

Spatiotemporal patterns in a porous catalyst

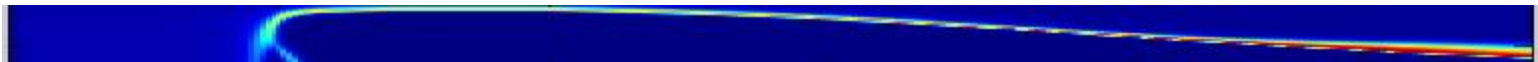
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Presented at 4th International Conference on Unsteady State Processes in Catalysis (USPC-4), Montreal, 2003.




Gradients in Porous Catalysts



- Composition
- Thermal
- Especially under dynamic conditions!

How to measure?

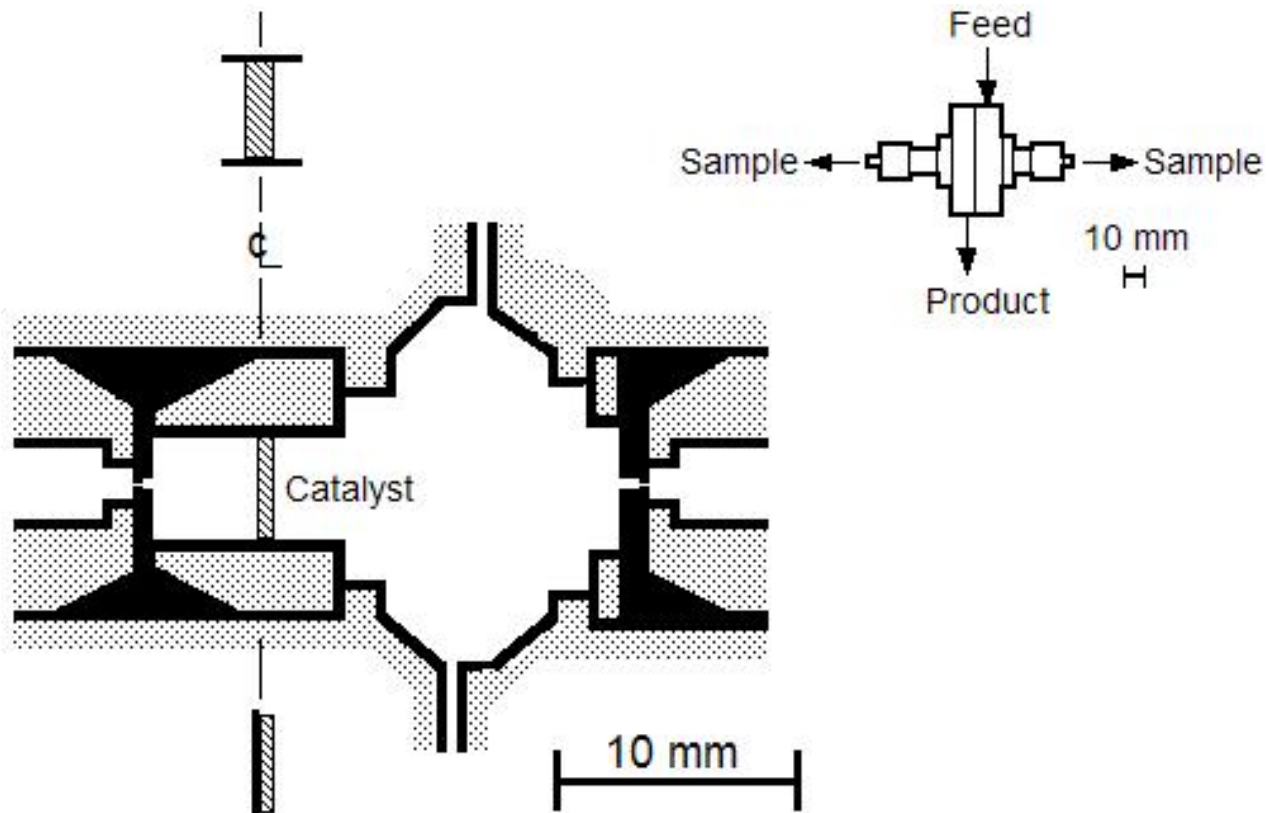
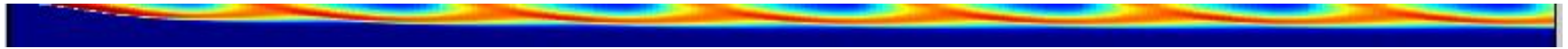
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- Single Pellet Diffusion Reactor at steady state, Petersen and students
 - Thermocouples in pellet, Butt and students
 - Related - infrared imaging parallel to pellet surface

Dynamic Diffusion Reactor

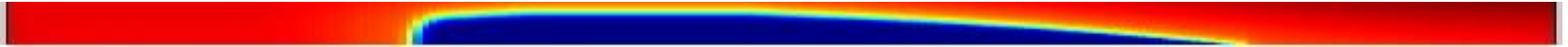


- Miniaturize single pellet diffusion reactor
- Minimize time constants
- Measure composition continuously

Dynamic Diffusion Reactor



DDR - Types of Experiments

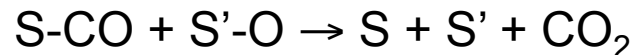
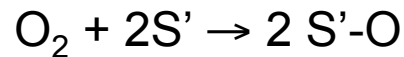
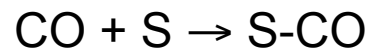


- Inert response →
diffusion coefficients
- Single species response →
adsorption coefficients
- Reaction response →
reaction-diffusion model

DDR - Approach



- Measure concentration gradients
- Model with detailed kinetics



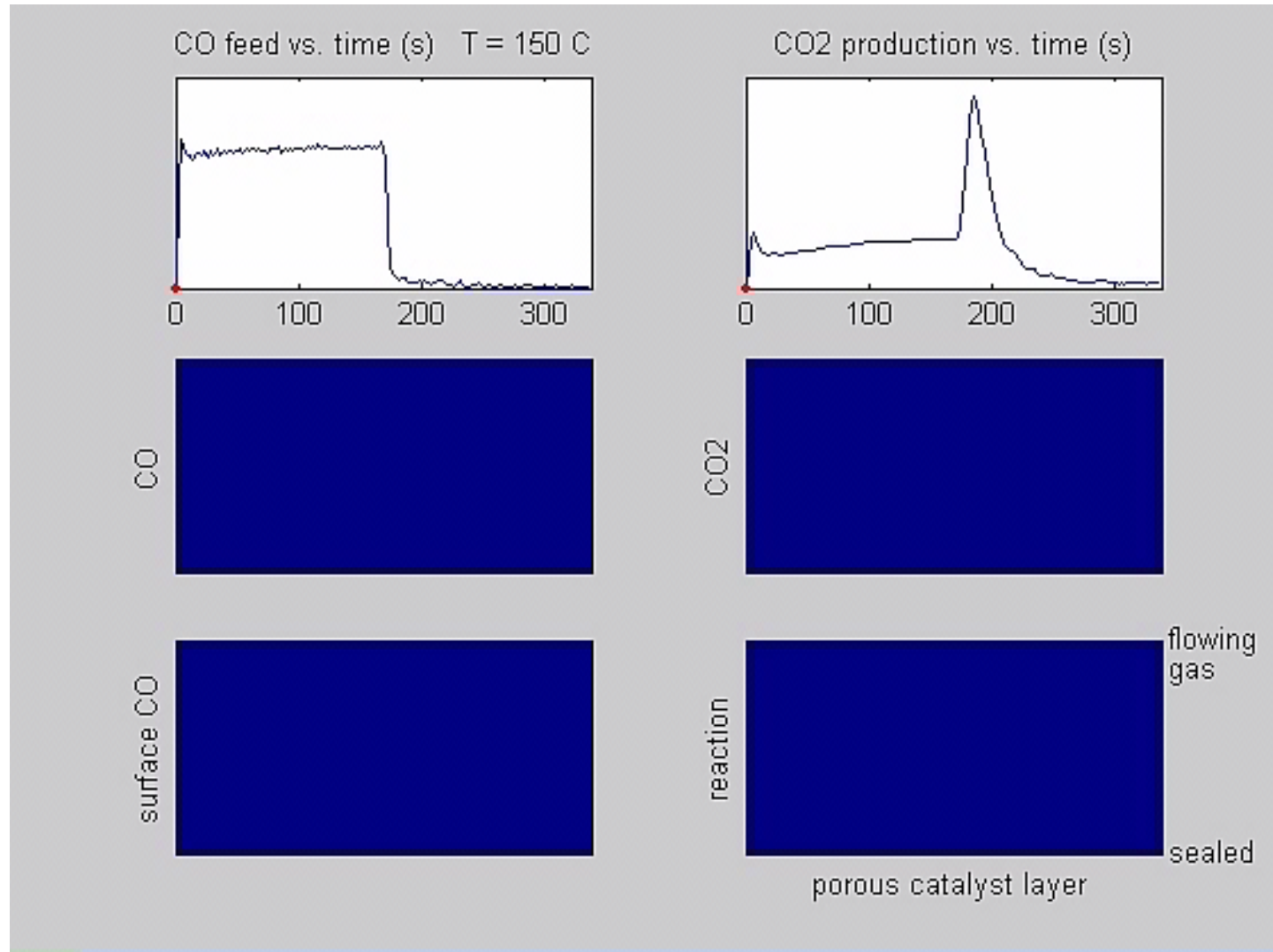
over multiple site types, don't assume adsorption equilibrium or rate-limiting step

