Chapter

Dual Response of Materials under Electric and Magnetic Fields

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Abstract

The electrorheological (ER) effect is known as the change in the rheological behaviors of ER fluids under applied electric field E. When an E is imposed, ER fluids show phase transition from a liquid to a solid-like state due to the interactions of polarized particles. This solid-like behavior of particles is due to the increasing viscosity of suspensions. ER materials belong to a family of controllable fluids. ER fluids are dispersions of solid particles in a hydrophobic insulating dispersion medium. These solid particles play a very important role in the ER activity of dispersions. As the dispersed phase, diverse materials such as polymer blends, gels, biodegradable materials, clays, graphene oxide, hybrid nanocomposites, copolymers, ionic liquids, and conducting polymers have been proposed. In the magnetorheological fluids, this control is provided with magnetic field. Various magnetic particles such as carbonyl iron and iron oxides have been suggested as MR material. The combined effect of magnetic and electric field produces intensified rheological changes in the suspensions. This synergic effect is termed as electromagnetorheological effect (EMR). The EMR effect provides a new strategy to control the rheological properties of dispersions.

Keywords: electrorheology, magnetorheology, nanocomposites, smart fluids, dual response

1. Introduction

The electrorheological (ER) effect is known as the change in the rheological behaviors of ER fluids under applied electric field E. When an E is imposed, ER fluids show phase transition from a liquid to a solid-like state due to the interactions of polarized particles. This solid-like behavior of particles is due to the increasing viscosity of suspensions. There are two main driving forces behind the viscosity increase in ER fluids. Fibrillar structure of polarized molecules with applied electric field and ion aggregation near the electrode surfaces are responsible for the viscosity increase. Essentially the ER effect in a liquid is related to the motions of ions or polar molecules [1, 2] (**Figure 1**).

In the nineteenth century, the apparent viscosity of pure nonconducting liquids was believed to increase with the applied E. This effect was called as electroviscous effect rather than the ER effect [3]. Many other solution systems showed that the electrolyte solutions have very much stronger electroviscous effect than the pure liquid. After this time, Winslow studied the E-induced viscosity increase of solid semiconducting particles dispersed in a low viscosity and high insulating oil medium [4]. The E-induced effect was much stronger than the electroviscous effect.

Therefore, the ER effect was termed as "Winslow effect." Winslow effect is regarded as the formation of fibrillated chain structures by the dispersed particles between the electrodes of a measuring system. Efforts have been carried out to elucidate the ER mechanism and to improve ER performance of dispersions that meets the industrial requirements.

ER fluids exhibit large reversible changes in their rheological properties when they are subjected to external E. Many studies have been carried out on the mechanism and application of ER fluid. One important property of the ER fluids is that it shows quick response to electric field. This inherent behavior has pioneered many researches to various engineering applications [5]. Some of these are machine mounts, shock absorbers, and smart structures. The rheological properties of ER fluids can be electrically controlled, and this control is beneficial to a wide range of application technologies such as active vibration, damping, and resistive force generation. Several commercial applications have been developed until today, and many potential applications especially in the automotive industry such as ER fluid-based engine mount, shock absorber, clutches, and seat dampers are still undiscovered [6].

Many materials have been used for the preparation of ER fluids. They can be classified as inorganic materials [7, 8] (such as TiO_2, $BaTiO_2$), organic materials [9] (such as chitosan, alginate, and its derivatives), and conducting polymers (such as polyaniline, polypyrrole). Among them conducting polymers have been widely studied, because of their ease of synthesis, less corrosive properties and high stability, and compatibility with the carrier liquid.

