Materials Research Express

OPEN ACCESS

PAPER

Electrical percolation and dynamic piezoresistivity of silver nanoparticle/polydimethylsiloxane films

Shan Heng1, Hongguang Liu1, Chengyuan Wang1,2, Chun Tang1,3, Ying Luo1 and Xuesen Wang2

1 Faculty of Civil Engineering and Mechanics, Jiangsu University, No. 301 Xuefu Road, Zhenjiang, Jiangsu 212013, People’s Republic of China
2 Changzhou Liduo Alloy Cooperation Limited, No.35 Leshan Road, Xinbei District, Changzhou 213125, People’s Republic of China
3 Authors to whom any correspondence should be addressed.

E-mail: cywang@ujs.edu.cn and tangchun@ujs.edu.cn

Keywords: nanoparticles, piezo-resistivity, electrical percolation, polymer, stress sensor

Abstract

Electrical percolation and piezo-resistivity under cyclic load were studied for silver nanoparticle (AgNP)/polydimethylsiloxane (PDMS) films. Attention was focused on the percolation process and the impacts of the percolation stage and load frequency on the piezoresistivity of the films. A power law was uncovered between the electrical resistance and the surface mass density (md) of AgNP layer. Dependence of piezoresistive sensitivity and linearity on md and cyclic load frequency were also investigated and interpreted in terms of the physical changes of the AgNP layer and the dynamics of the strain-induced microcracks. In particular, a peak gauge factor 75 was achieved at md = 0.32 mg cm−2 and the load frequency 0.5 Hz, and excellent linearity was found at the frequency 1 Hz or higher. These new findings provide important guidance for the design and applications of AgNP/PDMS films in body motion tracking, fatigue load monitoring and surface vibration detecting.

1. Introduction

Flexible electronics [1–4] has become a mainstream in the development of advanced technologies in multi-industries. Materials with high electrical conductivity but low elastic modulus and mass density play a crucial role in reaching this goal. One of the strategies used in this area is to distribute conductive nanofillers into polymer elastomers [5]. Due to the strong piezo-resistive effects of these materials, such nanocomposites show great potential for bendable and stretchable stress/strain sensors with high sensitivity [6, 7].

The last decade has witnessed the rapid development of these flexible and piezoresistive nanocomposites or double-layered films [5–11]. Comprehensive studies have been conducted to characterize their unique features, where the gauge factor (GF) in the order of 10 to 103 [8–11], work range (maximum strain) up to 100% [6, 8, 9] or even higher, piezoresistive linearity (R2) greater than 0.996 [9] and degree of hysteresis (DH) less than 12% [7, 9, 11] have been reported. Piezoresistive effect under cyclic loads has also been studied [6, 9] due to its important applications in body motion tracking, fatigue load measurement and stress/strain sensing of dynamic surface. The influence of the load frequency and electrical percolation process on dynamic piezo-resistivity, however, have not been discussed in detail so far. In addition, for the double-layered films, the electrical percolation law has not yet been established in terms of the relation between electrical resistance and the amount of the nano-conductors. Further investigations thus are necessary to study the percolation behaviour and examine the dependency of the piezoresistive sensitivity and linearity on the load frequency and percolation stage. The information and the underlying mechanisms are of importance for the design, fabrication and applications of these stress/strain sensing materials.

Up till now various nano-conductors and polymer substrates (matrixes) have been used, among which AgNP/PDMS films have attracted considerable attention due to the merits of metal NP-based nanocomposites, such as simple printing and sintering processes at low temperature, short fabrication time and compatibility.
with different matrixes [6], PDMS matrix also shows advantages over other polymer substrates due to its low elastic modulus, high corrosion resistance, high transparency and mechanical as well as thermal stability. These distinctive features provide the impetus to systematically examine the aforementioned issues for AgNP/PDMS double-layered films in the present work. The results and insights of this research are expected to provide new guidelines for designing high performance electronics based on flexible piezo-resistive stress/strain sensors. The layout of the paper is as follows. The experimental techniques and fabrication method of AgNP/PDMS films are briefly described in section 2. The results regarding the percolation, hysteresis and tensile test under cyclic strains are presented, and the physical mechanisms are discussed in section 3. Finally, the conclusions are summarized in section 4.