R. Herz & S. Marin, J.Catal. 65, 281 (1980)

- Fit model to experiments
- Perform numerical experiments...

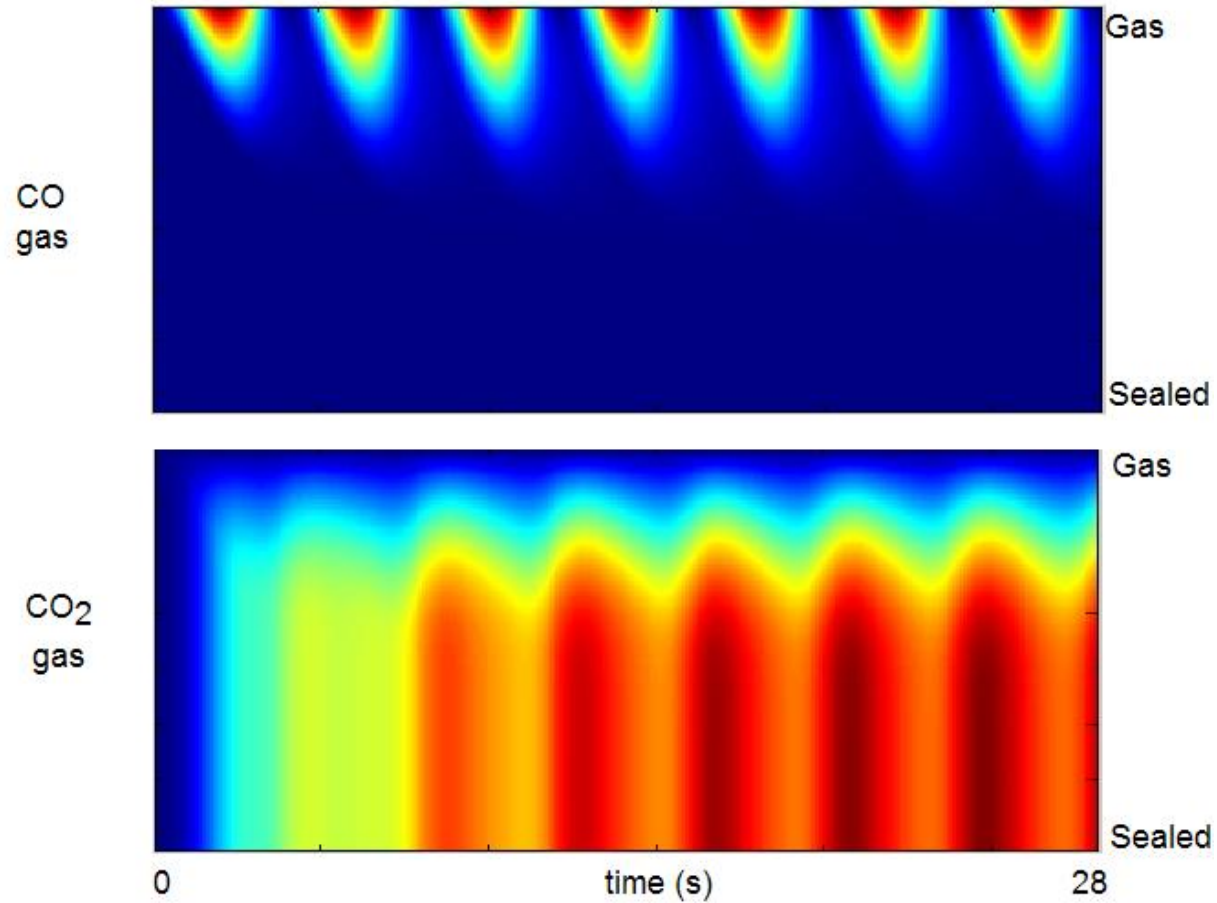
Step Responses

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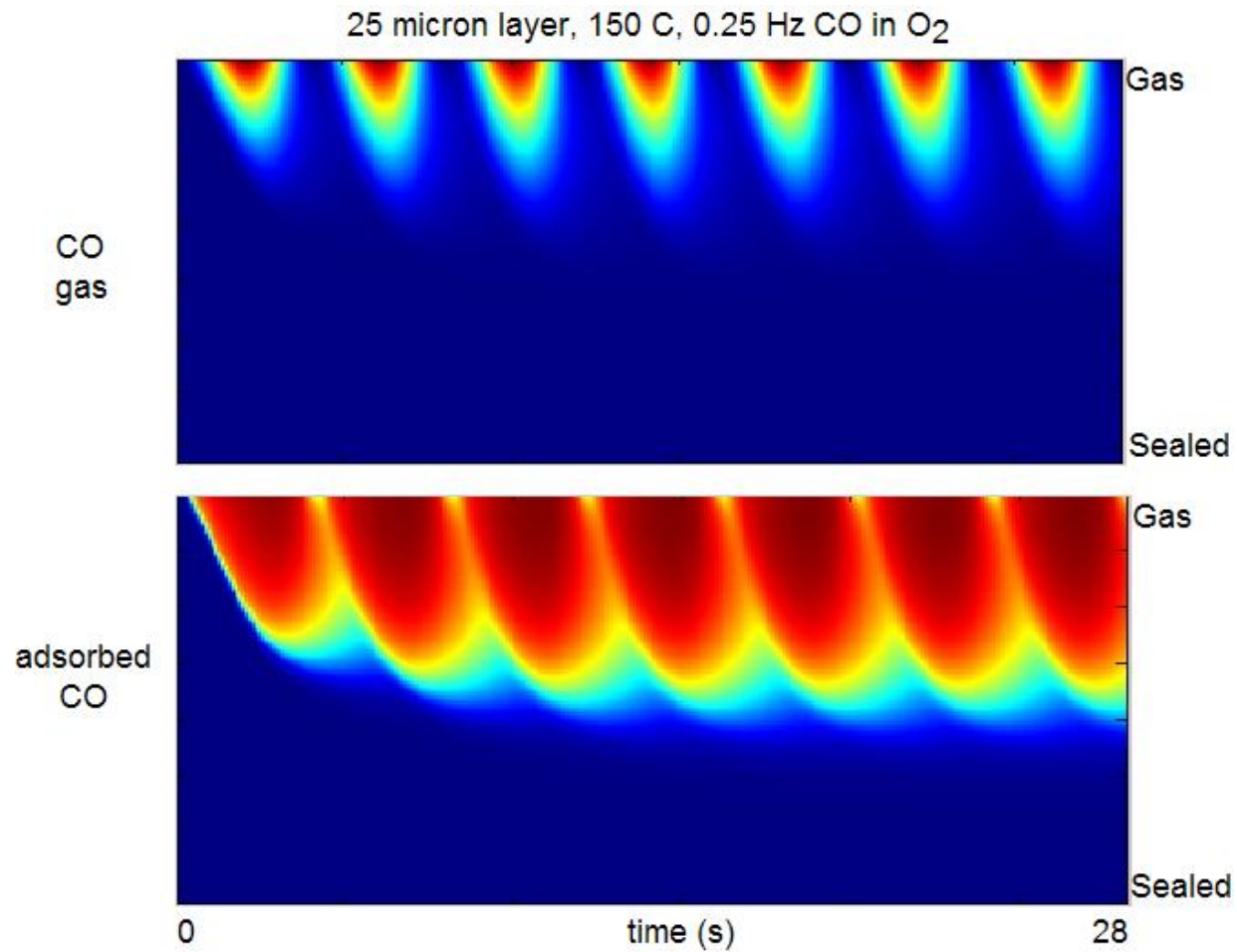