Yield shear stress is one of the most important parameters for ER fluids and can be varied by the applied E. Despite these properties, ER fluids have limited their application due to low yield stress, which yields poor mechanical performance that limits their application [1, 10]. The developing of the giant ER effect has revived interest in this area. When compared with traditional ER fluids, giant ER fluids show higher magnitude yield stresses. The polarization force of molecular dipoles between dispersed particles is responsible for the giant ER effect [11]. Many organic–/inorganic-structured giant ER materials have been synthesized. Wen et al. [12] reported the synthesis and ER properties of a new type of giant ER fluid consisting of polar group-modified nano-sized barium titanyl oxalate particles. Also, the problem associated with sedimentation of colloidal particles, as well as the lack of high-performance materials, has inhibited broad engineering applications [13, 14]. Giant ER fluids like other traditional ER fluids exhibit sedimentation drawback due to the density mismatch of the fluid and solid phases as well as the aggregation of the dispersed particles [15, 16]. To improve the sedimentation stability of the ER

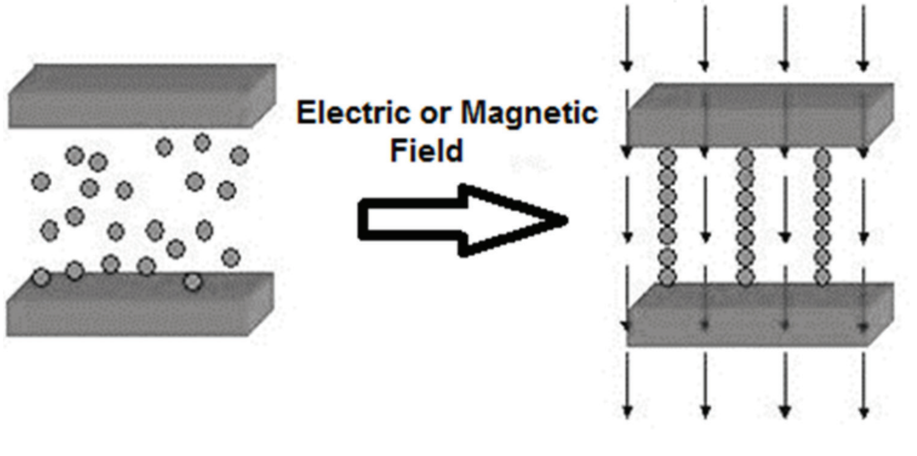

Figure 1.
Schematic illustration of particles into an ER fluid at absence and presence of electric or magnetic field.

fluids, several methods have been developed. They can be listed as follows by adding surfactant to the dispersion, making the particle phase less dense that decrease the density mismatch and modify the surface or particle morphology [17]. Li et al. [18] reported the fabrication of multiwall carbon nanotubes, urea-coated barium titanyl oxylate composite particles, that exhibit the giant ER effect (yield stress was 194 kPa at 5 kV/mm). The antisedimentation property enhanced dramatically in silicone oil due to multiwall carbon nanotubes. Cabuk et al. [16] reported ER response and temperature effect on antisedimentation stability of expanded perlite particles in silicone oil. The expanded perlite particles as porous ultra-lightweight materials showed typical ER properties under applied E, exhibiting a shear thinning non-Newtonian viscoelastic behavior (**Figure 2**).

When the mechanism of the ER fluids is examined, Most ER fluids fit the Bingham fluid model. At this model, an ER fluid has its own yield stress, and the flow motion of the fluid impedes when an applied external shear stress is lower than the yield stress [19]. The Bingham model has been extensively used as a viscoelastic equation:

$$\tau = \tau_y + \eta\dot{\gamma}, \tau \geq \tau_y \tag{1}$$

$$\dot{\gamma} = 0, \tau < \tau_y \tag{2}$$

In Eqs. (1) and (2), τ and τ_y are shear and yield stresses, $\dot{\gamma}$is a shear rate, and η is a shear viscosity.

Klass and Martinek [20] introduced a dielectric principle and a water-associated electrical double-layer model, to explain that the critical factor of the ER effect was the molecule in the wet-base ER fluids. The dispersed particles were polarized and distorted when the extra E was applied, and then the water molecules into the dispersion made an attractive bridge between dispersed particles, thereby producing a higher surface tension.

