Augmented Experiments in Material Engineering Using Machine Learning

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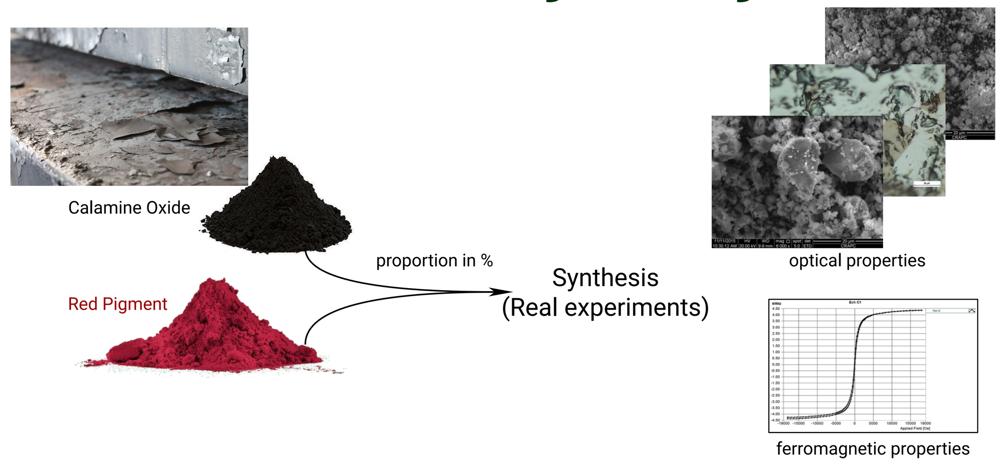
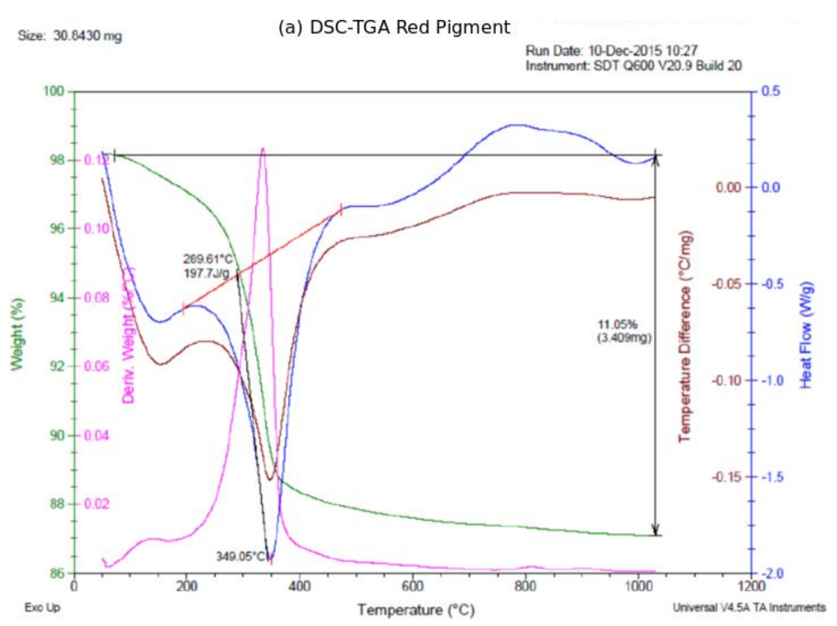
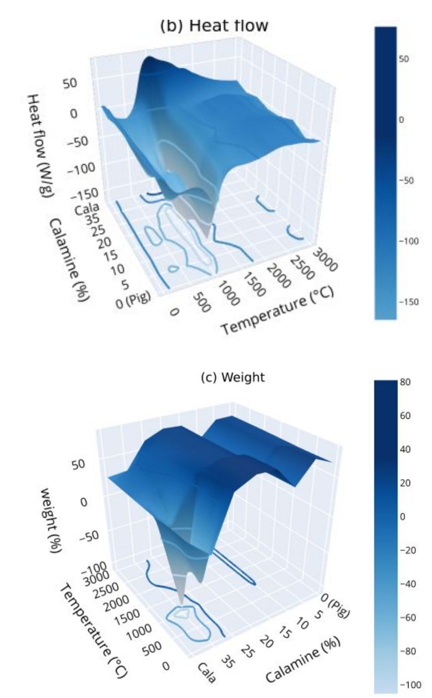
Synthesizing new material with desired properties in an industrial context requires several costly experiments. Our goal is to reduce the need to perform real experiments using a machine learning approach circumscribed by analytical models.



