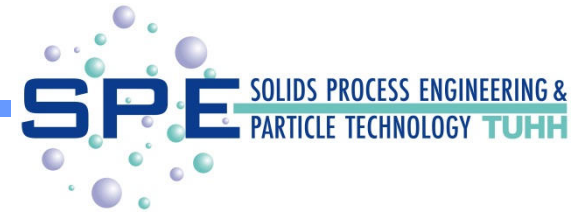


The Role of Attrition and Solids Recovery in a Chemical Looping Combustion Process

Marvin Kramp, Andreas Thon, Ernst-Ulrich Hartge,
Stefan Heinrich, Joachim Werther

Hamburg University of Technology
Institute of Solids Process Engineering and Particle Technology

The CLOCK Project (COORETEC)



- Investigates the Chemical Looping Combustion (CLC) of Coal for the CO₂ separation in atmospheric fluidized beds for a steam power process
- 3 institutes involved
 - Institute of Solids Process Engineering and Particle Technology (Hamburg)
 - Institute of Energy Systems (Hamburg)
 - Institute of Combustion and Power Plant Technology (Stuttgart)
- Funding by



(Federal Ministry of Economics and Technology)



Introduction

Goal of the present work:

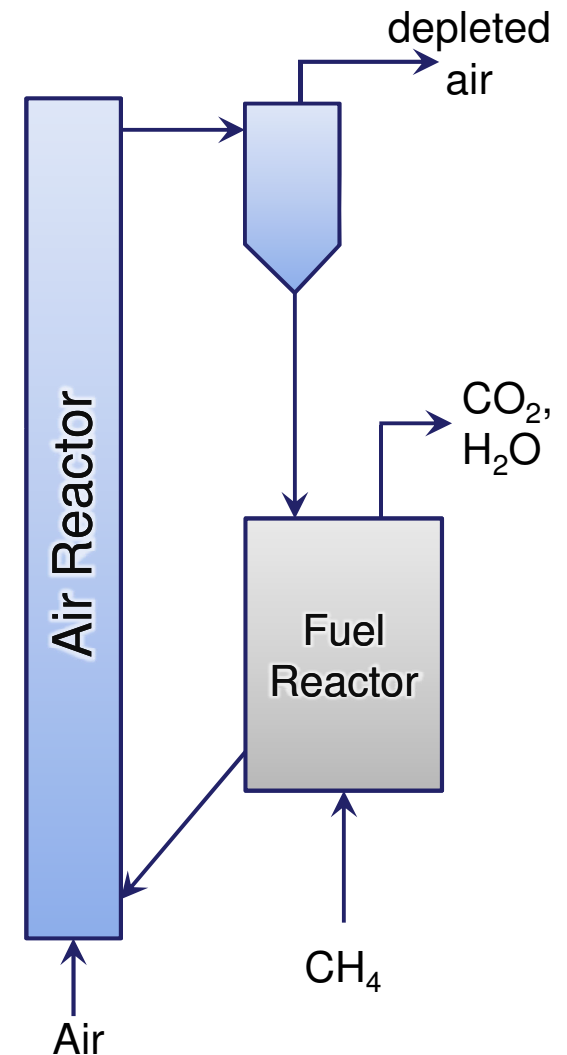
- Investigation of the role of attrition and solids recovery in a gas fired CLC process

Prediction of:

