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Multiscale additive manufacturing of electronics and biomedical devices

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ABSTRACT

Recent advances in 3D printing have enabled the creation of novel 3D constructs and devices with an unprecedented level of complexity, properties, and functionalities. In contrast to manufacturing techniques developed for mass production, 3D printing encompasses a broad class of fabrication technologies that can enable 1) the creation of highly customized and optimized 3D physical architectures from digital designs; 2) the synergistic integration of properties and functionalities of distinct classes of materials to create novel hybrid devices; and 3) a biocompatible fabrication approach that facilitates the creation and co-integration of biological constructs and systems. Developing the ability to 3D print various classes of materials possessing distinct properties could enable the freeform generation of active electronics in unique functional, interwoven architectures. Here we are developing a multiscale 3D printing approach that enables the integration of diverse classes of materials to create a variety of 3D printed electronics and functional devices with active properties that are not easily achieved using standard microfabrication techniques. In one of the examples, we demonstrate an approach to prolong the gastric residence of wireless electronics to weeks via multimaterial three-dimensional design and fabrication. The surgical-free approach to integrate biomedical electronics with the human body can revolutionize telemedicine by enabling a real-time diagnosis and delivery of therapeutic agents.

Keywords: 3D printing, additive manufacturing, ingestible electronics, gastric resident electronics, biomedical devices.

1. INTRODUCTION

Our group focuses on the development of novel additive manufacturing technologies to create unique functional, interwoven architectures and devices that cannot be created with conventional fabrication methods. We seek to advance the scientific understanding that enables the assembling and processing of functional nanomaterials as well as the seamless integration of diverse classes of materials. We develop a multiscale, multimaterial 3D printing approach that is fundamentally free from the constraint of the conventional two-dimensional, top-down fabrication methodologies[1]. The abilities to create freeform multiscale functional architectures and devices could overcome the geometrical, mechanical and material dichotomies between conventional manufacturing technologies and a broad range of three-dimensional systems. Ultimately, we strive to address unmet clinical needs by creating tailorable three-dimensional free-form biomedical devices with our advanced manufacturing technologies.

1.1 Multiscale Additive Manufacturing

Additive manufacturing (3D printing) is defined as a process by which material is joined or solidified under computer control to create a three-dimensional object. Additive manufacturing is a broad class of manufacturing technologies[2]: the first demonstration of such platform was stereolithography, invented as early as the 1980s by Chuck Hull. Over the past decades, a wide range of additive manufacturing technologies with unique capabilities has been developed. Novel technology such as two-photon polymerization can create micron-scale feature; while Continuous Liquid Interface Production (CLIP) can produce relatively seamless 3D printed parts at a faster speed. In general, additive manufacturing platform can create highly-customized three-dimensional physical architecture from digital design has been used as an industrial rapid prototyping tool. However, most of these platforms have a limited set of materials and are used to create passive, structural constructs.

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Extrusion-based additive manufacturing is an attractive subset of additive manufacturing – it is a versatile and affordable platform where materials are extruded through a nozzle. In contrast to other additive manufacturing methods, it can accommodate a wide range of viscosities (as high as 10^6 mPa·s) and with disparate properties. This has enabled the incorporation of different classes of materials from a wide range of length scales, such as nanomaterials, ceramics, metals, polymers, to biomaterials, cells, tissues, and organ. For example, functional nanomaterials can be formulated to form solution-processable inks[3], as shown in Figure 1A. Indeed, nanomaterials are materials particularly attractive building blocks for devices. These are materials where at least one of their length scales is approximately 1 – 100 nm. Nanomaterials properties are highly tunable – for example, below the Bohr exciton radius, the band gap of semiconducting nanomaterials is dependent on the size. Further, as the particle size decreases, the surface to volume ratio increases. This has important geometric effect: for instance, silver nanoparticles can be sintered at a lower temperature than its bulk counterpart. Importantly, nanomaterials approach length scales in which van der Waals interactions become dominant. This enables the patterning and coating of nanomaterials on an arbitrarily-shaped three-dimensional substrate with high adhesion[3], as shown in Figure 1B. The co-printing of functional inks with structural or biological construct using a multimaterial 3D printer can create macroscale hybrid construct and devices[3,4,5,6], as shown in Figure 1C.

