NANO LETTERS

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Stretchable Electrets: Nanoparticle−Elastomer Composites

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Cite This: https://dx.doi.org/10.1021/acs.nanolett.0c01434 Read Online

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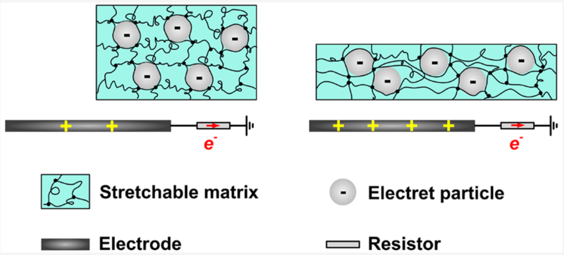

ABSTRACT: Manipulating charges is fundamental to numerous systems, and this ability is achieved through materials of diverse characteristics. This ability is achieved through materials of diverse characteristics. Electrets are dielectrics that trap charges or dipoles. Applications include electrophotography, microphones, air filters, and energy harvesters. To trap charges or dipoles for a long time, electrets are commonly made of hard dielectrics. Stretchable dielectrics are short−lived electrets. The two properties, longevity and stretchability, conflict; existing electrets struggle to attain both. This work describes an approach to developing stretchable electrets. Nanoparticles of a hard electret are immobilized in a matrix of dielectric elastomer. The composite divides the labor of two functions: the particles trap charges with longevity, and the matrix enables stretchability. The design considerably broadens the choice of materials to enable stretchable electrets. Silica nanoparticles in the polydimethylsiloxane elastomer achieve a charge density ∼ 4×10^{-5} C m^{-2} and a lifetime beyond 60 days. Long−lived, stretchable electrets open extensive opportunities.

KEYWORDS: *Electrets, Elastomers, Nanoparticles, Net charge, Electromechanical transduction*

The separation of positive and negative charges is prerequisite for electromechanical transduction. The mechanisms of charge separation vary with materials. A dielectric separates charges of different polarities that accumulate at the interfaces between the dielectric and two electrodes. A piezoelectric breaks the symmetry of the crystal, so that the center of positive charges and the center of negative charges separate on the scale of the unit cell of the crystal, giving a spontaneous polarization.[1,2] An electret traps injected charges separated on a macroscopic scale or traps molecular dipoles.[3−5] Contact electrification induces charges near the surface of a dielectric.[6,7] All of these materials are being developed to enable electromechanical transduction in stretchable electronics. Applications include human−computer interfaces and soft robots.

The development of a stretchable electret faces a fundamental challenge. An electret is commonly required to separate charges for the entire life of a device, often over the years. Whereas a piezoelectric is in a thermodynamic phase (i.e., the ferroelectric phase) and separates charges by a thermodynamic effect, an electret separates charges by a kinetic effect. All dielectrics can trap charges or dipoles and function as electrets. Some electrets are short−lived, and others are long−lived. To trap charges or dipoles for a long time, electrets are commonly made of hard dielectrics (e.g., inorganics and plastics).[8−13] Stretchable dielectrics such as elastomers are short−lived electrets: they discharge fast. The two properties, longevity and stretchability, conflict. Existing electrets struggle to attain both.

The struggle to resolve the longevity−stretchability conflict is playing out in the development of stretchable electrets. Hard electrets can be made soft by introducing pores.[14,15] The reduced stiffness increases a principal characteristic of electromechanical transduction, the ratio of the induced charge to the applied force. However, the porous inorganics and plastics have relatively small stretches, the low breakdown strength of the air in the pores limits charge density, and the pores and their surfaces provide paths of conduction for discharge. Hard electrets have been made flexible in the form of thin sheets[16−20] and made stretchable in the form of serpentines.[21] The serpentine-shaped electrets achieve both longevity and stretchability. Carbon nanotubes are introduced into an elastomer matrix to enable stretchable electrets.[22] Upon poling, the interfaces between the carbon nanotubes and the elastomer host charges, but do not trap charges, just like a conductor/dielectric interface in a capacitor. The poled composite discharges through the elastomer within hours. Porous elastomers have been studied as stretchable electrets in several papers.[23−26] Only one of these papers reported a lifetime, which went beyond 40 days.[23] This result is intriguing, given the low resistivity of elastomers. The result was unexplained in the original paper. We will discuss this result

