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A novel method to join carbon nanotube fibers with metal substrates

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ABSTRACT

Electrical and electronic applications of carbon nanotube (CNT) fibers require new methods to join them with other live parts of the circuits electrically and mechanically. In this study, the meniscus-confined localized electrochemical deposition (LECD) from a simple aqueous electrolyte was developed as a feasible method to precisely join CNT fibers with copper substrates in an open air environment successfully. During the LECD process, the meniscus remained stable without extending along the fiber surface. The deposited joints have regular shapes and clear boundaries with sizes close to the dispensing nozzle size. The copper deposit with a uniform and compact microstructure clings tightly to the fiber surface, with a distinct interface forming between the copper deposit and the fiber, even though the fiber surface is uneven. The original CNT network morphology remains largely unaffected. The joint surface roughness can be as low as 79.67 nm. The electrical resistance of an individual joint was evaluated to be 1.35 Ohms. With thick enough copper deposit, it can be considered that the joint has a tensile strength of more than 200 MPa. Moreover, this joining method has showed a considerable tolerance for the initial gap between the fiber and the substrate surface.

1. Introduction

Due to their excellent properties, carbon nanotubes (CNTs) have been very popular for building high-performance materials since their discovery in 1991. It has been considered that CNT-based materials are very competitive candidates to replace metal conductors like copper as next generation conductive materials [1,2]. Advances in production techniques of continuous fibers made purely of CNTs, have paved the way for new macro-scale applications which utilize the superior properties of individual CNTs. These wire-like macroscopic assemblies of axially aligned CNTs have showed obvious potential to be used in electrical wires and cables [3–6], flexible high-performance fiber supercapacitors and solar cells [7–10], and specialty composites [11–21]. CNT fibers can already provide significant advantages over metallic wiring for applications which require high performance to weight ratios, high flexure tolerance and corrosion resistance.

In spite of their functional and light-weight advantages, a necessary step for electrical and electronic applications of CNT fibers is to join them with other live parts of the circuits electrically and mechanically. In other words, the joining need provide a low-resistance and mechanically strong connection to other parts of the circuit for the CNT fibers. And the joining method needs to be simple, reliable and cost-efficient.

So far, much work has been done in the field of joining individual nanotubes and small assemblies of nanotubes. For example, superstrong low-resistant CNT-carbide-tungsten junctions were successfully fabricated by Wang et al. via combined electrical biasing of the CNT and focused electron-beam irradiation inside a transmission electron microscope [22,23]. However, most of the methods, such as ion beam irradiation [24,25], electron beam– or focused ion beam–based deposition [26,27], and focused ion beam lithography [28], are not applicable to macroscopic CNT fibers. Furthermore, these methods must be performed in a high-vacuum environment and the throughput is low.

On the other hand, it is well recognized that the CNT fibers differ from conventional metal-based wiring materials. Therefore, traditional methods of connecting wires, including soldering, conductive epoxy, mechanical crimping, and ultrasonic welding, are not always practical for the CNT fibers. In regard to soldering, not only the high temperature issue but also the formation of an interlayer of poorly conductive carbides between the metal and CNT-based

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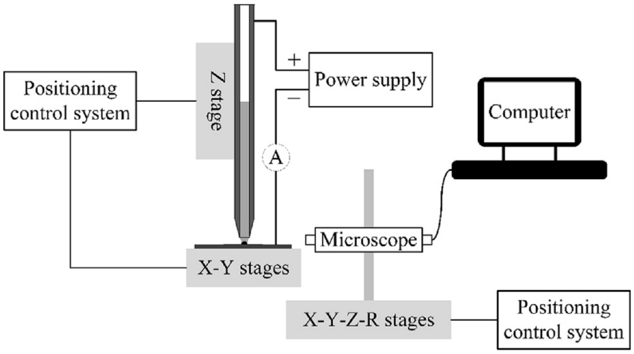

Fig. 1. Experimental set-up for the joining via meniscus-confined localized electrochemical deposition.

conductors is not acceptable. Additionally, conventional solders do not typically wet the CNTs, and therefore pre-metallization of the carbon surface is required in order to make a robust solder connection [3]. Using conductive silver paint is the most common laboratory method of connecting CNTs. However, the silver paint generally requires long curing times and could easily spread between contacts resulting in shunt paths in closely spaced electrodes. Furthermore, the binder and solvents in the paint can lead to higher contact resistances and contamination of the CNT networks. The connections made by mechanical crimping can be mechanically strong, but the contact resistance is dependent on the crimping force and the connections tend to be large and bulky [6]. Ultrasonic welding has been shown as a viable technique for bonding CNT sheet materials to copper foils, whereas the high frequency friction and the force applied during ultrasonic welding can damage the CNTs as well as the CNT fibers [29]. Schauerman et al. presented another detailed summary of the advantages and disadvantages of the four joining methods mentioned above for carbon-based materials [29]. Moreover, downscaling the above-mentioned traditional techniques to the order of a few micrometers has proven to be difficult. As a result, they are not able to make micro-joints for the CNT fibers as fine as a few microns in diameters, especially where precise joint sizes are needed, such as in the electronic devices.

