

Chemical Looping Technology for Hydrogen Production: Commercialization Prospect

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Ohio Hydrogen Technology Forum

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- World population → **10 bn** by 2050, requiring **15% higher energy**

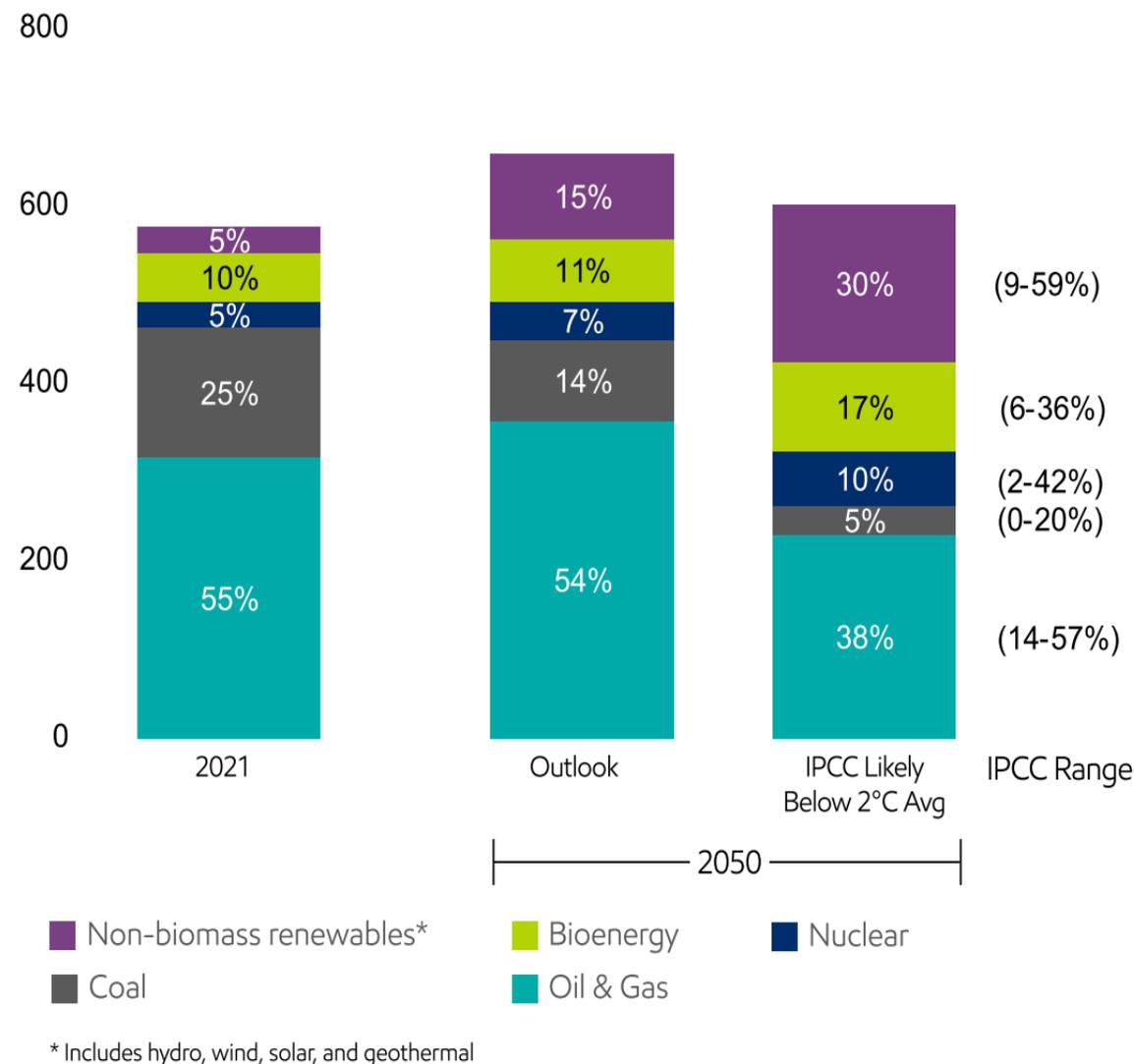
- Low-carbon energy technologies

- Three main technologies that have been identified to curb climate change to 2°C by 2050 are **Carbon Capture and Storage, Hydrogen, Biofuels**

- Chemical Looping:** a promising technology for reducing CO₂ emissions

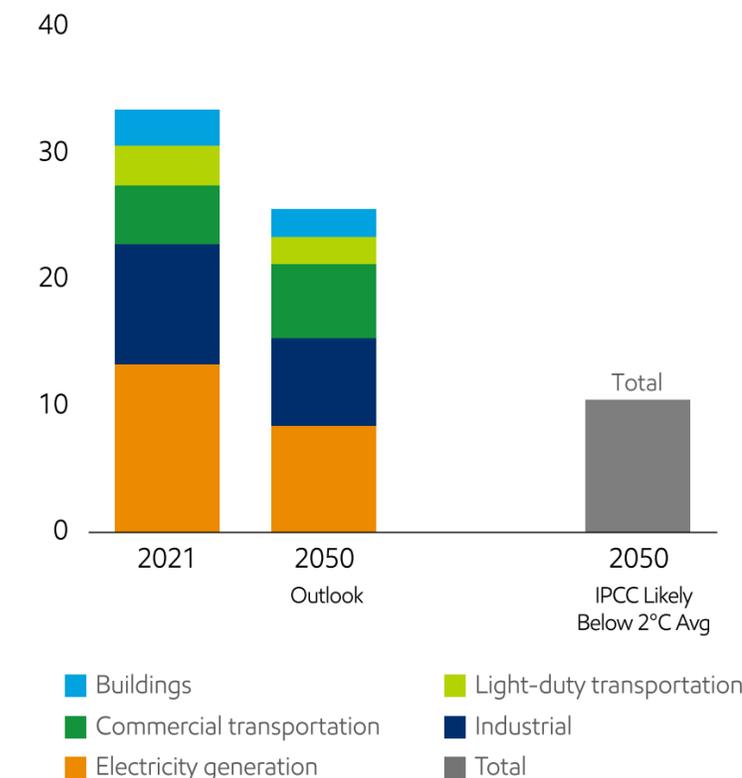
Global energy mix

Quadrillion Btu



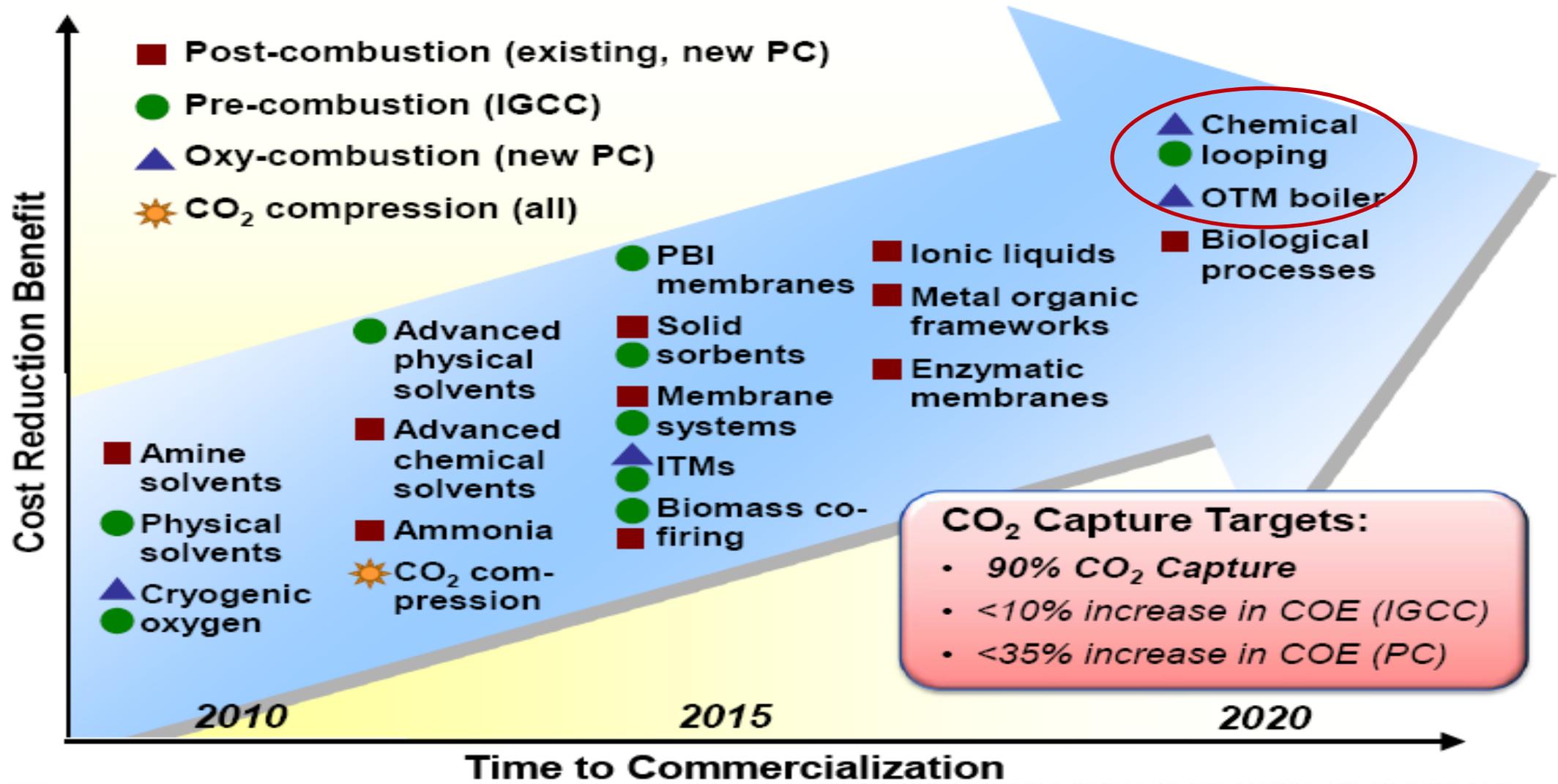
Energy-related emissions

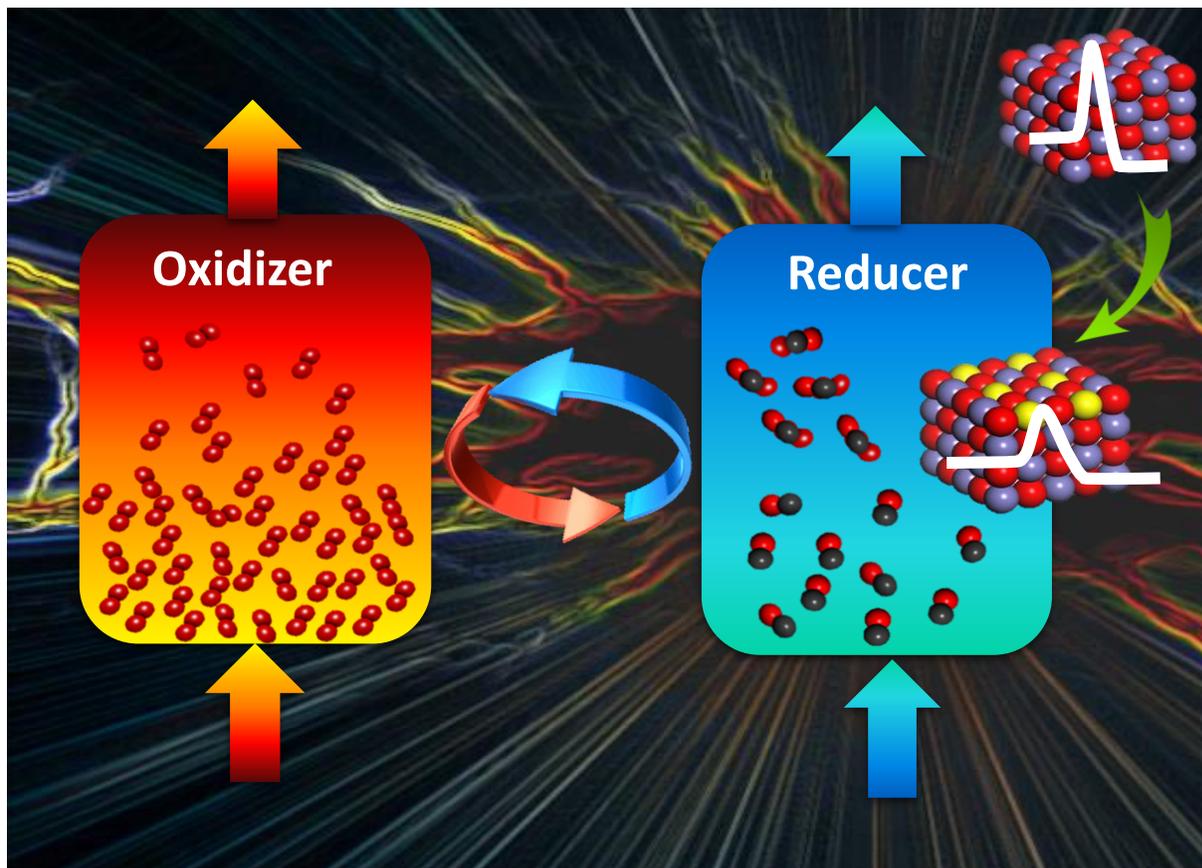
CO₂ Billion metric tons



CO₂ Capture from Fossil Energy

Technological Solutions





As featured in Journal of Materials
Chemistry A, Advance Article, 2017
DOI:10.1039/C7TA04228K

Showcasing a new
approach on improving the
reactivity of iron oxide
oxygen carriers using a
very small concentration of
the lanthanum dopant by
Professor Liang-Shih Fan's
research group at the Ohio
State University.

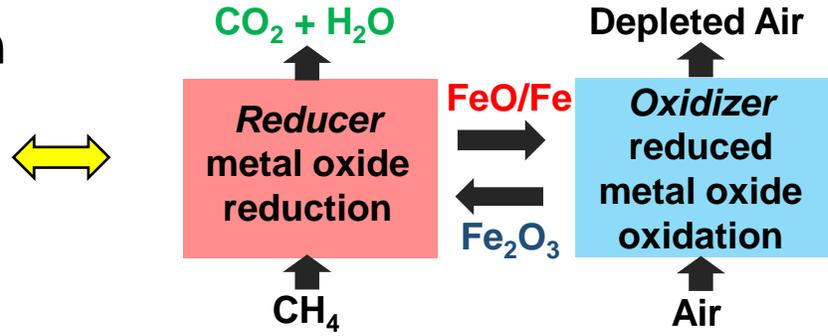
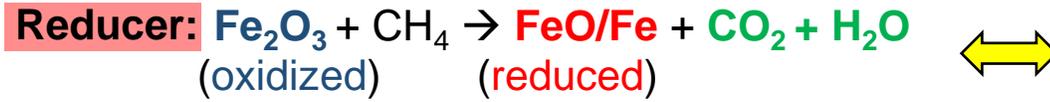
Title: Improved cyclic redox reactivity of lanthanum modified iron-based oxygen carriers in carbon monoxide chemical looping combustion

Oxygen carriers are required to have high reactivity and recyclability with low cost. A very low concentration of the lanthanum dopant can dramatically increase the reactivity of oxygen carriers in chemical looping combustion with carbonaceous fuels by reducing the reaction barriers. This methodology provides substantial performance improvements of oxygen carriers that are relatively simple to fabricate, and it will have an impact on chemical looping particle design and modification.

