grant, it is with a sense of accomplishment that we reflect on the transformative journey we have undertaken together over the past six years. During this time, we have supported the professional journey of many researchers and students, and it has been a pleasure to follow their progress to industry and academia. This certainly represents a cornerstone of our legacy beyond our significant academic contributions.

The future ahead is promising. During this Programme we set the foundations for the next generation of additive manufacturing. Latterly adopting a device-led approach, we have shown that – using specific applications upon which to focus our process, materials and computational research activity – we can clearly demonstrate the potential for multimaterial, multifunctional Additive Manufacturing (AM) across our target application areas of healthcare devices and 3D electronics. We have intensified our focus on extracting impactful outcomes in this area, marking the trail for future advancements. Our conviction in this strategy, particularly in the realms of multifunctional materials, and the evolving domain of devices in 3D electronics, clean energy and extreme environments, positions us well for future achievements.

As we officially close this Programme, our commitment to retaining our talented researchers and preserving our standing as industry leaders persists. We will continue to actively pursue funding opportunities from EPSRC and strategic partners, whilst deepening our collaboration with esteemed associates. Our dedication also extends to the continued dissemination of our impactful work through high-impact journal publications, as well as engaging with audiences using different channels. Though this final report marks the end of a chapter, it is also the beginning of a lasting legacy of this transformative multimaterial, multifunctional approach to additive manufacturing. We thank each advisory board member, contributor, collaborator, and partner for their invaluable role in making this a significant journey in additive manufacturing.



Governance

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René Waarsing
Canon Production Printing

Our partners and supporters

We thank all our partners who have supported the delivery of the Enabling Next Generation Additive Manufacturing Programme, including our funder EPSRC, industrial collaborators, allied research institutions and technology organisations. Our collaborative approach enabled co-creation and co-delivery of transformative solutions in the field of additive manufacturing.

Professor Christopher Barner-Kowolik
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