Use of CFD as a Design Tool for Scale-Up of Fluidized-bed Reactors

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RTI views CFD as a Critical Design Tool

- RTI has an extensive pipeline of fluidized-bed processes
- Many technologies are entering pilot-scale and commercial-scale demonstration phases

Bench scale

Pilot scale

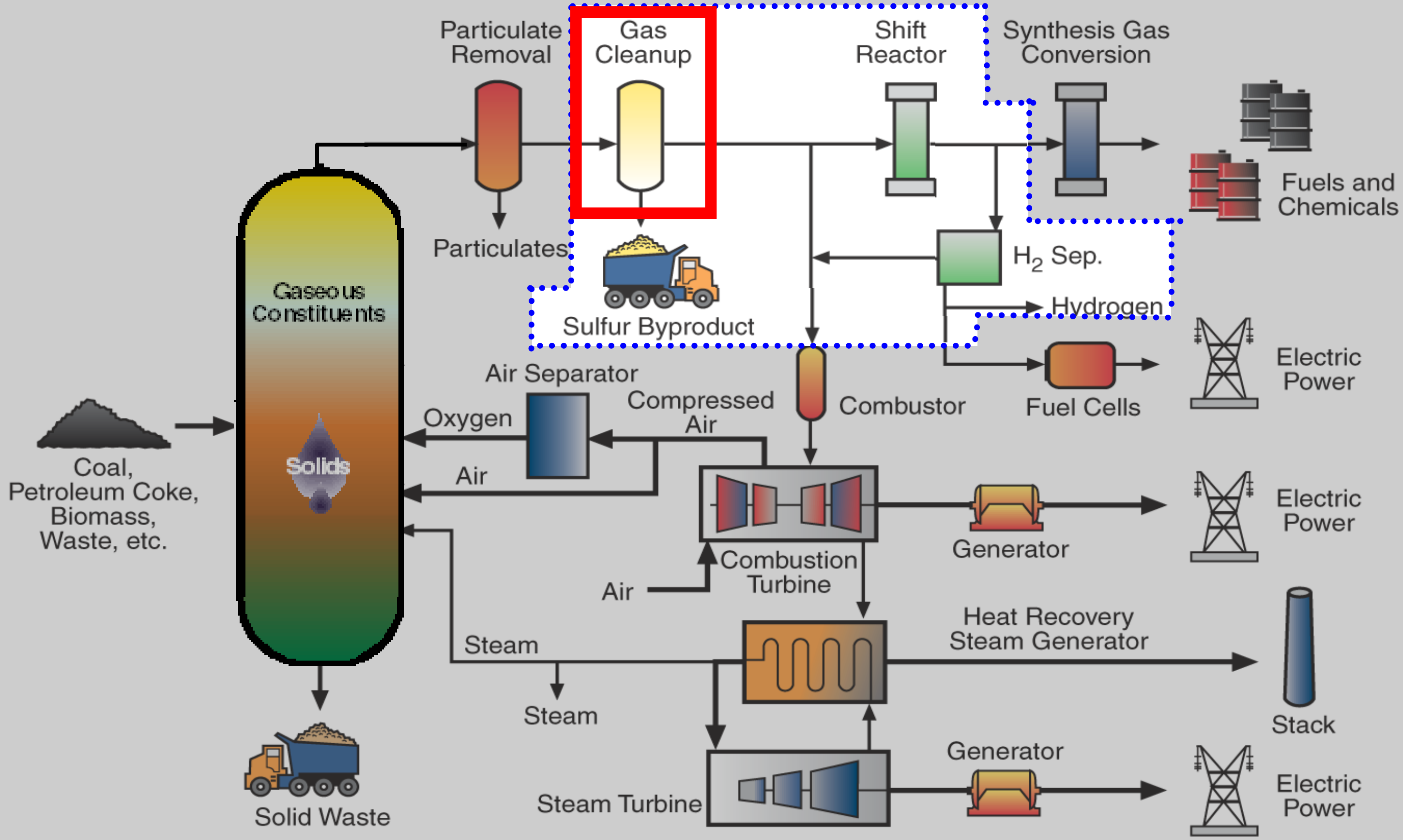
Commercial scale

CFD modeling offers tremendous benefits for RTI's scale-up and commercialization efforts

RTI's Fluidized-bed Reactor Technologies

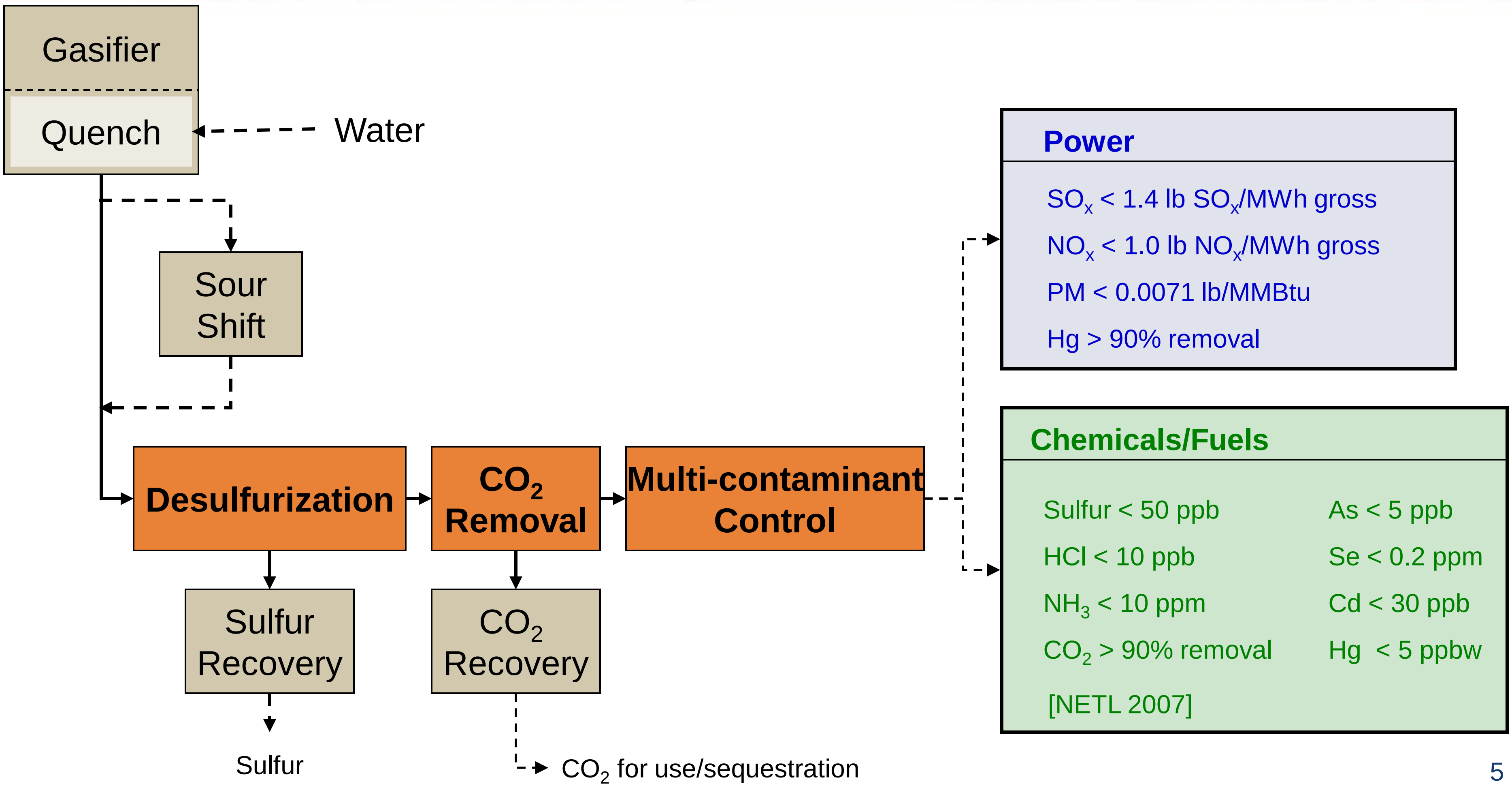
- **High temperature desulfurization process (HTDP)**
- **Dry carbonate CO_2 capture**
- **Transport reactor-based Sabatier process for CO_2 reuse**
- **SNG production with catalytic gasification in transport reactor**
- Hydrogen production using steam-iron process
- Syngas clean-up using 'Therminator' technology
- Chemical looping combustion
- Co-gasification (Coal & Biomass)
- Catalytic biomass pyrolysis

