University of Nottingham
UK | CHINA | MALAYSIA

UKRI Engineering and Physical Sciences Research Council

EPSRC Programme Grant

Enabling Next Generation Additive Manufacturing

Annual report 2021

University of Birmingham

Warwick
The University of Warwick

Foreword

Richard Hague
Principal Investigator

After completing the fourth year of this EPSRC Programme Grant (PG), Enabling Next Generation Additive Manufacturing, we are pleased to present our achievements during 2021.

Our Programme is based within the Centre for Additive Manufacturing (CfAM) at the University of Nottingham, with the University of Warwick and the University of Birmingham as partner institutions alongside a host of other academic and industrial partners. This 5-year Programme was launched in 2018, thanks to the support of £5.8 million from EPSRC and during this Programme, our focus has been on the development of next generation, multi-material additive manufacturing through strategic fundamental research. Overall, our vision is to drive disruptive change, rapid development and adoption of next generation additive manufacturing by establishing the fundamental knowledge and advanced methods of control to enable targeted 3D multifunctionality.

Despite the external scenario of Covid-19 our Programme team has fully resumed all lab-based activities and has made impressive progress on the four Research Challenges, continuously developing and implementing novel approaches to solve the challenge of controlled deposition of both functional and structural materials. The activities undertaken in 2021 were based on a review of the work carried out in the first half of its operation to ensure that the PG can maintain its strong momentum and success, as recognised by the overall score of 9 out of 10 in our mid-term review.

Building on the initial work of the PG, this year's research has seen strong progress within the core areas of modelling, interfacial analysis and multi-material processing of electronic, pharmaceutical and biological materials that leaves our team well-positioned to achieve significant impact in the remaining time that we have. I would like to sincerely thank the investigators, researchers, PhD students and support team for their dedication in keeping up the momentum of the Programme, as well as our advisory board for their guidance and EPSRC for their continuous support.

In the remaining time of the Programme, as indicated in our mid-term review, we will be using the manufacture of demonstrator devices to further drive the direction of research, particularly focusing on the core themes of 3D electronic, biomedical and pharmaceutical based devices that are of relevance to our industrial partners. These activities will be underpinned by the advancements made in materials analysis, modelling and processes that have been executed within our Research Challenges over the course of the PG.

Overall, we see our contribution as helping shape the future of UK additive manufacturing research strategy, where we are honoured to be acting on behalf of the UK academic science and engineering communities, whilst actively engaging with other EPSRC centres and leading research groups across the UK and globally.

I invite you to look at our successes, our initiatives and the work done in the past year!

Contents

Foreword by Richard Hague	**2**
Meet the team	**4**
Technical highlights of year four	**6**
Research Challenge 1	6
Research Challenge 2	10
Research Challenge 3	14
Research Challenge 4	18
Projects completed 2021	**22**
Aligned doctoral research	**24**
Outputs	**30**
Conference participation	30
Publications	32
Funding	34
Outreach events	38
The year ahead	**40**
Management structure	**42**
Our partners and collaborators	**43**

Cover image:
Jonathan Austin using 4-terminal micropositioners to measure conductivity of printed gold

Meet the team

Investigators

Professor Richard Hague

Professor of Additive Manufacturing

Director of the Centre for Additive Manufacturing, Faculty of Engineering, University of Nottingham

Principal Investigator and Joint RC1 Lead

Professor Clive Roberts

Professor of Pharmaceutical Nanotechnology

Head of the School of Pharmacy, University of Nottingham

Co-Investigator and Joint RC1 Lead

Professor Ian Ashcroft

Professor of Mechanics of Solids

Head of the Department of Mechanical, Materials and Manufacturing Engineering, Faculty of Engineering, University of Nottingham

Co-Investigator and Joint RC2 Lead

Dr James Sprittles

Associate Professor, Mathematics Institute, University of Warwick

Co-Investigator and Joint RC2 Lead

Professor Ricky Wildman

Professor of Multiphase Flow and Mechanics, Faculty of Engineering, University of Nottingham

Co-Investigator and Joint RC3 Lead

Professor Derek Irvine

Professor of Materials Chemistry, Faculty of Engineering, University of Nottingham

Co-Investigator and Joint RC3 Lead

Professor Chris Tuck

Professor of Materials Engineering

Associate Pro-Vice Chancellor for Research and Knowledge Exchange and Director of the EPSRC Centre for Doctoral Training in Additive Manufacturing and 3D Printing, Faculty of Engineering, University of Nottingham

Co-Investigator and Joint RC4 Lead

Professor Mark Fromhold

Professor of Physics

School of Physics and Astronomy, University of Nottingham

Co-Investigator and Joint RC4 Lead

Professor Yulii Shikhmurzaev

Professor of Applied Mathematics, School of Mathematics, University of Birmingham

Co-Investigator

Dr Lyudmila Turyanska

Associate Professor, Faculty of Engineering, University of Nottingham

Co-Investigator

Researchers

Dr Yinfeng He

Transitional Assistant Professor, Faculty of Engineering, University of Nottingham

Dr Laura Ruiz Cantu

Transitional Assistant Professor, Faculty of Engineering, University of Nottingham

Dr Jisun Im

Research Fellow, Faculty of Engineering, University of Nottingham

Dr Geoffrey Rivers

Research Fellow, Faculty of Engineering, University of Nottingham

Dr Feiran Wang

Research Fellow, Faculty of Engineering, University of Nottingham

Dr Mykyta Chubynsky

Research Fellow, Mathematics Institute, University of Warwick

Dr Peng Zhao

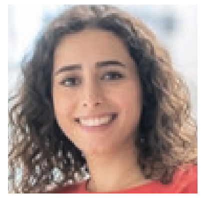
Research Fellow, Faculty of Engineering, University of Nottingham

Dr Carlos Galeano Rios

Research Fellow, Faculty of Engineering, University of Birmingham

Dr Anna Lion

Research Fellow, Faculty of Engineering, University of Nottingham



Negar Gilani

Research Associate, Faculty of Engineering, University of Nottingham

Research affiliates

Dr Marco Simonelli

Assistant Professor, Faculty of Engineering, University of Nottingham

Dr Nesma Aboulkhair

Lead of Additive Manufacturing, Technology Innovation Institute (TII)

Dr Gustavo Ferraz Trindade

Senior Research Scientist (NiCE-MSI), National Physical Laboratory (NPL)

Professor Ian Gilmore

Head of Science, National Physical Laboratory (NPL)

Professional and managerial staff

Flavia M. G. Villarroel

Programme Manager, Faculty of Engineering, University of Nottingham

Mark East

Senior Research Technician, Faculty of Engineering, University of Nottingham

Technical highlights of year four

Research challenge 01

The primary challenge for Research Challenge 1 (RC1) is to understand, at the micro or nano scales, the spatio-temporal interface/interphase evolution between successively deposited droplets or voxels.

Through precision experiments and development of new methodologies for ex-situ materials analysis, we have been solving some of the challenges to enable the inter and intra layer coalescence/bonding for functional-structural or functional-functional materials. One of the primary challenges in multi-material deposition is the potential differences in the co-deposited materials' physical state, chemistry and temperature at deposition or conversion.

Image: FIB-SEM image of a 3D structure in Ag fabricated with the MetalJet platform

In the first year, we established appropriate micro or nano scale analytical methods for chemical and physical properties of interfaces within samples, in particular utilising the 3D OrbiSIMS facilities at the National Physical Laboratory (NPL) and the University of Nottingham that enable exceptional spatial and chemical resolution. In the second and third years, we have implemented and integrated these methods across the Programme, thereby informing modelling (RC2) and build-optimisation strategies (RC3 and 4).

RC1's work has overall strengthened the internal collaborations with all other Research Challenges. This includes the study of commercial formulations and the development of new metallic nanoparticle ink formulation within RC4, alongside experiments related to multimaterial deposition that have helped progress RC3 activities, supporting both the chemistry and manufacturing aspects. Many of these collaborations have resulted in publications in the past year (see further details on the publications list).

Key findings and activities include:

- A study that uses a series of analytical methods to show that residual polymer stabiliser causes anisotropic electrical conductivity during inkjet printing of metal nanoparticles. These results inform new approaches across the RCs, including new nanoparticle sintering strategies, new nanoparticle ink formulations and applications with low-dimensional materials for electronic devices
- A collaboration with RC2 researchers on the influence of UV curing strategies to optimise inkjet 3D printing of organic materials. The RC1 team has developed a method to systematically measure the 3D distribution of vinyl group consumption of a printed material using Raman microscopy. These results were used to validate and create a predictive model for inkjet 3D printing
- In collaboration with RC3 researchers, the 3D OrbiSIMS instrument and advanced data analysis were used to study the interface between organic materials in 3D inkjet printed bacterial biofilm resistant composites
- 3D chemical imaging, using ToF-SIMS, was able to identify the distribution of polymers and drugs in 3D-printed polydrug implants developed in RC4
- High resolution mass spectrometry imaging and Raman microscopy was used to investigate the influence of printing parameters on multi-material two-photon polymerisation AM
- A comprehensive chemical analysis was undertaken to understand the role of additives within a new bespoke gold nanoparticles ink formulation developed in RC4 for improved cohesion and electrical conductivity
- Benchmark definition for morphology analysis of printed tracks was undertaken using focused ion beam scanning electron microscopy (FIB-SEM), profilometry, atomic force microscopy (AFM), and other techniques, before investigating material/substrate interfaces with scanning/transmission electron microscopy (SEM/TEM)
- Advancements on Atomic Force Microscope (AFM) characterisation for Low D materials
- Development of a MATLAB tool for the analysis of optical profilometry data
- Simulation of ToF-SIMS data generation for the correction of artefacts in 3D chemical maps of complex geometries (typically produced via inkjet printing or two-photon polymerisation)
- Analysis of degradation radiation detectors used in high energy physics experiments at CERN as a benchmark for the future analysis of 3D printed detector prototypes in collaboration with University of Sao Paulo and CERN
- Publication of findings related to the MetalJet thermomechanical behaviour of high-temperature metallic microdroplets during solidification and cooling using Cu as droplet and substrate

Ongoing projects

Materials interfaces in inkjet-printed organic electronic devices

Due to intermixing during inkjet printing, the interfaces of heterojunctions in electronic devices can have thickness variations and material gradients, resulting in loss of efficiency and responsivity. In collaboration with Programme partner, National Physical Laboratory (NPL), we are investigating inkjet-printed interfaces of functional materials using ToF-SIMs, FIB-SEM, and FIB-TEM to excavate and map the printed interfaces at the sub-micron scale. Understanding how the ink formulation, printing strategies, and post-treatments interact to produce various interfaces is critical to the development of high efficiency digital and optoelectronic devices. Following this phase, we will extend these protocols into characterisation of inkjet-printed 3D circuits, in which digital devices are layered to increase device density; a challenging scenario for maintaining well-defined interfaces at all z-axis layers.

Detection of organic residues that hinder biocompatibility of stents printed via multi-material 3D inkjet printing.

Supporting materials are an indispensable component for constructing complex 3D geometries in the multi-material 3D inkjet printing process. They are normally printed next to, or under, the structural and functional materials to provide temporary support for overhanging geometries and are washed away after processing is complete. However, it is apparent that such support materials can lead to surface contamination on the final product, and it is therefore necessary to investigate this to understand the influence on both mechanical and functional performance. For example, such contamination could impact on the printed functional devices of which its surface chemistry is important to its functionalities, e.g. bacterial biofilm resistance or biocompatibility of the printed medical devices. Therefore, are deploying advanced characterization techniques (e.g. 3D OrbiSIMS) to help investigate, understand and therefore solve such contamination issues, thereby leading to more reliable device performance.

