Augmented Experiments in Material Engineering Using Machine Learning

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Université Sorbonne Paris Nord

CRTI
Research Centre in Industrial Technologies

Synthesis of New Materials in Industry

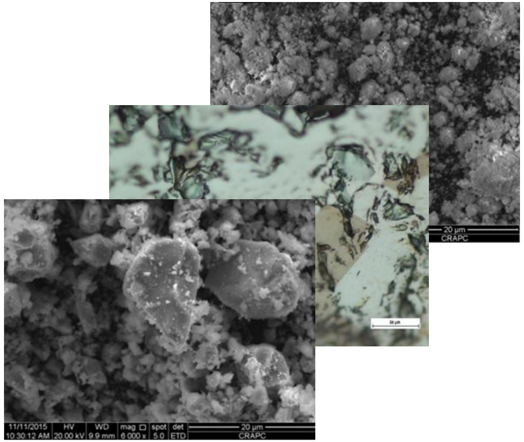
Binary Mixture and Target Material

Calamine Oxide

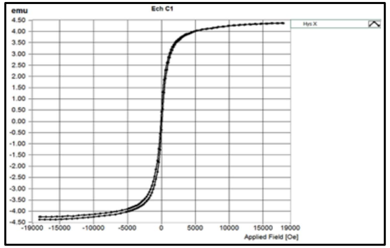
proportion in %

Red Pigment

Synthesis
(Real experiments)



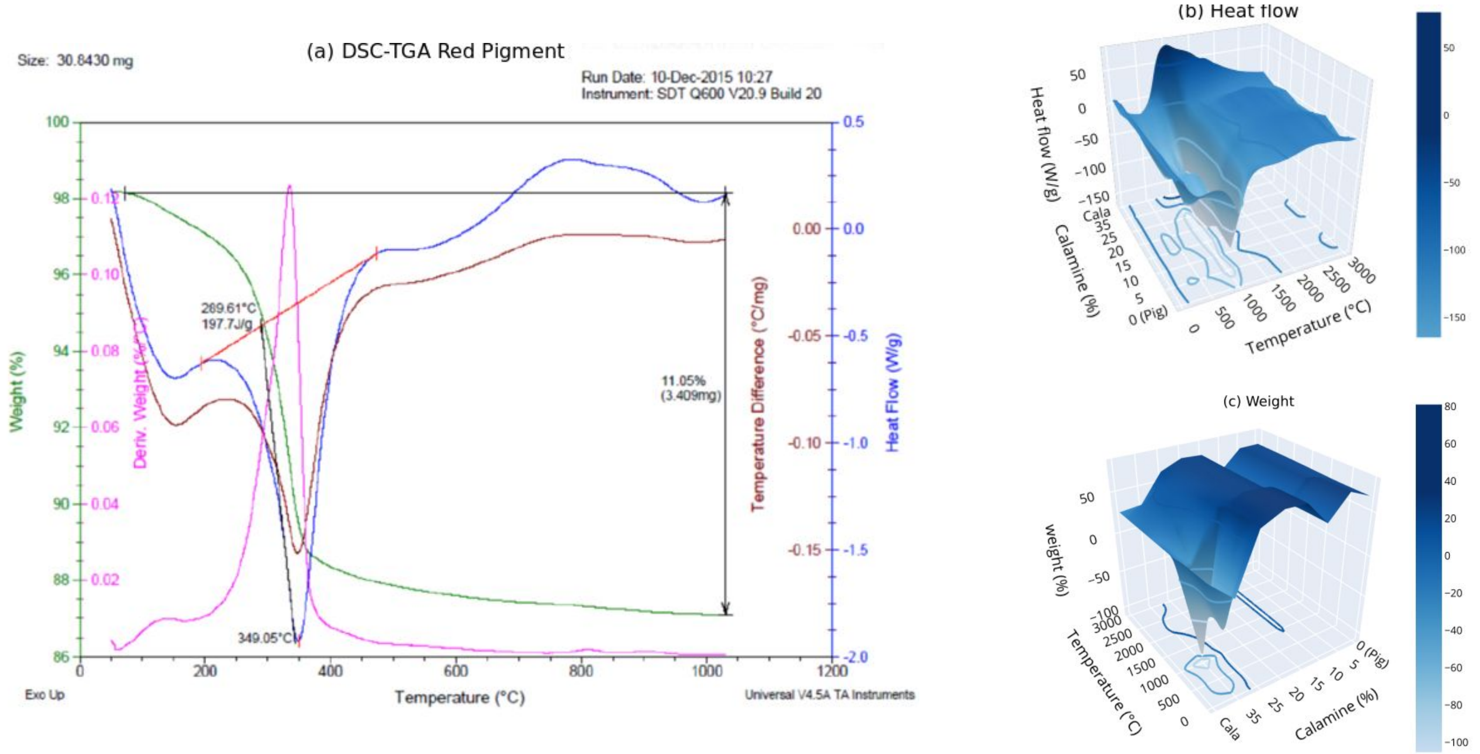
optical properties



ferromagnetic properties

AbedGhars, Mohamed Tayeb. Diss. Université Badji Mokhtar de Annaba, 2018.

