Deep learning modeling of the atomic structure and physical properties of alkali and alkaline-earth aluminosilicate glasses and melts

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**Abstract**

The quest for a general, usable model to predict atomic structure, thermodynamic and viscous properties of aluminosilicate melts and glasses, is critical for geo- and materials sciences. Recently, a deep learning greybox model, ‘i-Melt’, was proposed and trained to predict 18 different latent and observed properties of melts and glasses in the K2O-Na2O-Al2O3-SiO2 system, including melt configurational entropy and viscosity, as weel as glass optical refractive index, density, and Raman signals. Here, we extend the compositional range of the model to include CaO and MgO. The model now allows accurate prediction of observed and latent properties for glass-forming melts in the CaO-MgO-K2O-Na2O-Al2O3-SiO2 system, including melt viscosity (precision ≥ 0.4 log10 Pa·s in the 100-1015 log10 Pa·s range), configurational entropy at glass transition (XXX J mol-1 K-1), liquidus Tl (≤ 60 K) and glass transition temperature Tg (≤ 14 K) as well as glass density (≤ 0.02), optical refractive index (≤ 0.006), Abbe number (XXX), elastic modulus (XXX), coefficient of thermal expansion (XXXX) and Raman spectra (~19 %). The model offers new possibilities to explore how melt/glass properties change with composition and atomic structure. Among those, model predictions suggest that known changes in the coordination numbers of metal cations and aluminum correlate with variations in their predicted partial molar contributions to the glass molar volume.

**1. Introduction**

Aluminosilicate melts that contain alkali and calc-alkaline metal cations serve as the base composition in the glass-making industry and also constitute the liquid fraction of most of the Earth's magmas. The viscosity of these melts is critical as it determines their resistance to movement at high temperatures, thereby influencing their mobility. This property heavily impacts the fragmentation of magmas in volcanic edifices (e.g., see reviews of Gonnermann and Manga, 2013; Gonnermann, 2015), and working temperatures in industrial glass-making furnaces. Other properties of melts and glasses, such as density or optical refractive index, may also be of significant interest because they can influence not only the mobility of the melt, for example through buoyancy effects, but also the weight and optical properties of glass objects. The prediction of such physical properties is, therefore, essential in addressing problems ranging from the dynamics of volcanic eruptions to the development of novel glass materials.

Viscosity predictions can be performed with empirical (Bottinga and Weill, 1972; Shaw, 1972; Persikov, 1991; Hui and Zhang, 2007; Giordano et al., 2008) thermodynamic models (Sehlke and Whittington, 2016; Le Losq and Neuville, 2017; Starodub et al., 2019). Molecular dynamics (MD) simulations also provide estimates at very high temperature conditions (e.g., Guillot and Sator, 2007; Bauchy et al., 2013; Dufils et al., 2017). However, to date, those methods did not allow building a “universal” viscosity model that predicts glass-making and volcanic silicate melt viscosity over a very broad range of temperature and compositions. This is because they all present limitations: very short timescales and very high temperatures for MD simulations, a limited interpolative working set relying on pre-determined functions for empirical models, and our relatively limited understanding of the thermodynamics of silicate melts for thermodynamic models.

To circumvent existing limitations, a new set of models relying on machine learning have been proposed: greybox models. Those combine physical/thermodynamic equations with machine learning to predict melt/glass property, with good success to date (Tandia et al., 2019; Liu et al., 2019; Hwang et al., 2020; Cassar, 2021). Among published greybox models, i-Melt (Le Losq et al., 2021) is a multitask model that predicts not only melt viscosity through five different equations but also glass density, optical refractive index and Raman spectrum. It further provides access to latent properties such as the melt fragility or configurational entropy at the glass transition. i-Melt thus allows the systematic exploration of the links between the composition, the structure (through Raman spectra predictions), and the properties of melts and glasses (Le Losq et al., 2021). The downside of this model is that it currently is limited to the glass-forming domain of the Na2O-K2O-Al2O3-SiO2 quaternary system.

In this study, we present a new version of i-Melt that now includes CaO and MgO. i-Melt was trained on melt and glass compositions in the CaO-MgO-K2O-Na2O-Al2O3-SiO2 system, for which a fairly complete, albeit sparse, experimental dataset is available. In this communication, we present this new dataset, the improvements and performance of the updated i-Melt model, and we discuss its possibilities and limits.

**2. Methods**

***2.1 Datasets and data preparation***

The original database of i-Melt was completed by collecting existing Raman spectra, optical refractive index, density, Abbe number, elastic modulus, coefficient of thermal expansion (CTE) of glasses and liquid heat capacity, viscosity and liquidus temperatures of melts in the Na2O-K2O-CaO-MgO-Al2O3-SiO2 system. The data were selected via a review of the existing literature as well as of the SciGlass database. Melt viscosity, and glass density, Raman spectra and optical refractive index were selected by hand following this literature review. Data from the IPGP internal database includes Raman spectra, viscosity and density data acquired on glasses in the CaO-MgO-Al2O3-SiO2 system synthesized at IPGP in Paris (Neuville and Richet, 1991; Neuville, 1992; Richet and Neuville, 1992; Neuville et al., 2006, 2008c, 2014; Neuville and Le Losq, 2022). Abbe number, elastic modulus, CTE and liquidus temperature data in the Na2O-K2O-CaO-MgO-Al2O3-SiO2 system were extracted from the SciGlass database thanks to the GlassPy python package (Cassar, 2020). The data used to train the model as well as the associated references are provided in the database available in the Github software repository that hosts i-Melt (https://github.com/charlesll/i-Melt), except for unpublished viscosity data on five melt compositions (Neuville, pers. comm.) and unpublished Raman spectra of 19 glasses from Neuville (1992) that are under embargo.

