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Letter

Planar to cage mode switching in carbon nanostructure growth on bimetallic micro-scrolls

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ABSTRACT

The thermal stress generated by the bimetallic layer of the scroll induces the switching of the growth mode from being in a tightly stacking planar mode to a loosely stacking cage mode. Tightly stacked carbon layers grown at 300 °C are consisted of nanoscale graphite, but loosely stacked carbon strips, which are stacks of decoupled graphene layers, are grown at 350 °C. However, carbon structures grown on the scrolls at 375 °C are nano-onion.

Thin metallic layers on silicon substrates have been widely used in many areas including complementary metal oxide semiconductor technology, electromechanical devices and carbon nanostructure growth [1–3]. Often the metallic layers on silicon substrates are stable at low temperatures. While the stability of Ti/Ni bimetallic layers on silicon substrates with or without thick oxide layers has not been thoroughly explored at relatively low temperatures, it has been established that nanometer thick films on a substrate are often scrolled into nano and micro-scrolls due to the stress developed in the thin film [4–9].

If scrolls with a bimetallic structure are formed, they can be useful for many applications including the development of nano and micro-electromechanical devices and sensors because the scroll responds to any mechanical perturbation and can be electrically and thermally modulated. A novel function can be added to the scroll by coating the bimetallic surface with a functional material. In particular, carbon nanostructures are interesting to add to the scroll due to their distinct physical, electronic, optical, structural, chemical, thermal, and mechanical properties. Bimetallic scrolls with carbon nanostructures can be applied to the development of a new type of multifunctional devices [3,10]. These include three-dimensional cylindrical electronic, thermal, photonic, and electromechanical devices, chemical, biological and radiological sensors, charge storage, supercapacitors, solar cells and energy conversion devices, to name a few [2,11–14].

In this letter, we report simultaneous bimetallic scroll formation and carbon nanostructure growth on scrolls using very thin Ti/Ni bimetallic layers, which have a thickness of 120 nm. We were able to directly grow tightly stacked nano-carbon film (300 °C), loosely stacked nano-graphene strips (350 °C) and nano-onions (375 °C) by controlling the growth temperature as shown in Figs. 1–3. This occurred through the growth mode transition from the planar mode to the cage mode, which is driven by the stress acting on the catalyst. In particular, the loosely stacked carbon layers exhibit the same property as that of decoupled nanoscale graphene layers.

Bimetallic layers, composed of 20 nm thick Ni and 100 nm thick Ti films, were deposited on a boron-doped Si wafer with a 2 μm thick thermal silicon dioxide layer using an e-beam evaporator. The bimetallic layers were patterned by the lift-off method and were peeled off from the pattern edge as shown in Fig. 1a. These were loaded to a thermal chemical vapor deposition system with a 3″ quartz tube furnace for carbon nanostructure growth. The growth temperatures range from 300 °C to 375 °C. At the growth temperature, the mixture of 25 sccm of acetylene (C_2H_2) and 50 sccm of Ar was introduced, and the pressure of the quartz tube was maintained at 10 Torr for 20 min. The structure of the bimetal layers was studied by a Hitachi S-2400 scanning electron microscope (SEM), and the crystalline properties of carbon nanostructures were investigated by a JEOL-2010 high resolution transmission electron microscope (HRTEM). The temperature-dependent chemical composition of the Ni and Ti layers and the carbon nanostructures were investigated by a Renishaw inVia Raman spectrometer using excitation energy of 2.45 eV [15].

The deposited 120 nm thick Ti(100 nm)/Ni(20 nm) bimetallic layers were peeled and torn off from the substrate during the lift-off process as shown in Fig. 1a. The initial detachment can be attributed to

