Determination of Intrinsic Kinetic Models and Parameters for Product Quality Assurance

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Motivation

- Consumer products may be subject to prolonged storage and transportation times
- Ensuring Product Quality and Stability is a critical and time-consuming activity
  - Shelf-lives typically measured in years
  - Need to understand impact of formula design on product performance and stability
- Undesirable chemical reactions may lead to product degradation
- Develop a suite of modeling tools to support the development of new consumer products:
  - Simulate product chemistry
  - Design efficient experimental campaigns
  - Discriminate among alternative models and estimate model parameters
  - Understand the uncertainty associated with model predictions

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Ostace EWO Project: ***Heat transfer model of large shipping containers***

Problem Statement

- Given a set of alternative kinetic models and an experimental design space:
  - Generate an initial set of experiments
  - Perform sequential Design of Experiments for Model Discrimination based on posterior probability share
  - Determine Global Sensitivity of Model Parameters

$$S_i = \frac{V\left(E(Y|\theta_i)\right)}{V(Y)} \qquad S_{Ti} = 1 - \frac{V\left(E(Y|\boldsymbol{\theta}_{\sim i})\right)}{V(Y)} \qquad S_{Ti} \geq S_i \qquad \left\{S^{\sigma}_{\theta_i} = \frac{\sigma_{\theta_i}}{\sigma_Y}\frac{\partial Y}{\partial \theta_i}\right\}$$

  - Perform sequential Design of Experiments to improve Parameter Estimates guided by sensitivities
  - Perform Uncertainty Quantification

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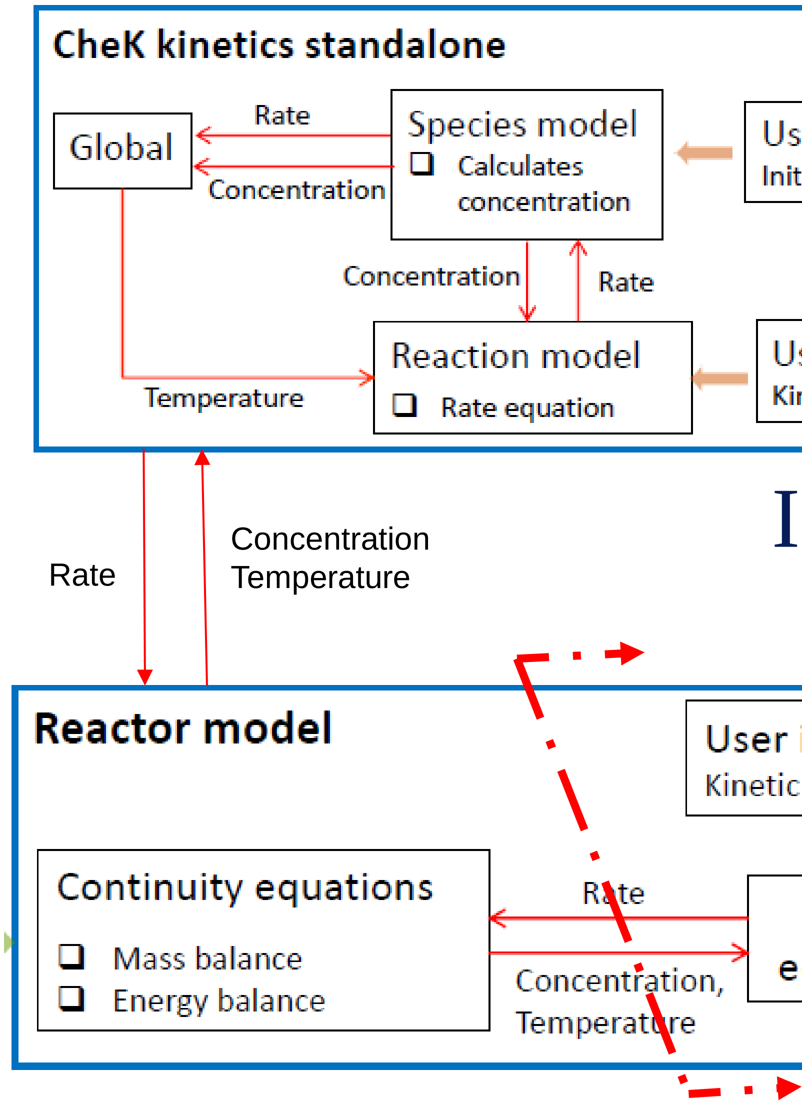
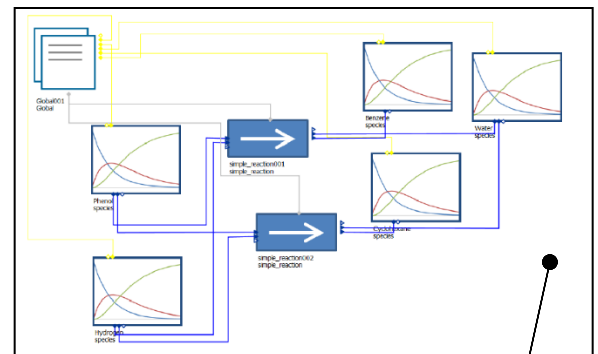
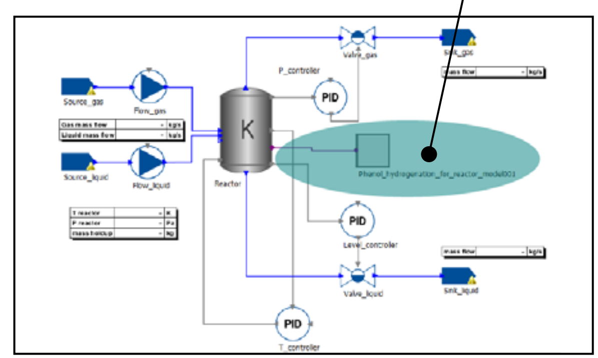
Leverage *gPROMS* Modeling Environment with CheK Library to Input Kinetic Models

