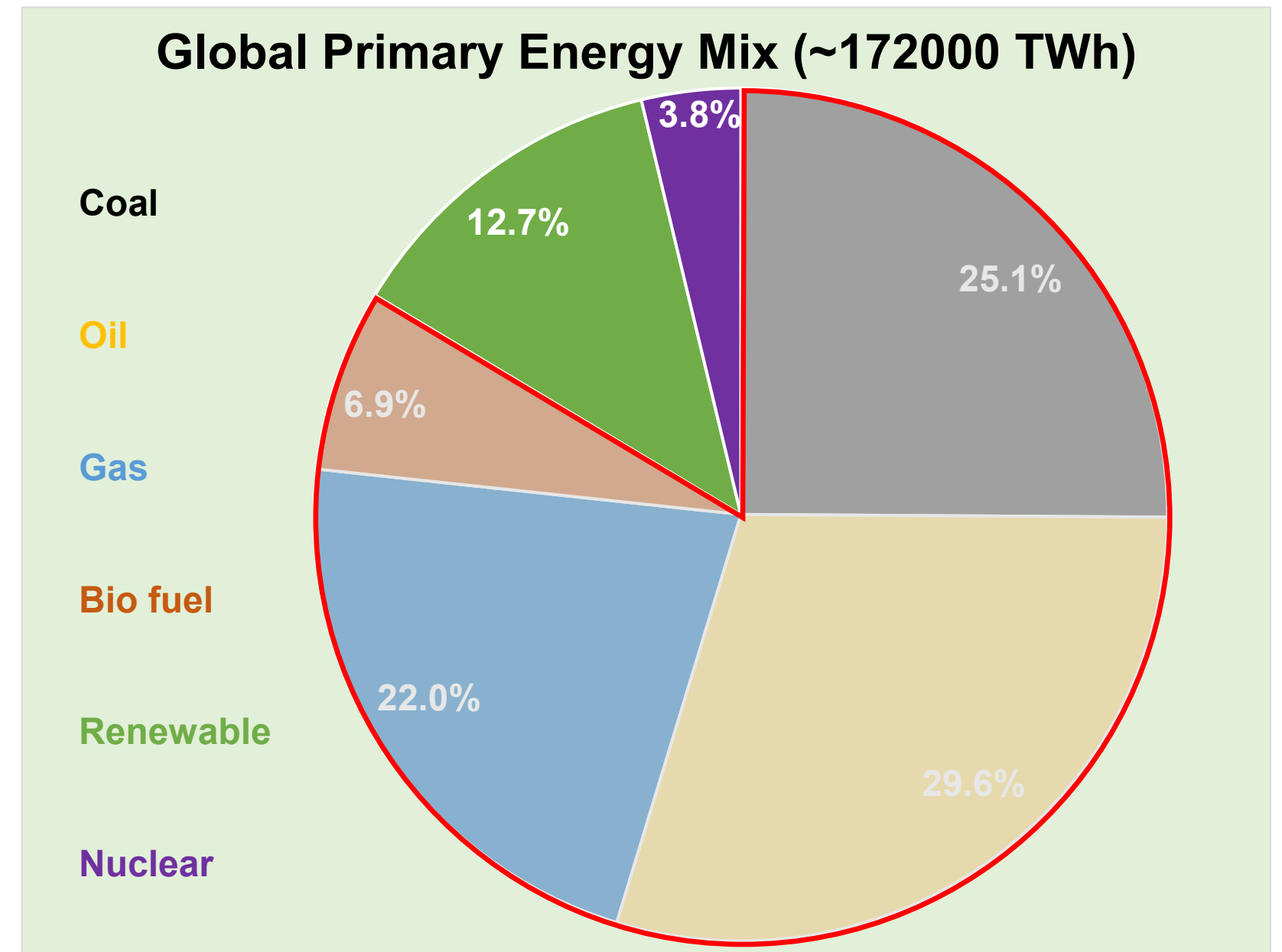
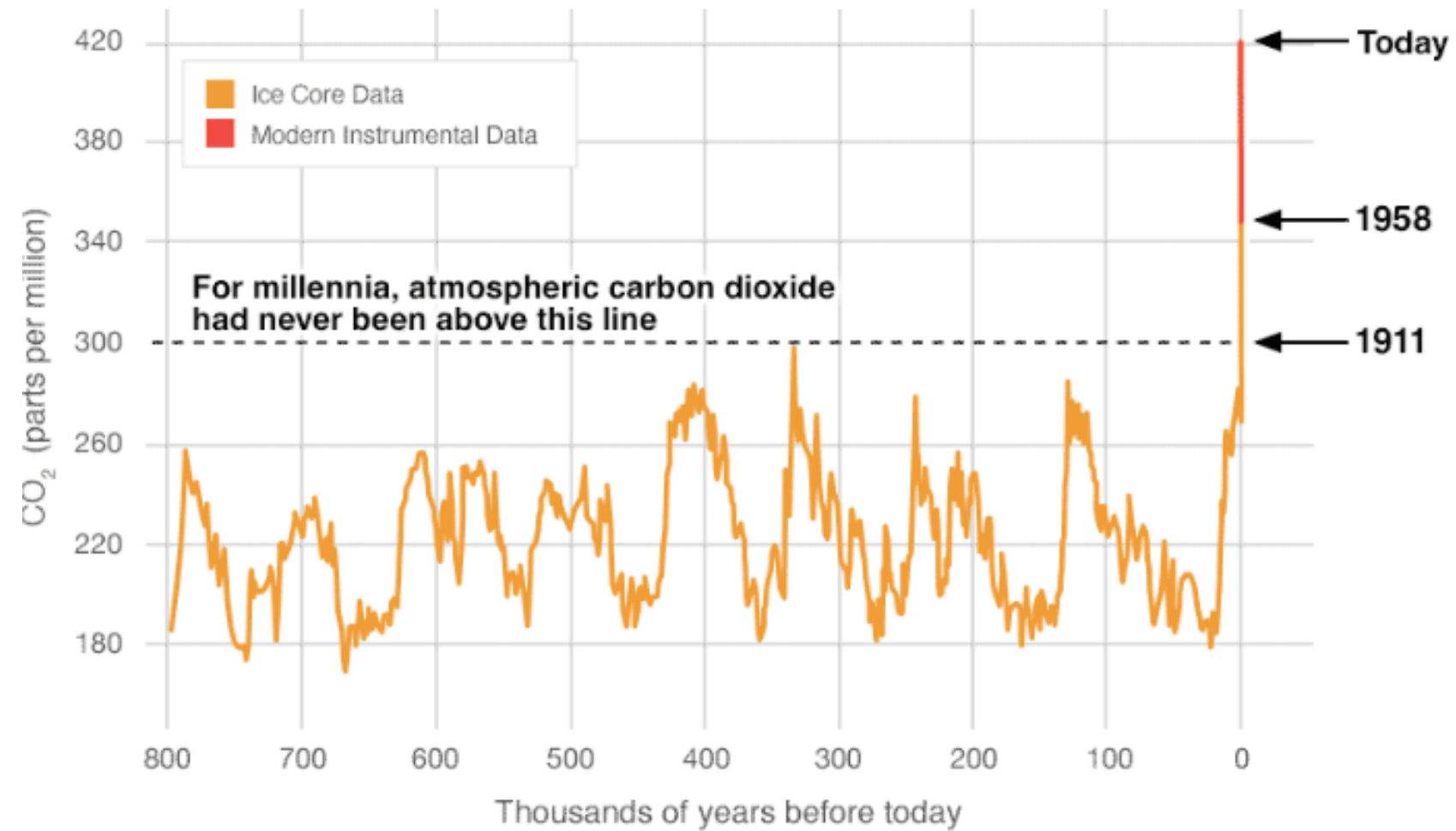


Electrothermal Fluidized Bed Reactor for Methane Pyrolysis: Toward Zero-Emission Hydrogen Production

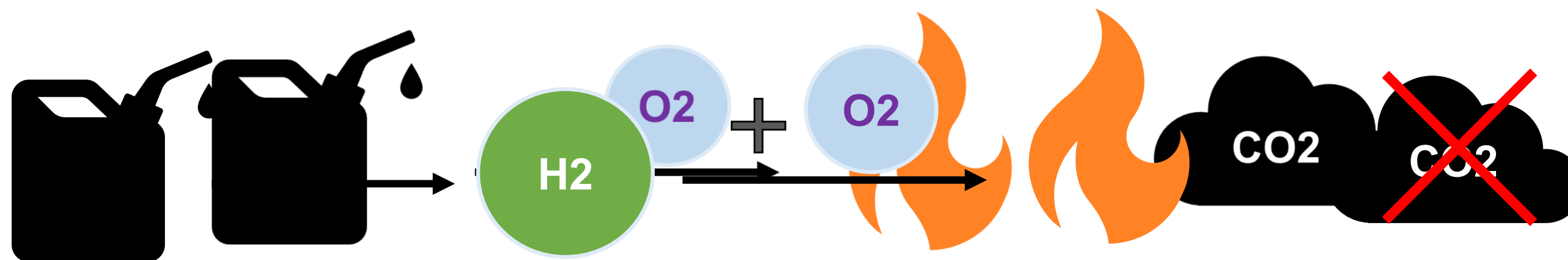
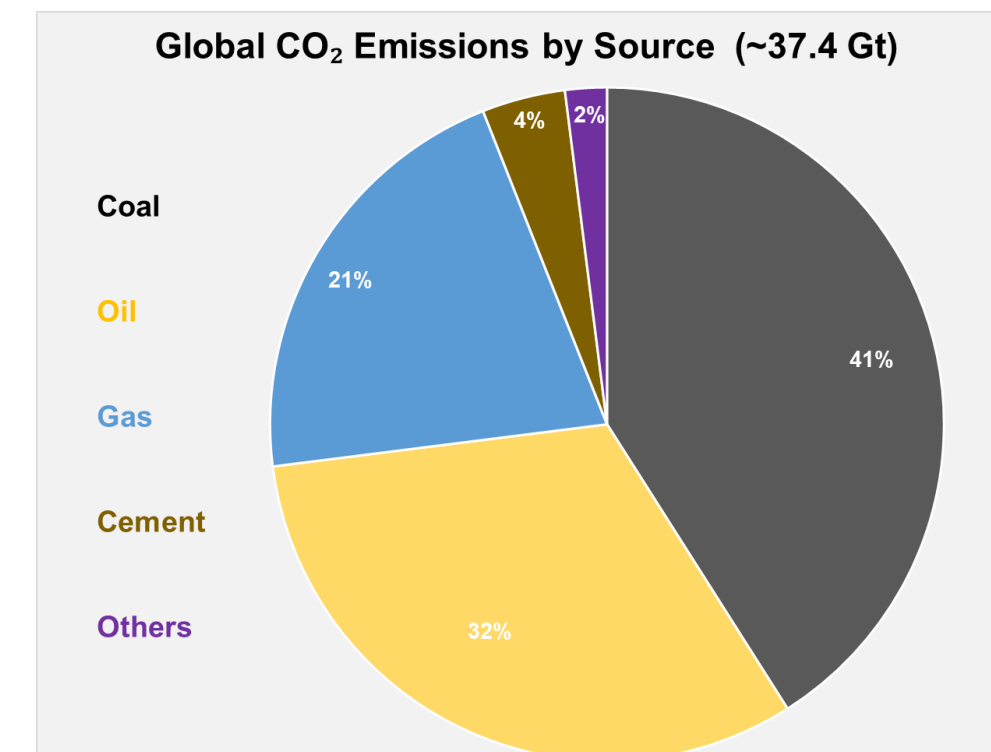
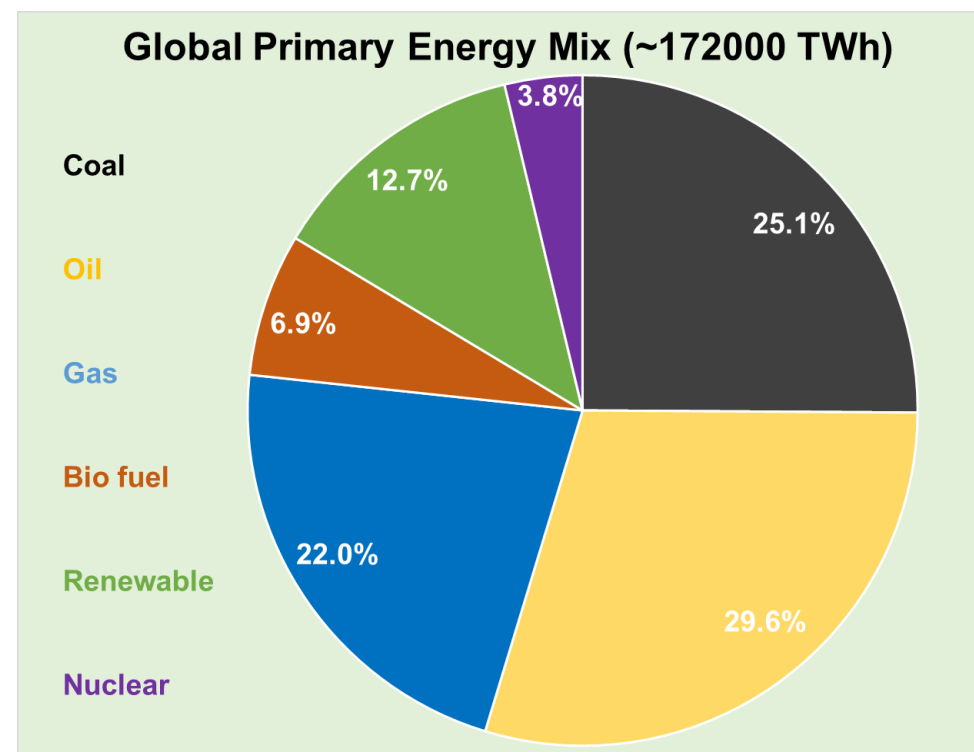
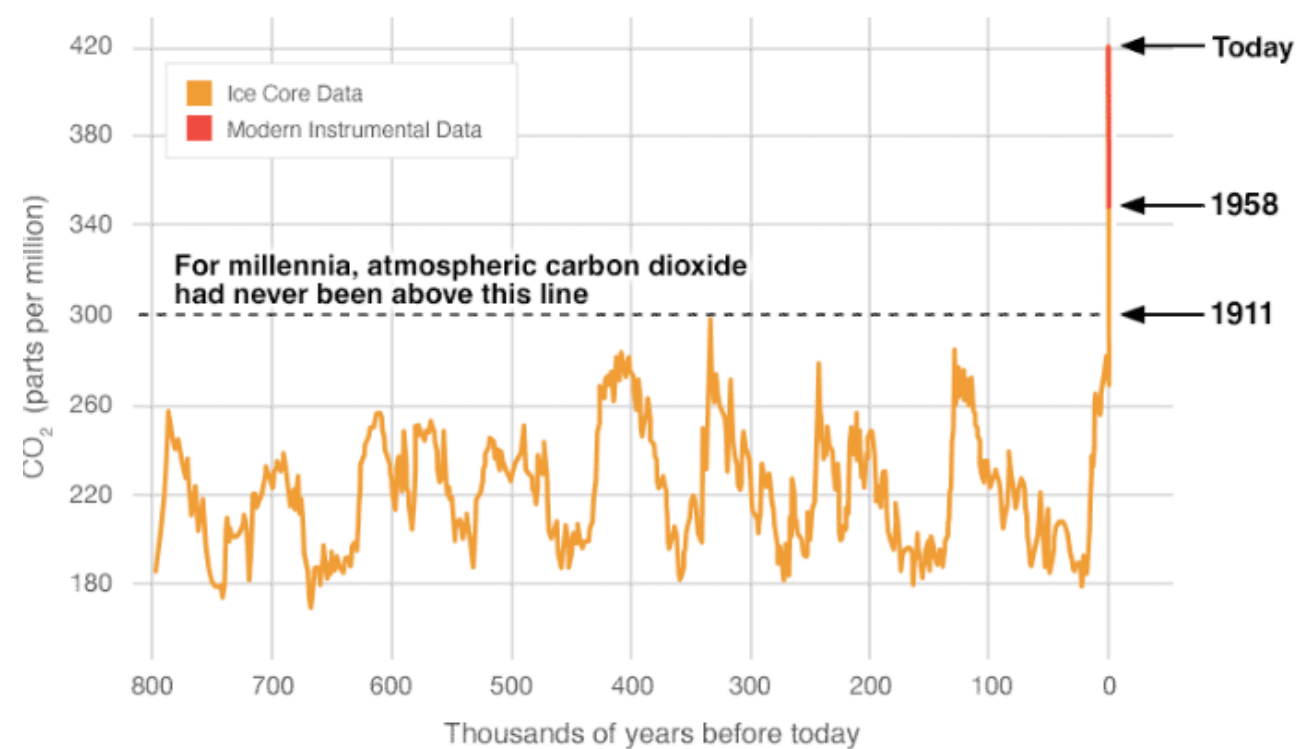
Soroush Zareghorbaei, Klaus Jacobs, Sébastien Siradze, Jeroen Lauwaert, Joris Thybaut

