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Citation for published version:
Wang, T, Carlberg, B, Jonsson, M, Jeong, G-H, Campbell, EEB & Liu, J 2007, 'Low temperature transfer and formation of carbon nanotube arrays by imprinted conductive adhesive', *Applied Physics Letters*, vol. 91, no. 9, 093123, pp. -. https://doi.org/10.1063/1.2776849

Digital Object Identifier (DOI):
10.1063/1.2776849

Published In:
Applied Physics Letters

Low temperature transfer and formation of carbon nanotube arrays by imprinted conductive adhesive

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(Received 15 May 2007; accepted 7 August 2007; published online 29 August 2007)

This letter demonstrates the transfer and formation of aligned carbon nanotube (CNT) arrays at low temperature by imprinted conductive adhesive. A thermoplastic isotropic conductive adhesive is patterned by an imprint and heat transfer process. The CNTs grown by thermal chemical vapor deposition are then transferred to another substrate by the conductive adhesive, forming predefined patterns. The current-voltage response of the transferred CNT bundles verifies that good electrical connection has been established. This process can enable the integration of CNTs into various temperature-sensitive processeses and materials. © *2007 American Institute of Physics*.
[DOI: 10.1063/1.2776849]

Aligned carbon nanotube (CNT) structures have been proposed for various applications, such as field-emission electron sources,[1] electrochemical energy storage and production,[2] and flip-chip bumps.[3] The most widely used fabrication method of the aligned CNTs is thermal or plasma-enhanced chemical vapor deposition, in which the carrier substrate of the CNT structures typically needs to be heated up to approximately 700 °C or more. Such high temperature is not compatible with some temperature-sensitive processes and materials, preventing the utilization of CNTs in these cases. For instance, putting integrated circuits under this temperature may destroy the active devices inside the chips. A possible solution is to transfer carbon nanotubes onto the target substance at low temperature after the growth process. Contact transfer of aligned CNT films was reported by Huang *et al.* using a transmission electron microscopy grid in the transfer process to create patterns of CNT films.[4] An improvement was made by selectively transferring CNTs to an adhesive sticky tape (Scotch tape) covered with a predefined mask layer.[5] The utilization of the grid and sticky tape in these two processes limits their application. CNT transfer to conductive substrates using unpatterned or patterned metal alloys has also been demonstrated.[6,7] However, the transfer of CNTs with patterned metal based alloys still requires relatively high temperature (250–300 °C) and long time (2 h).[7] Silver paste can also be used to transfer CNTs but the annealing is carried out at even higher temperature (530 °C).[8] A lift-off technique to transfer aligned CNT films was recently reported but the adhesion between CNT film and the substrate is weak and no patterning can be implemented in the process.[9] Using conductive adhesives to transfer a whole CNT film at low temperature has also recently been reported, but no patterning technique was applied.[10] For many applications, it is important to produce patterned CNT bundles, e.g., when using CNTs as interconnects.[3] In this letter, we demonstrate a method to transfer and form aligned CNT arrays using isotropic conductive adhesive (ICA), which is previously patterned by an imprinting and heat transfer process. Figure 1 illustrates the process, containing two main phases: patterning of the ICA and transfer of CNTs.

The patterning of the conductive adhesive is based on an imprinting process. The molds used to imprint the conductive adhesive are made by anisotropic wet etching into silicon substrates in tetramethylammonium hydroxide solution. The patterns are defined by a standard photolithography process and buffered etching of a silicon dioxide layer. The patterns demonstrated in this letter are arrays of squares with 50 μm long sides and 100 μm pitch. Pyramidlike cavities are formed by the four {111} planes of silicon. The silicon dioxide mask layer is removed after the silicon etching. Before the imprinting process, the silicon mold is spin coated with a thin layer of release agent, which prevents the conductive adhesive from sticking to the mold. The imprinting is carried out with the help of a flip-chip bonder, in which the pressure force, temperature on both the mold and the substrate side, and heating time can be accurately controlled. The ICA used in the present experiments needs to be melted repeatedly in different steps. For this reason, a silver filled thermoplastic ICA was used in the present study. A drop of

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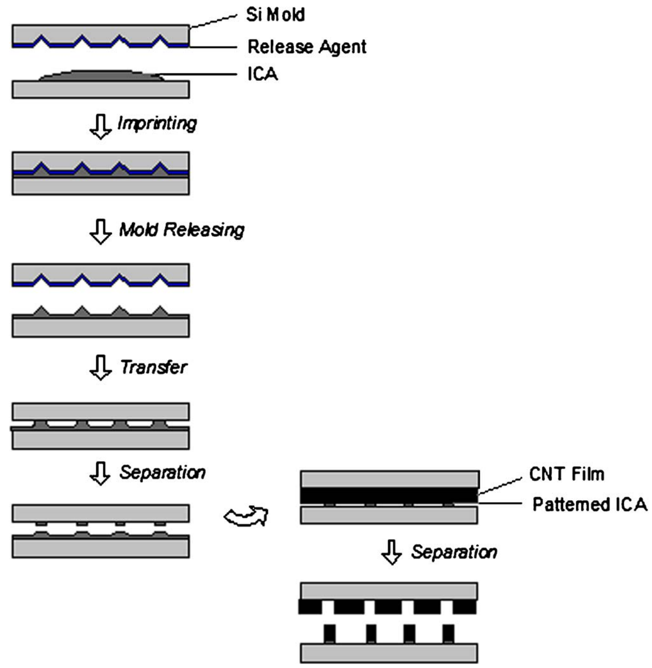

FIG. 1. (Color online) Schematic diagram of the experimental process. The ICA is patterned by an imprinting and heat transfer process and then utilized to transfer and form CNT bundles from a CNT film.