Several studies have reported that the electrochemical deposition (ECD) technique could be applied to successfully produce CNT-

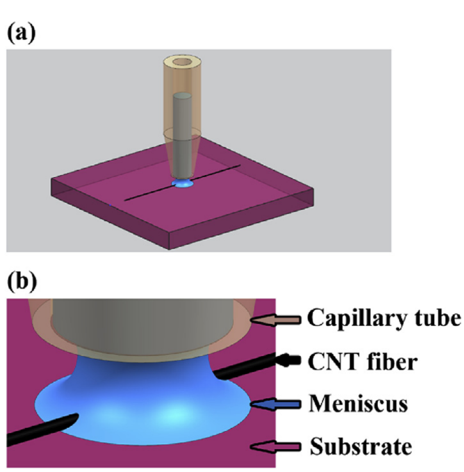

Fig. 2. (a) A schematic of the copper capillary tube-meniscus-CNT fiber-substrate system during LECD and (b) a close-up of the meniscus. (A colour version of this figure can be viewed online.)

metal composites with enhanced electrical properties [16–18,30]. Hannula et al. studied electrochemical deposition of copper on an aerogel-spun CNT fiber from a copper sulfate-sulfuric acid bath, and the resulting CNT-copper composite wire was found to have higher specific conductivity (as much as a nine-fold increase) than the pure CNT fiber. ECD can allow metals to deposit on the CNT surface and meanwhile have little influence on the original CNT network morphology, therefore a previously optimized CNT network can be applied as the substrate onto which metal is deposited [16], which is the advantage of ECD on CNT electrodes over other methods for fabricating CNT-metal composites. Nevertheless, the ECD processes reported were performed with the CNT fibers immersed in an electrolyte environment, and the deposition positions could not be controlled effectively.

At the same time, localized electrochemical deposition (LECD) has rapidly been a rising technology for fabricating three-dimensional microstructures since it was proposed in 1995 [31,32]. Given that the method of LECD is carried out via micro-anode guided electroplating techniques in the open air environment, high-end equipment and absolutely clean rooms are not required, which makes this method simple, economic and clean. Moreover, this method can fabricate intensive structures with high aspect ratio and complicated geometry, and also can deposit various materials, including metals, metal alloys, conducting polymers and even some semiconductor materials, which makes it compatible with electronic devices [33–37]. It has been demonstrated that LECD carried out in pulse current (PC) mode was better than that in direct current (DC) mode in order to obtain micron copper columns with uniform diameter, compact structure and a smooth surface [35]. In practice, the LECD is generally realized through the localization of the electric field near the end of a sharpened metal tip, and fabrication must be performed with both the workpiece and the tip immersed in an electrolyte environment. If the CNT fiber was immersed in the electrolyte during LECD, it would be filled with deposited metals as well as the electrolyte, similar to the result of the fabrication of CNT-metal composite via ECD.

Hu and Yu made an advance over common LECD technologies and developed a meniscus-confined ECD method that exploits the thermodynamic stability of a microscale or nanoscale liquid meniscus to “write” pure copper and platinum three-dimensional structures of designed shapes and sizes in an ambient air environment [38]. As an electrolyte-containing micropipette with a microscopic dispensing nozzle approaches a conductive substrate surface, a meniscus (i.e. liquid bridge) is established between the dispensing nozzle and the substrate surface. With the application of an appropriate electrical potential between the electrolyte contained in the micropipette and the substrate surface, electrochemical deposition is initiated within the substrate surface confined by the meniscus. This automated direct-writing wire-bonding technology enabled wire diameters of less than 1 μm and bond sizes of less than 10 μm^2. These wire bonds achieve a breakdown current density of more than 10^{11} A per square meter, six orders of magnitude higher than that for solder-based interconnects. Apart from high-density and high-quality interconnects, the technology can also be used to fabricate complex three-dimensional microscale and even nanoscale metallic structures.

In the present work, we report a novel method that utilizes the meniscus-confined LECD to precisely join CNT fibers with metal substrates electrically and mechanically. The ability to fabricate size-controlled joints and little impact on the original CNT network structure are the significant advantages of this method over the traditional methods of connecting wires. The morphology, microstructure and properties of the joints fabricated were evaluated