Labyrinthine Instability in Dielectric Fluids

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Abstract—The dual to recent work of the labyrinthine instability in magnetic fluids is demonstrated with analysis and measurements using dielectric fluids. Here polarizable fluid layers are placed within an initially uniform electric field tangential to the fluid interface separating two dielectric fluid layers. To eliminate electric space charge effects, alternating electric fields must be applied at a frequency much greater than the larger reciprocal dielectric relaxation time of the fluids. Past theory and measurements have shown that a flat interface in a uniform tangential field is stable if the interface is of infinite extent. If the fluid interface has a finite thickness in the direction of the applied field, reaction fields cause nonuniform field components tangential to the fluid interface. Above a critical magnitude the resultant field destabilizes the fluid interface forming labyrinthine patterns. An interfacial stability analysis and an energy minimization method are compared to measurements.

INTRODUCTION

A uniform magnetic field tangential to the interface between fluids of different magnetizability or a uniform electric field parallel to the interface between two different permittivity fluids is always stabilizing to small signal waves along the field, having no effect on interfacial waves transverse to the field [1]. However, recent work has demonstrated interfacial instability when magnetic fluid is confined with an immiscible nonmagnetic fluid between closely spaced parallel plates, with a uniform magnetic field applied normal to the plates and parallel to the thin dimension of the interface [2]–[7]. At a threshold field strength a comblike pattern develops which grows to a complicated labyrinth pattern of two simply connected but highly convoluted regions. The duality between magnetization and polarization phenomena when there is no space charge has been demonstrated whereby similar labyrinth patterns form between two dielectric fluids in an electric field [8], [9].

EXPERIMENTAL APPARATUS

The test cell is shown in Fig. 1 whereby transparent glass plates with a conducting coating act as parallel plate electrodes enclosing Plexiglas spacers and the two immiscible test fluids with a flat interface along the electric field. To eliminate space charge effects, alternating electric fields are applied at a frequency much greater than the larger reciprocal dielectric relaxation time of the fluids. For our experiments we used a frequency of 500 Hz. To verify that a flat interface in a uniform tangential field is stable, the Plexiglas spacer is removed (l = 0) so that the applied electric field is completely tangential to the interface. No instability developed for field strengths up to 28 kV/cm. The labyrinth would similarly not develop for magnetic fluids if the magnet pole-faces are placed without gap directly against the test fluids.

EXPERIMENTS

Physical parameters of test fluids used are listed in Table I. Figs. 2 and 3 show representative labyrinth patterns for various rms voltages and plate spacings l in horizontal and vertical cells with Plexiglas spacer length l = 1/8 in. In a vertical cell, larger voltages are necessary as the field instability must overcome the additional stabilizing effect of gravity with the more dense fluid below. For horizontal cells, the maximum spacing l was limited by gravity to 1/16 in. Larger spacings would not have a stable flat interface with zero voltage.

For some fluids a red dye was added to provide photographic contrast against the adjacent clear fluid. We note that the characteristic spacing of the instability and the onset voltages are a function of material and electrode spacings.

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TABLE I
DENSITY AND RELATIVE PERMITTIVITIES OF TEST DIELECTRIC FLUIDS AND PLEXIGLAS SPACER PERMITTIVITY \( \epsilon_p \)

<table>
<thead>
<tr>
<th>Fluid</th>
<th>Density (g/cm³)</th>
<th>Relative Dielectric Constant</th>
</tr>
</thead>
<tbody>
<tr>
<td>Castor Oil</td>
<td>0.945</td>
<td>4.5</td>
</tr>
<tr>
<td>Chain Lubricating Oil</td>
<td>0.925</td>
<td>2.45</td>
</tr>
<tr>
<td>Gear Lubricating Oil</td>
<td>0.89</td>
<td>2.33</td>
</tr>
<tr>
<td>Silicone Fluid</td>
<td>0.945</td>
<td>2.6</td>
</tr>
<tr>
<td>Corn Oil</td>
<td>0.925</td>
<td>3.1</td>
</tr>
<tr>
<td>Transformer Oil</td>
<td>0.89</td>
<td>2.3</td>
</tr>
<tr>
<td>Halowax Oil</td>
<td>1.28</td>
<td>4.7</td>
</tr>
<tr>
<td>Plexiglas spacer</td>
<td></td>
<td>3.3</td>
</tr>
</tbody>
</table>

