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BOUNDARY CONDITIONS FOR A CAPILLARY FLUID IN CONTACT WITH A WALL

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Abstract

Contact of a fluid with a solid or an elastic wall is investigated. The wall exerts *molecular forces* on the fluid which is locally strongly nonhomogeneous. The problem is approached with a fluid energy of the second gradient form and a wall surface energy depending on the value of the fluid density at the contact. From the virtual work principle are obtained limit conditions taking into account the fluid density, its normal derivative to the wall and the curvature of the surface.

1. Introduction

The phenomenon of surface wetting is a subject of many experiments [1]. Such experiments have been used to determine many important properties of the wetting behavior for liquid on low energy surface [2]. In fact the wetting transition of fluids in contact with solid surfaces is an important field of research both for mechanics and physical chemistry. In the recent paper [3], the

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first author using statistical methods proposed an explicit form for the energy of interaction between solid surfaces and liquids. This energy yields a bridge connecting statistical mechanics and continuum mechanics. To obtain boundary conditions between fluid and solid it is also necessary to know the behaviour of the fluid as well as the solid.

We propose a mechanical model similar to that used in the mean-field theory of capillarity that leads to the second gradient theory of continuous media in fluid mechanics [4]. The theory is conceptually more straightforward than the Laplace one to build a model of capillarity [5,6]. That theory takes into account systems in which fluid interfaces are present [7]. The internal capillarity is one of the simplest cases since we are able to calculate the superficial tension in the case of thin interfaces as well as in thick ones [8]. It is possible to obtain the nucleation of drops and bubbles [9].

It seems that the approximation of the mean-field theory is too simple to be quantitatively accurate. However, it does provide a qualitative understanding. Moreover, the point of view, that the fluid in interfacial region may be treated as a bulk phase with a local free energy density and an additional contribution arising from the nonuniformity which may be approximated by a gradient expansion truncated at the second order, is most likely to be successful and perhaps even quantitatively accurate near the critical point [10].

In this paper we connect both the interaction of a solid surface and a fluid phase by means of the virtual work principle. The distribution of fluid energy in the volume and the surface density energy on the solid surface yield boundary conditions. The conditions are different from those obtained for a classical fluid within the theory of gas dynamics. We obtain an embedding effect for the density of the fluid; moreover, the conditions take into account the curvature of the surface. The result is extended to the case of an elastic wall.

A discussion is obtained depending on the value of the density of the fluid at

the surface.

Let us use asterisk "*" to denote *conjugate* (or *transpose*) mappings or covectors (line vectors). For any vectors \mathbf{a}, \mathbf{b} we shall use the notation $\mathbf{a}^*\mathbf{b}$ for their *scalar product* (the line vector is multiplied by the column vector) and $\mathbf{a}\mathbf{b}^*$ or $\mathbf{a} \otimes \mathbf{b}$ for their *tensor product* (the column vector is multiplied by the line vector). The product of a mapping A by a vector \mathbf{a} is denoted by $A \mathbf{a}$. Notation $\mathbf{b}^* A$ means covector \mathbf{c}^* defined by the rule $\mathbf{c}^* = (A^* \mathbf{b})^*$. The divergence of a linear transformation A is the covector $\text{div}A$ such that, for any constant vector \mathbf{a} ,

$$\text{div}(A) \mathbf{a} = \text{div} (A \mathbf{a}).$$

If $f(\mathbf{x})$ is a scalar function of the vector \mathbf{x} associated with the Euler variables in the physical space, $\frac{\partial f}{\partial \mathbf{x}}$ is the linear form associated with the gradient of f and $\frac{\partial f}{\partial x^i} = (\frac{\partial f}{\partial \mathbf{x}})_i$. Consequently, $(\frac{\partial f}{\partial \mathbf{x}})^* = \text{grad } f$

2. Continuous mechanical model of capillary layers

We consider a fluid in contact with a solid. The fluid occupies the domain D and its boundary Σ which is common with the solid wall. Physical experiments prove that the fluid is nonhomogeneous in the neighbourhood of Σ [10]. It is also possible to consider the fluid as a continuous medium by taking into account a *capillary layer* existing in the vicinity of Σ and a form of its stress tensor [11]. One way to present the behaviour of such a fluid is to consider the specific internal energy ε as a function of the density ρ as well as $\text{grad } \rho$. Such an expression is known in continuum mechanics as *internal capillary energy*, see [4,5]. It is related to molecular models of strongly non homogeneous fluids in the frame of the mean field theory and is equivalent to the van der Waals model of capillarity (see the review by Rowlinson and Widom [10]). The energy ε is also a function of the specific entropy. In the case of isothermal media at a given temperature, the specific internal energy is replaced by the specific free energy.