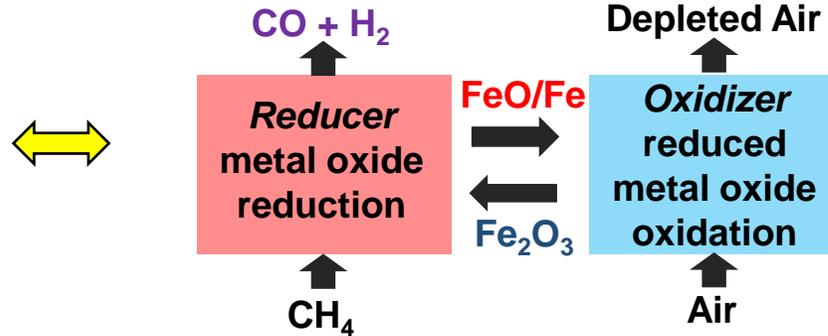
Redox Chemical Looping Technology

Metal Oxide as Oxygen Carrier

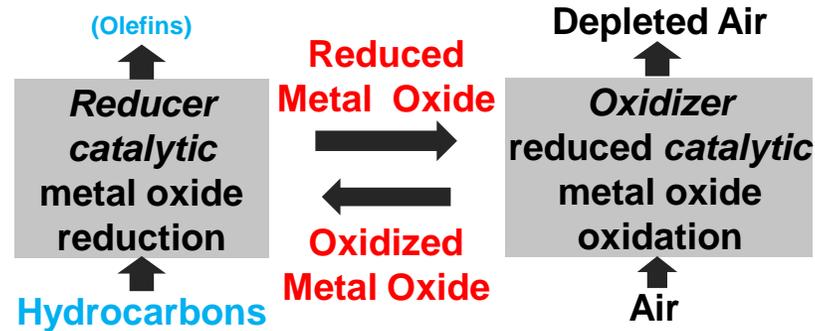
Combustion: Complete Fuel Oxidation



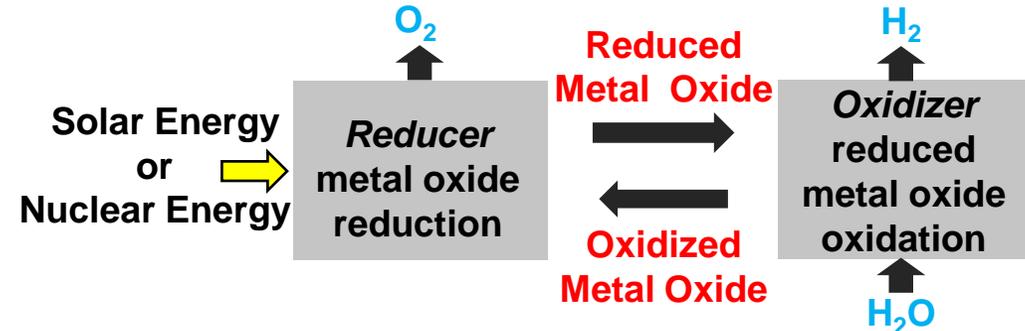
Gasification: Partial Fuel Oxidation



Chemicals



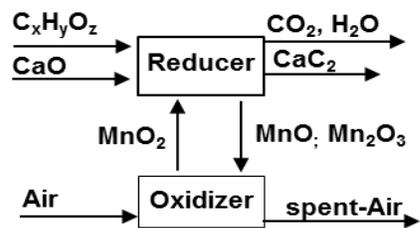
Chemicals Production: Selective Oxidation



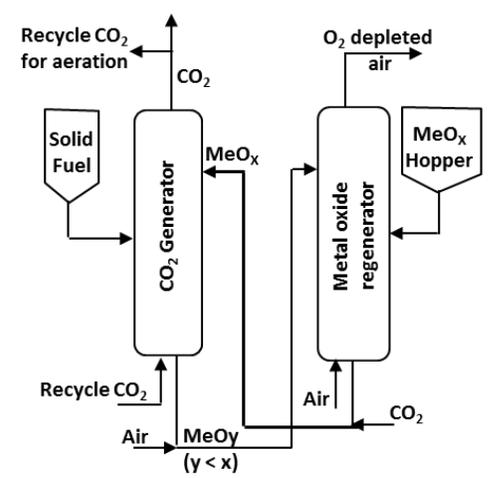
Solar/Nuclear Chemical Looping: Water Splitting

History of Redox of Chemical Looping Technology development

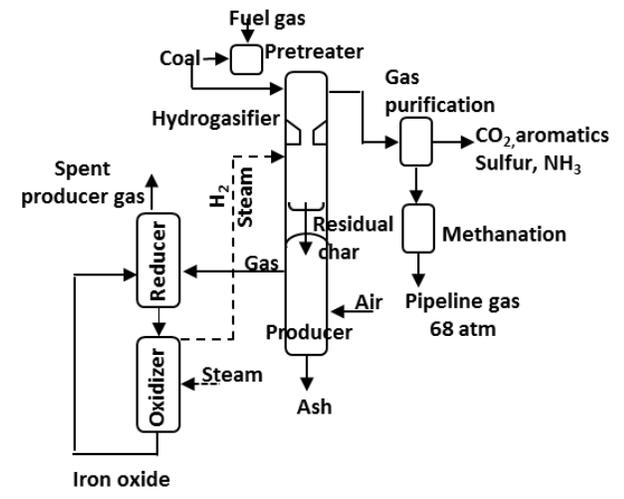
Technologies	Bergmann Process	Lane Process & Messerschmitt Process	Lewis and Gilliland Process	IGT HYGAS Process	CO ₂ Acceptor Process
Time	1897	1910	1950s	1970s	1970s
Looping Media	MnO ₂ /MnO/Mn ₂ O ₃	Fe/FeO/Fe ₃ O ₄	Cu ₂ O/CuO	FeO/Fe ₃ O ₄	CaO/CaCO ₃
Reactor Design	Blast Furnace	Fixed Bed	Fluidized Bed	Staged Fluidized Bed	Fluidized Bed



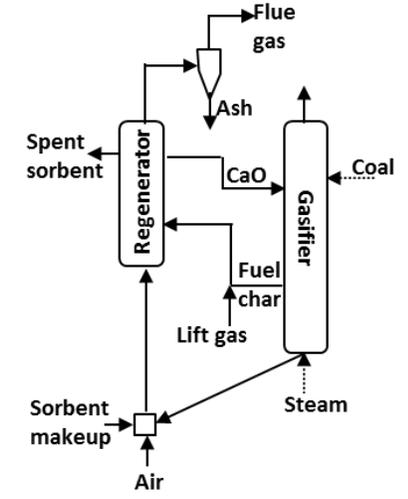
Bergmann Process



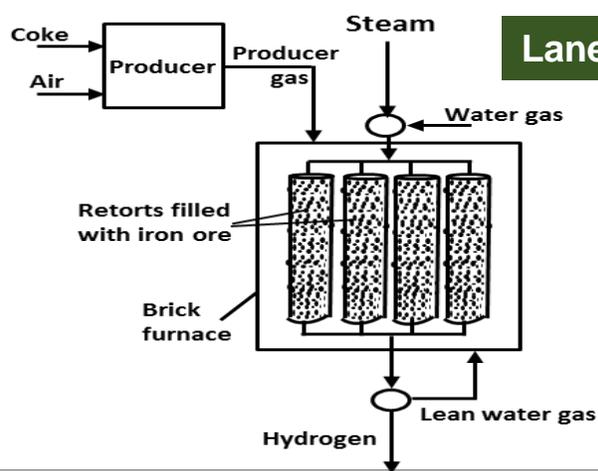
Lewis and Gilliland Process



IGT Process



CO₂ Acceptor Process

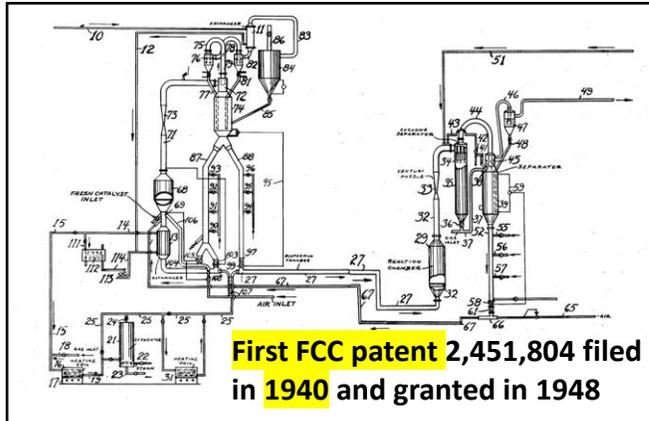


Lane Process

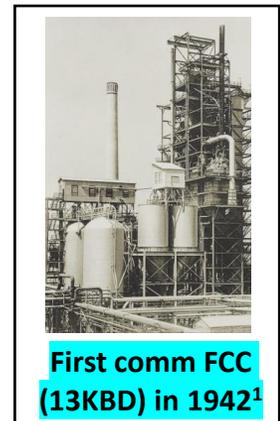


Fluid Catalytic Cracking (FCC) Process Development

2,3 Focused on catalyst improvement and additives development



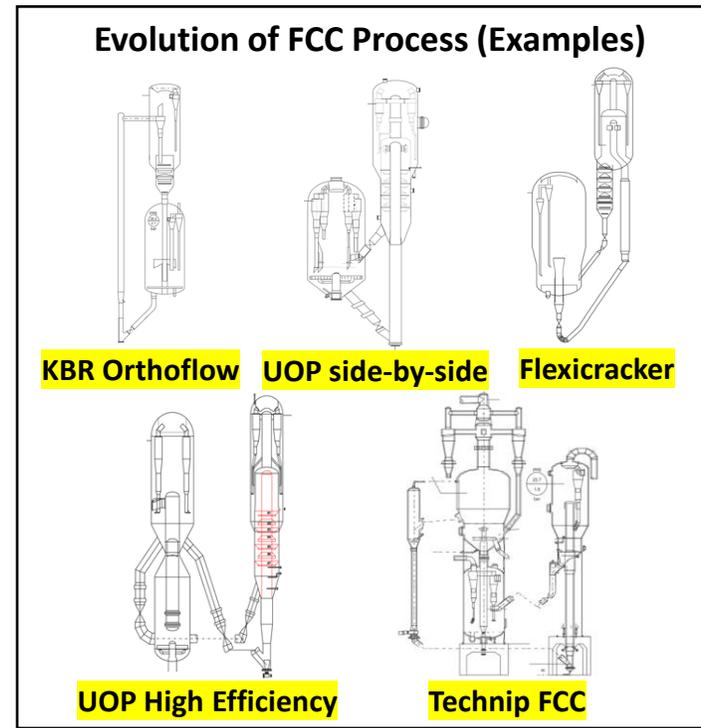
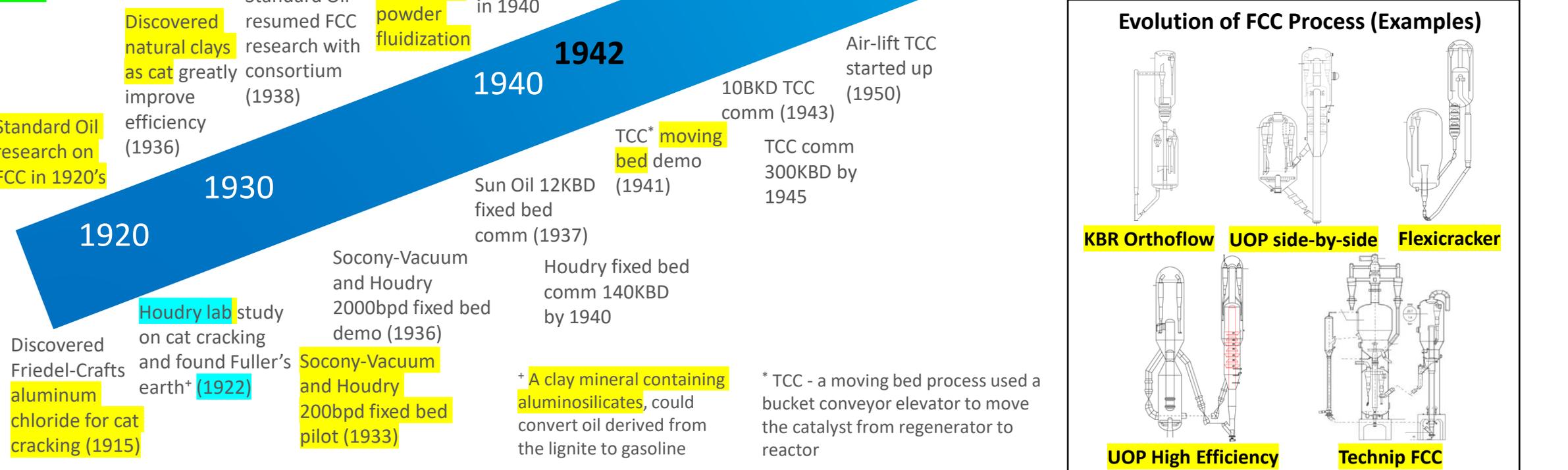
First FCC patent 2,451,804 filed in 1940 and granted in 1948



First comm FCC (13KBD) in 1942¹

- Reference:
1. A National Historical Chemical Landmark – The Fluid Bed Reactor, 1998
 2. Fluid Catalytic Cracking Handbook
 3. Wikipedia – Fluid Catalytic Cracking

Δt ≈ 20 years for first plant



* TCC - a moving bed process used a bucket conveyor elevator to move the catalyst from regenerator to reactor

+ A clay mineral containing aluminosilicates, could convert oil derived from the lignite to gasoline

Multi-Scale, Multiphase Technology

Development Approach

Powder Technology 439 (2024) 119654; LS Fan et al.,
 “Multiphase Entrepreneurship: An Academic Reflection”

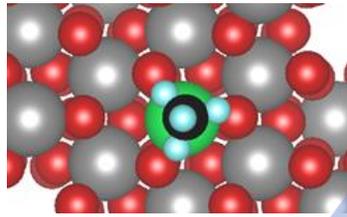
System scale (> 100m)

Reactor scale (m)

coupling of momentum, heat, mass transport

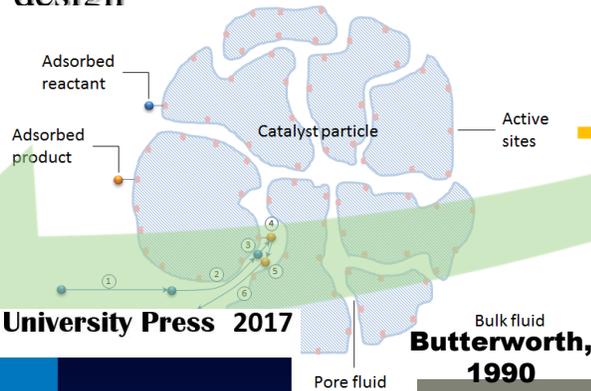
Particle, Droplet or Bubble scale ($\mu\text{m}\sim\text{mm}$)

external/internal diffusion, reaction, size and shape design



Molecular scale (\AA)

Surface chemistry, e.g., how chemical bonds are formed and cleaved. reaction mechanism, energetics, charge flow



Cambridge University Press 2017

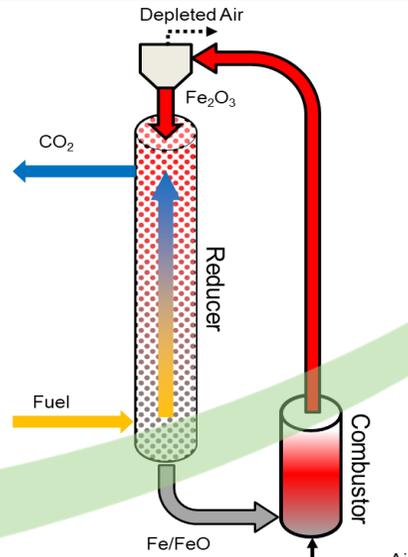
Butterworth, 1990

Cambridge University Press 1998

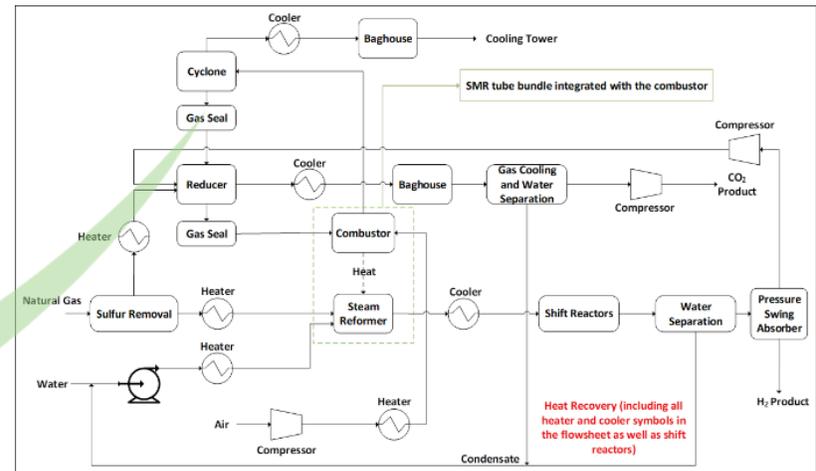
Butterworth, 1989

Cambridge University Press 2021

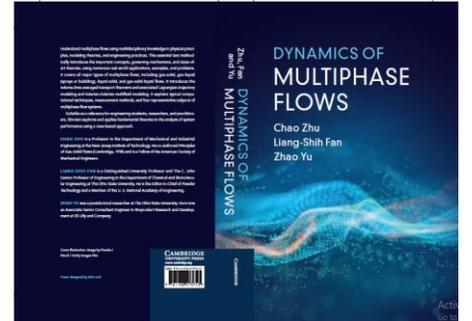
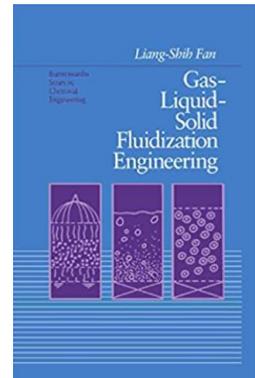
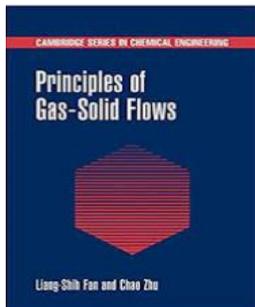
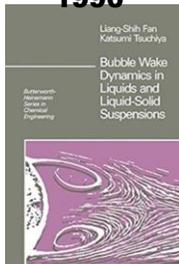
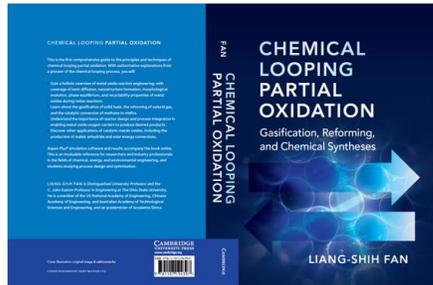
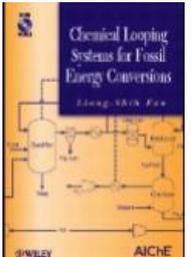
Counter-current: Full Combustion