The different streams of data are:

- *Dviscosity* (n= 793 compositions), the dataset of viscosity measurements, composed of *Xviscosity* chemical composition entries (mole fractions) as well as their associated temperatures (Kelvin) and *yviscosity* observations (log10 Pa·s);

- *Ddensity* (n= 668 compositions), the dataset of glass density measurements, composed of *Xdensity* chemical composition entries (mole fractions) and *ydensity* observations (g cm-3);

- *DRaman* (n= 250 compositions), the dataset of glass Raman spectra, composed of *XRaman* chemical composition entries (mole fractions) and *yRaman* spectra observations (normalised Raman intensities);

- *Doptical* (n= 610 compositions), the glass dataset of optical refractive index, composed of *Xoptical* chemical composition entries (mole fractions) as well as their associated wavelength (µm) and *yrefractive index* observations;

- *DCpl* (n= 95 compositions), the glass dataset of liquid heat capacities *Cpliquid*, composed of *XCpl* chemical composition entries (mole fractions) as well as their associated temperatures (Kelvin) and *yCpl* observations (J mol-1 K-1).

- *DAbbe* (n= 296 compositions), the glass dataset of Abbe Number, composed of *XAbbe* chemical composition entries (mole fractions) as well as their associated Abbe Numbers (no unit).

- *Delastic* (n= 1006 compositions), the glass dataset of elastic modulus, composed of *Xelastic* chemical composition entries (mole fractions) as well as their associated elastic modulus (GPa).

- *DCTE* (n= 2122 compositions), the glass dataset of elastic modulus, composed of *XCTE* chemical composition entries (mole fractions) as well as their associated coefficients of thermal expansion (K-1).

- *Dliquidus* (n= 4505 compositions), the dataset of liquidus temperatures, composed of *Xliquidus* chemical composition entries (mole fractions) as well as their associated liquidus temperatures (K).

The size of the *Dviscosity*, *Ddensity*, *Doptical*, *DCTE*, *Delastic and Dliquidus* datasets allow training i-Melt with a *“high performance”* mindset, because those datasets cover an important part of the glass-forming domain of alkali and alkaline-earth aluminosilicates (**Fig. 1**). The liquid heat capacity dataset *DCpl* is small (**Fig. 1e**), but actually this is not a problem because good fits of viscosity data with the Adam-Gibbs theory already require the prediction of consistent liquid heat capacities (see *Results* section). However, having a *Dcpl* dataset, even small, allowed fine-tuning the model and ensuring that *Cpliquid* predictions are consistent with existing *Cpliquid* data. *DAbbe* also is fairly limited (**Fig. 1f**), but it actually will depend on XXXXX. Regarding *DRaman*, it also covers a limited set of compositions (**Fig. 1b**) and we do not expect a very high precision on Raman spectra predictions. Raman data were actually used as a way of improving learning through a multitask approach (Caruana, 1997), because they encode structural information that could assist the network in learning physical properties, embedding a shared representation of the composition-structure-property links in melts and glasses. This actually is one important basis underlying the i-Melt model: the use of datasets of different glass/melt properties should allow leveraging their different compositional coverage to ensure better predictive performance of the model overall.

Following Le Losq et al. (2021) and prior to training i-Melt, the datasets were split by composition into three different, randomly chosen *training*, *validation* and *testing* subsets. While the *training* subsets are used for training the model (i.e. tuning its internal parameters), the *validation* subsets are used for monitoring overfit and triggering early stopping when the later occurs (Goodfellow et al., 2016). The final predictive abilities of the trained neural networks are evaluated using the *testing* data subsets. For performing the splits, we used a stratified group splitting approach. This approach was implemented because (i) we need to split the datasets by compositions to avoid data leackage (Kaufman et al., 2012), and (ii) we have imbalanced datasets. While the former point was taken into account in the original version of the model (Le Losq et al. 2021), the latter was not and is particularly important now. For instance, we have significantly less compositions including Na2O and K2O than other elements in the viscosity dataset, while the density dataset includes less MgO-bearing compositions (**Fig. 2**). A train-valid-test split by composition will work, but may not retain the proportions of each kind of compositions (e.g. sodium silicate, magnesium aluminosilicate, etc.) in the different train-valid-test splits. To solve this problem, we assigned to each type of compositions a class (e.g., for sodium silicate class 1, for sodium aluminosilicates class 2, etc.), and used a stratified splitting approach that aims at retaining, as much as possible, the proportions of each class in the different train-valid-test data subsets. In practice, this approach is implemented via a hack of the *StratifiedGroupKFold* function of scikit-learn library version 1.1.2 (Pedregosa et al., 2011). After data splitting, we systematically check that there is no sign of data leakage (all compositions in the train, valid and test subsets are different), and we visually checked that the coverage of the different splits is reasonable (**Fig. 1**). Train-valid-test splits were performed with 0.8/0.1/0.1 ratios for all datasets, except for *DRaman*. The latter was divided in only two *train* and *validation* subsets due to its small size, with a 0.85/0.15 ratio. This is not problematic, because we do not aim at precise predictions of Raman spectra but rather use this dataset as a way to improve the general predictive capacity of the trained neural network through multitask learning. Scaling of the datasets was done as described in Le Losq et al. (2021). After pre-processing, the different data subsets were saved in Hierarchical Data Format HDF5 files for their future use.

