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Calibration of Organic Material Decomposition Models Using Gradient-Based Optimization and MCMC

E. Le Wagman, S.N. Scott, A. Frankel, R. Keedy, V. Brunini, B. Houchens, T. Johnson

Sandia National Laboratories, California

Goal

- Calibrate organic material decomposition models
  - Improve **efficiency, accuracy, reproducibility** of estimating kinetic parameters from TGA data

Motivation

- Organic materials are everywhere
  - In large quantities: Spacecraft, car, furniture, house
  - Lightweight and strong, replace traditional engineering materials
  - Burn at low temperatures (250°C)
- Large-scale fire simulations
  - Decomposition kinetics model — a critical component [1]
- Complex materials of interest
  - Can't use a global one-step reaction model
  - Tedious traditional analytic approaches [2]
  - Shuffled Complex Evolution (SCE) recommended optimization algorithm, GA used most [3]
  - Both stochastic — different estimates with different random seeds
  - Problematic for high-risk applications

Data

- Thermogravimetric Analysis (TGA)
  - Mg size sample: Isolate chemical kinetics
  - Heat at constant heating rate β
  - Measure total *mass loss* as function of time/temperature
  - Postprocess to get *mass loss rate* (temperature derivative) — see inflection points
- Noisy
- Synthetic data — assess methods against target values

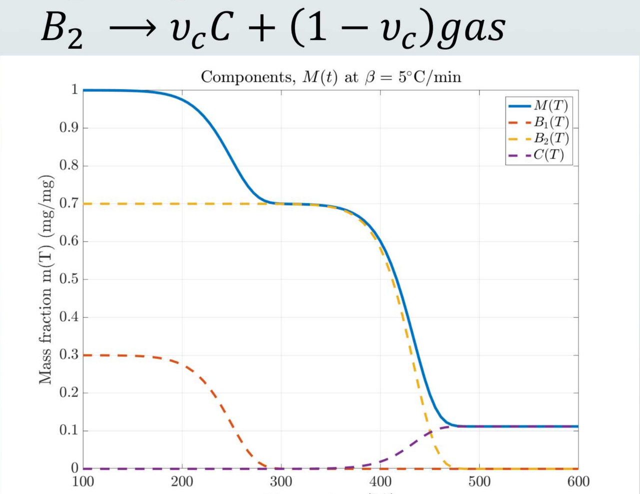
Model

Hypothetical charring material

Two parallel component model - allows for overlapping reactions

$B_1 \longrightarrow gas$

$B_2 \longrightarrow \nu_C C + (1-\nu_C) gas$



$$\frac{dB_1}{dt} = -A_1 \exp\left(-\frac{E_1}{RT}\right) B_1^{n1}$$
$$\frac{dB_2}{dt} = -A_2 \exp\left(-\frac{E_2}{RT}\right) B_2^{n2}$$
$$\frac{dC}{dt} = \nu_C\, A_2 \exp\left(-\frac{E_2}{RT}\right) B_2^{n2}$$
$$\frac{dT}{dt} = \beta$$

$$M = B_1 + B_2 + C$$
$$\frac{dB_1}{dT} + \frac{dB_2}{dT} + \frac{dC}{dT}$$

Approach

- Use/improve advanced gradient optimization – model may be complex, but still a smooth system of ODEs
  - Evolutionary algorithms as a last resort / initial run
  - Cited motivation was problem ill-posedness (multiple solutions), high-dimensionality [3]
  - Instead use a gradient-based method suited to ill-posed, high-dimensional inverse problems
- Improving optimization: Compare objective functions [4]
  - Misfit — *mass loss* or *mass loss rate* data?
- Best fit parameters are inherently uncertain
  - Quantify parameter uncertainty using Bayesian inverse problem framework
  - Incorporate subject matter expert subjective knowledge
  - Estimate probability of nearby parameters

Results – Deterministic Calibration

- $\boldsymbol{m} \in R^9$, 9 parameters:
  - $A_i, E_i, n_i, B_i(0)$ for each component B_i, plus char coefficient ν_C.
  - *Compensation effects,* esp. with kinetic triplets (A_i, E_i, n_i)
  - "$\mathrm{A_i} = \log(A_i)$", scale other parameters to $O(10)$