Chemical Looping H₂ Process system

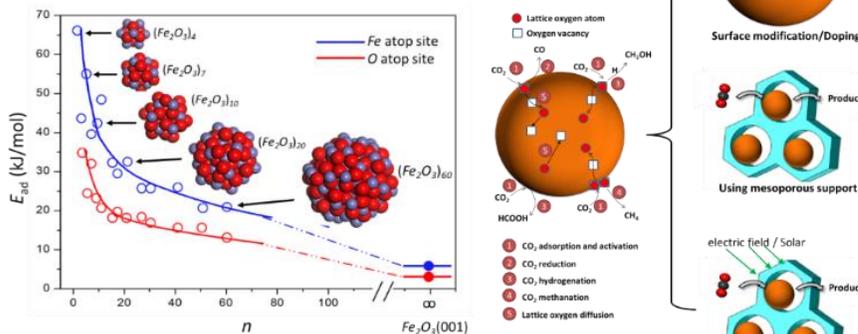


Wiley/AIChE 2010



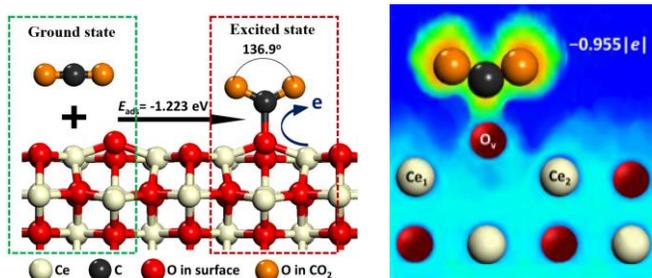
Molecular scale (Å)

Nanoparticle oxygen carrier adsorption energy

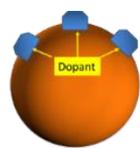


Nature Review Chemistry, 2018, 2, 349

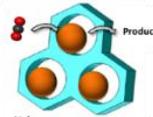
Molecular adsorption and charge transfer



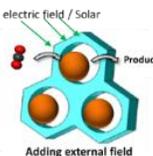
Nature Communications, 2019, 10, 1



Surface modification/Doping



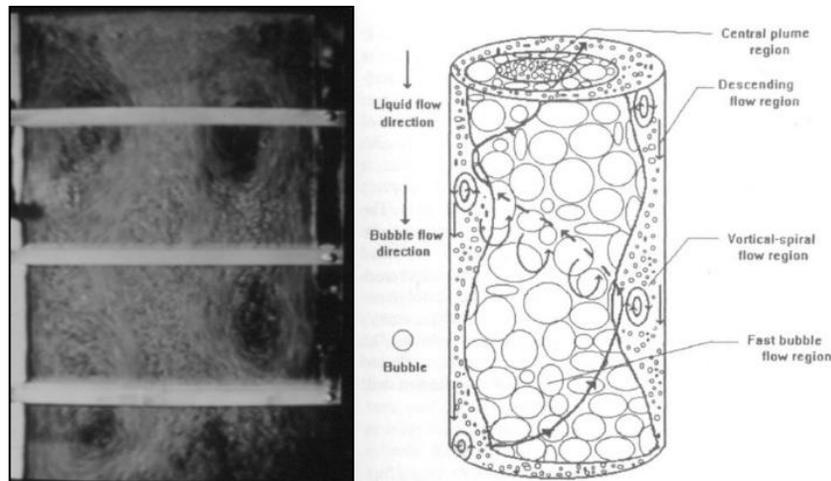
Using mesoporous support



Adding external field

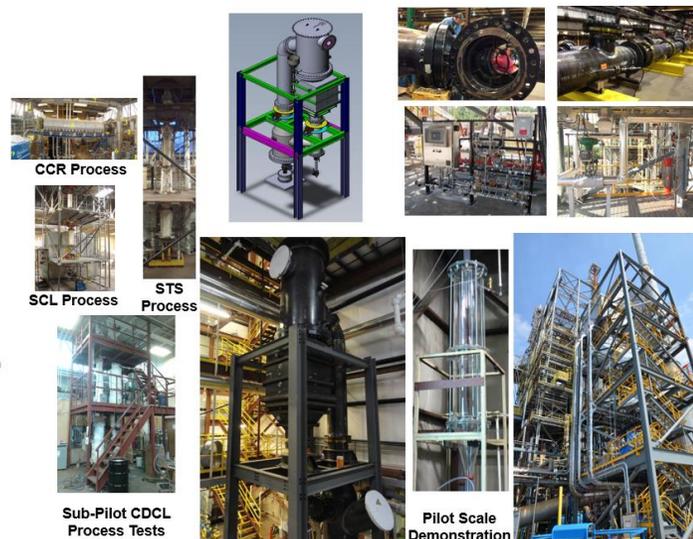
Reactor scale (m)

Flow structure of a 3D bubble column and 3D fluidized bed



AIChE Journal, 1994, 40, 1093

Sub-pilot and pilot scale demonstrations

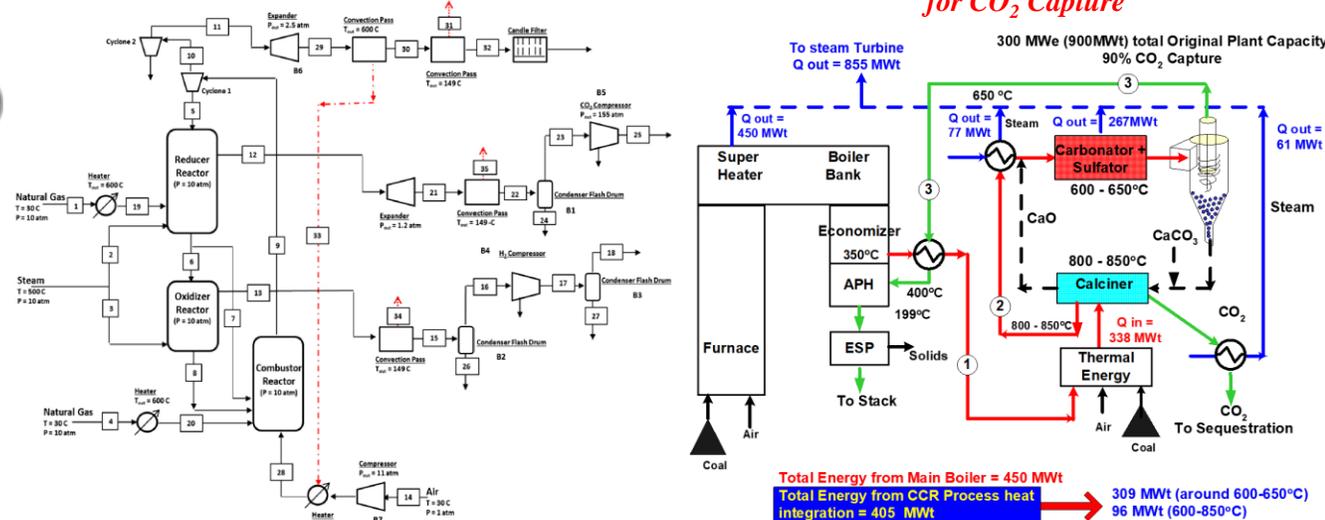


AIChE Journal, 2015, 61, 2

System scale (> 100m)

Process system analysis for H₂ production

Calcination-Carbonation-Reaction Process for CO₂ Capture



Applied Energy, 2016, 165, 183

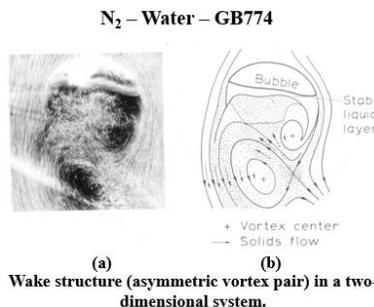
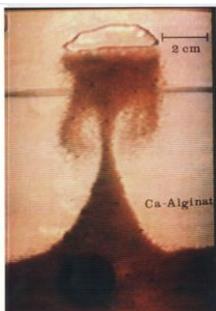
Total Energy from Main Boiler = 450 MWt
Total Energy from CCR Process heat integration = 405 MWt
309 MWt (around 600-650°C)
96 MWt (600-850°C)

Fuel, 2013, 104, 561

Particle, Droplet or Bubble scale (μm~mm)

Metal oxide redox chemistry

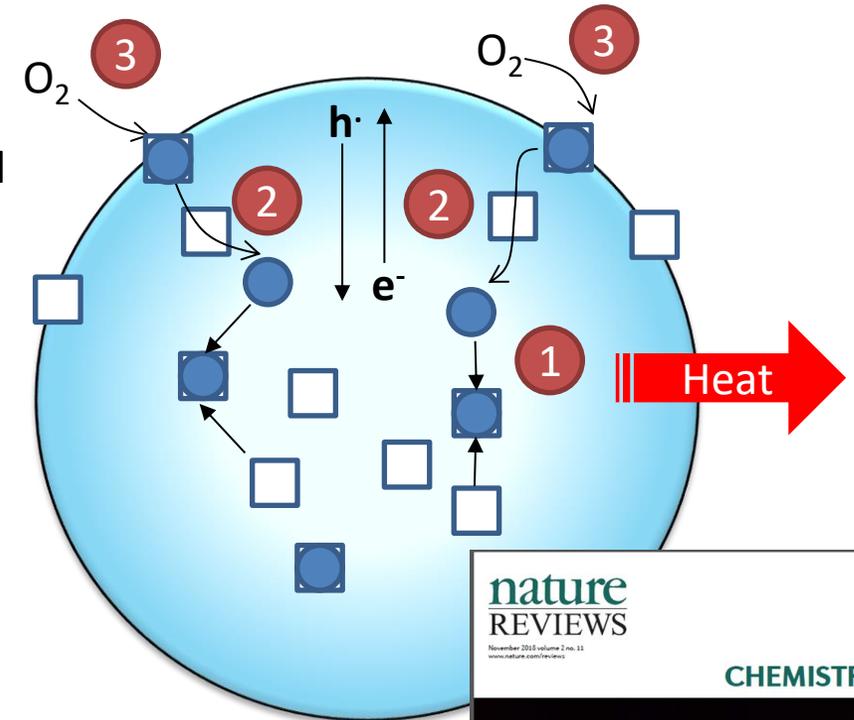
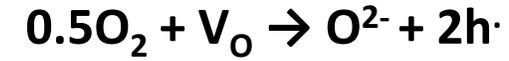
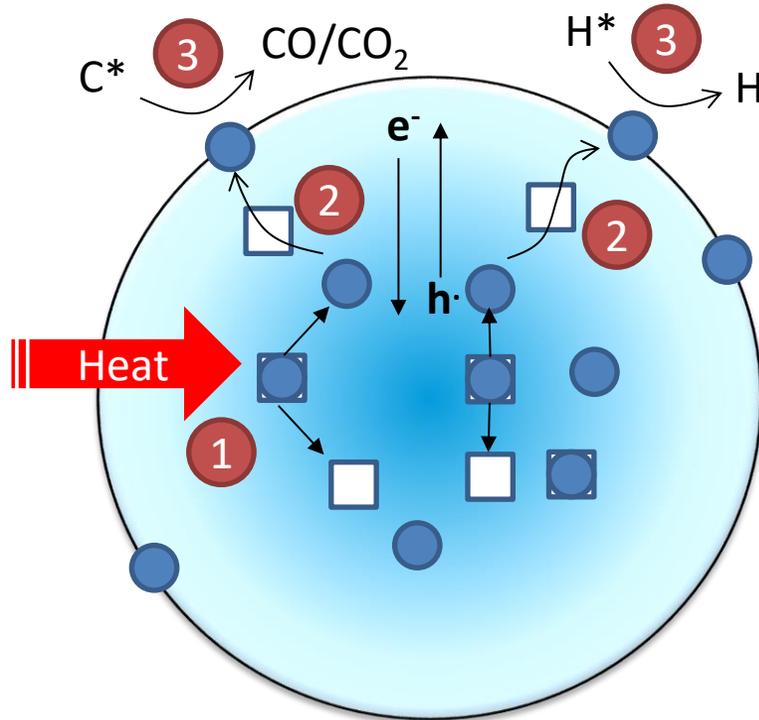
Bubble wake dynamics



Nature Reviews Chemistry, 2018, 2, 349

International J. of Multiphase Flow, 1990, 16, 187

Metal Oxide Redox Chemistry



Oxygen chemical potential /Oxygen partial pressure



Low

High

□ Oxygen vacancy, V_O

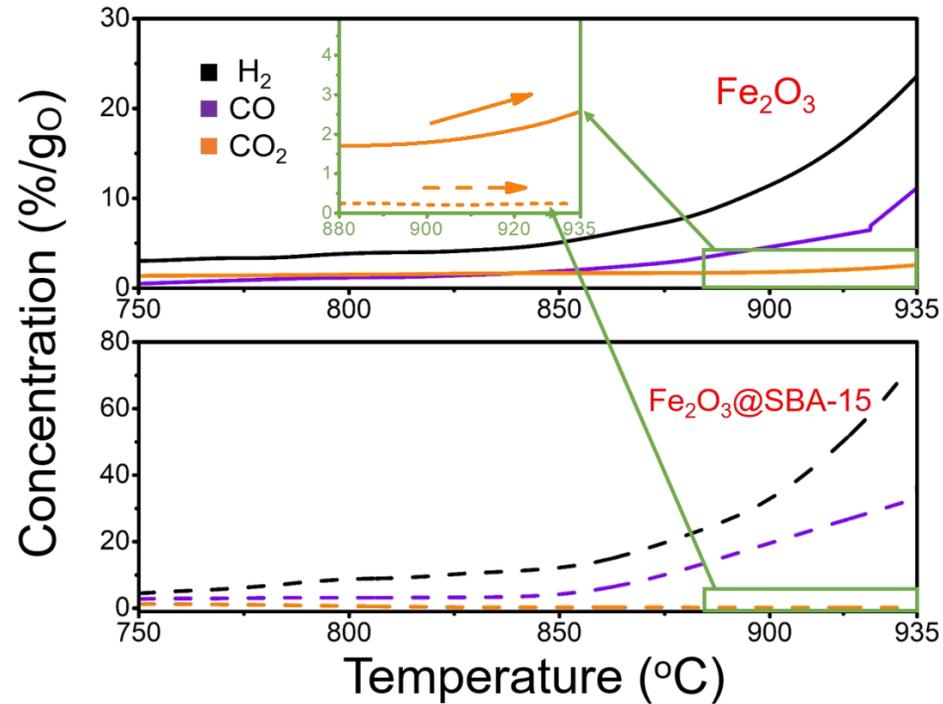
● Oxygen ion

1. Oxygen ion/vacancy generation
2. Bulk phase diffusion
3. Surface reaction

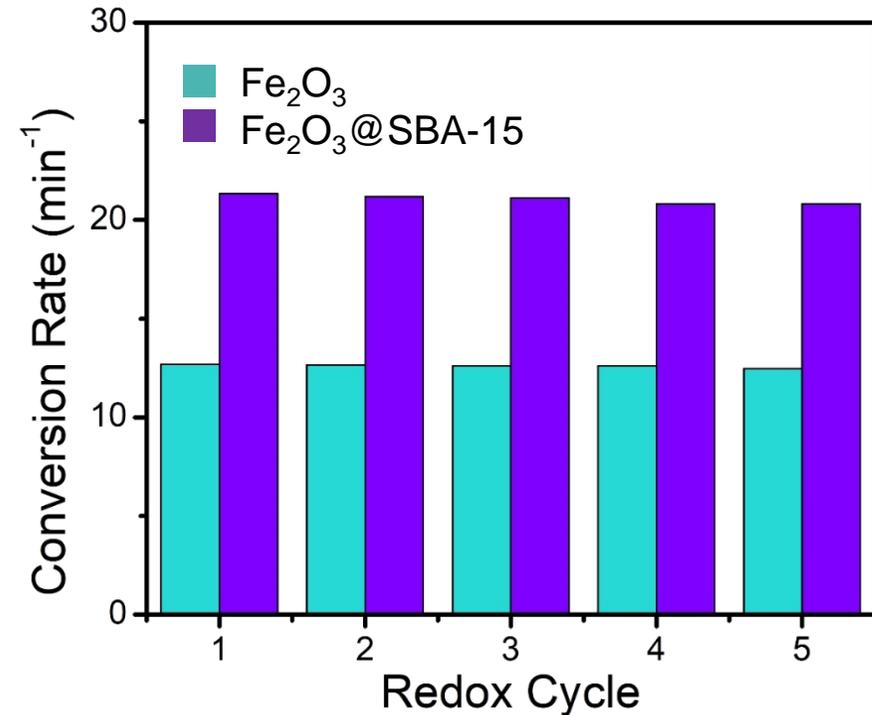


Oxygen Carrier Nanoparticles - $\text{Fe}_2\text{O}_3@SBA-15$

Temperature programmed reduction (TPR)