Thermal & Mass Loss Analysis



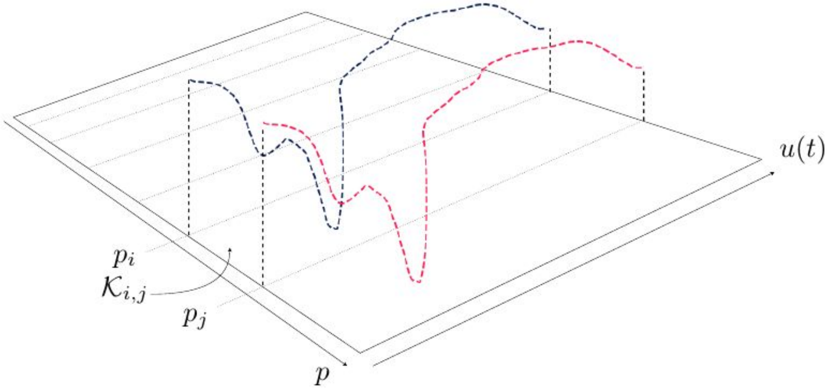
AbedGhars, M. T., S. Bouhouche, and M. Ghers. *Measurement* 109 (2017): 326-333.

State Space Partitioning & Evaluation Protocol

Reconstruction models:

- Inside circumscribed regions;
- Outside circumscribed regions

State space



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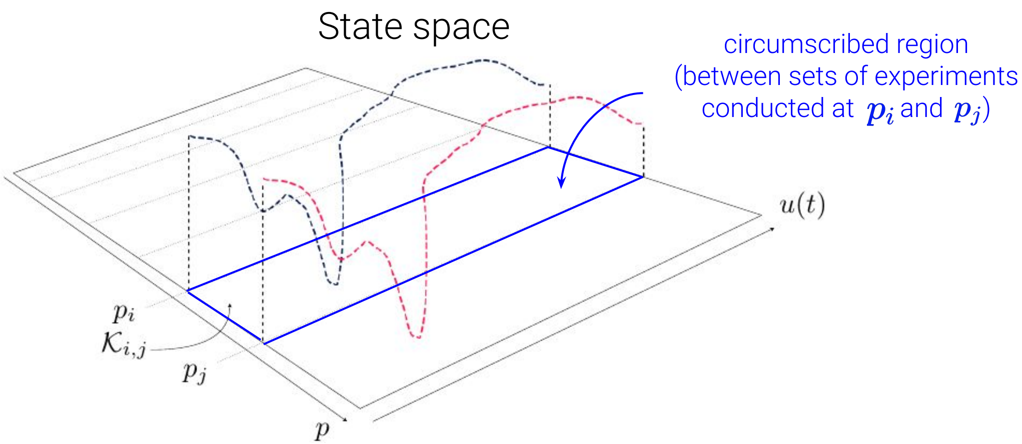

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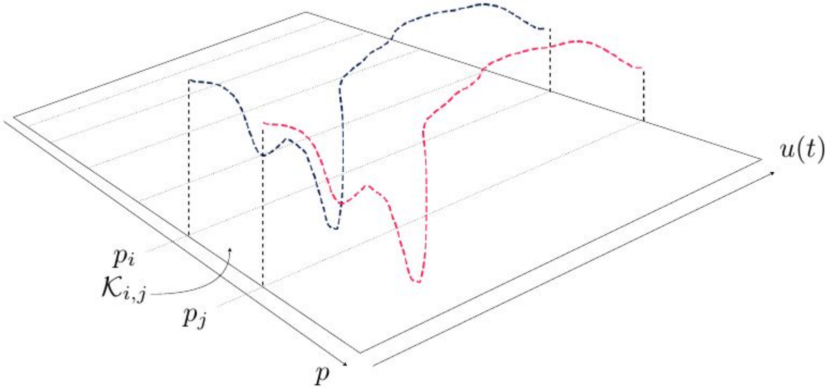
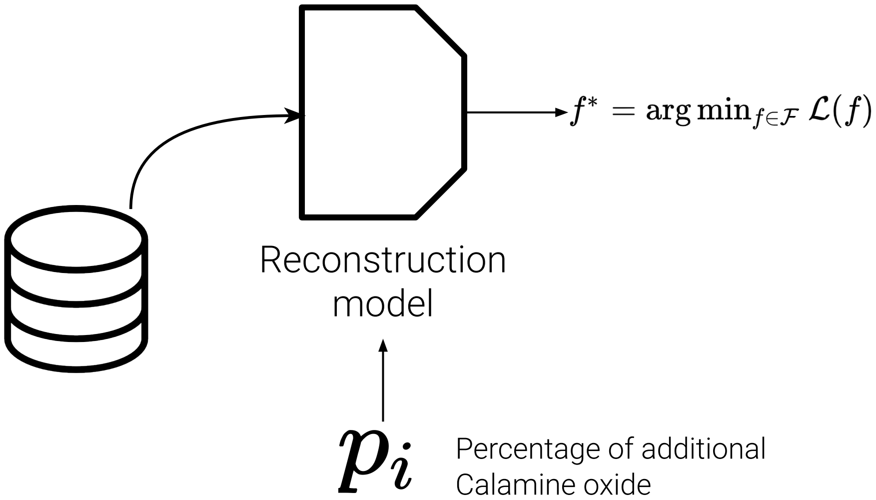




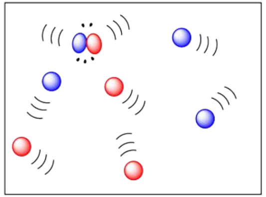
Combining Domain Models & Empirical Data

Combining Analytical Models and Real Experiments

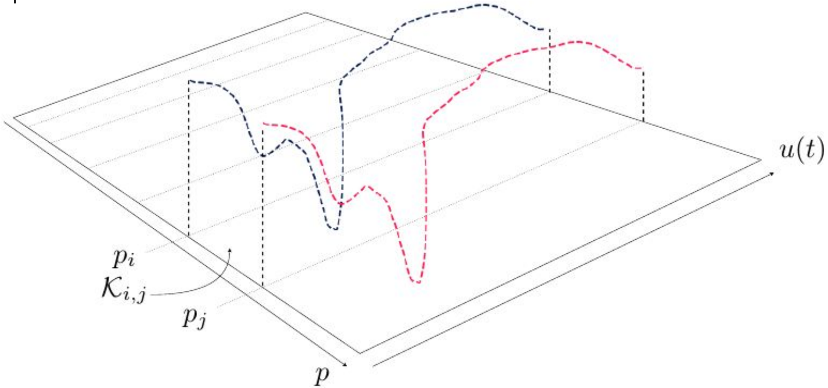
Rate of the reaction $\frac{\partial \alpha}{\partial t} = k(1-\alpha)^n$

