A mechanical view of capillarity

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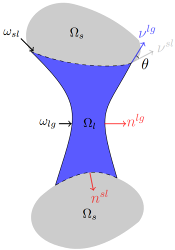
Definition of the system and associated energy

System: liquid occupying the domain Ω_l of volume V + liquid-gas interface



Internal energy of the system

$$E_{\Omega_l} = -P_{liq}V + \gamma_{lg}A_{lg} \tag{1}$$

- P_{liq} liquid pressure inside the capillary bridge
- A_{lg} surface of the liquid-gas interface
- γ_{lg} liquid-gas surface tension

Rem : the variation of liquid-gas surface energy may be seen as the one of a membrane with associated membrane stress tensor $n_t = \gamma_{lg} I_{T_{\omega_{lg}}}$

Definition of the system and associated energy

First law of thermodynamics in adiabatic conditions:

$$\delta E_{\Omega_l} - W_{F_{\mathrm{ext/sys}}} = 0 \tag{2}$$

$W_{F_{\mathrm{ext/sys}}}$ mechanical work of external forces acting on the system (liquid capillary bridge)

External work of pressure forces

$$dF^{p}_{\mathrm{ext}/\Omega_l} = -P_{\mathrm{g}}\ dS\ n_{\Omega_l} \tag{3}$$

n_{Ω_l} : unit external normal to Ω_l
P_{g} gas (air) pressure outside the capillary bridge

$$\begin{aligned} W_{F^{p}_{\mathrm{ext}/\Omega_l}} &= \int_{\partial\Omega_l} -P_{\mathrm{g}}\ n_{\Omega_l}.\delta u\ dS = -P_{\mathrm{g}} \int_{\omega_{lg}} n_{lg}.\delta u dS \\ &= -P_g\ \delta V \end{aligned} \tag{4}$$

Friction forces acting on the contact line

Friction forces exerted by the solid substrates Ω_s on the liquid Ω_l:

$$f^s_{\mathrm{ext}} = -\gamma_s \nu_{sl} - \alpha t_{slg} \tag{5}$$

• ν_{sl} and t_{slg} are the normal and tangent unit vectors to the contact ligne L_{slg} of Darboux basis $(t_{slg}, g_{sl}, n_{sl})$ of L_{slg}

• Friction forces in the tangent plane of the solid only

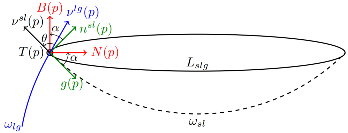


Figure: Contact line and Darboux basis

Definition of the system and associated energy

Work of external friction forces acting on the contact line

$$W_{f^s_{\text{ext}/\Omega_l}} = -\gamma_s \int_{L_{slg}} \nu_{sl}.\delta u dl - \ \alpha \int_{L_{slg}} t_{slg}.\delta u dl \tag{6}$$

We have exactly:

$$W_{f^s_{\text{ext}/\Omega_l}} = -\gamma_s \delta A_{sl} - \ \alpha \delta \mathcal{L}_{slg} \tag{7}$$

$\delta \mathcal{L}_{slg}$: variation of the the length $\mathcal{L}_{slg}$ of the contact line L_{slg}

Definition of the system and associated energy

Adhesion forces acting on the system

$$f_3^{ad} = (f_3^c + f_3^{sl}) n_{sl} \tag{8}$$

- f_3^c: normal adhesion forces located on the contact line $\mathcal{L}_{slg}$
- f_3^{sl}: normal adhesion forces located on ω_{sl}

Work of adhesion forces associated to the displacement δu :

$$W_{f_3^{ad}} = \int_{\omega_{sl}} f_3^{sl} v_3^{sl} \ d\omega + \int_{L_{slg}} f_3^c v_3^{sl} \ dl \tag{9}$$

Remark: We could also define a tangential friction/adhesion force f_t^{sl} acting on ω_{sl} with

$$W_{f_t^{sl}} = \int_{\omega_{sl}} f_t^{sl} . v_t^{sl} \ d\omega \tag{10}$$

At the end, we will prove from the final equilibrium equations that $f_t^{sl} = 0$