Laboratory for Chemical Technology (LCT), Ghent University, Technologiepark 125, 9052, Ghent , Belgium

CO₂ challenge and energy transition

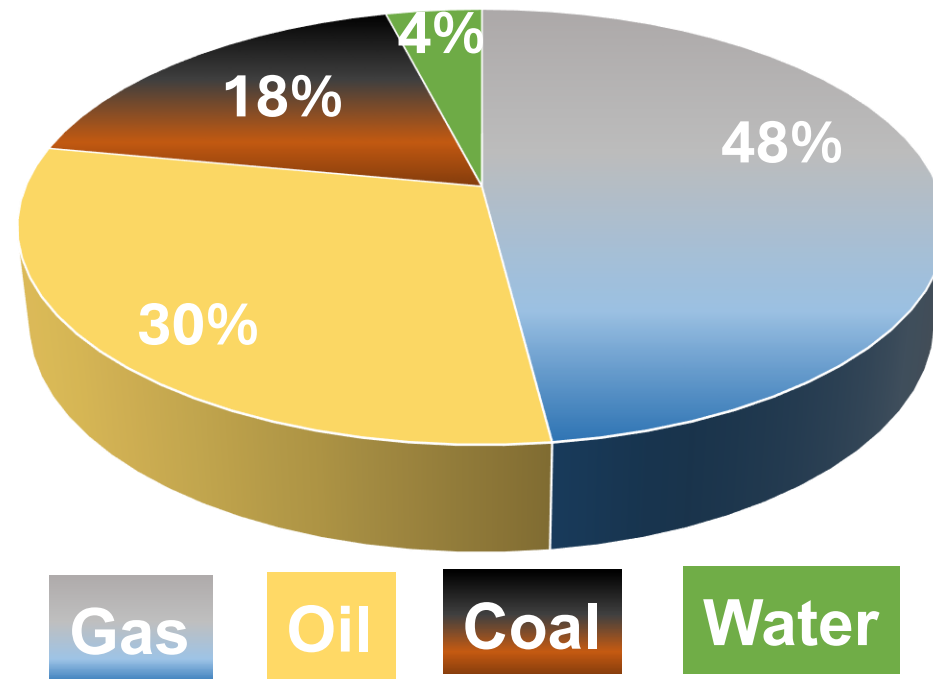


CO₂ challenge and energy transition



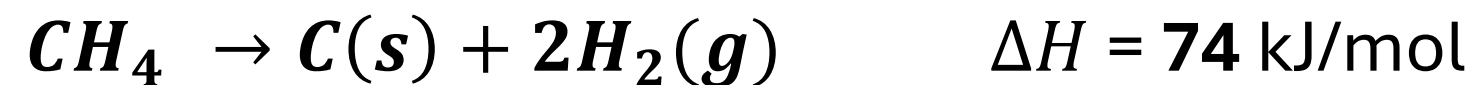
H₂ production technologies

H₂ production feedstocks



Technology	reaction	ΔH kJ/mol	cost (\$/kg)	GWP (kg CO ₂ eq)
Steam Methane Reforming SMR + CCS	$CH_4 + H_2O \leftrightarrow CO + 3H_2$	206	2.08	11.9
			2.27	3.7
Coal Gasification CG + CCS	$C + H_2O \leftrightarrow CO + H_2$	-130	1.34	22.9
			1.63	4.8
Electrolysis	$2H_2O \leftrightarrow 2H_2 + O_2$	285	>3	>0.7

Catalytic Methane Decomposition:



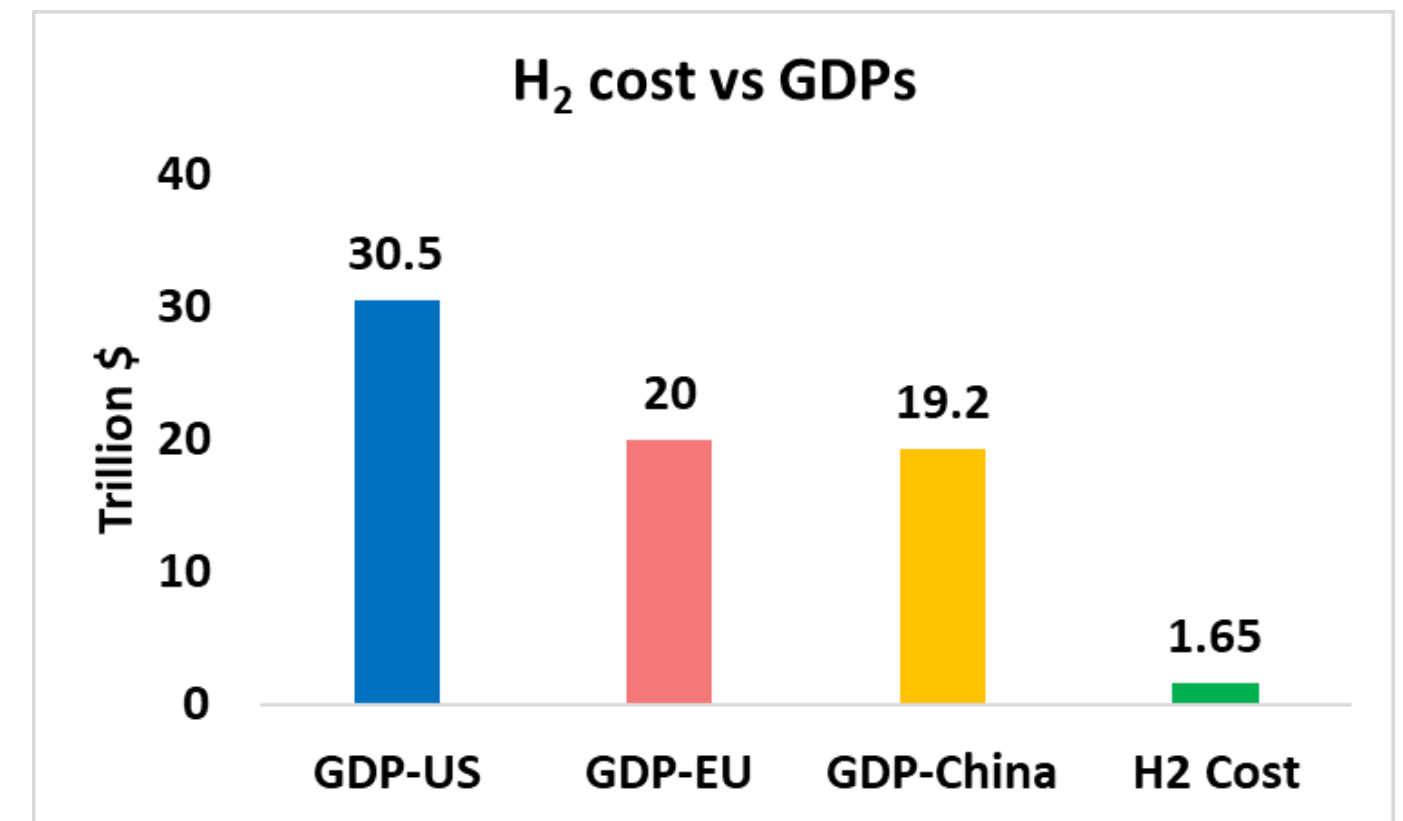
CDM on carbonaceous catalyst:

- ☐ less energy-intensive
- ☐ Less greenhouse gas emission
- ☐ Produces H₂ with high purity
- ☐ Cheap catalyst
- ☐ Sulfur-resistive catalyst

can H₂ replace methane affordably?



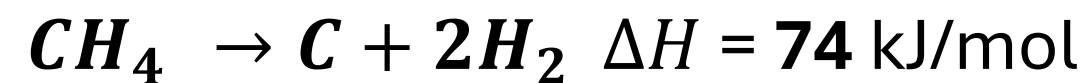
- CH₄ consumption in 2024: ~2.9 Gt
- **H₂ route needs ~50% more CH₄**
- H₂ production ~1.1 Gt
- **CO₂ avoided ~9 Gt/yr**
- H₂ production cost ~ 1.5 \$/kg
- **Annual H₂ cost ~\$1.65 trillion**



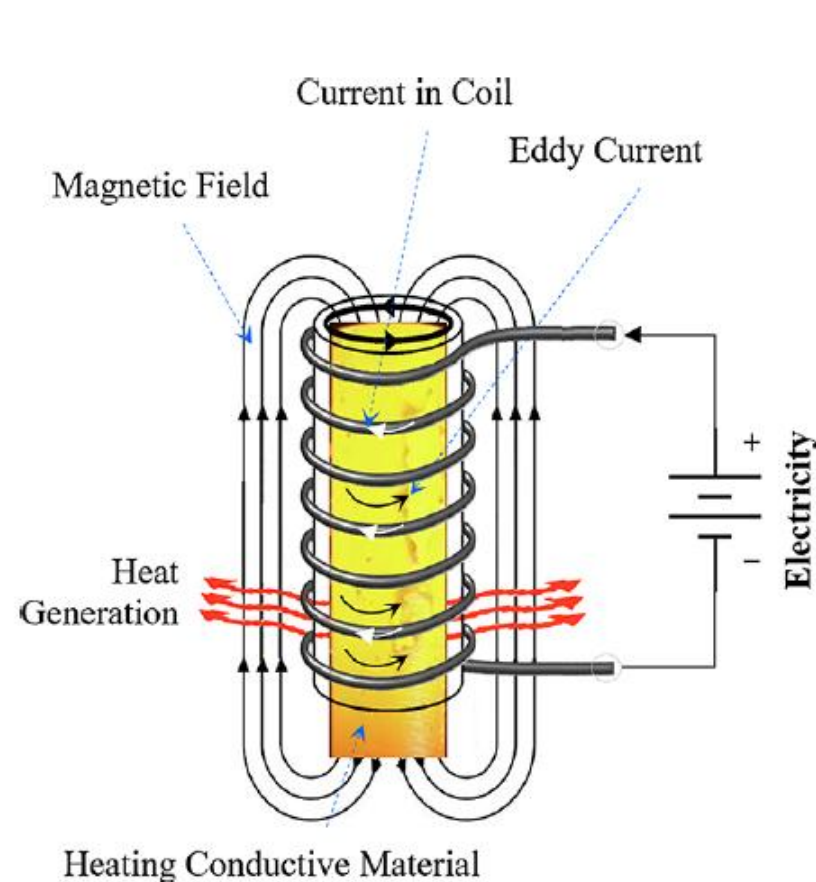
outline

- introduction
- model development
 - ElectroThermal Fluidized Bed reactor (ETFB)
- processes
 - e-CODUCT: simultaneous CO₂ and H₂S abatement
 - OBIWAN: biogas upcycling via H₂ production
- conclusions & perspectives