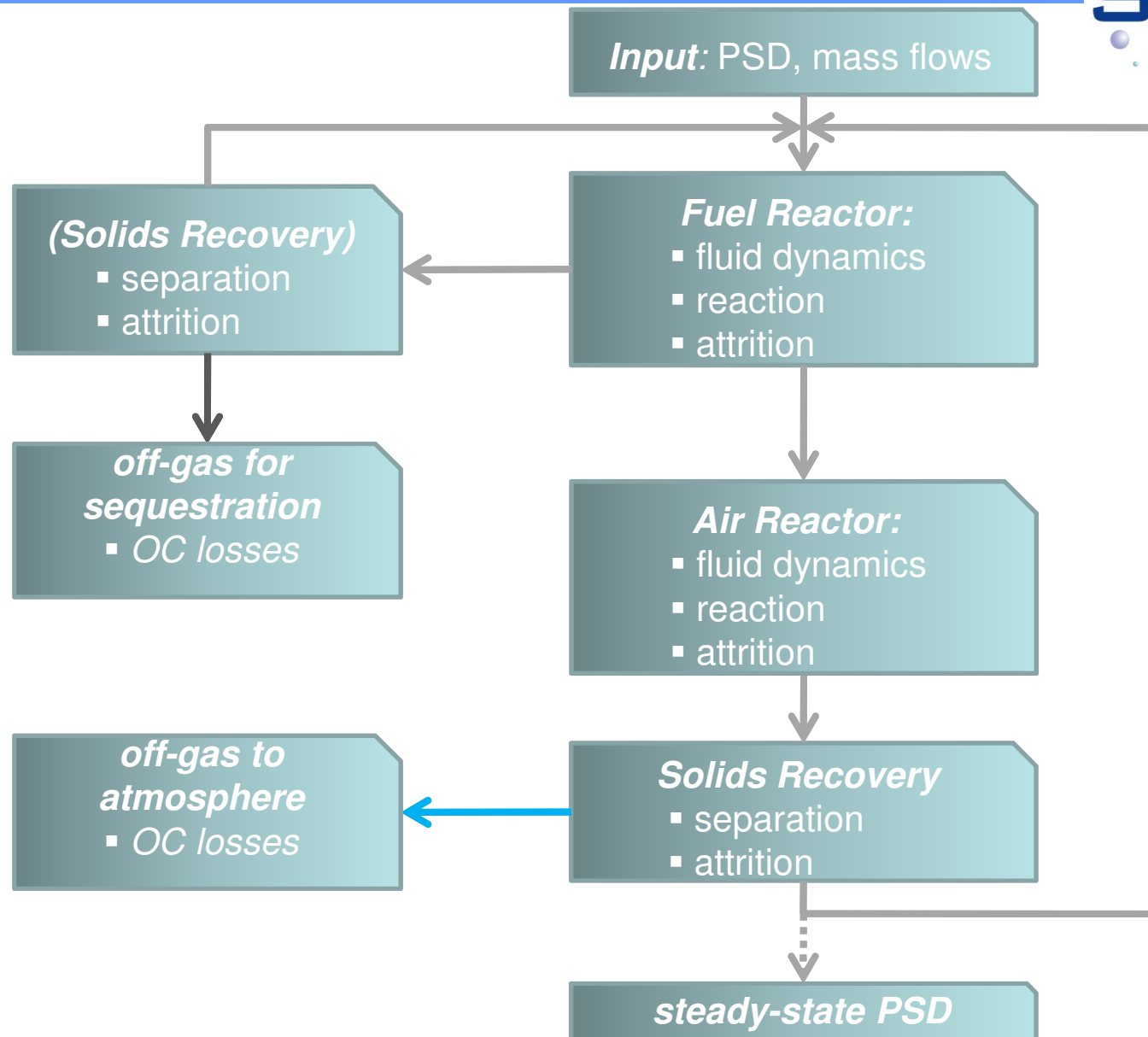
- circulation mass flow between reactors
- particle size distributions
- oxygen carrier losses

requires calculation of:

- fluid mechanics
 - influences attrition
- reactant conversion
 - influences fluid mechanics



Simulation Strategy



■ Fluid dynamics (1-D approach)

- **Bottom zone** (suspension and bubble phase according to Werther & Wein, 1994 / Hillgardt & Werther, 1986)
 - Considering diffusive (Sit & Grace, 1981) and convective **mass transfer** (Sitzmann et al., 1985) between suspension and bubbles
- **Freeboard** (elutriation constant approach e.g. Tasirin & Geldart (1998) and exponential decay of solids volume concentration according to Kunii & Levenspiel, 1990)

■ Heterogeneous reactions (shrinking core model according to Kunii & Levenspiel, 1991)

- Volume increase considered

■ Attrition (population balance approach according to Werther & Hartge, 2004)

- Bubble induced attrition (Merrick & Highly, 1974)
- Grid jet attrition (Werther & Reppenhagen, 2003)
- Cyclone attrition (Werther & Reppenhagen, 2003)