Conversion rate during redox at 800°C

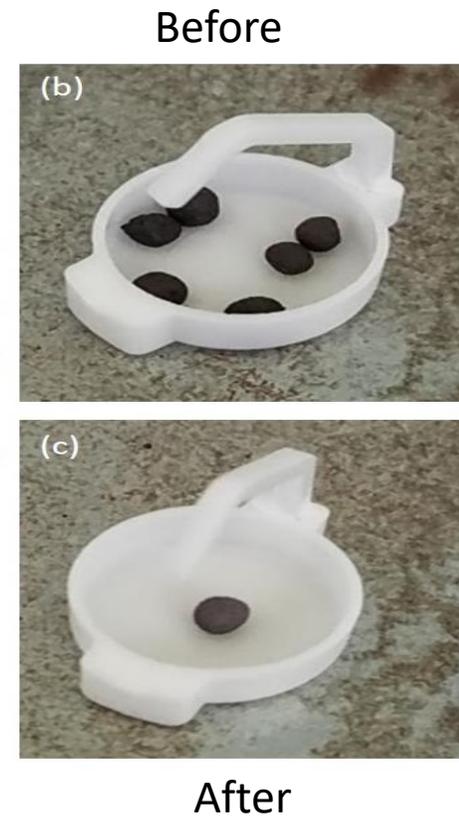
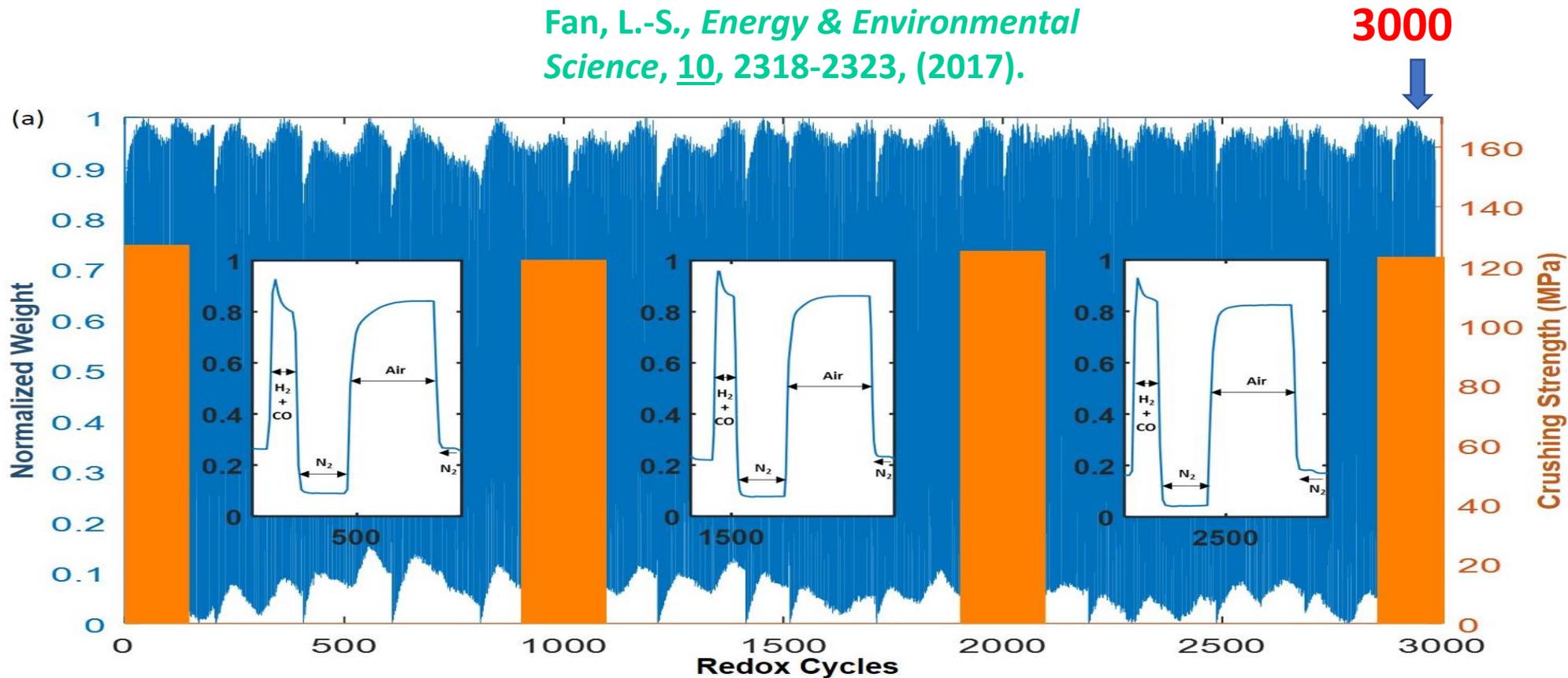


Fe_2O_3 nanoparticles@SBA-15 has a high CO selectivity of $\sim 100\%$, as well as a high reactivity, which is 66% higher than Fe_2O_3 particles.

Chemical Stability of OSU Oxygen Carrier – 3000 Redox Cycles

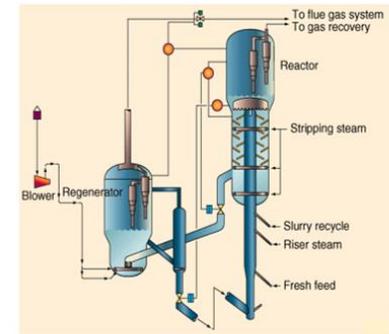
Both reactivity and strength (120 MPa) are sustained over 3000 redox cycles at 1000 °C with constant particle size of 1.5 mm

Chung, C., Qin, L., Shah, V., and Fan, L.-S., *Energy & Environmental Science*, 10, 2318-2323, (2017).



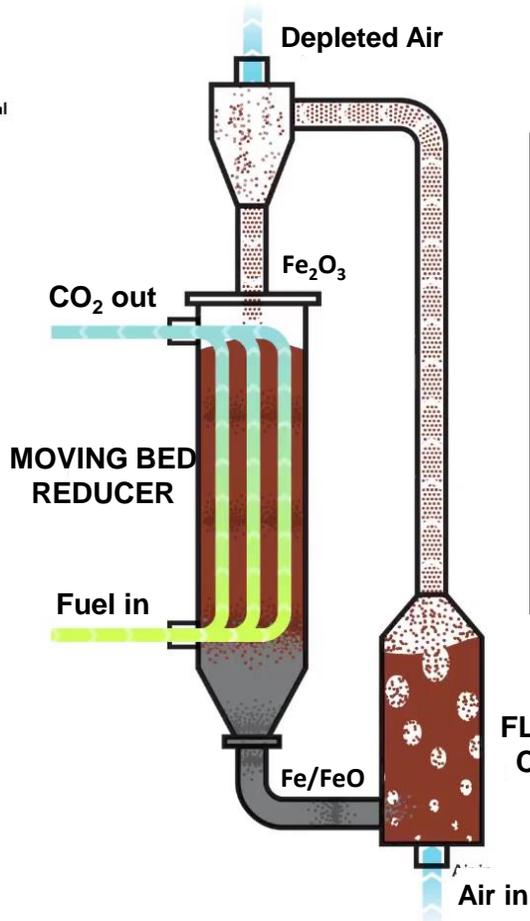
Latest results also indicates sustained physical and chemical stability at 1100 °C over 900 redox cycles

Understanding Chemical Looping Process – Reactor Configuration



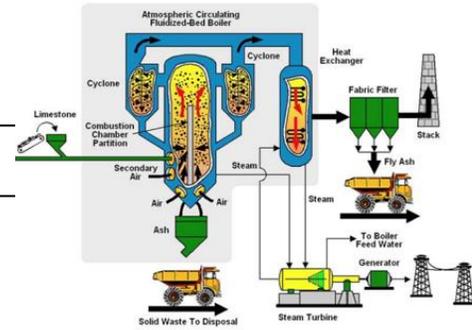
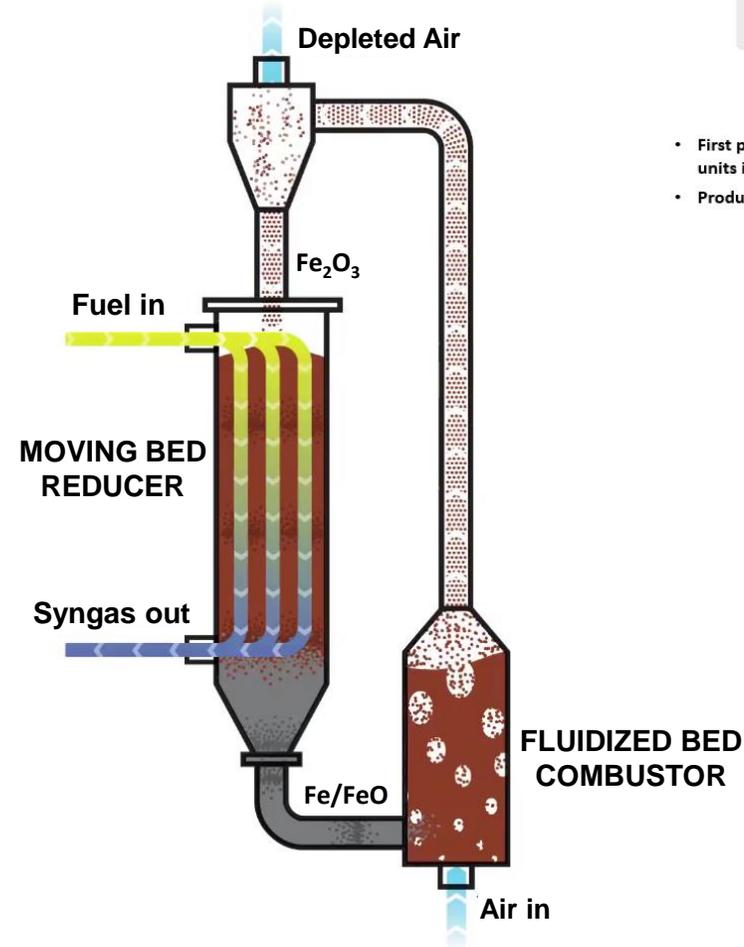
- First patent in 1940 with more than 350 commercial units in operation now
- Accounts for >50% of all transportation fuel and >30% gasoline production

Counter-current: Full Combustion



Simplicity:
One Loop
Unique Reducer
Configuration:
Moving Bed
Unique Flow
Controller:
Non-Mechanical L-Valve

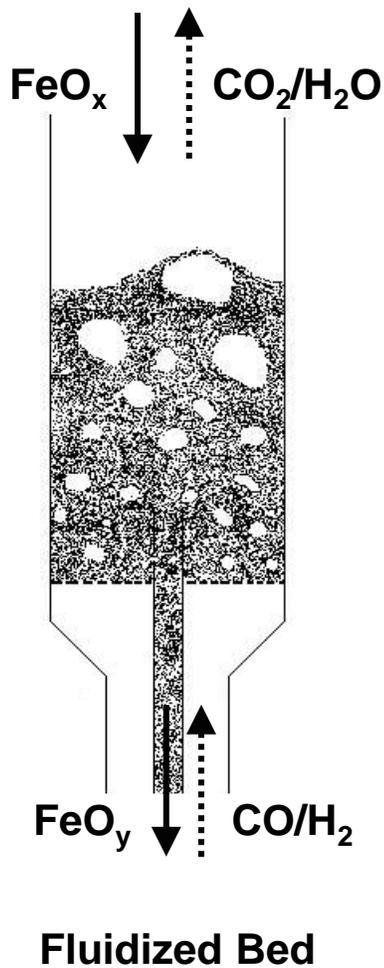
Co-current: Full Gasification



- First patent in 1976 with more than 600 commercial units in operation now
- Producing electricity with low sulfur dioxide emission

Reducer Design Concept: Combustion

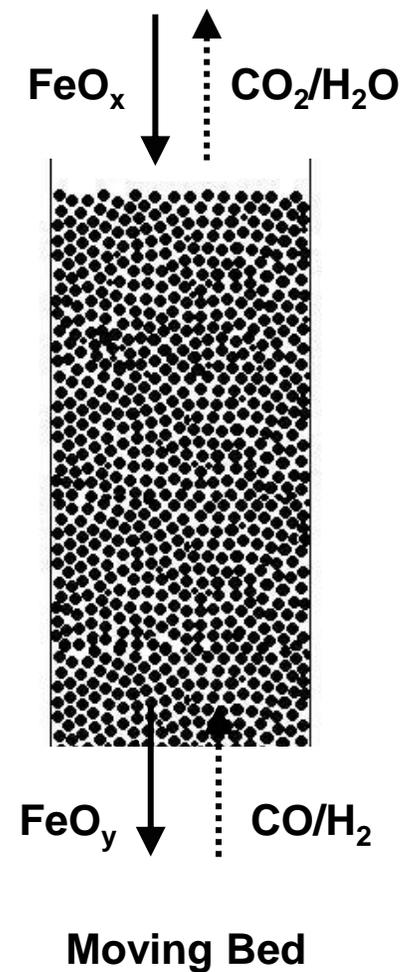
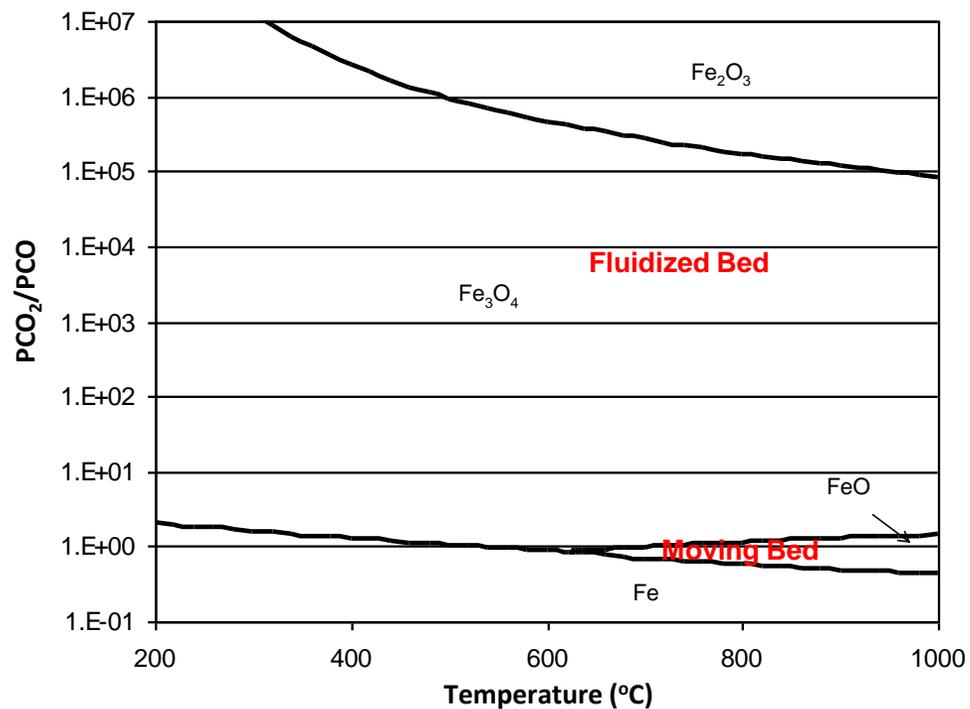
Fluidized Bed v.s. Moving Bed ($x > y$)



11.11% ← Maximum Solid Conversion → 50.00%

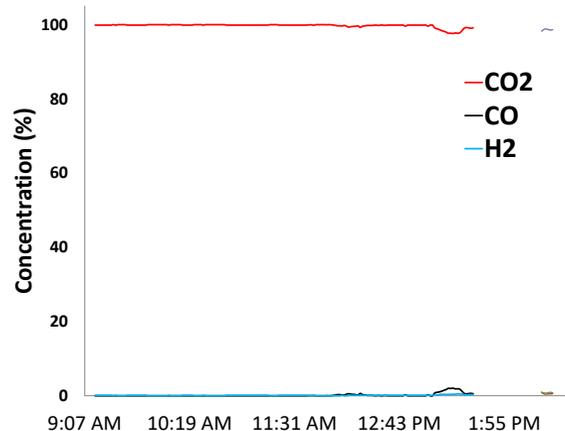
$> U_{mf}$ ← Gas Velocity → $< U_{mf}$

Small ← Particle Size → Large

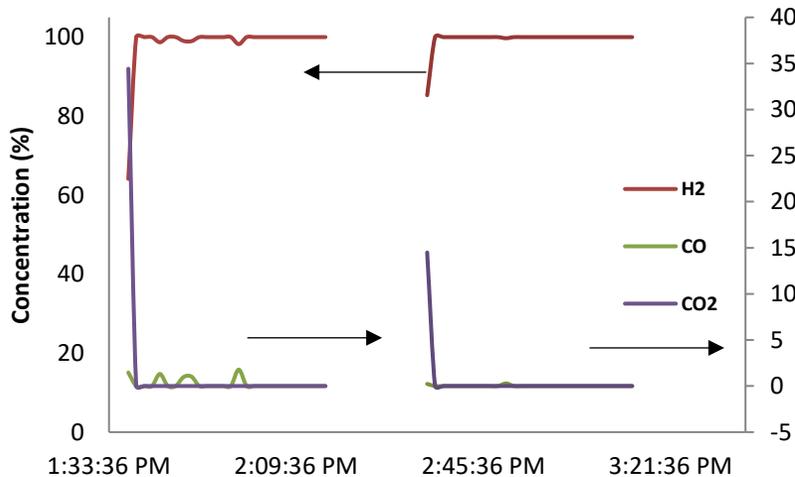


OSU 3-Reactor System for Hydrogen Production

> 99% CO₂ purity in Reducer

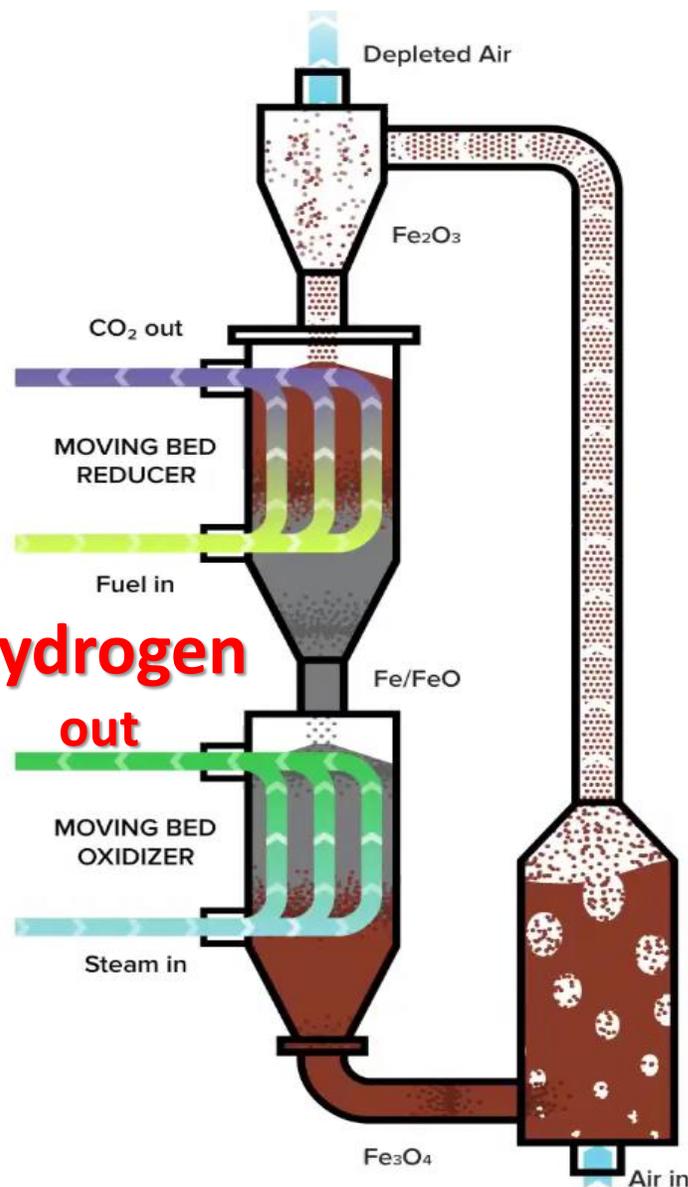


>99.99% H₂ purity in Oxidizer

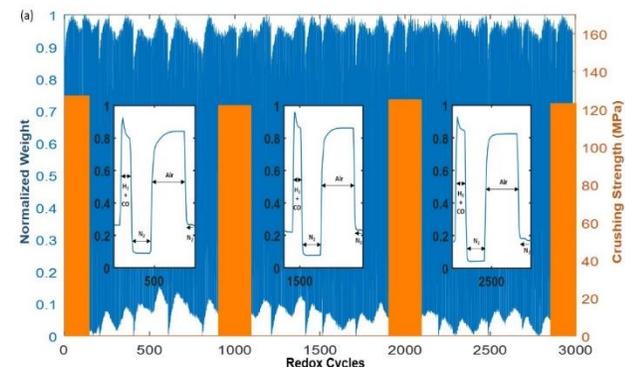


Hydrogen

out



Pilot demonstration at National Carbon Capture Center, AL



Cost of Low Pressure H₂ Production with CO₂ Capture Can be less than \$1.5/Kg H₂

Hsieh and Fan et al., Applied Energy, 2018.

