Conductivity Enhancement in Carbon Nanocone Adhesive by Electric Field Induced Formation of Aligned Assemblies

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ABSTRACT: We show how an alternating electric field can be used to assemble carbon nanocones (CNCs) and align these assemblies into microscopic wires in a commercial two-component adhesive. The wires form continuous pathways that may electrically connect the alignment electrodes, which leads to directional conductivity ($\sim 10^{-3}$ S/m) on a macroscopic scale. This procedure leads to conductivity enhancement of at least 2−3 orders of magnitude in the case where the CNC fraction ($\sim$0.2 vol %) is 1 order of magnitude below the percolation threshold ($\sim$2 vol %). The alignment and conductivity are maintained on curing that joins the alignment electrodes permanently together. If the aligned CNC wires are damaged before curing, they can be realigned by an extended alignment period. This concept has implications in areas such as electronic packaging technology.

KEYWORDS: carbon nanostructures, orientation, alignment, electric field
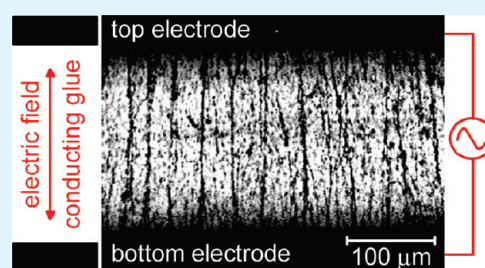
■ INTRODUCTION

Interconnections between flat electrodes in electronic packaging technology are conventionally formed by soldering. While providing superior electronic connection, soldering requires environmentally unfriendly heavy metals and can cause breakages due to mechanical stress. These factors become increasingly problematic with increasing environmental concern and decreasing device thickness. Therefore, soldering is being complemented by conductive adhesives,[1−4] for example, when connecting internal and external electrodes, busbars, and tabs in solar cells.[5,6] Isotropically conductive adhesives (ICAs)[7,8] contain conductive particles dispersed in a polymer matrix. In this, the particles form conductive pathways in all directions over macroscopic distances. These adhesives do not contain heavy metals and may provide stress-free connections. However, the required particle loading to form conductive pathways may be as high as tens of volume percentages. Large particle loading, while providing conductivity through unavoidable particle contacts, increases materials cost and may lead to loss of strength. Anisotropically conductive adhesives (ACAs)[9,10] contain in turn conductive particles that are large enough to reach from one electrode to another and whose fraction is so small that no lateral pathways are formed. The particles are distributed randomly, and the large particles may not hit small patches. Moreover, for the particles to just fit between the

electrodes and make a conductive bridge, nearly monodisperse particles are required. Thus ideally it would be advantageous to have conductive adhesives that would allow the small particle fraction for the particles smaller than the electrode−electrode distance.

Commercial conductive adhesives are usually based on silver loading[11] as this provides high conductivity. However, to provide inert all-organic materials, attention has also been placed on adhesives loaded by industrial-scale produced carbon particles such as carbon fibers[12] and carbon black.[13] There is also a scientific interest in adhesives with nonindustrial carbon particles, single-walled carbon nanotubes (SWCNTs),[14] multiwalled carbon nanotubes (MWCNTs),[15−22] and graphene oxide[23] in particular.

In the present study, we have used carbon nanocones and disks (hereafter CNCs)[24−28] as conductive filler particles. CNCs are manifested by a stacking of conically folded graphene sheets (Figure 1). The conical shape stems from 1 to 5 carbon pentagons incorporated into the graphene structure near the cone apex, giving rise to five discrete apex angles 112.9°, 83.6°, 60.0°, 38.9°, and 19.2°.[26,29] Nanodisks can be regarded as cones with an apex angle of 180°. CNCs can be synthesized in semi-industrial scale.[30] From an atomic level perspective,[25] the multilayered structures of
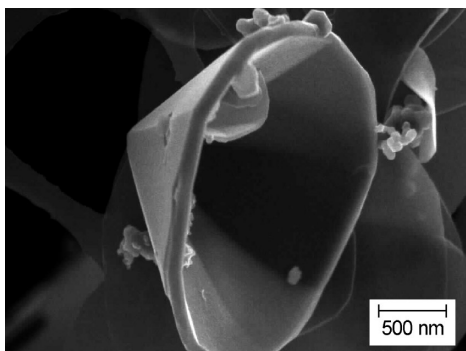
Figure 1. Scanning electron micrograph of CNC material used in the present study. A hollow bottom structure of the cone.

