INVITED PAPER

LABYRINTHINE INSTABILITY IN MAGNETIC AND DIELECTRIC FLUIDS

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The magnetic and electric duality of magnetizable and polarizable liquids is demonstrated for the labyrinthine instability that occurs in thin layers subjected to initially uniform fields. Theory is developed for the spacing of fully developed labyrinth patterns in horizontal cells, and measurements compared with theory.

When magnetic fluid is confined with an immiscible fluid between closely spaced parallel plates, and a uniform magnetic field is applied normal to the plates, the interface between the fluids exhibits a threshold instability in which a comblike pattern is established [1]. At supercritical intensities of the magnetic field a mutual invasion of the two phases occurs, in which fingers of magnetic fluid invade the nonmagnetic fluid and vice versa; a single finger bifurcates into two fingers as the invasion process continues [2]. The final state of this process is the formation of a labyrinth pattern in essence comprising two simply connected but highly convoluted regions [3]. When the cell containing the fluids is horizontal the labyrinth spacing is characteristically uniform, and when the cell is vertically oriented the pattern is graded more densely to the bottom [4].

Theory for onset of the instability has been developed by Maiorov and Tsebers for flat interfaces [1], drops [5] and bubbles [6]. In the present study, we develop a simple theory for spacing in the fully developed, horizontally oriented labyrinthine pattern and compare the theory to measurements. The effect is demonstrated to occur also for dielectric fluids subjected to an applied electric field. The magnetic and dielectric phenomena are illustrated in fig. 4.

2. Theory of labyrinth spacing

The horizontal labyrinth is idealized as a repeating pattern of parallel paths, see fig. 1. Thus, the presence of convolutions, nodes, cul-de-sacs, and other features of an actual labyrinth are ignored. The idealized pattern and its nomenclature are illustrated in fig. 1. The total energy of one period of the repeating pattern will be formulated as the sum of the magnetostatic energy \( U_m \) and the interfacial energy \( U_s \).

\[
U = (U_m + U_s)(w_f + w_t)^{-1}.
\]

From the geometry of the figure the essential portion of the interfacial energy per unit depth is given as

\[
U_s = 2\gamma t,
\]

where \( \gamma \) denotes interfacial tension. Magnetic field energy associated with the configuration can be written as

\[
U_m = -\frac{\mu_0}{2} \int_0^L MH_0 \, dV = -\frac{\mu_0}{2} MH_0 tw_t,
\]

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Computing the Demagnetization Factor

Fig. 1. Sketch of the labyrinth idealized as a periodic system of parallel strips. \( \gamma \) denotes interfacial tension of the fluids. The nonmagnetic fluid preferentially wets the walls.

where it is assumed that the magnetization is uniform at all points within the ferrofluid. Magnetization \( M = \chi H \) where \( H \), the field within a strip, is reduced from the intensity \( H_0 \) of the applied field due to the demagnetization field \( H_d \) of the strip and that of all the other strips in the system. Thus \( H = H_0 - H_d = H_0 - DM \), where \( D \) is the demagnetization coefficient and \( M = \chi H_0/(1 + \chi D) \), so that \( U_m \) from (3) may be written as

\[
U_m = -\frac{\mu_0 \chi H_0^2 tw_1}{2(1 + \chi D)}.
\]  

Substituting eqs. (4) and (2) into (1) gives, upon rearrangement,

\[
N_U = -\frac{\chi N_B}{1 + \chi D} \frac{2}{z},
\]

where the magnetic Bond number is \( N_B = \mu_0 H_0^2 t/2\gamma \). Other definitions introduced above are

\[
N_U = \frac{U(1 + r)}{\gamma}, \quad r = \frac{w_1}{w}, \quad z = \frac{w_1}{t}.
\]

Minimization of energy in (5) yields the expression

\[
\frac{dN_U}{dz} = \frac{\chi^2 N_B}{(1 + \chi D)^2} \cdot \frac{\partial D}{\partial z} \frac{2}{z^2} = 0.
\]

The demagnetization coefficient at the midline of a strip is found by integrating the demagnetizing field of the assumed uniform magnetic surface charge \( \pm \mu_0 M \) of each strip and \( N \) neighborhood strips on either side of the given strip as illustrated in fig. 1.

\[
D = \frac{2}{\pi} \left[ \tan^{-1} \frac{w_1}{t} + \sum_{n=0}^{N} \left( \tan^{-1} \frac{(n+1)w_1 + (n+\frac{3}{2})w_1}{t/2} - \tan^{-1} \frac{(n+1)w_1 + (n+\frac{1}{2})w_1}{t/2} \right) \right].
\]

From (7) and (8) the magnetic bond number at equilibrium is

\[
N_B = \frac{\pi}{\chi^2 z^2} \left\{ 1 + \frac{2\chi}{\pi} \left[ \tan^{-1} z + \sum_{n=0}^{N} \left( \tan^{-1} 2z \left( (n+1)r + n + \frac{3}{2} \right) - \tan^{-1} 2z \left( (n+1)r + n + \frac{1}{2} \right) \right) \right] \right\}^2.
\]

\[
N_B = \frac{1}{1 + z^2 + \sum_{n=0}^{N} \left[ \frac{(n+1)r + n + 3/2}{1 + 4z^2 \left( (n+1)r + n + 3/2 \right)^2} - \frac{(n+1)r + n + 1/2}{1 + 4z^2 \left( (n+1)r + n + 1/2 \right)^2} \right]}
\]
The general dependence of $N_B$ on $z$, $\chi$, and $r$ displayed in fig. 2 illustrates that a minimum value of Bond number is required to establish the labyrinthine pattern. A curious feature is that $z$ is double valued in the range of $N_B$ just above threshold; a more exact analysis is needed to determine if this feature is an artifact.

