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COUPLING BETWEEN A TRANSCONFORMATION SURFACE REACTION AND HYDRODYNAMIC MOTION

A. SANFELD and A. STEINCHEN
Université Libre de Bruxelles, Faculté des Sciences, Brussels, Belgium

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Conditions are given for chemical and hydrodynamic instability of a cooperative transconformation reaction at the interface between two immiscible liquids.

1. Introduction

The purpose of the present paper is to analyse the hydrodynamic and the chemical stability of an interface where a transconformation reaction takes place.

The system consists of a flat plane layer of macromolecules (polypeptides or proteins) spread at the interface of two immiscible fluids. External constraints of pH or of electrical potential may change the conformation of the spread molecules, giving rise to a change in surface tension.

This phenomenon was recently observed [1,2] for the helix–coil transformation of a layer of poly-L-benzyl glutamate, poly-L-benzyl aspartate and poly-L-alanin.

Changeux et al. have assumed [3] that the permeability of the biological membranes to ions could be correlated to the change of conformation of protein protomers. The same correlation between the structure of the carrier and the permeability to ions is observed in artificial lipid bilayers containing a cyclic antibiotic like, for example, allametycin [4]. In the system studied here, the permeability of the monolayer to the ions will be analyzed on the base of a carrying mechanism due to Changeux and Lefever [5] and Van Roten [6] for biological membranes.

Now, the question arises to know what the influence is of the change in conformation and of the ion transfer through an interface on the hydrodynamic stability.

Numerous experimental evidences [7,8] exist showing surface instabilities (surface turbulence, spontaneous emulsification) induced by the mass transfer of a surfactant. Sternling and Scriven [9] gave a theoretical interpretation of these instabilities in term of a Marangoni effect.

The same type of analysis will be carried out here to elucidate the connection between the surface reaction and the hydrodynamic stability of the system. A first approach was already preformed by the authors [10] for an autocatalytic reaction. From the biological point of view, the system studied here could provide a simplified model of the cell deformation during phagocytosis. The spread monolayer of macromolecules simulates the membrane, as it is now well known [11,12] that the living membranes have the consistency of a light oil in which proteins are free to move. The change in surface tension due to the transconformation reaction could give a good picture of the change in the mechanical properties of the biological membrane with the conformation of the proteins in the membrane [13]. We shall use here the same formalism as Glansdorff and Prigogine [14] and Nicolis [15] for the chemical instabilities and as Chandrasekhar [16] for the hydrodynamic processes.

2. Chemical aspect

This section will deal only with the kinetic processes occurring in the layer. The layer of thickness d is located at a mean position $z = z_s$. It contains the insoluble macromolecules and two electrical double layers and lies between two immiscible liquid phases (internal phase i and external phase e). We assume the following sequence of reactions to take place in the layer

$$\underset{C_3}{\bigcirc} \underset{k_c'}{\overset{k_c}{\rightleftharpoons}} \underset{C_1}{\square} \qquad \text{(A) transconformation}$$

$$\underset{C_1}{\square} + A \underset{k_d^i}{\overset{k_a^i}{\rightleftharpoons}} \underset{C_2}{\boxdot} \qquad \text{(B)}$$

$$\square + B \rightleftharpoons \boxdot \qquad \text{(C) fixation}$$

$$A_i \underset{k_{-p}^i}{\overset{k_p^i}{\rightleftharpoons}} A \qquad \text{(D)}$$

diffusion-migration from the bulk

$$B_e \underset{k_{-p}^e}{\overset{k_p^e}{\rightleftharpoons}} B \qquad \text{(E)}$$

where C is the concentration (in mass per unit volume) of the macromolecules in the layer; the subscripts 3, 2 and 1 refer respectively to the molecule in the closed conformation, and to the molecule in the open conformation filled with an ion or free from ion.

A and A_i are the concentrations of the ions respectively in the internal double layer and in the internal bulk phase, B and B_e for the external layer and bulk phase.

The transconformation step (A) is assumed to be characterized by a change in the dipole moment and, in consequence, the electric part of the standard affinity (or $-\Delta G$) is given by

$$\mathscr{A}_{el} = (\mu_1 - \mu_3)E/RT\,, \tag{1}$$

with E the electric field (assumed to be uniform and normal to the interface, throughout the layer) and μ_1 and μ_3, respectively, the average dipole moment of the open molecule and of the closed molecule.