In the mechanical case the entropy or the temperature are not concerned by the virtual variations of the medium. Consequently, for an isotropic fluid, it is assumed that

$$\varepsilon = f(\rho, \beta)$$

where $\beta = (\overrightarrow{\text{grad}} \rho)^2 = \text{grad} \rho \cdot \text{grad} \rho$ (the dot \cdot denotes the scalar product). The fluid is submitted to external forces represented by a force potential Ω per unit mass as a function of Euler variables \mathbf{x} .

We denote by $\mathbf{x} \in \Sigma \longrightarrow B(\mathbf{x}) \in R$ the surface density of energy of the solid wall. The total energy E of the fluid in D and its boundary Σ is the sum of the three potential energies: $E = E_f + E_p + E_S$ with,

$$E_f = \int \int \int_D \rho \varepsilon(\rho, \beta) dv, \quad E_p = \int \int \int_D \rho \Omega(\mathbf{x}) dv, \quad E_S = \int \int_{\Sigma} B ds$$

Let us denote by δ a variation of the position of the fluid as in [12]. The variation is associated with the *virtual displacement*

$$\mathbf{x} \in D \rightarrow \delta \mathbf{x} = \zeta(x)$$

We have the following results presented in Appendix,

$$\begin{aligned} \delta E_f &= \int \int \int_D (-\text{div } \sigma) \cdot \zeta dv + \\ &\int \int_{\Sigma} \left\{ -A \frac{d\zeta_n}{dn} + \left(\frac{2A}{R_m} \mathbf{n} + \text{grad}_{tg} A + \sigma \mathbf{n} \right) \cdot \zeta \right\} ds \end{aligned} \quad (1)$$

with

$$\sigma = -P\mathbf{I} - C \text{grad} \rho \otimes \text{grad} \rho = -P\mathbf{I} - C \left(\frac{\partial \rho}{\partial \mathbf{x}} \right)^* \frac{\partial \rho}{\partial \mathbf{x}}$$

where $C = 2\rho \varepsilon'_\beta$ and $P = \rho^2 \varepsilon'_\rho - \rho \text{div}(C \text{grad} \rho)$,

ε'_ρ denotes the partial derivative of ε with respect to ρ , $\zeta_n = \mathbf{n}^* \zeta$ where \mathbf{n} is the external unit normal to Σ and $A = C \rho \frac{d\rho}{dn}$ where $\frac{d\rho}{dn} = \frac{\partial \rho}{\partial \mathbf{x}} \cdot \mathbf{n}$.

The scalar R_m is the mean curvature of Σ and grad_{tg} is the tangential part of grad relatively to Σ .

Moreover,

$$\delta E_p = \int \int \int_D \rho \frac{\partial \Omega}{\partial \mathbf{x}} \zeta dv = \int \int \int_D \rho \text{grad} \Omega \cdot \zeta dv \quad (2)$$

and using the results presented in Appendix,

$$\delta E_S = \int \int_{\Sigma} \delta B - \left(\frac{2B}{R_m} \mathbf{n} + \text{grad}_{tg} B \right) \cdot \zeta \, ds \quad (3)$$

One assumes that the volume mass in the fluid has a limit value ρ_s at the wall Σ . One assumes also that B is a function of ρ_s only. These hypotheses are confirmed by results presented in [3]. Then

$$\delta B = B'(\rho_s) \delta \rho_s = -\rho_s B'(\rho_s) \text{div} \zeta$$

Let us denote $G = -\rho_s B'_{\rho_s}$. Consequently,

$$\int \int_{\Sigma} \delta B \, ds = \int \int_{\Sigma} G \text{div} \zeta \, ds = \int \int_{\Sigma} \left(G \frac{d\zeta_n}{dn} - \frac{2G}{R_m} \mathbf{n} \cdot \zeta - \text{grad}_{tg} G \cdot \zeta \right) ds$$

(see Appendix).

