Bayesian parameter inference for PICA devolatilization pyrolysis at high heating rates

Joffrey Coheur

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Motivation: atmospheric entry

Thermal protection systems (TPS)

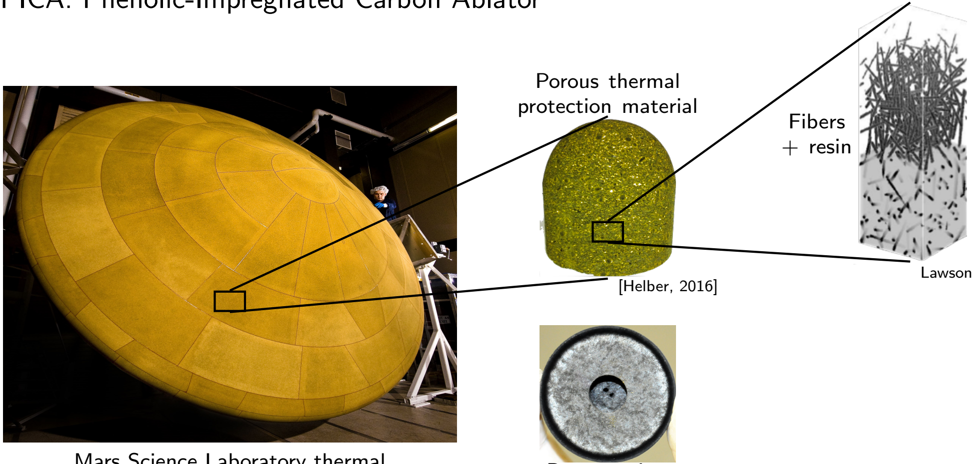
Dragon capsule (Space X)

Space debris

ATV-1 *Jules Verne* (ESA)

Ablative materials for thermal protection systems

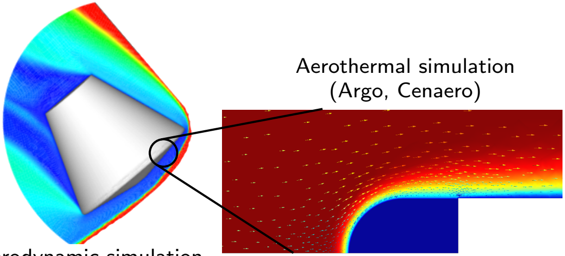
- PICA: Phenolic-Impregnated Carbon Ablator



Mars Science Laboratory thermal protection system (NASA)

Bottom view (after burn)

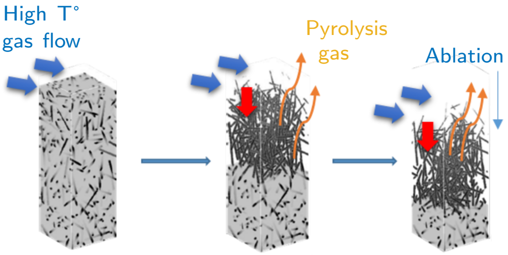
Modeling the pyrolysis of ablative thermal protection materials



Aerothermal simulation
(Argo, Cenaero)

Aerodynamic simulation
(CooLFluiD, VKI)

[Coheur, Schrooyen, 2017]



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

$$\frac{\partial \epsilon_g \langle \rho_i \rangle_g}{\partial t} + \nabla \cdot (\epsilon_g \langle \rho_i \rangle_g \langle u \rangle_g) = \nabla \cdot \langle J_i \rangle + \langle \dot{\omega}_i^{\text{pyro}} \rangle$$

- Objectives: deduce $\langle \dot{\omega}_i^{\text{pyro}} \rangle$ from dedicated pyrolysis experiments
- Methodology: Bayesian inference for a robust characterization with uncertainties

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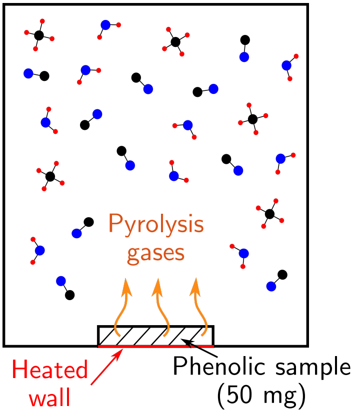
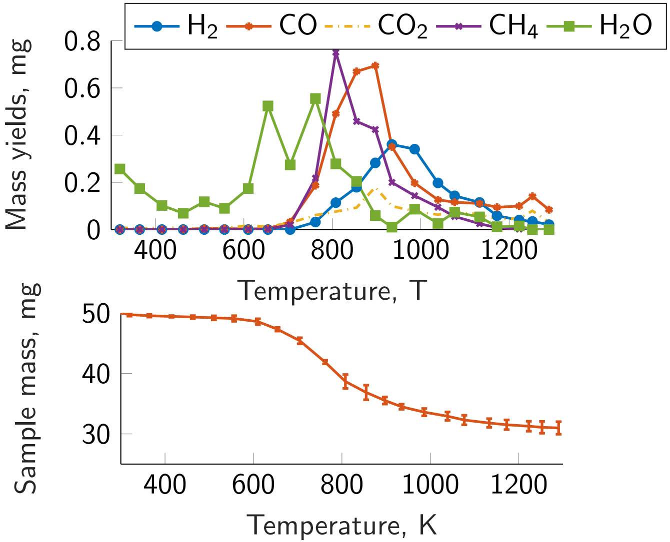
Pyrolysis experiments [Wong et al., 2015; Bessire and Minton, 2017]