***2.2 Deep learning model***

The model i-Melt, implemented in the Python programming language using the Pytorch machine learning library (Paszke et al., 2019), was presented in details in Le Losq et al. (2021). We refer the reader to this publication for an extensive presentation of the model. Here, we briefly present the model and its general architecture, and focus on describing improvements and new features.

i-Melt combines a deep artificial neural network with various dynamic and thermodynamic equations to predict latent and observed melt/glass properties. The model takes six inputs: the mole fractions of SiO2, Al2O3, Na2O, K2O, MgO and CaO. From these, new chemical descriptors are now calculated, such as the glass optical basicity and NBO/T, the Al/M ratio (with M the sum of metal cations), and the ratio of each element to another. A total of 39 descriptors, including initial melt composition, are fed into a neural network composed of *n* hidden layers, each one having *k* activation units (a.k.a neurons). The outputs of the hidden layers are fed into two different linear layers for outputs: the first one returns vectors that are Raman spectra, and the second one returns 34 different values:

- the parameters allowing the calculation of melt viscosity through five different theoretical and empirical equations, including Adam-Gibbs (Adam and Gibbs, 1965; Richet, 1984), MYEGA (Mauro et al., 2009), Avramov-Milchev (Avramov and Milchev, 1988), Tamman-Vogel-Fulcher and Free Volume Theory (Cohen and Grest, 1979, 1984);

- the partial molar volumes of each oxide components, for density calculations;

- the partial liquid heat capacity of each oxide component as well as two temperature-dependent terms for Al2O3 and K2O (following Richet and Bottinga, 1985) for *Cpliquid* calculations;

- the coefficients of the Sellmeier equation for optical refractive index calculation;

- the melt liquidus temperature;

- the glass Abbe number;

- the glass elastic modulus;

- and the coefficient of thermal expansion of the glass.

The artificial neural network allows us, therefore, to input chemical compositions and obtain predictions for:

* melt viscosity, within five distinct theoretical or empirical frameworks,
* melt heat capacity, including partial molar contributions of oxide components,
* liquidus and glass transition temperatures,
* latent variables like configurational entropy and fragility,
* glass density and partial molar volumes of oxide components,
* glass refractive index as a function of wavelength, its Abbe number and elastic modulus,
* and the glass Raman spectra.

The predictions depend on a large number of tuneable parameters integral to the neural network, as well as on the careful adjustment of the neural network hyper-parameters. Adjustment of model parameters (weights and bias of the activation units) was performed via batch training through gradient descent with using the ADAM optimizer. The global loss function was calculated from a weighted sum of the root-mean square errors (RMSE) between measurements and predictions for viscosity as well as liquid heat capacity, density, optical refractive index, Raman spectra, liquidus temperatures, elastic modulus, CTE, Abbe numbers and known glass configurational entropy *Sconf(Tg)* values. The weights assigned to the different tasks in the global loss function were learned during the optimization process, following the method proposed by Kendall et al. (2018). Back-propagation was performed using the automatic differentiation methods implemented in Pytorch (Paszke et al., 2019).

Good performance of the model can only be achieved upon finding optimal sets of model hyperparameters, including the optimizer learning rate, and the number of layers, the number of units per layer, and the type of activation functions in the artificial neural network. Regarding the activation functions, the initial version of i-Melt relied on Rectifier linear units (a.k.a. ReLU), but new tests showed that Gaussian error linear units (GELU, Hendrycks and Gimpel, 2020) – yield better generalization performance (less overfitting and better estimates on unseen samples). Therefore, the new version of i-Melt uses GELU units. To further prevent overfitting and help model generalisation, we also rely on early stopping (Goodfellow et al., 2016) and dropout (Srivastava et al., 2014). The dropout rate as well as the optimizer learning rate, the number of layer and that of activation units per layer in the artificial neural network were tuned using the hyperparameter optimization framework Ray Tune (Liaw et al., 2018), which allows distributed model selection and training. In practice, we used the Optuna algorithm (Akiba et al., 2019) that relied on monitoring the global loss on the validation datasets to guide the selection of the most promising models.

This approach allows obtaining a sample of trained models from which we can select the best ones. Typically, the architecture of the best models is moderately deep, with 4 to 6 layers, each containing 300 to 400 activation units. We also observed that the use of GELU units allowed obtaining good model prediction with dropout rates in the range 0.10-0.15. Best ADAM learning rates were generally in the 5e-3 - 9e-3 range. For final predictions, we average predictions of an ensemble of the 10 best models. Uncertainties on those predictions can also be estimated through the MC Dropout method (Gal and Ghahramani, 2016): for a given input, we leave the dropout active and ask for 100 different samples for each one of the 10 neural network. We thus obtain for each input a subset of 1000 predictions, each prediction being slightly different from another because coming from a different neural network, or from a different part of a given neural network. From this subset, we can provide mean values and standard deviations, approximating a Bayesian sampling of the model posterior probability function (Gal and Ghahramani, 2016).

**3. Results**

***3.1 Predictive performance evaluation***

Over the very broad range of compositions we investigate (**Fig. 1**), viscosity of melts are predicted with a good precision (**Fig. 3a**). Regardless of the chosen theory, the RMSE valuesare equal to 0.4 log10 Pa·s on the testing data subset (**Table 1**). Predictions in the supercooled temperature domain are affected by larger errors than predictions in the sub-liquidus to super-liquidus domain: testing RMSE values (all equations considered) are in the range 0.3-0.5 log10 Pa·s when considering only data in the 107-1015 Pa·s range, whereas they are in the range 0.2-0.3 log10 Pa·s for data below 107 Pa·s (**Table 1**). The present model uncertainties on viscosity are comparable with, or even better than those of the previous version of i-Melt for alkali aluminosilicate melts (~0.4 log10 Pa·s**)**, and are better than those affecting existing thermodynamic models for quaternary alkali aluminosilicate melts (Starodub et al., 2019; Neuville and Le Losq, 2022). The precision on viscosity predictions of the model actually approach, despite a much broader compositional dataset, that of the thermodynamic model of the viscosity of alkali silicate melts of Le Losq and Neuville (2017) (0.2 log10 Pa·s).