Cycling

25 micron layer, 150 C, 0.25 Hz CO in O₂

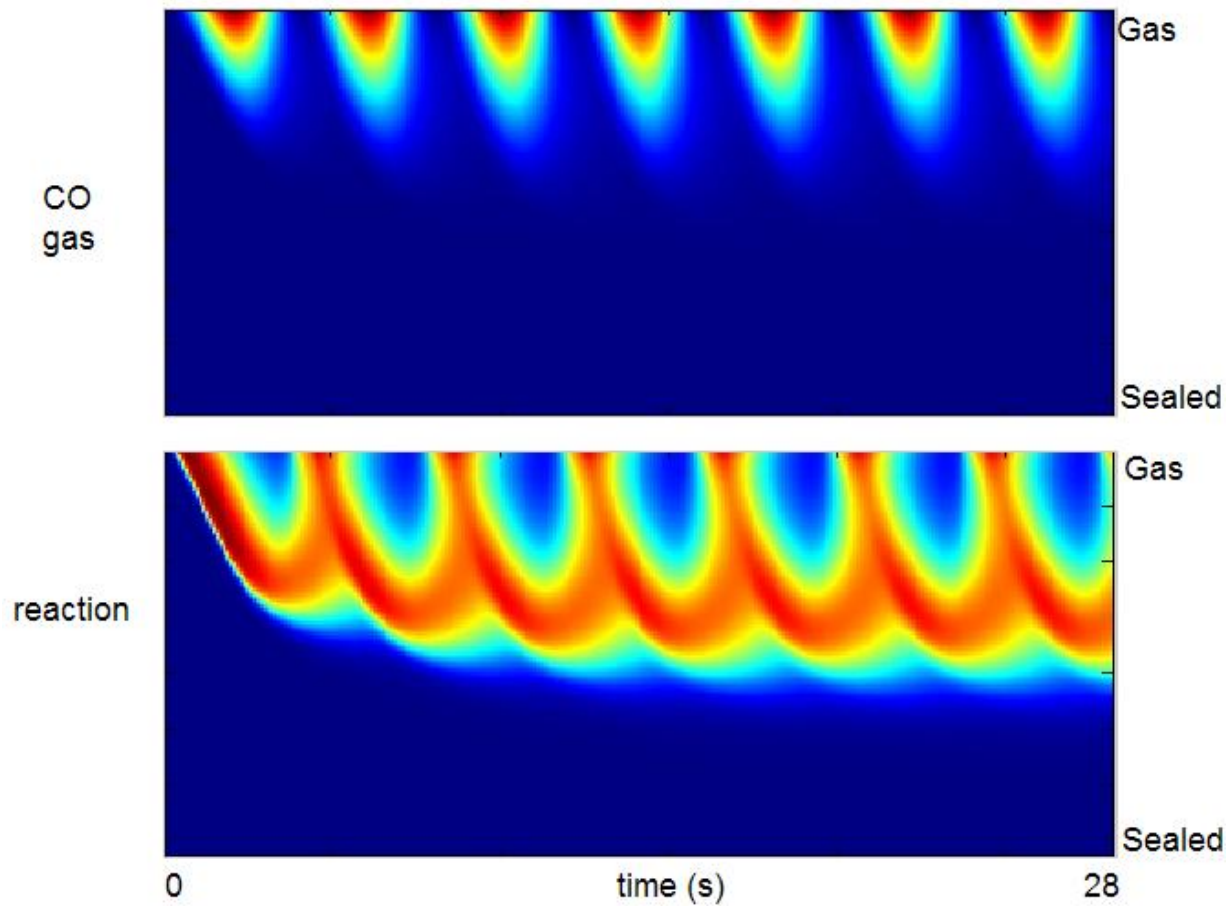


Cycling



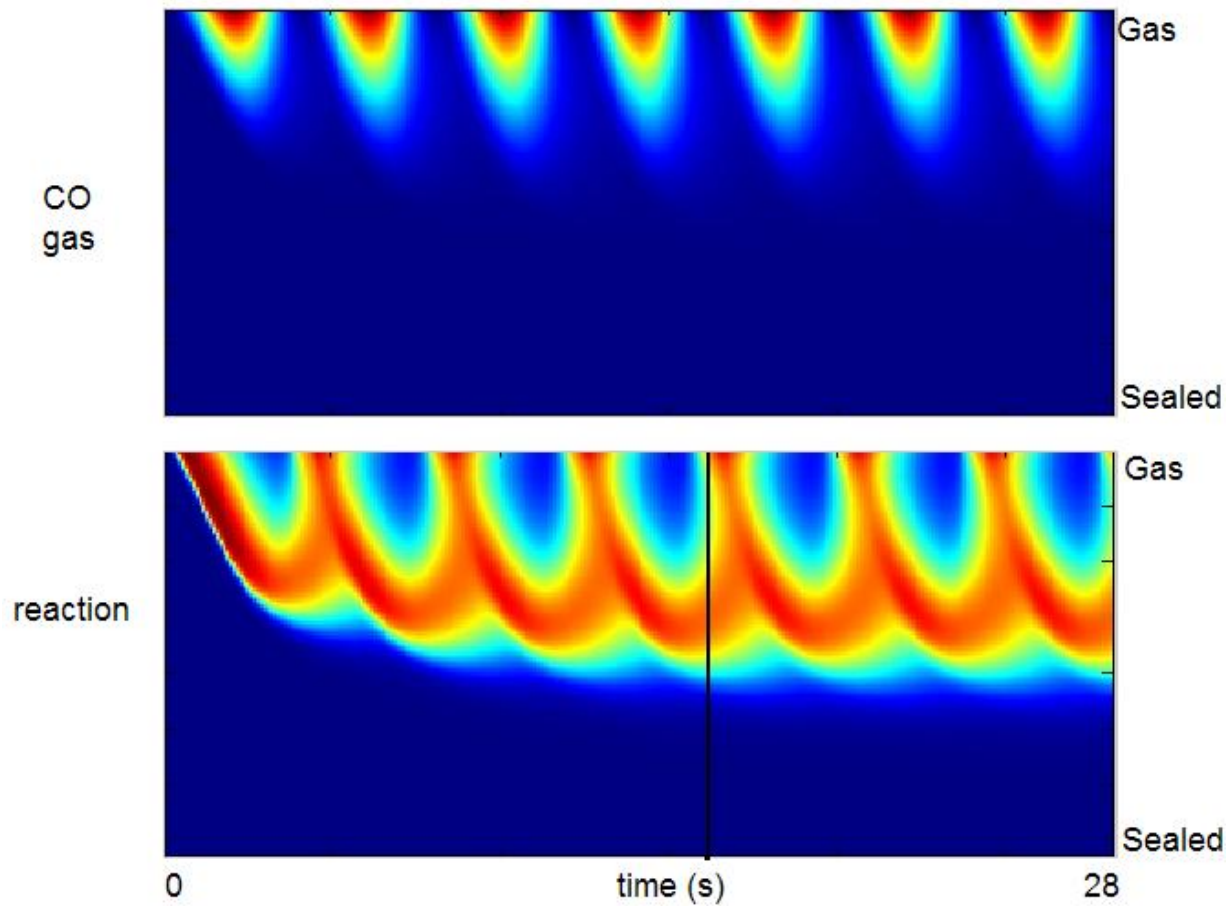