Received: April 1, 2020
Revised: May 14, 2020
Published: May 15, 2020

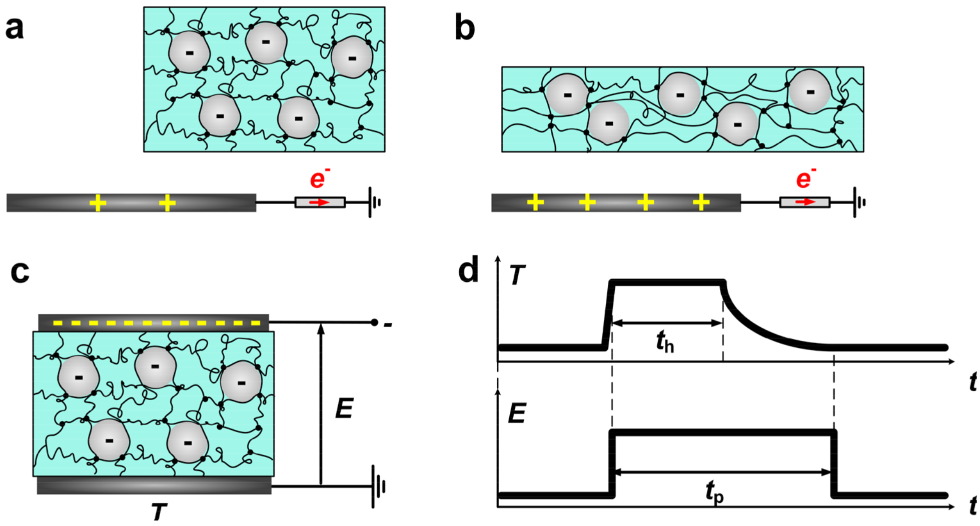

Figure 1. Principle of the long−lived, stretchable electret. (a) A matrix of a dielectric elastomer immobilizes nanoparticles of a hard electret. Upon poling, the electret particles trap charges. A rigid electrode is connected through a resistor to the ground. When the composite approaches the electrode, charges are induced on the electrode, drawn from the ground. (b) When the composite is stretched, the deformation of the elastomer carries the electret particles, changes the amount of induced charges on the electrode, and causes a current through the resistor. (c) During poling, the composite is attached with two electrodes, subject to an intense direct electric field, *E*, in a thermal environment of an elevated temperature, *T*. (d) The composite is kept at a constant elevated temperature by a heat plate for a period of heating time, t_h. After the heat plate is switched off, the composite cools to room temperature over some time. The poling electric field is applied for a period of poling time, t_p, typically until the sample cools to room temperature.

later on the basis of our own findings. Particles of a polymer with molecular dipoles are embedded in a matrix of a dielectric elastomer to enable stretchable electrets.[27,28] This approach can potentially achieve both longevity and stretchability, but the reported electrets discharge substantially within days. In a recent work, droplets of a liquid electret are embedded in a stretchable textile and are shielded by a molecular design to enhance lifetime.[29] This composite electret has demonstrated impressive longevity and stretchability. Block copolymers have also been used as stretchable electrets, where the hard blocks trap charges, and the soft blocks enable stretchability.[26,30] No lifetime of these materials has been reported.