Definition of the system and associated energy

First law of thermodynamics in adiabatic conditions:

$$\delta E_{\Omega_l} - W_{F_{\mathrm{ext/sys}}} = 0 \tag{11}$$

General form of the energy of the system

$$\gamma_{lg}\delta A_{lg} \ + \gamma_s \delta A_{sl} \pm \gamma_{slg}\delta L_{slg} - \Delta P \ \delta V - W_{F^s_{\mathrm{ext}/\Omega_l}} = 0 \tag{12}$$

with $W_{F^s_{\mathrm{ext}/\Omega_l}}$: work of other external action on Ω_l.

In general we have $\gamma_s = \gamma_{sl} - \gamma_{sg}$ which an be identified as **a tangential friction force** exerted by the solid substrates on the liquid Ω_l

Other possible definition of the system

Equivalently, we have

$$\delta E_{\Omega} - \Delta P \ \delta V \pm \gamma_{slg} L_{slg} = 0 \tag{13}$$

with

$$E_{\Omega} = \gamma_{lg} A_{lg} \ + \gamma_s A_{sl}$$

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Calculation of different contributions of energy of the system

- **Variation of volume**

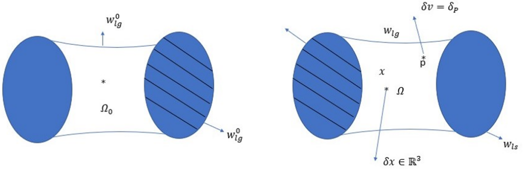

Figure: Initial and final configuration of the liquid domain

After some algebra, we can prove that we have

$$\delta V = \int_{\omega_{lg}} \delta p \cdot n_{lg} \, d\omega + \int_{\omega_{sl}} \delta p \cdot n_{sl} d\omega \tag{14}$$

Rem: Due to the use of Stokes theorem, the outer unit normals n_{lg} and n_{sl} must be considered pointing outer the liquid domain Ω_l

Calculation of different contributions of energy of the system

Decomposition of the displacement δp

$$\begin{array}{rcll} \delta p & = & v_t^{lg} + v_3^{lg} n_{lg} & \quad on \quad \omega_{lg} \\ \delta p & = & v_t^{sl} + v_3^{sl} n_{sl} & \quad on \quad \omega_{sl} \end{array} \tag{15}$$

into a tangential part and a normal part, both on ω_{sl} and ω_{lg}

General expression of the volume variation

$$\delta V = \int_{\omega_{lg}} v_3^{lg} \, d\omega + \int_{\omega_{sl}} v_3^{sl} d\omega \tag{16}$$

Calculation of different contributions of energy of the system

- **Variation of liquid gas area** A_{lg} (needs intrinsic differential geometry)

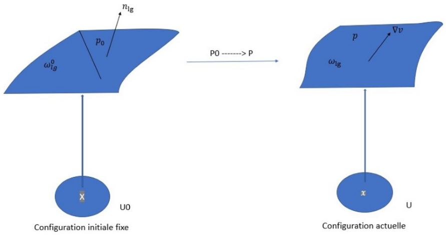

Figure: Initial and final configuration of liquid surface

Using the decomposition $\delta p = v_t^{lg} + v_3^{lg} n_{lg}$, we have exactly

$$\delta A_{lg} = -\int_{\omega_{lg}} v_3^{lg} \, \mathrm{Tr}\,(C)\, \mathrm{d}\omega + \int_{L_{slg}} v_t \cdot \nu_{lg} dl \tag{17}$$

Calculation of different contributions of energy of the system

- **Variation of solid-liquid area A_{sl}**

In a similar way, using the decomposition $\delta p = v_t^{sl} + v_3^{sl} n_{\omega_{sl}}$, we have exactly

$$\delta A_{sl} = -\int_{\omega_{sl}} v_3^{sl} \mathrm{Tr}\left(C^{sl}\right) d\omega + \int_{L_{slg}} v_t^{sl} \cdot \nu_{sl} dl \qquad (18)$$

