STEVENS
Institute of Technology

A Look At Microchemical Systems

What are MCS? What are some key limitations?

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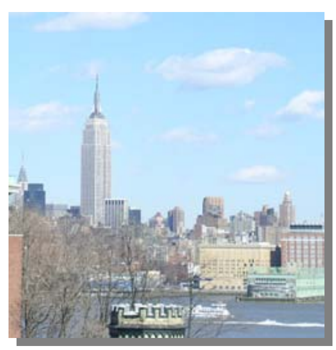
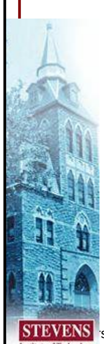
Stevens Institute of Technology



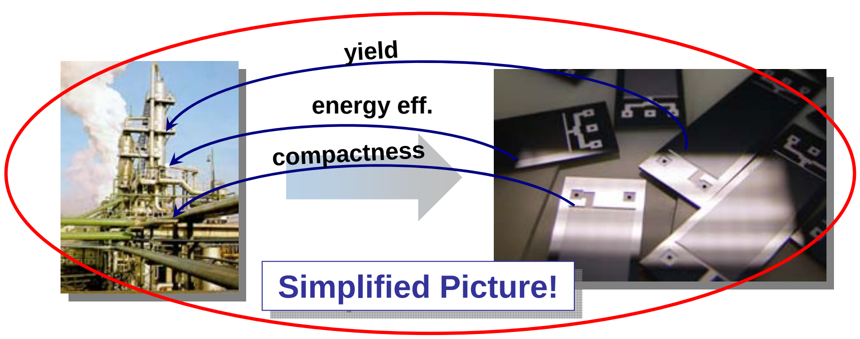


- Founded 1871
- Stevens family: Commercialized urban ferry transport in NYC
- 4500 students: 1700 trad. undergrad; 2800 grad (most p-t)
- Freshmen: 3.8 GPA; SAT: 1200-1400 (25%-75%)
- Chem/Biomed/Matls Engineering → *NJ Center for Microchemical Systems*

New Jersey Center for MicroChemical Systems (NJCMCS)

- Official start in September 2002
  - $10.0M commitments to date
- Vision
  - Leadership for microchemical device/fundamental understanding, design methodology and tools development
- Mission
  - Original research, education of new PhDs
- Systems-level concept demo with partners
  - Army-Picatinny, Bristol-Myers Squibb, FMC, Lucent-Bell Labs/NJNC
  - Applications: portable power, pharmaceutical processing, critical industrial chemicals, biomedicine

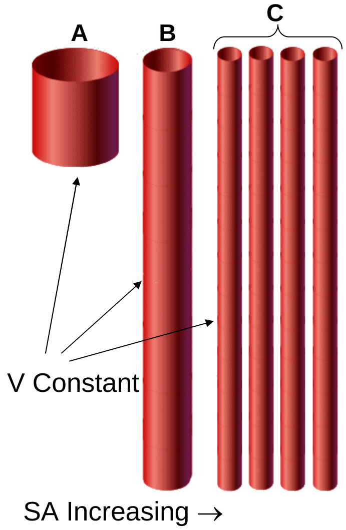
Microchemical Systems

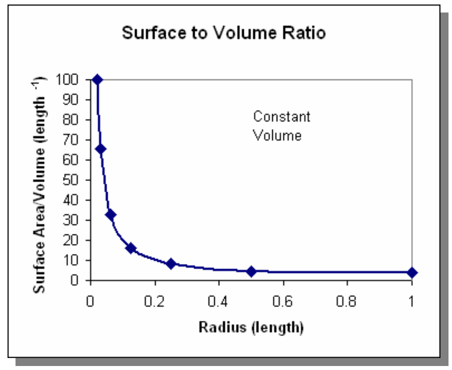


Miniature reaction and other unit operations, possessing ***specific advantages*** over conventional chemical systems

Not Lab-on-Chip: chemical <u>production</u> vs. analysis

Key Benefit: Surface to Volume





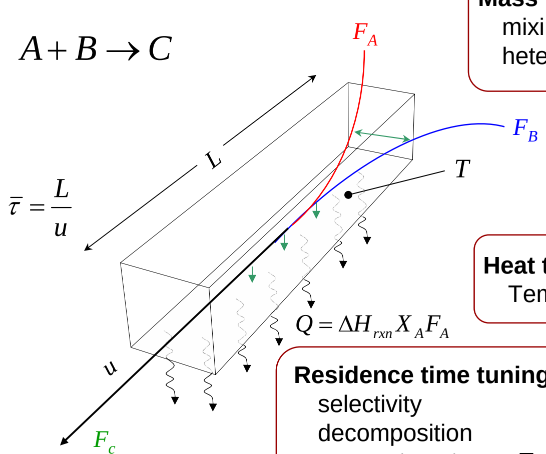
Heat Management

Mixing

Surface Reaction

Explosion-Safety

Transport and Residence Time Advantages



Mass transport limited
- mixing
- heterogeneous rxn

Heat transport limited
- Temp uniformity, control

Residence time tuning to limit exposure
- selectivity
- decomposition
- access to extreme T and P

