Using Time-Series Forecasting to Accelerate Materials Stability Assessments

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1. Introduction

1.1 Background

The stability of materials and devices is a crucial aspect of materials research. In numerous instances, material degradation occurs gradually and is nearly imperceptible, however, it is still important for continuous functionality. Accelerated ageing methodologies are essential to ascertain the longevity of a material under external stressors, including heat, light, and UV radiation. For example, optoelectronic devices, such as solar cells are expected to last for decades[1] to make them feasible as low-cost, green energy. In our group, we address challenges of this type, by integrating combinatorial materials testing with time-series forecasting models. Through these efforts, we aim to develop improved materials faster, while also furthering the understanding of their underlying properties.

1.2 Related work

Our work connects two major research areas of acceleration: High-throughput experimentation and time-series forecasting (TSF). High-throughput, combinatorial experimentation has been used for almost a century by now. Nowadays, a lot of work is focused on implementing machine learning techniques, such as Bayesian optimization[2] in order to find desired material properties in a vast compositional space faster. In many reports the focus is optimization rather than exploration.

TSF is well-established in fields, such as economics or weather forecasting. In materials science, a trade-off between generalizability and interpretability of approaches must be made. TSF has been used in material degradation studies for example in the fields of batteries[3–5] and photovoltaics[6,7]. These either rely on unimodal[6,7] or multimodal[3–5] datasets for their predictions. Transformer-based models have been used as well[5] with accurate predictions, but low generalisability and interpretability of the models. To address these challenges both a priori[4] and post-hoc[6] imbedding of domain knowledge, as well as the use of exergonic parameters[7] have been reported. Still there is a need to explore TSF for materials science in more detail.

2. Methodology

In our work[8] we address these challenges with three major novelties:

1) We combine high-throughput materials testing with TSF
2) We introduce interpretability into the model, by using a physics-informed featurization of the time-series data
3) We investigate to which extend our approach can be generalized to other chemistries or microstructures.

We studied metal halide perovskites (MHPs), which are a promising class of materials for both photovoltaic and light-emission applications. Within the MHP material class, we studied 4 significantly different groups to cover a wide range of potential optoelectronic applications: inorganic and organic-inorganic thin films for photovoltaic and detector applications, as well as inorganic and organic-inorganic nanocrystals for light-emission applications (here: inorg. TF, hybr. TF, inorg. NC, hybr. NC). All samples where stressed with a white light LED (100 mW cm^{-2}) and kept at 85 °C, while we tracked the photoluminescence (PL) automatically in a custom-built high-throughput setup[9]. From the PL data we extracted physically meaningful features, by fitting them with a model that includes a high-energy peak (HE peak) and a low-energy tail (LE tail). A representative set of PL spectra together with the fitting curves is shown in **Fig. 1A**.

In total, we extracted 20 features, making our approach multi-modal. We use all features as an input for the TSF, while only predicting the normalized PL area (Norm_Area) as a proxy for material quality. Our TSF approach is based on convolutional neural networks (CNN) and long-short-term-memory (LSTM) architecture. The data from all material groups was first split 8:2 into a training and test set. Then the training set was again split 8:2 into the training and validation set. We kept all hyperparameters fixed, except for the input time interval (in hours) and the time difference of the last input value and a discrete output value. To show some initial results, we focus here on the model with an input interval of 48 h and a predicted time point 72 h