- $\partial \omega_{sl}$ corresponds to the contact line L_{slg} ($\partial \omega_{lg}$ coincides also with L_{slg})
- ν_{sl} denotes the outer normal to the L_{slg}

Calculation of different contributions of energy of the system

First synthesis of the results

$$
\begin{aligned}
\delta V &= \int\limits_{\omega_{lg}} v_3^{lg}\, d\omega + \int\limits_{\omega_{sl}} v_3^{sl} d\omega \\
\delta A_{lg} &= -\int\limits_{\omega_{lg}} v_3^{lg}\, \mathrm{Tr}\,(C)\, \mathrm{d}\omega + \int\limits_{L_{slg}} v_t^{lg} \cdot \nu_{lg}\, dl \\
\delta A_{sl} &= -\int\limits_{\omega_{sl}} v_3^{sl} \mathrm{Tr}\left(C^{sl}\right) d\omega + \int\limits_{L_{slg}} v_t^{sl} \cdot \nu_{sl}\, dl
\end{aligned}
\tag{19}
$$

Calculation of different contributions of energy of the system

Minimization of energy

$$\int_{\omega_{lg}} \left(-\Delta P - \gamma_{lg} \mathrm{Tr}\left(C\right)\right) v_3^{lg} \mathrm{d}\omega + \int_{\omega_{sl}} \left(-\Delta P - \mathrm{Tr}\left(C^{sl}\right)\right) v_3^{sl} \mathrm{d}\omega$$

$$+ \int_{L_{slg}} \gamma_{lg}\, v_t^{lg} \cdot \nu_{lg} + v_t^{sl} \cdot \nu_{sl} dl = \int_{\omega_{sl}} f_3^{sl} v_3^{sl} \; d\omega \tag{20}$$

$$+ \int_{L_{slg}} f_3^{c} v_3^{sl} \; dl + \int_{\omega_{sl}} f_t^{sl} . v_t^{sl} \; d\omega$$

Necessity to decompose the displacement δp in the same basis

Calculation of different contributions of energy of the system

Decomposition of δp in the same basis

Continuity of the displacement on the contact line

$$v_t^{lg} + v_3^{lg} n_{lg} = v_t^{sl} + v_3^{sl} n_{sl} \qquad on \quad L_{slg} \tag{21}$$

General definition of the contact angle θ

$$\nu_{sl} \cdot \nu_{lg} = \cos\alpha = \sin\theta \tag{22}$$

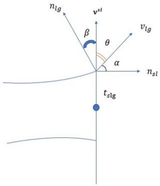

Figure: Definition of angle θ

Calculation of different contributions of energy of the system

One has

$$
\begin{aligned}
T &= \int\limits_{L_{slg}} \left(\gamma_{lg}\, v_t^{lg} \cdot \nu_{lg} + v_t^{sl} \cdot \nu_{sl} \right) dl \\
&= \int\limits_{L_{slg}} \left(\gamma_{lg}\, v_t^{sl} \cdot \nu_{lg} + v_t^{sl} \cdot \nu_{sl} \right) dl + \int\limits_{L_{slg}} \gamma_{lg}\ \sin\theta\ v_3^{sl} dl
\end{aligned}
\tag{23}
$$

and then after some algebra

$$
T = \int\limits_{L_{slg}} \left(\gamma_{lg} \cos\theta + \gamma_s \right)\ v_t^{sl} \cdot \nu_{sl}\ dl + \int\limits_{L_{slg}} \gamma_{lg}\ \sin\theta\, v_3^{sl} dl \tag{24}
$$