Li and Fan et al., AIChE Journal. 2010

Tong and Fan et al., Fuel. 2013.

Kong and Fan et al., Fuel. 2020

Evolution of Ohio State University Redox Chemical Looping Technology

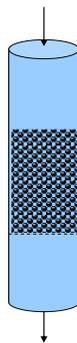


Particle Synthesis

1993



TGA Tests



Fixed Bed Tests

1998



Bench Scale Tests

2001



CCR Process



SCL Process

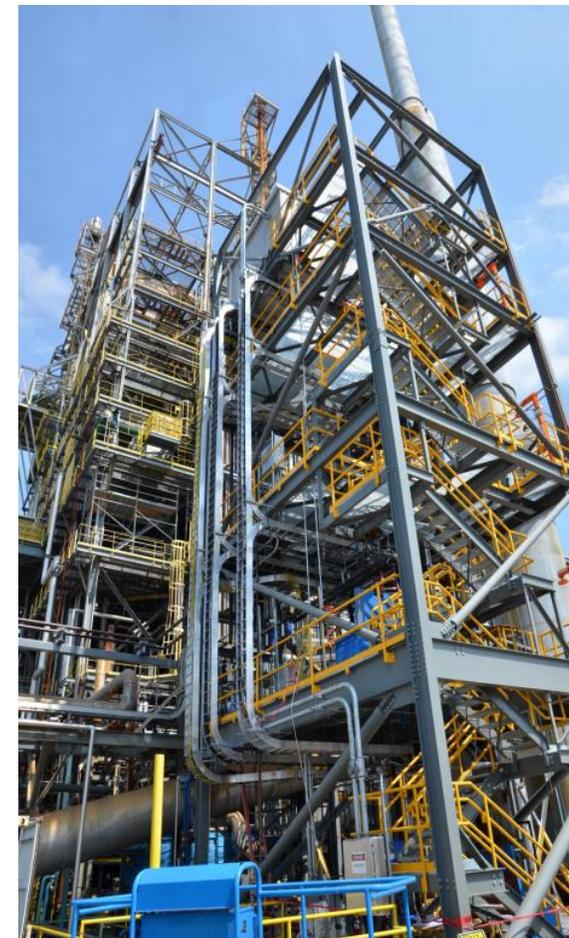


STS Process



Sub-Pilot CDCL Process Tests

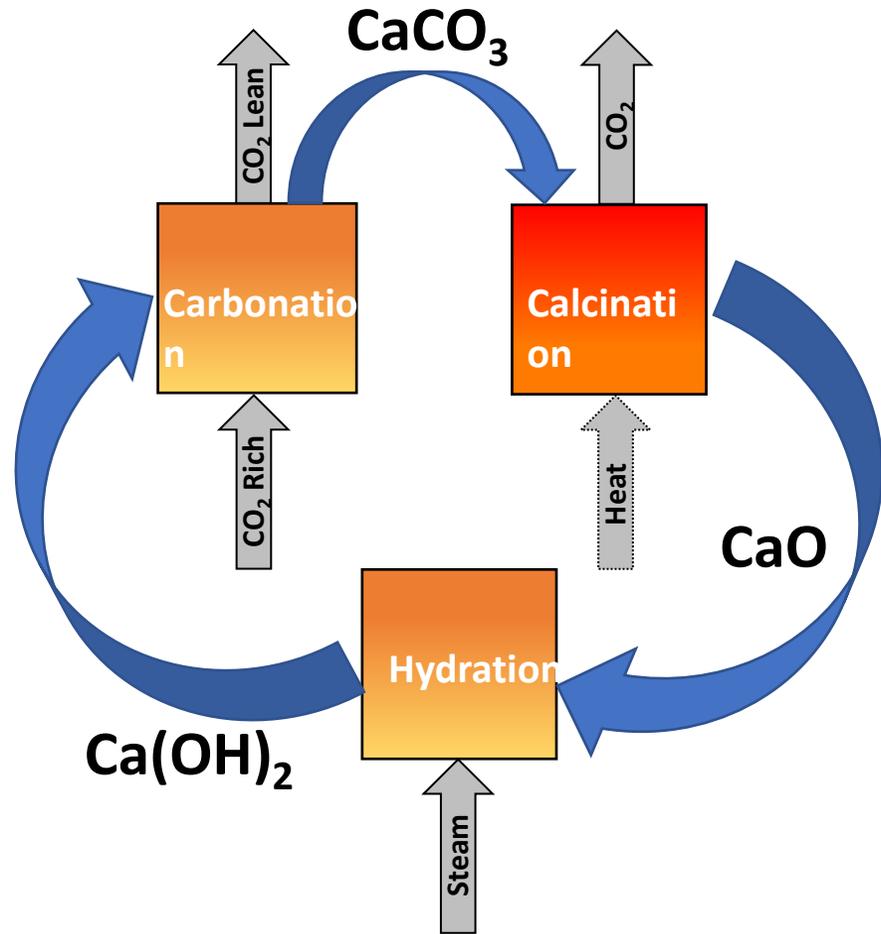
2007



Pilot Scale Demonstration for **Hydrogen production**

2010 to date

Calcium Chemical Looping Technology



OSU Carbonation-Calcination (1990s onwards) and Carbonation-Calcination-Hydration Looping Systems



ITRI Demonstration Plant (2015)

H. Gupta, L. S. Fan, U.S. Patent 7,067,456

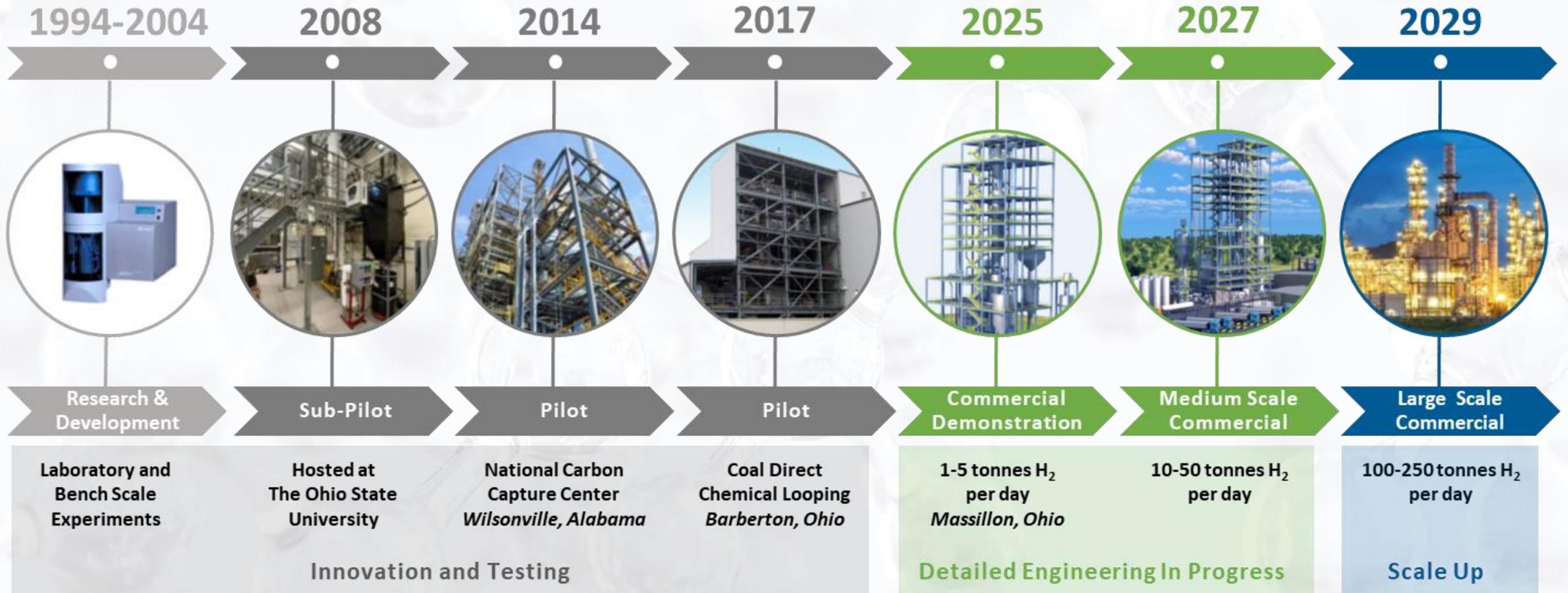
M. V. Iyer, H. Gupta, L. S. Fan: U.S. Patent 7,618,606

S. Ramkumar, L. S. Fan: U.S. Patent 8,496,909

R. Statnick, W. Wang, S. Ramkumar, L. S. Fan: U.S. Patent 8,512,661

N. Deshpande, N. Phalak, L. S. Fan: U.S. Patent us 8,877,150

BrightLoop™ Technology Evolution & Commercialization



BrightLoop™ Project: Massillon, Ohio, USA

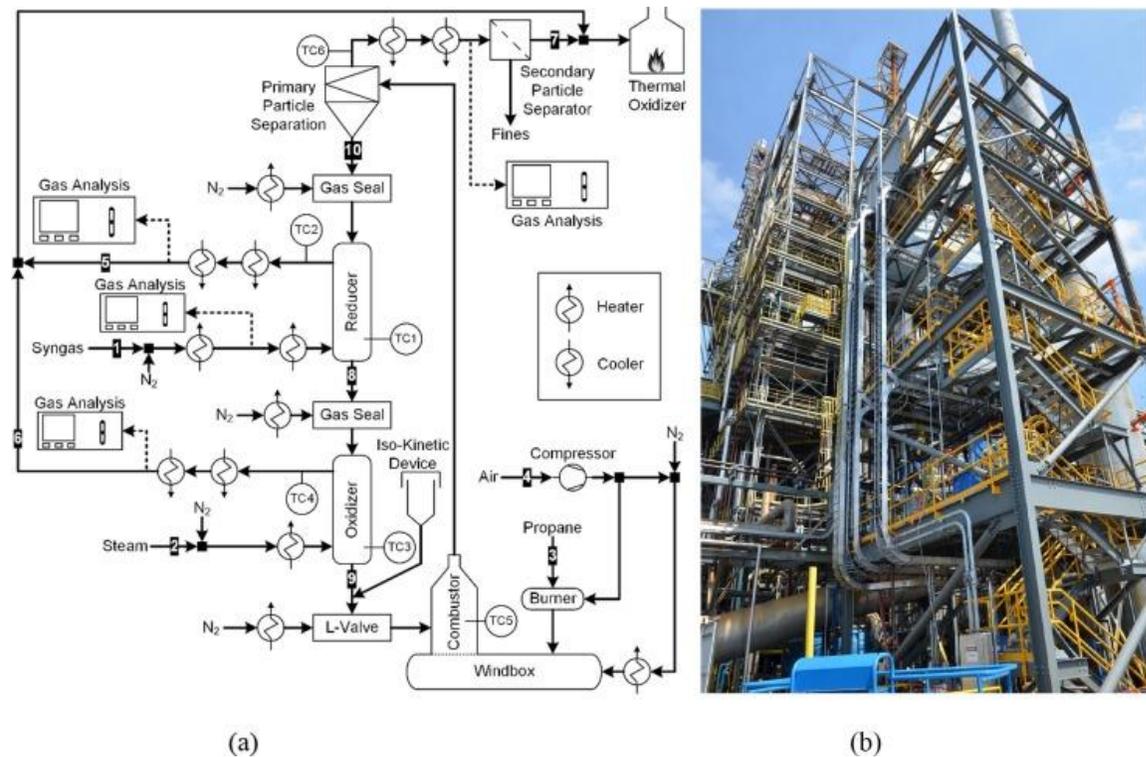
Natural Gas to Hydrogen

OUTPUT	
H ₂ from Natural Gas	1-5 tonnes/day
H ₂ production use	Industrial, Transportation

PROJECT DEVELOPMENT PLAN – Approximate Timeline	
Off-take agreement finalized	2Q 2024
Funding Commitment	3Q 2024
Permits issued	4Q 2024
Target first H ₂ production	1Q 2026



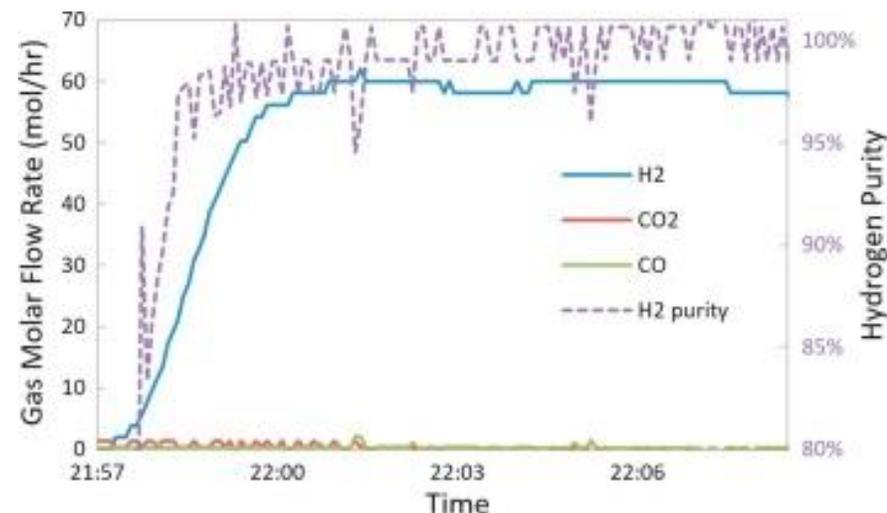
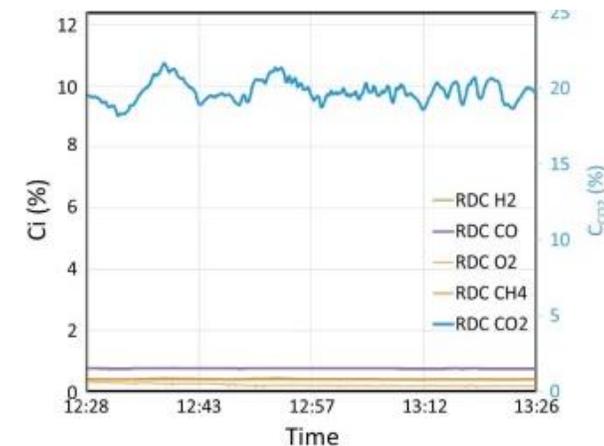
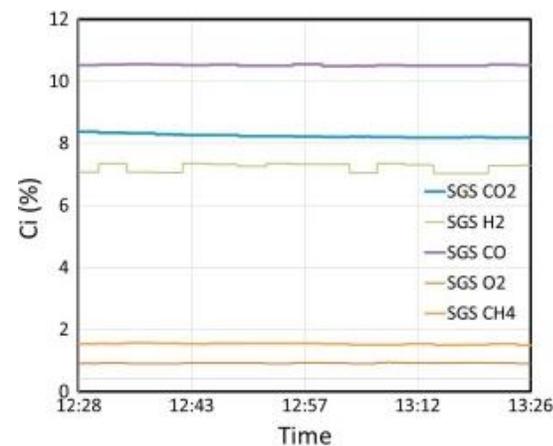
OSU 3-Reactor System for Hydrogen Production