2. Experimental methods

2.1. Chemical reagents and materials

The chemical reagents used in fabricating the AgNP/PDMS films include silver nitrate (AgNO3, 99.8%), anhydrous dextrose (C6H12O6, 99%), ammonia (NH5O, 25%-28% wt), dopamine (C8H11NO2·HCl, 98%), trihydroxymethyl aminomethane (Tris, 99.9%) and hydrochloric acid (HCl, 35%-38%). All of these reagents were purchased from Shanghai Macklin Biochemical Co., Ltd. The PDMS films (thickness 0.3 mm) were used here were purchased from Hangzhou Baoerde New Material Technology Co., Ltd.

2.2. AgNP/PDMS film fabrication

There are two steps in fabricating AgNP/PDMS films, i.e., (1) functionalizing the surface of PDMS films and (2) coating the functionalized surface of PDMS substrate with AgNP layer.

To functionalise PDMS surface, we first prepared 100 ml of Tris solution with concentration of 10 mM. At temperature 60 °C, HCl was introduced into the Tris solution to adjust its PH value to 8.5. This Tris-HCl solution was used as a buffer solution. Subsequently 200 mg of dopamine powder was dissolved into the Tris-HCl buffer solution. By stirring the solution for 5 min we obtained the final dopamine solution, which was kept at 60 °C for use. A PDMS film was then selected and washed for 10 min in an ultrasonic bath using ethanol solution and distilled water. After this, the film was submerged in the dopamine solution for 24 h. The functionalised PDMS film was subsequently cleaned with distilled water and dried in a vacuum drying cabinet at room temperature.

In the second step, 2% ammonia was added into 20 ml of AgNO3 solution (0.2 mM). When the solution became white and transparent we mixed it (volume 25 ml and concentration 0.5 mM) with glucose solution. The functionalized PDMS film was then submerged in the final solution for 5 to 17.5 min (i.e., coating time tc of AgNPs). In this process, the AgNPs were coated onto the surface of the functionalised PDMS substrate.

2.3. AgNP/PDMS film characterization

In this study, morphological analysis of the AgNP/PDMS films was carried out using FEI-Quanta 200FEG scanning electron microscope. Focus was first placed on understanding the morphological change of AgNP layer coated on the surface of the PDMS substrate. To monitor the percolation process, the mass of AgNPs coated on the PDMS was also measured (by the electronic scale (DV215CD) produced by OHAUS instrument) as the difference between the mass of the AgNP/PDMS film (after AgNP coating) and the corresponding PDMS film (before AgNP coating). The films were completely dried before weighting. For each AgNP/PDMS sample, the measurement was repeated for five times and the average value was reported as the mass of AgNPs on the PDMS substrate. The surface mass density mAg of AgNPs on the PDMS was then calculated by dividing the mass of AgNPs by the area of the PDMS surface.

The electrical resistance was also measured for the AgNP/PDMS films with different AgNP coating time tc and the surface mass density mAg of AgNPs. In doing this, a four-point probe (M-3) produced by Suzhou Crystal Electronics was employed. At each coating time tc, five samples were fabricated and five points on each sample were randomly selected for resistance measurement. The average value obtained was used as the electrical resistance of the AgNP/PDMS film associated with a given tc or mAg.

Finally, the piezoresistive sensitivity and linearity was quantified for the AgNP/PDMS films under cyclic tensile strain at different frequencies and amplitudes. The load frequency in this study varied from 0.1 Hz up to 2 Hz, and the maximum strain εranged from 1% up to 5%. A custom stress-strain-electrical resistance testing system was used here, which includes a universal extensometer (HS-3000 Shanghai Heshen Instrument), a digital multimeter (Keithley-2100) and a computer. Two ends of the rectangular AgNP/PDMS film (1 cm x 6 cm) were fixed on the insulating fixture of the extensometer. Here the stress-strain curves were recorded by using universal extensometer and the electrical resistance was measured in parallel using the digital multimeter for different load frequencies and maximum strains.
3. Results and discussion

