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Electrorheological Elastomers

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Abstract

Electrorheological elastomers (EREs) are smart elastomers with tunable dynamic properties by applying electric field. They are composed by polarizable particles dispersing within elastomers matrix. In comparison with electrorheological fluids, the particles in EREs do not undergo aggregation and sedimentation. Without the requirement of large-sized electromagnetic coils, the smart devices based on EREs have simpler structures than those based on magnetorheological (MR) elastomers. The unique properties and merits of EREs give rise to their bright future in various fields. During the past decades, great progress has been made by scholars around the world. In this chapter, the conception, preparation, physical mechanisms, classification, development, influence factors, and applications of electrorheological elastomers is reviewed.

Keywords: electrorheological (ER), elastomers, dynamic properties, smart materials, storage modulus

1. Introduction

Smart materials are reactive materials. Their properties can be changed by exposure to stimuli, such as magnetic and electric fields, temperature, pH value, and moisture [1, 2]. Shape memory alloys and polymers [3], piezoelectric materials [4], magnetostrictive materials [5], and magnetorheological fluids and elastomers [6] are typical smart materials. Electrorheological (ER) materials have been considered as another promising smart material. The pioneering example of ER materials is electrorheological fluids, whose rheological properties, such as viscosity and yield stress, can be reversibly changed by applying external electric field. Electrorheological fluids are usually composed with polarizable particles dispersing within an insulating carrier liquid [7–9]. Under an applied electric field, ER fluids will transfer from liquid-like state to semisolid-like state. Compared with their magnetorheological counterparts, MR fluids, ER fluids present faster response (within a millisecond) and simpler design, but

their field-induced yield stress cannot compete with MR fluids, and much higher electric field (several kV/mm) is required for notable changes. Besides, just like MR fluids, the aggregation of particles and the leakage of carrier liquid are two crucial shortcomings that prevent ER fluids from widely applications [10, 11].

To overcome those shortcomings, electrorheological elastomers (EREs) have developed in recent years. Typical ER elastomers consist of polarizable particles and elastomer matrix [12, 13]. According to the distribution of polarizable particles in the elastomer matrix, two kinds of ER elastomers can be distinguished as isotropic EREs and anisotropic EREs. The particles in the isotropic EREs are randomly distributed without order, while they are form ordered structures as chains or columns in the anisotropic ones. To get the anisotropic EREs, an electric field has to be applied during the crosslinking or curing or vulcanization process [14]. The ordered structures of particles are locked in the matrix. Compared with ER fluids, which are used in their postyield region, EREs are used in the preyield region. The viscoelastic properties, such as storage modulus and loss modulus, are electric field responsive, as shown in **Figure 1**. The most outstanding advantage of EREs with respect to ERFs is that the particles in ER elastomers do not undergo sedimentation. EREs also present obvious merits in comparison with magnetorheological elastomers (MREs). The EREs are driven by electric field rather than magnetic field, which is much easier to apply without the need of large-sized electromagnetic coils [15–17]. Therefore, the EREs can be used to design smart devices with simple structure and low weight. Due to those advantages, ER elastomers have been extensively studied in recent years and have presented promising future in applications.

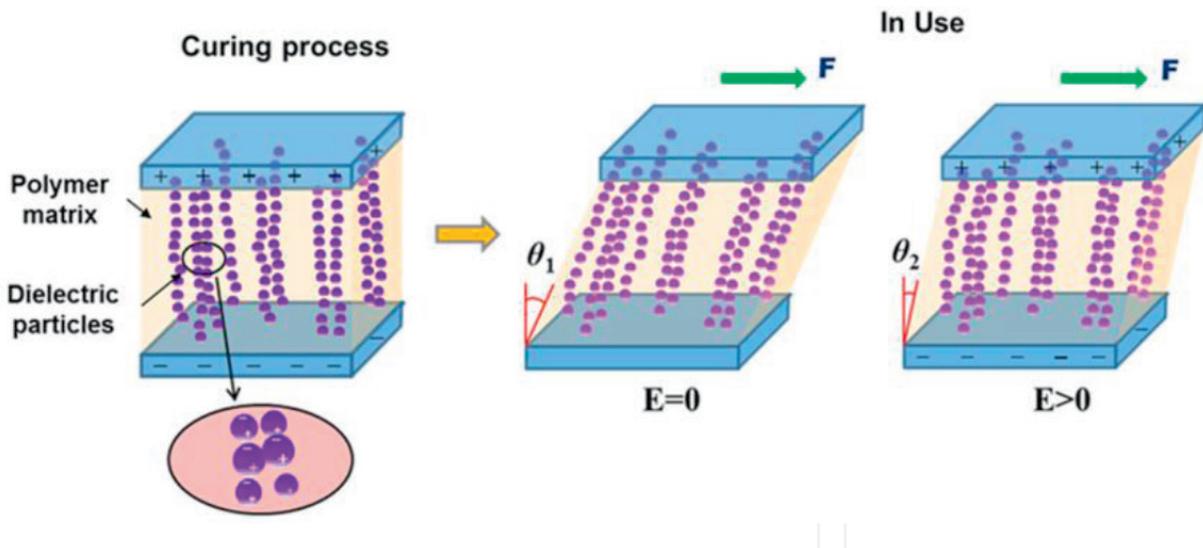


Figure 1. Schematic of EREs in curing process and in use.

2. Formulations and preparation of EREs

EREs are generally consisted of dielectric or semiconducting filler particles and polymer matrix. To prepare the EREs, filler particles are dispersed in the unvulcanized polymer and then locked in the matrix with the curing process. As mentioned above, the EREs will have isotropic structures if the particles randomly distribute in the matrix. In this case, the particles are dispersed at random and cured in the polymer without applying electric field. By curing the mixture of

polymer and particles in the presence of an electric field, field-induced interaction between adjacent particles promote them to form anisotropic microstructures like chains or columns aligned paralleling to the field direction. These structures will be retained in the matrix and that have been proved to be helpful for improving the field-dependence of EREs' mechanical performance when they are exposed to extra electric field. SEM photographs have been used to show the presence of chain structures of particles in the EREs, besides, wide-angle X-ray scattering technique is also used to observe the anisotropic structures in clay/gelatin EREs [18, 19].

