

Electrorheology of silicone oil suspensions of urea-modified poly[(glycidyl methacrylate)-*co*-(ethylene dimethacrylate)] particles

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Abstract

Two methods of modification of poly[(glycidyl methacrylate)-*co*-(ethylene dimethacrylate)] microspheres with urea were examined in this electrorheological (ER) study. First, the urea functional groups were introduced by chemical modification of the starting copolymer. A significant increase in storage modulus of the silicone oil suspension was observed compared with the loss modulus when an electric field is applied. On the other hand, the ER effect in suspension of particles coated with physically bonded urea was only slightly higher than that of untreated material. This is explained by poor coating of the hydrophobic microparticle surface with hydrophilic urea. According to dielectric measurements, the interfacial polarization was found to be responsible for the formation of chain structure of modified microspheres in the electric field resulting in the ER effect.

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1. Introduction

Electrorheological (ER) fluids—typically suspensions of electrically polarizable particles in non-conducting liquid media—rank among smart materials whose rheological characteristics can be controlled through the external electric field application. This effect described by Winslow [1] more than 50 years ago has attracted continuing interest due to great potential engineering applications. Since that time many studies have been performed and main results were summarized in several reviews [2–7]. The mechanism of the effect is closely related to particle polarization. Thus, on application of the electric field, the polarized particles become organized into chain-like fibrous structure along the stream lines. Consequently, a rapid increase in viscoelasticity of the suspension occurs. At high polarization, the interparticle forces can form a quasi-stiff structure and the yield stress of the fluid may appear. When the electric field is switched off, the polarization of particles disappears and the structure of the fluid returns to its original state.

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It has been concluded that the ER phenomenon is primarily controlled by the magnitude of charge separation in the particles and the rate at which the polarization proceeds. Hence, the dielectric and conducting properties of suspension particles are the crucial factors affecting their activity in the electric field. In the present studies, various organic and inorganic materials of suspension particles have been proposed. Among them a remarkable ER efficiency of the suspension of nanosized barium titanate particles treated with urea solution has been described [8–10]. This behavior was ascribed to high particle polarizability of urea on the particle surface due to a large molecular dipole moment and high permittivity.

In our preceding studies [11,12], we demonstrated that the intensity of the ER effect of nanosilica suspension considerably increases after particle treatment with aqueous solution of urea. Unfortunately, the strong aggregation tendency of fragmental silica particles in the silicone oil caused high low-field suspension viscosity at relatively low particle concentrations. In this work, microspheres of macroporous poly[(glycidyl methacrylate)-*co*-(ethylene dimethacrylate)] used in our previous paper [13] was selected as a more suitable model of globular particles for further investigation of the effect of urea modification. In addition to a simple particle treatment with urea solution, urea molecules were also chemically bonded to the particle surface.

2. Experimental

2.1. Materials

Glycidyl methacrylate (GMA; Fluka, Germany), ethylene dimethacrylate (EDMA; Ugilior, France), and cyclohexanol (Lachema Brno, Czech Republic) were purified by distillation under reduced pressure in nitrogen atmosphere, 2,2'-azobis(isobutyronitrile) (AIBN; Ferrak, Berlin, Germany) was recrystallized from ethanol. Dodecan-1-ol (Fluka, Germany), poly(vinyl alcohol) (PVA; Polyviol W 25/140; $M_w = 80,000$; Wacker, Germany), poly(1-vinyl-2-pyrrolidone) (PVP; K90, $M_w = 360,000$; Fluka, Germany), and potassium cyanate (Aldrich, USA) were used as received.

2.2. Preparation of *P(GMA-co-EDMA)* microspheres

Conventional suspension polymerization was used to prepare macroporous poly[(glycidyl methacrylate-*co*-ethylene dimethacrylate)] (*P(GMA-co-EDMA)*) microspheres. The dispersed monomer phase consisted of 72 g GMA, 48 g EDMA, 177 g cyclohexanol, and 3 g dodecan-1-ol and 0.6 g of the AIBN initiator. Cyclohexanol, a thermodynamically good solvent for the polymer, and dodecan-1-ol, a thermodynamically poor solvent of the polymer, served as diluents (porogens) leaving pores in the polymer structure after their removal by washing [14]. Continuous phase was formed by 900 ml of aqueous 2 wt% PVA and 1 wt% PVP solution. Both solutions were loaded into a 1.5 l reactor, purged with nitrogen for 10 min to remove oxygen and the mixture was polymerized under stirring (400 rpm) for 16 h at 70 °C. The resulting microspheres (Fig. 1) were repeatedly washed with water, methanol, toluene, methanol, and finally air-classified with a Multi-Plex Labor Zickzacksichter A 100 MZR instrument (Alpine, Germany). Combination of two steric stabilizers PVA and PVP provided the particle size in the range 10–16 μm after classification (Sample S1).

2.3. Treatment of *P(GMA-co-EDMA)* particles in urea solution

Ten grams of *P(GMA-co-EDMA)* particles were immersed in 500 ml of distilled water at 40–50 °C. After 1 h, 10 ml of 10 wt% of aqueous solution of urea (Aldrich, USA) was added and the suspension stirred for another 16 h. The suspension was filtered off, washed with ethanol, and dried at 60 °C in vacuum to constant weight (Sample S2).

2.4. Modification of *P(GMA-co-EDMA)* with urea

The advantage of *P(GMA-co-EDMA)* copolymers consists in the presence of reactive oxirane groups of GMA units; they are able to react with various nucleophilic groups, for example, to undergo hydrolysis, ammonolysis, oxidation, reaction with acids, or isocyanates. *P(GMA-co-EDMA)* microspheres were first

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