

# 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  $\Rightarrow$  promote renewable energies.
- ▶ **Avantages:** long lifetime + low costs.
- ▶ **Difficulties:** hydrodynamic phenomena (metal pad roll instability, thermal convection, etc.)  $\Rightarrow$  risk of short circuit + need for thermal efficiency improvement.
- ▶ **Example:** Li/CaBr<sub>2</sub>/Cd,  $T \approx 320^\circ\text{C}$

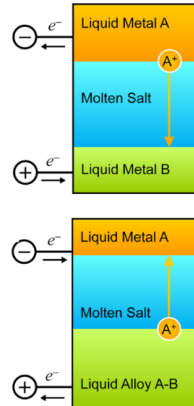


Figure: Diagram of an LMB in discharge (top) and in charge (bottom) from H Kim & al. Chem.

# 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

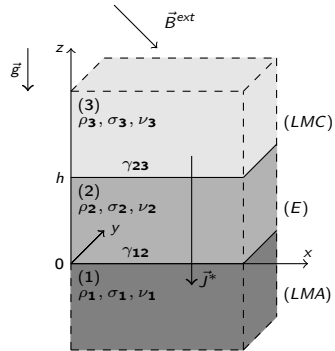


Figure: Sketch of the LMB model in the base state.

# 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

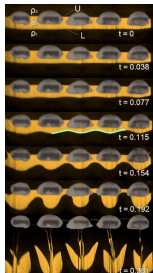
# 1 - Litterature for model 1

## Theoretical studies

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

## Experimental work

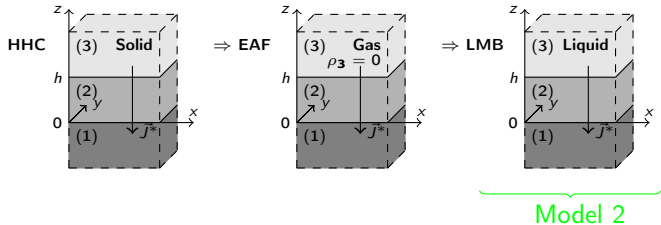
1. R Adkins & al., J. Phys. Rev. Fluids 2017



**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-Hérout 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.

## 2 - Formulation

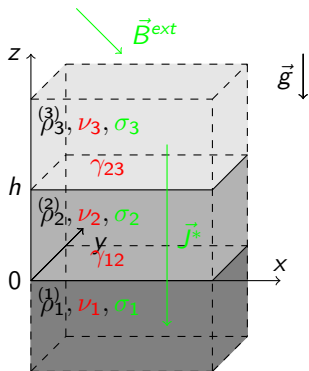


Figure: sketch of the 2 models.

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  $i \in \{1; 2; 3\}$ :

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

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

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

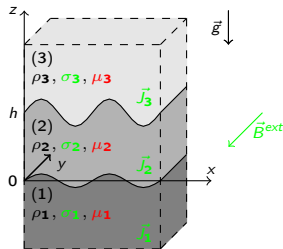
Maxwell equations and Ohm's law:

$$\vec{\nabla} \times \vec{B}_i = \mu_0 \vec{J}_i \quad (\text{Maxwell-Ampère})$$

$$\vec{\nabla} \times \vec{E}_i = \vec{0} \quad (\text{Maxwell-Faraday})$$

$$\vec{\nabla} \cdot \vec{B}_i = 0 \quad (\text{Maxwell-Thomson})$$

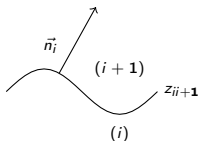
$$\vec{J}_i = \sigma_i \vec{E}_i \quad (\text{Ohm's law})$$





## 2 - Jump conditions

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



$$\vec{u}_i \cdot \vec{n}_i = \vec{v}_{ii+1} \cdot \vec{n}_i = \vec{u}_{i+1} \cdot \vec{n}_i \quad (\text{mass balance})$$

$$(\rho_{i+1} - \rho_i + \gamma_{ii+1} \kappa_{ii+1}) \vec{n}_i = (\bar{\tau}_{i+1} - \bar{\tau}_i) \cdot \vec{n}_i \quad (\text{momentum balance})$$

$$\vec{u}_i \times \vec{n}_i = \vec{u}_{i+1} \times \vec{n}_i \quad (\text{no slip})$$

$$\vec{B}_i \cdot \vec{n}_i = \vec{B}_{i+1} \cdot \vec{n}_i, \quad \vec{B}_i \times \vec{n}_i = \vec{B}_{i+1} \times \vec{n}_i \quad (\text{magnetic field})$$

$$\vec{J}_i \cdot \vec{n}_i = \vec{J}_{i+1} \cdot \vec{n}_i, \quad \vec{E}_i \times \vec{n}_i = \vec{E}_{i+1} \times \vec{n}_i \quad (\text{electric field})$$