Test Case

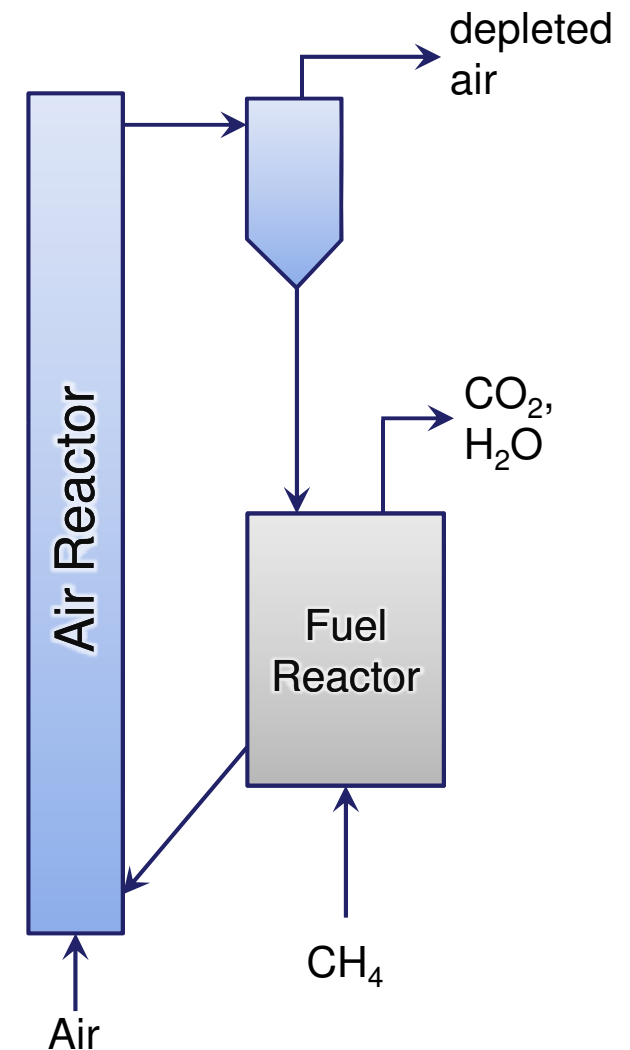
Application of the Model

Initial process setup:

- Setup as proposed by Lyngfelt et al. (2005) for gaseous fuels
- Air reactor: Circulating Fluidized Bed (CFB)
- Fuel reactor: Bubbling Fluidized Bed (BFB)
- 2 scales: Pilot (49,9 kW_{th}) and 100 MW_{th}

Investigation of setups changes:

- Different cyclone arrangements for improved solids recovery



Test Case

Oxygen Carrier

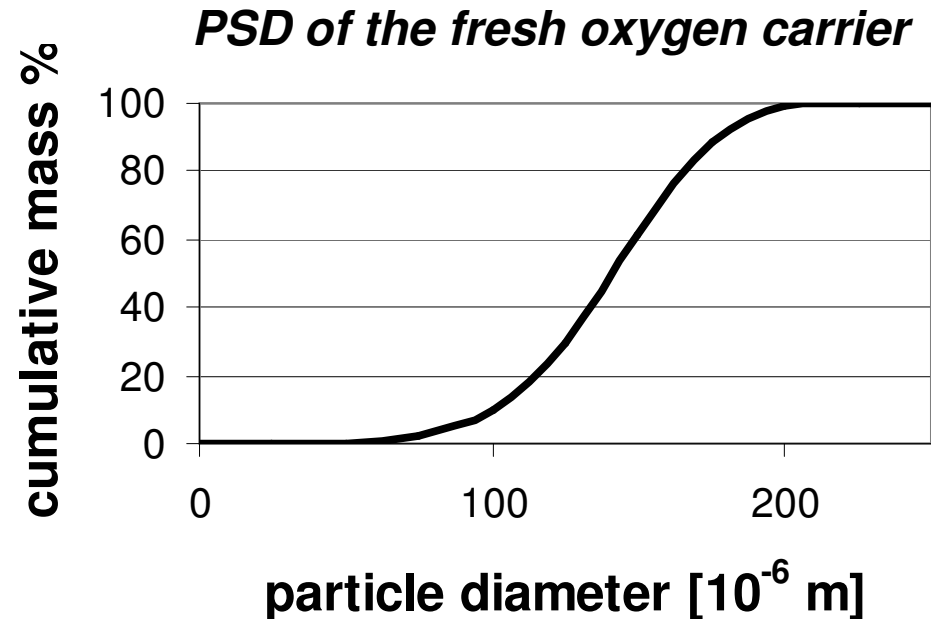
Oxygen Carrier (Cu10Al-I) information was taken from literature (Abad et al. 2007 / Garcia-Labiano et al. 2006)