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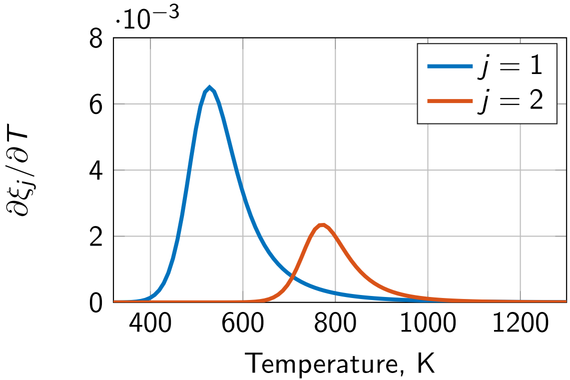
Phenomenological laws for pyrolysis mass loss and gas species production

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



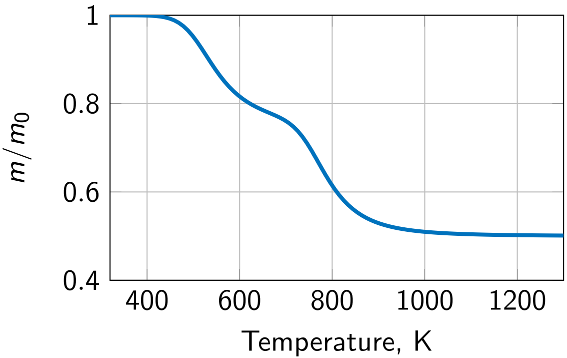
$$\langle \dot{\omega}_i^{\text{pyro}} \rangle = \sum_j^{N_\text{p}} F_{ij} \frac{\partial \xi_j}{\partial T} \tau m_0$$

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

- ξ_j: 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]



$$m = m_0 - \sum_{j}^{N_p} F_j \xi_j m_0$$

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

- ξ_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 $\mathbf{d}^{\text{obs}} = \{d_i \in \mathbb{R}, i = 1, \dots, n_{\text{obs}}\}$
- A model that predicts the measurements

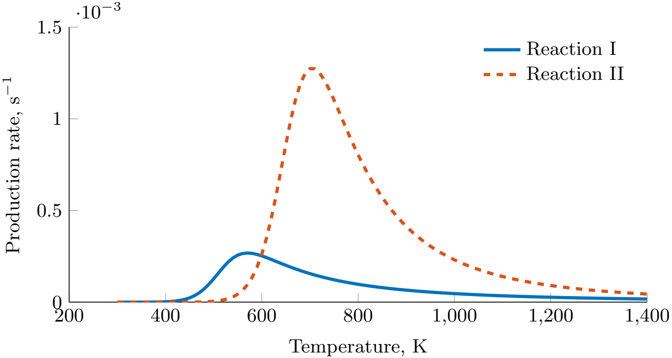
using Bayesian rule for improving our knowledge on $\mathbf{p}$

$$\pi(\mathbf{p}|\mathbf{d}^{\text{obs}}) = \frac{\pi(\mathbf{d}^{\text{obs}}|\mathbf{p})\pi_0(\mathbf{p})}{\int_{\mathbb{R}^p} \pi(\mathbf{d}^{\text{obs}}|\mathbf{p})\pi_0(\mathbf{p})d\mathbf{p}} \qquad \text{(Bayes' theorem)}$$