Integrated Gasification Combined Cycle (IGCC)



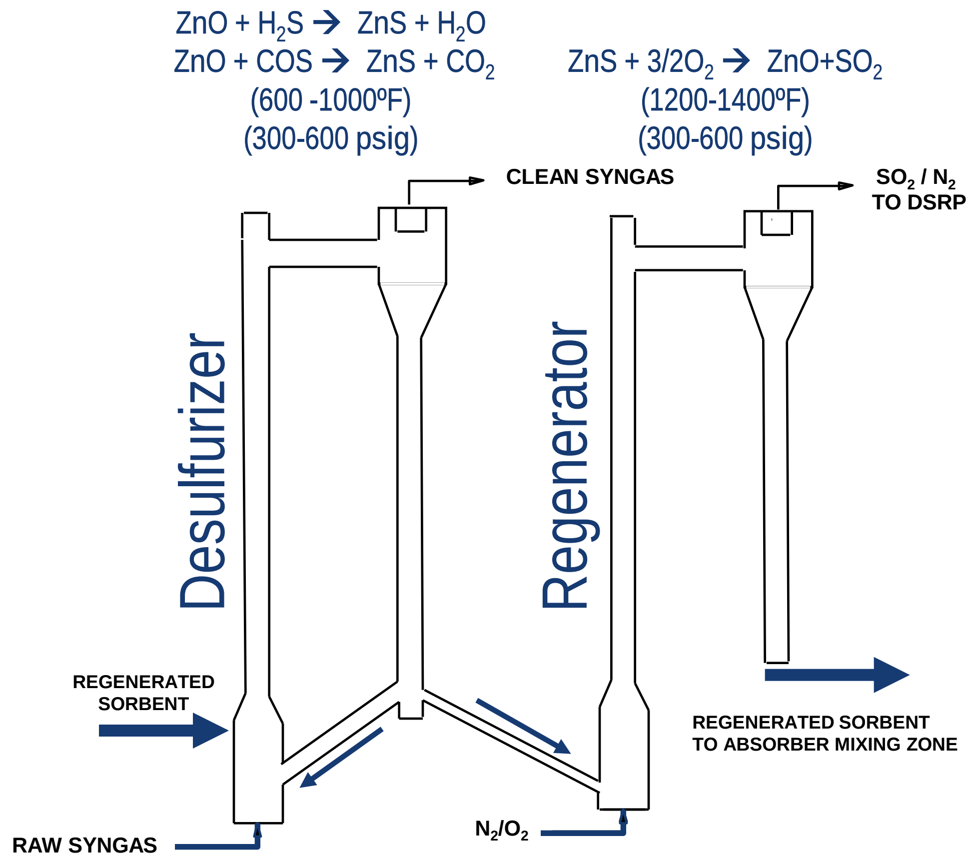
R&D objective: Platform of warm syngas cleaning technologies providing improved efficiency, environmental performance, and cost

Process Integration – Modular Approach



High Temperature Desulfurization Process (HTDP)

- Dual loop transport reactor system
- Similar to FCC process design
- Patented ZnO-based attrition-resistant sorbent (RTI-3)
- High temperature sulfur (H_2S and COS) removal

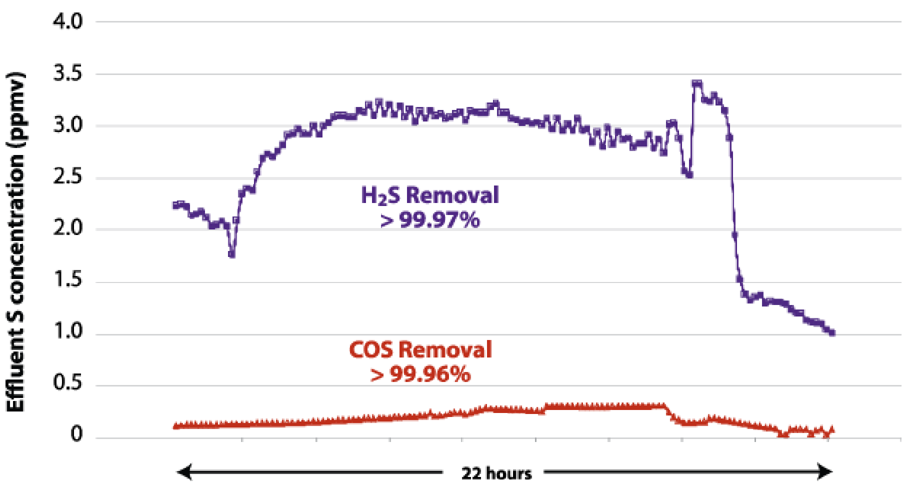


RTI – Eastman High Temperature Desulfurization Process

Pilot Plant Field Testing

Extensive pilot plant tests completed at Eastman Chemical Company with coal-derived syngas.

- High Temperature Desulfurization Process (HTDP)
  - >99.9% total sulfur removal (H_2S and COS) for >3,000 hours
  - Low attrition rates ~31 lb/million lb circulated



Pressure, psig	300	450	600
Inlet Concentration, S ppmv	8,661	7,023	8,436
Effluent Conc. S ppmv Range	5.9 0.4–9.3	10.7 2.4–20.6	5.7 3.3–18.1
S Removal, %	99.93	99.82	99.90

More than 3,000 hours of syngas operation

50 MW_e Protoype Unit of Warm Syngas Clean-up Technology

0.3 MWe Unit at
Eastman Chemical

50 MWe Unit to be commissioned in 2012 at Tampa Electric, Polk Power station

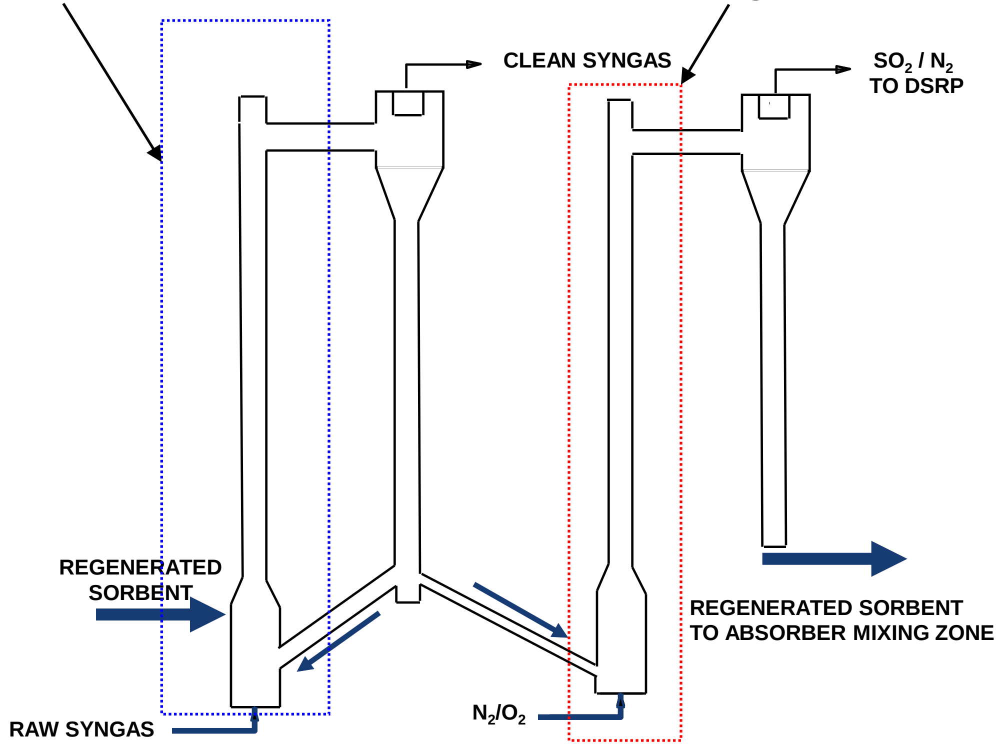
600 MWe
Unit ?

