Deep learning **model to predict the structure and properties of aluminosilicate glasses and melts**

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**Abstract**

The way aluminosilicate lavas flow and degas drives the dynamics of volcanic eruptions. In parallel, in industrial furnaces, similar aluminosilicate melts are used to produce glass, and their properties drive glass-forming processes and end-product characteristics. Despite such importance, no general model allows predicting the molecular structural, thermodynamic and viscous properties of aluminosilicate melts. Here, a deep learning framework that combines a deep artificial neural network with thermodynamic equations is used for understanding and predicting melts and glasses properties, including viscosity, optical refractive index, density, and Raman signals. Trained on alkali aluminosilicate compositions, it helps to understand, for instance, the link between nano-scale changes in lava composition/structure and the dynamic of mega-scale eruptions of silicic supervolcanoes like Yellowstone (U.S.A.). This is a glimpse into the possibilities offered by this approach, which provides a new way to build models of material properties for various applications.

**1. Introduction**

How do molten silicates move? How do they exchange heat with other media? How do they crystallize? These fundamental questions underpin many practical problems, including the dynamics of volcanic eruptions (Dingwell, 1996) , the formation of rocks (Bowen, 1956), and the manufacturing of novel glass, glass-ceramic and ceramic materials. Addressing them requires knowledge of physical properties such as viscosity, heat capacity and entropy, which are ultimately governed by the liquid’s composition and associated atomic/ionic structure (see for reviews Le Losq et al., 2019b; Mysen and Richet, 2019). While models of liquid and glass heat capacity exist (Stebbins et al., 1984; Richet and Bottinga, 1985; Richet, 1987; Russell and Giordano, 2017), other properties, such as viscosity, are more difficult to model due to their strong and non-linear dependence on intensive and extensive variables, particularly in the supercooled temperature domain near the glass transition domain that separates melts from glasses. Among such properties, viscosity is of great interest due to its influence on the dynamic of volcanic processes (Dingwell, 1996; Papale, 1999; Gonnermann and Manga, 2013; Gonnermann, 2015; Cassidy et al., 2018) and its importance for glass manufacturing [REF?].

Predictions of a property like viscosity can be performed, at present, with using (i) empirical models, (ii) thermodynamic ones, or (iii) molecular dynamics (MD) simulations. Focusing on viscosity, empirical models (Bottinga and Weill, 1972; Shaw, 1972; Persikov, 1991; Hess and Dingwell, 1996; Hui and Zhang, 2007; Giordano et al., 2008; Duan, 2014) rely on empirical equations, such as the Arrhenius or the Tammann-Vogel-Fulcher (TVF) equations, for predicting the melt viscosity. Those models are interpolative in nature, and bring no information about the links between structural, thermodynamic and dynamic properties of silicate melts. In term of precision, they usually claim errors of 0.6 log Pa·s or higher, but predictive uncertainties can reach much higher values in practice (e.g., Le Losq and Neuville, 2013).

Thermodynamic models solve some of the problems affecting empirical approaches. Regarding viscosity, initial models were limited to mixtures of specific melts composed of only a few oxide elements (Richet, 1984; Neuville and Richet, 1991; Neuville, 2006). Recent advances in the knowledge of the links between melt structure, thermodynamic properties and viscosity now allow building more complex models. For instance, the model of Le Losq and Neuville (2017) for melts in the Na2O-K2O-SiO2 system links experimental viscosity, structure in term of tetrahedral SiO4 *Qn* speciation (*Q* being a tetrahedral unit and *n* the number of bridging oxygen it carries; *4-n* thus gives the number of non-bridging oxygens), heat capacity and configurational entropy. This model, based on the Adam and Gibbs theory of relaxation processes (Adam and Gibbs, 1965, see section 2.5.2), brings important information about the viscous flow process in melts, and predicts precisely the viscosity (errors ση lower than 0.2 log Pa·s) and the structure of melts in the entire glass-forming domain of the Na2O-K2O-SiO2 phase diagram. Another thermodynamic model, combining an associate solution model of melt structure with the Avramov-Milchev equation of viscous flow (Avramov and Milchev, 1988), was proposed by Starodub et al. (2019) for the Na2O-K2O-Al2O3-SiO2 diagram. This model is a very interesting attempt that suffers from an important general difficulty that affects all structure-thermodynamic models: it is based on an internal structural model that remains unverified by experiments because the *Qn* speciation in Al-bearing silicate melts is difficult to determine as Al-Si interactions broaden the 29Si Nuclear Magnetic Resonance spectroscopy signals (e.g., Mysen et al., 2003). This difficulty and many other complex structural details, such as excess non bridging oxygens or highly coordinated Al species (see for a review Le Losq et al., 2019b), affects the extension of thermodynamic models to complex magmatic and industrial melt compositions. This explains why thermodynamic models exist only for compositions containing only a few oxide components.