- Al₂O₃ carrier impregnated with Copper

OC Properties	
Total CuO content [%-wt.]	13
Content of active CuO [%-wt.]	10
Apparent Density ρ_s [kg/m ³]	1800
Carrier Capacity, R_0	0.02

$$R_0 = \frac{m_{ox} - m_{red}}{m_{ox}}$$

Reactions	CH ₄ -Reaction	O ₂ -Reaction
molar density of solid reactant ρ_m [mol/m ³]	80402	140251
grain radius r_g [m]	4.0×10^{-10}	2.3×10^{-10}
k_0 [mol ¹⁻ⁿ m ³ⁿ⁻² s ⁻¹]	4.5×10^{-4}	4.7×10^{-6}
E_a [kJ/mol]	60	40
reaction order n [-]	0.4	1.0

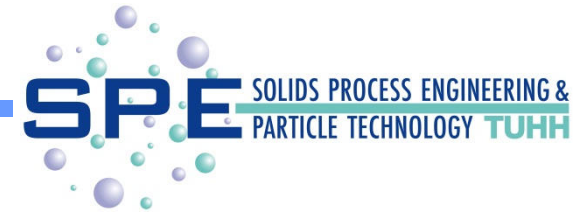


Attrition Constants	
C_{bubble} [1/m]	3.5×10^{-8}
C_{jet} [s ² /m ³]	4.5×10^{-3}
$C_{cyclone}$ [s ² /m ³]	2.2×10^{-5}

values determined by experiments for comparable catalyst

Test Case

Simulation Input Parameters



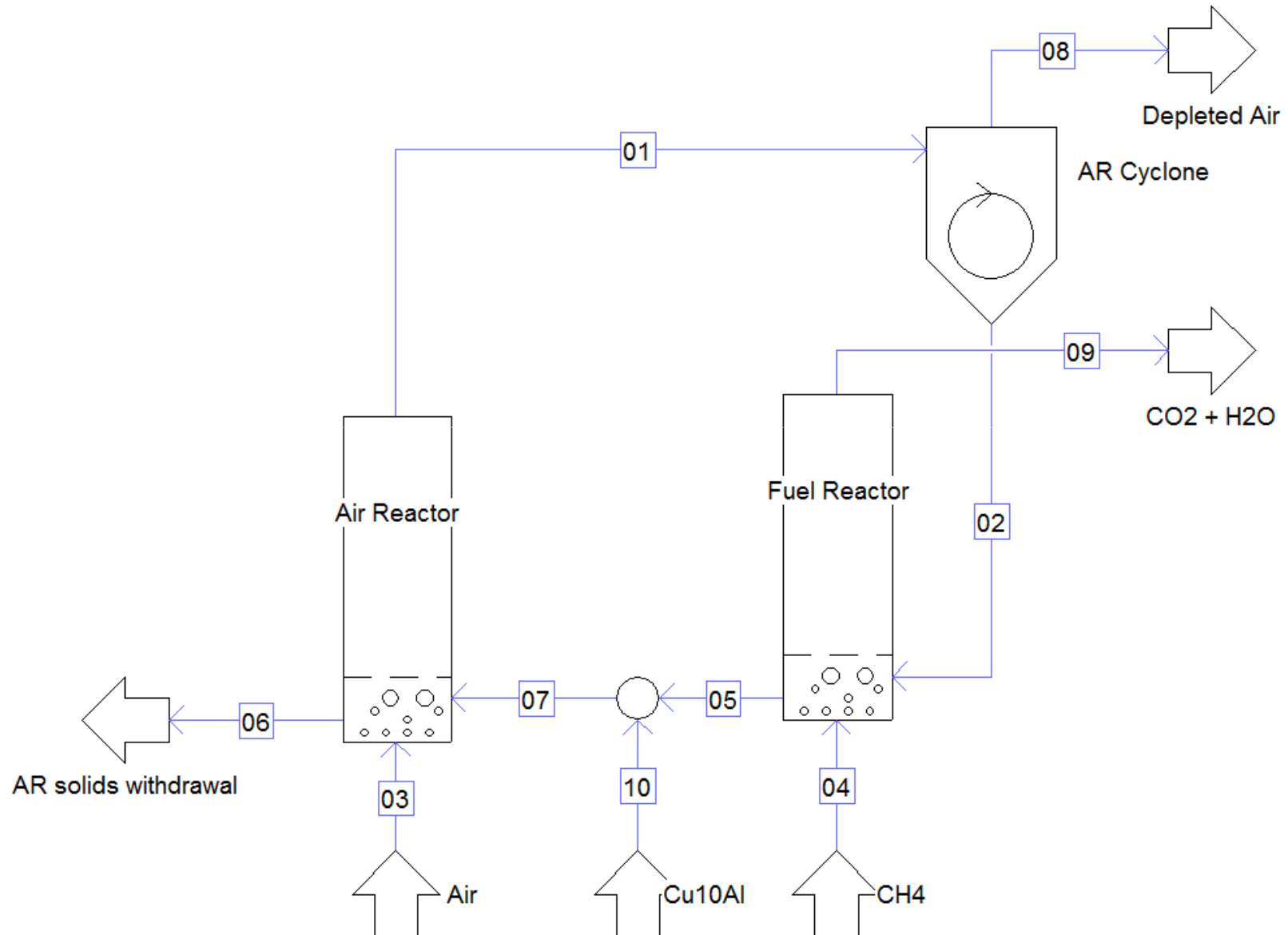
Fuel Reactor	Pilot Scale (50 kW _{th})	100 MW _{th} scale	dimension
height	2	12	m
diameter	0.3	10	m
Δp bed	2700	2700	Pa
Δp cyclone (if applicable)	900	900	Pa
temperature	850°C	850°C	°C