RTI and DOE/NETL’s Computational and Basic Sciences Group are using CFD modeling to assist in the design of HTPD for 50 MWe Prototype Unit

NETL's CFD Modeling Approach: Separate Desulfurization and Regeneration Reactor Models

Boundary of desulfurization reactor

Boundary of regenerator reactor



RTI – Eastman High Temperature Desulfurization Process

Comparison of NETL's Models Predictions with Pilot Plant Data

Desulfurization Reactor

Tag	*Descriptor*	*Unit*	*Measurement*	*CFD Predicted*
TIC-212	Mixing Zone T	°F	829	862
TIC-228	Riser Zone T	°F	856	865
PDT- 210	Mixing Zone Dp	in. H_2O	36	12
PDT-215	Transition Dp	in. H_2O	30	23
PDT-220	Riser Dp	in. H_2O	20	26

- Acceptable agreement between measured and predicted pressure drop values
- Agreement between measured and predicted temperature values are less favorable.
  - thermal boundary condition of the reactor walls
  - assumed constant temperature wall

Regeneration Reactor

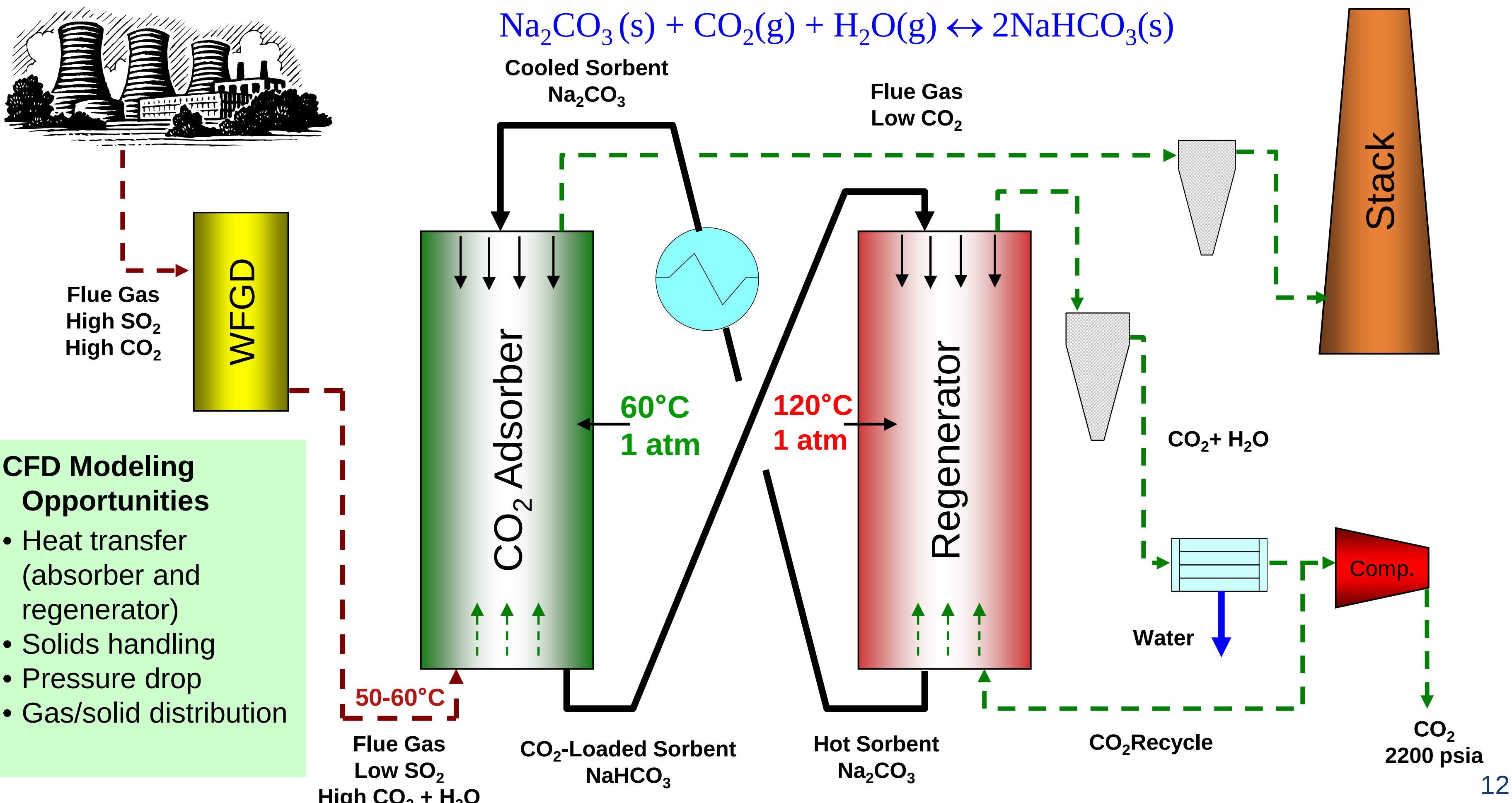
Tag	*Descriptor*	*Unit*	*Measurement*	*CFD Predicted*
TIC-312	Mixing Zone T	°F	1282	1350
TIC-313	Mixing Zone T	°F	1325	1404
TIC-314	Mixing Zone T	°F	1318	1400
TIC-315	Mixing Zone T	°F	1311	1396
TIC-316	Mixing Zone T	°F	1287	1391
TIC-328	Riser Zone T	°F	1252	1380
TIC-329	Riser Zone T	°F	1235	1375
PDT-310	Mixing Zone Dp	in. H_2O	87	42
PDT-315	Transition Dp	in. H_2O	56	34
PDT-320	Riser Dp	in. H_2O	8	25

Path Forward for HTDP CFD Modeling

- Validate NETL's CFD models with data from Eastman testing
- Use validated models to assist in design of 50 MW_e prototype unit
  - Reactor geometries (heights, diameters, return leg locations, gas/solid distribution)
  - Optimize hydrodynamics and reaction kinetics for maximum performance
  - Evaluate start up and shut down procedures for prototype unit
- Develop CFD modeling options for dual loop HTDP system
- Evaluate regenerator heat integration
  - Optimize use of exothermic regeneration heat to heat incoming solids
  - Evaluate options for system start up
    - Light off additives
    - Fuel injection
- Optimize solids feed control for regeneration reactor

RTI's Dry Carbonate Process

Capture and Purification of CO_2 from Power Plant Flue Gas



CFD Modeling Opportunities

- Heat transfer (absorber and regenerator)
- Solids handling
- Pressure drop
- Gas/solid distribution