For anisotropic EREs adopting gels or rubbers as matrix, the mixtures are cured in the mold made of glass or other insulating materials with two electrodes, which is shown as **Figure 2**. The electric field should be maintained during the entire curing process of the matrix, and the strength of the field is at least 1–2 kV/mm to ensure effective polarization of the particles. The appearance of EREs will be strongly influenced by the characteristics of the matrix raw materials. Some bubbles may be left in the mixture during the stirring process to achieve a ERE with voids, which will degrade the performance of the material. Thus, the mixture should be performed degassing to remove the bubbles within it.

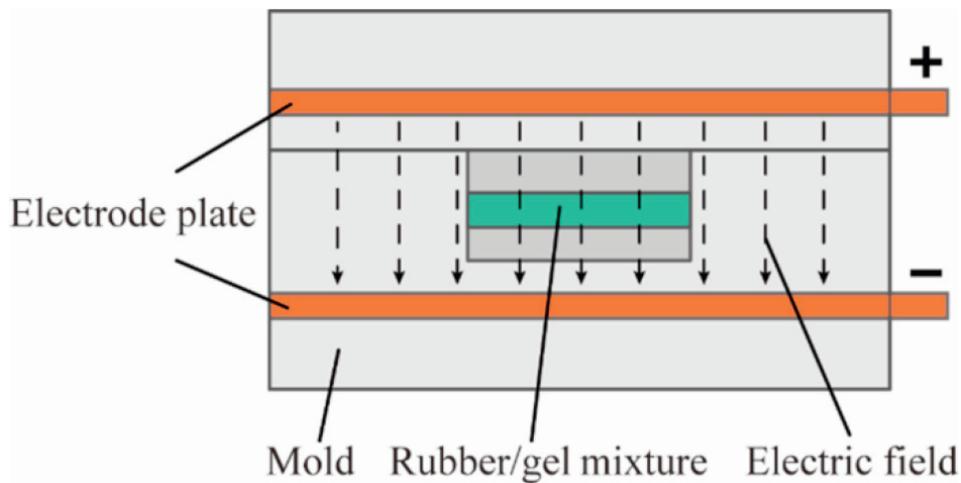


Figure 2. Schematic of ERE vulcanizing mold with DC electric field [18].

3. Physical mechanisms of EREs

The electroviscoelastic characteristics of EREs are mainly caused by the electro attraction between dispersed particles. Thus, the nature of the forces within adjacent particles is one of outstanding issues. Although some theories have been proposed, it is widely accepted that particle polarization forces play a dominant role in the ER effect [20, 21]. Klingenberg et al. first proposed the theoretical model to describe some aspects of ER suspensions with the one point dipole approximation model [22]. In this model, real ER solids could be considered as mono-disperse suspensions of dielectric hard spheres in a continuous polymer matrix. The polarization in the one point dipole approximation model that occurs inside the particles instead of on their surface, and the dipoles form within adjacent particles will be stronger as the dipoles come close to each other. Based on the classical theory, the polarization force of the particles under extra electric field can be given by [12]

$$F(R, \theta) = 12\pi r^2 \varepsilon_0 \varepsilon_m \kappa^2 E^2 (r/R)^4 [(3 \cos^2 \theta - 1) \mathbf{e}_r + \sin(2\theta) \mathbf{e}_\theta] \quad (1)$$

$$\kappa = (\varepsilon_p - \varepsilon_m) / (\varepsilon_p + 2\varepsilon_m) \quad (2)$$

where R is the center-to-center distance between the adjacent particles, θ is the angle between the applied electric field and the chain axis (**Figure 3**), r is the radius of the spheroidal particle, E is the strength of electric field, ε_0 is the permittivity in vacuum ($= 8.854 \times 10^{-12}$ F/m), ε_p and ε_m are the dielectric constants of the filler particles and matrix, respectively, \mathbf{e}_r and \mathbf{e}_θ are unit vectors in r and chain axis directions, respectively.

Eq. (1) is an expression obtained from solving Laplace's equation about a pair of spheroidal particles in the continuous unbounded medium. By assuming the spheres interaction was replaced by dipoles with the same intensity as the dielectric spheres in an electric field, they further obtained the point dipole moment, which can be written as

$$\mu = 4\pi r^3 \varepsilon_0 \varepsilon_m \kappa E \quad (3)$$

Eq. (2) shows the volume polarizability of one particle under applied field. If the adjacent two particles are parallel to the applied field and in contact with each other, the dipole interaction force between them can be rewritten as

$$F = (3/2)\pi r^2 \varepsilon_0 \varepsilon_m \kappa^2 E^2 \quad (4)$$

Based on the above results, Shiga et al. assumed that interactions between two adjacent particles mainly come from the dipole interaction between these two particles instead of the interaction between paths within them [23]. According to this assumption, macroscopic viscoelastic properties such as storage and loss modulus can be estimated by calculating the overall