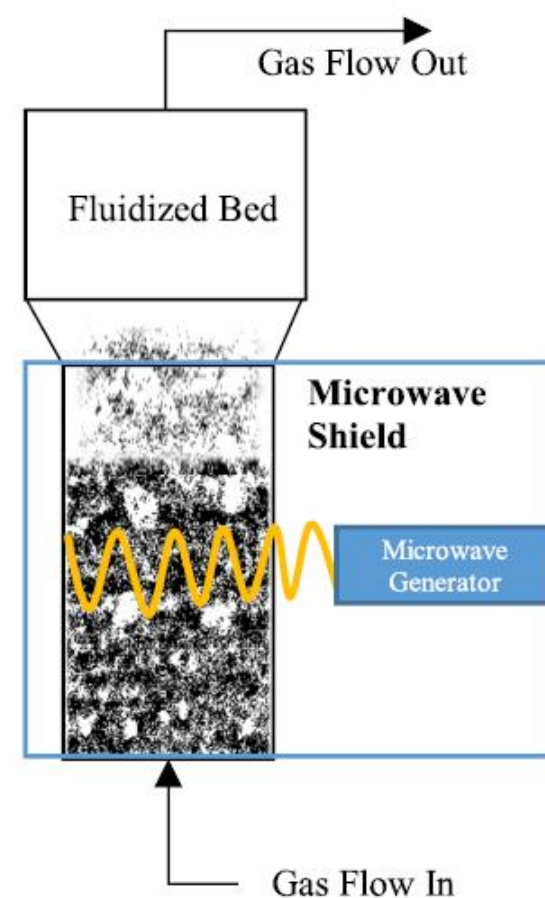
advantages of electrical heating



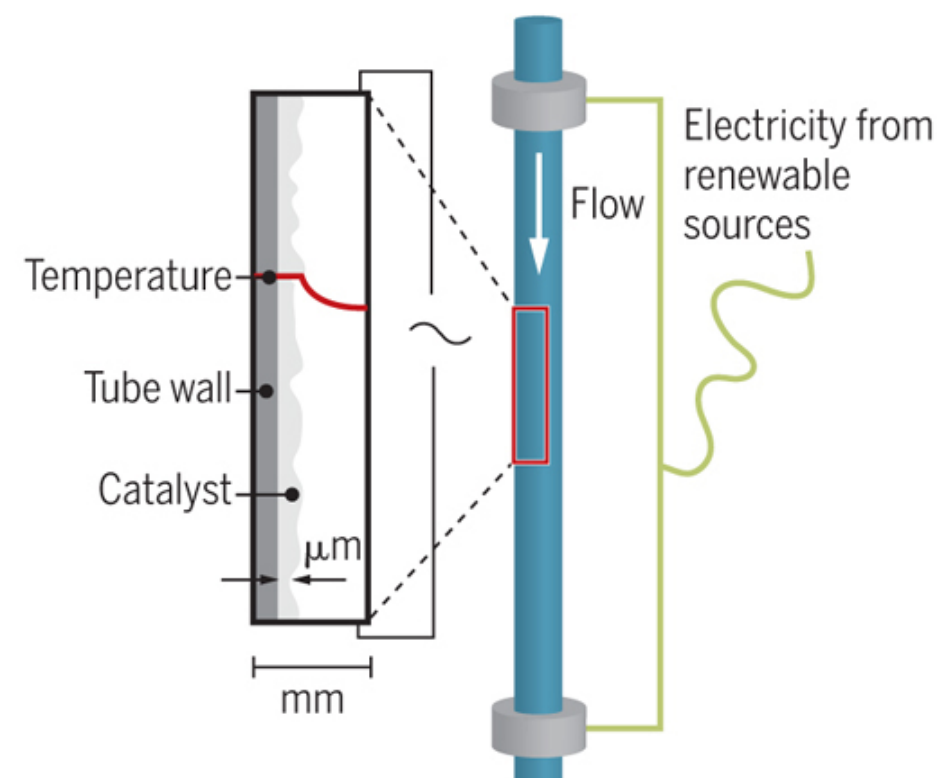
induction



microwave



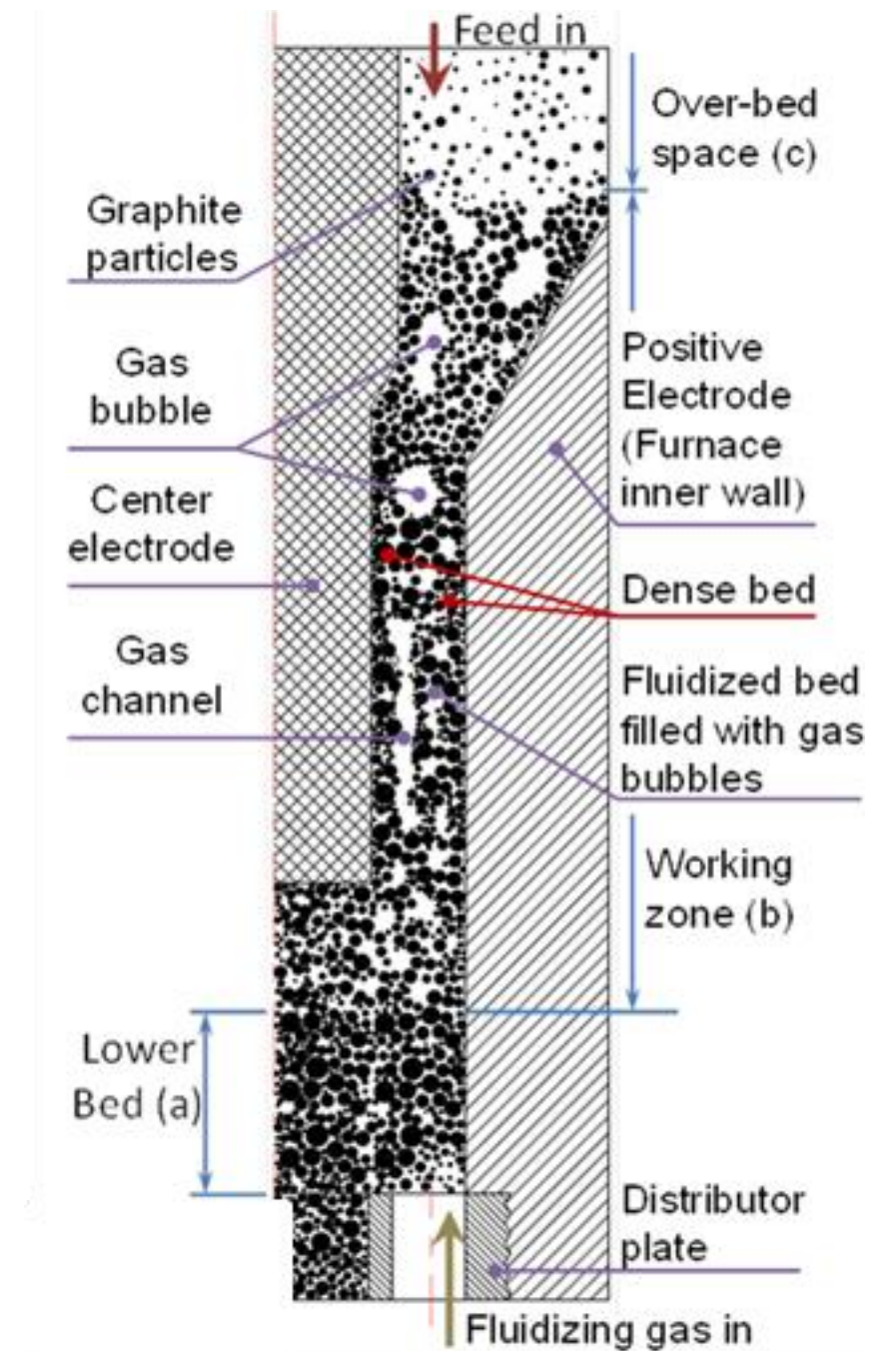
Joule



- zero emission
- energy efficient
- better T control
- rapid heating

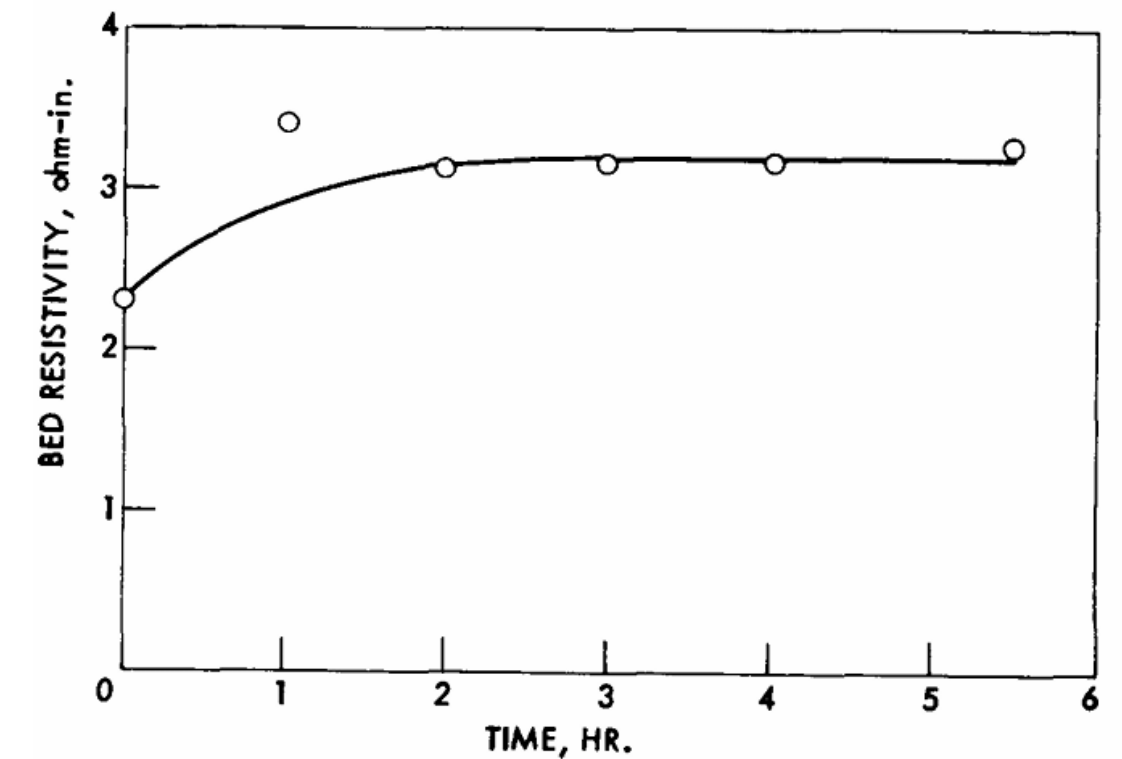
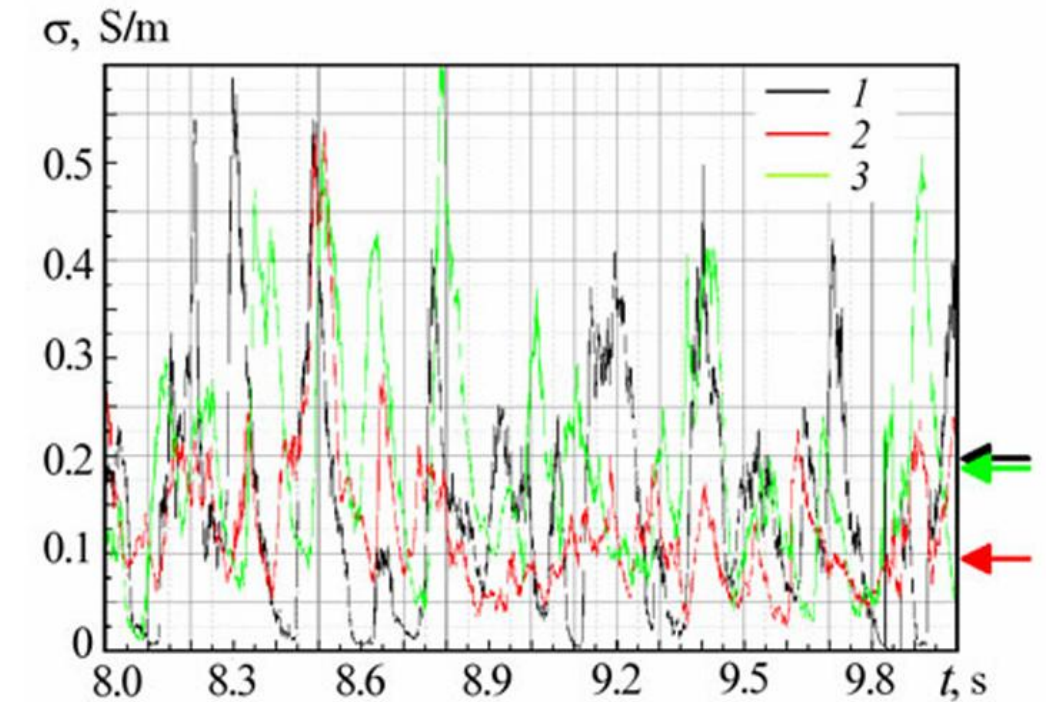
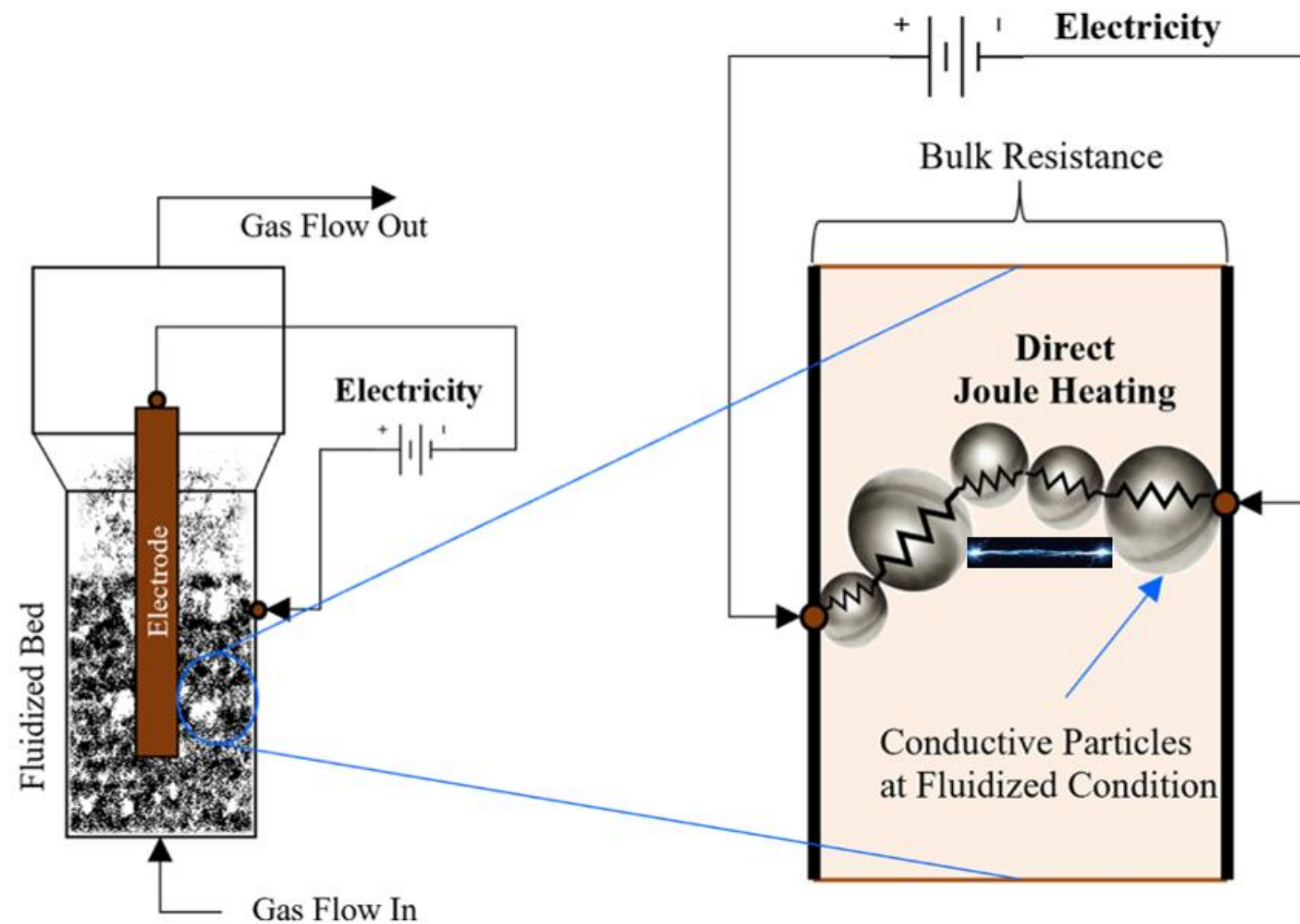
ElectroThermal Fluidized Bed reactor (ETFB)

- combination:
 - fluidization
 - Joule heating
- compared to conventional fluidized beds
 - better control over bed temperature
 - higher energy efficiency
 - rapid and uniform heating