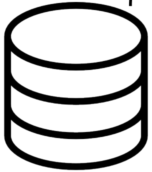
- Arrhenius equation $k = Ae^{-E_a/RT}$

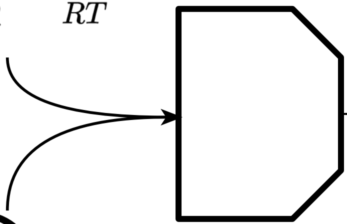
- Eyring equation $k = \frac{k_B T}{h} e^{\frac{\triangle S^{\ddagger}}{R}} e^{-\frac{\triangle H^{\ddagger}}{RT}}$



Pre-exponential factor







$f^* = \arg\min_{f\in\mathcal{F}} \mathcal{L}(f)$

Reconstruction model

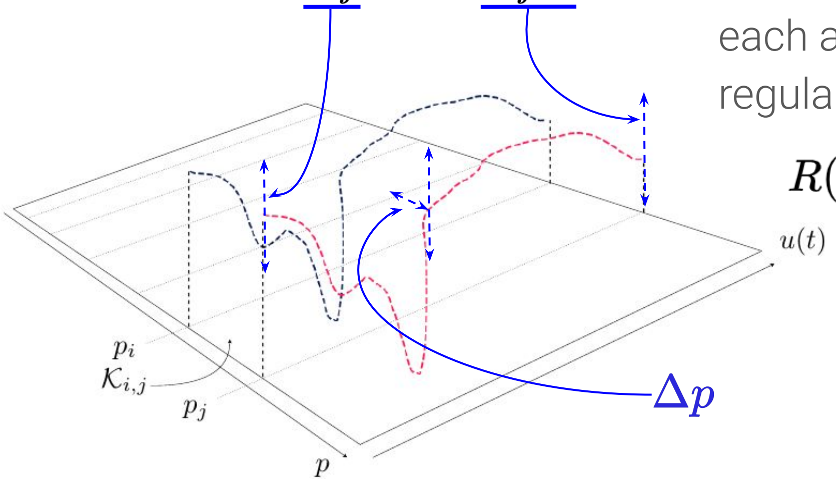
p_i Percentage of additional Calamine oxide

Laidler, Keith J. Journal of chemical Education 61.6 (1984): 494.
Lasaga, Antonio C. Rev. Mineral. 8 (1981).

Kinetic-Based Regularization

$$f^* = \arg\min_{f \in \mathcal{F}} \mathcal{L}(f) + \lambda R(f)$$

$$\mathbf{b}_j = [\Delta_j^{t_1}, \ldots, \Delta_j^{t_{max}}]$$



Using the neighboring points $p_i + \Delta p, p_i + 2\Delta p, p_i + 3\Delta p$ we derive a series of penalty bounds $\mathbf{b}_j = [\Delta_j^{t_1}, \ldots, \Delta_j^{t_{max}}]$ at each applied temperature $t_1, \ldots, t_{max}$. The regularization-like term becomes

$$R(f) = \frac{1}{P} \sum_{j=1}^{P} \mathbf{1}\{|f(p_i + j\Delta p) - \mathbf{b}_j| > \epsilon\}$$

Boyd, Stephen, Stephen P. Boyd, and Lieven Vandenberghe. Cambridge university press, 2004.
Ravi, Sathya N., et al. *Proceedings of the AAAI Conference on Artificial Intelligence*. Vol. 33. 2019.

Finding Pareto-Optimal Solutions

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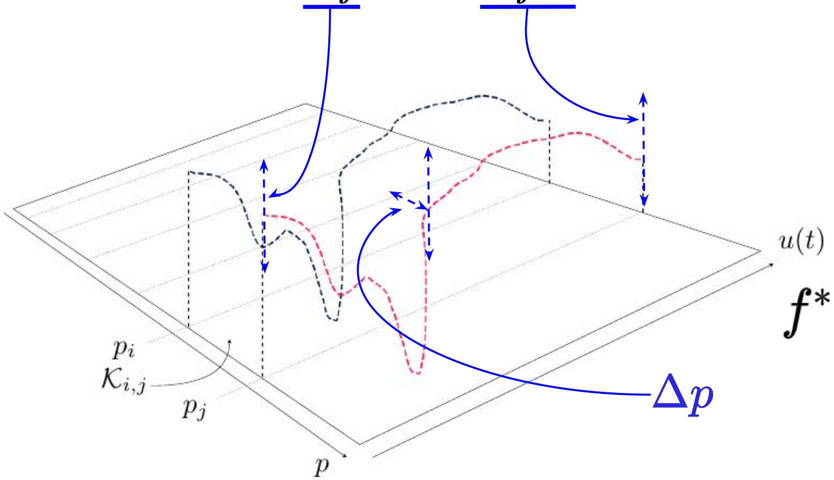

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Stochastic Gradient Descent

$$f^* = \arg\min_{f \in \mathcal{F}} \mathcal{L}(f) \text{ s.t. } R(f) \leq \mu$$

Conditional Gradient

Boyd, Stephen, Stephen P. Boyd, and Lieven Vandenberghe. Cambridge university press, 2004.
Ravi, Sathya N., et al. *Proceedings of the AAAI Conference on Artificial Intelligence*. Vol. 33. 2019.

