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ARTICLE

Highly stretchable conductive MWCNT-PDMS composite with self-enhanced conductivity

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Soft and highly stretchable, yet conductive elastomers are essential in development of next generation electronic devices. However, electrical conductivity and mechanical stretchability are normally mutually exclusive properties since good conductors are rigid, and stretchable materials are inherently soft. Furthermore, maintained conductivity at high elongation is difficult to achieve because of the particle break-up due to the imposed large mechanical stresses on the percolated conductive fillers in contrast to much lower stresses on the soft and compliant elastomer matrix. Herein, a novel, simple, and effective preparation method is presented for fabricating highly stretchable conductive elastomers, consisting of multi-walled carbon nanotubes (MWCNTs) as conductor and poly(dimethylsiloxane) (PDMS) as soft matrix. As a result of the preparation method, stretch-enhanced conductivity is observed due to the prepared “hairy” particles effectively constructing further conductive pathways upon elongation. The “hairy” structures result from entrapped MWCNTs uniformly dispersed into low-molecular weight silicone polymers that are lightly crosslinked before being introduced into a silicone network resulting from silicone polymers of longer length to create a so-called heterogeneous bimodal network. Through the advanced, yet simple, mixing procedure followed by curing of the silicone matrix, it is possible to prepare conductive MWCNT/PDMS elastomers that can be elongated up to 200%. The prepared system shows high initial conductivity of 0.038 S m^{-1} with 2.6 phr (parts per hundred rubber) MWCNTs and constant conductivity throughout 160% deformation range. The prepared MWCNTs/PDMS composite composes a new class of stretchable conductive materials with potential applications in next generation electronics.

Introduction

In recent years, stretchable conductive materials have attracted considerable attention because of their broad range of potential applications in e.g. wearable electronics, biomedical devices and flexible displays.^{1–9} One key technical challenge in realization of such soft devices is that the material is required to provide conductivity upon large deformations.^{10,11} Traditional conductive materials, such as noble metals, are stiff and brittle materials, intrinsically preventing them from undergoing large deformations and dynamic transformation into complex shapes.^{12,13} Conversely, stretchable conductive materials should be able to be stretched, bent, and twisted repeatedly with no loss of conductivity or mechanical integrity.¹³ Ideally, this capacity of maintaining electrical performance under large mechanical deformation (strains significantly above 50%) makes stretchable conductive materials well suited for flexible electronics.

As one of the most widely used polymer matrixes for stretchable conductive materials, crosslinked PDMS has gained

considerable traction because of its biocompatibility, high stability and superior elasticity.^{14–17} Although PDMS possesses a great ensemble of properties, the use of PDMS in electronic devices – besides as electrically insulating materials such as dielectric elastomers¹⁸ – has been limited, even though acceptable conductivities can be reached with particle filled composites, these generally exhibit a critical loss in conductivity upon stretching.^{19–21}

As promising conductive fillers, carbon particles such as carbon black, graphite and carbon nanotubes (CNTs) have been widely utilized in fabrication of stretchable conductive materials.^{22–25} One approach for preparation of stretchable conductive elastomers is to deposit the conductive filler in its pure form on a pre-stretched or freestanding PDMS film, where the deposition techniques can be e.g. shadow masking, stamping or printing, i.e. creating basically a structural composite.^{26–28} However, this approach, utilizing a direct distribution of conductive nanoparticles on a substrate, often results in poor long-term reliability due to the initial pre-stretch required to create the flake-like structures in addition to poor compatibility between conductive particles and the polymer matrix (limited to van der Waals forces). For coated flexible electrodes, a recent publication by Bele et al.²⁹ has shown that embedment into a silicone matrix followed by thin film spray coating can be applied as a method to establish flexible thin electrode material on a PDMS backing.

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For free standing conductive elastomers, the most popular pathway is to directly disperse conductive particles by sonication, magnetic stirring or high-shear mixing in a polymer matrix and subsequently entrapping the percolated particle network by chemical crosslinking.^{15,30-32} However, for these systems introduction of a percolated conductive particle network into the polymer matrix generally reduces the ultimate tensile strain, particularly for high aspect ratio particles, and cause a reduction in conductivity with strain.^{33,34}

Although extensive efforts have been devoted to prepare stretchable conductive materials, an industrially relevant protocol that can be up-scaled still needs to be demonstrated.³⁵ The complexity and preparation time required for the currently applied processes make them relevant for lab scale experiments, but substantially simpler processes are required before industrial applications of free standing stretchable conductive elastomers can be realized.