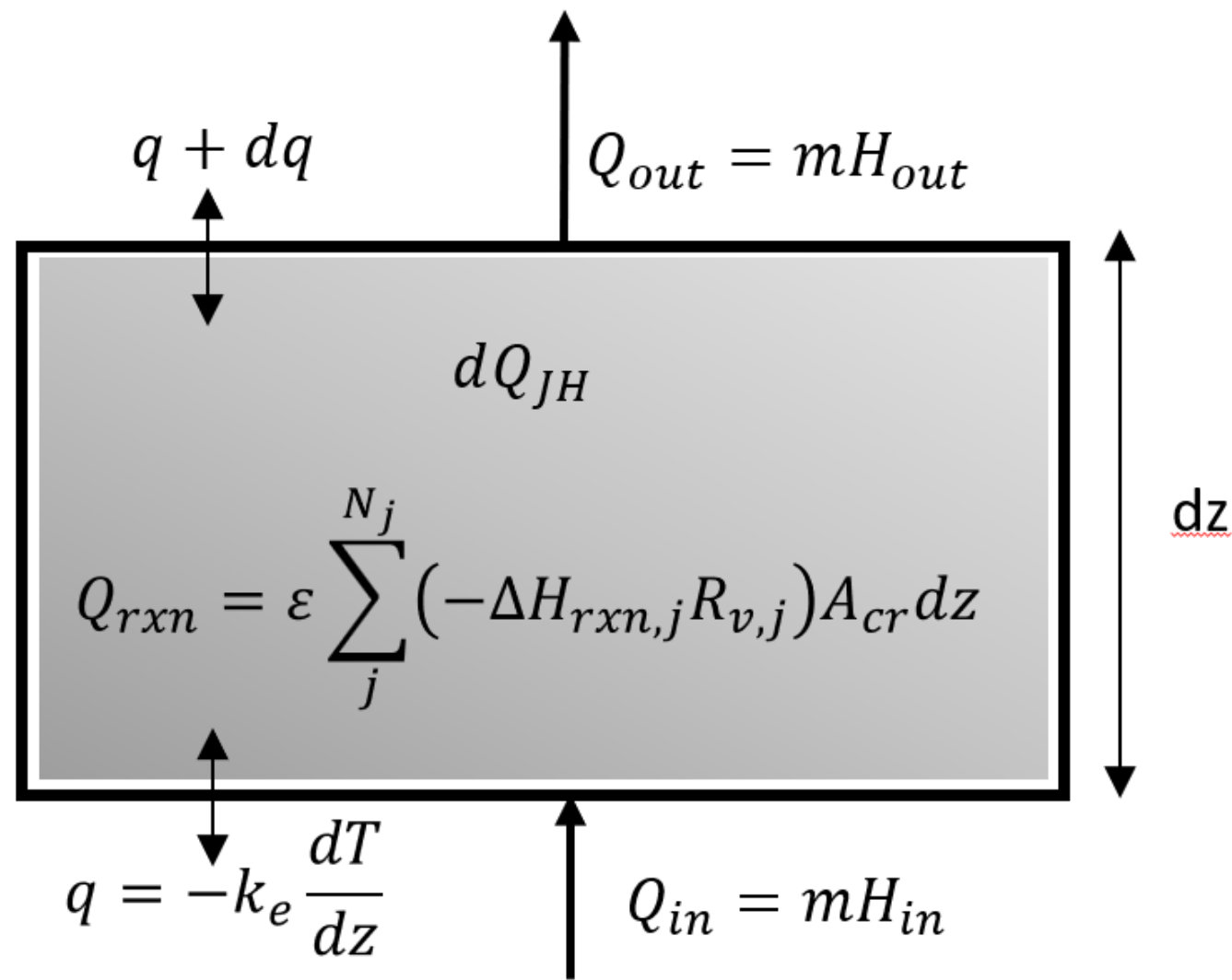


Fedorov *J. Fluids Eng.* 138 (2016) 044502

Joule heating in a fluidized bed



ETFB reactor heat balance



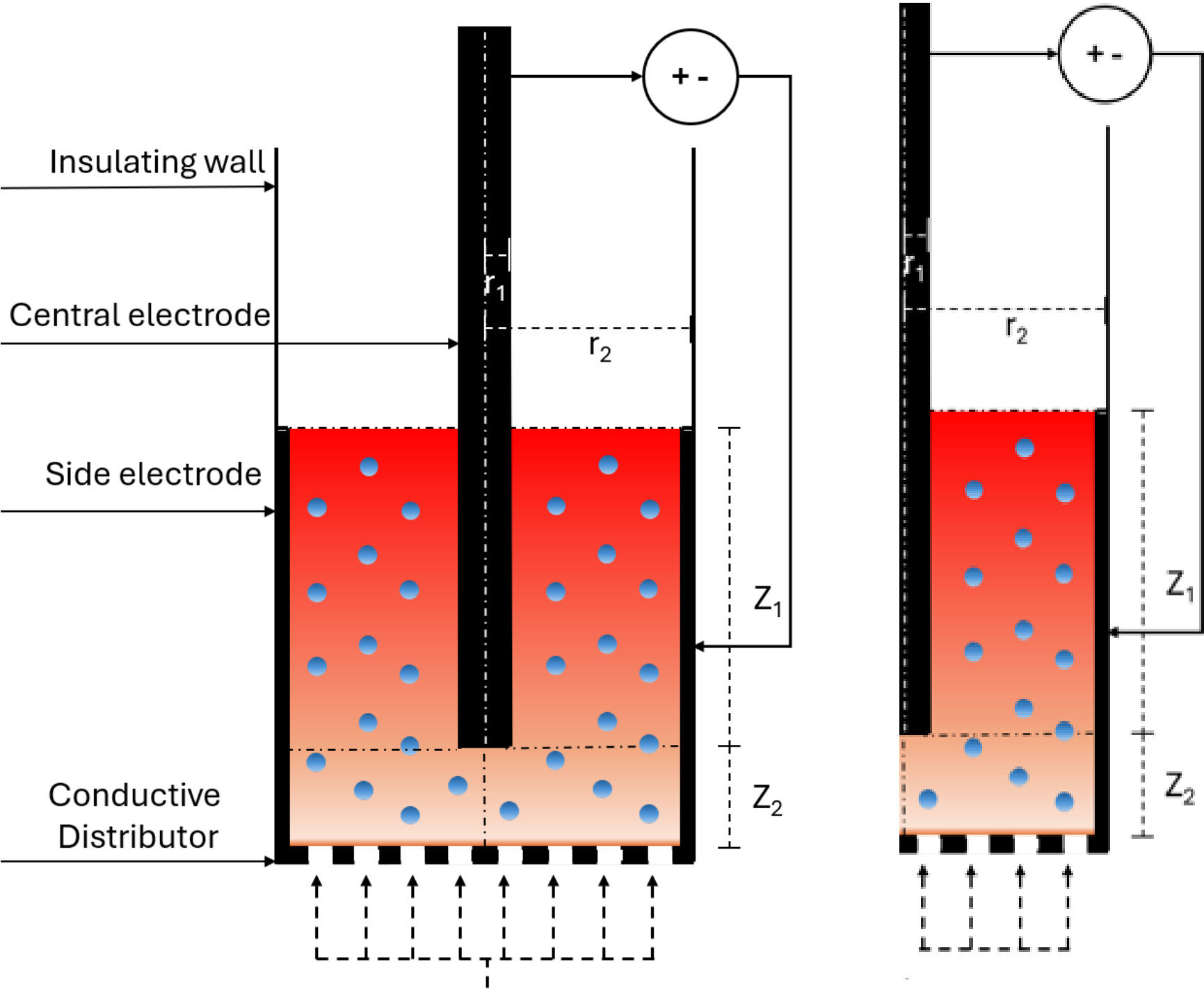
$$u\rho_g C p_g \frac{dT}{dz} - k_e \frac{d^2T}{dz^2} - \varepsilon \sum_j^{N_j} (-\Delta H_{rxn} R_{v,j}) - \frac{dQ_{JH}}{dz A c_{reactor}} = 0$$

convection conduction heat of reaction heat generation
 $\rightarrow$ Joule heating term

assumptions

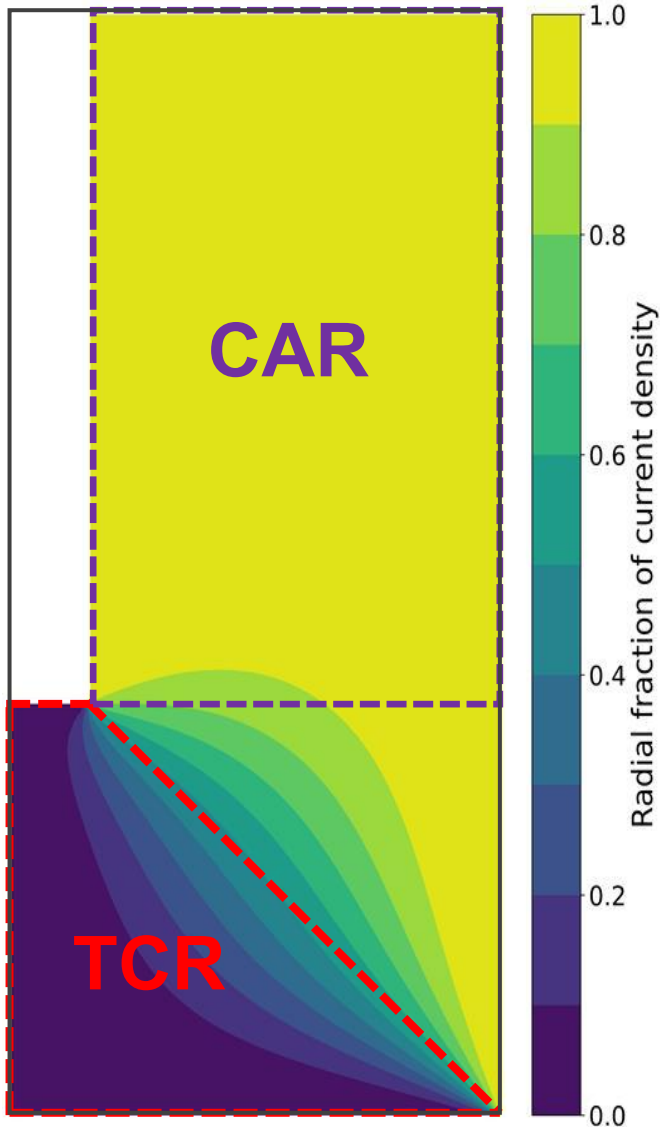
- bubbling fluidization regime
- low- and high-density phases are at the same temperature
- no radial profiles
- constant effective conductivity at average temperature and composition
- adiabatic operation
- steady state operation

ETFB electrical field



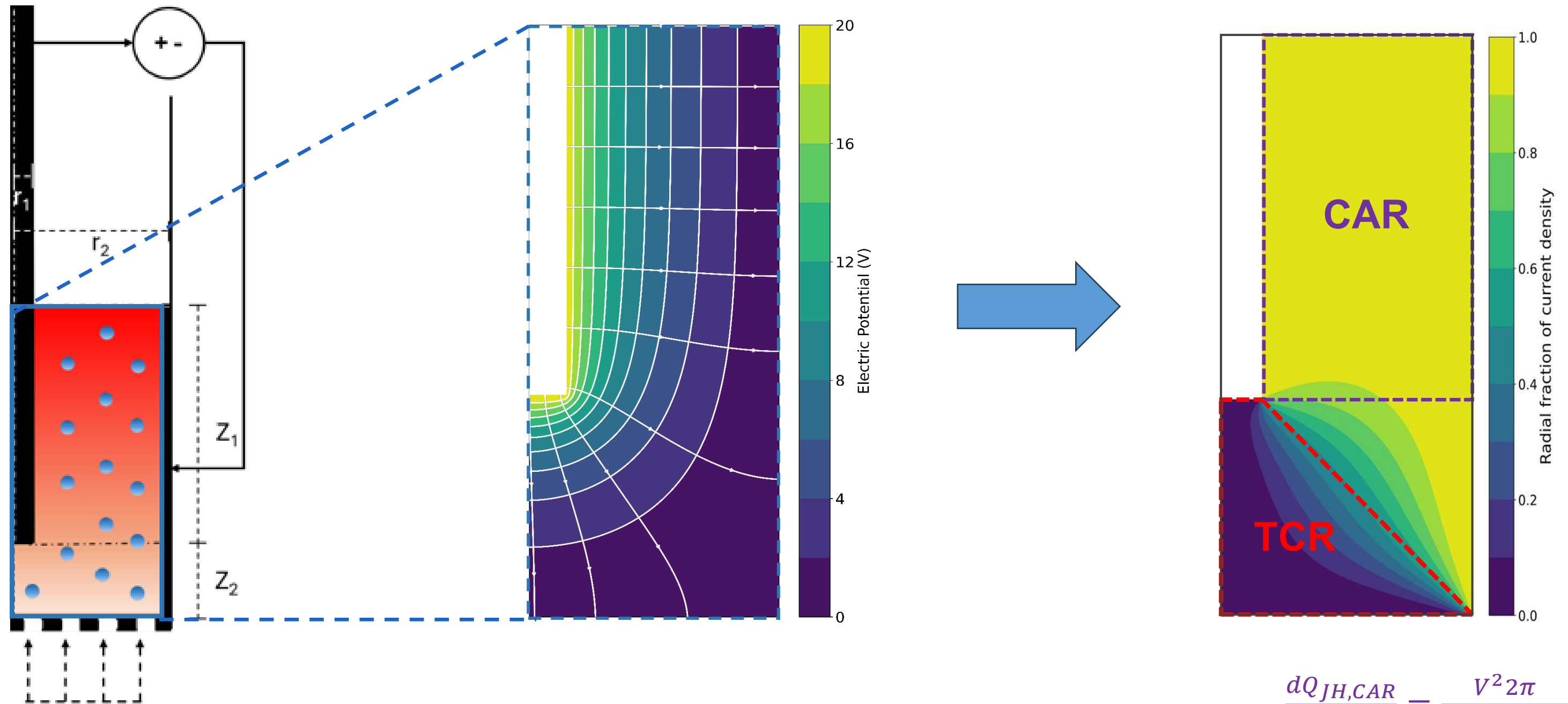
$$\frac{d^2V}{dr^2} + \frac{1}{r} \frac{dV}{dr} + \frac{d^2V}{dz^2} = 0$$

coaxial resistor



truncated cone resistor

Laplacian Filed Model vs Current Partition Model



LFM:
$$\frac{dQ_{JH}}{dz} = \int_0^R -\frac{2\pi}{s_{bed}} \left(\left(\frac{d\Delta V}{dr} \right)^2 + \left(\frac{d\Delta V}{dz} \right)^2 \right) r dr$$