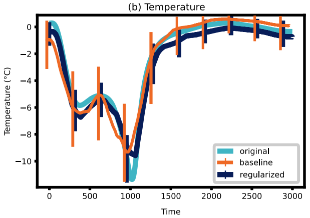
Experiments

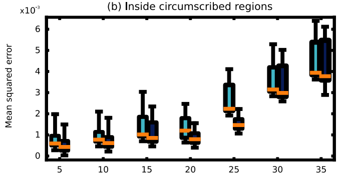
Experimental Setup

- Dataset
    - SDT-Q600 from TA-instruments version 20.9 build 20;
    - Monitored signals: temperature (°C), weight (mg), heat flow (mW), temperature difference(μV), sample purge flow (mL/min), etc.;
    - 3000 measurement points at a sampling rate of 2 Hz;
    - Real experiments conducted at 5, 10, 15, 20, 25, and 35 % of additional calamine oxide
- Training details
    - Stacking of Conv1d/ReLU/MaxPool blocks (Tensorflow);
    - Hyperparameter optimization (scikit-optimize/Microsoft NNI);
    - Kinetics regularization-like terms derived analytically (chempy)

Experimental Evaluation

(i) Reconstruction Process





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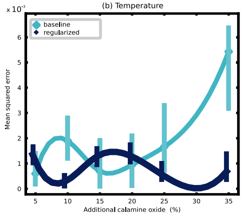

(iii) Reconstruction at specific percentages

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Eyring (E)	0.57 ± .0145 (10)	0.385 ± .0031 (5)	**0.228** ± **.0079** (10)	0.587 ± .0037 (**20**)
pig (P)	2.408 ± .0034 (10)	**0.408** ± **.015** (5)	1.188 ± .0061 (5)	2.408 ± .0042 (10)
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A+E+P+C	**0.192** ± **.0056** (15)	0.201 ± .0122 (5)	0.247 ± .0032 (10)	0.231 ± .0143 (15)

(iv) Trade-off between real experiments and analytical models

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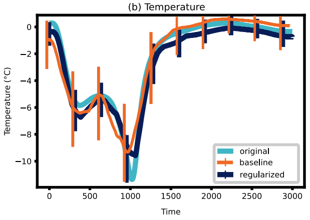

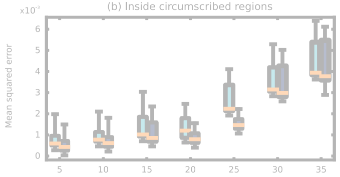

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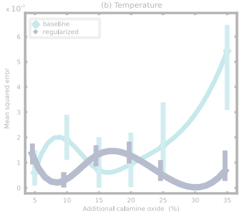

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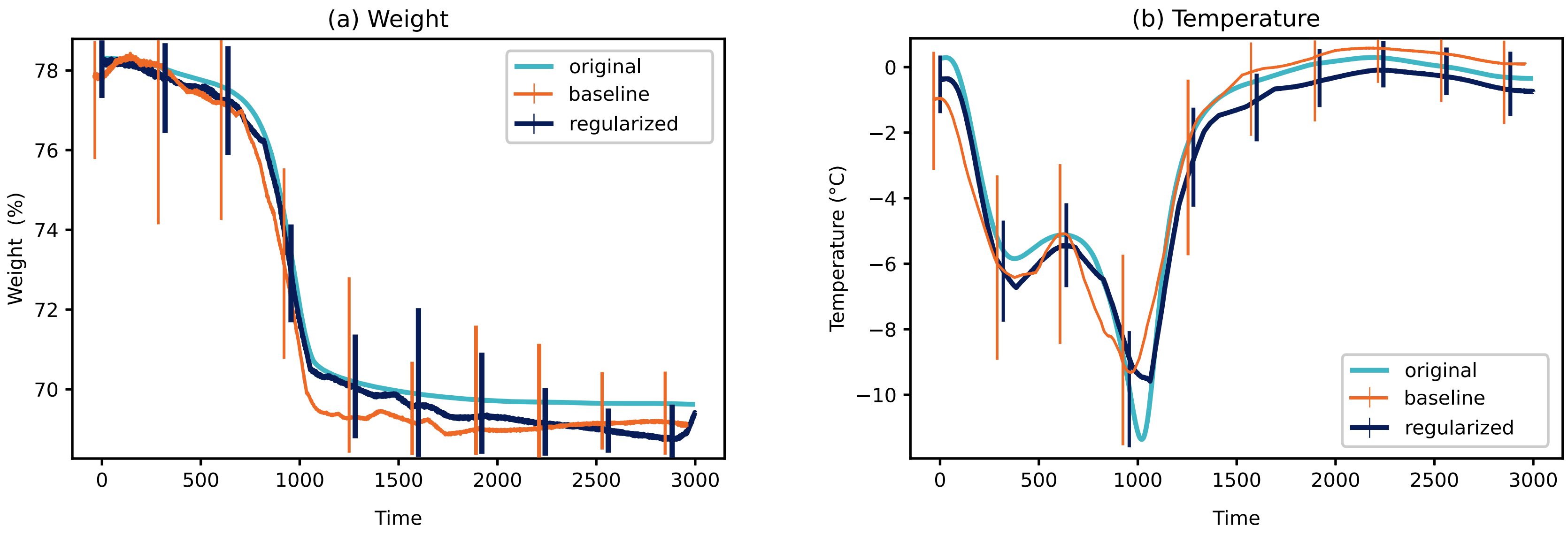

