Hands-on Tutorial:
Optimizing Experiments with Pyomo.DoE

`dowlinglab.github.io/pyomo-doe`

Prof. Alexander (Alex) Dowling

Chemical and Biomolecular Engineering
University of Notre Dame

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PYOMO DoE

Contributors to Pyomo.DoE: Jialu Wang (ND), Dan Laky (ND), Hailey Lynch (ND), John Siirola (SNL), Bethany Nicholson (SNL), Miranda Mundt (SNL)

Power of Adaptive Sequential Optimal Experiments

Self-Driving Laboratories

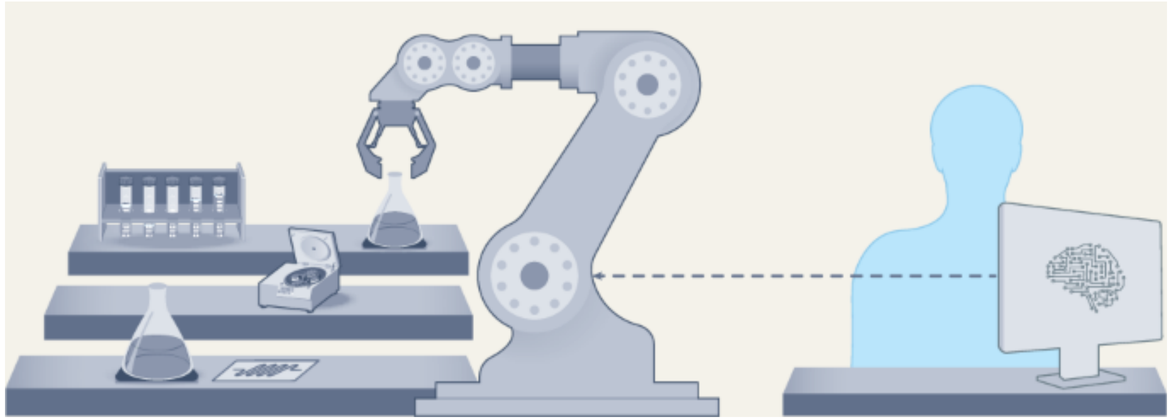
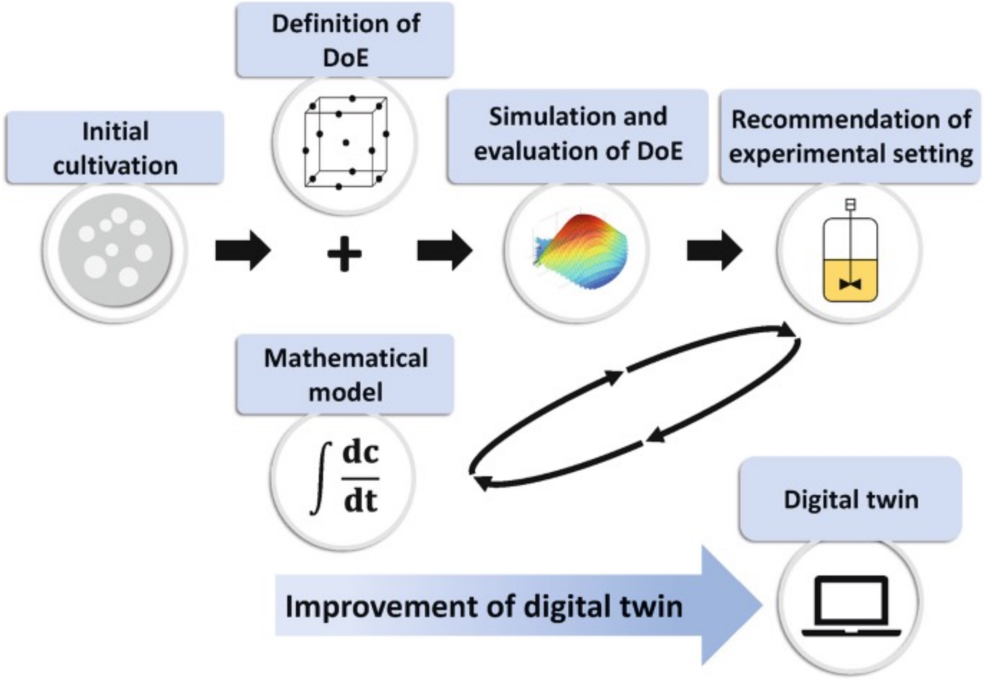


Figure: Abolhasani & Kumacheva (2023), *Nature Syn.*

Epps et al. (2022), *Advanced Materials*
MacLeod et al. (2020), *Science Advances*
MacLeod et al. (2022), *Nature Communications*
Hase, Roch, Aspuru-Guzik (2019), *Trends in Chemistry*
Seifrid et al. (2022), *Acc. Chem. Res.*

