Modelling of an Entrained Flow CaL Carbonator for CO_2 Capture in Cement Kilns

7th High Temperature Solid Looping Cycles Network Meeting
Luleå, Sweden, 4-5th September 2017

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Outline

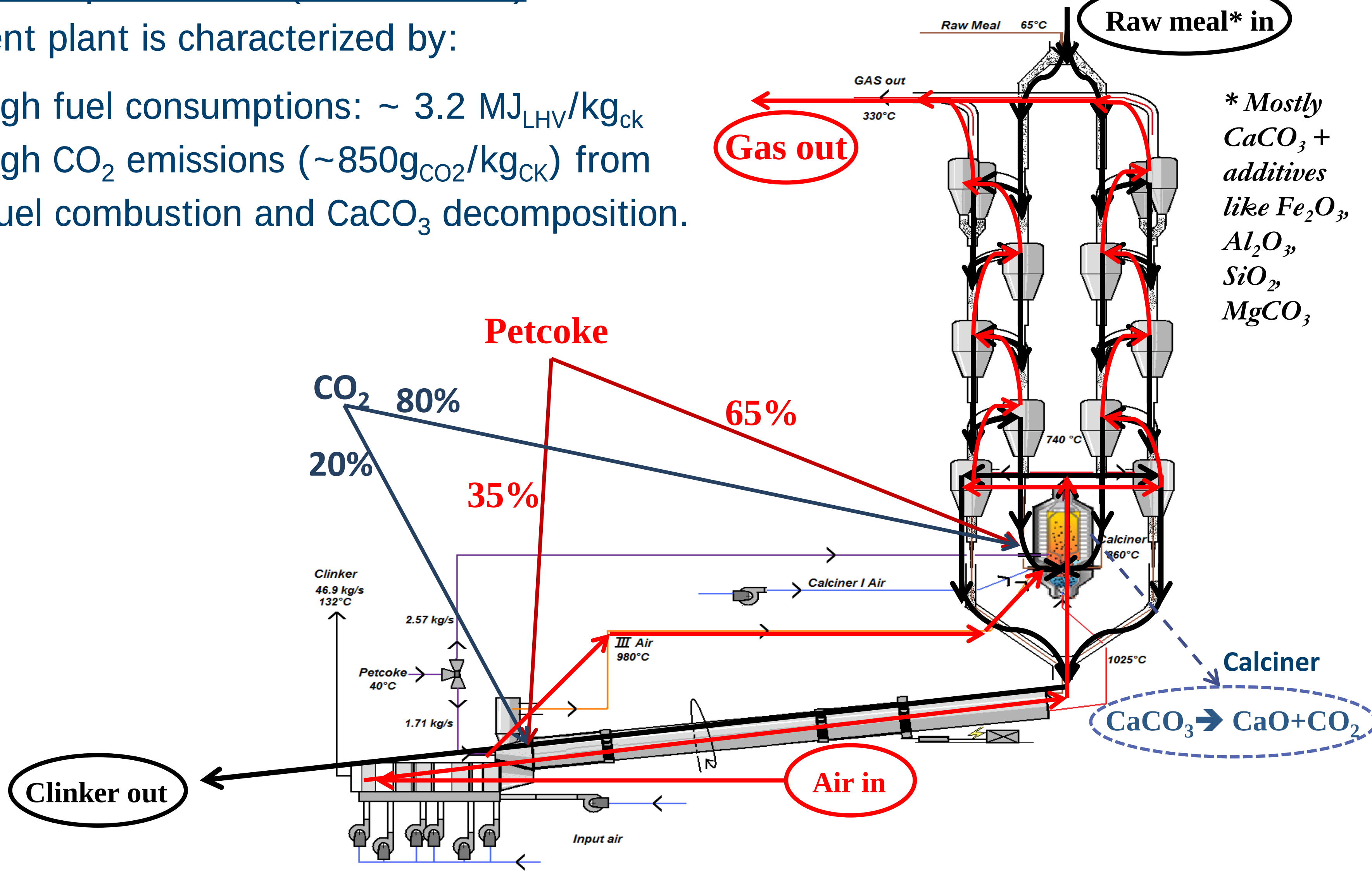
- Overview of CaL technology in cement plant:
  - Tail end
  - Integrated with EF carbonator
- 1D model of EF carbonator:
- Results of first sensitivity analysis
- Conclusion

Cement plant and related model

Reference plant ➔ GS (POLIMI code)

Cement plant is characterized by:

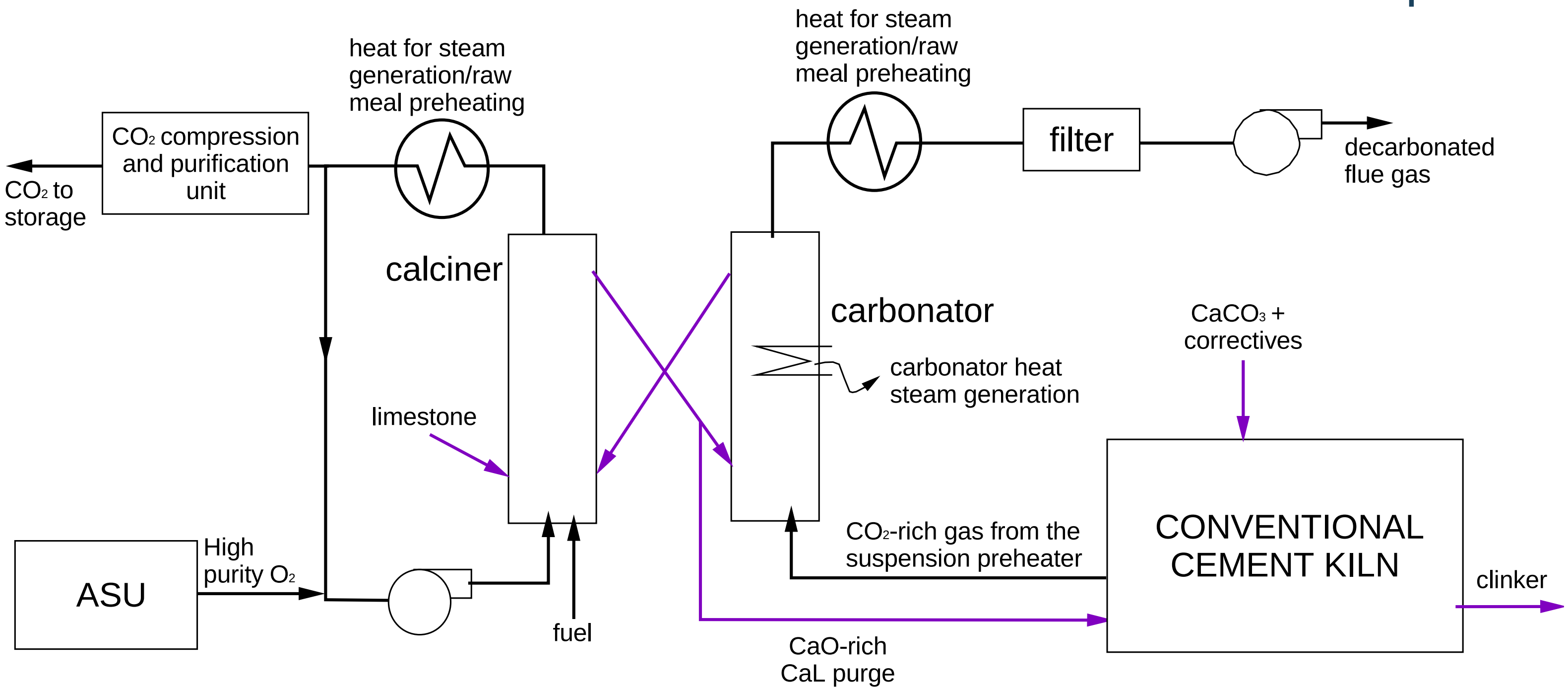
- ✓ high fuel consumptions: ~ 3.2 MJ_{LHV}/kg_{ck}
- ✓ high CO_2 emissions (~850g_{CO2}/kg_{CK}) from fuel combustion and $CaCO_3$ decomposition.



