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Introduction to nanomaterials for printed electronics

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 An introduction to the *Nanoscale* themed collection on nanomaterials for printed electronics, featuring exciting research on a variety of nanomaterials and techniques used for printed electronics.

The digital revolution enabled by the Internet of Things resulted in drastic changes in how goods are used, transported and stored, and how people manage their health and wellness with wearable devices, and has led to more efficient industrial production and cost reductions. In this framework, printed technologies are playing a crucial role, as they enable low-cost fabrication of devices such as transistors, sensors, supercapacitors and solar cells, to name a few examples, with minimal material waste and easy integration into flexible substrates, including paper, textiles and plastic.

The enticing potential of printing technologies has attracted significant interest from both academia and indus-

try, which has resulted in tremendous progress and the expansion of the libraries of ink compositions with new advanced nanomaterials, bringing additional functionalities and/or improved device performance, as compared to traditional materials, such as metallic inks. The family of printable nanomaterials now includes materials with different chemical compositions, from organic to inorganic or hybrids, and of different dimensionalities, from nanoparticles to nanowires and nanosheets. The entire range of electronic materials, from metallic to semiconducting, dielectric, thermoelectric and even superconductive inks, are now available, hence opening up the potential to fabricate the whole device by simply pressing a button on a printer, leading to the realization of customizable electronics accessible to everybody.

This collection of reviews, mini-reviews, communications and research articles covers the rational design, synthesis and characterization of nanomaterials tailored for different types of printed technologies, as well as their use in a wide range of applications.

Conductive inks are one of the most common functional materials in printed devices. Metallic nanoparticles, typically made of silver, are traditionally used as conductive inks. However, silver inks are relatively expensive, have limited flexibility when printed as a film and are also prone to electromigration. Hence, alternative materials are currently being

investigated. Amongst them, copper is very attractive, being abundant and cheap, and having good electrical conductivity. Yang *et al.* provide a review on the preparation and properties of copper nanoparticles, and their formulation into printable inks, as well as sintering processes and anti-oxidation strategies (<https://doi.org/10.1039/D2NR03990G>).

Silver nanowires, carbon nanotubes (CNTs), conductive polymers and metallic 2-dimensional materials, such as graphene and MXenes, can also be used as conductive inks. For example, Zhu *et al.* report on the development of a water-based silver nanowire inks suitable for screen printing, made with a relatively low conductive-material loading and a biodegradable binder, reaching conductivities up to $6.70 \times 10^6 \text{ S m}^{-1}$ (<https://doi.org/10.1039/D2NR05840E>).

Transparent electrodes are widely used for many applications, from sensors to electrochromic devices (ECDs). In this framework, Wu *et al.* (<https://doi.org/10.1039/D2NR03209K>) printed ECDs using a UV-curable solid-state electrolyte based on lithium bis(trifluoromethane-sulfonyl)imide (LiTFSI) and achieved a bleaching time of 0.6 s at 0.6 V and a coloring time of 1.4 s at -0.5 V . The ECDs also exhibited excellent stability, enduring up to 100 000 cycles of color switching, while still maintaining a 35% transmittance at 550 nm.

Amongst semiconducting inks, printed networks of CNTs are widely used as channels in transistors.

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Zaumseil *et al.* report on the use of a network of (6,5) single-walled CNTs as the transducing layer of water-gated transistors to detect Cu^{2+} ions over a wide range of concentrations in aqueous solutions (<https://doi.org/10.1039/D2NR02517E>). Remarkably, the (6,5) CNTs can be employed directly and without additional functionalization or ion-selective membranes. Zhao *et al.* demonstrate large-area ($8 \text{ cm} \times 14 \text{ cm}$) semiconducting single-walled carbon-nanotube thin films on flexible substrates made at a printing speed of 8 m min^{-1} with a roll-to-roll approach (<https://doi.org/10.1039/D2NR07209B>). Transistors made with such networks show a carrier mobility of $\sim 11.9 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, $I_{\text{on}}/I_{\text{off}}$ ratios of $\sim 10^6$, small hysteresis, and a subthreshold swing of $70\text{--}80 \text{ mV dec}^{-1}$ at low operating voltages ($\pm 1 \text{ V}$). In another work, Franklin *et al.* focus on different types of ionic dielectrics for printed transistors made of CNTs (<https://doi.org/10.1039/D2NR04206A>). Ionic dielectrics are very attractive as they enable the devices to operate at low voltages thanks to their high capacitance driven by the electric double layer. Dielectric inks can also be used for making other types of devices; for example, Lanza *et al.* demonstrate memristors made with h-BN-nanosheet inks (<https://doi.org/10.1039/D2NR06222D>), while Casiraghi *et al.* use anatase- TiO_2 -nanosheet water-based inks to demonstrate fully printed diodes on paper (<https://doi.org/10.1039/D2NR05786G>).

