Research into energy technologies and electronic devices is strongly governed by the available materials. More recently, carbon allotropes and carbon-rich molecules play an increasingly important role as electronic conductors, semiconductors and catalysts and are attractive alternatives to established organic and inorganic materials. The unique physical and chemical properties of the two-dimensional (2D) π-electron system graphene ask for its chemical synthesis.

We introduce a synthetic route to graphenes which is based upon the cyclodehydrogenation ("graphitization") of well-defined dendritic (3D) polyphenylene precursors. This approach is superior to physical methods of graphene formation such as chemical vapour deposition or exfoliation in terms of its (i) size and shape control, (ii) structural perfection, and (iii) processability (solution, melt, and even gas phase).

Columnar superstructures assembled from these nanographene discs serve as charge transport channels in electronic devices. Field-effect transistors (FETs), solar cells, and sensors are described as examples and their exemplary performance is discussed in terms of supramolecular order and interfacing.

Upon pyrolysis in confining geometries or "carbo-mesophases", the above carbon-rich 2D- and 3D-macromolecules transform into unprecedented carbon materials and their carbon-metal nanocomposites. Exciting applications are shown for battery cells and fuel cells. In the latter case, nitrogen-containing graphenes serve as catalysts for oxygen reduction whose efficiency is superior to that of platinum. Further, transparent and conducting window-electrodes are fabricated which can replace ITO.

References