**INTERFACIAL STABILITY THEORY**

**Interfacial Electric Fields**

The dielectric spacer in Fig. 1 gives rise to depolarizing fields that cause nonuniform field components tangential and perpendicular to the fluid interface. Far from the interface, the rms fields in each fluid region are

\[
E_a = \frac{V_{rms}}{t + 2l + \frac{1}{\epsilon_p}}, \quad E_b = \frac{V_{rms}}{t + 2l + \frac{1}{\epsilon_p}}.
\]

These fields are due to polarization charge \( \sigma_{pa} \) and \( \sigma_{pb} \) on the spacer–test fluid interface as well as free and polarization charge \( \sigma_a \) and \( \sigma_b \) on the electrode–spacer interface as illustrated in Fig. 1.

\[
\sigma_{pa} = \frac{\epsilon_0}{\epsilon_p}(\epsilon_p - \epsilon_a)E_a, \quad \sigma_{pb} = \frac{\epsilon_0}{\epsilon_p}(\epsilon_p - \epsilon_b)E_b
\]

\[
\sigma_a = \frac{\epsilon_0}{\epsilon_p}E_a, \quad \sigma_b = \frac{\epsilon_0}{\epsilon_p}E_b
\]

where we recognize that the electric field in the spacer \( E_p \) is related to the adjacent dielectric fields as \( \epsilon_p E_p = \epsilon_a E_a \) near dielectric (a) and \( \epsilon_p E_p = \epsilon_b E_b \) near dielectric (b).

As an approximation, we assume that these surface charge densities valid far from the interface uniformly extend to the interface in each region. By integrating the fields due to differential line charge elements we solve for the field and field gradient at the \((x = 0, z = 0)\) midline of the interface as

\[
\frac{dE_z}{dx} = \frac{8V_{rms}(\epsilon_b - \epsilon_a)(1 + l/t)}{\pi \epsilon_p (1 + 2l/t)} \left( t + \frac{2\epsilon_a}{\epsilon_p} \right) \left( t + \frac{2\epsilon_b}{\epsilon_p} \right)
\]

There is an \( x \)-directed normal field and field gradient along the interface which is zero along the midline. Those field components are much smaller than the tangential field and thus neglected here.

**Small Signal Interfacial Waves**

Past electrohydrodynamic work on the stability of fluid interfaces consider systems of infinite extent stressed by nonuniform electric fields tangential and normal to a flat interface. The dispersion relation for small signal perturbation of the form

\[
e^{i(a - \beta x + k z)}, \quad k = \sqrt{k_x^2 k_z^2}
\]

are obtained by interfacial balance of surface tension, pressure, and electrical forces. For our case, if we assume the fluids to be wet perfectly the thick spaced electrodes, the interface will have a radius of curvature equal to half the spacing even without electric field. The spacing \( t \) in Fig. 1 is also considered to be so small that no interfacial variations occur in the \( z \) direction beyond the equilibrium interfacial curvature due to wall wetting so that \( k_z = 0 \) in (4). There are nonzero \( x \) components of the electric field and field gradient along the interface, but at the \( z = 0 \) midpoint these terms are zero. The \( z \) component of the electric field and normal field gradient also vary with \( z \) along the interface but at midpoint \( z = 0 \) are given by (3). We also assume that the sinusoidal time variations in electric field are much faster than time for the fluids to move, that the interface only responds to the rms field values. Within these approximations, the dispersion relation is then [10], [11]

\[
(\eta_1 + \eta_2)s = -\gamma k^3 - k \left[ g(p_b - p_a) + 4\gamma_{w}/t^2 - (\epsilon_b - \epsilon_a)E_z \frac{dE_z}{dx} \right]
\]

where \( \eta_1 \) and \( \eta_2 \) are the Darcy’s drag coefficients for flow in a Hele–Shaw cell, \( \gamma \) is the interfacial tension between the two fluids, \( \gamma_w \) is the surface tension at the wall, and the interface has equilibrium radius of curvature \( t/2 \).

