

Title: Printed electronics

Standfirst: Felice Torrisi and Jonathan Coleman describe how graphene can be used in conductive inks to print electronic circuits.

Printed electronics<sup>1,2</sup> is an important new direction for electronics beyond conventional silicon-based technologies. Electronic devices including radio-frequency identification (RFID) tags, light emitting diodes, batteries, and transistors can be built by printing liquid-dispersed functional materials such as organic polymers, nanoparticles and nanotubes. Techniques including inkjet, roll-to-roll and spray deposition can pattern these materials onto either rigid or flexible substrates, at low temperature and over large areas, and most importantly at very low cost. However, the quality of printed devices is limited by that of the materials contained in the inks. Graphene and other 2D materials are solution-processable and have useful physical and chemical properties. Can they make an impact on printed electronics?

Electronic inks are mixtures of a functional material, stabilizers and rheology modifiers. Different inks are needed to print the different elements of devices: semiconducting inks (the functional materials) in the active layer, insulating materials for dielectrics, and conducting materials for electrodes. They must be stable, cheap and print easily on appropriate substrates to fabricate devices with high charge carrier mobility and long lifetime, without the need of aggressive post-treatments. Finding materials which fulfil these requirements is difficult. For example, conducting inks often contain metallic nanoparticles that must be sintered at high temperatures to maximize conductivity, making them unsuitable for polymer-based substrates. Furthermore, conducting-polymer-based inks are not stable enough for many applications, whereas most nanotube inks contain both semiconducting and metallic tubes.

Graphene fulfils most requirements for use in conducting inks thanks to its high carrier mobility, mechanical robustness, environmental stability and potential for low-cost production. Before an ink can be produced, graphene must first be dispersed as nanosheets in a liquid. Graphene suspensions can be produced from graphite by oxidization to produce graphene oxide, or by liquid phase exfoliation in solvents to give few-layer nanosheets.<sup>3</sup> Large quantities of nanosheets, typically a few microns across and ~1 nm thick, can be produced quickly and easily.<sup>4</sup> The resulting ink is stable, processable in ambient conditions and ideal for printing and coating.<sup>5</sup> Continuous networks of graphene nanosheets have been produced by methods such as spraying, filtration and rod coating and tend to have conductivities of up to  $10^4$  S/m,<sup>6</sup> even without high temperature treatments.<sup>7</sup> They have performed well in applications such as electrodes for supercapacitors, as well as transparent electrodes for liquid crystal displays, thin film transistors (TFT) and smart windows.<sup>5,8</sup> Patterned networks, with features as small as  $30\ \mu\text{m}$ ,<sup>9</sup> have been

produced by inkjet printing<sup>5,7</sup> and gravure coating,<sup>9</sup> and have shown promise in applications from supercapacitors<sup>10</sup> to antennae<sup>11</sup>.

Although graphene can be used as an active material in printed supercapacitors, its lack of a bandgap means that it cannot replace conjugated polymers<sup>2</sup> as the active material in other printed electronic devices. However, in addition to graphene, liquid phase exfoliation can produce suspensions of a range of other 2D nanosheets, including WS<sub>2</sub>, MoO<sub>3</sub> and BN.<sup>3</sup> These 2D materials have diverse properties, some being semiconductors or insulators, and some are electrochemically active, making their inks suitable for a wide range of applications as active materials in printed electronics. Spray coating<sup>12</sup> or inkjet printing<sup>13</sup> of MoS<sub>2</sub> nanosheets have been recently reported for supercapacitor and sensing applications.

However, the most exciting applications of 2D materials in printed electronics will come from printing devices where two or more nanomaterials are integrated together in well-defined structures. All-inkjet-printed heterostructure photodetectors and transistors have already been produced by sequential printing of graphene and WS<sub>2</sub>, MoS<sub>2</sub> or BN.<sup>7,14</sup> It is likely that many developments will stem from these pioneering works.

Nevertheless, a number of major hurdles remain before these developments can lead to commercialization of graphene-based inks. For example, although nanosheets exfoliated by liquid phase exfoliation can be made in large quantities, the yield of monolayers is very low and the flake size is poorly controlled. For inkjet printing, the nanosheet size and rheological properties of the suspension must be carefully tuned.<sup>5</sup> In addition, the high aspect ratio of nanosheets generally results in low concentration suspensions (~0.01-1 wt%) which are problematic for some printing techniques. Electrical performance is another issue. Printed devices will consist of networks of either graphene or inorganic nanosheets, and charge transport will be limited by inter-nanosheet junctions, resulting in mobilities typically two orders of magnitude lower than in individual nanosheets.<sup>15</sup> This will limit the achievable conductivity of graphene interconnects, and reduce the performance of devices based on semiconducting nanosheets. On the other hand, MoS<sub>2</sub> nanosheets have mobilities of hundreds of cm<sup>2</sup>/Vs, and production of MoS<sub>2</sub> networks with mobilities of a few cm<sup>2</sup>/Vs might be possible, making them competitive with printed conjugated-polymer-based devices.<sup>2</sup>

We believe that the future is bright for printed devices based on 2D materials although many problems remain. As new materials combinations are demonstrated, the range of printed devices and applications will expand. In addition, as the scaling up of ink production proceeds, costs will fall, eventually allowing printed devices to compete with traditional technologies on cost if not performance.

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