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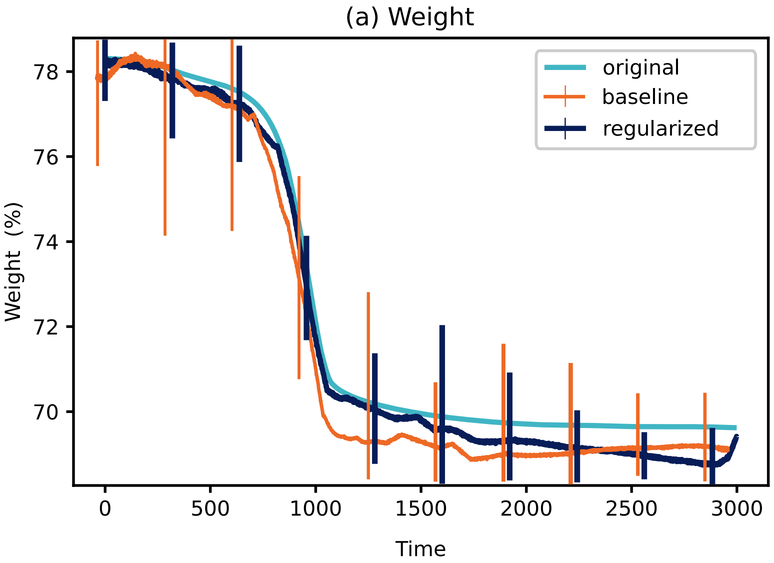

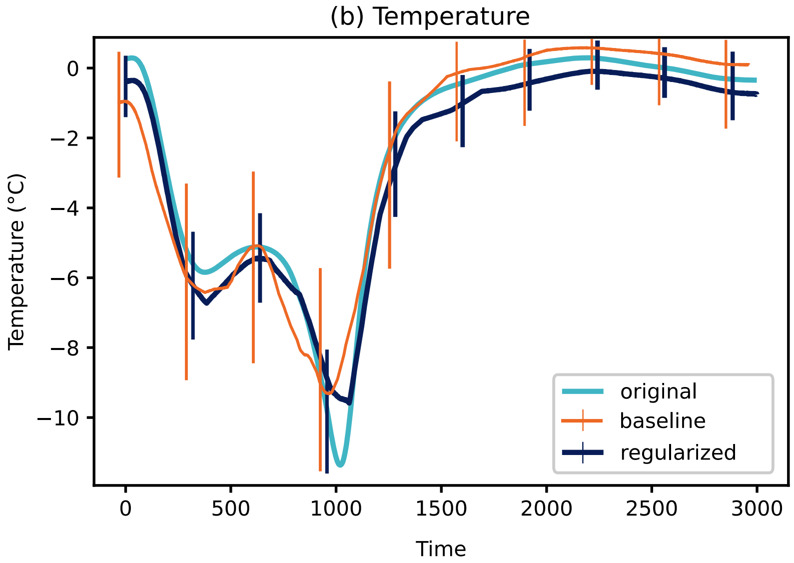

2.76±0.09 vs 3.29±0.15

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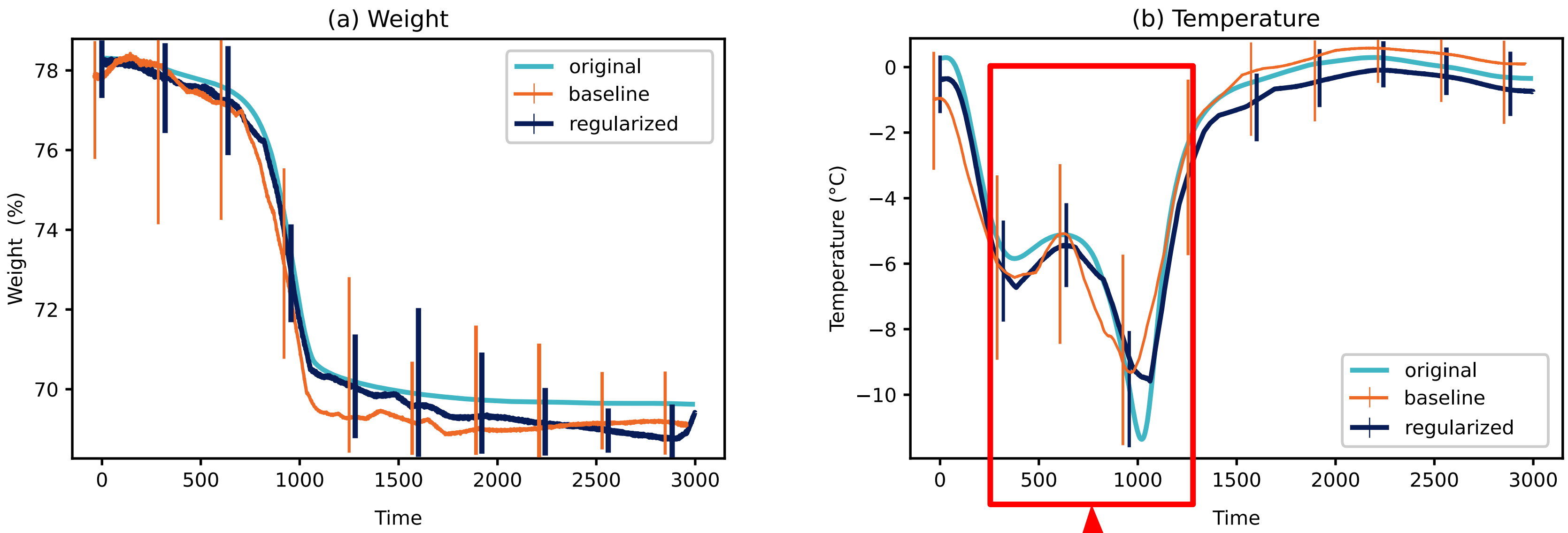

