Bayesian parameter inference for PICA devolatilization pyrolysis at high heating rates

Joffrey Coheur

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Motivation: atmospheric entry

Thermal protection systems (TPS)

Dragon capsule (Space X)

Space debris

ATV-1 Jules Verne (ESA)
Ablative materials for thermal protection systems

- PICA: Phenolic-Impregnated Carbon Ablator

Mars Science Laboratory thermal protection system (NASA)

Porous thermal protection material

Fibers + resin

[Helber, 2016]

Lawson et al.

Bottom view (after burn)
Modeling the pyrolysis of ablative thermal protection materials

CFD codes require accurate models for ablation [Lachaud, 2014; Schrooyen, 2016], e.g. conservation of mass species

\[ \frac{∂\rho_i}{∂t} + \nabla \cdot (\rho_i u_g) = \nabla \cdot (J_i) + ω_{pyro} \]

Objectives: deduce \( ω_{pyro} \) from dedicated pyrolysis experiments

Methodology: Bayesian inference for a robust characterization with uncertainties
**Table of Contents**

Introduction

Characterization of physico-chemical parameters relevant to pyrolysis reactions
- Experiments
- Modeling

Challenges
- Non-linear posterior and highly-correlated parameters
- Zero-variance data in the dataset

Application to complex pyrolysis model

Conclusions
Table of Contents

Introduction

Characterization of physico-chemical parameters relevant to pyrolysis reactions
  • Experiments
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Challenges
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Conclusions
Pyrolysis experiments [Wong et al., 2015; Bessire and Minton, 2017]

Phenolic sample (50 mg)

Pyrolysis gases

Heated wall

Temperature, T

Mass yields, mg

Sample mass, mg

Temperature, K

H₂, CO, CO₂, CH₄, H₂O
Table of Contents

Introduction

Characterization of physico-chemical parameters relevant to pyrolysis reactions
  • Experiments
  • Modeling

Challenges
  • Non-linear posterior and highly-correlated parameters
  • Zero-variance data in the dataset

Application to complex pyrolysis model

Conclusions
Phenomenological laws for pyrolysis mass loss and gas species production

Pyrolysis decomposition of ablative materials follows successive reaction rates [Goldstein, 1969; Trick, 1997]

\[ \langle \dot{\omega}_i^{\text{pyro}} \rangle = \sum_{j}^{N_p} F_{ij} \frac{\partial \xi_j}{\partial T} \tau m_0 \]

\[ \frac{\partial \xi_j}{\partial T} = (1 - \xi_j)^{n_j} \frac{A_j}{\tau} \exp \left( -\frac{E_j}{R T} \right) \]

\[ \xi_j: \text{ advancement of reaction of the fictitious resin component } j \]
Phenomenological laws for pyrolysis mass loss and gas species production

- Pyrolysis decomposition of ablative materials follows successive reaction rates [Goldstein, 1969; Trick, 1997]

\[ m = m_0 - \sum_{j} F_j \xi_j m_0 \]

\[ \frac{\partial \xi_j}{\partial T} = (1 - \xi_j)^{n_j} \frac{A_j}{\tau} \exp \left( - \frac{E_j}{RT} \right) \]

- \( \xi_j \): advancement of reaction of the fictitious resin component \( j \)
Bayesian inference for parameter calibration

Objective: Infer on a finite set of parameters $\mathbf{p} \in \mathbb{R}^q$ from

▶ A set of measurements $\mathbf{d}^{\text{obs}} = \{d_i \in \mathbb{R}, i = 1, \ldots, n_{\text{obs}}\}$
▶ A model that predicts the measurements

using Bayesian rule for improving our knowledge on $\mathbf{p}$

\[
\pi(\mathbf{p}|\mathbf{d}^{\text{obs}}) = \frac{\pi(\mathbf{d}^{\text{obs}}|\mathbf{p})\pi_0(\mathbf{p})}{\int_{\mathbb{R}^p} \pi(\mathbf{d}^{\text{obs}}|\mathbf{p})\pi_0(\mathbf{p})d\mathbf{p}} \quad \text{(Bayes’ theorem)}
\]

▶ Choice for the prior $\pi_0(\mathbf{p})$: uniform pdf (bounded support)
▶ Choice for the likelihood $\pi(\mathbf{d}^{\text{obs}}|\mathbf{p})$: additive Gaussian noise

\[
\pi(\mathbf{d}^{\text{obs}}|\mathbf{p}) = \frac{1}{\prod_i (2\pi \sigma_i^2)^{n/2}} \exp \left( -\sum_{i=1}^{n_{\text{s}}} \sum_{k=1}^{n} \frac{(d_{ik}^{\text{obs}} - \eta_i(\mathbf{x}_k, \mathbf{p}))^2}{2\sigma_i^2} \right)
\]
Benchmark: two-equation model