250 kW_{th} Pilot demonstration at National Carbon Capture Center, AL

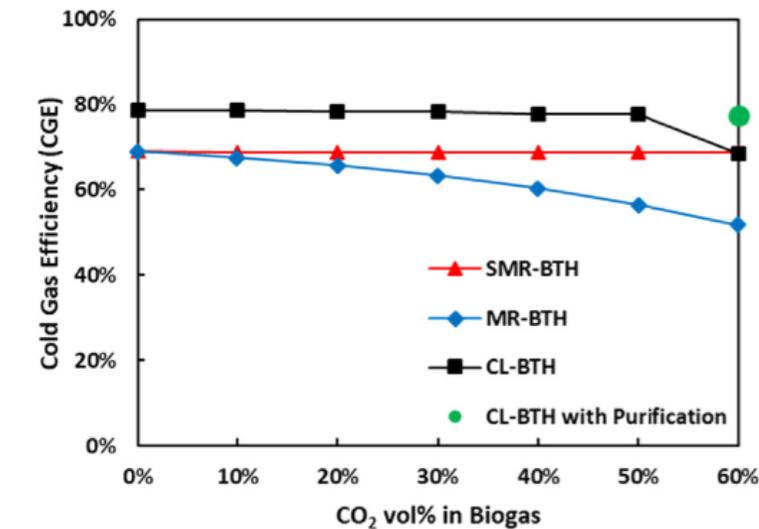
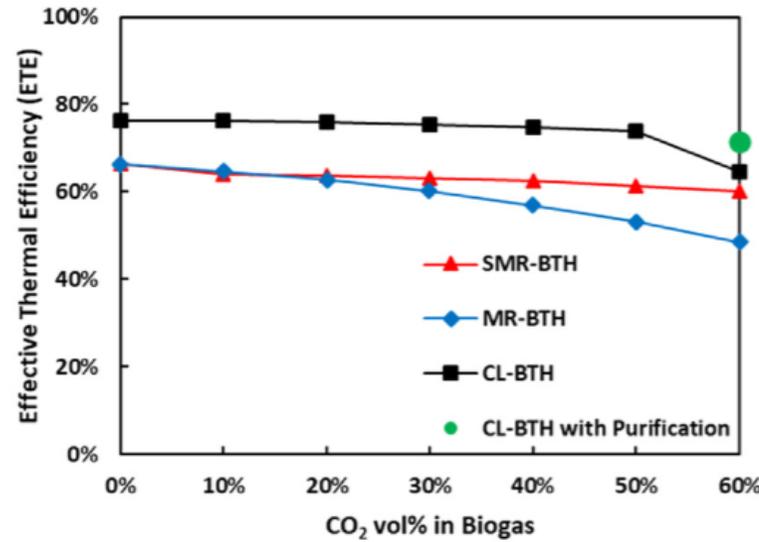
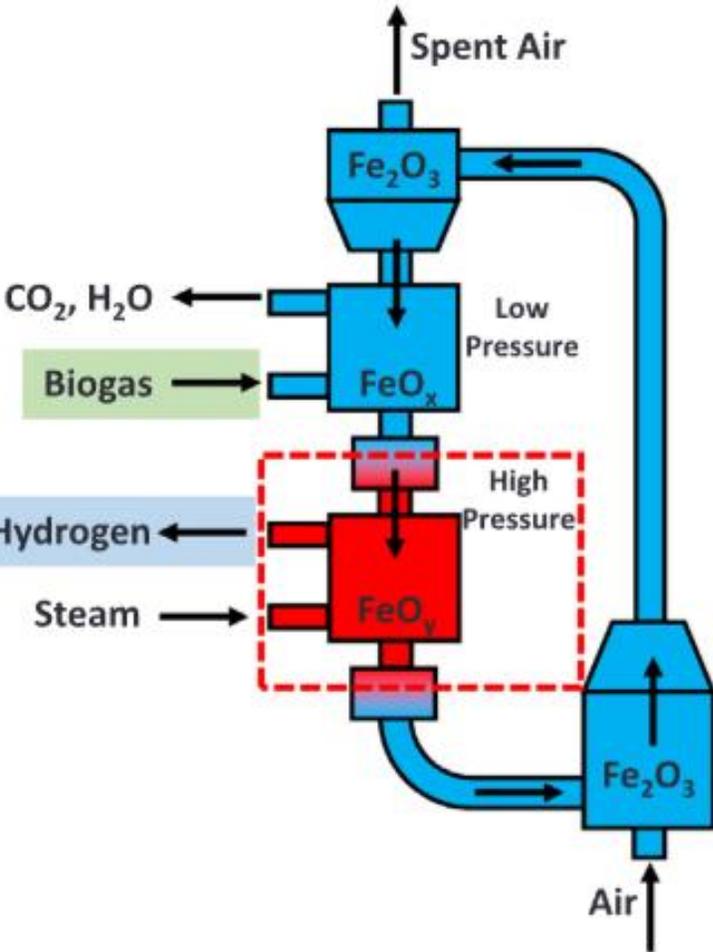
Operating parameters:

1. OC flow rate = 1320 kg/hr
2. Syngas inlet flow rate = 324.8 kg/hr
3. H₂ outlet flow rate = 3.61 kg/hr

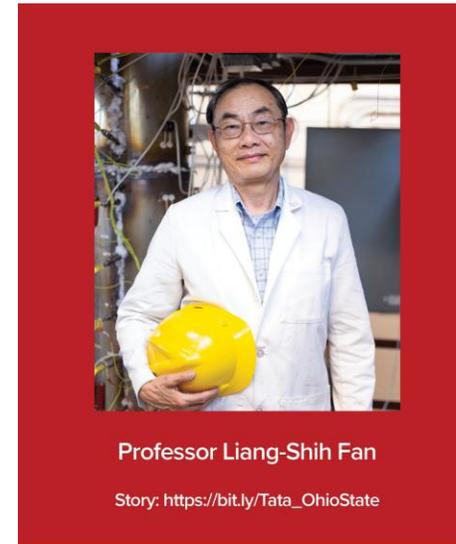
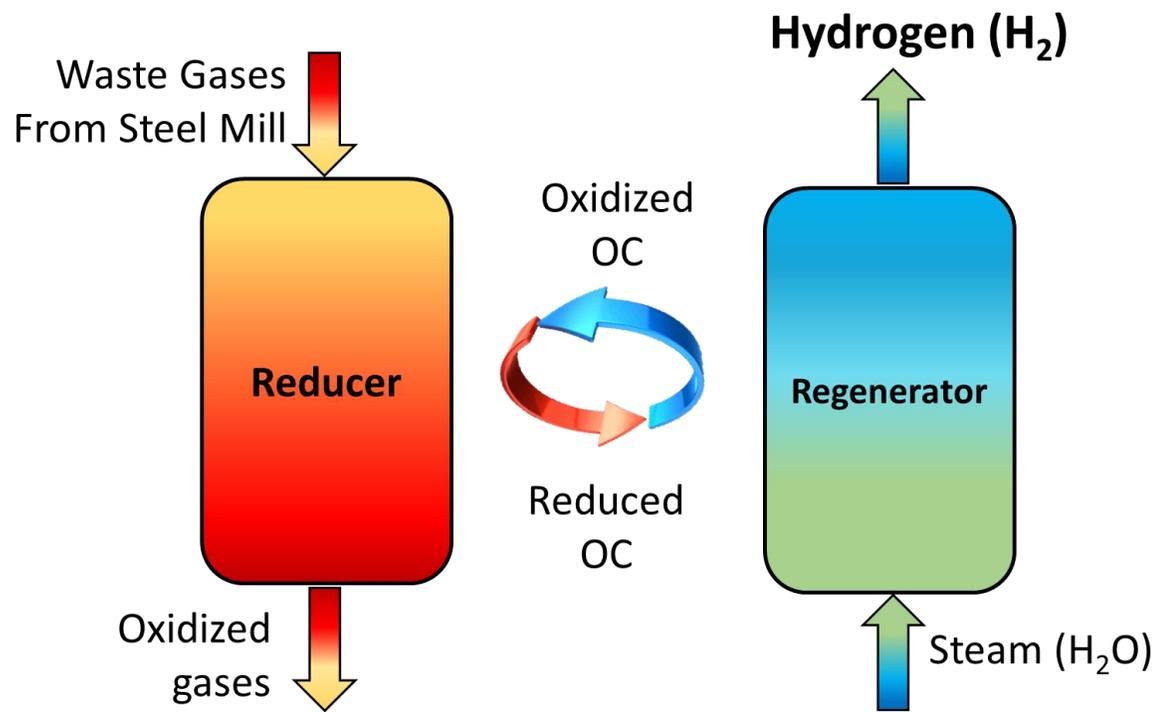


Gas profiles for pilot demonstration: Reducer (top) and Oxidizer (bottom)

Biogas to H₂ conversion with CO₂ capture using chemical looping technology



- The chemical looping process can directly handle biogas with 0-50% CO₂ volume ratio.
- Achieves 100% CO₂ capture, making it a carbon-negative hydrogen production method.
- Oxidizer pressure set at 3 MPa eliminates the need for hydrogen compression, reducing auxiliary power consumption.
- H₂ yield reaches 2.42 moles of H₂ per mole of CH₄, outperforming conventional methods.
- Cold Gas Efficiency (CGE) increases by 13-14% compared to conventional reforming processes.
- Effective Thermal Efficiency (ETE) improves by 15-20% demonstrating better energy utilization.



 THE OHIO STATE UNIVERSITY
COLLEGE OF ENGINEERING



Tata Steel licenses Ohio State Tech

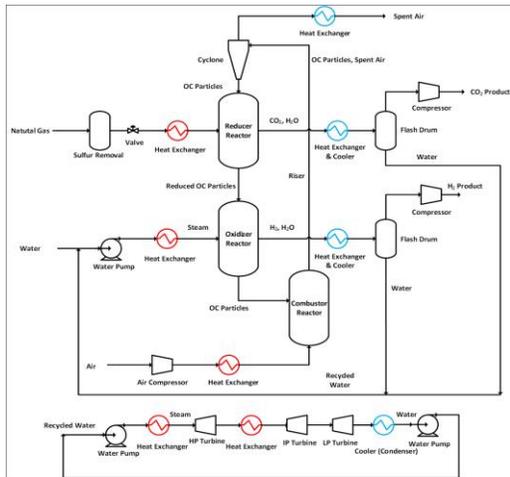
Applying Professor L.-S. Fan's revolutionary chemical looping Redox Energy Recovery (RER) system to the steel industry will produce sizable economic and environmental benefits by producing sustainable hydrogen while reducing carbon emissions.

William G. Lowrie Department of Chemical and Biomolecular Engineering

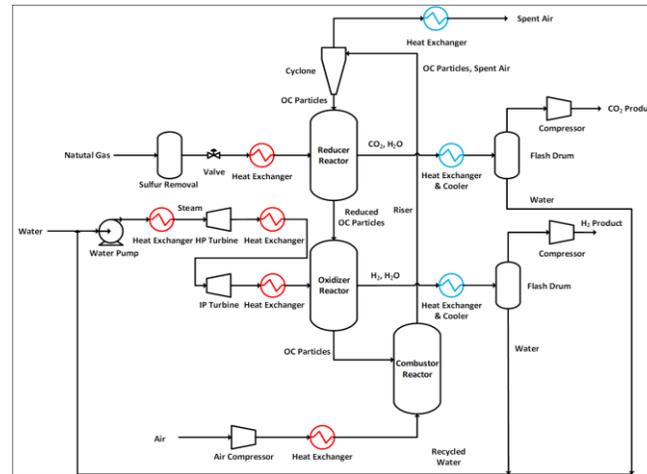
- Steelmaking: high contributions to GHG emissions
- Outlet gases contain H_2O , CO_2 , along with some amount of H_2 and CO , typically flared
- Leveraging the reducing potential to generate H_2 .
- Seamless integration into existing steel production processes with minimal modifications to plant infrastructure
- Lower carbon intensity steel production
- An effort towards a decarbonized steel production with Tata Steel, India.
- Current ongoing efforts for commercialization.

Co-generation of Electricity and Hydrogen Using OSU Chemical Looping Platform

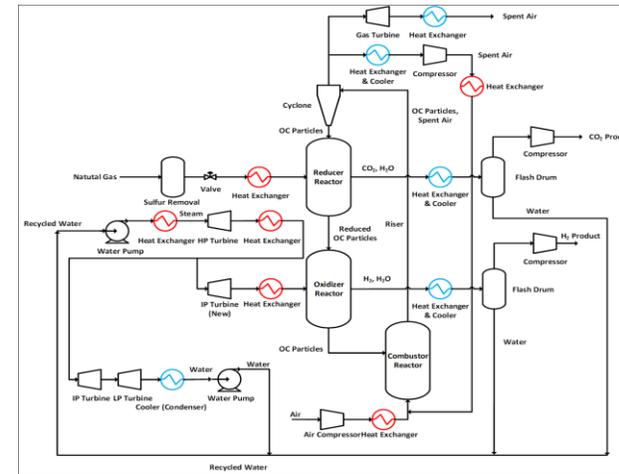
ITC-CLWS: integrated turbine combined chemical looping water splitting



Standard CLWS process & Rankine cycle (case 1)



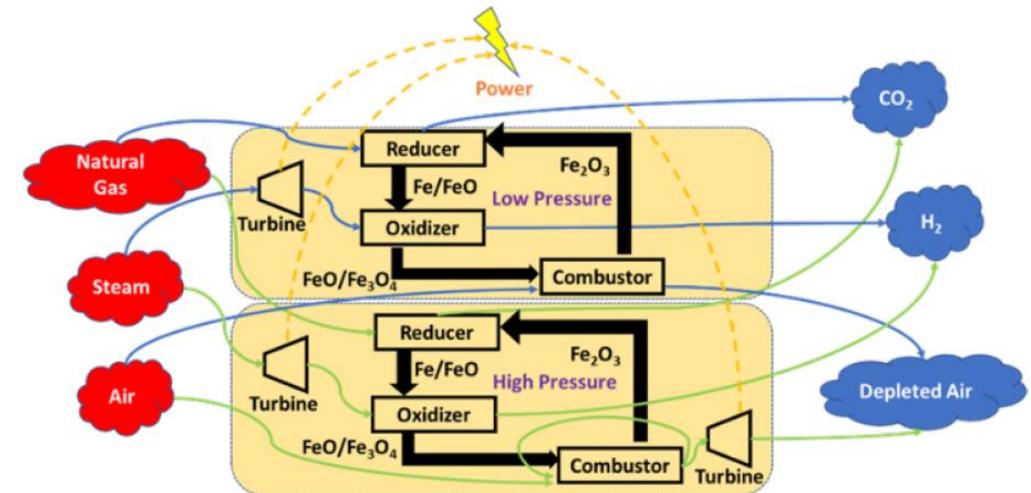
Basic ITC-CLWS process (case 2&3)

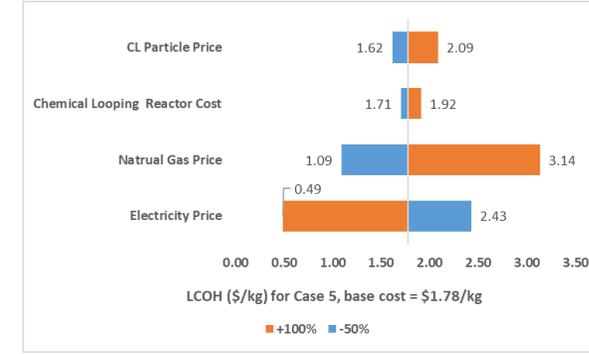
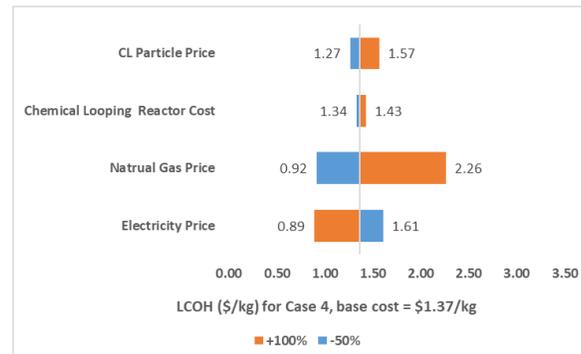
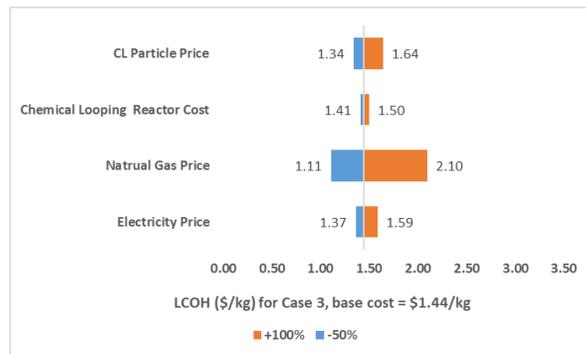
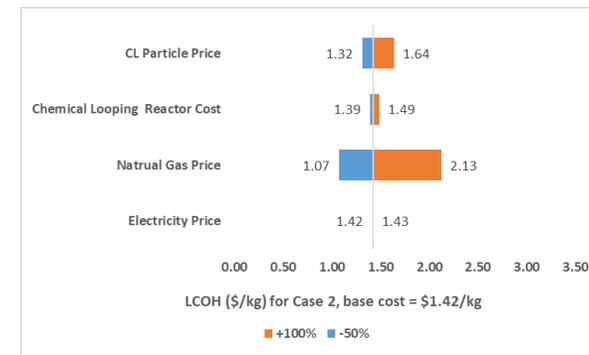
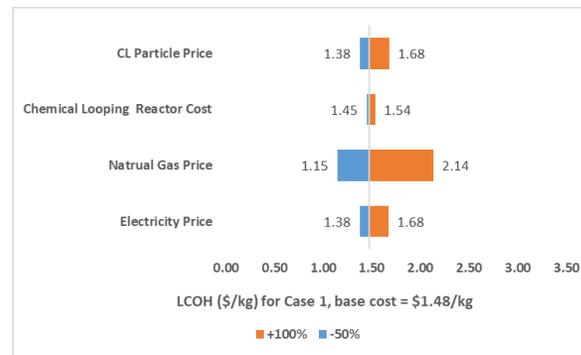
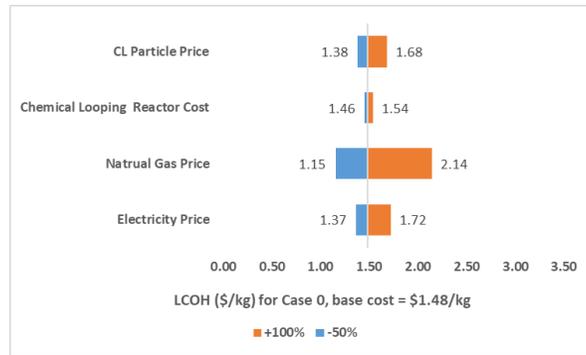


High-pressure ITC-CLWS processes (case 4&5)

Tax policies:

1. 45Q - The amount that a taxpayer may claim as a Section 45Q tax credit is computed per metric ton of qualified carbon dioxide captured and sequestered (\$27.61/MT CO₂ in 2023).
2. 45V - Credit is calculated based on the amount of CO₂ equivalent per kilogram of hydrogen. Used to incentivize clean H₂ production. Maximum tax credit can be \$3/kg H₂ if the process emits less than 0.45 kg CO₂ eq/kg H₂.
3. CO₂ emission tax - Penalty for emitting CO₂ into the atmosphere. It is considered to be \$37.7/MT CO₂ in 2018.
4. For this study emission tax is considered instead of 45Q and 45V.





