Multiphysics Modeling of a Fluorine Production Cell

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Introduction

To produce uranium hexafluoride (UF_6), gaseous fluorine (F_2) is essential. It is obtained on-site via the electrolysis of an HF-composed molten salt.

Simulation with COMSOL Multiphysics® has been used for years to understand better the physical phenomena occurring inside a fluorine production cell. To improve the model, experiments have been realized on a R&D cell at a semi-industrial scale.

Basics of the process

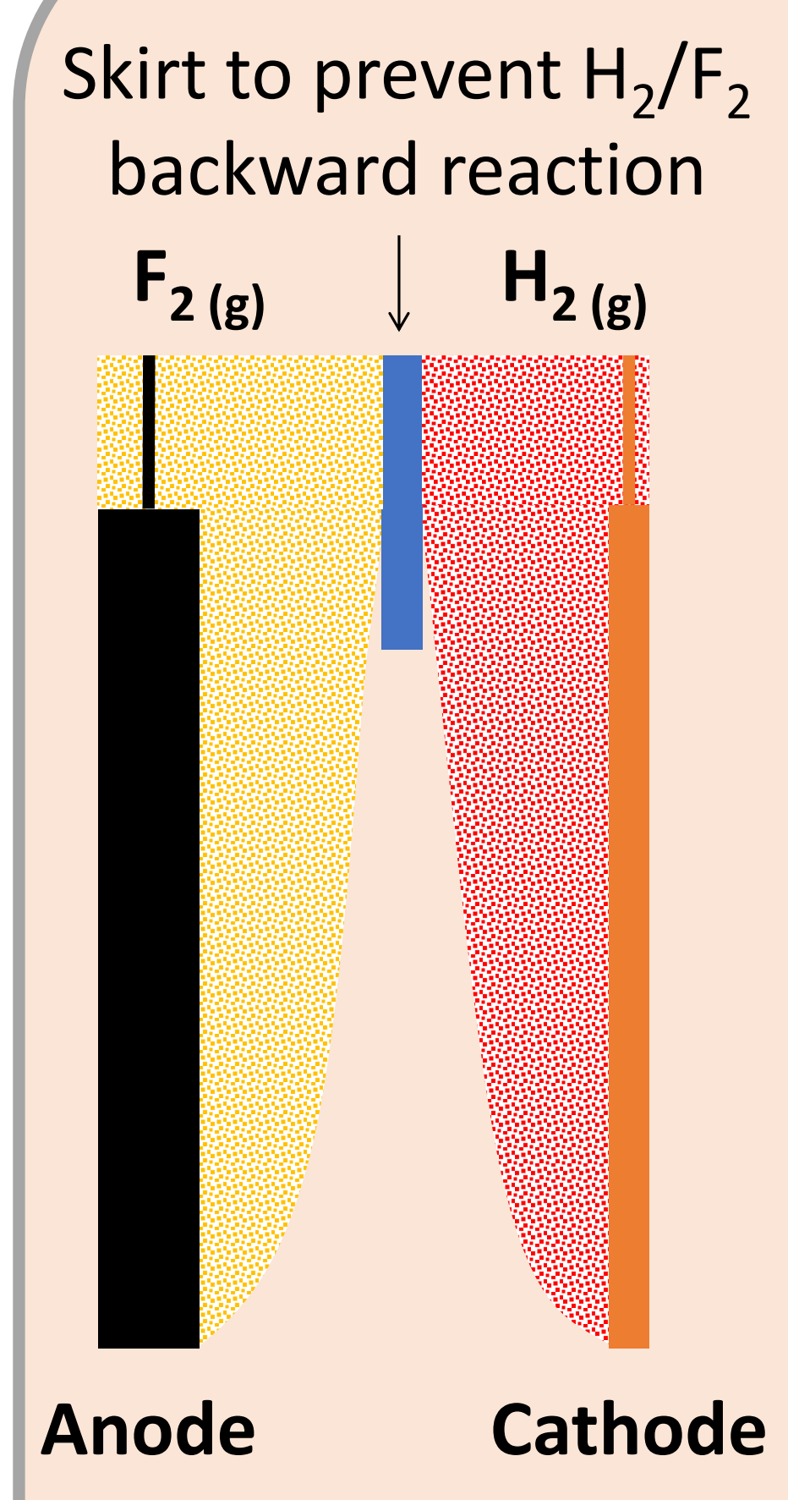

Figure 1. Schematic view of the R&D cell.