With this model, we thus see that the instability of Figs. 2 and 3 is driven by the nonuniform tangential electric field. If \( s \) has a positive real part the interface is unstable as any perturbation grows exponentially with time. Because the electric field term in (5) is positive, the electric field tends to destabilize the interface. The fastest growing unstable wave has wavelength \( \lambda^* = 2\pi/k^* \)

\[
\frac{\lambda^*}{\lambda_0} = \left( -1 + \frac{W}{t + \frac{2\epsilon_a}{t}} \right)^{1/2}
\]
Fig. 2. Representative labyrinth patterns using castor oil and gear lubricating oil for various voltages in horizontal and vertical test cells of various spacings. Note that, when \( t = 1/16 \) in and \( t = 1/32 \) in and 1/16 in, some dark lubricating oil slipped between electrode and Plexiglas spacer, but with no effect on labyrinth pattern.
Castor Oil (Clear) - $\epsilon_r = 4.5$, $\rho = 945$ g/cc
Lubricating Oil (Dark) - $\epsilon_r = 2.3$, $\rho = 89$ g/cc
Plexiglas Spacer $t = 16$ in, $\epsilon_r = 3.3$

Fig. 2. (Continued).

where we introduce nondimensional parameters

$$\tilde{\varepsilon}_a = \frac{\varepsilon_a}{\varepsilon_p}, \quad \tilde{\varepsilon}_b = \frac{\varepsilon_b}{\varepsilon_p}, \quad \tilde{t} = t/l, \quad W = \frac{4(\varepsilon_b - \varepsilon_a)^2 V_{rms}^2}{[4\gamma_w/t^2 + g(\rho_b - \rho_a)]\pi \epsilon_p l^3}$$

and $\lambda_0$ is related to the wavelength of gravity-capillary waves

$$\lambda_0 = \frac{2\pi}{\sqrt{[g(\rho_b - \rho_a) + 4\gamma_w/t^2]/3\gamma}}$$

The physical parameters of permittivity and density were measured for all tested fluids as listed in Table I. The interfacial tension $\gamma$ was measured by a ring surface tensiometer to be about 1 dyne/cm for all tested fluids. This was at the extreme lower end of the scale and was not accurate. Similarly, wall surface tension $\gamma_w$ could not be accurately measured.

Our procedure was to note first in (6) that there is a critical value of $W$ for the instability to onset

$$W_c = \frac{\tilde{l}^2 \left[ 1 + \frac{2\tilde{\varepsilon}_a}{\tilde{t}} \right] \left[ 1 + \frac{2\tilde{\varepsilon}_b}{\tilde{t}} \right]}{\left[ 1 + \frac{\tilde{\varepsilon}_a + \tilde{\varepsilon}_b}{\tilde{t}} \right] \left[ 1 + \frac{1}{1 + 2/\tilde{t}} \right]}.$$
\( \gamma_w \) using (7) and (9). For the castor/gear case we did this measurement for two values of \( t \) (\( t = 1/32 \) in, 1/16 in) and two values of \( l(l = 1/16 \) in, 1/8 in). We averaged the four values to obtain \( \gamma_w = 0.4 \) dyn/cm. The results for a horizontal cell comparing predicted and measured values of \( W_c \) are shown in Fig. 4 by the open symbols for various fluids. Using the value of \( \gamma_w \) obtained for a horizontal cell, measurements were repeated in a vertical cell and plotted in Fig. 4 with darkened symbols for various fluids in reasonable agreement with analysis. Note that if wall surface tension was not included in our analysis \( (\gamma_w = 0) \), the instability in a horizontal cell \((g = 0)\) would onset at zero voltage, contrary to measurements.