Here we demonstrate a general approach to make long−lived, stretchable electrets. We immobilize nanoparticles of a hard electret in a matrix of a dielectric elastomer (Figure 1a). Upon poling, the volume of the particles and the interfaces between the particles and matrix trap charges. Particles in different locations in the matrix may trap charges of different amounts and different polarities. Charges trapped in the elastomer, if any, are short−lived. Like any electret, the composite electret can be used with no electrode, one electrode, or multiple attached electrodes. Here we illustrate the principle of stretchable electret in noncontact electromechanical transduction. An electrode is connected through a resistor to the ground. When the composite electret approaches the electrode, charges are induced on the electrode, drawn from the ground (Movie S1). The stretchability of the composite electret enables another mode of operation. When the composite electret is stretched, the deformation of the elastomer carries the electret particles, changes the amount of induced charge on the electrode, and causes a current through the resistor (Figure 1b, Movie S2). Both rigid−body motion and deformation of the composite electret induce charges on the electrode. Such a stretchable electret divides the labor of two functions: the particles of a hard electret trap charge for a long time, and the matrix of a dielectric elastomer enables stretchability. This approach considerably broadens the choice of materials to enable stretchable electrets. Many inorganics and plastics can be used as particles to trap charges with longevity, and many dielectric elastomers can be used as matrices to enable stretchability.

To illustrate this approach, we use silica nanoparticles (AEROSIL−200 hydrophilic) as the electret and polydimethylsiloxane (PDMS, SYLGARD 184) as the dielectric elastomer. Several methods can be used to pole the composite; here, we adopt thermal poling.[3,4] The composite is maintained at temperature, *T*, using a heat plate for a period of heating time, t_h, and, after the heat plate is switched off, is cooled naturally to room temperature (Figure 1c). A direct electric field, *E*, is applied through the top and bottom electrodes to the composite and is kept for a certain poling time, t_p, typically until the sample cools to room temperature (Figure 1d). The experimental setup of the thermal poling can be found in Figure S1.

We characterize the composite electret using an established method (Figure 2a).[3,4] The electret, thickness, *h*, is attached with a bottom electrode. The top electrode is not attached to the electret. As the top electrode moves down and up, we record the charge flowing between the two electrodes as a function of time, using a charge meter (B&K 2635A) and an oscilloscope (KEYSIGHT DSO1004A) (Figure 2b). The flow of charge is highly reversible. The peak charge is a measure of effective charges trapped in the electret. The two peaks of the same amplitude but opposite polarities represent the contact and the detachment between the top electrode and the electret. The decay of the charge over time corresponds to the *RC* delay of the charge meter, $10/2\pi \sim 1.6$ s, governed by its internal impedance.

The nominal charge density of an electret, *q*, is defined by the peak charge divided by the area of the top electrode. Unless otherwise specified, the PDMS is the 10:1 SYLGARD

