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Enhanced electrical conductivity and piezoresistive sensing in multi-wall carbon nanotubes/polydimethylsiloxane nanocomposites via the construction of a self-segregated structure

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Formation of highly conductive networks is essential for achieving flexible conductive polymer composites (CPCs) with high force sensitivity and high electrical conductivity. In this study, self-segregated structures were constructed in polydimethylsiloxane/multi-wall carbon nanotube (PDMS/MWCNT) nanocomposites, which then exhibited high piezoresistive sensitivity and low percolation threshold without sacrificing their mechanical properties. First, PDMS was cured and pulverized into 40–60 mesh-sized particles (with the size range of 250–425 μm) as an optimum self-segregated phase to improve the subsequent electrical conductivity. Then, the uncured PDMS/MWCNT base together with the curing agent was mixed with the abovementioned PDMS particles, serving as the segregated phase. Finally, the mixture was cured again to form the PDMS/MWCNT nanocomposites with self-segregated structures. The morphological evaluation indicated that MWCNTs were located in the second cured three-dimensional (3D) continuous PDMS phase, resulting in an ultralow percolation threshold of 0.003 vol% MWCNTs. The nanocomposites with self-segregated structures with 0.2 vol% MWCNTs achieved a high electrical conductivity of 0.003 S m$^{-1}$, whereas only 4.87 × 10$^{-10}$ S m$^{-1}$ was achieved for the conventional samples with 0.2 vol% MWCNTs. The gauge factor GF of the self-segregated samples was 7.4-fold that of the conventional samples at 30% compression strain. Furthermore, the self-segregated samples also showed higher compression modulus and strength as compared to the conventional samples. These enhanced properties were attributed to the construction of 3D self-segregated structures, concentrated distribution of MWCNTs, and strong interfacial interaction between the segregated phase and the continuous phase with chemical bonds formed during the second curing process. These self-segregated structures provide a new insight into the fabrication of elastomers with high electrical conductivity and piezoresistive sensitivity for flexible force-sensitive materials.

1. Introduction

Flexible conductive polymer composites (F-CPCs), which can easily convert external stresses (tensile, compression, bending, twisting, etc.) into electrical signals, exhibit potential applications in electronic skins,1–3 wearable electronics,4–6 and flexible touch sensors.7–9 Recently, F-CPCs have been designed to achieve various functions, such as piezoresistive sensing,10–12 solvent/gas detection,13–15 health monitoring,16–18 and movement detection,19–21 because of their flexibility, light weight, easy processing, and tunable sensitivity towards the applied stress. In particular, the piezoresistive sensors that transduce the compression stress into electrical resistance signals have attracted significant attention and have also been effectively used in many industrial fields.22–27 For example, a polyurethane sponge coated with carbon black can be used for a human–machine interface platform.28 Normally, high-quality piezoresistive sensing depends on a highly flexible polymer matrix and highly efficient conductive filler networks. First, the selection of a polymer matrix with favorable stretchability and excellent processability is important for piezoresistive sensors. Polyurethane (PU)29 and its foams,30 polydimethylsiloxane (PDMS),31 natural rubber,32 and polyvinylidene fluoride (PVDF)33 have been intensively investigated for piezoresistive sensors. For example, PVDF/
The most important challenge for achieving high quality piezoresistive sensors is to form highly efficient conductive networks in the polymer matrix. Essentially, the variation in the resistance during compression indicates the change in the electron transport property. Thus, the piezoresistive behaviors of F-CPCs can be efficiently tuned by changing the conductive networks (i.e., the transport pathways) in the polymer matrix. Graphene networks, \( \text{carbon nanotubes networks,} \) \( \text{carbon black networks,} \) and networks of their mixtures have been successfully fabricated in the polymer matrix and have exhibited good piezoresistive properties. However, to achieve good electrical conductivity and piezoresistive sensitivity, a large amount of conductive fillers need to be added into the insulating polymers, normally leading to poor processability, low economic affordability, and inferior flexible properties. With the conductive fillers randomly distributed in the insulating polymer matrix, the only way to improve the electrical conductivity of the composites is to reach or exceed the percolation threshold.