3.1. Electric percolation and hysteresis

In this section, the attention was first focused on the electrical percolation of the AgNPs on the surface of the PDMS substrate. The whole process can be monitored by the surface morphology, the coating time $t_c$ and the surface mass density $m_d$ of AgNPs. Herein the scanning electron microscope images of the surface morphology are shown in figures 1(a)–(c) for the AgNP layer with $t_c$ equal to 5 min, 12.5 min and 17.5 min, respectively. It is seen from figure 1(a) that at $t_c = 5$ min (or $m_d = 0.16 \text{ kg m}^{-2}$) AgNPs are relatively small and the gaps between adjacent AgNP clusters can be observed. When the coating time increased from 5 min to 10 min (or $m_d$ grew from 0.16 kg m$^{-2}$ to 0.27 kg m$^{-2}$), the average size of AgNPs rose from around 140 nm to 200 nm (supplementary materials is available online at stacks.iop.org/MRX/7/045701/mmedia). In figures 1(b) and (c), at $t_c = 12.5$ min (or $m_d = 0.32 \text{ kg m}^{-2}$) and $t_c = 17.5$ min (or $m_d = 0.38 \text{ kg m}^{-2}$) the size of AgNPs further increased leading to a nearly continuous coating layer. After this, AgNPs may start piling up to form additional AgNP layers. Obviously, the mass density $m_d$ of AgNPs on PDMS surface increases with the rising coating time.

In figure 2 we demonstrate how the mass density $m_d$ of AgNPs grows when the coating time $t_c$ increases from 5 min, to 7.5 min, 10 min, 12.5 min and 17.5 min. An almost linear relation was obtained, i.e., $m_d = 0.017t + 0.089$ (mg cm$^{-2}$), showing a steady mass growth stage at $5 \text{ min} \leq t_c \leq 17.5 \text{ min}$ characterized by a nearly constant rate of the density change of $0.017 \text{ mg cm}^{-2} \text{ min}^{-1}$. Prior to this steady stage, there should be a transition zone at $0 \leq t_c < 5$ min, where a highly nonlinear $m_d$ and $t_c$ relation can be expected. These results, to
some extent, quantitatively describe the dynamic behavior of AgNP nucleation and coalescence on the surface of the PDMS substrate functionalised by the dopamine solution.

The dependence of the electrical resistance $R$ on the surface mass density $md$ is plotted in figure 3 for the AgNP/PDMS films. As expected, $R$ is found to decrease with rising surface mass density $md$. In particular, a power law $R = 2.81 \times 10^4 e^{-23.8md}$ (Ω) is achieved between $R$ and $md$, which governs the percolation behavior of the AgNP layer on the surface of the PDMS substrate. Here, the surface mass density $md$ controls the electrical resistance as it determines the fraction of AgNPs in physical contacts and the tunneling distance between AgNPs [12–16].

Specifically, the electrical percolation threshold of AgNPs on the PDMS surface is found to be around $md = 0.16$ mg cm$^{-2}$ as $R$ decreases rapidly from 59 Ω to 1.04 Ω by nearly 60 times when $md$ grows from 0.16 mg cm$^{-2}$ to 0.32 mg cm$^{-2}$ by a factor of 2. This indicates a transition percolation stage with a relatively wide range, i.e., $0.16 \text{mg cm}^{-2} \leq md \leq 0.32 \text{mg cm}^{-2}$, a distinctive feature of nano-particle percolation [17, 18]. In this transition stage the main conduction mechanism switches from electron tunneling at $md = 0.16$ mg cm$^{-2}$ to contact conductance at $md = 0.32$ mg cm$^{-2}$ [17, 18]. $R$ changes rapidly with $md$ in the transition stage. The rate of change however decreases with growing $md$ and approaches zero (or $R$ approaches an asymptotic value 1.1Ω) at $md > 0.32$ mg cm$^{-2}$. In other words, beyond the transition stage the AgNP/PDMS films move into a steady stage where the dependence of the electrical resistance on the surface mass density $md$ turns out to be trivial.

Within the transition stage ($0.16 \text{mg cm}^{-2} \leq md < 0.32 \text{mg cm}^{-2}$), the number of AgNPs in physical contact is small. Therefore, raising the surface mass density $md$ via the nucleation and/or growth of AgNPs would efficiently decrease the distance between AgNPs (or AgNP clusters) and increase the fraction of AgNPs in
physical contact. Specifically, consistent with the observations in figure 1, at $m_d = 0.32 \text{ mg cm}^{-2}$ the density of the conductive paths on AgNP layer almost reaches its maximum value. Further raising $m_d$ in the steady stage ($m_d > 0.32 \text{ mg cm}^{-2}$) may result in the accumulation of AgNPs. This however would not substantially improve the density of the conductive paths.