* Mostly $CaCO_3$ + additives like Fe_2O_3, Al_2O_3, SiO_2, $MgCO_3$

Tail-end CaL configuration

- Carbonator removes CO_2 from cement plant flue gas → highly suitable for retrofit
- CaO-rich purge from CaL calciner used as feed for the cement kiln
- CFB CaL reactors: d_{50}=100-250 µm
  Particle size for clinker production d_{50}=10-20 µm } → CaL purge milled in the raw mill at low temperature

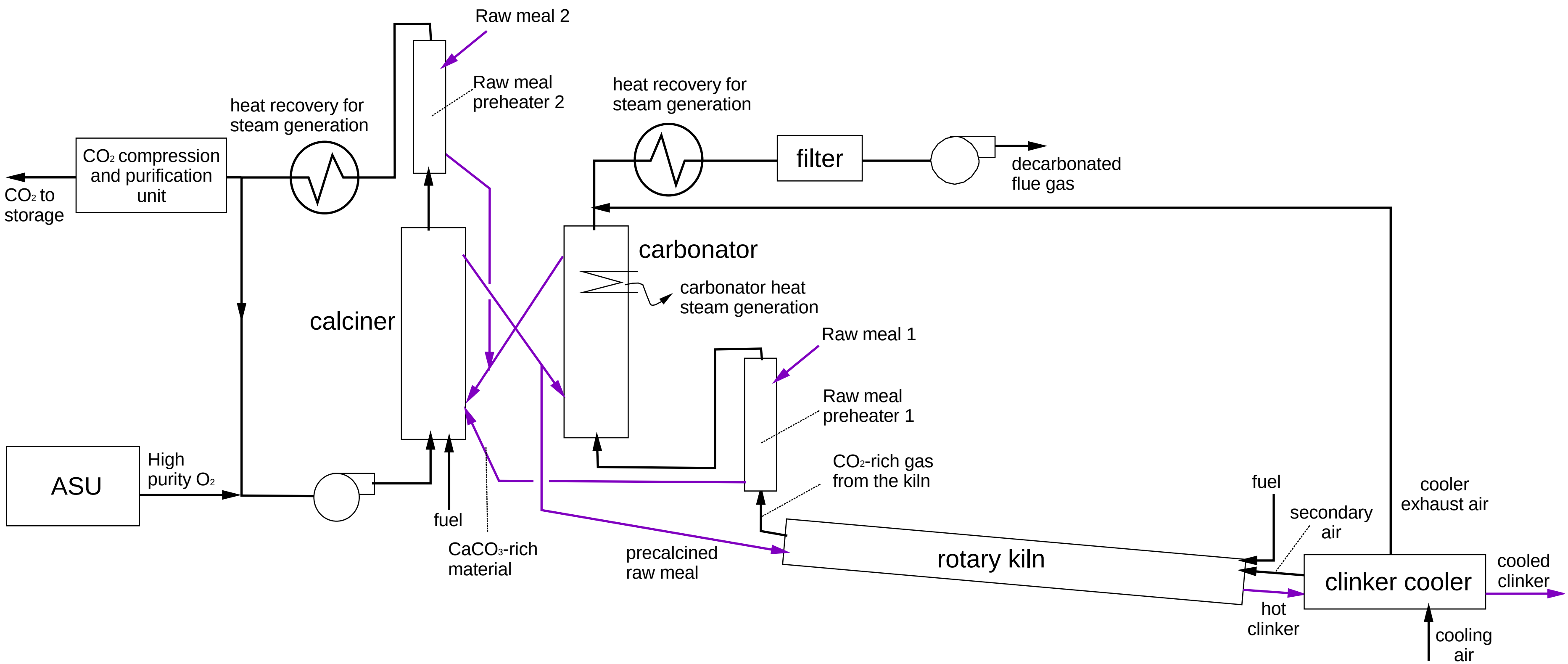


1) Spinelli et al., "**Integration of Ca-Looping systems for CO_2 capture in cement plants**" GHGT13 conference paper, Energy Procedia 114 (2017) 6206 – 6214

2) De Lena et al., "**Process integration study of tail-end Ca-Looping process for CO_2 capture in cement plants**" Submitted to **International Journal of Greenhouse Gas Control**

Integrated CaL configuration

- CaL carbonator highly integrated within the preheating tower, on rotary kiln gas
- CaL calciner coincides with the cement kiln pre-calciner
- Calcined raw meal as CO_2 sorbent in the carbonator
- Sorbent has small particle size (d_{50}=10-20 μm) → entrained flow reactors