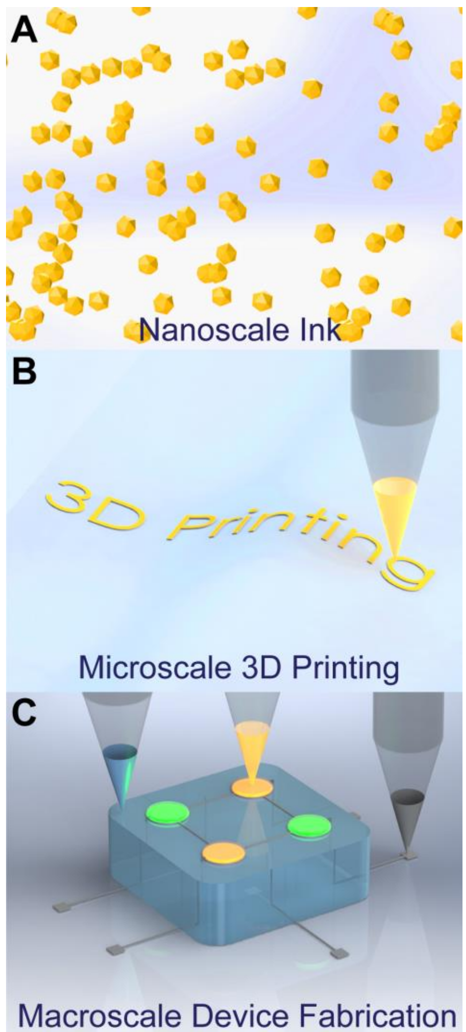

Figure 1: Multiscale additive manufacturing. (A) Solution-processable inks can be created with functional nanomaterials or polymer. (B) The inks are then patterned at microscales via microextrusion. (C) Macroscale functional devices can be created via the three-dimensional co-printing of various classes of materials. Reproduced with permission from Ref. [3]. Copyright 2016 Elsevier.

2. MULTISCALE ADDITIVE MANUFACTURING OF ACTIVE ELECTRONICS AND DEVICES

The ability to 3D print various classes of materials possessing distinct properties could enable the freeform generation of active electronics in unique functional, interwoven architectures. Achieving seamless integration of diverse materials with additive manufacturing is a significant challenge, which requires overcoming discrepancies in material properties in addition to ensuring that all the materials are compatible with the 3D printing process. Indeed, to date, 3D printing has been limited to specific plastics, passive conductors, and a few biological materials. Here we are developing a multiscale extrusion-based 3D printing approach that enables the integration of a diverse class of materials to create a variety of 3D printed electronics and functional devices with active properties that are not easily achieved using standard microfabrication techniques.

2.1 Microscale patterning

Solution-processed functional polymer and nanomaterial inks can enable the creation of devices where the film properties (thickness, morphology, etc.) is critical to determine the device performance. In most microfabricated electronics and devices, such properties are modulated with processes such as spin-coating, in which most solution is expelled through centrifugal forces to create a uniform thin film[3]. However, unlike such film, a colloidal droplet or function polymer printed on a surface typically do not form a uniform layer due to a complex drying and assembling process of the particles. For example, as shown in **Figure 2** (left), “coffee-ring” is formed where the enhanced drying rate at the edge of a pinned contact line has driven the suspended nanomaterials or functional polymers to deposit at the edge of the printed droplet.