Other melt/glass properties are also well predicted by i-Melt (**Fig. 3 and Table 1**). Known viscous glass transition temperatures *Tg* and configurational entropy at *Tg*, *Sconf(Tg),* are predicted within 13 K and 1.0 J mol-1 K-1, respectively. For *Tg*, such a precision is better than that achieved by the first version of i-Melt (~19 K), while for *Sconf(Tg)* it is comparable. Melt liquidus temperatures are predicted to within 60 K, an uncertainty that approaches those of dedicated polynomial and machine learning models (e.g., Dreyfus and Dreyfus, 2003). The melt heat capacities are predicted within 2%, a precision that can be compared with that of 5% achieved when using the model of Richet and Bottinga (1985) corrected with the Al2O3 partial *Cpliquid* value of Courtial and Richet (1993), following Giordano and Russell (2017). Glass density and refractive index are predicted to within 0.02 g cm-3 and 0.005, respectively. Such values are comparable to, or better than those for the original i-Melt version for alkali aluminosilicate compositions (Le Losq et al., 2021). For glass density, the model standard error further compares very well with that reported by dedicated parametric models such as that of Fluegel (2007). Glass elastic modulus is predicted within 7 GPa, a precision that also compares well with those achieved by topological models for instance (e.g., Wilkinson et al., 2019).

Global variations in glass Raman signals remain well-captured despite the very small experimental Raman dataset (Fig. 1b): the relative least square errors on the training and validation subsets are of 20 % and 19 %, respectively. Such errors actually are better than those affecting the original version of i-Melt, which were of ~25%. **Figure 4** shows example of Raman spectra predictions. We observe that the general shape of the spectra has been well captured by the model. In details, there remains visible deviations between predictions and observations, particularly for small features in the spectra such as small shoulders and peaks. To get better Raman spectra predictions, the model could benefit from a broader training dataset, as this actually is one of the smallest training dataset (Fig. 1). Despite this, predictions can be useful (e.g., see Le Losq et al. (2021) for examples of usage) but the uncertainties should be taken into considerations upon interpreting the obtained results. Fortunately, the new ability of the model to return uncertainties on predictions allow users to systematically take uncertainties into considerations.

***3.2 Model internal consistency***

Among the predicted parameters, i-Melt returns the melt fragility *m*, which corresponds to the derivative of the viscosity against temperature curve at *Tg*. Melt fragility (*m*) is related to structural and thermodynamic melt properties, and allows distinguishing “strong” melts from “fragile” ones that show a strongly non-Arrhenian dependence of their viscosity against temperature (Angell, 1991). Indeed, the Adam-Gibbs theory (Adam and Gibbs, 1965) relates melt dynamic and thermodynamic properties through the equation (Richet, 1984):

, (*2*)

with *T (K)* the temperature, *Ae* (Pa·s) a high-temperature limit, *Be* (J mol-1) a term proportional to the energy barriers opposed to molecular re-arrangements, and *Sconf* (J mol-1 K-1) and *Cpconf* (J mol-1) the melt configurational entropy and heat capacity, respectively. *Cpconf* is equal to (Richet, 1984):

, (*3*)

with the glass heat capacity at Tg and the liquid heat capacity at T. In the model, is predicted via the Dulong and Petit limit (see discussion in Richet, 1987). is predicted from the ponderated sum of neural network predicted partial molar heat capacities of the oxide components, and neural network predicted temperature terms for Al2O3 and K2O (see for a discussion regarding those terms Richet and Bottinga, 1985; Courtial and Richet, 1993).

Melt fragility is expected to be proportional to the ratio of the configurational heat capacity at *Tg* over the configurational entropy at *Tg, Cpconf(Tg)/Sconf(Tg), as* *(Toplis et al., 1997):*

. (3)

The correlation expected from eq. (3) has been experimentally observed for melts presenting relatively restrained compositional ranges (Webb, 2008; Russell and Giordano, 2017). Therefore, if i-Melt is internally consistent, i.e. if it predicts physically realistic melt fragilities, heat capacities and configurational entropies, we should also observe the correlation expected from eq. (3). This is clearly the case, as observed in **Figure 5**. We observe a general trend located in between those reported in Webb (2008) and Russel and Giordano (2017), the first study concerning sodium and calcium aluminosilicate melts and the second one iron-bearing aluminosilicate melts of geological compositions.

In the past version of i-Melt, focused on predictions in the Na2O-K2O-Al2O3-SiO2 system, the correlation expected from eq. 3 was observed but with a significant scatter (see Figure 6 in Le Losq et al., 2021). Here, the scatter is much more limited, despite the fact that predictions cover a much broader compositional space. A systematic, albeit not fully linear, trend between melt fragility and *Cpconf(Tg)/Sconf(Tg)* is observed (**Fig. 5**). The improvement in internal consistency is actually due to predicting through the artificial neural network. Indeed, we tried modeling with using partial molar liquid heat capacity values for the different oxide components from several studies (e.g., Richet and Bottinga, 1985; Courtial and Richet, 1993; Tangeman and Lange, 1998; Webb, 2008). However, the present predictive range of the model covers a very wide compositional range (**Fig. 1**), including Na-rich and Mg-rich ones for which non-linear dependence of partial molar has been reported (e.g., Courtial and Richet, 1993; Tangeman and Lange, 1998). Artificial neural network predictions of partial molar for the different oxide components, and their associated temperature dependence for Al2O3 and K2O, allowed largely increasing the consistency of the model, while yielding good predictions of (**Figs. 3e, 5**).

In **figure 5**, we observe that there is clear compositional mapping between melt fragility and *Cpconf(Tg)/Sconf(Tg)*. Si-rich and alkali-bearing compositions systematically present low fragilities and *Cpconf(Tg)/Sconf(Tg)*, while calcalkaline compositions present higher ones. Ca aluminate compositions present the highest fragilities and *Cpconf(Tg)/Sconf(Tg)* (magenta symbols in **Fig. 5**). In parallel, the general trend between melt fragility and *Cpconf(Tg)/Sconf(Tg)* is almost linear, but deviates from linearity at high fragilities and *Cpconf(Tg)/Sconf(Tg)* ratios.