Objective functions

(100-0) *Mass loss* objective

$$f_1(\boldsymbol{m}) = \sum_{\beta} \sum_{t_i} (M(\boldsymbol{m}) - M_i)^2$$

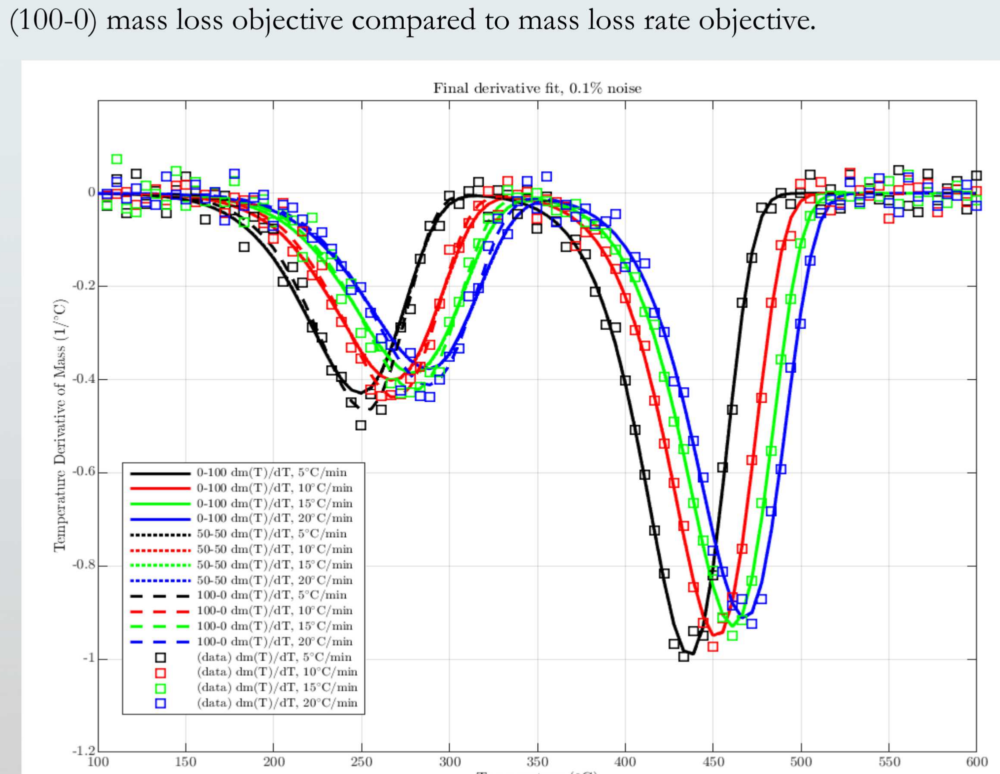
(0-100) *Mass loss rate* objective

$$f_2(\boldsymbol{m}) = \sum_{\beta} \sum_{T_i} \left(\frac{dM(\boldsymbol{m})}{dT} - \left(\frac{dM}{dT}\right)_i \right)^2$$

(50-50) Hybrid objective [5]

$$f_3(\boldsymbol{m}) = \lambda f_1 + (1-\lambda) f_2, \quad \lambda = 0.5$$

See improved fit at peak mass loss rate / matching of rate data at first reaction with (100-0) mass loss objective compared to mass loss rate objective.