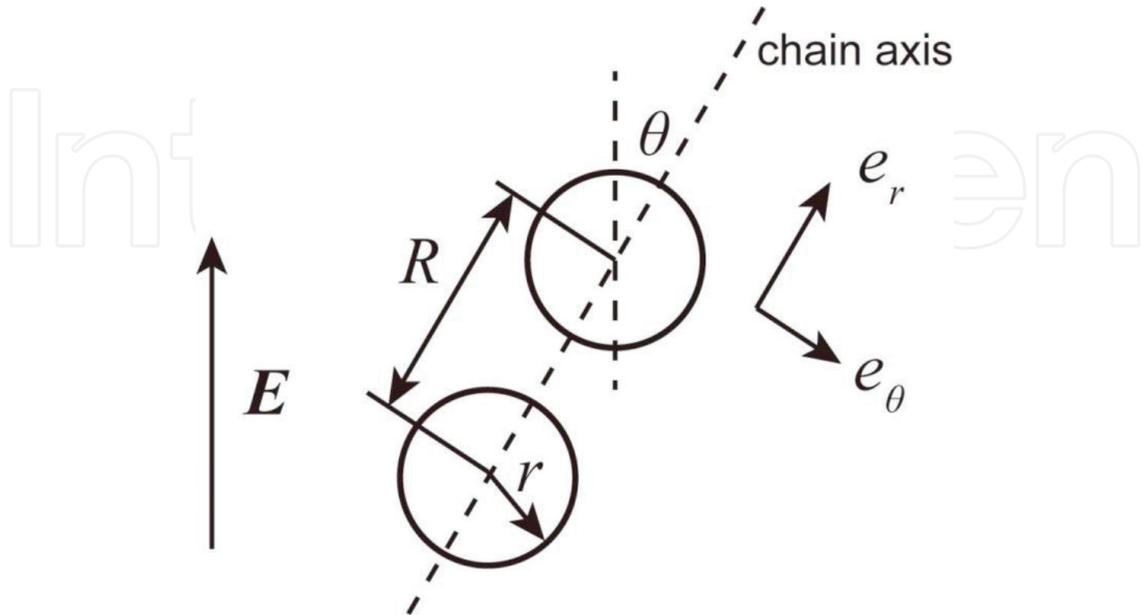


Figure 3. Two adjacent particles in electric field.

electrostatic interaction force within the adjacent particles in the matrix. Thus, the following equation can be used to calculate the increase in storage modulus with varied extra electric field:

$$\Delta G = (9/4)\phi\epsilon_m\kappa^2E^2 \quad (5)$$

where ΔG is the change of storage modulus, ϕ is the volume fraction of dielectric particles. It can be inferred from Eq. (5) that ΔG is proportional to the particle content and electric field intensity.

Liu et al. investigated the change of electrorheological and mechanical properties of EREs with anisotropically filled dielectric particles by numerical simulations [24]. Their simulations mainly focus on the distribution of electrostatic energy between the tips of two adjacent particles in a chain, as shown in **Figure 4**. According to their research, when the dielectric constant of filler particles is much greater than the matrix, the intensity of electric field will be concentrated between these two particles. This implied that electrostatic stress is located on the particle surface and the local fields between the tips of particles play an important role in determining the strength of restoring force.

The importance of local field strength between the particles on ER materials was further proved in the study of Lu et al., and a polar molecule (PM) dominated model had been proposed by them to explain the enhanced ER performance of the novel ER suspension [25, 26]. For this kind of ER suspensions, the polar molecules may orientate along the applied field in the tips of adjacent particles caused by the local field, which is three orders of magnitude larger than external electric field under certain condition. Once the polar molecules on particle surface are orientated under electric field, the PM-ER effect mainly comes from the attractive force f_{m-e} of polar molecules and polarization charges on the nearby particle (**Figure 5**). The overall attractive force F_{m-e} of molecule-charge in a unit area that perpendicular to applied electric field is given by

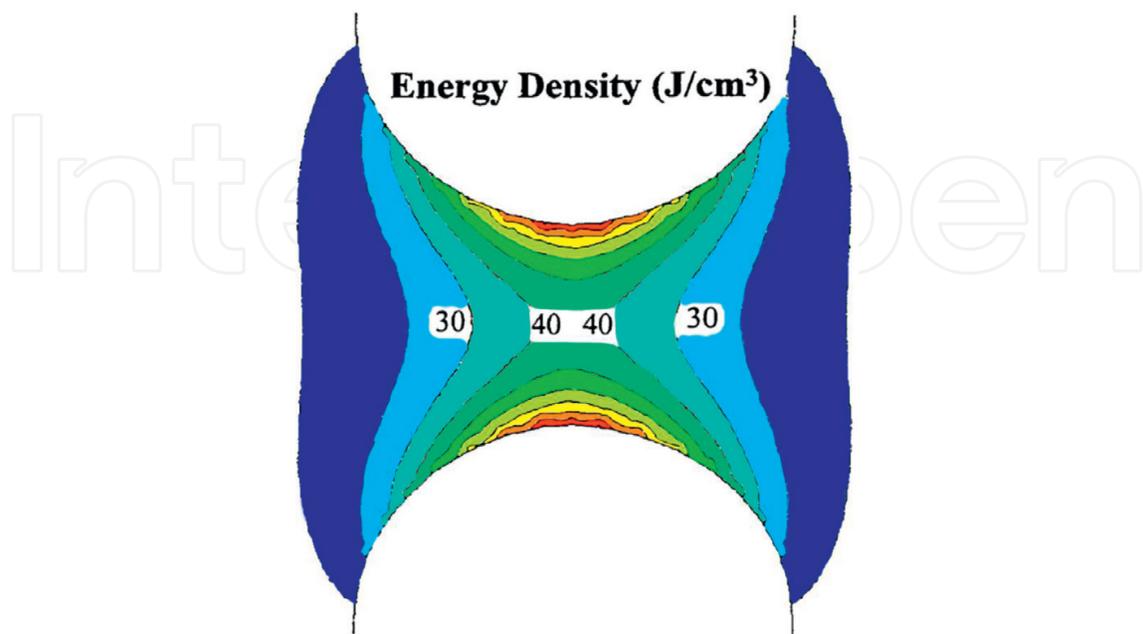


Figure 4. Contour of electrostatic energy distribution for rod-like filler particles in a 2-D axisymmetric simulation [24].

