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Embedded Error Bayesian Calibration of Thermal Decomposition of Organic Materials

PRESENTED BY

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Overview

Organic Material Decomposition and Thermogravimetric Analysis

Pyrolysis Modeling

Embedded Error Formulation and Numerical Methods

Results

Organic Material Decomposition and Thermogravimetric Analysis

Organic Materials

Organic materials are increasingly being used as structural materials, replacing metals and other heavy materials

- Lightweight, easy to shape
- Reduced corrosion and fatigue
- Versatile applications

But may decompose at lower temperatures

- Pose fire and smoke risk (deflagration or smoldering)
- Structural integrity issues

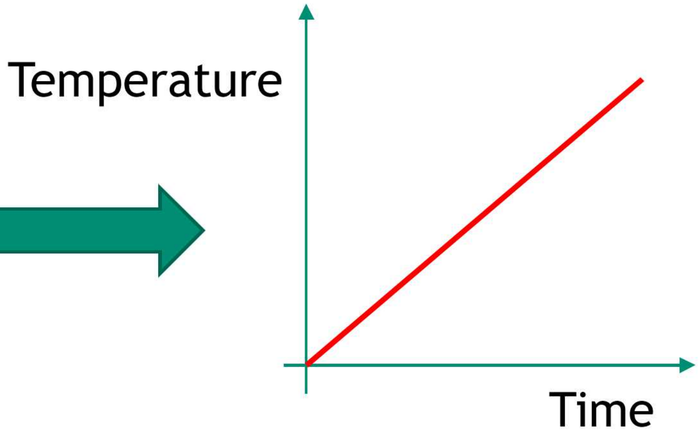
Boeing 787 Dreamliner airframe is 50% Carbon fiber epoxy composites

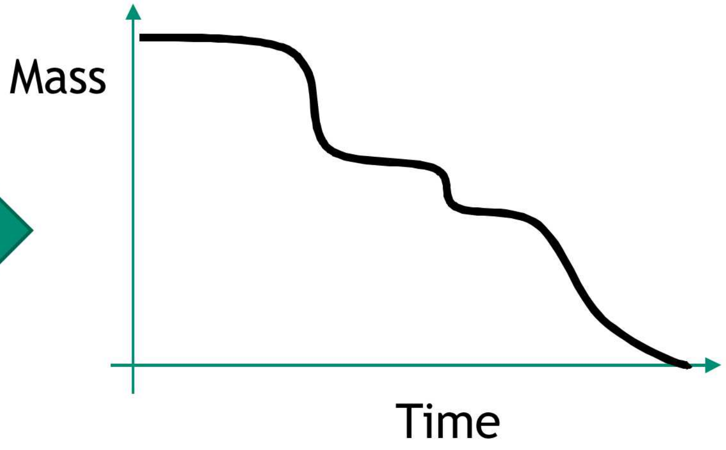
Thermogravimetric Analysis (TGA)

Goal is to analyze the safety limits and potential accident scenarios involving exposure of organic materials to high temperatures

Can explore decomposition rates of materials at different temperatures using TGA

TGA analysis





Pyrolysis Modeling

Pyrolysis Models

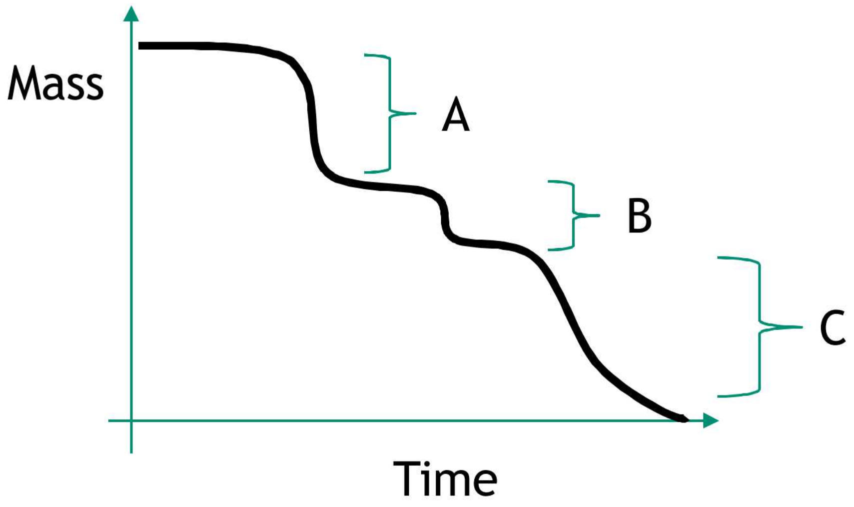
Chemistry can be very complex in pyrolysis

