A machine learning model to predict non-metallic inclusion dissolution in the metallurgical slag

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ABSTRACT

Dissolution of non-metallic inclusions in the metallurgical slag is of vital importance for cleanliness control of the steel manufacturing. With the development of high temperature confocal laser scanning microscope (HT-CLSM), new insights have been obtained due to its in-situ observation characteristics, higher resolution, and precise control. However, HT-CLSM measurement has the limitation, e.g. the slag composition cannot include high amount of transition metal oxides. In addition, it is time consuming for the experimental procedure and not so simple to succeed for every measurement. It is known that digitalization has made a significant progress in recent years. Machine learning (ML), a sub-domain of artificial intelligence (AI), is the key enabling technology for the digitalization of the material science and industry. The database for ML model is collected using almost all available HT-CLSM experimental data, and subsequently the established database is trained by different ML methods. Unseen data is used as the benchmark of the ML model. Al_2O_3 dissolution is the main process to be predicted in the current study. A good agreement between the HT-CLSM data and the ML model prediction results show the possibility to apply ML in process metallurgy.

INTRODUCTION

Inclusion (oxide) dissolution in the metallurgical slag is one of the key issues for the clean steel production. On the one hand, the main components in refractories, e.g. alumina, magnesia, etc. have the potency to be dissolved in the slag during contacting. Subsequently, the eroded refractory has a much lower chemical and physical stability at high temperature, and tends to be a source for exogenous inclusions formation in the steel. On the other hand, the formed inclusion particles tend to flow up to pass the steel/slag interface and need to be dissolved quickly in the slag (Park & Zhang; 2020; Park & Kang, 2017; Reis, et al., 2014; Webler and Pistorius, 2020). If the remained inclusions entrapped back into the liquid steel, they may have the risk to make agglomerations resulting in serious industrial problems e.g. nozzle clogging. In this case, the study of inclusion dissolution attracts the attention of metallurgists during the past decades. Finger rotating test (also named as dip test) is the conventional method to investigate the dissolution of oxide (refractory or synthetic inclusion) in the liquid slag. This method provides a good freedom of varying the experimental conditions (slag composition, temperature, atmosphere, etc.), however the sample size is much larger than the real inclusion so the obtained knowledge cannot connect to the real steelmaking process directly (Cooper Jr & Kingery, 1964; Aneziris et al., 2013).

With the development of the instrumentation in recent twenty-five years, high temperature confocal laser scanning microscope (HT-CLSM) offers a path to the in-situ observation of dissolution of inclusion particles in the steelmaking slag in real time. To track the particle dissolution, a quasi-three-dimensional images with sharp boundary and good contrast could be taken from the video by the CCD camera equipped with HT-CLSM. Specifically, the size of the used particle which ranges from a few tens to several hundreds of microns fits well with the actual inclusion size. Al_2O_3 dissolution in the slag is the most comprehensively studied by HT-CLSM, and the early work can refer to Sridhar and Cramb (2000). They investigated the effect of the temperature on the dissolution of alumina particles in the CaO-Al_2O_3-SiO_2-MgO (CASM) slag. Subsequently, comprehensive research activities focusing on the effect of inclusion type, slag composition, slag viscosity, temperature, etc. are performed. Specifically, 1630 °C is the maximum temperature which could be achieved so far to observe oxide dissolution (Liu et al., 2007). Due to the development of furnace in HT-CLSM, this maximum temperature could be further challenged. CASM slag without transition metal (Fe, Mn, etc.) oxides is the mainly selected system since the slag needs to be transparent to enable the in-situ observation, which is one main limitation for the application of HT-CLSM in this field. By utilizing the advantage of HT-CLSM, dissolution kinetics of different kinds of non-metallic inclusions besides Al_2O_3, i.e. MgO, $MgAl_2O_4$, TiO_2, Al_2TiO_5, $CaO\cdot 2Al_2O_3$ (CA2); SiO_2, SiC, TiN, etc. (Liu et al., 2007a,b; Miao et al., 2018; Michellic et al., 2016; Park et al., 2018; Ren et al., 2022; Sharma, et al., 2018; Yi et al., 2003) have been investigated. Except for the chemistry of inclusion, other processing parameters, e.g. dissolution temperature, particle size, slag composition, have been studied, however, there are still many parameters need to worth further investigated. For instance, the HT-CLSM can only observe the particle dissolution in the transparent slag, which means the influence of some component, e.g. Fe_tO cannot be studied quantitatively, especially the Fe_tO content in slag is high. In this case, the simulation and other technique needs to be performed to understand the

inclusion dissolution mechanism with the comprehensive conditions.