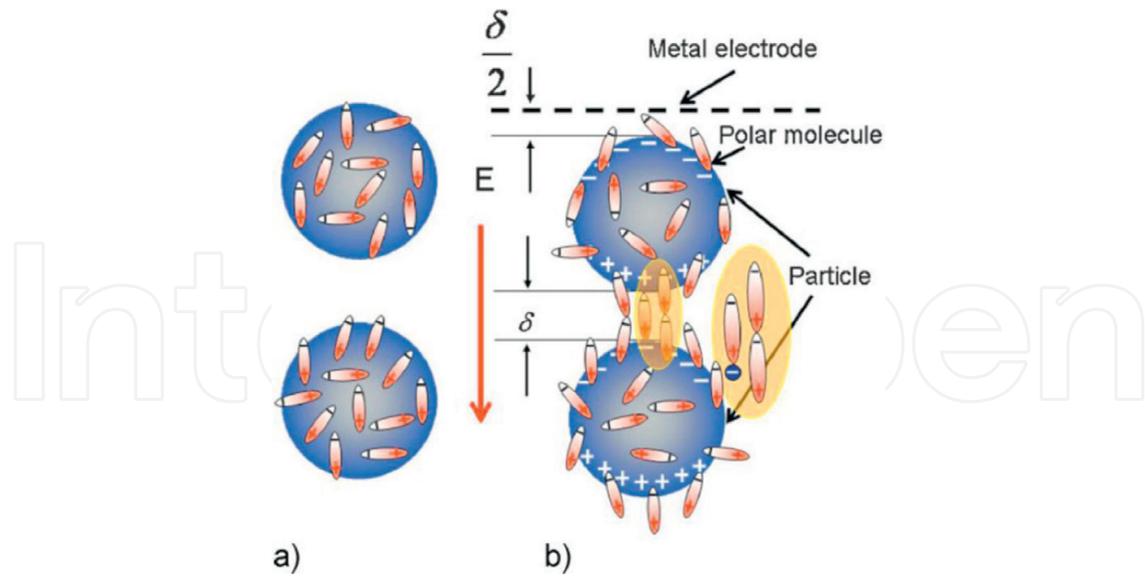


Figure 5. Schematic of the polar molecules aligned in the gap of adjacent particles. (a) The polar molecules adsorbed on particle surface are arbitrarily oriented when $E = 0$. (b) The polar molecules in the gap of the particles are oriented parallel to the field when E is high enough [26].

$$F_{m-e} = \frac{3\phi}{2\pi r^2} N f_{m-e} = A \frac{3\phi \rho_m e \mu^2 E}{\pi r \epsilon_0 \epsilon_m d^2} \quad (6)$$

in which

$$f_{m-e} = \frac{e\mu}{2\pi \epsilon_0 \epsilon_m d_{m-e}^3} \quad (7)$$

where A is a factor that related to the properties of the particle and oil, ϕ is the volume fraction of dielectric particles, ρ_m is the area density of the polar particles on the particle surface, e is the fundamental charge, r is the radius of filler particle, μ and d are the dipole moment and size of the polar molecules, respectively. Polar molecule-dominated model was initially used to explain the mechanical properties of PM-ER fluids, while it also opened the door to the innovation of novel ER elastomers filled with polar molecule modified particles.

One recent study by Zhao et al. proposed a theory on the mesostructured design of EREs for gaining giant tunable stiffness [27]. The EREs in this theory consist of two kinds of layers with defined thickness and dielectric constant, and the effective dielectric constant of the layered structure $\bar{\epsilon}$ can be regarded as a function of variable θ with coefficient ϵ , n , and m , as shown in **Figure 6**. By connecting the thermodynamic system with the geometrical parameters of EREs, they can calculate the electrorheological coefficient as

$$K_{ER} = 4\alpha \cos(2\theta) \left(\frac{\partial \theta}{\partial \Delta \theta} \right)^2 + 2\alpha \sin(2\theta) \frac{\partial^2 \theta}{\partial \Delta \theta^2} \quad (8)$$

$$f(\theta) = \alpha \cos(2\theta) + \beta \quad (9)$$

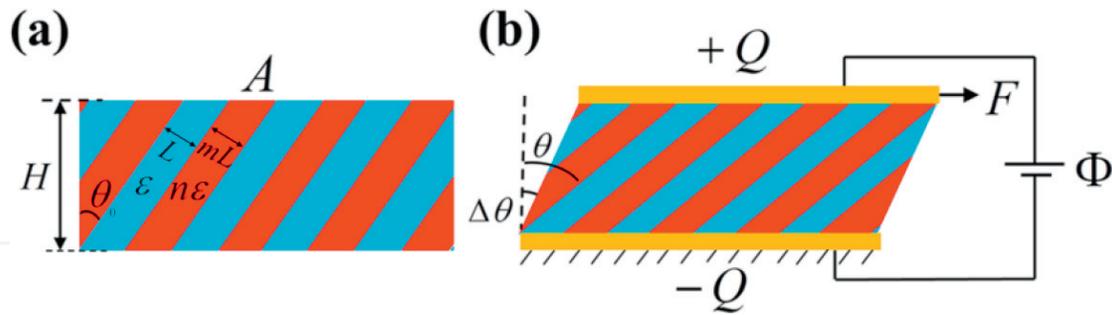


Figure 6. Schematic of one ERE with layered mesostructures at undeformed state (a) and undergoing simple shear deformation by an applied force and a voltage (b) [27].

where $\alpha = [f(0) - f(\pi/2)]/2$ and $\beta = [f(0) + f(\pi/2)]/2$. The EREs will have giant tunable stiffness when the coefficient K_{ER} reaches the maximum value. Finally, they found when $m = \sqrt{n}$, K_{ER} reaches its maximum with a given m . The K_{ER} can be expressed as

$$K_{ER}^{\max}(0) = 2(\sqrt{n} - 1)^2 \quad (10)$$

In their conclusion, the EREs with a layered mesostructured arranged in parallel configuration will have the most widely tunable stiffness among all the structures with different orientations.