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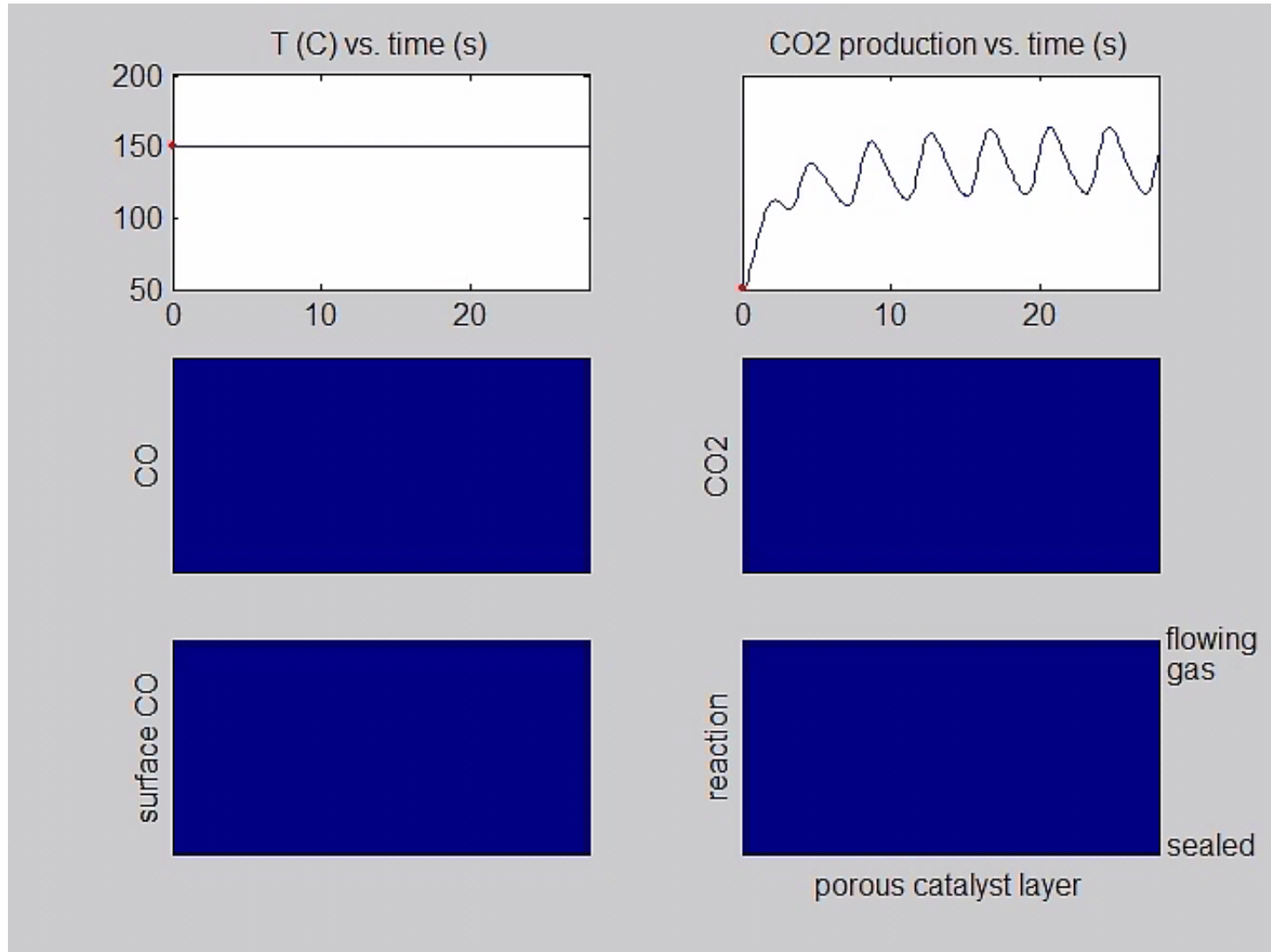
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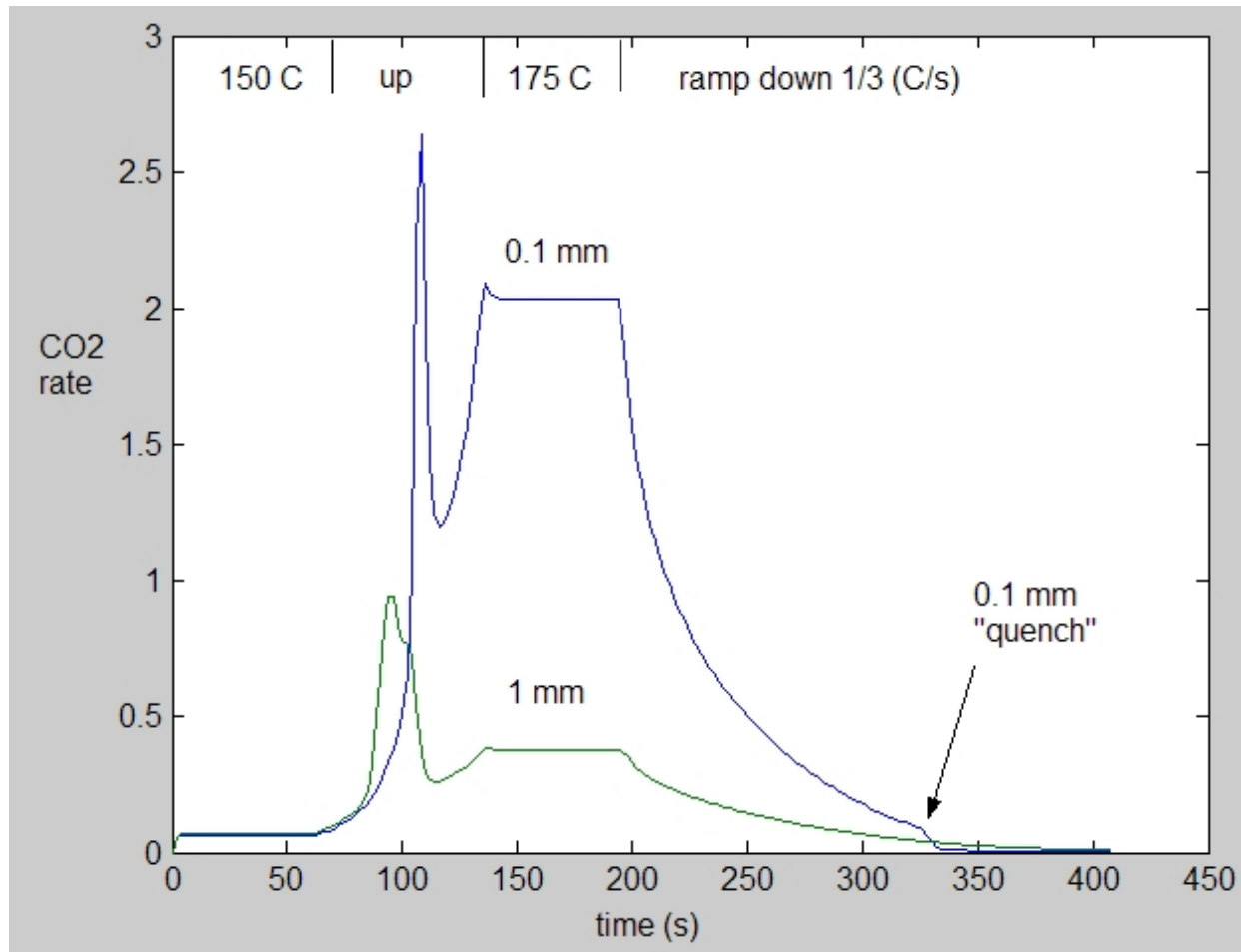