Surface to Volume: Superb Transport

Example: Overall Heat Transfer Coefficient

Hx Type	*U (W/m²K)*
Tubular	150-1200
Spiral	700-2500
Plate	1000-4000

Microchannel: 3800-6800 W/(m^2K)

(Stevens undergrad design project)

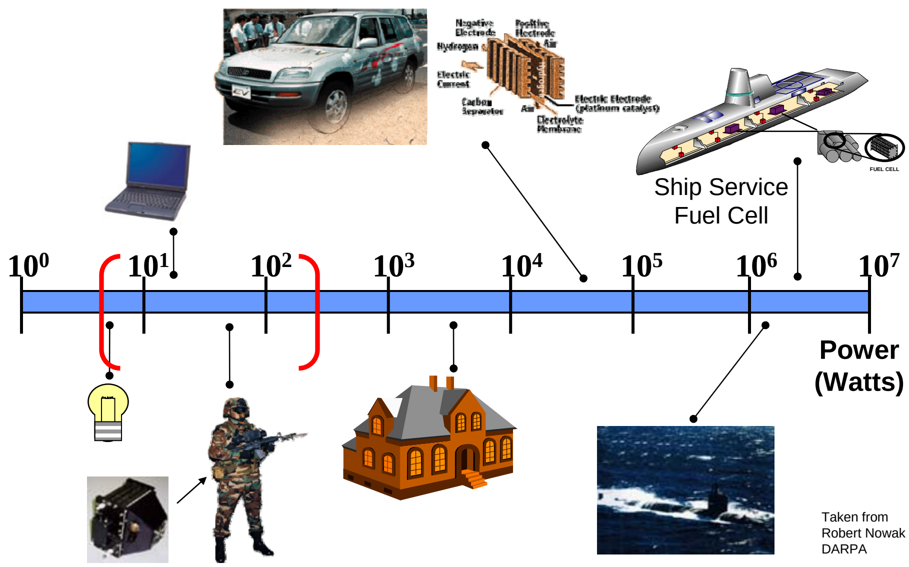
(500x500 μm^2 x 1.5 cm channels)

- SIT and NJCMCS
- MCS: a Definition

Key Limitations in MCS and Examples of How We Are Exploring Them

- Geometry Limitation in Heat Removal
- Thermal Integration Limitation in a Multiunit System
- Multiphase Mixing Limitation
- Integrated Process Control Limitation

Fuel Cells: Applications & Power Ranges



Taken from
Robert Nowak
DARPA

Rutgers, 2/23/06

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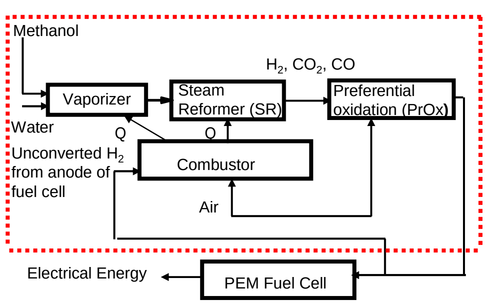
Next Generation Portable Power Sources
Fuel Cells & Fuel Processing

- ❖ As alternatives to batteries
- ❖ Offers high energy density. Allows portable devices to operate for longer times with less recharging

Milliken, DOE, 2002

ONR, S and T



Methanol to hydrogen: Components of fuel processor

- ❖ Generating hydrogen by processing of easily stored liquid hydrocarbons
- ❖ Methanol reforming: attractive source of H_2 for portable PEMFC

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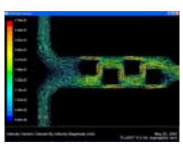
Geometry Limitation in Heat Removal

- Exothermic reactions at high processing rates generate lots of heat
- Thermal resistance leads to temperature non-uniformities (“hot spots”)
- Critical reactions are sensitive to temperature variations
- Can MCS help?--How small is small enough?

