

Electrorheological fluids based on polymer electrolytes

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Abstract

The phenomenon of electrorheological activity taking part in so called electrorheological fluids (ERFs) relies on strong and reversible changes of fluid viscosity upon application of electric field and finds interesting technical applications. ERFs typically comprise dispersions of polarisable solid particles in liquid matrices. The paper describes studies on complexes of polyacrylonitrile with various salts of alkaline elements. The materials in a powder form were dispersed in silicone oil as well as in active matrices containing a liquid crystalline polymer. It was found that these novel systems were substantially anhydrous and electrorheologically active. The observed ER effect was relatively high and accompanied by very low current consumption. The magnitude of the ER effect was correlated with bulk ionic conductivity of the studied materials. The optimal bulk conductivity giving the highest ER effect at reasonably low currents amounted to about 10^{-5} S/cm. Higher conductivities resulted in higher currents only and saturation of the yield stress values. It was also shown that dispersions of the polymer complexes in a solution of poly(*n*-hexyl isocyanate) in xylene manifested enhanced ER activity.

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1. Introduction

Electrorheological fluids (ERFs) belong to the extensively studied group of so called intelligent materials whose properties can be controlled by external factors. ERFs increase their viscosity or even solidify upon application of an electric field. These changes are fast and reversible and form a basis of numerous potential technical applications, especially in automotive, aerospace and medical industries [1]. In general, there are two main types of ERFs. The first and more common one comprises heterogeneous suspensions of fine polarisable solid particles in inert liquid matrices of low electric permittivity. A specific fibrous or lamellar microstructure is formed when the field is on and chains of particles bridge the electrodes contributing to the observed changes of the ER system rheology. The second type includes homogeneous solutions of low molecular weight [2] or polymeric [3,4] liquid crystalline substances as well as ferroelectric polymers

[5]. The observed increase of viscosity under electric field is caused by reorientation of mesogenic groups in parallel to the field. There are also some attempts to combine these two groups and prepare a hybrid ER fluid being a dispersion of solid particles in a liquid crystalline matrix [6,7]. The ER effect in heterogeneous and hybrid fluids is related to electric polarisation processes occurring in the bulk or on the surface of the dispersed grains, therefore, a proper choice of the properties of a solid is of great importance for the preparation of technically useful systems. A lot of solid materials have been investigated in order to determine their applicability for the ER technology. Most of them were inorganic or organic polar substances whose activity was promoted by water present in a system. Their commercial applications were, however, limited because of the narrow temperature range and undesirably high levels of conductance [8,9]. On the other hand, in recently developed anhydrous ER fluids consisting of semi-conducting polymers, such as poly(acenequinones) [10], polyaniline [11], poly(*p*-phenylene) [12] and pyrolysed polyacrylonitrile [13] the required polarisation is much less related to the presence of water, and it results from polar

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functional groups present on the grain surface or mobile ions that move within the solid state. The studies on polyelectrolyte (ion exchanging resins)-based ER systems with reduced amount of water are also worth to acknowledge here [14]. Unfortunately, these new systems are also often too conductive and many technical problems related to the ER technology still remain unsolved. Therefore, it is a great need to find some new substances manifesting high bulk or surface ionic polarisation at electric field without presence of water and even without a significant affinity to moisture.

Looking for novel systems of possible ER activity, we have turned our attention to water-free solid polymer electrolytes. Many types of these substances have been developed and characterised recently mainly because of their potential applications in electrochemical power sources, but only in a very few cases, their ER activity was investigated. Intrinsic mobility of ions in polymer electrolytes can result in interesting ER properties without a need of application of any promoters like water or other low molecular compounds. Taking into account such requirements like moderate level of conductivity, limited capability of absorbing moisture, possibility of preparation in a powder form and high thermal and environmental stability, we focused our interest on systems based on complexes of polyacrylonitrile with various salts.

The direct aim of our work was to synthesise polyacrylonitrile-based electrolytes, prepare ER fluids comprising these materials, test the obtained fluids and look for correlations between their rheological behaviour under electric field and material properties of the electrolytes.