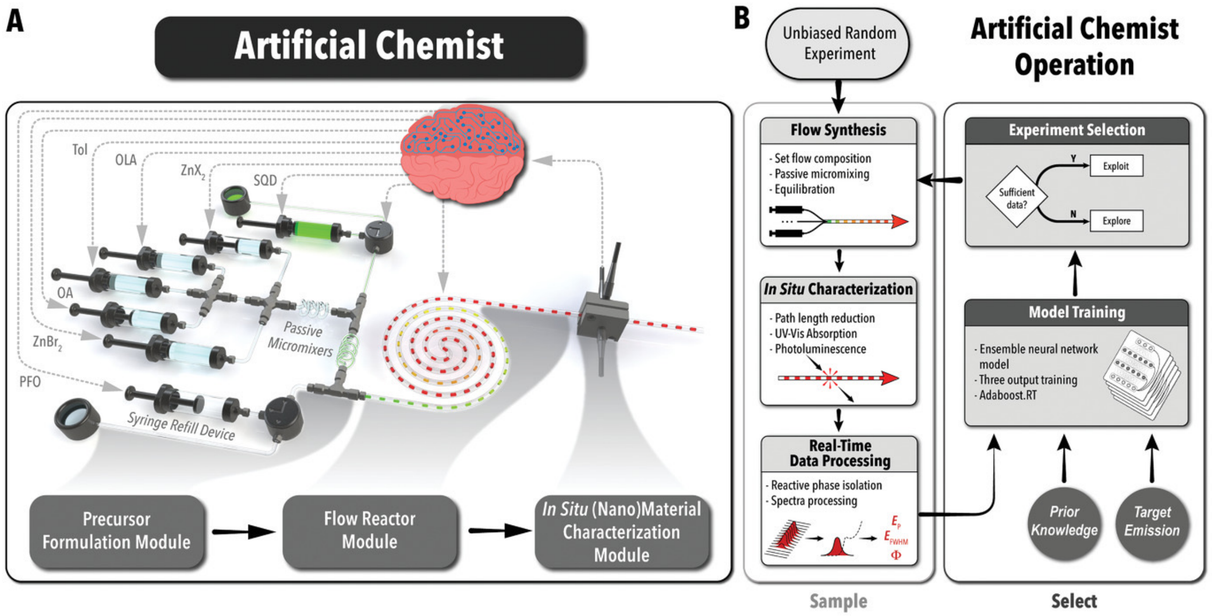
Automation + Model-Based Design of Experiments



Kuchemuller et al. (2020), *Digital Twins*

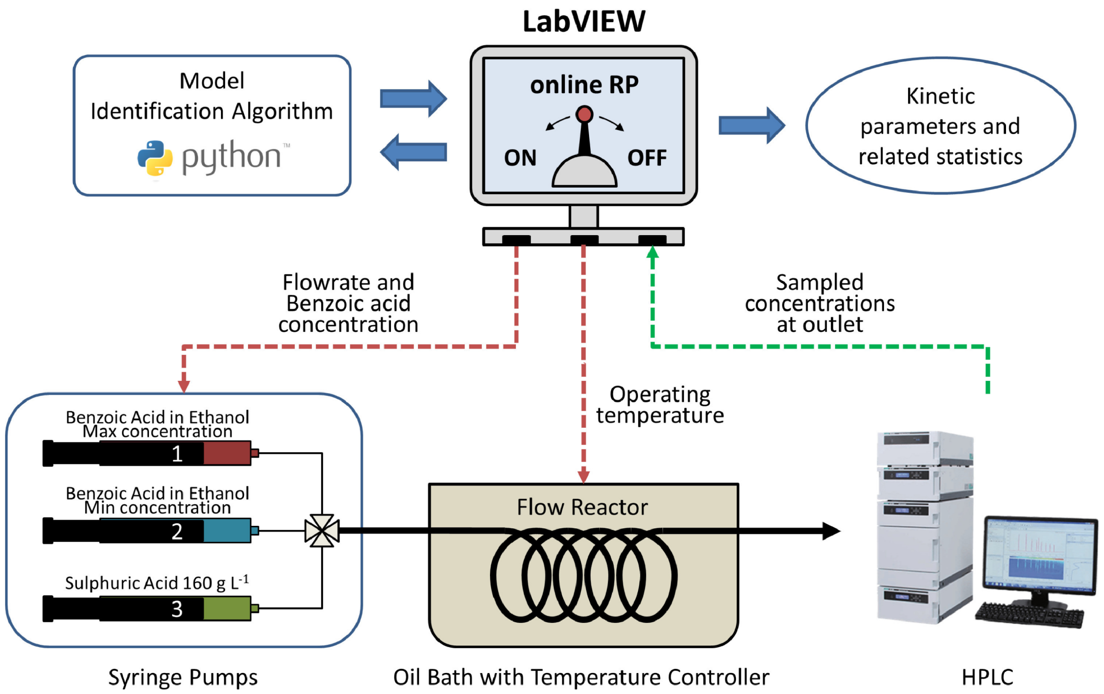
Many Recent Examples of Sequential Optimal Experiments

Quantum Dots
(Machine Learning)