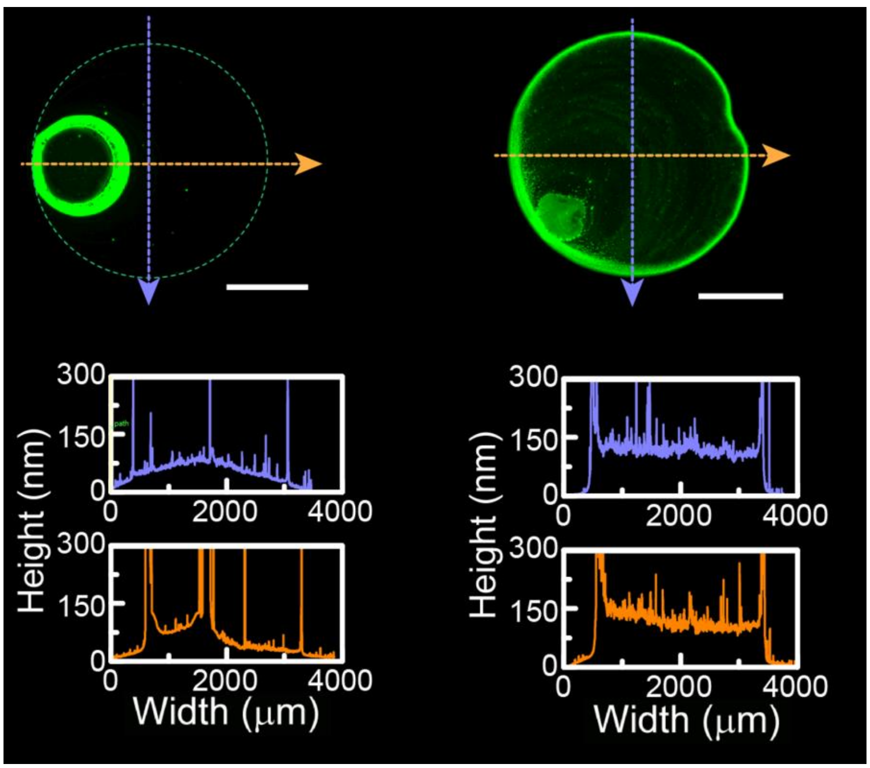

Figure 2: Non-uniformity can be reduced by introducing a co-solvent. The left figure shows the deposition of quantum dots from pure toluene, while the right figure shows an improvement in the morphology via the introduction of 20% dichlorobenzene. Scale bar is 1 mm. Reproduced with permission from Ref. [7]. Copyright 2014 American Chemical Society.

Several strategies can improve the uniformity of the ink to overcome the non-homogeneous deposition. For example, directed or self-assembly with external forces such as magnetic, ultrasound or electric field have been previously demonstrated[3]. Co-solvent can also introduce a surface tension gradient that generates the Marangoni flow, which at the right composition can mitigate the effect. For example, Figure 2B shows a significant improvement in uniformity due to the addition of co-solvent. The root-mean-square roughness of the resulting quantum dots film decreases by 9-fold in comparison to a deposition with pure toluene [7].

2.2 3D printed quantum-dots light-emitting diode (QD-LED)