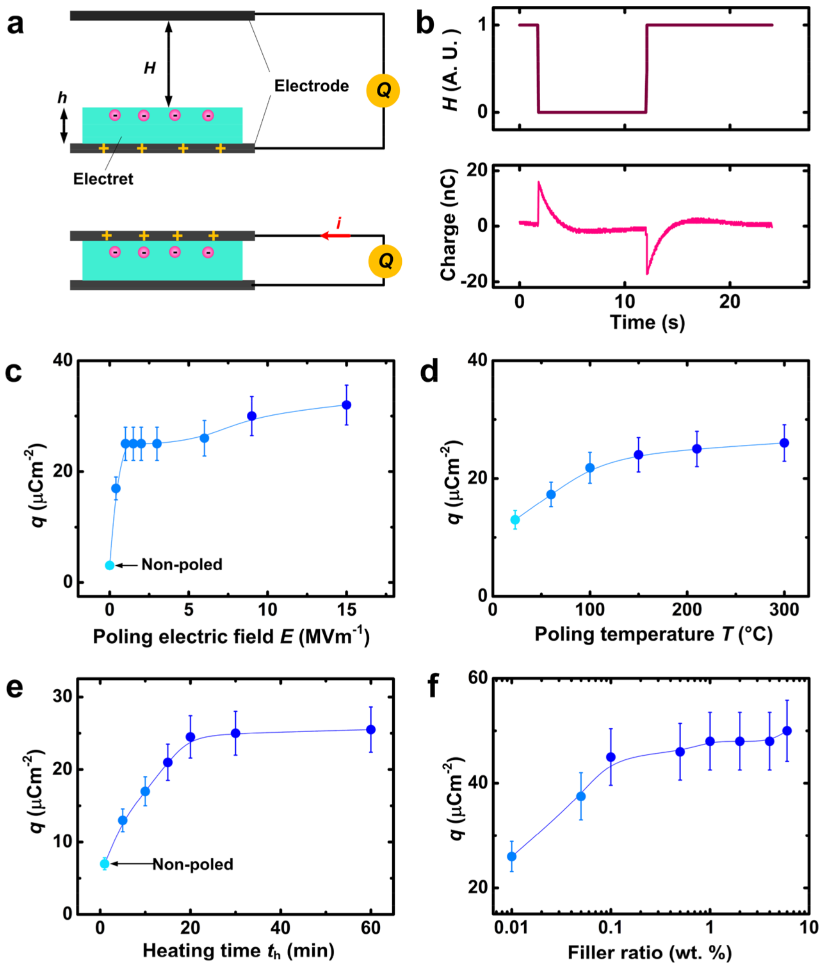

Figure 2. Poling characteristics. (a) Schematic of the experimental setup. (b) When the top electrode moves toward and away from the electret, charges flow through a charge meter, Q, which connects the two electrodes. The nominal charge density, q, as a function of (c) the poling field, (d) the poling temperature, (e) the heating time, t_h, and (f) the filler ratio. The measured nominal charge densities are negative, but we plot the absolute values. All data represent the mean and standard deviation.

184, the silica−to−PDMS weight ratio is 2.0%, the area of the top electrode is 6.25×10^{-4} m^2, t_h = 30 min, and t_p = 120 min.

The nominal charge density q increases with the poling electric field, E, and attains a level insensitive to the further change of the electric field (Figure 2c). To study the effect of poling temperature, we set the poling field, E = 3 MV m^{-1}, and the heating time, t_h = 30 min. When the poling temperature increases, the surface charge density also increases (Figure 2d). We then set the poling field, E = 3 MV m^{-1}, and the poling temperature, T = 210 °C, and measure the nominal charge density as a function of poling time (Figure 2e). We vary t_h and set, $t_p = t_h + 90$ min. When the heating time is sufficiently long, the charge density reaches a plateau. The suitable heating time is determined empirically by the material and the geometry. The nominal charge density increases with the amount of silica in the electret (Figure 2f). We have also obtained similar results with polytetrafluoroethylene (PTFE) particles embedded in PDMS matrices (Figure S2).

The observed "plateau charge density" is understood as follows. The electrical breakdown strength of PDMS is much larger than that of the air.[31,32] Consequently, when the composite is poled and the top electrode is removed, the nominal charge density of the electret is limited by the breakdown of air, not by the breakdown of PDMS. Given the electric breakdown strength of air, $E_B \sim 10^6$ V m^{-1} and the permittivity, $\varepsilon \sim 10^{-11}$ F m^{-1}, the upper limit of the nominal charge density of the electret, $q \sim \varepsilon E_B \sim 10^{-5}$ C m^{-2}. This order of magnitude is consistent with the experimentally observed upper bound.

We measure the nominal charge density of an electret as a function of the storage time after poling. For a given sample, the nominal charge density is measured once a day in the first 7 days and then in days 15, 30, and 60. Between the measurements, the sample without electrode is sealed in a plastic bag at room temperature. (We will measure charges of the same samples over months and years when laboratories open after the COVID−19 pandemic.) The charge density depends on the amount of silica nanoparticles, and remains nearly constant during the period of 60 days (Figure 3a). When the filler ratio is beyond 2.0%, the mechanical properties of the composite become poor, and the materials can hardly be handled (Figure S6). We then fix the silica−to−PDMS weight ratio as 2.0% and change the cross−linking density of PDMS. The capability of charge retention decreases when the cross−linking densities of PDMS decreases (Figure 3b). We surmise that the elastomers with sparse cross-links are unable to immobilize the electret particles. We estimate the mesh size at various cross−linking densities and compare them with the size of a silica particle (see details in the Supporting Information). We cyclically stretch the electret with amplitude λ = 1.2 and