In parallel of practical empirical and thermodynamic models, MD simulations (Rapaport, 2004) also allow us to infer the structure and properties of complex melts. They bring important structural, dynamic and thermodynamic information by simulating atomic movements over picoseconds timescales

(Guillot and Sator, 2007; Vuilleumier et al., 2009; Bauchy et al., 2013; Wang et al., 2014; Dufils et al., 2017). They allow observing how melt behaves at high to very high temperatures, how atoms move, interact, and how this affects their physical properties like viscosity and density. However, predictions in temperature ranges of volcanic eruptions (typically 700-1300 °C) are not possible, and the calculation timescale is very limited (tens of picoseconds at most for classical MD models, and shorter timescales for *ab initio* calculations).

The above discussion rises the following question: can we build a model that links our knowledge of the joint variations of intensive variables such as temperature (*T*), melt composition (*x*), structure and physical properties, in order to perform accurate predictions of the melt properties of interest for volcanology and material sciences? In this publication, we show that this is possible. We propose the i-MELT model that combines theoretical equations and deep learning to predict melt properties under a wide range of realistic circumstances. It further enables systematic investigation of the fundamental links between material composition, structure and properties. As a concrete example, we focus on melt and glass compositions in the K2O-Na2O-Al2O3-SiO2 system, central to understanding the activity of silicic supervolcanoes like Yellowstone or Long Valley (U.S.A.) (Di Genova et al., 2017), and for which a fairly complete dataset is available (Fig. 1). Viscosity is key to this problem (Di Genova et al., 2017), but the underlying structural and thermodynamic controls have yet to be properly characterized. A better understanding of – and ability to predict – the properties of alkali aluminosilicate glasses is also valuable beyond volcanology. For example, it can aid the development of technological glass materials such as smartphone screens (Varshneya and Bihuniak, 2017).

**2. Methods**

***2.1 Experimental Design***

Developing the model required the collection and compilation of viscosity, density, refractive index data, and Raman spectra for glasses and melts in the K2O-Na2O-Al2O3-SiO2 quaternary diagram (Fig. 1). The viscosity of supercooled melts for peralkaline compositions in this diagram remains poorly-understood, and we conducted additional experiments to complement the existing dataset. We further compiled existing data as specified below, prior to developing the i-MELT framework in the Python programming language using the PyTorch library. The codes can be run using Jupyter Notebooks. All codes and data necessary to reproduce this study can be found on the software repository Github at the web address <https://github.com/charlesll/neuravi>.

***2.2 Datasets***

Existing Raman spectra and observations of optical refractive index, density and viscosity of alkali aluminosilicate glasses were selected by hand via a review of the existing literature. Cross-validation of the accuracy of viscosity data from different studies is critical and was checked on compositions including sodium trisilicate, albite and jadeite. Publications presenting deviations larger than 0.1 log Pa·s compared to the general literature trend on such compositions were discarded. Density and refractive index come from various publications reviewed in Mazurin et al. (1987). Raman spectra are published data from the IPGP and Carnegie Institution for Science laboratories (see below for details). Four different streams of data are thus present:

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

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

- *DRaman*, the dataset of Raman spectra, composed of *XRaman* chemical composition entries (mole fractions) and *yRaman* spectra observations (min-max scaled Raman intensities);

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

*Dviscosity*, *Ddensity* and *Doptical* cover an important part of the glass-forming domain of alkali aluminosilicates (Fig. 1); they were thus used to train the artificial neural network with a performance oriented mindset. *DRaman* covers a more limited set of compositions (Fig. 1). It was used as a way of improving multitask learning as well as a way of introducing structural information in the deep learning framework.

***2.3 Sample synthesis and viscosity-density measurements***

To extend the viscosity dataset for peralkaline aluminosilicate melts, new compositions were synthesized at IPGP in Paris from reagent-grade K2CO3, Na2CO3, Al2O3 and SiO2 dried oxide powders, following the protocol described in Le Losq and Neuville (2013). Viscosity and density measurements follow the standard protocol used in the Géomaterial laboratory at IPGP and described in (Neuville, 2006; Le Losq and Neuville, 2013; Le Losq et al., 2014). Chemical compositions (Table 1) have been measured using a Cameca SX50 electron microprobe, with a 30 nA current, U = 30 kV, and 5 seconds of counting. Beam-induced alkali loss was minimized by working with a defocused beam that was moved continuously during the analysis. The mean and standard deviation values reported in Table 1 are calculated from 10-20 individual measurements on each sample. The corresponding viscosity measurements are provided in Table 2, and are affected by an error lower or equal to 0.03 log Pa·s.

***2.4 Raman spectroscopy***

Raman spectra of silicate and aluminosilicate glasses acquired at IPGP in Paris were recorded using a T64000 Jobin-Yvon® Raman spectrometer equipped with a confocal system, a 1024 charge-couple detector (CCD) cooled by liquid nitrogen and an Olympus® microscope. The optimal spatial resolution allowed by the confocal system is 1-2 μm2 with a ×100 Olympus® objective, and the spectral resolution is 0.7 cm-1. A Coherent® laser 70-C5 Ar+, having a wavelength of 488.1 or 514.532 nm, has been used as the excitation line. Unpolarized Raman spectra were acquired between 20 and 1500 cm-1 on pieces of glass from the starting materials that were excited with a laser power of 100-150 mW on the sample.