Moreover we will assume in agreement with Changeux–Lefever [5] and Van Roten [6] that the transconfor-

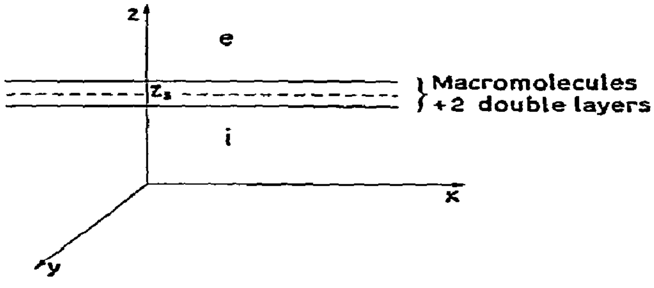

Fig. 1.

mation is cooperative, which means that the standard chemical affinity reads

$$\mathcal{A}_{\text{chem}} = \mathcal{A}^* + \eta(C_1 + C_2)\,, \tag{2}$$

where η is a positive constant and $\mathcal{A}^*$ the standard affinity in the absence of cooperativity. The standard affinity $\mathcal{A}$ is then given by

$$\mathcal{A} = \mathcal{A}_{\text{chem}} + \mathcal{A}_{\text{el}} = \mathcal{A}^* + \eta(C_1 + C_2) + (\mu_1 - \mu_3)E\,. \tag{3}$$

The pseudo-equilibrium constant k_c'/k_c then reads

$$\frac{k_c'}{k_c} \approx \exp\left(-\frac{1}{RT}\,[\mathcal{A}^* + (\mu_1 - \mu_3)E]\right)\,\exp\left(-\frac{\eta}{RT}\,(C_1 + C_2)\right). \tag{4}$$

For the sake of simplicity let us write (4) as

$$\frac{k_c'}{k_c} = l\Lambda^{C_\mathrm{R}} = l^* \mathrm{e}^{\Delta\mu E/RT}\Lambda^{C_\mathrm{R}}\,, \tag{5}$$

where

$$l = \exp\left(-\frac{1}{RT}\,[\mathcal{A}^* + (\mu_1 - \mu_3)E]\right), \quad l^* = \mathrm{e}^{-\mathcal{A}^*/RT}, \quad \Lambda = \mathrm{e}^{-\eta/RT}\,;$$

and

$$C_1 + C_2 = C_\mathrm{R} \tag{6}$$

is the total concentration of open macromolecules.

The kinetic equations are then

$$\frac{\partial C_1}{\partial t} = k_c(C_3 - l\Lambda^{C_\mathrm{R}}C_1) + k_\mathrm{d}^\mathrm{i}C_2 - k_\mathrm{a}^\mathrm{i}C_1A + k_\mathrm{d}^\mathrm{e}C_2 - k_\mathrm{a}^\mathrm{e}C_1B - \operatorname{div} v^\mathrm{s}C_1 + D_1\Delta^\mathrm{s}C_1\,, \tag{7}$$

$$\frac{\partial C_3}{\partial t} = -k_c(C_3 - l\Lambda^{C_\mathrm{R}}C_1) - \operatorname{div} v^\mathrm{s}C_3 + D_3\Delta^\mathrm{s}C_3\,, \tag{8}$$

$$\frac{\partial C_2}{\partial t} = k_\mathrm{a}^\mathrm{i}AC_1 - k_\mathrm{d}^\mathrm{i}C_2 + k_\mathrm{a}^\mathrm{e}BC_1 - k_\mathrm{d}^\mathrm{e}C_2 - \operatorname{div} v^\mathrm{s}C_2 + D_2\Delta^\mathrm{s}C_2\,, \tag{9}$$

$$\frac{\partial A}{\partial t} = k_p^\mathrm{i}A_\mathrm{i} - k_{-p}^\mathrm{i}A + k_\mathrm{d}^\mathrm{i}C_2 - k_\mathrm{a}^\mathrm{i}AC_1 - \operatorname{div} v^\mathrm{s}A + D_A\Delta^\mathrm{s}A\,, \tag{10}$$