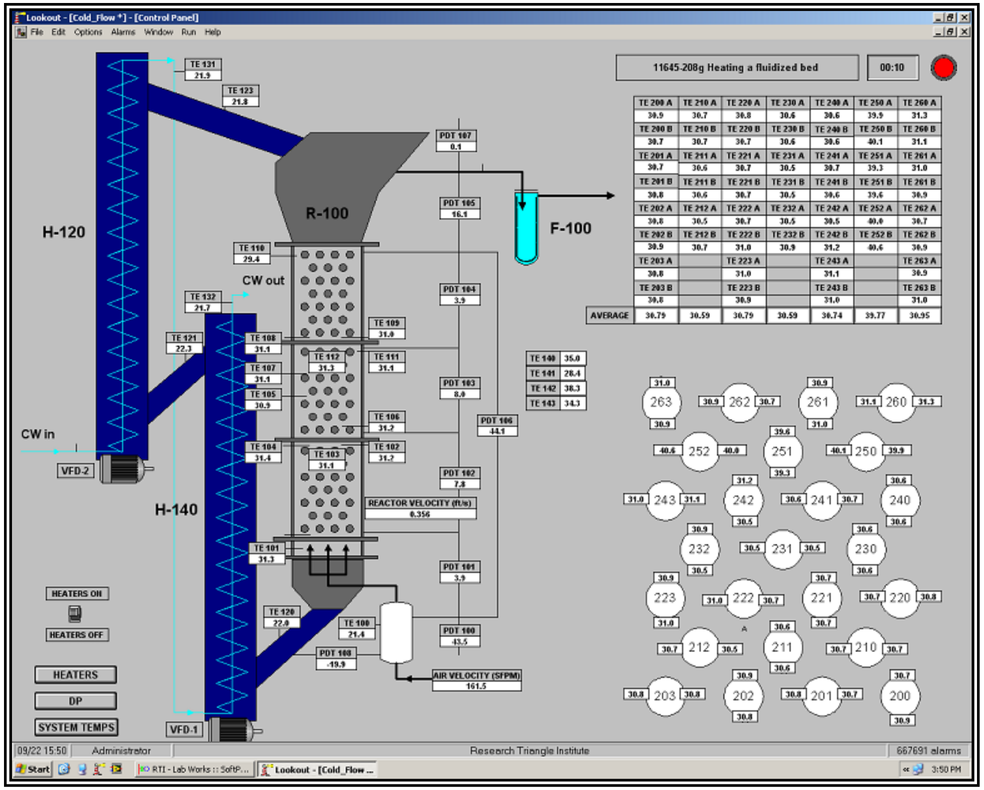
Heat Transfer – Reactor Design

Parameters	Uncertainties
• Tube Layout • Superficial Gas Velocity • Solids Circulation Rate • Gas Distributor Layout • Sorbent Properties (Particle size, shape, density, thermal conductivity, etc.)	• Fluid dynamics in reactor • System ΔP • Solid density distribution • Thermal gradients • Heat transfer coefficients • Mass transfer rates • Mixing, bubbling phenomena

Combination of CFD modeling and actual prototype testing will be needed to optimize reactor design

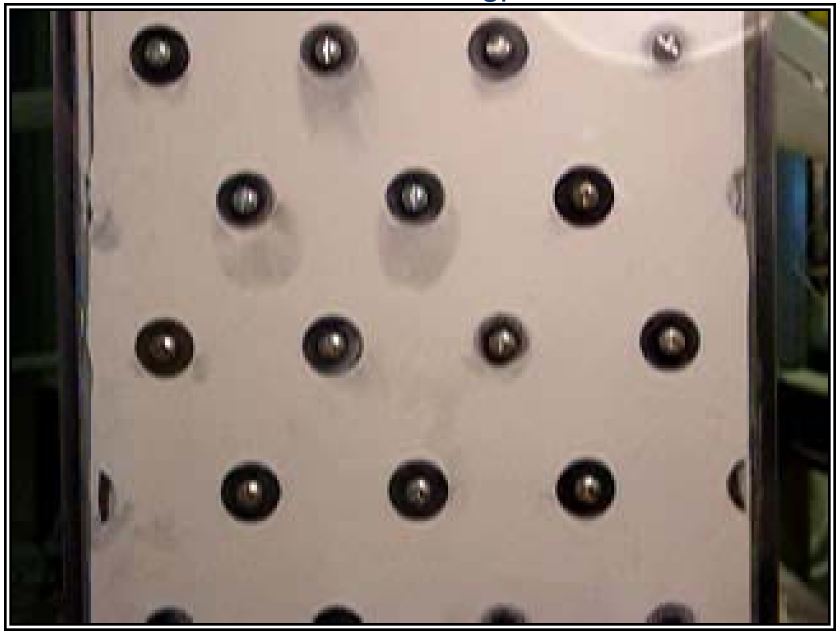
RTI’s Prototype Test Unit

HTU Process Control Interface



Highly instrumented unit allows for ‘tuning’ and validation of CFD models

Fluidization at u_{sf}=1.5 ft/s

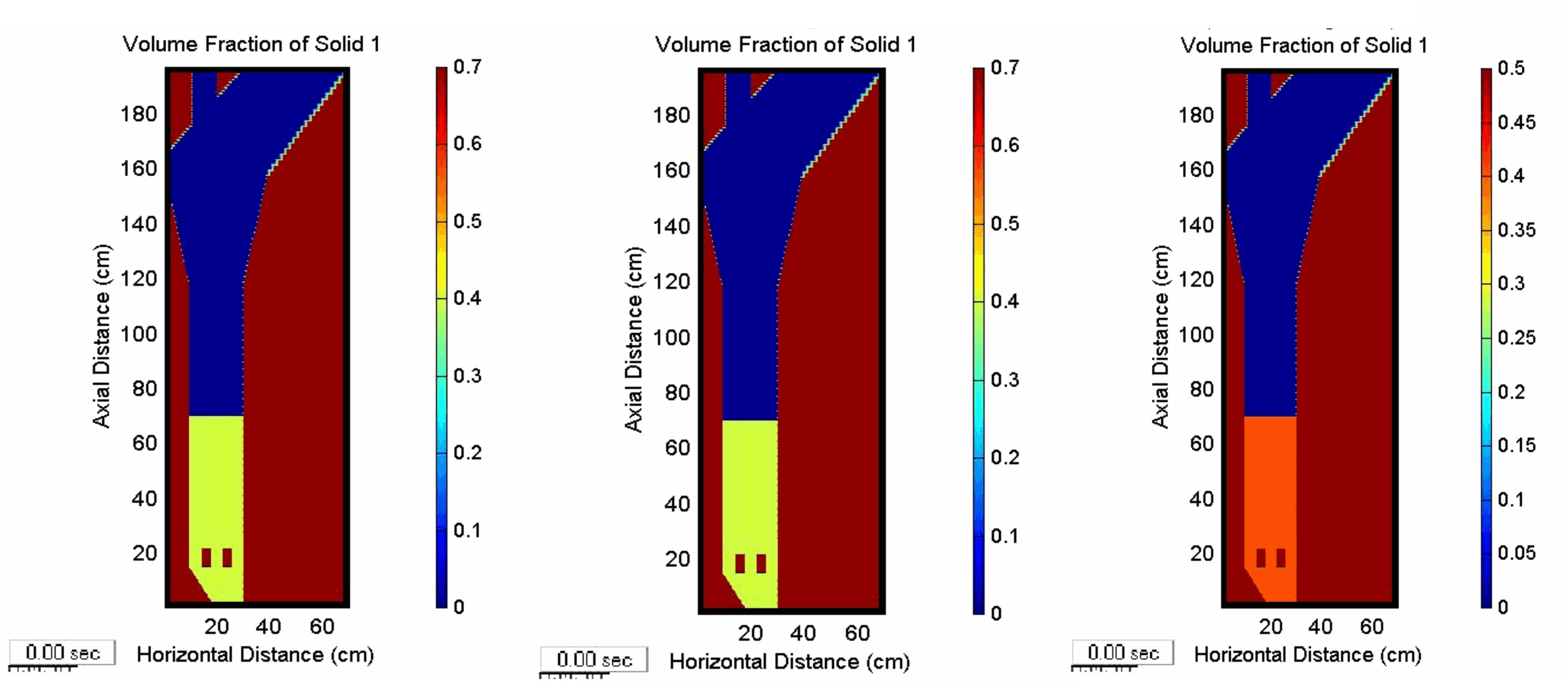


Constructed from clear, high melting point, polycarbonate to allow for visual observation to visually compare HTU and CFD hydrodynamics