*Spinelli et al., "**Integration of Ca-Looping systems for CO_2 capture in cement plants**"*
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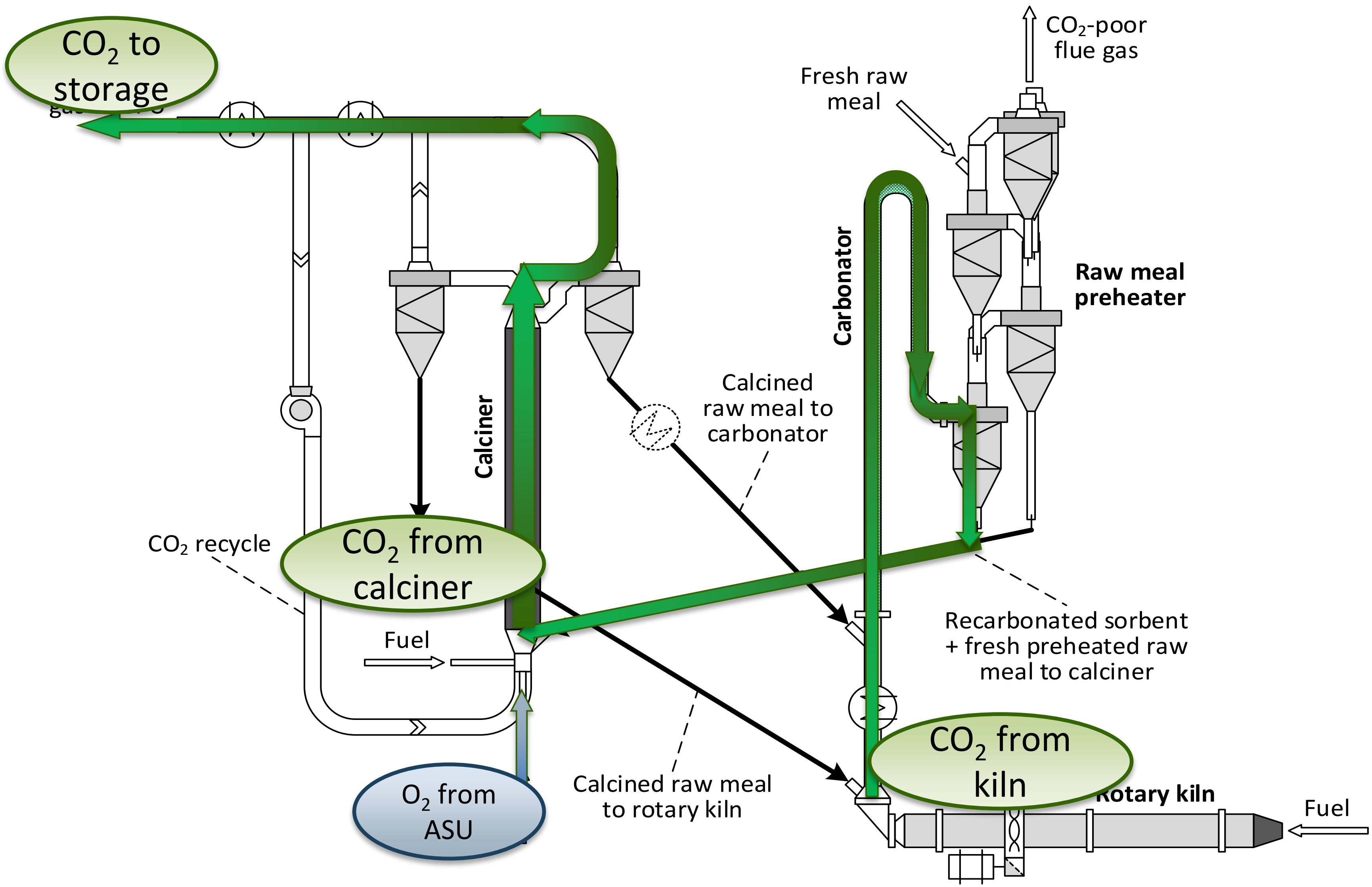
Integrated CaL concept: entrained flow (EF) reactors



Integrated / Tail End CaL concepts: results

	Reference cement plant w/o CO_2 capture	Tail-end CaL configuration	Integrated CaL configuration
Direct CO_2 emissions [kg_{CO2}/t_{clk}]	863.1	143.2	**71.4**
Indirect CO_2 emissions [kg_{CO2}/t_{clk}]	105.2	-123.5	**128.7**
Equivalent CO_2 emissions [kg_{CO2}/t_{clk}]	968.3	19.7	**200.1**
Equivalent CO_2 avoided [%]	--	98.0	**79.3**
SPECCA [MJ_{LHV}/kg_{CO2}]	**--**	**3.26**	**2.32**