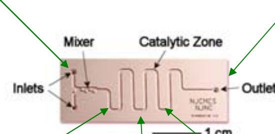
Looking for Improvements with MCS: Preferential Oxidation of CO_2

$$CO \rightarrow CO_2$$



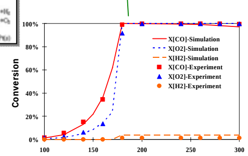
CFD Simulation

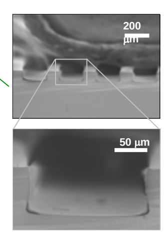
Microkinetic Array



Si Test Reactor

Chemical Kinetics Simulation



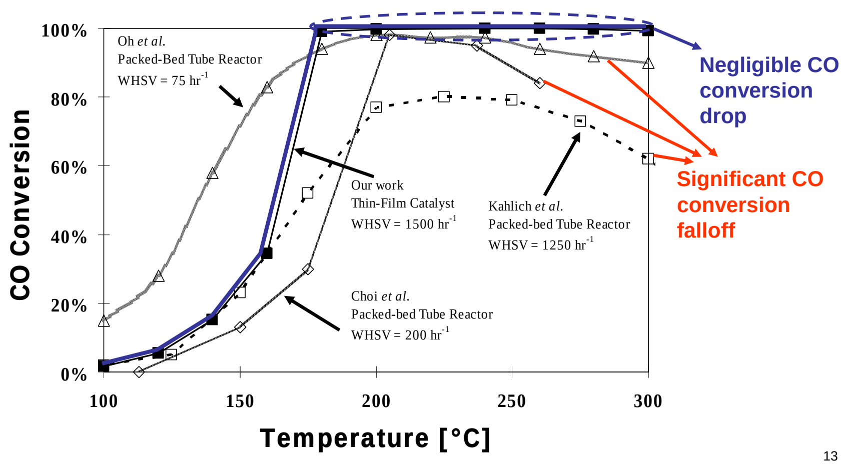


Thin-Film Catalyst Coating

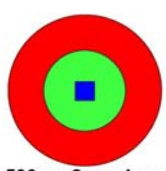
X. Ouyang, L. Bednarova, P. Ho, and R.S. Besser, ***AIChE J.***, 51 (6), 2005.

$$CO + \frac{1}{2}O_2 \xrightarrow{\Delta H = -67 kcal/mol} CO_2$$

$$H_2 + \frac{1}{2}O_2 \xrightarrow{\Delta H = -58 kcal/mol} H_2O$$

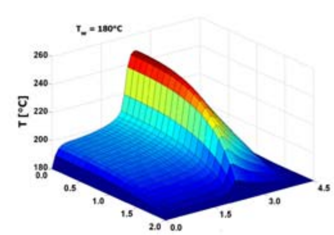


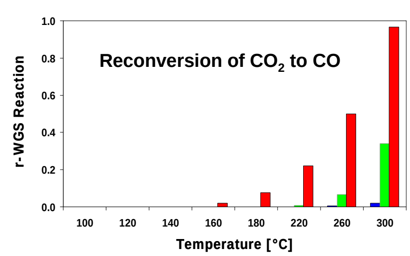
3D Reactor Model



500µm,2mm,4mm

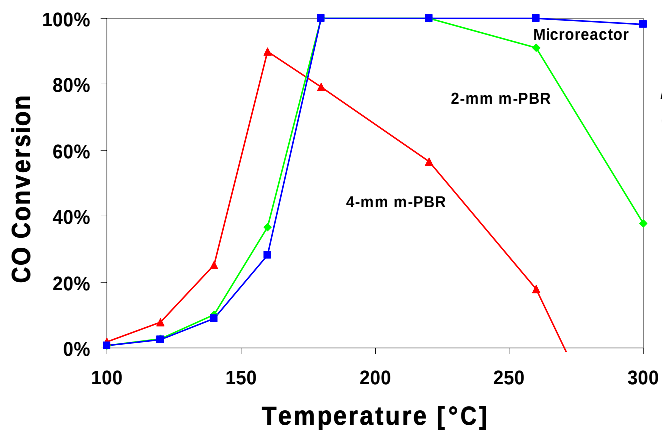
$$CO_2 + H_2 \xrightarrow{\Delta H = 9.8 kcal/mol} CO + H_2O$$





X. Ouyang and R.S. Besser, **J. Power Sources**, 141 (1), 39-46 (2005).

Temperature Dependence of X_{CO}



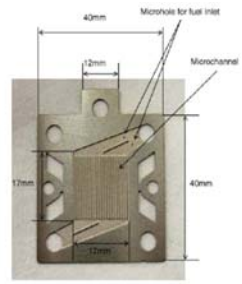
Importance of Geometry

Greater diameter → More severe hot spot formation → Greater r-WGS activity

Key Limitations in MCS and Examples of How We Are Exploring Them

- Geometry Limitation in Heat Removal
- Thermal Integration Limitation in a Multiunit System
- Multiphase Mixing Limitation
- Integrated Process Control Limitation

Microfabricated reactors are flat and sheet-like

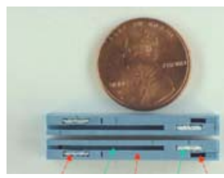


1. **High Surface to Volume**
2. **Lossy, bad for heat retention**
3. **Good for inter-unit heat transfer**
4. **But units need to operate at their <u>individual</u> <u>optimal</u> <u>temperatures</u>**

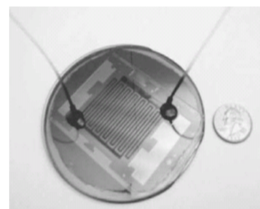
Assembled cat. Combustor/reformer (S.K. Ryi et al. Chemical Engineering Journal 113 2005 47-53)

Fuel processor module. L. Thompson, Univ. Mich.