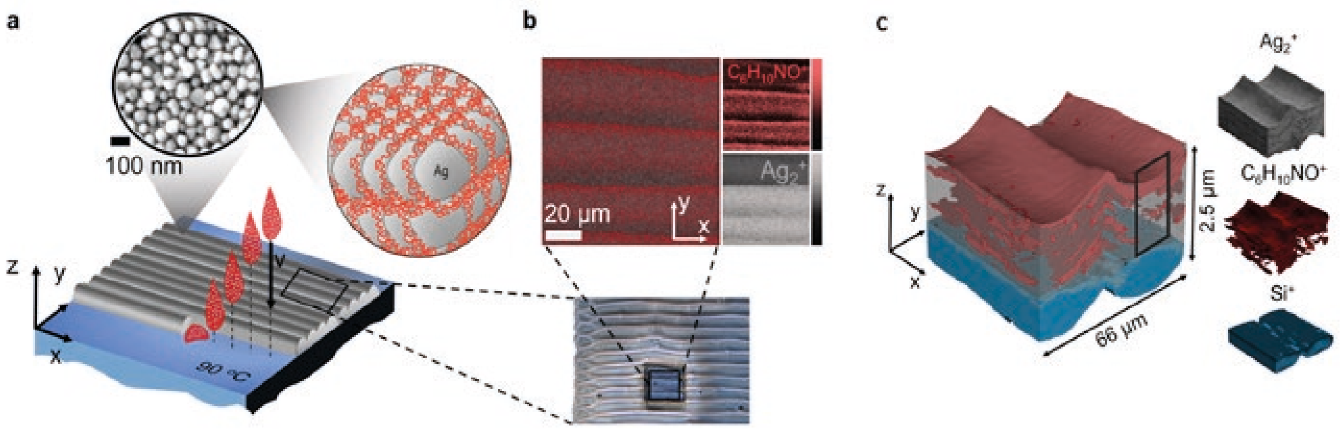


Image: Digital inkjet printing of inks containing metal nanoparticles with in-situ solvent evaporation (pinning) (a) and optical and chemical images of a printed layer of silver nanoparticles showing organic residues at the surface (b). Distribution of organic stabiliser residues within 3D printed layers of silver nanoparticles (c)

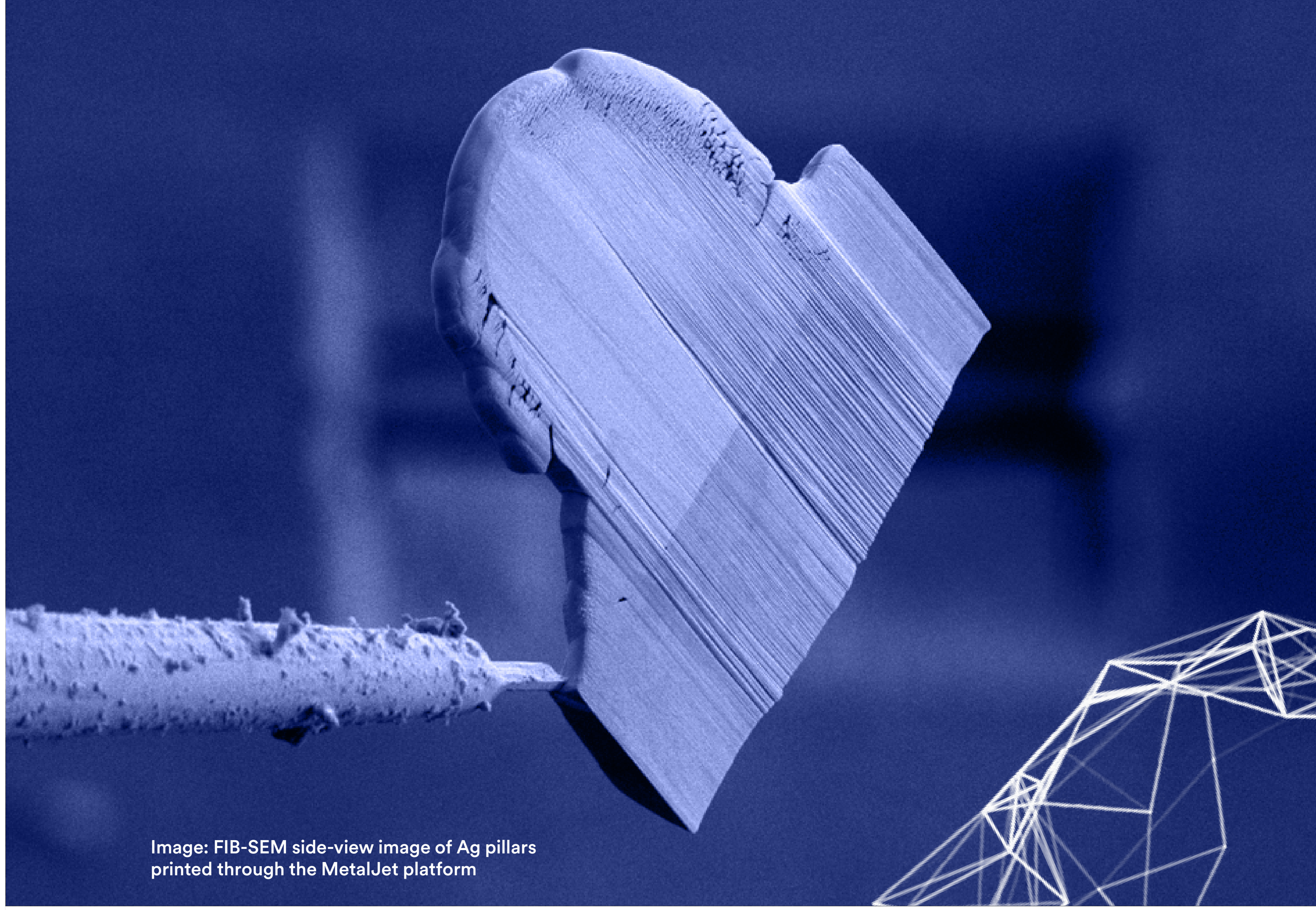


Image: FIB-SEM side-view image of Ag pillars printed through the MetalJet platform

Nanocomposite metamaterials fabricated using two photon polymerisation and micro stereolithography

We have previously demonstrated that nanocomposite metamaterials containing plasmonic gold nanoparticles can be fabricated by two photon polymerisation. Notably gold nanoparticles can be selectively deposited on target regions to create functional nanocomposites by tailoring resin functionality. Selectively gold nanoparticles-decorated 3D structures were confirmed using ToF-SIMS 3D chemical imaging. We will continue to investigate the characterisation of gold nanoparticles on the surface and within nanocomposites at macro/micro/sub-micron scale using focused ion beam scanning electron microscopy (FIB-SEM) in combination with transmission electron microscopy (TEM). The effect of gold nanoparticles on polymerisation during 2PP process will be also investigated using Raman spectroscopy.

MetalJet: droplet-substrate interface characterisation

Understanding the underlying physics of interface formation between the droplet and substrate and between successively deposited droplets in the MetalJet process is an essential requirement of understanding how to successfully construct 3D objects, since it sets the consistency of printed parts through this process. However, under certain sub-optimal processing conditions, MetalJet droplets can be weakly bonded to the substrate or other droplets, therefore rendering traditional metallurgical preparation methods unsuitable. Consequently, we will further focus on using stress-free Focused Ion Beam (FIB) milling to cross section droplets and their interfaces/interphases with both the substrate and each other. Post FIB milling, we will perform characterisations such as FIB-SEM, EDX (Energy Dispersive X-Ray Spectroscopy), EBSD (Electron BackScatter Diffraction), and TEM (Transmission Electron Microscopy). Following this phase, we will be developing a serial FIB milling to reconstruct the 3D microstructure of individual droplets and their interface with the substrate and each other.

Image: Dr Peng Zhao carrying out AM modelling research

Research challenge 02

This research challenge aims to develop a multifunctional Additive Manufacture Computational Framework that will guide the manufacturing strategies to be employed to create functional objects and is delivered across all the academic partners at Warwick, Birmingham and Nottingham.

RC2 provides underpinning understanding across all processes, with a framework that leads to greater design capability for multifunctional additive manufacturing. It has been collaborating closely across the other more experimentally driven RCs, both at the micro scale in RC1 and the device level scales of RC3 and 4 that is giving the Programme a strong, multi-scale understanding.

Progress on modelling of inkjet processes:

- Development of a systematic descriptive and predictive model to determine and optimise the quality of the inkjet-based 3D printing, incorporating critical processing parameters, such as UV source pathway, UV intensity, printing strategy, and interlayer attenuation. The model can predict the degree of ink conversion throughout the product and can be used to generate control over the material microstructure, as well as guide an optimised build strategy
- Development and validation of the CFD framework to represent the process of deposition, consolidation and solidification during inkjet-based 3D printing
- Development and validation of a molecular dynamics to in silico screen viscosity of potential 3D printing inks
- A new model to capture transport mechanisms in ink jet printed graphene

Progress on MetalJet processes:

- Overcoming singularity issues in the computational codes to adapt the modelling to both spontaneous and forced spreading
- Incorporation of unsteady temperature distributions into the thermo-mechanical modelling of droplets
- Dynamic Wetting: numerical implementation of seamless transition between methods for acute and obtuse contact angles with interface formation effects and new implementation of mass balance equation at the contact line, leading to increased robustness of the method
- Solidification: 1D-model implementation of a unified framework for solidification and interface formation effects

Progress on two photon polymerisation processes:

- Development of a kinetic model to accommodate spatial variation in two photon polymerisation-based manufacture
- Creation of a coarse-grained model to be able to rapidly simulate many voxels
- Interaction with Nanoscribe to be able to adopt model into a commercial environment

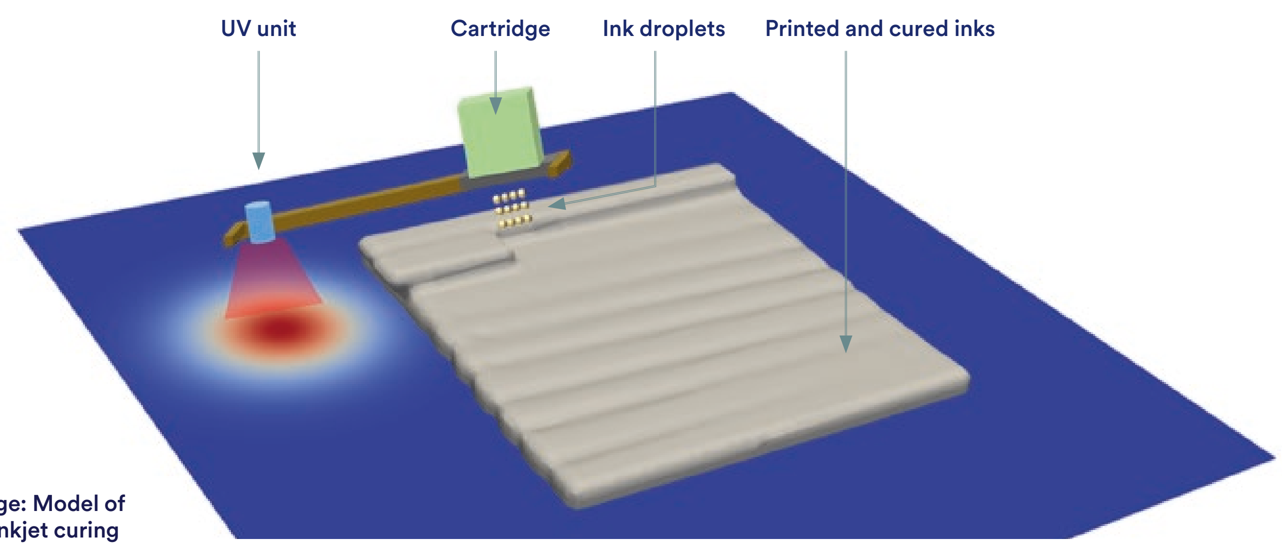


Image: Model of 3D inkjet curing

Ongoing projects

Inkjet: molecular dynamics model to predict viscosity and reactivity of 3D printing inks

A bottleneck in identifying whether materials are suitable for 3D printing, is determining whether they have the right rheology and reactivity to support manufacture in suitable timeframes. We propose a first step, where we will develop a molecular dynamics model that will be used to identify the viscosity of a given candidate ink. This model will use Green-Kubo relations to be able to extract viscosity of *in silico* described inks. As we proceed, we will move from validation to prediction, allowing us to predict ‘printability’ of previously unknown materials and screen the molecular space rapidly.

Two-photon polymerisation (2PP): towards full-structure simulations

Our previous extension of a single-voxel model of two-photon polymerisation (2PP) makes it possible to easily simulate just a few dozen / hundreds of voxels.However, real-life structures produced by 2PP 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, but are not feasible at full resolution. 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 some interest from industry. While the immediate plan is to publish these results, we intend to continue interacting with experimentalists, both within the Program Grant and outside, to further develop and validate our methods.

MetalJet: modelling the microstructure evolution during the droplet deposition

The microstructure of individual droplets partially sets the final properties of printed parts through DoD-MJ; hence it is important to understand how the microstructure is developed to better control it. Droplets solidify from the interface with the substrate or previously deposited droplets, and concurrently the grains nucleate and grow from the solidified sections to form the final structure of the part. The process can be simulated by combining the temperature field obtained from previous thermal models and setting metallurgical rules. The CAFE (Cellular Automata Finite Element) modelling approach will be used to simulate the microstructure evolution.

MetalJet: dynamic wetting with heat transfer and phase transitions for drop-on-demand additive manufacturing

The shape of solidified droplets, in drop-on-demand additive manufacturing, depends mainly on how much these droplets spread before they solidify. This, in turn, depends on the interaction of dynamic wetting with heat transfer and phase transition effects. Consequently, models that can effectively inform innovation must integrate all these effects.

The interface formation model for capillary flows is the natural foundation for such a modelling framework, and the first big challenge is to put together, in a single implementation, all of its features and the adaptations needed to deal with singularities that appear in the flows of our interest. This task is well advanced, and yielding its first complete results. Ongoing work includes expanding and publishing the outcomes of the dynamic wetting advances and, subsequently, initiating the inclusion of heat transfer and phase transition effects.

