Study of an interfacial instability in a 3-layer fluid system.

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1 - Liquid Metal Battery (LMB)

- **Definition**: system of 3 superimposed fluid layers (anode/electrolyte/cathode) + a strong electric current + a relatively strong temperature.
- **Interest**: energy storage for stationary applications ⇒ promote renewable energies.
- **Avantages**: long lifetime + low costs.
- **Difficulties**: hydrodynamic phenomena (metal pad roll instability, thermal convection, etc.)⇒ risk of short circuit + need for thermal efficiency improvement.
- **Example**: Li/CaBr2/Cd, $T \approx 320^{\circ} C$

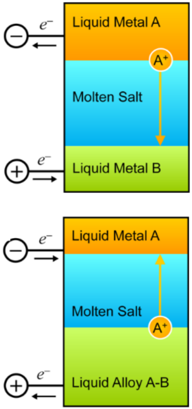


Figure: Diagram of an LMB in discharge (top) and in charge (bottom) from H Kim & al. Chem. Rev.

1 - Simplified representation of a LMB

- **Incompressible** fluids.
- **Non miscible** fluids.
- **Vertical** and **homogeneous** electric current in the base state ($*$)
- Electric conductivity of the electrolyte « electric conductivity of the electrodes
- Thickness of the electrolyte « thickness of the electrodes
- Width of the layers » wavelength of the disturbance

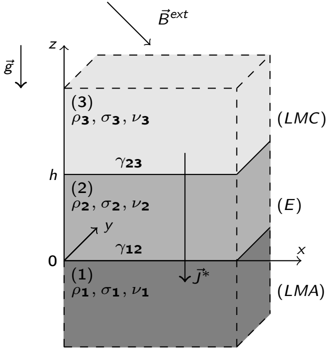


Figure: Sketch of the LMB model in the base state.

1 - Complexities

- Consider viscosity:

  ⇒ Numerical solution for the eigenvalue problem

- Add a third layer:

  ⇒ Coupling between the 2 interfaces ⇒ All the modes considered for the initial value problem

- Consider the Lorentz force:

  ⇒ Coupling between the Navier-Stokes (Fluid Mechanics) equations and the Maxwell equations (Electromagnetism)

Model 1

Model 2

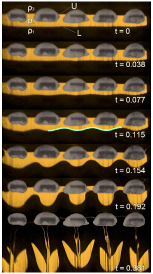
1 - Litterature for model 1

Theoretical studies

1. KO Mikaelian Phys. Rev. A 1983
   $\rightarrow$ inviscid model, N layers, analytical solutions.
2. S Parhi & G Nath Int. J. Engng Sci. 1991
   $\rightarrow$ viscous model, solving method, stability criterion.
3. KO Mikaelian Phys. Rev. E 1996
   $\rightarrow$ viscous model, 2 finite layers, solving method.

Experimental work

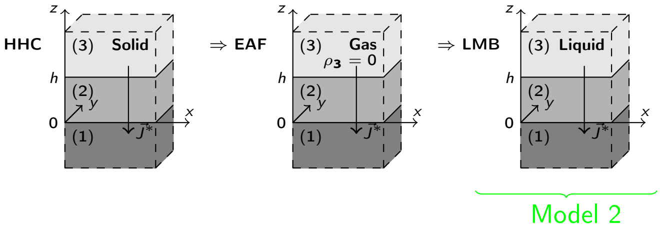
1. R Adkins & al.,J. Phys. Rev. Fluids 2017



Model 1: Mikaelian (3 layers) + consider viscosity

1 - Litterature for model 2

1. A D Sneyd J. Fluid Mech. 1985
   → Linear stability analysis of two inviscid systems close to LMB: Hall-Héroult Cell (HHC) and Electric Arc Furnace (EAF) + far magnetic field.



   HHC: the local magnetic field has only a stabilizing effect.
   EAF: the local magnetic field can destabilize the system.

