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Aerosol Jet Printed Nanocarbons on Heat Sink Materials †

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Abstract: Graphene- and carbon nanotube (CNT)-based inks have been printed on relevant heat sink materials by Aerosol jet. The thickness of the layers varied between ~100 and ~1.500 nm. The inks' viscosity ranged from <20 up to 600 cps at a solid content between 0.18 and 3% and wide particle sizes from 5 nm up to 5 µm. The printed layers could be interesting for rather high-power and high-temperature applications including thermal heat spreaders, resistive heaters, high-current carrying interconnectors, temperature sensors and ordnance fuze technology.

Keywords: aerosol-jet printing; nanocarbons; graphene; carbon nanotube (CNT); heat sink; electronic devices; Raman spectroscopy

1. Introduction

Additive manufacturing techniques, such as inkjet and aerosol jet printing (AJP), are being explored as alternative fabrication methods, for example for flexible and wearable devices [1]. Such direct writing techniques allow for rapid customization and prototyping. The high heat generation of sophisticated micro- and nanoelectronic components not only impairs their performance, but also shortens their service life. The development of low-cost, high heat-conducting materials is necessary here. In particular, graphene and carbon nanotubes (CNT), whose heat conductivity is greater than that of reference materials such as graphite, copper and diamond, are suitable for thermal applications.

This work presents graphene and CNT-based inks deposited by AJP on heat sink materials for high-power LED lighting devices. Physical properties like thickness, surface, structure, chemistry and thermal conductivity of printed nanocarbon layers and their relations in a wide printing parameter range have been investigated and are discussed.

2. Materials and Methods

Graphene and carbon nanotubes (CNT) inks have been used to print test patterns at printing speeds between 0.5 and 3 mm/s by an Optomec Aerosol-jet printer on glass slides, (100) polished silicon wafers, and on heat sink materials, i.e., Plasma-electrolytic oxidation (PEO) aluminum (Al) oxide coated and hexagonal boron-nitride (h-BN)/polymer composite coated Al blocks. Additionally, 5 nm SiN membranes were printed for transmission electron microscopy (TEM). The inks' viscosity was measured by viscosimetry; both substrates and printed samples have been characterized by stylus profilometry, atomic force microscopy, Raman spectroscopy, light and scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM-EDS) and X-ray diffractometry (XRD).

3. Results and Discussions

CNTs aqueous inks showed good line definition over a wide range of the AJP parameter space. Coverage was largely continuous at low printing speed, including coverage of µm substrate hole but non-continuous at high speeds. Raman spectroscopy showed that CNTS retained high quality during ink preparation and printing. TEM investigations confirmed that the CNTs are randomly "spaghetti-like" oriented and single-wall nanotubes (SWNTs) are forming bundles in the printed layers. Residual crystalline particles, formed from sodium-cholate surfactant remnants from ink preparation, are embedded in the printed CNT films.

Line definition, coverage and quality of few-layer graphene (FLG) within patterns printed from aqueous graphene inks are similar to CNTs aqueous inks. SEM and TEM images showed a layered morphology of FLG flakes parallel to the substrate with crystalline (002) texture. Some graphene flakes bend out of the substrate plane at their edges.

Lines printed from graphene inks dispersed in H_2O and *N*,*N*-Dimethylformamide showed poor definition at high speeds, and good definition at lower speeds. In the latter case, coverage was also satisfactory. FLG flakes in the printed patterns are of lower quality compared to those printed from pure aqueous inks. Consistently, the films show a layered morphology parallel to the substrate but crystalline texture is less pronounced. The appearance of the printed patterns is more compact compared to those printed from aqueous graphene inks.

4. Conclusions

AJP patterns were successfully deposited on flat and rough ceramic and polymeric substrates. Printing from aqueous inks is possible in a wide AJP parameter range and results in good line definition, coverage and good quality of CNTs and FLG flakes in the patterns. Patterns printed from aqueous inks with added *N*,*N*-Dimethylformamide dispersing agent must be printed at lower speed and suffer from lower CNTs and FLG flake quality, but show a more compact appearance.

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