

Nonstationary electro-osmotic flow in closed cylindrical capillaries. Theory and experiment

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Received 1 December 2006; accepted 12 February 2007

Available online 21 February 2007

Abstract

Both from the experimental and theoretical viewpoints it is of fundamental importance to know precisely which are the fluid flow characteristics in a (cylindrical, say) closed cell under the action of an externally applied electric field, parallel to the cell axis. This is so because in many cases the experimental determination of the electrophoretic mobility of dispersed particles is carried out in closed cells, whereby the motion of the particles in the laboratory reference system is the result of the superposition of their electrophoretic migration plus the liquid motion with respect to the cell. This makes it of utmost importance to analyze the above-mentioned fluid and particle movements. If, in particular, this evaluation is carried out in the presence of alternating fields of different frequencies, information about the dynamics and time scales of the processes involved can be obtained for different frequencies of the applied field. In the present contribution, we discuss experimental results based on the determination of the velocity of polystyrene latex particles in a closed, cylindrical electrophoresis cell, and compare them to our previous theoretical analysis of the problem. It is concluded that the theory explains with great accuracy the observed particle velocities. In addition to the use of the particles as probes for the fluid velocity distribution, this work intends to give additional clues on the frequencies and positions for which electrophoretic mobility measurements in closed cells can be more reliable.

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Keywords: Back flow; Closed capillary; Electro-osmosis; Electrophoresis; Stepwise pulses; Velocity profiles

1. Introduction

Both theoretical and experimental investigations of electrokinetic phenomena are usually carried out in conditions of stationary liquid flow regimes. However, studies carried out in nonstationary conditions may allow a better understanding of the stationary processes and provide additional information about their main characteristics. Furthermore, features arising from the application of a nonstationary electric field, either periodic or not, can also play an important role in different technologies, particularly electrorheology or electroacoustics.

Previously published papers on the subject of nonstationary electrokinetic processes, although mostly of great interest, are not very numerous and have mainly focused on sinusoidal applied electric fields, used either in the evaluation of peri-

odical flow regimes [1–6], or of the dielectric dispersion of colloidal systems and its relation with the polarizability of the solid/liquid interface [7–10]. Another fundamental application of sinusoidal fields that can be mentioned is the measurement of electrophoretic velocity [11,12], because the application of cyclically varying fields prevents or reduces electrode polarization effects. In addition, some kinds of nonstationary fields, namely, asymmetrical stepwise pulses, have also been used for the experimental study of the nonlinear component of electrophoresis [13–15]. However, the theoretical analysis of the obtained results was only qualitative because of the lack of a rigorous theoretical treatment of the problem, that was published considerably later [16,17]. This points to the need of a theoretical and experimental analysis of processes occurring at stepwise periodical regime in high field strengths.

In this work we present data for low electric field strength, i.e., for linear processes, since even in this simpler case the nonstationary regime of electro-osmosis strongly affects the move-

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ment of particles and leads to some peculiarities in the obtained experimental dependencies that have not received sufficient attention.

2. Experimental

Our aim is to analyze the dynamic electro-osmosis in the cylindrical channel of an electrophoresis cell, and the associated electrophoretic motion of dispersed particles under the action of the applied field. A Malvern Zetasizer IIc instrument (Malvern, England) was modified to control the type of voltage that is applied (frequency and shape of signal). The modifications performed included disconnecting the main electrodes of the electrophoresis cell from the power source of the instrument and connecting them to an external power source, consisting of a Promax GFD917 (Spain) signal generator connected to a Trek 6096-E (USA) high-voltage amplifier.

Like in standard electrophoretic mobility measurements, the measured velocity will consist of the superposition of the electro-osmotic velocity of the liquid at the selected position in the channel and the electrophoretic velocity of the particle with respect to the liquid. No changes were introduced in the cell characteristics themselves; the commercial one is a fused quartz cylinder 2 mm in radius and 4 cm in length, surrounded by a thermostatted water bath. The suspensions studied were in all cases polystyrene latex spheres 1 μm in diameter, dispersed in 4×10^{-4} mol/L KCl solution. The voltage applied to the cell was a square signal with 200 V amplitude and frequency ω ranging between 1 and 20 Hz.