A number of strategies have been well-demonstrated to control the distribution of the conductive fillers and further form highly efficient conductive networks in the polymer matrix. For example, double percolation has been used to fabricate highly efficient conductive networks via selectively dispersing conductive fillers in one of the polymer phases. The volume exclusion effect has been utilized to concentrate the conductive fillers in the continuous amorphous phase. Nanoconfinement has also been developed to design highly efficient conductive networks in rubber-like polymers. Recently, the formation of segregated structures in the polymer matrix has become a promising strategy to enhance the electrical property of the CPCs. The conductive fillers can be restrictively located in the continuous phase or at its interfaces around the segregated particles. As a result, dense conductive networks can be formed using this strategy. The CPCs with segregated structures usually exhibited superior electrical conductivity, low percolation concentration, and high piezoresistive sensitivity at ultralow filler loadings.

The segregated structures are usually obtained via two different polymers: one is used as the segregated phase with a high processing temperature and the other is the continuous phase with a low processing temperature. A number of interfaces have been created using this processing method. The interfacial interaction at these interfaces is usually very weak, which leads to poor mechanical properties. Thus, the challenge to achieve segregated structures is how to improve the interfacial interaction between two polymers. The problem can be solved by constructing self-segregated structures with the same components in an individual polymer matrix. However, the related studies have rarely been reported. Yang’s group used chemically cross-linked polyolefin elastomer (POE) granules as the segregated phase and the uncrosslinked POE/multiwall carbon nanotubes (POE/MWCNTs) as the continuous phase to form the segregated elastomeric composites. The obtained conductive elastomeric composites were reported to exhibit not only greatly reduced percolation thresholds (from 9.0 vol% to 1.3 vol%) but also improved mechanical properties (more than 12% and 30% increase in the stress at 100% and 300% strain, respectively). However, because of the high viscosity of the POE and the weak interfacial interaction between the POE and MWCNTs, high dispersion of MWCNTs in the POE phase became very hard to achieve. Thus, the percolation threshold of the segregated samples is still much higher than that of the usual CPCs with segregated structures such as 0.0085 vol% in the PLLA/PCL/MWCNTs nanocomposites with a segregated structure. Furthermore, the processing procedure for the segregated structures is more complex and difficult to control, and POE has poor biocompatibility.

Herein, PDMS/MWCNTs nanocomposite-based piezoresistive sensors were fabricated with low percolation threshold and high sensitivity without sacrificing the interfacial interaction through the formation of self-segregated structures. PDMS was first cured and pulverized into 40–60 mesh-sized particles, with the sizes ranging from 250 to 425 μm, as a self-segregated phase. The uncured PDMS/MWCNTs base together with the curing agent was used as a continuous phase and finally cured after coating with the PDMS particles to form the PDMS/MWCNT nanocomposites with self-segregated structures. The classical percolation threshold theory was applied to investigate the electrically conductive properties. The gauge factor was introduced to evaluate the piezoresistive sensitivity of the nanocomposites. A damage model, in which the self-segregated particles blocked the conductive pathways during compression, was used to discuss the piezoresistive mechanism of the nanocomposites with self-segregated structures.

2. Experimental

2.1 Materials

Polymethylsiloxane (PDMS, Sylgard 184), consisting of a base elastomer and a curing agent, was bought from Dow Corning (USA). The base elastomer was vinyl-terminated PDMS chains, and the curing agent was a mixture of methyl-hydroisiloxane copolymer chains with a Pt catalyst and an inhibitor. The MWCNTs (NC 7000) with an average diameter of 9.5 nm, average length of 1.5 μm, specific surface area of 250–300 m² g⁻¹, and carbon purity of 90% were purchased from Nanocyl S.A (Belgium).

2.2 Preparation of the PDMS/MWCNT nanocomposites with self-segregated structure

First, the PDMS/MWCNT samples with 0, 0.1, 0.2, 0.3, 0.45, 0.5, 0.7, and 1.0 vol% random distribution of MWCNTs were
first prepared. The processing procedure is shown in Fig. 1a. The MWCNTs and PDMS/THF solution were directly mixed to form a uniform mixture, and then, THF was removed to obtain the PDMS/MWCNT suspension. The curing agent was then added, mixed, degassed, and finally cured at 80 °C for 2 h to obtain the PDMS/MWCNT samples with randomly distributed MWCNTs.