Regarding the study of dissolution mechanism, shrinking core (SC) model is believed to be the most frequently applied approach. Using this model, the dissolution mechanism is usually identified through a proper fitting between the SC model prediction and the experimental observation data. If the observed dissolution profile is linear, the dissolution process is considered as the reaction-controlled dissolution (RCD). If the dissolution profile is in a parabolic shape, the boundary layer diffusion-controlled dissolution (BLDD) is the mechanism. When the dissolution profile shows a sigmoidal shape, shell layer diffusion-controlled dissolution (SLDD) may be the controlled mechanism. Alternatively, stationary interface diffusion model (SIM) could also obtain a sigmoidal profile which quite closes to SLDD. For the classical SC models, the experimental profile sometimes could not fit any mechanism curve which is a clear drawback, a few attempts have been made to modify the SC model. For instance, Feichtinger et al. (2014) introduced a factor 'f' in the SIM model (also named 'invariant interface diffusion control'), which improves the model performance significantly, and 'f' is reported to relate to slag viscosity. Very recently, a modified SC-based physical model, i.e. diffusion-distance-controlled dissolution (DDD) model (Xuan and Mu, 2021) is developed to investigate the shape origin and variation of oxide dissolution in the slag. It is reported that the diffusion-related particle dissolution is actually one mechanism but not a few, the shape of the dissolution profile is controlled by the distance of diffusion region. This model has been verified with HT-CLSM data of different kinds of inclusions dissolution. However, the comprehensive application of this model is relied on the input of physical parameters to present different slag compositions, currently this is the only limitation for the DDD model.

In order to further extend the application of the theoretical model to predict inclusion dissolution in the comprehensive processing conditions, machine learning (ML) has been considered to use to combine with the current DDD physical model. Digitalization has brought many significant changes to the manufacturing in the recent decade. ML is a sub-domain of artificial intelligence, which is the key enabling technology for industrial-driven research, e.g. material engineering and metallurgy. The database for ML model is collected using HT-CLSM experimental data, DDD model is used for the augmentation of the current database, to have a continuous distribution of data. Unseen experimental data is used as the benchmark of the ML model. Al_2O_3 is the most common inclusion, so its dissolution in the slag is performed in the current study, to test the accuracy of the current hybrid modelling methodology. The successful implementation of this model can be applied to predict the dissolution of other kinds of inclusions, e.g., MgO, SiO_2, $MgAl_2O_4$, $CaO\cdot 2Al_2O_3$, etc. in the further study.

METHODOLOGY

The current ML dataset contains 1506 data points collected from the open literatures (Hagemann et al., 2010; Lee et al., 2001; Liu et al., 2007a, b; Monaghan et al., 2005; Michellic et al., 2016; Sharma et al., 2018; Soll-Morris et al.,2009; Sridhar and Cramb, 2000; Yi et al., 2003), DDD physical model is used for the data augmentation, this combination methodology can be seen in the report by Mu et al. (2021). The collecting dataset is split into 80% for training and 20% for testing. Furthermore, 252 unseen data which are not included in the training and testing datasets (Liu et al., 2007a,b; Yi et al., 2003; Michellic et al., 2016) is used as the validation dataset for the benchmark. There are ten parameters including in the dataset to predict Al_2O_3 dissolution, i.e. original inclusion size (radius), temperature, concentration of five components in slag (SiO_2, CaO, Al_2O_3, MgO and Fe_tO), density of inclusion, slag viscosity and radius change (R/R_0, R represents the radius at the certain time and R_0 is the initial radius). The time required for the dissolution of Al_2O_3 particles was selected as the output. For the further work to predict different kinds of inclusion dissolution, one more parameter, inclusion type needs to be added. The detailed information of training & testing dataset as well as validation dataset are listed in Tables 1 and 2.

