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Extrusion printing was investigated as a wet-processing method for fabrication of robust, flexible conducting structures. Layer resistance values of 7 - 8 kΩ/cm were obtained for one printed layer on flexible substrates. Increasing the number of extrusion printed layer significantly improved resistance.

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Extrusion, printing, conducting, gel, carbon, nanotube, structures, upon, flexible, substrates

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Extrusion printing conducting gel-carbon nanotube structures upon flexible substrates.

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1 Introduction:
Soft and flexible electronic materials [1] will play a significant role in the future development of medical implants and prosthetic devices. One way of approaching this problem is to investigate combinations of conducting fillers and “soft” biopolymer matrix materials. The challenges include (but are not limited to): developing reliable wet-process fabrication methods and achieving suitable characteristics such as low electrical resistance as well as the ability to function under strain.

Carbon nanotubes (CNTs) show great promise as a conducting filler, but are difficult to wet-process due to their extreme hydrophobic nature [2]. A range of biopolymers have been identified as suitable hosts for CNTs. For example, gellan gum (LAGG) is a biopolymer approved by both the US FDA and the EU (E418) for food and medical usage and is showing promise as a bio-material [3].

Wet-processing methods such as inkjet and extrusion printing are emerging as a suitable method for fabricating conducting materials [4]. Of these, extrusion printing offers rapid prototyping, as it can quickly and reliably deposit large volumes through cheap and replaceable parts.

In this work, gellan gum is used to prepare inks with a high CNT loading fraction. These inks are then used to extrusion print multiple layers of conducting structures on rigid and flexible substrates.

2 Experimental Details:
2.1 Ink preparation: Solutions of low acyl gellan gum (LAGG, CP Kelco Gelzan CM, Lot #9K6968A) were prepared by homogenising 2 g in 100 mL of 70 °C Milli-Q water at 4,000 rpm (Wisemix HG-15D with a HD 1018 Tool Head) and diluted to the required concentration.

Multi-walled carbon nanotubes (MWNT, Nanocyl 3100, Batch 090901) were dispersed in LAGG solutions through sonication (Branson 450 digital sonifier, 30 min, 0.5 s pulse on/off, 20 W, 1 mm tip diameter). The viscosity of the dispersions was increased through mixing with 2 wt% LAGG.

2.2 Extrusion Printing: LAGG-MWNT inks were extruded using a custom-built syringe printer. This consists of a gas pressure controller (EFD Ultimus I, under Nitrogen) connected to a syringe assembly (3 cc syringe with luer lock 100 μm stainless steel tips) strapped to the vertical axis of a CNC three dimensional stage (Sherline 8020 CNC 8-Direction Vertical Mill). The pressure and substrate feed rate was controlled to deliver 0.5 μL/cm. Substrates included glass (microscope slides), photo-paper (Kodak Premium Matt) and gel films prepared by evaporative casting (10 mL of 2wt% LAGG dried inside a 100 x 50 x 3 mm3 tray at 50 °C for 12 hrs).

2.3 Characterisation: Ink rheology was measured using the Anton Paar Physica MCR301 Rheometer (PP25 head). Line widths were measured using the Leica Z16APO digital microscope. DC electrical resistance was measured using copper-adhesive tape contacts (3M electrical tape) and a waveform generator (Agilent 33220A) interfaced with a digital multimeter (Agilent 34410A) under controlled am-
bient conditions (21 °C and 45 % relative humidity).

3 Results and Discussion: A high MWNT loading fraction ink of 0.5 wt% LAGG and 0.5 wt% MWNT was prepared through sonication, and diluted with 2 wt% LAGG to a MWNT:LAGG ratio of 1:1.7. The resulting rheology (170 cP at 100 s⁻¹ shear and yield point 3.42 Pa) was appropriate for printing (at 5 PSI).

The total resistance ($R_T$) of extruded lines was found to scale linearly according to:

$$ R_T = l (\sigma A)^{-1} + R_c, $$

where $l$, $A$, $\sigma$ and $R_c$ are the sample’s length, cross-sectional area, electrical conductivity and electrode-sample contact resistance, respectively. The slope of the straight line fits is inversely proportional to the conductivity. Fig. 1 shows that increasing the number of extruded layers (on glass) improves the layer resistance $(\sigma A)^{-1}$ from 3.0 ± 0.5 kΩ/cm to 0.7 ± 0.2 kΩ/cm.

Printing upon two flexible substrates (regular photo paper and a LAGG film) produced a radical decrease in line thickness with a modest increase of layer resistance (Fig 2). The printed structures on photo paper and LAGG displayed robust flexibility, as they can be bent without significantly affecting performance (results not shown).

Fig. 2. A) Layer resistance (diamonds) and layer width (squares) of extrusion printed structures on glass, photo paper and LAGG substrates; B) and C) Photographs of bended layers on photo paper and LAGG, respectively.

4 Conclusion: The fabrication of robust flexible conducting structures by extrusion printing is reported. It was established that gellan gum is a good rheology modifier for the MWNT inks. Layer resistance values of 7 – 8 kΩ/cm were obtained for one printed layer on the flexible substrates. Increasing the number of extrusion printed layer significantly improved resistance. This work contributes to the development of flexible and conducting materials.

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