Synthesis of New Materials in Industry — Mixtures of Red Pigment and Calamine Oxide

Red Pigment is a natural form of mineral composed mainly of iron oxide;
Calamine Oxide is a steel by-product obtained during continuous casting or heating of slabs and billets;
The synthesis of new materials is obtained by the contribution of the calamine in this process by ensuring a sufficient quantity of Fe_2O_3 and increasing the density of the synthesized pigment. The goal is to get materials with some desirable qualitative properties including optical properties, ferromagnetic properties, etc.

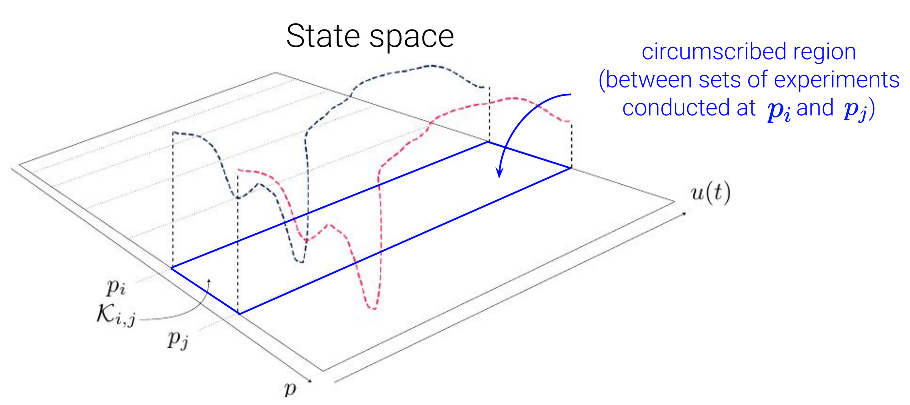




Dataset Description

- Dataset consists of thermal analysis of raw materials collected with an SDT-Q600 version 20.9 build 20 industrial instrument that monitors the calcination of the mixtures;
- Various signals are monitored by the instrument, including, temperature (℃), weight (mg), heat flow (mW), temperature difference (μV), sample purge flow (mL/min), etc.;
- In addition to the theoretical curves of the red pigment (pig) and the calamine oxide ($cala$) that were obtained separately, we perform calcination of mixtures with various percentages, $p_i \in \{5, 10, 15, 20, 25, 35\}$, of additional calamine oxide to the red pigment.