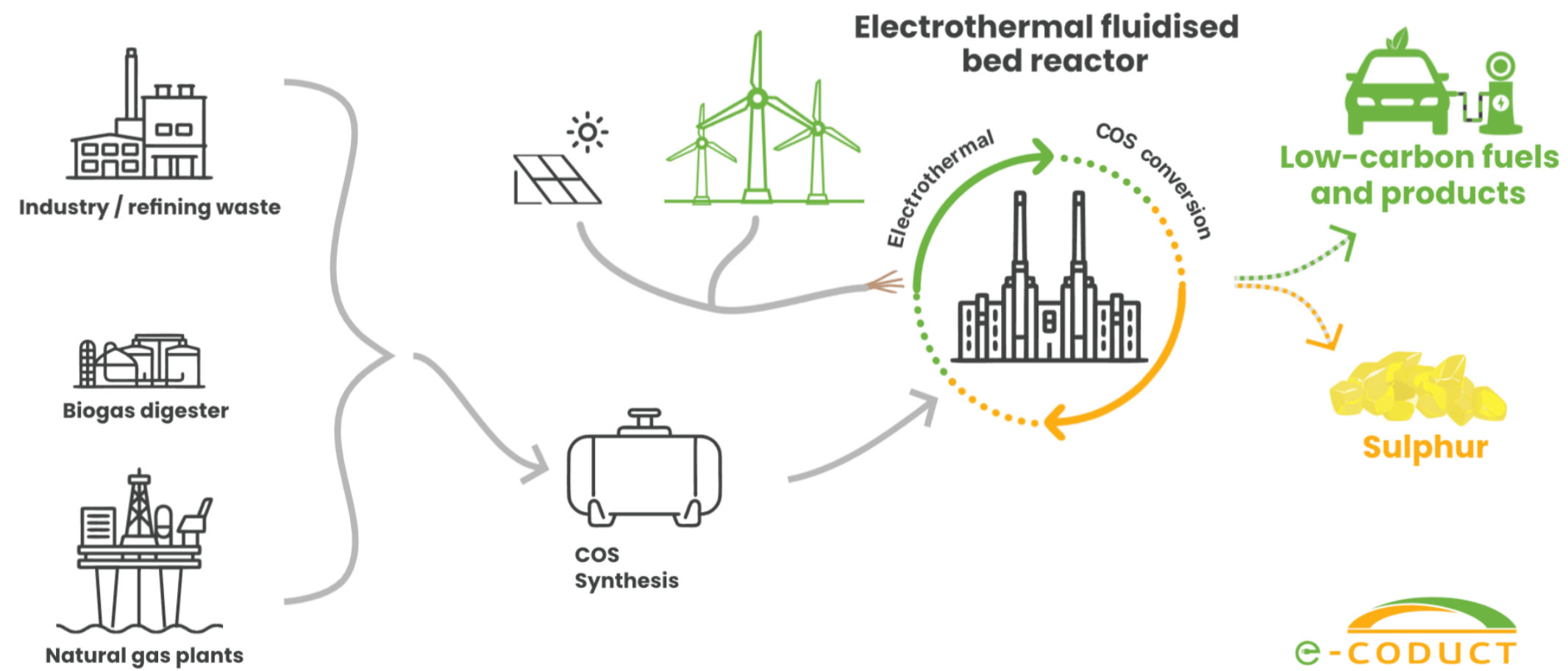
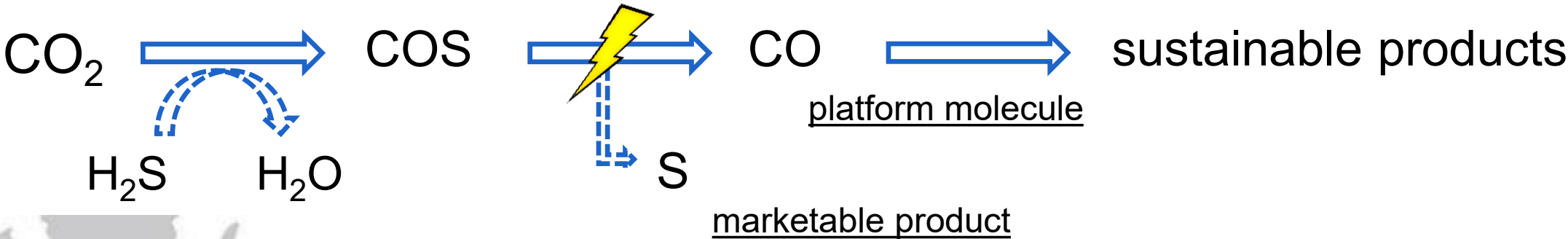
CPM:
$$\frac{dQ_{JH,CAR}}{dz} = \frac{V^2 2\pi}{s_{bed} \ln\left(\frac{r_2}{r_1}\right)}$$

$$\frac{dQ_{JH,CAR}}{dz} = \frac{I_{TCR}^2 s_{bed}}{\pi r^2}$$

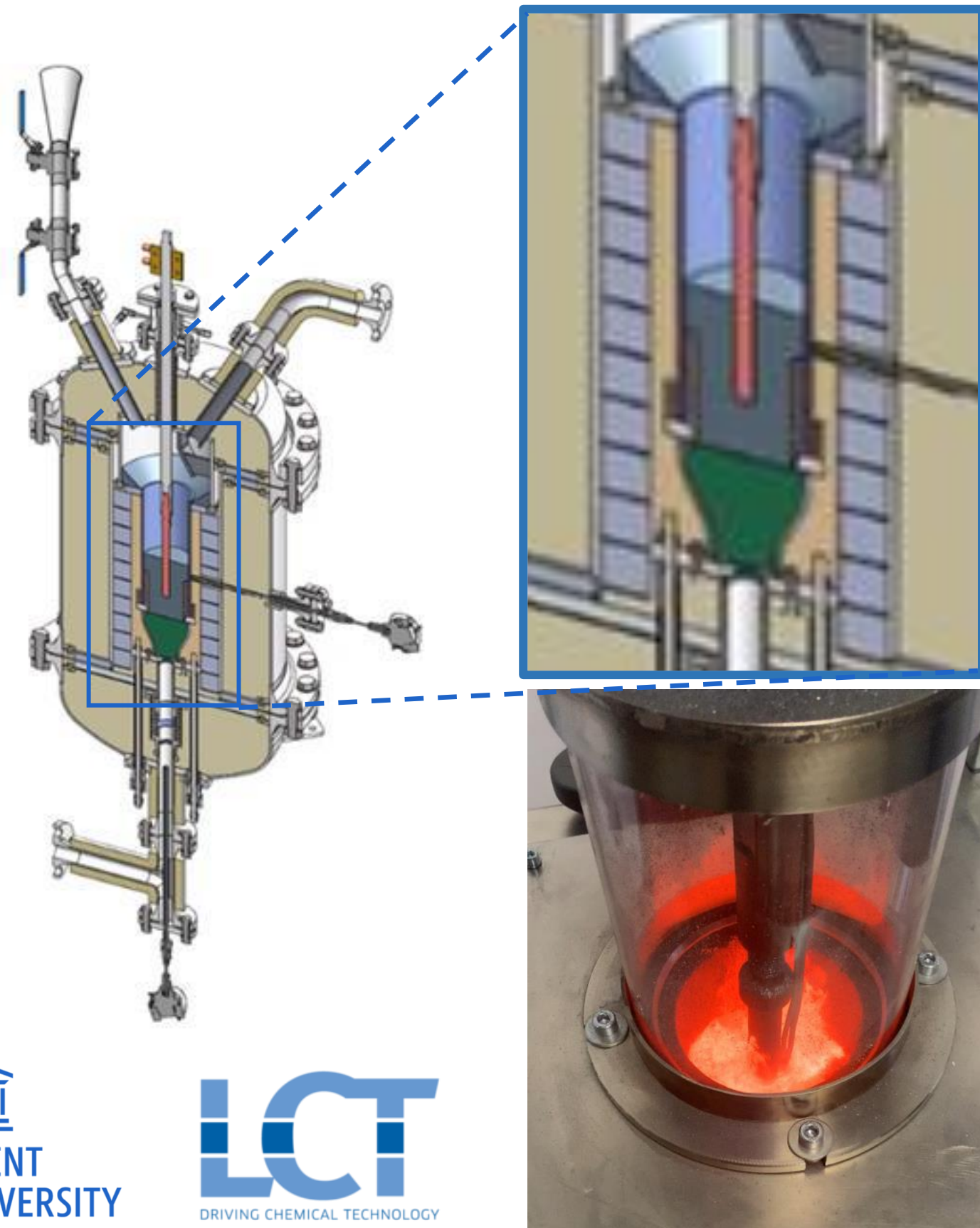
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e-CODUCT: process lay-out

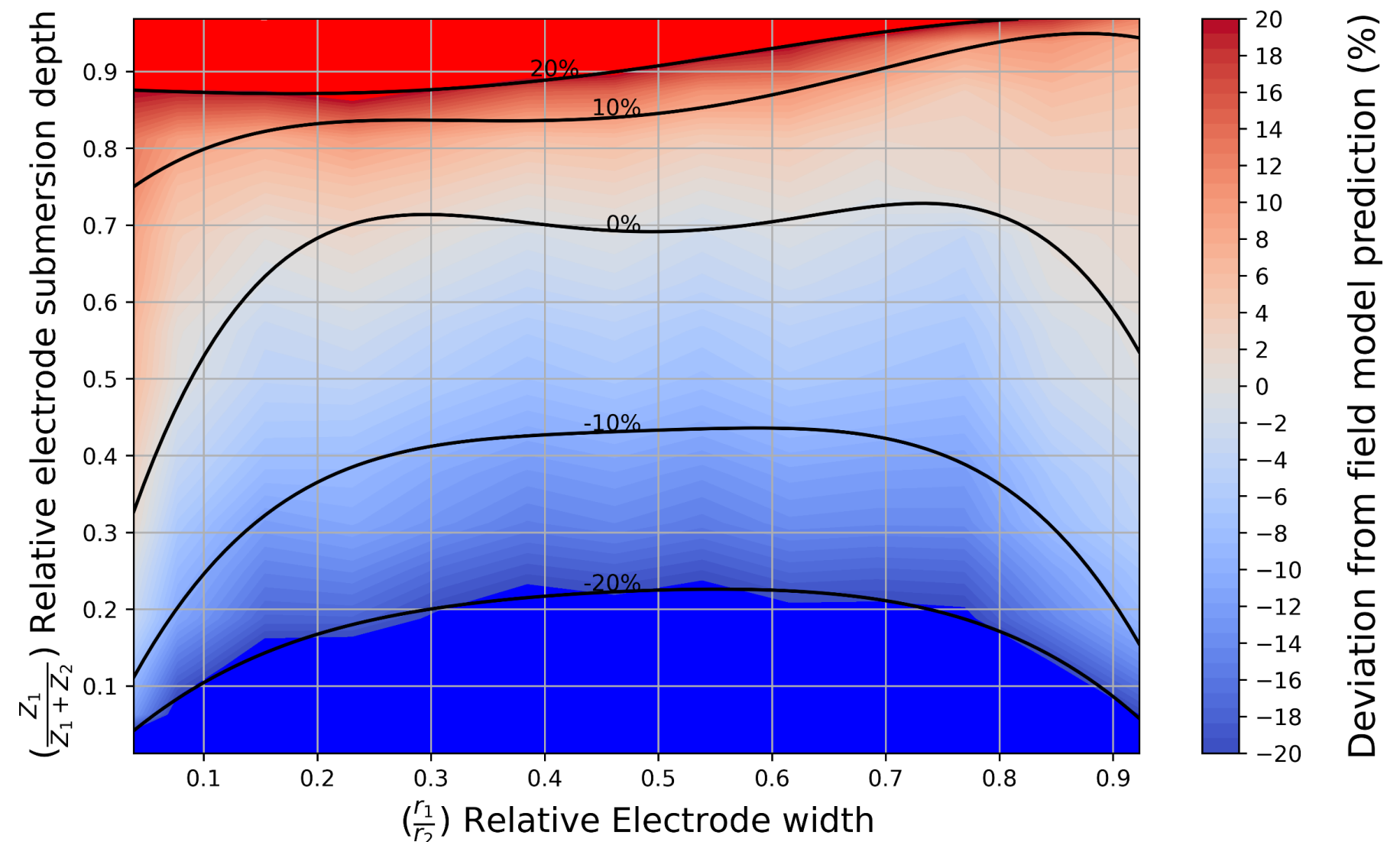


COS decomposition

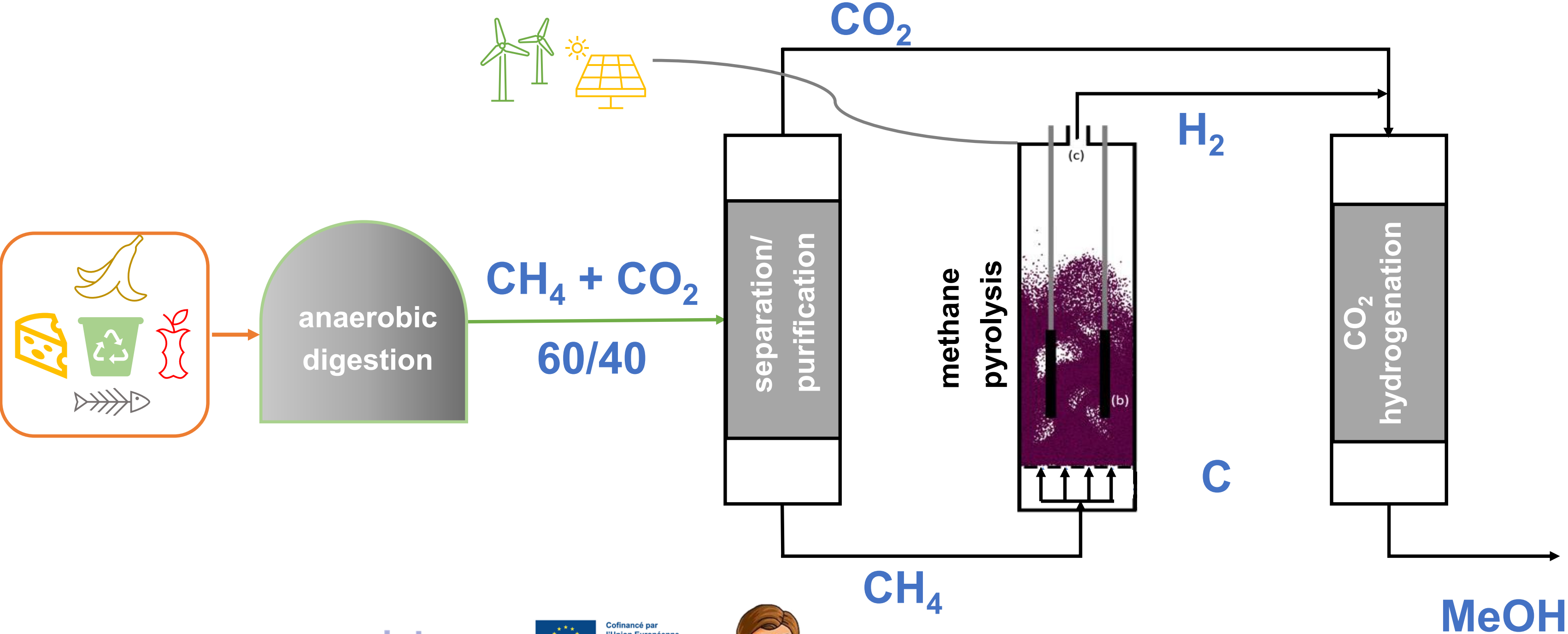


- COS decomposition to CO and sulphur
- temperatures up to 800-1200 °C
- in situ heat generation by joule heating