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

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

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:

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

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

## 2 - Basic state

Admissible magnetic fields:

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

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:

$$p_1^*(x, y, z) = -\rho_1 g z + p_c^* + p_M^*(x, y)$$

$$p_2^*(x, y, z) = -\rho_2 g z + p_c^* + p_M^*(x, y)$$

$$p_3^*(x, y, z) = -\rho_3 g z + g h (\rho_3 - \rho_2) + p_c^* + p_M^*(x, y)$$

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

## 2 - Perturbed state

Definition:

$$\vec{u}_i(x, y, z, t) = \vec{u}_i^* + \vec{u}'_i(x, y, z, t) = \vec{u}'_i \text{ with } u'_i \ll 1$$

$$p_i(x, y, z, t) = p_i^*(z) + p'_i(x, y, z, t) \text{ with } p'_i \ll p_i^*$$

$$z_{ii+1}(x, y, t) = z_{ii+1}^* + z'_{ii+1}(x, y, t) \text{ with } z'_{ii+1} \ll z_{ii+1}^*$$

$$\vec{J}_i(x, y, z, t) = \vec{J}_i^* + \vec{J}'_i(x, y, z, t) \text{ with } J'_i \ll J_i^*$$

$$\vec{B}_i(x, y, z, t) = \vec{B}_i^*(x, y, z) + \vec{B}'_i(x, y, z, t) \text{ with } B'_i \ll B_i^*$$

$$\vec{E}_i(x, y, z, t) = \vec{E}_i^*(z) + \vec{E}'_i(x, y, z, t) \text{ with } E'_i \ll E_i^*$$

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:

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

## 2 - Perturbed state

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

$$u_{iz} = z'_{ii+1,t} = u_{i+1z}$$

$$u_{ix} = u_{i+1x}$$

$$u_{iy} = u_{i+1y}$$

$$p'_{i+1} - p'_i - (\rho_{i+1} - \rho_i)gz'_{ii+1} - \gamma_{ii+1}(z'_{ii+1,x,x} + z'_{ii+1,y,y}) = 2(\mu_{i+1}u_{i+1z,z} - \mu_i u_{iz,z})$$

$$\mu_{i+1}(u_{i+1z,x} + u_{i+1x,z}) = \mu_i(u_{iz,x} + u_{ix,z})$$

$$\mu_{i+1}(u_{i+1z,y} + u_{i+1y,z}) = \mu_i(u_{iz,y} + u_{iy,z})$$

$$\vec{B}'_i \cdot \vec{n}_i = \vec{B}'_{i+1} \cdot \vec{n}_i$$

$$\vec{B}'_i \times \vec{n}_i = \vec{B}'_{i+1} \times \vec{n}_i$$

$$\vec{J}'_i \cdot \vec{n}_i = \vec{J}'_{i+1} \cdot \vec{n}_i$$

$$\vec{E}'_i \times \vec{n}_i = \vec{E}'_{i+1} \times \vec{n}_i$$

## 2 - Dimensionless numbers

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  $\omega$  the time coefficient (complex number).

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

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

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

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

With  $k_c$  the cut-off wave number

$$K = kh \quad (\text{thickness})$$

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

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

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

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

$$|M| = 0$$

## 2 - Solutions

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:

$$f_4 = \rho_{r12}(\rho_{r12} \sinh(K) + \cosh(K)) + \rho_{r13}(\rho_{r12} \cosh(K) + \sinh(K))$$