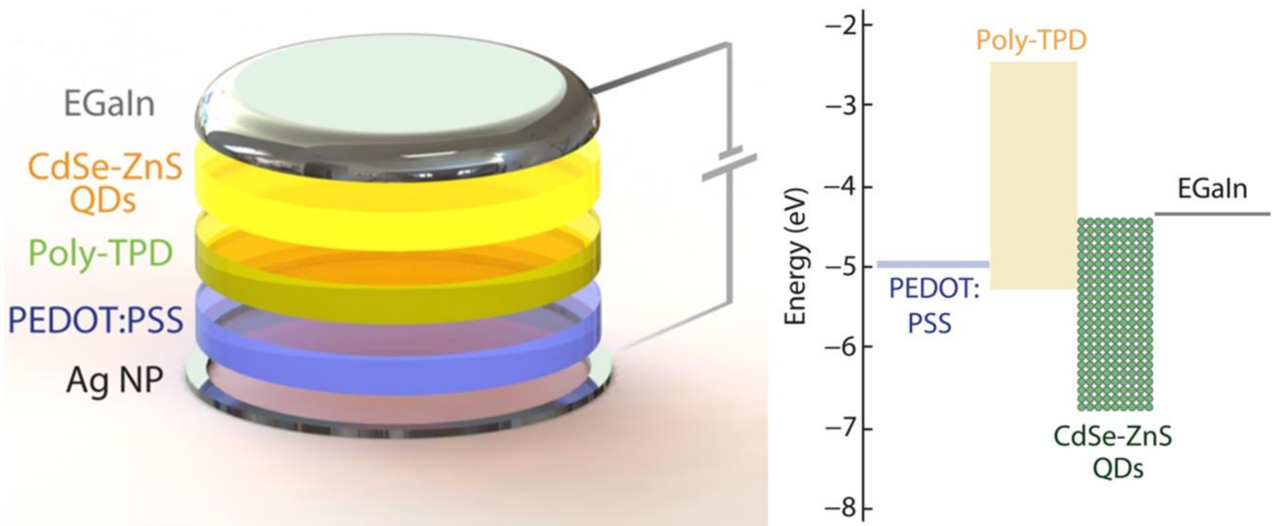

Figure 3: Schematic of the QD-LED components (left) and the energy level diagram (right). Reproduced with permission from Ref. [7]. Copyright 2014 American Chemical Society.

For example, as a proof of concept, we developed an entirely 3D printed quantum dots light emitting diode by integrating five different materials[7] including (1) emissive semiconducting inorganic nanoparticles, (2) an elastomeric matrix, (3) charge transport layers, (4) metal and (5) UV adhesive substrate layer. Specifically, we designed the electronics with the following layer as shown in Figure 3 with (1) cadmium selenide/zinc sulfide (CdSe/ZnS) core-shell QDs as the emission layer, (2) poly[N,N′-bis(4-butylphenyl)-N,N′-bis(phenyl)-benzidine] (poly-TPD) as the hole transport layer, (3) poly(ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS) as the transparent anode, surrounded by (4) a sintered silver nanoparticle (AgNP) ring metallic interconnect, and (5) a eutectic gallium indium liquid metal (EGaIn) cathode.

The printed QD-LED has a maximum brightness of 250 cd/m^2 at 5 V for the green LED as shown in **Figure 4**A. The green QD-LED can emit 100 cd/m^2 at 4.5 V, in comparison to 9.3 V required for direct-current driven partially inkjet-printed based QD-LEDs, or 12.2V for mist-coated QD-LED.

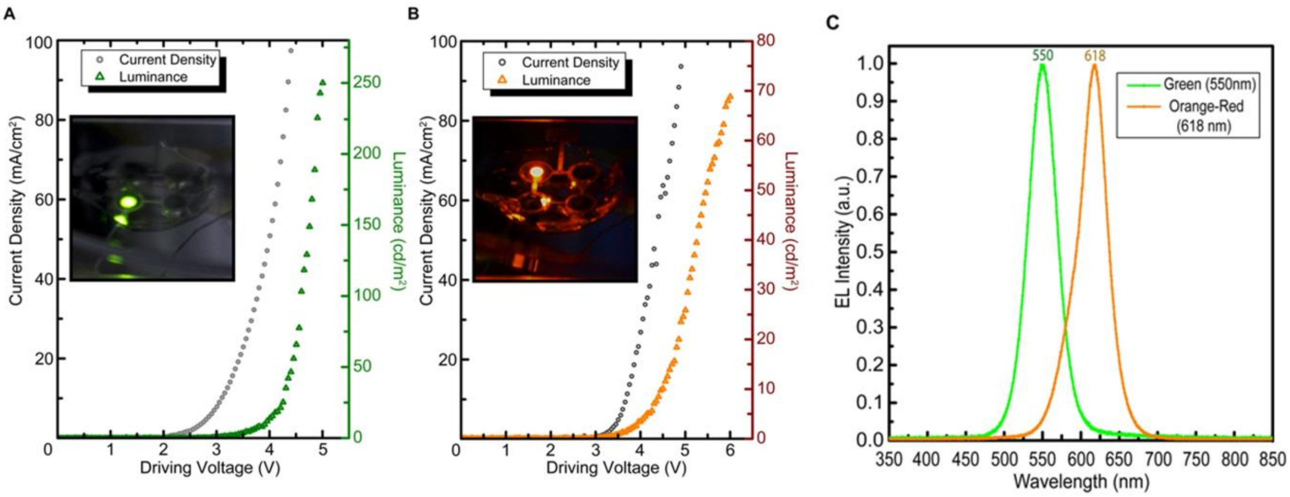

Figure 4: Performance of 3D printed QD-LEDs. Current density-voltage curves and forward luminance output of printed (A) green and (B) orange-red QD LEDs. Inset shows the printed QD-LED. (C) Normalized electroluminescence spectra with peak emission intensities. Reproduced with permission from Ref. [7]. Copyright 2014 American Chemical Society.