Epps et al. (2022), *Advanced Materials*

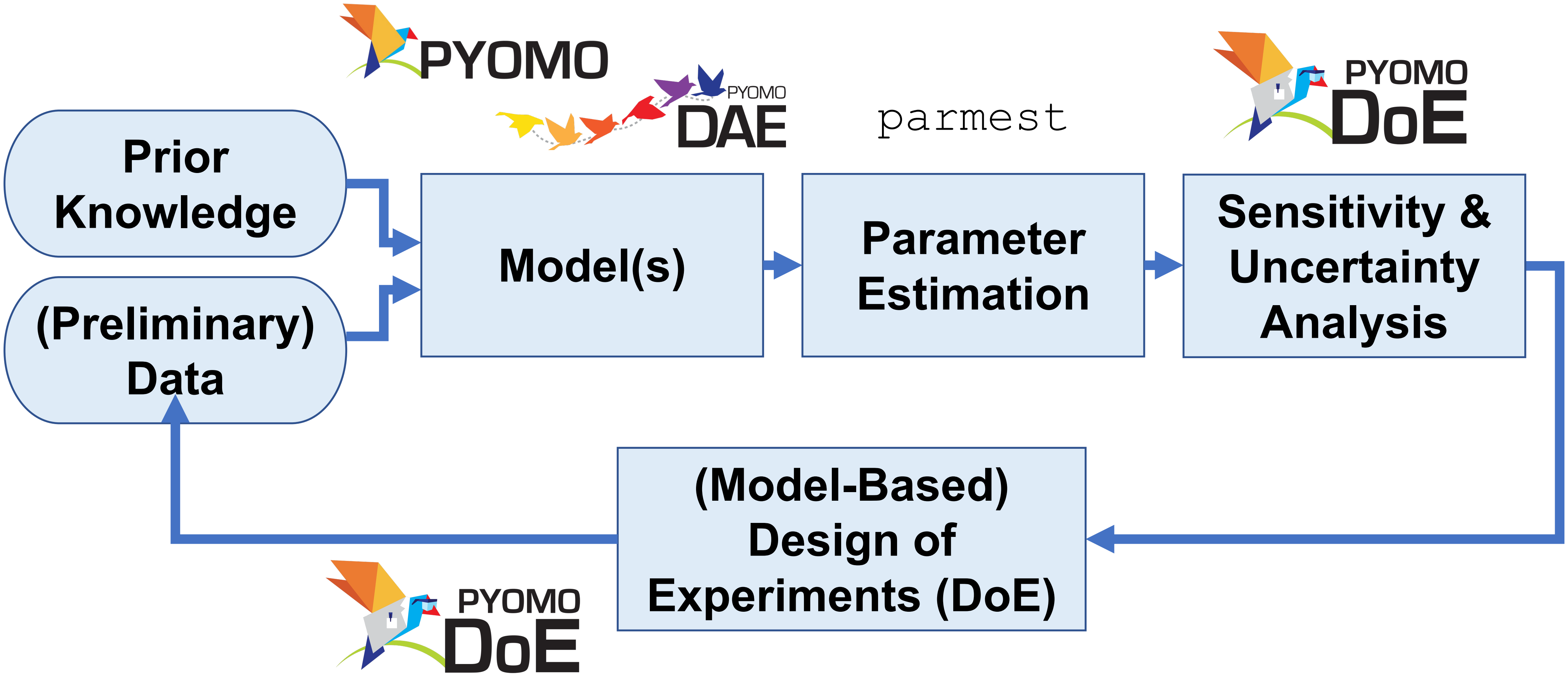
Reaction Engineering
(Science-based Models)



Quaglio et al. (2019), *Comp. & Chem. Eng.*

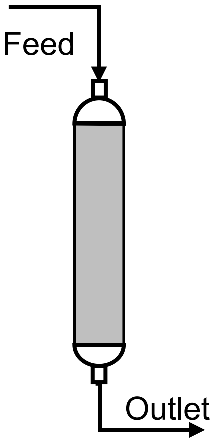
Science-based Data Analytics Workflow

Goal: iteratively develop and validate predictive models (**digital twins**) based on engineering science



MBDoE Facilitates Collaborations

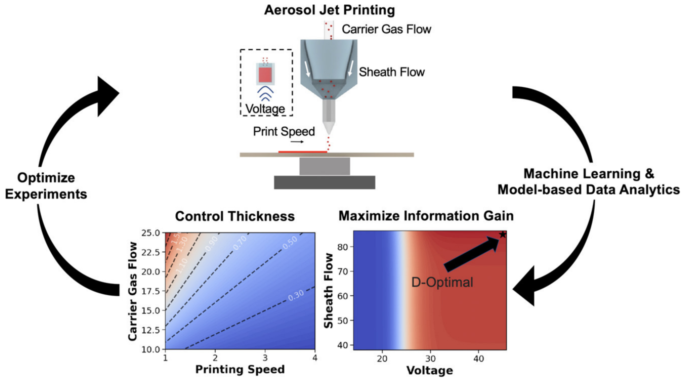
CO_2 Capture



Jialu Wang

Wang, J. and Dowling, A.W. (2022), *AIChE J.* e17813.

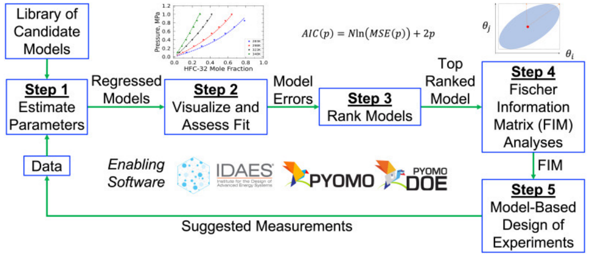
Additive Manufacturing of Thermoelectric Devices



Ke Wang

Wang K., Zhang M., Wang, J., Shang, W., Zhang, Y., Luo, T., Dowling, A.W. (2023), *Digital Chemical Engineering*

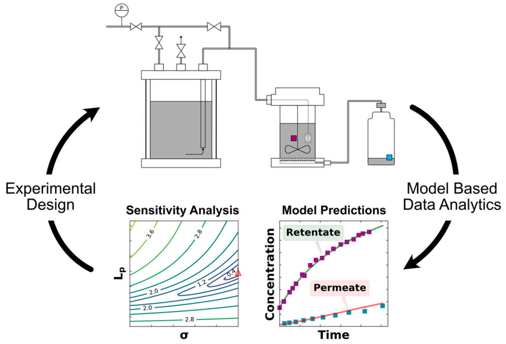
Thermodynamic Modeling (Refrigerants)



