A TRANSIENT ANALYSIS FOR THE ELECTRODEFORMATION OF DROPLETS AND VESICLES

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Summary A transient analysis for prolate droplet and vesicle deformation under dc electric fields is presented. The Taylor-Melcher leaky dielectric model is adopted, and the equations are written in a rotational spheroidal coordinate system to allow solution and reduction into an ODE governing the evolution of the aspect ratio. For both droplets and vesicles, the results are compared with experimental data to reveal good quantitative agreements.

INTRODUCTION

When subject to an applied electric field, a droplet undergoes deformation due to the electrostatic stresses exerted on the interface. Research on electrohydrodynamic (EHD) deformation of droplets has wide industrial applications including EHD atomization, oil purification etc. Historically, this phenomenon has been extensively investigated, and steady-state solutions have been obtained for both small and large deformations [1, 2] using the Taylor-Melcher leaky dielectric model. However, to capture the transient dynamics, especially in the large deformation regime, numerical simulations had to be employed [3].

A similar problem is the electrodeformation of vesicles, where a lipid membrane separating two fluids phases needs to be considered in addition. This phenomenon can be harnessed to effectively probe membrane structures and properties, and has received broad attention in the past decades. A large amount of quantitative data has been generated in recent years, capturing dynamic and steady-state responses of vesicles under both ac and dc electric fields [4]. In contrast, theoretical developments are relatively few. Vlahovska *et al.* developed a small-deformation theory for vesicles under ac electric fields [5], whereas one of us (HL) presented a semi-empirical model for large deformations [6].

For both cases, a comprehensive transient analysis is presently lacking, and is much needed. The development of such a model can generate valuable results to directly compare with experimental data, to provide physical insights and in-depth understanding. In this work, we present such an analysis for both droplet and vesicle deformations. Following the Bentenitis and Krause [2], we assume that the droplets and vesicles are prolate spheroids such that the problems can be solved under a rotational spheroidal coordinate transformation. Our results provide good agreements with both numerical simulations and experimental measurements.

ANALYTICAL SOLUTION

We first develop a transient analysis for droplet deformation. The droplet is assumed to be spheroidal in shape with volume conservation. Under these assumptions, the droplet can be characterized with a single parameter, namely, the aspect ratio of the primary to secondary axes. The droplet interface separates two fluids phases with arbitrary properties including viscosity, permittivity, and conductivity. The Taylor-Melcher leaky dielectric model is adopted, where the hydrodynamic problem (the Stokes equations) and the electrical problem (the Ohmic equation) can be solved separately, and then coupled at the interface with stress (including surface tension) matching conditions. The equations are recast into a rotational spheroidal coordinate system, and reduced to the single ODE governing the change of the aspect ratio as a function of time.

The vesicle problem is dealt with in a similar manner, with the additional consideration of the lipid membrane. The membrane is considered to be both impermeable and non-conductible, and both a capacitive charging and a mechanical model [7] are implemented. The latter replaces the surface tension in the droplet model and employs properties such as initial tension, bending rigidity, and elastic modulus. The solution leads to two ODEs governing the aspect ratio and the transmembrane potential. The details of the derivations are presented in [8, 9].

RESULTS

We present exemplary results on both droplet and vesicle electrodeformations, and directly compare them with available experimental data [10, 11]. Fig. 1*a* shows the evolution of the aspect ratio, *a/b*, as a function of time for a water-ethanol droplet immersed in silicone oil subject to a constant electric field of 4.5 kV/cm in strength. The exterior-to-interior ratio of the conductivity, permittivity, and viscosity are 10^{-5}, 0.05, and 23.3, respectively. Good quantitative agreements are found between our theoretical predictions and experimental data. The process is demonstrated to be highly nonlinear. The aspect ratio grows rapidly followed by a plateau. At the final stage, an exponential growth occurs, indicating droplet breakup. The entire process is well captured by our model.

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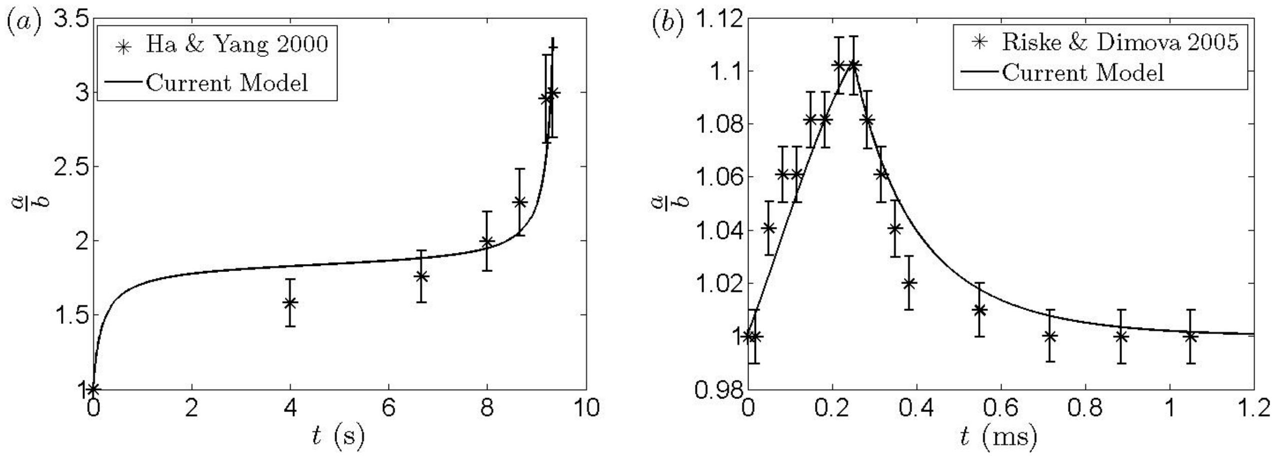

FIGURE 1. COMPARISON OF MODEL PREDICTION WITH EXPERIMENTAL DATA FOR (a) DROPLET, AND (b) VESICLE ELECTRODEFORMATION.

Fig. 1*b* shows the time-evolution of an L-α phosphatidylcholine vesicle under a finite electric pulse of 1 kV/cm in strength and 250 μs in duration. The extravesicular and intravesicular conductivities are 4.5×10^{-4} S/m and 6×10^{-4} S/m, respectively. The permittivities and viscosities are all taken to be those of water. For the membrane properties, the initial tension is 1.5×10^{-4} N/m, the bending rigidity is 2.47×10^{-20} J, and the elastic modulus is 0.14 N/m. Again, we found that the model quantitatively captures the evolution of the aspect ratio, including both the growth during the pulse, and the relaxation post-pulsation. In addition, the model provides useful insights on the timescales involved: we found that the two separate timescales discussed by Dimova *et al.* [4] can be collapsed into a single one related with viscosity, initial tension, initial vesicle radius, and bending rigidity.

In brief summary, our model is capable of predicting both droplet and vesicle electrodeformation with quantitative accuracy. Our results are validated by comparisons with both numerical simulations (not shown here) and experimental data, and are subject to further analysis to review useful insights on the physical mechanisms of both phenomena. This work is an important step toward a quantitative prediction for complex, EHD-driven processes.

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