Cano and Goda (2014)



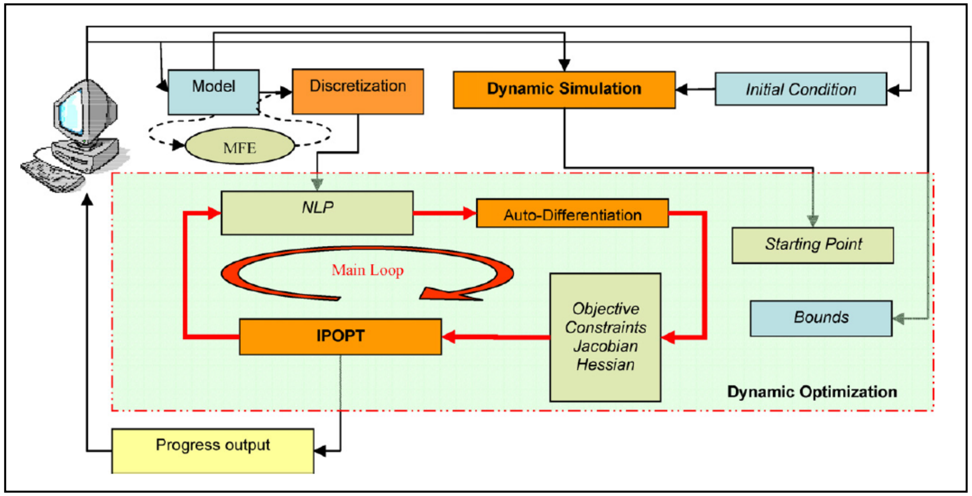


Integrate with Detailed Reactor Models

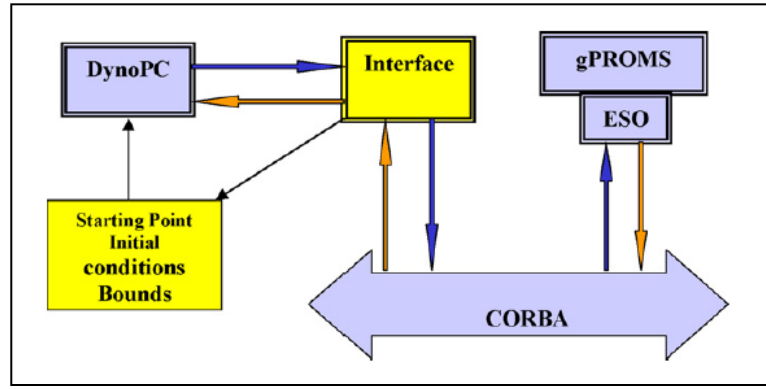


Leverage Prior Interface Work Between *gPROMS* and *DynOpt* (Collocation + IPOPT)