Hysteresis describes the reliability or the stability of the stress/strain sensing and is one of the major issues in the development of stress/strain sensing materials. To quantify this property, the tensile tests were performed, where the stress-strain curves were recorded in three loading and unloading cycles along longitudinal direction. In transverse direction, the hysteresis properties would be similar as AgNPs are randomly distributed on the isotropic PDMS substrate. Here, the film was selected with $t_c = 12.5 \text{ min}$ ($m_d = 0.16 \text{ mg cm}^{-2}$) and the stretching rate was fixed at 50 mm min$^{-1}$. The results are shown in figure 4 for three cycles where maximum strain is 30%, 50% and 95%, respectively. The small area enclosed by the loading and unloading stress-strain curves measures the energy dissipation during the loading-unloading cycle due to the internal friction between the large molecule chains of the viscous PDMS substrate\[19, 20\]. In addition, some residual strain was also obtained in all three cycles. Specifically, the energy dissipation and residual strain are found to increase gradually with the growing maximum strain. The mechanical hysteresis rate $H_m$ defined as the ratio between the energy lost in a loading-unloading cycle and the input work was calculated for the films based on equation (1).\[21\]

$$H_m = \frac{W_{\text{load}} - W_{\text{unload}}}{W_{\text{load}}} \times 100\%$$

where $W_{\text{load}} = \int_0^{x_{\text{max}}} F_{\text{load}} \, dx$, $W_{\text{unload}} = \int_0^{x_{\text{max}}} -F_{\text{unload}} \, dx$, $F_{\text{load}}$ and $F_{\text{unload}}$ are the forces applied in loading and unloading processes, respectively. When the maximum strain rises from 30% to 60% and 100%, $H_m$ grows from 9.36% to 9.43% and then 11.57%, indicating excellent mechanical stability of the AgNP/PDMS films. These properties show the advantages of the AgNP/PDMS films when comparing to the recent experimental data of carbon nanotube coated PDMS films, where $H_m$ around 20% was obtained at maximum compression 50%\[21\]. Here, it should be pointed out that the hysteresis cycle would change due to the variation of the mass density of AgNPs. This change however would not be very large as the hysteresis cycle is mainly determined by the material properties of the PDMS substrate.

3.2. Piezo-resistive sensitivity and linearity

Herein, the piezo-resistive sensitivity and linearity were studied for the AgNP/PDMS double-layered films subjected to cyclic tensile strains. Two important issues were examined, i.e., the effects of the percolation stage and the cyclic strain frequency (CSF) on the above-mentioned piezo-resistive properties. The percolation stage was described by the surface mass density $m_d$ of AgNPs on the PDMS substrate. The relative change in electrical resistance $\Delta R / R_0$ was measured for the AgNP/PDMS films under cyclic strain with maximum value $\varepsilon$ of 1% to 5%. Here $R_0$ is the baseline electrical resistance and $\Delta R$ is the change of resistance due to the applied strain.

Measurements were first conducted at constant CSF of 1 Hz and the results are shown in figure 5 for the AgNP/PDMS film with $m_d = 0.32 \text{ mg cm}^{-2}$. As seen in figure 5, for each $\varepsilon$, $\Delta R / R_0$ varies slightly with the increasing number of cycles (or time) and in general, fluctuates around a constant average value, which

![Figure 4. Mechanical hysteresis cycles obtained for AgNP/PDMS with surface mass density $m_d = 0.32 \text{ mg cm}^{-2}$ and the maximum strain of 30%, 50% and 95%, respectively.](image-url)
monotonically increases with rising $\varepsilon$. For example, the average $\Delta R / R_0$ rises from 0.38 to 0.89, 1.21, 1.67 and 2.09, as the maximum strain increases from 1% to 2%, 3%, 4% and 5%. The relation can be approximated by $\Delta R / R_0 \propto 41.9 \varepsilon$, where the slope represents the average piezo-resistive sensitivity of the AgNP/PDMS films.