LFM vs CPM results

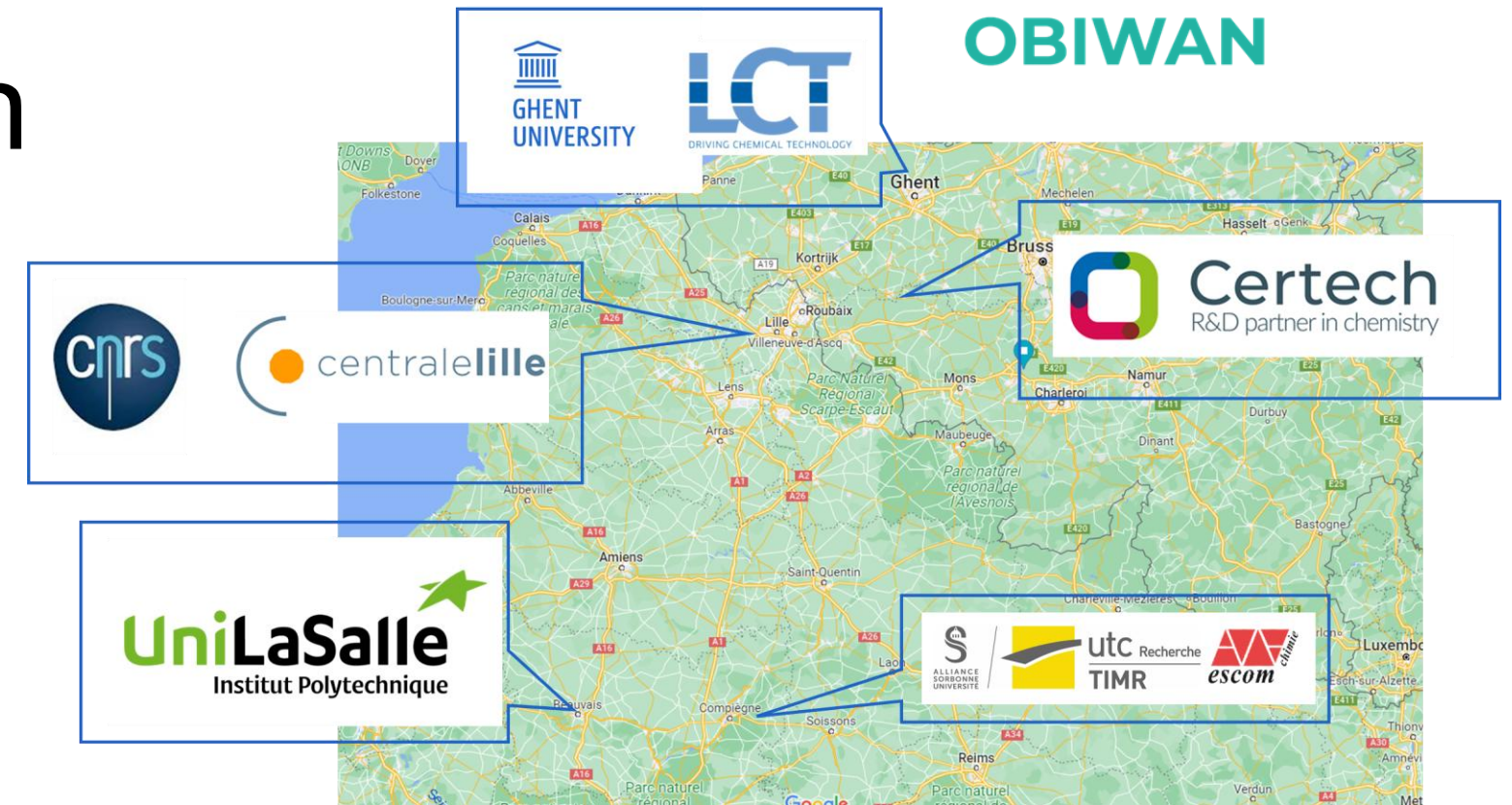


OBIWAN: process lay-out



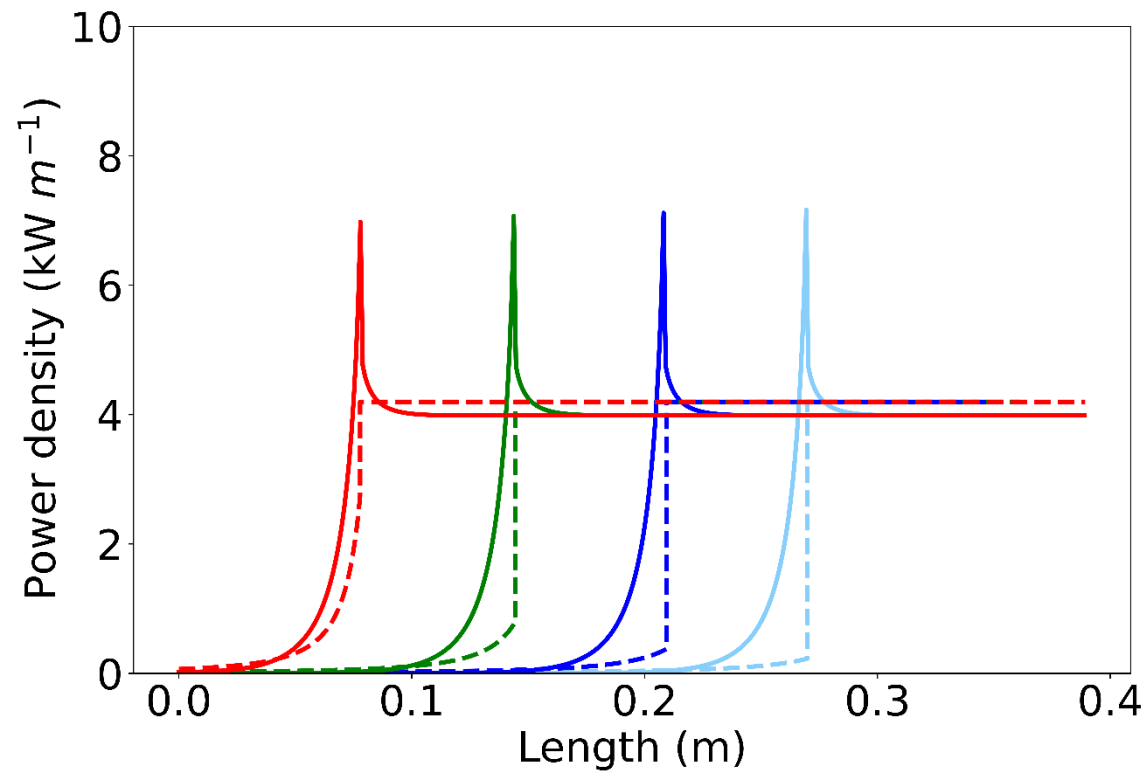
OBIWAN: context

- biogas ($\text{CH}_4 + \text{CO}_2$) valorization
 - calorific
 - to chemicals/(sustainable aviation fuels)



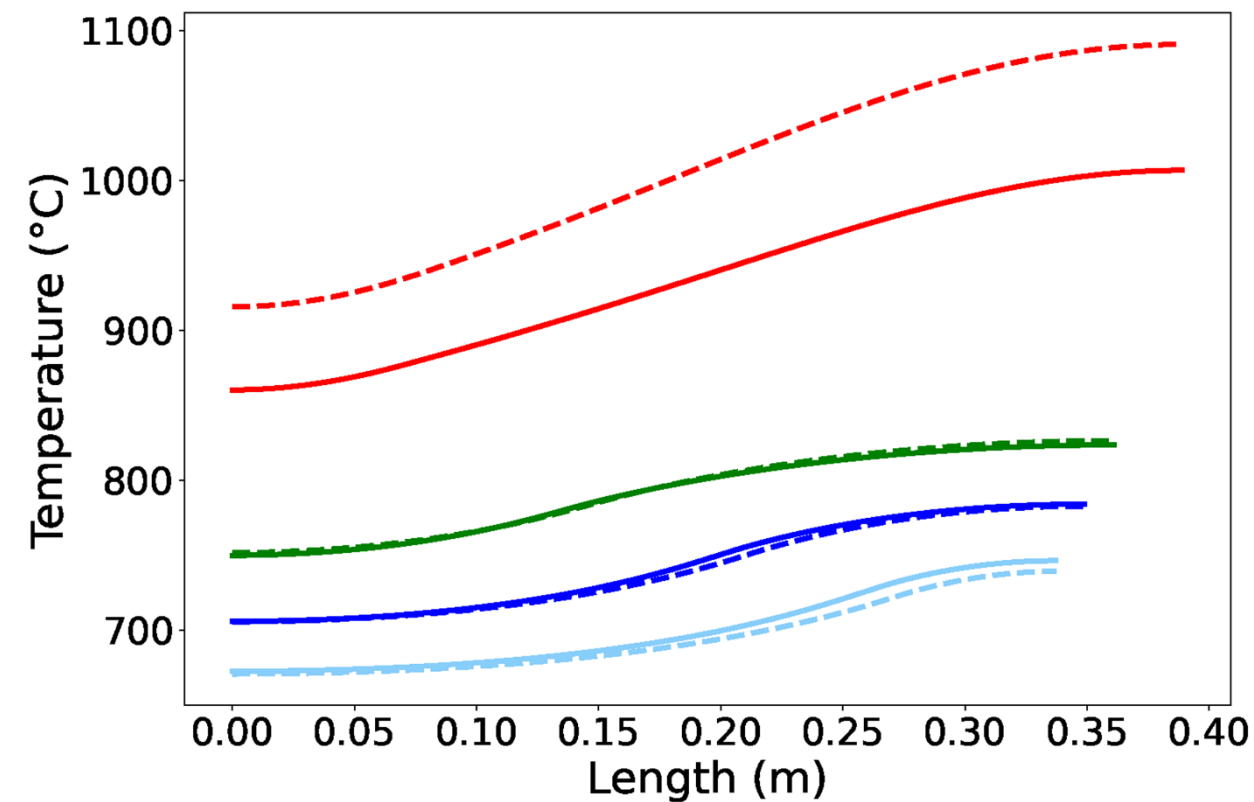
CH₄ pyrolysis in ETFBR

Effect of Electrode Depth on linear power density



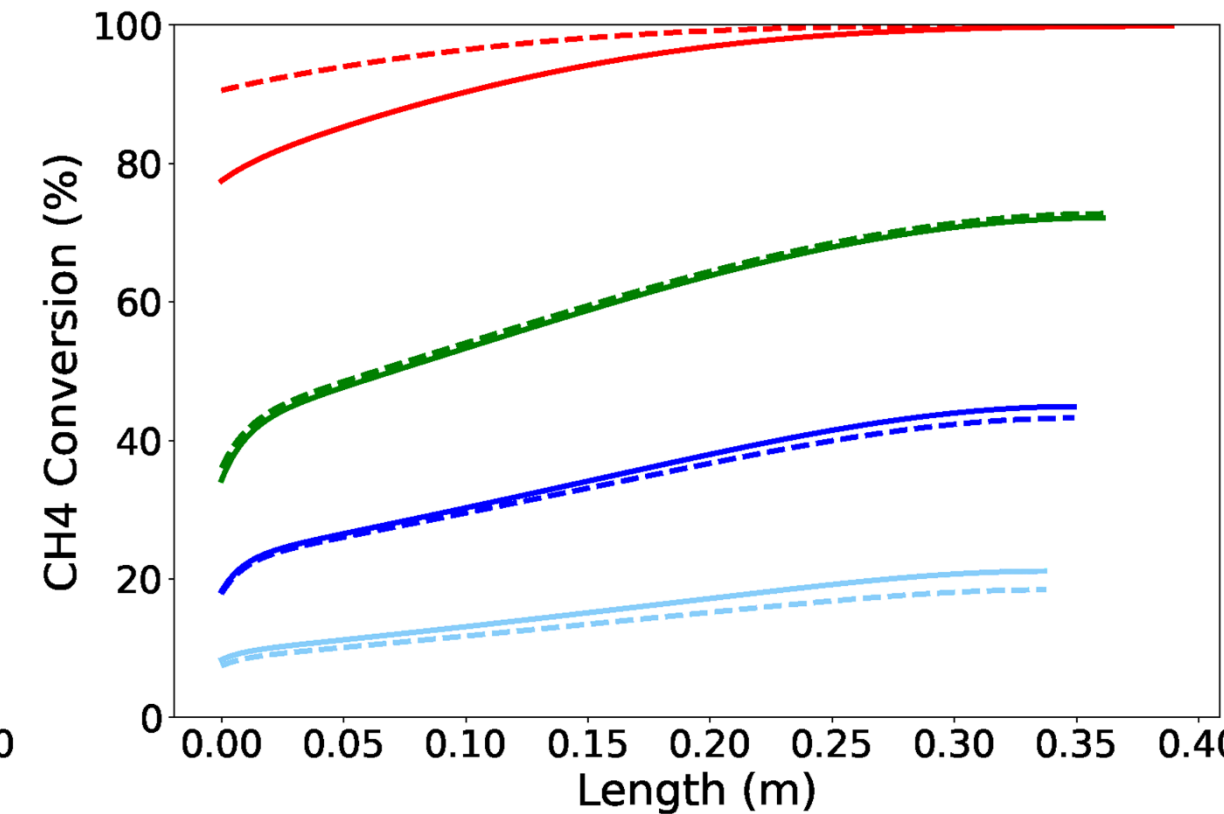
68 mm
 140 mm
 217 mm
 312 mm
 Current Partition Model
 Laplacian Field Model

Effect of Electrode Depth on temperature



68 mm
 140 mm
 217 mm
 312 mm
 Current Partition Model
 Laplacian Field Model

Effect of Electrode Depth on conversion



68 mm
 140 mm
 217 mm
 312 mm
 Current Partition Model
 Laplacian Field Model

CH₄ = 0.72 kg hr⁻¹, T_{in} = 600°C, P = 1atm, ST = 1 kg h m⁻³, σ = 50 Ω . cm, ΔV = 20V

conclusions, opportunities and perspectives

- “electricity is in the catalyst”
 - reactor electrification more than connecting an electric heater to the grid
 - reasoning from the inside: ElectroThermal Fluidized Bed (ETFB) reactor
- opportunities (for endothermic reactions):
 - acid gas conversion (decomposition of COS)
 - bio-gas upcycling (CH_4 pyrolysis)
 - ...
- challenges
 - (renewable) electricity availability
 - impact on chemistry
 - ...

acknowledgments



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Interreg



Cofinancé par
l'Union Européenne
Medegefinancierd door
de Europese Unie

France - Wallonie - Vlaanderen



OBIWAN



Interreg



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l'Union Européenne
Medegefinancierd door
de Europese Unie

France – Wallonie – Vlaanderen

The OBIWAN is funded under Grant Agreement
n°0100186

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LABORATORY FOR CHEMICAL TECHNOLOGY