2. J W Herreman & al. J. Fluid Mech. 2019 and 2023 to be published
   → Metal pad roll instability with a perturbative method in a cylindrical model of LMB.

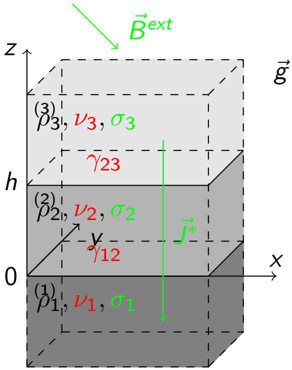


Figure: sketch of the 2 models.

Assumptions:

- **Isothermal** conditions.
- Extreme layers of **infinite** depth and **infinite** width.
- **Newtonian** fluids.
- **No slip** conditions.
- Slowly variable regime (quasistatic approximation).
- Coulomb force disregarded.
- Magnetic field not bounded at the interfaces.

2 - Volume equations

Volume equations in each phase $i \in \{1;2;3\}$:

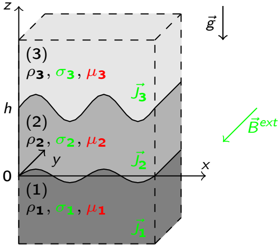
$$\vec{\nabla} \cdot \vec{u}_i = 0 \quad \text{(mass balance)}$$

$$\rho_i \left(\vec{u}_{i,t} + (\vec{u}_i \cdot \vec{\nabla}) \vec{u}_i \right) = -\vec{\nabla} p_i + \mu_i \vec{\Delta} \vec{u}_i + \vec{f}_{Li} + \rho_i \vec{g} \quad \text{(momentum balance)}$$

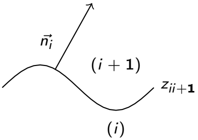
With $\vec{f}_{Li} = \vec{J}_i \times \vec{B}_i$, the Lorentz force in the layer (i)

Maxwell equations and Ohm's law:

$$\vec{\nabla} \times \vec{B}_i = \mu_0 \vec{J}_i \quad \text{(Maxwell-Ampère)}$$
$$\vec{\nabla} \times \vec{E}_i = \vec{0} \quad \text{(Maxwell-Faraday)}$$
$$\vec{\nabla} \cdot \vec{B}_i = 0 \quad \text{(Maxwell-Thomson)}$$
$$\vec{J}_i = \sigma_i \vec{E}_i \quad \text{(Ohm's law)}$$



Jump conditions at each interface $ii+1$ with $i \in \{1;2\}$:



$$\vec{u}_i \cdot \vec{n}_i = \vec{v}_{ii+1} \cdot \vec{n}_i = \vec{u}_{i+1} \cdot \vec{n}_i \quad \text{(mass balance)}$$
$$(p_{i+1} - p_i + \gamma_{ii+1}\kappa_{ii+1})\,\vec{n}_i = (\bar{\bar{\tau}}_{i+1} - \bar{\bar{\tau}}_i) \cdot \vec{n}_i \quad \text{(momentum balance)}$$
$$\vec{u}_i \times \vec{n}_i = \vec{u}_{i+1} \times \vec{n}_i \quad \text{(no slip)}$$
$$\vec{B}_i \cdot \vec{n}_i = \vec{B}_{i+1} \cdot \vec{n}_i \;,\quad \vec{B}_i \times \vec{n}_i = \vec{B}_{i+1} \times \vec{n}_i \quad \text{(magnetic field)}$$
$$\vec{J}_i \cdot \vec{n}_i = \vec{J}_{i+1} \cdot \vec{n}_i \;,\quad \vec{E}_i \times \vec{n}_i = \vec{E}_{i+1} \times \vec{n}_i \quad \text{(electric field)}$$

with $\vec{n}_i = \left(-z_{ii+1,x}, -z_{ii+1,y}, 1\right)^T$, $\kappa_{ii+1} = \frac{z_{ii+1,x,x}}{\left(1+z^2_{ii+1,x}\right)^{\frac{3}{2}}} + \frac{z_{ii+1,y,y}}{\left(1+z^2_{ii+1,y}\right)^{\frac{3}{2}}}$ and