4. Development and diversification of EREs

4.1. Inorganic particles filled EREs

Inorganic particles are most frequently employed as the dispersion phase as their outstanding properties, including high permittivity, easy synthesis, and good environmental stability. A range of inorganic micro/nanosize particles, such as $PbTiO_3$ [28], silica [29], montmorillonite clay [19], titanium dioxide [30], and barium titanate [31] have been applied as the candidates of dispersed phase in EREs. Gao and Zhao investigated the mechanical properties of hydrogel elastomers containing barium titanate particles [32]. They found the aligned particle chain effect could induce large compression modulus that can be controlled by applied field strength. While in the research of Mitsumata on swollen silicone gel containing barium titanate, a negative electrorheological effect appeared for microscopic phase separation in the elastomers [33]. To enhance the electric responses, Gao et al. also prepared $BaTiO_3$ particles coated with many kinds of polymers, such as polyimide, chitosan, polystyrene, poly acrylic acid, and poly acrylamide [34]. They found the hydrous elastomer filled with $BaTiO_3$ /polyimide particles show strongest electric response, which is closely related to hydrophilic surface and permittivity of the particles.

In order to improve the permittivity and conductivity of the particles, Kossi et al. prepared the TiO_2 doped with acetylacetonate dipolar molecules (Acac), and the EREs prepared by dispersing

TiO₂-Acac in PDMS achieved a relative storage modulus change higher than 500 kPa at 2 kV/mm [30]. The relative storage modulus change is defined by the following equation

$$\frac{\Delta G'}{G'_0} = \frac{G' - G'_0}{G'_0} 100\% \quad (11)$$

where G' and G'_0 are the storage modulus with and without an electric field, $\Delta G'$ is the change of storage modulus. Inspired by the study of Wen et al. on polar molecules dominated ER materials, Dong et al. proposed a kind of elastomer containing polar molecule-modified particles, which is expected to have much enhanced ER activities [18]. The elastomers synthesized by dispersing urea-coated titanium dioxide (TiO₂/urea) particles in silicon rubber presented high ER performance, the relative ER effects of which achieves 266% at 3 kV/mm (Figure 7).

4.2. Organic particles filled EREs

Organic hydrous particles, such as starch and cellulose, have been widely used in ER materials and shown strong ER performance. When Shiga et al. prepared the earliest elastomers with ER activity, the filler particles they adopted were polymethacrylic acid cobalt (II) salt with a small amount of absorbed water [21]. This kind of organic particles will present stable dispersion properties in polymer matrix and relatively good ER behaviors. However, the EREs containing organic hydrous particles will suffer from some defects. First, the leakage current density of EREs would be large if too much water is absorbed on the

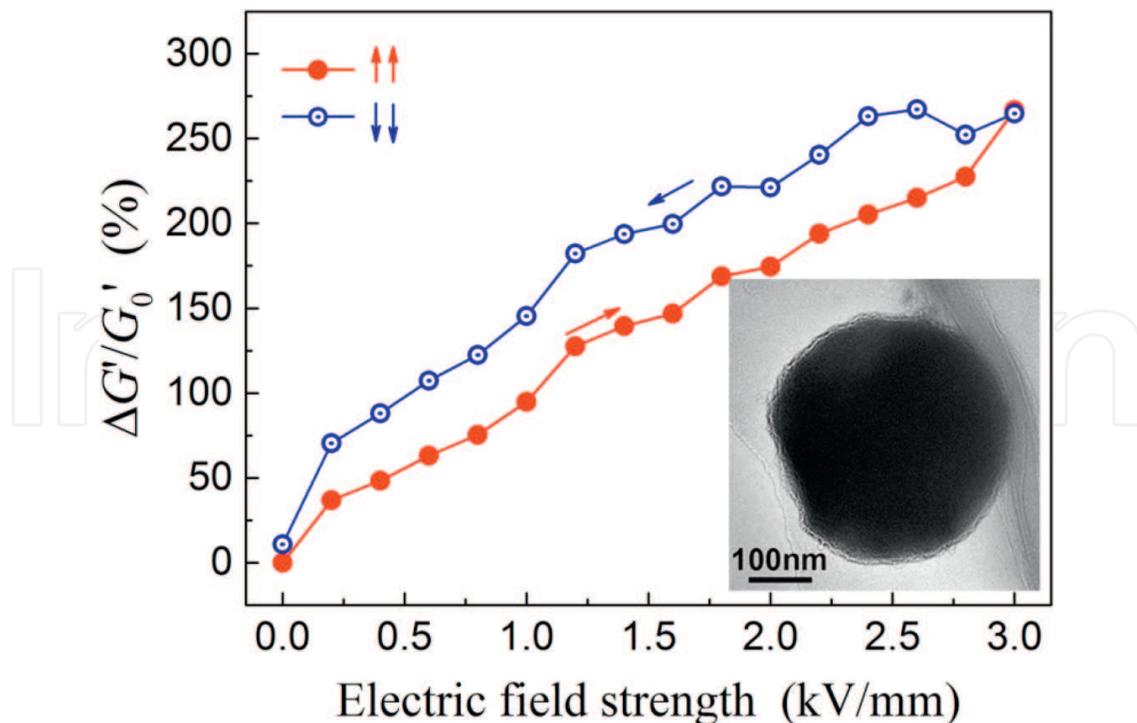


Figure 7. Relative ER effect of TiO₂/urea particles filled elastomers as a function of electric field strength, inset figure is the TEM images of TiO₂/urea particles.

particles, which will be unable to apply high voltage on the samples. In addition, the EREs containing hydrous particles are sensitive to working conditions, especially the operating temperature. Besides, the moisture in the EREs is also possible to cause corrosion problems on the electrode [35].