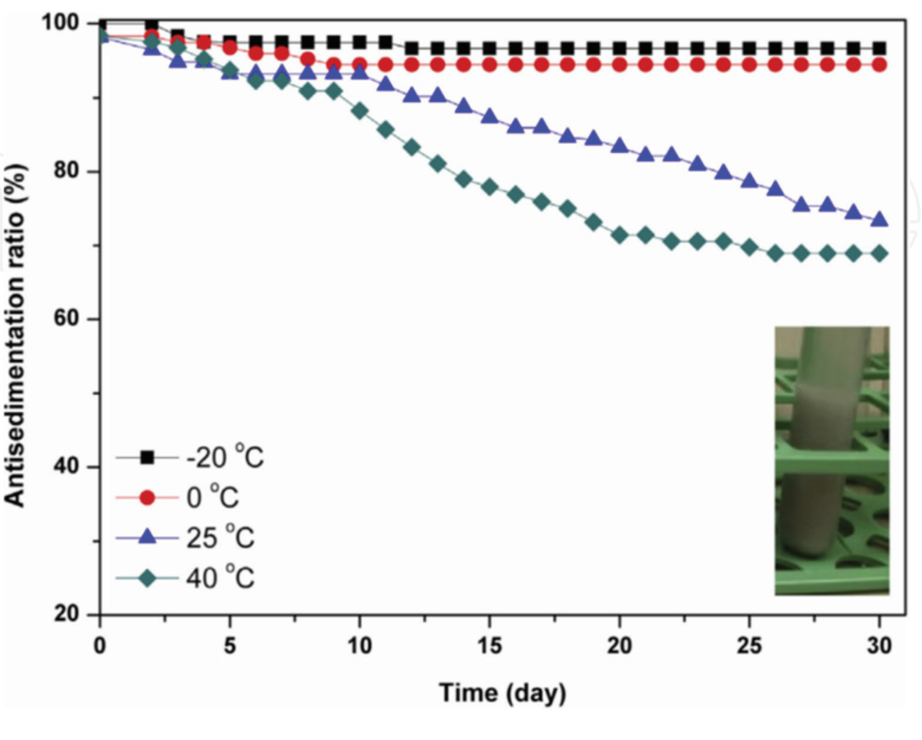

Figure 2.
Antisedimentation stabilities of expanded perlite/silicone oil colloidal dispersions at various temperatures (φ = 10%) [16].

Cho-Choi-Jhon (CCJ) model is an empirical formula with six parameters to explain the ER characteristics in a whole shear rate range [21]. The CCJ equation is given as follows:

$$\tau = \frac{\tau y}{1 + (t_1\dot{\gamma})^{\alpha}} + \eta_{\infty}\left(1 + \frac{1}{(t_2\dot{\gamma})^{\beta}}\right)\dot{\gamma} \tag{3}$$

The first term expresses the decrease in the shear stress at the low shear rate area, and the next term shows a shear-thinning behavior at the high shear rate region. The t_1 and t_2 are time constants, and η_{∞} is a shear viscosity at the infinite shear rate. The exponent α corresponds the shear stress decrease, and b is located between 0 and 1. The CCJ model is observed to be a good indication of the overall behavior of many ER fluids, including the reduced shear stress behavior.

Magnetorheological (MR) fluids consist of dispersed magnetic particles and nonmagnetic fluids such as an hydrocarbon, aqueous carrier fluid, or silicone oil [22, 23]. MR fluids have a Newtonian-like flow behavior with applied magnetic field. However, without an external magnetic field, they show a phase change from a liquid-like to a solid-like due to the formation of a fibrillar structure along the direction parallel to the direction of the applied magnetic field. This behavior is due to the magnetic polarization between the dispersed magnetic particles into the MR dispersion. A hybrid of conducting and magnetic particle-based materials can display an improved dual stimuli–response under electric and magnetic fields [24]. These kinds of fluids also show typical ER and MR behaviors.