Synthetic data using:

\[ \sigma_I = 0.149 \text{ s}^{-1} \]
\[ \sigma_{II} = 0.5382 \text{ s}^{-1} \]

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<th>E</th>
<th>m</th>
<th>F</th>
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<td>103680</td>
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Random-Walk Metropolis-Hastings: bivariate posterior PDFs
Challenges for the inference of pyrolysis kinetic parameters

- Non-linear posterior and highly-correlated parameters: random-walk Metropolis-Hastings may fail or be very slow, tuning of the proposal covariance hard for high dimensional problems
  
  * Reparametrization of parameters space adapted to the physical model
  
  * Gradient-based algorithms (Itô-SDE)

- Fitting unimportant data may biased the results
  
  * Hyperparameter choice
  
  * Data feature selection
# Table of Contents

## Introduction

## Characterization of physico-chemical parameters relevant to pyrolysis reactions
- Experiments
- Modeling

## Challenges
- Non-linear posterior and highly-correlated parameters
- Zero-variance data in the dataset

## Application to complex pyrolysis model

## Conclusions
Reparametrization of parameters space adapted to the physical model

- More complex models become prohibitive in terms of number of iterations required
- Need to improve the mixing of the Markov chains

Non-linear transformation of the parameter space

\[
\begin{align*}
\tilde{A}_i &= \ln A_i - E_i/(R \bar{T}_i), \\
\tilde{E}_i &= E_i/E_i, \\
\tilde{m}_i &= m_i/m_i, \\
\tilde{F}_i &= F_{ij}/\bar{F}_{ij}.
\end{align*}
\]

Reaction rate:

\[
k_i = \exp \left(\tilde{A}_i + \tilde{E}_i \tilde{T}_i(t)\right)
\]

Local reciprocal temperature:

\[
\tilde{T}_i = \bar{E}_i/R(1/T + 1/\bar{T}_i)
\]
Benchmark: two-equation model

Synthetic data using:

\[ \sigma_I = 0.149 \text{ s}^{-1} \]
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Bivariate posterior PDFs (initial parameter space)
Bivariate posterior PDFs (rescaled parameter space)
We use the Itô Stochastic Differential Equation (ISDE) introduced by Soize (2008)

\[
d\Xi = [\hat{C}]H dt \\
dH = -\nabla_\Xi \phi(\Xi) dt - \frac{1}{2} \zeta_0 H dt + \sqrt{\zeta_0} [L_{\hat{C}}]^{-T} dW
\]

with \( \phi = -\log(\pi(d^{obs}|p)) \), \([\hat{C}]\): approximation of the covariance matrix, \( \zeta_0 \): free parameter (Arnst, Soize (2019)). Discretization in time using a Stormer-Verlet method (Soize, Ghanem (2016))

\[
\Xi(\ell+\frac{1}{2}) = \Xi(\ell) + \frac{\Delta t}{2} [\hat{C}]H^{(\ell)} \\
H^{(\ell+1)} = \frac{1-b}{1+b} H^{(\ell)} - \frac{\Delta t}{1+b} \nabla_\Xi \phi(\Xi(\ell+\frac{1}{2})) + \frac{\sqrt{\zeta_0}}{1+b} [L_{\hat{C}}]^{-T} \Delta W^{(\ell+1)} \\
\Xi(\ell+1) = \Xi(\ell+\frac{1}{2}) + \frac{\Delta t}{2} [\hat{C}]H^{(\ell+1)}
\]
Table of Contents

Introduction

Characterization of physico-chemical parameters relevant to pyrolysis reactions
  • Experiments
  • Modeling

Challenges
  • Non-linear posterior and highly-correlated parameters
  • Zero-variance data in the dataset

Application to complex pyrolysis model

Conclusions
Zero variance data in the data-set

- Hyperparameter choice

\[ \pi(d_{\text{obs}} | p) = \frac{1}{\prod_i (2\pi \sigma_i^2)^{n/2}} \exp \left( -\sum_{i=1}^{n_s} \sum_{k=1}^{n} \frac{(d_{ik}^{\text{obs}} - \eta_i(x_k, p))^2}{2\sigma_i^2} \right) \]