Calculation of different contributions of energy of the system

We finally obtain the following equilibrium equation **or variational principle** without line energy

$$
\begin{aligned}
&\int_{\omega_{lg}} \left(-\Delta P - \gamma_{lg}\,\mathrm{Tr}\,(C)\right)\, v_3^{lg}\,\mathrm{d}\omega + \int_{L_{slg}} (\cos\theta\ \gamma_{lg} + \gamma_s) v_t^{sl} \cdot \nu_{sl}\, dl \\
&\quad + \int_{\omega_{sl}} \left(-\Delta P - \mathrm{Tr}\left(C^{sl}\right)\right)\, v_3^{sl}\,\mathrm{d}\omega + \int_{L_{slg}} \sin\theta\gamma_{lg}\ v_3^{sl}\, dl = \int_{\omega_{sl}} f_3^{sl} v_3^{sl}\, d\omega \\
&\quad + \int f_3^{c} v_3^{sl}\ dl + \int_{\omega_{sl}} f_t^{sl} . v_t^{sl}\ d\omega
\end{aligned}
\tag{25}
$$

▷ 3 degrees of freedom: v_3^{lg}, $v_t^{sl} \cdot \nu_{sl}$ and v_3^{sl}

Calculation of different contributions of energy of the system

The degrees of freedom involved in the variational principle are :

- v_3^{lg} that plays the role of $\delta y(x)$ in the particular case considered of an axisymmetric capillary bridge between two plates or other geometries
- $v_t^{sl} \cdot \nu_{sl}$ that plays the role of δy_c
- v_3^{sl} that plays the role of δx_c

and must/can be considered as independent in the minimization problem.

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Young-Laplace equation

Eq. (25) must be satisfied for all virtual displacements v_3^{lg} defined on ω_{lg}, considering $v_t^{sl} = 0$ and $v_3^{sl} = 0$ defined on ω_{sl}

$$\int_{\omega_{lg}} \left(-\Delta P - \gamma_{lg} \mathrm{Tr}\left(C\right)\right) \, v_3^{lg} \mathrm{d}\omega = 0 \quad \forall v_3^{lg}$$

We obtain the classical Young-Laplace equation in its general intrinsic form:

$$\mathrm{Tr}\left(C\right) = -\frac{\Delta P}{\gamma_{lg}} \tag{26}$$

Associated equilibrium equations without line energy

"Young equation" and its interpretation

Coming back to Eq. (25), and considering any non vanishing v_t^{sl} defined on ω_{sl} (with $v_3^{sl} = 0$), we obtain :

$$\int_{L_{slg}} (\cos\theta\ \gamma_{lg} + \gamma_s) v_t^{sl} \cdot \nu_{sl}\ dl = 0 \quad \forall v_t^{sl}$$

$$-\gamma_s = \gamma_{lg} \cos\theta \tag{27}$$

which provides the expression of the local tangentiel friction force on the contact line (5):

$$f^s_{\text{ext}} = -\gamma_s \nu_{sl} = \gamma_{lg} \cos\theta\ \nu_{sl} \tag{28}$$

It is generally written on the form

$$\gamma_s + \gamma_{lg} \cos\theta\ = 0$$

and called Young equation.

General expression of associated capillary force

Work of the adhesion force during the displacement v_3^{sl}

$$
\begin{aligned}
W_{f_3 ad} &= \int_{\omega_{sl}} f_3^{sl} v_3^{sl} \; d\omega + \int_{L_{slg}} f_3^{c} v_3^{sl} \; dl \\
&= \int\limits_{\omega_{sl}} \left(-\Delta P - \gamma_s \mathrm{Tr}\left(C^{sl} \right) \right) \; v_3^{sl} \mathrm{d}\omega + \int\limits_{L_{slg}} \gamma_{lg} \sin\theta \; v_3^{sl} dl
\end{aligned}
\tag{29}
$$

By identification, we may define the "local" adhesion forces:

$$
\begin{aligned}
f_3^{sl} &= \gamma_{lg} \sin\theta \\
f_3^{c} &= -\Delta P - \mathrm{Tr}\left(C^{sl} \right)
\end{aligned}
\tag{30}
$$

- Classical for plane surface when $Tr(C^{sl}) = 0$.
- First term often called "capillary force" and second one Laplace pressure force
- This identification is "local" whereas the definition of the capillary force that can be measured is "global", resulting from an integration on ω_{sl} and L_{slg}