2. ER effect and types

On the basis of changes in ER properties of the dispersions, such as viscosity, shear stress, elastic and viscous moduli, and creep recovery, they can be classified as positive or negative ER material depending on their response to the external electric field imposed. In addition to ER effect, depending on the type of applied external stimuli photo, MR and electromagnetorheological effects are also investigated and used in various industrial applications.

2.1 Positive ER effect

An increase in rheological property with applied E is known as the positive ER effect. The fibrillar structure of ER particles is observed in positive ER fluids under E. This dramatic increase in viscosity is due to the change from liquid to solid-like state which is caused by the aggregation of polarized particles into fibrous structures under electric filed. These fibrous structures restrict the motion of the dispersed particles inside the base fluid under the shear flow conditions and result in enhanced viscosity (**Figure 3**).

The positive ER effect is the most common ER phenomenon encountered in the literatures. Conducting polymers (polyaniline, polypyrrole, etc.) [26–28], liquid crystalline polymer/poly(dimethylsiloxane) blends [29], smart polymer/carbon nanotube nanocomposites [30], PPy-tin oxide nanocomposite [31], PAni-coated mesoporous silica [32], core−/shell-structured SiO_2/PPy nanoparticles [33], graphene oxide/PAni composites [34], and biodegradable chitosan/bentonite composites [35] were reported to show positive ER effect.

2.2 Negative ER effect

On the contrary to positive ER effect, for the negative ER effect, the ER properties (i.e., electric field-induced viscosity) are decreased with applied E. the first

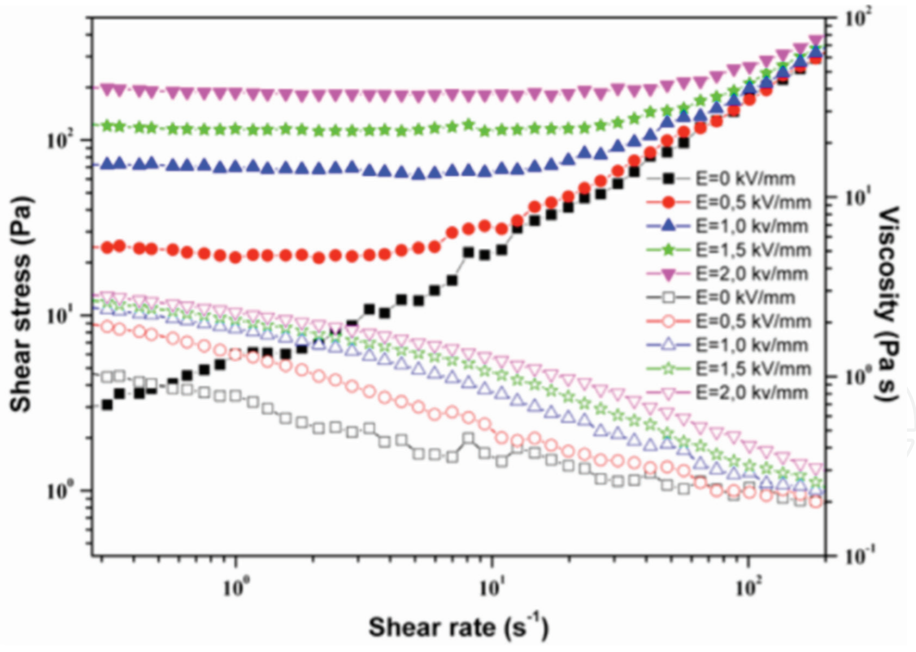

Figure 3.
Change of viscosity and shear stress with shear rate under various E for polyaniline/bentonite composite [25].