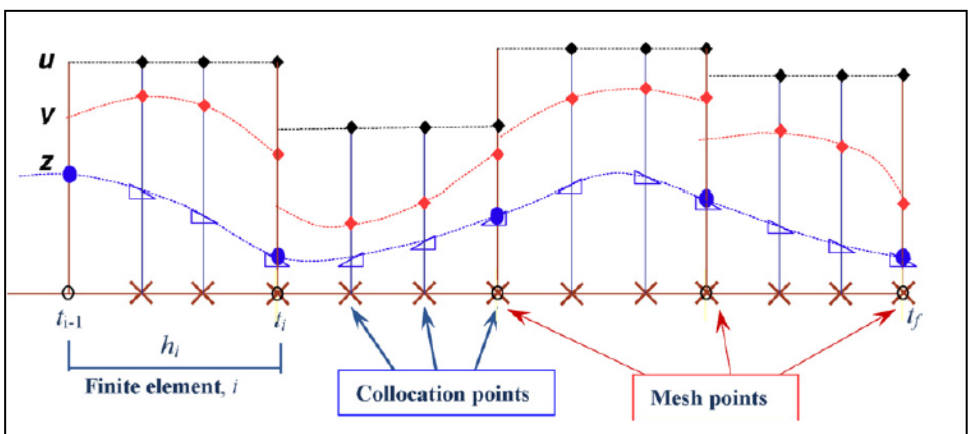
Lang and Biegler (2005, 2007)



Convert DAE to NLP Using Collocation on Finite Elements



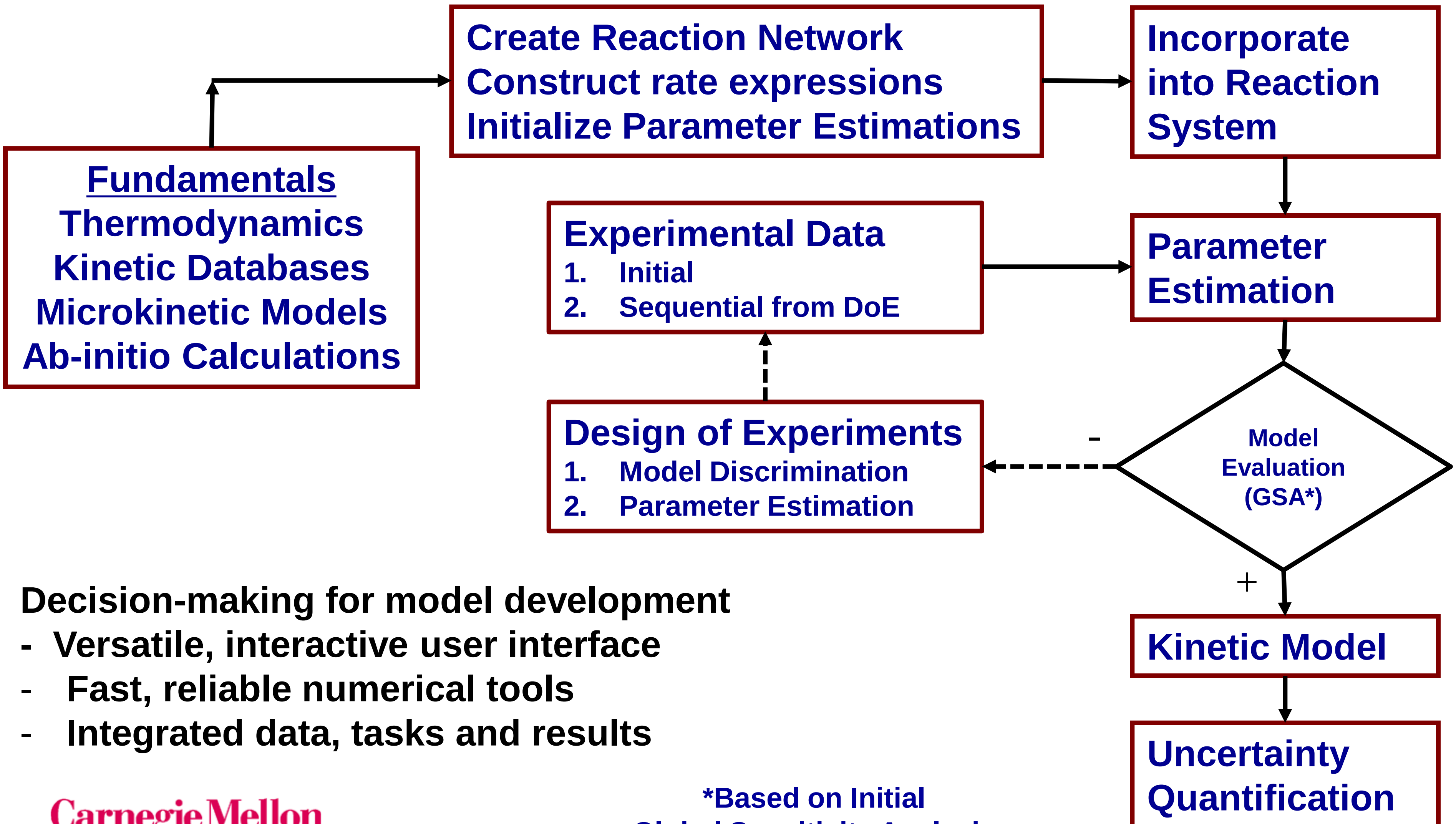
ESO provides Function Evaluations, Jacobian and Residuals at Collocation Points. DynOpt returns Values of the Optimization Variables.



