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ABSTRACT

Transient currents of reverse micelles in a non-polar solvent from voltage step stimuli were studied to investigate the electrophoretic behavior of the charges. It showed various time-dependent transients depending on the applied voltage and the charge content. A one-dimensional drift-diffusion model could reproduce the behaviors for various conditions.

1. INTRODUCTION

Electrophoretic control of charged particles has been used for reflective displays and is getting increasing attention. Since charge motion results in an electric current, the transient current is a useful tool to understand how the charges move in the display cell. Previous efforts [1-3] to interpret the transient current behaviors and to extract useful information have not resulted in a thorough understanding of all the phenomena. In this talk we will present transient current measurement data for various voltages and surfactant concentrations and compare them with numerical simulations of a one-dimensional model.

2. EXPERIMENTS

Dispersions of poly-isobutylene succinimide (OLOA11000, Chevron) in an iso-paraffinic solvent in various concentrations were used as model electrophoretic fluids. According to a popular interpretation, reverse micelles formed by the OLOA11000 disproportionate into the charged species in this system. The test cells used had a parallel-plates geometry. The current through the test cells was measured after application and removal of a voltage step of various magnitudes.

3. NUMERICAL MODELING

A one-dimensional model has been built and solved numerically using COMSOL Multiphysics. The charge flux is a sum of the drift and diffusion contributions. The concentration of i -th species, c_i , satisfies the local mass conservation equation, $\partial c_i / \partial t + \nabla \cdot (-D_i \nabla c_i - (\mu_i \nabla \phi) c_i) = 0$, where D_i is the diffusion coefficient, μ_i the electrophoretic mobility, and ϕ the electrostatic potential. The electrostatic potential is governed by the Poisson

equation, $-\varepsilon_0 \varepsilon_r \nabla \cdot \nabla \phi = q(c_+ - c_-)$, where ε_0 is the dielectric constant of vacuum, ε_r the relative dielectric constant of the solution, and q the elementary charge. It is assumed that the mobilities of the positive and the negative charges are identical and that each charge has one elementary charge only. Linear elastic compression is assumed for charges closer than 50 nm to one another.

4. RESULTS AND DISCUSSION

Fig.1 shows transient currents from 3 wt% OLOA11000 dispersion for various voltages. After the voltage is applied, the current shows a sharp peak with a height proportional to the voltage and then decreases to zero. After 5 seconds the voltage is brought down to zero volts. The reverse transient current that follows shows a different behavior from the forward transient depending on the magnitude of the step voltage. When the voltage is lower than 0.5 V, the reverse current decays monotonically in a similar way to the forward current. At step voltages higher than 0.5 V the reverse current develops a broad hump. The peak of the hump shifts to later times as the voltage increases up to 1 V. After that, the shape and the position of the peak does not change significantly. Previous attempts to explain the origin of the delayed hump [1,2] were not conclusive.

The one-dimensional drift-diffusion model has been solved for a system with various amounts of charge and a given electrophoretic mobility. All important phenomena in the transient current of the experiments have been reproduced, including the delayed hump in the reverse transient as shown in Fig. 2.

The forward transient behavior can be easily explained by a series resistor-capacitor model with a limited charge content. At the early stage of the transition all charges in the cell are moved by the applied electric field, which produces a high peak current with a magnitude proportional to the voltage. As the charges arrive at the collecting electrodes, they develop space charge layers that generate an electric field against the applied field, which lowers the driving electric field in the middle of the cell. The accumulated charges do not contribute to the current anymore because the diffusion flux developed by the accumulated charge balances the

electrophoretic drift. The current decreases to zero either when the space charge completely screens the applied field at low voltages, or when all mobile charges are accumulated at the electrodes at high voltages. At equilibrium, the diffusion and drift fluxes completely balance each other throughout the length of the cell.

When the applied voltage is removed (at $t = 5$ s), the balance between the drift and diffusion is broken, which gives rise to a reverse current. The driving force for the drift is the electric field generated by the accumulated charges. At the early stages of the reversal the current from the charges moving away from the electrodes is small because most of the charges are still held near the electrodes where they are being pushed against the electrodes by local electric fields generated by the spatial distribution of the charges, as shown in Fig. 3 ($t = 5.01$ s). The charges diffuse toward the cell center where they are more susceptible to the local electric field that pushes them toward the opposite electrodes ($t = 5.43$ s), which results in increase in the reverse current. As charges of two opposite polarities mix in the bulk of the cell, the space charge, which is the main driving force of the reverse transient, diminishes with time. The charge distribution becomes more even throughout the cell, which also reduces the diffusion fluxes. Fig. 6 shows the drift flux which is the product of the local electric field and the charge concentration at the center of the cell. This agrees closely with the external current. In the early stage of the reversal the increase in current is mainly driven by the increase in the charge concentration rather than by the change in the local electric field. In the later stage ($t > 5.43$ s), the current decreases because the electric field decreases faster than the charge concentration increases.

