COMBINING PROCESS SHORT CUTS AND ARTIFICIAL NEURAL NETWORKS FOR PREDICTIVE LIFE CYCLE ASSESSMENT OF CHEMICALS

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Abstract Overview

The increasing awareness of climate change moves the focus of the chemical industry to aim not only for economics but also for ecologically optimal processes. However, the design of ecologically optimal processes requires an assessment of environmental impacts that can be applied in early process development stages. In early stages of process design, an environmental impact assessment is limited by the lack of data. In this work, we propose a fully predictive Life Cycle Assessment (LCA) framework using neural networks. Input data are only molecular properties from quantum mechanics calculations and process information available from pinch-based process models. The resulting framework is fully automatized including feature selection and hyperparameter optimization, and shown to be able to predict 17 environmental impact categories.

Keywords

Artificial neural networks, predictive life cycle assessment, early process development stage, comparison of pathway alternatives

Introduction

The assessment of environmental impacts is key for the chemical industry to meet their goals on climate protection. To assess environmental impacts, an accepted method is Life Cycle Assessment (LCA), which is an ISO-normed, holistic approach taking into account all phases of the life cycle (DIN EN ISO 14044: 2018-5). However, LCA requires detailed data on energy and mass balances, which is usually limited in early process development stage. Therefore, streamlined approaches have been proposed, that either reducing the scope or simplify the required data (Casamayor and Su 2012).

A promising streamlining approach uses machine learning techniques to predict environmental impacts based on the molecular structure of the product (Wernet et al. 2009, Song et al. 2017). Since this approach requires only a training set consisting of LCA results with already assessed chemicals and the molecular structure of the novel product to be predicted, this prediction approach is highly suitable to be integrated in early process development stages. However, using only the molecular structure as input, results in component-specific predictions. The approach is not able to distinguish between process alternatives leading to the same product.

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An alternative streamlining approach uses simplified process design methods to estimate energy demands (Righi et al. 2018). However, on the one hand, Righi et al. stated that building a preliminary process model requires knowledge and effort to estimate energy demands well, and on the other hand, the use of oversimplified models might produce significant discrepancies in the results. Thus, preliminary process models can generate data for LCAs in early development stage but require much more knowledge for sound results. However, the resulting LCA results can be used to compare process alternatives.

To combine the advantages of both streamlining approaches, we propose a fully automated framework to predict the LCA environmental impacts. The framework integrated pinch-based process models (Redepenning et al. 2017) for automated process design of chemical processes with artificial neural networks. Since the framework is fully automated, it can serve as screening tool for environmentally favourable process alternatives in the chemical industry.

Fully Automated Algorithm for Predictive Life Cycle Assessment

We propose an algorithm consisting of four steps:

1. Feature collection
2. Feature selection
3. Hyperparameter optimization
4. Impact prediction

In the first step, the algorithm requires two inputs from the user: the SMILES-Code of the desired product describing the molecular structure (Weininger 1988) and its reaction equation. Optional inputs are the reaction temperature and pressure, if these reaction conditions are already known. In a next step, thermodynamic properties of the product and the reaction mixture, such as the reaction enthalpy, are obtained from quantum mechanical calculations using geometries and frequencies from b3lyp/TVZP and energies from b2plyp/aug-cc-pVQZ level of theory. Pinch-based process models are used to calculate process-specific mixture properties such as the mole fraction in liquid-liquid equilibria. All features are passed to the feature selection step 2. In step 3, a genetic algorithm (GA) generates automatically a feedforward artificial neural network (ANN) for a given training set. In step 4, the optimized ANN is used to predict the environmental impact of the regarded process. The algorithm is implemented in Python 3.6 using Tensorflow and Deap.

Feature selection

We consider 218 possible features in total, consisting of molecular descriptors of the molecular structure and physical properties, and process descriptors obtained by the Douglas hierarchy and based on the process indicators proposed by Patel et al. (2012). Examples are the molecular weight as molecular descriptor or the mole fraction of the product in reactor equilibrium as process descriptor.

In Step 2 of our algorithm, we use recursive feature elimination with cross validation as feature selection algorithm. We compared several feature selection algorithms, including: principal component analysis, non-linear principal component analysis (utilizing kernel method), selecting the best singular features, recursive feature elimination, recursive feature elimination with cross validation, exhaustive search, sequential feature selection, sequential floating feature selection, sequential backward elimination, RReliefF, feature selection and feature selection using genetic algorithms (both as wrapper and embedded methods). The comparison has shown that recursive feature elimination with cross validation delivers the best performance in terms of minimum root mean squared error (RMSE) and maximum coefficient of determination (R_P^2), whilst requiring small computational time.