Cycling

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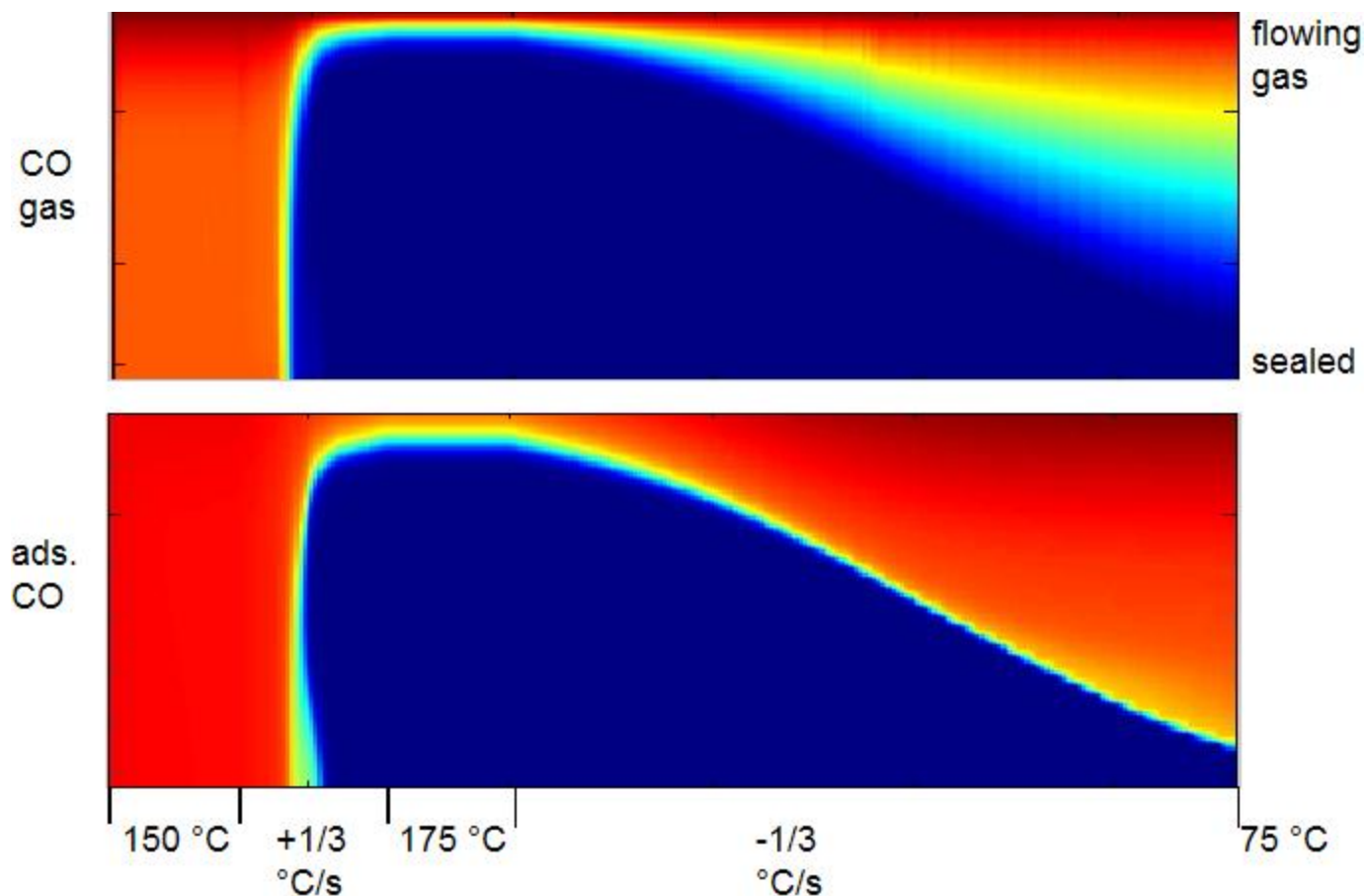