Associated equilibrium equations without line energy

General expression of associated capillary (adhesion) force

Consequently we write

$$F_3^{sl} = \int_{\omega_{sl}} \left(-\Delta P + \gamma_{lg} \cos(\theta) Tr(C^{sl}) \right) n^{sl} dS$$

$$F_3^{c} = \int_{L_{slg}} \gamma_{lg} \sin(\theta) n^{sl} dl$$

The capillary (adhesion) force is then given by

$$F_3^{adh} = \int_{\omega_{sl}} \left(-\Delta P + \gamma_{lg} \cos(\theta) Tr(C^{sl}) \right) n^{sl} dS + \int_{L_{slg}} \gamma_{lg} \sin(\theta) n^{sl} dl \quad (31)$$

▷ The capillary-adhesion force, is defined as the adhesion force exerted **by** the solid **on** the liquid

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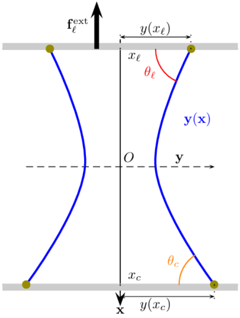
Explicit calculation of capillary force for simple geometries

Axisymmetric capillary bridge between two parallel planes



- $C^{sl} = 0$

- In that particular case, $n^{sl} = -\boldsymbol{e}_x$ for the upper plate and $\boldsymbol{e}_x$ for the lower plate.

Explicit calculation of capillary force for simple geometries

Lower plate case

One has

$$F_3^{adh} = \int_{\omega_{sl}} -\Delta P \boldsymbol{e}_x dS + \int_{L_{slg}} \gamma_{lg} \sin(\theta) \boldsymbol{e}_x dl \tag{32}$$

We obtain

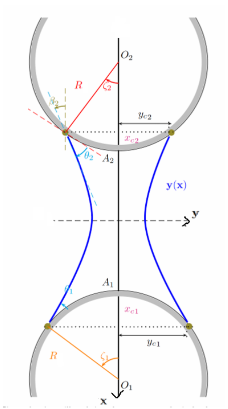
$$F_3^{adh} = \left(-\Delta P \ \pi y_c^2 + 2\pi y_c \gamma_{lg} \sin\theta\right) \boldsymbol{e}_x \tag{33}$$

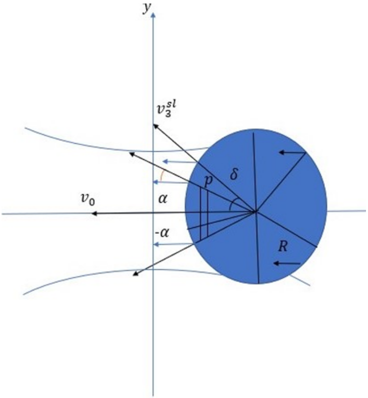
▷ **Classical expression of literature**

Note that $F_3^{adh} = \left(-\Delta P \ \pi y_c^2 + 2\pi y_c \gamma_{lg} \sin\theta\right) (-\boldsymbol{e}_x)$ for the upper plate.

Explicit calculation of capillary force for simple geometries

Axisymmetrical capillary bridge between two spheres





- For spheres $Tr\ C^{sl} = \frac{2}{R}$, with $R > 0$ the radius of the spheres.

Explicit calculation of capillary force for simple geometries

Lower sphere case

One has

$$F_3^{adh} = \left(-\Delta P + \gamma_{lg}\cos(\theta)\frac{2}{R}\right)\int_{\omega_{sl}} n^{sl}\,dS + \gamma_{lg}\sin(\theta)\int_{L_{slg}} n^{sl}\,dl \qquad (34)$$

Here $\displaystyle\int_{\omega_{sl}} n^{sl}\,dS = \pi R^2 \sin^2\delta\ \boldsymbol{e}_x$ and $\displaystyle\int_{L_{slg}} n^{sl}\,dS = 2\pi R\sin\delta\cos\delta\ \boldsymbol{e}_x$.