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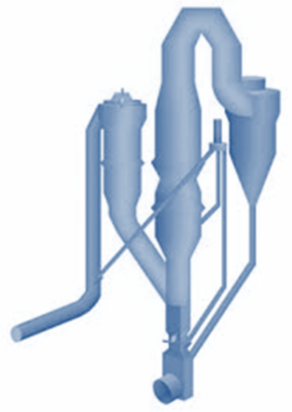
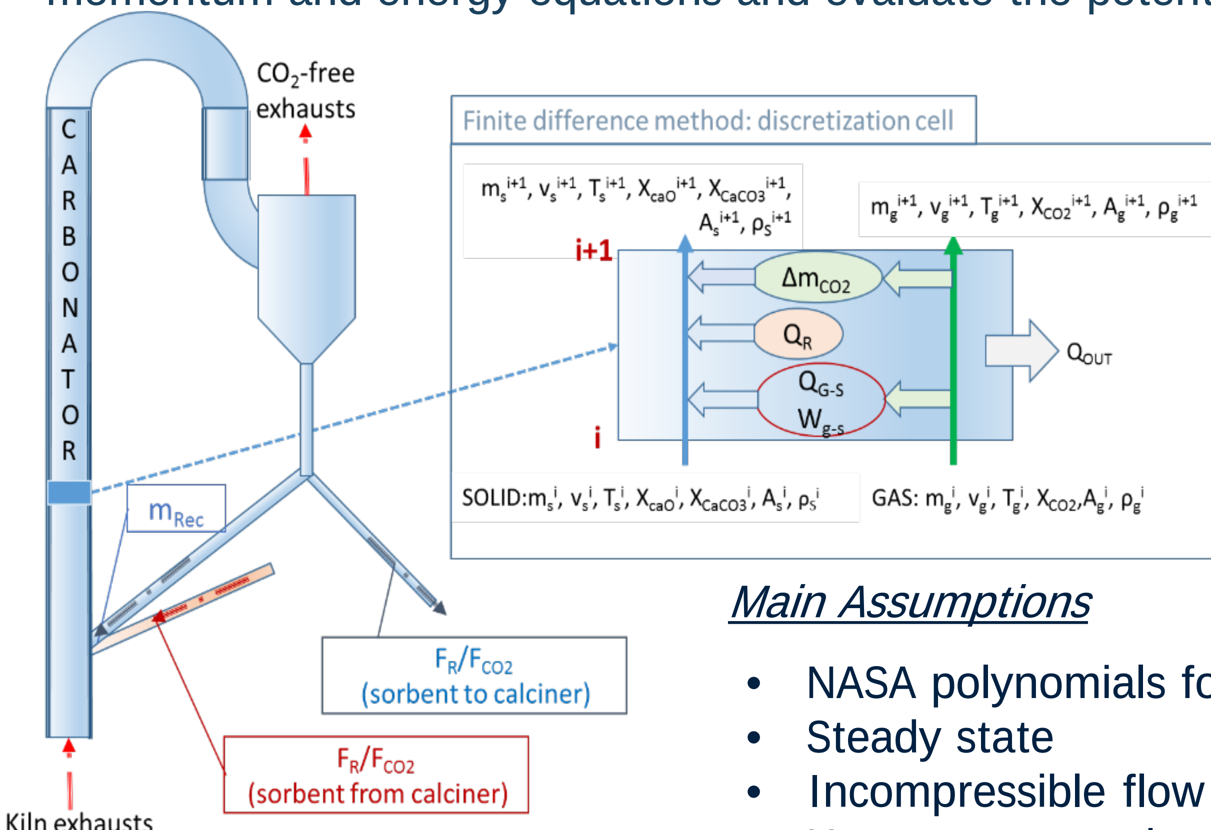
Entrained flow carbonator model



Entrained flow CaL carbonator modeling

Dilute reactor is the most suitable option for the cement plant CaL application, because of the **experience** with entrained flow technologies and the **low particle size**.

A simple, finite-difference model (axial discretization) has been developed to solve mass, momentum and energy equations and evaluate the potential CO_2 capture rate.



- CaL kinetics
- Gas-solid drag ➔ velocities
- Interphase heat transfer
- External heat transfer
- Pressure losses
- Fluid-dynamic check
- Internal sorbent recycle

Main Assumptions

- NASA polynomials for gas /solid TDN properties
- Steady state
- Incompressible flow
- Homogeneous mixtures
- Mass transfer effect neglected (low *Da* numbers)

EF carbonator modeling – Equations

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Mass

$$1-Gas: \quad \frac{d\dot{m}_g}{dx} = -\frac{\dot{M}_{s,a}}{u_s} \cdot \frac{dX}{dt} \cdot M_{CO2} \qquad 2-Solid: \quad \frac{d\dot{m}_s}{dx} = -\frac{d\dot{m}_g}{dx}$$

Carbonation kinetics

Random Pore model with X_{max} ***imposed***

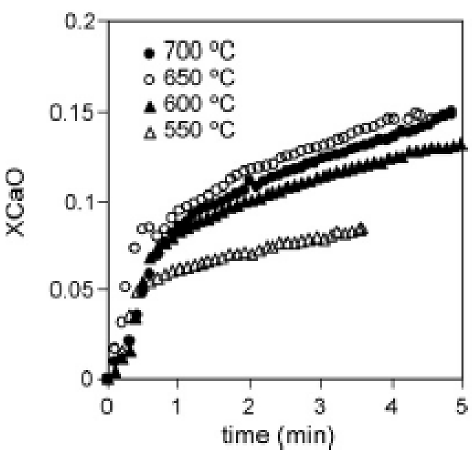
Temperature influence

X_{max} influence

$$CaO_{(s)} + CO_{2(g)} \xrightarrow{\Delta h+} CaCO_{3(s)}$$

$$\frac{dX}{dt} = \frac{k_s \cdot S_N}{(1-\varepsilon_P)} \cdot (1-X) \cdot \left(C_{CO_2} - C_{CO_2,eq}\right) \cdot \sqrt{1-\Psi\, ln(1-X)}$$

$$k_s = k_{s,0} \cdot \exp\frac{-20300}{T_R \cdot R_u} \qquad S_N = S_0 \cdot X_{max}$$



G. Grasa et al., *"**Application of the random pore model to the carbonation cyclic reaction**", AIChE J. 55 (2009)*

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Mass

1 – *Gas*: $\frac{d\dot{m}_g}{dx} = -\frac{\dot{M}_{s,a}}{u_s} \cdot \frac{dX}{dt} \cdot M_{CO2}$

2 – *Solid*: $\frac{d\dot{m}_s}{dx} = -\frac{d\dot{m}_g}{dx}$

Momentum

3-*Gas:* $\frac{d(\dot{m}_g \cdot u_g)}{dx} + A \cdot \frac{dp}{dx} = -I_G \cdot A_g \cdot \rho_g \cdot g - F_{fg} - F_{gs}$

4 – *Solid*: $\frac{d(\dot{m}_s \cdot u_s)}{dx} = -I_G \cdot A_s \cdot \rho_s \cdot g - F_{fs} + F_{gs}$

Energy

5 – *Gas*: $\frac{d\left(\dot{m}_g \cdot h_g + 0.5 \cdot \dot{m}_g \cdot u_g^2 + I_G \cdot \dot{m}_g \cdot g \cdot x\right)}{dx} = -\dot{w}_{gs} - \dot{q}_{gw} - \dot{q}_{gs} - \dot{q}_{CO2,carb}$

6 – *Solid*: $\frac{d(\dot{m}_s \cdot h_s + 0.5 \cdot \dot{m}_s \cdot u_s^2 + I_G \cdot \dot{m}_s \cdot g \cdot x)}{dx} = \dot{w}_{gs} - \dot{q}_{sw} + \dot{q}_{gs} + \dot{q}_{r,carb} + \dot{q}_{CO2,carb}$

Complementary correlations

$$\dot{Q}_g^i = -\dot{Q}_s^i = N_s \cdot A \cdot \pi \cdot D_p^2 \cdot h_{gp} \cdot \Delta T_{ml,g-s}$$