***3.3 Uncertainty estimations and model extrapolation***

Obtaining uncertainty estimations from machine learning models can be challenging. While several methods exist to address this issue, there is no one-size-fits-all approach for obtaining "straightforward" uncertainty estimations. In the case of deep neural networks, there are at least three established methods for obtaining uncertainty estimations on model predictions. These methods include using Bayesian neural networks (e.g., Kononenko, 1989; Izmailov et al., 2021), the MC dropout method (Gal and Ghahramani, 2016) or conformal predictions (Angelopoulos and Bates, 2021).

Here, we performed a systematic analysis of the confidence intervals provided using the MC Dropout method, which relies on the use of dropout. During training, dropout consists in randomly turning off activation units of the artificial neural network at each iteration with a probability *p*. This regularization method is recognized to help generalization in deep learning models. At inference, it can also be used to generate multiple, slightly different predictions for a given input. The obtained sample of predictions can then be used to produce an estimate of uncertainties (Gal and Ghahramani, 2016).

To assess the reliability of the uncertainties estimated by the MC Dropout method, we computed the 2.5th and 97.5th percentiles of the predictive distributions for the density and viscosity datasets and compared them to the corresponding observed values. Ideally, the calculated confidence intervals should encompass 95% of the observations in the datasets. To further evaluate the efficacy of the model in accurately capturing uncertainties, we also generated confidence interval predictions using both a single model and an ensemble of ten models, as per i-Melt's usual inference protocol.

**Table 2** displays the percentage of observed viscosity and density values that fall within the 95% confidence intervals estimated using MC Dropout with either one or ten neural networks. Remarkably, the proportion of data points falling within the calculated confidence intervals is consistently within ±1% of the expected 95% value, regardless of the number of neural networks employed. Hence, both the single-model and ensemble-based predictive distributions, as well as their associated uncertainties, are highly consistent and reliable.

In the interpolative regime (50 mol% < SiO2 < 100 mol%), the confidence intervals and mean predicted values obtained using one or ten neural networks are comparable and closely align with the available data. However, upon extrapolation (SiO2 < 50 mol%), we observe a rapid expansion in the calculated confidence intervals. For instance, using one neural network, the confidence intervals quickly widen as Na2O content increases beyond the training set. This abrupt increase in calculated uncertainty indicates that the model is in the extrapolative regime, and the predictions are subject to significant uncertainties. Leveraging multiple networks to calculate the confidence intervals strengthens this observation, as each network generates distinct predictions when extrapolating (as shown in **Fig. 6a**). This leads to a substantial rise in the calculated confidence intervals and facilitates the detection of highly uncertain extrapolative predictions. Overall, the present results demonstrate that, in addition to delivering trustworthy interpolative uncertainties, the MC Dropout methodology incorporated into the updated version of i-Melt allows for the identification of poor extrapolation performance by a considerable increase in predicted uncertainties.

**4. Discussion**

The inclusion of MgO and CaO in the i-Melt model has been accomplished without compromising its predictive accuracy. In fact, as shown in **Table 1**, the error metrics for the extended model are comparable or even superior to those of the original version that focused only on the Na2O-K2O-Al2O3-SiO2 system. This is attributed to several factors, including a significantly enlarged database, the incorporation of new chemical descriptors as inputs to the artificial neural network, and the use of GELU activation units with a larger dropout rate, enabling moderately deep artificial neural networks to generalize well and produce accurate predictions. Consequently, the updated model enables systematic the predictions of the properties for melts and glasses in the Na2O-K2O-MgO-CaO-Al2O3-SiO2 system. It covers a broad compositional range that encompasses the entire glass-forming domain of this system and its related sub-systems (**Fig. 1**). Furthermore, in addition to high predictive performances (**Figs. 3, 4**), the model provides reliable uncertainty estimates using MC Dropout (**Table X, Fig. 6**).

Future extension of the model may include the addition of new properties and oxide elements. However, it should be noted that the addition of a new oxide element may require significantly more compositions to be added to the database to maintain the achieved level of precision. At the moment, it is difficult to estimate how much data needs to be added to maintain the model precision when adding new elements. Despite this, the fact that the new version of i-Melt is as precise as, or better than the original one focused on a quaternary system is encouraging and suggests that this problem is not impossible to solve. One point will require a particular focus: the addition of Raman spectra. This dataset is currently the smallest, and higher predictive precision may be achieved by adding more Raman spectra. Data on heterogeneous samples, such as Raman maps along diffusion profiles, could largely benefit model predictions, and are very welcomed.

With the new version of i-Melt, it is possible to predict many different latent and observed thermodynamic, physical and structural properties of glasses and melts. This opens up a range of possibilities for studying alkali and calc-alkaline aluminosilicate melts and glasses, such as exploring correlations between structure and properties, investigating properties in ternary and quaternary systems, and studying cation mixing effects. These possibilities were exhaustively highlighted in Le Losq et al. (2021), and examples are provided in the i-Melt repository online. Here, we focus on the noteworthy ability of the new version of i-Melt to predict partial molar volumes and heat capacities of oxide components, which has important implications for understanding the thermodynamics and transport properties of melts and glasses.

Predicted partial molar volumes of oxide components fall close to those reported in previous publications (**Table 3**), but the model predicts that they depend on composition, particularly on the Al content. A similar comment can be made for partial molar *Cpliquid*. Such a compositional dependence is not surprising, because it has been reported in several publications for density (e.g., Doweidar, 1998, 1999; Neuville and Le Losq, 2022) and for heat capacity (Courtial and Richet, 1993; Tangeman and Lange, 1998).

