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Predicting the dissolution kinetics of silicate glasses using machine learning

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ABSTRACT

Predicting the dissolution rates of silicate glasses in aqueous conditions is a complex task as the underlying mechanism(s) remain poorly understood and the dissolution kinetics can depend on a large number of intrinsic and extrinsic factors. Here, we assess the potential of data-driven models based on machine learning to predict the dissolution rates of various aluminosilicate glasses exposed to a wide range of solution pH values, from acidic to caustic conditions. Four classes of machine learning methods are investigated, namely, linear regression, support vector machine regression, random forest, and artificial neural network. We observe that, although linear methods all fail to describe the dissolution kinetics, the artificial neural network approach offers excellent predictions, even for untrained data, thanks to its inherent ability to handle non-linear data. We further note that the predictive ability of simpler methods, such as linear regression, could be improved using additional physics-based constraints. Such methods, called as physics-informed machine learning can be used to extrapolate the behavior of untrained compositions as well. Overall, we suggest that a more extensive use of machine learning approaches could significantly accelerate the design of novel glasses with tailored properties.

1. Introduction

Silicate glasses are often exposed to water—from the manufacturing stage to their service lifetime—which can result in corrosion and dissolution [1–5]. The durability of glasses in aqueous environments plays a critical role in various applications and processes, including bioactive glasses, laboratory glassware, atmospheric weathering of outdoor glasses, post-manufacturing treatment, nuclear waste immobilization, geological processes, or dissolution-precipitation-induced creep [6–16].

Depending on each application, glass dissolution may be desirable or not. As such, developing novel glasses with tailored dissolution rates requires an accurate prediction of their dissolution kinetics. However, despite decades of research, the lack of reliable models predicting how the dissolution kinetics depends on intrinsic (glass composition, structure, surface geometry, etc.) and extrinsic conditions (temperature, pressure, solvent chemistry, etc.) [1,13,17–20] suggests that improved predictive models are required. Elucidating composition–durability relationships in silicate glasses is further complicated by the facts that glasses can exhibit a virtually infinite number of possible compositions and that dissolution kinetics is highly non-additive with respect to composition [20–23]. These issues are further complicated by the fact that the rate-limiting mechanism of dissolution can change over time, as exemplified in the cases of silicate minerals [8] or nuclear waste glasses [10,12]. To this end, various empirical and mechanism-based models have been suggested to predict the dissolution rate of oxide glasses [1,13,17,19,24–28]. However, these models are usually applicable only for prescribed glass composition envelopes, solvent chemistry (e.g., acidic or caustic), thermodynamic conditions (e.g., temperature range). Although a mechanistic model of dissolution transferable to a broad range of glass compositions would be highly desirable, this task might not be realistic due to the structural complexity of glasses and the fact that several dissolution mechanisms can be observed, individually or in combination.

As an alternative route, data-driven models relying on machine learning are a promising tool to predict composition–property relationships in glasses based on analysis of the large quantities of

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experimental data that are already available [29–34]. Data-driven predictive models range from simple regression-based methods to highly non-linear methods, such as artificial neural networks—usability of which depends on the complexity of the mechanism involved [35–38]. Such methods exploit available databases of high-quality measurements to develop semi-empirical models to improve predictive capabilities [35]. These methods have been used for a wide range of applications, ranging from face recognition [39] to infrastructure lifespan prediction [40,41] or the design of novel composites [42]. However, very few studies have been carried out and published for the use these methods for predicting the properties of glasses [30,43,44].

Here, we investigate the ability of some machine learning approaches to predict the dissolution kinetics of a selection of sodium aluminosilicate glasses. In particular, we use linear regression (LR), random forest (RF), support vector machine regression (SVM), and artificial neural network (ANN), which represent four different classes of machine learning techniques. On account of the intrinsically non-linear character of the composition–dissolution relationship, we demonstrate that the ANN approach offers the most reliable prediction of the SiO_2 leaching rate over a wide range of glass compositions.

2. Methodology

2.1. Data set

To test the predictive capability of different machine learning methods, we rely on the database of dissolution rates reported by Hamilton [23]. The experiments were conducted on eight different sodium aluminosilicate glasses, including albite glass (Na_2O–Al_2O_3–$6SiO_2$), jadeite glass (Na_2O–Al_2O_3–$4SiO_2$), nepheline glass (Na_2O–Al_2O_3–$2SiO_2$), and Na_2O–xAl_2O_3–$(3-x)SiO_2$ glasses, where $x = 0.0$, 0.2, 0.4, 0.6, and 0.8. The composition range thus covers both tectosilicate and peralkaline compositions, with varying ratio of bridging to non-bridging oxygens. For each composition, the dissolution experiments were carried out on glass powders with grain sizes ranging from 74-to-149 μm. The dissolution kinetics was assessed both in acidic and caustic conditions, specifically, pH = 1, 2, 4, 6.4, 9, and 12. These experiments were conducted in static conditions at a surface area to solution volume ratio (SA/V) of approximately 1.4 to 2.0 cm^{-1} [23]. For each pH, the extent of dissolution was assessed from the concentration of leached SiO_2 in solution at five to seven regular intervals (e.g., 24, 49, 96, 168, and 336 h) of solvent contact. The complete dataset used for the present study is provided in Supplementary Information. In each case, the pH was recorded before any dissolution and at the time of the dissolution measurement. All the experiments were conducted at 25 °C and ambient pressure. For a detailed description of the measurements, the reader is invited to refer to Ref. [23].