Fig. 5 plots the fastest-growing unstable wavelength \( \lambda/\lambda_0 \) versus \( W \) from (6) for voltages in excess of the critical voltage for various values of spacing \( t \). Experimental data for various fluids are also plotted in Fig. 5, with values \( \gamma_w \) and \( \gamma \) chosen to give a good fit with analysis. A troublesome difference between analysis and experiment is that the analysis of (6) predicts the fastest growing unstable wavelength to decrease smoothly from infinity as \( W \) is increased above \( W_c \). As the voltage is increased from zero, our measurements show some long wavelength interfacial distortion at an incipient voltage from which we calculate \( W_c \). As the voltage is further increased, the interfacial disturbances have a well-defined spacing that does not smoothly change with voltage.

Relation to Magnetic Fluid Analysis

The magnetic dual problem, treated by an energy minimization analysis [2] considered only one fluid weakly magnetizable with the field source in free space at infinity. The interesting feature of that analysis was the prediction of a finite wavelength of instability as the field strength was increased, in agreement with observations. However, that analysis neglected the magnetic body force due to the field gradient near the interface. Including this body force, their result for the threshold of instability converted to the dielectric analog problem in SI units in the present notation has \( \epsilon_b = \epsilon_a = \epsilon_0, \epsilon_b - \epsilon_0 \ll \epsilon_0 \), and \( l \to \infty \) with \( E = V/2l \) finite:

\[
\frac{\gamma \pi \epsilon_0}{2P^2} = \frac{1 + \frac{1}{2} \left[ \Gamma + \ln \frac{kt}{2} + K_0(kt) \right]}{(kt)^2 + (k_0 t)^2}
\]

where \( P = (\epsilon_b - \epsilon_0)E \) is the dielectric polarization, and \( \Gamma = \ldots \)
0.5772 is the Euler constant,

\[ k_0 = \left\{ \frac{g(\rho_0 - \rho_2)}{4\gamma \omega / l^2} \right\}^{1/2} \]

is the gravity capillary wavenumber augmented by wall surface tension, and \( K_0 \) is the zeroth-order modified Bessel function.

In the same limits, (9) gives the onset of instability as

\[ \frac{\pi \sigma_0 \gamma}{2P_0^2} \left( k_0 \right)^2 + (k_0) \]

Both (10) and (11) are plotted in Fig. 6 and are seen to be fairly close and thus probably not resolvable by experiment.

**LIMITING LABYRINTH STATE**

The interfacial stability analysis provides a good measure for the onset of instability and its dependence on material properties, geometry, and field strength. However, it provides no information on the limiting labyrinth state once the interface is no longer flat. Then there is a mutual invasion of the two phases in which fringes continually bifurcate, finally reaching a steady-state labyrinth pattern comprising two simply connected but highly convoluted regions.

To solve for the limiting state of a horizontal labyrinth, we extend the energy method analysis developed by Rosenweig for magnetic fluids [8]. We neglect the presence of convolutions, nodes, cul-de-sacs, and other complicating features of the real labyrinth and consider a repeating pattern of parallel fluid strips shown in Fig. 7.

The total energy of one period of the repeating pattern of width \( w_a + w_b \) is due to the electrostatic, \( W_E \), and surface tension \( W_s \) energies

\[ W_T(w_a + w_b) = W_E + W_s \]

where \( W_T \) is the average energy per unit length in the z direction.

The interfacial energy for a system of height \( h \) is

\[ W_s = 2h[w_a \gamma_{ap} + w_b \gamma_{bp} + \gamma_{ab}] \]

where \( \gamma_{il} \) is the appropriate surface tension at each of the interfaces.