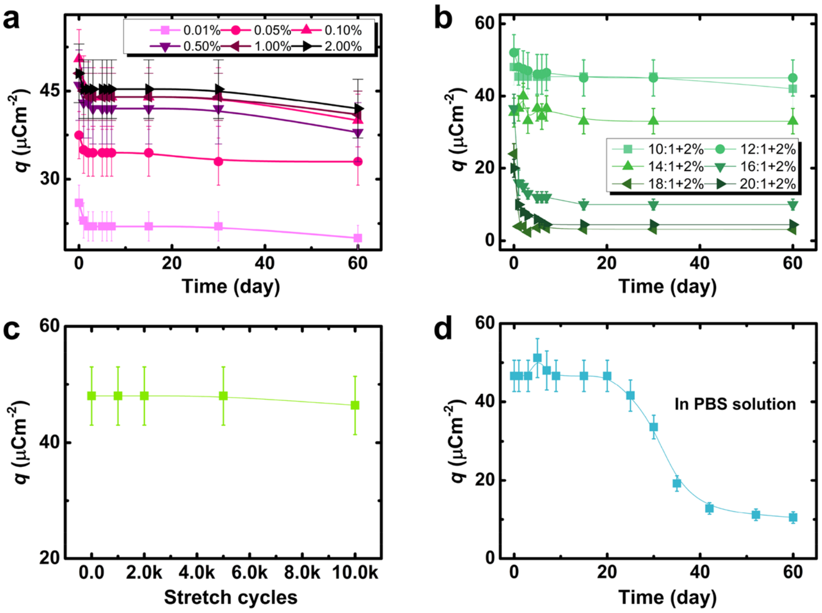

Figure 3. Charge retention. Unless otherwise specified, the experiments are conducted using a 1 mm thick PDMS SYLGARD film, with test area 6.25×10^{-4} m^2, external electric field, $E = 3$ MV m^{-1}, heating temperature, $T = 210$ °C, and heating time, $t_h = 30$ min. (a) Charge retention of PDMS SYLGARD 10:1 with different filler weight ratios. It is seen that a larger filler weight ratio gives higher charge density. (b) Charge retention of the 2.0% weight ratio of fillers and different cross−linking densities of PDMS SYLGARD as 10:1, 12:1, 14:1, 16:1, 18:1, and 20:1. (c) Charge stability of PDMS SYLGARD 10:1 with the 1.0% weight ratio of fillers after cyclic stretch. (d) Charge decay of PDMS SYLGARD 10:1 with the 1.0% weight ratio of fillers into phosphate−buffered saline solution (150 mL) at room temperature. All data represent the mean and standard deviation.

frequency 2 Hz. After 10^4 cycles of stretch, the charge density is still 85% of the initial−poled value (Figure 3c).

Stretchable electrets are promising in biological applications due to the reduced mechanical mismatch between electrets and soft living tissues. To test the lifetime of the stretchable electrets in biofluidics, we soak the 1 mm thick electret in the 1× phosphate−buffered saline (PBS) solutions and measure its nominal charge density as a function of time (Figure 3d). The charge density remains constant for 20 days and then starts to decrease. It reaches another steady state after 40 days. The ions in PBS may diffuse into the PDMS, accumulate at the PDMS/silica interfaces, and neutralize the electret. The discharge of the electret soaked in the ionic solution indicates the need for an elastomer matrix with a low conductivity of the charge carriers present in the environment. The discharge can also be retarded by using a stretchable seal.[33,34] A recent paper has reported the impedance of several elastomers after soaking in the PBS solution.[35] It is likely that some elastomers may have markedly low conductivity than PDMS, and can be developed as more suitable matrices or seals of stretchable electrets. In addition, factors such as hydrolysis[36] and other chemical degradation may also limit the longevity of the stretchable electret. Using long−term stable elastomers in ionic solution (e.g., fluorinated elastomers) as an elastomeric matrix could increase the longevity of the stretchable electrets in biological applications.

Like any electrets, stretchable electrets enable both contact and noncontact electromechanical transduction. Furthermore, stretchable electrets induce electrical signals by both rigid−body motion and deformation. For a stretchable electret attached with one or multiple electrodes, we use carbon grease (Digi−key 846) as the stretchable electrode. The stretchable electret is sealed in 0.5 mm thick Ecoflex (00−30) at both the bottom and top.