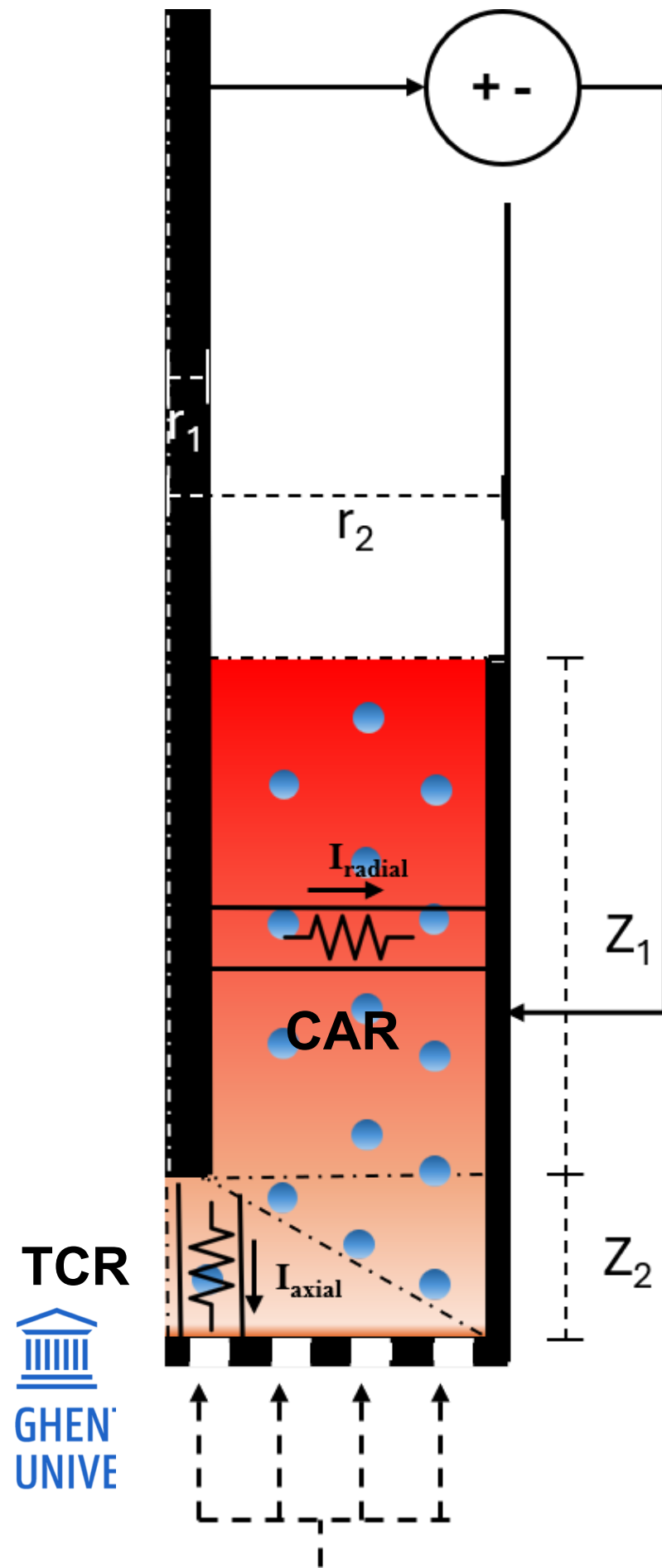
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CAR and TCR resistor formulation



- Identification of the characteristic length (L) and the cross-sectional area to the flow of the current (A_c) for both resistors

$$R = \frac{s_{bed} L}{A_c} \rightarrow L_{CAR} = r, A_{c,CAR} = 2\pi r z$$

$$\rightarrow L_{TCR} = z, A_{c,TCR} = \pi r^2$$

- Coaxial resistor

$$dR_{CAR} = \frac{s_{bed}}{2\pi dz_1} \ln\left(\frac{r_2}{r_1}\right)$$

resistances in parallel



Replace resistances expressions in the Joule heating equation

$$dQ_{JH,CAR} = \frac{V^2 2\pi dz_1}{s_{bed} \ln\left(\frac{r_2}{r_1}\right)}$$

- Truncated cone resistor

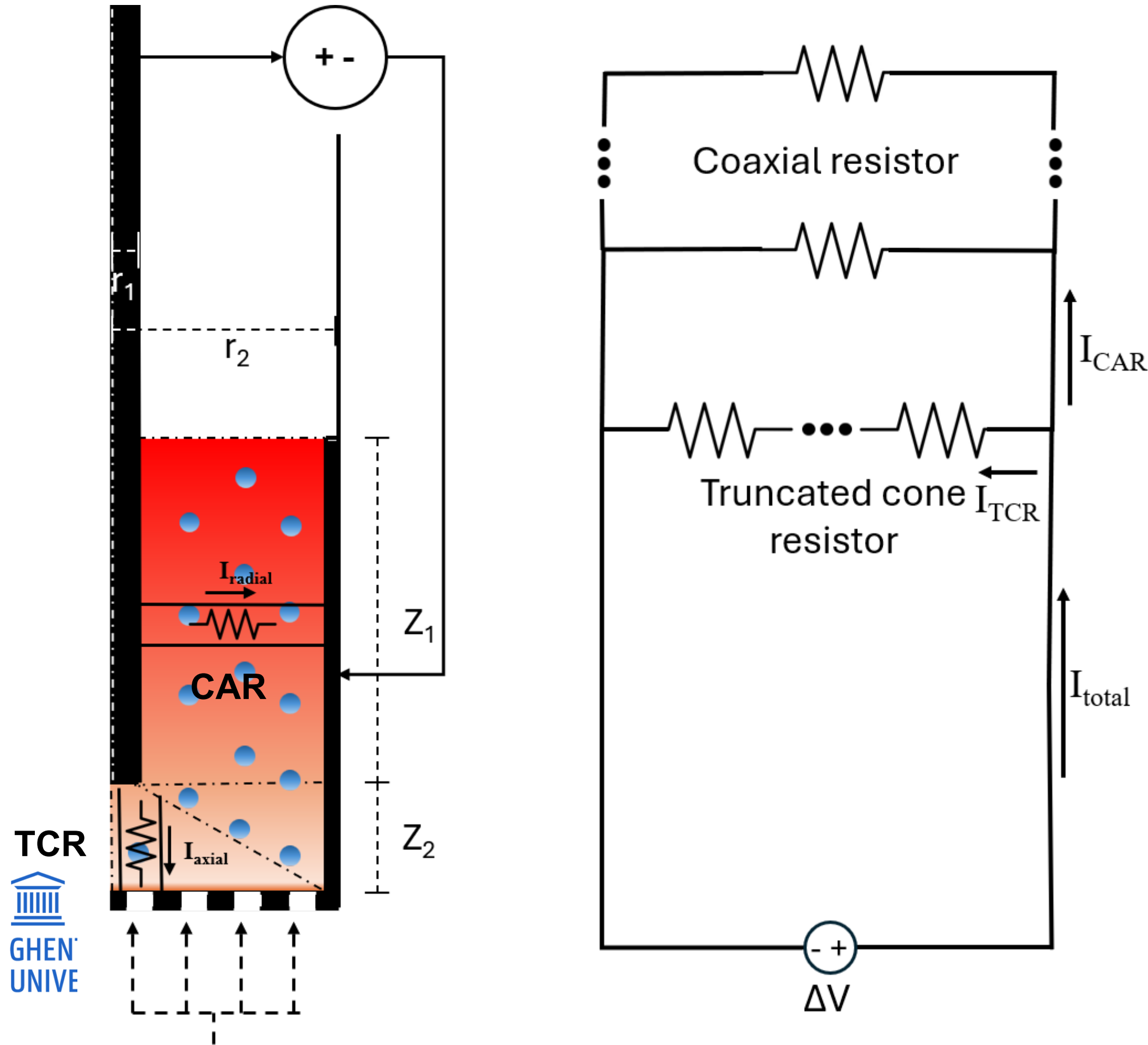
$$dR_{TCR} = \frac{s_{bed} dz_2}{\pi r^2}$$

resistances in series



$$dQ_{JH,TCR} = \frac{I_{TCR}^2 s_{bed} dz_2}{\pi r^2}$$

Dual zone electrothermal model

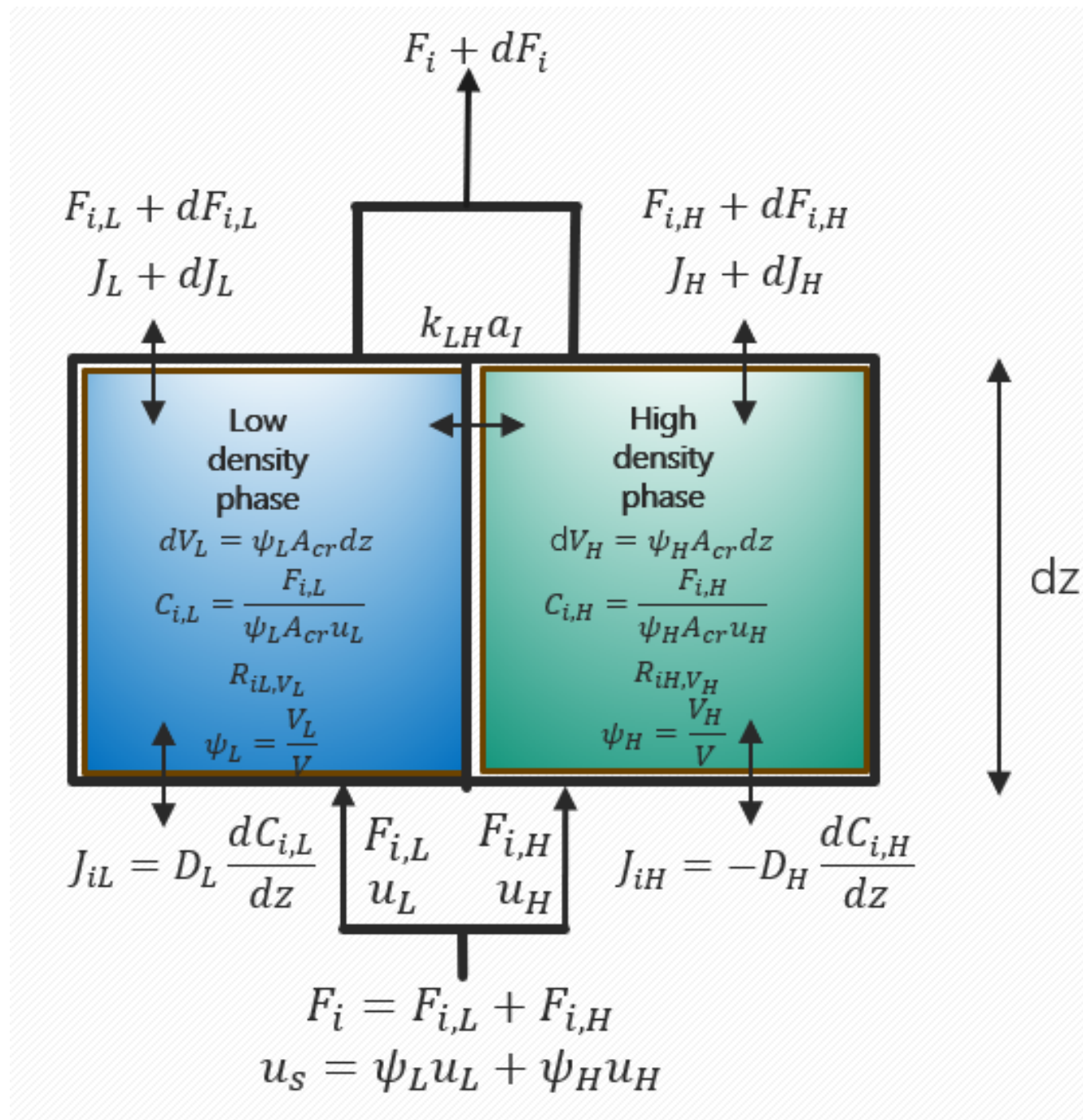


- Assuming contact resistances with the electrodes are negligible
- CARs and TCRs are represented as two resistors in parallel

$$R_{\text{total}} = \left(\frac{1}{R_{\text{CAR}}} + \frac{1}{R_{\text{TCR}}} \right)^{-1}$$

$$Q_{\text{JH},\text{total}} = \frac{V^2}{R_{\text{total}}} = I_{\text{total}}^2 R_{\text{total}} = V I_{\text{total}}$$

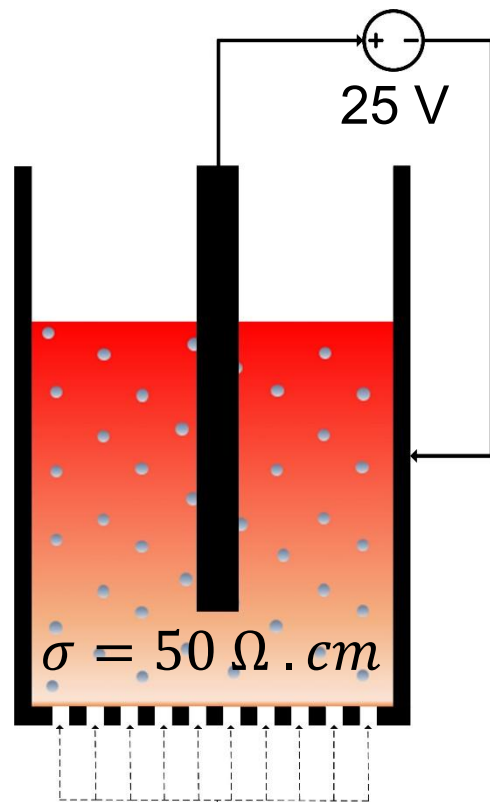
ETFBR Mass balance



Assumptions

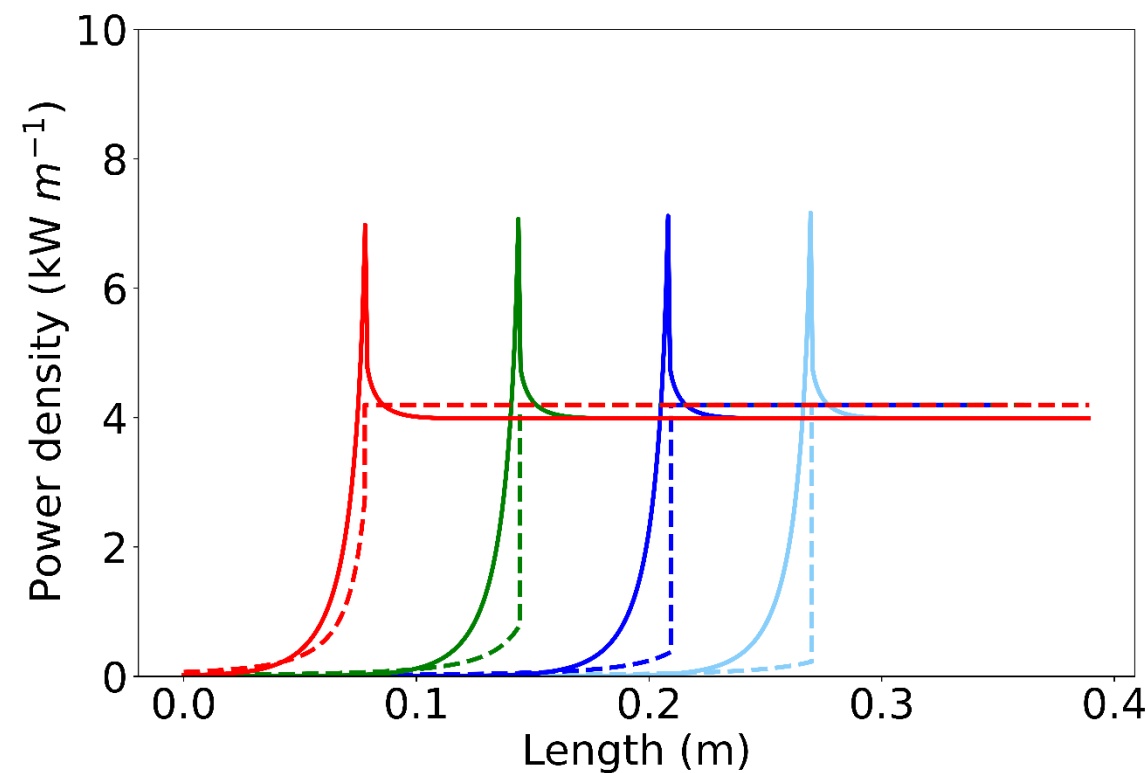
- Bed is divided into two phases
 - High density phase
 - Low density phase
- No radial profiles
- Constant properties
 - At average bed temperature and average composition
- High density phase at minimum fluidization condition
- Steady state operation
- Bubbling fluidization regime