Now, $H = B(\rho_s) - \rho_s B'_{\rho_s}(\rho_s)$ is the Legendre transformation of B with respect to ρ_s . Then,

$$\delta E_S = \int \int_{\Sigma} G \frac{d\zeta_n}{dn} - \left(2H \frac{\mathbf{n}}{R_m} + \text{grad}_{tg} H \right) \cdot \zeta \, ds \quad (4)$$

The d'Alembert-Lagrange principle of virtuals works is expressed in the form [12] :

$$\forall \mathbf{x} \in D \rightarrow \zeta(\mathbf{x}), \quad \delta E = 0 \quad (5)$$

Consequently, from the fundamental lemma of variation calculus, we obtain the balance equation in the fluid D and the boundary conditions on the solid wall Σ .

Equilibrium equations :

From any arbitrary variation $\mathbf{x} \in D \rightarrow \zeta(\mathbf{x})$ such that $\zeta = \mathbf{0}$ on Σ , we take first

$$\int \int \int_D \left(\rho \frac{\partial \Omega}{\partial \mathbf{x}} - \text{div} \sigma \right) \zeta \, dv = 0$$

Consequently,

$$-\text{div} \sigma + \rho \frac{\partial \Omega}{\partial \mathbf{x}} = 0 \quad (6)$$

This equation is the well known equilibrium equation [5,7,9]

Boundary conditions :

a) Case of a rigid (undeformed) wall.

We consider a rigid wall. Consequently, the virtual displacements satisfy on Σ the condition $\mathbf{n}^* \zeta = 0$. Then, at the rigid wall

$$\int \int_{\Sigma} (G - A) \frac{d\zeta_n}{dn} + \left\{ \frac{2(A - H)}{R_m} \mathbf{n} + \text{grad}_{tg}(A - H) + \sigma \mathbf{n} \right\} \cdot \zeta \, d\sigma = 0$$

Hence, we deduce the boundary conditions at the rigid wall

$$\text{For } \mathbf{x} \in \Sigma, \quad G - A = 0 \quad (7)$$

and moreover, there exists a Lagrange multiplier $\mathbf{x} \in \Sigma \rightarrow \lambda(\mathbf{x}) \in R$ such that

$$\frac{2(A - H)}{R_m} \mathbf{n} + \text{grad}_{tg}(A - H) + \sigma \mathbf{n} = \lambda \mathbf{n} \quad (8)$$

b) Case of a elastic (non-rigid) solid wall.

In such a case the equilibrium equation(6) is unchanged. On Σ the condition (7) is also unchanged. The only different condition comes from the fact that we do not have anymore the slipping condition for the virtual displacement ($\mathbf{n}^* \zeta = 0$).

Due to the possible deformation of the wall, the virtual work of mechanical stresses on Σ is

$$\delta E_e = \int \int_{\Sigma} \mathbf{T}^* \zeta \, ds$$

with $\mathbf{T} = Q \mathbf{n}$ representing the stress (loading) vector, where Q is the value of the Cauchy stress tensor of the wall on the boundary Σ .

Relation (8) is replaced by :

$$2 \frac{(A - H)}{R_m} \mathbf{n} + \text{grad}_{tg}(A - H) + \sigma \mathbf{n} = -\mathbf{T} \quad (9)$$

3. Analysis of the boundary conditions

Relation (7) yields :

$$C \frac{d\rho}{dn} + B'_\rho = 0 \quad (10)$$

and we obtain,

$$H - A = B.$$

Consequently, from the definition of σ ,

$$\sigma \mathbf{n} = P \mathbf{n} - C \frac{d\rho}{dn} \text{grad}\rho.$$

The tangential part of equation (8) is always verified and equation (8) yields the value of the Lagrange multiplier λ .