Common choice: fit reduced-order mechanisms to independent components, use Arrhenius rate law

$$\frac{dm}{dt} = -A \exp\left(-\frac{E}{RT}\right) m^n$$

Alternate form of Arrhenius: define characteristic temperature T_o such that $\log A = \frac{E}{RT_o}$

Reduces parameter correlations and model non-identifiability

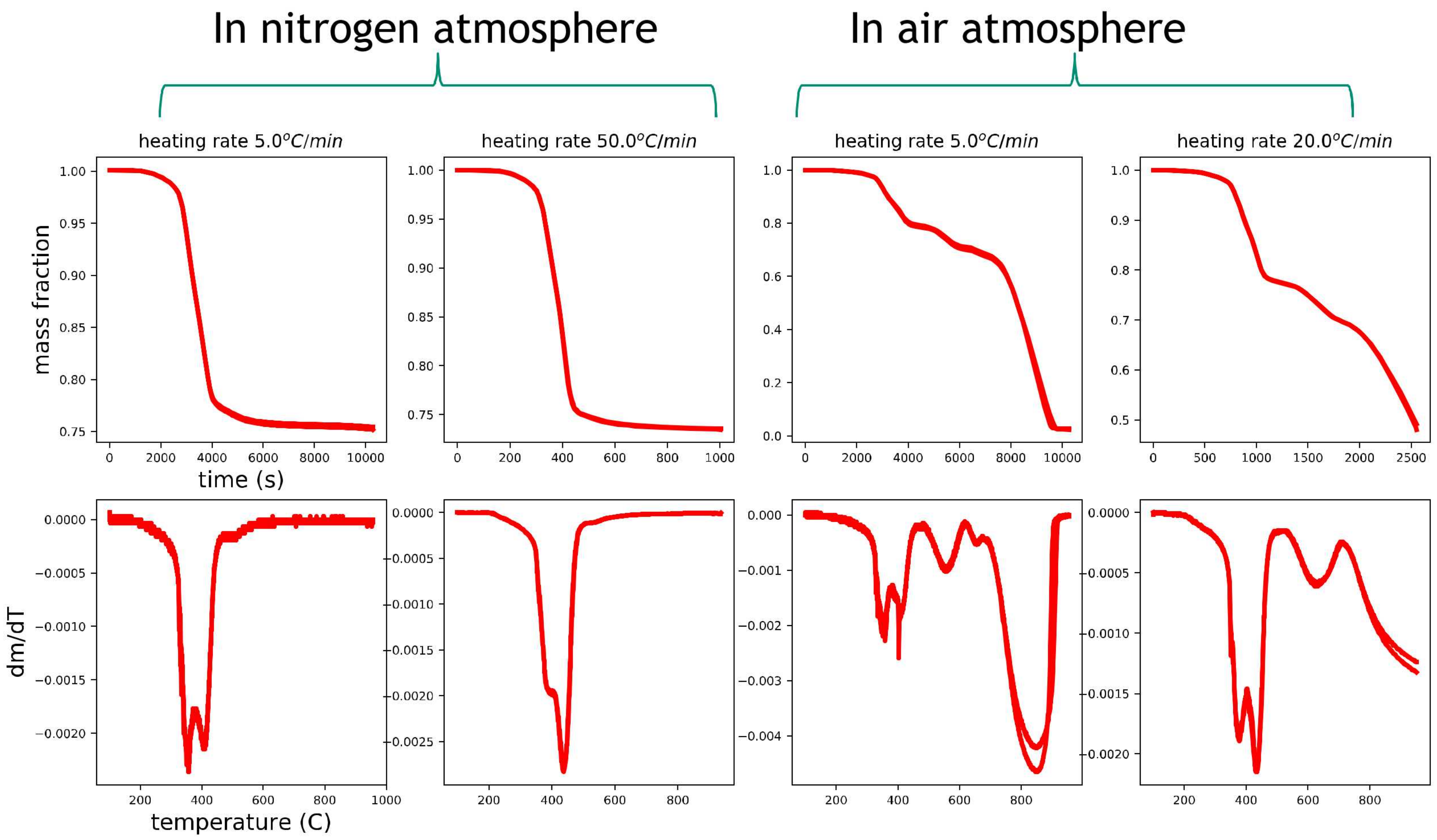


$$\frac{dm_A}{dt} = -\exp\left(\frac{E_A}{R}\left(\frac{1}{T_{oA}} - \frac{1}{T}\right)\right) m_A^{n_A} \qquad m_A(t=0) = m_{Ao}$$