negative ER effect was reported by Boissy and his co-workers for the poly(methyl methacrylate) PMMA powder/SO ER fluid [36]. Also, magnesium hydroxide, poly(tetrafluoroethylene) [37], hematite [38], and fumed silica [39] were reported to show the negative ER effect. Urethane-modified polyether liquids exhibit either positive or negative ER effect. Negative ER effect is increased by introducing a branched structure in the polyether main chain or a hard segment at the center of the linear polyether [40]. In a study, colemanite/SO ER fluid showed positive ER effect until E = 1.5 kV/mm and exhibited negative ER effect above E = 1.5 kV/mm. On the other hand, polyindene/colemanite composites dispersed in silicone oil were reported to show negative ER effect with all applied E. This negative ER activity of both the materials was attributed to the electromigration of the particles to one of the electrodes which resulted in phase separation of the dispersion. The presence of Triton X-100 nonionic surfactant converted a negative ER effect to a positive one with improving ER activity. The water/surfactant-bridge formation model was attributed [41]. In another study, the ER properties of urethane-modified polypropylene glycol (UPPG)/PDMS were reported. When the viscosity of UPPG is higher than viscosity of PDMS, ER effect of the suspension is positive. When the viscosity of UPPG is lower than viscosity of PDMS, the ER effect of the suspension is negative [42].

Negative ER effect is explained with two reasons:

- Electrophoresis of particles which migrate to one of the electrodes under the applied electric field leading to phase separation.

- Quincke rotation—Accumulation of charges on the particle surface after the polarization under applied E: the charges on the top and bottom of the surface of particles have the same sign as that of the upper and lower electrodes, and this makes the dispersion unstable and causes the particles to rotate under E.

ER fluids have been used in various industrial areas such as mechanical sensors, damping systems, e-ink, brakes, haptic services, human muscle simulators, and polishing media. Their main advantage is ability to control of mechanical properties with E. These fluids can also exhibit Newtonian fluid characteristics which is the advantage of negative ER fluids [43].

2.3 Photo effect

Light can induce polarization of dispersed particles similar to that of an applied E. Not only positive ER effect but also negative ER effect can be improved by ultraviolet illumination in some ER dispersions. This behavior is called photo-ER effect [44]. Photo-generated carriers are responsible for changing the conductivity of the ER materials and improving their ER response. Photo ER materials show an important relationship between charge mobility and ER activity. Komoda and co-workers studied the photo-ER of TiO_2 nanoparticle dispersions [45]. These dispersions showed positive photo-induced ER effect at low water content, while negative photo-induced ER effect was observed at high water content. Besides, some dye particles (Monastral Green B and copper phthalocyanine), titania nanoparticles, and phenothiazine were reported to show photo ER effect [46, 47].

3. ER and MR materials and their properties

ER materials belong to a family of controllable fluids. They are typically dispersions of solid particles in an insulating liquid. Since the discovery of ER fluids, various types of ER material composition have been proposed [48, 49].

Optical microscopy (OM) studies provide observation of fibrillation of dispersed solid particles under an externally applied E which is a characteristic property of ER phenomenon (**Figure 4**).

The dispersed particles have an importance in the ER activity. As the dispersed phase, diverse materials such as polymer blends, gels, biodegradable materials, clays, boron-containing polymers, hybrid nanocomposites, copolymers, ionic liquids, and conducting polymers have been proposed. In recent years, the synthesized ER materials are polystyrene/laponite composites [51], polypyrrole-g-chitosan copolymer [52], polythiophene/polyisoprene composite [53], graphene oxide/titania nanocomposite [54], chitosan/bentonite composite [9], and poly(vinyl chloride)/polyindole composite [55].

The yield stress of MR fluids is generally higher than that of ER fluids. Therefore, MR fluids have a potential for active development. MR fluids show reversible specific properties. They can magnetize under magnetic field and demagnetize in the absence of magnetic field. Magnetite and carbonyl iron (CI) as magnetic particles have been used in the industrial applications of MR fluids [56]. Among the magnetic particles, soft magnetic micron-sized CI particles have an attention for superior MR fluids due to its high saturation magnetization value, suitable size, spherical shape, and low cost [57].