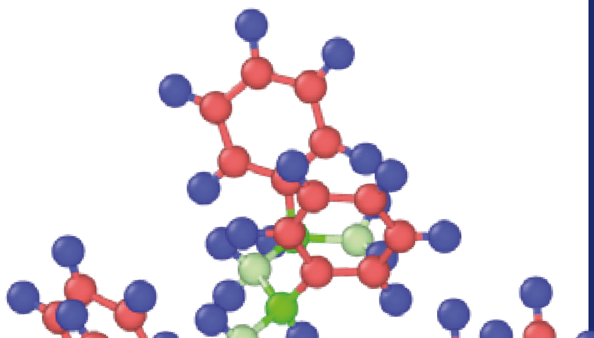


Image: Molecular simulation of polystyrene

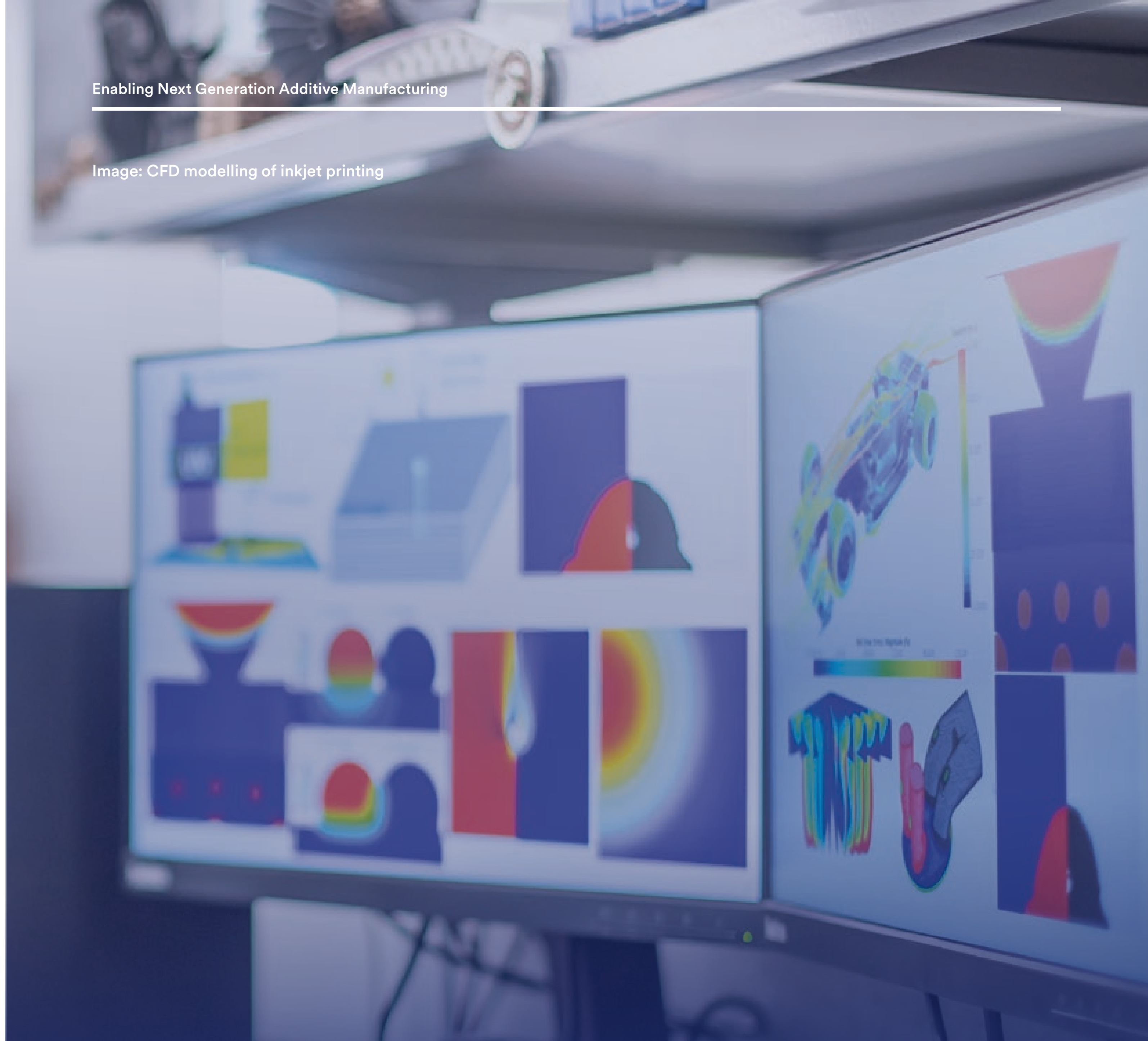


Image: CFD modelling of inkjet printing

Inkjet: CFD modelling framework to predict and optimise the inkjet-based 3D printing

To date, modelling of the ink jet processes has ignored dynamic, wetting and coalescence effects, and accepts as an input perfectly formed layers of material. This approach is being extended with a CFD based modelling framework, with a specific target to be able to capture ‘sagging’ and other non-desirable fluid effects that disrupt the precision of ink jet printing. We have developed and will validate different sub-models, including fluid dynamics, heat transfer, photochemistry, light absorption, polymer rheology, and solidification. Once all sub models have been validated, the model will be used in an inverse approach to be able to provide a design tool to those seeking to perfect the ink jet process.

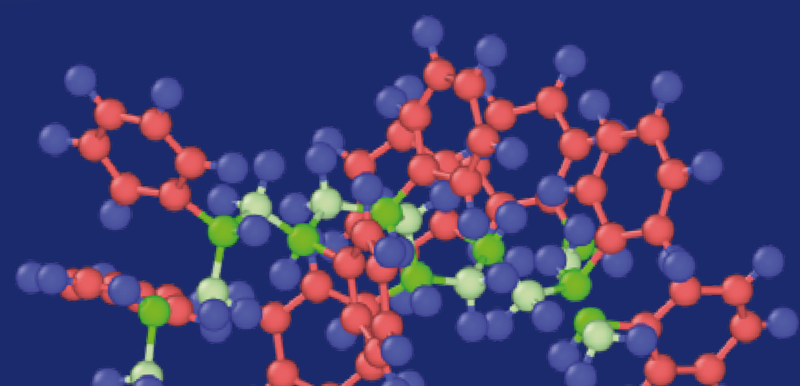


Image: PiXDro LP50 Inkjet Printer internal view (yellow tint due to film added to the window to prevent UV light entering)

Research challenge 03

Research Challenge 3 aims to control the connectivity of additively manufactured objects in order to enable its full functionalisation.

In the initial stages of the Programme, we had a focus on processes and material library development for the two photon polymerisation (2PP). Latterly, we have made significant advancements in both the 3D inkjetting and MetalJet processes and continued development of 2PP processes and multi-materials. In the past year, we have focused on the manufacturing process itself so as to offer a toolkit for RC2's interface study and set the foundations for RC4's device manufacturing.

Some of the main achievements for 2021 include:

- A Computed Axial Lithography (CAL) system has been constructed and is now fully set up and functional at Nottingham. The CAL system's rotatory platform was adapted in order to have a fully closed system for printing with cells
- A compact Intense Pulse Light (IPL) has been developed as a photonic post-process treatment unit and subsequently installed on the LP50 jetting platform with the aim of providing an effective and efficient in-situ sintering methodology for 0D metal nanoparticle inks, alongside annealing of 2D graphene inks, paving the way for fabricating multi-material electronic devices
- The efficacy of MetalJet's new stage heating system in improving the bonding level of low to medium temperature metallic materials (<1000°C) has been successfully demonstrated. With an increase from 500°C to 800°C plus, this represents a significant step forward in the direct metal printing of functional multi-material components with improved consistencies
- Completed the set-up, modification and demonstration of the feasibility of multimaterial printing using the multi-printhead Toucan system, to enable our Programme Grant team to contemporaneously deposit up to six functional/structural formulations developed

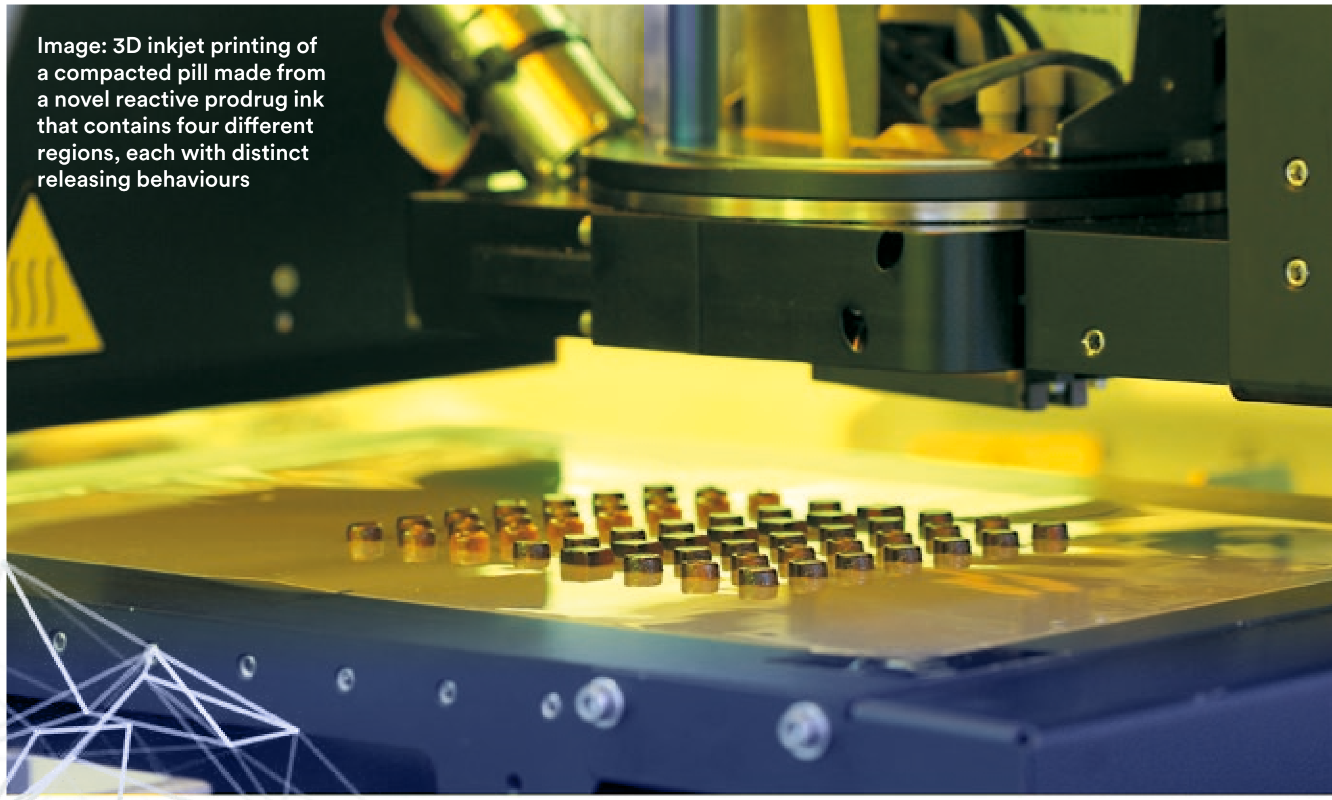


Image: 3D inkjet printing of a compacted pill made from a novel reactive prodrug ink that contains four different regions, each with distinct releasing behaviours

Ongoing projects

Six printheads inkjet system for medical device production delivery and printed electronics

As a drop-on-demand manufacturing process, multi-material inkjet 3D printing offers the ultimate flexibility to manufacture complex bespoke devices with tailored hybrid functionalities. This capability is of particular interest for medical devices and printed electronics , the central application focus of our research.

Our recently enhanced multi-printheads inkjet 3D print system (Toucan) enables us to integrate up to six different functionalities into one single device. Combined with the new functional formulations developed within the team, we have been able to reliably process materials during the past year without any printhead failure.

For example, batches of printed pills with hybrid delivery behaviours and biofilm resistant implants that can self-assemble after application have been successfully manufactured using this system. Our next step is to introduce design tools to guide the distribution of multiple functional materials alongside utilising metallic nanoparticle and dielectric inks for metamaterial antenna application.

Development of photo-cleavable ligand functionalised nanoparticles

In 3D printing, it is essential to control the surface and interface of materials during material deposition; in this regard, we have successfully functionalised gold nanoparticles with photo-cleavable ligands that can labile upon UV light exposure. This enables the tunable electrical conductivity via light induced on-off mechanism and in-situ printing and sintering for multimaterial deposition. Currently, we are optimising the ink formulation for material jetting and demonstrating multifunctional structures through multimaterial co-deposition and in-situ UV sintering.

Low dimensional nanomaterials for the two-photon polymerisation process

Micro/nanoscale multifunctional structures offer great potential in the fields of photonics, microelectromechanical systems, metamaterials, sensors, actuators and biomedical devices. Currently, we are developing two strategic methodologies that enable the fine control for the positioning of plasmonic nanomaterials in the 3D microstructure, firstly via optimised intermolecular interaction between nanomaterials, and secondly a polymer matrix for the two-photon polymerisation process. This will allow us to fabricate plasmonic metamaterials in micro/nanoscale for potential use in optoelectronics and sensors

Self-cyclising diacrylates: inkjet materials with programmable crosslink degradation

To enable recyclability of crosslinked polymers, there is a need to enable a simple means of degrading them in a controlled manner at temperatures below 140°C. This project is developing an acrylate crosslinker which undergoes thermal degradation at a tuned temperature range, for formulation of polymeric inks that print with a high crosslink density (a rigid structure) that can then degraded to soft or dissolvable materials with low crosslink density.