$$\frac{\partial B}{\partial t} = k_p^\mathrm{e}B_\mathrm{e} - k_{-p}^\mathrm{e}B + k_\mathrm{d}^\mathrm{e}C_2 - k_\mathrm{a}^\mathrm{e}BC_1 - \operatorname{div} v^\mathrm{s}B + D_B\Delta^\mathrm{s}B\,, \tag{11}$$

where v^s is the mean surface velocity and Δ^s a two dimensional Laplace operator in the plane of the interface. The surface layer (macromolecules + double layers) is assumed to have a mean dielectric constant ϵ, and the position of the fixation site in the layer is characterized by a parameter p. This quantity is the relative width of the layer occupied by the two double layers. According to Lefever–Changeux [5] and Van Roten [6], we then write the kinetic constants in terms of the reduced electric potential difference through the layer, i.e.,

$$k_\mathrm{a}^\mathrm{i} = k_\mathrm{a}^{\mathrm{i}*}\mathrm{e}^{(p-1)\chi/4}\,, \quad k_\mathrm{d}^\mathrm{i} = k_\mathrm{d}^{\mathrm{i}*}\,\mathrm{e}^{(1-p)\chi/4}\,, \quad k_\mathrm{a}^\mathrm{e} = k_\mathrm{a}^{\mathrm{e}*}\,\mathrm{e}^{(1-p)\chi/4}\,, \quad k_\mathrm{d}^\mathrm{e} = k_d^{\mathrm{e}*}\,\mathrm{e}^{(p-1)\chi/4}\,, \tag{12}$$

$$k_p^{\mathrm{i}} = k_p^{\mathrm{i}*}\,\mathrm{e}^{-p\chi/4}\,, \quad k_{-p}^{\mathrm{i}} = k_p^{\mathrm{i}*}\,\mathrm{e}^{p\chi/4}\,, \tag{13}$$

$$k_p^{\mathrm{e}} = k_p^{\mathrm{e}*}\,\mathrm{e}^{p\chi/4}\,, \quad k_{-p}^{\mathrm{e}} = k_p^{\mathrm{e}*}\,\mathrm{e}^{-p\chi/4}\,, \tag{14}$$

where

$$\chi = z\mathcal{F}\Delta V/RT \tag{15}$$

is the reduced electric potential difference.

For an incompressible layer, the conservation law of the macromolecular species reads

$$C_1 + C_2 + C_3 = C_{\mathrm{T}}\,, \tag{16}$$

where C_{T} is a constant.

Moreover, we will assume here that the fixation steps (B) and (C) are always in a quasi-steady state, i.e., that

$$\frac{\partial C_2}{\partial t} \approx 0\,. \tag{17}$$

With relations (5), (6), (10), (11), (12) and (16) the kinetic equations reduce to

$$\frac{\partial C_{\mathrm{R}}}{\partial t} = k_{\mathrm{c}}[C_{\mathrm{T}} - C_{\mathrm{R}} - l^*\mathrm{e}^{\Delta\mu E/RT}\,\Lambda^{C_{\mathrm{R}}}(C_{\mathrm{R}} - C_2)] - \operatorname{div} v^{\mathrm{s}}C_{\mathrm{R}} + D_{\mathrm{R}}\Delta^{\mathrm{s}}C_{\mathrm{R}}\,, \tag{18}$$

$$\frac{\partial A}{\partial t} = k_p^{\mathrm{i}*}(A_{\mathrm{i}}\mathrm{e}^{-p\chi/4} - A\,\mathrm{e}^{p\chi/4}) + k_{\mathrm{d}}^{\mathrm{i}*}C_2\mathrm{e}^{(1-p)\chi/4} - k_{\mathrm{a}}^{\mathrm{i}*}A(C_{\mathrm{R}} - C_2)\,\mathrm{e}^{(p-1)\chi/4} - \operatorname{div} v^{\mathrm{s}}A + D_A\Delta^{\mathrm{s}}A\,, \tag{19}$$

$$\frac{\partial B}{\partial t} = k_p^{\mathrm{e}*}[B_{\mathrm{e}}\mathrm{e}^{p\chi/4} - B\mathrm{e}^{-p\chi/4}] + k_{\mathrm{d}}^{\mathrm{e}*}C_2\mathrm{e}^{(p-1)\chi/4} - k_{\mathrm{a}}^{\mathrm{e}*}B(C_{\mathrm{R}} - C_2)\,\mathrm{e}^{(1-p)\chi/4} - \operatorname{div} v^{\mathrm{s}}B + D_B\Delta^{\mathrm{s}}B\,, \tag{20}$$

where $D_{\mathrm{R}} = D_1 = D_2$.