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

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

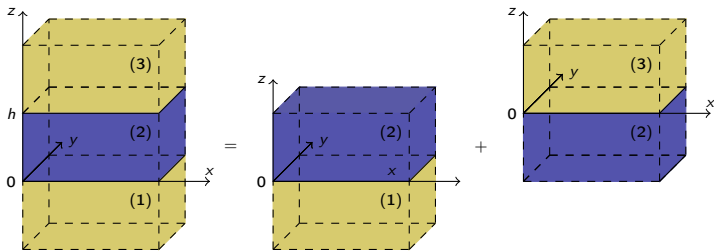


### 3 - 2 limit cases for comparison for model 1

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

(3)/(2)/(1) is compared to (2)/(1) and (3)/(2) with  $(3) \equiv (1)$

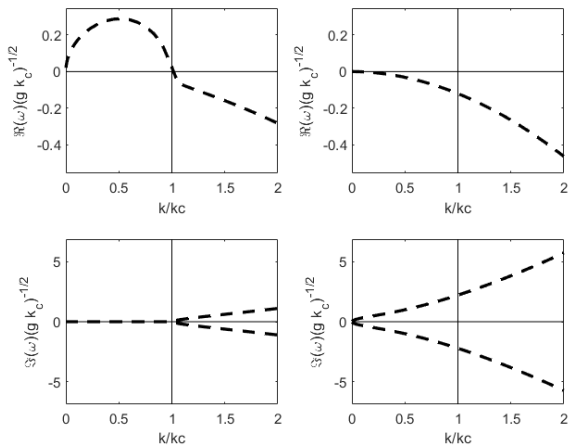
- ▶ (2)/(1) is gravitationally **unstable**.
- ▶ (3)/(2) is gravitationally **stable**.



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

### 3 - Solutions for 2 separate 2-layer fluid systems

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



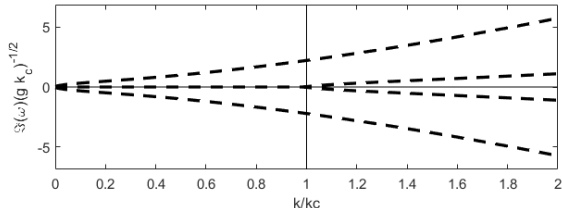
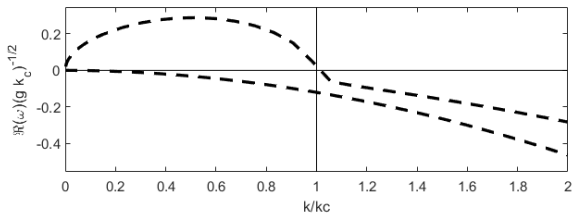
$k_c$ : cutoff wavenumber due to surface tension.

$\Re(w) > 0 \rightarrow$  unstable solution;  $\Im(w) \neq 0 \rightarrow$  oscillations.

### 3 - All solutions on the same graph!

$k < k_c$ : 1 unstable + 2 stables

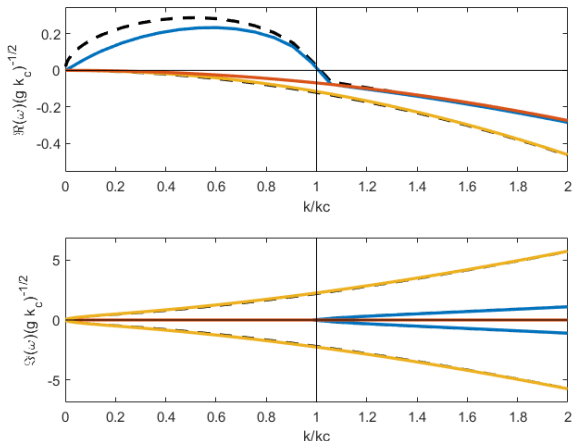
$k > k_c$ : 4 stables



### 3 - Solutions for the 3-layer fluid system, $h=1$ mm

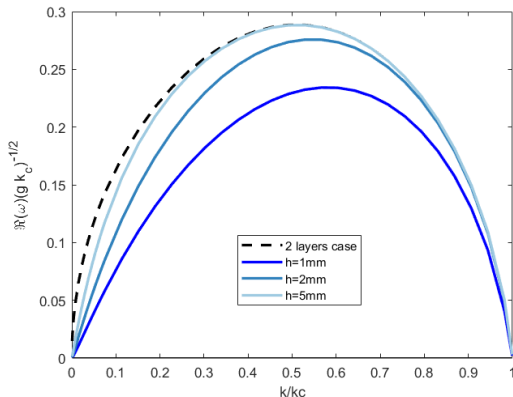
3 totally **stable** solutions: 1 **analytically**, 2 **numerically** determined