Development of multi-material projection micro stereolithography 3D printing systems

The projection micro stereolithography (PμSLA) approach gives a manufacturing precision up to 100 times higher than an inkjet system. As a result, this is a potentially powerful technique for high resolution manufacturing (<10μm) applications. However, PμSLA has a reduced capability to produce hybrid functional composites due to the single material nature of the process. To increase its versatility and subsequently the range of applications, we are developing two multi-material projection micro stereolithography systems: a top-down fluid chamber design and a bottom-up multi-vat system.

The top-down fluid chamber system, created in collaboration with Boston Micro Fabrication Limited, offers an ultimate printing resolution of 1-2 μm at the expense of high material waste. The bottom-up multi-vats system offers a compromise solution, where printing resolution is sacrificed (10μm) to reduce ink consumption. The success of these two systems will enable the PG to explore an extended range of devices at scales in between those possible with ink jetting and 2 Photon Lithography, including the manufacture of multi-drug microneedle patches, customisable micro-lenses array and programmable scaffold for bioengineering applications, for example.

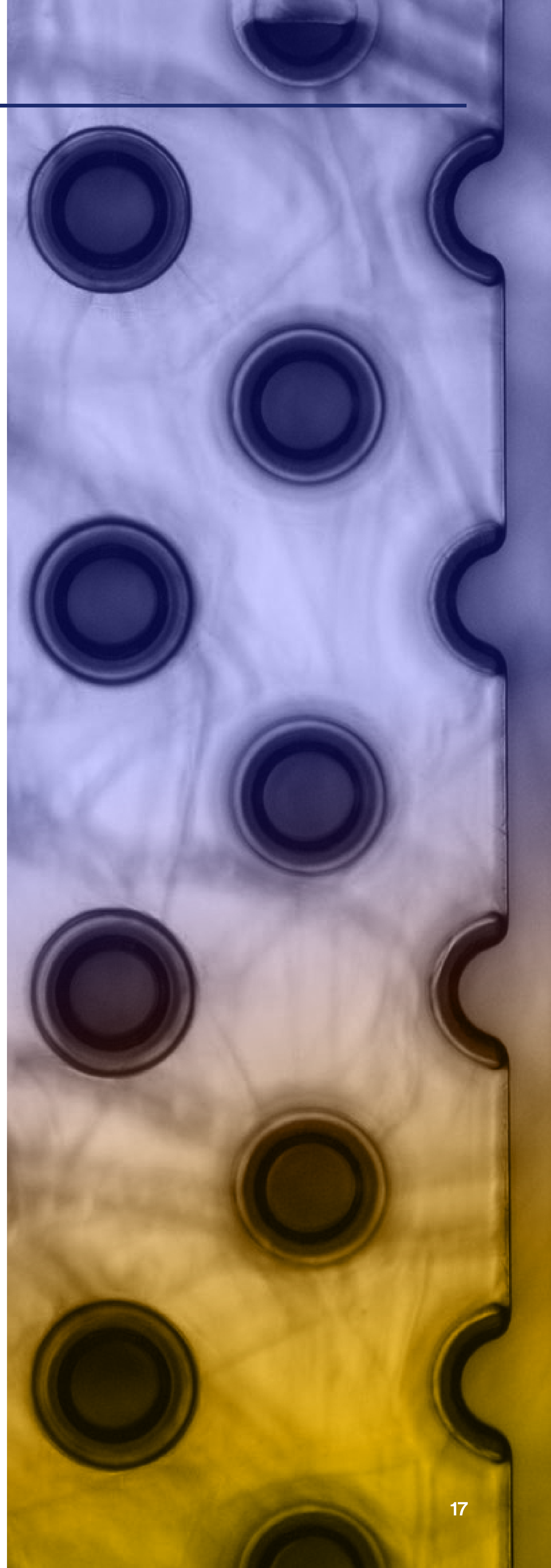


Image: PuSLA Gas Electron Multiplier

Research Challenge 04

Research Challenge 4 (RC4) aims to investigate strategies for the macroscale co-deposition of functional and structural materials via piezo-driven inkjetting, high temperature metaljetting and functionalised multiphoton techniques, with a focus on producing electronic devices and healthcare demonstrators.

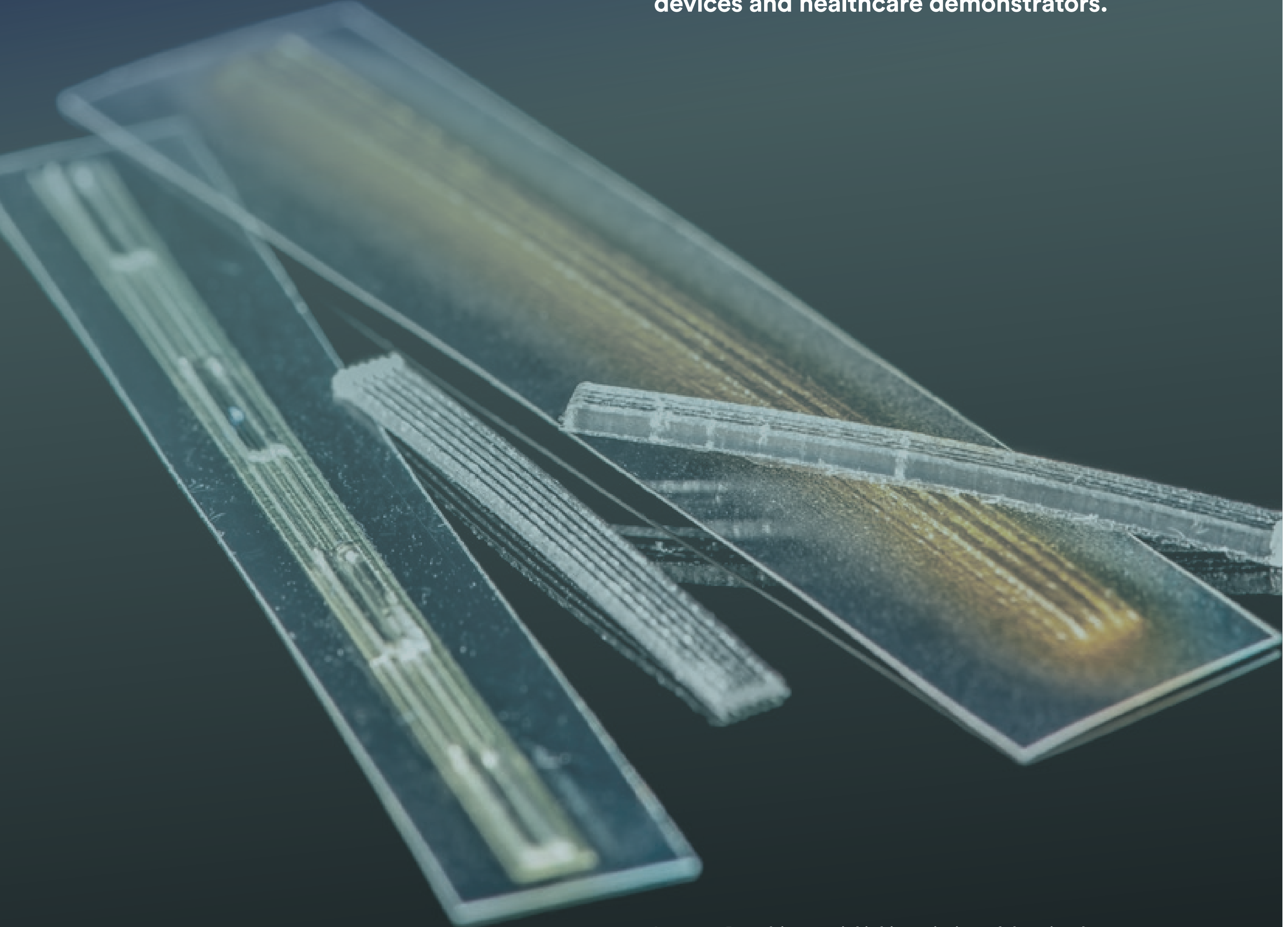


Image: 3D multi-material inkjet printing of drug implants

In RC4 we integrate novel materials and printing strategies to produce multimaterial devices and heterostructures with tailored functionalities to elucidate the opportunities of multimaterial, multifunctional additive manufacturing (AM) which are informed by the analysis, material, processing and modelling developments of RC1 to RC3.

Highlights on electronic materials and devices include:

- Improvement in the inkjet stability of PEDOT:PSS through addition of glycerol carbonate as a co-solvent. Glycerol carbonate provides a more environmentally benign solution and improved conductivity in the printed film. Manipulation of print strategy obtained improved flatness of the printed ink, resulting in an improved sheet resistance and conductivity
- Demonstration of a newly formulated conductive thiol-based gold nanoparticulate ink for inkjetting with enhanced cohesion for flexible electronics and bioelectronics application
- Integration of functional materials with two-photon polymerisation for fine control of positioning plasmonic nanomaterials in the 3D microstructure
- Development of an inkjet-printed multifunctional graphene sensor using resistance and capacitance change in response to the environment. The sensor exhibited a quick, reliable and recoverable response to temperature, humidity and pressure and has great potential for further exploration for applications in healthcare devices
- Initiation of the development of optically active perovskite nanocrystal inks developed for 3D inkjet deposition

Highlights on biomedical materials and applications include:

- Experimental replication of shape-like functional units of liver (liver lobules) and bone (osteons) manufactured with the bioink developed for PμSLA. Both the lobules and the osteons were fabricated with integrated microchannels to mimic blood vessels. It was also demonstrated that osteoblast cells attach and grow on the printed osteons
- Development of a new biocompatible hydrogel formulation for the Computed Axial Lithography (CAL) system. This formulation will be used for fabricating flexible wound fillings that will deliver analgesics and antiseptics into the wound
- Development of a new fully water-soluble pharmaceutical excipient that is compatible with the multi-material inkjet-based 3D Printing (MM-IJ3DP) process for batch production of bespoke pills
- By combining the MM-IJ3DP technology and the new pharmaceutical excipient formulations, pills with tuneable drug loading, dissolving and therefore releasing behaviours are achieved within a standard size of pill (6-8mm)
- Development of a photo-reactive, water-soluble hybrid material ink, natural polymer gelatin, amphiphilic copolymer, for better biocompatibility

Image: Post-doc researcher working with the Cellink+ system in the clean room

Ongoing projects

Inkjet printing perovskite nanocrystals towards flexible display devices

Ceaseum lead halide perovskite nanocrystals (PeNCs) are promising materials for applications in optoelectronics owing to their tunable optical properties, narrow emission bandwidth, and high photoluminescence quantum yield. By formulating inks containing PeNCs and developing the inkjet printing protocol, photodetectors have been fabricated on graphene layers and achieved a high photoresponsivity of 1000A/W. We have identified the route to a fully inkjet-printed LED for flexible display devices and have formulated and tested the required materials for functional layers, including injection layers and transport layers. Currently, charge optimisation of layer deposition is underway to achieve required interface properties for enhanced performance.

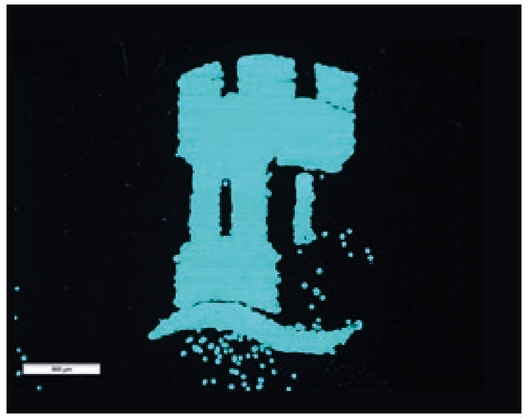


Image: University of Nottingham logo inkjet printed using all-inorganic perovskite nanocrystals with green photoluminescence

Inkjet printed stretchable electrodes for wearable devices

Wearable devices require their components to be stretchable and conductive. Unfortunately, the stiffness of most highly conductive materials needed in electrodes results in cracking when stretched, negatively impacting conductivity. To combat this, a fully inkjet-printed composite electrode is under development. It incorporates silver nanoparticle ink with a novel stretchable PEDOT:PSS ink to act as an inter-crack conductor, and is printed onto a newly developed inkjet printed elastomer.

3D printing the micro and macro architecture of complex tissues

The aim of this project is to develop a multi-technology biofabrication platform that will replicate the complex hierarchical architecture of full-sized solid organs with integrated vasculature to create functional organs. As proof of concept we selected two types of tissues: bone and liver. To replicate the microarchitecture (osteons and lobules) we selected pμSLA as the printing technology and for the macro architecture we selected CAL. Initially, we synthesized and formulated a synthetic biodegradable ink that enhances cells adhesion. Following this, we demonstrated that we can achieve a 10 um resolution with this formulation and manufacture osteons and lobules with integrated micro channels of 30 um diameter, which has not been accomplished before. Currently, we are developing a gelatin-based formulation to be used with the CAL system for macro scale.