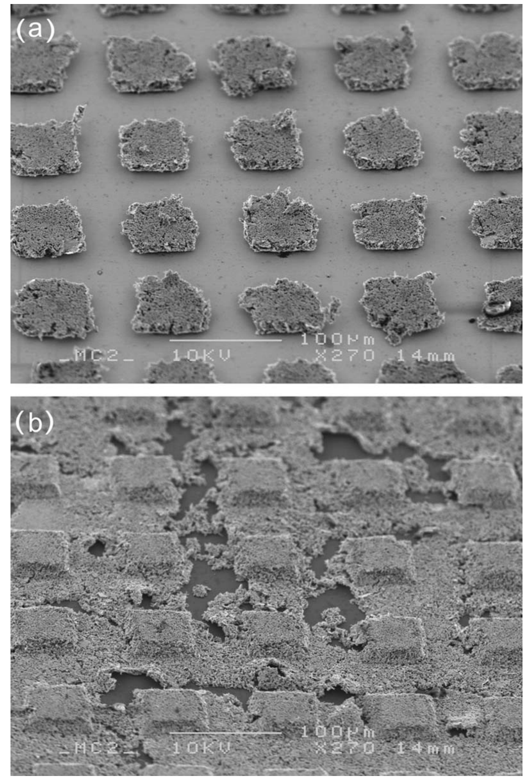

FIG. 2. (a) SEM image of discrete ICA patterns. These patterns are transferred from the top of the original imprinted pyramid-like ICA structures. (b) The imprinted ICA pyramids with flattened tops are left on the original substrate after the heat transfer process. The residual layer connecting the ICA blocks can also be seen.

the ICA was placed in the center of a piece of silicon and then imprinted by the mold. The imprinting of the ICA contains two key steps: making the adhesive flow under elevated temperature and high pressure, and curing of ICA at high temperature and moderate pressure. The flow phase is performed under a pressure force of 40 N at 60 °C for 90 s. The adhesive is then cured under 20 N at 150 °C for 90 s. After this, the mold is removed and the ICA is formed as uniform pyramidlike structures. Limited by the microstructure of the adhesive, it is impossible to get completely rid of the residual layer connecting the pyramidlike structures even if a very high pressure is applied for a long time. As an isotropic conductive adhesive is utilized in this work, it is necessary to produce discrete adhesive blocks without electrical connection via a residual layer. A heat transfer process is implemented to produce discrete ICA patterns without residual layer. A second silicon substrate is placed close to the original substrate. This target substrate is previously sputtered with a thin titanium/gold layer which assists the electrical test in the latter stage. The two substrates are kept parallel and the gap between the two substrates is fixed at 20 μm during the transfer process. Considering that the height of the ICA pyramids is roughly 35 μm ($25 \tan 54.7°$), the target substrate can touch and flatten the adhesive pyramids without reaching the residual layer. The temperature of the original substrate was set to 60 °C and that of the target substrate was raised to 150 °C for 90 s. This configuration allows the thermoplastic adhesive to reflow only at the surface of the target substrate. The two substrates are separated after a 90 s transfer process and an array of discrete thin ICA blocks is achieved on the target substrate. Figure 2 shows the scanning electron microscopy (SEM) images of patterned adhesive on the target substrate (a) and the flattened pyramidlike adhesive structures connected by the residual layer left on the original substrate (b).

The carbon nanotube films used here consist of well-aligned multiwalled CNTs prepared by thermal chemical vapor deposition. 10/1 nm thick Al_2O_3/Fe is deposited onto a silicon wafer as the catalyst layer by an electron beam evaporator. The sample is then inserted into a quartz tube furnace. Under a flow of 100 SCCM (SCCM denotes cubic centimeter per minute at STP) H_2 and 900 SCCM Ar at atmospheric pressure, the furnace is heated to 750 °C. When the furnace has reached the set temperature for 15 min, an additional flow of 5 SCCM of C_2H_2 is added as the carbon source and the gas composition of H_2 and Ar are changed to 500:500 SCCM. After 15 min growth, the sample is cooled down to room temperature in a flow of Ar before being removed from the quartz tube.