CNCs may lead to the electrical properties resembling MWCNTs. However, the properties of CNCs do not reach those of MWCNTs, which can be attributed to their lower molecular anisotropy. Therefore, from both a fundamental and applications point of view, it is advantageous to develop production procedures that would increase the order and anisotropy in CNC materials. This might improve the electronic properties of their macroscopic samples up to the phenomenologial level of MWCNTs.

Various authors have shown how alignment of MWCNTs,[31−33] carbon fibers[12,34] and carbon black,[35,36] and related systems such as conjugated polymers[37] influences macroscopic conductivity and charge carrier mobility. It has been demonstrated how SWCNTs[38] or CNCs[39,40] can be aligned in fluid-like dispersion. In this method, the carbon material is dispersed in a liquid under an ac electric field that leads to the formation and alignment of micrometer and submicrometer size channels. Osuji and co-workers aligned SWCNTs in a liquid polymer precursor by magnetic field[41] and Park and co-workers[42] by ac electric field and obtained aligned SWCNT channels in a polymer composite by subsequent curing. Sancaktar and Dilsiz[43] aligned nickel particles in an epoxy matrix by magnetic field, forming thus a conductive adhesive with anisotropic conductivity, its strength increasing with the increasing magnetic field. Following the guidelines of these reports, we begun to develop these methods further and found evidence for alignment of CNCs in polymer composites.[44]

In this paper, we elaborate on the electric field assembly and alignment process with conducting adhesive applications in mind. We demonstrate enhanced conductivity in the CNC-adhesive, when the CNC filling is particularly low. The CNCs are dispersed in a commercial adhesive, with the particle loading being significantly under the percolation threshold of an isotropic mixture. Before treatment, this material is an isotropic CNC dispersion of isolated particles without particle−particle contacts or significant macroscopic conductivity. The CNC particles are assembled and aligned by electric field into chainlike wires, which act as conductive pathways. This leads to both directional conductivity and significant conductivity enhancement, which are maintained in the subsequent curing, which connects the electrodes together. Moreover, we show that this alignment can be restored by continued alignment if the sample suffers from a mechanical breakage. Overall, this example shows how increased order in CNC composite with a low particle fraction leads to significant enhancement in its electronic performance. This concept may have useful applications in areas like electronic packaging technology.

■ EXPERIMENTAL SECTION

Materials. The CNC material (Figure 1) was provided by n-Tec AS (Norway). The material was synthesized using the so-called Kvaerner Carbon Black and Hydrogen Process.[30] The material contained 70% carbon disks, 20% carbon cones, and 10% soot and was annealed at 2700 °C in argon prior to use. This improves the crystallinity of material and removes a disordered layer from the surface of the particles.[27,28] The quality of the material after annealing was assessed using a Hitachi S-4800 scanning electron microscope (SEM). The two component adhesive consisted of Araldite AY 105-1 (Huntsman Advanced Materials GmbH) epoxy resin and Ren HY 5160 (Vantico AG) amine hardener. The epoxy resin was based on bisphenol A. The hardener contained 50−60% polyoxypropylenediamine, 14−20% benzylalcohol, 10−18% isophorone diamine, and 1−5% trimethyl-hexamethylenediamine. Both components were of technical quality and supplied by Lindberg & Lund AS (Norway). The mixing ratio of the epoxy and the hardener was 2:1 parts per weight. The CNCs were mixed with the adhesive by stirring them at 60 rpm for 30 min at room temperature. After this period, a completely uniform CNC−adhesive dispersion was formed with a low CNC fraction (≤2 vol %). It turned out that it was not possible to prepare a uniform mixture for too high CNC fraction, above 2 vol %, as a certain fraction of the particles remained segregated in this case.

Alignment. Two separate setups were used in this study: (i) Optical microscopy inspection, and (ii) dc and ac conductivity measurements. In the optical microscopy study, the CNC−adhesive mixture was injected between two copper electrodes with a typical spacing of 0.25−2 mm. The lateral size of this layer could be varied from 1 mm to several centimeters. The alignment in connection with the dc and ac conductivity measurements was performed with a measurement cell equipped with two flat stainless steel out-of-plane electrodes located parallel and on top of each other with a rubber O-ring or a polyester film as a spacer (the out-of-plane configuration). The area of electrodes was typically 10 cm^2, and spacing was 0.25−0.75 mm. The ac electric field was generated by a custom-made ac source[39,40] or by a system that consisted of a handheld CIE 555 RC oscillator connected to a Denon PMA-725R stereo amplifier. In the latter case, the sinusoidal input signal was monitored by an oscilloscope. In a typical procedure, a 1 kHz ac field of 0.5−1 kV/cm (rms value) was applied for 1−60 min at room temperature. The curing was performed immediately afterwards, typically at room temperature for 6 h or at 100 °C for 6 min.