Dr. Bridgette Befort

Befort, B.J., Garciadiego, A., Wang, J., Wang, K., Maginn, E.J., Dowling, A.W. (2023), *Fluid Phase Equilibria.*

Rapid/Automated Membrane Characterization



Xinhong Liu

Ouimet, J.A, Xinhong, L., Brown, D.J., Eugene, E.A., Popps, T., Muetzel, Z.W., Dowling, A.W., Phillip, W.A., (2022). *J. Membrane Science.*

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CCSI²
Carbon Capture Simulation for Industry Impact

dowlinglab.nd.edu

Pyomo Team (SNL):

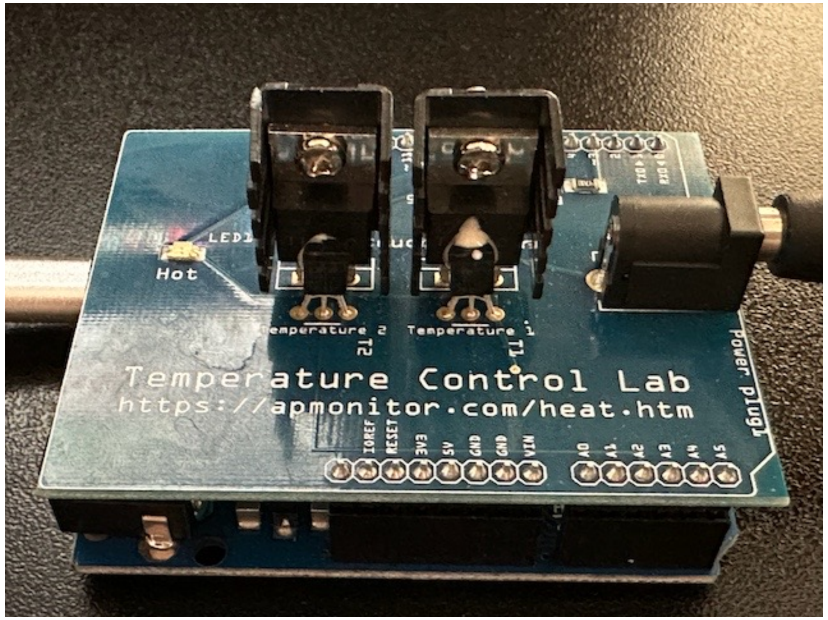
- Bethany Nicholson
- John Siirola
- Miranda Mundt

Contributors (ND):

- Dr. Jialu Wang
- Dr. Dan Laky
- Hailey Lynch

Thank you to Prof. Jeff Kantor (1954-2023) for the TCLab examples and so much more.

Agenda for Today



Welcome and Overview

TCLab Example and Pyomo (1:05 pm)

Parameter Estimation with ParmEst (1:30 pm)

Break (2:10 pm)

Optimizing Experiments with Pyomo.DoE (2:20 pm)

Parameter Estimation and Uncertainty Basics

Assume a model and error structure:

measurement

input variables

observation error (i.i.d.)

$$y_i = m(\boldsymbol{x}_i, \boldsymbol{\theta}) + \epsilon_i \qquad \epsilon_i \sim N(0, \sigma_\epsilon^2)$$

model

parameters

What values of model parameters $\boldsymbol{\theta}$ best fit the data $\boldsymbol{X}$ and $\boldsymbol{y}$?

$$\widehat{\boldsymbol{\theta}} = \arg\min_{\boldsymbol{\theta}} \Psi \coloneqq \frac{1}{2} \sum_i [y_i - m(\boldsymbol{x}_i, \boldsymbol{\theta})]^2$$

best fit estimates

How sensitive are the least-squares objective Ψ to perturbations in $\boldsymbol{\theta}$?

$$\boldsymbol{H} = \begin{bmatrix} \frac{\partial^2 \Psi}{\partial \theta_1^2} & \cdots & \frac{\partial^2 \Psi}{\partial \theta_n \partial \theta_1} \\ \vdots & \ddots & \vdots \\ \frac{\partial^2 \Psi}{\partial \theta_1 \partial \theta_m} & \cdots & \frac{\partial^2 \Psi}{\partial \theta_m^2} \end{bmatrix} \qquad \boldsymbol{Q}(\boldsymbol{\theta}) = \begin{bmatrix} \frac{\partial m(x_1, \boldsymbol{\theta})}{\partial \theta_1} & \cdots & \frac{\partial m(x_1, \boldsymbol{\theta})}{\partial \theta_m} \\ \vdots & \ddots & \vdots \\ \frac{\partial m(x_n, \boldsymbol{\theta})}{\partial \theta_1} & \cdots & \frac{\partial m(x_n, \boldsymbol{\theta})}{\partial \theta_m} \end{bmatrix}$$

Hessian matrix $\qquad \boldsymbol{H} \approx \boldsymbol{Q}^T \boldsymbol{Q} \qquad$ sensitivity matrix

How does measurement uncertainty ϵ propagate into uncertainty about the regressed parameters $\hat{\theta}$?

covariance matrix for $\widehat{\boldsymbol{\theta}}$

$$\boldsymbol{V}_{\widehat{\boldsymbol{\theta}}} \approx \sigma_\epsilon^2 \boldsymbol{H}^{-1} \approx \sigma_\epsilon^2 (\boldsymbol{Q}^T \boldsymbol{Q})^{-1}$$