To evaluate the influence of the percolation stages on the piezoresistive linearity and sensitivity we carried out the tensile tests for the AgNP/PDMS films at different percolation stages or different $m_d$. The CSF was again fixed at 1 Hz. The relation between $\Delta R / R_0$ and $\varepsilon$ is plotted in figure 6 for all the AgNP/PDMS films considered. The linear fitting to the experimental data is also shown in the figure. Clearly, under a cyclic strain, $\Delta R / R_0$ increases almost linearly with the maximum strain $\varepsilon$ at all percolation stages. The piezoresistive linearity characterized by R2 grows from 0.993 to 0.996 and 0.999 when $m_d$ rises from 0.27 mg cm$^{-2}$ to 0.32 mg cm$^{-2}$, 0.34 mg cm$^{-2}$ and 0.38 mg cm$^{-2}$. It seems that the piezo-resistive linearity tends to increase slightly with the rising surface mass density $m_d$ of AgNPs. A relatively lower value 0.942 is obtained at $m_d = 0.34$ mg cm$^{-2}$, possibly due to the uncertainty in the measurement.

In figure 7, the gauge factor (GF) of the films considered in figure 6 was calculated as a function of $\varepsilon$ by using equation (2).

$$\text{GF} = [(R - R_0) / R_0] / \varepsilon$$  \hspace{1cm} (2)
Here $R$ is the electrical resistance at the maximum strain $\varepsilon$ of the cyclic load. The average GF for a given $m_{d}$ is also represented by dashed lines in figure 7 where the inset shows the tendency of the average GF to change with the surface mass density $m_{d}$. As shown in figure 7 and the inset with CSF fixed at 1 Hz, a large GF close to 41 is achieved at $m_{d} = 0.32$ mg cm$^{-2}$. It then decreases drastically to 13 when $m_{d}$ reduces to 2.7 mg cm$^{-2}$ within the transition stage of the percolation, and declines gradually to 32 and 34, as $m_{d}$ increases to 0.34 mg cm$^{-2}$ and 0.38 mg cm$^{-2}$ at the steady percolation stage. It is noted that GF shown here is comparable to the value of 10 obtained for AgNP/PDMS films [22], 32 achieved for P3HT-NF/PDMS semiconductor composites [9] and AgNP thin films [6], and 50 to 60 of AgNP/PET films [23].

Here, the piezo-resistivity may originate from the breakup of the conductive paths due to the tension-induced separation of AgNPs and/or the micro-cracks found in the AgNP layer [24]. The microcracks were indeed observed in previous studies [6, 12, 20, 25] and the present work for the double-layered films (figure 8) mainly due to the mismatch of deformability between the PDMS substrate and conductive coating layer. The nucleation and propagation of the micro-cracks would result in the strain relaxation in the local area of the substrate. Thus AgNP separation (without cracks) due to the strain of the PDMS substrate would generally become much less significant.

Following the above analyses, we believe that the high piezoresistive sensitivity of the AgNP/PDMS films should be primarily attributed to the microcracks on the AgNP layer [6, 12, 20, 25]. The greatest impact of the micro-cracks occurs at the interface ($m_{d} = 0.32$ mg cm$^{-2}$) between the transition and steady percolation stages, where the electrical circuits of AgNPs almost saturate the surface of the PDMS substrate and the electrons are transported primarily via the contact conductance, instead of electron tunneling between AgNPs [26]. Thus, at $m_{d} = 0.32$ mg cm$^{-2}$ the cracks can most efficiently reduce the density of the conductive paths and lead to high piezoresistive sensitivity. At the transition stage (i.e., $0.16 \text{ mg cm}^{-2} \leq m_{d} < 0.32 \text{ mg cm}^{-2}$) the number of electrically connected AgNPs per unit area of PDMS surface (or the surface density of AgNP conductive paths) should be much smaller than that at $m_{d} = 0.32$ mg cm$^{-2}$. As a result, the cracks formed on the AgNP layer would become much less efficient in decreasing the surface density of the electrically connected AgNPs. This in turn results in substantially lower piezoresistive sensitivity. On the other hand, at the steady stage (i.e., $m_{d} > 0.32$ mg cm$^{-2}$) more AgNPs added to the PDMS surface may result in thicker and stronger AgNP layer, which could decrease the density of the microcracks formed on AgNP layer during cyclic straining and ultimately reduce the piezoresistive sensitivity or GF value.

It is worth mentioning that the physical mechanisms of piezo-resistivity should be different for double-layered AgNP/PDMS films and AgNP/PDMS nanocomposite where AgNPs are randomly distributed in the PDMS matrix. For the double-layered films, the micro-cracks of the AgNP layer play a key role in determining the piezoresistive sensitivity while for the nanocomposite the strain-induced separation of AgNPs should be the main physical origin [24, 27]. Accordingly, the nanocomposite should show the highest sensitivity at the transition stage where the electron tunneling (highly sensitive to the distance change between AgNPs) is predominant [28].