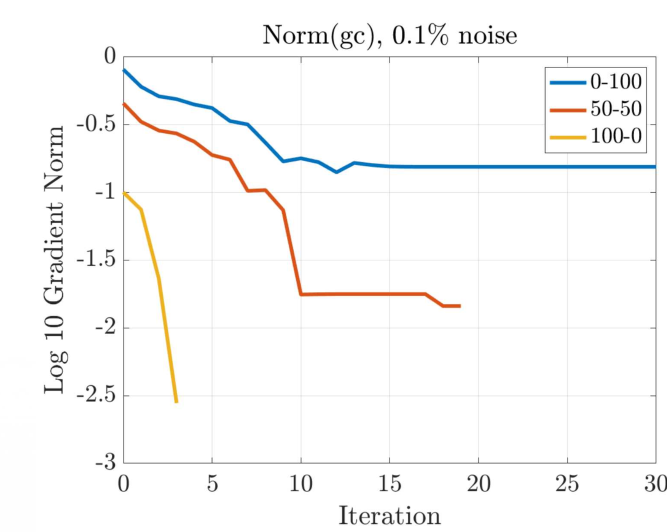


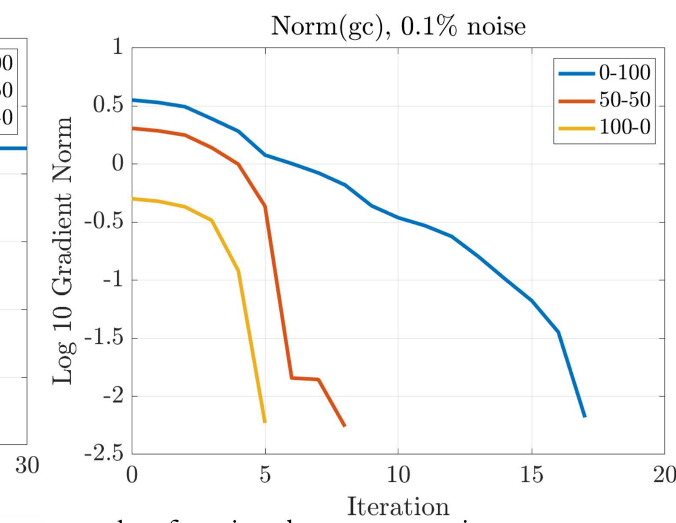
Results – Computational Efficiency

Computational cost history, 0.1% noise:

IC 1	#Model runs	#Newton	Cpu Time (s)
0-100	1293	30	285.53
50-50	816	19	179.69
100-0	125	3	27.45

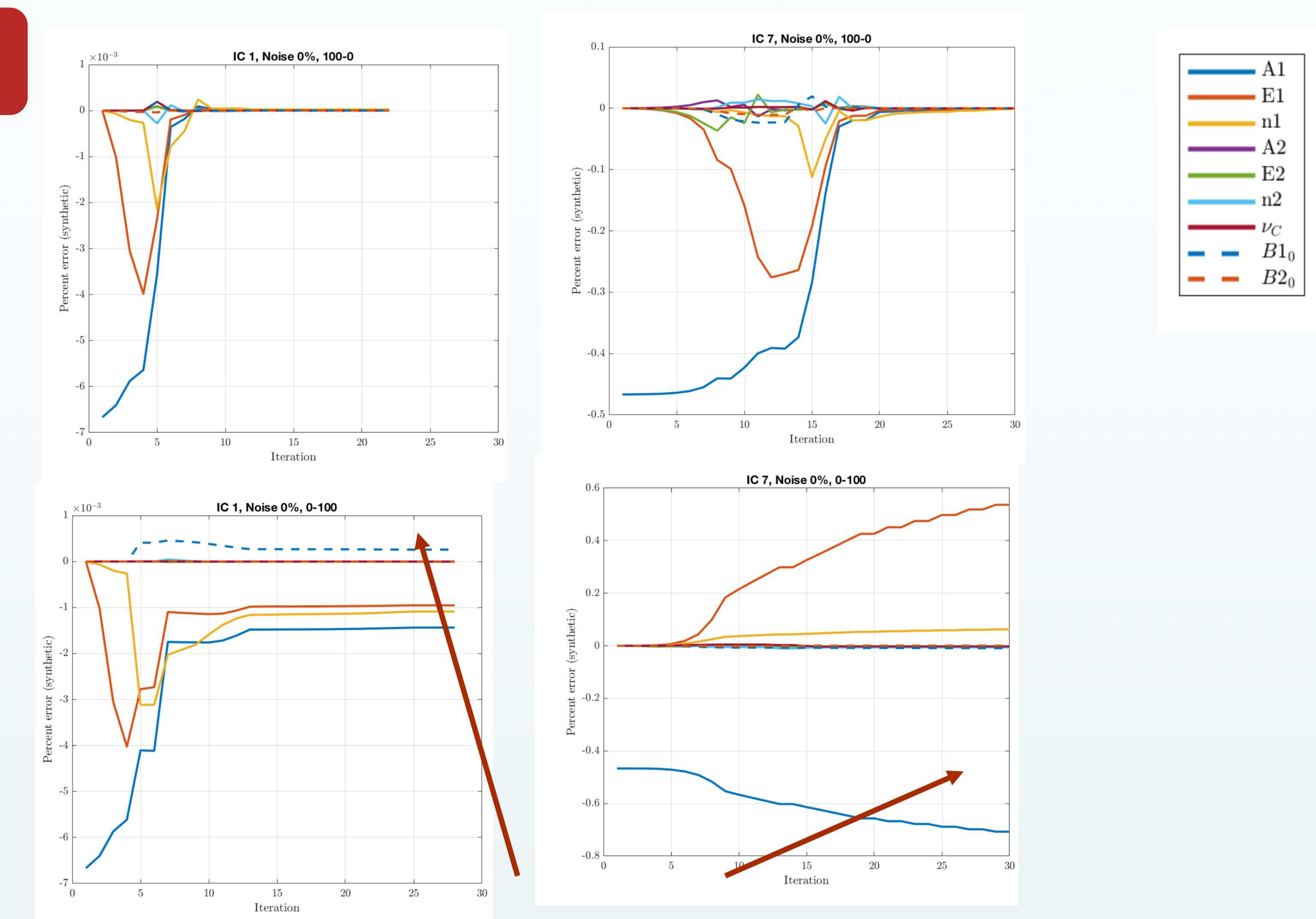
IC 2	#Model runs	#Newton	Cpu Time (s)
0-100	736	17	145.36
50-50	336	8	59.73
100-0	165	5	26.84