Fisher information matrix for $\widehat{\boldsymbol{\theta}}$

$$\boldsymbol{M}_{\widehat{\boldsymbol{\theta}}} \approx \boldsymbol{V}_{\widehat{\boldsymbol{\theta}}}^{-1} \approx \frac{1}{\sigma_\epsilon^2} (\boldsymbol{Q}^T \boldsymbol{Q})$$

Bard (1974)
Bates and Watts (1988)
Pirnay, Lopez-Negrete, Biegler (2012)

Extensions not shown: sophisticated error structures, Bayesian or MLE inference, …

Model-Based DoE Optimization Formulation

$$\max_{\boldsymbol{\varphi}} \quad \Psi[\,\boldsymbol{M}(\,\hat{\boldsymbol{\theta}}\,,\boldsymbol{\varphi}\,)\,]$$

$$\text{s.t.} \quad \left.\begin{array}{c} \left.\begin{array}{c} \dot{\boldsymbol{x}}(t) = \boldsymbol{f}(\,\boldsymbol{x}(t), \boldsymbol{z}(t), \boldsymbol{u}(t), \overline{\boldsymbol{w}}, \hat{\boldsymbol{\theta}}\,) \\ \boldsymbol{g}(\,\boldsymbol{x}(t), \boldsymbol{z}(t), \boldsymbol{u}(t), \overline{\boldsymbol{w}}, \hat{\boldsymbol{\theta}}\,) = \boldsymbol{0} \\ \boldsymbol{y}(t) = \boldsymbol{h}(\,\boldsymbol{x}(t), \boldsymbol{z}(t), \hat{\boldsymbol{\theta}}) \end{array}\right\} \text{DAE System} \\ \left.\begin{array}{c} \boldsymbol{f}^{\boldsymbol{0}}(\dot{\boldsymbol{x}}(t_0), \boldsymbol{x}(t_0), \boldsymbol{z}(t_0), \boldsymbol{u}(t_0), \overline{\boldsymbol{w}}, \hat{\boldsymbol{\theta}}) = \boldsymbol{0} \\ \boldsymbol{g}^{\boldsymbol{0}}(\,\boldsymbol{x}(t_0), \boldsymbol{z}(t_0), \boldsymbol{u}(t_0), \overline{\boldsymbol{w}}, \hat{\boldsymbol{\theta}}) = \boldsymbol{0} \\ \boldsymbol{y}^{\boldsymbol{0}}(t_0) = \boldsymbol{h}(\,\boldsymbol{x}(t_0), \boldsymbol{z}(t_0), \hat{\boldsymbol{\theta}}) \end{array}\right\} \begin{array}{l}\text{Initial} \\ \text{Conditions}\end{array} \end{array}\right\} \boldsymbol{m}(\,\boldsymbol{x}(t), \boldsymbol{y}(t), \boldsymbol{z}(t), \boldsymbol{u}(t), \overline{\boldsymbol{w}}, \hat{\boldsymbol{\theta}}\,) = \boldsymbol{0}$$

$\boldsymbol{y}$	Measurements (model responses)
$\hat{\boldsymbol{\theta}}$	Estimated parameters
$\boldsymbol{x}$	Time-dependent differential state variables
$\boldsymbol{z}$	Time-dependent algebraic state variables
$\boldsymbol{u}$	Time-varying control variables
$\overline{\boldsymbol{w}}$	Time-invariant control variable

Fisher information matrix (FIM):

$$\boldsymbol{M} \approx \boldsymbol{V}_{\hat{\boldsymbol{\theta}}}^{-1} \approx \sigma_{\epsilon}^{-2}\boldsymbol{H} \approx \sigma_{\epsilon}^{-2}\boldsymbol{Q}^T\boldsymbol{Q}$$

MBDoE Decisions:

$$\boldsymbol{\varphi} = (\,\boldsymbol{u}(t), \boldsymbol{x}(t_0), \boldsymbol{z}(t_0), \overline{\boldsymbol{w}}, \mathbf{t}\,)$$

Franceschini, G., & Macchietto, S. (2008). Model-based design of experiments for parameter precision: State of the art. *Chemical Engineering Science*, *63*(19), 4846-4872.

Alphabetic Design Criteria Measure Information Content

Figure adapted from: Franceschini, G., & Macchietto, S. (2008). *Chem. Eng. Sci.*, *63*(19), 4846-4872.

A-optimality

max trace(**M**)

enclosing box volume

poor choice for highly correlated **θ**

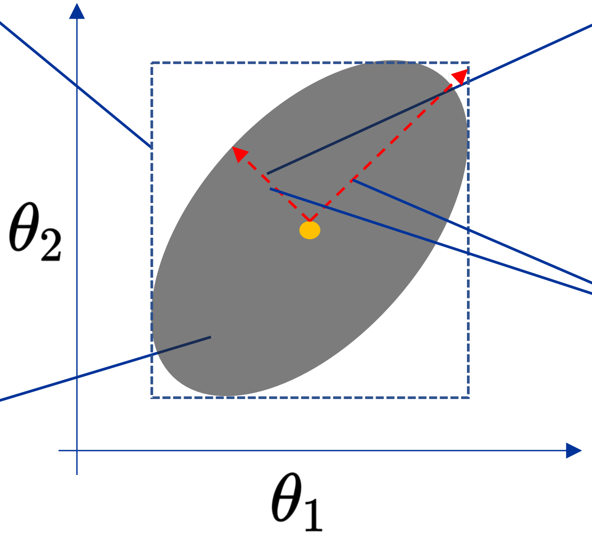
D-optimality

max det(**M**)