The liquid phase should have high electrical resistivity and hydrophobicity. Liquids such as mineral oil, kerosene, toluene, petroleum fractions, and silicone oil

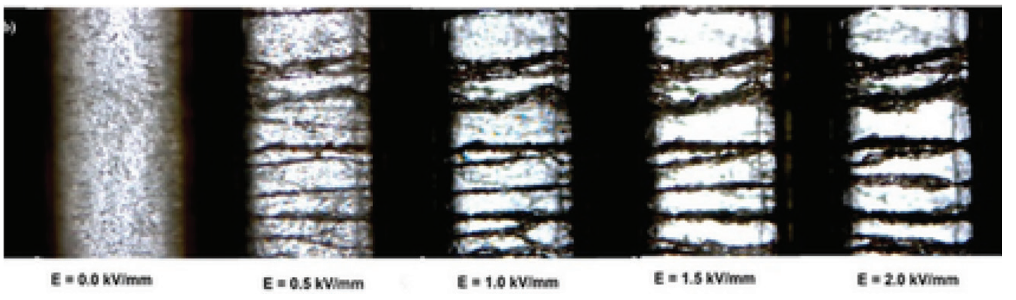

Figure 4.
OM images of fibrillation of dispersed solid nanocube-TiO_2/P_3OT particles [50].

(dimethyl siloxane), dibutyl sebacate, dioctyl phthalate, trioctyl trimellitate, and chlorinated paraffin have been used as dispersing phase [58].

ER fluids are divided into two categories as dry-base ER system and wet-base ER system. The dry-base ER system is anhydrous dispersion and shows ER activity without adding any polar promoter. The wet-base ER system is hydrous dispersion and needs a polar promoter to show ER activity. When these systems compare, dry-base ER system has more attention than wet-base ER system due to its physicochemical characteristics. Many materials such as conducting polymers, biopolymers, and polymer/clay nanocomposites exhibit dry-base ER properties [59, 60].

To enhance the ER effect and to increase the colloidal stability of the dispersions, various types of additives (water, ethyl alcohol, diethylene glycol, surfactants, etc.), called activators, have been added into the dispersions. They most certainly affect the particulate surface and the dispersing liquid.

Surfactants can be added to ER dispersions to improve ER activity and used to tailor dispersion properties. They effect the colloidal stability of dispersions and keep the particles from irreversibly flocculating. Also, they promote the rheological properties of dispersions in the absence an E. While some dispersions show little or no ER effect with adding of water or surfactant, some dispersions display improved ER response with water or surfactant [61, 62]. The adding of water as promoter has several problems such as the low of operation temperature, corrosion, and dispersion conductivity. Some surfactants used in ER dispersions are SDS, CTAB, Triton X, sodium oleate, glycerol, borax, and fatty amines [63–65].

Other additives into the ER dispersions effect the liquid phase by chancing the conductivity. The additives are generally used to increase the rheological characteristics of the dispersions. Some additives used in ER dispersions are water, NaCl, NaOH, acetic acid, amines, oxalic acid, diethylene glycol, glycerine [62]. In a study, the effect of water and surfactant was tested and both in rheological and electrical properties of the dispersions changed by adding small amounts of additives. The yield stress of the ER fluid increased with increasing water content. After the optimal water amount, a reduction in ER activity was observed. This tendency was explained by the water bridge model. On the other hand, the current density of ER fluid increased by adding water and Brij 30. ER activity of Brij 30-activated dispersions decreased under high E conditions. It can be attributed to the formation of surfactant droplets enclosing the particles [66] (**Table 1**).

Antisedimentation stability (or colloidal stability) is one of the important characteristics for the usability of ER dispersions at different environmental conditions. When charged particles in an ER fluid are dispersed, the particle interaction between dispersed particles increases, and rheological properties of ER fluid change. Surface charge of dispersed particles is determined by ζ-potential measurements. Therefore, ζ-potential can be used to determine the colloidal stability of ER fluids.