Temperature ramps



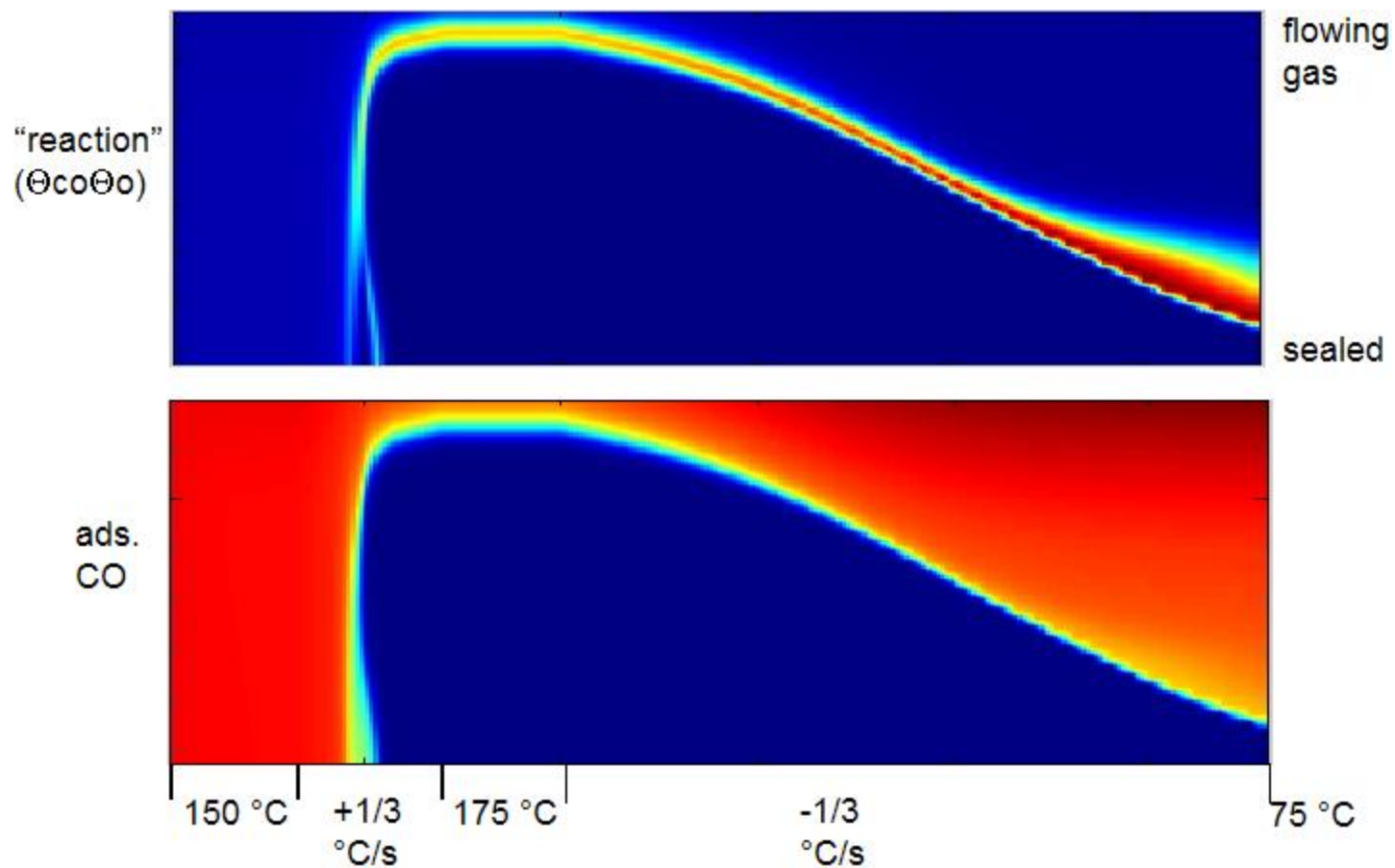
Temperature Ramps

1 mm layer



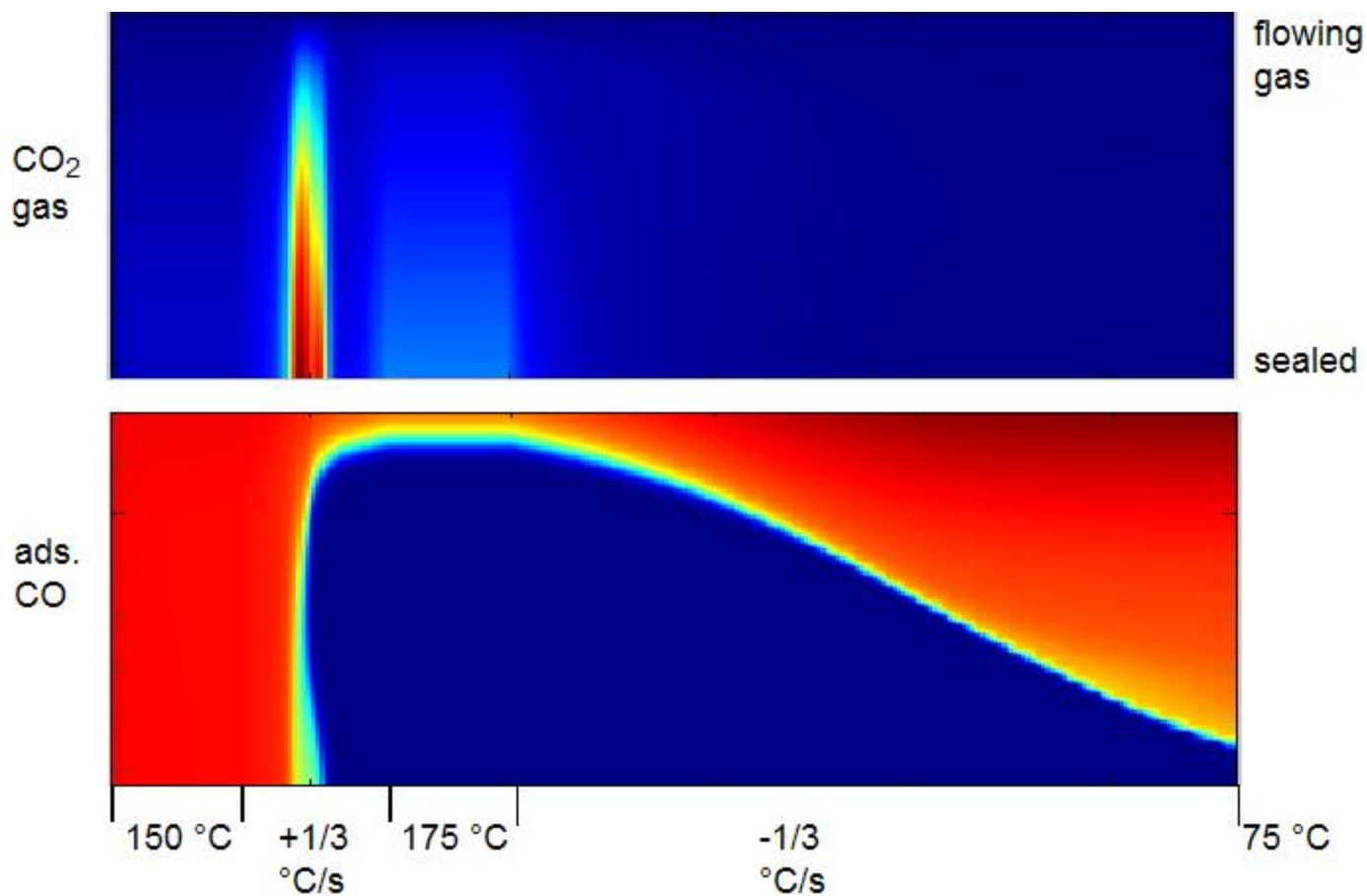
Temperature Ramps

1 mm layer



Temperature Ramps

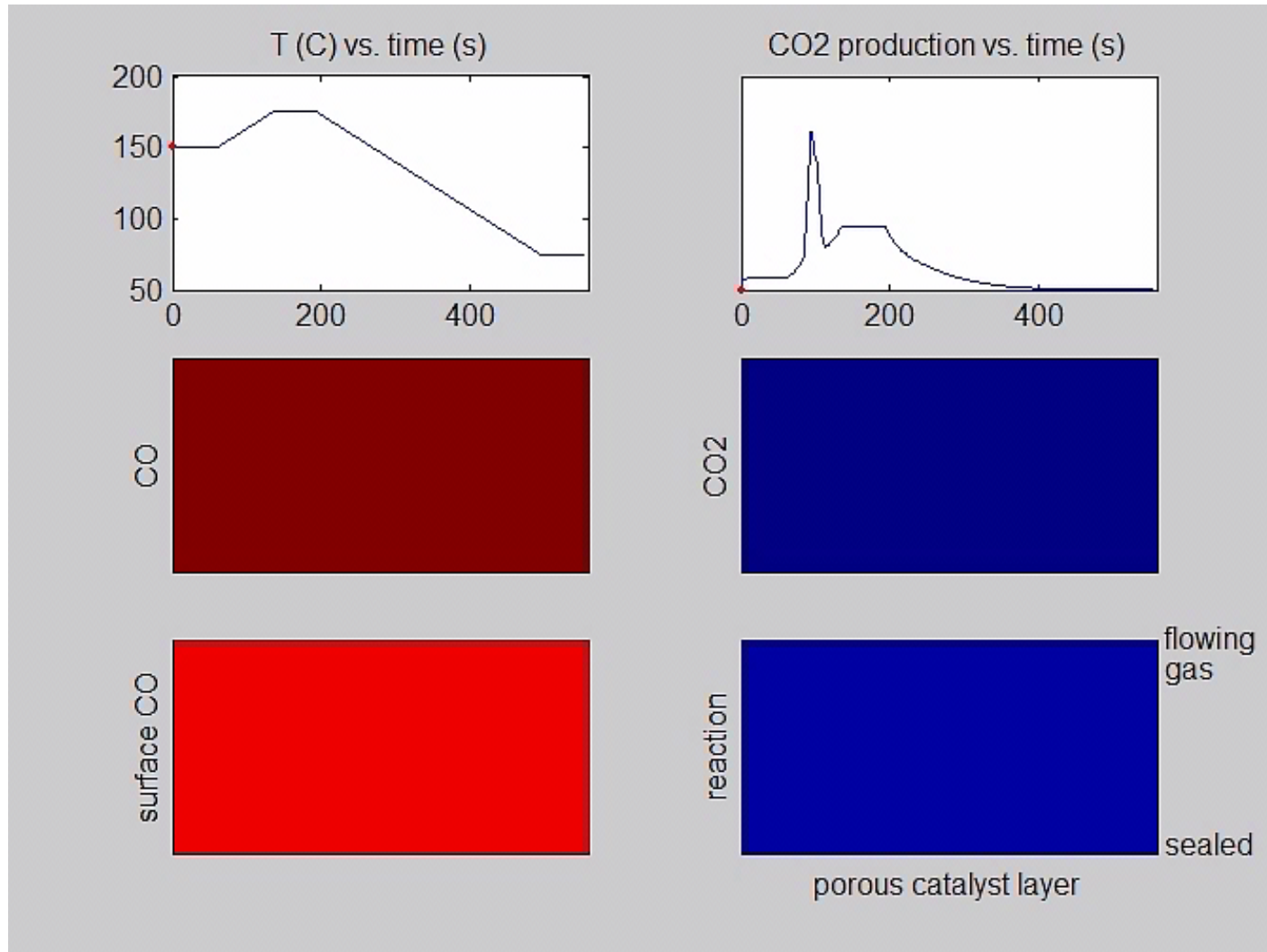