The model allows further understanding the potential origin of such non-linear dependence on composition. For partial *Vm*, an explanation may come from changes in the average oxygen coordination number (CN) of the cations. For instance, increasing Al2O3/Mx+2/xO in aluminosilicate glasses induces a change in the role of the metal cation Mx+2/x, from network modifier (breaking Si-O-Si bonds) to charge compensator of Al3+ (e.g., Mysen et al., 2003; Le Losq et al., 2014; Bechgaard et al., 2017). In sodium aluminosilicate glasses with 75 mol% SiO2, 23Na Nuclear Magnetic Resonance (NMR) spectroscopy document an increase in Na+ CN as Al2O3/(Na2O+Al2O3) ratio increases (Le Losq et al., 2014). A similar conclusion was drawn from calorimetry data on sodium aluminosilicate compositions by Richet et al. (1993), who proposed an increase in Na+ CN from 6 to 9 as Al is introduced into the glass network. Therefore, for metal cations such as Na+, we may expect a distribution of partial *Vm* and Cpliquid values as a function of glass composition that reflects changes in the local environment of the cations.

To test this hypothesis, we explore how the partial molar *Vm* and Cpliquid of Mx+2/x O, Al2O3 and SiO2 vary with the ratio XAl = Al2O3/(Mx+2/x O+Al2O3) in glasses containing 50 mol% SiO2 (**Fig. 7**). Upon increasing XAl, the partial molar Vm of SiO2 barely vary. That of Al2O3 decreases from ~ 38-40 cm3 mol-1 down to 35 cm3 mol-1 when XAl becomes larger than 0.3. Vm Al2O3 appears to be slightly lower in calc-alkaline systems than in alkaline ones when XAl < ~0.3. The partial Vm of Na2O and K2O both significantly increase of ~3 and 5 cm3 mol-1, respectively, when XAl becomes higher than 0.3. On the contrary, the partial Vm of MgO and CaO do not vary significantly with XAl.

Similar observations can be made for partial Cpliquid values (**Fig. 7**). Whereas that of SiO2 barely vary, those of MgO and CaO slightly and gradually increase of 6 J mol-1 K-1 as XAl increases. The partial Cpliquid values of Na2O and K2O show distinct decreases of 12 and 20 J mol-1 K-1, respectively, as XAl increases from 0.3 to 0.5. In parallel, in alkali melts, the partial Cpliquid Al2O3 slightly decreases from ~111-113 J mol-1 K-1 down to 106-107 J mol-1 K-1 as XAl increases from 0.0 to 0.3, then increases back to ~113-115 J mol-1 K-1 as XAl further increases. In calcalkaline melts, Cpliquid Al2O3 is significantly higher, and nearly linearly increases with XAl from 122-127 J mol-1 K-1 to 134-135 J mol-1 K-1.

Based on previous works (e.g., Richet et al., 1993; Cormier and Neuville, 2004; Le Losq et al., 2014), we hypothesize that the observed Vm changes in alkali system occur as K+ and Na+ forms compensating complexes with AlO45- tetrahedral units: this results in variations in the CNs of the alkali metal cations that may induce variations in their partial molar volume. For alkaline-earth metal cations, we expect limited variations in their CNs (e.g., Cormier and Neuville, 2004; Gambuzzi et al., 2015; Deng et al., 2022), explaining the relatively small variations in their partial *Vm*. In parallel, the change, or lack of change in the local environments of alkali and alkaline-earth cations also seems related to the variations, or lack of variations of their partial *Cpliquid*values with *XAl*.

The case of Al2O3 is more complex. First, there is a large dependence of partial Al2O3 Cpliquid to temperature: the higher glass transition temperatures of Al-rich and alkaline-earth bearing melts naturally incur higher values of Al2O3 Cpliquid. Besides, Al in CN 5 and 6 is detected in tectosilicate and peraluminous glasses (McMillan and Kirkpatrick, 1992; Stebbins et al., 2000; Toplis et al., 2000; Neuville et al., 2008b; Stebbins et al., 2008; Thompson and Stebbins, 2011, 2012, 2013; Le Losq et al., 2014), and may further induce variations in its partial contributions to Vm and Cpliquid. To test the latter hypothesis, we used the compilation of the fractions of Al in CN 4, CN 5 and CN 6 in magnesium and calcium aluminosilicate glasses made by Neuville and Le Losq (2021), and report the cumulated fraction of Al in CN 5 and 6 against Al2O3 partial molar Vm and Cpliquid at Tg. (**Fig. 8**). While no systematic relationship is observed between partial Al2O3 *Cpliquid* and the fraction of [5]Al and [6]Al, a systematic trend is observed for Al2O3 partial molar volume. In general, Al2O3 *Vm* seem to tend toward the value of 25.575 cm3 mol-1 for corundum, where Al is in CN 6.

The above discussion indicates that changes in the partial molar *Vm* of oxide components in glasses occur as the CNs of cations evolves with glass composition. This process also should be particularly important in melts, in which changes in the coordination numbers of cations are occurring not only as a function of composition, but also as a function of temperature (e.g., Allwardt et al., 2005a; Kanehashi and Stebbins, 2007; Neuville et al., 2008a; Le Losq et al., 2014) and pressure (e.g., Allwardt et al., 2005b; C. Sanloup et al., 2013; Chrystèle Sanloup et al., 2013; Drewitt et al., 2015; Sanloup, 2016; Lee et al., 2020). Therefore, systematic and precise predictions of melt molar volume and density, a key area to solve questions related to the presence and behavior of deep silicate melts in the inner Earth near the mantle transition zone or the mantle-core boundary (e.g., see Sanloup, 2016), requires further knowledge regarding the links between cationic environment and molar volumes. For a given cation, combining i-Melt predictions of cationic *Vm* with experimental data regarding the cationic environment may result in producing interesting data to better constrain the links between the cation partial molar volume, local atomic environment and glass/melt density.