Effect of Gas velocity on Solid Volume Fraction Distribution (MFIX simulations)

Inlet Gas: Air at atmospheric pressure

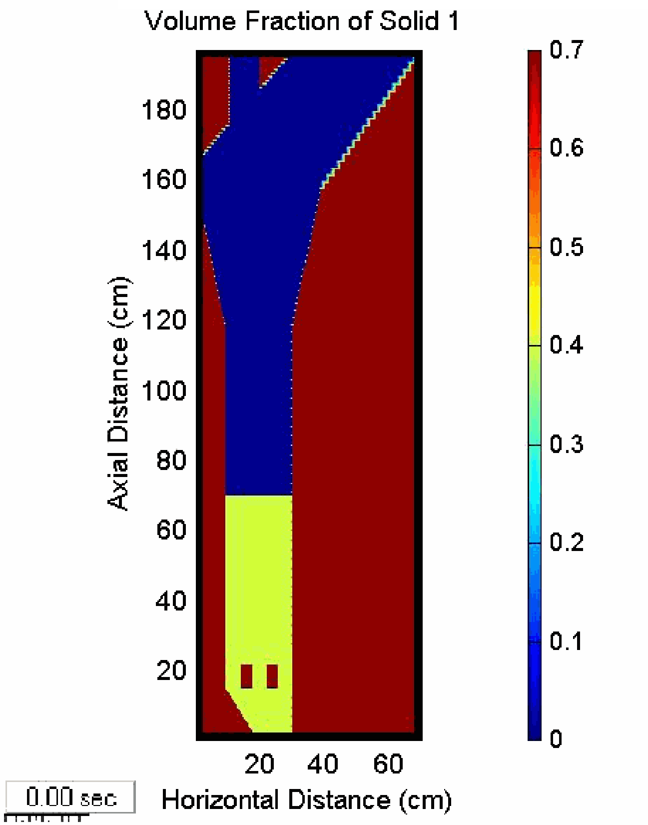
Gas Inlet Vel.: 0.39 ft/s 0.65 ft/s 1.3 ft/s



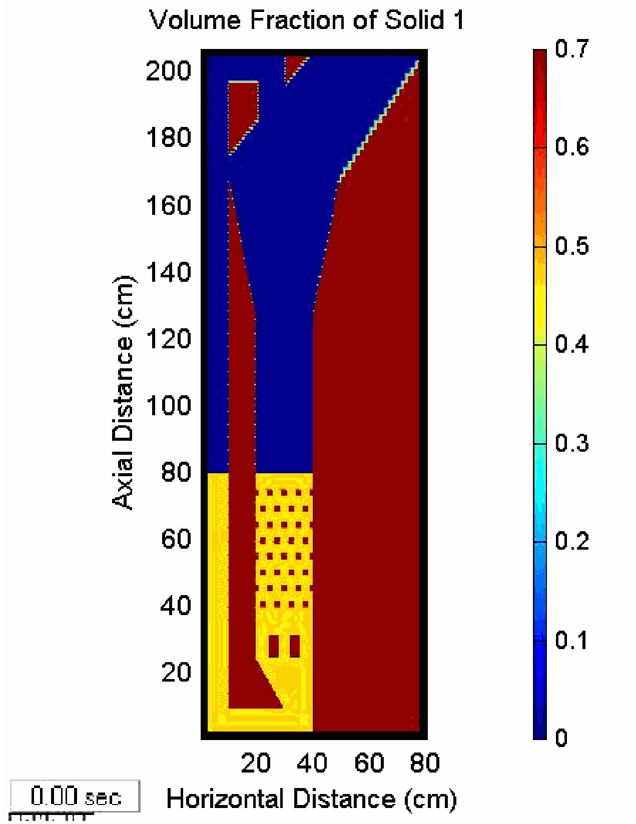
Particle density	1600 kg/m^3	Restitution coefficient	0.9	Specularity coefficient	0.6
Particle Diameter	70 μm	Restitution coefficient at the wall	0.6		

Effect of Heat Transfer Tubes on Hydrodynamics (MFIX Simulations)

With out Tube Bank



With Tube Bank



RTI's CFD Modeling Efforts For Dry Carbonate Process

Completed Work:

- Developed CFD model of contactor designs using MFIX
  - Evaluated effect of superficial gas velocity
  - Evaluated effect of heat internals
  - Completed actual testing with prototype unit
- Developing CFD model of contactor designs using Fluent

Planned Effort:

- Validate CFD models with prototype data
- Utilize validated CFD models to rapidly screen heat transfer configurations
- Utilize CFD models to optimize reactor design
  - Minimize pressure drop
  - Maximize gas throughput
  - Optimize recovery of reaction enthalpy (heat management)

Transport Reactor-based CO_2 Reuse Process

Sabatier Reaction

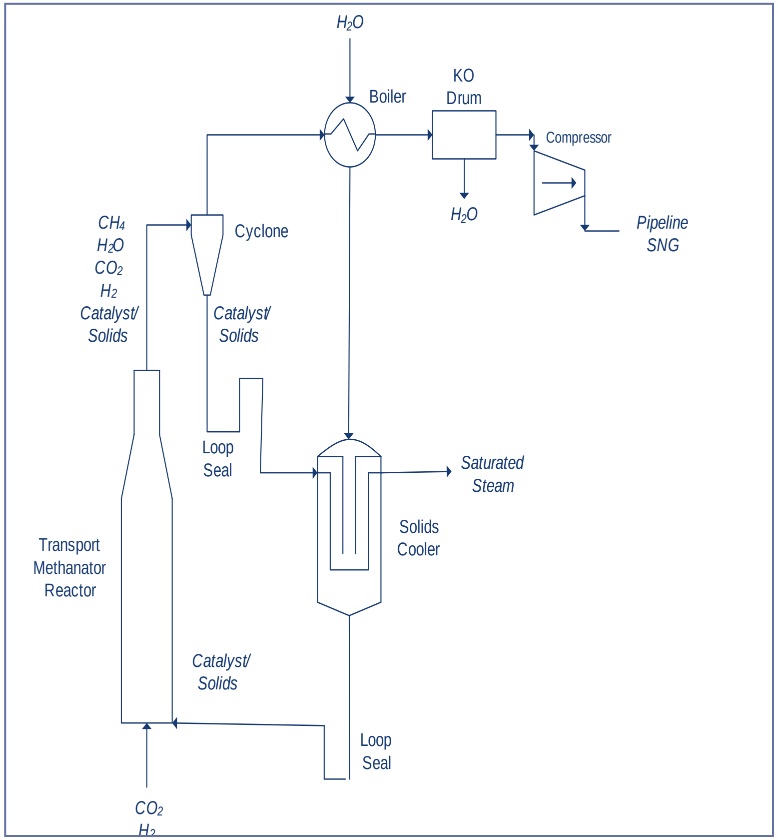
$CO_2 + 4H_2 = CH_4 + 2H_2O \quad \Delta H = -165 kJ/mol\ CO_2$

Current Process Technology

- Multiple reactors and heat exchangers in series
- Reactant gas dilution with product (recycle product)

Innovation: Improved single-pass conversion

- Improved management of exothermic reaction enthalpy
- Highly-active, attrition resistant, fluidized catalyst