- 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

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

Benchmark: two-equation model



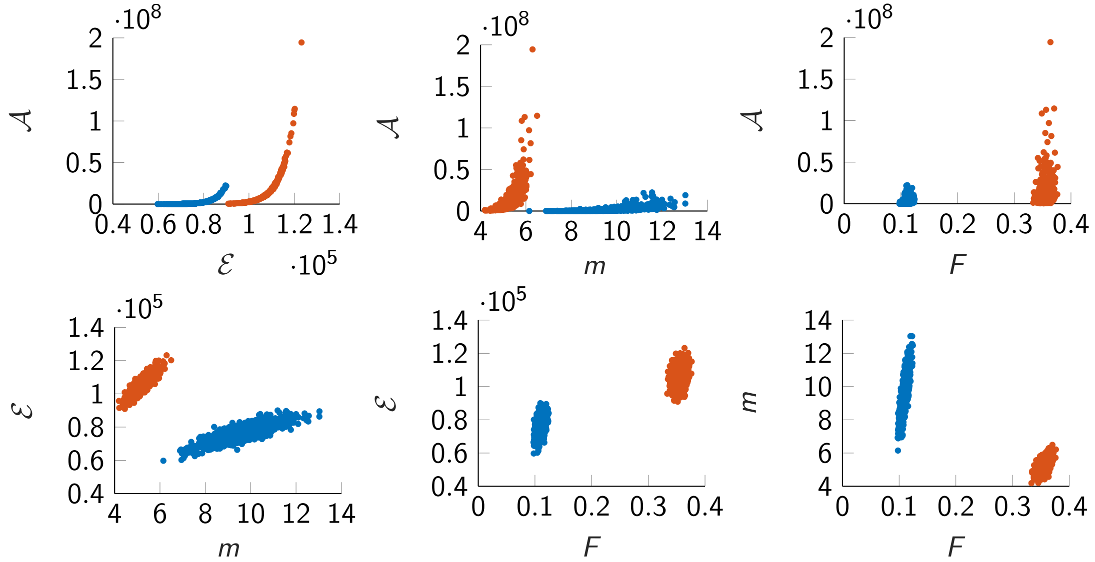
Synthetic data using:

$$\sigma_{\mathrm{I}} = 0.149 \text{ s}^{-1}$$

$$\sigma_{\mathrm{II}} = 0.5382 \text{ s}^{-1}$$

	A	E	m	F
Reaction I	541218	74046.7	9.08	0.11
Reaction II	5360000	103680	5.07	0.35

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

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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

$$
\begin{aligned}
\tilde{A}_i &= \ln A_i - E_i/(\mathcal{R}\overline{T}_i), \\
\tilde{E}_i &= E_i/\overline{E}_i, \\
\tilde{m}_i &= m_i/\bar{m}_i, \\
\tilde{F}_i &= F_{ij}/\overline{F}_{ij}.
\end{aligned}
$$

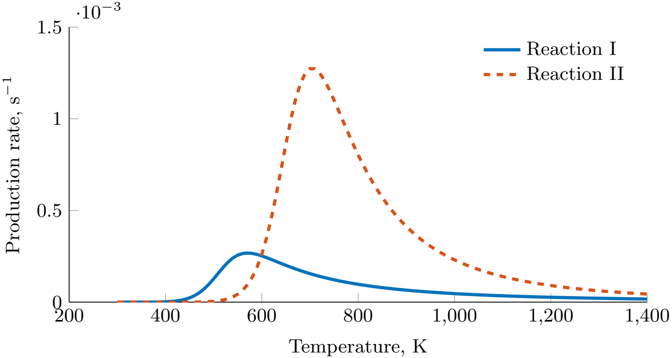
Reaction rate:

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

Local reciprocal temperature:

$$\tilde{T}_i = \overline{\tilde{E}}_i/\mathcal{R}(1/T + 1/\overline{T}_i)$$

Benchmark: two-equation model



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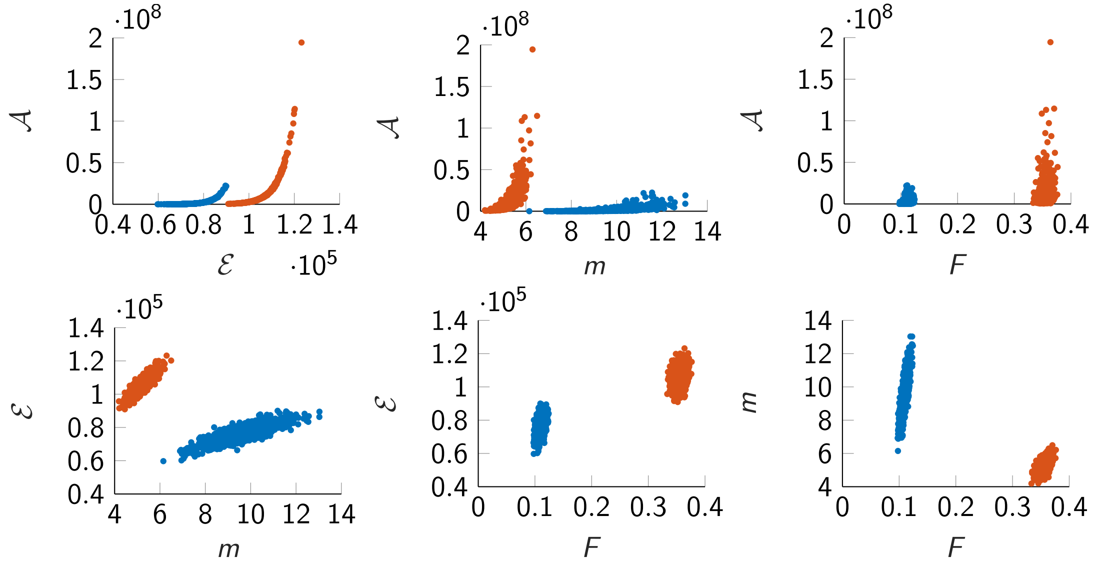
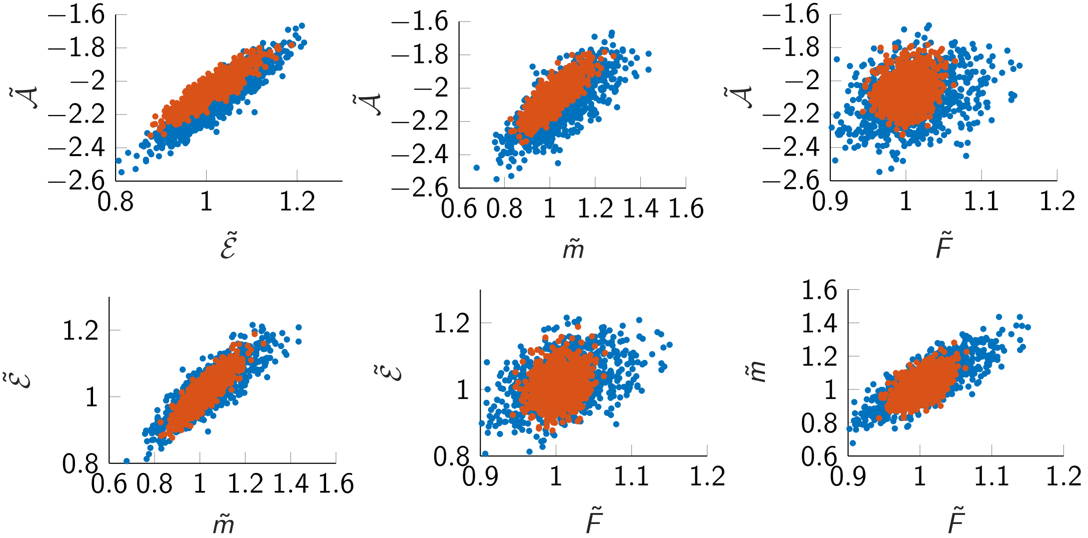
Bivariate posterior PDFs (initial parameter space)