Detection of Alignment and Conductivity Enhancement. The conductivities of the nonaligned mixtures at a high CNC fraction of ≥2 vol % were measured using a Fluke 179 multimeter connected to thin 3 mm long gold electrodes on glass (ABTECH Scientific, Inc.). The spacing of the electrodes was 10 μm. The particle alignment was seen in the optical microscope (Nikon Optiphot) and detected by the abrupt increase of conductivity. The conductivities as a function of alignment time were measured in the custom-made steel cell described above. The dc conductivity was measured directly using the Keithley 2000 multimeter, and the alignment was paused for each measurement. The impedance analysis was carried out with a Solartron SI 1260 with a Solartron 1294 interface in a two-terminal configuration. The frequency range for the sweep was 1 Hz to 1 MHz, with an applied measuring voltage of 100 mV rms. These sweeps were done after applying the alternating electric
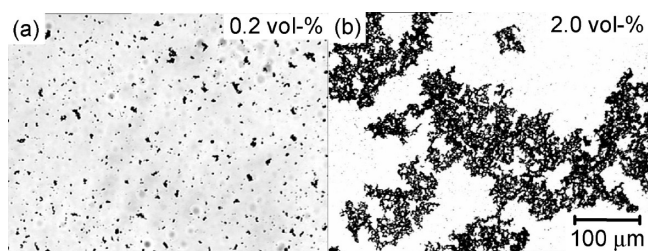
Figure 2. Optical micrographs of CNC−adhesive. CNC loadings were 0.2 vol % (a) and 2.0 vol % (b).
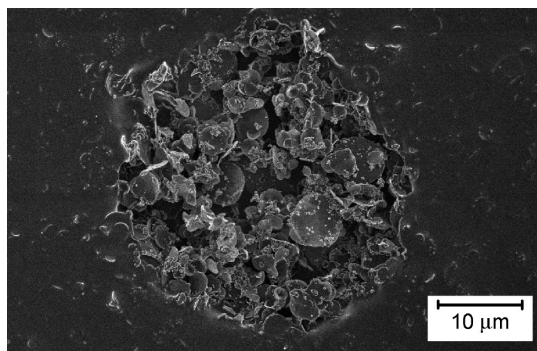

Figure 3. SEM image of CNC−agglomerate when the CNC loading was 2.1 vol %.

field for alignment in 30 s or 1 min intervals, and the alignment was paused for each measurement. The dc conductivity was estimated by extrapolating the ac conductivity to the zero-frequency limit.

■ RESULTS AND DISCUSSION

Figure 2 shows optical micrographs of the CNC material mixed with the adhesive. When the CNC fraction is below about 2 vol %, the mixture appears as a uniform dispersion of isolated particles (Figure 2a). Typical particle size is about 10 μm or less, and the visual particles contained one or a few CNCs. All particles appear to be wetted by the adhesive. This dispersion is easily formed by simple stirring in 15−30 min. No detergents, sonication, functionalization, or further purification of the raw material are needed. This is an apparent benefit compared to CNTs that may require a pretreament until a good quality polymer dispersion can be formed.[45] When the CNC fraction is increased over ∼2 vol %, the CNC particles begin to aggregate, forming continuous networks in similarly prepared mixtures (Figure 2b). When the CNC networks are formed, the mixture seems still uniform beyond the length scale of 100 μm. When the networks are formed, at least a part of the aggregates are not wetted by the adhesive. A SEM image of such particle agglomerate is shown in Figure 3.

Figure 4 shows the dependence of dc conductivity σ of a nominally isotropic mixture on volume fraction p of CNCs. The curve consists of three parts. At low CNC fraction ($p \leq 0.02$), the conductivity is on the order of 10^{-6} S/m. The conductivity of pure epoxy is not attainable using our instrument, but the hardener is a moderate ionic conductor ($\sim 10^{-6}$ S/m). Therefore, the observed conductivity at low p is attributed to the ionic conductivity of the hardener. Next, the curve shows a sharp increase approaching a plateau with increasing CNC fraction, which may
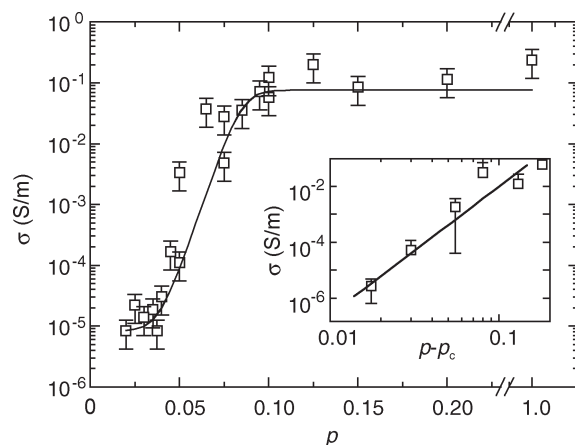