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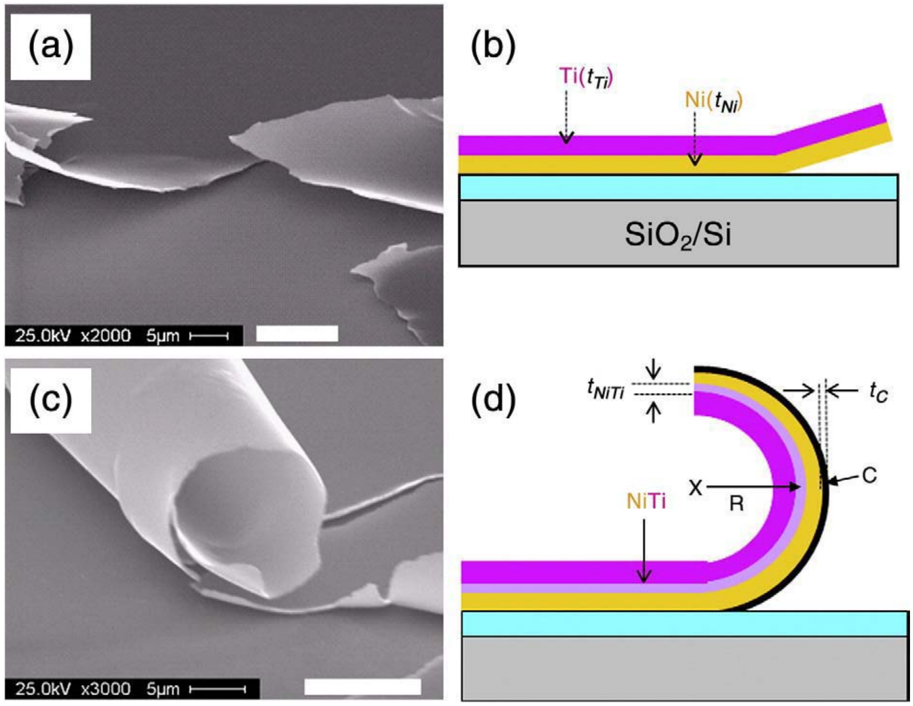

Fig. 1. a) An SEM image of a peeled and torn off Ti/Ni bimetal layer and b) a schematic showing the layer structure on a silicon substrate. c) An SEM image of a scroll formed in the carbon nanostructure growth atmosphere at 300 °C, and d) a schematic showing the structure of the scroll. The white scale bars in SEM images correspond to 10 μm.

the weak adhesion between the bimetallic layer and the thick silicon dioxide layer as well as the interfacial residual stress accumulated in the bimetallic layer during the film deposition. This triggers the peeling off of the bimetallic layer from the substrate, starting at the edge of the bilayer, and then the thin bimetallic layer is torn off during the peeling as shown in Fig. 1a and b.

The peeled off bimetallic layer structures (Fig. 1a) were annealed at 300 °C in the carbon nanostructure growth atmosphere. The

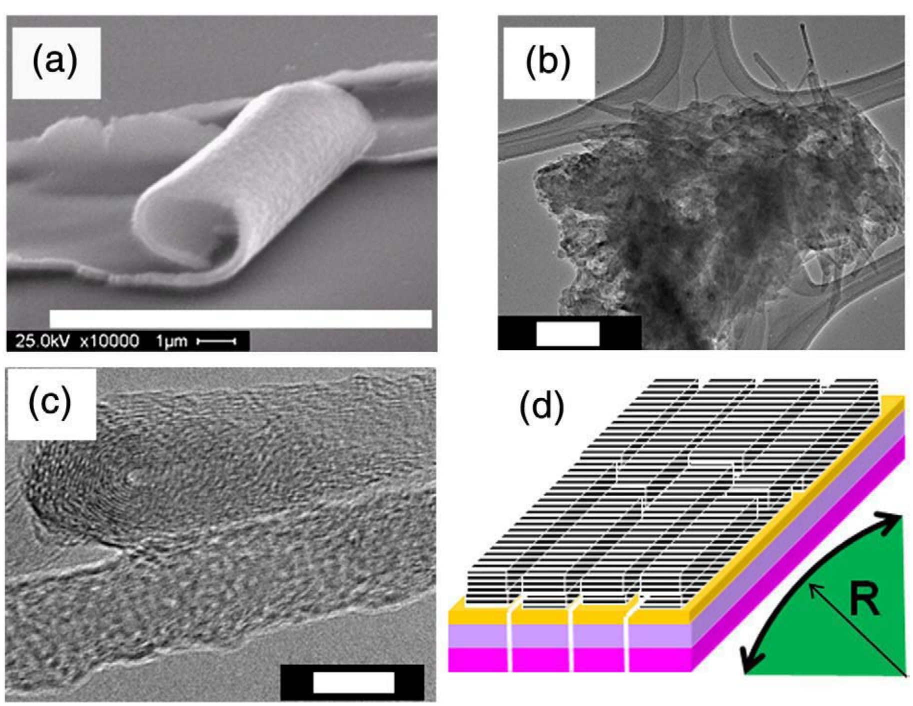

Fig. 2. a) SEM, and b) and c) HRTEM images of carbon nanostructures detached from a scroll formed at 350 °C. The white scale bar for the SEM image of a) corresponds to 10 μm, and those for the TEM image of b) correspond to 200 nm and of c) correspond to 10 nm, respectively. d) A schematic showing the growth of nano-graphene strips.