Bivariate posterior PDFs (rescaled parameter space)



Itô Stochastic Differential Equation based MCMC method

We use the Itô Stochastic Differential Equation (ISDE) introduced by Soize (2008)

$$
\begin{aligned}
d\Xi &= [\widehat{C}]\boldsymbol{H}dt \\
d\boldsymbol{H} &= -\boldsymbol{\nabla}_\xi\phi(\boldsymbol{\Xi})dt - \frac{1}{2}\zeta_0\boldsymbol{H}dt + \sqrt{\zeta_0}\left[L_{\hat{C}}\right]^{-\mathrm{T}}d\boldsymbol{W}
\end{aligned}
$$

with $\phi = -\log(\pi(\mathbf{d}^{\text{obs}}|\mathbf{p}))$, $[\widehat{C}]$: approximation of the covariance matrix, ζ_0: free parameter (Arnst, Soize (2019)). Discretization in time using a Stormer-Verlet method (Soize, Ghanem (2016))

$$
\begin{aligned}
\Xi^{\left(\ell+\frac{1}{2}\right)} &= \Xi^{(\ell)} + \tfrac{\triangle t}{2}[\widehat{C}]\boldsymbol{H}^{(\ell)} \\
\boldsymbol{H}^{(\ell+1)} &= \tfrac{1-b}{1+b}\boldsymbol{H}^{(\ell)} - \tfrac{\triangle t}{1+b}\boldsymbol{\nabla}_\xi\phi\left(\boldsymbol{\Xi}^{\left(\ell+\frac{1}{2}\right)}\right) + \tfrac{\sqrt{\zeta_0}}{1+b}\left[L_{\widehat{C}}\right]^{-\mathrm{T}}\Delta\boldsymbol{W}^{(\ell+1)} \\
\boldsymbol{\Xi}^{(\ell+1)} &= \boldsymbol{\Xi}^{\left(\ell+\frac{1}{2}\right)} + \tfrac{\triangle t}{2}[\widehat{C}]\boldsymbol{H}^{(\ell+1)}
\end{aligned}
$$

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Zero variance data in the data-set

- Hyperparameter choice

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

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

- Feedback on the experiments: fit only important features (activation temperature, maximum production peaks)

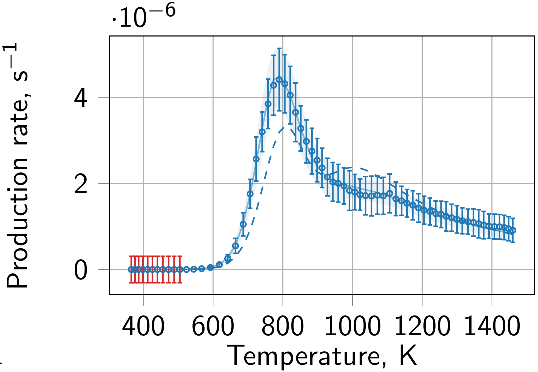
Illustration using experimental data: two-reaction model with one species