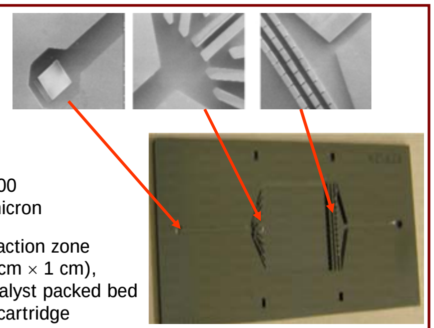
Vaporizer-reformer stack (O.J. Kwon et al.)

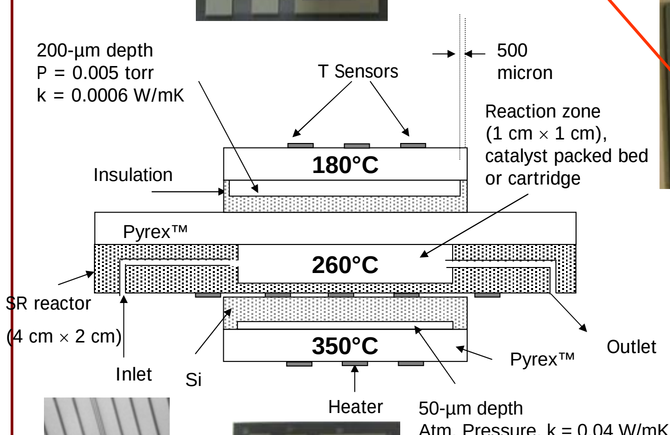


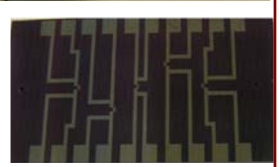
Solid State Research Center, Motorola Labs



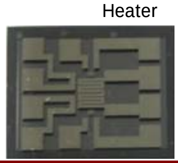
Pattekar-Kothare, IEEE-JMEMS

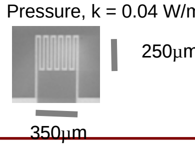




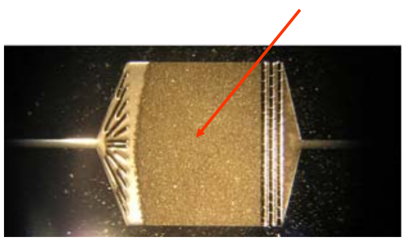


Temp.sensor-center of heater

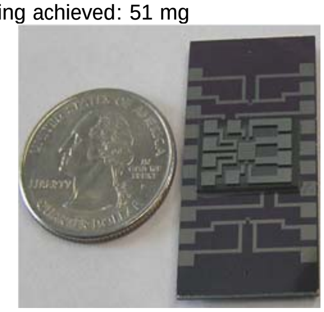




Integrated Fuel Processor

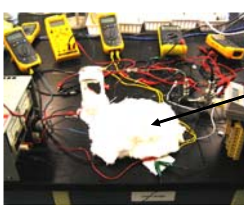


Incorporation of catalyst in the form of packed bed by vacuum loading. Catalyst loading achieved: 51 mg



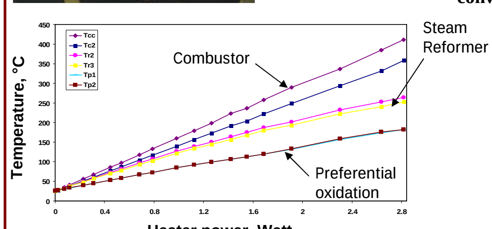
Thermal Characterization

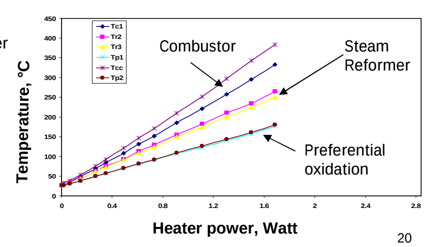
Thermal Characterization: Measure critical thermal parameters like Q required, transfer of heat between components, temperature profiles in different components, insulation effectiveness, and heat loss mechanisms