- Taking all the data might lead to inaccurate results, although zero variance data (before activation temperature or after reaction) are obtained experimentally

- Feedback on the experiments: fit only important features (activation temperature, maximum production peaks)
Illustration using experimental data: two-reaction model with one species

- Observable (data) $d_{H_2k}^{obs}$: mass yields of species $H_2$ at each temperature. $N_p = 2$.

\[ d_{H_2k} = \sum_j^{N_p} F_{H_2j} m_0 \left( \xi_j^{(k)} - \xi_j^{(k-1)} \right), \]

\[ \frac{\partial \xi_j^{(k)}}{\partial t} = \left( 1 - \xi_j^{(k)} \right)^n_j A_j \exp \left( -\frac{E_j}{RT_k} \right), \]

\[ \xi_j^{(0)} = 0. \]

- Simple point-mass model (0D), $k = 1, \ldots, n_T$

- $p = \{ A_1, E_1, n_1, F_{H_2,1}, A_2, E_2, n_2, F_{H_2,2} \}$

- Initial guess obtained from genetic algorithm
Comparison of the different approaches

- Comparison of posterior samples obtained from the Markov chains (length = 1e4) using Itô-SDE (blue dots) and random-walk Metropolis-Hastings in the reparametrized parameter space (orange dots) and in the initial parameter space (green dots)

![Graph showing comparison of different approaches]
Table of Contents

Introduction

Characterization of physico-chemical parameters relevant to pyrolysis reactions
• Experiments
• Modeling

Challenges
• Non-linear posterior and highly-correlated parameters
• Zero-variance data in the dataset

Application to complex pyrolysis model

Conclusions
Proposed five-equation model (38 unknown parameters):

\[ R_1(s) \xrightarrow{k_1} F_{1,1}H_2O + F_{1,2}CO_2 + F_{1,3}(1\text{-propanol}) \]

\[ R_2(s) \xrightarrow{k_2} F_{2,1}H_2O + F_{2,2}CO + F_{2,3}CO_2 + F_{2,4}\text{Phenol} + F_{2,5}\text{Cresol} + F_{2,6}(\text{Dimethyl Phenol}) + F_{2,7}(\text{Trimethyl Phenol}) \]

\[ R_3(s) \xrightarrow{k_3} F_{3,1}\text{CH}_4 + F_{3,2}H_2O + F_{3,3}CO + F_{3,4}H_2 + F_{3,5}CO_2 + F_{3,6}\text{Xylene} + F_{3,7}\text{Benzene} + F_{3,8}\text{Toluene} \]

\[ R_4(s) \xrightarrow{k_4} F_{4,1}H_2 \]

\[ R_5(s) \xrightarrow{k_5} F_{5,1}H_2 + F_{5,2}\text{CH}_4 + F_{5,3}H_2O + F_{5,4}CO \]
Propagation results: production rate curves

Temperature, K

Production rate, $s^{-1}$

$H_2O$

$H_2$

$CH_4$

$CO$

$3 \cdot 10^{-6}$

$6 \cdot 10^{-6}$
Propagation results: production rate curves

Temperature, K

Production rate, s\(^{-1}\)

- Phenol
- Dimethyl phenol
- Trimethyl phenol

CO\(_2\)
Cresol

Production rate, s\(^{-1}\)

0
0.1
0.2
0.3
0.4

4 \times 10^{-7}

Propagation results: production rate curves

Temperature, K

Production rate, s$^{-1}$

- 1-propanol
- Benzene

- Xylene
- Toluene
Table of Contents

Introduction

Characterization of physico-chemical parameters relevant to pyrolysis reactions
  • Experiments
  • Modeling

Challenges
  • Non-linear posterior and highly-correlated parameters
  • Zero-variance data in the dataset

Application to complex pyrolysis model

Conclusions
Conclusions

- Application of a simple model to simulate pyrolysis experiments.
- Inference on the parameters of pyrolysis reactions using Bayesian approach.
- Efficient method to infer on kinetic parameters and characterize their uncertainties. However, lack of identifiability for $A$ and $E$.
- Rescaling the parameter space to their posterior distribution based on model features reduce significantly the tuning of the proposal covariance
- Ito-SDE method enables no proposal tuning and samples efficiently the posterior distribution, but requires the computation of gradients
- Simulations of gas production rate and mass loss with error bars lead to good agreement with experimental curves.
Future work