The versatility of this approach is demonstrated by printing the QD-LEDs on polyimide tape, which can then be transferred to a wide range of substrates including paper, polycarbonate and nitrile glove.

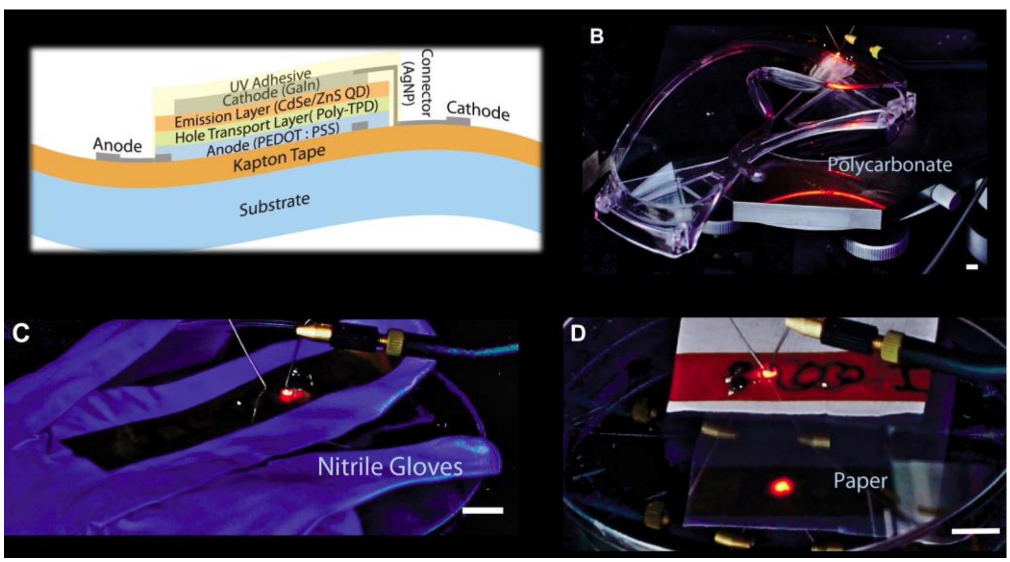

Figure 5: The printing and integration of QD-LED on a wide range of surfaces including polyimide (Kapton) tape, which allows interfacing with polycarbonate, nitrile gloves, and paper.[7] Reproduced with permission from Ref.[7]. Copyright 2014 American Chemical Society.

Further, 3D scanning can be used to acquire the geometry of a nonplanar object, which can be integrated in the Computer Aided Design (CAD) of the electronics. The QD-LED can then be precisely designed to match the topology and upon 3D printing can conformally interface with the 3D surface. Specifically, here 3D structured-light scanning is used to create a compatible computer model of the target surface geometry (a contact lens), which can then be used as a substrate template for the 3D printing process (Figure 6). The ability to 3D print an entire electronics can also allow the creation and fabrication of devices in a layer-by-layer process, which can enable the creation of electronics elements within a 3D printed construct (Figure 6, bottom).