Figure 4. The dc conductivity of the CNC−adhesive as a function of CNC loading p. Solid line (sigmoidal fit) is a guide to the eye. Inset shows the linear relationships between conductivity and $p - p_c$, with the estimated percolation threshold $p_c = 2$ vol %.

be attributed to the electronic conductivity of the emerging CNC network. If the particle loading is high (>7 vol %), the conductivity approaches that of the pure CNC powder ($\sim 10^{-1}$ S/m). The behavior shown in Figure 4 can be understood in terms of a percolation model.[8] In this simple model, the percolation threshold of the isotropic mixture, p_c, and the conductivity as a function of the volume fraction is approximated as $\sigma \approx \sigma_0 (p - p_c)^t$, where σ_0 and t are constants. The percolation threshold of the isotropic CNC−adhesive is estimated to be $p_c \sim 2$ vol %. The scaling parameter $t \sim 2$ is related to the dimensionality of the percolation, whereas the fitting parameter $\sigma_0 \sim 10$ S/m represents the conductivity of the percolative network, and this is not the conductivity of individual particles. As the CNC powder is granular in nature, the conductivity of CNC powder ($\sim 10^{-1}$ S/m) remains lower than σ_0.

The data shown in Figure 4 are similar to those reported elsewhere for CNCs mixed with polyvinylalcohol (PVA),[46] which shows the threshold at ∼2.1 vol % and a conductivity plateau at ∼10^{-1} S/m. In contrast, this threshold is higher than typical thresholds of isotropic MWCNTs mixed with adhesives such as epoxies (0.05 vol %[15−18] or even 0.0025 wt %[22]) or polyurethanes (∼1 vol %[19]) or with technical polymers such as PVA (∼0.4 vol %)[46] or polyimide (∼0.05 vol %).[47] SWCNTs exhibit similar values (0.1−0.2 wt %), for instance, when mixed with poly(methyl metachrylate) (PMMA).[48] This difference stems from the lower particle aspect ratio as shown for example by Elliott and co-workers.[49] The threshold for CNCs is also higher than that of many graphene composites (∼0.6 vol %),[50] where graphene is mixed with polymers such as poly(vinyl chloride).

Even though the percolation threshold of isotropic mixtures decreases with an increasing aspect ratio, this does not imply that in the aligned systems the particle fraction needed to get the electrical conductivity would decrease with an increasing aspect ratio. Quite the contrary, long fibers or tubes may become isolated once aligned, which leads to a decrease in conductivity unless the particle fraction is increased.[32,33] If the aspect ratio of tubes is fixed, the conductivity does increase with modest alignment but decreases with further alignment.[51] If the system contains a mixture of tubes of different aspect ratios, the percolation threshold of isotropic mixtures decreases with its aspect ratio distribution moving toward a lower value.[49] However, in the aligned systems,
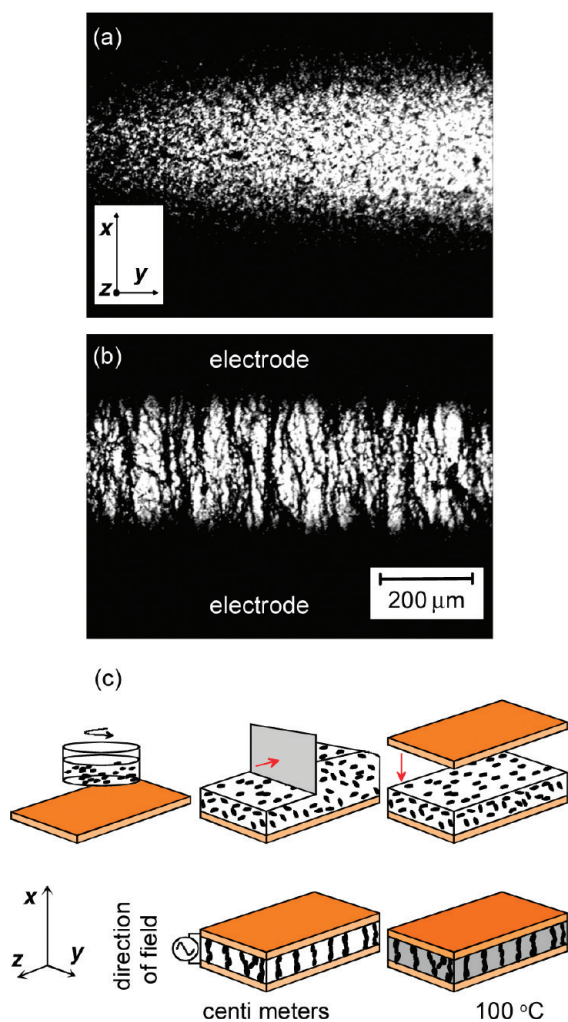
Figure 5. The concept for preparing a conductive adhesive with aligned CNC channels. Optical micrographs of CNCs dispersed in a two-component adhesive: (a) Essentially nonconductive isotropic dispersion of 0.2 vol % CNC loading. (b) Material after assembly, alignment and curing. (c) Schematics illustrating experimental procedure and out-of-plane alignment geometry.