The density mismatch between the solid and fluid phases in an ER dispersion causes the sedimentation. It can be due to the particle aggregation through van der Walls interaction between the particles and the non-favorable particle-solvent interactions. As a result, the dispersed particles settle down according to Stokes' law [67]. To overcome this traditional problem in ER fluids, several solutions have been suggested.

Surfactants can be used to improve the colloidal stability of the ER fluids. They reduce the sedimentation and aggregation of dispersed particles in an ER fluid via adsorption on the particle surface. This method is still necessary for the long-term stability of ER dispersions. In a reported study, for the synthesis of graft copolymerization with aniline, tailor-made stabilizer was used, and polyaniline particle dispersions with improved stability were obtained [68]. In addition, porosity [69],

Properties	Magnitude
Zero-field viscosity (E = 0 kV/mm)	Low (<1 Pas)
High yield strength	τ = 5 kPa at E = 2 kV/mm
Short response time	<5 ms
Wide working temperature	(−30)–(+125°C)
Low density	d < 1–2 g/cm^3
Chemical resistance	High
Electric field resistance	High
ER materials	Inert, hydrophobic, and cheap
Particle size	<10 μm
Current density	<10 μA/cm^2 at E = 4 kV/mm

Table 1.
Preferred properties of an ideal ER fluid.

particle size [70], and apparent density [52] of dispersed particles play an important role on colloidal stability of the ER fluids.

For MR fluids, sedimentation is an important problem due to the limit of their technological applications. Several approaches to improving the colloidal stability of the dispersed magnetic particles have been suggested. They can be the dispersion stabilizers or additives, viscoplastic carrier medium, and the modification of magnetic particles with either polymers or inorganic coatings [71, 72]. Because of much lower density and relatively good magnetic properties than CI particles, Fe_2O_3 and Fe_3O_4 have been used as MR materials [73].

3.1 Dual response of materials under electric and magnetic fields

Basically the particles used to make MR suspensions cannot be used for ER systems because magnetic particles are more conductive; however, the magnetic particles can be coated with an insulating agent, which renders the suspension applicable in both MR and ER systems. The combined effect of magnetic and electric field produces intensified rheological changes in the suspensions. This synergic effect is termed as electromagnetorheological effect (EMR) [34]. **Figure 5** shows the flow curves of a reported study [74]. As shown, suspension behaves like a Newtonian fluid in the absence of an E or magnetic field, where the shear stress increases linearly with increasing shear rate. On the other hand, it acted as a Bingham fluid under an external E or magnetic field with a yield stress due to the formed particle cluster or chains by polarization forces.

The EMR technique is a new strategy to control the rheological properties of fluid materials. The rheological properties of some fluids can be enhanced with the combination of an electric field and a magnetic field [75]. At this technique, superimposed electric and magnetic fields are applied to a fluid, and the strength/direction of each field can be independently changed during the measurements. Also, the change of magnetic field direction is easier rather than that of the electric field. In a study, a comparison of EMR effects was carried out with the parallel-field and cross-field systems [76]. It was observed that a marked EMR effect was obtained with applied parallel field for the spherical iron dispersion. These EMR effect has been attributed to the different reorientations of the dispersed particles according to field directions. In a reported study, simultaneous impact of electric and magnetic fields on fluids containing a two-component dispersed phase (SiO_2 and CI) was investigated. The fluids showed the strong synergistic effect in the entire investigated range of intensities of electric and magnetic fields. Also, the ferromagnetic particles