- Observable (data) $d^{\mathrm{obs}}_{\mathrm{H}_2 k}$: mass yields of species H_2 at each temperature. $N_\mathrm{p} = 2$.

$$d_{\mathrm{H}_2 k} = \sum_{j}^{N_\mathrm{p}} F_{\mathrm{H}_2,j}\, m_0 \left(\xi_j^{(k)} - \xi_j^{(k-1)}\right),$$

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

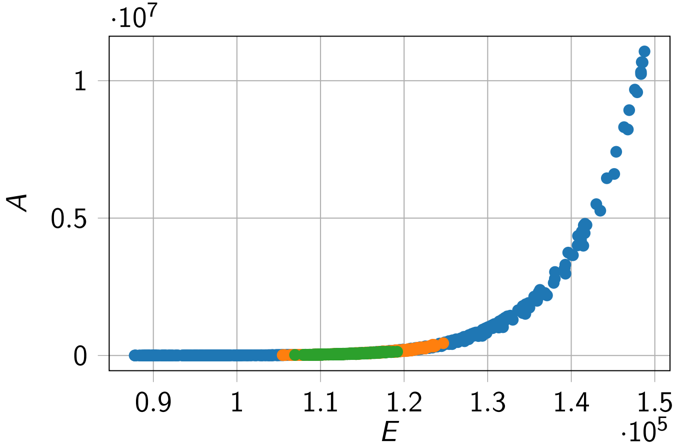
$$\xi_j^{(0)} = 0.$$



- Simple point-mass model (0D), $k = 1, \ldots, n_\mathrm{T}$
- $\mathbf{p} = \{A_1, E_1, n_1, F_{\mathrm{H}_2,1}, A_2, E_2, n_2, F_{\mathrm{H}_2,2}\}$
- 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)



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Application to pyrolysis experiments [Bessire and Minton, 2015]

Proposed five-equation model (38 unknown parameters):

$$\mathrm{R}_1(s) \xrightarrow{k_1} F_{1,1}\mathrm{H_2O} + F_{1,2}\mathrm{CO_2} + F_{1,3}(\text{1-propanol})$$

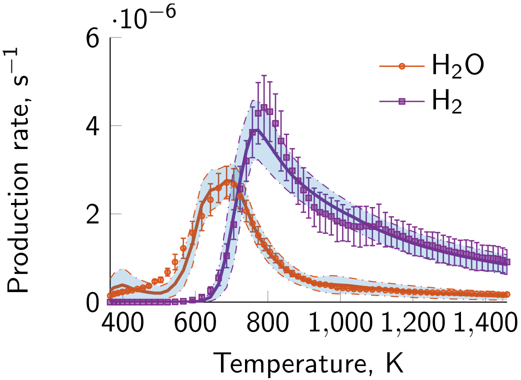
$$\mathrm{R}_2(s) \xrightarrow{k_2} \begin{array}{l} F_{2,1}\mathrm{H_2O} + F_{2,2}\mathrm{CO} + F_{2,3}\mathrm{CO_2} + F_{2,4}\text{Phenol} + F_{2,5}\text{Cresol} + \\ F_{2,6}(\text{Dimethyl Phenol}) + F_{2,7}(\text{Trimethyl Phenol}) \end{array}$$

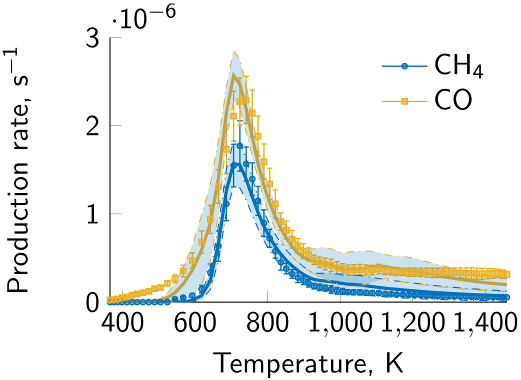
$$\mathrm{R}_3(s) \xrightarrow{k_3} \begin{array}{l} F_{3,1}\mathrm{CH_4} + F_{3,2}\mathrm{H_2O} + F_{3,3}\mathrm{CO} + F_{3,4}\mathrm{H_2} + F_{3,5}\mathrm{CO_2} + \\ F_{3,6}\text{Xylene} + F_{3,7}\text{Benzene} + F_{3,8}\text{Toluene} \end{array}$$

$$\mathrm{R}_4(s) \xrightarrow{k_4} F_{4,1}\mathrm{H_2}$$

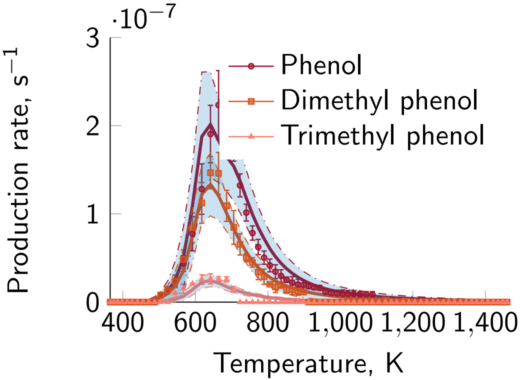
$$\mathrm{R}_5(s) \xrightarrow{k_5} F_{5,1}\mathrm{H_2} + F_{5,2}\mathrm{CH_4} + F_{5,3}\mathrm{H_2O} + F_{5,4}\mathrm{CO}$$