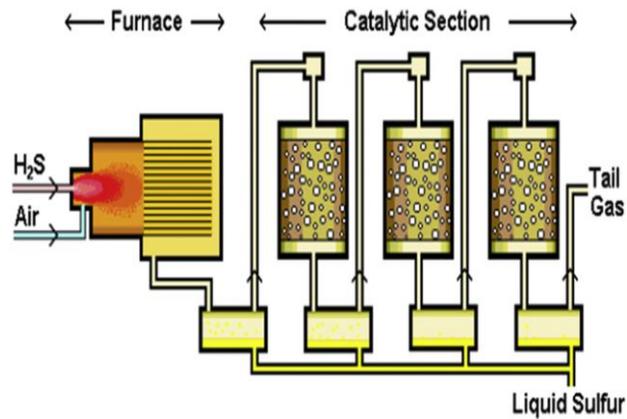
LCOH sensitivity diagrams for CLWS/ITC-CLWS Cases 0-5 (a-f)

Case index	0	1	2	3	4	5	ATR	SMR
Hydrogen Price, 2018 \$/kg	1.48	1.48	1.42	1.44	1.37	1.78	2.23	1.61

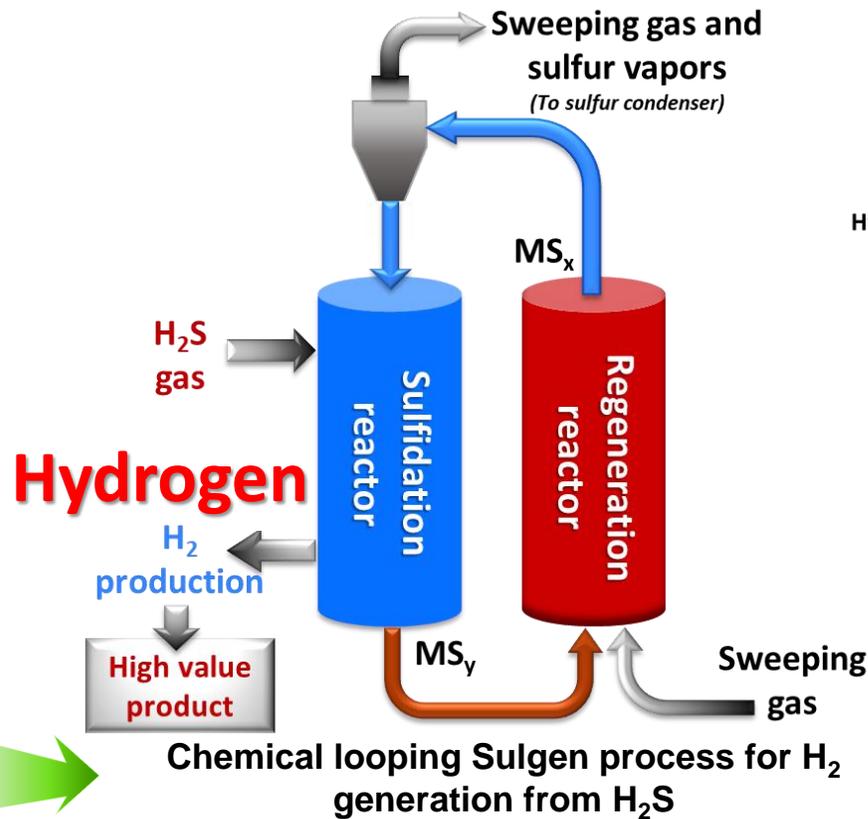
LCOH comparison of Cases 0-5 and ATR, SMR with 103.2 ton/hr natural gas as feedstock

LCOH: Levelized Cost of Hydrogen

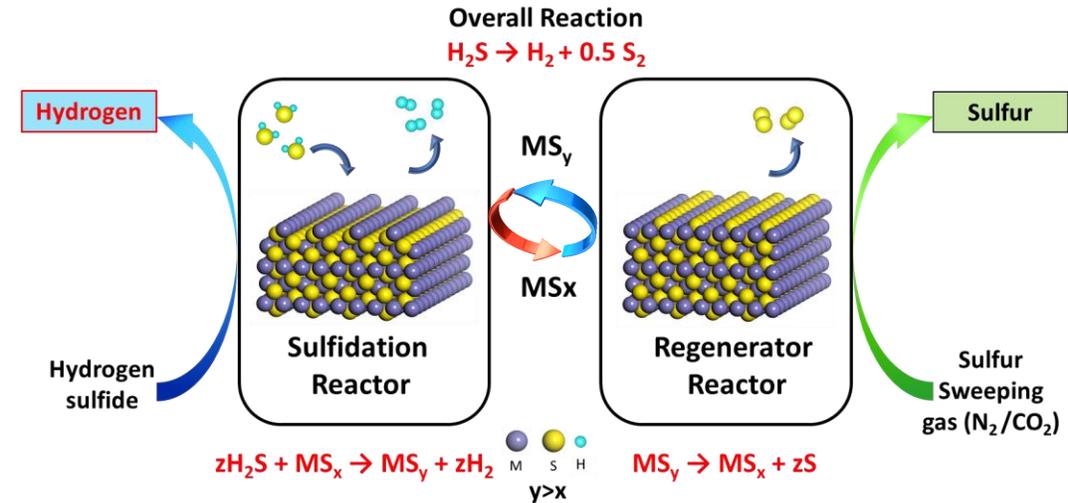
OSU Chemical Looping Sulgen Process for Hydrogen Generation from Hydrogen Sulfide



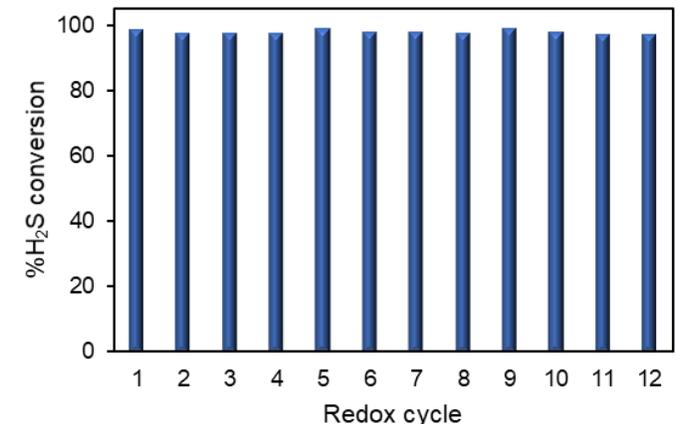
Conventional H₂S treatment using the Claus process



Chemical looping Sulgen process for H₂ generation from H₂S



>98% H₂S conversion in Sulfidation step



H₂S conversion into H₂ over 12 sulfidation (T: 400°C) and regeneration (T: 950°C) cycles using iron-based sulfur carrier

Nadgouda SG, Jangam KV, Fan L.-S. Systems, methods and materials for hydrogen sulfide conversion. 2018 (62/716,705 (US), patent pending).

Jangam and Fan et al., ACS Sustainable Chem. Eng. 2021

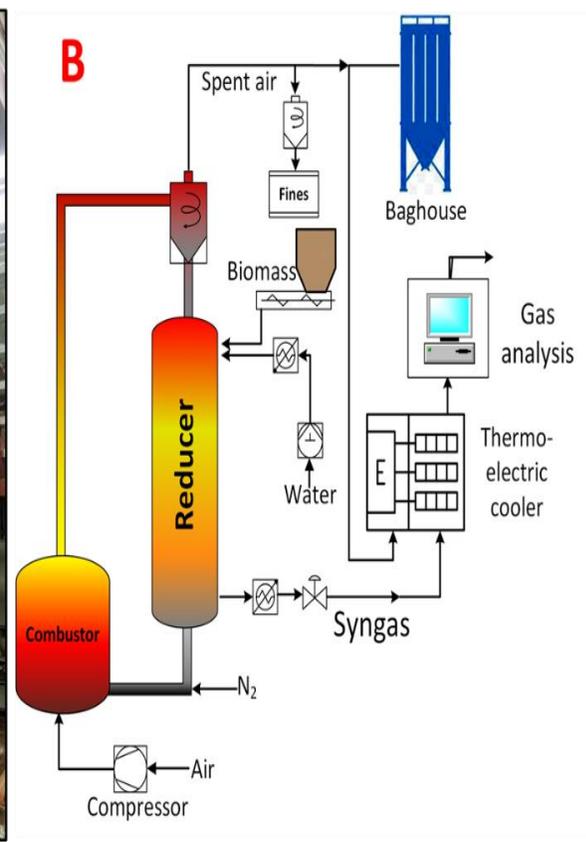
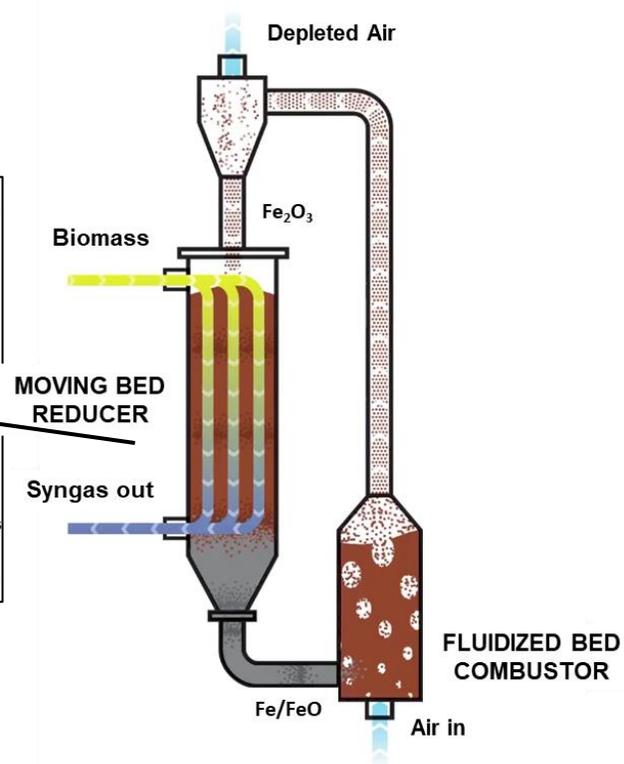
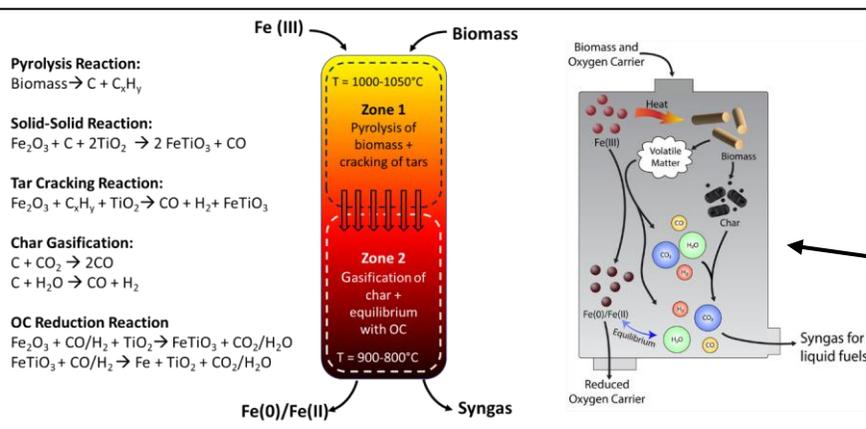
Jangam and Fan et al., Chem. Eng. J., 2021

Sassi and Gupta, Am. J. Environ. Sci., 2008

Key advantages over the Claus process:

- Production of H₂ instead of steam
- ~99% reactive separation of H₂S into H₂ from syngas, natural gas, acid gas and hydrocarbon (C₂-C₄) stream
- Significant reduction in processing units, cost and energy requirement

Biomass to Syngas (BTS) Technology



Picture of the actual sub-pilot unit. B) Schematic of the experimental setup with accessories

- 15 kW_{th} unit has been built at OSU with the capacity to process 3.6 kg/hr of biomass
- The unit has been successfully run for over 600 hours with 150 hours of continuous operation
- The system exhibits the ability to process various types of biomass feedstocks

Comparison with Conventional Process

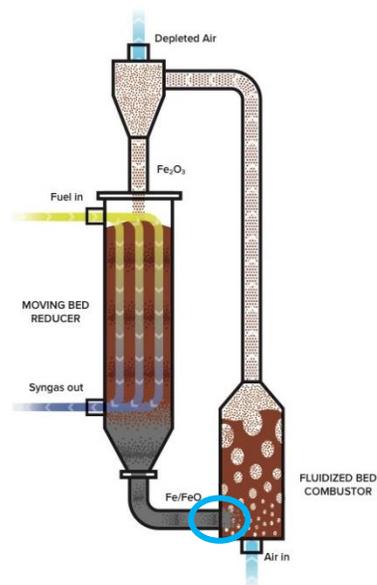
SYSTEM	INDIRECT GASIFIER	BTS
BIOMASS IN, AS RECEIVED (SHORT TPD)	81464	70152
GASIFICATION CARBON EFFICIENCY (%)	69.7	89.4
SYNGAS PURITY	87.9	82.9
PRODUCT (BBL/DAY)	49958	49957
JET FUEL	36944	36943
DIESEL	8614	8613
NAPHTHA	1178	1178
LPG	3222	3222
NET POWER EXPORT (MW)	545.6	410.2
THERMAL EFFICIENCY (%)	48.8	54.5

SYSTEM	INDIRECT GASIFIER	BTS
SYNGAS COMPRESSION DUTY	324	387.7
COMBUSTOR COMPRESSION DUTY	96.5	87.4
ASU ELECTRICITY DEMAND	114.6	-
DRYER AIR BLOWER	-	2.7
F-T SYNTHESIS NET EXPORT POWER	966	887
NET PLANT EXPORT POWER	545.6	410.2

- To validate the superior performance of BTS, it was compared with Indirect gasification for liquid fuel production through FT synthesis
- The results indicate that BTS has higher thermal and carbon efficiency than indirect gasification
- Furthermore, BTS achieves process intensification by eliminating the energy-intensive Air Separation Unit

ECVT Commercial Applications

60" Sensor



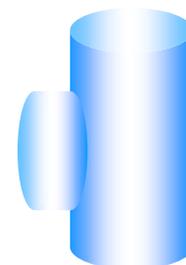
(a)



(b)



(c)



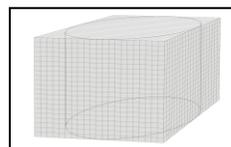
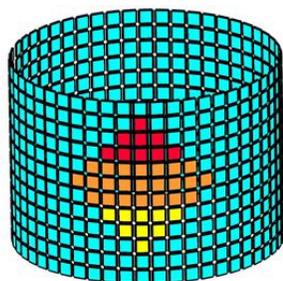
(d)



(e)



(f)