Electrolyte: KF-xHF

Electrochemical reactions

At the anode: $F(HF)_n^- \rightarrow \frac{1}{2}\boldsymbol{F_{2\,(g)}} + nHF_{(l)} + e^-$

At the cathode: $(n+1)HF_{(l)} + e^- \rightarrow \frac{1}{2}\boldsymbol{H_{2\,(g)}} + F(HF)_n^-$

Physics involved

- Electrochemistry
- Two-phase flow
- Heat transfer with phase change
- Species transport

} In the model

Goals of the present work:

- Build a 3D fully coupled model of the cell,
- Assess its precision against experimental data.

Computational method

Equations

$$\nabla(\sigma_{\text{electrode}}V_{\text{electrode}}) = 0$$
$$\nabla(\sigma_{\text{electrolyte}}V_{electrolyte}) = 0$$

Current conservation: **EC**

$$\alpha_c\rho_c\frac{\partial \mathbf{u_c}}{\partial t} + \alpha_c\rho_c\mathbf{u_c}\cdot\nabla(\mathbf{u_c}) = -\nabla(p) + \nabla\cdot(\alpha_c\boldsymbol{\tau}_\mathbf{c}) + \alpha_c\rho_c\mathbf{g}$$
$$\frac{\partial(\alpha_c\rho_c + \alpha_d\rho_d)}{\partial t} + \nabla\cdot(\alpha_c\rho_c\mathbf{u_c} + \alpha_d\rho_d\boldsymbol{u}_\mathbf{d}) = 0$$
$$\frac{\partial\alpha_d\rho_d}{\partial t} + \nabla\cdot(\alpha_d\rho_d\boldsymbol{u}_\mathbf{d}) = 0$$

Bubbly flow: **BF**

$$\rho_c C_{p,c}\frac{\partial T}{dt} + \rho_c C_{p,c}\mathbf{u_c}\nabla T - \nabla\cdot(k_c\nabla T) = Q$$

Heat transfer with phase change: **HT**

Couplings & boundary conditions

Physics are intertwined in many ways in this system, via physical properties or source terms:

	EC	BF	HT
EC	**	J_a and J_c	Ohmic drop
BF	Impact of void fraction on σ	**	Two-phase macroconvection
HT	Impact of temperature on σ	Impact of temperature on μ and ρ	**

Table 1. Links between physics. For example: EC impacts BF via J_a and J_c, the anodic and cathodic current densities at the electrodes' surfaces.

Study

Transient studies are performed until a pseudo-steady state is reached for three output parameters: cell voltage, gaseous outflow and power evacuated by the cooling system.

Results

Electric current and bubbly flow

	Cell voltage (V)					
I (A)	*Exp.*	*Sim.*	*Exp.*	*Sim.*	*Exp.*	*Sim.*
22.5	6.9	6.5	6.4	6.3	6.3	6.2
34	7.8	7.5	7.3	7.2	7.1	7.0
45.2	8.7	8.4	8.0	8.1	7.8	7.8
HF (%)	**39.2**		**40.8**		**42.2**	

Table 2. Simulated and measured cell voltage for various intensities and HF contents.