- ESO functionality not currently accessible in *gPROMS*
- Project will provide Proof-of-Concept Solution using PyOMO
- Long-term goal remains to leverage CheK with IPOPT capabilities

Proposed Solution Method

Adapted from www.Eurokin.org



Decision-making for model development

- Versatile, interactive user interface
- Fast, reliable numerical tools
- Integrated data, tasks and results

*Based on Initial Global Sensitivity Analysis

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Model Discrimination Methodology

Schwaab *et al.* (2008)

$$\underset{\mathbf{x}=\mathbf{x}_{N+1}}{\text{maximize}}\,\widehat{D}$$

$$\widehat{D} = \max_{m,n\neq m} \widehat{D}_{m,n}(\mathbf{x}_{N+1})$$

$$\widehat{D}_{m,n}(x_{N+1}) = (P_m P_n)^Z[\mathbf{y}_m(\mathbf{x}_{N+1}) - \mathbf{y}_n(\mathbf{x}_{N+1})]^T \widehat{\mathbf{V}}_{m,n}^{-1}(\mathbf{x}_{N+1})[\mathbf{y}_m(\mathbf{x}_{N+1}) - \mathbf{y}_n(\mathbf{x}_{N+1})]$$

$$0 \leq Z \leq 2\ (use\ Z = 1)$$

$$P_m = \frac{\phi_m}{\sum_{n=1}^{M}\phi_n}$$

$$\phi_m = 1 - p\left[\chi^2_{v_m} \leq F_m\right]$$

$$v_m = NE - NP_m$$

$$F_m = \underset{\boldsymbol{\theta}_m}{\text{minimize}} \sum_{i=1}^{N} \left(\mathbf{y}_{i,m} - \mathbf{y}_i^{exp}\right)^T \mathbf{V}_i^{-1}\left(\mathbf{y}_{i,m} - \mathbf{y}_i^{exp}\right)$$

$$\widehat{\mathbf{V}}_{m,n}(\mathbf{x}_{N+1}) = 2\mathbf{V}(\mathbf{x}_{N+1}) + \widehat{\mathbf{V}}_m(\mathbf{x}_{N+1}) + \widehat{\mathbf{V}}_n(\mathbf{x}_{N+1})$$