Additional Raman spectra acquired at the Geophysical Laboratory on glasses along the K2Si4O9-K2(KAl)4O9 and K2Si4O9-K2(KAl)4O9 joins were added to the database. Those spectra were acquired with a Dilor XY confocal microRaman spectrometer equipped with a cryogenic Thompson Model 4000 CCD. The 488 nm line of a SpectraPhysics model 2025 Ar+ laser operating at several hundred mW at the sample was used for sample excitation.

Preprocessing of the spectra was kept to minimum: (i) a linear baseline was adjusted to the minima in the 700-800 and 1200-1300 cm-1 portions of the spectra and then subtracted to obtained baseline-corrected spectra, (ii) the spectra were then corrected from temperature and excitation line effects (see details and references in Le Losq and Neuville, 2013; Le Losq et al., 2014), and (iii) the spectra were normalised to their maximum intensity such that the intensity in each spectrum varies between 0 and 1. Only signals in the 400-1250 cm-1 range were retained as different spectra had different starting and ending Raman shift values. After pre-processing, spectra were saved in a HDF5 file for their future use.

***2.5 Deep learning model***

*2.5.1 Overview*

The i-MELT framework (Fig. 2) combines a deep artificial neural network with various dynamic and thermodynamic equations. This strategy allows developing an intelligent model that links together different observables from the same object (melt/glass). The artificial neural network is a feed forward network with multiple interconnected hidden layers (Murphy, 2012; Goodfellow et al., 2016), it either predicts directly-observable glass properties including density, refractive index and Raman spectra, or outputs the latent variables (such as configurational entropy, *Sconf*, a property that reflects the melt structure) required to predict properties like melt viscosity*.* In the present case, this offers the ability to predict the melt viscosity within five theoretical and empirical models commonly used for reproducing experimentally-observed variations of viscosity with temperature for silicate melts: Adam-Gibbs, MYEGA, Avramov-Milchev, Tamman-Vogel-Fulcher and Free Volume Theory. We will present the possibility of performing such trans-theoretical predictions in the next section.

*2.5.2 Trans-theoretical predictions*

No consensus exists regarding a “best theory” to explain and reproduce the variations of liquid viscosity with parameters such as temperature, pressure or composition. Despite this, some models may be preferred. For instance, the Adam-Gibbs theory (Adam and Gibbs, 1965) has been particularly successful in reproducing relaxation and viscosity data of silicate melts (Richet, 1984; Scherer, 1984; Neuville and Richet, 1991; Bottinga et al., 1995; Bottinga and Richet, 1996). It assumes that liquid movements occur through cooperative molecular re-arrangements; viscosity (*η)* can be expressed as a function of temperature (*T)* and composition (*x*) via

, (*1*)

with *Ae* representing a high-temperature limit, *Be* a term proportional to the energy barriers opposed to molecular re-arrangements, and *Sconf* and *Cpconf* the melt configurational entropy and heat capacity, respectively. *Tg* is the glass transition temperature, at which melt is frozen-in into glass upon quench. For convenience, we adopt hereafter the empirical definition of *Tg* as equal to the temperature for which *η* = 1012 Pa·s.

Alternatively, one might adopt the Free Volume theory (Cohen and Grest, 1979; Cohen and Grest, 1984), which states that melts present liquid-like and solid-like molecular cells, their mobility being ensured by atomic diffusivity within/between liquid-like cells. This takes the form

**,** (*2*)

with again representing the high-temperature limit, and , and latent variables related to free volumes’ properties like percolation threshold, size and numbers.

Beyond the Free Volume and Adam-Gibbs models, many other theories have been proposed to describe the viscous flow of liquids. Among those, some are empirical like the Tamman-Vogel-Fulcher (TVF) equation:

, (*3*)

with ATVF, BTVF and T1 adjustable parameters. The semi-empirical model proposed by Avramov and Milchev (1988) (AM) also relates viscosity to temperature as

, (4)

with AAM a pre-exponential terms proportional to , *Tg(x)* and *m(x)* the melt glass transition temperature and fragility. The MYEGA equation (Mauro et al., 2009) directly derives from eq. 1, and takes the form:

**,** (5)

with *Ae*  a pre-exponential term proportional to that was taken as equal to that in the Adam-Gibbs theory (eq. 1) because the MYEGA equation is a daughter product of the Adam-Gibbs theory. Equations 3 to 5 remain empirical or semi-empirical as unlike the Adam-Gibbs or Free Volume equations, they are not expressed in terms of measurable physical quantities, such as heat capacity. However, they still represent the viscosity dependence upon temperature very well.

While the equations presented above are popular for silicate melts, no strong consensus towards any one model appropriate for all liquids exists. In fact, some of those models rely on very different theoretical backgrounds. The i-MELT approach circumvents the problem of choosing one particular theory by proposing a trans-theoretical approach: the artificial neural network simultaneously enables predictions across multiple theories, and is trained to provide best average performance across the selected theories. i-MELT also provides optimal common values for parameters that appear in multiple theories, such as the glass transition temperature *Tg*.