2.76±0.09 vs 3.29±0.15

Phase transitions between ~ 250°C and 1250°C

Experimental Evaluation

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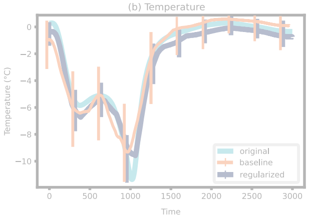

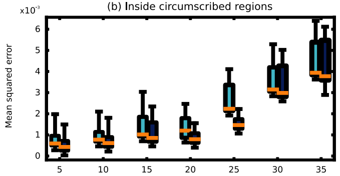

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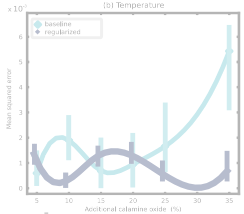

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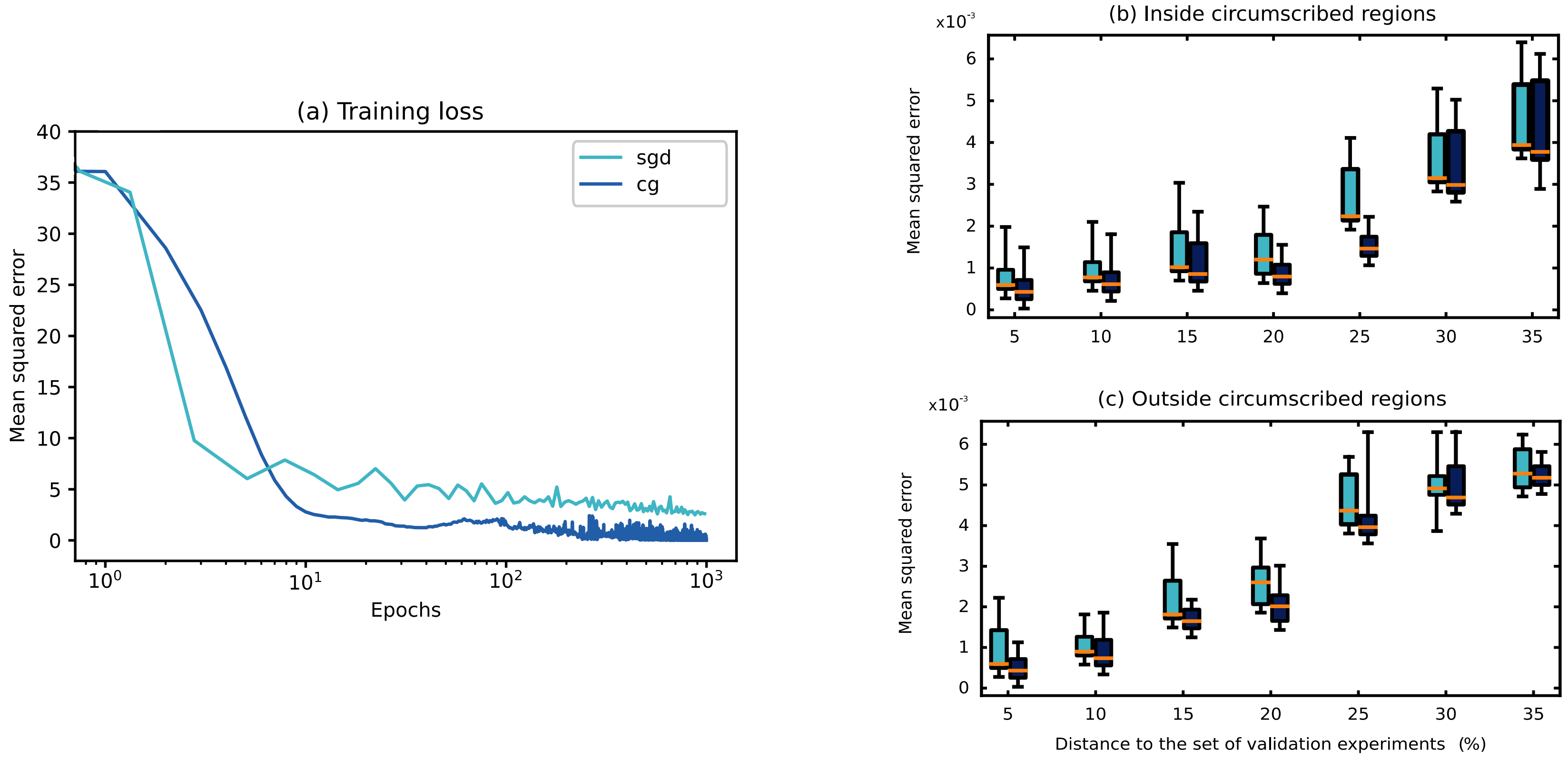

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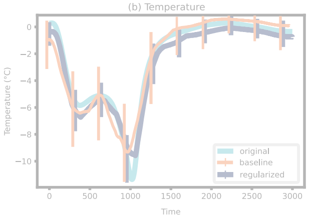

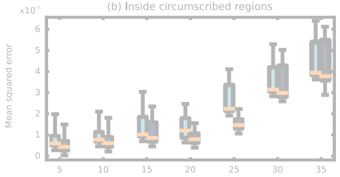

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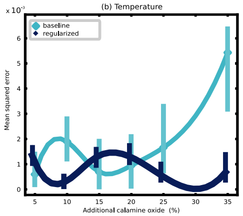