This leads to

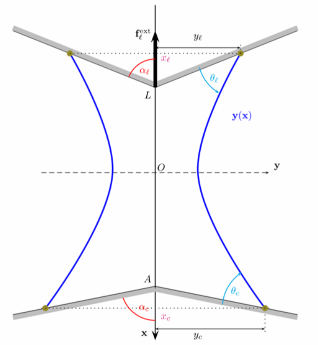
$$F_3^{adh} = \left(-\Delta\, P\pi R^2\sin^2\delta + 2\pi R\gamma_{lg}\sin\delta\sin(\theta+\delta)\right)\boldsymbol{e}_x \qquad (35)$$

▷ **Classical expression of literature**

Note that $F_3^{adh} = \left(-\Delta\, P\pi R^2\sin^2\delta + 2\pi R\gamma_{lg}\sin\delta\sin(\theta+\delta)\right)(-\boldsymbol{e}_x)$ for the upper sphere.

Explicit calculation of capillary force for simple geometries

Axisymmetric capillary bridge between two identical cones of opening angle α_c



- Imposed virtual displacement of the upper cone in x-direction.
- Here on the upper cone $n^{sl} = \cos\beta\ (\text{-}\boldsymbol{e}_x)$ where $\beta = \pi/2 - \alpha_c$ with $\alpha_c = \alpha_l$

Explicit calculation of capillary force for simple geometries

General expression of work of normal adhesion forces

$$F_3^{adh} = \int\limits_{\omega_{sl}} \left(-\Delta P - \gamma_s \mathrm{Tr}\,(C)^{sl} \right) \; n^{sl} \mathrm{d}\omega + \int\limits_{L_{slg}} \gamma_{lg} \sin\theta \; n^{sl}\, dl$$
$$= \int\limits_{\omega_{sl}} \left(-\Delta\, p - \gamma_s\, TrC^{sl} \right) \sin\alpha_c \; \mathrm{d}\omega \; (-\boldsymbol{e}_x) + \int\limits_{\gamma_{slg}} \gamma_{lg} \sin\theta \sin\alpha_c \;\; dl \; (-\boldsymbol{e}_x) \tag{36}$$

Using Young equation and the relation $Tr \;\; C^{sl} = \frac{\cos\alpha_c}{y(x)}$ for a cone, we have

$$F_3^{adh} = \left(\int\limits_{\omega_{sl}} \left(-\Delta\, p + \frac{\gamma_{lg} \cos\theta \cos\alpha_c}{y(x)} \right) \mathrm{d}\;\omega + \int\limits_{\gamma_{slg}} \gamma_{lg} \sin\theta \;\; dl \right) \sin\alpha_c \;\; (-\boldsymbol{e}_x) \tag{37}$$

Explicit calculation of capillary force for simple geometries

After some algebra, we obtain

$$F_3^{adh} = \Big(-\Delta\, p\ \pi y_c^2 + 2\pi\gamma_{lg} y_c \cos(\theta - \alpha_c)\Big) \pm \boldsymbol{e}_x \tag{38}$$

▷ General expression of the normal capillary force for a capillary bridge between two cones

$$F_{cap} = -\Delta\, p\ \pi y_c^2 + 2\pi\gamma_{lg} y_c \cos(\theta - \alpha_c) \tag{39}$$

- For $\alpha_c = \pi/2$ we recover the expression of F_{cap} for two parallel plates
- New expression obtained also by direct parametric (explicit) calculation $x \mapsto y(x)$ for axisymmetrical meridian

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Energy minimization with line energy

Back to initial energy minimization problem

$$\gamma_{lg}\delta A_{lg} \; + \gamma_s \delta A_{sl} \pm \gamma_{slg}\delta \mathcal{L}_{slg} - \Delta P \; \delta V - W_{F^s_{\mathrm{ext}/\Omega_l}} = 0 \qquad (40)$$

▷ How to compute $\delta \mathcal{L}_{slg}$ in the general case?