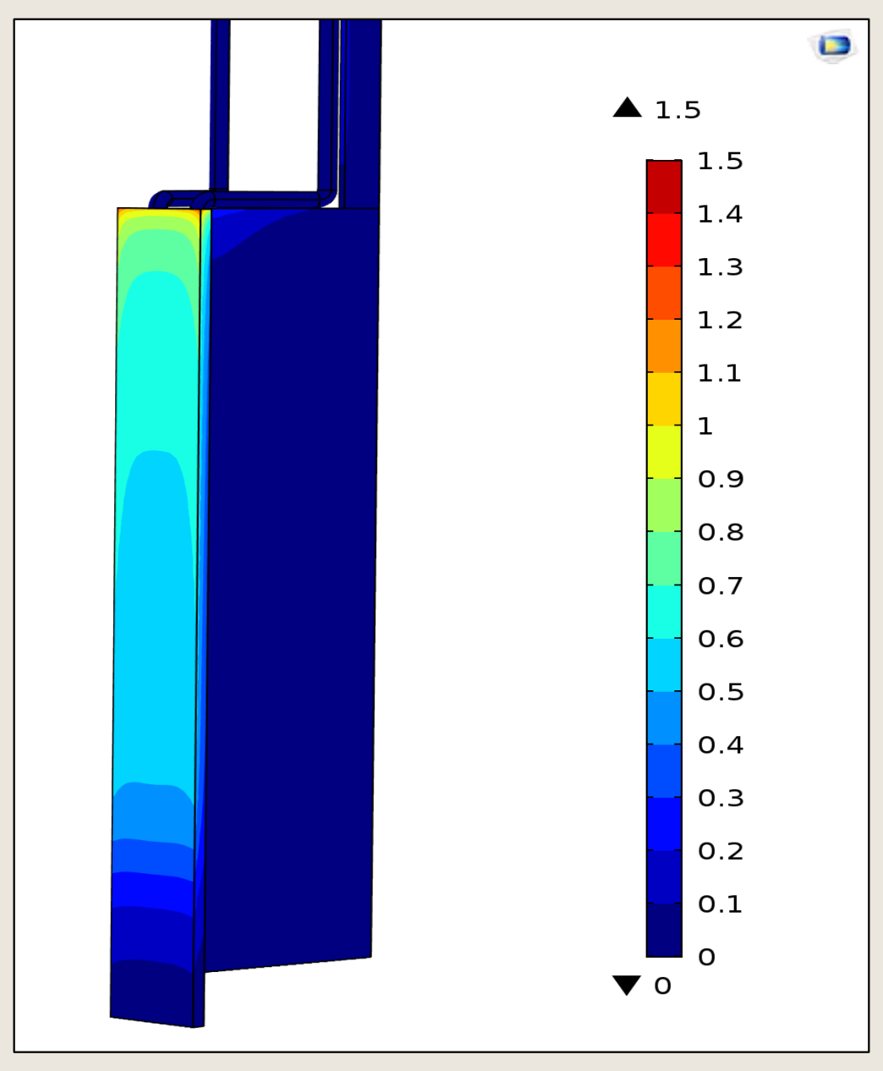

Figure 2. Normalized current density at the cathode.

- Cell voltage values and trends are well modelled, with less than 5% error.
- Possibility to access local current densities at the electrodes' surfaces.

F_2 going to the right collector vs total F_2 produced → F_2 yield:

- exp. 91.0 % → mean value, small variations measured.
- sim. 92.4 % → no variation at all in the model.