1) Gas/solid heat transfer: several correlations evaluated:

$$Nu_P = \frac{h_{gp} \cdot D_p}{k_g} = a_1 \cdot Re_P{}^{a_2} \cdot Pr_P{}^{a_3}$$

Gas/limestone system in pneumatic transport regime

$$Nu_p = Nu_{pL,CaCO_3} \cdot 0.1374\ \Phi_s^{-0.8243} + 0.15 \cdot Re_p$$

Evaluation of limiting Nusselt number as a function of particle nature and solid loading

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Mass

$$1 - Gas: \quad \frac{d\dot{m}_g}{dx} = -\frac{\dot{M}_{s,a}}{u_s} \cdot \frac{dX}{dt} \cdot M_{CO2} \qquad 2 - Solid: \quad \frac{d\dot{m}_s}{dx} = -\frac{d\dot{m}_g}{dx}$$

Momentum

$$3\text{-}Gas: \quad \frac{d(\dot{m}_g \cdot u_g)}{dx} + A \cdot \frac{dp}{dx} = -I_G \cdot A_g \cdot \rho_g \cdot g - F_{fg} - F_{gs}$$

$$4 - Solid: \quad \frac{d(\dot{m}_s \cdot u_s)}{dx} = -I_G \cdot A_s \cdot \rho_s \cdot g - F_{fs} + F_{gs}$$

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$$6 - Solid: \quad \frac{d(\dot{m}_s \cdot h_s + 0.5 \cdot \dot{m}_s \cdot u_s^2 + I_G \cdot \dot{m}_s \cdot g \cdot x)}{dx} = \dot{w}_{gs} - \dot{q}_{sw} + \dot{q}_{gs} + \dot{q}_{r,carb} + \dot{q}_{CO2,carb}$$

Complementary correlations

2) Gas/wall heat transfer – consider the external heat transfer improvement in dilute suspension flows

$$Nu_g = \frac{h_{gw,0} \cdot D}{k_g} = 0.023 \cdot Re^{0.8} \cdot Pr^{0.3} \qquad \frac{h_{gw}}{h_{gw,0}} = 1 + 4 \cdot Re^{-0.32} \cdot \frac{\dot{m}_s}{\dot{m}_g} \cdot \frac{c_{p,s}}{c_{p,g}}$$

"Analysis and correlation of heat-transfer coefficient and friction factor data for dilute gas-solid suspensions", NASA technical note, 1966

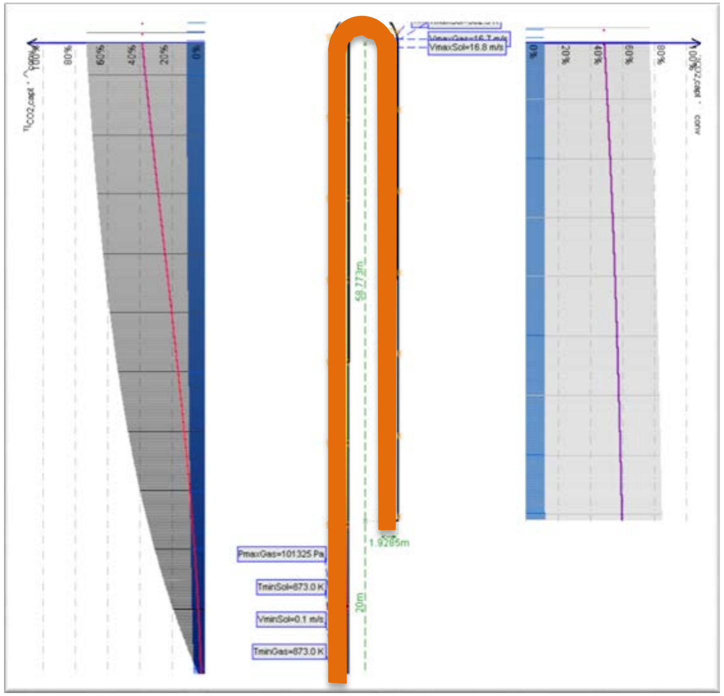
The higher is the solid loading, the higher the heat transfer coefficient (especially at low Reynolds)

Methodology and scope

1) Definition of reactor boundary conditions and simplified design;
2) Calculation of the CO_2 capture rate as a function of kinetic models;
3) Identification of the most promising operating parameters.

Simulation assumptions:

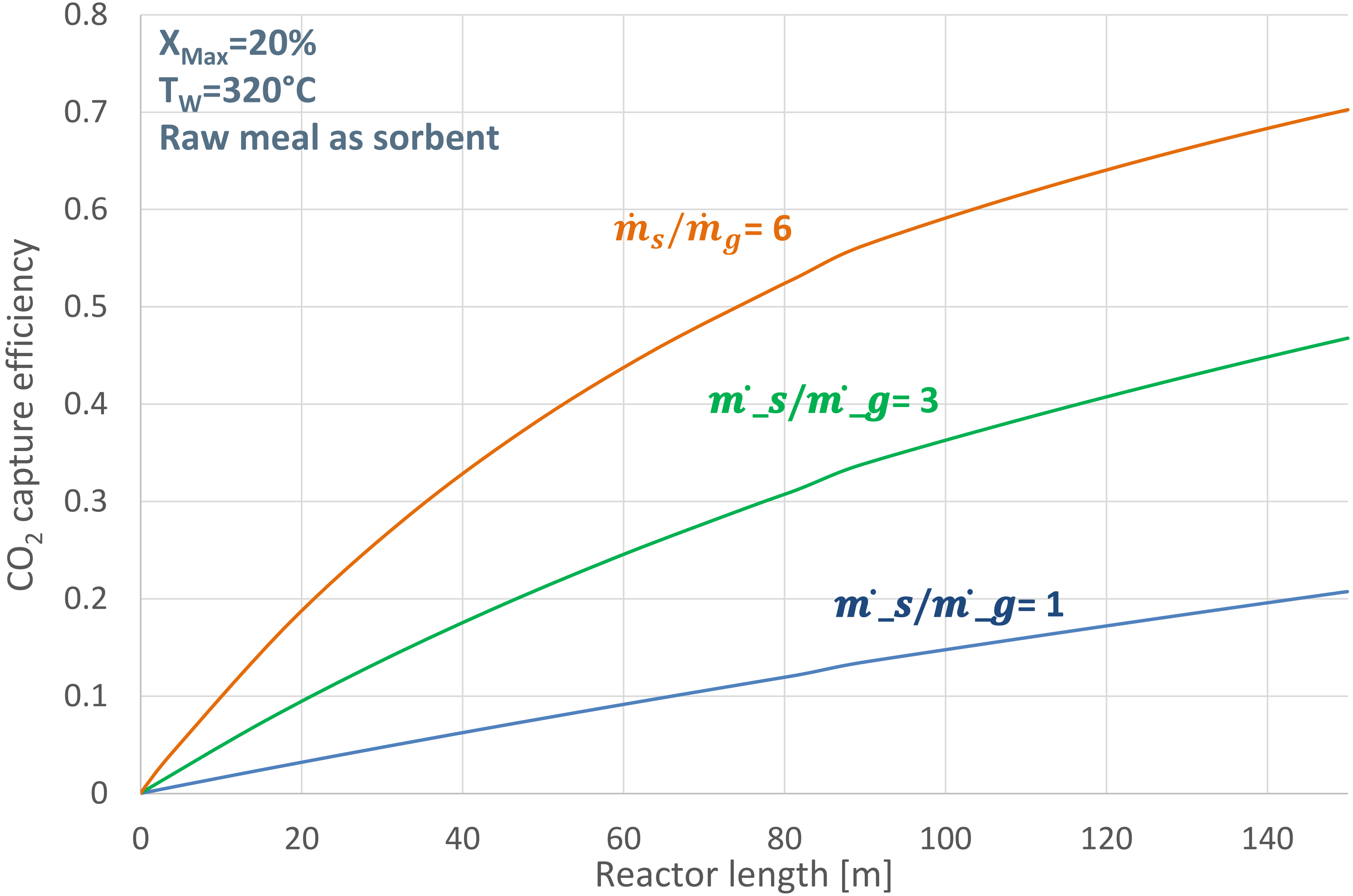
- Reactor length: 150 m
- Sorbent nature: calcined raw meal
  (*66.1% CaO, 4%$CaCO_3$, 21% SIO_2, 3.1% Al_2O_3, 1.1% Fe_2O_3, 3.6% MgO, 1.1%$CaSO_4$*)
- Inlet solid/gas velocities: 0/15 m/s
- Inlet solid/gas temperatures: 600/600°C
- Isothermal reactor walls.



Sensitivity analysis on:

- Solid loading ($\dot{m}_s/\dot{m}_g = 1/3/6$);
- Reactor wall temperature (T_W=260/320/500°C)
- Carbonators number (1/4)
- Sorbent maximum conversion (X_{MAX}=10/20/30%).

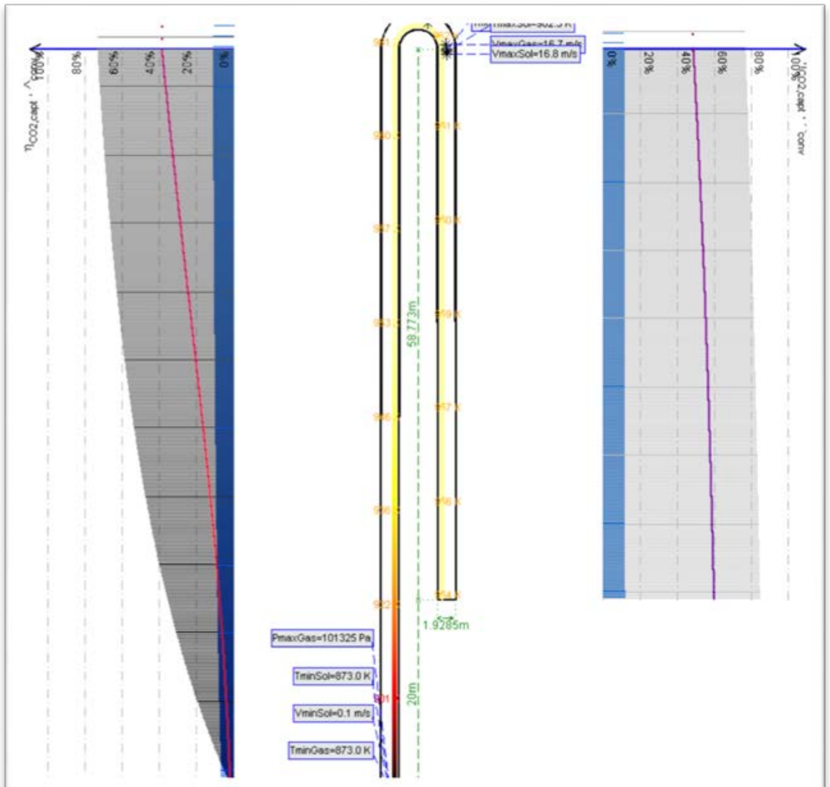
Simulation results (i) – Effect of solid loading ($\dot{m}_s/\dot{m}_g$)