State Space Partitioning and Evaluation Protocol



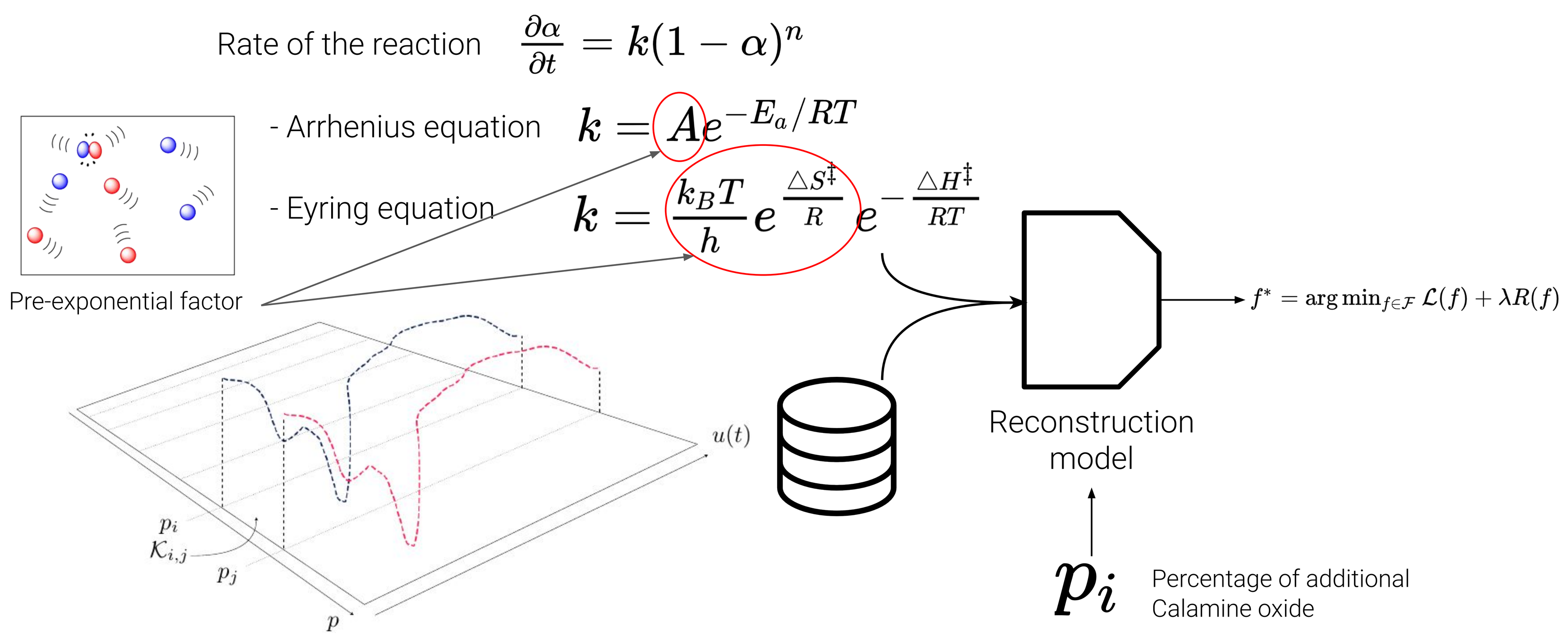
- The set of all real experiments obtained for every mixture percentage and every heating temperature input form what we refer to as the state space;
- Using the sequences of real experiments ordered by the percentage of additional calamine oxide in the mixture, we divide the state space into contiguous partitions;
- As a result, we consider two kinds of machine learning models: models that approximate experiments (Valid.) in the partition circumscribed by two given sets of real experiments (Train.); models that approximate experiments for regions located beyond a fixed partition.