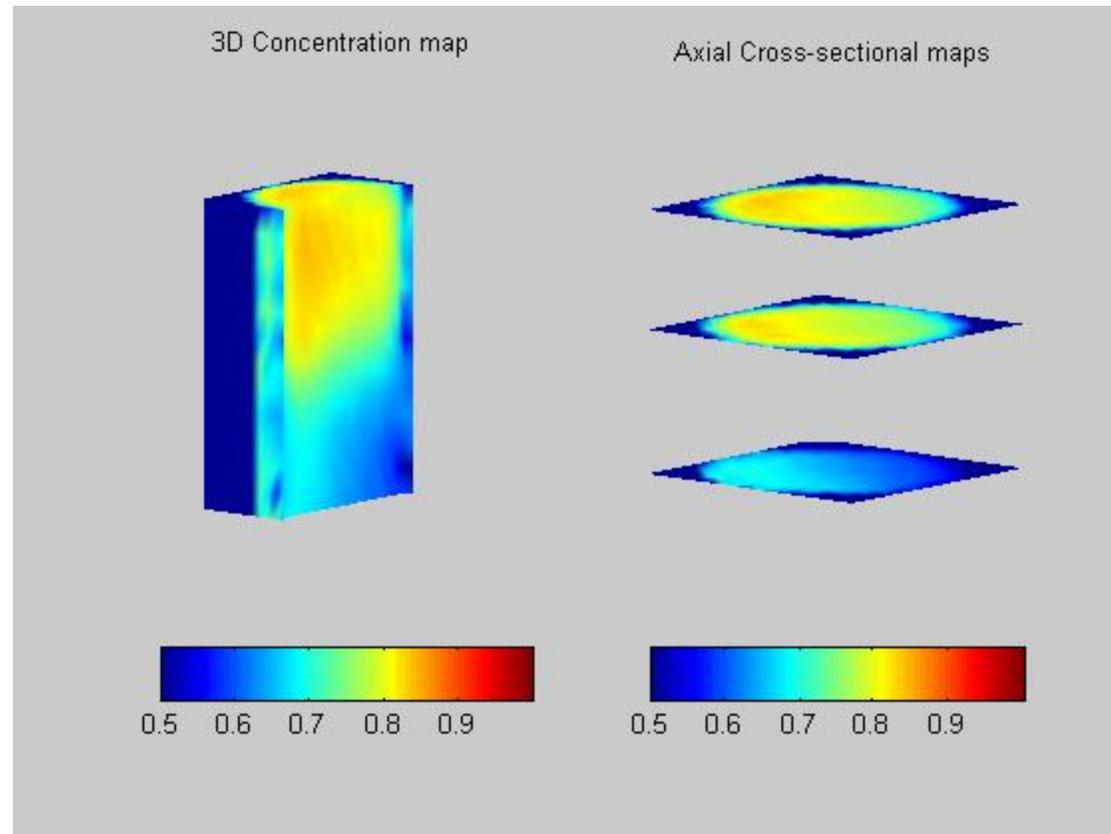
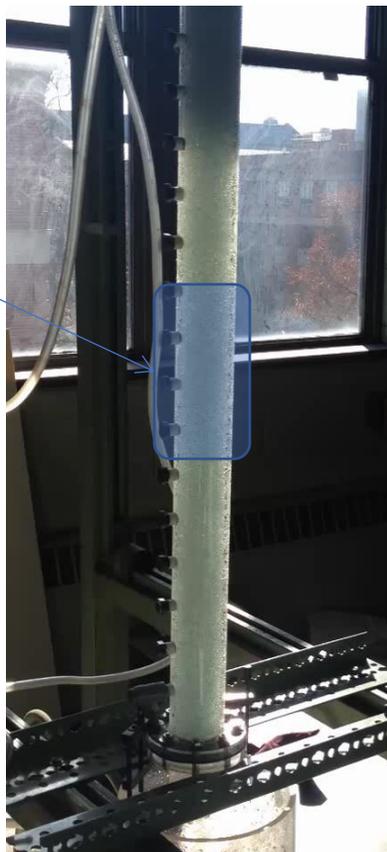
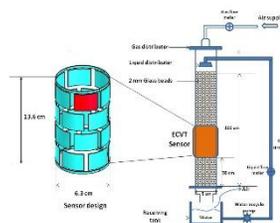


20 × 20 × 20



Electrical Capacitance Volume Tomography Videos for pulsation flow (G: 0.454 kg/m²s, L:21.7 kg/m²s)

ECVT Sensor location



Original video in normal speed

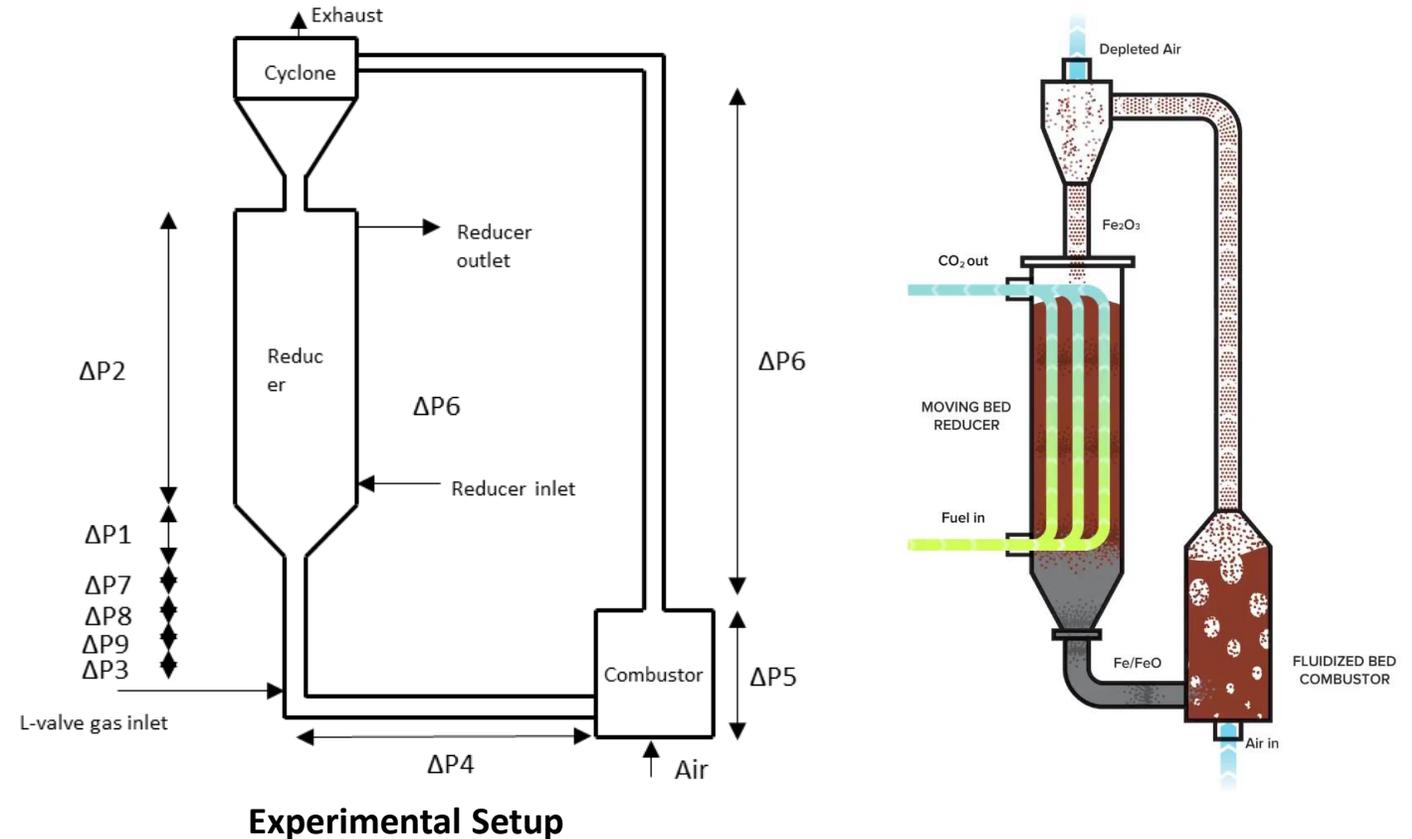
ECVT reconstructed video in normal speed (50fps)

Recurrent Neural Network (AI) Based Metal Oxide *Attrition* Risk Assessment on Arching in Chemical Looping Systems

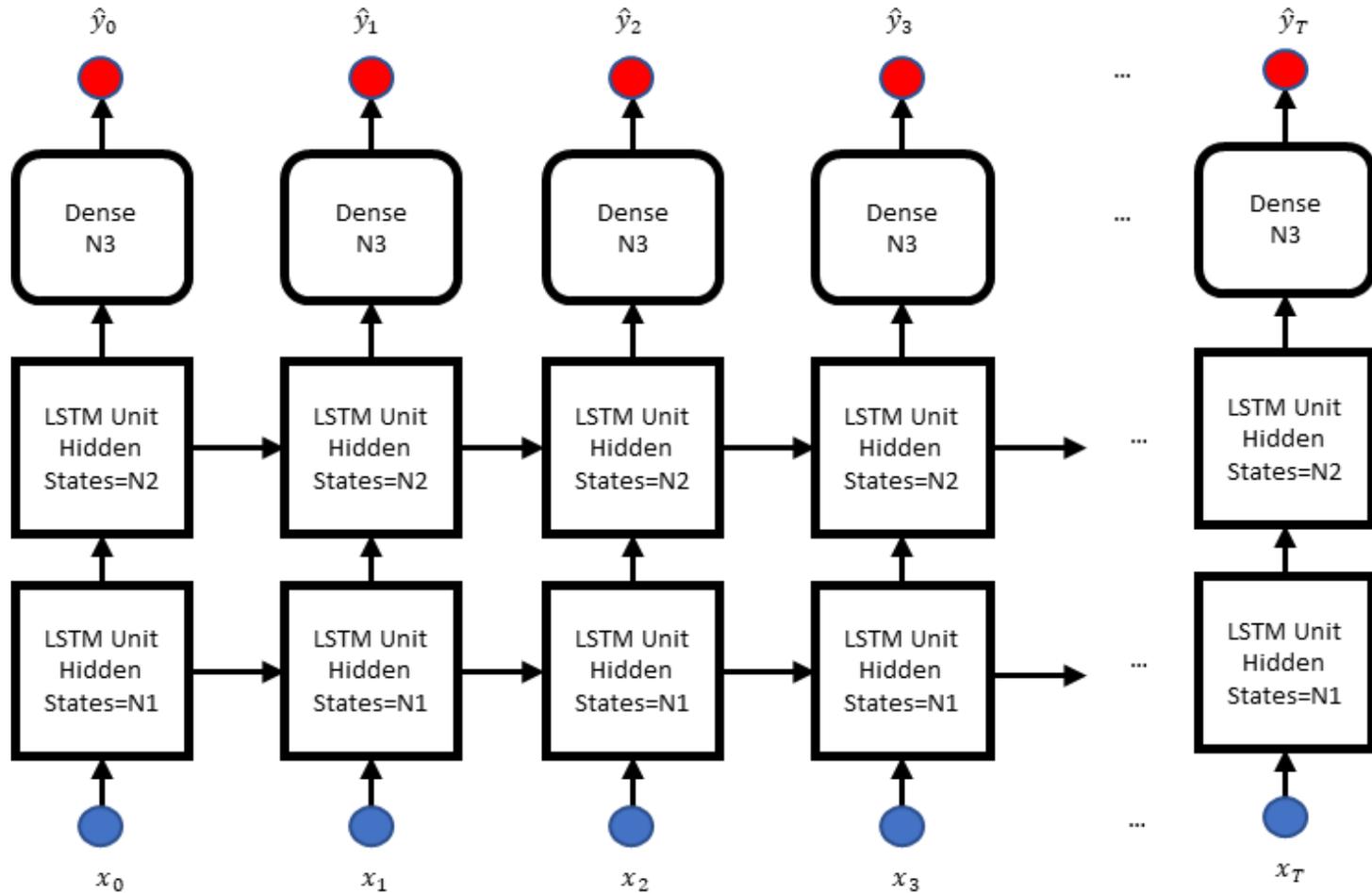
- 9 different pressure transducers ($\Delta P1 - 9$) were installed across the cold flow model
- Three different gas inputs: L-valve, Combustor and Reducer
- ITCMO particles with particle density of 2500 kg/m³ and particle diameter of 1.5 mm

Experimental conditions:

Reducer	2 SCFM
L-valve	2 SCFM
Combustor	1000 SLPM



Recurrent NN model Setup



- Layers in different time steps share the same weights
- Activation function in the dense layer is the rectifier function:

$$f(x) = \max(0, x)$$

- Activation function in the output layer is the sigmoid function:

$$f(x) = \frac{1}{1+e^{-x}}$$

- **Loss function is binary cross entropy**

$$\frac{-1}{T+1} \sum_{t=0}^T (1 - y_t) \log(1 - \hat{y}_t) + y_t \log(\hat{y}_t)$$

LSTM Scheme

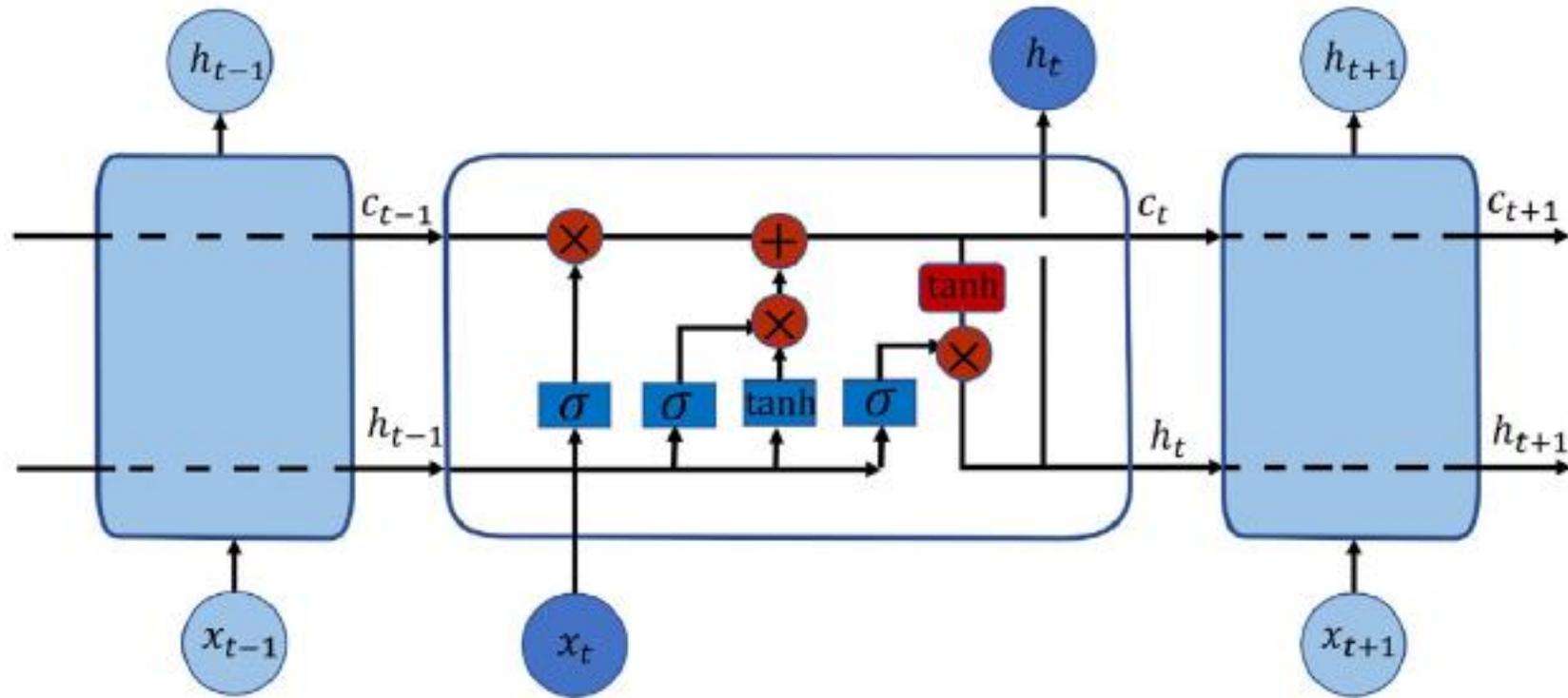


Fig. 5. Flow chart of the LSTM unit, referenced from Olah [33].

Training, Validation and Testing Results

[N1,N2; N3]	Training	Validation	Testing Dataset				
	Loss	Loss	Loss	Accuracy	Precision	Recall	F1 Score
[128,64;32]	0.0361	0.0932	0.108	0.960	0.689	0.838	0.757
[128,32;32]	0.0380	0.0961	0.152	0.9399	0.559	0.896	0.688
[64,32;32]	0.0374	0.0962	0.122	0.959	0.680	0.841	0.752
[64,32;16]	0.0370	0.0931	0.141	0.948	0.607	0.861	0.712
[64,16;16]	0.0364	0.0947	0.125	0.952	0.624	0.876	0.729
[128;32]	0.0358	0.0907	0.119	0.951	0.621	0.882	0.729
[128;16]	0.0342	0.0905	0.154	0.942	0.569	0.873	0.688

$$Accuracy = \frac{TP + TN}{TP + TN + FP + FN}$$

$$Precision = \frac{TP}{TP + FP}$$

$$Recall = \frac{TP}{TP + FN}$$

$$F1 = \frac{2}{1/Precision + 1/Recall}$$

TP: True Positive (correctly predict it happens); TN: True Negative; FP: False Positive (incorrectly predict it happens); FN: False Negative (incorrectly predict it did not happen); True Positive – correct prediction of bubbles (1); true negative – correct prediction of no bubbles (0)

Early stopping was used to choose weights corresponding to the lowest validation loss

Concluding Remarks

- **Chemical Looping** concept and practice **started 125 years** with the Bergmann Process, followed by some pilot plant demonstrations along with hundreds of labs worldwide working on metal oxide redox materials and bench-scale processes for generation of various products. Despite its long history of development efforts, **no commercial plant is yet in operation.**
- Chemical Looping is an **enabling platform** technology with **high Exergy efficiency** for generation of a variety of products such as hydrogen, syngas, fuels and chemicals. It is particularly attractive for a process operation **with decarbonization requirement.**
- Chemical Looping **multi-scale** approaches encompassing four essential scale levels - **molecular, particle, reactor, and system - in synergism interaction** propel successful chemical looping and other new energy and fuel technology development. **Interfacing** between the levels is of the area where exciting research continues to evolve.
- The **first Chemical Looping commercial plant** is expected to be in operation **within one years by Babcock and Wilcox Company**, a licensee of the technology from the Ohio State University **based on unique moving-bed reducer configuration and iron-based metal oxide oxygen carrier for hydrogen production.**