**5. Conclusion**

The new implementation of i-Melt, a greybox model combining artificial neural networks with physical equations, allows systematic and precise predictions of alkali and alkaline-earth aluminosilicate melt and glass properties, including configurational entropy, liquid heat capacity and partial molar contributions from the different oxide components, configurational heat capacity, glass transition temperature, fragility, viscosity, density and partial molar volumes of oxide components, optical refractive index and Raman spectra. Reliable uncertainty estimates are now provided with using MC dropout.

The new abilities of the model, i.e. the prediction of partial oxide values for glass molar volumes and liquid heat capacities, allow better understanding the links between the role and environment of cations in glasses and melts, and variations in melt/glass properties. For instance, at fixed SiO2 but varying Al/Na2O, Na2O and Al2O3 partial glass molar volumes and liquid heat capacities at Tg vary as a function of the role of Na in the glass network (i.e., if acting as a network modifier or charge compensator), as well as a function of the fractions of Al in 4, 5 and 6 fold coordination. Such a demonstration is only a glimpse into the possibilities offered by machine-learning powered models such as i-Melt. Their development will help understanding the properties and structure of melts under various conditions. The open source and free nature of i-Melt implies that future developments also can greatly benefit from inputs of users. Contributions can range from sending new data for their integration in the database to code development.

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**Author contributions:** CLL designed the study, collected the data, performed Raman and viscosity experiments, and developed the deep learning framework and the associated computer code. AV and BOM helped in the design of the neural network. CLL and AV worked on improving and testing of the computer code. CLL, BOM and DN performed Raman measurements. CLL and AV drafted the manuscript. All authors contributed to the final version of the manuscript.

**Competing interests:** Authors declare no competing interests.

**Materials & Correspondence:** All the data are available in the main text or the supplementary materials. The computer code to reproduce the results of this study is available as a Python library at the web address [https://github.com/charlesll/i-melt](https://github.com/charlesll/neuravi). Correspondence can be addressed to the corresponding author.

Table 1: Root Mean Square Error (RMSE) and Median Absolute Error (MAE) between predictions and measurements.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Data subset |  | Training | Validation | Testing |
| Viscosity, log10 Pa·s (Adam-Gibbs theory) | RMSE | 0.3 | 0.3 | 0.3 |
| MAE |  |  |  |
| Viscosity, log10 Pa·s (Free Volume theory) | RMSE | 0.3 | 0.3 | 0.3 |
| MAE |  |  |  |
| Viscosity, log10 Pa·s (Vogel-Tamman-Fulcher equation) | RMSE | 0.3 | 0.4 | 0.3 |
| MAE |  |  |  |
| Viscosity, log10 Pa·s (MYEGA equation) | RMSE | 0.2 | 0.3 | 0.3 |
| MAE |  |  |  |
| Viscosity, log10 Pa·s (Avramov-Milchev theory) | RMSE | 0.2 | 0.4 | 0.3 |
| MAE |  |  |  |
| Density, g cm-3 | RMSE | 0.01 | 0.01 | 0.02 |
| MAE |  |  |  |
| Raman spectra (%, relative RMSE) | RMSE | 20 | 19 | - |
| MAE |  |  |  |
| Refractive index | RMSE | 0.003 | 0.005 | 0.006 |
| MAE |  |  |  |
| CTE, 1e-7 K-1 | RMSE | 1.1 | 1.0 | 1.2 |
|  | MAE |  |  |  |
| Elastic Modulus, GPa | RMSE | 4 | 8 | 6 |
|  | MAE |  |  |  |
| Abbe number | RMSE | 1.2 | 0.5 | 4.0 |
|  | MAE |  |  |  |
| Liquidus temperature | RMSE | 12 | 16 | 12 |
|  | MAE |  |  |  |

Table 2: Percentage of data comprised within the 2-sigma MC dropout uncertainties

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
|  |  | Training | Validation | Testing |
| Density | MC Dropout –  1 network | 0.96 | 0.95 | 0.94 |
|  | MC Dropout –  ten network |  |  |  |
| Viscosity | MC Dropout –  1 network | 0.93 | 0.93 | 0.95 |
|  | MC Dropout –  10 network |  |  |  |

Table 3: Average partial molar volumes Vm and liquid heat capacities Cpliquid of oxide components calculated using the density and heat capacity datasets. Important error bars indicate compositional dependence of those values, and do not reflect model uncertainties. Ranges of reported values are from (Bottinga et al., 1983; Lange and Carmichael, 1987; Liu, 2006; Neuville and Le Losq, 2022) for partial molar volumes and (Richet and Bottinga, 1985; Courtial and Richet, 1993) for partial liquid heat capacities.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
|  | Partial molar volume, cm3 mol-1 | | | | Liquid heat capacity, J mol-1 | |
| Oxide | 2.5th | 50th | 97.5th | Range of reported values | Average model value | Reported value |
| SiO2 |  | 25.8 |  | 26.0 - 27.5 | 79 ± 2 | 81.37 |
| Al2O3 |  | 38.2 |  | 37.0 – 39.0 | 105 ± 10 +  0.026 ± 0.006 \* T | 130.2 +  0.0357 \* T |
| Na2O |  | 23.5 |  | 25.0 – 29.0 | 98 ± 5 | 100.6 |
| K2O |  | 37.5 |  | 40.0 – 46.0 | 66 ± 10 +  0.017 ± 0.003 \* T | 50.13 +  0.01578 \* T |
| MgO |  | 13.5 |  | 11.0 – 13.0 | 81 ± 10 | 85.78 |
| CaO |  | 15.9 |  | 14.0 – 18.0 | 89 ± 5 | 86.05 |

**Figure 1.** Datasets for melt viscosity **(a)**, glass Raman spectroscopy **(b)**, glass density **(c),** glass refractive index **(d),** liquid heat capacity **(e)**, Glass Abbe Number **(f)**, glass Elastic Modulus **(g)**, glass coefficient of thermal expansion **(h)**, and liquidus temperature **(i)**.Each symbol corresponds to a sample. The glass-forming domain at usual laboratory cooling rates is indicated in grey.

