ABSTRACT: In this study, a new preparation method is developed to include thermochromic complex ions in polydimethylsiloxane (PDMS) as a stretchable composite. Ethylene glycol (EG) droplets down to the nanometer scale were dispersed using a reverse micelle method to form a stable suspension in PDMS precursor solution. After curing, the EG nanodroplets were well encapsulated in the cured PDMS elastomer. The EG/PDMS composite exhibited great stability after thermal heating at 100 °C for 2 hours. The deformable liquid droplets helped maintaining the composite structures under severe stretching conditions, and thus the stretched composite exhibited great transparency without any fractures or delamination. Ionic dyes, such as methylene blue and Congo red, can be added in the EG droplets to color the composite. Moreover, complex ions with thermochromic properties can also be used in the composite. Upon thermal heating, the reconfiguration of the complex ions in the liquid dispersed phase led to obvious color changes, and the color remained unchanged up to 50% tensile strain after more than 1000 stretch cycles.

INTRODUCTION

With the increasing need for wearable electronic devices, stretchable polymer substrates have drawn much attention recently. Among many stretchable polymers, polydimethylsiloxane (PDMS) is widely used for stretchable applications due to its great elasticity, optical transparency, nonflammability, and chemically inert nature. With the addition of nanofillers, such as carbon nanotubes, graphene, and silver nanowires, in PDMS, many studies have demonstrated the flexibility and versatility of PDMS composites. However, due to the incompatible elasticity between solids and PDMS elastomers, PDMS composites with solid fillers exhibit poor long-term low durability under stretching conditions. After multiple repetitive stretching cycles, separation between the solid fillers and PDMS occurs and results in material delamination or local fractures. To resolve this problem, in a recent study by Guan et al., a liquid filler is used to form deformable liquid packets within PDMS. With great flexibility of liquids under large-scale deformation conditions, the composite exhibits stable electrochemical characteristics under extensive stretching conditions.

On the other hand, it is difficult to prepare uniform and well-encapsulated droplets in the PDMS composite due to the hydrophobicity of PDMS. Without complete encapsulation, liquid fillers potentially evaporate and leave cavities in the composite. Moreover, diffusion of the dye molecules out of the PDMS matrix occurs and leads to undesired porous composites. Direct mixing of ionic solutions in PDMS usually results in aggregation, delamination, or even phase separation and thus leads to reduction in mechanical or stretchability of the composites. Moreover, because of the disparity in refractive indices, addition of either liquid or solid fillers usually leads to severe light diffraction or scattering and thus results in low transparency of PDMS composites. To reduce both...
aggregation and light scattering effects, i.e., in order to maintain transparency and elastic properties, a general way is to reduce the filler size into nanometer scale.\textsuperscript{19} One of the commonly used dispersion methods for stable nanometer droplets is the emulsion process.\textsuperscript{20,21} Nanoemulsions are kinetically stable liquid-in-liquid dispersions with droplet sizes in the nanometer scale.\textsuperscript{22} High-energy methods are usually needed to prepare nanoemulsions, including high-pressure homogenization, ultrasonication, phase inversion temperature, and the emulsion inversion point method.\textsuperscript{22–25} Besides preparation methods for nanoemulsion, surfactants also play a very important role.\textsuperscript{26,27} Surfactants can significantly reduce the interfacial tension between the two liquids to improve the miscibility.\textsuperscript{27} In a recent study,\textsuperscript{28} water droplets in the micrometer scale were dispersed and stabilized in a PDMS precursor solution with a surfactant. After the thermal curing process, water evaporates and leaves a porous elastic structure to create pressure-sensitive devices. These studies show that surfactant addition can be quite efficient for dispersing liquid fillers in the PDMS elastomer.

In this study, a new formulation method is developed to include ionic materials for elastic and transparent PDMS composites. First, liquids with refractive indices close to PDMS are chosen as the dispersed phase. After surfactant addition, a high-energy method is then used to create droplets of the nanometer scale for enhancement in both dispersion stability and transparency of the composite material.\textsuperscript{22,28,30} Ionic dyes, methylene blue and Congo red, will be mixed in the liquid dispersed phase and embedded in the PDMS composite to understand the microstructures of the encapsulated liquid droplets in PDMS. The optical properties of the composite materials can also be tested to evaluate the effectiveness of this composite formulation method in droplet encapsulation, which helps maintaining the composite structures under repeating stretching cycles. Furthermore, the ionic materials are well protected in the PDMS film at different conditions such as low and high pH. Finally, a complex ion with thermochromic properties\textsuperscript{1,31} is mixed in the liquid dispersed phase, and the color change performance of the composite film under stretching or thermal cycles will be also tested to show the possibility of using this material for stretchable thermochromic applications.