3. Fundamentals

Since nonstationary processes during the measurement of electrophoresis involve two nonstationary sub-processes—electro-osmosis and electrophoresis, each with its own time scale—the correct interpretation of experimental data can only be done after a careful theoretical analysis of each of these components. First of all, it can be easily shown that the electrophoretic movement of not too big charged particles reaches a stationary state rather quickly, and therefore at relatively low frequencies of the pulsed field (or, equivalently, a short time after a voltage step is applied), a steady electrophoretic velocity is reached. Hence, the nonstationary particle motion can only be caused by nonstationary electro-osmosis. This is the situation that we will consider, as it is the one of practical interest, and, in addition, it allows to single out one instead of the two nonstationary processes, with their different characteristic times.

3.1. Electrophoresis under periodic applied fields

It can be easily understood that the motion of a colloidal particle under the action of a nonstationary electric field will be nonsteady because of both the inertial properties of the particle and the liquid and the finite time needed for the electrical double layer to get polarized. Only if the characteristic time required for the particle to achieve a steady motion is considerably shorter than the duration of the applied voltage pulse, can the double-layer polarization kinetics be supposed to have

a negligible effect on the measured particle velocity. Otherwise, such velocity will be strongly dependent on the field frequency.

The characteristic times associated to the inertial properties of the particle, τ_p , and the liquid, τ_l , are related to the dynamic viscosity of the liquid, η , the particle radius, a , and the density of the particle, ρ_p , and the liquid, ρ_l , as follows [11,18]:

$$\tau_{p,l} = \frac{\rho_{p,l} a^2}{\eta}.$$

Concerning the role of the double-layer polarization, we have two contributions. One is the Maxwell–Wagner polarization, associated to the mismatch between the conductivities and permittivities of the medium and the particles. We will call τ_{MW} the time typically needed for this polarization to develop. The second contribution is the so-called concentration polarization (gradient of neutral electrolyte concentration around the particle), with characteristic time τ_{cp} [11,19–23]. Both are related to the diffusion coefficient of ions (we assume a common value for both counterions and coions), D , the thickness of the double layer (or Debye length) κ^{-1} , and the particle radius:

$$\tau_{MW} = \frac{\kappa^{-2}}{D}, \quad \tau_{cp} = \frac{a^2}{D}.$$

Assuming that the double layer is thin compared to the particle size ($\kappa a \gg 1$), it is immediate to see that the latter is the longest of the three characteristic times, i.e., $\tau_{cp} \gg \tau_p, \tau_{MW}$. Indeed, for typical values of the densities of the particles and the liquid, the viscosity of the latter, and the electrolyte concentration, $\tau_{p,l}$ and τ_{MW} are on the order of microseconds. Concerning τ_{cp} , considering that inorganic ions have typically $D = (1 \div 2) \times 10^{-9}$ m²/s, it can take values between 0.5 and 1 μs even for $a = 1$ μm . This time rapidly increases with the particle size (for example, $\tau_{cp} = 5$ or 50 μs , for radii 3 or 10 μm , respectively), so that for larger particles the role of nonstationarity will increase. That is why in our experimental investigation we used particles with radius $a = 0.5$ μm , as in this case even for a pulse frequency of 100 Hz the above cited relaxation times are at most about 1.25–2.5% of one pulse duration, and therefore the concentration polarization will be well developed right from the beginning of any pulse. In terms of frequencies, the characteristic frequency below which concentration polarization can be fully developed is about 10^4 Hz for our suspensions.

3.2. Electro-osmosis at a periodic regime

It is important to consider that the application of a periodic electric field should affect the electro-osmotic component of the particle movement considerably more strongly than its electrophoretic component. This is so because the characteristic time required for the development of a steady flow of liquid in a cylindrical capillary of radius R is given by [11,16,17]

$$\tau_{eo} = \frac{\rho}{\eta} R^2.$$

Considering the typical values of R and a , one can see that $\tau_{eo} \gg \tau_{cp} \gg \tau_p, \tau_{MW}$. For example, for a capillary with $R = 2$ mm, as used in our Malvern equipment, $\tau_{eo} = 4$ s, that is, between 16,000 and 32,000 times longer than τ_{cp} for $a = 0.5$ μm .

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