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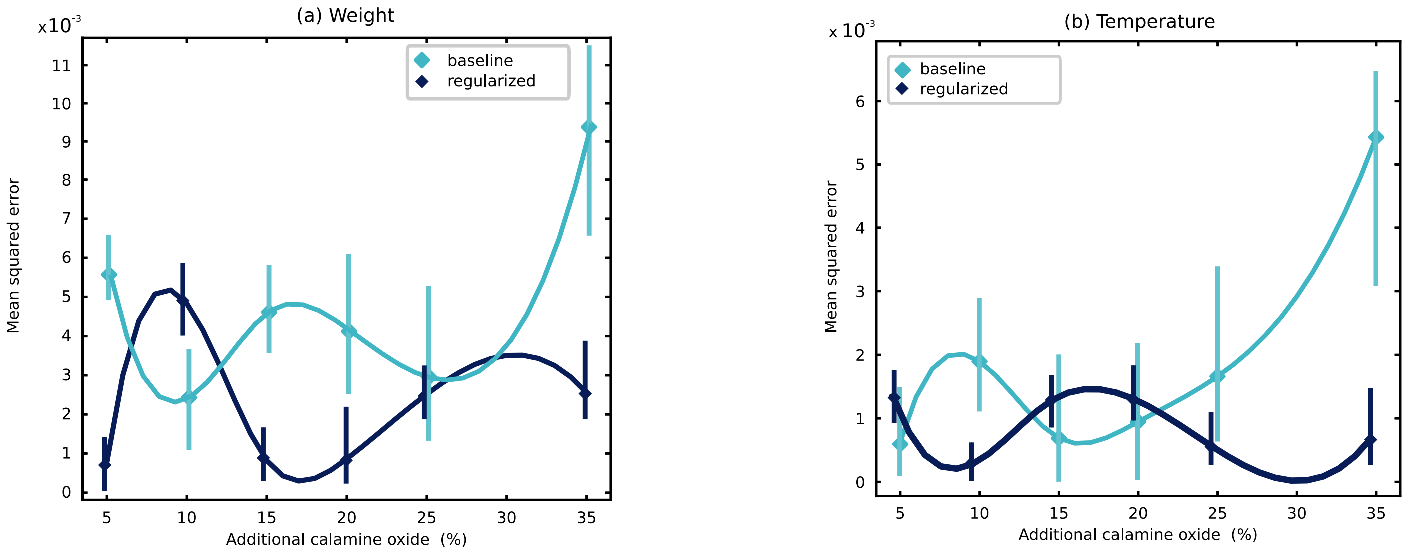

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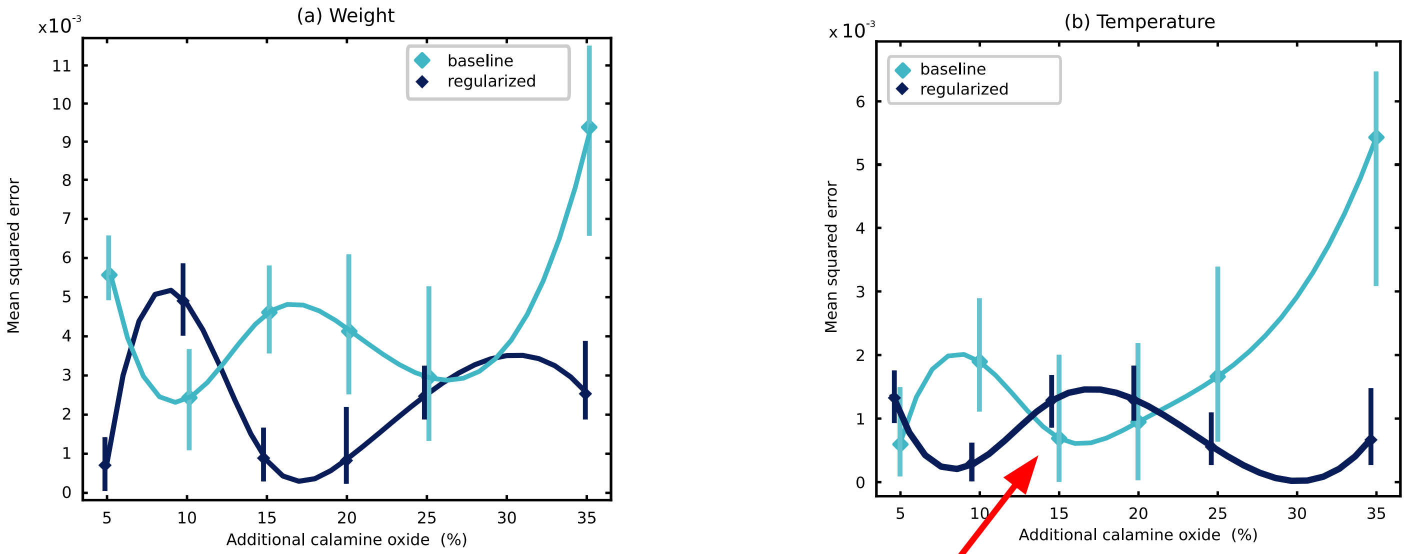
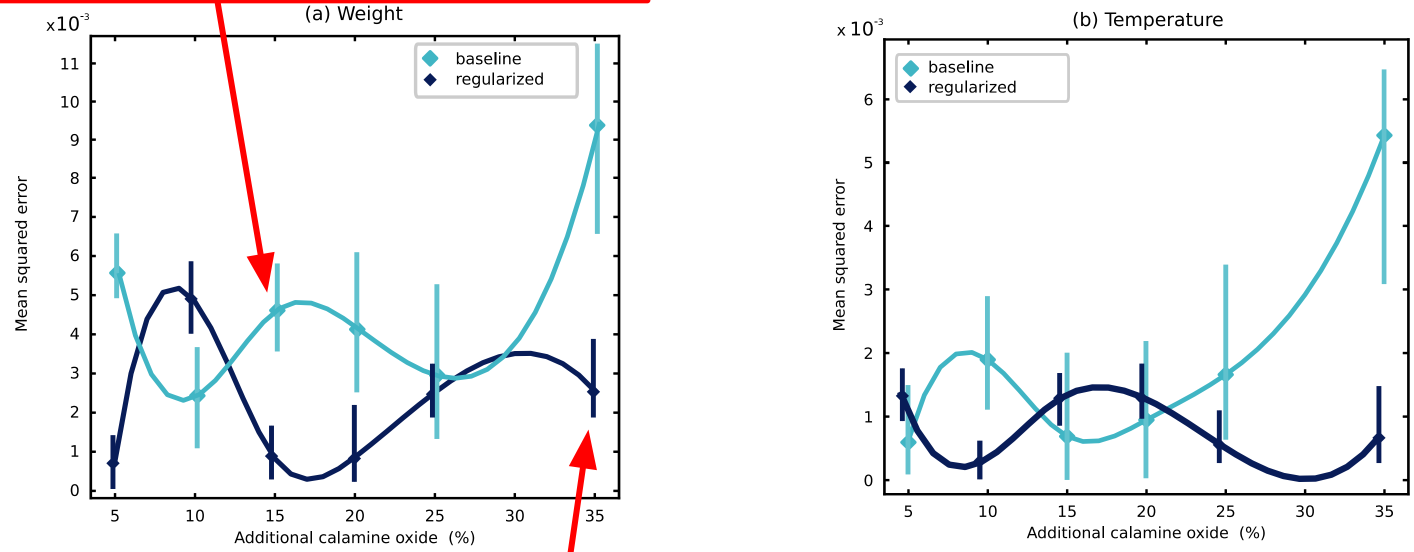