Standard normalization was used in this work in order to eliminate the difference in numeric scale of features. Based on the previous modelling results, it is found that the prediction of the trained ML model fluctuates as the different partition of training dataset and testing dataset, so multiple hold-out method is used in order to reliably evaluate model's performance (Shen et al., 2019). In this study, the dataset was randomly divided into training and testing sets by 50 times to develop 50 ML models. Moreover, common mean absolute error (MAE) and square correlation coefficient (R^2) were used to

evaluate the model's performance (Pal, 2017). Besides, the suitable selection of ML algorithm is also important for good performance. In this work, six common ML algorithms were employed to develop three models, including Support Vector Regression (SVR) (Liu et al., 2021), Multi-Layer Perception (MLP) (Del Campo et al., 2021) and three kinds of ensemble learning algorithms, i.e. AdaBoost (Ada), decision tree regression (DTR), random forest regression (RFR) (Sammut et al., 2011), and the modelling results for testing data are plotted in Fig. 1. It is seen that ensemble algorithms have the higher prediction accuracy than MLP and SVR algorithms. The R^2 values of these models were greater than 98%, meaning a very good prediction performance. Finally, RFR algorithm was selected to predict the dissolve curve of Al_2O_3 particle owing to the relatively lowest MAE value.

TABLE 1 - Data distribution in present dataset (including training and testing sets).

Parameters	Min.	Max.	Mean	Std.
Radius [μm]	41.3	250.2	176.6	76.9
Temperature [°C]	1673	1873	1772.3	56.7
SiO_2 [wt.%]	0	62.4	33.4	19.8
CaO [wt.%]	22.4	57.2	35.2	9.1
Al_2O_3 [wt.%]	13.3	50.0	29.0	11.6
MgO [wt.%]	0	7.5	2.0	3.1
Fe_tO [wt.%]	0	9.2	0.4	1.7
Slag Density [$kg.m^{-3}$]	3568	3640	3604	20.4
Slag viscosity [Pa.s]	0.1	100.5	4.7	16.4
R/R_0 [-]	0	1	0.7	0.3
Dissolution time [sec]	0	4490	925	1099

TABLE 2 - Data distribution in validation set

Parameters	Min.	Max.	Mean	Std.
Radius [μm]	95.3	249.1	226.6	47.3
Temperature [°C]	1723	1873	1778.2	50.1
SiO_2 [wt.%]	0	46.2	39.2	16.5
CaO [wt.%]	29.7	49.5	32.7	7.1
Al_2O_3 [wt.%]	24.1	50.0	28.0	9.3
MgO [wt.%]	0	0.5	0.08	0.18
Fe_tO [wt.%]	0	0	0	0
Slag Density [$kg.m^{-3}$]	3568	3622	3602	18.0
Slag viscosity [Pa.s]	0.8	3.6	2.5	1.1
R/R_0 [-]	0	1	0.56	0.26
Dissolution time [sec]	0	4490	1527	1290

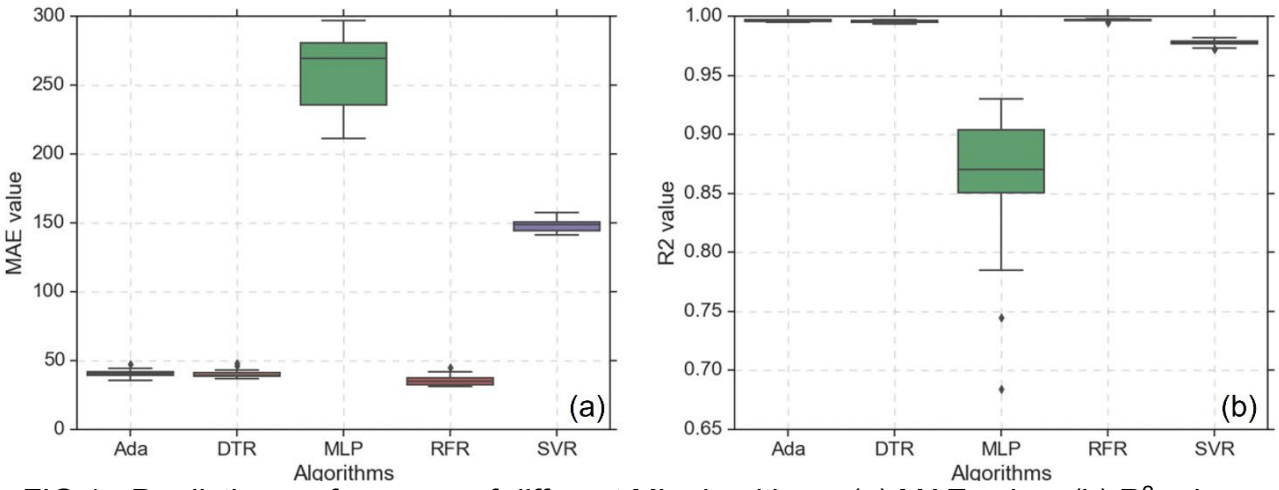

FIG 1 - Prediction performance of different ML algorithms: (a) MAE value; (b) R^2 value.