$\bar{\bar{\tau}}_i = \mu_i(\vec{\nabla}\vec{u}_i + (\vec{\nabla}\vec{u}_i)^T)$

The unit vector normal to z_{ii+1}, the curvature of z_{ii+1} and the viscous stress-tensor in the layer (i)

2 - Basic state

Definition:

- Fluids at **rest**: $\vec{u}_i^* = \vec{0}$
- Interface 12 **plane**: $z_{12}^* = 0$
- Interface 23 **plane**: $z_{23}^* = h$

Volume equations:

$$-\vec{\nabla}(p_i^* + \rho_i g z) + J^* \vec{e}_z \times \vec{B}_i^* = \vec{0} \quad \text{(momentum balance)}$$
$$\vec{\nabla} \times \vec{B}_i^* = \mu_0^* \vec{e}_z \quad \text{(Maxwell-Ampère)}$$
$$\vec{\nabla} \cdot \vec{B}_i^* = 0 \quad \text{(Maxwell-Thomson)}$$

Jump conditions:

$$p_{i+1}^* - p_i^* = 0 \text{ at } z = (i-1)h$$
$$\vec{B}_{i+1}^* - \vec{B}_i^* = 0 \text{ at } z = (i-1)h$$

Admissible magnetic fields:

$$\vec{B}_i^* = \vec{B}_C^* + \alpha \cdot \vec{x} \ \ \textit{with}\ \alpha = \mu_0 J^* \begin{pmatrix} Q & R-\frac{1}{2} & 0 \\ R+\frac{1}{2} & -Q & 0 \\ 0 & 0 & 0 \end{pmatrix} = \alpha_{local} + \alpha_{far}$$

With $\vec{B}_C^*$, a constant vector and $\vec{x} = (x, y, z)^T$.
Q and R refer to the far magnetic field.

Solution for the pressure:

$$\begin{aligned} p_1^*(x,y,z) &= -\rho_1 gz + p_c^* + p_M^*(x,y) \\ p_2^*(x,y,z) &= -\rho_2 gz + p_c^* + p_M^*(x,y) \\ p_3^*(x,y,z) &= -\rho_3 gz + gh(\rho_3-\rho_2) + p_c^* + p_M^*(x,y) \end{aligned}$$

with $p_c^* = p_1^*(z=0)$ a constant and $p_M^* = \mu_0 J^* \left(y\vec{B}_c^* \cdot \vec{e}_x - x\vec{B}_c^* \cdot \vec{e}_y \right) + \mu_0 J^{*2} \left(Qxy - \frac{1}{4}\left(x^2+y^2\right) + \frac{1}{2}R\left(y^2-x^2\right)\right)$, the magnetic pressure in the base state.

2 - Perturbed state

Definition:

$$\vec{u}_i(x,y,z,t) = \vec{u}^*_i + \vec{u}'_i(x,y,z,t) = \vec{u}'_i \text{ with } u'_i \ll 1$$
$$p_i(x,y,z,t) = p^*_i(z) + p'_i(x,y,z,t) \text{ with } p'_i \ll p^*_i$$
$$z_{ii+1}(x,y,t) = z^*_{ii+1} + z'_{ii+1}(x,y,t) \text{ with } z'_{ii+1} \ll z^*_{ii+1}$$
$$\vec{J}_i(x,y,z,t) = \vec{J}^* + \vec{J}'_i(x,y,z,t) \text{ with } J'_i << J^*$$
$$\vec{B}_i(x,y,z,t) = \vec{B}^*_i(x,y,z) + \vec{B}'_i(x,y,z,t) \text{ with } B'_i << B^*_i$$
$$\vec{E}_i(x,y,z,t) = \vec{E}^*_i(z) + \vec{E}'_i(x,y,z,t) \text{ with } E'_i << E^*_i$$