The electric energy stored in the system is [12]

\[ W_E = -\frac{1}{2} \int V (e - e_0) E \cdot E^T dV \]

\[ = -\frac{E_0}{2} [(e_a - e_0) w_a E_a + (e_b - e_0) w_b E_b] h \]

where \( E^T = E_{d, E} \) is the electric field in the test cell with electrode charge per unit area \( Q \) in the absence of dielectric fluids

\[ E_0 = \frac{Q}{e_0} \]

and \( E_a \) and \( E_b \) are the average final electric fields when both dielectric fluids are in place with the same value of electrode charge per unit area \( Q \). It is important to express \( W_E \) as a state function of \( Q \) and not the terminal voltage. By keeping \( Q \) constant, no electrical energy can be input so that the system is closed and the fluids will arrange themselves to minimize the total energy. For the same charge \( Q \), the terminal voltage will be different with and without dielectric fluids. A similar type of energy analysis has been applied to bubble elongation in a liquid capacitor [13].

The final electric fields \( E_a \) and \( E_b \) are actually complicated functions of position to be integrated in (14). However, as a simplification we assume in (15) that \( E_a \) and \( E_b \) are uniform in each dielectric being the value at the midpoint of each dielectric including the depolarizing effects of polarization charge \( \sigma_{2b} \), \( \sigma_{2b} \) on the spacer–dielectric fluid interface and free and polarization charge \( \sigma_{1a} \), \( \sigma_{1b} \) on the electrode/spacer interface.

With both dielectrics in place and stressed by a constant voltage \( V_0 \) with average charge per unit area \( Q \), the average field in each dielectric is

\[ E_{d0} = \frac{e_p V_0}{e_{pd} + 2e_{d0}} \]

\[ = \frac{Q}{(e_{pd} + 2e_{d0})} \left[ \frac{e_a}{e_{pd} + 2e_{d0}} \left( 1 + \frac{e_b}{w_f} \right) \right] \]

\[ E_{b0} = \frac{e_p V_0}{e_{pd} + 2e_{d0}} \]

\[ = \frac{Q}{(e_{pd} + 2e_{d0})} \left[ \frac{e_a}{e_{pd} + 2e_{d0}} \left( 1 + \frac{e_b}{w_f} \right) \right] \]

The interfacial charges in Fig. 7 at dielectric–spacer and electrode–spacer interfaces are then

\[ \sigma_{1a} = \frac{e_p}{e_{pd}} E_{d0} \]

\[ \sigma_{1b} = \frac{e_p}{e_{pd}} E_{b0} \]

\[ \sigma_{2a} = e_0 \left( 1 - \frac{e_a}{e_p} \right) E_{d0} \]

\[ \sigma_{2b} = e_0 \left( 1 - \frac{e_b}{e_p} \right) E_{b0} \]
Fig. 7. Sketch of idealized labyrinth geometry as periodic system of parallel strips of alternating dielectric fluids. Widths \( w_a \) and \( w_b \) will be determined by those values that minimize sum of surface tension and electric field energies.

The fields at the midpoint of each dielectric are then the integrated sum of fields due to these four alternating surface charges of the form

\[
E_a = A_{aa}E_{a0} + A_{ab}E_{b0}
\]

\[
E_b = A_{ba}E_{a0} + A_{bb}E_{b0}
\]

\[
A_{aa} = \frac{2}{\pi} \left\{ \tan^{-1} \left( \frac{w_a}{t} \right) + \frac{w_a}{t} + \frac{w_a}{t} \right\} + \frac{\sum_{n=1}^{n} \left( \tan^{-1} \left( \frac{w_a + w_b}{t} \right) \right)}{l + 1/2} - \frac{n(w_a + w_b) - w_a/2}{l + 1/2}
\]

\[
A_{ab} = \frac{2}{\pi} \left\{ \tan^{-1} \left( \frac{w_b}{t} \right) + \frac{w_b}{t} + \frac{w_b}{t} \right\} + \frac{\sum_{n=1}^{n} \left( \tan^{-1} \left( \frac{w_b + w_a}{t} \right) \right)}{l + 1/2} - \frac{n(w_b + w_a) - w_b/2}{l + 1/2}
\]

\[
A_{bb} = \frac{2}{\pi} \left\{ \tan^{-1} \left( \frac{w_b}{t} \right) + \frac{w_b}{t} + \frac{w_b}{t} \right\} + \frac{\sum_{n=1}^{n} \left( \tan^{-1} \left( \frac{w_b + w_a}{t} \right) \right)}{l + 1/2} - \frac{n(w_b + w_a) - w_b/2}{l + 1/2}
\]