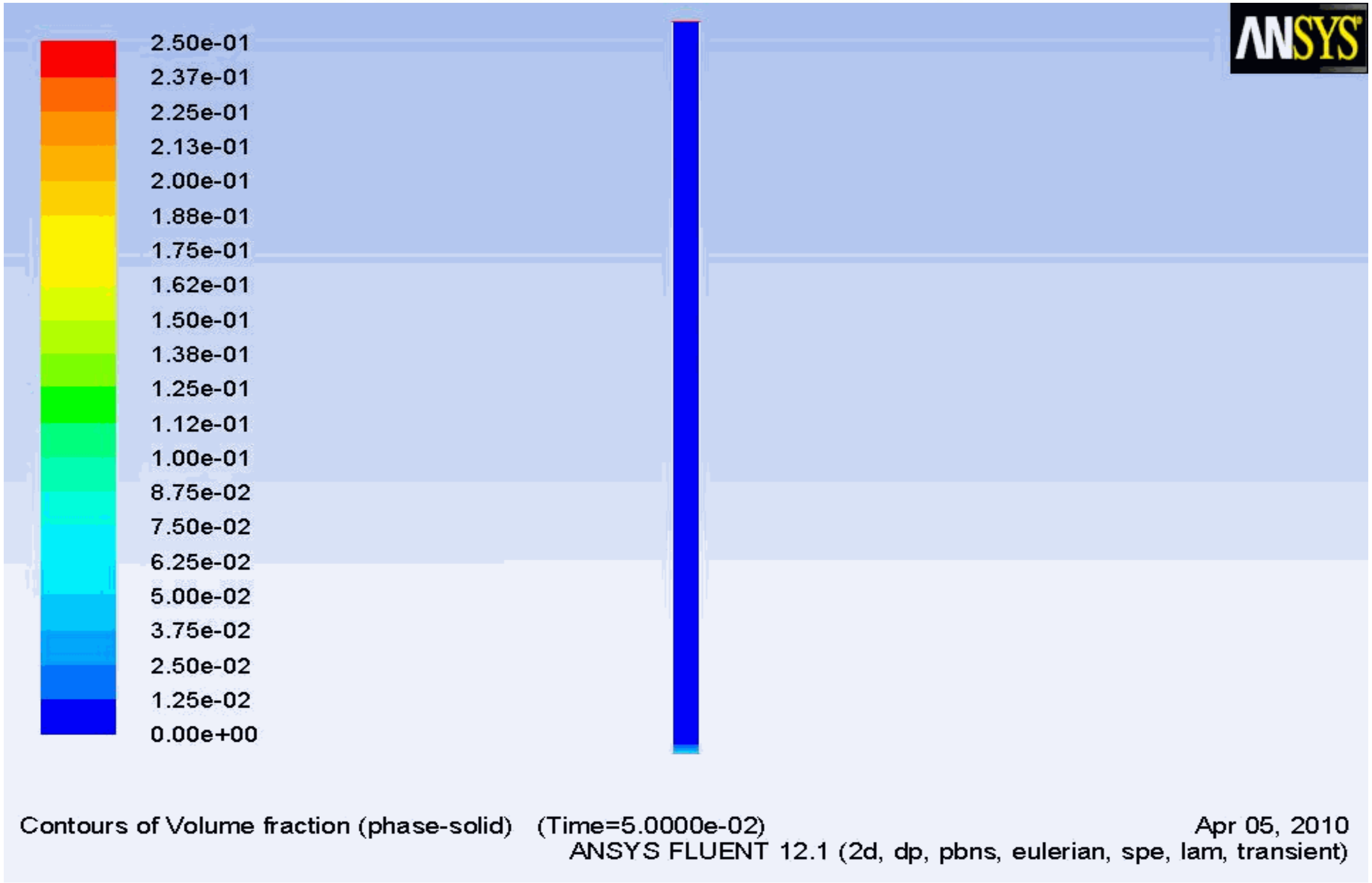


CFD Modeling Opportunities

Optimize single pass conversion through modeling of

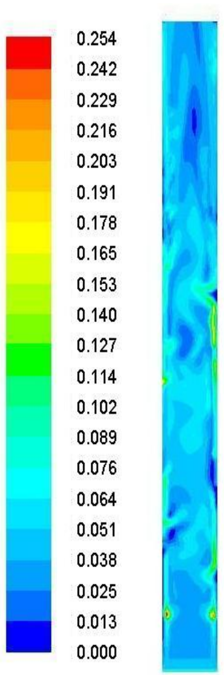
- Hydrodynamic properties
- Reaction kinetics,
- Heat and mass transfer

Catalyst Distribution in Reactor
(12” ID & 60 ft High)

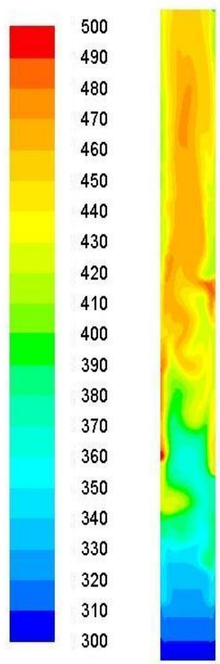


Spatial Profiles of Various Bed Properties
(12” ID & 60 ft High)

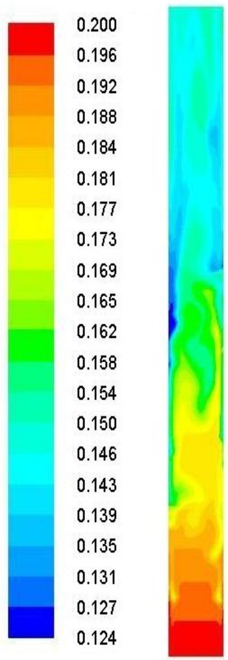
Catalyst volume fraction



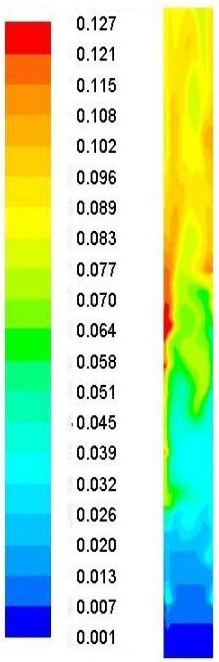
Gas Temperature (°C)



CO_2 Mole Fraction



CH_4 Mole Fraction



25% CO_2 conversion with less than 150°C

Summary

- RTI is actively engaged in RD&D of a number of fluidized-bed processes at different stages of development (bench-scale through large prototype-scale)
- RTI is developing CFD capabilities to assist in these RD&D efforts
- RTI's goal is to couple CFD modeling with actual test results to enhance research efforts
  - Reactor design issues for HTDP
  - Desulfurization performance based hydrodynamics and reaction kinetics for HTDP
  - Heat transfer for the RTI's dry carbonate process
  - Solids handling, pressure drop and gas/solid distribution for dry carbonate process
  - Heat management for Sabatier-base CO_2 reuse process

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Governing Equations

Continuity equations (g-gas, s-solid):

$$\frac{\partial(\rho_g \varepsilon_g)}{\partial t} + \nabla \cdot (\rho_g \varepsilon_g \vec{\upsilon}_g) = \sum_{n=1}^{N} R_{gn}$$

$$\frac{\partial(\rho_s \varepsilon_s)}{\partial t} + \nabla \cdot (\rho_s \varepsilon_s \vec{\upsilon}_s) = \sum_{n=1}^{N} R_{sn}$$