For the self-segregated PDMS/MWCNT samples, the PDMS base and curing agent with a weight ratio of 10 : 1 were first mixed well, degassed, and then cured at 80 °C for 2 h to obtain PDMS sheets of ca. 5 mm thickness. The sheets were pulverized using a high-speed mechanical pulverizer (FW100, Tianjin Taisite Ltd, China) and then meshed with a screening sifter. The pulverized cross-linked particles with the mesh size between 40 and 60 (with the size range of 250–425 μm) were utilized for the segregated phase, as shown in Fig. 1b. Different concentrations of PDMS and MWCNTs were first mixed via ultrasonication in tetrahydrofuran (THF) at room temperature. The uncrosslinked PDMS/MWCNT base was obtained by the evaporation of THF. After this, the curing agent and cross-linked particles were simultaneously added to the uncrosslinked PDMS/MWCNT base, mixed well, degassed, and then cured at 80 °C for 2 h to obtain the PDMS/MWCNT nanocomposites with a self-segregated structure (Fig. 1c). Sheets with a diameter of 64 mm and thickness of 1.5 mm were fabricated for investigating the conductivity and mechanical properties. Samples with a diameter of 12 mm and thickness of 3.7 mm were prepared for the piezoresistive evaluation.

For all the self-segregated samples, the uncrosslinked PDMS/MWCNT base and curing agent were kept at a weight ratio of 10 : 1; the cross-linked PDMS particles and the uncrosslinked PDMS/MWCNTs base had a weight ratio of 80 : 20. The content of MWCNTs in the self-segregated samples was kept at 0, 0.01, 0.02, 0.04, 0.08, 0.12, 0.16, 0.2, and 0.5 vol% for the evaluation of the percolation threshold.

2.3 Characterization

The self-segregated structure and MWCNTs distribution in the PDMS elastomers were investigated using a field-emission scanning electron microscope (SEM, JEOL, JSM-7800F) with an accelerating voltage of 10 kV. The samples were first cryofractured in liquid nitrogen and then coated with a layer of platinum in a vacuum chamber before SEM observation for better imaging.

The electrical conductivity and percolation threshold of the samples were evaluated using a digital high resistance machine (PC68, Shanghai Precision Instrument Manufacture, China) at room temperature. At least five specimens were tested and the average data were obtained.

The compression property was investigated using an electrical universal testing machine (Sansi CMT6503 Universal Testing Machine) at the compression speed of 0.5 mm min⁻¹ at 23 ± 2 °C. The varying resistance of the composites was simultaneously obtained using a precision digital resistor (Model TH2683A, Changzhou Tonghui Electronics Co. Ltd, China) under a constant voltage of 10 V to evaluate their piezoresistive
properties. Silver paste was used to ensure good contact between the electrode and the samples. The precision digital resistor and the universal testing machine were coupled with a computer to online investigate the piezoresistive property upon cyclic loading. At least five specimens were tested for each sample, and the average results were evaluated.

3. Results and discussion

3.1 PDMS/MWCNT nanocomposites with self-segregated structures

The distribution of MWCNTs in the conventional PDMS/MWCNT samples was first investigated via SEM. The MWCNTs can be observed to be randomly distributed in the PDMS matrix, as shown in Fig. 2. However, the surface morphology of the samples with the self-segregated structure is very different from that of the conventional samples. For the samples without MWCNTs, the pre-cured PDMS particles and the later cured continuous PDMS phase were found in the self-segregated samples. Interestingly, the cryofractured surface of the PDMS self-segregated phase was much smoother than that of the continuous PDMS phase, as shown in Fig. 3a. Different fracture behaviors of these two phases were ascribed to different mechanical properties of these two phases. Since the PDMS self-segregated phase undergoes two curing processes, it becomes more brittle than the PDMS continuous phase, which undergoes only one curing process. It has been reported that the modulus of PDMS increases with the increasing curing time, which has been discussed hereinafter.

![Fig. 2](image-url) SEM images of the (a) conventional PDMS/0.2 vol% MWCNTs, (b) magnification of a.

![Fig. 3](image-url) SEM images of the (a) self-segregated neat PDMS, (b) self-segregated PDMS/0.2 vol% nanocomposites, (b') medium magnification of b, and (b'') higher magnification of b'. The areas around the red dotted lines indicate the segregated PDMS particles.
For the self-segregated samples with MWCNTs, there are also two different phases: the pre-cured PDMS particles with smooth surfaces and the later cured PDMS/MWCNT continuous phase with rough surfaces. The pre-cured PDMS particles are indicated by the red dotted lines in Fig. 3b and b'. Furthermore, the interfaces of the pre-cured PDMS particles and the later cured continuous phase are shown to be very tight, suggesting high interfacial interaction between them. The high interfacial interaction is probably ascribed to the chemical bonds developed during the second curing process. In addition, it was also found that the MWCNTs were confined in the continuous phase, which developed into 3D conductive networks by constructing a self-segregated structure. Furthermore, the MWCNTs are well dispersed in the PDMS continuous phase, as shown in Fig. 3b'', indicating the high interfacial interaction between the PDMS matrix and MWCNTs.