$$\frac{dm_B}{dt} = -\exp\left(\frac{E_B}{R}\left(\frac{1}{T_{oB}} - \frac{1}{T}\right)\right) m_B^{n_B} \qquad m_B(t=0) = m_{Bo}$$

$$\frac{dm_C}{dt} = -\exp\left(\frac{E_C}{R}\left(\frac{1}{T_{oC}} - \frac{1}{T}\right)\right) m_C^{n_C} \qquad m_C(t=0) = m_{Co}$$

Carbon Fiber Epoxy Composite: TGA Data and Model



Carbon Fiber Epoxy Composite: TGA Data and Model

Choose to fit 3 major, separately identifiable reactions

One reaction is pyrolysis without oxygen, two reactions require oxygen

Epoxy is a proprietary component; no knowledge of major chemical decomposition mechanisms

Carbon fiber is known to oxidize mostly to completion at very high temperature, off-gas as CO/CO_2

$$A \rightarrow \text{gas} \quad \frac{dm_A}{dt} = -\exp\left(\frac{E_A}{R}\left(\frac{1}{T_{oA}} - \frac{1}{T}\right)\right) m_A^{n_A} \qquad m_A(t=0) = m_{Ao}$$

$$B \xrightarrow{O_2} \text{gas} \quad \frac{dm_B}{dt} = -\delta_{O_2} \exp\left(\frac{E_B}{R}\left(\frac{1}{T_{oB}} - \frac{1}{T}\right)\right) m_B^{n_B} \qquad m_B(t=0) = m_{Bo}$$

$$C \xrightarrow{O_2} (1-\nu)\text{gas} + \nu\,\text{residue} \quad \frac{dm_C}{dt} = -\delta_{O_2} \exp\left(\frac{E_C}{R}\left(\frac{1}{T_{oC}} - \frac{1}{T}\right)\right) m_C^{n_C} \qquad m_C(t=0) = m_{Co}$$

$$\frac{dm_r}{dt} = -\nu \frac{dm_C}{dt} \qquad m_r(t=0) = 0 \qquad \delta_{O_2} = \begin{cases} 1 & \text{in air} \\ 0 & \text{in } N_2 \end{cases}$$

$$m_{TGA} = m_A + m_B + m_C + m_r \qquad T = T_i + \beta t$$

Carbon Fiber Epoxy Composite

This gives us 12 parameters to calibrate:

- 3 activation energies
- 3 characteristic temperatures (related to log-Arrhenius factor)
- 3 reaction orders
- 2 mass fractions + 1 stoichiometric coefficient

Main approaches in literature:

- Use optimizer to target close fit between model data and TGA data
- Choose various objectives: mass, mass-derivative, combination, least-squares, least-deviations, etc…
- Choose various optimizers: Newton, quasi-Newton, genetic algorithms…
- Return best fit parameters

Challenges:

- Models of this kind are generally too simplistic to be fully trusted
- Optimization alone cannot give uncertainty estimates for each parameter from limited data/model form error

Embedded Error Formulation

Embedded Error Formulation

Bayesian inference

- Given prior knowledge of parameter ranges and data set, return posterior distribution for credible parameter values
- Considered state-of-the-art in many disciplines for statistical inference with uncertainty

Additive error formulation used in "normal" calibration:

$$m_{TGA} = m_{model}(\theta) + \epsilon$$

- Parameters are **random variables**
- Experimental observations are equal to model plus some experiment noise
- Given a lot of data, will eventually converge to "true" parameter values, even if noise in experiments is large

Embedded error formulation (Sargsyan et al, 2019):

$$m_{TGA} = m_{model}(\theta(\xi)) + \epsilon$$

- Parameters are **random distributions**
- Experimental observations are equal to random model evaluations plus some experiment noise
- Given a lot of data, will eventually converge to a distribution of parameter values, cover experimental variation