Momentum Equations:

$$\frac{\partial(\rho_g \varepsilon_g \vec{\upsilon}_g)}{\partial t} + \nabla \cdot (\rho_g \varepsilon_g \vec{\upsilon}_g \vec{\upsilon}_g) = \varepsilon_g \rho_g \vec{g} - \varepsilon_g \nabla P + \nabla \cdot \overline{\overline{\tau}}_g + \beta(\vec{\upsilon}_s - \vec{\upsilon}_g) - R_{gs}(\xi_{gs} \vec{\upsilon}_s + \overline{\xi}_{gs} \vec{\upsilon}_g)$$

$$\frac{\partial(\rho_s \varepsilon_s \vec{\upsilon}_s)}{\partial t} + \nabla \cdot (\rho_s \varepsilon_s \vec{\upsilon}_s \vec{\upsilon}_s) = \varepsilon_s \rho_s \vec{g} - \varepsilon_s \nabla P - \nabla P_s + \nabla \cdot \overline{\overline{\tau}}_s + \beta(\vec{\upsilon}_g - \vec{\upsilon}_s) + R_{gs}(\xi_{gs} \vec{\upsilon}_s + \overline{\xi}_{gs} \vec{\upsilon}_g)$$

Fluctuating energy equation:

$$\frac{3}{2}\left[\frac{\partial(\rho_s \varepsilon_s \theta)}{\partial t} + \nabla \cdot (\rho_s \varepsilon_s \theta \vec{\upsilon}_s)\right] = \left(-P_s \overline{\overline{\mathrm{I}}} + \overline{\overline{\tau}}_s\right) : \nabla \vec{\upsilon}_s + \nabla \cdot (\kappa_s \nabla \theta) - \gamma + \Pi_s$$

Constitutive Equations

Solid pressure:

$$P_s = \varepsilon_s \rho_s \theta \left[1 + 2(1+e)\varepsilon_s g_0\right]$$

Stress tensor:

$$\vec{\vec{\tau}}_g = \mu_g \left(\nabla \vec{\upsilon}_g + \nabla \vec{\upsilon}_g^{\,T}\right) - \frac{2}{3}\mu_g \nabla \cdot \vec{\upsilon}_g \vec{\vec{\mathrm{I}}} \qquad \vec{\vec{\tau}}_s = \mu_s \left(\nabla \vec{\upsilon}_s + \nabla \vec{\upsilon}_s^{\,T}\right) + \left(\xi_s - \frac{2}{3}\mu_s\right)\nabla \cdot \vec{\upsilon}_s \vec{\vec{\mathrm{I}}}$$

Viscosity:

MFIX:

$$\mu_s = \left(\frac{2+\alpha}{3}\right)\left[\frac{\mu_s^*}{g_0\eta(2-\eta)}\left(1+\frac{8}{5}\eta\varepsilon_s g_0\right)\left(1+\frac{8}{5}\eta(3\eta-2)\varepsilon_s g_0\right)+\frac{3}{5}\eta\mu_b\right]$$

$$\mu_s^* = \frac{\rho_s \varepsilon_s g_0 \theta \mu}{\rho_s \varepsilon_s g_0 \theta + \left(\frac{2\beta\mu}{\rho_s \varepsilon_s}\right)} \qquad \mu = \frac{5}{96}\rho_s d_p \sqrt{\pi\theta} \qquad \xi_s = \frac{256}{5\pi}\eta\mu\varepsilon_s^{\,2} g_0$$

FLUENT:

$$\mu_s = \frac{4}{5}\varepsilon_s^{\,2}\rho_s d_p g_0 (1+e)\left(\frac{\theta}{\pi}\right)^{1/2} + \frac{10\rho_s d_p \varepsilon_s \sqrt{\theta\pi}}{96(1+e)g_0}\left[1+\frac{4}{5}g_0\varepsilon_s(1+e)\right]^2$$

$$\xi_s = \frac{4}{3}\varepsilon_s^{\,2}\rho_s d_p g_0 (1+e)\left(\frac{\theta}{\pi}\right)^{1/2}$$

Constitutive Equations (Contd..)

Granular conductivity:

MFIX:

$$\kappa_s = \frac{\kappa_s^*}{g_0}\left[\left(1+\frac{12}{5}\eta\varepsilon_s g_0\right)\left(1+\frac{12}{5}\eta^2(4\eta-3)\varepsilon_s g_0\right)+\frac{64}{25\pi}(41-33\eta)\eta^2\varepsilon_s{}^2 g_0{}^2\right]$$

$$\kappa_s^* = \frac{\rho_s\varepsilon_s g_0\theta\kappa}{\rho_s\varepsilon_s g_0\theta+\frac{6\beta\kappa}{5\rho_s\varepsilon_s}} \qquad \kappa = \frac{75\rho_s d_p\sqrt{\pi\theta}}{48\eta(41-33\eta)}$$

FLUENT:

$$\kappa_s = \frac{150\rho_s d_p\varepsilon_s\sqrt{\theta\pi}}{384(1+e)g_0}\left[1+\frac{6}{5}\varepsilon_s g_0(1+e)\right]^2+2\rho_s\varepsilon_s{}^2 d_p(1+e)g_0\sqrt{\frac{\theta}{\pi}}$$

Constitutive Equations (Contd..)

Collisional dissipation:

MFIX:

$$\gamma = \frac{12}{\sqrt{\pi}}\left(1-e^2\right)\frac{\varepsilon_s g_0}{d_p}\theta^{3/2}$$

FLUENT:

$$\gamma = 3\left(1-e^2\right)g_0\rho_s\varepsilon_s{}^2\theta\left(\frac{4}{d_p}\sqrt{\frac{\theta}{\pi}}-\nabla\cdot\vec{\upsilon}_s\right)$$

Exchange term:

$$\Pi_s = -3\beta\theta + \frac{81\varepsilon_s\mu_g{}^2\left|\vec{\upsilon}_g-\vec{\upsilon}_s\right|^2}{g_0 d_p{}^3\rho_s\sqrt{\pi\theta}}$$

Radial distribution function:

MFIX:

$$g_0 = \frac{1}{\varepsilon_g} + \frac{3\left(\sum_{\lambda=1}^{M}\varepsilon_\lambda / d_{p\lambda}\right)d_{pl}d_{pm}}{\varepsilon_g^2\left(d_{pl}+d_{pm}\right)}$$

FLUENT:

$$g_0 = \left[1-\left(\frac{\varepsilon_s}{\varepsilon_{s,\max}}\right)^{1/3}\right]^{-1}$$

Species and Energy Balance and Constitutive models

Species balance equations:

$$\frac{\partial\left(\rho_g \varepsilon_g X_{gn}\right)}{\partial t} + \nabla \cdot \left(\rho_g \varepsilon_g \vec{\upsilon}_g X_{gn}\right) = R_{gn}$$

$$\frac{\partial\left(\rho_s \varepsilon_s X_{sn}\right)}{\partial t} + \nabla \cdot \left(\rho_s \varepsilon_s \vec{\upsilon}_s X_{sn}\right) = R_{sn}$$

Energy balance equations:

$$\varepsilon_g \rho_g C_{pg}\left[\frac{\partial T_g}{\partial t} + \vec{\upsilon}_g \cdot \nabla T_g\right] = \cancel{-\varepsilon_g \frac{\partial P}{\partial t}} + \overline{\overline{\tau}}_g : \nabla \vec{\upsilon}_g + \nabla \cdot \left(k_g \nabla T_g\right) + \gamma_{gs}\left(T_s - T_g\right) - \Delta H_g + \gamma_{Rg}\left(T_{Rg}^4 - T_g^4\right)$$

$$\varepsilon_s \rho_s C_{ps}\left[\frac{\partial T_s}{\partial t} + \vec{\upsilon}_s \cdot \nabla T_s\right] = \cancel{-\varepsilon_s \frac{\partial P}{\partial t}} + \overline{\overline{\tau}}_s : \nabla \vec{\upsilon}_s + \nabla \cdot \left(k_s \nabla T_s\right) + \gamma_{gs}\left(T_g - T_s\right) - \Delta H_s + \gamma_{Rs}\left(T_{Rs}^4 - T_s^4\right)$$

$$\gamma_{gs} = \frac{C_{pg} R_{os}}{\exp\left(C_{pg} \, {R_{0s}} / {\gamma_{gs}^0}\right) - 1}$$

$$\varepsilon_g \rho_g C_{pg}\left[\frac{\partial T_g}{\partial t} + \vec{\upsilon}_g \cdot \nabla T_g\right] = \overline{\overline{\tau}}_g : \nabla \vec{\upsilon}_g + \nabla \cdot \left(k_g \nabla T_g\right) - \Delta H_g$$

$$\varepsilon_s \rho_s C_{ps}\left[\frac{\partial T_s}{\partial t} + \vec{\upsilon}_s \cdot \nabla T_s\right] = \overline{\overline{\tau}}_s : \nabla \vec{\upsilon}_s + \nabla \cdot \left(k_s \nabla T_s\right) - \Delta H_s$$