2. Experimental

The commercial acrylic fibres (DRALON[®], 1.7DTX, Bayer AG) reported as a co-polymer of acrylonitrile (about 90%) and methyl methacrylate (about 10%) were used as an initial polymer for the ER fluids preparation. It was dissolved in dimethylformamide solutions of sodium (NaSCN) and potassium thiocyanate (KSCN), potassium perchlorate (KClO₄), lithium triflate (LiTF) and tetrabutylammonium iodide (TEBAD). The solvent was vacuum evaporated at 80 °C and foils containing 5, 10, 15 and 20% (m/m) of the salt were obtained. Their electric conductivities were calculated on the basis of impedance spectra taken on the cast out samples. Then, the foils were milled and dried again in high vacuum at ambient temperature over a period of 30 h. The obtained grains were irregular in shape and non-porous as can be seen in the example SEM image shown in Fig. 1. The grain-size distribution of each sample was estimated by means of picture analysis software on the basis of optical microscopy pictures taken for diluted suspensions. The typical distribution is shown in Fig. 2 proving that almost all of the particles were below 30 μm in diameter with most of them not exceeding a few micrometers. All prepared samples looked very similarly and no significant differences in their SEM images or grain-size distributions were found.

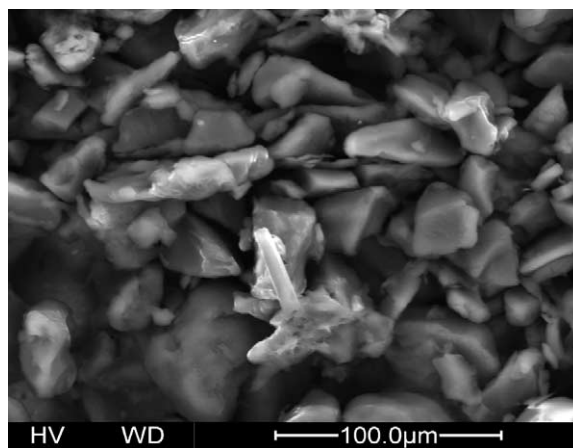


Fig. 1. SEM image of PAN/KSCN polymer electrolyte grains.

The liquid crystalline polymer – poly(*n*-hexyl isocyanate) (PHIC) – used for preparation of the active matrix ER fluids was obtained by anionic polymerisation of hexyl isocyanate in *N,N*-dimethylformamide/toluene mixture according to Aharoni and Walsh [15]. The reaction was carried out at –78 °C in presence of a NaCN catalyst and terminated with methanol. The product was purified by recrystallisation from toluene, vacuum dried for 48 h and analysed by ¹H NMR, GPC and FT-IR methods. The molecular weight of the polymer was $M_w = 76000$ with polydispersity M_w/M_n equal to 2.01.

To obtain the ER fluids weighted amounts of the prepared powders were dispersed in a dried silicone oil (polydimethylsiloxane, PDMS) of 0,1 Pa s viscosity or in xylene solution of PHIC (1.4%, w/w). Concentration of the solid phase in the all studied dispersions was constant and amounted to 15% (w/w).

ER properties of the suspensions were examined by means of a modified PC-controlled viscometer Bohlin Visco 88 operating in two concentric cylinders geometry and capable to apply dc fields up to 5.0 kV/mm. The gap between electrodes was 1 mm. Flow curves were recorded within the shear rate

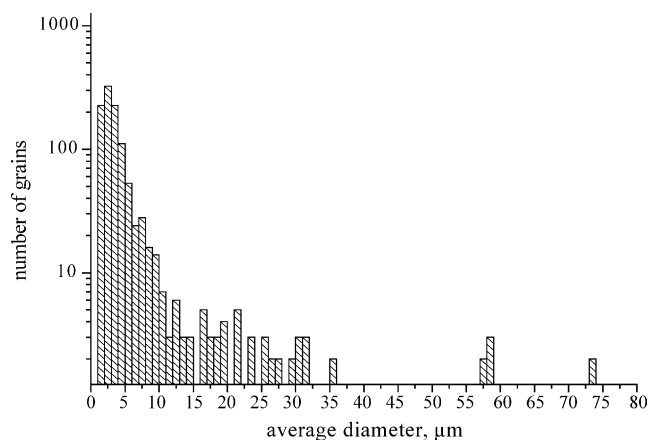


Fig. 2. Typical grain-size distribution of PAN/KSCN polymer electrolyte.

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