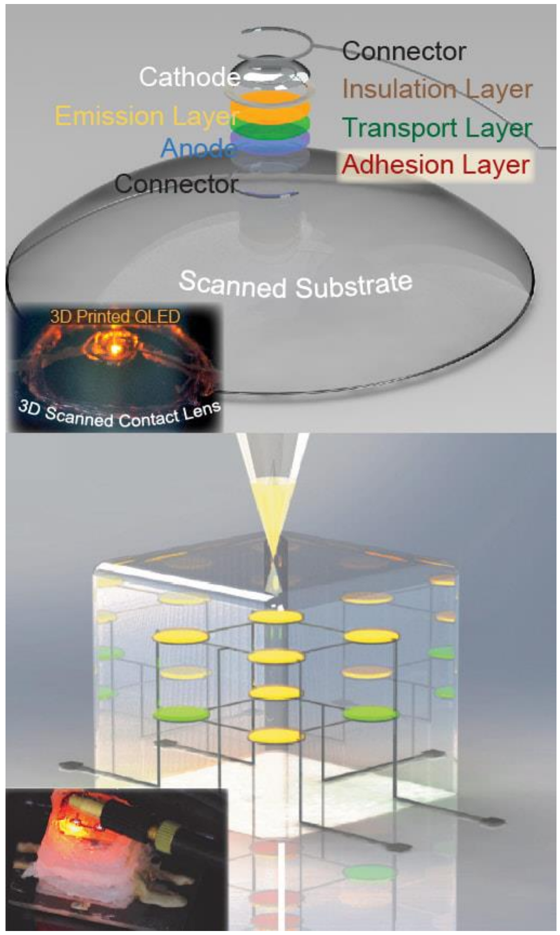

Figure 6: Top image: 3D printed QD-LED on a 3D scanned contact lens: CAD model shows the QD-LED components. Inset shows the electroluminescence of 3D Printed QD-LED on the contact lens. Bottom image: 3D printed multidimensional arrays of QD-LEDs: CAD model of the 2 x 2 x 2 model and inset shows the electroluminescence of 3D Printed QD-LED.[7] Reproduced with permission from Ref. [7]. Copyright 2014 American Chemical Society.

Indeed, developing the ability to govern the assemblies of functional nanomaterials and polymers can impart an otherwise passive three-dimensional construct with active functionalities. We study complex fluids mechanism and soft matter physics phenomenon to govern a variety of complex deposition process to achieve multiscale functional device printing at a variety of constructs[8,9]. For instance, micro-scale printing of nanomaterials can be achieved with directed or self-assembly based methods to generate a functional architecture without the need for conventional fabrication processes that are typically incompatible with a three-dimensional construct. The understanding of the mechanism of the deposition process, such as the evaporation kinetics, colloidal drying and assemblies phenomena could enable the creation of functional meso-scale architecture on or inside a variety of three-dimensional constructs that cannot be fabricated otherwise. Ultimately, the synergistic integration of the micro-scale assemblies of functional materials with advanced manufacturing technologies could realize the creation of unique functional devices.

3. INGESTIBLE GASTRIC RESIDENT ELECTRONICS

In addition to promoting interfacing with a wide range of three-dimensional systems via multiscale additive manufacturing, we are developing wireless ingestible biomedical electronics platform as the next generation remote monitoring, diagnosis, and treatment platform[10,11]. The surgical-free biomedical electronics integration with the human body can revolutionize telemedicine by enabling a real-time diagnosis and delivery of therapeutic agents. Towards this aim, we create functional materials, design unique architectures, and develop a hybrid fabrication approach to enable the creation of highly-functional and safe ingestible biomedical electronics. For example, we invented the concept of gastric resident electronics (GRE) as described in Figure 7. We leverage the gastric environment to allow, for the first time, a long-term (at least multi-weeks long) gastric residence of an orally delivered ingestible electronics. Advancement in multimaterial additive manufacturing enables the GRE to reside in the hostile gastric environment for a maximum of 36 days (Figure 8) with at least 15 days (Figure 9) of bilateral wireless electronic communication.