*2.5.2 Data preparation: Train-Validation-Testing split and standardisation*

Overfitting refers to the fact that a model reproduces very well its training dataset, but fails at making reliable predictions on new, unseen datasets. This is a common problem affecting machine learning models. Several strategies are deployed in this work to prevent overfitting. The first one is to monitor this phenomenon. To do so, the available datasets were split in three different, randomly chosen *training*, *validation* and *testing* subsets (Fig. 1). During the training process, the *training* subset was used for training the model while the *validation* subset was used for monitoring overfit and trigger early stopping. The final predictive abilities of the trained neural networks were then evaluated using the unseen *testing* data subset. In the present study, the data were randomly separated by composition (Le Losq et al., 2019a) to avoid the pitfall of having the same composition in the different *training*, *validation* and *testing* subsets (a phenomenon known as ‘data leakage’). While *Dviscosity, Doptical* and *Ddensity* were each separated in three splits following the above protocol (Fig. 1), *DRaman* was divided in only two *train* and *validation* subsets due to its small size. 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 predictive capacity of the trained neural network and to introduce structural knowledge.

After train-validation-test splitting, an important step in any machine learning data preprocessing is standardization of the data. In practice, appropriate data scaling is often essential to obtaining good convergence within algorithms (Goodfellow et al., 2016). The goal of re-scaling is to promote feature variations close to unity and ensure that all features have comparable numerical ranges: failure to do so tends to lead to instabilities in the gradient back-propagation process that is central to training neural networks. In the present study, we have implemented a custom approach. All chemical compositions inputs are in mole fractions, which corresponds to a modification of min-max (0-1) scaling. Raman spectra were normalised to be comprised between 0 and 1. Viscosity, density and refractive index were not scaled, as scaling the outputs was not found to affect network convergence. However, as we will see, when outputs are unscaled it is essential to properly initialise the bias of the output layer of the neural network to match the expected range of the predictions to be made, as done for instance for Mixture Density Network (Bishop, 2006).

After pre-processing, the different scaled *training*, *validation* and *testing* data subsets were saved in HDF5 files for their future use.

*2.5.3 i-MELT model technical implementation*

i-MELT is implemented in the Python programming language, using the Pytorch machine learning library (Paszke et al., 2019). It takes four inputs: the mole fractions of SiO2, Al2O3, Na2O and K2O. These are fed into to a neural network composed of *k* hidden layers, each one having a given number of activation units (a.k.a neurons). Changing the number of activation units between different hidden layers in a single network did not improve training convergence or final predictive abilities, such that we chose to keep thesamenumber of activation units in the different hidden layers for simplicity. Having explored various alternatives, we adopted the now-popular rectifier function (Glorot et al., 2011) as the activation function of activation units, so that an activation unit receiving input *x* returns output *y = max(0,x)*. The outputs of the hidden layers were finally fed into two output linear layers. The first output layer returns vectors that we regard as Raman spectra, calculated from the linear sum of the last neural network hidden layer. The second output linear layer returns 16 different values:

- the parameters *Ae*, *AAM*, *AFV* and *ATVF* (eqs. 1 to 5), as well as the coefficients *B1* to *B3* and *C1* to *C3* of the Sellmeir equation (see eq. 6 below) for the calculation of the glass refractive index *n* are directly given by the linear outputs; and

- the natural logarithms of *Sconf(Tg)*, *CFV*, *Tg*, *To*, *T1*, the melt fragility *m*, and the glass density *d*.

The use of the logarithm in the latter case was inspired by a strategy proposed by Bishop (2006) for Mixture Density Networks: it ensures that quantities are assigned positive values in accordance with their physical meaning. We also found that it aids rapid convergence during training. One trick for this method to work is to properly set the initial values of the biases of the last output layer to realistic values when creating the network. Other terms like *Be,BFV and BTVF*  were calculated from eqs. 1 and 5 and the knowledge of the other parameters.

Neural network predictions can be used in equations 1 to 5 to obtain trans-theoretical predictions of melt viscosity, and also directly provide different observables like glass density, glass transition temperature or Raman spectra. Furthermore, the coefficients *B1* to *B3* and *C1* to *C3* are used to predict the refractive index at given wavelength, , via the Sellmeier equation:

*. (6)*

Our neural network therefore allows us to input chemical compositions and obtain predictions for:

* melt viscosity, within five distinct theoretical or empirical frameworks,
* glass transition temperature,
* latent variables like configurational entropy and fragility,
* glass density,
* glass refractive index as a function of wavelength, and
* glass Raman spectra.

These predictions depend on a large number of tuneable parameters embedded within the neural network. During network training, we use our database of observed glass properties to optimize these parameters, seeking good average predictive performance.