Heat transfer

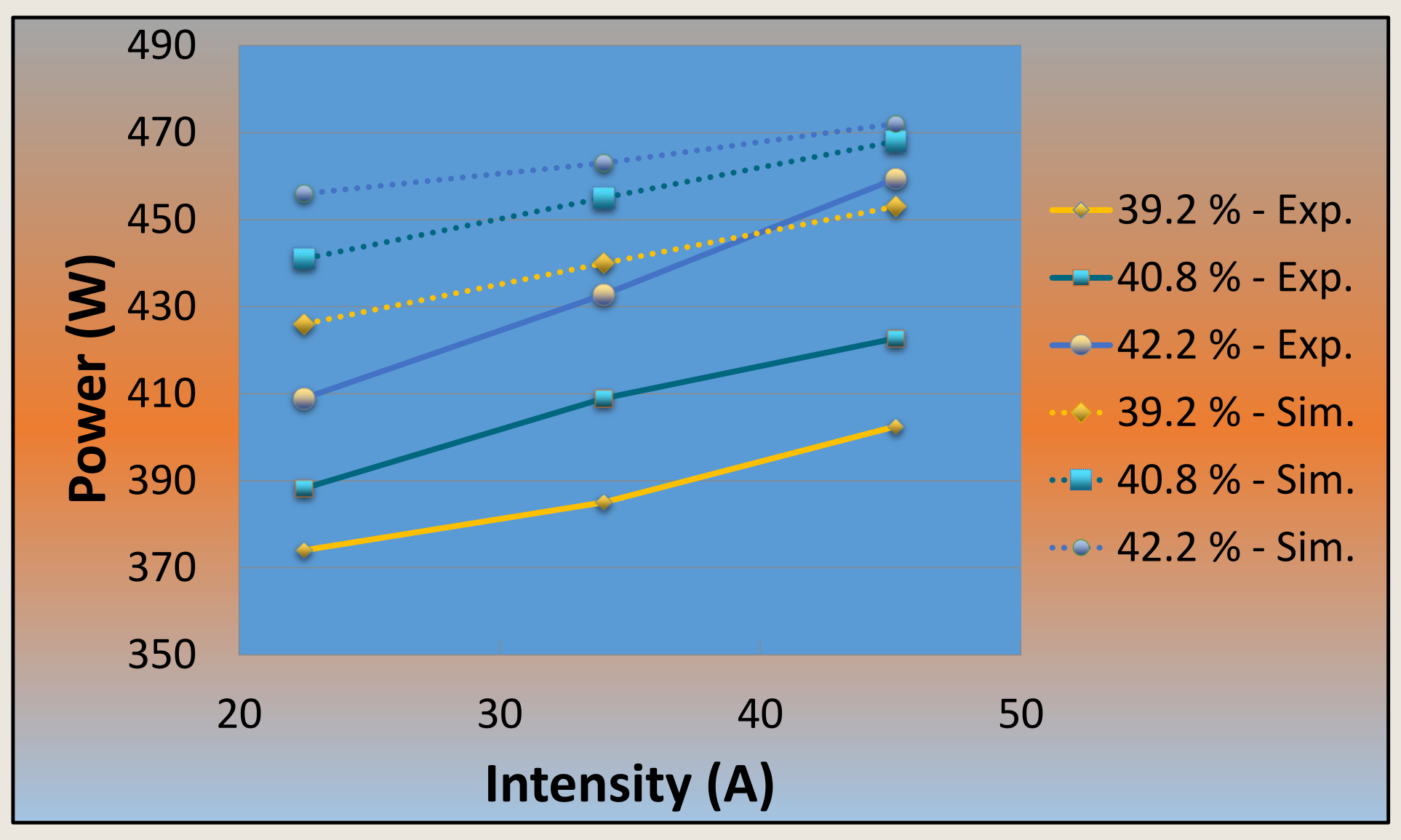

Figure 3. Simulated and measured power evacuated by the cooling system.

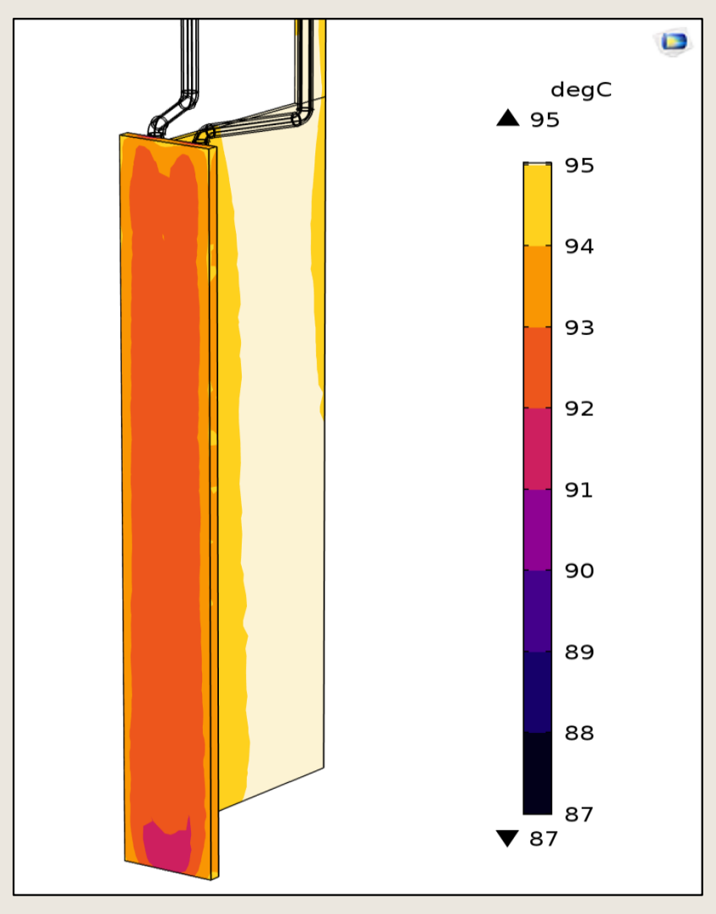

Figure 4. Temperature of the cathode with the cooling tube.

Major trends captured by the model:

- higher intensity → increased convection and ohmic drop,
- higher HF content → decreased ohmic drop but also decreased viscosity → better heat transfer between the cooling system and the electrolyte.

Several hypothesis to explain small gaps between experimental and simulated results:

- errors in physical properties,
- wrong flow field around the cooling tube in the electrolyte,
- inaccurate simulation of the solidified electrolyte on the cooling tube.

Conclusion: the 3D-fully coupled model developed shows good agreement with experimental data and can be a tool to understand better the fluorine production process.

Next steps: improving the two-phase flow model and the heat transfer close to the cooling tube.

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