Volume equations in each phase $i \in \{1;2;3\}$: Mass balance: $u_{ij,j} = 0$

Linearized momentum balance: $\rho_i u_{ij,t} = -p'_{i,j} + \mu_i \Delta u_{ij} + f'_{Lij}$
$j \in \{x;y;z\}$ with $f'_{Lij} = \epsilon_{jlm} J^*_{il} B'_{im} + \epsilon_{jlm} J'_{il} B^*_{im}$, the disturbed Lorentz force

Linearized Maxwell equations and Ohm's law:

$$\epsilon_{jlm} B'_{im,l} = \mu_0 \vec{J}'_{ij} \quad , \quad \epsilon_{jlm} E'_{im,l} = \vec{0}$$
$$B'_{ij,j} = 0 \quad , \quad J'_{ij} = \sigma_i E'_{ij}$$

Linearized jump conditions at $z = (i-1)h$, $i \in \{1;2\}$:

$$u_{iz} = z'_{ii+1,t} = u_{i+1z}$$
$$u_{ix} = u_{i+1x}$$
$$u_{iy} = u_{i+1y}$$
$$p'_{i+1} - p'_i - (\rho_{i+1} - \rho_i) g z'_{ii+1} - \gamma_{ii+1}(z'_{ii+1,x,x} + z'_{ii+1,y,y}) =$$
$$2(\mu_{i+1} u_{i+1z,z} - \mu_i u_{iz,z})$$
$$\mu_{i+1}(u_{i+1z,x} + u_{i+1x,z}) = \mu_i(u_{iz,x} + u_{ix,z})$$
$$\mu_{i+1}(u_{i+1z,y} + u_{i+1y,z}) = \mu_i(u_{iz,y} + u_{iy,z})$$
$$\vec{B}'_i \cdot \vec{n}_i = \vec{B}'_{i+1} \cdot \vec{n}_i$$
$$\vec{B}'_i \times \vec{n}_i = \vec{B}'_{i+1} \times \vec{n}_i$$
$$\vec{J}'_i \cdot \vec{n}_i = \vec{J}'_{i+1} \cdot \vec{n}_i$$
$$\vec{E}'_i \times \vec{n}_i = \vec{E}'_{i+1} \times \vec{n}_i$$

Solutions searched under the form: $e^{(ik_x x + ik_y y + \omega t)}$ with $k^2 = k_x^2 + k_y^2$ the wavenumber of the perturbation and ω the time coefficient (complex number).

Vaschy-Buckingham theorem $\Rightarrow$ 9 dimensionless numbers for model 1 and 5 dimensionless numbers for model 2

$\rho_{r12} = \frac{\rho_2}{\rho_1}$, $\rho_{r13} = \frac{\rho_3}{\rho_1}$ (densities)

$Re_i = \sqrt{\frac{g}{k^3}} \frac{1}{\nu_i}$ (viscosities)

$Bo_{ii+1} = \frac{(\rho_{i+1} - \rho_i) g}{\gamma_{ii+1} k^2} = \frac{k_c^2}{k^2}$ (surface tensions)

With k_c the cut-off wave number

$K = kh$ (thickness)

$\Omega = \omega \sqrt{gk}$ (pulsation)

$J = J_{local} + J_{far}$ (magnetic field)

With $J_{local} = -\mu_0 J^{*2} / \rho_1 g$ and $J_{far} = J_{far}(Q, R)$

Dispersion relation: $f(\rho_{r12}, \rho_{r23}, Re_i, Bo_{ii+1}, J, K, \Omega) = 0$

2 - Dispersion relation

- Volume equations $\rightarrow u_{iz}$:

$$u_{1z} = (C_1 e^{kz} + D_1 e^{q_1 z}) e^{(ik_x x + ik_y y + \omega t)}$$
$$u_{2z} = (A_2 e^{-kz} + B_2 e^{-q_2 z} + C_2 e^{kz} + D_2 e^{q_2 z}) e^{(ik_x x + ik_y y + \omega t)}$$
$$u_{3z} = (A_3 e^{-kz} + B_3 e^{-q_3 z}) e^{(ik_x x + ik_y y + \omega t)}$$

  with C_1, D_1, A_2, B_2, C_2, D_2, A_3 and B_3 **8 unknown** coefficients and $q_i = \sqrt{k^2 + \omega\rho_i/\mu_i}$ the **modified** wavenumber in phase i.