When the applied voltage is not large enough to polarize the entire population of charged species, a significant amount of charge is left in the middle of the cell. As a result, it can contribute to the electrophoretic drift from the beginning of the reversal, which produces a high initial reverse current. ($V = 0.25 - 0.5$ V in Fig.4(a) and (b)) As the polarization becomes more complete with increasing bias, less charge is available for the initial drift and it takes longer to reach the peak current. ($V = 0.75 - 1.25$ V in Fig.4(a) and (b)) However, once the voltage reaches the value that is sufficient to polarize the entire charge in the system, no further increase in the bias will change the charge distribution in the cell. The reverse current response stops changing because the entire process is determined by the initial charge distribution profile. ($V = 1.25 - 2$ V in Fig.4(a) and (b)) Therefore, the degree of the reverse peak development is a good measure of the completeness of polarization.

Individual charge interaction plays an important role in charge compaction at the electrodes. Though the model describes the overall behavior very well, it starts to deviate from the experimental data if the charge-to-charge

interaction is ignored. As seen in Fig.4 and Fig.5, the reverse peak positions appear earlier than the model without inter-charge interaction predicts. This is because the model assumes point charges, which permits unrealistically high local charge concentrations in the vicinity of the electrodes. This makes it underestimate the electric field buildup and predict complete polarization at a lower voltage than reality. And the lower electric field estimation from the space charge slows down the rise of the reverse drift, which ends up with the shift of the peak position to a later time. This error gets worse as the charge content increases as in Fig.5. The model with point-charge assumption predicts later appearance of the peak as the total charge content increases while the experimental data shows that the peak stops shifting as the charge concentration increases higher than 1.3×10^{-5} C/cm³. However, when linear elastic compression is assumed when the charges are compacted closer than 50nm to one another, (Fig.4(a) and Fig.5(a)) the model fits the experimental results very closely. The charge concentrations used in the model was determined from experimental data according to the method described below and the electrophoretic mobility was calculated from the conductivity from the initial forward transient current using the charge concentration.

Under complete polarization, the time integral of the forward or reverse transient current is equal to the half of the total charge content (Fig. 7). Therefore, one can easily estimate the charge content by integrating the experimental data under a sufficiently large voltage. The experimental data of the 3 wt% OLOA11000 dispersion showed the exact same trend as predicted by the model (Fig. 7) for which the single polarity charge concentration of 4.6×10^{-5} C/cm³ is obtained. Using the conductivity of 4.6×10^{-10} S/cm obtained from the initial forward currents, which is in agreement with impedance spectra measurements, the mobility of the charge carriers is estimated as 1.0×10^{-5} cm²/Vs. A good agreement between the model and the experimental results confirms the absence of any significant charge generation or charge injection, as assumed in the model.

5. SUMMARY

Step-voltage transient current responses of surfactant dispersions have been measured and analyzed. At step voltages above a certain value, the reverse current shows a broad hump after voltage is removed. The behavior can be reproduced by a one-dimensional drift-diffusion model. It is explained as a competition between an increasing number of charges available for drift and a decreasing electric field resulting from mixing of opposite charges. From the measurement data the total charge content and the electrophoretic mobility has been determined.

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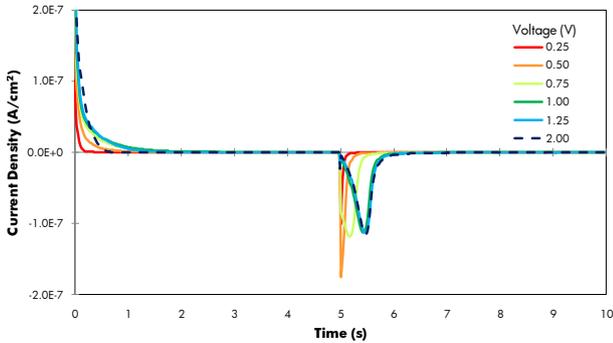


Fig. 1 Transient current responses of 3 wt% OLOA11000 dispersion

The currents were measured with parallel-plates electrodes. The distance between the electrodes was 1×10^{-3} cm.

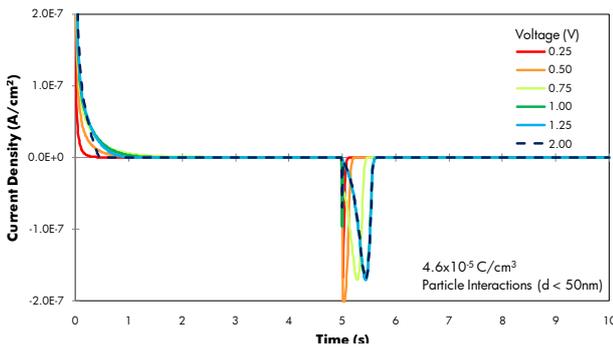


Fig. 2 Transient currents from 1-D drift-diffusion model
Parameters used: total charge concentration 4.6×10^{-5} C/cm³, charge mobility 1×10^{-5} cm²/Vs, electrode spacing 1×10^{-3} cm, elastic particle interactions when closer than 50nm