introducing tubes with a lower aspect ratio actually increases the conductivity.

We turn next to the conductivity investigations. The motivation for our work was to increase the order and anisotropy of CNC materials for potentially achieving electrical conductivity comparable to the isotropic MWCNT composites with similar particle fractions. Therefore, we studied a CNC—adhesive with a CNC loading of 0.2 vol %, which is of the same order of magnitude as the typical percolation threshold of an isotropic MWCNT blend. This is an order of magnitude lower than the estimated p_c for CNCs. This mixture has essentially no particle—particle contacts as shown in Figure 2a, and its dc conductivity is very low ($\sim 10^{-6}$ S/m) as shown in Figure 4.

Figure 5 illustrates our concept for electric field assembled and aligned CNC wires to form conductive adhesives from nonconductive isotropic dispersion. In this procedure, the CNC material shown in Figure 1 is dispersed into the liquid adhesive to form an isotropic mixture without particle—particle contacts (Figure 5a). When a 1 kHz alternating electric field of 1 kV/cm is switched on, the CNC particles gather into assemblies that align into wires
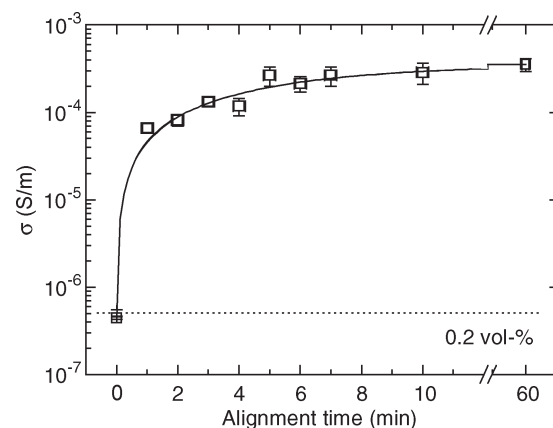

Figure 6. The dc conductivity of the field aligned 0.2 vol % CNC—adhesive as a function of alignment time (open squares). Dotted horizontal line shows the dc conductivity of pure liquid adhesive for comparison. Solid line (sigmoidal fit) is a guide to the eye.

forming continuous pathways over the electrode spacing of ~ 0.4 mm in 10 min. The somewhat asymmetric particles are not only aligned with the field but also with respect to each other. The procedure is analogous to that described for SWCNTs by Park et al.[42] It differs, for example, from that of Tsuda and Sakka[52], who aligned MWCNTs in composites by magnetic field through rotation without forming continuous pathways through the macroscopic sample. This step was followed by curing at 100 °C for 6 min, which stabilizes the formed wires in the solid composite (Figure 5b). The scheme in Figure 5c shows the applied alignment geometry, where the width of the electrodes is at least an order of magnitude larger than the electrode spacing, which is in turn at least an order of magnitude larger than the particle size.

Figure 6 shows the dc conductivity parallel to the alignment direction of a CNC—adhesive as a function of alignment time. The CNC loading was 0.2 vol %. The alignment was performed using the out-of-plane geometry with an electrode distance of 0.75 mm and a 1 kHz field of rms amplitude 1 kV/cm, which corresponds to the situation shown in Figure 5. The initial conductivity $\sim 5 \times 10^{-7}$ S/m corresponds to the ionic conductivity of the liquid hardener. When the alignment is started, the conductivity increases sharply and saturates in a few minutes approaching the level $\sim 10^{-3}$ S/m, which is comparable to the results of Park et al.[42] No further increase in conductivity is seen, if the field is kept on for 1 h. From the optical observations, we know that at room temperature a clear formation of fibrous CNC wires is visible within 1—3 min time, depending on the electrode spacing. Complete electrode-to-electrode connections are formed in about 10 min. Therefore, it seems fully plausible that the emerging CNC wires act as conductive channels and are responsible for the enhanced conductivity observed during the alignment process.