**Other solutions**: 2 **stable** for  $k > k_c$ , 1 **unstable** for  $k < k_c$ .



### 3 - Effect of $h$ on the unstable solution

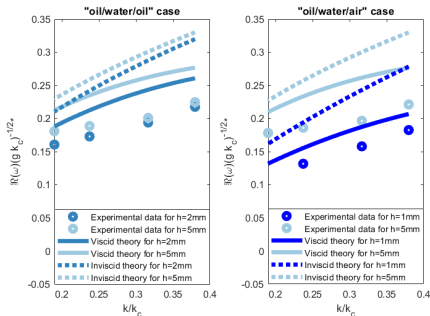
Effect of  $h$ :



► Increasing  $h$  (i.e. decreasing interface coupling) increases  $\omega$ .

### 3 - Comparison to experimental values

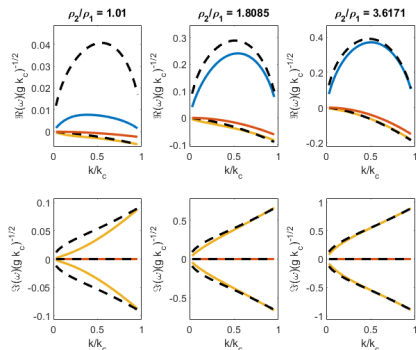
Effect of  $h$  on Adkins data compared with **model 1** and Mikaelian's theory:



► The viscous model improves the prediction as  $h$  is reduced.

### 3 - Effect of densities on the eigenvalues

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

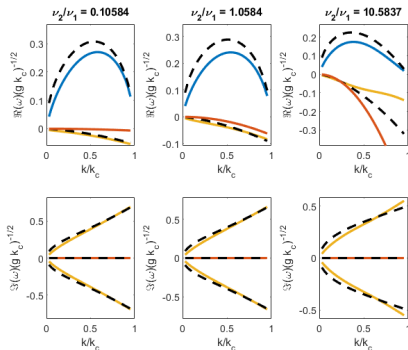


for  $h = 1\text{mm}$

- ▶ 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



### 3 - Conditions of stability for the model 2

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

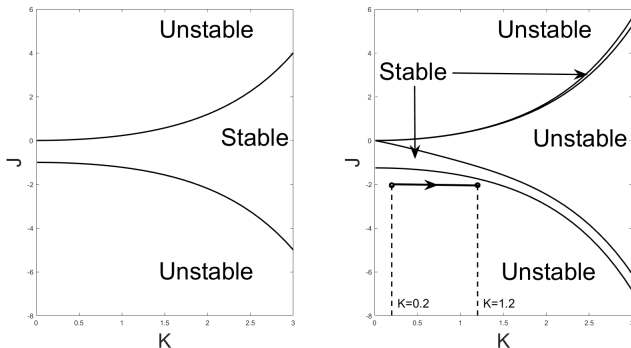


Figure: Stability diagram for an EAF model where  $\rho_{r13} = 0$  (left) and the present LMB model (right)

- ▶ 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  $\rho_1$  is with respect to  $\rho_2$  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  
⇒ perform numerical simulations.
- ▶ Archer (In-house code) has been chosen. 3 steps missing:
  1. 3 phases on Archer (J. C. Brändle de Motta)
  2. Magneto-static (R. Canu)

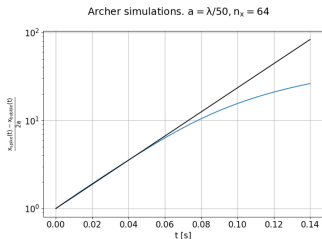
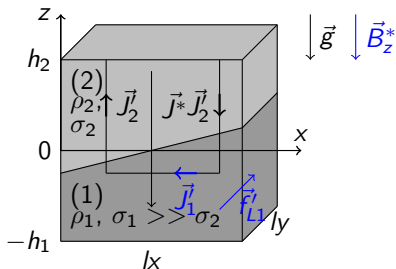


Figure: Comparison between theory and simulations for a 2-layer fluid system.

## 4 - Outlooks for model 2

- ▶ Study the standing wave  $\Rightarrow$  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?*