- 4 equations at each interface $\rightarrow$ **8** equations $\rightarrow$ **closed** problem

Dispersion relation: solution of $M \cdot (C_1, D_1, A_2, B_2, C_2, D_2, A_3, B_3)^T = 0$

Non-trivial solutions are solutions of

$$|M| = 0$$

For model 1:

There is one **analytical** solution, stable $\forall k$:

$$\Omega_1 = \frac{\omega_1}{\sqrt{gk}} = \sqrt{\frac{g}{k^3}}\frac{\rho_i}{\mu_i} = \boxed{-Re_2^{-1}}$$

And 3 or 4 other solutions, depending on k, determined **numerically**.

For model 2:

$$\boxed{\Omega^4 f_4(K, \rho_{r12}, \rho_{r13}) + \Omega^2 f_2(K, \rho_{r12}, \rho_{r13}, J) + f_0(K, \rho_{r12}, \rho_{r13}, J) = 0}$$

With:

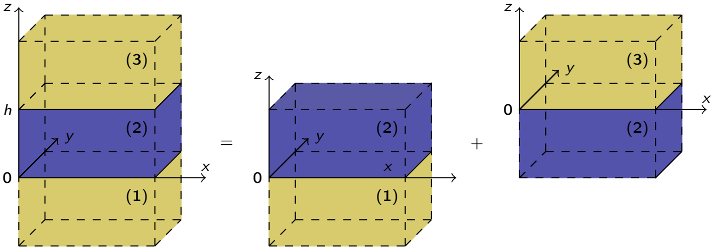
$$f_4 = \rho_{r12}(\rho_{r12}\sinh(K) + \cosh(K)) + \rho_{r13}(\rho_{r12}\cosh(K) + \sinh(K))$$
$$f_2 = \rho_{r12}(1-\rho_{r13})(\sinh(K)+\cosh(K)) - (1+\rho_{r13})J\sinh(K)^{-1}$$
$$f_0 = (\rho_{r12}+\rho_{r13})(1-\rho_{r12})\sinh(K) - (1+\rho_{r13})J\sinh(K)^{-1} - J^2\sinh(K)^{-1}$$

③ - 2 limit cases for comparison for model 1

1. 2 separate **2-layer** fluid systems (considering the fluid viscosities)

   (3)/(2)/(1) is compared to (2)/(1) and (3)/(2) with (3)≡(1)

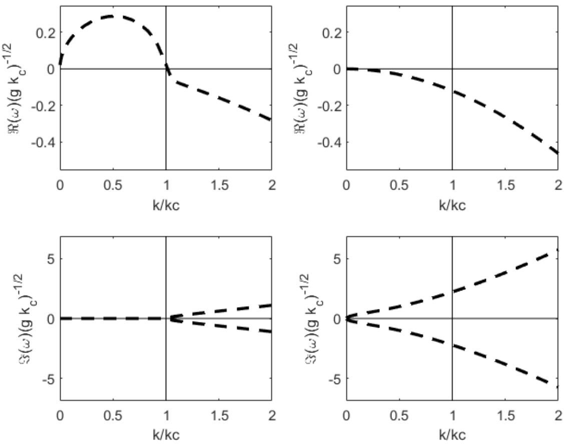
   - (2)/(1) is gravitationally **unstable**.
   - (3)/(2) is gravitationally **stable**.