## RESULTS AND DISCUSSION

### Dispersed Phase Material Selection

Ethylene glycol mixing with PDMS can result in a stretchable composite with great transparency. To fabricate homogeneous stretchable composites with ionic compounds, it is necessary to mix polar solvents with PDMS, and the mixture was allowed to remain in the liquid state after the curing process. Several commonly used solvents are listed in Table 1. Based on the curing temperature of the PDMS precursor, 80 °C, only water and ethylene glycol can be used here without total evaporation. As shown in the first column of Figure 1, obvious phase separation is observed due to the polarity disparity between the two solvents and PDMS precursors. After putting in a sonication bath for 10 min, uniform gel-like solutions are obtained (the second column in Figure 1), indicating the existence of droplets at the micrometer scale.\textsuperscript{33} To further enhance the uniformity and stability of the PDMS/solvent mixtures, ultrasonication at high energy is used to reduce the droplet size.\textsuperscript{22,30} After ultrasonication for 15 min, the PDMS/water mixture remains as a milky gel-like solution, but the PDMS/EG mixture becomes a translucent solution, indicating that the droplets of ethylene glycol are made into sizes in the range of visible light wavelength.\textsuperscript{30} Moreover, because the refractive index of EG is very close to that of PDMS (Table 1), the less light scattering in the EG/PDMS mixture leads to better transparency.\textsuperscript{35} The EG/PDMS and water/PDMS mixtures after ultrasonication are cured thermally at 80 °C for 1 h. As indicated in the literature,\textsuperscript{28} the PDMS/water mixture shows porous structures due to water evaporation in the curing process. On the other hand, ethylene glycol has a much higher boiling point and the EG/PDMS mixture becomes a uniform transparent film. Considering the transparency and evaporation, ethylene glycol is selected as the liquid disperse phase in the following sections.

The mixture, however, is not quite stable at high EG content. As shown in Figure 1(b), phase separation happens after the EG/PDMS mixtures set still for several hours. Moreover, after curing, the EG liquids are found over the cured PDMS films (Figure 1(b)), indicating strong phase separation in the heating process. After washing, a maximum EG/PDMS composite formulation method in droplet encapsulation, which results in a highly porous PDMS film (Figure 1(b)), indicating strong phase separation after curing.

### Table 1. Comparison of Refractive Indices and Boiling Points for Solvents and PDMS

| Solvent         | Refractive Index | Boiling Point (°C) |
|-----------------|-----------------|--------------------|
| Water           | 1.3333          | 100                |
| Ethanol         | 1.3590          | 78                 |
| Acetone         | 1.3592          | 56                 |
| Ethylene glycol | 1.4233          | 198                |
| PDMS            | 1.4260          | NA                 |

![Figure 1](https://dx.doi.org/10.1021/acsomega.0c00668)

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ratio of 0.1 is found. Therefore, addition of surfactants is necessary to stabilize the interfaces between the EG and PDMS precursor for better dispersion formation.

**Surfactant Addition for Droplet Size Reduction.** To improve the dispersion of EG in PDMS, the surfactant was added into the EG/PDMS mixture. The surfactant can significantly reduce the surface tension of the solution and can also reduce the interfacial tension between the two liquids to improve miscibility. In this study, IGEPAL CO-520 was selected as the emulsifier because of its good solubility in PDMS. Figure 2 shows the interfacial tension measured by drop shape analysis at various surfactant concentrations in EG. The addition of CO-520 in EG leads to a dramatic tension decrease at the EG/PDMS interface and the variation of interfacial tension $\gamma$ can be described by a typical Langmuir isotherm curve:

$$\gamma = \gamma_0 - \Gamma_m aRT \ln \left( 1 + \frac{C}{a} \right)$$

where $\gamma_0$ is the original interfacial tension without surfactant, $\Gamma_m$ is the maximum surface density of the surfactant, $R$ is the gas constant, $T$ is the temperature, $a$ is the adsorption constant, and $C$ is the surfactant concentration. The best fitted curve indicates $a = 0.00335$ M, and $\Gamma_m$ = 1.52 x $10^{-6}$ mol/m$^2$ or a polar head group area of ~1.1 nm$^2$, which is close to the size of CO-520. The tension reaches a plateau when the concentration is larger than 0.01 M, indicating that a critical micellar concentration (cmc) is reached. To disperse EG in PDMS more effectively, a concentration of 10 cmc is used afterward.