Due to limited LCA data availability for the training, the number of selected features is limited to 10 % of the number of training samples to avoid overfitting.

Hyperparameter optimization

In Step 3, the hyperparameters, including the number of hidden layers, the number of neurons in each hidden layer, and the regularization parameter are optimized using a genetic algorithm (GA). The objective of the GA the ratio between RMSE and R_P^2. We propose to minimize the ratio of the two error measures to minimize the absolute error (RMSE) as well as possible trends (R_2^P). Minimizing only the RMSE tends to lead to constant predictions of the average impact value, while minimizing only the coefficient of determination improves predictions of trends but increases absolute prediction errors.

To validate the proposed network architecture as well as the final prediction quality, the available training data is divided into 3 sets: (1) a training set, (2) a validation set and (3) a final test set. The training set contains 85 % of the available training data and is used to train the ANN in each GA loop. The validation set contains 10 % of the training samples and is used to quantify the generalization ability of the regarded set of hyperparameters in each GA loop. The calculated objective on the validation set is passed as feedback to the GA. The test set consists of 5 % of the training samples and is used to calculate the final objective value and to validate the overall performance of our algorithm. Due to the local nature of the GA, we use a multistart approach, running several GA instances in parallel. After the GA converges, the optimal architecture is used to estimate environmental impacts of the regarded process.

Case study: Component vs. Process-specific Networks

The proposed framework is applied to generate 17 neural networks, each predicting one of 17 Recipe v1.08 (H) Midpoint categories (Goedkoop et al., 2009). For training purposes, we obtain 220 training samples from the LCA database Ecoinvent (Wernet et al. 2016).

Here, we exemplify the framework for comparing the CO_2-based production of methanol and formic acid with their fossil-based productions, respectively (Figure 1). We consider two scenarios: "today", assuming hydrogen supply by steam methane reforming and European grid mix for electricity, and "future", where hydrogen is supplied by electrolysis and electricity is produced by wind power. Carbon dioxide is the carbon source for the CO_2-based processes and captured from an ammonia plant in both scenarios. The fossil-based processes utilize synthesis gas as carbon source. The results are compared to the results obtained with a component-specific ANN and with literature.

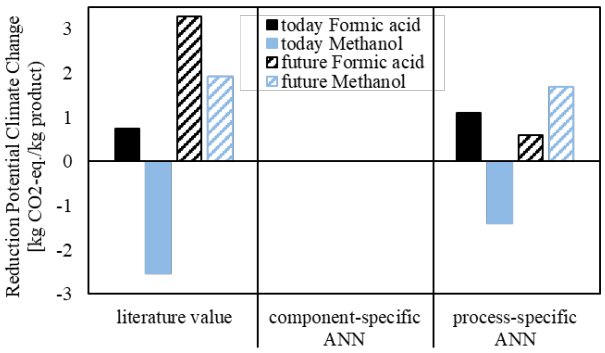

Figure 1. Comparison of the prediction quality of climate change impacts by the proposed process-specific ANN and a component-specific ANN to literature values.

In literature (Artz et al. 2018), the CO_2-based production of formic acid is environmentally beneficial in comparison to the fossil-based production for both scenarios, indicated by the positive sign of the reduction potential. In contrast, the CO_2-based production of methanol is environmentally worse in the today-scenario but becomes promising in the future as indicated by the change of sign in the reduction potential. The component-specific ANN is not able to distinguish between production alternatives and thus cannot be used to predict a reduction potential due to changing the production process.

In contrast, the process-specific ANN can resolve process alternatives and correctly predicts a positive reduction potential for CO_2-based formic acid in both scenarios. The CO_2-based methanol is correctly predicted to have a negative reduction potential in today turning into a positive reduction potential in the future-scenario. Although the absolute potentials are not predicted correctly, the process-specific ANN is able to predict the sign for each comparison correctly and thus can be used for a first screening in early process development stages.

Conclusion

A fully automatized framework is presented to predict process-specific life-cycle assessments by combining simplified process design with artificial neural networks. The framework is exemplified in a case study comparing CO_2-based production to fossil-based production of both methanol and formic acid. The results show that the proposed ANN is able to predict whether a technology change has the potential to reduce climate change impacts. Thus, the proposed framework can be integrated as a screening tool in early process development stages.

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