The electret can be used as a stretch sensor. A poled PDMS SYLGARD 10:1 film, dimensions 2.5 cm × 4.0 cm × 1 mm, is attached with the carbon grease electrode at the bottom and is uncovered by the electrode at the top. The sealed electret is cyclically stretched to amplitude $\lambda = 1.1$ (Figure 4a) and $\lambda = 1.3$ (Figure 4b) by hands in gloves, and the induced charge flow is recorded over time. The change in the amount of induced charge on the electrode increases with the stretch. The peak-to-peak change is 0.96 nC for $\lambda = 1.1$ and 3.48 nC for $\lambda = 1.3$. It is seen from the linear electro-mechanical response that the stretchable electret could be a sensitive and self-powered stretch sensor.

The stretchable electret can also be used as a pressure sensor (Figure 4c). The pressure is applied cyclically by a finger in glove to an area ∼1 cm^2. In each cycle, the pressure is kept for 2 s before release. The applied pressure sensor also illustrates a high sensitivity: the flow of charge (Figure 4d) is as much as ∼2 nC within a finger load amplitude.

We further demonstrate noncontact stretch sensing (Figure 4e). A poled PDMS SYLGARD 10:1 film, dimensions 2.5 cm × 8.0 cm × 1 mm, is used without any attached electrode. An electrode is made of an aluminum film of 3.0 cm × 5.0 cm, placed 1.5 cm below the stretchable electret. The left end of the electret is aligned with the right end of the electrode. During the stretch, the right end of the electret is fixed, and the left end is stretched with various stretches. To prevent the random electrostatic charge noises from dielectrics such as gloves, the left end of the electret is stretched with a grounded metal. The amount of induced charge depends on the stretch (Figure 4f).

We have also poled several other elastomers and measured their nominal charge densities over storage time (Figure S3). Polybutadiene (BR), Ecoflex (00−30), and VHB (3 M 4910) discharge within days. These findings confirm that elastomers