2.1. Stationary states

In the absence of diffusion migration and convection, the eqs. (9), (18), (19) and (20) read at the steady state (superscript 0)

$$C_2^0 = \frac{k_{\mathrm{a}}^*[A^0\mathrm{e}^{(p-1)\chi/4} + B^0\mathrm{e}^{(1-p)\chi/4}]\,C_{\mathrm{R}}^0}{k_{\mathrm{a}}^*[A^0\mathrm{e}^{(p-1)\chi/4} + B^0\mathrm{e}^{(1-p)\chi/4}] + 2k_{\mathrm{d}}^*\cosh(1-p)\chi/4}\,, \tag{21}$$

$$k_{\mathrm{c}}[C_{\mathrm{T}} - C_{\mathrm{R}}^0 - l^*\mathrm{e}^{\Delta\mu^*\chi}\Lambda^{C_{\mathrm{R}}^0}(C_{\mathrm{R}}^0 - C_2^0)] = 0\,, \tag{22}$$

$$k_p^{\mathrm{i}*}[A_{\mathrm{i}}\mathrm{e}^{-p\chi/4} - A^0\mathrm{e}^{p\chi/4}] + k_{\mathrm{d}}^*C_2^0\,\mathrm{e}^{(1-p)\chi/4} - k_{\mathrm{a}}^*A^0(C_{\mathrm{R}}^0 - C_2^0)\,\mathrm{e}^{(p-1)\chi/4} = 0\,, \tag{23}$$

$$k_p^{\mathrm{e}*}[B_{\mathrm{e}}\mathrm{e}^{p\chi/4} - B^0\mathrm{e}^{-p\chi/4}] + k_{\mathrm{d}}^*C_2^0\,\mathrm{e}^{(p-1)\chi/4} - k_{\mathrm{a}}^*B^0(C_{\mathrm{R}}^0 - C_2^0)\,\mathrm{e}^{(1-p)\chi/4} = 0\,, \tag{24}$$

with

$$\Delta\mu^* = (\mu_1 - \mu_3)/z\mathcal{F}d\,, \tag{25}$$

$$k_{\mathrm{a}}^{\mathrm{i}*} = k_{\mathrm{a}}^{\mathrm{e}*} = k_{\mathrm{a}}^{*} \quad \text{and} \quad k_{\mathrm{d}}^{\mathrm{i}*} = k_{\mathrm{d}}^{\mathrm{e}*} = k_{\mathrm{d}}^{*} \,. \tag{26}$$

Van Roten (cf. [6] fig. 11) shows, that for peculiar values of the constants, the system is characterized by multiple steady states (fig. 2).

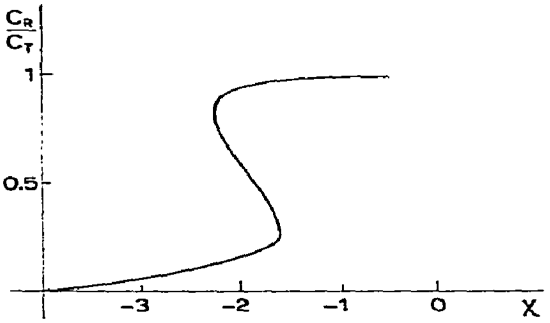

Fig. 2. Mole fraction $C_{\mathrm{R}}/C_{\mathrm{T}}$ of the open macromolecules versus the reduced potential χ (see ref. [6]); here

$$\frac{C_{\mathrm{T}}\eta}{RT} = 5\,, \quad A_{\mathrm{i}}\frac{k_{\mathrm{a}}^{*}}{k_{\mathrm{d}}^{*}} = 1.26\,, \quad Be\frac{k_{\mathrm{a}}^{*}}{k_{\mathrm{d}}^{*}} = 11.81\,, \quad C_{\mathrm{T}}k_{\mathrm{a}}^{*} = 1\,,$$

