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Computational modeling of structure formation during dielectrophoresis in particulate composites

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In this paper a novel numerical model is proposed for qualitative simulation of the structure formation during dielectrophoresis in a medium of dielectric particles suspended in a liquid between parallel-plate electrodes. Upon application of an electric field particles reorient with respect to the imposed electric field, followed by rotational and axial displacement of particles interaction until chains are formed. The performance of the model is illustrated in a number of fundamental cases. The influence of parameters such as size, aspect ratio and heterogeneity of the particles is studied for the purpose of obtaining insight in the ideal conditions required to obtain the desired structure. In a multi-particle system, the relative particle size is shown to be a key parameter in chain evolution process. The quality of the structure evolution is investigated using a set of geometric parameters. A new topological parameter, chain perfection degree, relating the microstructure to the overall performance of the piezoelectric composite, is proposed and calculated for the fundamental case-studies.

1. Introduction

Piezoelectric ceramics widely used in sensing, actuation and energy harvesting applications due to their high permittivity, thermal stability, and electromechanical coupling suffer from inherent brittleness. Furthermore they are difficult to manufacture which limits their use for applications in which mechanical flexibility is required. Ductile and flexible piezoelectric polymers which have a modest sensitivity and a low thermal stability are found in less demanding applications. By embedding piezoelectric ceramics in a polymer matrix one can combine the superior piezoelectric and mechanical properties allowing for high piezoelectric coupling characteristics with increased flexibility and toughness. In such multiphase composites the volume fraction of the phases, their morphology, the pattern of connectivity as well as the dielectric and electrical properties of the two phases control the overall physical and electromechanical properties [1–3]. Newnham et al. [4] distinguished 9 different types of connectivity. The random particle distribution, 0–3 composites with unconnected equiaxed particles in a fully self-connected matrix, and a fibrous distribution, 1–3 composites with continuous ceramic rods fully aligned in one dimension, present the lower and upper bounds as far as the final properties of the composites are concerned. An intermediate state between the 0–3 (particulate) and 1–3 (fibrous) state can be obtained by dielectrophoresis (DEP) treatment on a semi-dilute solution of particles in a viscous matrix (such as the most polymer systems prior to curing) [1,2,5]. As a result of the local electromechanical field the particles reorient and reposition themselves in thread like structures. Under ideal processing conditions, long and well separated particulate threads form which even span the full electrode to electrode spacing, closely approaching the topology of perfect 1–3 composites. For a given particulate concentration the alignment of the particles in the thread like structure leads to a marked increase in piezoelectric properties while the high mechanical flexibility due to the polymer matrix is maintained [2,6–8]. The quality of particle alignment in the thread like structure is the key parameter in controlling the overall properties of the composites [8–11]. Enhancing the alignment quality by means of decreasing the inter-particle distance has been shown to significantly improve the dielectric, piezoelectric and pyroelectric properties of particulate composites [5]. The effect of processing parameters on thread formation is experimentally illustrated for high aspect ratio particles aligned in a thermoset matrix [12]. The orientation angle of the particles in the DEP composites has

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been shown to be dependent on the applied electric field, time, and aspect ratio of the particles. To date all the research into DEP structuring of piezoceramic particulate-polymer composites has been of an experimental nature. Furthermore the quality of the structure formation has only been determined after consolidation of the polymer matrix and additional processing. It is the aim of the present paper to present a 2D computational model for the structuring of particulate composites when the matrix is still in its fluid state. The performance of the model is illustrated for a number of fundamental cases. The influence of parameters such as size, aspect ratio and heterogeneity of the particles on the thread or chain formation is analyzed. The quality of the chain like structure created is investigated using a set of geometric parameters, defined as a chain perfection degree, that correlates to the overall performance of the piezoelectric composite.

2. Dielectrophoresis modeling

2.1. Background

The dielectrophoretic force is defined as the action experienced by a polarized particle immersed in an electric field $\mathbf{E}$. In the ideal case that the particle can be modeled by means of an infinitesimal dipole with moment $\mathbf{p}$ it will experience a force

$$\mathbf{F} = \mathbf{p} \cdot \nabla \mathbf{E} \tag{1}$$

and a torque

$$\mathbf{M} = \mathbf{p} \times \mathbf{E}. \tag{2}$$

Further to the infinitesimal dipole idealization, these equations are based on the assumption that the electric field itself is not influenced by the dipole [13,14].

For the case of structure formation during dielectrophoresis, however, none of the above assumptions hold. Particles need to be viewed as finite domains with an own polarization field $\mathbf{P}$ that will contribute to the electric field $\mathbf{E}$ in the surrounding medium. This contribution will also induce a force on neighboring particles. This situation can be treated analytically by means of higher-order multipoles [13,15–17]. Alternatively, and especially for the purpose of simulation of structure forming with several particles involved in which emphasis is given to performance the qualitative behavior of the system rather to the accuracy of the results, numerical methods such as finite elements provide a meaningful alternative.