1 mm layer



Temperature Ramps

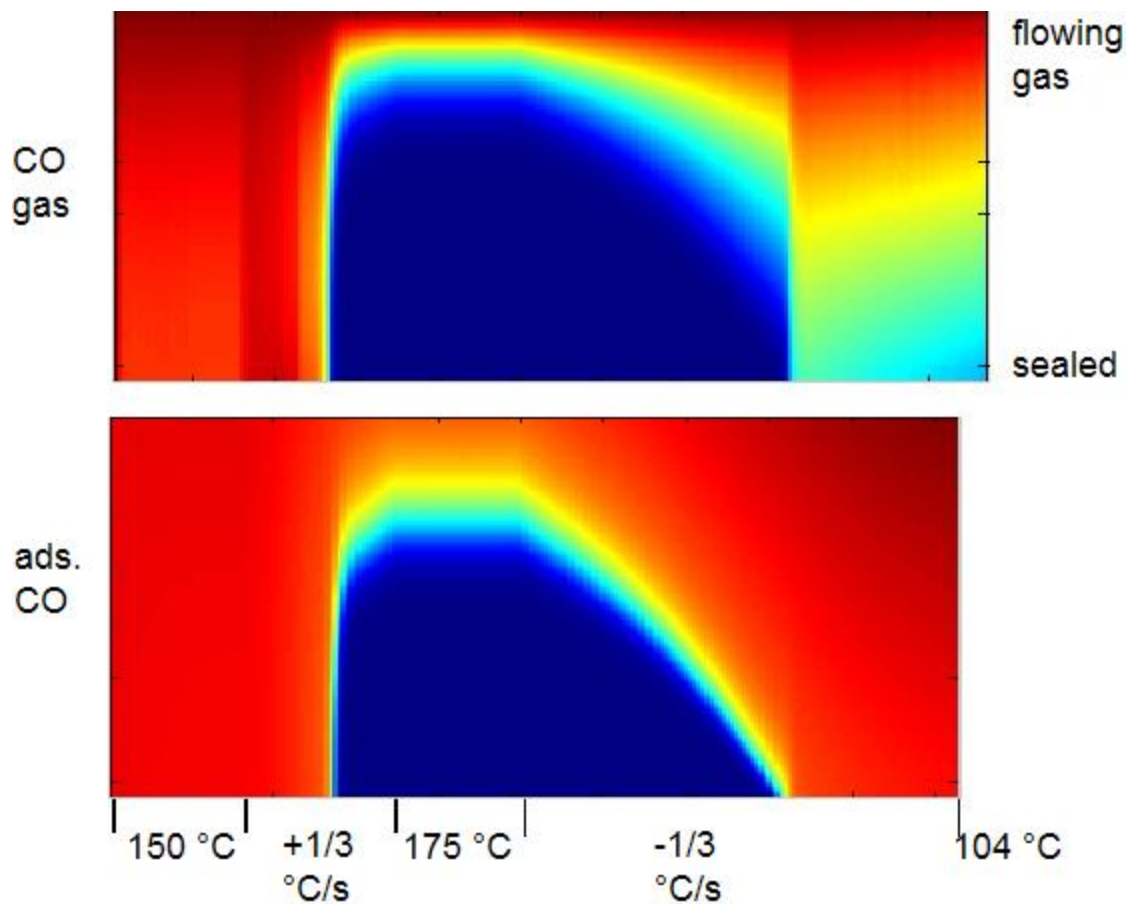
1 mm layer

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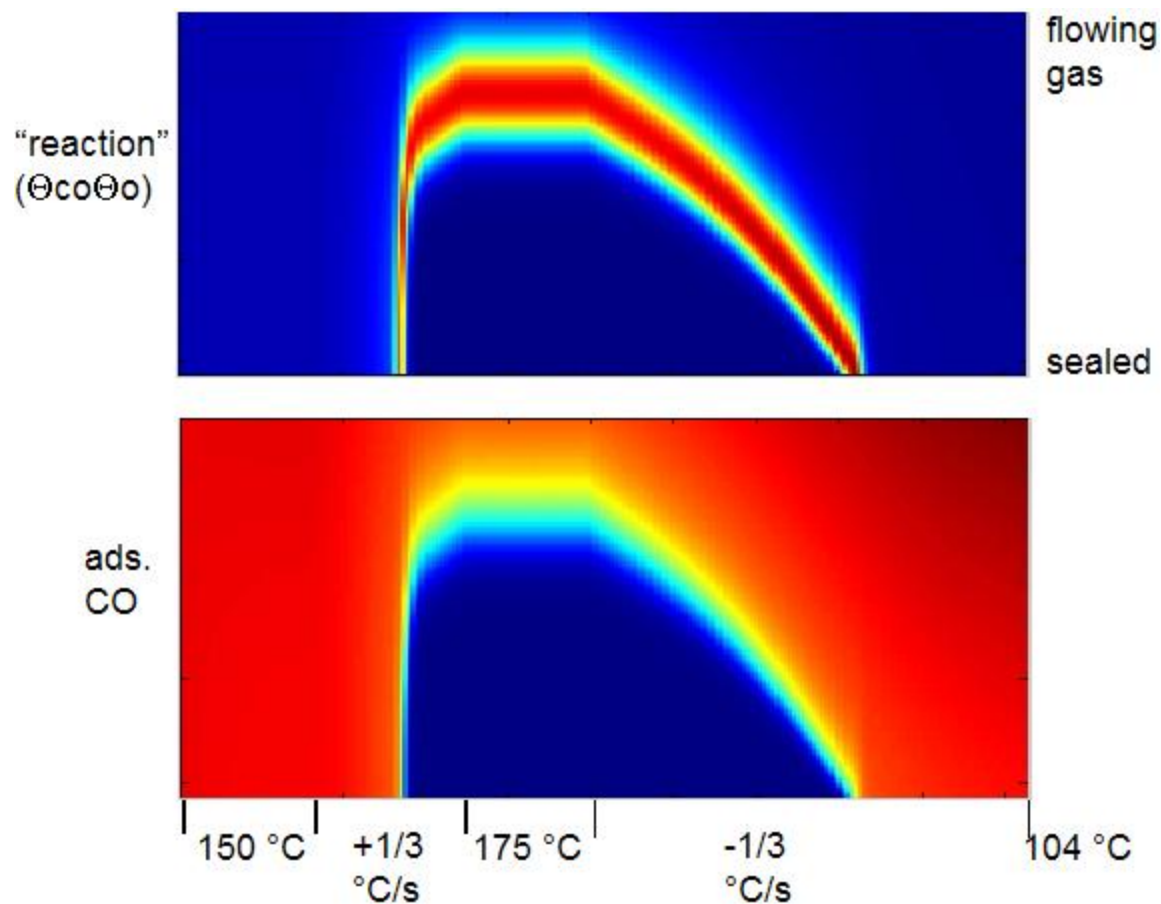
Temperature Ramps