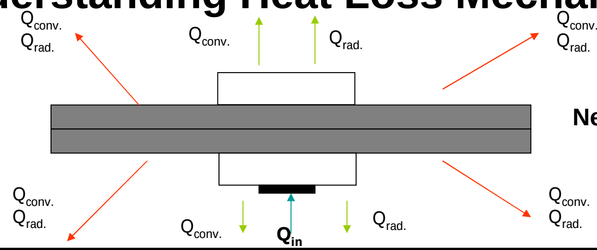
Ambient pressure testing. Device placed in bowl of insulation

Vacuum testing to distinguish multiple modes of heat transport in the system by eliminating convective losses





Thermal Characterization
Understanding Heat Loss Mechanism



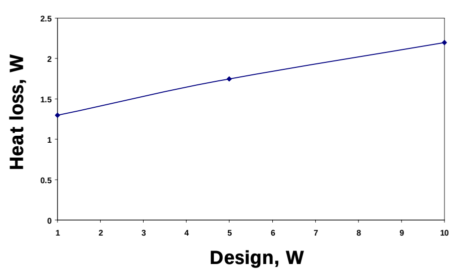
New correlation

Experiments	Q_{total}	T_{comb}	$T_{reactor}$	T_{PrOx}	$Q_{cond.}$	$Q_{conv.}$	$Q_{rad.}$	$h_{conv.}$
Vacuum suspended	3.4	443	260	181	—	0.3	2.9	1.5
Atm. suspended	5.6	517	259	166	—	2.6	3.1	10.3
Vacuum with Al foil	2.2	404	262	190	—	0.3	1.9	1.5
Atm. with Al foil	4.7	476	261	166	—	1.9	2.8	10.3
Vacuum, covered with fiberglass insulation	1.7	386	263	181	1.5	0.2	—	0.5
Atm., covered with fiberglass insulation	2.8	411	263	181	1.7	1.1	—	7.0

Thermal Characterization: Conclusion

1. **Both radiation and convection important**
2. **2. Combination insulation (vac, low-k, reflective) is most effective (insulation design)**

3. Think about 3D architectures instead of sheets (packaging)



Design (W)	Loss (W)
1.3	1.3
5	1.8
10	2.2

Key Limitations in MCS and Examples of How We Are Exploring Them

- Geometry Limitation in Heat Removal
- Thermal Integration Limitation in a Multiunit System
- Multiphase Mixing Limitation
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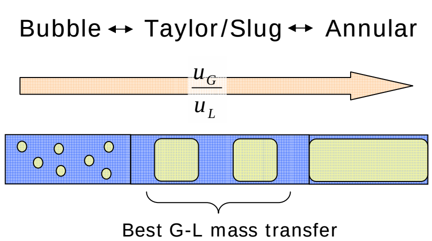
Multiphase (S,L&G) Mixing Is Difficult Without Turbulence

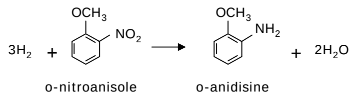
And the pressure drop becomes enormous

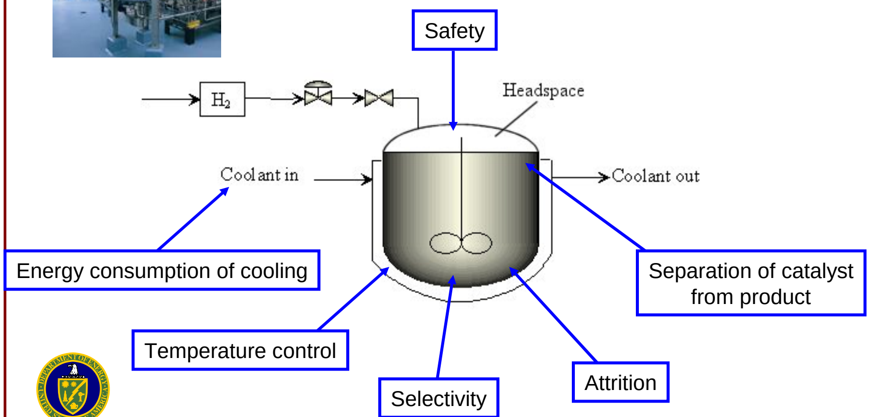
Two-phase flow



- What flow regimes exist within the microreactor?
- How do flow regimes affect the reaction conversion?
- Can we design a microreactor to operate in bubble mode and still be kinetically-controlled?