Here we have employed a mixing scheme for preparation of heterogeneous bimodal networks³⁶⁻³⁸ to selectively distribute MWCNTs in silicones. Heterogeneous bimodal networks are prepared in a 2-step process as shown in Fig.1, where two hyperbranched intermediates are prepared by separate reaction of a long-chain PDMS (LC-PDMS, 49500 g/mol) as well as a short-chain PDMS (SC-PDMS, 800 g/mol) in two pre-reactions employing sub-stoichiometric amounts of crosslinker. These two hyperbranched systems can then subsequently be mixed and crosslinked with the required amount of crosslinker into a highly flexible and stable elastomer. Such networks are inherently reinforced through their heterogeneous nature, coming from the long chain (LC-PDMS) and short-chain (SC-PDMS) regions in the elastomer. Due to this fabrication procedure, it is possible to add fillers to either of or both of the two hyperbranched parts during their preparation. These systems are therefore prime candidates for selective distribution of particles in PDMS elastomers.³⁹ The purpose of this study was to prepare a stretchable conductive PDMS, employing the heterogenous bimodal network preparation method to distribute highly conductive domains throughout a PDMS elastomer matrix. Through sequential mixing, these high conductivity domains can be mixed with a low-conductivity and flexible matrix (the LC domains), efficiently combining high conductivity of the internal phase (SC domain) with elongation and stretchability.

Experimental Section

Raw materials

Short chain PDMS (SC-PDMS, α , ω -vinyl-terminated PDMS, DMS-V05) and long chain PDMS (LC-PDMS, α , ω -vinyl-terminated PDMS, DMS-V35) with $M_n = 800 \text{ g mol}^{-1}$ and 49500 g mol^{-1} were purchased from Gelest Inc. Multi-walled carbon nanotubes (MWCNTs, NC 7000TM) were supplied by Nanocyl S.A. The average diameter and average length of the MWCNTs are 9.5 nm and 1.5 μm , respectively. Crosslinker (eight functional, HMS-301) was purchased from Gelest Inc. The platinum catalyst (platinum cyclovinyldimethyl-siloxane complex,

511) were obtained from Hanse Chemie AG. All other chemicals were acquired from Sigma-Aldrich, and used as received.

Preparation of MWCNTs/PDMS composites (2.6 phr MWCNTs as an example)

Preparation of hyper-branched SC-domains with MWCNTs First, 0.1 g MWCNTs and 0.5 g SC-PDMS (1,25 mmol functional group) were dispersed in 30 mL toluene by using a sonication probe (200 W, 24 kHz, UP200S, Hielscher– Ultrasound Technology). After ultrasonic dispersion for 15 minutes, 0.0305 g crosslinker (0.125 mmol functional group, stoichiometry $r=0.1$, where stoichiometry r is given by the molar ratio between hydride and vinyl functionalities) and 0.08 mg platinum catalyst were added to the mixture. Then the whole mixture was mixed in the speedmixer (DAC 150 FVZ, Hauschild Corporation) at 3000 rpm for 2 minutes to entrap MWCNTs in hyper-branched short chain PDMS.

Preparation of hyper-branched LC-PDMS with MWCNTs 0.0044 g crosslinker (0.018 mmol functional group, $r=0.1$), 0.6 mg platinum catalyst and small amount of MWCNTs (0.03 g) were dispersed in 4.5 g LC-PDMS (0.18 mmol functional group) by speed-mixing at 3000 rpm for 2 minutes to prepare hyper-branched long chain PDMS.

Preparation of conductive elastomer film The mixtures prepared in step 1 and 2 were mixed in the speedmixer at 3000 rpm for 2 minutes, and residual solvent was allowed to evaporate in a fumehood. Additional crosslinker (0.4712 g, 1,94 mmol functional group, resulting in an overall $r=1.5$) and platinum catalyst (0.76 mg) were added into the mixture and mixed at 3000 rpm for 1 minute. After this, the material was poured into a 1 mm thick steel mold and cured at 120 °C for 1 hour to make film. Several composites of different MWCNTs concentrations were prepared.