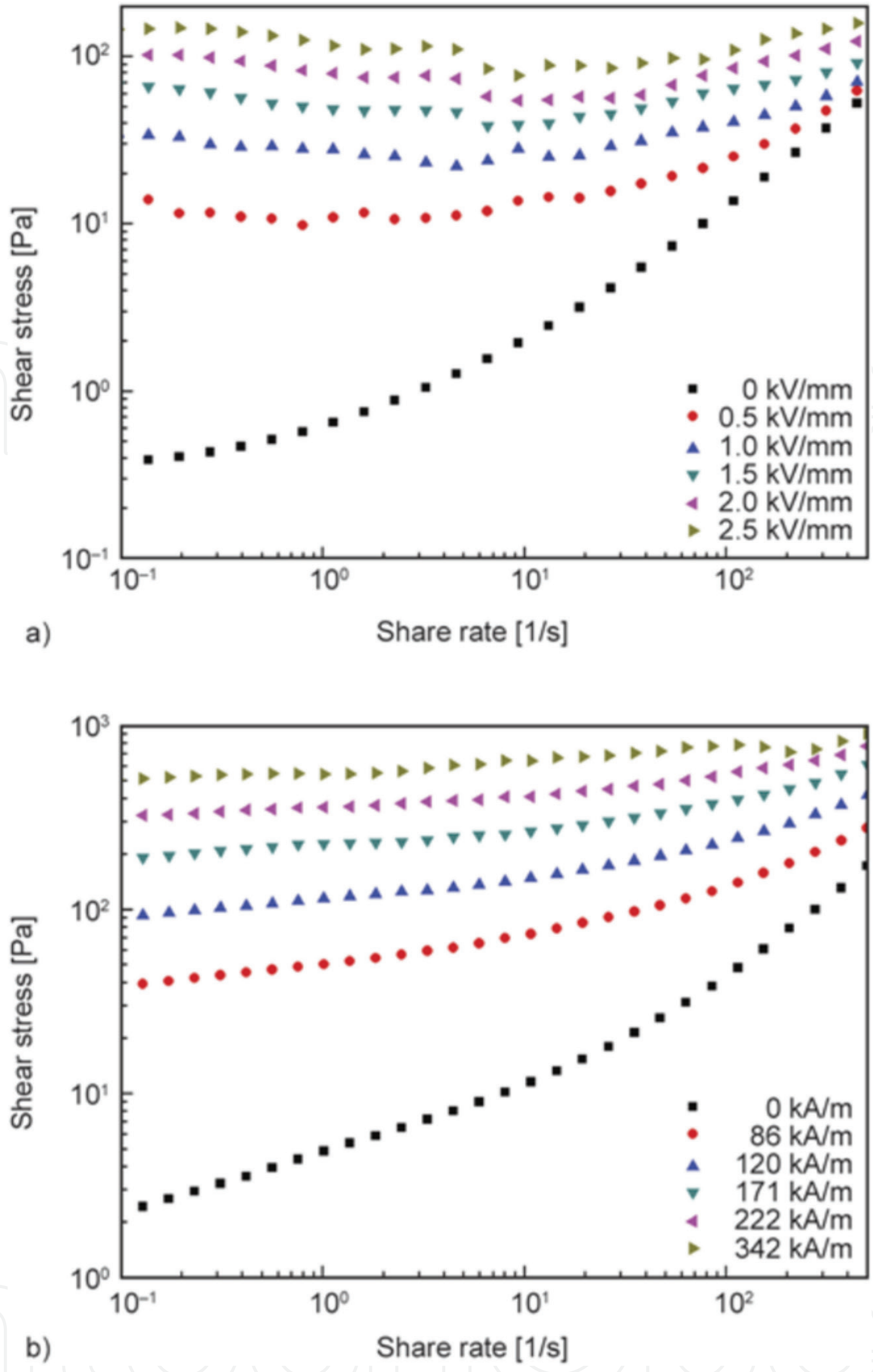

Figure 5.
Flow curves of CI@PANI/SO ER fluid under (a) electric and (b) magnetic fields [75].

(γ-Fe_2O_3) sensitive to the impact of an E was added to the fluids, and an increasing synergistic effect was observed until reaching saturation magnetization [77].

4. Conclusions

Various industrial applications such as the automotive, production sector, and robotic controls are full-potential ER and MR fluid applications. EMR fluids can change the particle interaction and the mechanical properties of dispersed materials. It means that they can allow an increase in the shear stress under two fields. Therefore, EMR fluids can expand the application areas of both ER and MR fluids due to their synergic effects.

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