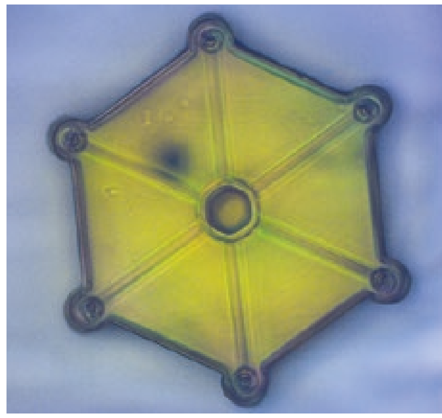


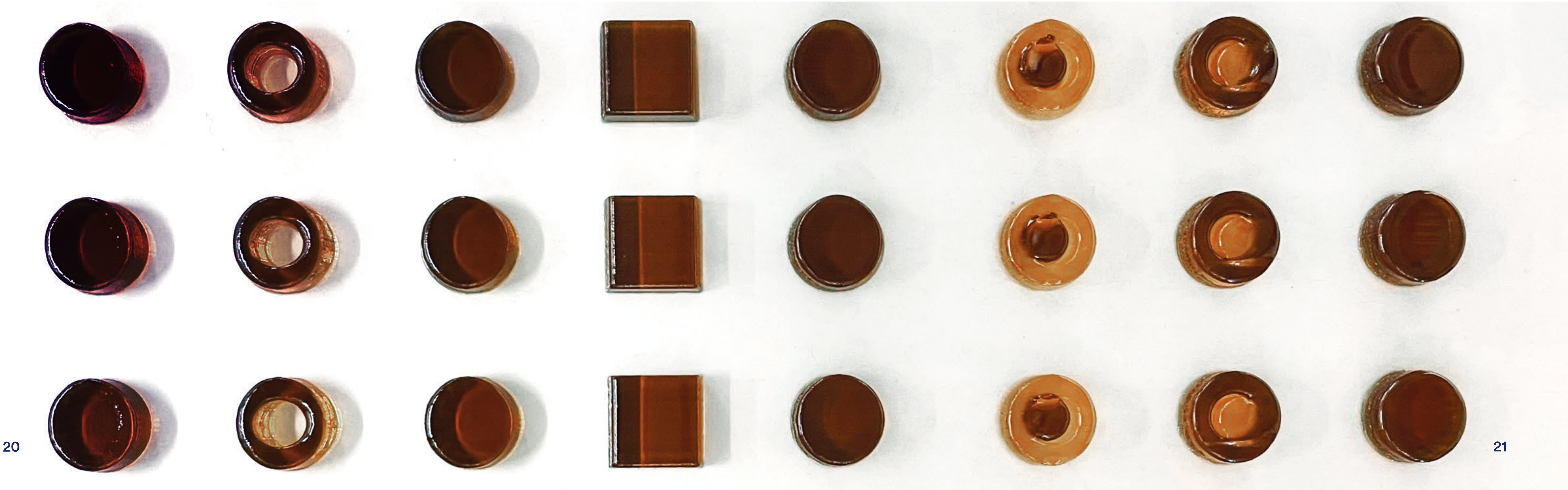
Image: Liver lobule upper tube printed using PμSLA

Batch production of customised pills for the patient through multi-material inkjet-based 3D printing (MM-IJ3DP)

3D printing of pills allows patients to receive more targeted therapeutics and improved treatments. However, achieving this goal requires a suitable 3D print technique that is able to handle multiple drug loaded excipients for the pill to deliver a choice of drug 'loads' in a designed and controlled way. In addition, the technique needs to be feasible for batch production to offer pills with an affordable price.

We have demonstrated the use of 3D inkjet printing printing (IJ3DP) for customised pill production that has enabled us to produce batches of pills (50-100), each with individualised, customised design and drug loading, without the need for any process adjustment. We continue to work on expanding the database of IJ3DP compatible excipient formulations and optimising our multi-material printing system to achieve more controlled drug release, more complex drug loading, and a more rapid turnaround.

Image below: Batch of 3D printed polypills in different shapes and composition



Projects completed in 2021

Investigating the presence of residual polymer stabilisers in nano particulate inks deposited by 3D inkjet printing

Inkjet printing of metal nanoparticles allows for design flexibility, rapid processing and enables the 3D printing of functional electronic devices through co-deposition of multiple materials. However, the performance of printed devices, especially their electrical conductivity, is often lower than those made by traditional manufacturing methods and is not fully understood. In this project we revealed that anisotropic electrical conductivity of printed metal nanoparticles is caused by organic residuals from their inks. We employed a combination of electrical resistivity tests, morphological analysis and 3D nanoscale (orbiSIMS) chemical analysis of printed devices using silver nanoparticles to show that the polymer stabiliser polyvinylpyrrolidone tends to concentrate between vertically stacked nanoparticle layers as well as at dielectric/conductive interfaces. Understanding the behaviour of organic residues in printed nanoparticles revealed potential new strategies to improve nanomaterial ink formulations for functional printed electronics.

A predictive model to determine and optimise quality of the inkjet based 3D printing

On this project, a predictive model was developed to assist in the design and manufacture of structures by inkjet-based 3D printing (IJ3DP) additive manufacturing. We developed and validated a predictive model that incorporates the critical processing parameters, including UV source pathway, UV intensity, printing strategy, and interlayer attenuation, such that we are able to predict the degree of ink conversion throughout the product. We showed how this model can then be used to guide users by demonstrating the coupling of this description with a cost model and illustrating how printing strategy affects descriptors of both the quality and cost of production.

Integrating solidification with interface formation

Interfaces are transitional regions between two phases with different properties; consequently, the study of the process of solidification, which takes place over an interfacial region, requires resolving the interfacial scales. Moreover, if solidification happens as other interface-dominated behaviour (such as dynamic wetting) occurs, models need to capture the interaction between solidification and these other effects.

To produce predictive models for drop-on-demand additive manufacturing, a new integrated framework for solidification and interface-forming capillary flows has been developed. This implementation resulted in one accepted paper, whose results will inform the construction of the overall modelling framework.

Development of Gold conductive ink for flexible electronics and bioelectronics

The development of gold nanoparticle (AuNP) ink is of particular interest to enable additive manufacturing of electronic components and devices, due to its high electrical conductivity, chemical stability, and biocompatibility. On this project, we worked on the formulation of conductive gold ink with a multifunctional thiol to prevent the formation of microcracks and pores, as these can lead to poor integrity of a printed electronic component and electrical failure under external mechanical deformation. The successful outputs of this project includes high electrical conductivity and stable electrical properties under repeated cycles of mechanical deformation, offering exciting potential for applications in flexible devices and wearable and healthcare electronics.

Improvement of inkjet stability of PEDOT:PSS through co-solvent selection

A new ink formulation, referred to as green PEDOT:PSS ink, uses two bio-renewable chemicals (glycerol carbonate and cyrene) as co-solvents and displays improved storage stability and stability of jetting, without clogs or misplaced droplets. The improved jetting stability has enabled accurate printing of narrow (approx. 0.1 mm wide) conductive traces over a large 11 × 7cm area, exceeding print areas in the open literature. This co-solvent change also enabled an improvement to the precision of droplet placement, allowing advanced printing strategies to be employed. One of these strategies improved the flatness and reduced the sheet resistance of the printed films, a useful feature for future implementation in multi-layered optoelectronic devices.

Inkjet printing multifunction graphene sensors

Graphene sheets display fulfils all-around sensing performance and will be important in applications such as wearable devices. Recently, we developed a multifunction graphene sensor using inkjet printing technology of low dimensional materials that we had previously established. The device, fabricated by printing two pairs of graphene tracks separated by a dielectric TPGDA layer on flexible Kapton film, provides a sensing platform, which allows scaling up as well as adding other sensing parameters, such as gases, ions, chemicals, or bacteria. A theoretical model explained the response quantitatively, providing insight into electron transport on these devices.

Aligned doctoral research

Developing our future researchers

This Programme Grant (PG) 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, where academic guidance from PG academic and researcher supervisors, alongside access to the state-of-the-art research equipment is provided. As a result, these PhD students have been producing valuable inputs to the Programme progress.

In addition, the Programme Grant has also generated opportunities for some of these students to advance their research careers and we are extremely proud that two students were awarded a research position within the group in 2021.

Anna Lion
Research Fellow at University of Nottingham since October 2021
—
Graduated December 2021

Development of a multi-material hot-melt inkjet 3D printing platform towards complex and personalised dosage form

Anna's project focused on developing a multi-material hot-melt inkjet 3D printer for complex and personalised dosage form. This involved designing and assembling an additional heated reservoir alongside a support system to house the two dispersing units inside the main body of a commercially available LP50 printer. This was undertaken in SolidWorks, the reservoir was then produced at the in-house workshop using traditional tooling while the support structure was manufactured using a laser sintering process from Nylon PA12 to reduce weight to the minimum. In support, a temperature control system was developed and original software was updated to allow control of the additional components.

In parallel, Anna worked on expanding the library of compatible materials that can be used to produce successful prints, raising the number of available carriers from one, found in the literature, to six. Finally, a set of tablet geometries displaying immediate, extended, delayed and pulsatile drug release were produced, including both single and multi-material complex 3D patterns with designed local drug loading, where the drug-free ink is used as a release-retarding material.

The work carried out during this PhD set the foundations for Anna's appointment as a Research Fellow within this Programme where she is currently developing two multi-material micro stereo lithography systems aimed at the production of multi-drug microneedle patches, customizable micro-lenses array and programmable scaffold for bioengineering applications.

Negar Gilani
Research Associate at University of Nottingham since October 2021
—
Started October 2018

Control of high temperature drop-on-demand metaljetting through numerical modelling and experiments

Drop-on-demand metaljetting (DoD-MJ) is a new exciting fabrication technique for single and multi-metal components at high resolutions which avoids the use of powders or complicated post-processing. MetalJet, a bespoke DoD-MJ platform, can produce molten microdroplets (<70 µm) at high temperatures (up to 2000 °C) to form single and multi-material objects.

Negar's PhD project focused on understanding the deposition of metallic microdroplets produced by MetalJet through an integrated computational and experimental approach. To achieve this, a thermomechanical Finite Element model was developed to predict the temperature and residual stress evolution during the solidification and cooling of single and multiple droplets. In parallel, printing experiments were performed, and characteristics of micro-droplets were investigated using various characterisation techniques such as FIB-SEM, EBSD, and TEM. The research provided insight into the underlying physics of the morphology of metal jet droplets after deposition and solidification, the interface formed by such droplets, the microstructure formation, and the residual stress evolution, all of which define the consistency and quality of printed parts. The knowledge obtained through this research permitted the optimisation of the jetting and deposition parameters involved in producing consolidated 3D prints.

The research represented a step forward in the direct metal printing of high resolution functional multi-material components and set the foundations for Negar's current post as a research fellow, where she continues to investigate the fundamentals behind drop-on-demand MetalJet.

Jonathan Gosling
Final year
PhD Student,
University of
Nottingham
—
Started October
2018

Electronic properties of graphene-based electronics

Inkjet printing of 2D materials involves depositing individual flakes into a matrix form. Many features of printed heterostructures influence the performance of conductive components, from the disorder of flakes to the properties of surrounding dissimilar materials.

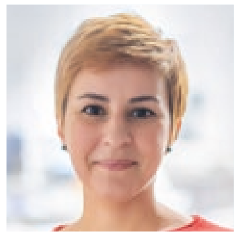
In this project, the electrical properties of dissimilar materials were predicted by quantum transport simulations under multiple sources of electronic scattering. Jonathan also developed a model that explains charge transport in graphene-based heterostructures, including percolation and disorder effects. The model accurately reproduces the observed properties of inkjet-printed graphene. The size and density of flakes, and amount of disorder were found to have significant effects. Latest developments include modelling of the effects of temperature and interlayer twisting on the transport properties of graphene based heterostructures. The models developed aim to inform the next stages of 2D material printing with a deeper understanding of the mechanics that govern transport between 2D films and within constituent flakes.

Eric Lehder
Final year PhD
Student, University
of Nottingham
—
Started October
2018

Optimising the geometry of a fracture healing assembly including a cell seeded scaffold and a stiffness graded auxetic fixation plate

Additively manufactured scaffolds with biocompatible, biodegradable and bone matched mechanical properties could improve the outcomes of bone healing and regeneration. To keep the bone-scaffold assembly stable while osteo-blast cells develop into mature bone, a fracture fixation plate is needed, therefore, it is important to optimize both elements. Triply Perdiodic Minimal Surface (TPMS) scaffolds offer a large surface-to-volume ratio and ideal curvature profiles for cell growth and stiffness grading and auxetic structures are promising solutions for the problem of stress shielding with stiff plates.

During his research, Eric worked on the development of a computational method to address this problem, by using a curvature dependent model to optimise a TPMS geometry bone regeneration scaffold, maximising the regenerative capacity; and a mechanical performance finite element model to optimise the plate geometry, minimising the stress shielding. Lately, he worked on cell culture experiments and compared the results with the cell growth model and carried out a compression strain test to verify the plate optimisation.



**Maria Ines
Evangelista
Barreiros**
CDT Student,
*University of
Nottingham
—
Started February
2019

Design and manufacture of drug delivery platforms

Whilst there are several different 3DP methods and many of these have been used to produce oral solid dosage forms, Ines's project focusses on material extrusion and its different modalities using the BioX by Cellink printer. This way, by using one single drug as a model drug and the same printer, a reliable comparison of the release profile using different printing modalities and excipients can be established.

During this project, Ines is working on three different 3D extrusion printing modalities: a paste-based approach, direct powder printing, and a photocurable formulation, comparing the processes and produced tablets. To date, she has nearly completed the paste-based and direct powder printing and next, will focus on the optimisation of the photocurable formulation and its printing process.