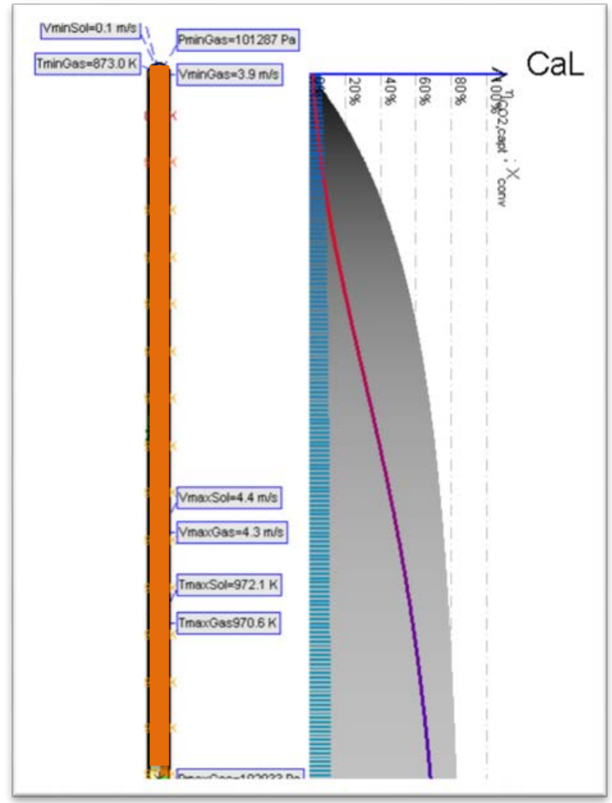


Alternative geometry: downdraft carbonator

L=150 m , v=15 m/s

L=60 m , v=4 m/s

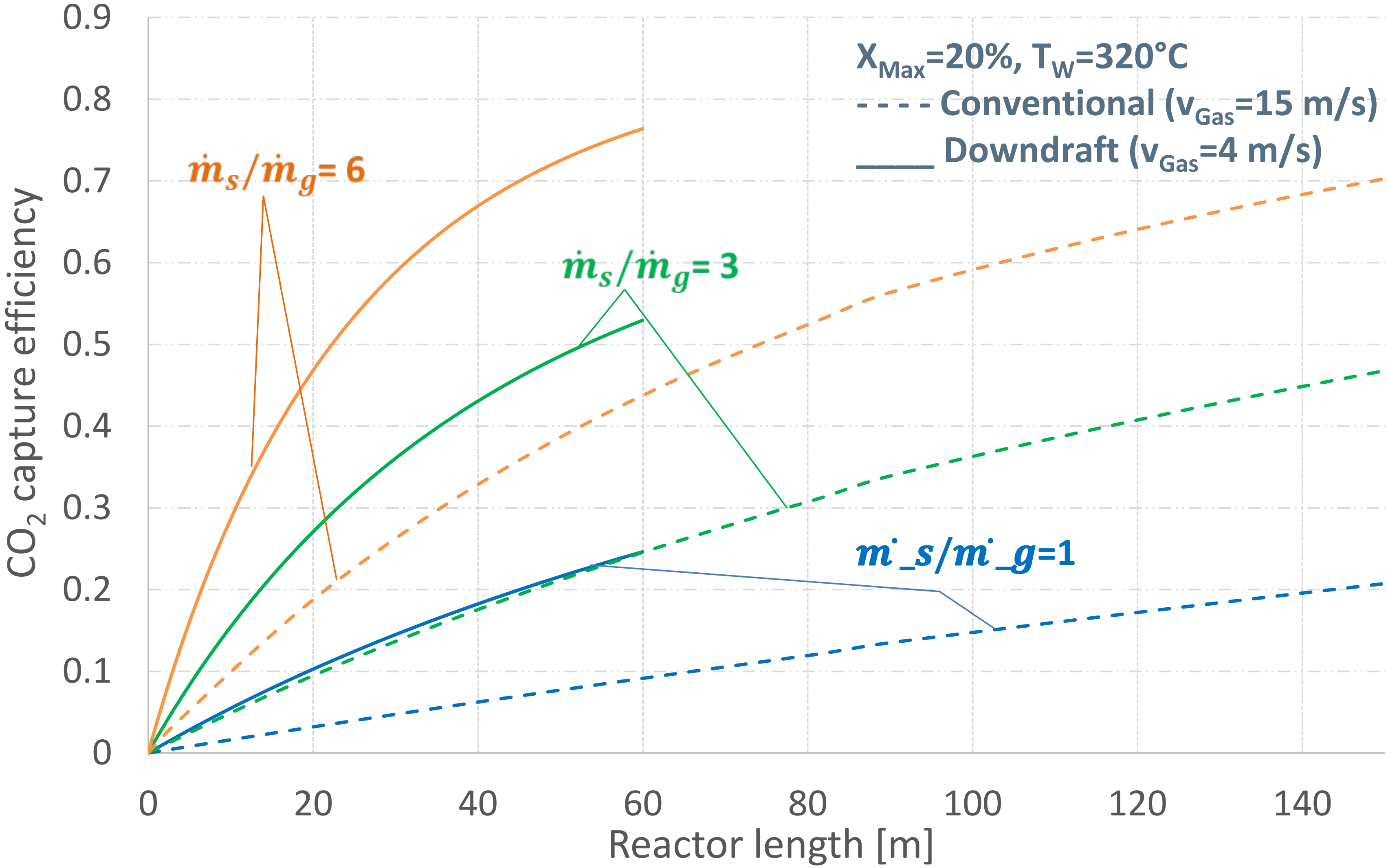




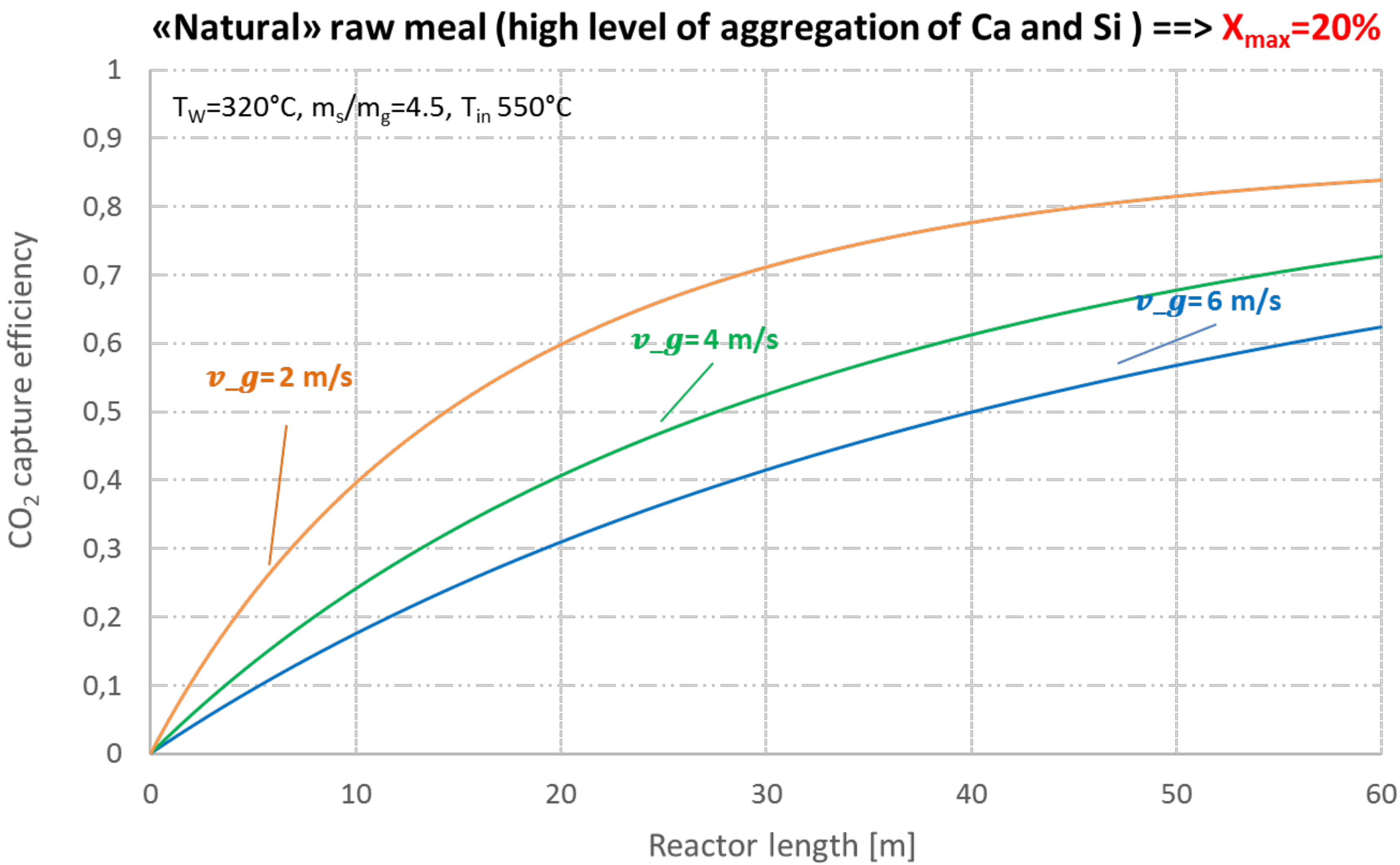
UPDRAFT

DOWNDRAFT

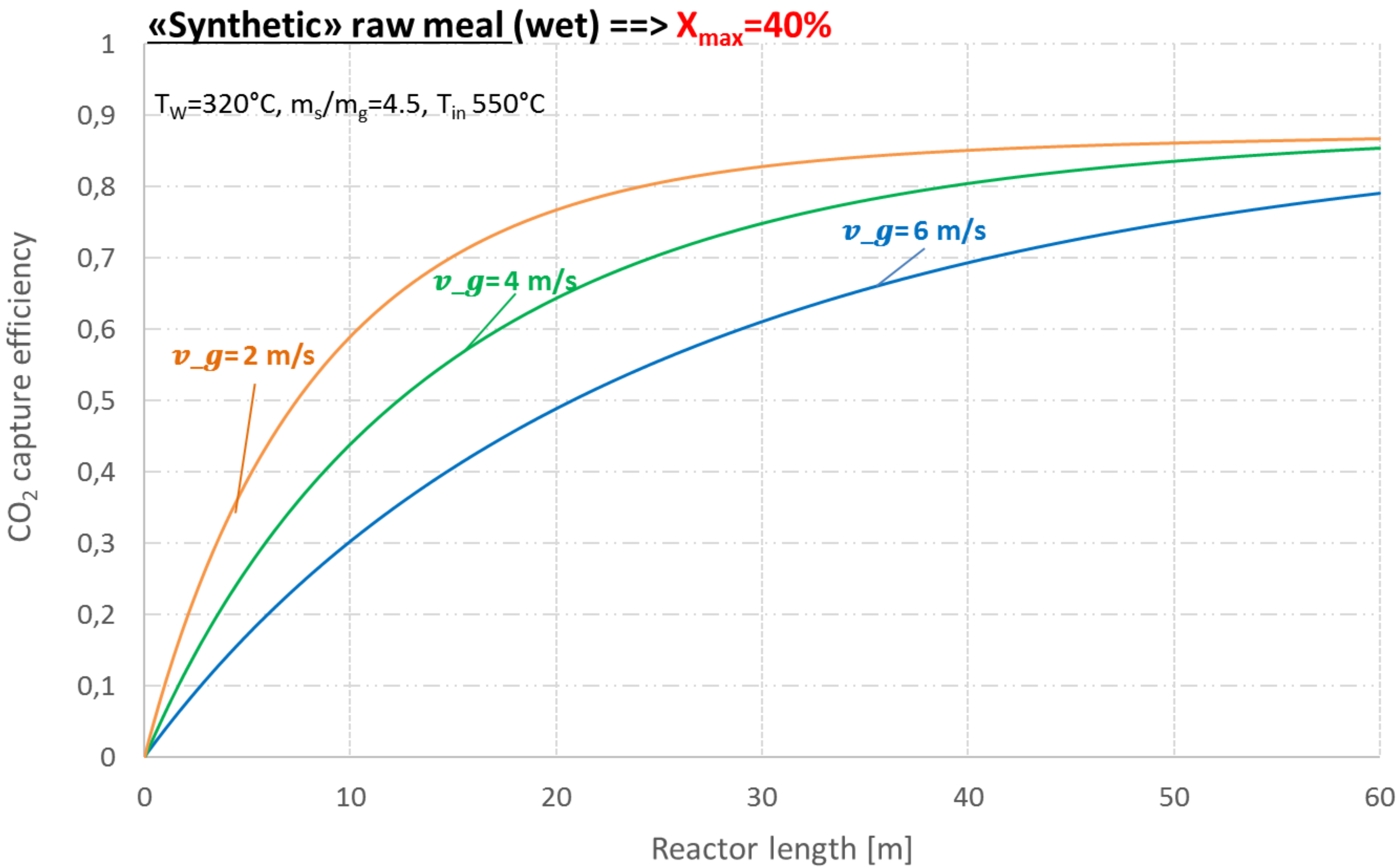
Simulation results (ii) – Downdraft vs updraft



Simulation results (iii) : Natural raw meal as sorbent



Simulation results (iv) : Synthetic raw meal as sorbent



Conclusions

- A relatively high solid loading (m_s/m_g=6÷10) is required for obtaining high capture rates;
- Sorbent capacity (➔raw meal nature, calcination condition) has a significant impact on carbon capture rate;
- Downdraft option allows for higher residence time and higher sorbent loadings ➔ improves capture rates;

Further research needs :

-properties of different raw meals and calcination conditions on CaO sorbent performance

CEMCAP

-fluid dynamics of EF reactor at high solid/gas ratio
-experimental validation of the concept under realistic conditions

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Thank you for your attention!

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ACKNOWLEDGMENTS

Activities related to Cemcap project have received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 641185.

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Co-funded by the European Union