Embedded Error Formulation

In equation:

$$-\log p(m_{TGA}|\xi) = \sum_{i=1}^{N_{exp}} w_i \left[\sum_{j=1}^{T} \frac{(m_{ij,TGA} - E_\xi(m_{ij,model}))^2}{2(V_\xi(m_{ij,model}) + \sigma_n^2)} + \frac{1}{2}\log(2\pi(V_\xi(m_{ij,model}) + \sigma_n^2) \right]$$

Assume each variable is independently Gaussian distributed with uncertain mean and uncertain variance

Algorithm:

- Sample values for mean and variance for each parameter
- Draw many samples of realizations of the parameters from those means and variances using quasi-Monte Carlo sampling scheme
- Evaluate the model at each set of parameters for each set of experiments
- Evaluate mean and variance of the mass from the model over all samples
- Compute log-likelihood using above equation, comparing against all experimental data

Maximum A Posteriori Inference

Bayesian inference

$$p(\xi|m_{TGA}) \sim p(m_{TGA}|\xi)p(\xi)$$

Due to expense of evaluating likelihood, use MAP inference for now

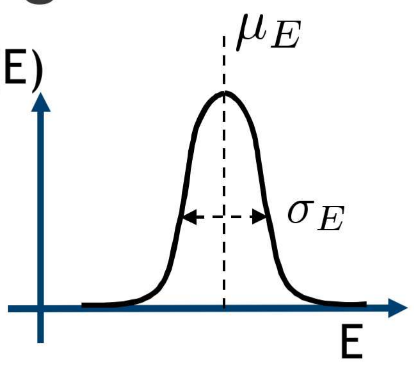
$$\xi^* = \arg\max_{\xi} p(m_{TGA}|\xi)p(\xi)$$

Flat prior for the uncertain means over wide interval, Jeffreys prior for the variances

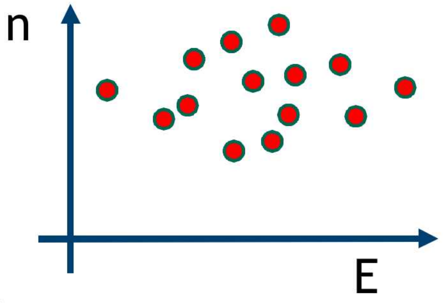
Optimize parameters using differential evolution in SciPy, initial MCMC to fine-tune values

Overall algorithm

$$\mu_E = \ldots$$
$$\sigma_E^2 = \ldots$$



Pick values of means/variances

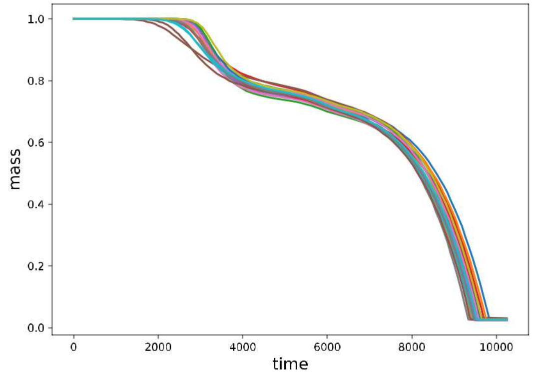


Draw samples of parameters from the distributions

$$\frac{dm_A}{dt} = -\exp\left(\frac{E_A}{R}\left(\frac{1}{T_{oA}} - \frac{1}{T}\right)\right) m_A^{n_A}$$

Evaluate the model at each set of parameters (adaptive RK solver)

Evaluate mean/variance of mass versus time



Compute log-likelihood from experimental data, evaluate objective function

$$E_\xi(m_{model}(t,\xi))$$
$$V_\xi(m_{model}(t,\xi))$$
$$p(m_{TGA}|\xi)p(\xi)$$

Optimization algorithm converged?