2. 3-layer **inviscid** model of Mikaelian

③ - Solutions for 2 separate 2-layer fluid systems

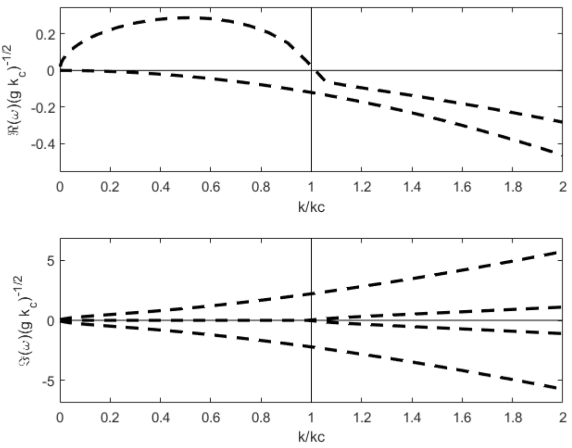
$\vec{g}$-unstable vs $\vec{g}$-stable



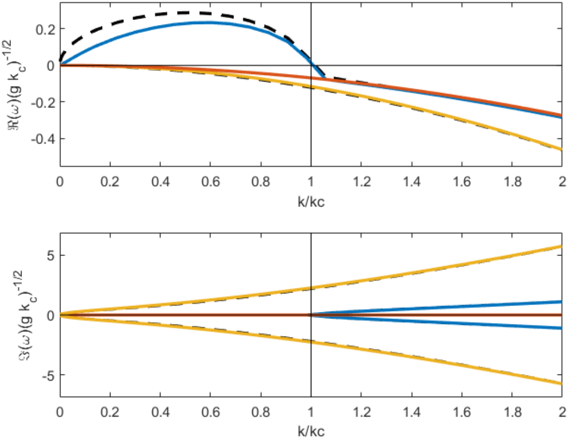
k_c: cutoff wavenumber due to surface tension.
$\mathfrak{Re}(w) > 0 \rightarrow$ unstable solution; $\mathfrak{Im}(w) \neq 0 \rightarrow$ oscillations.

③ - All solutions on the same graph!

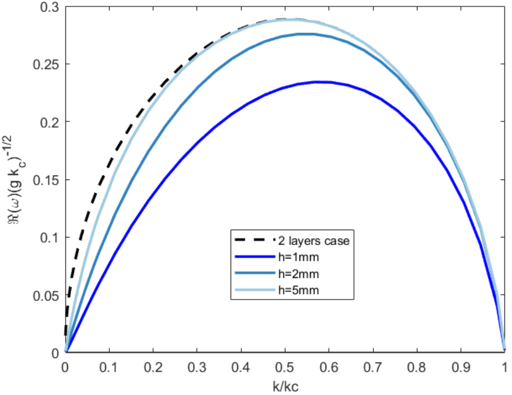
$k < k_c$: 1 unstable + 2 stables
$k > k_c$: 4 stables



3 totally **stable** solutions: 1 analytically, 2 numerically determined
Other solutions: 2 **stable** for $k > k_c$, 1 **unstable** for $k < k_c$.

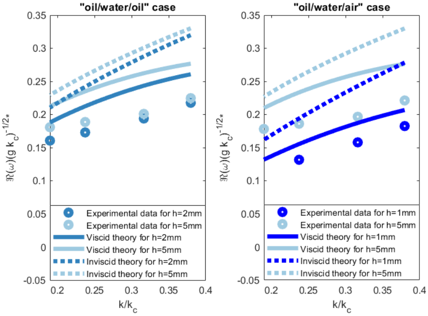


Effect of h:



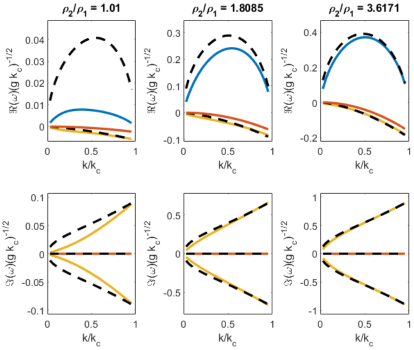
- Increasing h (i.e. decreasing interface coupling) increases ω.

Effect of *h* on Adkins data compared with model 1 and Mikaelian's theory:



- The viscous model improves the prediction as *h* is reduced.