$$\frac{k_p^{*}}{C_{\mathrm{T}}k_{\mathrm{a}}^{*}} = 5 \times 10^{3}\,, \quad p = 0.95\,, \quad \frac{\mu_3 - \mu_1}{z\mathcal{F}d} = 2\,,$$

$$l^{*} = 4\,, \quad k_{\mathrm{c}} = 1\,.$$

3. Hydrodynamical aspect

Let is now consider the hydrodynamic motion in the two homogeneous phases i and e, where the electric field is zero. For an infinitesimal perturbation of the steady state (at rest), the Navier–Stokes equations read

$$\rho\frac{\partial v_x}{\partial t} = -\frac{\partial\,\delta p}{\partial x} + \mu\Delta v_x\,, \tag{27}$$

$$\rho\frac{\partial v_y}{\partial t} = -\frac{\partial\,\delta p}{\partial y} + \mu\Delta v_y\,, \tag{28}$$

$$\rho\frac{\partial v_z}{\partial t} = -g\,\delta\rho - \frac{\partial}{\partial z}\delta p + \mu\Delta v_z\,, \tag{29}$$

where μ is the viscosity coefficient, p the pressure and ρ the density.

The stability of the reference steady state will be tested by means of a normal mode analysis (see for example [16, chap. 10]). Using the continuity equation, the incompressibility condition and, the following boundary conditions

$$v_z^{\mathrm{i}} = v_z^{\mathrm{e}} = v_z^{\mathrm{s}}\,, \tag{30}$$

$$\text{at } z = z_{\mathrm{s}}\,,$$

$$\mathrm{D}v_z^{\mathrm{i}} = \mathrm{D}v_z^{\mathrm{e}} = (\mathrm{D}v_z)^{\mathrm{s}}\,, \tag{31}$$

where D = d/dz,

$$\left\{\rho^{\mathrm{e}} - \frac{\mu^{\mathrm{e}}}{\omega}(\mathrm{D}^2 - k^2)(\mathrm{D}v_z)^{\mathrm{s}}\right\} - \left\{\rho^{\mathrm{i}} - \frac{\mu^{\mathrm{i}}}{\omega}(\mathrm{D}^2 - k^2)(\mathrm{D}v_z)^{\mathrm{s}}\right\} =$$

$$-\frac{k^2}{\omega}g(\rho^{\mathrm{e}} - \rho^{\mathrm{i}})v_z^{s} + \frac{k^4}{\omega}\sigma^0 v_z^{s} - \frac{2k^2}{\omega}(\mathrm{D}v_z)^{s}(\mu^{\mathrm{e}} - \mu^{\mathrm{i}})\,, \tag{32}$$

$$\mu^{e}\left(\frac{\partial v_x}{\partial z}+\frac{\partial v_z}{\partial x}\right)^{e}-\mu^{i}\left(\frac{\partial \mu_x}{\partial z}+\frac{\partial v_z}{\partial x}\right)^{i}=\frac{\partial\,\delta\sigma}{\partial x}\,, \tag{33}$$

(Landau–Levich conditions [17]). (A similar equation is also writen for the y coordinate.) Here k is the wave number and σ the surface tension depending on the electric field [18].

Let us now assume that the fluctuation of surface tension is related to the fluctuations of the concentrations of the chemical species by a state equation (local equilibrium [14]):

$$\delta\sigma = a_1\delta C_{\mathrm{R}} + a_2\delta A + a_3\delta B\,, \tag{34}$$

where a_1, a_2, a_3 are constants.

The solutions for the two volume phases can be read, for the amplitude of the velocity perturbation

$$\tilde{v}_z^{i} = \mathcal{A}_1 e^{kz} + \mathcal{B}_1 e^{q_1 z}\,, \quad z<0\,, \tag{35}$$

$$\tilde{v}_z^{e} = \mathcal{A}_2 e^{-kz} + \mathcal{B}_2 e^{-q_2 z}\,, \quad z>0\,, \tag{36}$$

where $q_1 = (k^2+\omega\rho^{i}/\mu^{i})^{1/2}$ and $q_2 = (k^2+\omega\rho^{e}/\mu^{e})^{1/2}$. These solutions must satisfy the boundary conditions (30)–(33). i.e.,

