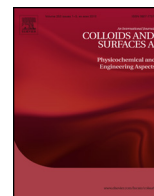




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A study of electric field-induced conductive aligned network formation in high structure carbon black/silicone oil fluids

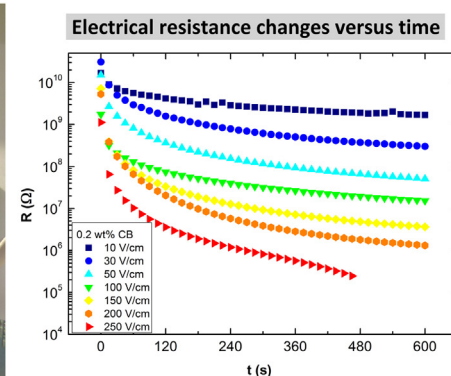
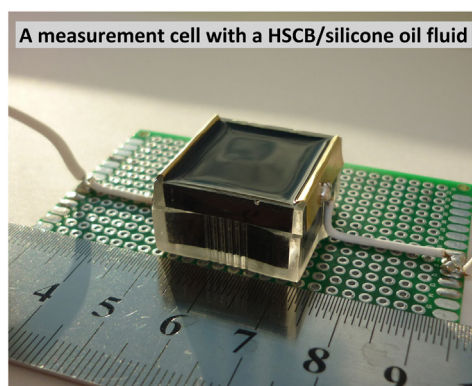
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HIGHLIGHTS

- Electric field induced 60 fold percolation threshold decrease has been observed for nanofluid.
- Tunneling-percolation model for description of nanofluid aligning process has been proposed.
- The model well describes electrical properties of aligned nanofluids.

GRAPHICAL ABSTRACT



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ABSTRACT

An electric field-induced alignment characteristics of high structure carbon black (HSCB) nanoparticles in silicone oil base fluids with different viscosities were investigated in this work. Resistivity change versus time of HSCB/silicone oil fluids with different concentrations of HSCB filler and different silicone oil viscosities was determined at different electric field strengths. Measurement results were analyzed using percolation theory by calculating critical concentration at which rapid increase of conductivity appears. An analytic model based on tunneling percolation theory was developed and successfully applied to explain the experimental results. The obtained results potentially can be used in the development of polymer composites with aligned nanoparticle structure.

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1. Introduction

Fluids [1–5] and polymers [6–20] filled with electroconductive nano- or microparticles and processed with electric field have drawn attention of scientists for years. It has resulted in both

theoretical modelling attempts [2–6,18] as well as experiments on the alignment of conductive particles in liquids using electric field [1–3,5–17]. Different theoretical models based on combinations of thermodynamics, classical percolation theory, colloidal particle kinetics, electrostatics as well as electroforetics have been developed to describe the conductive nano- and microparticle alignment. Tai et al. [6] researched dynamics of an electric field-induced carbon black (CB) particle alignment in a low-density polyethylene (LDPE) matrix. CB/LDPE samples were put into a

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high temperature cell and DC electric fields of various strengths were applied. Time dependence of the electrical resistance for different electric field strengths was measured during isothermal processing at different annealing temperatures and subsequently for different viscosities of melt. During the isothermal process CB particles aggregated and formed conductive network. Authors [6] based their theoretical considerations on a description of the kinetics of colloidal particle coagulation, the thermodynamics of an equilibrium state, and a classical percolation theory. The percolation concentration for a thermodynamic equilibrium state as well as the electric field strength for a transition from a three-dimensional isotropic network to a one-dimensional chain was calculated. Authors concluded that the external electric field greatly accelerated the conductive network formation [6].

Oliva-Alviles et al. [2] experimentally investigated two suspensions based on two liquids with different viscosities (deionized water and polysulfone solution in chloroform) where multi-walled carbon nanotubes (MWCNTs) were first dispersed and then aligned using both a DC and an AC field. The influence of the electric field strength and frequency on the MWCNT network formation was studied. Migration of the MWCNTs towards the negative electrode was observed while the DC electric field was applied whereas only formation of the aligned MWCNT network was obtained while applying the AC electric field. An increase of the electric field frequency facilitated a faster formation of the aligned MWCNT network and created thinner MWCNT bundles. Higher viscosity of the liquid medium resulted in slower MWCNT alignment [2]. An analytic model based on the electrophoresis-induced torque and which accounts for the viscosity of the medium was proposed to explain the dynamics of the MWCNT alignment. This model was further advanced by the same authors in their recent solely theoretical work [3,4] where two-dimensional coupled rotational and translational motion of interacting carbon nanotubes (CNTs) in an AC electric field was theoretically investigated. In their work [3] three mechanisms have been modelled: the CNT rotation in the electric field, the translational motion due to the CNT-to-CNT Coulombic interaction, and the CNT migration towards an electrode. The coupling between two adjacent CNTs is achieved by the interacting Coulombic forces and torques. Results of the modelling showed that the rotational motion of the CNTs occurs faster than the translational motion rendering first a fast alignment with the direction of the electric field followed by a collinear translation of the CNTs until a tip-to-tip “contact” occurs [3,4].