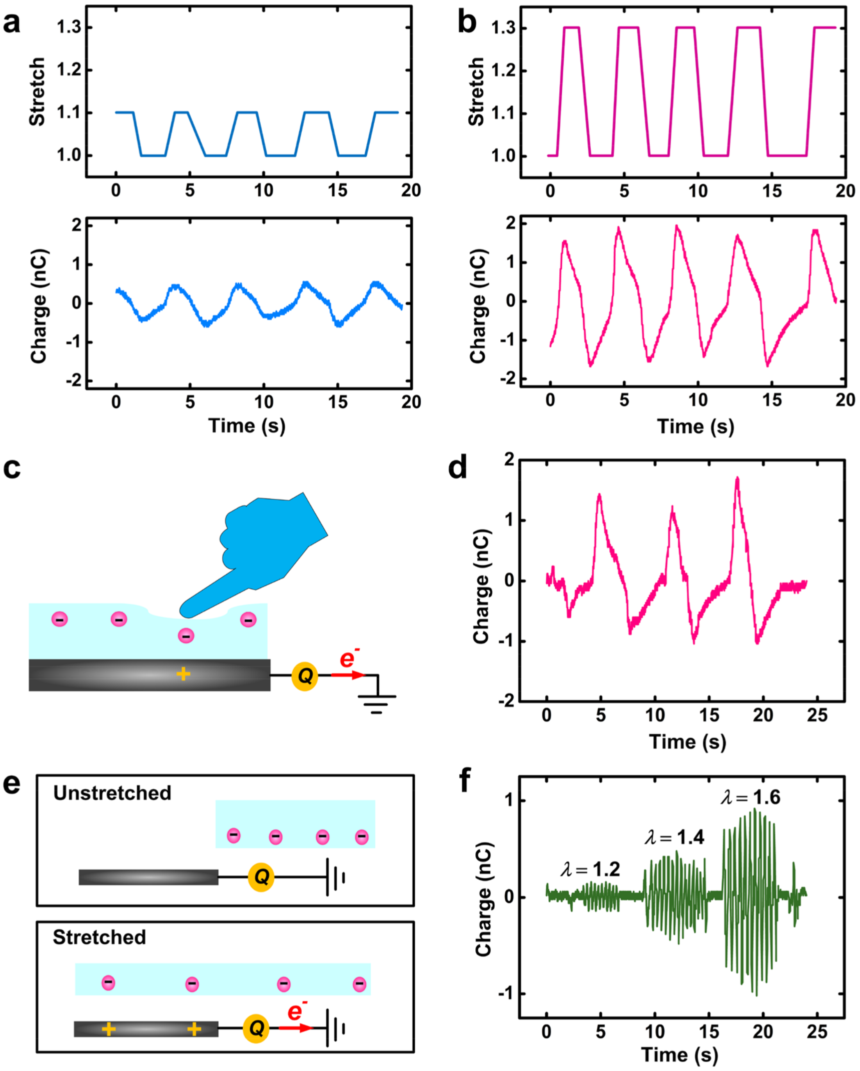

Figure 4. Electromechanical transduction. (a) Induced charge when a sample is cyclically stretched to the amplitude of (a) λ = 1.1 and (b) λ = 1.3. (c) Schematic of the sensing of the compression due to a finger. (d) Charge flow as a function of time during finger compression. (e) Schematic of noncontact stretch sensing. The distance from the electret to the electrode is 1.5 cm. (f) Induced charge as a function of time during noncontact stretch. Stretch with multiple amplitudes are applied as λ = 1.2, λ = 1.4, and λ = 1.6.

by themselves are short–lived electrets. A cross–linked BR containing silica nanoparticles retains charges, but the silica–free cross–linked BR discharges within days, just like un–cross–linked BR and other measured elastomers.

The PDMS SYLGARD 184 precursor contains some amount of silica.[37] To confirm the effect of silica nanoparticles, we make a silica–free PDMS, and compare it with PDMS SYLGARD 10:1 without AEROSIL silica, and PDMS SYLGARD 10:1 with 2.0% AEROSIL silica (Figure S4). The nominal charge densities of the three materials are 3×10^{-6} C m^{-2}, 2.6×10^{-5} C m^{-2}, and 4.8×10^{-5} C m^{-2}. The values remain nearly constant over 60 days. These findings provide an explanation for the report that a porous elastomer after poling retains charges beyond 40 days.[23] The elastomer used in that paper was SYLGARD 184, which contains some amount of silica.

We have demonstrated the stretchable electrets using thermal poling. Other methods of poling can also be used. For example, corona poling may enlarge the nominal charge density[3] and lower poling temperature, which prevents the elastomer from degradation.[38,39] Even in thermal poling, the poling electric field may be increased by one or 2 orders of magnitude to further enlarge the nominal charge density.[31,32,40] The breakdown strength of elastomers is the limit of initial-poled charge density, and the breakdown strength of the storage environment (open air in this work) is the limit of charge retention. The poled charge generates an electric field in the environment and may cause the breakdown of the environment, reducing charge density.[5] We have adopted the contact–detachment procedure to measure charges trapped in an electret. This procedure may induce the breakdown of air and reduce the nominal charge density.[3] Further results are presented in Figures S5–8.

In our experiments, the poling electric field is set at 3 MV m^{-1}, within the breakdown of open air, and the poling temperature is set at 210 °C so that PDMS will not suffer thermal degradation. These conditions are below the values commonly used to pole silica (500 MV m^{-1} and 400 °C).[38,39] It is likely that the charges are trapped on the silica–PDMS interfaces, rather than in the volumes of the silica nanoparticles. It is conceivable that, with suitable hard electrets, dielectric elastomers, and poling methods, charges can be trapped in the volumes of the hard electret particles. In principle, this approach should enable stretchable electrets to achieve the same longevity as the hard electret itself. On the other hand, the enormous total area of the interfaces between particles and the elastomer may itself be an ideal trap for charges. These considerations provide fertile grounds for further research. In addition, the particles can be distributed in the matrix homogeneously or with a pattern of design. Patterned charges may provide interesting properties and extend the scope of applications of the stretchable electrets. Examples of patterned charges include electrophotography and electrostatic assembly.[7]