**Figure 2.** Examples of the numbers of compositions including the different oxide components in the viscosity and density datasets.

**Figure 3.** Predictions against measured values for the different available observations. Blue squares, orange circles and green diamonds are used for distinguishing the train, validation and testing data subsets.

**Figure 4.** Examples of predicted Raman spectra (red dashed curves) represented on top of the measured ones (black solid lines) in the training (left) and validation (right) data subset.

**Figure 5.** Glass fragility *versus* melt *Cpconf(Tg)/Sconf(Tg)* ratio. Symbols are predictions of the deep learning framework on the different subsets of the *Dviscosity* dataset (mean of predictions from 10 models). The dashed line is the relationship observed by Webb (2008, abbreviated W2008 in the figure) using experimental heat capacity data, and the dotted line is that observed by Russell and Giordano (2017, abbreviated RG2017 in the figure). Symbols are coloured according to the melt chemical compositions: black symbols are for melts with more than 95 mol% SiO2, blue ones for SiO2-Al2O3 melts, red ones for calc-alkaline silicate and aluminosilicate melts, cyan ones for alkaline silicate and aluminosilicate melts, green symbols for silicate and aluminosilicate melts mixing all metal cations, and magenta symbols are for aluminate melts.

**Figure 7:** Deep learning framework predicted variations in **(a,b)** glass transition temperature *Tg*, **(c,d)** relative density and **(e,f)** refractive index at 589 nm in the upper part (SiO2 > 50 mol%) of the ternary Na (left) and K (right) aluminosilicate systems.

**Figure 8:** Melt fragility, *m*, of melts in the upper part (SiO2 > 50 mol%) of the K2O-Na2O-Al2O3-SiO2 system. Fragility is represented in the upper part of the ternary sodium **(a)** and potassium **(b)** aluminosilicate systems, as well as as a function of the silica fraction and the potassium to total alkali ratio of silicate, peralkaline and tectosilicate melts **(c, d, e, f)**.

**Figure 9:** Configurational entropy at *Tg*, *Sconf(Tg),* of melts in the upper part (SiO2 > 50 mol%) of the K2O-Na2O-Al2O3-SiO2 system. *Sconf(Tg)* vary non-linearly with oxide contents in the ternary diagrams **(a)** Na2O-Al2O3-SiO2 and **(b)** K2O-Al2O3-SiO2. In silicate melts **(c)**, a mixed alkali effect (MAE) is observed upon Na-K mixing. It disappears as [Al]/[Na+K] increases **(d, e, f)**.

**Figure 10:** Melt and glass properties vary in a complex way with glass network topology. i-Melt reveals that parameters such as **(a)** the viscous glass transition temperature *Tg* and **(b)** *BFV,*an activation energy term in the Free Volume theory (eq. 2)**,** correlate broadly with R*Raman*. Other terms also show more complex variations, influenced by cationic mixing interactions and steric hindrance effects, such as the glass configurational entropy *Sconf(Tg)* **(c)**or the free volume parameter *CFV* **(d).** Each symbol represents the calculation for a randomly generated composition (n=10,000) in the glass-forming domain of the Na2O-K2O-Al2O3-SiO2 system (Fig. 1).

**Figure 11:** Spearman correlation matrix between the different variables predicted by i-Melt or calculated from those predictions. The large the circles, the larger the correlation. A correlation of 1 or -1 means a perfect monotonic (possibly non linear) correlation. Spearman correlation coefficients were calculated from the predictions for the 10,000 randomly generated composition in the glass-forming domain of the Na2O-K2O-Al2O3-SiO2 system (inset in Fig. 10).

**Figure 12:** High temperature viscosity limit. **(a)** histograms of the high temperature viscosity limits *AVFT*, *Ae*, *AFV* and *AAM* predicted for 10,000 melt composition randomly selected from the glass-forming domain of the Na2O-K2O-Al2O3-SiO2 system. **(b)** *Ae* *versus* Al/(Al+Si) diagram highlighting a possible compositional dependence of *Ae*. The ternary diagram shows again the randomly selected compositions.

**Figure 13: Influence of Al and K/(K+Na) ratios on rhyolite structure and properties. (a)** configurational entropy maps as a function of the ratio K/(K+Na) and the rheological agpaitic index, calculated as (Na2O + K2O + CaO + MgO + FeO)/(Al2O3 + Fe2O3). On top of the maps, symbols of rhyolite effusive (purple circles) and explosive (black squares) eruptions from Di Genova et al. (2017) are represented. Some scatter in the Sconf(Tg) contour map is visible and results from model noise. The cyan to pink line is a compositional transect used in (b) to (d). **(b)** Raman spectra predicted for melts along the cyan-pink transect shown in (a). **(c)** Example of peak-fitting of the Raman spectra with four gaussian peaks (see text). **(d)** Evolution of the NBO/T calculated from melt composition (dashed grey line) and from *Q3* peak areas, converted using the Q3 Raman cross-section from Mysen (2007).

**Figure 14: Influence of the K/(K+Na) ratio on the viscosity and atomic structure of alkali aluminosilicate melts, mimicking the evolution from a basanite-like mafic pole to a phonolite-like pole. (a)** Viscosity (log10 Pa·s) at 1100 °C of the melts as a function of their silica mol fraction. Two trends are visible for Na-rich melts with K/(K+Na) = 0.33, and K-rich melts with K/(K+Na) = 0.66. **(b)** and **(c)** Predicted evolution of the Raman spectra of Na-rich and K-rich melt compositions, respectively. Mean predictions are generated feeding the 10 models with 100 melt compositions generated linearly between the end-member poles (see text for details).