With the addition of surfactants, the EG droplets can be easily dispersed in the PDMS precursor solution and yield in a nearly transparent solution after ultrasonication. To further increase dispersion stability, a high-energy method is used to reduce the EG droplet sizes. Because the mixture has a large viscosity (~ 3.5 Pa s), it is quite difficult to measure the size using a regular dynamic light scattering tool. Thus, an ultracentrifuge method is used to measure the droplet size, and the size distribution of the droplets is summarized in Figure 3(a). With a longer ultrasonication process time, the EG droplet size in the dispersed phase decreases, and the mixture becomes more translucent. Upon sonication over 15 min, the droplet size is significantly reduced to 545 nm, and a nearly transparent solution is obtained, indicating an effective enhancement in the droplet dispersion.

To verify the droplet sizes, the mixture is thermally cured, and the cross-linked samples are examined by SEM. As shown in Figure 3(b,c), scattered spherical cavities, or the space occupied by EG in the reverse micelles, are observed in the cross-sectional SEM images of cured thin films, indicating that the EG droplets are well embedded in PDMS. Moreover, the size of these cavities also becomes smaller as the sonication time increases (Figure 3(a)), and they exhibit the same size as those measured by sedimentation methods. However, because of the limitation of SEM on polymeric materials, it is difficult to observe the cavities at the nanometer size. Nevertheless, the droplet sizes inferred from the sedimentation method are verified and acceptable.

**Optical, Mechanical, and Chemical Properties.** The cured EG/PDMS thin films show great transparency, and the embedded EG droplets are well encapsulated for a long period of time. EG has a refractive index close to that of PDMS. Thus, after embedding in PDMS, the small EG droplets result in light scattering and leads to a transparent film (Figure 4(a)). The UV spectra of the EG/PDMS films show that a high transmittance of 99% can be achieved. The EG/PDMS films also exhibit a similar elastic behavior as the pristine PDMS (Figure 4(b)). Liquid-type materials have poor ductility, but a liquid filler can form deformable liquid packets within PDMS. It can avoid material delamination or local fractures. Since we added EG in the PDMS composite, the liquid additive would reduce the elongation for 10% for 10 wt % EG was added. However, it still has high strain elongation above 0.5 that is enough for most conditions. After stretching or baking, the films still maintain the transparency, indicating the great encapsulation of the EG droplets. To understand the thermal stability of the EG encapsulation, the cured EG/PDMS composite was heated at 120 °C to test the evaporation rate of the encapsulated EG droplets. As shown in Figure 5, there is nearly no EG evaporation (< 0.1 wt % loss) after baking for 2
h, indicating that the EG droplets are well concealed in the PDMS after curing. This demonstrates that the PDMS composites in this work could remain stable at 120 °C and can be used for coloration application.

**Coloration with Ionic Dyes.** Ionic dyes can be easily added in the encapsulated EG droplets for coloration applications. To demonstrate the feasibility, two ionic dyes, Congo red and methylene blue, are dissolved in EG before the suspension/sonication process. As shown in Figure 6(a), both ionic dyes are well dispersed, and the cured composite films show uniform red or blue colors with great transparency, indicating the well-dispersed EG droplets in PDMS. One can also change the color by varying the concentration of dyes (Figure 6(b)). Moreover, the color of Congo red switches to blue at low pH, but the composite films with Congo red remain red in color while soaked in acidic solution (Figure 6(c)), showing that the dyes in EG droplets are well insulated in the EG/PDMS composite films. This shows their potential application in optical devices for stretchable applications such as wearable electronics.