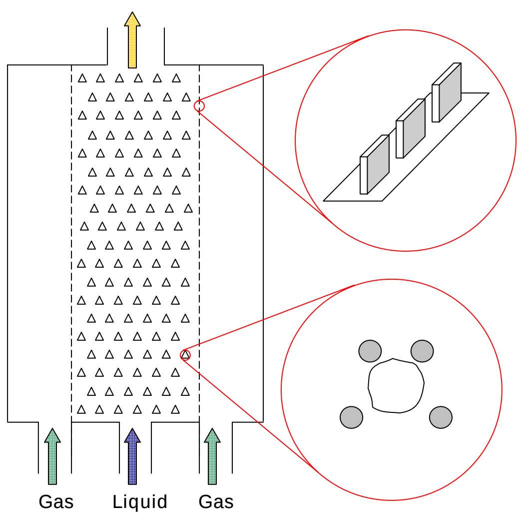
Pharmaceutical Hydrogenation



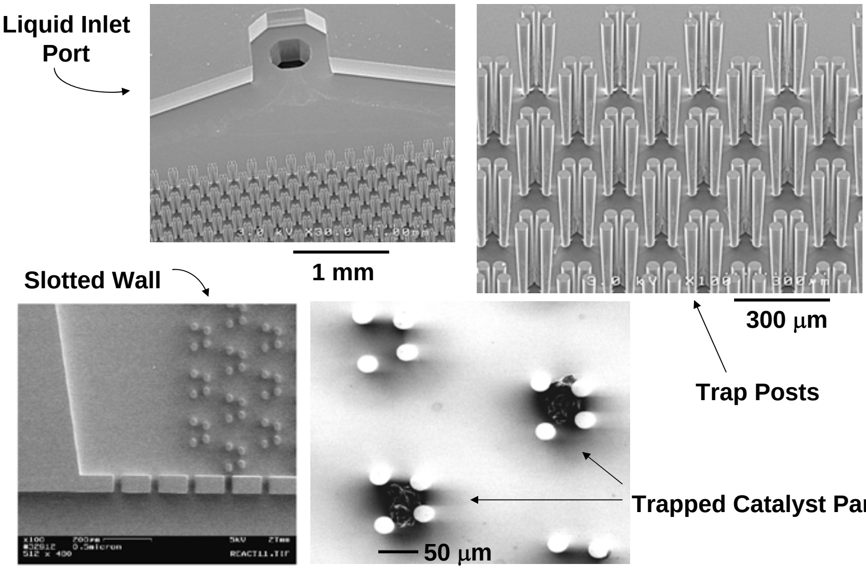


Reactor Concept

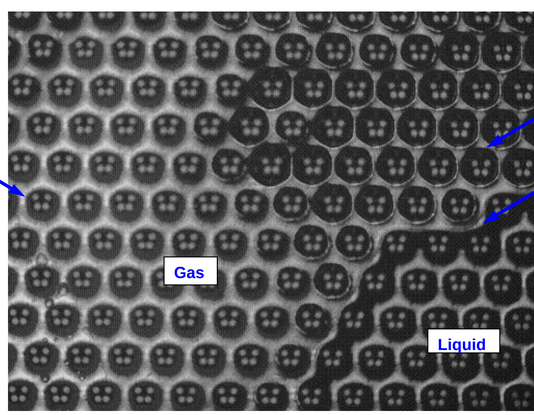
- Separate gas & liquid channels
- Slotted walls-allow gas access along entire length
- Catalyst traps-engineering porosity for ΔP
- Access to range of u_G/u_L



Catalyst Trap Microreactor

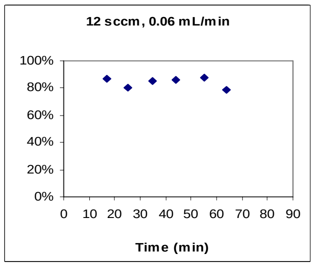


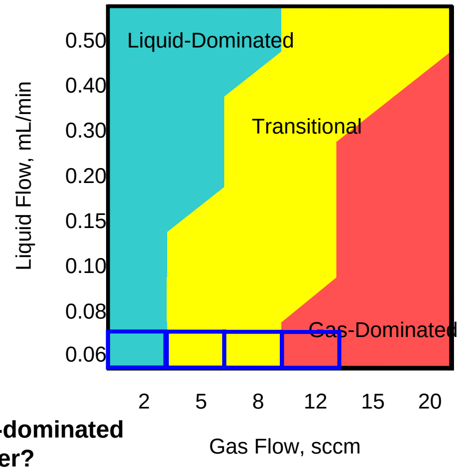
Wetted Traps



- Indication of the type of flow regime present.
- Traps function as a network of “mini-batch” reactors. Measured conversion exceeds model prediction.

Conversion Results (in progress)





Still limited by G/L area?

Can we achieve higher yields in L-dominated region where productivity is greater?

Key Limitations in MCS and Examples of How We Are Exploring Them

- Geometry Limitation in Heat Removal
- Thermal Integration Limitation in a Multiunit System
- Multiphase Mixing Limitation
- Integrated Process Control Limitation

Integrated Process Control Needs for Portable MCS-Based Systems

- Integrated sensing (temperature, flow, pressure, etc.)
- Integrated actuation (valves, heaters, etc.)
- Control strategy
- Miniature
- Autonomous

We Need a MicroFlow Sensor. Build One.