### 3.2 Mechanical properties of the PDMS/MWCNT nanocomposites with self-segregated structures

Fig. 4a shows the compression stress–strain curves of PDMS and its nanocomposites with different structures and different MWCNT loadings. The mechanical properties were observed to be significantly improved in the PDMS/MWCNT nanocomposites with a self-segregated structure as compared to those of the conventional samples with a random distribution of the MWCNTs. For example, the compression strength of neat conventional PDMS at 30% strain shows only ~15%, ~33%, and ~44% enhancement upon the incorporation of 0.08, 0.2, and 0.3 vol% MWCNTs, respectively. However, the neat self-segregated PDMS samples have a ~170% higher compression strength at 30% strain as compared to that of the neat conventional PDMS samples. The compression strength at 30% strain further increased from 1.7 MPa to 1.88, 1.89, and 2.09 MPa upon the addition of 0.08, 0.2, and 0.3 vol% MWCNTs, respectively, as shown in Fig. 4b.

The compression strength at 80% strain and the compression modulus were also abruptly improved via the construction of the self-segregated structure in the PDMS/MWCNT nanocomposites, as shown in Fig. 4c and d. The compression strength of the self-segregated samples at 80% strain was ~203%, ~202%, ~237%, and ~221% higher than that of the corresponding conventional samples with 0, 0.08, 0.2, and 0.3 vol% MWCNTs, respectively. Furthermore, the compression modulus of the self-segregated samples was ~41%, ~53%, ~38%, and ~52% higher than that of the conventional samples with 0, 0.08, 0.2, and 0.3 vol% MWCNTs, respectively. The mechanical enhancement was ascribed to the high interfacial interaction between the pre-cured PDMS particles and the continuous PDMS phase and the high mechanical property of the pre-cured PDMS particles, which experienced four hours curing in total.71,72

![Compression stress–strain curves of neat PDMS and its nanocomposites with MWCNT loadings](image)

**Fig. 4** Representative compression stress–strain curves (a), the compression strength at 30% strain (b) and at 80% strain (c), and compression modulus of the PDMS and its nanocomposites (including conventional mixed samples and the self-segregated samples) with MWCNT loadings of 0.08, 0.2, and 0.3 vol% (d).
2 hours, respectively. In the self-segregated samples, the pre-cured PDMS particles underwent curing for almost 4 hours, whereas the continuous PDMS phase underwent a 2-hour curing. As a result, the pre-cured PDMS particles were more brittle than those of the continuous PDMS phase, which was consistent with the SEM images. We also believe that the different mechanical properties of these two phases lead to high piezoresistive sensitivity.

3.3 Percolation behavior of the PDMS/MWCNT nanocomposites with self-segregated structures

Fig. 6 shows the comparison between the electrically conductive properties of the PDMS/MWCNT nanocomposites with a self-segregated structure and the conventional nanocomposites. The electrical conductivity of these two types of nanocomposites increases with the increasing filler loading and also shows the classical percolation behavior. As was expected, the electrical conductivity of the self-segregated samples was obviously enhanced because of the concentrated distribution of MWCNTs in the continuous PDMS phase as compared to that of the conventional samples, as shown in Fig. 6a. The electrical conductivity of the self-segregated samples could reach $1.33 \times 10^{-6}$ S m$^{-1}$ upon the addition of only 0.01 vol% MWCNTs. However, the electrical conductivity is only $4.87 \times 10^{-10}$ S m$^{-1}$ for the conventional samples with 0.2 vol% MWCNTs. Moreover, the electrical conductivity of the self-segregated samples could reach 0.003 S m$^{-1}$ upon the addition of 0.2 vol% MWCNTs. The results indicate that the conventional samples exhibit good electrical insulation at low MWCNT loadings, whereas the self-segregated samples show outstanding electrical conduction. The high electrical conductivity of the self-segregated samples is ascribed to the dense and confined distribution of the MWCNTs in the continuous PDMS phase by the self-segregated structures.