Characterization

Mechanical and conductivity testing under strain were performed on a Hounsfield H20K-W. Cyclic bending tests were conducted on an ElectroForce 3200 from TA Instruments. A Fluke 189 digital multimeter was used to record the change in resistance using two-probe measurements. The conductivity was calculated by the following equation:

$$\sigma = \frac{L}{RWt}$$

Where the σ , L , R , W and t are conductivity, length, resistance, width and thickness of samples, respectively.^{40,41} The thicknesses of the samples during elongation were calculated as shown below, assuming volume conservation during elongation (t_1 , t_2 and ϵ are the thicknesses of the sample before and after elongation and strain, respectively):

$$t_2 = \frac{t_1}{\sqrt{(1+\epsilon)}}$$

The micro morphology of entrapped MWCNTs, dispersed MWCNTs and bimodal networks were characterized by optical microscopy (Leica DM LB optical microscopy) and scanning

Fig. 1 Mixing procedure for preparation of heterogeneous bimodal networks containing high amounts of MWCNTs in the SC-domains, bridged by free MWCNTs dispersed in the LC-domains.

electron microscopy (ESEM, Quanta, FEG 250). All measurement data was acquired under ambient atmosphere.

Results and Discussion

The highly conductive internal phase was prepared from an industrial grade MWCNT, which was uniformly dispersed in toluene by sonication together with the SC-PDMS, leading to a premix containing approx. 20 wt% MWCNT. Toluene is a necessity in this step to ensure sufficiently effective dispersion of the large quantity of MWCNT in the SC-PDMS. The premix was mixed with deficient amounts of crosslinker, which was heated and allowed to react under platinum catalysis, resulting in formation of a hyper-branched, highly conductive liquid mixture. Due to high concentration of MWCNTs in the SC-domain, it is not possible to show the actually formed structures by optical microscopy. A sample of the hyper-branched SC-domain was therefore diluted with vinyl PDMS and crosslinked into a thin solid sample that was sufficiently transparent to be investigated by optical microscopy (Fig.2a). Here a characteristic microstructure of entrapped MWCNTs can be observed, which appears as “hairy” conductive aggregates in the transparent PDMS matrix.

Separately, LC-PDMS was hyper-branched by reacting it with deficient amounts of crosslinker. In this step, small amounts of MWCNTs (0.4-0.6 phr, overall) was added in order to enable connections between the highly conductive SC-domains to be formed. The dispersion of small amounts of additional MWCNTs in LC-PDMS can easily be achieved by speed-mixing without the use of solvent, due to the large shear forces generated during mixing. In Fig.2b, optical microscopy images show a dilution of the filled hyperbranched LC domains crosslinked into a PDMS matrix. Finally, the two hyperbranched mixtures were combined (optical image of the dilution in Fig.2c) with a sufficient amount of crosslinker and the mixture was cured, resulting in a flexible and conductive elastomer.

A short parameter study, was initially conducted to establish the most favourable overall composition of the elastomers and the percolation threshold for the various compositions (see compositions in SI-Table 1 and percolation threshold in SI-Fig 1). Without addition of MWCNT in the LC domains (at least 0.4 phr, overall), it was not possible to remain above the percolation threshold and to maintain a sufficient conductivity under elongation of the samples. From a mechanical point of view, it is desirable to keep the MWCNT content as low as possible, and it was therefore limited to 0.4-0.6 phr in overall elastomer composition. Similarly, it is from a practical point of view not possible to increase the MWCNT loading above 2 phr by increasing the content in the SC domains, since this would prevent curing of the elastomer.

Scanning electron micrographs of the MWCNT/PDMS composite with an overall concentration of 2+0.6 phr MWCNTs (2 phr MWCNTs in the SC domain and 0.6 phr MWCNTs in the LC domain) are shown in Fig.2d-e. Here the heterogeneous nature of the sample is clearly visible, with many small separate domains or “islands” being identified throughout the sample. In addition, it is possible to discern the uniformly dispersed MWCNTs from the LC-domains, which are dispersed homogeneously in the polymer matrix. The existence of the “islands” demonstrates that the mixing methodology has allowed a combination of the two domains, where the bimodal structure contains both the highly conductive domain as well as the bridging MWCNTs.

The most important property of an inherently stretchable conductor is how the conductivity changes as a function of elongation. The electric properties of prepared MWCNTs/PDMS nanocomposites were characterized with different loading of MWCNTs in both domains, while the samples were stretched up to 200%. The normalized resistances (R/R_0 , where R is resistance at a given strain and R_0 is the resistance at zero stress state) of MWCNT/PDMS nanocomposites during strain were measured as shown in Fig.3a and b.