Note quadratic shape for (100-0) convergence – a smoother function, better properties for Newton optimizer

Parameter iteration histories (difference to global min/target), no noise:



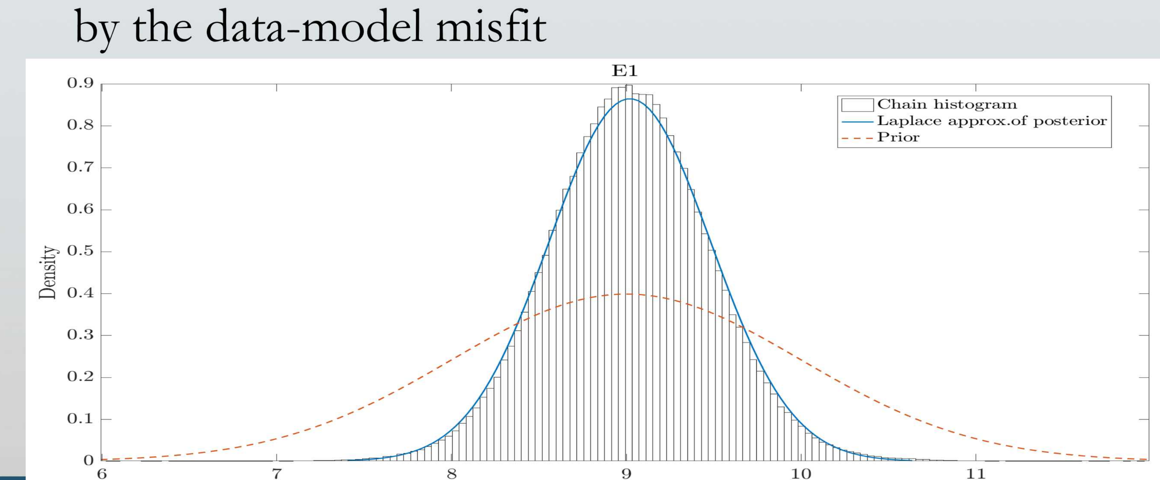
(0-100) Slower or diverges

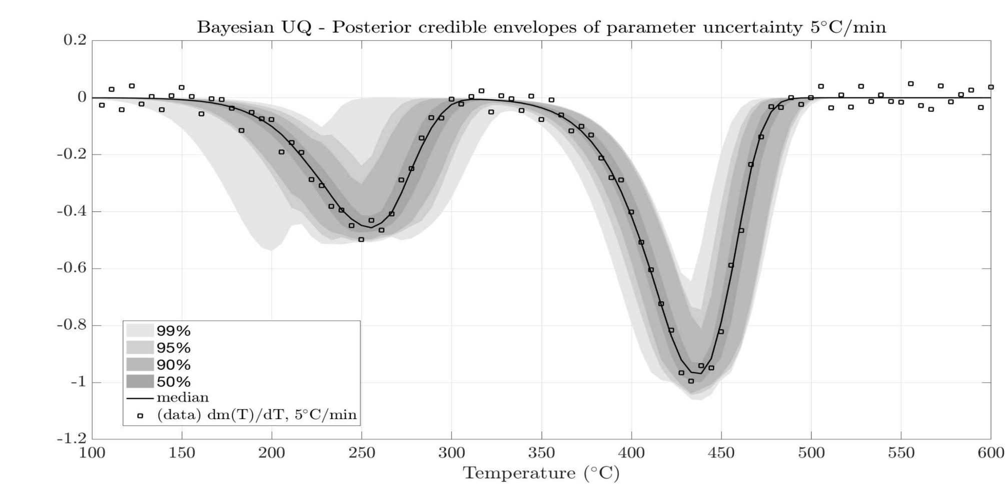
In each of the 6 initial conditions (ICs) where the *mass loss* objective (100-0) converges to the global min (the synthetic target values), the *mass loss rate* objective (0-100) converges more slowly or diverges.

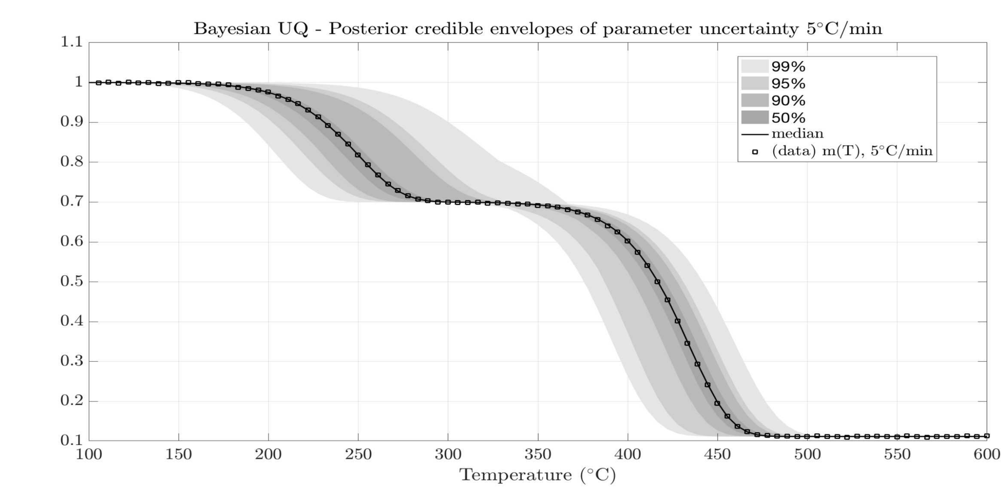
Results – Bayesian Uncertainty Quantification

Using DRAM, Gaussian priors with finite variance, .1% noise

- Posterior Gaussian density estimate plot, only E_i 's are *well-constrained* (posterior range is half of the prior range) by the data-model misfit







- Shown are point-wise posterior credible envelopes containing the given parameter uncertainty, incorporate prior knowledge + data misfit
- Dashed orange line is prior – what we thought the range / probability for E_1 was. Solid line – converged posterior probability for E_1, after 750,000 samples – what we can infer now from Bayes theorem (incorporate information prior, data, model likelihood)

Conclusions

- Quadratic convergence, better matching of noisy *mass loss rate* data, with *mass loss* objective
- Bayesian inverse problem — a mathematical framework, can integrate expert knowledge with experimental data, model misfit, do parameter uncertainty quantification
- Ill-posedness and compensation effects — intended application drives the recommended method / accuracy

Current Work

- Stan: Hamiltonian MC (HMC)
  - Compare convergence, information obtained w/ DRAM
  - Improve convergence of uniform prior or inf.-variance Gaussian
- Analytic gradient
  - Address difficulties with order term and extinguishing mass components

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