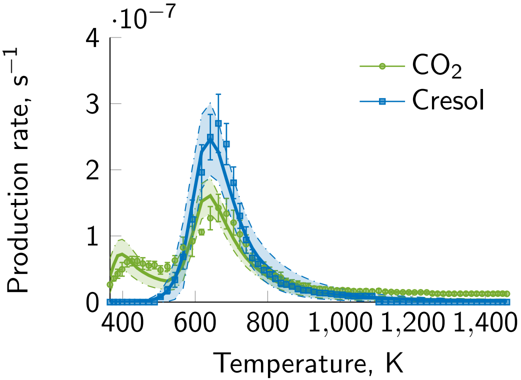
Propagation results: production rate curves



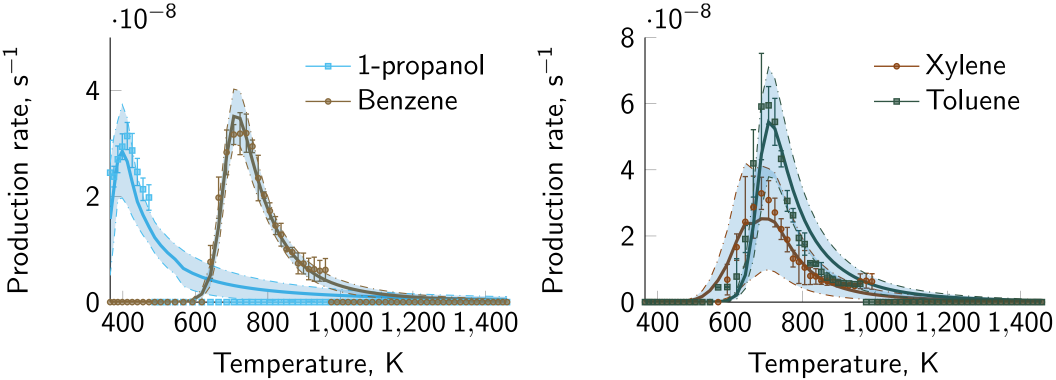


Propagation results: production rate curves





Propagation results: production rate curves



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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 indentifiability 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) $\rightarrow$ model inference
- Results interpretation and their used in CFD codes; *uncertainty propagation for the validation of CFD codes*

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- F. Torres (VKI), B. Helber (VKI), J. Lachaud (I2M Bordeaux), H.-W. Wong (UMassachusetts Lowell), George Bellas-C. (VKI), J.-B. Scoggins (VKI)

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Additional Information

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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, $\mathrm{Bi} = \frac{L_c h}{k} << 1$).

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

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

- $T_{\text{out}} = \tau t$, τ heating rate

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

$$C = \frac{(1-\xi_0)^{1-n}}{1-n} + \frac{A}{\tau}T_0\exp\left(\frac{-E}{RT_0}\right) + \mathrm{Ei}\left(\frac{-E}{RT_0}\right)\frac{EA}{\tau}, \quad \text{where } \mathrm{Ei}(x) \equiv \int_{-\infty}^{-x}\frac{\exp(v)}{v}dv$$

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).

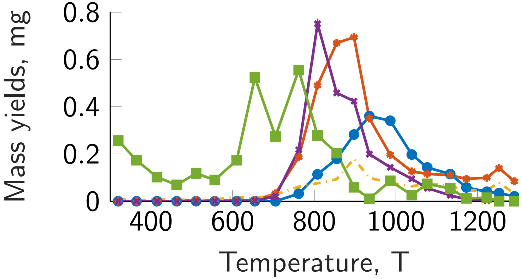
Numerical set-up: 3 reactants model with 5 species

- Observable (data) d_{ik}^{obs} with $i = \{H_2, CO, CO_2, CH_4, H_2O\}$: mass yields at each temperature iteration k. $N_p = 3$.