*2.5.4 Training i-MELT*

During training, we monitored the least-square deviations between measurements and predictions for the viscosity from eqs. 1 to 5 as well as density, optical and Raman spectra. We further added loss functions for known viscous *Tg* and *Sconf(Tg)* values in the dataset *Dviscosity*. This enabled to better constrain the estimates of *Sconf(Tg)* because this parameter is hard to evaluate as equation 1 does not have a well-defined, unique solution but multiple optimal solutions due to strong correlations between *Be* and *Sconf(Tg).* This strong correlation originates from the intervention of the intrinsic entropy *Sc\** of the molecular subunits involved in the melt viscous flow / relaxation process in both *Be* and *Sconf(Tg)*(Adam and Gibbs, 1965; Toplis, 1998; Toplis, 2001).

Batch training was performed using the Adam optimizer with a learning rate of 0.001, and monitoring the global loss on the *training* and *validation* data subsets. Early stopping (Goodfellow et al., 2016) was used to avoid overfitting: when the global loss function on the *validation* data subset stopped decreasing for more than 50 epochs, training was stopped and the network presenting the best validation loss was saved. Dropout (Srivastava et al., 2014), a method that consists in randomly turning off a given fraction of activation units at each training iteration, was also applied to promote generalisation and reduce overfitting as much as possible.

**3. Results**

***3.1 Optimization of the artificial neural network architecture***

The i-MELT framework uses a feed-forward deep neural network (Fig. 2). Such network feeds its inputs to several fully connected hidden layers composed of a given number of activation units, a.k.a. neurons or perceptrons. Before presenting any results regarding the performance of the model, we first document the optimal architecture and the way we searched for it. This optimal architecture is important as it determines its performance to fit the existing data, its sensitivity to overfitting, and its generalisation ability (i.e. its ability to provide precise and accurate predictions for new, unseen compositions).

The architecture of the hidden layers was optimized via a random search process (Bergstra and Bengio, 2012). We monitored how the number of hidden layers and of hidden activation units affected the neural network overall performance by training 3000 artificial neural networks under the same conditions on the same datasets. We also varied during those trainings the dropout parameter, which affects the *p* percent of neurons per layer turned off at each training iteration in order to prevent overfitting (Srivastava et al., 2014), to observe its influence on the training process. We documented the neural network performances using the Root-Mean-Square-Error (RMSE) between viscosity data predictions and measurements. In parallel, we also generated training datasets with different numbers of samples to observe how the number of samples affects the training of a given neural network architecture. The results of those tests are reported in figure 3.

Figure 3a reveals that the RMSE on the different training, validation and testing datasets become constant with more than ~70 sample compositions. This implies that, in the quaternary alkali aluminosilicate system, data from at least 70 different chemical compositions are necessary to train efficiently the model, i.e. to get a model providing good predictions while avoiding overfitting. Turning to the neural network architecture, the RMSE of the different data subsets all converge to values of ~0.4 log Pa·s for moderately deep neural network with 3 to 5 layers and 200-300 units per layer; accordingly, best performances are in general reached with more than 1000 neurons (Fig. 3b,c,d). Those results confirm that moderately deep neural network still generalizes better than shallow ones on this problem with small datasets. The dropout method, slightly helps preventing overfitting but is not a critical feature in the present case (Fig. 3e). It actually seems that low to very-low dropout values should be preferred: the different of the RMSE between the training, testing and validation data subsets are lowest for *p* < 0.1.

From this random search, we selected the 10 best neural networks with the lowest error on the validation data subset. Given the results presented in Fig. 3, overfitting by each network should be very limited but may be still present. To help further limiting this issue, all reported predictions by i-MELT are calculated from the average of those from the 10 best neural networks. This method is called bagging (Breiman and Breiman, 1996) and promotes generalization (good predictions on new samples) of machine learning algorithms. For i-MELT, generalization was further promoted by the adopted training protocol and by the multi-task learning strategy. Indeed, artificial neural networks learning to predict multiple related features/observables tend to show better prediction abilities compared to those trained to predict only a given task/parameter/feature (Caruana, 1997). The overall adopted training strategy allowed i-MELT to perform good predictions on new samples despite our small experimental datasets.

***3.2 Melt and glass property predictions***

Trans-theoretical predictions of *η* (Figs. 4, 5) are possible with good precision (RMSElower than 0.4 log Pa·s on unseen test data, Table 3; for comparison, the RMSE of the best empirical magma viscosity models typicallyare higher than or equal to0.6 log Pa·s (e.g. Giordano et al., 2008). There is no strong predictive performance differences between the different theories (Fig. 5), except at very low viscosities where predictions through the free volume theory or the TVF equation seem affected by some bias in the training dataset; however, this is not observed in the other validation and testing datasets. Overall, the Adam-Gibbs, MYEGA and Avramov-Milchev equations appears to make the most consistent predictions as no systematic outliers are visible in the RMSE histograms (Fig. 5a,c,d, iv).

The glass properties are also well predicted by i-MELT. Known viscous *Tg* and *Sconf(Tg)* are predicted within 16 K and 0.8 J mol-1 K-1, respectively. The glass density and refractive index are also predicted within 0.05 g cm-3 and 0.005, respectively (Fig. 6, Table 3). For the two latter properties, a few outliers are visible and correspond to extreme compositions along the SiO2-Al2O3 join (Figs. 1, 6c,d) for which only a few data points are available.