Fig. 2 Optical images (a-c) of highly diluted samples of the different stages of the mixing process. a) MWCNTs entrapped in hyper-branched SC-PDMS. b) MWCNTs dispersed in LC-PDMS. c) Bimodal mixture before crosslinking. SEM images of bimodal networks prepared with an overall concentration of 2.6 phr MWCNTs (2 phr in SC-domain and 0.6 phr in LC-domain). For a comparison to a homogeneously dispersed sample of 2.6 phr MWCNT in PDMS, see SI-Fig 7.

For the bimodal system having overall 1+0.4 phr MWCNTs (1 phr MWCNTs from the SC domain and 0.4 phr MWCNTs from the LC domain) and the bimodal system with overall 1+0.6 phr MWCNTs (1 phr MWCNTs from the SC domain and 0.6 phr MWCNTs from the LC domain), the normalized resistances of both samples initially decrease slightly with elongation, where after they are constant until the strain exceeds 120%, where the 1+0.6 phr bimodal system fractures. At this point, the normalized resistance of the 1+0.4 phr MWCNT bimodal system starts to increase until it finally fractures at 160% strain. Similarly, the bimodal systems with 2 phr MWCNT from the SC-domain and 0.4 or 0.6 phr from the LC-domain (Fig.3b), show higher elongations at break, and a constant resistance up to 100% and 160% strain, respectively. This is in both cases followed by a dramatic increase in resistance prior to their respective breaking points at 140% and 200% elongation. We assume that this increase in elongation at break for the 2+0.6 phr MWCNT sample can be attributed to a slightly less efficient crosslinking for the sample with a higher loading, which is corroborated by a minor increase in $\tan(\delta)$, as shown in the rheological data in SI-Fig 3.

Contrary to conventional nano-size particle based composites, it is interesting to note that the prepared MWCNT/PDMS composites in this study show a reduction in resistance with increased strain, which means that the conductivity of MWCNT/PDMS composites is increased by elongation of the samples (data replotted to show conductivity is shown in SI-Fig 2). This interesting property is attributed to the bimodal

network structure, enabling new percolated paths to form during elongation, resulting in a dynamic system where many different conductive pathways can be established in the material.

From previous studies, it is known that high aspect ratio particles like carbon nanotubes will significantly decrease the flexibility of prepared composite,^{33,42} or results in a significant viscous loss in the systems due to poor incorporation of the conductive filler.⁴³ This is also not seen for the prepared conductive bimodal systems, where dynamic mechanical analysis by rheology showed that all the samples are fully cured and exhibit no significant viscous loss (SI-Fig 3). In addition, the samples also show excellent bending properties. For the optimal composition of 2+0.6 phr MWCNT, a bending test is conducted, where the sample is bent to different radii (r), and the resistance is measured at each radius (Fig.3c). The normalized resistance of the 2+0.6 phr MWCNT system is stable throughout the measuring range of the instrument (bending radius from infinite to 0.71 mm). Fig.3d shows the influence of bending cycles and bending radii on the normalized resistance. It can be seen that the normalized resistance remain unchanged within 1000 bending cycles when bending radius is larger than 0.88 mm. However, as the bending radius decreases to 0.71mm, the normalized resistance increases slightly during the first 100 bending cycles and thereafter remains constant until 1000 bending cycles. Because most of the MWCNTs are entrapped in the SC-PDMS domain and only a relatively small amount of MWCNTs are dispersed in the LC-domain, the overall

Fig. 3 Electrical properties of MWCNTs/PDMS composites and reference samples: a) Normalized resistance of MWCNTs/PDMS composites with 1+0.4 phr and 1+0.6 phr MWCNTs under different strains. b) Normalized resistance of MWCNTs/PDMS composites with 2+0.4 phr and 2+0.6 phr MWCNTs under different strains. c) Normalized resistances of MWCNTs/PDMS composites with 2+0.6 phr MWCNTs at different bending radii. d) Normalized resistances of MWCNTs/PDMS composites with 2+0.6 phr MWCNTs at different bending radii and bending cycles.