The material remains aligned after curing, and a conductivity level of $\sim 10^{-3}$ S/m parallel to the alignment direction is maintained. Our experimental setup does not allow in situ measurement perpendicular to the alignment direction, but this conductivity is very low after curing, corresponding to that of hardened epoxy and may not be reliably measured using our setup ($\leq 10^{-9}$ S/m). While the obtained conductivity level along the alignment direction greatly exceeds that of isotropic dispersion, it is below the level expected for pure CNC material. The conductivity of the aligned material normalized to its CNC
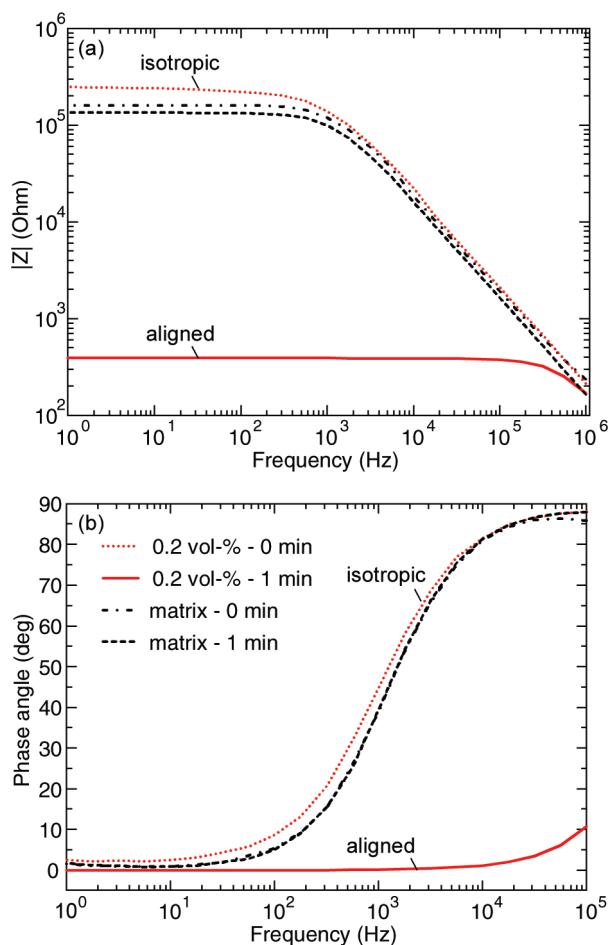
Figure 7. (a) Absolute value of the complex impedance, Z, and (b) phase angle of the studied adhesive with 0.2 vol % CNC loading before (dotted red line) and after (solid red line) 1 min of alignment. Data for pure adhesive before (dashed and dotted black line) and after (dashed black line) 1 min of identical treatments are shown for comparison.

Figure 8. The dc conductivity estimated from the ac conductivities by zero-frequency extrapolation for the aligned CNC−adhesive as a function of alignment time (open squares). Data of identically treated pure matrix are shown for comparison (open circles). Solid lines (sigmoidal fits) are guides to the eye. Data correspond to those shown in Figure 7.

fraction corresponds to that expected for pure CNC powder. The data clearly indicate that the formation of aligned CNC wires leads to a significant conductivity enhancement as compared to the isotropic dispersion. This enhancement occurs well below the percolation limit of the isotropic mixture.

The dc conductivity measurements were complemented by ac impedance spectroscopy, widely used for carbon black−polymer composites.[53] Figure 7a shows an example of such impedance measurements of a 0.2 vol % CNC−adhesive before alignment and after 1 min of alignment. The alignment was performed using an alternating electric field (1 kHz, 1 kV/cm) in the out-of-plane geometry with an electrode distance of 0.25 mm and an electrode area of 10.8 cm^2. Figure 7b shows the corresponding phase angle. The CNC loading and the alignment conditions are similar to those shown in Figures 5 and 6. The data of a similarly treated adhesive mixture without CNCs are shown for comparison. The impedance of the CNC adhesive before alignment is constant for low frequencies dropping for frequencies above 1 kHz, indicating capacitive conductivity of an ionic conductor. When the electric field of 1 kV/cm is turned on, the impedance level drops almost 3 orders of magnitude in 1 min. No further change is seen, if the alignment is continued for 10 min (data not shown here). The impedance of the aligned sample also stays constant up to 100
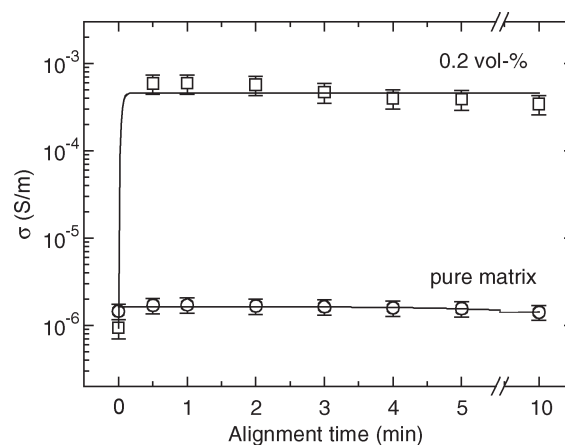
kHz. The data of the adhesive without CNCs are similar to the data of the CNC−adhesive before alignment, and no changes are observed if the electric field is turned on for 1 or 10 min. This indicates that the alignment has a major effect not only on dc conductivity but also on ac conductivity. In our interpretation, the conductivity before alignment is due to the hardener solvents, whereas the conductivity after alignment stems from the CNC wires. If this is correct, the shift in capacitance with frequency should be understood in terms of different conductivity mechanisms in organic solvent and graphitic CNCs.