RESULTS AND DISCISSION

Pearson correlation is one of the simplest methods to determine the linear relation between features and the dependent variable. According to the method, the features that are uncorrelated with the dependent variable are good candidates to exclude from the dataset before training the model (Rahaman et al., 2019). Here we apply the same methodology to determine the correlation between variables (radius, temp., slag compositions, inclusion density, slag viscosity, and R/R_0) with dissolution time. This method measures the linear correlation between two variables and the resulting value lies between -1 and 1. Negative values mean negative correlation; alternatively, positive values mean the opposite; 0 means that there is no linear correlation between the two variables. The Pearson's correlation coefficient for the inclusion dissolution dataset is represented as a heat map in Fig.2. It can be seen that some parameters, e.g. original inclusion radius and SiO_2 have clear positive correlation with the dissolution time. It is obvious that the increasing inclusion original size as well as the SiO_2 content in slag will increase the dissolution time. The parameter, e.g. inclusion density has a less influence on the dissolution time. However, since the role of Pearson correlation is to check if some parameters can be removed but not the feature importance, we will discuss deeply on each parameter's linear correlation, also we keep all parameters in this ML model. For the current work, the data amount is not really large, more parameters are always good to use for the accuracy of ML modelling containing small size dataset.

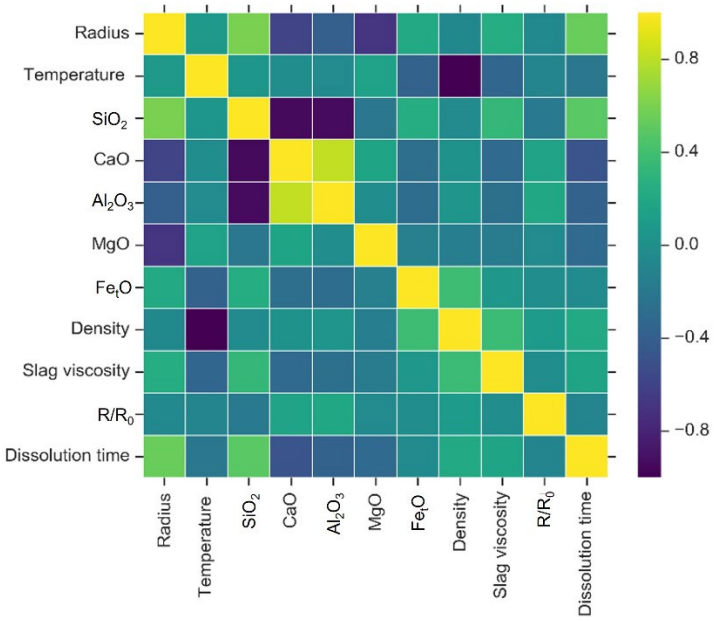

FIG 2 – Pearson correlation

Fig. 3 displays the prediction results of inclusion dissolution. The output is dissolution time of Al_2O_3 inclusion in the slag, and the X and Y axis strand for the actual dissolution time and predicted time by ML model, respectively. Fig. 3 (a) shows the actual value *vs.* predicted value for training data. It can be seen that all data points basically concentrated around the diagonal, showing a very high prediction accuracy. The MAE and R^2 values are 17.0 second and 99.8%, demonstrating that the ML model has learned the reliable relation between dissolution time of inclusions and all input features based on training samples. In order to test the application ability of trained model, 20% of the total data in the testing set were inputted into model, and the prediction result is shown in Fig. 3 (b). It is observed that most data points are also located in the line with the slope of 1, showing a good robustness of trained model. The MAE and R^2 values are 30.2 second and 98.7%. It is also seen that small number of data points deviates from the diagonal, but the largest MAE value is still less than 150s, which is within the acceptable scale. In order to further validate the robustness for new data, trained model was applied to the unseen 252 data in the validation dataset with different parameters. The prediction result is shown in Fig. 4. It can be seen that like training and testing data, present model also has a high prediction accuracy on validation data, and most data points locate around the diagonal. The MAE and R^2 values are 70.8s and 97.7%, respectively. Compared to training and testing data, the predicted accuracy on validation data slightly reduces, but present prediction level is still high enough to guide the actual experiments.