- Apply Itô-SDE method to the full set of experimental data

- Efficient computation of model gradients, e.g. adjoint-based

- Use more general models, e.g. include competitive reaction schemes (F. Torres, J. Blondeau), include more data (different heating rates) → model inference

- Results interpretation and their used in CFD codes; uncertainty propagation for the validation of CFD codes
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Additional Information
Table of Contents

Bibliography

Mathematical model
  • Model Selection
Bibliography

Simple model for simulating pyrolysis experiments

For point-mass material, conduction is assumed to be instantaneous and \( T \) is uniform inside the material (lumped capacitance model, \( Bi = \frac{Lc h}{k} << 1 \)).

\[ T = T_i = \text{cte in } \Delta t = [0, t_f] \]

\[ \xi(t) = 1 - \left[ (1 - n)(C_i - A \exp \left( -\frac{E}{R T_i} t \right)) \right]^{1/(n-1)}, \quad \text{with } C_i = \frac{(1 - \xi(0))^{1-n}}{1-n} \]

\[ T_{\text{out}} = \tau t, \quad \tau \text{ heating rate} \]

\[ \xi(T) = 1 - \left\{ (1 - n) \left[ -\frac{A}{\tau} T \exp \left( -\frac{E}{R T} \right) - \frac{A E}{\tau R} \text{Ei} \left( -\frac{E}{R T} \right) + C \right] \right\}^{\frac{1}{1-n}} \]

\[ C = \frac{(1 - \xi_0)^{1-n}}{1-n} + \frac{A}{\tau} T_0 \exp \left( -\frac{E}{R T_0} \right) + \text{Ei} \left( -\frac{E}{R T_0} \right) \frac{EA}{\tau}, \quad \text{where } \text{Ei}(x) \equiv \int_{-\infty}^{-x} \exp(v) \frac{dv}{v} \]

For more complex temperature evolution, integration should be performed numerically. When \( Bi > 1 \) (\( L_c, h \nearrow \), or \( k \searrow \)) more complex model should be used (Argo).
Numerical set-up: 3 reactants model with 5 species

Observable (data) $d_{ik}^{\text{obs}}$ with $i = \{\text{H}_2, \text{CO, CO}_2, \text{CH}_4, \text{H}_2\text{O}\}$: mass yields at each temperature iteration $k$. $N_p = 3$.

$$d_{ik} = \sum_j N_p F_{ij} m_0 \left( \xi_j^{(k)} - \xi_j^{(k-1)} \right),$$

$$\frac{\partial \xi_j^{(k)}}{\partial t} = \left(1 - \xi_j^{(k)}\right)^{n_j} A_j \exp \left(- \frac{E_j}{RT_k}\right),$$

$$\xi_j^{(0)} = 0.$$

Simple point-mass model (0D), $k = 1, \ldots, n_T$

$$p = \{ \{A_3, E_3, n_3, F_{\text{H}_2\text{O},3}\}, \{A_2, E_2, n_2, F_{\text{CO},2}, F_{\text{CH}_4,2}, F_{\text{H}_2\text{O},2}\}, \{A_1, E_1, n_1, F_{\text{CO},1}, F_{\text{CO}_2,1}, F_{\text{H}_2,1}\}\}$$

$R_1 \rightarrow \text{H}_2\text{O}$

$R_2 \rightarrow \text{CO} + \text{CH}_4 + \text{H}_2\text{O}$

$R_3 \rightarrow \text{CO} + \text{CO}_2 + \text{H}_2$
Posterior predictive checks: 3 reactants model with 5 species
Posterior predictive checks: 3 equations, 5 species
How to select an appropriate model?

- Principle of Parsimony (Occam’s razor): “Shave away all that is unnecessary”
  
  \[ \text{Accuracy} \gg \text{complexity} \]

- Kullback-Leibler information

  \[ I(f, g) = \int f(x) \log \left( \frac{f(x)}{g(x|\theta)} \right) \, dx \]

- Information criteria

  \[ \text{AIC} = -2 \log(\mathcal{L}(y|\hat{\theta})) + 2K \]
  \[ \text{BIC} = -2 \log(\mathcal{L}(y|\hat{\theta})) + K \log n \]
Illustration with the two-equation benchmark

- Approximate model: one-equation model
Illustration with the two-equation benchmark

- Approximate model: two-equation model
Illustration with the two-equation benchmark

- Approximate model: three-equation model

![Graph showing production rate vs. temperature for different models](image-url)
### Illustration with the two-equation benchmark

#### Computation of information criteria:

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<th>$N_{\text{samples}}$</th>
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