**Thermochromic Performance.** The well encapsulated EG droplets in PDMS can be further used to host thermochromic materials. Nickel complex ions, a thermochromic material 31, is adopted here as an example. Transition metal salts, nickel halides, can form complex ions in ethylene glycol. These complex ions transfer from tetrahedral to octahedral configurations during repeated heating−cooling cycles and lead to significant changes in the color of the complex ions. As shown in Figure 7(a), the composite with NiCl/EG/PDMS has a color of pale green at room temperature, which transfers to dark turquoise at 90 °C. The thermochromic properties of the composite remain unchanged after repetitive stretching. Due to deformability of liquid EG droplets, which host the thermochromic complex ions, the colors of the composite remain quite stable under stretching conditions. As shown in the inset pictures in Figure 7(b), no obvious color difference is observed when a tensile strain is applied to the composite film. By using a colorimeter, the colors at different strains are quantified as RGB values. The colors have a little variation of less than 1% even at a strain of 50%. The optical density (OD) can be calculated at a wavelength of 700 nm. At this wavelength, there is an obvious absorbance difference between low (25 °C) and high (90 °C) temperatures.32

In addition, the color of the thermochromic composite is quite stable and remains almost the same after 1000 stretching cycles with a peak strain of 50% (Figure 8(a)). Moreover, because of its good thermal stability, the thermochromic performance of the composite is quite stable. As shown in Figure 8(b), the color contrast (O.D. value) remains almost...
the same after 1000 heating and cooling cycles, indicating that the encapsulated EG droplets in PDMS remain in the liquid state, allowing ion reconfiguration for color changes, even during temperature variation cycles.

CONCLUSIONS

In this study, by using EG as the dispersion phase, a reverse-micelle emulsion with a high-energy method is developed to disperse ionic fillers in PDMS. Ethylene glycol, with a close refractive index as PDMS, is dispersed using a reverse micelle method to form a stable suspension in PDMS precursor solution. After curing, the EG nanodroplets are well encapsulated in the cured PDMS elastomer. The EG/PDMS composite exhibits great stability after thermal heating at 100 °C for 2 hours. The deformability of the encapsulated EG liquid droplets helps maintaining the composite structures under severe stretching conditions. Thus, the EG/PDMS composite exhibits great transparency without any fractures or delamination under stretching conditions. Ionic dyes, such as methylene blue and Congo red, can be added in the EG droplets to color the composite. Moreover, complex ions with thermochromic properties are also used in the composite. Upon thermal heating, the reconfiguration of the complex ions in the liquid dispersed phase led to obvious color changes, and the color remained unchanged up to 50% tensile strain after more than 1000 stretch cycles. In summary, this new emulsion formulation provides a new synthetic method for transparent composites and can be further extended for many other stretchable composite applications.

EXPERIMENTS

Materials. IGEPAI CO-520, choline chloride, Congo red, and methylene blue were purchased from Sigma Aldrich, USA. NiCl$_2$·6H$_2$O was purchased from Ferak, Germany. Ethylene glycol (EG) was purchased from J.T. Baker, USA. The PDMS precursor and curing agent (Sylgard 184) were purchased from Dow Corning, USA. All chemicals were used as purchased without further purification.

Fabrication of Composite Films. To prepare ionic material composite films, 0.05 g of methylene blue (or Congo red) powder was added to 20 g ethylene glycol at room temperature and stirred until dissolved. The solution was then added into the PDMS precursor at a weight ratio of 1:4 and stirred vigorously. Then, the curing agent was added to this solution at a weight ratio of 1:10 to the PDMS amount. Vigorous stirring and de-foaming were also performed by using a planetary centrifugal mixer. To prepare thermochromic films, 7 g of choline chloride powder was mixed with 6.2 g ethylene glycol at room temperature. NiCl$_2$·6H$_2$O (0.663 g) was then added into the liquid mixture. After complete mixing, the solution was added to the PDMS precursor (with a weight ratio of 1:4) and stirred. Then, the curing agent was added to the PDMS solution (with a weight ratio of 1:10) to obtain a pale green colloidal dispersion. The dispersion was further emulsified using an ultrasonic homogenizer (Ruptor 4000, Omnin) at a power of 200 W for 20 min. The solutions were poured into a glass Petri dish and dried in an oven at 80 °C for 1 h to obtain composite films.

Characterization. Absorbance measurements were carried out at 25 °C using a spectrophotometer (V-670, Jasco), which was equipped with a pulsed near infrared light-emitting diode (865 nm) as a light source. The morphology and microstructures of the samples were observed using a scanning electron microscope (SEM, Nova NanoSEM 230). Color analysis was performed using a colorimeter (Precision colorimeter NR110, 3nh). The weight loss of the composite films was measured by using an electronic moisture balance (MOC-120H, SHIMADUZ). Stirring and de-foaming processes were performed using a planetary centrifugal mixer (ARE-310, Thinky). The droplet size calculation was performed using a Lumisizer (LUMiSizer 6112, LUM).
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Notes
The authors declare no competing financial interest.

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