

Over the past two decades research in the field of nanotechnology has fostered the development of new carbon allotropes including individual sheets of graphite, known as graphene, as well as its tubular form known as carbon nanotubes (CNTs). These materials have properties unlike any other engineering material. For instance, they combine sizes below a nanometer in thickness (or diameter) and reach several centimetres in length; hereby bridging molecular and macroscopic scales. Typical features of CNTs and graphene include a Young's Modulus of 1 TPa and a tensile strength of 100 GPa (i.e. higher than steel at only a fraction of its weight), high currents carrying capacity of up to 10^9 A/cm², and thermal conductivities of up to 3500 Wm⁻¹K⁻¹.

These unique properties, along with the development of cost effective CNT mass production processes have sparked not only academic but also a strong industrial interest in carbon nanomaterials. Early on, CNTs were processed with existing high-throughput manufacturing methods such as for instance by injection molding and ink-jet printing. While these techniques have supported the commercialisation and scale-up of these materials, they result in un-organized CNT arrangements whose figures of merit typically drop by an order of magnitude compared to what is measured in individual nanoparticles.

This Marie Curie Career Integration Grant supported the purchase of equipment and consumables to better assemble CNTs in controlled structures and to develop next generation CNT devices. In this work, we found a hierarchical approach that simultaneously optimises the organisation of CNTs at the nano, micro and macro scale to be particularly attractive. To this end we have successfully fabricated a Chemical Vapour Deposition (CVD) tool able to tailor make CNT structures. We have used these structures to (i) fabricate new electrodes for flexible batteries and (ii) for recording electrogenic cells.

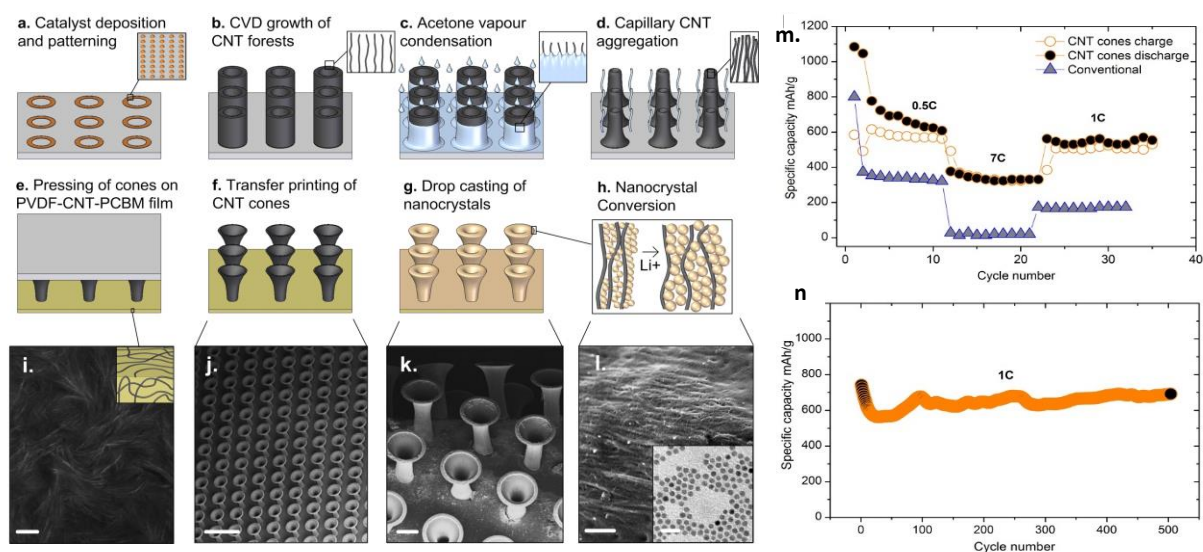


Figure 1. a-h) Schematic fabrication process of hierarchical CNT structures. i-l) SEM image of the flexible CNT electrodes at different stages of the fabrication process. m-n) Electrochemical battery performance of hierarchical and reference electrodes.

(i) Flexible Batteries: The development of flexible batteries is important to support one of the most fascinating paradigm shifts in modern electronics, namely, the development of soft and flexible electronic devices. This development is fuelled by a continuous search for more compact and intuitive consumer electronics, medical implants, and the emergence of the “Internet of Things.” While considerable progress has been made in the fabrication of flexible and stretchable circuits, radio-frequency identification (RFID) tags, and displays, the flexible batteries needed to power these devices remain challenging. Existing flexible batteries are often too heavy, bulky, and rigid, and require a radical redesign of the battery architecture to address these issues. The CANA project demonstrated that extremely flexible batteries can be achieved by designing 3D carbon nanotube (CNT) microstructures

which decouple the stress induced during bending in the collector electrode from stress in the energy-storage material (Fe₂O₃ anodes and LNCO cathodes in this work). The 3D CNT trumpets shown in Figure 1, are particularly interesting for this application because they have a slender base, which reduces stress transfer from the bent collector electrode, and they have a wide crown to load active material. This battery architecture not only imparts excellent flexibility (bending radius $\approx 300\ \mu\text{m}$), but also high capacity ($>650\ \text{mAh/g}$), rate ($20\ \text{A g}^{-1}$), cycling stability (See Figure 1 m-n).

