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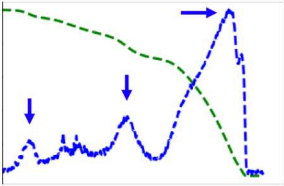

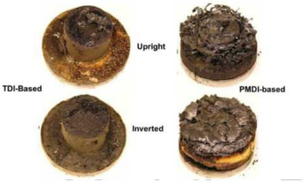


# Advanced Calibration of Decomposition Kinetics Models

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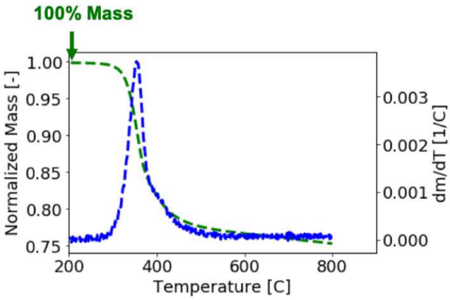
# Intro: Organic material decomposition

# Organic materials are everywhere, unfortunately they burn


- A downside - decompose @ $\sim 250^{\circ}$C
- Motivation: Fire safety
  - Temperatures $> 1000^{\circ}$C in building fires, jet-fuel fires
- **Thermal decomposition**: Material $\rightarrow$ Char $+$ Gas

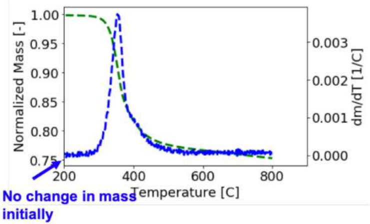
# TGA data: Mass loss history

- Thermogravimetric Analysis (TGA): Mg size sample, isolates chemical kinetics
- Record **mass loss history** $M(T(t))$, time, temperature



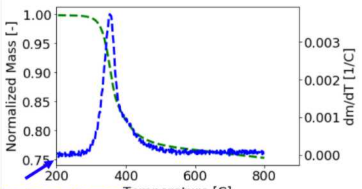
# Derivative data: Mass-loss derivative with respect to temperature

- Finite-differenced **mass-loss derivative** history $\frac{dM}{dT}$

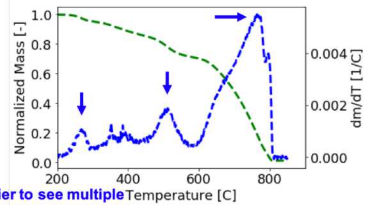


# Derivative data: Mass-loss derivative with respect to temperature

- Finite-differenced **mass-loss derivative** history $\frac{dM}{dT}$
- Easier to see separate reactions




**No change in mass initially**




**Easier to see multiple reactions**

# Objective

- Given
  - TGA data
  - A proposed kinetics mechanism
  - State-of-the-art calibration methods

Part 1. **Q: How can we improve calibration results?**

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  - State-of-the-art calibration methods

Part 1. **Q: How can we improve calibration results?**

Part 2. A: Exploit mathematical properties for fast, robust, **deterministic calibration**

Part 3. A: Gain UQ insights using **Bayesian calibration**

# Uncertain parameters to calibrate

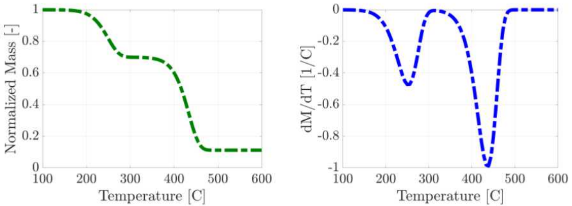
$$Material_1 \longrightarrow Gas$$
$$Material_2 \longrightarrow \boldsymbol{v_c} Char + (1 - v_c) Gas$$

$$\frac{dM_i}{dt} = -\boldsymbol{A_i} \exp\left(-\frac{\boldsymbol{E_i}}{RT}\right) M_i^{\boldsymbol{n_i}}$$

- Total of 9 parameters to calibrate
  - $(A_i, E_i, n_i)$ for each reaction $i$
  - *(Prefactor, activation energy, order)*
  - Initial mass fractions
  - Char rate coefficient $\nu_C$
- Method: Globalized Newton-type (*optpp_fd_newton*, 'trust_region'; *rol*)
- **Data is synthetic**—Assess results against a "truth set" of synthetic parameters

# How to quantify goodness of fit

- Need a **data misfit** metric
- Objective function choices: Which data/features to prioritize fitting?