The percolation threshold $\phi_c$ of the nanocomposites was also evaluated using the classical percolation theory, which was calculated by fitting the variation in the electrical conductivity $\sigma$ with the content of the MWCNTs $\phi$.

$$\sigma = \sigma_0 (\phi - \phi_c)^t$$

where $\sigma_0$ and $t$ are the plateau conductivity of the fully loaded composites and the scaling exponent, respectively. The $t$ value can also be used to predict the conductive mechanism of the composites. Generally, the values of $t \approx 2$ and $t \approx 1.3$ indicate three-dimensional and two-dimensional conductive networks, respectively.74 Herein, the calculated values of $t$ for the self-segregated samples and the conventional samples are 2.12 and 1.42, respectively, as shown in Fig. 6b. The results indicated that the two-dimensional conductive networks were formed in the conventional samples, whereas three-dimensional conductive networks were formed in the self-segregated samples to enhance the electrical conductivity.

Furthermore, an ultralow percolation threshold of 0.003 vol% (0.0054 wt%) was found in the self-segregated samples. However, the percolation threshold of the conventional samples is about 0.44 vol%, which is about 147-fold that of the self-segregated samples. Moreover, Table 1 provides the reported percolation threshold in the flexible polymer/MWCNT nanocomposites. The results show that the percola-

![Fig. 5](image-url)  
**Fig. 5** Representative compression stress–strain curves of the neat PDMS cured for different times at 80 °C.

![Fig. 6](image-url)  
**Fig. 6** (a) Electrical conductivity of the PDMS/MWCNT nanocomposites with random distribution of MWCNTs and self-segregated structure as a function of the MWCNT loadings. (b) The fitting lines of the nanocomposites obtained via the classical percolation theory.
The piezoresistive properties of the PDMS/MWCNT nanocomposites with self-segregated structures were also evaluated using an electrical universal testing machine. As is known, the piezoresistive properties are mainly related to the variation in the conductive networks (conductive pathways). However, the conductive networks usually very weakly respond to external stimuli when the filler content is close to the percolation threshold. Thus, to ensure stable conductive networks, the stimuli when the filler content is close to the percolation threshold are usually used to investigate the piezoresistive properties. Herein, the conventional samples with 0.6 vol% MWCNTs and the self-segregated samples with 0.12 vol% MWCNTs were chosen to evaluate the resistance variation under compression. Note that the content of MWCNTs in the continuous PDMS phase of the self-segregated samples is also 0.6 vol%, which is the same as that in the conventional samples. In addition, the initial electrical resistivity of the conventional samples with 0.6 vol% MWCNTs and the self-segregated samples with 0.12 vol% MWCNTs is \(7.4 \times 10^{-4}\) and \(2.6 \times 10^{-4}\) S m\(^{-1}\), respectively. The approximate initial electrical resistivity can also provide more reasonable results to perform a comparative investigation.

Fig. 7a shows the relative resistance \(\Delta R/R_0\) (\(\Delta R\) is the change from zero-strain resistance \(R_0\)) due to an applied strain variation for the 1st cycle at 30% compression strain at a compression rate of 0.5 mm min\(^{-1}\). The results show that the value of \(\Delta R/R_0\) increases with the increasing compression strain, which is defined as the positive strain effect, because of the breakdown of the electrically conductive pathways and the gradual increase in the separation between fillers. Furthermore, the \(\Delta R/R_0\) curves at the stretching stage do not overlap those at the release stage, indicating a hysteresis behavior in the unloading process because of the viscoelastic property of the PDMS-based composites. For the conventional samples, the value of \(\Delta R/R_0\) slightly increases, from 0 (zero strain) to 5.2 (30% strain), with the increasing strain suggesting that the positive strain effect is very weak in these conductive networks. However, the value of \(\Delta R/R_0\) for the self-segregated samples obviously increases, from 0 (zero strain) to 42.5 (30% strain), upon increasing the strain. The value of \(\Delta R/R_0\) for the self-segregated samples at 30% strain is nearly 717% higher than that of the conventional samples at 30% strain, indicating the higher piezoresistive sensitivity in the self-segregated samples.