ellipsoid volume

robust to linear transformations

confidence ellipsoid for covariance matrix **V** = **M**$^{-1}$



E-optimality

max min(eig(**M**))

major axis

recommended if M is ill-conditioned

ME-optimality

min κ(**M**) = max(eig(**M**)) / min(eig(**M**))

ratio of major to minor axes

recommended if **M** is ill-conditioned

Model Discrimination

Hunter, W.G. and Reiner, A.M., 1965. Designs for discriminating between two rival models. *Technometrics*, *7*(3), pp.307-323.

Buzzi-Ferraris, G. and Forzatti, P., 1983. A new sequential experimental design procedure for discriminating among rival models. *Chemical engineering science*, *38*(2), pp.225-232.

Ferraris, G.B., Forzatti, P., Emig, G. and Hofmann, H., 1984. Sequential experimental design for model discrimination in the case of multiple responses. *Chemical engineering science*, *39*(1), pp.81-85.

Joint Parameter Precision and Model Discrimination

Alberton, A.L., Schwaab, M., Lobão, M.W.N. and Pinto, J.C., 2011. Experimental design for the joint model discrimination and precise parameter estimation through information measures. *Chemical Engineering Science*, *66*(9), pp.1940-1952.

Galvanin, F., Cao, E., Al-Rifai, N., Gavriilidis, A. and Dua, V., 2016. A joint model-based experimental design approach for the identification of kinetic models in continuous flow laboratory reactors. *Computers & Chemical Engineering*, *95*, pp.202-215.

Galvanin, F., Cao, E., Al-Rifai, N., Dua, V. and Gavriilidis, A., 2015. Optimal design of experiments for the identification of kinetic models of methanol oxidation over silver catalyst. *Chimica Oggi-Chemistry Today*, *33*(3), pp.51-56.

Pankajakshan, A., Waldron, C., Quaglio, M., Gavriilidis, A. and Galvanin, F., 2019. A Multi-Objective Optimal Experimental Design Framework for Enhancing the Efficiency of Online Model Identification Platforms. *Engineering*, *5*(6), pp.1049-1059.

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Pyomo.DoE Formulation: MBDoE as 2-Stage Program

max $\quad \log\det\left(\mathbf{M}(\hat{\boldsymbol{\theta}}, \boldsymbol{\varphi})\right) = 2\sum_{i=1}^{N_p} \log L_{ii}$ D-optimality

s.t. $\quad \mathbf{M} = \sum_{r}\sum_{r'} \tilde{\sigma}_{r,r'} \mathbf{Q}_r^{\mathrm{T}} \mathbf{Q}_{r'}$

Stage 1

$\mathbf{M} = \mathbf{L}\mathbf{L}^{\mathrm{T}}, \qquad L_{ii} \geq \epsilon$ Cholesky factorization

$q_{r,p}(t) = \frac{y_{r,p}^{+}(t) - y_{r,p}^{-}(t)}{2\,\epsilon_p}$ Central finite difference

$\boldsymbol{m}\left(\boldsymbol{x}_p^{+}(t), \boldsymbol{y}_p^{+}(t), \boldsymbol{z}_p^{+}(t), \boldsymbol{u}(t), \overline{\boldsymbol{w}}, \boldsymbol{\theta}_p^{+}\right) = \mathbf{0}$

$\boldsymbol{m}\left(\boldsymbol{x}_p^{-}(t), \boldsymbol{y}_p^{-}(t), \boldsymbol{z}_p^{-}(t), \boldsymbol{u}(t), \overline{\boldsymbol{w}}, \boldsymbol{\theta}_p^{-}\right) = \mathbf{0}$

Two model evaluations

$\boldsymbol{\theta}_p^{+} = \hat{\boldsymbol{\theta}} + \boldsymbol{e}_p \epsilon_p$

$\boldsymbol{\theta}_p^{-} = \hat{\boldsymbol{\theta}} - \boldsymbol{e}_p \epsilon_p$

Up and down perturbations

Stage 2

$\forall p \in \{1, \dots, N_p\}$

Model Sensitivity

$$\mathbf{Q}_r = \begin{bmatrix} \frac{\partial y_r(t_1)}{\partial \theta_1} & \cdots & \frac{\partial y_r(t_1)}{\partial \theta_{N_p}} \\ \vdots & \ddots & \vdots \\ \frac{\partial y_r(t_n)}{\partial \theta_1} & \cdots & \frac{\partial y_r(t_n)}{\partial \theta_{N_p}} \end{bmatrix} = \begin{bmatrix} \mathbf{q}_{r,1} & \cdots & \mathbf{q}_{r,N_p} \end{bmatrix}$$

$$\mathbf{q}_{r,p} = \begin{bmatrix} \frac{\partial y_r(t_1)}{\partial \theta_p} & \cdots & \frac{\partial y_r(t_n)}{\partial \theta_p} \end{bmatrix}^{\mathrm{T}}$$