***EPSRC Centre for Doctoral Training in Additive Manufacturing**

Joseph Sefton
PhD Student,
University of
Nottingham
—
Started October
2019

Production and use of oligomers in additive manufacturing

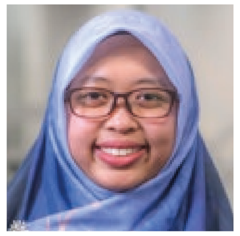
A key issue in photopolymer-based additive manufacture (AM) is a limitation of the materials palette; methacrylate monomers are regarded as "slow" and are disfavoured compared to acrylate monomers. To introduce methacrylic material with less negative impact, Joseph has utilised an in-situ approach to catalytic chain transfer polymerisation (CCTP) to synthesise methacrylic oligomers (low molecular weight polymers). Oligomers produced via CCTP contain a vinyl end-group, which renders the oligomers reactive in further radical polymerisations. He has also explored the copolymerisation of oligomers and some model acrylates used in additive manufacturing. This work has highlighted that oligomer structure and loading highly influence the molecular architecture of the final copolymer. Next is the translation of these results into photopolymer AM processes, such as stereolithography, to generate materials with novel and tuneable properties.

Kristian Plender
PhD Student,
University of
Nottingham
—
Started October
2019

Novel approaches to the long term release of biomacromolecules

Long-term delivery of therapeutic biomacromolecules has seen increasing interest for the provision of chronic disease treatment. However, retaining therapeutic function until final elution from a delivery device is a multi-factorial challenge. Additive manufacturing techniques present a high degree of flexibility regarding delivery device geometry and control over spatial location of bioactives to achieve and optimise release for intended patient groups.

Kristian's project focusses on encapsulation of model proteins within formulations that are printable using UV stereolithography (SLA). He has fabricated biodegradable implant structures to understand the biomacromolecule release over sustained periods, over 1 to 3 months, via the tailoring of formulations and design changes. Future work will establish the fundamental factors influencing release, potentially indicating appropriate manufacturing protocols and strategies to be adopted for delivering higher cost therapeutic biomacromolecules exhibiting similar characteristics (molecular weight, hydrodynamic radius, isoelectric point etc).



**Nur Rofiqoh
Eviana Putri**
PhD Student,
University of
Nottingham
—
Started
December 2019

Additive manufacture of vascularised bioactive scaffolds for bone tissue engineering

In tissue engineering, the lack of vasculature invasion into scaffolds leads to cell death due to nutrient and oxygen shortage. It is still challenging to replicate the complex vasculature using conventional methods. To solve these problems, additive manufacture offers high resolution for the potential to mimic the capillaries. Nur is working on the development of bone tissue analogues with integrated vasculature using ink-jet 3D printing and biodegradable materials.

To date, she has developed an amphiphilic diacrylated triblock copolymer of Poly-Trimethylene Carbonate and Poly(Ethylene Glycol) combined with Gelatin-methacryloyl ink, which is water-soluble and, as the use of a toxic solvent can be avoided, has better biocompatibility. Recently she has used this formulated ink on an inkjet system to replicate a vascularised scaffold. The next steps will be to focus on the characterisation of printed scaffolds for bone tissue formation.

Tien Quach
PHD Student, University of Nottingham
—
Started December 2019

Novel micro/nano scale characterisation of interfaces in multi-material additive manufacturing

Additive manufacturing (or 3D-printing) has many different applications in automotive, engineering, healthcare, aerospace, and defence. However, the main challenge to enable the next generation of 3D printing technology relates to incompatibility in physical and chemical properties between materials. To help overcome these problems, I am investigating the interfaces/surfaces between ink formulations and substrates by designing and the (standard) interface patterns from co-printing the water-soluble and structured inks.

A high-speed camera with new embedded coding was shown to be able to acquire in-situ morphological data of the 3D-printed samples whilst Interferometry with auto-stitching is shown to be effective for measuring the thickness of a single printed layer. Thus, after progressing the optimal ink-jet printing, we will be able to develop the analytical framework for interface and surface study of multi-materials additive manufacturing. Further characterisation will be carried out to better expose and analyse the interfaces/surfaces between material-material and material-substrate in real-time.

Jonathan Austin
PhD Student, University of Nottingham
—
Started October 2020

Additive manufacturing based on hybrid low dimensional 0D/2D heterostructures

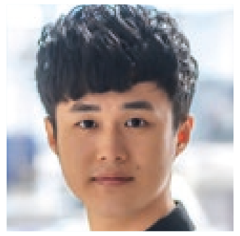
Jonathan's project aims to develop low-dimensional optoelectronic devices such as photodetectors, solar cells, and LEDs using inkjet printing. Inkjet printing enables efficient and upscaled production of electronic components, however, there are still major challenges in printing the multi-layered and multi-material heterostructures that are required for optoelectronic devices. To date, he has developed a perovskite nanocrystals ink and demonstrated fully printed photodetectors based on perovskite and graphene. He is now currently developing a fully inkjet-printed perovskite LED, which has not previously been achieved, which will include several other materials developed within this Programme, such as PEDOT:PSS and gold nanoparticles. Towards this goal, he has also formulated inks for two charge transport polymers and characterised the material interfaces that are important for device functionality.

Frederick Temple
PhD Student, University of Nottingham
—
Started December 2020

Predictive modelling and design optimisation of erodible drug delivery systems

Being able to make decisions on the best type of medicine to prescribe to a patient based on the patient's personal parameters would facilitate the prescription of medicines. Making accurate predictions through modelling different types of pharmaceutical tablets could potentially reduce the experimental costs by indicating what type of matrix is needed as a drug carrier in order to deliver the drug to the correct receptor and in the most efficient way.

Frederick's PhD project aims to understand the process and create a model for describing drug delivery via absorption, swelling and erosion. This model, currently being developed using finite elements, will support the creation of the most efficient pharmaceutical tablets, considering dimensions, matrix tablet and any other relevant parameters using established theory. In the next steps, he'll be carrying out some experiments and gathering data to validate the model.

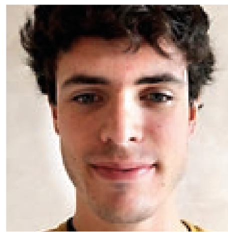


Xiangyun Gao
PhD Student, University of Nottingham
—
Started April 2021

Multi-material metaljetting: developing a colour metal printer

The metaljetting platform that is being investigated within the Programme, in collaboration with industrial partner Canon, shows great promise for the deposition high temperature metals, for example with recent progress being made with the deposition of copper. This ability to deposit 'real' metals, as opposed to nano-particulate materials that are usually found in drop-on-demand systems enables additional functionality (e.g. conductivity) to be gained in parts produced using this approach.

Xiangyun's project is focused on the investigation of multi-metal jet printing – effectively a unique colour metal printer. As no system currently exists for such research, his PhD has first concentrated on the development of a MetalJet platform capable of depositing two materials with melt points more than 1000°C. In his first year, Xiangyun has developed, with the technical support from CfAM, all the software needed to drive the system. Once commissioned, he will move on to the experimental stage of his PhD to investigate the challenges of multi metal deposition.



Nathan Coombs
PhD Student, University of Warwick
—
Started October 2021

Computational modelling of 3D inkjet printing: PEDOT residue patterns

PEDOT is a conductive polymer commonly used for printed electronics. In practice, nanoparticles of PEDOT surrounded by PSS are first dissolved in the printing medium where, after jetting onto the substrate, this medium then evaporates to leave a residue. The caveat of this method is that PSS has relatively poor conductivity: if the residual profile contains PSS-rich regions with very little PEDOT, this will significantly reduce overall conductivity.

Nathan's research project aims to capture the entire PEDOT deposition process in a mathematical model, considering how the PEDOT:PSS nanoparticles are transported during solvent evaporation and how they will interact with the solvent in the later stages of the process. Focusing initially on the former of these, he is investigating how physical phenomena such as capillary flow can influence the morphology of the residue. Nathan has been developing this work in collaboration with Geoffrey Rivers, from the University of Nottingham, who is providing experimental data for benchmarking of his simulations.

Ana Valeria Gonzalez Abrego
PhD Student, University of Nottingham
—
Started November 2021

3D printing of multi-functional scaffold and tissue models through multimaterial pμSLA

Bioprinting has been widely used and shown great promise for creating models and implantable artificial organ development, but the technology to incorporate various materials into the final printed articles still needs considerable investigation. Valeria's project is focussed on the development of a projection micro-SLA printer that will enable the use of different bioinks during the same printing process. Having this technique in place will allow the investigation of a hepatic model and to replicate the natural hepatic micro and macro environment using different biomaterials within the same structure.

Outputs

As Covid-19 restrictions were reduced throughout the year, our team was able to partially resume external activities. Yet, due to the uncertainty around in-person events and local restrictions, most conferences and external events continued to be hosted online.

Image: Dr Laura Ruiz Cantu (left) and Dr Yinfeng He (right)

The Royal Society Summer Science Exhibition

How to 3D print your wonder pill

The Summer Science Exhibition is the prestigious Royal Society flagship annual public engagement event that takes place in early July. Led by Programme Investigator, Professor Ricky Wildman, we were delighted to take part in the 2021 exhibition, when Dr Laura Ruiz Cantu and Dr Yinfeng He delivered a pre-recorded session in 3D printing pills to an on-demand audience and a workshop which had 1,250 households registered.

Recent pharmacogenetics research has shown that people react differently to drugs depending on their genetics, age, size and gender and how the field of medicine is moving towards personalised medicine. In the short video, our team talked about how 3D printing can enable pills to be produced in different shapes, materials and in a variety of doses of each drug to suit each person's needs.

Will future doctors prescribe a perfect pill for each patient? Will future pharmacists be ready to 3D print custom-made pills for every person? Until then, why don't you try our game and pretend to be a pharmacist of the future that assists with the demand of a queue of patients?

https://limitless-beach-00824.herokuapp.com/

This lecture was presented live during the summer festival and has had over 17,000 views to date. If you missed it, you can still find it on the RSC YouTube page.

Background image: 3D-printed polypills with controlled distribution and release of medication in different shapes

Aligned to the Research Publication Framework and CfAM's publication strategy, the group has been striving to produce high quality publications which translate the great quality research developed within the Programme. This means, the team follow a long-term publication plan, which considers different audiences and a range of publications for high leading journals. In 2020, there were 38 manuscripts in the pipeline, of which 15 publications aligned with the theme of the Programme were submitted, accepted and published in 2021.

Publications

Universal mobility characteristics of graphene originating from charge scattering by ionised impurities	Jonathan Gosling, Oleg Makarovsky, Feiran Wang, Nathan Cottam, Mark T. Greenaway, Amalia Patanè, Ricky Wildman, Christopher J. Tuck, Lyudmila Turyanska and T. Mark Fromhold. Nat Commun Phys 4, 30 (2021). https://doi.org/10.1038/s42005-021-00518-2
Core/shell metal halide perovskite nanocrystals for optoelectronic applications	Chengxi Zhang, Jiayi Chen, Lingmei Kong, Lin Wang, Sheng Wang, Wei Chen, Rundong Mao, Lyudmila Turyanska, Guohua Jia, Xuyong Yang. Adv. Funct. Mater. 2021, 31, 2100438. https://doi.org/10.1002/adfm.202100438
Modulating the biological function of protein by tailoring the adsorption orientation on nanoparticles	Journal of Colloid and Interface Science. Akhil Jain, Gustavo F. Trindade, Jacqueline M. Hicks, Jordan C. Potts, Ruman Rahman, Richard J.M. Hague, David B. Amabilino, Lluïsa Pérez-García, Frankie J. Rawson, Volume 587, 2021, Pages 150-161, ISSN 0021-9797. https://doi.org/10.1016/j.jcis.2020.12.025
Light-induced stark effect and reversible photoluminescence quenching in inorganic perovskite nanocrystals	Nathan D. Cottam, Chengxi Zhang, Joni L. Wildman, Amalia Patanè, Lyudmila Turyanska, Oleg Makarovsky. Adv. Optical Mater. 2021, 9, 2100104. https://doi.org/10.1002/adom.202100104
Residual polymer stabiliser causes anisotropic electrical conductivity during inkjet printing of metal nanoparticles	Gustavo F. Trindade, Feiran Wang, Yinfeng He, Jisun Im, Adam Balogh, Ian Gilmore, Mariavitalia Tiddia, David Pervan, Ehab Saleh, Lyudmila Turyanska, Christopher Tuck, Ricky Wildman, Richard Hague, Clive J. Roberts. Communications Materials 2, 47 (2021). https://doi.org/10.1038/s43246-021-00151-0
Exploiting generative design for 3D printing of bacterial biofilm resistant composite devices	Yinfeng He, Meisam Abdi, Gustavo F. Trindade, Belén Begines, Jean-Frédéric Dubern, Elisabetta Prina, Andrew L. Hook, Gabriel Y. H. Choong, Javier Ledesma, Christopher J. Tuck, Felicity R. A. J. Rose, Richard J. M. Hague, Clive J. Roberts, Davide S. A. De Focatiis, Ian A. Ashcroft, Paul Williams, Derek J. Irvine, Morgan R. Alexander, Ricky D. Wildman. Adv. Sci. 2021, 8, 2100249. https://doi.org/10.1002/advs.202100249
Evaluation of photoanode materials used in biophotovoltaic systems for renewable energy generation	Maira Anam, Helena I. Gomes, Geoffrey Rivers, Rachel L. Gomes, Ricky Wildman. Sustainable Energy Fuels, 2021, 5, 4209. https://doi.org/10.1039/D1SE00396H