Air Reactor	Pilot Scale (50 kW _{th})	100 MW _{th} scale	
height	10	17	m
diameter	0.1	4.2	m
Δp bed:	7000	7000	Pa
Δp cyclone(s)	1200	1200	Pa
temperature	900°C	900°C	°C

General	Pilot Scale (50 kW _{th})	100 MW _{th} scale	
gas distributors	porous plate	bubble cap	
gas inlet velocity of cyclones	≈ 20	≈ 20	m/s

Test Case

Screenshot of the SolidSim Flowsheet



Simulation Results

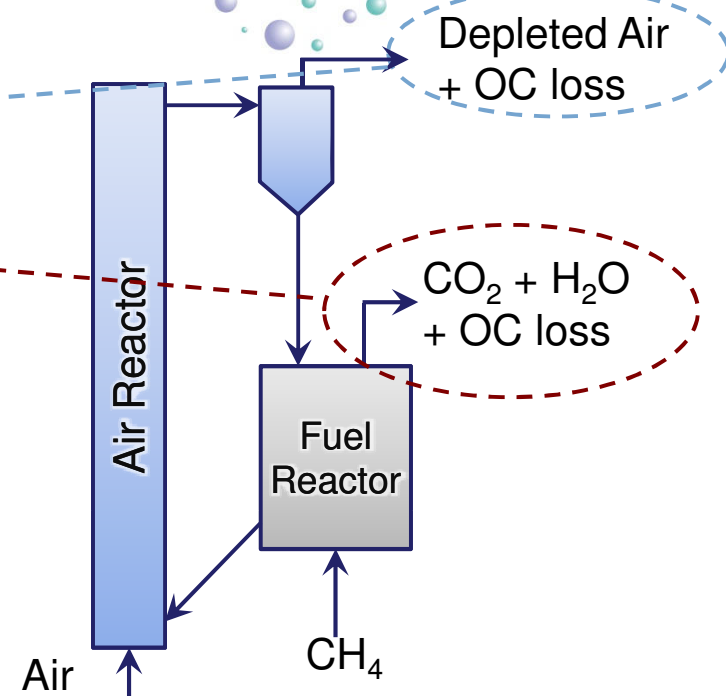
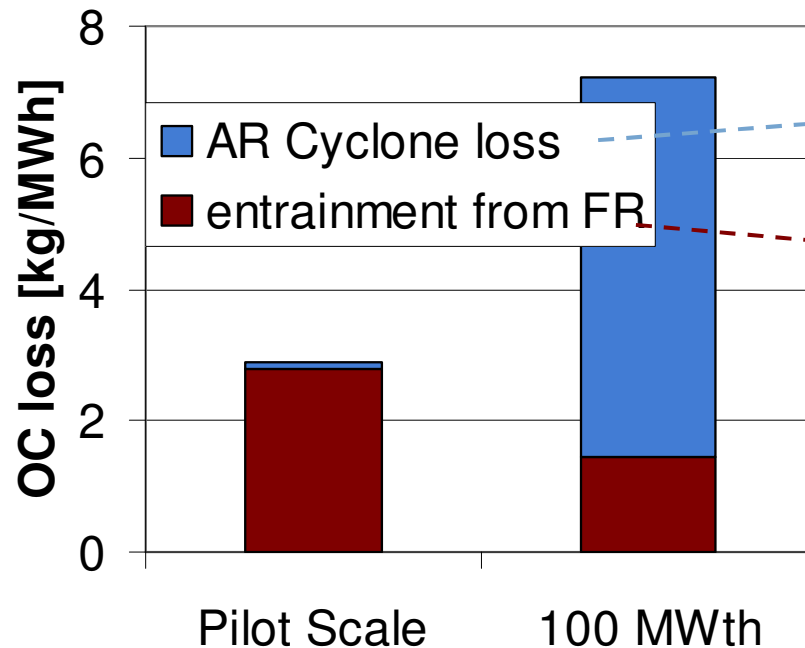
Results without Attrition Considered