ETFBR for CH₄ pyrolysis



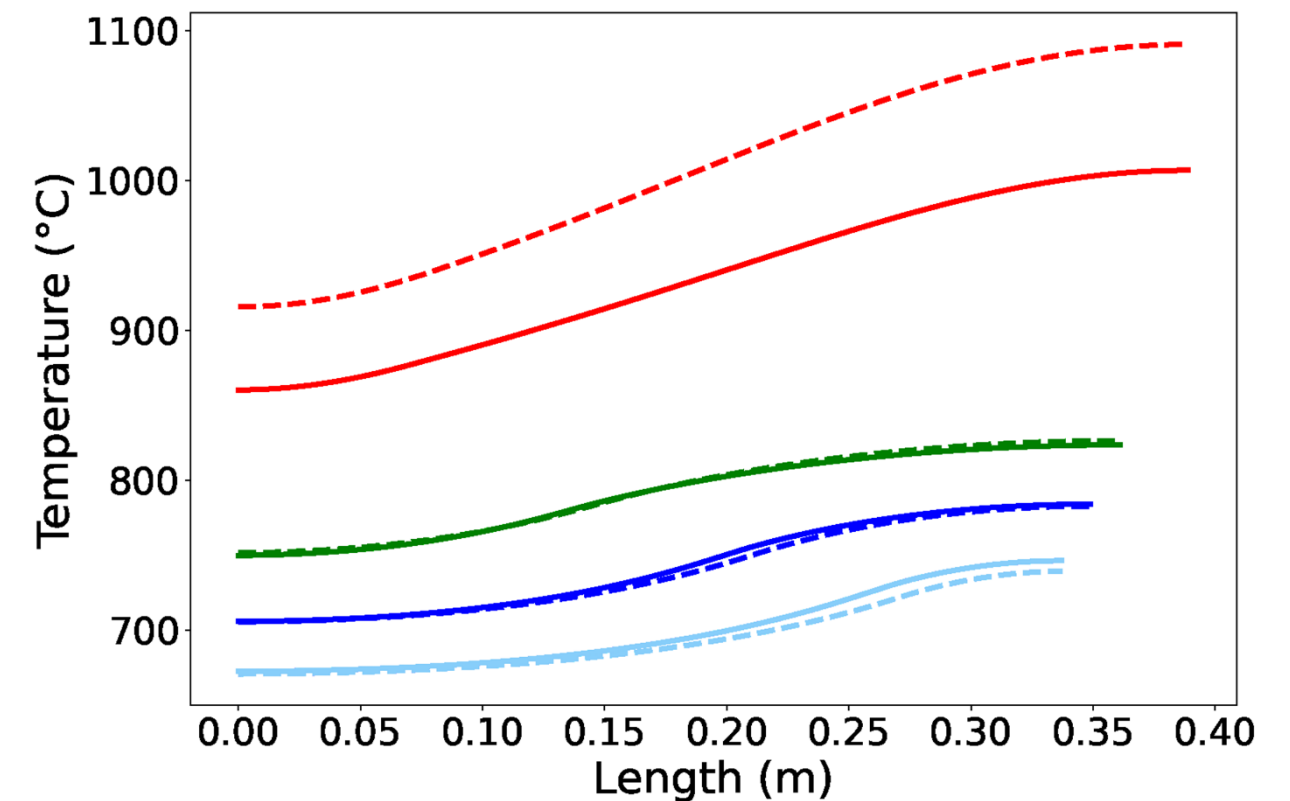
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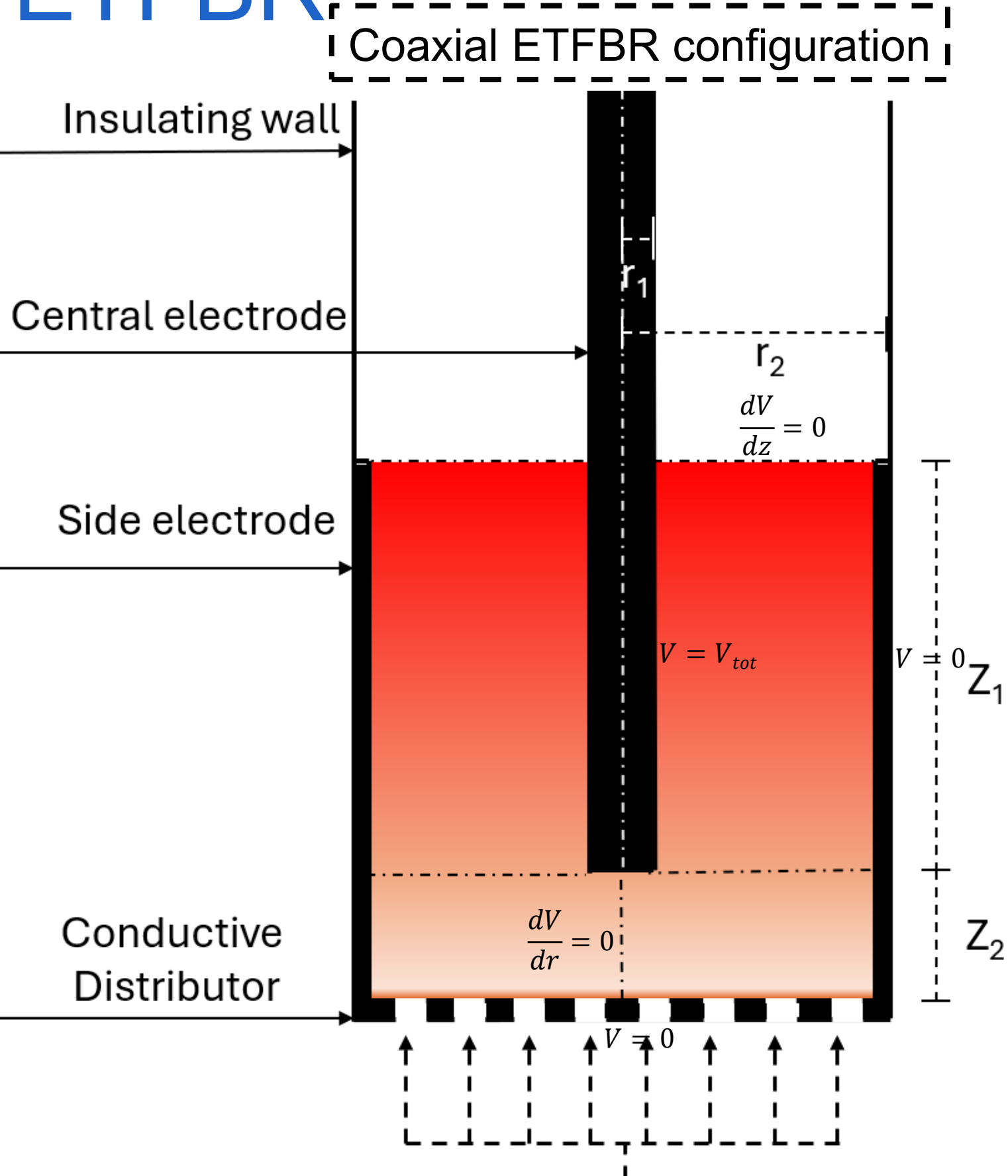
— 68 mm — 312 mm
 — 140 mm - - - - Current Partition Model
 — 217 mm — Laplacian Field Model

Effect of Electrode Depth on temperature



— 68 mm — 312 mm
 — 140 mm - - - - Current Partition Model
 — 217 mm — Laplacian Field Model

Field characteristics of the electrical properties in an ETFBR



- Electric potential field determination
 - Uniform resistivity of the bed
 - Voltage is uniform along the electrode surfaces
 - 2D Laplace equation over cylindrical coordinates

$$\frac{d^2V}{dr^2} + \frac{1}{r} \frac{dV}{dr} + \frac{d^2V}{dz^2} = 0$$

- Current density

$$J_r = -\frac{1}{s_{bed}} \frac{dV}{dr}, \quad J_z = -\frac{1}{s_{bed}} \frac{dV}{dz}$$

- Current

$$I_r = \int_0^{2\pi} \int_0^H J_r r dz d\Theta, \quad I_z = \int_0^{2\pi} \int_0^R J_z r dr d\Theta$$

- Joule heating

$$P_r = \int_0^{2\pi} \int_0^H \int_0^R -\frac{1}{s_{bed}} \left(\frac{dV}{dr} \right)^2 r dr dz d\Theta$$

$$P_z = \int_0^{2\pi} \int_0^H \int_0^R -\frac{1}{s_{bed}} \left(\frac{dV}{dz} \right)^2 r dr dz d\Theta$$

Derivation of the Coaxial Resistor Expression

- Identification of the characteristic length (L) and the cross-sectional area to the flow of the current ($A_{c,I}$)

$$R = \frac{s_{bed} L}{A_c} \rightarrow L = r, A_{c,I} = 2\pi r Z_1$$

- Derivation of the Coaxial resistor expression

$$R_{CAR} = \frac{s_{bed}}{2\pi Z_1} \ln\left(\frac{r_2}{r_1}\right) = \left(\sum_1^N \frac{1}{\frac{s_{bed}}{2\pi dz_1} \ln\left(\frac{r_2}{r_1}\right)}\right)^{-1} \overrightarrow{\text{over } dz_1} dR_{CAR} = \frac{s_{bed}}{2\pi dz_1} \ln\left(\frac{r_2}{r_1}\right)$$

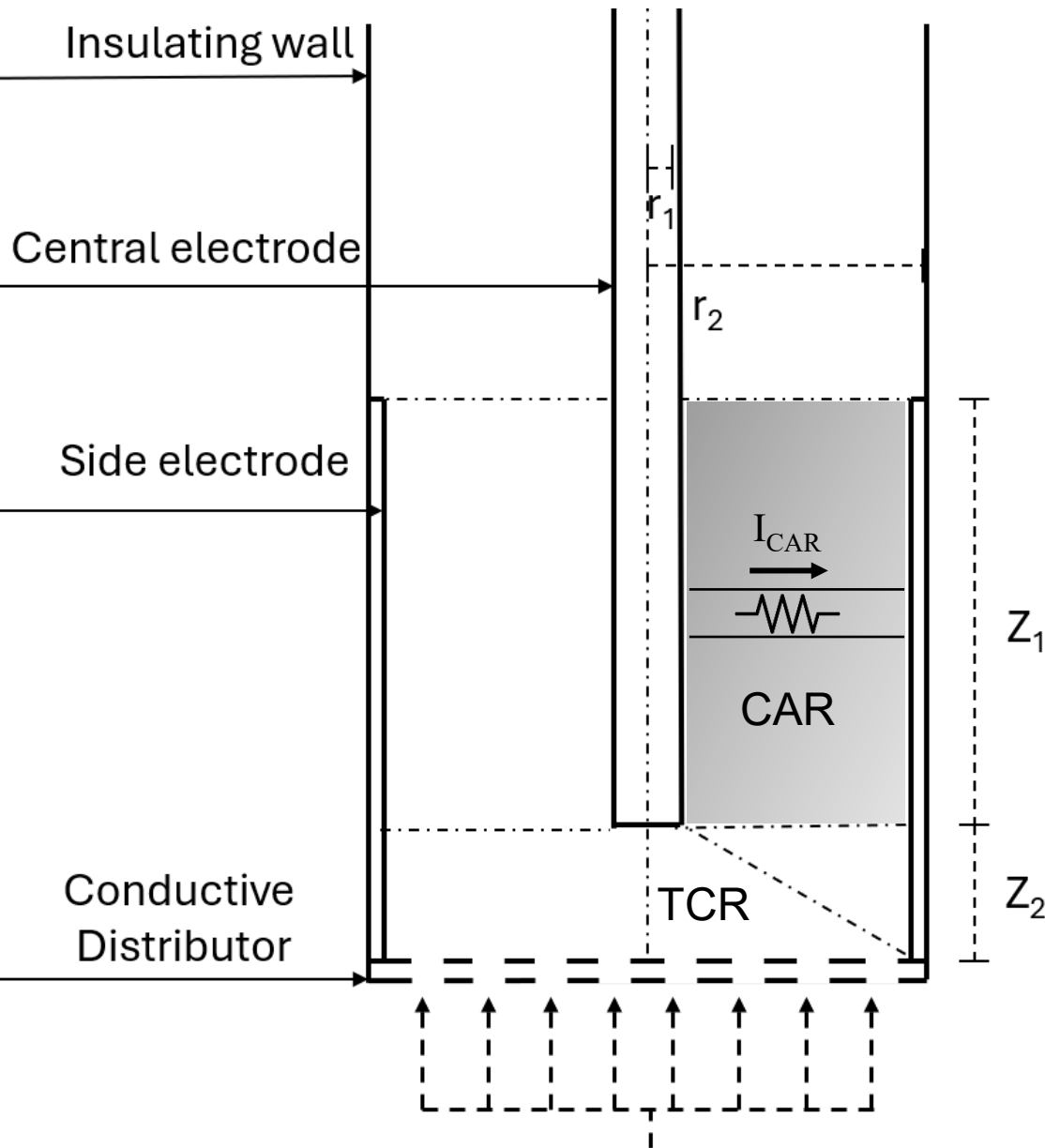
If V is constant along the axial direction → Differential resistances in the coaxial resistor can be treated as resistances in parallel



- Replace the coaxial resistance in the Joule heating equation

$$Q_{JH,CAR} = \frac{V^2}{R_{CAR}} = \frac{V^2 2\pi Z_1}{s_{bed} \ln\left(\frac{r_2}{r_1}\right)}$$

$$dQ_{JH,CAR} = \frac{V^2 2\pi dz_1}{s_{bed} \ln\left(\frac{r_2}{r_1}\right)}$$



Derivation of the Truncated Cone Resistor Expression

- Identification of the characteristic length (L) and the cross-sectional area to the flow of the current ($A_{c,I}$)

$$R = \frac{s_{bed}L}{A_c} \rightarrow L = z A_{c,I} = \pi r^2$$

- Derivation of the Coaxial resistor expression

$$R_{TCR} = \frac{s_{bed}Z_2}{\pi r_1 r_2} = \sum \frac{s_{bed}dz_2}{\pi r_1 r_2} \overrightarrow{\text{over } dz_2} dR_{TCR} = \frac{s_{bed}dz_2}{\pi r_1 r_2}$$

If I is constant along the axial direction → Differential resistances in the truncated cone resistor can be treated as resistances in series



- Replace the truncated cone resistance in the Joule heating equation

$$Q_{JH,TCR} = I_{TCR}^2 R_{TCR} = \frac{I_{TCR}^2 s_{bed} Z_2}{\pi r^2}$$

$$dQ_{JH,TCR} = \frac{I_{TCR}^2 s_{bed} dz_2}{\pi r^2}$$