***3.3 Structural information through Raman spectra predictions***

In addition to physical and thermodynamic properties, i-MELT has the ability to predict structure-dependent features such as glass Raman spectra. Considering the very small experimental Raman dataset (Fig. 1), global variations of Raman signals have been captured well (Fig. 4b, 6e) and can be predicted within ~18 % (average mean absolute deviation between observed and predicted spectra from the validation data subset). i-MELT thus embeds structural information, and allows estimation of structural parameters from Raman spectra, including the ratio of intra- and inter-tetrahedral aluminosilicate vibrations, *RRaman*. This ratio is calculated from the Raman intensities *IR* observed at the Raman shifts ω, *IRω,* as :

. (7)

The numerator integrates the signals assigned to bending/stretching of *Qn*-*Qn* intertetrahedral vibrations in the glass network (Bell et al., 1968; Sen and Thorpe, 1977; Furukawa et al., 1981; McMillan, 1984), and the denominator those assigned to stretching of Al-O and Si-O bonds in *Qn* units (Brawer and White, 1975; Brawer and White, 1977; Virgo et al., 1980; Furukawa et al., 1981; Mysen et al., 1982; McMillan, 1984; Neuville et al., 2014). The integration boundaries were selected looking at all the spectra plotted together, and correspond to common limits that delimitate the intertetrahedral *Qn*-*Qn* and intratetrahedral *Qn* vibrational regions. While some small changes could be make in some cases, our selection of common boundaries for all glass Raman spectra robustly captures the general trend.

The addition of network modifier metal cations in silica glass is accompanied by increases in the fractions of depolymerised *Qn* units (like Q2 and Q3 units) and in parallel in the number of non- bridging oxygens per tetrahedral unit (NBO/T); this results in a strong decrease in the ratio *RRaman* (Giordano and Russell, 2018). *RRaman* thus can serve as a proxy for the glass network connectivity and topology, i.e. the 3D connectivity of the polyhedral SiO2-AlO2 network (e.g., Greaves et al., 2011). The higher *Rraman* is, the higher the aluminosilicate network connectivity, the lower the NBO/T. Because of such link, *RRaman* is linked to variations in melt properties (Giordano and Russell, 2018). i-MELT can predict *Rraman* within 15 %. We see that there is a strong gap in *Rraman* between ~2 and ~4, the latter value corresponding to that of the silica glass. Consequently, only one spectrum (that of silica) constrains the model at high *RRaman* values. This is because the dataset lacks spectra above 90 mol% SiO2, as glasses are difficult to obtain experimentally in this range. Indeed, unmixing problems exist along the M2O-SiO2 binaries at high silica concentrations (M= Li, Na, K) (e.g., Jarry and Richet, 2001), and high liquidus temperatures make the synthesis of glasses difficult for Al-bearing compositions (Schairer and Bowen, 1955; Schairer and Bowen, 1956). Nevertheless, information from new experiments in silica-rich melts could be important to bring information to the model about melt/glass structural behavior between an extreme composition like SiO2 and multicomponent melts/glasses.

***3.4 Model internal consistency***

The ability to predict the melt fragility *m* allows further testing of the internal consistency of i-MELT. Indeed, experimental data indicate that a direct correlation between *m* and the ratio between the configurational heat capacity at *Tg* and *Sconf*(*Tg), Cpconf(Tg)/Sconf(Tg)* shall be observed (Webb, 2008; Russell and Giordano, 2017). i-MELT predicts this linear correlation (Figure 7). This indicates that the model is internally consistent (Russell and Giordano, 2017). Some scatter is visible, and most probably arises from the way melt and glass *Cpconf* values are determined in the model. Indeed, i-MELT internally calculates *Cpconf(Tg)* as *Cpliquid(Tg) – Cpglass(Tg),* with *Cpliquid(Tg)* predicted from the model of *(Richet and Bottinga, 1985)* and *Cpglass(Tg)* calculated from the Dulong-Petit limit of 3R, with R the perfect gas constant*.* Those *Cpliquid(Tg)* and *Cpglass(Tg)*  predictions are affected by some errors, explaining the scatter observed in Fig. 7.Other studies that used experimental values of *Cp* observe a better correlation (Webb, 2008; Russell and Giordano, 2017). In turn, this result indicates a critical need for better heat capacity models of aluminosilicate melts*.*

**4. Discussion**

The developed framework offers unique opportunities to objectively explore the links between chemical, structural, thermodynamic and dynamic parameters in materials.Such task is traditionally done by humans, and this situation is far from being ideal as we are influenced by various biases (Kaptchuk, 2003). The use of machine learning may be a new way of limiting such caveat. Here it allows performing systematic predictions of melt and glass properties. It thus is a novel and additional tool to perform a systematic exploration of the links between melt composition, molecular structure and physical/thermodynamic properties.