The piezoresistive sensitivity of the F-CPCs can also be evaluated using the gauge factor (GF), which is defined as \(GF = \frac{\Delta R}{R_0 \epsilon}\), where \(\epsilon\) represents the applied strain. Fig. 7b shows the variation of the GF value at different applied strains. For the conventional samples, the GF value, which shows slight
change with the increasing strain, is $\sim 17$ over the strain range from 0 to 30%. However, the GF value of the self-segregated samples, which exhibit an obvious increase with the increasing strain, increases from $\sim 2$ at zero strain to $\sim 142$ at 30% strain. The GF value of the self-segregated samples at 30% strain is 7.4-fold that of the conventional samples, demonstrating that the piezoresistive behavior of the self-segregated samples is more sensitive than that of the conventional samples.

The durability of the piezoresistive properties was also investigated via the 100 stretching–releasing cycles test with a maximum strain of 30% at a 0.5 mm min$^{-1}$ stain rate. Fig. 8 shows the relative resistance ($\Delta R/R_0$) variation during the first 20 stretching and releasing cycles for the self-segregated samples with 0.12 vol% MWCNTs and the conventional samples with 0.6 vol% MWCNTs. The relative resistance monotonically increases with the increasing strain and decreases as the strain decreases, indicating that the damage-reconstruction of the electrically conductive pathways happens during the stretching–releasing cycles. It was also found that the maximum and minimum values of the relative resistance were close at different cycles for both the conventional samples and the self-segregated samples; this suggests that the piezoresistive properties were very steady and had superior durability against repeated loading–unloading cycles in both the samples despite the difference in sensitivity.

The higher piezoresistive sensitivity of the self-segregated samples can be probably ascribed to the quicker damage of the conductive pathways during the compression. Fig. 9 shows the damage model for the conductive pathways that was used to discuss the piezoresistive behavior of the conventional samples and the self-segregated samples. For the conventional samples, some detachment of the MWCNTs in the conductive pathways happens during compression, which leads to increase in the resistance, as shown Fig. 9a. However, the conductive pathways were blocked by the PDMS pre-cured particles under compression, which also increased the resistance, as shown in Fig. 9b. In the self-segregated samples, the modulus of the PDMS particles is higher than that of the PDMS/MWCNT continuous phase. As a result, the distance between PDMS particles is abruptly reduced by the compression stress, which will block the conductive pathways in the PDMS/MWCNT continuous phase. Before compression, the distance between PDMS particles was sufficient for electronic conduction along the MWCNT networks between PDMS particles, like the MWCNT networks among 1, 2, 3, and 4 particles, as shown in Fig. 9b. A conductive pathway is indicated via a red arrow route. After compression, the distance between PDMS particles (such as 1, 2, 3, and 4 particles) is abruptly reduced, and the conductive pathway is broken, which blocks the effective transportation of electrons. Because of the more efficient damage of the conductive pathways via the blocking mechanism, the self-segregated samples exhibit higher increase in the resistance and also higher piezoresistive sensitivity.

4. Conclusions

In this study, the PDMS/MWCNT nanocomposites with self-segregated structures were successfully fabricated via the two-step curing processing: the first curing was carried out to produce cross-linked PDMS particles and the second curing was carried out to form self-segregated structures. The ultralow percolation threshold of 0.003 vol% (0.0054 wt%) is found in the self-segregated samples, which is about 147-fold lower than that of the conventional samples (0.44 vol%). In addition,
the electrical conductivity of the self-segregated samples can reach $1.33 \times 10^{-6}$ S m$^{-1}$ upon the addition of only 0.01 vol% MWCNTs, whereas the electrical conductivity is only $4.87 \times 10^{-10}$ S m$^{-1}$ for the conventional samples with 0.2 vol% MWCNTs. The ultralow percolation threshold and high electrical conductivity of the self-segregated samples are attributed to the 3D conductive networks in the samples. Furthermore, the GF value of the self-segregated samples at 30% strain is 7.4-fold that of the conventional samples, demonstrating that the piezoresistive behavior of the self-segregated samples is more sensitive than that of the conventional samples. The self-segregated samples not only exhibit high piezoresistive sensitivity and low percolation threshold but also show mechanical enhancement. For example, the self-segregated samples have ~170% higher compression strength at 30% strain than that of the conventional samples. We believe that these self-segregated structures can provide a new insight into the fabrication of elastomers with high electrical conductivity and piezoresistive sensitivity for flexible piezoresistive sensing, health monitoring, and movement detection.

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Notes and references