$$\widehat{\mathbf{V}}_m(\mathbf{x}_{N+1}) = \mathbf{B}_m(\mathbf{x}_{N+1})\widehat{\mathbf{V}}_{\theta,m}(\mathbf{x}_{N+1})\mathbf{B}_m^T(\mathbf{x}_{N+1})$$

$$\mathbf{B}_m(\mathbf{x}_{N+1}) = \frac{\partial \mathbf{y}_m(\mathbf{x}_{N+1})}{\partial \boldsymbol{\theta}_m}$$

$$\widehat{\mathbf{V}}_{\theta,m}(\mathbf{x}_{N+1}) = \left[\mathbf{B}_m^T(\mathbf{x}_{N+1})\mathbf{V}^{-1}(\mathbf{x}_{N+1})\mathbf{B}_m(\mathbf{x}_{N+1}) + \widehat{\mathbf{V}}_{\theta,m}^{-1}\right]^{-1}$$

$$\widehat{\mathbf{V}}_{\theta,m}^{-1} = \left[\sum_{i=1}^{N} \mathbf{B}_m^T(\mathbf{x}_i)\mathbf{V}^{-1}(\mathbf{x}_i)\mathbf{B}_m(\mathbf{x}_i)\right]^{-1}$$

Key contributions of the method:

1. **Probability weighting to focus on discriminating between the two best models**
2. **Calculating the posterior covariance matrix of model parameters at x_{N+1}**
3. **(2) is used to calculate the posterior covariance matrix of predictions at x_{N+1}**
4. **Discriminant is based on large posterior prediction differences and/or small variances**
5. **No further discrimination possible if, for Z=0, $D_{m,n}(x_{N+1})$ < # of responses**

Unique Project Aspects

- Optimization with s-IPOPT using Analytic Hessian in PyOMO
  - Determine parameter sensitivity to noise in the data
  - Identify problematic 2nd-order conditions:
    - Zero curvature: unbounded confidence region
    - Indefinite curvature: saddle point (rare)
  - Create ability to compare Analytic Hessian results with:
    - Gauss-Newton: ~*gPROMS* ESO with IPOPT
    - BFGS: ~Internal *gPROMS* Hessian for DoE/PE
- Orthogonal Collocation on Finite Elements ⇒ DAE to NLP
  - Restricts unstable nodes within finite element (**Robust**)
    - Sequential simulation/optimization may fail if IVP open loop unstable
  - Efficient computation using simultaneous approach (**Fast**)
    - Avoids overhead of sensitivity calculations from DAE solver
  - Avoids convergence failure due to large gradient errors (**Accurate**)
    - DAE solver's internal convergence loops → "convergence noise"

Novelty/Significance of Work

- Leverages contributions from several fields:
  - Math Programming-based Bayesian methodology (e.g. Warren Stewart, G.E.P Box) for Design of Experiments for
    - Model Discrimination
    - Parameter Estimation
  - Incorporation of "Robust" (i.e. distribution-free) alternative methods
    - May be particularly advantageous for small number of experiments
  - Global Sensitivity Analysis (Stochastic Simulation)
    - Accounts for both Numerical Sensitivity and Parameter Uncertainty
    - Provides relative ranking of importance of each parameter
  - Uncertainty Quantification (Stochastic Simulation)
    - Propagation of Parameter Uncertainty
    - Quantification of model discrepancies

Potential Impact for Industrial Applications

- Biegler group has solved large and/or complex parameter estimation problems using IPOPT
  - Zavala and Biegler (2006) 54 parameters for homo- and copolymer reactions included Error-in-Variables-Measured (EVM)
  - Zavala, Laird and Biegler (2008) 57 parameters for LDPE tubular reactor, including EVM. Solved in parallel using a Schur complement decomposition approach
  - Lin, Biegler, Jacobsen (2010) 15 parameters to predict particle growth dynamics, polymerization rate and particle average molecular weight for a seeded suspension polymerization process
- Working with Python-based PyOMO removes limitation on available *gPROMS* licenses to perform parallel simulations for Global Sensitivity Analysis and Uncertainty Quantification
- Bock's group at University of Heidelberg has had success with Robust methods working with BASF