Determination of the Liquidus Temperature of Cryolitic Baths by Applying different Machine Learning Approaches

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Introduction

- **Primary Aluminum production is based on the Hall-Herroult process,** in which alumina is dissolved into a molten cryolite bath. AlF_3, CaF_2, Al_2O_3, KF, MgF_2 and LiF are constituents of the bath, except for cryolite and the liquidus temperature is affected by the presence of the compounds that can be calculated by numerous equations [1,2].
- **Thermodynamic calculations of ionic liquids,** can be obtained using the Factsage7.0 software. Properties such as density, viscosity and mainly, the liquidus point of the cryolitic bath are estimated by the Modified Quasichemical Model with the Quadruplet Approximation[3,4,5].
- In this study, a liquidus temperature equation emerged through machine learning approaches with a huge collection of thermodynamically data based on the cryolite bath that is used in the primary Aluminum production, using Greek bauxite ores.

Methodology

Three different machine learning (ML) methods were compared regarding their capability to predict the liquidus temperature of the cryolitic bath, in respect to its composition based on the seven different constituents. For the development of ML models, a large dataset was created based only on thermodynamic calculations carried out by means of Factsage software for a comprehensive amount of different bath compositions (2860 data samples in total) within a reasonable range. Apart from training and testing the ML models in order to determine their accuracy and generalization strength, an additional dataset (148 data samples in total) was created in order to evaluate their capability for generalization for composition values beyond the defined range (extrapolation).

The first model which was developed was relevant to Multiple Linear Regression (MLR). Apart from the linear terms, interaction terms were also added for AlF_3, CaF_2, Al_2O_3, LiF and KF based on experience and suggestions from the relevant literature. Moreover, statistical analysis by means of Analysis of Variance (ANOVA) method was performed afterwards to remove statistically insignificant terms. This model allows for better interpretability as it produces an explicit equation correlating the composition of the bath and liquidus temperature and is simple and fast to use. The other models which were developed were a Support Vector Regression (SVR) and a Multilayer Perceptron (MLP) model. In contrast to MLR, these models are considered as “black-boxes”, as they provide the predicted values without yielding equations in explicit form. All models were evaluated based on R2, Root mean squared error (RMSE) and Mean absolute percentage error (MAPE).

Results

- The different models for the prediction of liquidus temperature of the cryolitic bath (Tliq) were compared based on their performance regarding both datasets.
  - Tliq(℃) = 1018.0 - 1.277 $xAlF_3$ - 1.440 $xCaF_2$ - 4.441 xAl_2O_3 - 9.256 xLiF - 6.405 $xMgF_2$ - 0.2562 $xAlF_3$*$xCaF_2$ - 0.3089 $xAlF_3$*xAl_2O_3 - 0.6099 $xAlF_3$*xKF + 0.2541 $xCaF_2$*xAl_2O_3 + 1.190 $xCaF_2$*xKF + 0.798 xAl_2O_3*xKF - 2.319 xLiF*xKF,

with an R^2 value of 94.14%, R^2-adjusted value of 94.11% and R^2-predicted value of 93.53%, whereas RMSE and MAPE values were 5.814 ℃ and 0.574%, respectively. The values of these metrics indicate a sufficient level of accuracy for the model.

- The MLP model which exhibited the best performance included 60 hidden nodes in a single hidden layer. The MLP model with the highest performance exhibited the following values for the performance metrics: R^2(train): 98.23%, R^2(test): 96.55%, RMSE(train): 1.249 ℃, RMSE (test): 1.408 ℃, MAPE(train): 0.043%, MAPE(test): 0.045%.
- The optimum SVR model with RBF kernel and hyper-parameters values C: 85.0 and ε 0.5 exhibited the following values for the performance metrics: R2(train): 94.75%, R2(test): 93.92%, RMSE (train): 1.557 ℃, RMSE (test): 1.881 ℃, MAPE(train): 0.04%, MAPE(test): 0.05%.
- From these results, depicted also in Figure 1, it can be deduced that the MLP model had superior predictive ability as it achieved the highest R^2 and lowest RMSE and MAPE values (especially for the test dataset, which indicates high generalization capability). The SVR model and the MLR model followed with relatively lower values in most metrics.
- Finally, regarding the comparison between the performance of the three models on the additional dataset, used for evaluating the extrapolation capability of the models, the MLR model had RMSE value of 15.212 ℃ and MAPE value of 1.467%, whereas the MLP model had RMSE value of 48.816 ℃ and MAPE of 3.785% and the SVR model had RMSE value of 59.500 ℃ and MAPE of 6.32%. These results, depicted also in Figure 2, clearly indicate that, while the black box models are superior for predicting the Tliq values between the composition ranges, when they are used to predict Tliq for compositions beyond this range, their performance is considerably inferior to that of the MLR model, which showed an average deviation of 15oC in these cases.

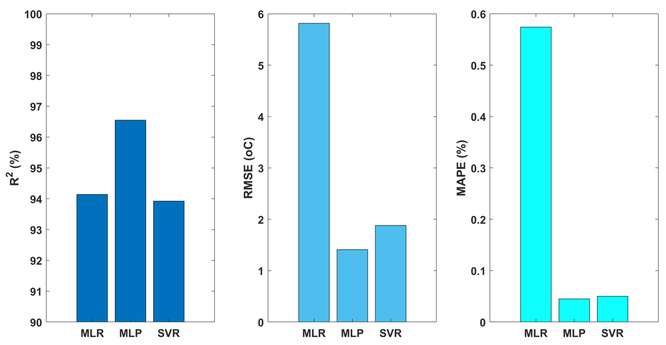

Fig. 1 Values of R^2, RMSE and MAPE performance metrics of the three models.

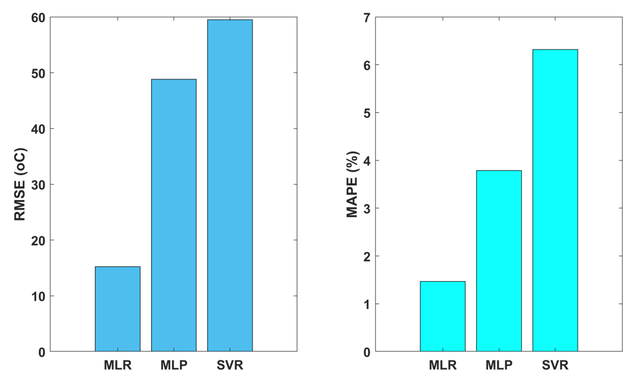

Fig. 2 Value of RMSE and MAPE performance metrics of the three models (additional dataset).

Conclusions

- In this work, three different ML models were compared in the case of cryolitic bath liquidus temperature prediction. The models were trained based only on data from thermodynamic calculations and were evaluated also based on different metrics.
- It was found that all models are capable of predicting the liquidus temperature with an average deviation of around 2 ℃ for a wide range of cryolitic bath composition values, with the MLP model exhibiting the highest performance than the SVR and MLR models. However, for cases with composition values outside the usual ranges, only the MLR model can provide an acceptable estimation of Tliq, with an average deviation of around 15 ℃. Thus, although the black-box models can be more accurate for the prediction of Tliq within the usual composition ranges, the MLR model, which provides an explicit equation, can be useful in practical applications when cryolitic bath composition is out of range in order to achieve a decent approximation of Tliq in this case.

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