Design and Optimization of Chemical Looping Combustion Processes

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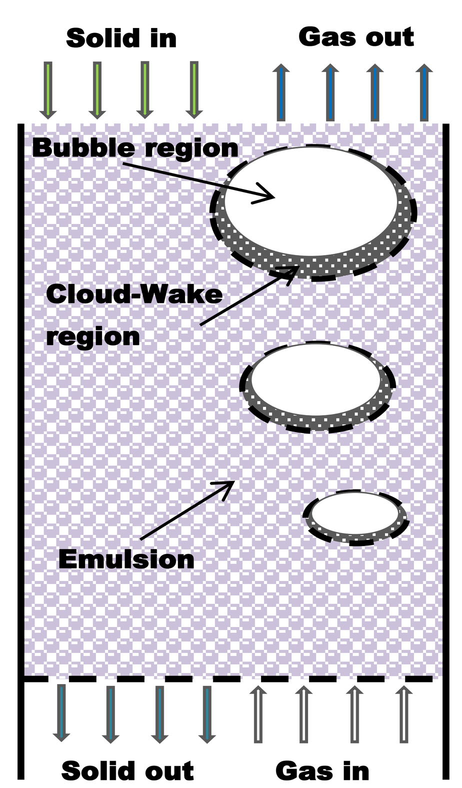
Chemical Looping Combustion (CLC)

- Advanced combustion system with inherent CO_2 separation
- Potential cost savings compared with conventional power technologies with CO_2 capture systems
- A fuel reactor is coupled to an air reactor with a solid oxygen carrier (OC) undergoing reduction-oxidation reactions as it circulates between the reactors
- Models must capture both micro and macro-scale phenomena
  - Micro-scale phenomena i.e. the effect of OC particle characteristics on reaction, mass and heat transfer
  - Macro-scale phenomena i.e. effect of reactor geometry, and OC circulation rate on process operating and capital costs

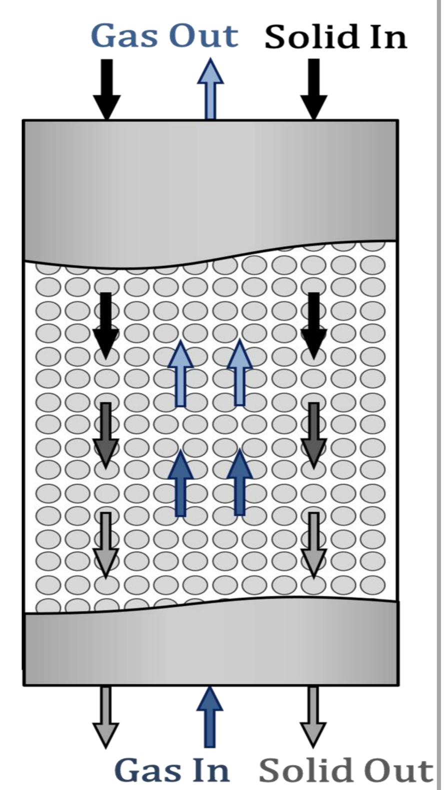
IDAES Advanced Models – Solid/Fluid Contactors



Bubbling Fluidized Bed

Model Features

- Captures multi-scale phenomena
- System of PDAEs
- First-principles mass & energy balance
- Empirical momentum balance
- Heat & mass transfer correlations
- Modular property implementation



Moving Bed

CLC Property Package

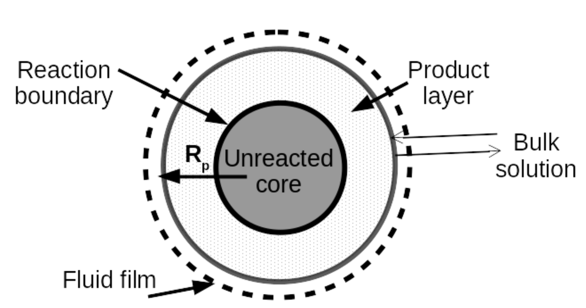
Gas phase properties – diffusion, viscosity, thermal conductivity

Solid phase properties – particle size distribution, density, sphericity, minimum fluidization velocity & porosity, terminal velocity, thermal conductivity

Thermodynamics – heat of formation, heat capacity, equation of state

Kinetics – Shrinking core kinetic model for spherical grain geometry [1]

$$\frac{dX_s}{dt} = \frac{3bkC_g^n}{\rho_m r_g}(1 - X_s)^{2/3}$$



1. Abad A., et al., Mapping of the range of operational conditions for Cu-, Fe-, and Ni-based oxygen carriers in CLC, Chem. Eng. Sci. 62 (2007) 533–549.