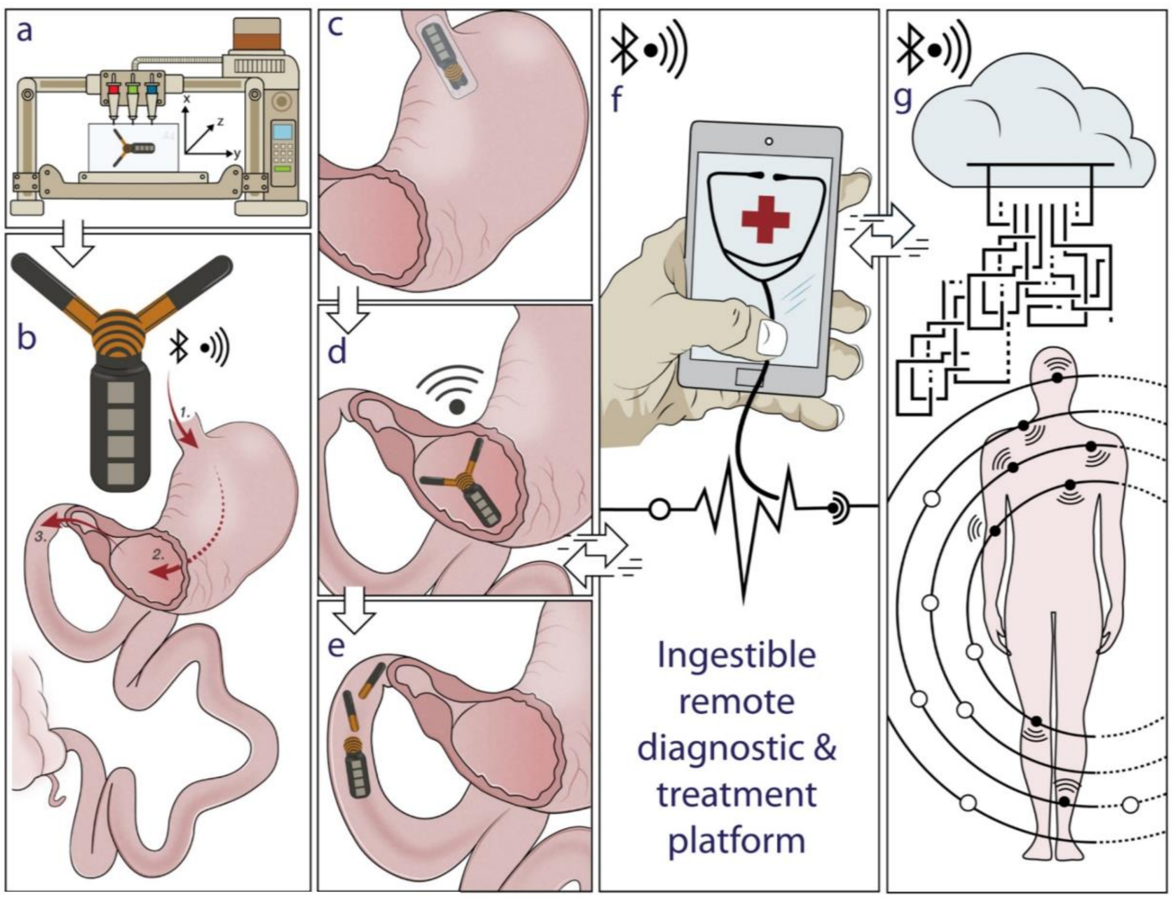

Figure 7: Gastric resident electronics (GRE). Illustration describes the GRE concept, (a) which can be achieved with multimaterial additive manufacturing. (b) For example, patient-specific GRE can be 3D printed and designed to be delivered orally (1.) and reside in the gastric space (2.) before disintegrated and removed from (3.). (c) Specifically, the GRE can transform between a capsule-size dosage form and (d) an expanded dimension enabling gastric residence. (e) Eventually, the device will disintegrate to enables the removal of GRE from the stomach. (f) This can enable long-term wireless inter-communication with a smartphone, for instance through Bluetooth. (g) The ability to achieve real-time, wireless communication with a smartphone enables interconnection of GRE with the digital cloud. Reproduced from Ref. [10] under the terms of the Creative Commons Attribution License (CC BY). Copyright 2018 John Wiley and Sons.