Finally, neglecting neighbors by setting $N = 0$ in (9) simplifies the relation greatly, as the summations disappear, while the calculated Bond number is reduced typically by only 15–20%. However, in computing the curves of fig. 2 the value $N = 100$ is used.

3. Magnetic fluid experiments

Colloidal magnetic fluid in hydrocarbon carrier (UC) had saturation magnetization of 25 860 A/m, initial susceptibility of 1.6, and specific gravity 1.23. The immiscible fluid was distilled water containing 100 ppm of a surfactant (ethomeen 18/60) to provide preferential wetting of the cell window by the transparent aqueous phase. Surface and interfacial tensions determined with a ring tensiometer gave the following values: ferrofluid/air 0.029 N/m; aqueous/air 0.050 N/m; aqueous/ferrofluid 0.0088 N/m (freshly contacted); aqueous/ferrofluid 0.0043 N/m (24 h equilibration). The equilibrated value of 0.0043 N/m was used to correlate the data.
The test cell consisted of two glass plates separated with gaskets defining a 7.62 cm square area clamped together with a screw fixture. The spacers were of different thicknesses and could be interchanged. Air core electromagnets served as the source of uniform applied vertically oriented magnetic field of intensity up to

![Fig. 4. Equilibrium labyrinthine patterns in horizontal layers for magnetic and dielectric fluids. Captions indicate applied field intensity and gap spacing; see text and fig. 5 for materials and property values.](image_url)
0.245 T. The cells were operated horizontally so that gravity would not affect the labyrinth patterns.

The labyrinth pattern filled the cell within less than a minute after field was applied. An additional time duration of 15 to 30 min was allowed for the pattern to anneal. In one instance a 24 h annealing period was allowed but no further change in the pattern could be observed.

Although the cell was filled with equal volumes of the two fluids, the fully developed labyrinth patterns displayed unequal thicknesses of magnetic fluid “walls” compared to the aqueous “lanes”. The magnetic fluid walls always appeared of lesser thickness due apparently to accumulation of magnetic fluid in nodes where three walls meet. In order to compare experiment with theory, it was decided to assign the local value $r$ as the ratio of lane to wall thickness, and to utilize this value of phase ratio $r$ in computing the experimental Bond number. The comparison to the values computed from eq. (9) of the theory is shown in fig. 3. Measured Bond numbers increase with increase in applied field and plate spacing; the theory predicts these directional trends well, and the value of the Bond number with rough accuracy. Additional data with narrow spacing to $t = 0.15$ mm fit the plot equally well.

4. Dielectric fluids

There is a complete duality between magnetization and polarization phenomena if there is no space charge by replacing the magnetic field $H$ by electric field $E$ and magnetic permeability $\mu$ by dielectric permittivity $\varepsilon$. To eliminate space charge effects, alternating electric fields must be applied at a frequency greater than the larger reciprocal dielectric relaxation time of the fluids. This duality is demonstrated in fig. 4 using the test apparatus and dielectric liquids shown in fig. 5.

Past theory and measurements have shown that a flat interface in a uniform tangential field is stable [8]. This was verified with the apparatus in fig. 5 by removing the spacer ($l = 0$) so that the applied electric field is completely tangential to the interface. No labyrinth pattern developed for field strengths up to 28 kV/cm.
When the spacer is reinserted, the labyrinth pattern occurs, growing more convoluted as the voltage is increased. The dielectric spacer gives rise to depolarizing fields with non-uniform field components tangential and perpendicular to the fluid interface causing interfacial instability.

The labyrinth would similarly not develop for the magnetic fluid case if the pole-faces of a magnet are placed without gap directly against the test fluids. Then the demagnetizing coefficient $D$ would be zero as the $H$ field in both magnetic and non-magnetic fluids would equal the applied field $H_0$. With $D = 0$, the energy minimization solution to (7) is $z = \infty$, indicating that the magnetic fluid layer does not form the labyrinth.

To predict the labyrinth spacing, the energy minimization analysis for dielectric fluids needs to be extended as now both fluids are polarizable giving rise to depolarizing fields in each dielectric strip that depend on the properties and geometry of both fluids. A further complication is that the source electrodes are nearby, so that we must also consider the energy stored in the electric field outside the cell. For the magnetic fluid case the magnet pole faces which are the source of $H_0$ are assumed to be very far away compared to the test cell thickness. For the dielectric case, this would require an unreasonably high voltage to create a sufficiently strong electric field. These additional complications make the energy minimization analysis for dielectric fluids more difficult and thus still a current topic of investigation.

References