0.1 mm layer



Temperature Ramps

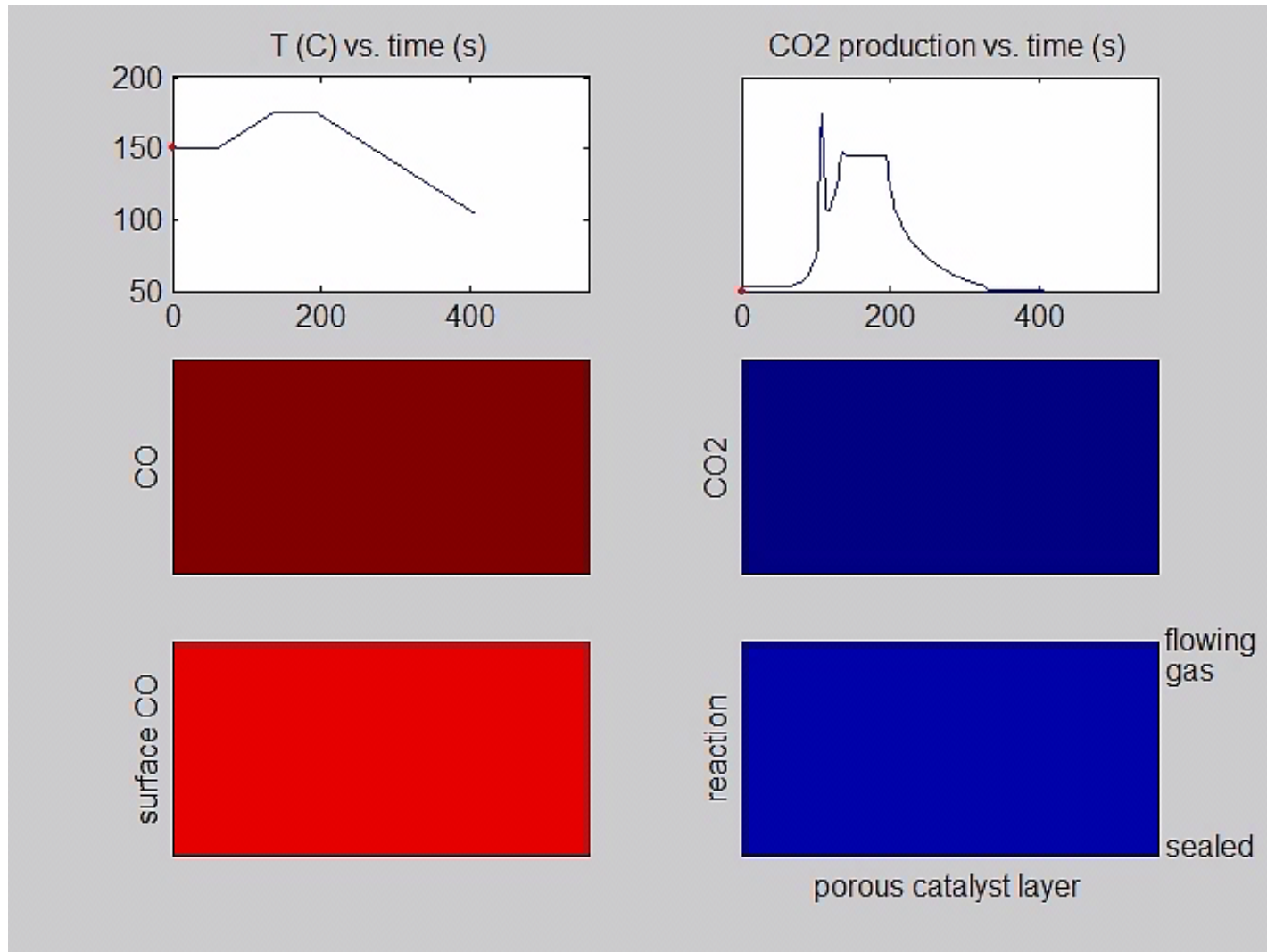
0.1 mm layer



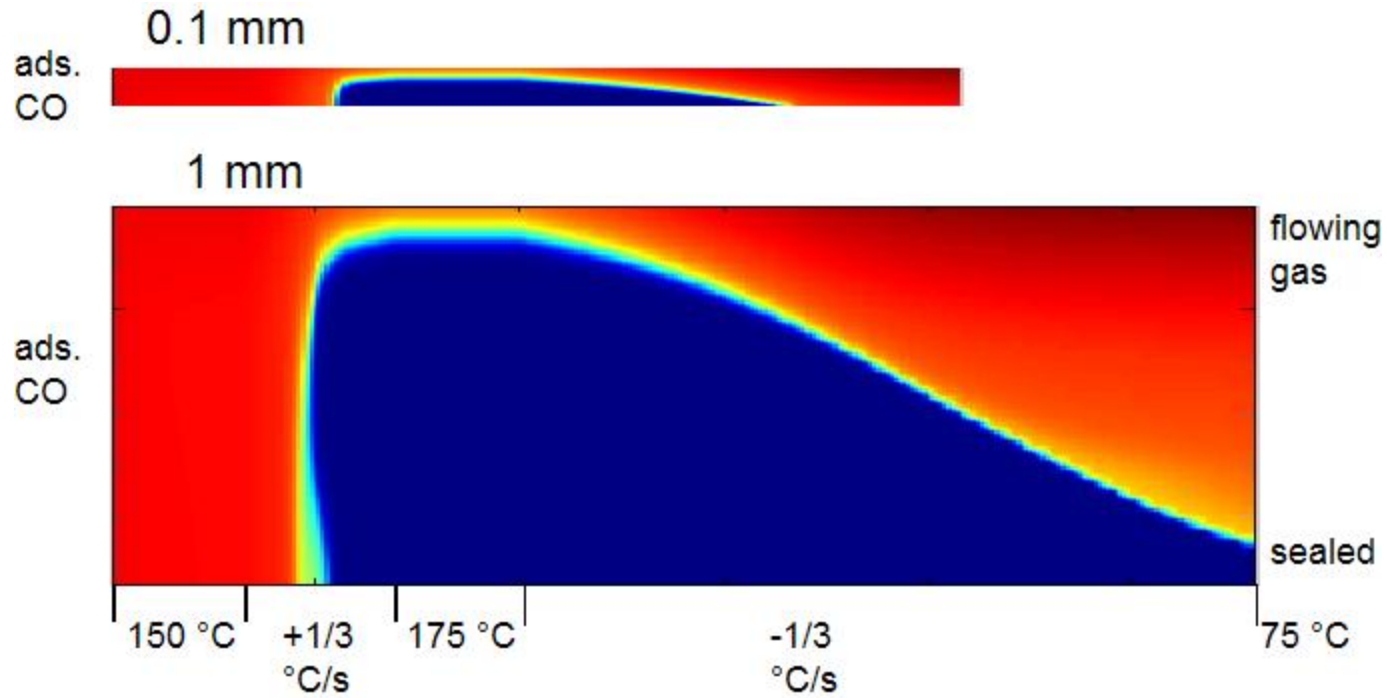
Temperature Ramps

0.1 mm layer

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Temperature Ramps

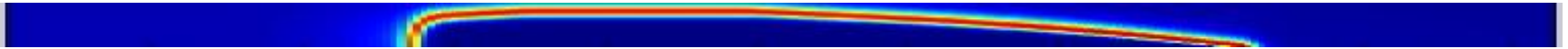


Temperature Ramps



- At constant total Pt, the following increase as catalyst thickness decreases:
 - overall reaction rate
 - “light off” temperature
 - “quench” temperature
- Thus, the catalyst is more active but over a narrow temperature range.

Temperature Ramps



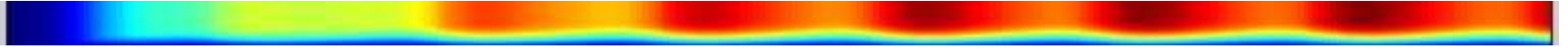
Patterns change with catalyst thickness:

1 mm – double reaction fronts move from
center on “light-off”

0.1 mm – reaction “quenches” as CO front
reaches center

≤ 0.06 mm – CO gas peaks in center, reaction
front starts at exterior

Spatiotemporal Patterns



- Questions?

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