Combining Analytical Models and Real Experiments

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

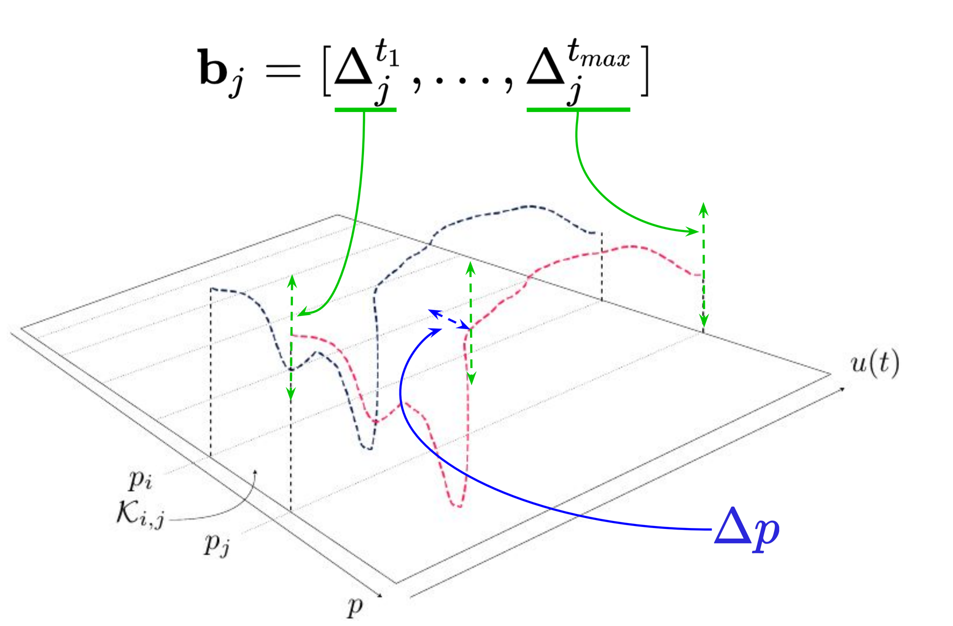
- Arrhenius equation $k = Ae^{-E_a/RT}$

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



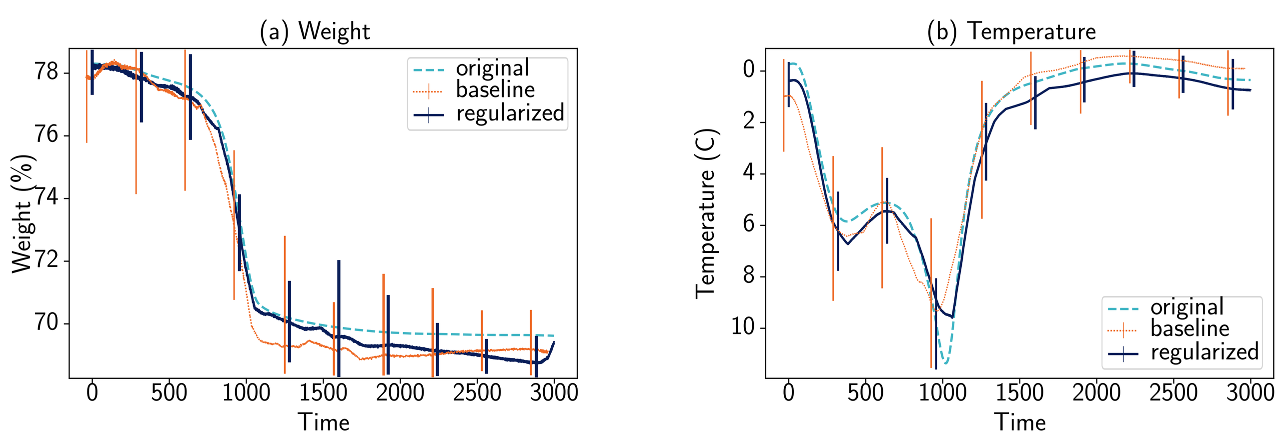
Kinetic-Based Regularization

- Given a set of experiments, 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 $R(f) = \frac{1}{P}\sum_{j=1}^{P} \mathbb{1}\{|f(p_i + j\Delta p) - \mathbf{b}_j| > \epsilon\}$. This additional term provides a necessary constraint, which our model must satisfy;
- Finding solutions satisfying both terms of the objective function, i.e. Pareto-optimal solutions, is not ensured using gradient descent. We ensure the fulfillment of the derived constraints using conditional gradient descent.



(1) Reconstruction Performances

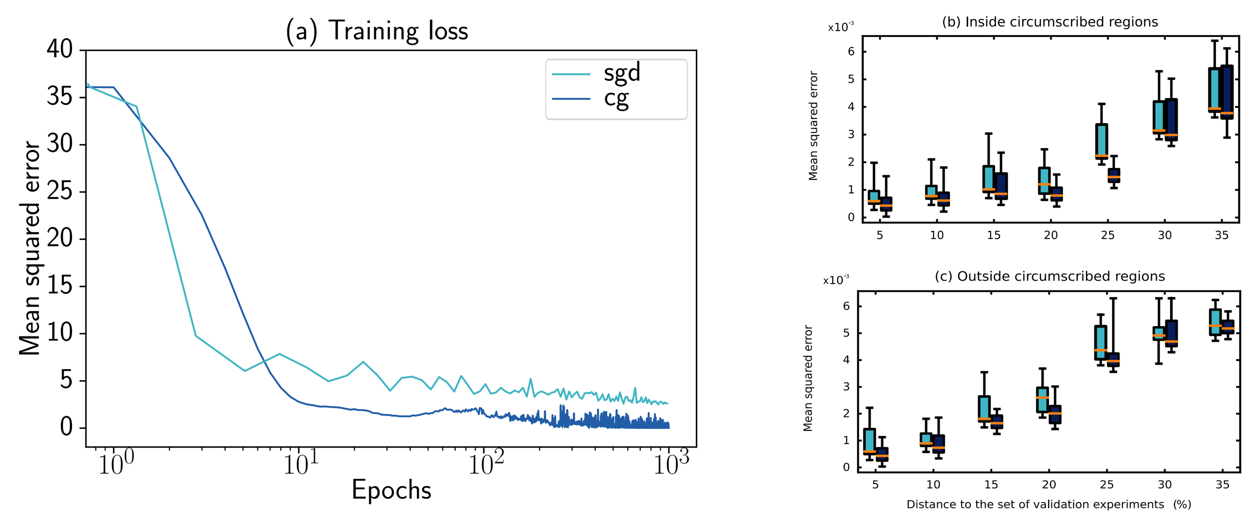
- We evaluate the reconstructions obtained using different configurations of the real experiments based on the setting described in the state space partitioning and evaluation protocol;
- We report reconstructions averaged over all evaluation setups and their corresponding perplexity. As references, we also report the reconstructions obtained (under the same evaluation setups) using the baseline.