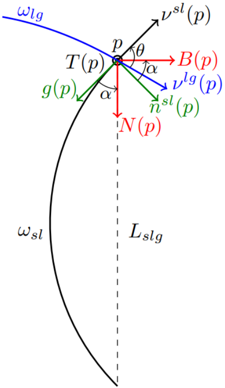
Very general result for any curve (plane or non planar)

$$\delta \mathcal{L}_{slg} = \Big[t_{slg} \cdot v_t^{sl} \Big]_{t_2}^{t_1} - \int_{L_{slg}} k \; \delta u \cdot N_{slg} \; dl \qquad (41)$$

- k is the curvature of L_{slg} defined in Frénet basis

- δu is the elementary displacement corresponding to the variation δ defined on L_{slg}

Darboux and frenet Basis on the triple line



- Frenet basis $(t_{slg}, N_{slg}, B_{sl})$ at any point p of L_{slg}
- Bardoux basis $(t_{slg}, g_{slg}, n_{sl})$ with $g_{slg} = -\nu_{sl}$
- Relation between Darboux en Frenet basis

$$N_{slg} = \cos(\alpha) g_{slg} - \sin(\alpha) n_{sl} \tag{42}$$

Energy minimization with line energy

General variational principle accounting with line energy

$$\int_{\omega_{lg}} (-\Delta P - \gamma_{lg}\mathrm{Tr}(C))\ v_3^{lg}\mathrm{d}\omega + \int_{L_{slg}} (\gamma_{lg}\ \cos\theta\ + \gamma_s \pm \gamma_{slg}\, k \cos(\alpha))\ v_t^{sl} \cdot \nu_{sl}\, dl$$

$$+ \int_{\omega_{sl}} \left(-\Delta P - \gamma_s \mathrm{Tr}\left(C^{sl}\right)\right)\ v_3^{sl}\mathrm{d}\omega + \int_{L_{slg}} (\gamma_{lg} \sin\theta \pm \gamma_{slg}\, k \sin(\alpha))\, v_3^{sl}\, dl$$

$$\pm \gamma_{slg}\ \left[t_{slg} \cdot v_t^{sl}\right]_{t_2}^{t_1} = \int_{\omega_{sl}} f_3^{sl} v_3^{sl}\ d\omega + \int_{L_{slg}} f_3^c v_3^{sl}\ dl + \int_{\omega_{sl}} f_t^{sl} . v_t^{sl}\ d\omega \tag{43}$$

Energy minimization with line energy

Associated equilibrium equations

- Young-Laplace equation unchanged

$$\mathrm{Tr}\,(C) = -\frac{\Delta P}{\gamma_{lg}} \tag{44}$$

- Generalized Young equation

$$\gamma_{lg} \cos\theta \ + \gamma_s \pm \gamma_{slg}\, k \cos\alpha = 0 \tag{45}$$

▷ **Supplementary term involving line energy**

Energy minimization with line energy

- Axisymmetrical capillary bridge with plane substrates ($k = \frac{1}{y_c}$)

$$\cos\theta \ + \frac{\gamma_s}{\gamma_{lg}} \pm \frac{\gamma_{slg}}{\gamma_{lg} y_c} = 0 \tag{46}$$

- Axisymmetrical capillary bridge between two spheres of same radius ($k = \frac{1}{y_c} = \frac{1}{Rsin\zeta}$ and $\alpha = -\zeta$)

$$\cos\theta \ + \frac{\gamma_s}{\gamma_{lg}} \pm \frac{\gamma_{slg}}{\gamma_{lg} y_c} \cos\zeta = 0 \tag{47}$$

- Axisymmetrical capillary bridge with two cones of same opening angles α_c ($k = \frac{1}{y_c}$ and $\alpha = pi/2 - \alpha_c$)

$$\cos\theta \ + \frac{\gamma_s}{\gamma_{lg}} \pm \frac{\gamma_{slg}}{\gamma_{lg} y_c} \sin\alpha_c = 0 \tag{48}$$

▷ Same expressions obtained by a direct calculation for axisymmetrical meridian

Energy minimization with line energy

Associated expression of capillary forces

• To be detailed

• For a simple geometries, line energy does not add a supplementary contribution! In the general case. . . ?

Thanks for your attention