Disclaimer

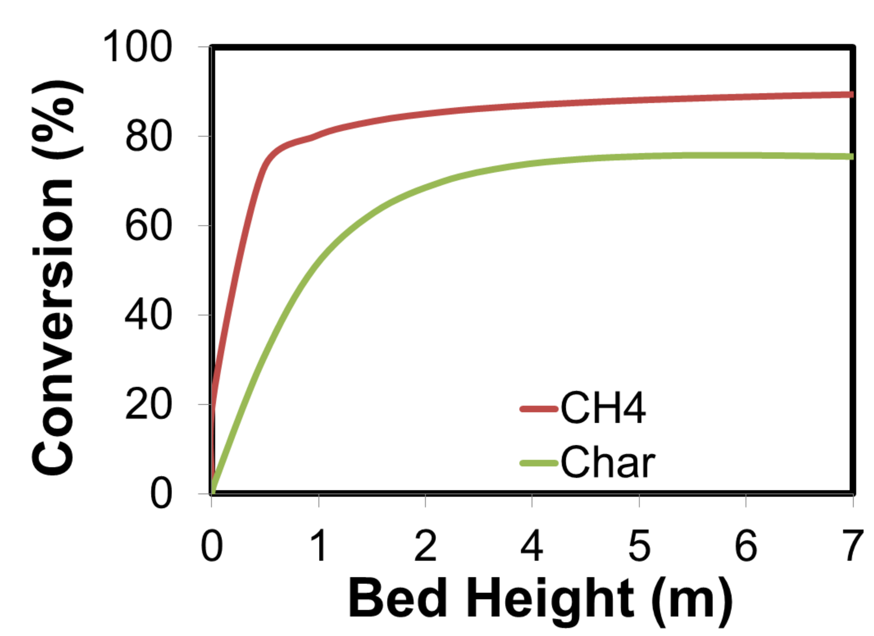
Bubbling Fluidized Bed Case Study

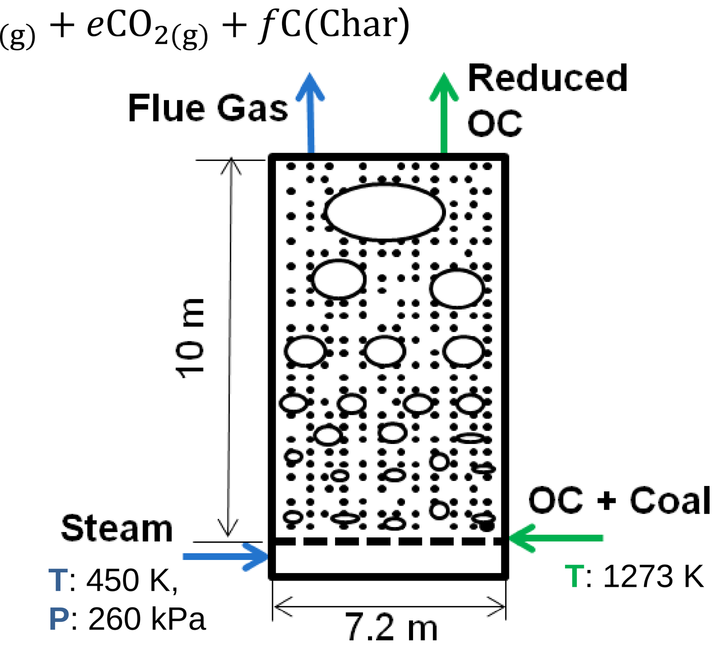
Design of a coal-fed BFB-CLC fuel reactor

- OC (922 kg/s): Fe_2O_3/Al_2O_3
- Fuel (3.4 kg/s): Illinois no. 6 coal – LHV = 29.5 MJ/kg
- Fluidizing gas (202 mol/s): steam
- **Key challenge**: modeling of multiple solid particles (OC and coal)

Design heuristics[2] and model assumptions

- Well mixed OC and coal feed
  - OC >> coal - OC properties used for hydrodynamic calculations
- Reactions – Coal devolatilization, OC reduction, char gasification, water gas shift
- Coal devolatilization occurs instantaneously at coal feed point
  - Products are calculated using Merrick's yield model[3]
  - $\mathrm{Coal}_{(s)} \rightarrow a\mathrm{CO}_{(g)} + b\mathrm{H}_{2(g)} + c\mathrm{CH}_{4(g)} + d\mathrm{H_2O}_{(g)} + e\mathrm{CO}_{2(g)} + f\mathrm{C(Char)}$