Finally, this collection also highlights the large variety of wet deposition techniques available, such as spin, spray, dip and bar coating, as well as various printing methods, like inkjet and aerosol-jet printing. Device fabrication may involve a combination of different printing techniques for the consecutive device layers, depending on the element being fabricated. Coleman *et al.* report the use of spray coating to make a diode integrat-

ing semiconducting CNTs and WS_2 nanosheets (<https://doi.org/10.1039/D2NR04196K>). Cicoira *et al.* describe the fabrication of fully stretchable organic electrochemical transistors by dispenser-printing all components of the device (<https://doi.org/10.1039/D2NR06731E>). To achieve the stretchability, a printed planar gate electrode and polyvinyl alcohol (PVA) hydrogel electrolyte were employed.

Sustainability is also important in developing new inks for printed electronics, making water the best choice of solvent. In this framework, Nielsen *et al.* introduced oligoether side-chain modifications to the polymer backbone to achieve stable nanoparticle dispersions in water without the addition of surfactants or additives (<https://doi.org/10.1039/D2NR06024H>).

The fast progress in the formulation of conductive inks also impacted the field of bioelectronics, as discussed in the mini-review by Guha and collaborators, focused on the self-assembly process of dipeptides (<https://doi.org/10.1039/D2NR03750E>).

Sensing is another area where printing is emerging as an effective process for low-cost electronics. In this collection, we include one example of non-intrusive sensors attached to marine species to monitor the impact of environmental changes on their behaviour and well-being, described by Ng *et al.* (<https://doi.org/10.1039/D2NR04382C>), as well as one example on environmental humidity sensors introduced by Jurchescu *et al.* (<https://doi.org/10.1039/D2NR04498F>). The high surface-to-volume ratio and its exceptionally high mobility make graphene very attractive for chemical sensing. In this collection, Torrisi *et al.* report the easy, low-cost and scalable production of an electrolyte-gated graphene field-effect transistor for chemical-sensor test strips (<https://doi.org/10.1039/D2NR05838C>).

The platform is enabled by the low-boiling-point, low-surface-tension sprayable graphene ink deposited on a substrate manufactured using a commercial printed-circuit-board process.

Aerosol jet printing, in which the ink is first aerosolized, then focused by a second sheath-gas stream and deposited through a tip onto the surface of the substrate, is gaining increasing popularity as it enables printing of inks with a larger range of viscosities, as compared to traditional piezoelectric inkjet printers. Secor *et al.* report the development of titanium hydride nanoparticles and nanoinks for an aerosol jet printer (<https://doi.org/10.1039/D2NR03571E>). Nanoparticles of TiH_2 were generated by heating TiH_2 particles in octylamine followed by wet ball milling. The platelets were then formulated into inks and printed on glass and polyimide. Post-annealing enables reaching a sheet resistance of $\sim 150 \Omega \square^{-1}$.

Finally, 3D printing is also gaining attention, as it enables the fabrication of three-dimensional devices. In this collection, Słoma provides a review on nanomaterials for 3D printing, covering the different types of techniques as well as applications of 3D printing of nanomaterials, ranging from energy-storage devices to sensors and micromechanical systems (<https://doi.org/10.1039/D2NR06771D>).

As guest editors of this themed collection, we would like to thank all of the authors for the high quality of their contributions. Furthermore, we would like to thank the editorial staff from *Nanoscale* for their support, as well as our reviewers. We hope that this themed issue will create an efficient platform for scientists working with different nanomaterials for printed technologies to share their experience, skills and results in order to further accelerate the development and use of printing technologies.



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Prof. Casiraghi holds a Chair in Nanoscience at the Department of Chemistry, University of Manchester (UK). She received her B.Sc. and M.Sc. in Nuclear Engineering from Politecnico di Milano (Italy) and her Ph.D. in Electrical Engineering from the University of Cambridge (UK). In 2005, she was awarded with an Oppenheimer Fellowship, followed by the Humboldt Research Fellowship and the prestigious Kovalevskaja Award (€1.5M). In 2010 she joined the Department of Chemistry at the University of Manchester. Her current research work focuses on the development of biocompatible 2D inks for printed electronics and biomedical applications. She has published more than 100 works, collecting more than 36 000 citations, and has an h-index of 56. She is a leading expert on Raman spectroscopy of carbon nanostructures, as recognised by the RSC Marlow Award (2014). She is also a recipient of the Leverhulme Award in Engineering (2016), and the recent RSC Gibson–Fawcett Award (2020), in recognition of the development of water-based 2D inks. She was also awarded an ERC Consolidator grant (2015), ERC Proof of Concept grant (2020) and ERC Advanced grant (2021). She serves as an editorial board member of Nanoscale and Nanoscale Advances.



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Shlomo Magdassi is a professor at The Hebrew University of Jerusalem's Institute of Chemistry and serves as the academic director for the university's Center for Functional and 3D Printing. His research centers on micro- and nanomaterials, with a focus on their applications in functional 2D and 3D printing. Over the course of his career, he has published more than 340 papers and edited four books, and holds approximately 300 patents and applications. His research outcomes include the creation of numerous commercial activities, including start-up companies, licensing agreements, and worldwide sales. In recognition of his contributions, he was awarded the 2022 Johann Gutenberg Prize by the Society for Imaging Science and Technology, and he is also a Fellow of the National Academy of Inventors.



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