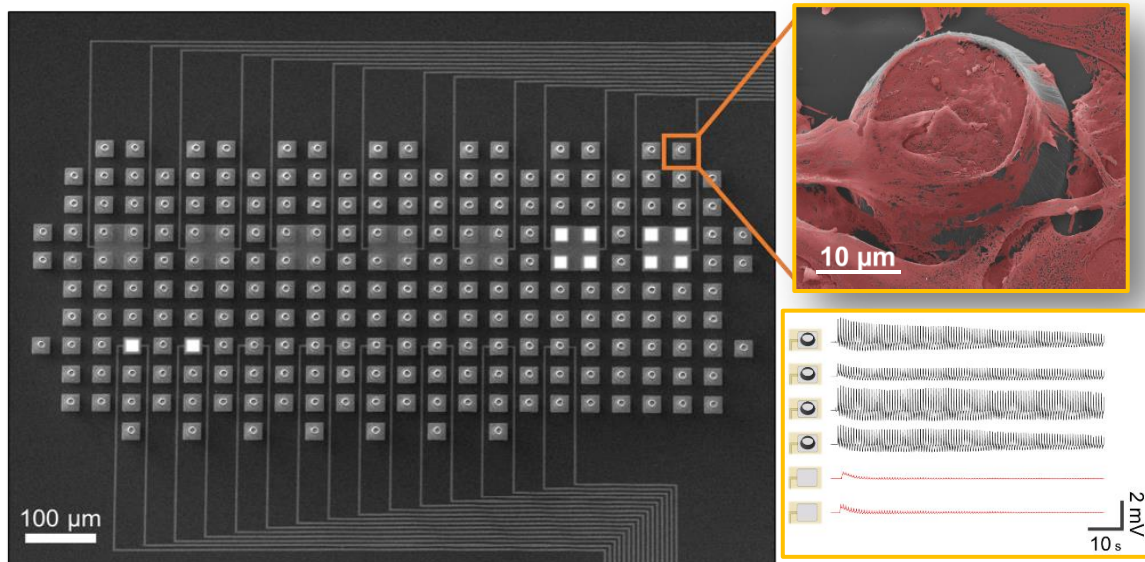


Figure 2. Left: Array of 3D CNT electrodes. Right: SEM image of a cell inside a 3D CNT electrode and cell recording signals from CNT enhanced and reference electrodes.

(ii) Cell Recording: We then discovered that the 3D electrodes developed for flexible batteries also offer a unique opportunity to record electrogenic cells such as cardiomyocytes and neurons. These cells rely on electrical signals for intercellular communication, and the development of Microelectrode Arrays (MEAs) to record such cells is key for understanding the behaviour of electrogenic cells. On the longer term this will contribute to a better understanding of for instance the operation of the brain. To date, MEAs are relying on flat or needle-shaped electrode surfaces, mainly due to limitations in the lithographic processes used to fabricate these devices. The 3D CNT structures developed in CANA are particularly interesting for MEA because i) CNT microwells of the right diameter preferentially trap individual cells, which facilitates single cell recording without the need for clamping cells or signal deconvolution, and ii) once the cells are trapped inside of the CNT wells, this 3D environment surrounds the cell, which increases the cell–electrode contact area. These structures were developed in collaboration with colleagues at imec, Belgium who found that the recorded cell output voltages increase significantly using 3D CNT electrodes (more than 200%) as illustrated in figure 2 right. Therefore, this fabrication process paves the way for future study of complex interactions between electrogenic cells and 3D recording electrodes.

To sum up, CANA achieved the goals that were set out in the project proposal, namely developing advanced CNT applications by harnessing recent developments in: (i) Lithography techniques to control macro- and microscale organization of carbon nanotubematerials (ii) Self-assembly of nanomaterials to create complex microscale geometries (iii) Nanocarbon chemistry to modify the surface of carbon nanomaterials, at nano and sub-nanometer lengthscale. This resulted in the development of state of the art flexible electrodes and cell recording arrays. This work was published in leading academic journals such as *Advanced Materials* and *Advanced Functional Materials*. A third publication is currently in review.

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