The current study is limited to a two-dimensional rectangular domain, which can be viewed as an idealization of a thin three-dimensional quadrangular domain Ω^l filled with a viscous liquid with dielectric permittivity ε^l and viscosity η, confined between two plates separated by a distance d and containing a single layer of particles $\{\Omega_i^p\}_{i=1}^n$ with dielectric permittivity ε^p and mass density γ restricted to move on the mid-plane. The particles have thickness l. This renders a computationally treatable model of an experimental setup [2] used to observe structure forming in dielectrophoretic manufacturing of piezoelectric polymers. A schematic representation of the geometry of this model is given in Figs. 1 and 2.

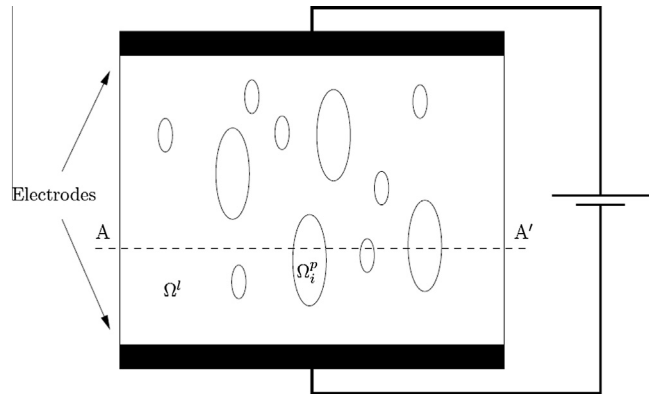

Fig. 1. Schematic representation of the geometrical model.

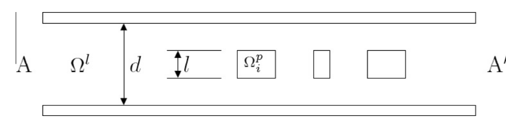

Fig. 2. Cross section of the geometrical model along line AA′ of Fig. 1.

2.2. Mechanical modeling

In the context of the simplification introduced in Section 2.1 the particles Ω_i^p can be modeled as linear elastic solids in plane-stress conditions. The motion of each individual particle is obtained by solving Cauchy equation

$$\nabla \cdot \mathbf{T} + \mathbf{b} = \gamma \ddot{\mathbf{u}} \qquad \text{in } \Omega_i^p; \tag{3}$$

$$\mathbf{Tn} = \mathbf{t} \qquad \text{on } \partial\Omega_i^p, \tag{4}$$

where $\mathbf{T}$ is the stress tensor, $\mathbf{b}$ is the body force density, γ is the mass density, $\mathbf{u}$ is the displacement field, $\mathbf{t}$ is the boundary traction and $\mathbf{n}$ is the outward normal vector at the boundary $\partial\Omega_i^p$. For a proper description of the particle motion geometrical nonlinearity is considered.

The body force $\mathbf{b}$ consists of a dielectrophoretic contribution $\mathbf{b}_d$ and an equivalent viscous drag term $\mathbf{b}_v$ while the boundary traction $\mathbf{t}$ is of a purely dielectrophoretic nature. These forces will be characterized in Sections 2.3 and 2.4.

2.3. Dielectrophoretic force

Laplace equation

$$\nabla^2 V = 0 \tag{5}$$

is solved for the potential V in $\bigcup_{i=1}^n \Omega_i^p \cup \Omega^l$ with adequate boundary conditions to obtain the electric field $\mathbf{E}$ and the polarization field $\mathbf{P}$ as derived quantities. This polarization is associated to the bound volume and surface charge densities in the particles according to the following expressions

$$\rho = -\nabla \cdot \mathbf{P} \qquad \text{in } \Omega_i^p; \tag{6}$$

$$\sigma = \mathbf{P} \cdot \mathbf{n} \qquad \text{on } \partial\Omega_i^p. \tag{7}$$

From these charge densities dielectrophoretic body and surface forces are computed as

$$\mathbf{b}_d = \rho\mathbf{E} \qquad \text{and} \qquad \mathbf{t} = \sigma\mathbf{E}. \tag{8}$$

2.4. Viscous drag

As the particles $\{\Omega_i^p\}_{i=1}^n$ are immersed in a fluid they will experience viscous drag upon motion. A simplified model is adopted for this purpose. It is assumed that the fluid interacts with the particle along the top and bottom flat surfaces and that the rate of shear is linear between the particle and the upper and lower confining plates, see Fig. 3. If a Newtonian fluid is considered this leads to a distributed shearing force on the surface amounting to

$$\tau = \eta \frac{v}{(d-l)/2}. \tag{9}$$