To overcome the shortcomings of the EREs filled with organic hydrous particles, some systems with anhydrous polymer particles have been investigated in recent years. Conducting polymers that contains conjugated π bonds have been used as ER materials due to their polarization properties under applied electric field. Besides, polymers with polarizable functional groups on the molecules backbone belong to another kind of anhydrous organics that adopted as ER materials. PANI is one kind of conducting polymers that has been regarded as an appropriate candidate for EREs. It has many excellent properties such as high conductivity and good environment stability, which is important for application in ER materials. Hiamtup and Sirivat investigated the electrorheological response of EREs filled with PANI particles, and they found the storage modulus of this kind of ERE will increase about 10–50% with the ascending field strength [36]. Other conducting polymers, such as polythiophene [37], poly(p-phenylene) [38], and polypyrrole [39] have also been studied as the dispersed phase of EREs. Compared with the inorganic particles, the polymeric dispersion particles are softer and have better compliance with the matrix.

5. Influence factors of ER performance

Since EREs are usually made of two constituent materials with different properties, the influence factors mainly come from three parts in them: the filler particles, matrix, and interface of these two phases.

5.1. Effects of particle properties

As the dispersion phase of EREs, particles play the most important role in determining the ER performance. According to the theory in previous study, the dielectric constant mismatch is closely related to polarization of EREs [40]. The polarization force is positively related to the coefficient κ in Eq. (2), which infers the difference between the dielectric constant of particles and matrix. When the permittivity of filler particle is much larger than that of polymer matrix, the ER materials will have stronger ER activity.

The content of particles filled in the EREs also has a great influence on their ER performance, and it has been investigated by many researchers theoretically and experimentally. As described in Eq. (5), the change of storage modulus of EREs is $\Delta G'$ is proportional to the content of filler particles [23]. In the study of Lu et al., the total interaction force between polarized molecule and charge in a unit area is also proportional to the particle content [25]. In 2006, Hiamtup and Sirivat investigated the viscoelastic properties of EREs with the particle volume fraction ranging from 0 to 20%, and they got the conclusion that both storage and loss moduli increased with the particle concentration, as shown in **Figure 8** [36].

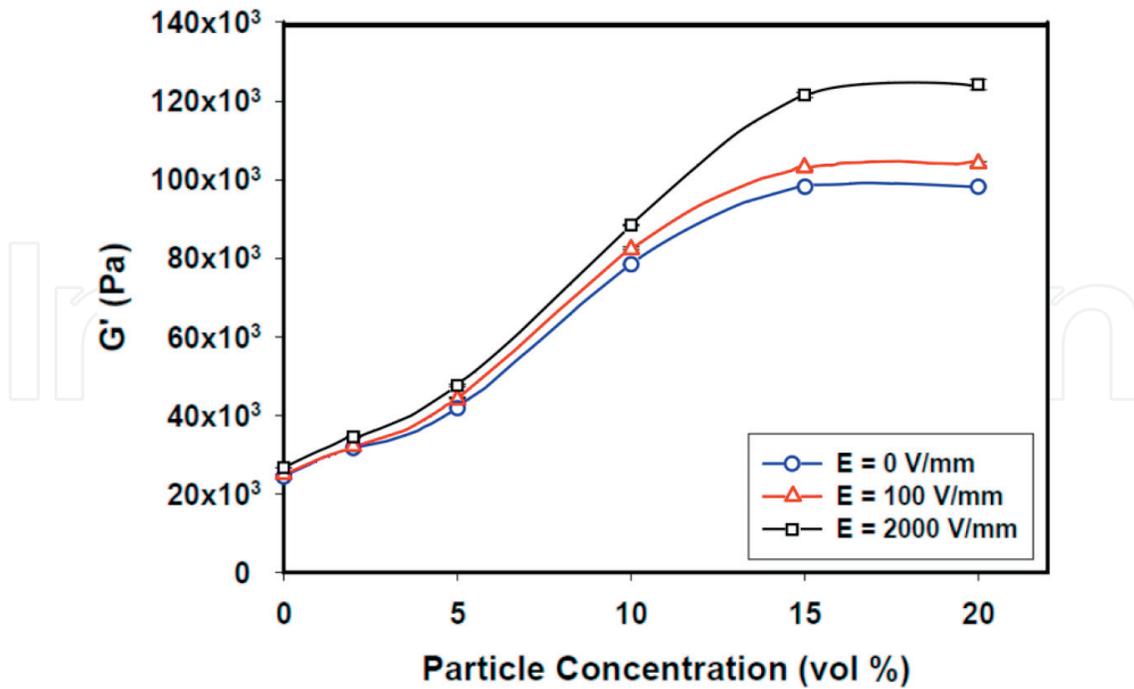


Figure 8. Particle concentration dependence of storage modulus of PANI/PDMS EREs under various electric field [36].

The morphology of particles is another important factor influences the viscoelastic properties of EREs under electric field. Kanu and Shaw systematically studied the role of particle geometry on ER response based on the dipole theory [41]. They found the polarization of the particles with larger aspect ratios will be stronger and these particles will participate in enhanced dipole-dipole interactions. Liu et al. computed the electrostatic forces between the particles with three kinds of geometries (sphere, capsule, and pointed capsule) in the EREs using numerical simulations [14]. Their calculation inferred the EREs containing the particles with a higher aspect ratio will present higher ER response for the formation of more particle chains. Jang et al. investigated the ER activity of the suspension containing pure silica or PANI coated mesoporous silica particles with different aspect ratio [42, 43]. According to their research, the ER materials with higher aspect ratios will have improved dielectric properties of large polarizability and short relaxation time. Therefore, the synergistic contribution of geometric effects and dielectric properties resulted in stronger ER activity.