Our approach contributes to a substantial reduction of this perplexity (e.g. 2.76±0.09 vs 3.29±0.15 for weight). The perplexity here can be related to 2 factors: the spacing of the real experiments; and the presence of phase transitions in some ranges.

(2) Distance between Train. and Valid. Experiments

- We measure the extent of reconstructions, i.e., how a model can satisfactorily reconstruct the state variables as a function of the distance between the real experiments it was trained on and the target mixture percentage that has to be reconstructed;
- These figures illustrate also the impact of using Conditional Gradient on the fulfillment of the constraints that are imposed on the models.

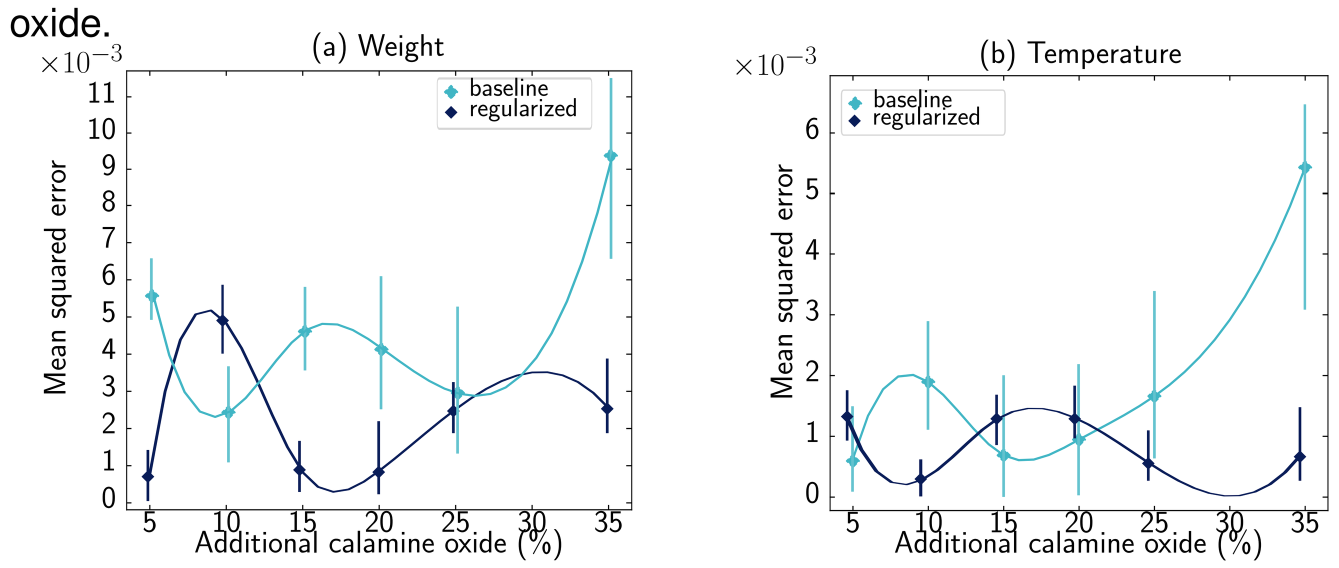


We can see a noticeable effect of Conditional Gradient on the reconstruction performances up to an extent of 25% inside circumscribed regions and 30% outside circumscribed regions.

This translates the ability of Conditional Gradient to converge towards solutions that take into account the regularization terms whereas Stochastic Gradient Descent tends to push towards solely satisfying the first term of the cost function at the expense of providing constrained reconstructions.

(3) Reconstruction at Specific Percentages

We further investigate the extent of reconstructions at specific percentages of additional calamine oxide.



- Some percentages, for example, reconstructions of temperature curves at 15%, no matter how far apart the set of training experiments are, the reconstructions are satisfactory with or without the addition of analytical models;
- On the other hand, for 35% for example, the reconstruction errors are greater using the baseline model. In this case, our approach is able to significantly improve upon the baseline model and over all percentages both in terms of approximation and perplexity

Incorporated analytical models contribute substantially to reducing the accompanied perplexity.