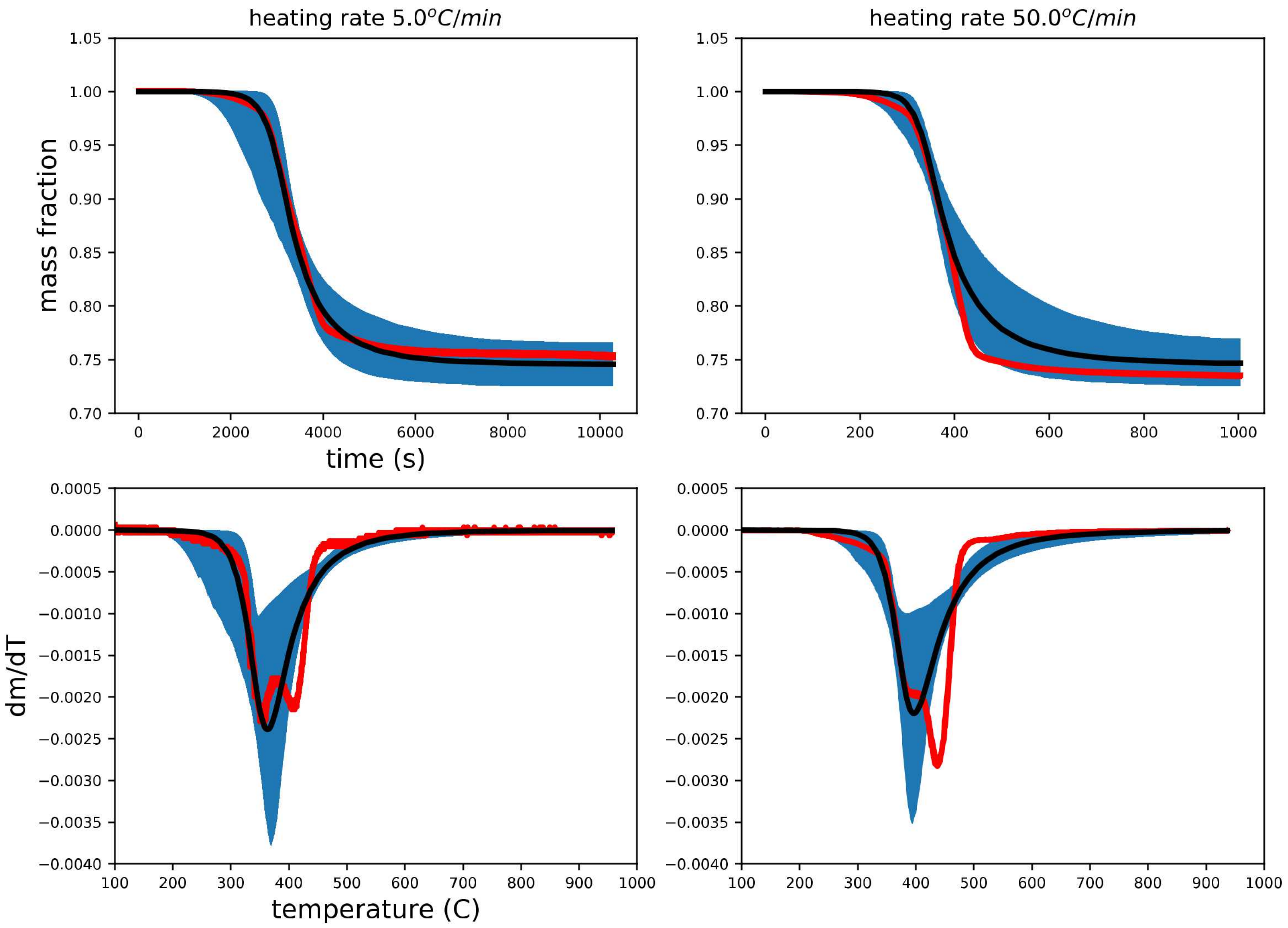
Nope!

Yup!