For full list of publications, please check: **www.nottingham.ac.uk/research/groups/cfam/major-epsrc-funding/programme-grant-next-gen-am/outputs-and-impacts/publications-and-conferences.aspx**

Publications

Solidification and dynamic wetting: a unified modeling framework	Yulii D. Shikhmurzaev. Physics of Fluids 33, 072101 (2021) https://doi.org/10.1063/5.0054431
UV-curable silicone materials with tuneable mechanical properties for 3D printing	Aleksandra Foerster, Vinotharan Annarasa, Anna Terry, Ricky Wildman, Richard Hague, Derek Irvine, Davide S.A. De Focatiis, Christopher Tuck, Materials & Design, Volume 205, 2021, 109681, ISSN 0264-1275, https://doi.org/10.1016/j.matdes.2021.109681
Poly (glycerol adipate) (PGA) backbone modifications with a library of functional diols: Chemical and physical effects	Philippa L. Jacob, Laura A. Ruiz Cantu, Amanda K. Pearce, Yinfeng He, Joachim C. Lentz, Jonathan C. Moore, Fabricio Machado, Geoffrey Rivers, Edward Apebende, Maria Romero Fernandez, Iolanda Francolini, Ricky Wildman, Steven M. Howdle, Vincenzo Taresco. Polymer, Volume 228, 2021, 123912, ISSN 0032-3861. https://doi.org/10.1016/j.polymer.2021.123912
A multiscale optimisation method for bone growth scaffolds based on triply periodic minimal surfaces	Eric Lehder, Laura Ruiz-Cantu, Ian Maskery, Ian Ashcroft, Ricky Wildman. Biomech Model Mechanobiol 20, 2085–2096 (2021). https://doi.org/10.1007/s10237-021-01496-8
Bespoke 3D printed polydrug implants created via microstructural control of oligomers	Laura Ruiz-Cantu, Gustavo F Trindade, Vincenzo Taresco, Zuoxin Zhou, Yinfeng He, Laurence Burroughs, Elizabeth A. Clark, Felicity R.A.J. Rose, Christopher Tuck, Richard Hague, Clive J. Roberts, Morgan Alexander, Derek J. Irvine, and Ricky D. Wildman. ACS Applied Materials & Interfaces 2021 13 (33), 38969-38978. https://doi.org/10.1021/acsami.1c07850
Modelling the influence of UV curing strategies for optimisation of inkjet based 3D printing	Peng Zhao, Yinfeng He, Gustavo F. Trindade, Martin Baumers, Derek J. Irvine, Richard J.M. Hague, Ian A. Ashcroft, Ricky D. Wildman. Materials & Design, Volume 208, 2021, 109889, ISSN 0264-1275, https://doi.org/10.1016/j.matdes.2021.109889
Insights into drop-on-demand metal additive manufacturing through an integrated experimental and computational study	Negar Gilani, Nesma T. Aboulkhair, Marco Simonelli, Mark East, Ian Ashcroft, Richard J. M. Hague. Additive Manufacturing, Volume 48, Part B, 2021, 102402, ISSN 2214-8604. https://doi.org/10.1016/j.addma.2021.102402
Impact of Dielectric Substrates on Chipless RFID Tag Performance	Amjad Ali, Christopher Smartt, Jisun Im, Roderick Mackenzie, Orla Williams, Ed Lester, Steve Greedy. SSRN, 2021. http://dx.doi.org/10.2139/ssrn.3980406

Related funding

Funder	Project/programme/partnership title	Investigator	Value
EPSRC	Quiet aerofoil with adaptive porous surfaces (QUADPORS)	Choi / Tuck	£531,142
DSTL	Design optimisation of low SWaP magnetic field generating, shielding and atom trapping subsystems for quantum sensors of magnetic fields, gravity and rotation	Fromhold	£120,000
DSTL	CMOS atom chips: a platform for scalable ITAR-free manufacture of fully integrated cold-atom trapping and control systems for quantum sensors	Fromhold	£142,000
EPSRC	UK National Quantum Technology Hub in sensing and timing	Fromhold	£2,700,000
Innovate UK	3 x IUK with various industrial partners	Fromhold	£3,300,000
EPSRC	Next generation rehabilitation technologies	Goodridge	£831,040
EPSRC	Additive manufacturing and 3D printing in clinical practice	Goodridge	£158,815
EPSRC - IAA	Accelerated commercialisation of the Nottingham reactive 3D printing process	He	£24,868
EPSRC - IAA	Scaling up and optimization of the printing process for a patented powder based 3D reactive inkjet technology	He	£62,277
iCURE (Innovate UK)	3D reactive jetting technology	He	£15,000
EPSRC - IAA	Accelerated commercialisation of the Nottingham reactive 3D printing process	He	£24,868
RHiN	Ultrafast 3D printed personalised wound fillings for immediate stabilization and regeneration of soft tissues	He/Cantu/ Wildman/Rose	£80,405
EPSRC - IAA	PoC Polymerisation method development for the manufacture of novel, high-performance, compostable and recyclable heteroaromatic bioplastics for Industrial prototyping-BioPolyMethod	Irvine	£35,697
IDRI	Engineering sustainable squalene analogues for novel vaccine adjuvant emulsions	Irvine	n/d
BBSRC	Development and exploitation of a bioactives-free technology for tackling fungal threats to food security, goods and health	Irvine / Avery	£224,967
EPSRC	PoC- Dundas - Improving the processability of polymers via spray-drying	Irvine / Dundas	£35,000
iCure	Reactive fusion	Ma/He/Rice/ McLeod/Winton	£27,166

Related funding

Funder	Project/programme/partnership title	Investigator	Value
InnovateUK	Reactive fusion - Revolutionary technology for 3D printing	Ma/He/ Tuck/Hague/ Wildman	£282,930
EPSRC	Wireless communication with cells towards bioelectronic treatments of the future	Rawson / Hague	£974,695
EPSRC - IAA	Ultrafast volumetric printing of wound fillings for immediate stabilization of soft tissues	Ruiz Cantu	£64,324
EPSRC - IAA	Enhancing osseo integration of additively manufactured implants by optimising surface architecture	Ruiz Cantu	£6,939
Wellcome Trust	Development of a fatigue detection device to prevent musculoskeletal injuries in football	Ruiz Cantu	£14,900
Wellcome Trust	Mechano-chemical integration at the osteochondral interface	Ruiz Cantu	£14,900
EPSRC	Quantum Technology Hub phase 2 – Optimisation/AM/ Rapid prototyping WP	Wildman/ Fromhold	£177,444
Impact acceleration program	IEG - Evaluation of novel tyre manufacturing formulation	Wang	£8,000
US Army	Low-cost Titanium for Combat Protective Equipment	Simonelli	£30,000
EPSRC - NanoPrime	Design of novel thermal treatments for additive manufactured titanium alloys through high-temperature microscopy	Simonelli	£2,000
EPSRC	An automated hardness tester and mapper	Simonelli / Aboulkhair	£60,000
Industry	Confidential Industry – MetalJet x 3 projects	Simonelli / Aboulkhair	£36,166
EPSRC	Dynamic wetting and interfacial transitions in three dimensions	Sprittles	£870,000
EPSRC	Dial-up Engineered Microstructures for Advanced Additively Manufactured Metals (DEMAMM)	Tuck	£1,809,056
QinetiQ Group PLC	Inkjet printing for antenna – Phase 1 scoping study	Tuck	£9,600
DSTL	Inkjet printing for fabrication of tailored electromagnetic materials	Tuck	£142,638
AWE	Silicone jetting phase 2 Micro-SLA	Tuck	£356,477
EPSRC	Intelligent structures for low noise environments: a Prosperity Partnership	Tuck	£715,093

Related funding

Funder	Project/programme/partnership title	Investigator	Value
EPSRC (NSF)	A Transatlantic Institute for volumetric powder bed fusion	Tuck / Hague	£254,070
EPSRC	Equipment grant for a suite of multimaterial AM facilities	Tuck	£1,900,000
EPSRC - IAA	Volumetric sintering of metal/ceramic implants	Tuck / Sohaib	£22,293
Industry	Silicone jetting phase 2 Micro-SLA	Tuck / Hague	£3,566,477
DSTL	Inkjet printing for fabrication of tailored electromagnetic materials	Tuck / He	£137,078
EPSRC - IAA	PoC – Development of gold conductive inks for material	Tuck / Im	£33,728
DSTL	Quantum bio-sensing for next generation health diagnostics	Turyanska	£114,725
DSTL	Graphene based UV sensor	Turyanska	£98,000
NBCRC	Investigating in vivo efficacy of apoferritin-encapsulated antitumour benzothiazole 5F203 against human-derived breast cancer xenografts	Turyanska	£10,000
EPSRC - Euro MagNET	Magneto-optical studies of all-inorganic perovskite nanocrystals	Turyanska	£50,400
EPSRC	Capital award for core equipment 2020/21	Wildman	£550,000
EPSRC	Prosperity Partnership (GSK) – Accelerated discovery and development of new medicines: Prosperity Partnership for a healthier nation	Wildman	£562,538
EPSRC	Dialling up performance for on demand manufacturing	Wildman	£5,865,536
MRC Confidence in Concept Award (CiC)	Preparing for first in man trials of 3D printed solid dosage forms	Wildman	£76,083
Astra Zeneca	Feasibility study: printotyping project	Wildman	£13,111
Prosperity GSK	Accelerated discovery and development of new medicines	Wildman	£446,285
EOARD USAF	Complex materials for advanced device fabrication	Wilman / Tuck	£191,070

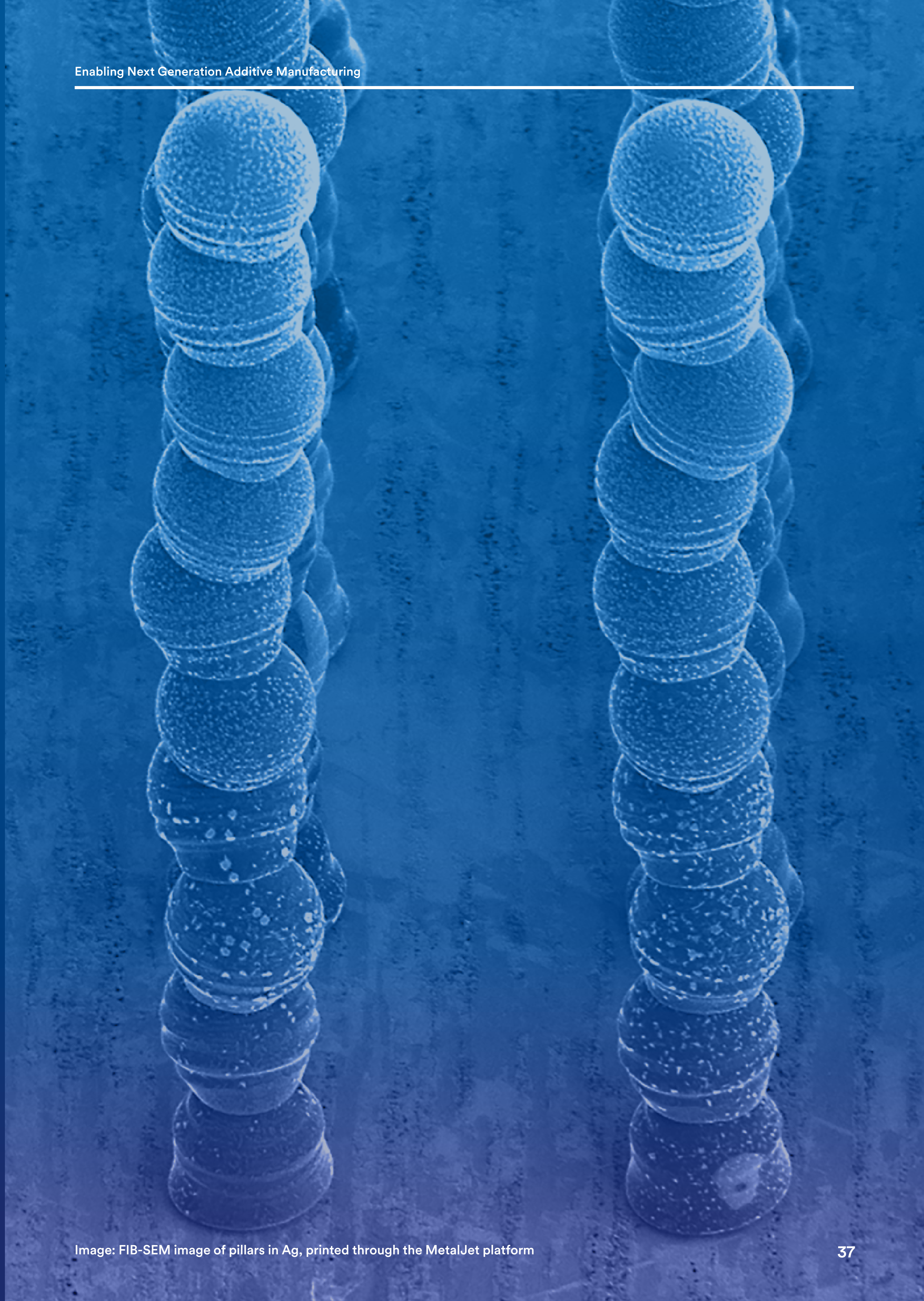


Image: FIB-SEM image of pillars in Ag, printed through the MetalJet platform

Outreach events

As Covid-19 restrictions were reduced throughout the year, our team was able to partially resume external activities. Yet, due to the uncertainty around in-person events and local restrictions, most conferences and external events continued to be hosted online.