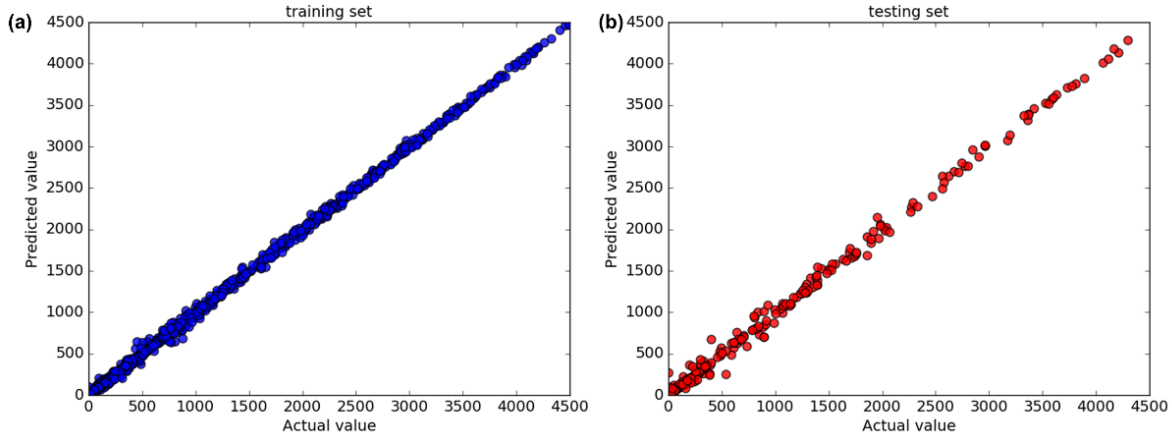

FIG 3 - Predicted results of dissolution time of inclusion using (a) training and (b) testing dataset.

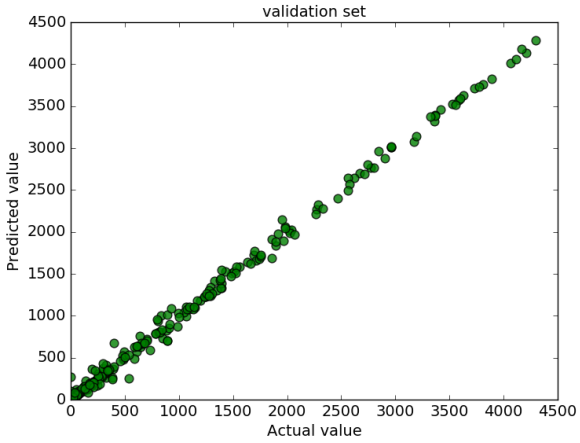

FIG 4 - Predicted results of dissolution time of inclusion using validation dataset.

It is known that the parameters, e.g. slag viscosity is a joint function of temperature and slag composition, and the inclusion density is also influenced by the temperature. Since these parameters are already included in the current database, new attempt to remove slag viscosity and inclusion density in the database has been made to see the model prediction performance. It seems the model accuracy decreases slightly but it is still quite acceptable (i.e. 41.66 of MAE and 98.41% of R^2 for using testing dataset in new model). In this case, the new model can be considered to use by inputting slag composition, temperature, etc. but without using physical parameters, e.g. slag viscosity, inclusion density, etc. which will be even easier to operate the model for the application perspective.

In addition, it needs to be mentioned that the correlation importance between dissolution time and each input parameter can be analysed. This type analysis is usually called feature importance (FI). FI is used to evaluate which parameter plays a more important role in within the specific dataset (Rahaman et al., 2019; Mu et al., 2021). The importance of the same parameter can be different if the dataset is changed. However, the current dataset includes the experimental data are all collected from the HT-CLSM measurement. In this case, some parameters e.g. contents of different components in the slag are not such comprehensive due to the limitation of HT-CLSM (e.g. Fe_tO content should be almost zero). In this case, the feature importance may show different tendency compare with the physical understanding of inclusion dissolution, so FI analysis is not included in the current work.

CONCLUSIONS

The current hybrid methodology in combination of machine learning and physical modelling has been established to predict Al_2O_3 dissolution in the steelmaking slag. The successful implementation of applying the method to predict of dissolution time provides the possibility to further application of the model. Different kinds of inclusion dissolution data observed by HT-CLSM and measured by other methodology is needed to be used to establish a robust dissolution model.

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