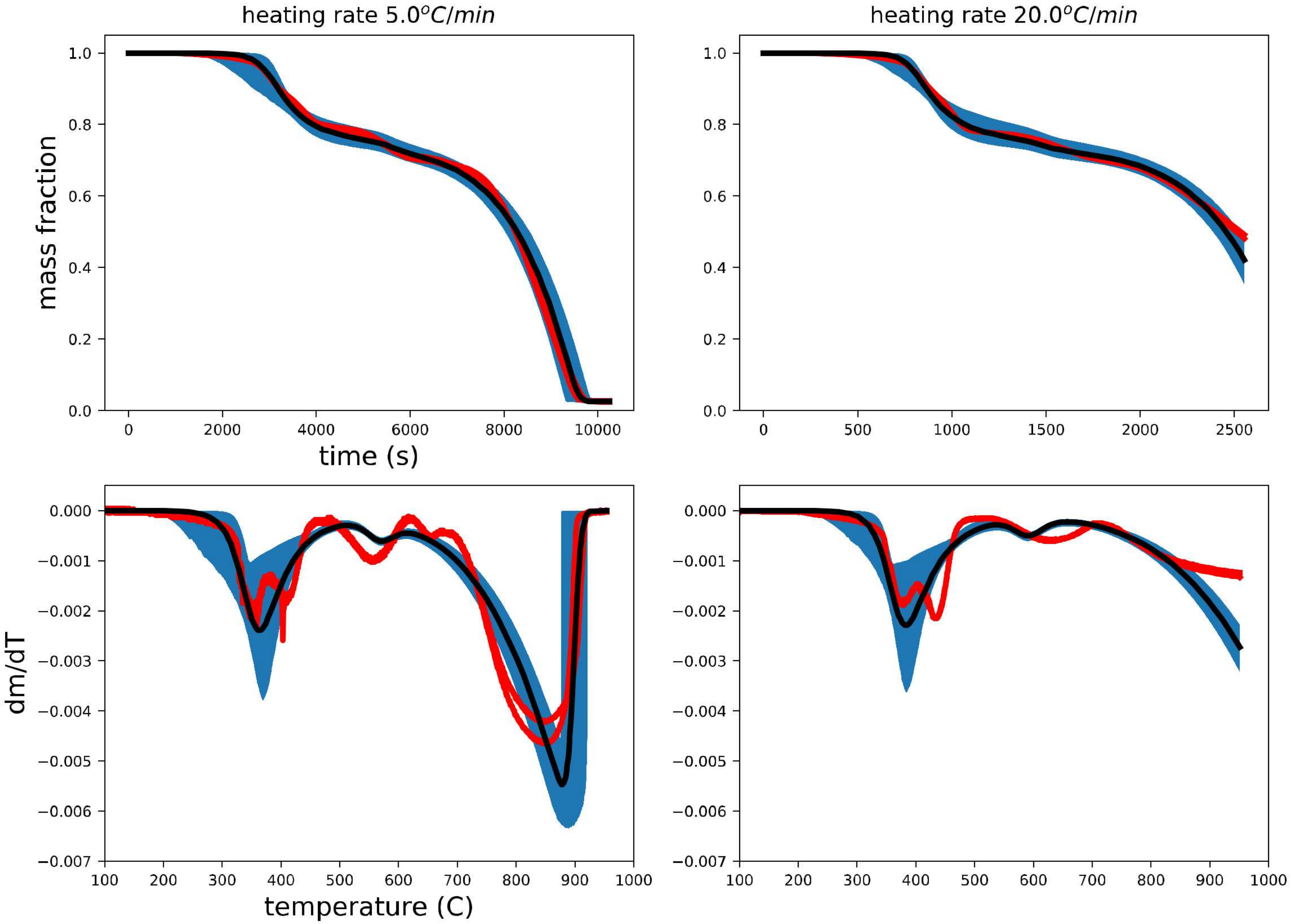
Return final means/variances

Results

TGA: Nitrogen



TGA: Air


heating rate 5.0$^oC/min$
heating rate 20.0$^oC/min$
mass fraction
time (s)
dm/dT
temperature (C)

Calibrated Parameters (MAP)

Noise std dev: 2.99×10^{-4}

Reaction A	Mean	Standard Deviation	Coefficient of Variation
T_o (K)	652	1.24	0.200%
E/R (x 10^{-3}) (K)	24.7	6.25	25.2%
n	4.15	0.174	3.99%
IC	0.255	0.00806	3.16%

Reaction B	Mean	Standard Deviation	Coefficient of Variation
T_o	954	2.62	0.184%
E/R (x 10^{-3})	35.3	0.0196	0.0590%
n	1.41	~0	~0
IC	0.0291	5.33×10^{-4}	1.65%

Reaction C	Mean	Standard Deviation	Coefficient of Variation
T_o	3870	~0	~0
E/R (x 10^{-3})	12.3	0.150	1.23%
n	0.0111	0.00309	27.6%
Stoich. Coeff.	0.0349	~0	~0

Conclusions and Next Steps

Conclusions:

- Developed an embedded-error framework for calibrating general physical models with arbitrary outputs
- Demonstrated application to TGA calibration of organic composite with uncertainty in parameters
- Good coverage of experimental data despite inconsistency in experiments, crude model form

Next steps:

- Fully Bayesian treatment of parameters (in progress)
- Accelerate the optimization process (require very fine precision in parameters)
- Finite sample sensitivity of likelihood function (sampling variance of model outputs)
- Choice of data-form to calibrate against (mass vs. mass-derivative vs. combination)