5.2. Effects of polymer matrix

The tunable stiffness range is one of the most important properties for EREs, which can be evaluated by the relative ER effect as Eq. (11). Many research studies have focused on the materials used for the matrix of EREs due to their roles in determining the initial elastic modulus. Sakurai et al. adopted silicone elastomers with different moduli as the matrix of EREs and investigated how the ER properties depend on the elasticity of the elastomers [44]. They found the largest relative increase in modulus under electric field occurs when silicone

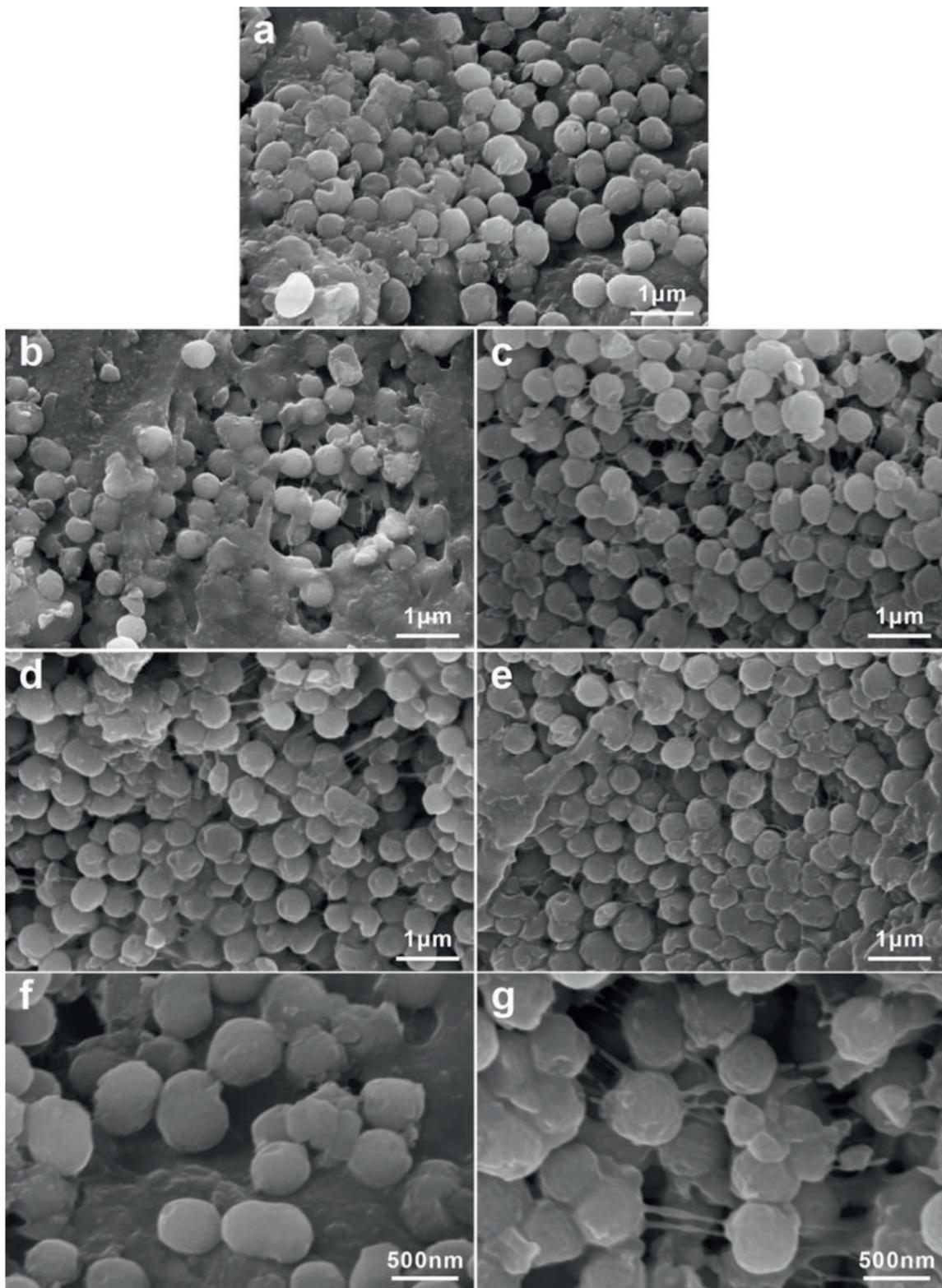


Figure 9. Section SEM microphotographs of (a) TiO_2 , (b) $\text{TiO}_2/\text{A1}$, (c) $\text{TiO}_2/\text{A3}$, (d) $\text{TiO}_2/\text{V1}$, and (e) $\text{TiO}_2/\text{V3}$ particles filled elastomers have been soaked in toluene and (f and g) the magnification images of (a and d). A1 means the weight fraction of A174 is 1% to the TiO_2 particles, the rest symbols represent similar meaning [48].

elastomer with low elasticity is used. Except for silicone rubbers, many elastomers with low initial moduli have been used as the matrix of EREs, such as acrylic and polyisoprene elastomer [37, 45]. To further decrease the initial moduli of the EREs, Gao et al. also tried to use the gels like gelatin and chitosan as the matrix [46, 47]. However, most of the EREs used gels as the matrix can only work in compression mode due to their mechanic properties. Besides, it is mentioned in the study of Liu et al. that the conductivity of polymer matrix can play an important role in the electric field distribution of EREs under high field strength [14]. It is implied that the polarization of filler particles in high field may be affected by the conductivity of the matrix.

5.3. Effect of particles/matrix interface

As a composite, the interface between filler particles and matrix plays an important role in the ER performance of EREs, which has been overlooked in previous studies. To improve the interfacial bond strength in the EREs and find out how does the interface influence the ER performance, Dong et al. adopted two coupling agents, 3-(trimethoxysilyl)propyl methacrylate (A174), and triethoxyvinylsilane (VTEO), to modify the surface of the TiO_2 particles [48]. Compared with the bare TiO_2 particle filled sample, the modified TiO_2 particle filled elastomers present bridging structures between the particles and the matrix, enhanced mechanical properties, and a higher dielectric constant, indicating enhanced interfacial bond strength (Figure 9).