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

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

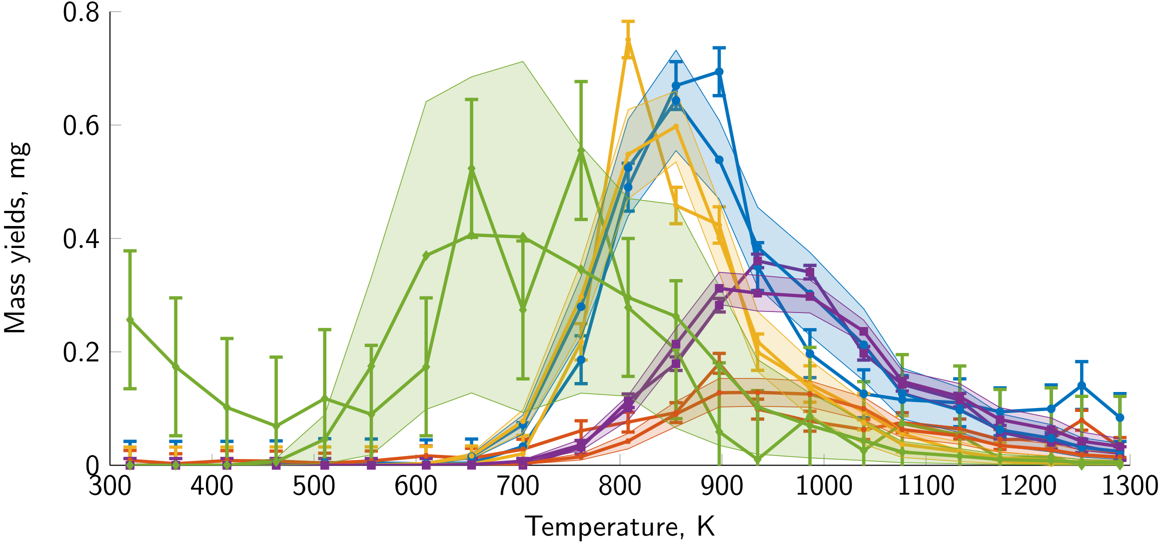
$$\xi_j^{(0)} = 0.$$



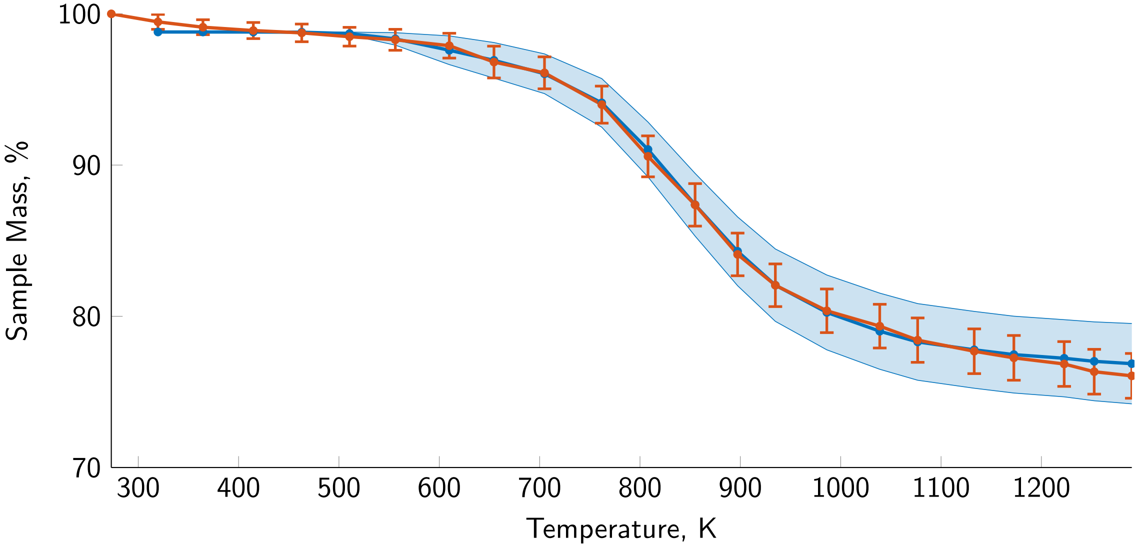
- Simple point-mass model (0D), $k = 1, \ldots, n_T$

$$\mathbf{p} = \{ \{A_3, E_3, n_3, F_{H_2O,3}, \} \quad R_1 \to H_2O$$
$$\{A_2, E_2, n_2, F_{CO,2}, F_{CH_4,2}, F_{H_2O,2}\}, \quad R_2 \to CO + CH_4 + H_2O$$
$$\{A_1, E_1, n_1, F_{CO,1}, F_{CO_2,1}, F_{H_2,1}\} \} \quad R_3 \to CO + CO_2 + H_2$$

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"

$$\textit{Accuracy} >< \textit{complexity}$$

- Kullback-Leibler information

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

- Information criteria

$$\text{AIC} = -2\log(\mathcal{L}(\boldsymbol{y}|\hat{\theta})) + 2K$$
$$\text{BIC} = -2\log(\mathcal{L}(\boldsymbol{y}|\hat{\theta})) + K\log n$$