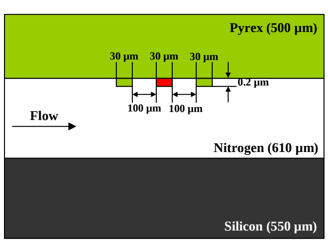
Principle of a Simple Flow Sensor: Calorimetric Sensor

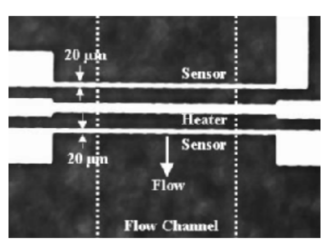
1. Heat generation by Joule dissipation in center resistor.

2. Trigger temperature changes of sensing resistors by convective movement of the fluid stream.

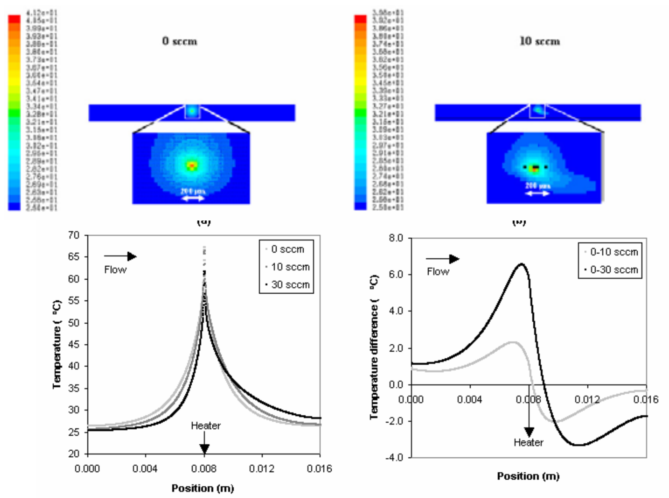
3. Temperature changes lead to resistance changes.

4. Monitoring resistance changes through voltage drop.





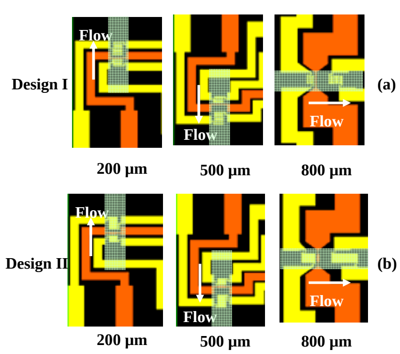
Will It Work and How (CFD Simulation)

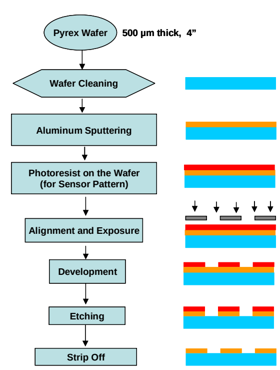


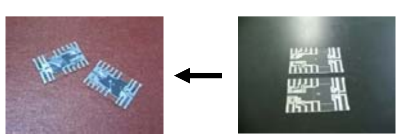
Temperature profiles with different flow rate

Temperature difference between static and flow conditions

Design & Fabrication

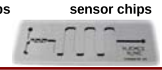




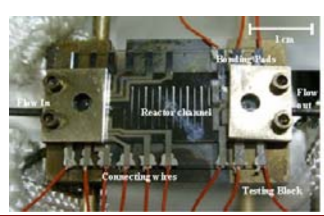


Anodic-bonded sensor chips with microreactor

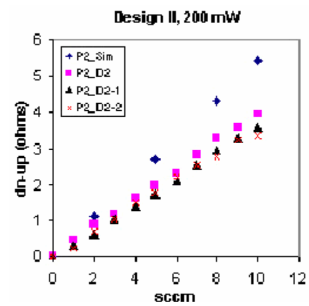
sensor chips



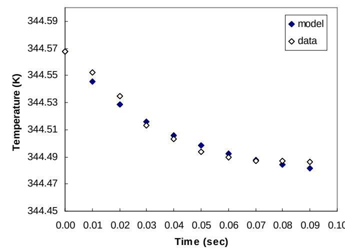
microreactor



Flow Sensor Performance Limitations



<u>Sensitivity</u>
Limited primarily by resistance of sense resistors within thermally affected zone

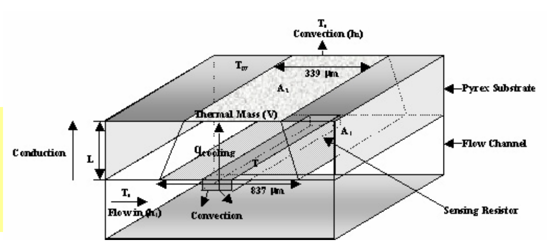


<u>Response time ≅ 70 msec</u>
Limited primarily by mass of Pyrex substrate above thermally affected zone