As the impedance phase angle is nearly zero for low frequencies (Figure 7b), this allowed us to estimate the nominal dc conductivities by extrapolating ac conductivities to zero-frequency. Figure 8 shows so obtained conductivities of CNC−adhesives as a function of alignment time. The initial conductivity of CNC−adhesive is near to that of a pure adhesive mixture whose conductivity remains unchanged by the electric field (lower data). The extrapolated dc conductivity increases sharply in 30 s time and saturates in a few minutes (upper data). This effect is concomitant with the alignment (Figure 5) and with the increase in the dc conductivity (Figure 6). The final level of conductivity does not depend on the experimental technique.

Figure 9 shows the dc conductivity before and after 10 min of treatment (1 kHz, 500 V/cm) for the CNC−adhesive as a function of CNC fraction. When the particle fraction is 0.2 vol %, the situation corresponds to that shown in Figure 8. The electrode distance was 0.25 mm, and the electrode area was 10.8 cm^2. The conductivity of the isotropic mixture is close to that of the pure adhesive ($\sim 10^{-6}$ S/m). A distinctive conductivity jump of nearly 3 orders of magnitude up to $\sim 10^{-3}$ S/m is observed on alignment. This situation is altered with increasing particle fraction. The conductivity level increases sharply with increasing CNC fraction, which stems from the particle aggregation and emerging conductive pathways (Figure 2). If the particle fraction is increased from 0.2 to 2 vol %, the nonaligned mixture shows a 100-fold increase in conductivity, and the final conductivity level ($\sim 10^{-5}$ S/m) corresponds to the value shown in Figure 4. In contrast, the conductivity level after alignment increases slightly less with increasing CNC fraction, and a
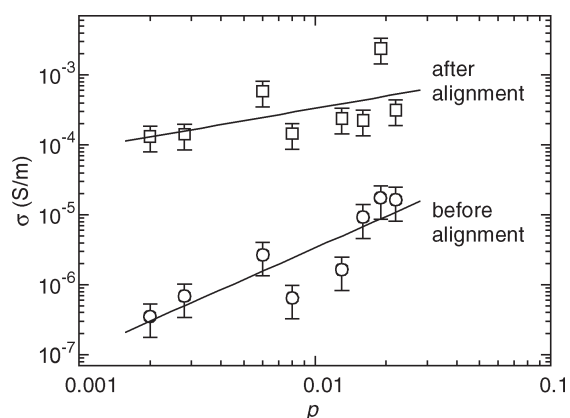
Figure 9. The dc conductivity of the CNC−adhesive as a function of CNC loading before and after assembly and alignment using an alternating electric field (500 V/cm, 1 kHz) for 10 min. The dc conductivity was estimated from the ac conductivities by zero-frequency extrapolation. Solid lines are guides to the eye.
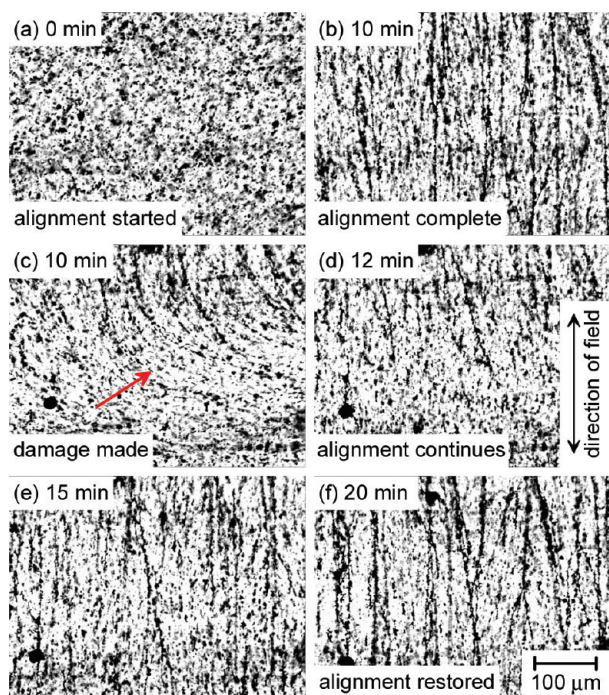