***4.1 i-MELT and the exploration of composition-structure-property links***

i-MELT allows performing a systematic exploration of the links between different observed and latent variables. Through this way, we can for example document the known division between topological (i.e, the geometry and interconnectivity of the T-O-T network, with T= Si,Al) and chemical effects (i.e., effects resulting from mixing different cations in similar sites) on different properties (Neuville and Richet, 1991). We observe that *RRaman*, hence the glass network topology, correlates with the glass transition temperature (Fig. 8a), and with quantities proportional to energy barriers opposed to ionic mobility in melts like the *BFV* term of the Free Volume viscosity equation (Fig. 8b, eq. 2). This agrees very well with the general knowledge of the influence of the topology/connectivity of the aluminosilicate network on melt transport properties (e.g., see reviews of Mysen and Richet, 2019; Le Losq et al., 2019b).

However, in details, we observe some compositional mapping in the *RRaman* *versus* glass transition temperature *Tg* plot (Fig. 8a). Such compositional sensitivity probably originates from metal cation chemical mixing effects that can affect *Tg* (Isard, 1969; Day, 1976)*.* While cationic mixing effects only slightly influences properties directly linked to the aluminosilicate network connectivity (Le Losq and Neuville, 2017), they strongly affect properties influenced by cationic interactions and steric hindrance effects, such as the configurational entropy *Sconf(Tg)* (Fig. 8c) (Richet, 1984; Neuville and Richet, 1991; Neuville and Mysen, 1996; Le Losq and Neuville, 2017). i-MELT results demonstrate that chemical mixing effects also affect the term *CFV* (eq. 2, Fig. 8d). This is explained by the fact that this term encompasses local cationic influences on melt free volumes in the Free Volume theory (Cohen and Grest, 1979). Results actually suggest a link between *CFV* and *Sconf(Tg)*, as corroborated by a spearman correlation coefficient of 0.93 between the two variables.This agrees with the possibility of building a free volume version of the Adam-Gibbs viscosity equation, as discussed below in section 4.2.

***4.2 Links between the Adam-Gibbs and the Free Volume theories***

The trans-theoretical character of i-MELT allows observing the link between two theories: Adam-Gibbs and Free Volume. In the Free Volume theory, solid-like and liquid-like molecular cells are distinguished and separated by a critical volume *v\**, and viscous flow occurs via cooperative molecular movements between liquid-like cells. In the Adam-Gibbs theory, viscous flow occurs via cooperative motions of molecular segments of a size *z\*(T),* characterized by an intrisic entropy *Sc\**. The two theories thus share some common philosophical background: viscous flow is assumed to occur via some sort of cooperative movements of molecular entities in the melt. This background can be retrieved when diving into the details of the parameters of eqs. 1 and 2. Indeed, *BCG* embeds some structural information because it depends on *v\**:

*BFV = v\* zo, (8)*

where *zo* is an adjustable parameter. Similarly, the ratio *Be/Sconf(Tg)* embeds molecular subunit length-scale information as

*Be/Sconf(Tg) = [ Δµ z\*(Tg) ] / R, (9)*

with *Δµ* the energy barriers opposed to the rearrangement of molecular subunits of size *z\*(Tg)*, and R the perfect gas constant. We can therefore consider *v\** and *z\** as structural parameters embedding information about the volume or length-scale of the cooperative molecular regions. In this case, they should directly depend on melt or glass structure. This is confirmed by the fact that both *BFV* and *Be/Sconf(Tg)* correlate very well with *RRaman* (spearman correlation coefficients of 0.990 and 0.987, respectively). In detail, this implies that it actually should be possible to develop a Free Volume version of the Adam-Gibbs theory, as it has been proposed (Hodge, 1994; Liu et al., 2015). More generally, the links between *BFV,* *Be/Sconf(Tg)* and *RRaman* support the general hypothesis that melt viscous flow occurs when a critical molecular length-scale is reached. This length-scale can be determined from Raman spectra (Fig. 8b) and strongly influences the glass transition temperature *Tg* (Fig. 8a).