- Solids inventory of 17.1 kg (50 kW_{th}) / 47,800 kg (100 MW_{th})
- Circulation mass flow: 0.36 kg/s (50 kW_{th}) / 647 kg/s (100 MW_{th})
 - $G_s = 46.2 \text{ kg/m}^2\text{s}$ in the AR
- Fuel gas conversion: 99.4 % (50 kW_{th}) / 84.7 % (100 MW_{th})
 - Too low conversion in fuel reactor for large scale application due to less efficient solids / gas contact in large scale
 - New simulation with higher Δp_{bed} (10,000 Pa instead of 2,700 Pa) and lower gas velocity (0.25 m/s instead of 0.37 m/s) by increased reactor diameter
 - New conversion: 93.1 %

Simulation Results

Oxygen Carrier Losses and their Origin

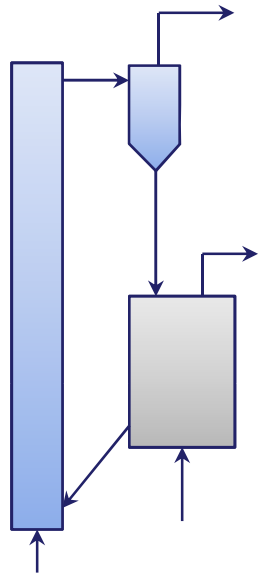


Results:

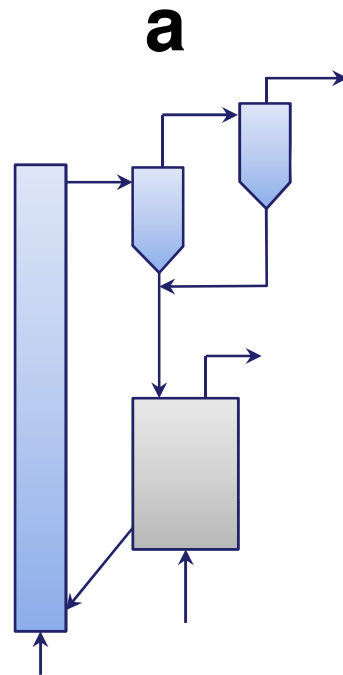
- The 100 MW_{th} scale process shows considerably higher losses
 - Lower separation efficiency of the large scale cyclone
- Major source of losses in the 100 MW_{th}-scale process is the air reactor cyclone
- Losses in the pilot scale originate mainly from elutriation from the fuel reactor

Simulation Results

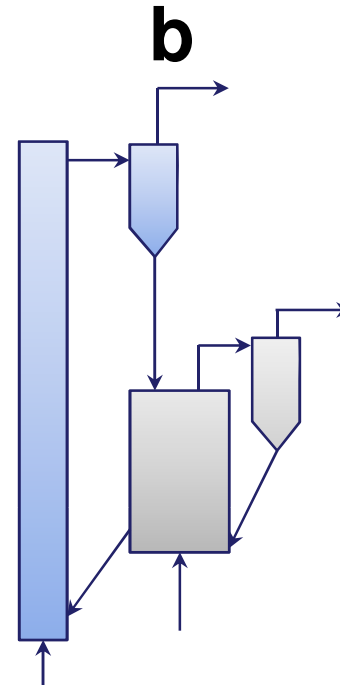
Investigated Arrangements of Solids Recovery