For an elastic (non-rigid) solid wall we obtain

$$T_{tg}^* = 0 \quad \text{and} \quad T_n = \frac{2B}{R_m} + P - B'_\rho \frac{d\rho}{dn} \quad (11)$$

where T_{tg}^* and T_n are respectively the tangential and the normal components of \mathbf{T} . Taking into account equation (10), $T_n = P + \frac{2B}{R_m} + \frac{1}{C}(B'_\rho)^2$ and equations (11) yield the value of the stresses in the elastic (non rigid) medium. The only new condition comes from equation (10).

We have the consequences:

In [3] we propose the surface energy in the form $B(\rho) = -\gamma_1\rho + \frac{\gamma_2}{2}\rho^2$ with γ_1 , and γ_2 as two positive constants. We obtain the condition for the fluid density on the wall

$$C \frac{d\rho}{dn} = \gamma_1 - \gamma_2\rho \quad (12)$$

Denoting again ρ_s the value of ρ on the wall Σ (the limit value of the volumic mass of the fluid on the wall Σ), we obtain that $\frac{d\rho}{dn}$ is positive (or negative) in the vicinity of the wall if $\rho_s < \rho_i$ (or $\rho_s > \rho_i$) with $\rho_i = \frac{\gamma_1}{\gamma_2}$ which is the *bifurcation fluid density* at the wall.

If $\rho_s < \rho_i$ we have a lack of fluid density at the wall. If $\rho_s > \rho_i$ we have an excess of fluid density at the wall.

Conclusion

For conservative medium, the first gradient theory corresponds to the case of compressibility. To take into account superficial effects acting between solids and fluids, we propose to use the model of fluids endowed with capillarity. The theory interprets the capillarity in a continuous way and contains Laplace's theory. The model corresponds for solids to *elastic materials with couple stresses* indicated by Toupin in [13].

We notice that the extension to the dynamic case is straightforward: by virtual work principle, equation (6) takes the form:

$$\rho\gamma^* - \operatorname{div} \sigma + \rho \frac{\partial \Omega}{\partial \mathbf{x}} = 0,$$

where γ denotes the acceleration of the fluid. Equations (10), (11), (12) and consequences in paragraph 3 are unchanged.

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Appendix

First of all we recall the following fact issued from differential geometry:

Let Σ be a surface in the 3-dimensional space and \mathbf{n} its external normal. For any vector field ζ ,

$$\mathbf{n}^* \text{rot}(\mathbf{n} \times \zeta) = \text{div} \zeta + \frac{2}{R_m} \mathbf{n}^* \zeta - \mathbf{n}^* \frac{\partial \zeta}{\partial \mathbf{x}} \mathbf{n}.$$

Then, for any scalar field A , we obtain:

$$\begin{aligned} A \text{div} \zeta &= A \frac{d\zeta_n}{dn} - \frac{2A}{R_m} \zeta_n - (\text{grad}_{tg}^* A) \zeta + \mathbf{n}^* \text{rot} (A \mathbf{n} \times \zeta) \\ &= \text{tr} \left[\left(\frac{\partial A}{\partial \mathbf{x}} (\mathbf{nn}^* - \mathbf{1}) - \frac{2A}{R_m} \mathbf{n}^* \right) \zeta \right] + A \frac{d\zeta_n}{dn} + \mathbf{n}^* \text{rot}(A \mathbf{n} \times \zeta) \end{aligned} \quad (\text{A.1})$$

Let us calculate δE_f : since D is a material volume,

$$E_f = \int \int \int_D \rho \varepsilon \, dv \quad \Rightarrow \quad \delta E_f = \int \int \int_D \rho \delta \varepsilon \, dv$$

$$\text{with } \delta \varepsilon = \frac{\partial \varepsilon}{\partial \rho} \delta \rho + \frac{\partial \varepsilon}{\partial \beta} \delta \beta.$$

From $\delta \frac{\partial \rho}{\partial \mathbf{x}} = \frac{\partial \delta \rho}{\partial \mathbf{x}} - \frac{\partial \rho}{\partial \mathbf{x}} \frac{\partial \zeta}{\partial \mathbf{x}}$, we deduce

$$\rho \varepsilon'_\beta \delta \beta = 2\rho \varepsilon'_\beta \delta \left(\frac{\partial \rho}{\partial \mathbf{x}} \right) \frac{\partial \rho}{\partial \mathbf{x}} = C \left(\frac{\partial \delta \rho}{\partial \mathbf{x}} - \frac{\partial \rho}{\partial \mathbf{x}} \frac{\partial \zeta}{\partial \mathbf{x}} \right) \frac{\partial \rho}{\partial \mathbf{x}}$$

with $2\rho \varepsilon'_\beta = C$.