The silicon chips carrying CNT films are then used in the CNT transfer process. A chip with CNT film is pressed onto the chip carrying the ICA array under a pressure force of 20 N. The chips are heated up to 150 °C for 120 s, and then separated after cooling down. Because the CNTs are aligned parallel in the film and only attracted to each other by weak van der Waals forces, they are easily separated by applying external forces. Transfer of CNTs and formation of CNT patterns are achieved simultaneously. The nanotubes held by the adhesion provided by ICA are transferred to the target substrate and the remaining CNTs are left on the carrier substrate. A 2–3 μm thick adhesive piece proved to be sufficient to transfer CNT bundles higher than 20 μm. An example of transferred and patterned CNT bundles is shown in Fig. 3(a) and the cavities formed in the original CNT film are shown in Fig. 3(b).

To verify the establishment of electrical connection, the *I-V* curve of the transferred CNT-ICA structure was mea-

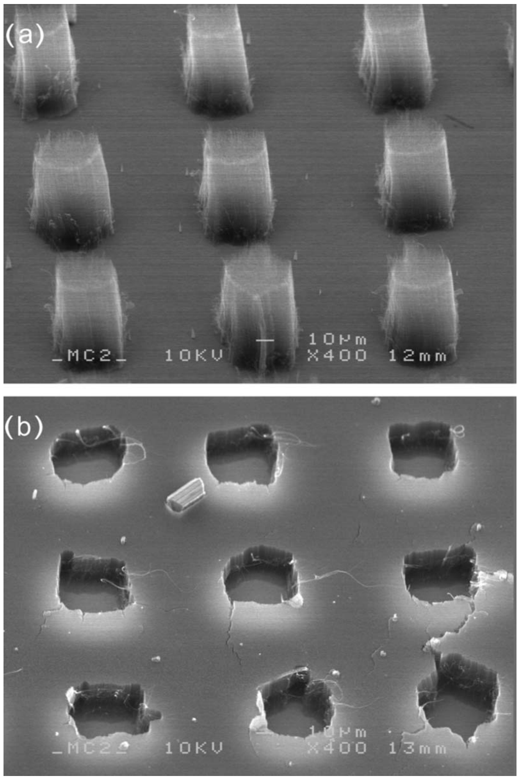

FIG. 3. (a) SEM image of an array of transferred CNT bundles with ICA underneath. (b) Cavities are left on the original CNT film.

sured with a multimeter connected to a probe station. One probe was placed on the gold conducting layer, close to the bundle under test. The other probe was placed on the top of the CNT bundle. The two probes were connected to a voltage source scanning from −0.5 to +0.5 V and the current was measured. No damage of the CNT bundles was observed under a microscope after removing the probe, indicating that the measurement results are valid. The *I*-*V* curve of one transferred CNT-ICA block is shown in Fig. 4. The current-voltage response is approximately linear in the measurement range and the resistance of this structure is around 100 Ω. The *I*-*V* measurement confirms that a good electrical connection has been made during the transfer process.

In conclusion, we have developed an efficient, scalable, and low-cost process to transfer and form CNT bundles at a low temperature using patterned isotropic conductive adhesive. The patterning of ICA is performed by an imprinting and heat transfer process, which can be applied for a variety of interconnect purposes. Arrays of fine pitch CNT bundles were produced.*I*-*V* measurements verify that good electrical contact between CNTs and ICA was established in the transfer process. The demonstrated transfer technique enables the integration of CNTs into various devices and processes that cannot withstand the high temperature of CNT growth.

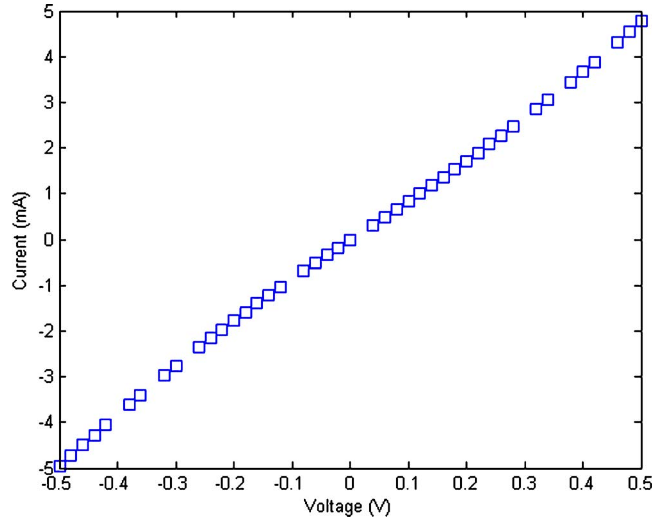

FIG. 4. (Color online) *I*-*V* curve of a transferred CNT-ICA block. The current-voltage response is approximately linear and resistance is around 100 Ω.

The authors thank Micro Joining KB for kindly providing the conductive adhesive used in this work. Three of the authors (T.W., B.C., and J.L.) are grateful to the financial support from EC-GEPRO project under the Contract No. of SSA 033349. One of the authors was (J.L.) also grateful to the support of Shanghai Science and Technology under the Pu Jiang Program (05PJ14045) and to the support of Intel under the higher education program with the Contract No. CNDA 4126007. Two of the authors (E.E.B.C. and M.J.) acknowledge financial support from the Swedish Strategic Research Foundation via the CARAMEL Materials Research Consortium.

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