0.00192±.00081 vs. 0.0076±.0023

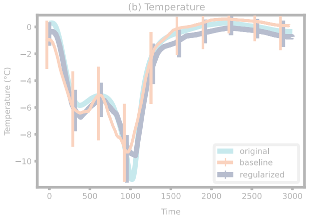
(iii) Reconstruction at Specific Percentages

0.00087±.00122 vs. 0.00477±.0021



0.00246±.002 vs. 0.00932±.0056

Experimental Evaluation



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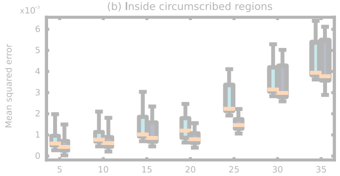

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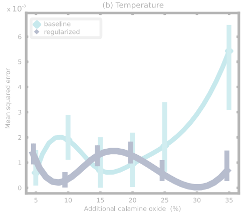

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pig (P)	2.408 ± .0034 (10)	**0.408 ± .015** (5)	1.188 ± .0061 (5)	2.408 ± .0042 (10)
cala (C)	0.533 ± .0112 (15)	0.512 ± .0055 (**20**)	0.524 ± .0047 (5)	**0.504 ± .0125** (10)
A+E	**0.188 ± .0058** (5)	0.197 ± .0079 (**20**)	0.214 ± .0051 (10)	0.204 ± .0147 (15)
P+C	0.318 ± .0012 (5)	**0.289 ± .0044** (10)	0.309 ± .0108 (10)	0.320 ± .0086 (10)
A+E+P+C	**0.192 ± .0056** (15)	0.201 ± .0122 (5)	0.247 ± .0032 (10)	0.231 ± .0143 (15)

(iv) Real Experiments & Richness of Domain Models

Analytical model(s)	Reconstruction error avg.$\pm$std. $\times 10^{-2}$ (best extent %)			
	$\lambda = 0.001$	$\lambda = 0.01$	$\lambda = 0.1$	$\lambda = 1$
Arrhenius (A)	$0.933 \pm .0073$ (5)	$0.988 \pm .0023$ (15)	$\mathbf{0.39} \pm \mathbf{.0157}$ (15)	$0.776 \pm .0027$ (5)
Eyring (E)	$0.57 \pm .0145$ (10)	$0.385 \pm .0031$ (5)	$\mathbf{0.228} \pm \mathbf{.0079}$ (10)	$0.587 \pm .0037$ (**20**)
pig (P)	$2.408 \pm .0034$ (10)	$\mathbf{0.408} \pm \mathbf{.015}$ (5)	$1.188 \pm .0061$ (5)	$2.408 \pm .0042$ (10)
cala (C)	$0.533 \pm .0112$ (15)	$0.512 \pm .0055$ (**20**)	$0.524 \pm .0047$ (5)	$\mathbf{0.504} \pm \mathbf{.0125}$ (10)
A+E	$\mathbf{0.188} \pm \mathbf{.0058}$ (5)	$0.197 \pm .0079$ (**20**)	$0.214 \pm .0051$ (10)	$0.204 \pm .0147$ (15)
P+C	$0.318 \pm .0012$ (5)	$\mathbf{0.289} \pm \mathbf{.0044}$ (10)	$0.309 \pm .0108$ (10)	$0.320 \pm .0086$ (10)
A+E+P+C	$\mathbf{0.192} \pm \mathbf{.0056}$ (15)	$0.201 \pm .0122$ (5)	$0.247 \pm .0032$ (10)	$0.231 \pm .0143$ (15)

Summary

- Evaluation of a real-world application of material engineering;
- Incorporation of domain analytical models via regularization-like terms;
- Converge to Pareto-optimal solutions using conditional gradient descent;
- Extensive experimental analysis reveal remarkable efficiency improvement;

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