In the mean-field molecular theory, the quantity C is assumed constant [10], but it is not necessary for our calculations. One can suppose the scalar C is a general function of ρ and even β . Then

$$\rho \varepsilon'_\beta \delta \beta = \text{div}(C \text{grad} \rho \delta \rho) - \text{div}(C \text{grad} \rho) \delta \rho - \text{tr} \left(C \text{grad} \rho \text{grad}^* \rho \frac{\partial \zeta}{\partial \mathbf{x}} \right)$$

Due to the fact that $\delta \rho = -\rho \text{div} \zeta$ (see [12]),

$$\begin{aligned} \rho \delta \varepsilon &= \text{div}(C \text{grad} \rho \delta \rho) - \left(\rho^2 \varepsilon'_\rho - \rho \text{div}(C \text{grad} \rho) \right) \text{div} \zeta \\ &\quad - \text{div}(C \text{grad} \rho \text{grad}^* \rho \zeta) + \text{div}(C \text{grad} \rho \text{grad}^* \rho) \zeta \end{aligned}$$

$$\begin{aligned}\rho \delta \varepsilon &= \operatorname{div} \left(C \operatorname{grad} \rho \delta \rho - (C \operatorname{grad} \rho \operatorname{grad}^* \rho) \zeta - P \zeta \right) \\ &\quad + \frac{\partial P}{\partial \mathbf{x}} \zeta + \operatorname{div} (C \operatorname{grad} \rho \operatorname{grad}^* \rho) \zeta\end{aligned}$$

Then

$$\begin{aligned}\delta E_f &= \int \int \int_D \left(\frac{\partial P}{\partial \mathbf{x}} + \operatorname{div} (C \operatorname{grad} \rho \operatorname{grad}^* \rho) \right) \zeta dv \\ &\quad - \int \int \int_D \operatorname{div} \left(C \rho \operatorname{grad} \rho \operatorname{div} \zeta + C \operatorname{grad} \rho \operatorname{grad}^* \rho \zeta + P \zeta \right) dv \\ &= \int \int \int_D -(\operatorname{div} \sigma) \zeta dv + \int \int_{\Sigma} (-A \operatorname{div} \zeta + \mathbf{n}^* \sigma \zeta) ds\end{aligned}$$

Taking into account (A.1), we deduce immediately

$$\begin{aligned}\delta E_f &= \int \int \int_D -(\operatorname{div} \sigma) \zeta dv \\ &\quad + \int \int_{\Sigma} \left(-A \frac{d\zeta_n}{dn} + \left(\frac{2A}{R_m} \mathbf{n}^* + \operatorname{grad}_{t_g}^* A + \mathbf{n}^* \sigma \right) \zeta \right) ds + \int \int_{\Sigma} \mathbf{n}^* \operatorname{rot} (A \mathbf{n} \times \zeta) ds \\ \text{But } \int \int_{\Sigma} \mathbf{n}^* \operatorname{rot} (A \mathbf{n} \times \zeta) ds &= \int_{\Gamma} \mathbf{A} \mathbf{t} \cdot (\mathbf{n} \times \zeta) d\ell = \int_{\Gamma} A(\mathbf{t}, \mathbf{n}, \zeta) d\ell\end{aligned}$$

where Γ is the line boundary of Σ and \mathbf{t} its tangent unit vector. If $\mathbf{n}' = \mathbf{t} \times \mathbf{n}$ we obtain the relation

$$\begin{aligned}\delta E_f &= \int \int \int_D (-\operatorname{div} \sigma) \zeta dv + \\ &\quad \int \int_{\Sigma} \left(-A \frac{d\zeta_n}{dn} + \left(\frac{2A}{R_m} \mathbf{n}^* + \operatorname{grad}_{t_g}^* A + \mathbf{n}^* \sigma \right) \zeta \right) ds + \int_{\Gamma} A \mathbf{n}' \cdot \zeta d\ell \quad (\text{A.2})\end{aligned}$$

In the following we assume that Σ has no boundary and consequently, the term associated with Γ vanishes.