$$\mathcal{A}_1+\mathcal{B}_1-\mathcal{A}_2-\mathcal{B}_2=0\,, \tag{37}$$

$$k\mathcal{A}_1+q_1\mathcal{B}_1+k\mathcal{A}_2+q_2\mathcal{B}_2=0\,, \tag{38}$$

$$\begin{aligned}&\left\{k\rho^{i}+\frac{k^4\sigma^0}{2\omega^2}+\frac{gk^2}{2\omega^2}(\rho^{i}-\rho^{e})+\frac{k^3}{\omega}(\mu^{i}-\mu^{e})\right\}\mathcal{A}_1+\left\{\frac{k^4\sigma^0}{2\omega^2}+\frac{gk^2}{2\omega^2}(\rho^{i}-\rho^{e})+\frac{k^2}{\omega}q_1(\mu^{i}-\mu^{e})\right\}\mathcal{B}_1\\&\quad+\left\{k\rho^{e}+\frac{k^4\sigma^0}{2\omega^2}+\frac{gk^2}{2\omega^2}(\rho^{i}-\rho^{e})+\frac{k^3}{\omega}(\mu^{i}-\mu^{e})\right\}\mathcal{A}_2+\left\{\frac{k^4\sigma^0}{2\omega^2}+\frac{gk^2}{2\omega^2}(\rho^{i}-\rho^{e})-\frac{k^2}{\omega}q_2(\mu^{i}-\mu^{e})\right\}\mathcal{B}_2=0\,.\end{aligned} \tag{39}$$

$$2\mu^{i}k^2\mathcal{A}_1+\mu^{i}(q_1^2+k^2)\mathcal{B}_1-2\mu^{e}k^2\mathcal{A}_2-\mu^{e}(q_2^2+k^2)\mathcal{B}_2-k^2a_1\mathcal{C}_1-k^2a_2\mathcal{C}_2-k^2a_3\mathcal{C}_3=0\,, \tag{40}$$

where $\mathcal{C}_1, \mathcal{C}_2, \mathcal{C}_3$ are the amplitude respectively of $\delta C_{\mathrm{R}}, \delta A$ and, δB.

Other boundary conditions are related to the kinetics of the reactions in the surface layer. From eqs. (19), (20), (20) and (22) one writes for one normal mode

$$\{\omega+k_{\mathrm{c}}[1-\eta^{*}(1-r^0)+l^{*}\Lambda^{*r^0}e^{\Delta\mu^{*}\chi}]+k^2D_{\mathrm{R}}\}\,\mathcal{C}_1=0\,, \tag{41}$$

$$k_{\mathrm{a}}^{*}A^0e^{(p-1)\chi/4}\,\mathcal{C}_1+\left\{\omega+k_p^{*}e^{p\chi/4}+C_{\mathrm{T}}k_{\mathrm{a}}^{*}\frac{(1-r^0)\,e^{(p-1)\chi/4}}{l^{*}\Lambda^{*r^0}e^{\Delta\mu^{*}\chi}}\right\}\mathcal{C}_2=0\,, \tag{42}$$

$$k_{\mathrm{a}}^{*}B^0e^{(1-p)\chi/4}\,\mathcal{C}_1+\left\{\omega+k_p^{*}e^{-p\chi/4}+k_{\mathrm{a}}^{*}C_{\mathrm{T}}\frac{(1-r^0)\,e^{(1-p)\chi/4}}{l^{*}\Lambda^{*r^0}e^{\Delta\mu^{*}\chi}}\right\}\mathcal{C}_3=0\,, \tag{43}$$

where $r^0 = C_{\mathrm{R}}^0/C_{\mathrm{T}}$, $\eta^{*} = \eta C_{\mathrm{T}}/RT$, $\Lambda^{*r^0} = \Lambda^{C_{\mathrm{R}}^0}$. For the sake of simplicity, we have assumed here that all the terms in δC_2 are small in comparison with the other terms. The determinant of the system of eqs. (37)–(43) gives rise to a secular equation which reads

$$\mathcal{D}_H(\omega + k_c[1 - \eta^*(1 - r^0) + l^*\Lambda^{*r^0}\, e^{\Delta\mu^*\chi}] + k^2 D_R)$$

$$\times\left(\omega + k_p^* e^{p\chi/4} + C_T k_a^* \frac{(1 - r^0)\, e^{(p-1)\chi/4}}{l^*\Lambda^{*r^0}\, e^{\Delta\mu^*\chi}}\right)\left(\omega + k_p^* e^{-p\chi/4} + k_a^* C_T \frac{(1 - r^0)\, e^{(1-p)\chi/4}}{l^*\Lambda^{*r^0}\, e^{\Delta\mu^*\chi}}\right) = 0\,, \qquad (44)$$

where $\mathcal{D}_H$ is the determinant of the hydrodynamic eqs. (37)–(40).