2.2. Inputs and outputs

Here, our goal is to develop a predictive model of the dissolution kinetics of silicate glasses. The output of the model is chosen as being the SiO_2 leaching rate (in units of log[mol $SiO_2/cm^2/s$]) as this quantity captures the dissolution of the silicate skeleton of the glass. This gives a total of 299 data points. However, the methodology developed herein is general and can be applied to other outputs (e.g., the Na_2O leaching rate or the glass weight loss rate). Based on the information contained in the selected database, the following variables are used as inputs: (i) the composition of the glass, (ii) the initial pH of the solution, and (iii) the pH at the time of measurement. Note that the dissolution rate was found to be fairly constant over the measured period, so that time was not included as an input. The temperature is not used since it is assumed to be constant over time.

2.3. Machine learning methodology

For most machine learning methods, the available data (inputs and outputs) is randomly divided into (i) a training set and (ii) a test set. The training set and test set are scattered within the whole area occupied by data due to the random sampling. The training and test set are chosen in such a way that the characteristics of the overall dataset are preserved in both the subsets. In other words, a test set is typically a dataset that is independent of the training set but follows the same probability distribution as that of the training set. Thus, all the compositions and pH values are represented with the same probability in both the training and test set. The training set is first used to train the model, that is, to optimize the parameters that relate the inputs to the outputs. The test set, which is fully unknown to the model, is then used to assess the performance of the model—by comparing the outcomes of the model for inputs the model has not been explicitly trained for to reference outputs. Such division of data into training and test sets helps to avoid any potential overfitting, which is a common problem when the entire data set is used to training the model. Here, 70% and 30% of the data are attributed to the training and test sets, respectively. Note that, in the case of the ANN method, a more elaborated data classification is used, as discussed below. In the following, we provide a brief description of the predictive methodologies used herein.

2.3.1. Linear regression

2.3.1.1. Simple linear regression. The linear regression (or least squares fitting) is the simplest form of regression technique. It consists of finding the best fitting straight line through a set of points. For a given input vector $X = (X_1, X_2, \ldots, X_p)$ and an output Y, the linear regression has the following form:

$$Y = \beta_0 + \sum_{j=1}^{p} X_j \beta_j \tag{1}$$

where β_j are the fitting parameters of the model and p the number of such parameters. The β_j values are usually obtained by minimizing the error of predicted values with respect to the actual values, which is represented by the residual sum of squares (RSS). Thus, for a given training data set $(x_1, y_1), \ldots, (x_N, y_N)$with N points, the RSS can be obtained by:

$$RSS(\beta) = \sum_{i=1}^{N} \left(y_i - \beta_0 - \sum_{j=1}^{p} x_{ij} \beta_j \right)^2 \tag{2}$$

where y_i is the measured value at the ith observation with features $x_{ij} = (x_{i1}, x_{i2}, \ldots, x_{ip})$. The least square estimate of the parameters β_j has the smallest variance among all linear unbiased estimates and, hence, is used commonly for linear regression. Note that, in unbiased estimates, all the input variables have non-zero coefficients, irrespective of whether they affect the output significantly or not.

2.3.1.2. Lasso regression. As an unbiased estimate, one of the major drawbacks of the least square estimate is its large variance. Such variance can be reduced by introducing a small bias, wherein the unimportant input variables are neglected. To this extent, we use the lasso regression method, which typically improves the prediction accuracy of linear regression by introducing a bias and shrinking the coefficients of insignificant variables to zero. In other words, lasso regression identifies the important variables that affect the prediction significantly. To achieve this, the lasso regression method introduces a constraint on the regression coefficients using a penalty factor λ as:

$$\hat{\beta}_{lasso} = \underset{\beta}{\operatorname{argmin}} \left\{ \sum_{i=1}^{N} \left(y_i - \beta_0 - \sum_{j=1}^{p} x_{ij} \beta_j \right)^2 + \lambda \sum_{j=1}^{p} |\beta_j| \right\} \tag{3}$$

Note that in the lasso regression method, the penalty is imposed on