Effect of the dimensionless density $\rho_{12}^{-1} = \frac{\rho_2}{\rho_1}$:

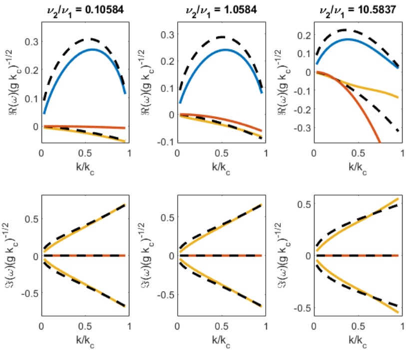


for $h = 1mm$

- The density ratio has a strong effect on the unstable eigenvalues

3 - Effect of viscosities on the eigenvalues

Effect of the ratio $\frac{Re_1}{Re_2} = \frac{\nu_2}{\nu_1}$:



for $h = 1mm$

- The viscosity ratio has a strong effect on the most stable eigenvalue

$\rho_{r12} = 0.27$ and $\rho_{r13} = \{0; \mathbf{0.25}\}$ **(values of Herreman & al.)**:

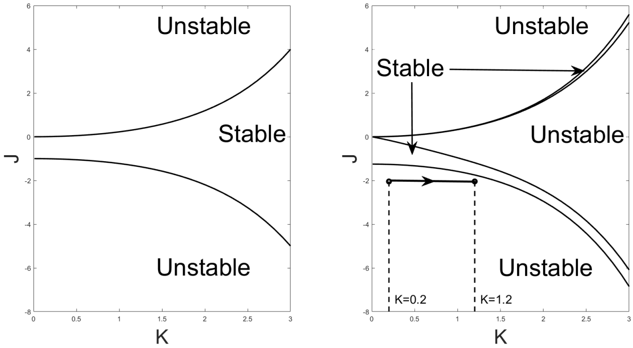


Figure: Stability diagram for an EAF model where $\rho_{r13} = 0$ (left) and the present LMB model (right)

- An unstable region appears in the middle of the stable one $\Rightarrow$ making difficult the conception of such a battery.

4 - Conclusion

- Development of 2 models for a 3-layer fluid system:

1. One that considers the viscosity of the fluids
    - The instability is less important when the interfaces are coupled
    - The lower ρ_1 is with respect to ρ_2 the less the instability will be important

2. One that considers the Lorentz force acting on the fluids
    - Find a liquid metal as light as possible for the top layer

4 - Outlooks for model 1

- Solve the initial value problem $\Rightarrow$ perform numerical simulations.
- Archer (In-house code) has been chosen. 2 steps are missing:
    1. 3 phases (J. C. Brändle de Motta)
    2. Magneto-static (R. Canu)

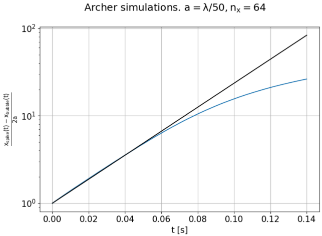
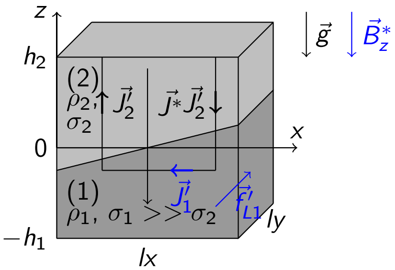


Figure: Comparison between theory and simulations for a 2-layer fluid system.

4 - Outlooks for model 2

- Study the standing wave ⇒ determine a more general Sele criterion.



Sele's criterion:

$$\beta = \frac{J^* B_z^* l_x l_y}{g(\rho_1 - \rho_2) h_1 h_2} > \beta_{cr} = 12$$

- Cross-validate the theoretical results with numerical simulations.
- Develop a viscid model for LMB.
- Develop a weakly non linear theory to extend the time of validity of the model.

Thank you for your attention.

Any questions?