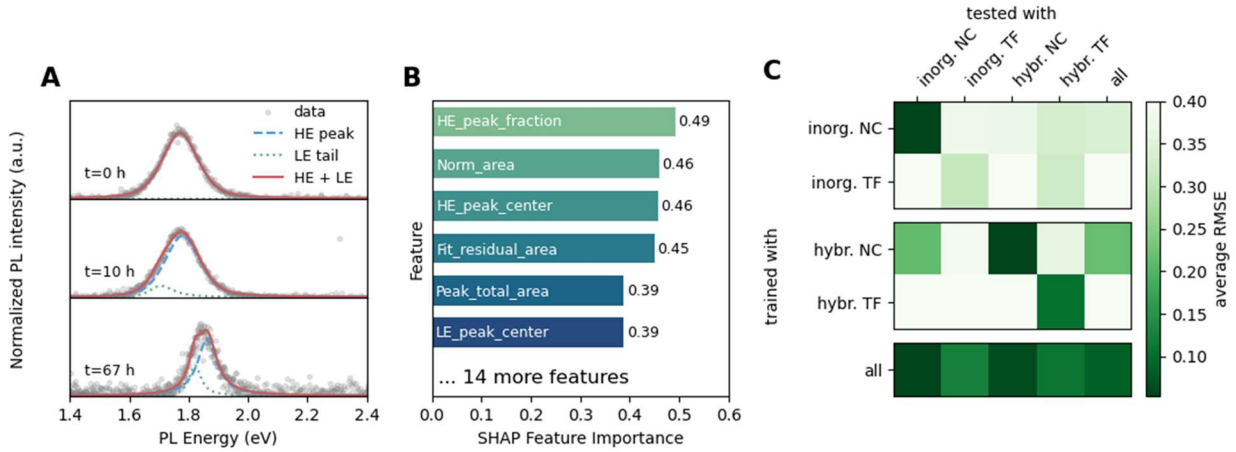

Fig. 1: **A** Results for automated segmentation of PL features into a high energy (HE) and low energy (LE) feature at various points in time. **B** SHAP feature importances for the six most important features of the CNN-LSTM time-series forecasting model. The depicted values were calculated from the mean of the input. **C** Average root-mean-square-error (RMSE) of the model trained and tested with subsets of the full dataset. The result is shown for the same 5 train/test splits.

after the end of the input. It is worth noting that we augmented our training data, with time-shifted data, so that input interval does not necessarily need to start at t = 0 for the predictions to be valid. We then repeated the training process for 5 random train/test splits.

3. Results

3.1 Model Performance

At first, we test the viability of our approach. From the 5 train/test splits we find an average root-mean-square-error (RMSE) for the test sets of 0.06. We compare this value to two benchmarks: first the naïve benchmark, where the predicted value is the last value of the input interval and a time-shuffle benchmark, where the values in the input interval are shuffled in time. We find average RMSEs of the same 5 test sets of 0.43 and 0.09 for the naïve and time-shuffle benchmarks, respectively. This gives us high confidence that our model used the temporal information for the predictions.

3.2 *Physics-Informed SHAP Analysis*

The use of the physics-informed featurization allowed us to use SHAP (SHapley Additive exPlanations) analysis in the end to close the loop and to further our understanding. In **Fig. 1B** we present the SHAP importance values for the first 6 features. We can see that features related to the HE peak seem to score quite high. Specifically, the HE_peak_center, the energy of the PL peak, can be seen as an encoder for both the chemistry and microstructure of the MHPs.

3.3 Investigating Learning Transfer

We thus investigated whether there was some learning transfer across different chemistries (inorg. /hybr.) or microstructures (NC/TF). For this, we repeated the model training with the individual datasets of a single material group and then tested them on all data sub-sets as well as the complete set (all). In **Fig. 1C** we show the resulting average RMSE from the 5 random train/test splits. We could confirm that training with all data would allow good predictions in all material groups. In contrast, when training with just a single material group we could not observe any significant learning transfer to the others. This result showcases that it is not guaranteed that a TSF model can predict the degradation behaviour of a new material based on the training on known materials. In fact, the current model will have to be trained materials with at least some similarity to the target material. It will be an important challenge in the future to address this limitation of TSF models for materials science.

4. Conclusions

We have demonstrated in this work, that TSF can be combined with combinatorial experimentation to yield both degradation predictions and insights into some underlying mechanisms. We believe that this type of approach will be beneficial to address future challenges in materials science and acceleration.

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