Let us calculate δE_S

$$E_S = \int \int_{\Sigma} B ds$$

Then

$$\delta E_S = \int \int_{\Sigma} \delta B - \left(\mathbf{n}^* \frac{2B}{R_m} + \operatorname{grad}^* B (\mathbf{1} - \mathbf{nn}^*) \right) \zeta ds + \int_{\Gamma} A \mathbf{n}' \cdot \zeta d\ell \quad (\text{A.3})$$

We notice that $\operatorname{grad}^* B (\mathbf{1} - \mathbf{nn}^*)$ belong to the tangent plane to Σ .

Let us proof equation (A.3): If we write $E_S = \int \int_{\Sigma} B \det (\mathbf{n}, d_1 \mathbf{x}, d_2 \mathbf{x})$, where $d_1 \mathbf{x}$ and $d_2 \mathbf{x}$ are the coordinate lines of Σ , we may write,

$$E_S = \int \int_{\Sigma_0} B \det F \det (F^{-1} \mathbf{n}, d_1 \mathbf{X}, d_2 \mathbf{X})$$

where Σ_0 is the image of Σ in a reference space in Lagrangian coordinates \mathbf{X} and F the deformation gradient tensor $\frac{\partial \mathbf{x}}{\partial \mathbf{X}}$.

Then,

$$\delta E_S = \int \int_{\Sigma_0} \delta B \det F \det (F^{-1} \mathbf{n}, d_1 \mathbf{X}, d_2 \mathbf{X}) + \int \int_{\Sigma_0} B \delta \left(\det F \det (F^{-1} \mathbf{n}, d_1 \mathbf{X}, d_2 \mathbf{X}) \right)$$

Moreover,

$$\begin{aligned} & \int \int_{\Sigma_0} B \delta \left(\det F \det (F^{-1} \mathbf{n}, d_1 \mathbf{X}, d_2 \mathbf{X}) \right) = \\ & \int \int_{\Sigma} B \operatorname{div} \zeta \det (\mathbf{n}, d_1 \mathbf{x}, d_2 \mathbf{x}) + B \det \left(\frac{\partial \mathbf{n}}{\partial \mathbf{x}} \zeta, d_1 \mathbf{x}, d_2 \mathbf{x} \right) - B \det \left(\frac{\partial \zeta}{\partial \mathbf{x}} \mathbf{n}, d_1 \mathbf{x}, d_2 \mathbf{x} \right) \\ & = \int \int_{\Sigma} \left(\operatorname{div} (B \zeta) - \operatorname{grad}^* B \zeta - B \mathbf{n}^* \frac{\partial \zeta}{\partial \mathbf{x}} \mathbf{n} \right) ds \end{aligned}$$

From (A.1) we obtain,

$$\operatorname{div} (B \zeta) - B (\operatorname{div} \mathbf{n}) \mathbf{n}^* \zeta - \mathbf{n}^* \frac{\partial B \zeta}{\partial \mathbf{x}} \mathbf{n} = \mathbf{n}^* \operatorname{rot} (B \mathbf{n} \times \zeta)$$

Then,

$$\begin{aligned} & \int \int_{\Sigma_0} B \delta \left(\det F \det (F^{-1} \mathbf{n}, d_1 \mathbf{X}, d_2 \mathbf{X}) \right) = \\ & \int \int_{\Sigma_0} \left(B (\operatorname{div} \mathbf{n}) \mathbf{n}^* + \operatorname{grad}^* B (\mathbf{n} \mathbf{n}^* - \mathbf{1}) \right) \zeta ds + \int \int_{\Sigma} \mathbf{n}^* \operatorname{rot} (B \mathbf{n} \times \zeta) ds \end{aligned}$$

and we obtain equation (A.3) with $\operatorname{div} \mathbf{n} = -\frac{2}{R_m}$.

We assume that Σ has no boundary and consequently, the term associated with Γ is null.