4. Chemical and hydrodynamic stability

In the present paper we will restrict our analysis to an inviscid system. It is easy to see that the contributions to the instability are only given by the product

$$\mathcal{D}_H(\omega + k_c[1 - \eta^*(1 - r^0) + l^*\Lambda^{*r^0}\, e^{\Delta\mu^*\chi}] + D_R/\lambda^2)\,,$$

where λ is the wavelength $\lambda = 1/k$. The pure hydrodynamic condition (Rayleigh–Tailor instability [16, p.435]) is:

$$\omega^2 = \frac{g}{\lambda}\left\{\frac{\rho^e - \rho^i}{\rho^e + \rho^i} - \frac{\sigma^0}{\lambda^2 g(\rho^e + \rho^i)}\right\}\,. \qquad (45)$$

If $\rho^e < \rho^i$ the arrangement is stable; if $\rho^e > \rho^i$ the arrangement is stable for $\lambda < \lambda_c$ where

$$\lambda_c = [(\rho^e - \rho^i)g/\sigma^0]^{-1/2}\,, \qquad (46)$$

while for $0 > \lambda > \lambda_c$, with $\rho^e > \rho^i$, the arrangement is unstable. The mode of maximum instability is reached for a wavelength λ_* and a frequency ω_*.

$$\lambda_* = \sqrt{3}\,\lambda_c\,, \qquad (47)$$

$$\omega_* = [2(\rho^e - \rho^i)^{3/2} g^{3/2}/3^{3/2}(\rho^e + \rho^i)(\sigma^0)^{1/2}]^{1/2}\,. \qquad (48)$$

The surface tension stabilizes the system. The pure chemical condition is

$$\omega = -k_c[1 - \eta^*(1 - r^0) + l^*\Lambda^{*r^0}\, e^{\Delta\mu^*\chi}] - D_R/\lambda^2\,; \qquad (49)$$

the chemical state is stable if

$$k_c\eta^*(1 - r^0) < k_c(1 + l\Lambda^{*r^0}) + D_R/\lambda^2\,, \qquad (50)$$

and unstable if the sign is reversed. The diffusion has a stabilizing character. After the instability threshold, the pure chemical system has an infinite wavelength for the mode of maximum instability. In the absence of diffusion and convection, the intermediate steady state is unstable while the two other ones are stable (for example if $\chi = -2, \eta^* = 5, l = 221$).

However when we consider the convection, the two stable states can become unstable if $\rho^e > \rho^i$ with $0 > \lambda > \lambda_c$. Furthermore the concentrations A and B become dependent with the critical wavelength λ_*. On the other hand, even if the pure hydrodynamic stability conditions are fulfilled, an unstable chemical intermediate state may induce convective motion in the two homogeneous liquid phases. It means that the surface layer undergoes an infinitesimal deformation δz_s (because $v_z^s/\omega = \delta z_s$) that will grow in time.

Remark: Calculations have been made without the assumption on the smallness of the terms in δC_2. The pure chemical stability condition is then different from (49) and after the instability threshold, the system could reach a mode of maximum instability for a finite critical wavelength. The chemical instability could then induce a hydrodynamic instability (i.e., a deformation of the interface) of the same wavelength.

5. Conclusions

The present analysis shows that a chemical instability of a reaction occurring at the interface between two immiscible fluids is able to induce a mechanical instability.

Reciprocally, a mechanical instability may induce a chemical instability in a reaction taking place at the interface. Moreover, the velocity patterns appearing after the instability threshold induce chemical patterns of the species at the surface.

The model studied here could give a picture of the deformation of the biological cells when they receive a chemical "message".

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