In conclusion, we have described a general approach to developing long−lived, stretchable electrets. A matrix of dielectric elastomer immobilizes nanoparticles of hard electret when the mesh of the polymer matrix is smaller than the particles, or when the elastomer adheres to the particles. The elastomer matrix enables stretchability, and the electret particles enable longevity. The approach can recruit numerous materials in the struggle to resolve the longevity−stretchability conflict. Many dielectric elastomers can be used as matrices, and many long−lived electrets can be used as particles. Both the particle volumes and the particle/matrix interfaces may trap charges of high density and longevity. The enormous diversity expands property space, enables unusual combinations of attributes, and opens doors to the applications of stretchable electrets in engineering and medicine. The long−lived, stretchable electret opens immediate opportunities in stretchable electronics and soft robots. Potential applications include stretchable and transparent touchpads and noncontact electromechanical transducers. This work once again highlights an unmet challenge in the broad field of stretchable electronics: the development of stretchable seals.

Experimental Section. *PDMS Containing Silica.* The PDMS is made using SYLGARD 184, and silica nanoparticles are AEROSIL−200 hydrophilic, with a surface−to−mass ratio of 200 $m^2\ g^{-1}$. Silica particles are weighed in a hood and mixed into hexane (0.1 g into 10 mL). The ratio of silica−to−PDMS SYLGARD is by weight. The silica−hexane mixture is stirred by a vortex mixer (VWR International as 3200 rpm) for 2 min and then added to the precursor of PDMS SYLGARD. This mixture is stirred by the same vortex mixer for 2 min and degassed in a sonicator (Branson 1800) for 5 min. Then the composite is poured into a plastic dish. Hexane evaporates, and PDMS SYLGARD cures at room temperature for 48 h in a hood.

Silica−free PDMS. Part A is made by dispersing the cross-linker (HMS−301(68037−59−2, gelest)) in DMS−V35 with 10 wt %. Part B is made by dispersing a Pt catalyst (SIP6831.2(68478−92−2, gelest)) in DMS−V35 with 0.15 wt %. Parts A and B are mixed in the weight ratio of 1:3. The mixture is stirred by a vortex mixer for 30 s, poured into a mold, degassed in a vacuum chamber for 10 min, and cured in a chamber at 65 °C for 24 h.

Thermal Poling. The temperature is maintained by a heat plate, and the poling electric field is applied using a high voltage source (Trek 610 E) with a bias output. Poling electrodes are copper films with 6.0 cm × 2.5 cm × 0.5 mm.

Electrical Measurement. The charge is measured by a charge amplifier (B&K−2635A), whose amplification ratio is set to be 0.1 mV pC^{-1}. An oscilloscope (KEYSIGHT DSO1004A) is connected in series to detect the charge flow. The DC coupling mode of the oscilloscope is used for Figure 2b as an illustration, and the AC coupling mode of the oscilloscope is used in the measurements of peak values. The measuring electrodes are copper−zinc alloy films with 10.0 cm × 2.5 cm × 1.0 mm. The measured area is $6.25 \times 10^{-4}\ m^2$.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.nanolett.0c01434.

Scheme of thermal poling on stretchable electrets; nominal charge density of PTFE−PDMS composite electret; charge measured as a function of time in multiple material systems; charge retention of the PDMS; nominal charge density of large size silica−PDMS matrix; photo image of the silica−PDMS composite of (a) 6.0% and (b) 10.0% filler mass ratio; sign of charge density; piezoelectric response of stretchable electret (PDF)

Movie S1 (MP4)

Movie S2 (MP4)

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Author Contributions

The manuscript was written through the contributions of all authors. All authors have given approval to the final version of the manuscript.

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This work is supported by NSF MRSEC (DMR−14−20570). X.Y. is funded by the National Natural Science Foundation of China (21905077). X.X.Y. was a visiting student at Harvard University, supported by the China Scholarship Council.

ABBREVIATIONS

PDMS, polydimethylsiloxane; PBS, phosphate-buffered saline

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