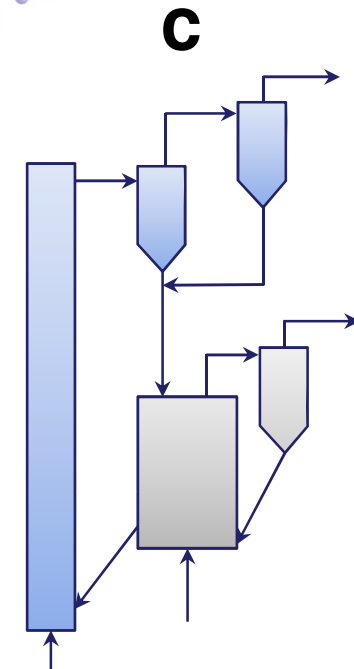
Standard case



Additional secondary cyclone stage after the air reactor



Additional cyclone after the fuel reactor with return leg to fuel reactor

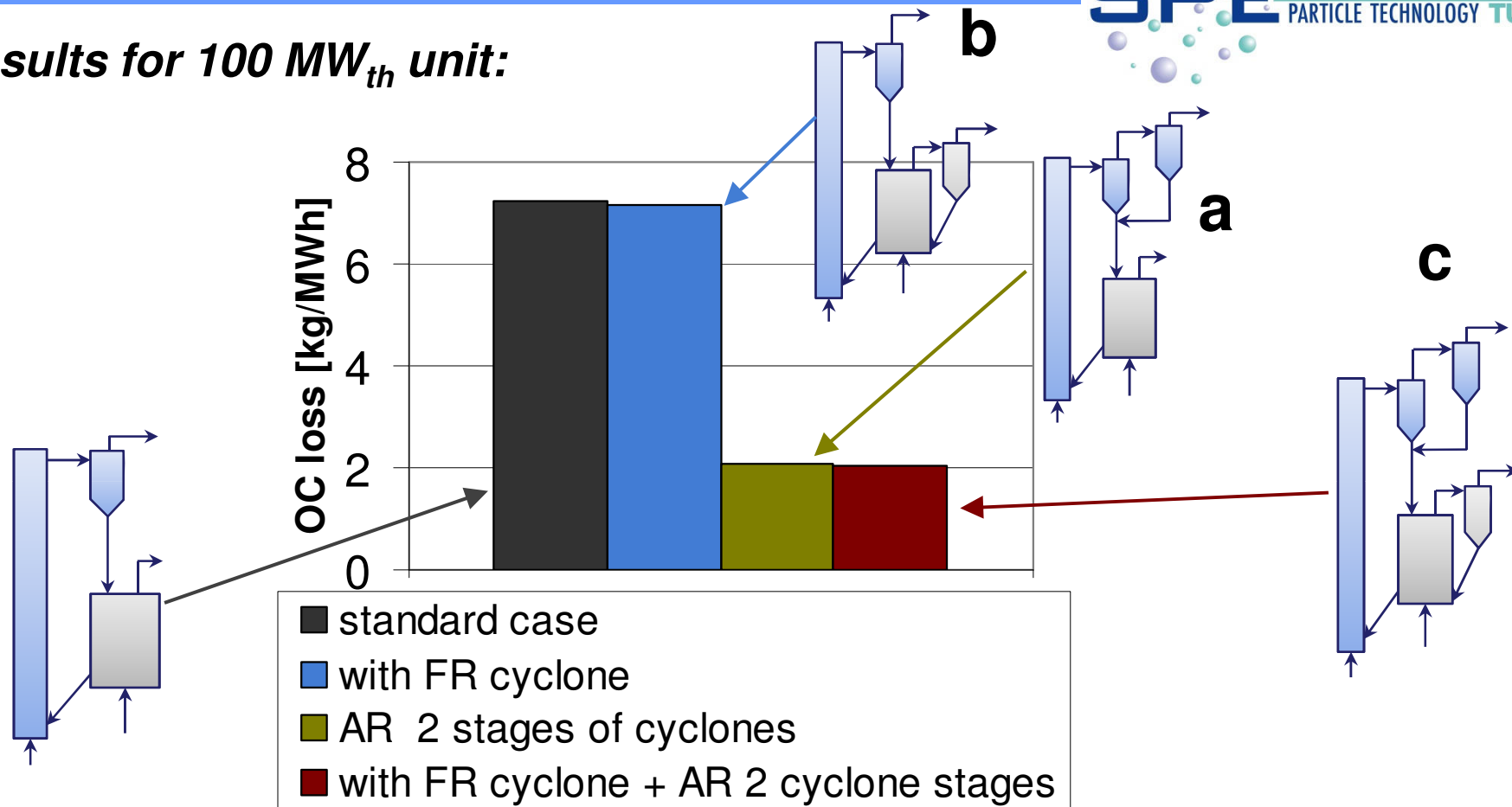


Combination of case a and b

Simulation Results

OC losses in Altered Setups

Results for 100 MW_{th} unit:

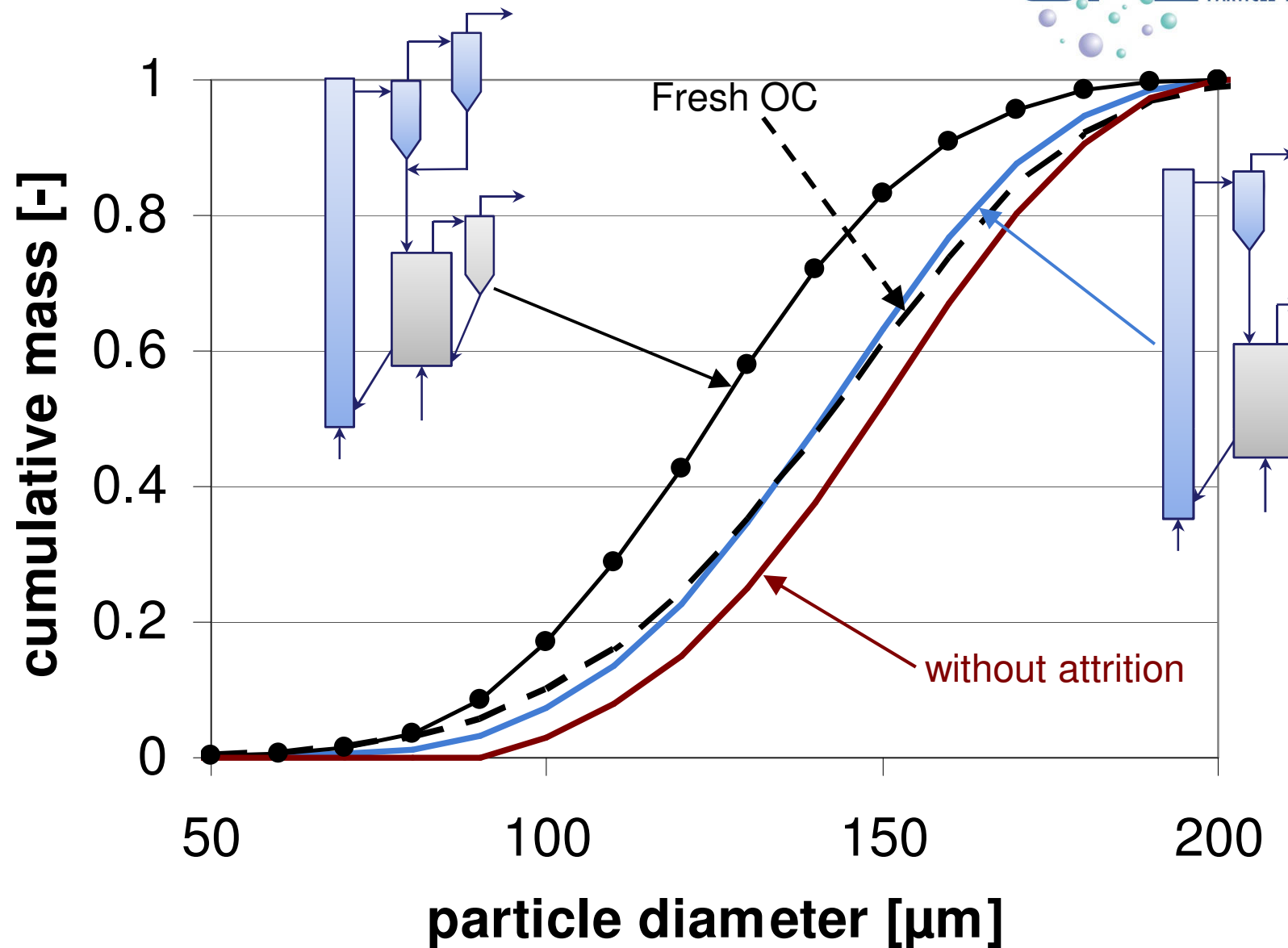


Results for 100 MW_{th} scale:

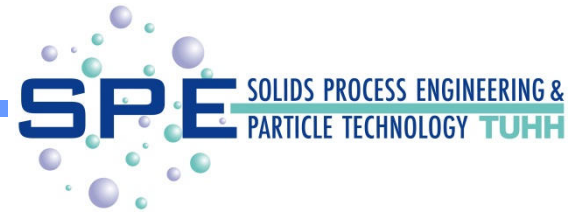
- Second cyclone stage after air reactor reduces the OC-losses significantly
- Additional cyclone after the fuel reactor has hardly any influence

Simulation Results

Steady-State Particle Size Distributions



Summary



- Attrition and solids separation influence the OC losses in a complex manner
- Results from measurements from lab or pilot scale cannot be directly applied to the large scale
- Large scale performance can only be predicted by simulations that consider fluid dynamics and reactions