The viscoelastic properties of the bare and the modified TiO_2 particle filled elastomers were tested and compared in this study. The results indicated that the ER performance, including the field-induced storage modulus and the storage modulus sensitivity, can be promoted by improving the bond strength between the TiO_2 particles and the silicon rubber, as shown in Figure 10.

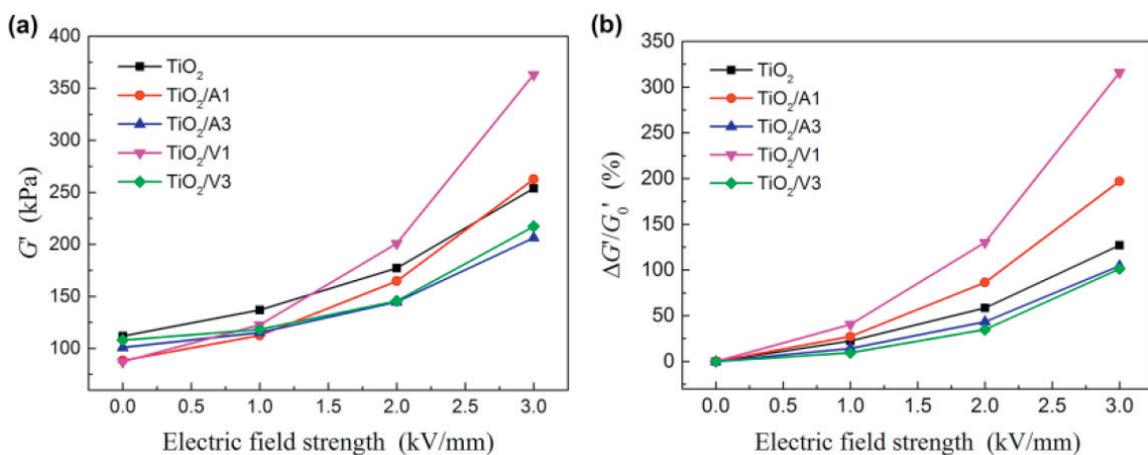


Figure 10. Storage modulus G' (a) and relative ER effect (b) of five types of particle filled elastomers as a function of electric field strength [48].

6. Applications of EREs

On the basis of the ER effect and special mechanic properties, EREs can be applied in several fields, including dampers, automotive industries, building base-isolation, and smart skins in bionic technology [12, 49]. Although commercialization of ERE-based devices has not been implemented, the tentative application of EREs has been done in some laboratory research studies. Biggerstaff and Kosmatka used the silicone gel based EREs as active dampers [50]. It is shown in the harmonic tests that the EREs reveal a factor six increase in stiffening and a factor of three decreases in damping under electric field. Koyanagi et al. created a novel prototype linear actuator with ERE drum and found the response time of the EREs was fast enough for mechatronics or robotics applications as physical supporting in welfare situations, while some problems like the relation between the pressure on the surface of the EREs and generative force were left for further investigation [51]. Besides, Wei et al. investigated the vibration characteristics and control capabilities of a cantilever sandwich beam at different electric fields [52]. The results indicated that the natural frequencies of the ERE sandwich beam increase and the vibration amplitudes at natural frequencies of the beam decrease with the ascending electric field intensity. Since the vibration characteristics of ERE beams can be controlled by changing the strength of applied field, the EREs are useful for applications in engineering structures where variable performance is desired. In 2013, Zhu et al. designed a shear mode ERE shock absorber and evaluated its vibration response performance under various excitation frequencies with and without external electric field [17]. Their experimental results showed that the stiffness and damping properties of the shock absorber could be controlled by changing the strength of applied electric field, whose macrofeatures was that the damping coefficient increased with the ascending electric field strength, and the damping effect in high frequency was better comparing with that in low frequency.

7. Summary and outlook

With respect to the scientific papers on EREs published in the past few years, methods of preparing new EREs and improving the dynamic mechanical properties, and the modeling approach for simulating the electrorheological and viscoelastic mechanical behavior of these EREs are of increasing scientific and technological importance. Based on the investigations about ER fluids, the point dipole approximation model and its modified model are most widely used to describe the viscoelastic properties controlled by electric field, and the meso-scale structure of EREs is also analyzed with micromechanics of composites. In addition, researchers have made considerable progress in developing new materials in this field, including inorganic particles, organic hydrous, and organic anhydrous particles filled EREs. For different types of EREs, the influencing factors on ER performance related to the filler particles, polymer matrix, and the interface between particles and matrix are studied experimentally or theoretically. In recent years, some preliminary explorations in the application of EREs have been carried out and some valuable conclusions are obtained.

Even though many reports focus on the mechanical and ER performance of EREs, some defects still prevent them from practical applications. The most important problem of EREs is the rather high voltage must be applied for obtaining large relative storage modulus, which will increase the security risks. The relatively low elastic modulus of the EREs makes them unable to sustain something of great weight, which is another defect that will narrow the application fields for EREs. Apart from that, considerable work is needed to broaden the practical application of EREs and make them more reliable under long service period. The EREs containing polar molecules modified particles have presented enhanced ER activities under relatively low field strength in previous studies, which indicates that to increase the polarization abilities of particles under the guidance of polar molecules dominated ER effect model is an effective method to improve ER performance of the composites. Therefore, research studies of EREs are still widely open for extensive works on theoretical analysis of basic mechanisms on electroresponsive behavior of the particles based on their dielectric characteristics, and dynamic viscoelastic mechanical properties. Besides, new generation of EREs should have both large relative storage modulus and mechanical properties. On the other hand, it is important to design the devices with full insulation protection and safety power supply aiming at commercial applications. When these problems are solved, EREs should be one kind of promising smart materials that serve for modern society.

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