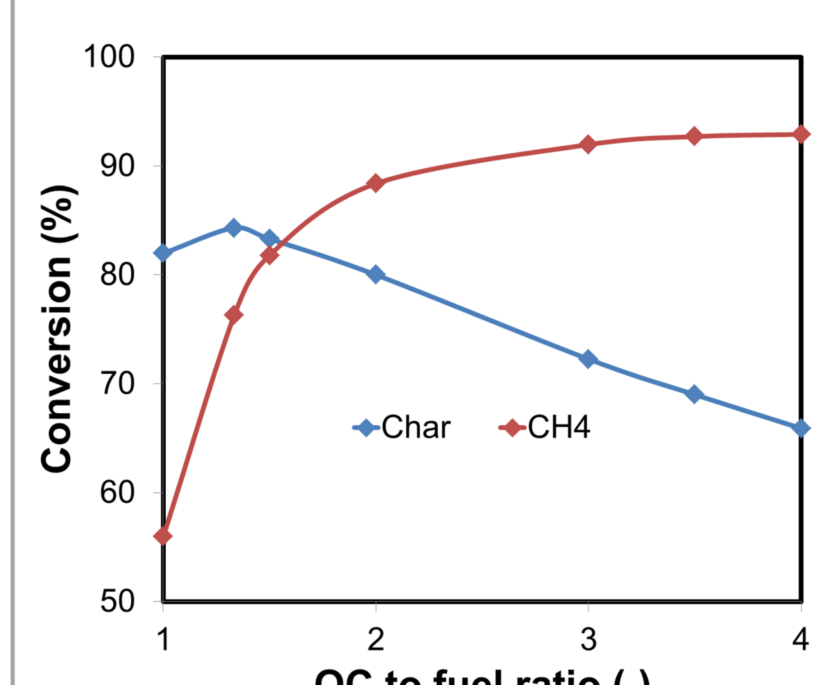




Species	OC(s)	Char(s)	CH_4(g)	CO(g)	H_2(g)
% Conversion	45	76	91	98	100

Sensitivity Analysis Results

Parameter	Variation	Char Conv. (%)	CH4 Conv. (%)	OC Conv. (%)
Bed Height	12.5 m (+25%)	79.3	91.1	46.4
(10 m)	7.5 m (-25%)	70.1	90.0	43.0
Bed Diameter	8.64 m (+20%)	86.3	95.3	49.3
(7.2 m)	5.76 m (-20%)	60.7	82.5	39.1
OC to fuel	3.5	69.0	92.7	30.5
ratio (2.5)	1.5	83.3	81.8	78.3



Trade-off between char(s) and CH_4(g) conversion for a given OC to fuel ratio

Increase in OC to fuel ratio decreases the residence time of char while increasing the contact between the OC and CH_4

2. Okoli C., et al., 2019, A framework for the optimization of chemical looping combustion processes, J. Pow. Tech. *In Press*.
3. R. Monaghan and A. Ghoniem, 2012, A dynamic reduced order model for simulating entrained flow gasifiers Part I: Model development and description, Fuel, 91, 61 – 80.

Moving Bed Case Study

Optimal design of a natural gas CLC process using interconnected MB reactors, with three different oxygen carriers.

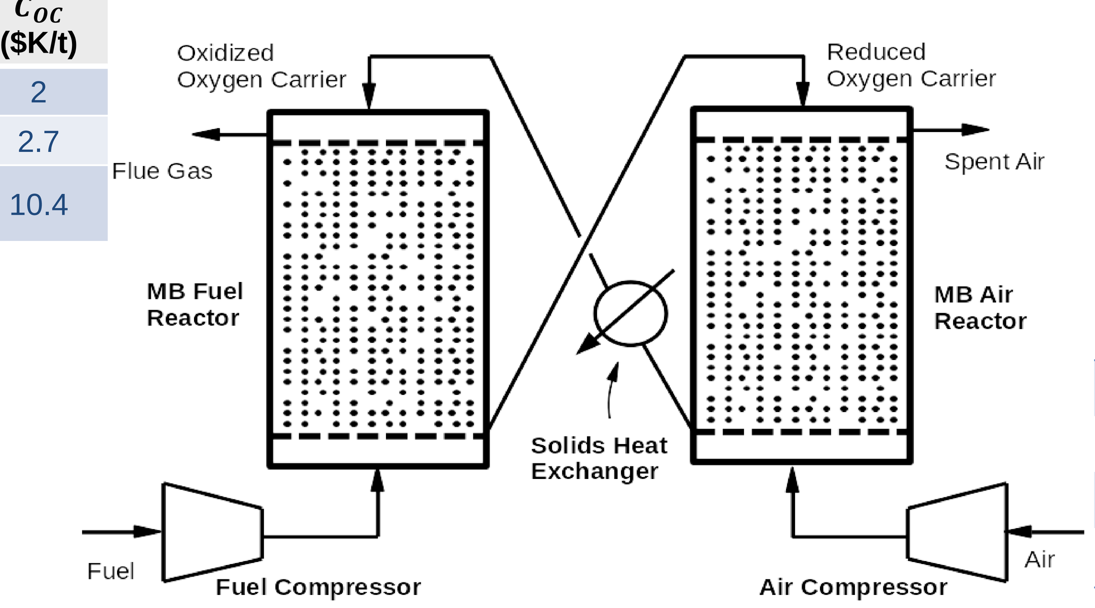
Objective: Minimize TAC of CLC process for varying heat production