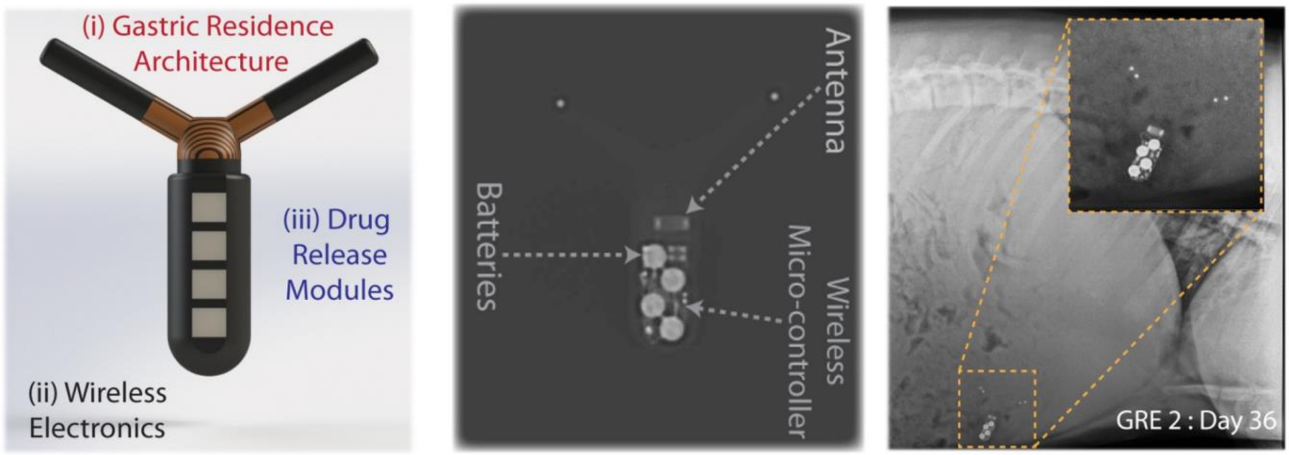

Figure 8: Gastric Resident Electronics (GRE): (Left) Schematic of the GRE, which consists of gastric residence architecture, wireless electronics, and drug release modules. (Center) X-ray image of the device, showing electrical components that contains batteries, wireless Bluetooth micro-controller. (Right) GRE that achieved a maximum gastric residence of 36 days. Reproduced from Ref. [10] under the terms of the Creative Commons Attribution License (CC BY). Copyright 2018 John Wiley and Sons.

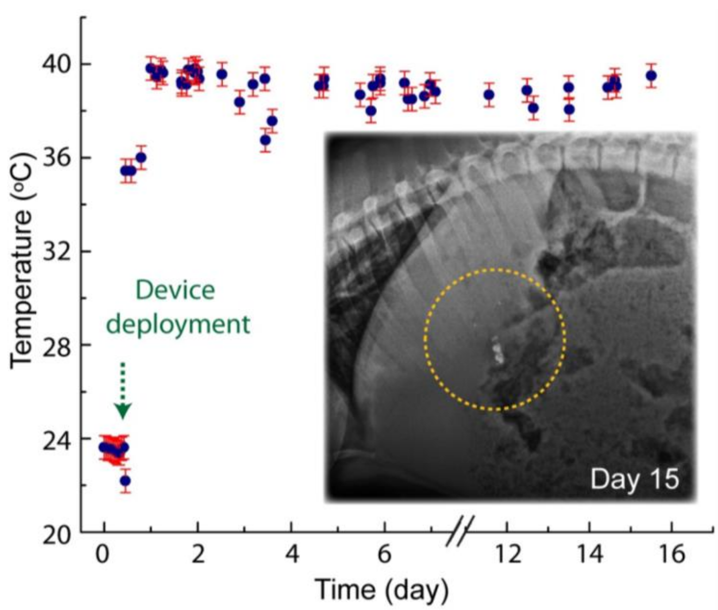

Figure 9: *In vivo*, real-time, bi-lateral communication and control of GRE with a personal electronics. As proof of concept, we demonstrated the first gastric resident electronics in a large animal model. Reproduced from Ref. [10] under the terms of the Creative Commons Attribution License (CC BY). Copyright 2018 John Wiley and Sons.

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