$\boldsymbol{y}$	Measurements (model responses)
$\mathbf{Q_r}$	Dynamic sensitivity for response *r*
$\boldsymbol{m}(\,)$	DAE model
$\hat{\boldsymbol{\theta}} \in \mathbb{R}^P$	Estimate for parameters
$\mathbf{M} \in \mathbb{R}^{P \times P}$	Fisher information matrix
$\mathbf{L} \in \mathbb{R}^{P \times P}$	Lower triangular Cholesky factorization
ϵ_p	Small perturbation for parameter *p*
$\boldsymbol{e}_p \in \mathbb{R}^P$	Unit vector with “1” in position *p*

Wang and Dowling, 2022. *AIChE J.*

Pyomo.DoE Extends `parmest` Interface

`create_model`

Create Pyomo model for DAE compatible with `parmest`

`DesignVariables`

Specify MBDoE degrees of freedom with bounds

`MeasurementVariables`

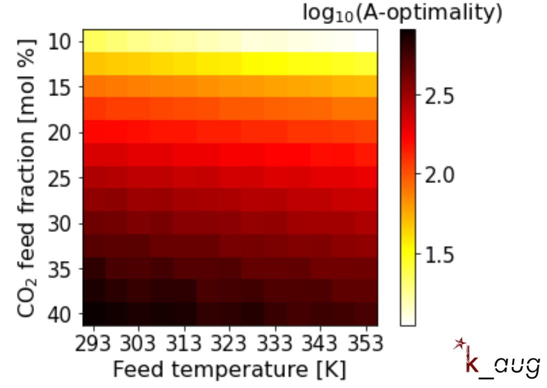
Specify MBDoE measurement variables and observation error covariance