RTI's Dry Carbonate Process

Support for Development of 1 TPD CO_2 Pilot Unit

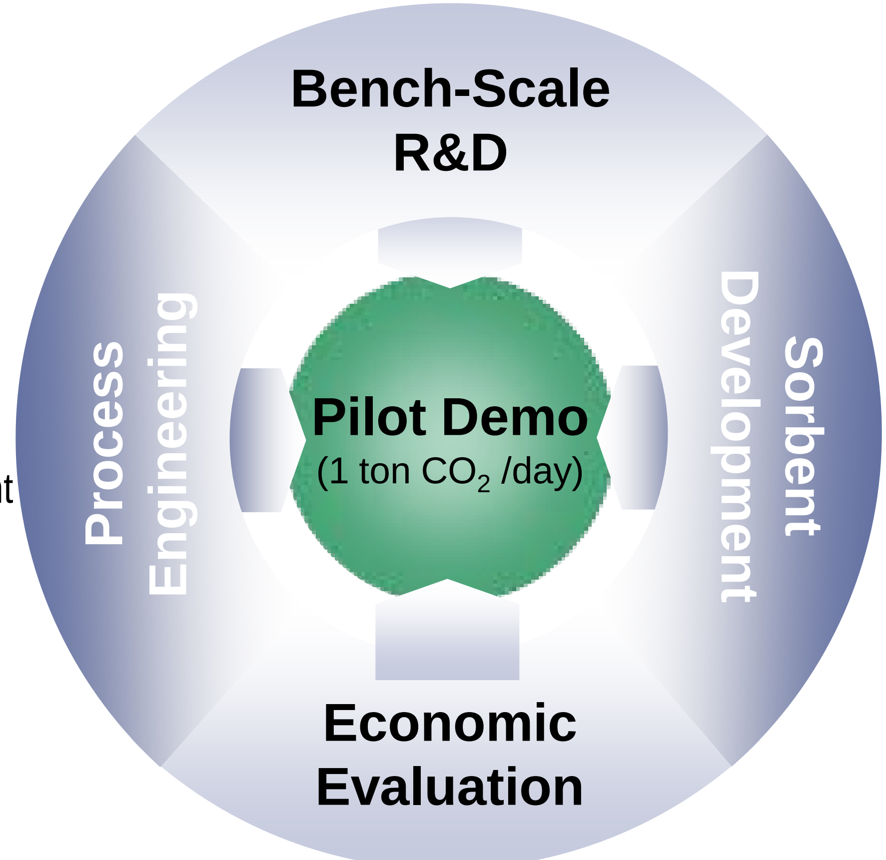
Fluidized, Moving-bed, Heat Transfer Unit

- Designed to address uncertainties in the 1TPD design
- Develop a fundamental understanding of how heat transfer internals affect the hydrodynamic and heat transfer characteristics of a moving, fluidized-bed reactor
- Hydrodynamic Properties
  - ΔP across bed and process, gas-solids mixing
  - Regions of operability (solids and gas flow)
  - Gas-solids separation
- Heat Transfer Characteristics
  - Effect of gas and solids flow rates on overall heat transfer coefficient
  - Effect of tube layout on overall heat transfer coefficient

RTI's Dry Carbonate Process

Current R&D Efforts – 1 TPD CO_2 scale Testing

Process and Sorbent Evaluation Efforts



Fluidized, Moving-Bed

- Existing technology
- Effective heat transfer
- Good gas-solid contact
- Acceptable ΔP
- Sufficient Residence time to load ~20 wt% CO_2 on sorbent

Solids Handling

- Move 10^6s lb/h
- Existing technology

Engineered-Na_2CO_3

- Inexpensive materials
- High CO_2 loadings
- Maintain high reactivity
- Acceptable physical properties
  - Attrition resistance
  - Particle density
  - Surface area

Internal/External Cost and Performance Evaluations

Basis for CFD Simulations

Reaction: $CO_2+4H_2=CH_4+2H_2O$

Kinetic equation (r: mmoles/g h kPa; P: kPa):

$$r = k_1 P_{CO_2} P_{H_2}^{0.5} / \left(P_{H_2}^{0.5} + k_2 P_{CO_2}\right)$$

$$k_1 = 1.46E9 \exp(-9460/T)$$

$$k_2 = 1.18E-3 \exp(3710/T)$$

Heat of reaction: -165 kJ/mol

Transport reactor:

Diameter: 2” – 12”

Height: 60 ft

Material property:

Gas: Incompressible ideal gas

Catalyst:

Particle size: 80 µm

Density: 1600 kg/m3

Specific heat: 940 j/kg k

Thermal conductivity: 130 W/m K

Boundary conditions:

Wall:

Gas: No slip

Catalyst: Johnson Jackson partial slip

No heat flux

Inlet:

Mass flow rate: 448.378 lb/hr

Mass ratio (solid : gas) = 10 :1

Gas composition (mole fraction):

CO_2= 0.1851; H_2= 0.7317; CH_4= 0.0832

Gas temperature: 250°C

Catalyst temperature: 250°C

Outlet:

Prescribed pressure: 314.70 psi

Initial conditions:

Temperature: 300°C or 250°C

No catalyst

Gas velocity: 0

Gas composition (mole fraction):

CO_2= 0.1851; H_2= 0.7317; CH_4= 0.0832

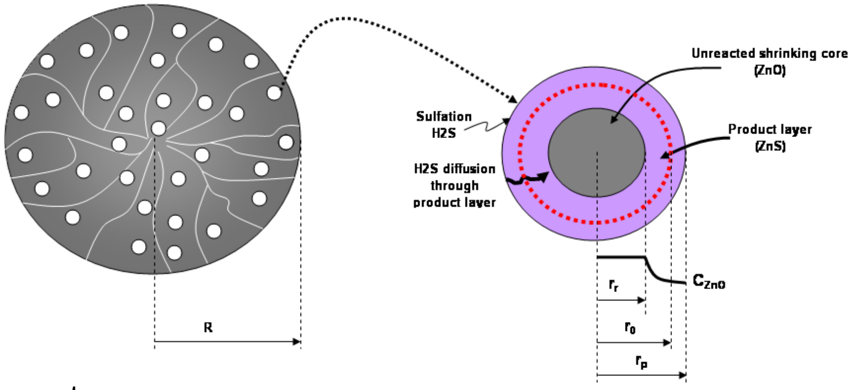
Development of CFD Model for HTDP

DOE/NETL CRADA tasks

- Develop a CFD model
- Validate CFD model with 0.3 MW_e HTDP pilot plant data from Eastman testing
- Use validated model to optimize design of 50 MW_e demonstration HTDP system

Model Development

- Reaction Models
  - Overlapping grain model
  - Arrhenius rate expression
  - H_2S diffusion in RTI-3 pore structure
  - Knowledge of RTI-3 pore structure
- Fluid Dynamics (Fluent)
  - Continuity, momentum, and energy for each phase
  - Coupling phases achieved through inter-phase exchange terms
  - Mass transfer between phases modeled through heterogeneous reaction schemes
- System Geometry



RTI INTERNATIONAL 50 YEARS