In what follows, we further examined the impacts of CSF on the piezoresistive linearity and sensitivity. In doing this, the tensile tests were performed for the AgNP/PDMS films (with $m_{d} = 0.32$ mg cm$^{-2}$) subjected to

![Figure 7](image-url)
cyclic strains with CSF of 0.1 Hz, 0.5 Hz, 1 Hz and 2 Hz, respectively. The relative resistance change \( \Delta R / R_0 \) is plotted in figure 9 as a function of \( \varepsilon \). The linear fitting is also plotted in figure 9 and represented by the dashed lines. It is seen that the high piezoresistive linearity characterized by \( R^2 \geq 0.993 \) is achieved when CSF is 1 Hz or higher. Substantially lower linearity associated with \( R^2 = 0.789 \) and 0.863 is obtained when the lower frequencies 0.1 Hz and 0.5 Hz were considered. In other words, the piezoresistive linearity of the AgNP/PDMS films generally increases with rising CSF with excellent linearity achieved at CSF \( \geq 1 \) Hz. In addition to the linearity, GF of the AgNP/PDMS film \( (m_d = 0.32 \text{ mg cm}^{-2}) \) was calculated in figure 10 against \( \varepsilon \) for the CSFs considered in figure 8. At \( \varepsilon \leq 2\% \), GF of the film is found to increase monotonically from around 30 at CSF of 1 Hz to around 42 at CSF of 1 Hz and reach its maximum value about 75 at CSF of 0.5 Hz. It then decreases to around 60 when CSF further decreases to 0.1 Hz. Similar CSF-dependence is also observed for GF associated with \( \varepsilon \) rising from 2% up to 5% but the effect of the CSF turns out to be smaller.

The above results show the substantial effects of CSF on the piezoresistive sensitivity of the AgNP/PEMS films. This in fact reflects the influence of the loading and unloading frequency on the dynamics of the
microcracks of the AgNP layer, i.e., the opening, propagation and relaxation of the micro-cracks [22]. The microcracks open, grow and relax when tensile load is applied and close when the external load is removed. At CSF much higher than 0.5 Hz, the micro-cracks may close rapidly and do not have enough time to propagate. This would lead to the shorter length and lower density of the micro-cracks, and thus the lower sensitivity or GF of the AgNP/PDMS films. In contrast, at CSF much lower than 0.5 Hz, there would be sufficient time for the relaxation (e.g., crack bridging and length reduction [6, 12]) of the micro-cracks, which would again decrease the density of the microcracks and eventually impair the piezo-resistive sensitivity or GF of the films.

4. Conclusions

In this work, experimental techniques were used to study the electrical percolation and dynamic piezo-resistivity of the AgNP/PDMS films. The emphases were placed on the percolation behavior and the effects of the percolation stage and frequency of cyclic load on film piezo-resistivity.

In coating the PDMS substrate with AgNP layer the surface mass density $m_d$ of AgNPs increases almost linearly with the coating time $t_c$ after an initial transition stage. In this process, the electrical resistance $R$ of the AgNP layer decreases with rising surface mass density $m_d$ by following a power law relationship, $R \propto e^{-23.8m_d} (\Omega)$. The percolation thus shows a transition stage where $R$ grows rapidly with $m_d$ followed by a steady stage with $R$ nearly independent of $m_d$.

A peak gauge factor 42 is achieved at $m_d = 0.32 \text{ mg cm}^{-2}$, where the strain-induced micro-cracks can most efficiently reduce the density of the AgNP conductive paths. The gauge factor however decreases rapidly in the transition stage due to the lower density of AgNP conductive paths and declines gradually in the steady stage possibly due to the thicker and/or stronger AgNP layer with a lower density of micro-cracks.

The peak gauge factor associated with $m_d = 0.32 \text{ mg cm}^{-2}$ also changes with the cyclic load frequency and reaches its global maximum 75 at 0.5 Hz. It then declines when the frequency shifts away from the value as the frequency shifts have substantial impacts on the micro-crack dynamics and finally lead to the shorter length or lower density of micro-cracks.