Presentations

January 2021		
Additive manufacture of vascularized biocompatible scaffold for bone tissue engineering	Nur Rofiqoh Eviana Putri	RSC Biomaterials Chemistry Group Annual Meeting (virtual)
February 2021		
3D-printing of medicines and implants: making the formulations work	Clive Roberts	12th World Meeting on Pharmaceutics and Biopharmaceutics, Vienna, Austria
Micro/nanoscale characterisation of the interfaces of 3D-printed multi-materials	Tien Thuy Quach	Invited talk at monthly meeting of Polymer club (virtual)
ToF-SIMS to measure residual metal nanoparticle stabiliser in 3D printed electronics	Gustavo F Trindade	SIMS Workshops (virtual)
March 2021		
High-temperature drop-on-demand metaljetting, MetalJet	Negar Gilani	TMS2021: The Minerals, Metals & Materials Society Annual Meeting 2021 (virtual)
May 2021		
Surface analysis of degraded GEMs	Gustavo F Trindade	Invited talk at USP-UFF-UoN-CERN MPGD meeting, Sao Paulo, Brazil (virtual)
Metal additive manufacturing activities at the Centre for Additive Manufacturing, University of Nottingham	Nesma Aboulkhair	Invited Seminar for the University of Brescia, Italy (virtual)
June 2021		
Secondary ion mass spectrometry applications in highly multidisciplinary research	Gustavo F Trindade	Invited talk at LAMFI seminar series for University of Sao Paulo, Brazil (virtual)
August 2021		
Interface characterisation for the next generation of multi-materials additive manufacturing	Tien Thuy Quach	European Advanced Materials Congress (EAMC 2021), Stockholm, Sweden (attended online)
Gold conductive ink formulation with enhanced cohesion for material jetting	Jisun Im	Solid Freeform Fabrication Symposium 2021 (virtual)
Inkjet printing graphene for devices: from contacts to functional layers	Feiran Wang	Solid Freeform Fabrication Symposium 2021 (virtual)
September 2021		
Bioprinting webinar: from benchside to bedside	Laura Ruiz	Medilink East Midlands event (virtual)
Additive manufacturing technologies for regenerative medicine	Laura Ruiz	EPSRC Special Focus Day: Engineering and Physical Sciences in Regenerative Medicine (virtual)
Quantum tunneling of charge carriers in 3D printed graphene functional devices	Jonathan Gosling	Materials Research Society Fall Meeting 2021 – Boston, M, USA

Posters

Developing micro/nanoscale analyses for next-generation multi-functional 3D-printed products	Tien Thuy Quach	Allied Health Professional Postgraduate Research Conference 2021, University of Nottingham
3D-printing of medicines and implants: making the formulations work	Clive Roberts	12th World Meeting on Pharmaceutics and Biopharmaceutics
Additive manufacture of vascularized biocompatible scaffold for bone tissue engineering	Nur Rofiqoh Eviana Putri	RSC Biomaterials Chemistry Group Annual Meeting
Micro/nanoscale analyses of the interfaces of the next-generation 3D-printed multi-materials	Tien Thuy Quach	Formative Formulation 2 Conference
Micro/nanoscale characterisation for the next-generation 3D-printed multi-materials	Tien Thuy Quach	The FORGE: Characterization of Pharmaceutical Formulations
Additive manufacture of vascularised hybrid scaffolds for bone tissue engineering	Nur Rofiqoh Eviana Putri	Tissue and Cell Engineering Society (TCES) 2021 Virtual Conference
Tuning the release of multidrug personalized implants by microstructural control of polymers during 3D printing	Laura Ruiz Cantu	Tissue Engineering and Regenerative Medicine Congress 2021
Inter-flake quantum transport in inkjet-printed graphene devices: from contacts to functional layers	Feiran Wang	Euromat 2021
2D quantum transport in graphene: from exfoliated single layer flakes to inkjet-printed	Feiran Wang	The 24th International Conference on Electronic Properties of Two-Dimensional Systems

Additive International spotlight event

Advances in Multi-Material, Multifunctional Additive Manufacturing for 3D Printed Electronics and Pharmaceuticals – 18 March 2021, Nottingham, UK

This online conference, open to all, was aimed at disseminating the research of the Programme to a wider audience beyond just academia. In total, over 250 people joined this event from around the world. The speakers included:

Understanding how multi-materials interact: novel multiscale chemical and physical analysis techniques for multifunctional AM	Gustavo F Trindade
Can we print pills? Advances in multi-material 3D inkjet printing for pharmaceutical devices	Yinfeng He
Hot metal: how do we develop 'colour' metal printing?	Nesma Aboulkhair
Modelling for (multi) functionality – understanding how to optimise deposition in 3D	Mykyta Chubynsky
3D electronics: building blocks for 3D printed sensors and computers	Geoff Rivers, Jisun Im and Fieran Wang

The year ahead

The Programme continues to run well and to schedule, where much progress within the exciting field of multimaterial additive manufacturing (AM) and the understanding of how to interface dissimilar materials has been made – this very much puts the UK at the forefront of this next generation AM research domain.

As the Programme moves into its fifth year, the development of our PhD students and post-doctoral researchers into leading scientists and engineers in industry and academia remains one of our key priorities. As highlighted within this report, we have already seen graduated PhDs become post-doctoral researchers within the Programme as well as our transitional fellows, Yinfeng He and Laura Ruiz Cantu, making the move to become full academics, joining the Faculty of Engineering at Nottingham as assistant professors.

As highlighted within our midterm review, both the researchers and aligned cohort of PhD students will increasingly focus on gaining impact from the Programme by using a device-led approach to drive the direction of their research. We believe that this approach – which will use specific applications upon which to focus our research activity – will result in us gaining significant traction in work associated with our application areas of pharmaceutical devices, 3D electronics and, increasingly, regenerative medicine and devices.

In order to retain our talented researchers and maintain our position as leaders in this field, we will continue to pursue related funding applications with EPSRC and other strategic partners and build on the strong relationships with our high-profile collaborators. We will continue to communicate our work by publishing in high-impact journals and taking part in outreach and engagement activities. Now that Covid-19 restrictions are subsiding, we also intend to reinstate our exchange programme with institutions such as Lawrence Livermore National Laboratories in the US and will encourage researchers to disseminate their work at high quality research conferences.

In the year ahead the Programme will concentrate on:

Analytical methods

- Continue to exploit 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 NPL and concentrating on devices, further explore the use of the 3D orbiSIMS instrument to exploit its potential for multimaterial AM
- Develop analytical methods for new kinds of multimaterials produced within the Programme

Processes

- Install new multimaterial jetting system based on Xaar 1003 printheads to enable printing of more viscous ink formulations
- Develop a low-cost multimaterial jetting platform based on Xaar 128 printheads expedite functional material development and formulation
- Complete the construction of our multi-head MetalJet platform to co-deposit dissimilar materials in the same print volume, for multi-functionality
- Develop new metal jetting materials based on alloys (Cu-based and Al-based)
- Investigate the integration of a high-speed camera into the single material MetalJet platform to characterise and model ejection and impact dynamics
- Complete the development of two multimaterial PμSLA systems to enable the co-printing of multiple materials for biochip and implant applications
- Continue to expand the database of photo reactive functional ink formulations for PμSLA to enable the production of high-performance devices for biomedical and electronic applications
- Further optimise the Toucan multi-material inkjetting platform to enable printing of complex multi-material geometries
- Develop 2PP deposition of functional materials and/or ordered sub-micron arrays for applications in electronic and optoelectronic devices.
- Continue to explore opportunities for the post-deposition functionalisation of 2PP structures with nanomaterials
- Develop the volumetric CAL printing system and investigate the potential for multimaterial deposition

Modelling

- Investigate a predicative model for the microstructure of droplets in MetalJet
- Incorporate heat transfer and phase transition effects into predictive modelling for MetalJet drop spreading/solidification and validate the resulting models by comparing their predictions to the results of the experimental researchers
- Further optimise inkjet printing by incorporating CFD modelling into the recently developed model to give detailed predictions, including for single and compound drop formation and coalescence, and UV cure and solidification
- Further develop the 2PP theory into a computational modelling tool, carry out experimental validation of its predictions and use it to inform future manufacturing strategies

Functional materials and demonstrators

- Concentrating on the creation of functional demonstrators, alongside the investigation of both conductive and dielectric materials for inkjet printing, we will further investigate the development of inkjet printing for perovskite nanocrystals and printing of photodetectors and LEDs
- Additionally, we will further investigate post-deposition functionalisation of low dimensional materials to inkjet print other electronic and optoelectronic devices such as capacitive sensors, digital processors with multiple vertically stacked 2D material layers, and photon sensors with graphene functionalised by optically active materials
- In terms of healthcare applications, alongside our leading work in the field of pharmaceutical polypills, we will extend our bio printing activities to investigate the printing an artificial liver model
- Though at a low TRL level, we will investigate the use of multi-metal deposition from high temperature MetalJet for the creation of functional electro-mechanical devices and sensors
- Further investigate green chemistry PEDOT:PSS materials
- Provide prototypes of functional electronics including a drug release monitoring RF sensor for a transdermal skin patch, Surface Enhanced Raman Scattering (SERS) based sensors via selective gold decoration on 2PP nanostructures, and piezoelectric energy harvesters.
- Demonstrate the personalisable manufacture of multimaterial inkjet 3D devices printing through generative design guided co-deposition of inks to create functional composites that are both resistant to bacterial biofilm formation and achieve a specific deformation profile

Management structure

Executive team

Professor Richard Hague
University of Nottingham
Principal Investigator

Professor Clive Roberts
University of Nottingham
Co-Investigator

Professor Ian Ashcroft
University of Nottingham
Co-Investigator

Dr James Sprittles
University of Warwick
Co-Investigator

Professor Ricky Wildman
University of Nottingham
Co-Investigator

Professor Derek Irvine
University of Nottingham
Co-Investigator

Professor Chris Tuck
University of Nottingham
Co-Investigator

Professor Mark Fromhold
University of Nottingham
Co-Investigator

Professor Yulii Shikhmurzaev
University of Birmingham
Co-Investigator

Dr Lyudmila Turyanska
University of Nottingham
Co-Investigator

Flavia Guzman Villarroel
University of Nottingham
Programme Manager

Advisory board

Dr Will Barton
Chair, WillB Consulting

Rebecca Cheesbrough
Engineering and Physical Science Research Council (EPSRC)

Professor David Rosen
Georgia Tech and SUTD

Rebecca Mangham
Defence Science and Technology Laboratory (dstl)

Mark Swan
Atomic Weapons Establishment (AWE)

Dr Jonathan Booth
AstraZeneca

Professor Ian Gilmore
National Physical Laboratory (NPL)

Dr Christopher Spadaccini
Lawrence Livermore National Laboratory (LLNL)

René Waarsing
Canon Production Printing

Our partners and collaborators

We thank all our partners who have supported the delivery of the Enabling Next Generation Additive Manufacturing Programme, including our industrial collaborators, allied research institutions and technology organisations and our funder, EPSRC.

Professor Christopher Barner-Kowolik
Queensland University of Technology (QUT), Karlsruhe Institute of Technology (KIT)

Professor Martin Wegener
Karlsruhe Institute of Technology (KIT)

Dr Christopher Spadaccini
Lawrence Livermore National Laboratory (LLNL)

Professor Ian Gilmore
National Physical Laboratory (NPL)

Dr Paul Gellert
AstraZeneca

Martin Hermatschweiler
Nanoscribe GmbH

Jörg Smolenski
Nanoscribe GmbH

Trevor Polidore
Rogers Corporation

Mark Swan
Atomic Weapons Establishment (AWE)

René Waarsing
Canon Production Printing

Dr Benjamin Cook
Texas Instruments

Sean Chang
Texas Instruments

Laura Jones
Defence Science and Technology Laboratory (dstl)

Rebecca Mangham
Defence Science and Technology Laboratory (dstl)

David Hallam
Defence Science and Technology Laboratory (dstl)

Dr Brian Henry
Pfizer

Dr Martin Rowland
Pfizer

For general enquires please contact:

Flavia M. G. Villarroel
Programme Manager
Centre for Additive Manufacturing
Faculty of Engineering
Advanced Manufacturing Building
Jubilee Campus
Nottingham
NG8 1BB

Flavia.GuzmanVillarroel@nottingham.ac.uk

nottingham.ac.uk/NextGenAM

This publication is available in alternative formats:

+44 (0)115 951 5559

Printed June 2022.