flexibility of prepared MWCNTs/PDMS composites will not be significantly reduced by addition of MWCNTs and the composite will still be flexible. When the bending radius is large, the conductive network can adapt to the bending deformation, and bending deformation will not impose serious strain on the conductive network. However, as the bending radius decreases to 0.71 mm, we hypothesize that it is difficult for the conductive network to accommodate such large deformations, and the least flexible elements in the conductive path are interrupted during the initial bending cycles (at this bending radius). After this initial event, the normalized resistance stabilizes at a slightly higher level and remains constant during the subsequent bending cycles. The long-time performance of prepared MWCNT/PDMS sample at was similarly investigated through cyclic load tests up to 30% strain as illustrated in SI-Fig. S4. The conductivity of MWCNTs/PDMS composites decrease gradually within first 1000 cycles, where after it remains constant until the test was concluded after 5000 cycles, demonstrating excellent long-term stability of the system. To further understand the conductive structures formed during the preparation of the bimodal networks, reference samples with uniformly dispersed MWCNTs were prepared by roll milling and ultra-sonication (conventional system). The conductivity and break down strain of these systems were compared with

the bimodal system as shown in SI-Fig 5. By extensive dispersion of the MWCNTs, it is possible to prepare samples with similar initial conductivity. The homogeneously dispersed sample prepared by ultrasonication shows a higher conductivity than the bimodal system, which illustrates that the heterogeneous distribution of MWCNTs with a low content of particles in the continuous matrix indeed was achieved through the bimodal methodology. However, in elongation at break, the bimodal system can be elongated up to 200%, compared to the conventional and roll-milled samples that can only be extended up to 70%. For all of the homogeneously dispersed samples, there is a clear tendency that the resistance increases with strain, contrary to the bimodal samples, which all show either constant or increasing conductivity. In all cases, the obtained initial conductivity is comparable to what has been obtained by direct dispersion of conductive particles to prepare freestanding conductive elastomers (see Table S2), though in our case it is achieved with a lower overall content of conductive filler due to the distribution between the two domains.

The structures formed during the initial hyper-branching of the SC-domain have a "hairy" appearance as illustrated in the diluted samples (Fig.2). In addition to having separate domains, this branched appearance of the internal domains was

Fig. 4 Microstructures of MWCNTs/PDMS composites at high dilution embedded in PDMS: a) LC-domain dispersed MWCNTs at zero strain. b) LC-domain dispersed MWCNTs at 100% strain. c) SC-domain MWCNTs at zero strain. d) SC-domain MWCNTs at 100% strain.

hypothesized to be directly related to the reformation of percolative paths through orientation of branches and free MWCNTs during stretching. This was corroborated by optical microscopy of samples diluted in PDMS, in THEIR relaxed state and at 100 % elongation, respectively, as shown in Fig.4.

For the LC-domain it is clear that the free MWCNTs are homogenous dispersed in the matrix (Fig.4a). As the sample is elongated to 100%, the carbon nanotubes orientate along the force direction (Fig.4b), confirming the hypothesis that carbon nanotubes are reorganized in the elongated state. Contrary to this, the SC-domains containing the much higher concentration of carbon nanotubes (Fig.4c), are hard and cannot deform to the same large extent. Only the branched structures on the surface of the SC-domains can orientate along the force direction, as shown for the sample at 100% elongation (Fig.4d). We therefore conclude that it is this combination of MWCNT in the branches on the surface of the SC-domains and the dispersed MWCNTs in the LC-domains that is essential in forming and reforming the percolative pathways.

It is well known that MWCNTs form specific structures by dispersion in solvents, and that some solvents are more efficient in breaking down aggregates of MWCNTs than others. In an attempt to greatly simplify the method presented here, a simple attempt of forming similar structures was therefore conducted using heptane and toluene in combination with roll-milling. By sonication in toluene or heptane the initial agglomerates are broken down, leading to cloud-like structures forming over time in the solution (matured dispersions), similar to the structures observed from the SC-domains. Subsequent mixing of these matured dispersions with PDMS and crosslinking of the systems (after evaporation of the respective solvents), led to elastomers, that showed slightly better elongation at break compared to classical systems, though they still exhibit a decrease in conductivity with elongation (SI-Fig 6), underlining the importance of the bimodal distribution.

Conclusions

A new stretchable conductive material can be obtained by incorporating MWCNTs into a PDMS matrix by a novel, simple, and effective method. The initial electrical conductivity of prepared MWCNTs/PDMS composite can reach 0.038 S m^{-1} with only 2.6 phr MWCNTs and the conductivity keeps constant up to 160% tensile strain. Moreover, instead of reduction of conductivity under strain, increased conductivity can be achieved, when the composites are exposed to large strain, which is a remarkable characteristic compared with other carbon nanotubes based conductive composites. The unique properties indicate that material reported here is a promising material for a variety of electronics applications. In addition to this, the simplicity of the preparation method and the fact that all of the required steps can easily be up-scaled enables this as a feasible fabrication route for large-scale production.

Conflicts of interest

There are no conflicts to declare.

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