Figure 10. Optical snapshot micrographs illustrating the concept of an electric field "healing" of macroscopic damage in the aligned CNC−adhesive. Complete assembly and alignment of CNC wires has been obtained from isotropic dispersion (a) using 1 mm electrode spacing (at top and bottom outside field of view) in 10 min (b). The wires were mechanically cut by a perpendicular incision as shown by a red arrow in panel (c). When the field is maintained after this step, the alignment reappears (d−f).

10-fold increase in the CNC fraction corresponds roughly to a 10-fold increase in conductivity of the aligned material. This indicates that the amount of conductive pathways in the aligned material is more or less directly proportional to the CNC fraction. Thus, the alignment shows the largest relative gain at a low CNC fraction level (0.2 vol %) with the matrix dominating the initial conductivity.

This method turns out to be extremely robust, and the alignment of CNCs can readily be achieved with moderate fields (0.5−1 kV/cm). Furthermore, the electric field may not only be applied to align isotropic CNC−adhesive but also to "heal" scratches and defects, if they exist before curing. This is illustrated in the next example, which uses a 0.2 vol % loading and an alignment field of 1 kHz and 500 V/cm. Figure 10 shows optical micrographs of an alignment process, whereby a defect has been introduced and subsequently healed by the continued alignment process. In this case, the electrode distance is 1 mm, and the alignment of the initially isotropic CNC−adhesive (Figure 10a) is completed in 10 min (Figure 10b). The scratch is made by a scalpel in the aligned material (Figure 10c). This defect is repaired, and the full alignment is restored with an additional 10 min of treatment (Figures 10d−f). The present concept can be contrasted to typical polymer alignment atop rubbed substrates,[37] where a new alignment of an aligned and defect material cannot be continued. The ability to cure these kinds of defects is of importance in production lines where the samples are moved continuously.

The alignment of CNC dispersions has several practical implications. The material costs can be decreased compared to nonaligned samples with similar performance. High CNC fraction can potentially impair mechanical properties and reduce transparency of the CNC−adhesive. The increased order and anisotropy may render the anisotropic conductivity of CNC composite approaching those of isotropic MWCNT composite. The conductivity values are moderate, but we have not optimized the CNC composite in terms of particle purity, degree of alignment, or polymer environment. We expect that CNC wires may be used for thermal management as shown for SWNTs in PMMA[14] and will attempt to disperse CNCs in conjugated polymers as shown previously for nano and microscale graphite.[54]

CONCLUSIONS

We have demonstrated that an alternating electric field can be used to assemble and align CNCs into microscopic wires in commercial two-component adhesive. In this procedure, the CNC particles are aligned both with the field and with the respect to each other, forming continuous wires that may connect the plane−parallel electrodes, resulting in anisotropic directional conductivity ($\sim 10^{-3}$ S/m) between the electrodes. This out-of-plane geometry corresponds, for example, to a solar cell tab and its busbar. The alignment of CNCs leads to a pronounced conductivity enhancement of 2−3 orders of magnitude in the case where CNC loading ($\sim$0.2 vol %) is significantly below the estimated percolation threshold ($\sim$2 vol %). If the wires that are prepared in this way are damaged before curing, they can be healed by an extended alignment period. The particle loading used in the alignment corresponds to the typical percolation threshold in MWCNT composites with similar conductivity levels. Thus, this study demonstrates how the properties of aligned CNC−adhesive can approach those of isotropic MWCNT−adhesive. Though phenomenological, we propose that this concept may have implications in areas such as electronic packaging technology, for example, to form possibly stress-free electric interconnections without environmentally unfriendly heavy metals used in soldering methods. We expect, moreover, that a higher conductivity level may be obtained by optimizing raw materials and interfacial properties between CNC particles and between carbon wires and the electrodes.

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dx.doi.org/10.1021/am100990c |ACS Appl. Mater. Interfaces 2011, 3, 378–384

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■ ACKNOWLEDGMENT

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