The high piezo-resistive linearity is only achieved at a load frequency equal to or higher than 1 Hz. In particular, when the frequency falls in this range, the piezo-resistive linearity seems to increase with the growing surface density $m_d$ of the AgNP layer. These results suggest that in fabricating the double-layered films AgNP coating time is a key parameter that can be used to control the surface mass density of AgNPs and thus the piezo-resistive sensitivity as well as the electrical conductivity. In addition, the film-based strain/stress sensors will show higher sensitivity when the AgNP surface mass density of the film approaches 0.32 mg cm$^{-2}$ and the sensors are used to detect the cyclic loads at frequency closer to 0.5 Hz. The sensors will also exhibit excellent piezoresistive linearity when the load frequency is higher than 1 Hz.
ORCID iDs

Chengyuan Wang  https://orcid.org/0000-0002-9790-4342
Chun Tang  https://orcid.org/0000-0002-7767-2126

References

[1] Rogers J A, Someya T and Huang Y 2010 Science 327 1603
[2] Harris K D, Elias A L and Chung H J 2016 J. Mater Sci. 51 2771
[3] Wan Y B, Wang Y and Guo C F 2017 Mater. Today Phys. 1 61
[4] Sim K S, Rao Z Y, Ershad F H and Yu C J 2019 Adv. Mater. 32 1902417
[5] Yao S and Zhu Y 2015 Adv. Mater. 27 1480
[6] Kim S Y, Lee J M, Park J H, Lee J Y, Kim J H and Inkyu Park I K 2019 Funct. Compos. Struct. 1 035006
[7] Park K, Kim S, Lee H, Park I and Kim J 2019 Sens. Actuators A Physical 295 541
[8] Shih X, Liu S, Sun Y, Liang J J and Chen Y S 2018 Adv. Funct. Mater 28 1800850
[9] Kim H J, Thukrai A and Yu C 2018 ACS Appl. Mater. Interfaces 10 5000–6
[10] Zheng Y, Li Y, Li Z, Wang Y L, Dai K and Zheng G Q 2017 Compos. Sci. Technol 139 64
[11] Yi L Z, Jiao W H, Wu K, Quo I H, Yu X X, Xia Q, Mao K M, Yuan S L, Wang S and Jiang Y T 2015 Nano Research 8 2978
[12] Olichner N, Leib E W, Halfar A H, Petrov A and Vossmeyer T 2012 ACS Appl. Mater. Interfaces 4 6151
[13] Terrill R H et al 1995 J. Am. Chem. Soc. 117 12537
[14] Steinecker W H, Rowe M P and Zellers E T 2007 Anal. Chem. 79 4977
[15] Krasteva N, Fogel Y, Bauer R E, Müllen K, Josephy Y, Matsuzawa N, Yasuda A and Vossmeyer T 2007 Adv. Funct. Mater. 17 881
[16] Kim Y, Zhu J, Yeon B J, Prima M D, Su X L, Kim J G, Yoo S J, Uber C and Kotov N A Nature 500 59
[17] Wang G T, Wang C Y, Tang C, Zhang F L, Sun T and Yu X Z 2018 J. Phys. Chem. 122 8614
[18] Zhang F L, Wang G T, Wang C Y, Tang C, Sun T and Yu X Z 2018 Appl. Phys. Letts. 113 253105
[19] Hilyard N C and Cunningham A 1994 Low Density Cellular Plastics (Physical Basis of Behavior) (Dordrecht: Springer)
[20] Alger M 2017 Polymer Science Dictionary (London, Weinhein New York Tokyo Melbourne Mardas: Springer)
[21] Park K, Kim S H, Lee H S, Park I and Kim J 2019 Sens. Actuators A Phys. 295 541
[22] Lee J, Kim S H, Lee J J, Yang D J, Park B C, Ryu S and Park I 2014 Nanoscale 6 11932
[23] Zhan Z L and Si T T 2017 Sens. Actuators A Phys. 264 188
[24] Duan J Y, D’hooge D R and Cardon L 2019 Prog. Mater. Sci. 100617 (Available online)
[25] Li X et al 2012 Sci. Rep. 2 870
[26] Bao W S, Meguid S A, Zhu Z H and Weng G J 2012 J. Appl. Phys. 111 093726
[27] Liu H, Li Y L, Da K, Zheng G Q, Liu C T, Shen C Y, Yan X R, Guo J and Guo Z H 2016 J. Mater. Chem. C 4 157
[28] Wang G T, Wang C Y, Tang C, Zhang F L and Yu X Z 2018 Comp. Mater. Sci. 150 102