The ratio of fluid widths \( r = w_a/w_b \) is constant and equal to the ratio of fluid volumes. We wish to find that value of \( w_a \) or \( w_b \) with constant \( r \) that minimizes the total energy per unit length \( W_T \).
Using (13)-(24) in (12) yields
\[
W_T = 2h \left[ \frac{\gamma_\rho + \gamma_\varphi}{1 + 1/r} + \frac{w_{ab}}{w_b(1 + r)} \right]
- \frac{1}{2} \alpha E_0 \left[ \frac{\epsilon_\varphi - \epsilon_0}{1 + 1/r} \right] (A_{ab}E_{ab} + A_{bb}E_{bb})
+ \frac{(\epsilon_\varphi - \epsilon_0)}{(1 + r)} (A_{ab}E_{ab} + A_{bb}E_{bb})
\]
\[\text{(25)}\]

Note in (25) that \(E_0, E_{ab}, \) and \(E_{bb}\) do not depend on \(w_b\) but only on the constant \(r\) so that the energy minimization condition will depend directly on the depolarization factors of (21)-(24) yielding
\[
\frac{dW_T}{\delta w_b} = 0 \Rightarrow N_E = \frac{\epsilon_p E_0 V_0}{4\gamma_{ab}}
\]
\[\text{(26)}\]

where we introduce the nondimensional electric Bond number \(N_E\) and normalize all permittivities to \(\epsilon_p\) as in (7) and all lengths to \(t\)
\[\tilde{t} = t \tilde{w}_b = \frac{w_b}{w_b/t}.\]
\[\text{(27)}\]

Fig. 8 plots the dependence of the nondimensional electric Bond number \(N_E\) as a function of \(\tilde{w}_b = w_b/t\) for various values of \(r = w_a/w_b\) and \(\tilde{t} = t/\tilde{w}_b\) for permittivity parameters corresponding to dielectric fluids of castor oil and the lubricating oil with a Plexiglas dielectric spacer. This plot was obtained using (26), taking 2000 terms in the infinite series of (21)-(24), and illustrates a minimum value of electric Bond number necessary to establish the labyrinth pattern. For the representative values in our experiments, with \(r = w_a/w_b = 1\), the minimum value of \(N_E\) is about 25 with \(\tilde{w}_b = 1\) to 2. Higher values of voltage increase \(N_E\) and thus decrease \(\tilde{w}_b\).

A curious feature of Fig. 8 is that \(\tilde{w}_b\) is double valued in the the range of \(N_E\) just above threshold. This is most likely a result of the approximations made in computing the electric fields in each fluid strip. These features of Fig. 8 are similar to that found in the analogous magnetic fluid problem where only one fluid was magnetizable and the magnet pole faces were at infinity [8]. Here, the analysis is more complicated because both fluids and spacers are polarizable and the electrodes are a finite distance from the fluids.

Table II shows the predicted critical voltage of \(\approx 1.5-2\) kV necessary for labyrinth formation in a horizontal cell using the fluids listed in Fig. 8 to be in good agreement with measurements. The surface tension value of 1 dyne/cm for \(\gamma_{ab}\) was used since this value best fit the data of Fig. 5.

### CONCLUDING REMARKS

The linear stability analysis of (4)-(9) predicts the voltage necessary for a flat interface to become first unstable including the effects of gravity and wall surface tension. For a horizontal cell so that gravity has no effect and with no wall surface tension, this incipient voltage is zero. However, in this limit, the minimum energy analysis of (12)-(27) shows that a threshold voltage is still necessary for full labyrinth development, in agreement with measurements. Lesser voltages disturb the flat interface but do not have complete interpenetration of fluids.

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REFERENCES


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