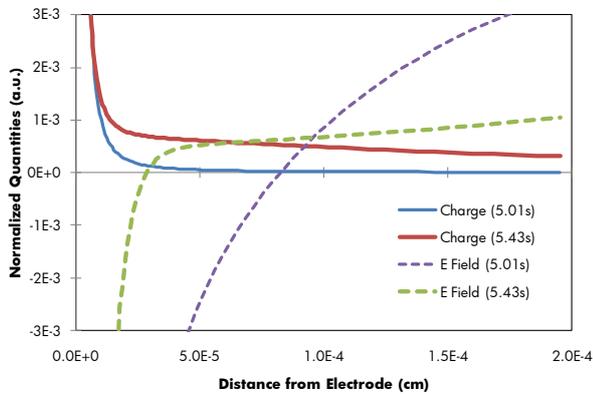


Fig. 3 Normalized positive charge concentration and electric field near an electrode

Same parameters as Fig. 2. The applied voltage is 2 V.

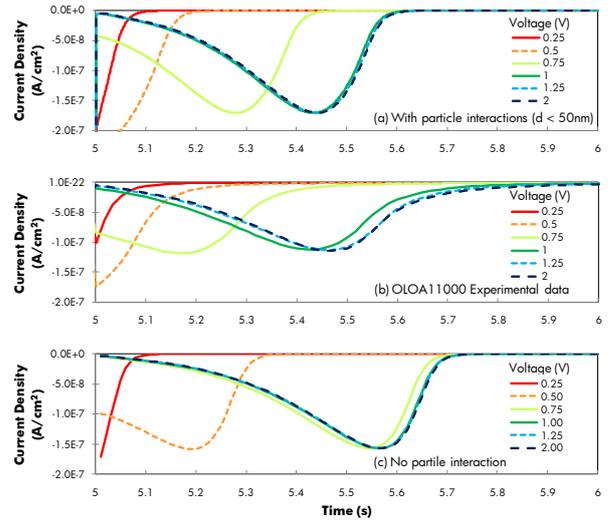


Fig. 4 Reverse transient currents from experiments and models for various applied voltages

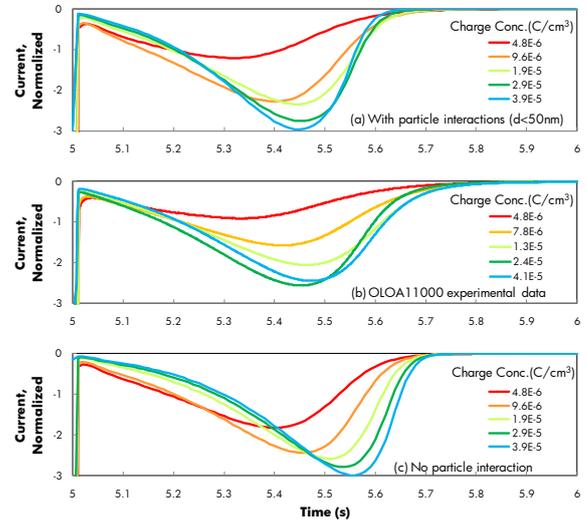


Fig. 5 Reverse transient currents from experiments and models for various charge concentrations

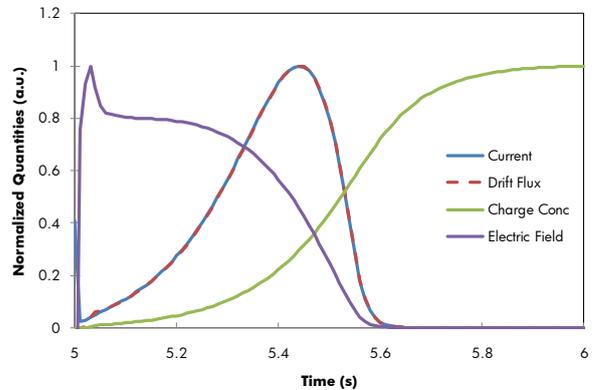


Fig. 6 Two major contributions to reverse transient current behavior

The reverse transient current is compared with the drift flux at the center of the cell. 2 V is removed at 5 s.

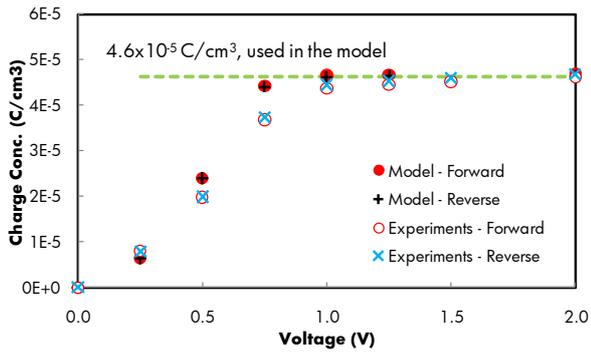


Fig. 7 Charge concentrations from transient current integral of 1-D model and experiments

Same parameters as Fig. 2, the total single-polarity charge concentration of $4.6 \times 10^{-5} \text{ C/cm}^3$ was used. Experimental data are from 3 wt% OLOA11000 dispersion