**(100-0)** **Mass** loss misfit objective

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

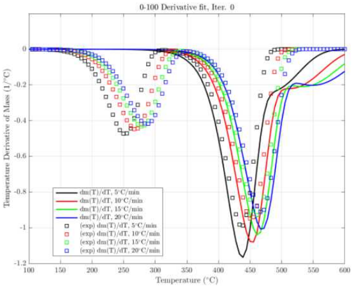
**(0-100)** Mass-loss **derivative** misfit objective

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

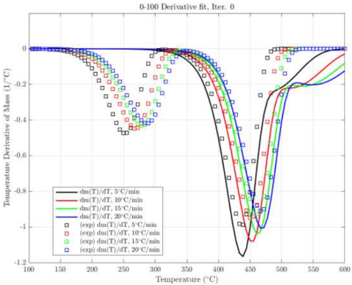
**(50-50)** Hybrid misfit

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

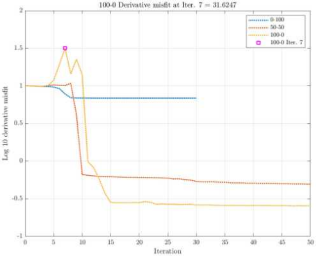
# An unintuitive result: Derivative objective does not lead to the best derivative data fit

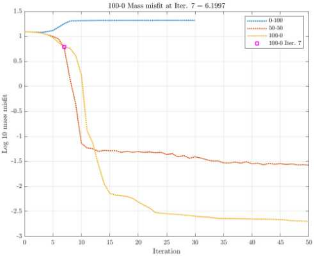


# Optimization history with derivative misfit

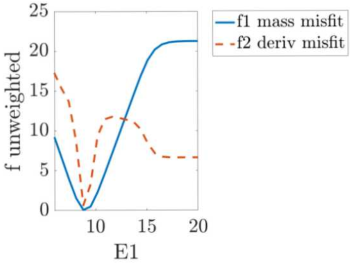


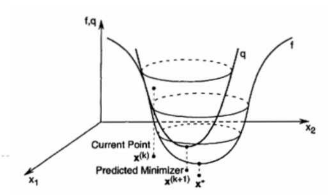
# Comparison of fit value history, different objective functions, same IC



# Contour slices: Why derivative misfit is worse

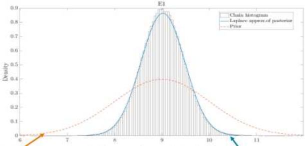



- Both objective functions have the same minimum
- But, mass misfit convex on a wider interval
- **Advanced methods shine when objective function is bowl-like.**

# Parameter UQ w/ Bayesian calibration

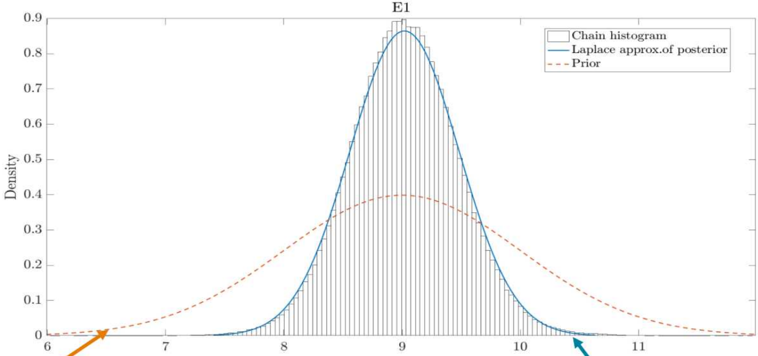
# Why and how: Bayesian UQ with DRAM




**Prior:** What we thought the range / probability for $E_1$ was.
**Posterior:** (After 750,000 samples) – What we can infer now about $E_1$, using Bayes theorem (incorporate information in prior, data, model likelihood)

- Important differences—no truth, subjective prior, simplicity
- TGA: Characterize uncertainty around parameter choices (from deterministic calibration)
- Bayes theorem: *Posterior* $\propto$ *Prior* $\times$ *Likelihood*
- New in Dakota: DRAM for Bayesian calibration

# Bayesian calibration: Data reduced the uncertainty

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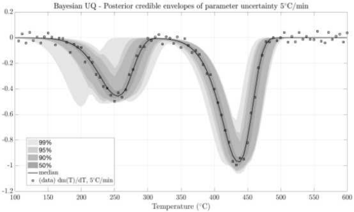
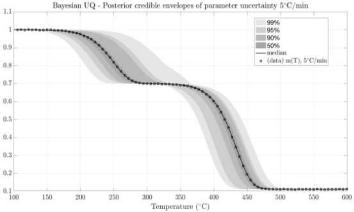
# Usefulness beyond posterior pdfs, application-dependent




- Is risk management the UQ goal?
- Posterior credibility envelopes contain a snapshot of **current state of knowledge**
- E.g., probability of Organic Material X losing mass before 150°C is 1%.

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  - More convex objective function
- **Bayesian calibration**
  - A new/different UQ tool
  - Understand uncertain parameters for fire safety analysis
- Fire safety application: Sometimes, a large gap between advanced tools and their use in engineering...
- ...Also means there are a lot of opportunities!