Lima et al. [1] used DC electrical conductivity measurements to investigate the alignment of CNTs in chloroform suspensions. Based on classical percolation theory authors described the formation of a dynamic percolating network [19,20] and concluded that the measurements of the electrical properties of the chloroform/CNT suspensions can be used as a new method for CNT characterization. They found a good correlation with the results obtained by methods commonly used for CNT characterization such as Raman spectroscopy, transmission electron microscopy and electrical conductivity measurements of free standing CNTs [1].

Matzui et al. [17] using in situ optical microscopy while curing a MWCNTs/epoxy mixture in an electric field found that both AC and DC fields induce a formation of aligned carbon nanotube networks and that the resistivity of the MWCNTs/epoxy composite is about 5–6 orders of magnitude lower than when curing without the application of the AC electric field. Authors explained the observed aligning effect with direct CNT-CNT Coulombic interactions in the lateral direction and a formation of bundles, which arises due to the movement of adjacent nanotubes closer to each other because of dipole–dipole interactions. In recent work, Matzui et al. [18] investigated the effect of filler alignment on conductive and shielding properties of MWCNTs/epoxy nanocomposites containing 0.05–1 wt% of MWCNTs. The filler alignment was induced

by applying an external AC electric field during the hardening phase of the polymer. Two percolation thresholds with different physical origins were identified for the nanocomposites. The appearance of the upper threshold (at 1.45 wt% of MWCNTs) was explained by a statistical percolation theory. The existence of the lower threshold (at 0.45 wt% of MWCNTs) was substantiated by dynamic or kinetic percolation, which is determined by the parameters of the MWCNTs, the characteristics of the electric field, and the viscosity of the polymer matrix. It was found that the timescale of the CNT rotation is about 10 s and is independent of the CNT concentration. It is more than two orders of magnitude shorter than the characteristic time scales of the CNT-to-CNT Coulombic interactions and the CNT migration towards an electrode [3,17].

Dimaki et al. [19] obtained highly ordered networks of single-walled carbon nanotubes (SWCNTs) on chips with a microelectrode arrays using dielectrophoresis as an assembly method. Each chip contained two identical and individually connected structures with 14 equally spaced (3 μm gap) electrode pairs. A 3 μl droplet of SWCNTs/aqueous sodium dodecyl sulfate solution was placed on top of the chip. Then a 14.5 V_{pp} AC voltage was applied for 2 min using four different frequencies: 10 kHz, 100 kHz, 1 MHz and 10 MHz. It was found that at higher frequencies the alignment of the nanotubes was more pronounced.

Knaapila et al. [20] published a comprehensive review about main aspects of preparation and properties of string-like carbon particle assemblies using dielectrophoresis in alternating electric fields.

In our recent study [21] the development of a percolative electro-conductive structure in a polyisoprene/nanostructured carbon composite during vulcanisation and without an application of aligning electric field was studied (weak electric field was applied to enable resistance measurements). It was found that during the vulcanisation phase (at constant temperature) a dynamic percolation process takes place and the conductivity of the composite increases by seven orders of magnitude.

Our current paper is focussed on studies of DC electric field-induced alignment of HSCB in silicone oil. Due to the elongated form of the primary particle aggregates, their graphitised structure and, consequently, high electrical conductivity [22] HSCB is an excellent choice of filler for this purpose. HSCB/silicone oil fluids were chosen as an inexpensive convenient system for rapid and repeatable measurements. The time dependencies of electrical resistance for different silicone oil viscosities and HSCB concentrations are measured. The percolation thresholds for different strengths of the electric field and different HSCB concentrations with constant viscosity of the silicone oil are calculated. An analytic model based on a tunneling percolation theory is developed. In this model change of the tunneling current intensity between adjacent conductive nanoparticles during the alignment process has been taken into account. The acquired results could potentially be used for the development of solid HSCB/polymer composites with anisotropic properties.

2. Materials and methods

Three types of silicone oil (AK100, AK500 and AK1000 from Wacker Chemie AG) with different viscosities (100, 500 and 1000 mPa·s, respectively) were used as base fluids to prepare the HSCB/silicone oil fluids. HSCB (Printex EX2, obtained from Degussa AG) with an average particle size of 30 nm (surface area 950 m²/g) was used as an electroconductive filler. The fluids were prepared in the following way. First HSCB was crushed in a mortar to pulverize any inherent agglomerates. Silicone oil was put into a beaker and stirred with a magnetic stirrer. While stirring the silicone oil, the necessary amount of the pulverized HSCB was gradually added to

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