`DesignOfExperiment`

PYOMO DoE

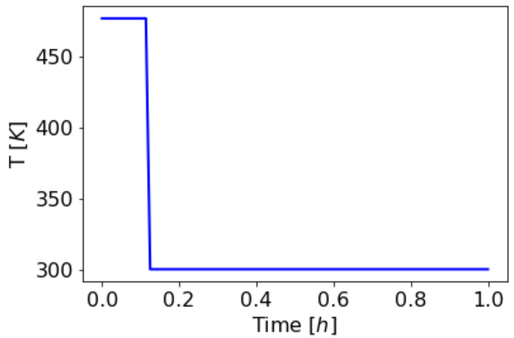
`compute_FIM`

Fast exploratory analysis



`stochastic_program`

Dynamic optimization



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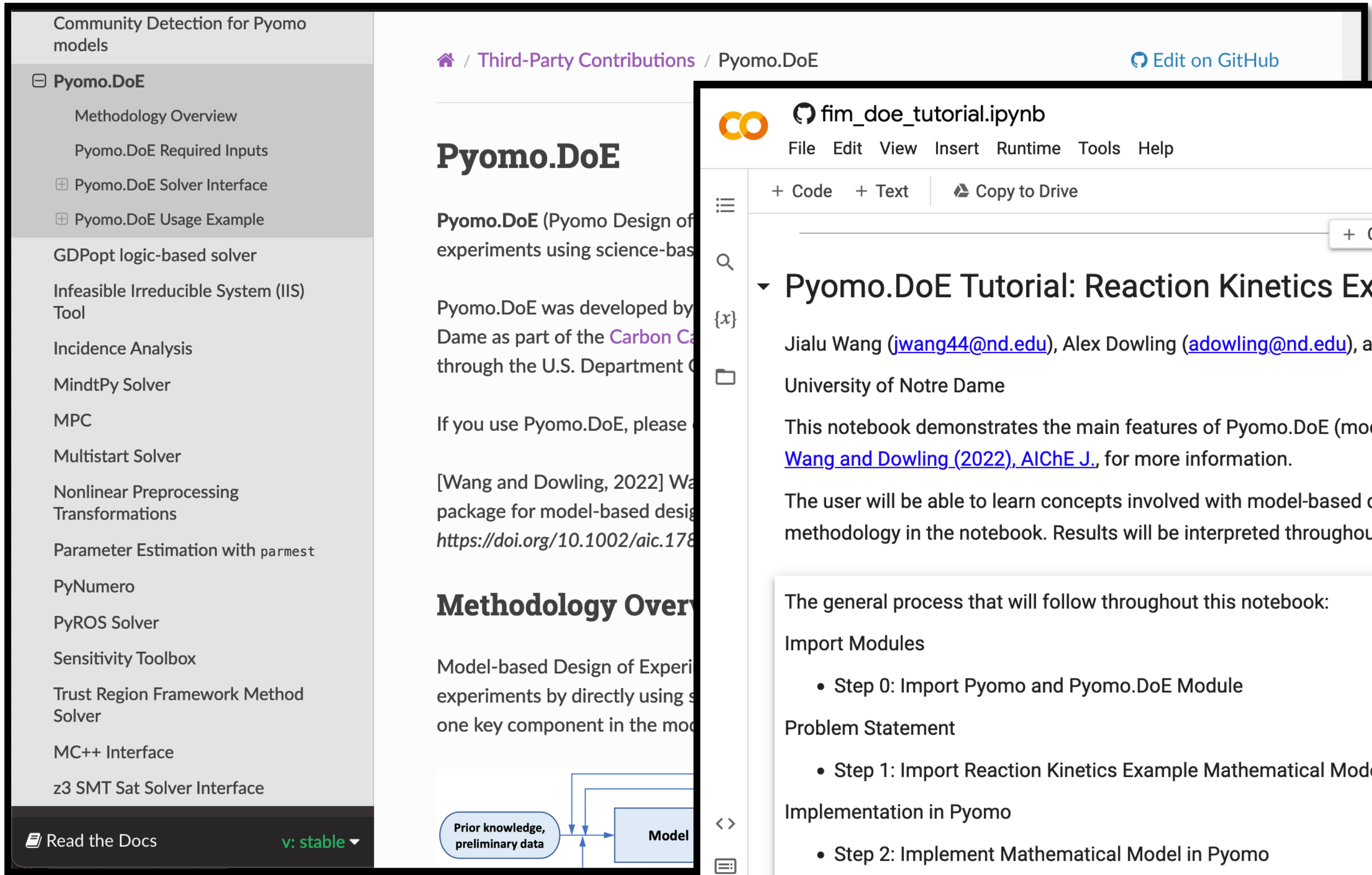
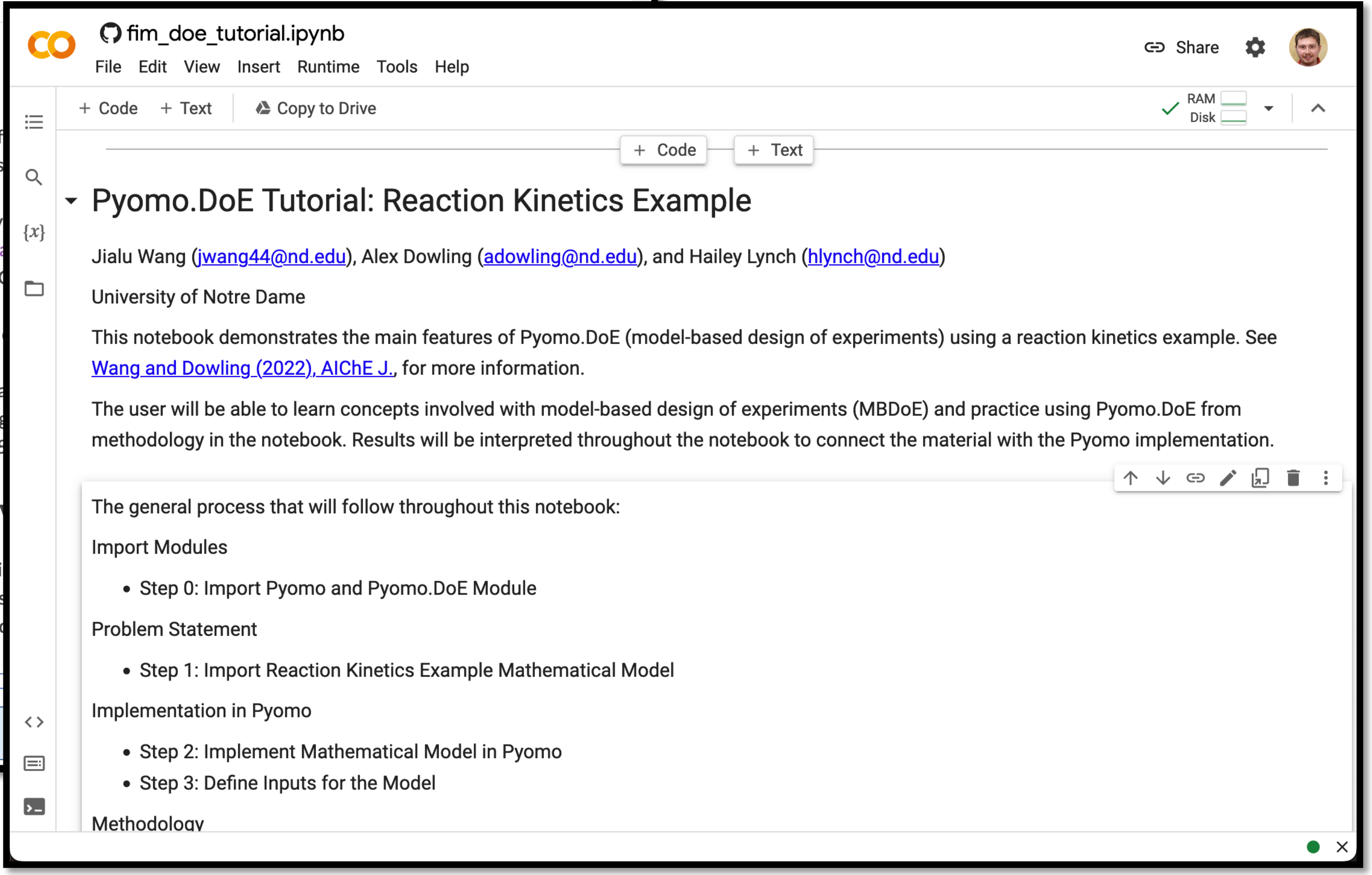
Wang and Dowling, 2022. Pyomo.DoE: An open-source package for model-based design of experiments in Python. *AIChE J. 68(12),* e17813.

Getting Started with Pyomo.DoE

Documentation: https://pyomo.readthedocs.io/en/stable/contributed_packages/doe/doe.html
Tutorial: https://colab.research.google.com/github/Pyomo/pyomo/blob/main/pyomo/contrib/doe/examples/fim_doe_tutorial.ipynb





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ParmEst and Pyomo.DoE Development Plans

Coming soon:

- New modeling abstraction and interface
- Improved initialization
- Improved optimization performance
- More applications and examples

PYOMO DoE

This tutorial (https://dowlinglab.github.io/pyomo-doe/) will be updated in Fall 2024 to reflect these major enhancements in ParmEst and Pyomo.DoE.