Decision Variables

- Size of beds (diameter, height)
- Oxygen carrier circulation rate
- Solids inlet temperature
- Air flow rate

Oxygen Carrier	R_O	C_{OC} ($K/t)
Fe_2O_3/Fe_3O_4	0.03	2
CuO/Cu	0.20	2.7
NiO/Ni	0. 21	10.4

F_g (mol/s)	128.2
y_{CH4}	0.975
y_{CO2}	0.025
P (bar)	1.37
T_{gas} (K)	293



y_{O2}	0.21
y_{N2}	0.79
P (bar)	1.37
T_{gas} (K)	293

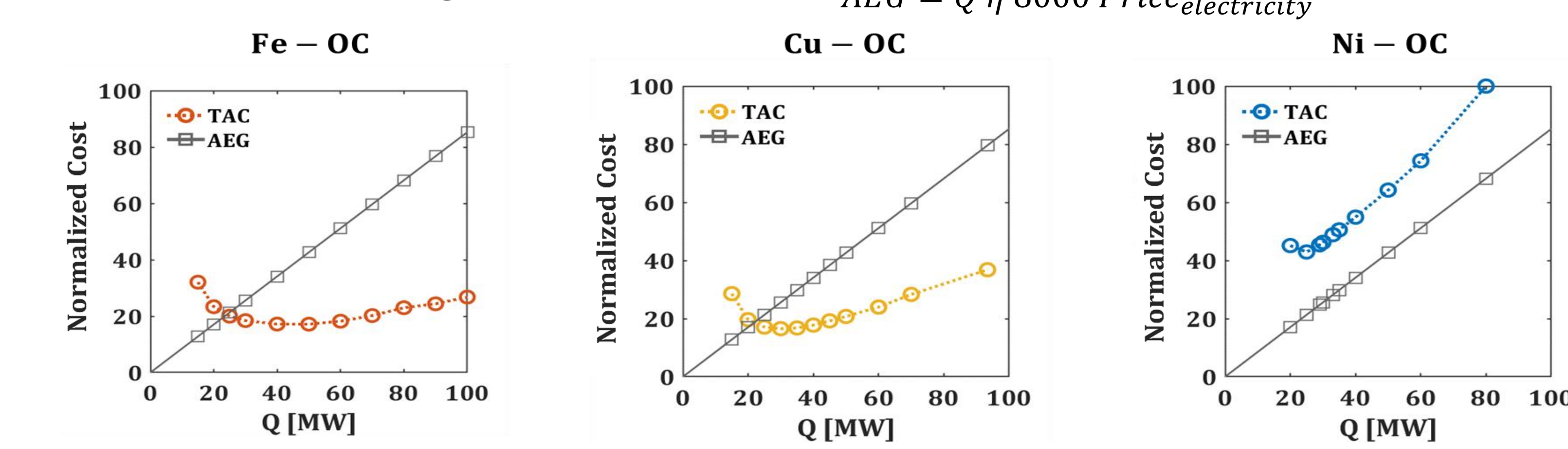
Flowsheet of interconnected Moving Bed CLC reactors

MB-CLC Results

Different values of Q_{target} [MW]

AEG ($/yr): annual electricity generated

$AEG = Q\,\eta\,8000\,Price_{electricity}$



Normalized TAC and AEG (w.r.t. $\max \mathrm{TAC}_{\mathrm{Ni-OC}}$) vs. usable heat from CLC system.

Optimal results for $Q_{target} = 40\ MW$	Fe - OC		Cu - OC		Ni - OC	
	FR	AR	FR	AR	FR	AR
Design decision variables						
Reactor bed diameter (m)	5.7	10.9	6.4	12.8	4.4	11.3
Reactor bed height (m)	3.3	2.4	1.8	2.6	0.9	5.8
Operating decision variables						
OC circulation rate (kg/s)	554		409		168	
Air flowrate (mol/s)		1182		1253		1660

Concluding Remarks

- Solve time: 22 s for BFB simulation case; 180 – 300 s for most MB optimization cases
- Pyomo.DAE provides various options for model discretization:
  - Orthogonal collocation scheme used but finite difference methods also applicable
- Models are optimization ready, and flexible for using property packages of different OCs
- IDAES enables fast solution of **rigorous models** that are suitable for both simulation and optimization
  - Can be leveraged to **accelerate CLC development** by rapidly screening alternative OC and reactor configurations

Future work

- Ongoing collaboration with Ohio State University to validate the models with pilot-scale data
- Focus in the future will be on optimizing a complete CLC system for power production
  - Including a complete steam system
  - Also considering a detailed techno-economic analysis
- Ongoing work on developing dynamic models of the reactors