## Large-Scale Graphene Production for Environmentally Friendly and Low-Cost Energy Storage Production, Coating, and Applications

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Faculty of Science, Technology and Media Thesis for Doctoral Degree in Engineering Physics Mid Sweden University

Sundsvall, 2019-05-10

## ABSTRACT

There is great demand for energy-efficient, environmentally sustainable, and cost-effective electrical energy storage devices. One important aspect of this demand is the need for automotive electrification to achieve more energy-efficient transportation at a reasonable cost, thus supporting a fossil-fuel free society.

Another important aspect is the requirement for energy storage in the growing field of renewable energy production from wind and solar sources, which generates an irregular supply of electricity due to weather conditions. Much of the research in this area has been conducted in the field of battery technology with impressive results, but the need for rapid storage devices such as supercapacitors is growing. Due to the excellent ability of supercapacitors to handle short peak power pulses with high efficiency along with their long lifetime and superior cyclability, their implementations range from small consumer electronics to electric vehicles and stationary grid applications. Supercapacitors also have the potential to complement batteries to improve pulse efficiency and lifetime of the system, however, the cost of supercapacitors is a significant issue for large-scale commercial use, leading to a demand for sustainable, low-cost materials and simplified manufacturing processes. An important way to address this need is to develop a cost-efficient and environment-friendly large-scale process to produce highly conductive nanographites, such as graphene and graphite nanoplatelets, along with methods to manufacture low-cost electrodes from large area coating.

In this thesis, I present a novel process to mechanically exfoliate industrial quantities of nanographite from graphite in an aqueous environment with lowenergy consumption and at controlled shear conditions. The process is based on hydrodynamic tube-shearing and can produce both multilayer graphene and nanometer-thick and micrometer-wide flakes of nanographite. I also describe the production of highly conductive and robust carbon composites based on the addition of nanocellulose during production; these are suitable as electrodes in applications ranging from supercapacitors and batteries to printed electronics and solar cells.

Furthermore I demonstrate a scalable route for roll-to-roll coating of the nanographite-nanocellulose electrode material and propose a novel aqueous, low-cost, and metal-free supercapacitor concept with graphite foil functioning as the current collector. The supercapacitors possessed more than half the specific capacitance of commercial units but achieved a material cost reduction of more than 90 %, demonstrating an environment-friendly, low-cost alternative to conventional supercapacitors.