Illustration with the two-equation benchmark

- Approximate model: one-equation model

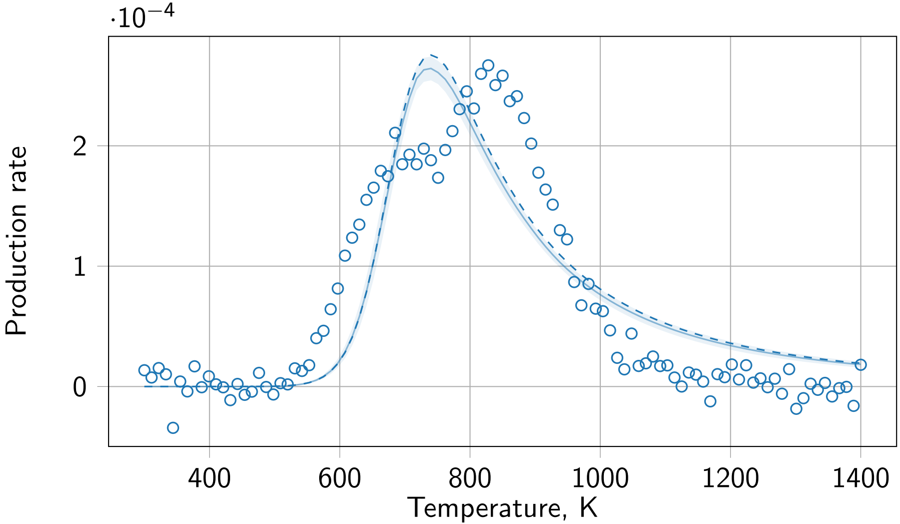


Illustration with the two-equation benchmark

- Approximate model: two-equation model

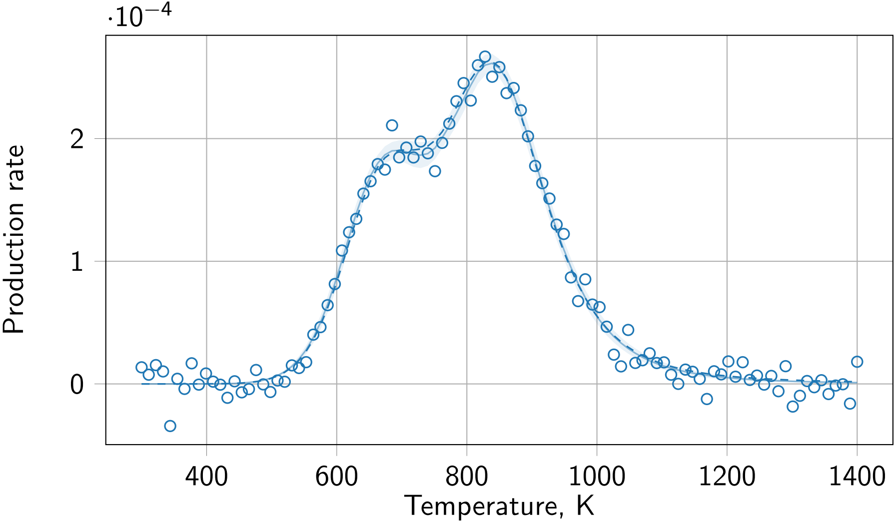


Illustration with the two-equation benchmark

- Approximate model: three-equation model

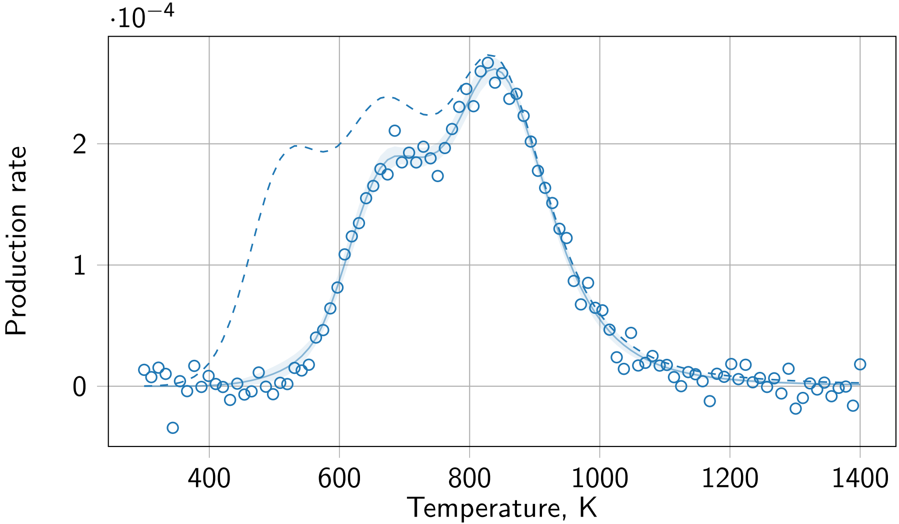


Illustration with the two-equation benchmark

- Computation of information criteria:

Model	log-like	N_{params}	N_{samples}	AIC	AIC_c	BIC
1 reaction	-851.59	4	101	1711.172	1711.588	1711.188
2 reactions	-46.08	8	101	108.1501	109.7153	108.1847
3 reactions	-46.51	12	101	117.0258	120.5712	117.1078
3 reactions (2)	-46.27	12	101	116.54	120.0855	116.592