Flow Sensor Development (Transient Thermal Response)

Lumped Thermal Capacitance Model:

Biot number << 1
(Internal Conduction Resistance/ External Convection Resistance)



Thermal model of sensor

Heat loss from thermal mass = Convection to gas flow + Conduction to substrate

$$Vc\rho(\frac{dT}{dt}) = q_{convection} + q_{conduction}$$

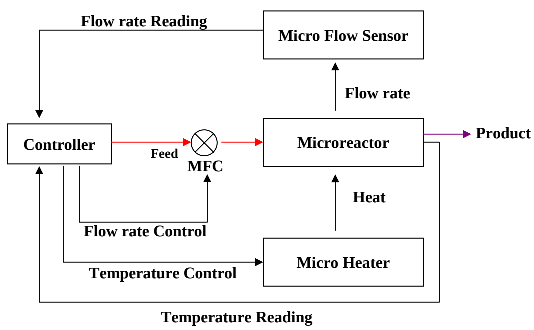
$$Vc\rho(\frac{dT}{dt}) = -A_1 h_1 (T - T_s) - LK_{Py}(T - T_{Py})$$

Flow Sensor Development Summary

- Satisfactory sensitivity
- Matched to the flow range of microreactor (10 sccm)
- Limitations on sensitivity
  - R of the sensing resistor, separation of heater and sensor, and the heating power
- Limitations on response time
  - With reduction of Pyrex thickness, response time could be reduced to < 10 msec
- Integratability, manufacturability, cost

W.C. Shin and R.S. Besser, "Micromachined Thin-film Gas Flow Sensor for Micro Channel Chemical Reactor," J. of Micromechanics and Microengineering, 2006, in press.

A Control Scheme is Needed



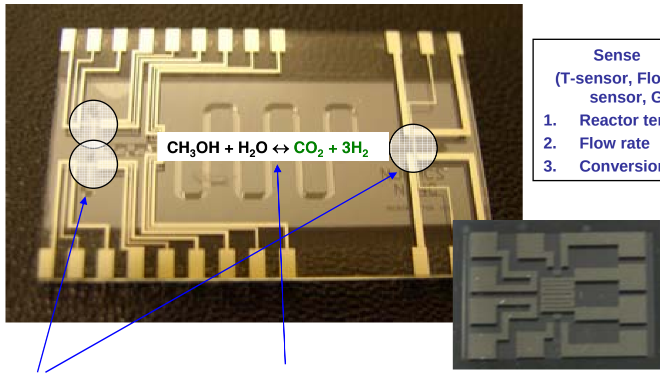
Block diagram of feedback control system for microreactor system

Microchemical System Control

Experimental Plan

Control Unit
(PID, Fuzzy, etc.)
1. Labview algorithm implementation

Actuation
1. Microheater
2. External prop. valve*



Sense
(T-sensor, Flow-sensor, GC)
1. Reactor temp.
2. Flow rate
3. Conversion

Sensing Zone
(Flow & Temp.)

Reaction Zone
(Steam Reforming of MeOH)

Micro Heater
(beneath reactor)

1. Perform the reaction with different control schemes. (Fuzzy logic, PID, etc.)
2. Demonstrating effective control scheme and condition for SR-reaction.

Geometric Limitation on Heat Transfer

1. 500 μm vs. 2 mm made a big difference

Thermal Integration of Separate Units

1. Minimum design 1.3 W

Multiphase Mixing

1. Do novel flow regimes offer an advantage?

Miniature Integrated Control

1. Limitations of integrated flow sensor manageable.

Microchemical Systems

1. Surface to volume ratio

Acknowledgements

People
Prof. Woo Lee, SIT
Prof. A. Lawal, SIT
Dr. Pauline Ho, Reaction Design
Dr. C.S. Pai, NJNC
Mr. B. Mansfield, NJNC
Mr. Ashley Taylor, NJNC
Dr. Stanley Pau, ASU
Prof. Suphan Koven, SIT
Dr. D. Ivanov, NJIT
Dr. K.R. Farmer, Fisher
Mr. Steve Nicolich, TACOM
Dr. Art Kaufman
X. Ouyang, Ida-Tech
L. Bednarova, Ultracell

Agencies, Institutions
U.S. Department of Energy
Defense Advanced Research Projects Agency
New Jersey Commission on Science and Technology
Stevens Institute of Technology
TACOM-ARDEC
Office of Naval Research
Cornell Nanofabrication Facility (NSF)

People (cont.)
K. Shah, W. Shin
S. McGovern, H. Gadre