***4.3 Configurational entropy of aluminosilicate melts***

Cationic mixing effects result in excess entropy and have a fundamental influence on variables such as *Sconf(Tg)*. Mixing between two cations in silicate and aluminosilicate melts can be random (Neuville and Richet, 1991; Neuville and Mysen, 1996) or not (Lee, 2005; Neuville, 2006), and can occurs between Si and Al “network formers” (Neuville and Mysen, 1996), between “network modifier” metal cations (Richet, 1984; Neuville and Richet, 1991; Lee et al., 2003) or between the charge compensator metal cations that compensate AlO4- units in aluminosilicate compositions (Neuville and Richet, 1991). It also can be non-existant, like between Na and K in tectosilicate melts (Le Losq and Neuville, 2013; Le Losq et al., 2017), or present peculiar variations like those observed between Ca and Na in silicate melts (Neuville, 2006). In all cases, such effect is usually difficult to predict, and even subject to interpretation. i-MELT solves this problem by enabling systematic quantification and visualization of such phenomena. For example, we can observe in Figure 9 how *Sconf(Tg)* varies as a function of the chemical composition of alkali aluminosilicate melts. Increasing Al concentration leads to decreasing *Sconf(Tg)* (Fig. 9a,b). The Al/(Na+K) ratio also largely affects the way Na and K mix: without Al, their interaction results in an entropy excess (Fig. 9c) and, hence, in large decreases in melt viscosity as it is proportional to the inverse of *Sconf(Tg)* (eq. 1). Increasing Al/(Na+K) leads to changing the role of alkali metals in the network (Le Losq et al., 2014). In presence of Al, Na and K segregate in different molecular nano-environments (Le Losq and Neuville, 2013; Le Losq and Neuville, 2017), inducing less and less excess entropy of mixing as Al/(Na+K) increases. As a result, *Sconf(Tg)* varies more and more linearly upon mixing Na and K (Fig. 9d,e,f). Finally, i-MELT predicts small *Sconf(Tg)* values for K-rich and Al-rich melts (Fig. 9b,f), a finding explained by the fact that Al and K respectively promote network polymerisation and nano-structuration in aluminosilicate glasses (Le Losq et al., 2014; Le Losq et al., 2017). The variations in *Sconf(Tg)* with the composition of aluminosilicate melts thus depend largely on (i) how metal cations interact together, (ii) how those interactions are affected by the presence of Al, and by Si-Al interactions (Neuville and Mysen, 1996).

***4.4 Melt fragility***

Melt fragility vary smoothly with the SiO2 and Al2O3 concentrations (Fig. 10a,b). It does not strongly depend on the mixing of Na and K (Fig. 10c,d,e,f). Al-rich melts present generally higher fragilities, a fact that can be explained by the change in the oxygen coordination of Al in Al-rich melts as temperature increases. Indeed, in Al-rich alkali aluminosilicate melts, the Al coordination number increases with increasing temperature (Le Losq et al., 2014). It was proposed that species like Al in five-fold coordination participate in polymerizing the glass network (Neuville et al., 2008) and, as a paradox, may change their role at high temperature and ensure the mobility of strongly polymerized melts (see section 4.3 in Le Losq et al., 2014 and references cited therein). Such change in the role of Al can explain why the fragility of Al-rich melts appears to be particularly high in comparison with those of depolymerised Al-free silicate melts (Fig. 10a,b).

***4.5 Glass properties***

i-MELT predicts the viscous glass transition temperature well (Fig. 6a), and allows systematic exploration of the variations of this parameter with melt composition (Fig. 11a,b). Its variations are complex as it can be expected, because they depend on melt structure and are also affected by chemical mixing effects (Fig. 8a). Glass density or optical refractive index display simpler variations with glass composition. Interestingly, fragility correlates well with the glass density (spearman correlation coefficient of 0.908). Similar variations are thus visible when comparing melt fragility and glass density in the ternary sodium and potassium aluminosilicate diagrams (Figs. 10, 11). Glass density largely depends on the concentrations of Na2O and Al2O3 added in SiO2. The variations in the glass optical refractive index show a different pattern, the addition of Al2O3 having a greater effect than that of Na2O at comparable molar contents (Fig. 11). The optical refractive index actually does not correlate strongly with any of the thermodynamic/dynamic variables. The strongest spearman correlation observed is that with *RRaman*: it is equal to -0.581. It reflects that, while glass structure has a slight effect on its optical properties, this variable is mostly controlled by the electronic properties of the atoms present in the glass.

***4.6 Volcanological implications***

i-MELT puts us in a position to understand why eruptions of silicic volcanoes may be more explosive if the magma is rich in K and Al (Di Genova et al., 2017). Indeed, among the usual critical parameters driving the dynamic of volcanic eruptions such as pre-eruptive volatile content, degassing path and nano-cristallization (Andújar and Scaillet, 2012; Di Genova et al., 2017; Cáceres et al., 2020), explosive or effusive eruptions of silicic volcanoes respectively appear to involve lavas more or less rich in Al and K (Figure 12). Most of the lavas emitted at silicic volcanic systems, like Yellowstone or Long Valley (U.S.A.), present dry chemical compositions made of more than 95 % of Na2O, K2O, Al2O3 and SiO2. Melts in the Na2O-K2O-Al2O3-SiO2 system can thus be considered as simplified analogues of the lavas involved in silicic volcanic eruptions, such that i-MELT can be used to glimpse the links between eruptive dynamics and magma composition, structure, and properties.

From model predictions, the transition between effusive and explosive silicic eruptions correlates with a decrease in *Sconf(Tg)* driven by ongoing network connectivity and nano-structuration as [Al] and [K] increase (Fig. 12). The chemical separation between effusive and explosive eruptions at silicic volcanic centers is associated, according to the present work, with limits in *RRaman* and *Sconf(Tg)* of ~1.5 and ~8.0 J mol-1 K-1, respectively. However, it should be emphasize that the structural and associated entropic transition, which drive an important increase in melt viscosity (eq. 1), is actually smooth. This observation raises the question of a direct link between melt structure and the eruptive dynamic; an indirect link could be seen in the way the Al/(Na+K) ratio affects the speciation and valence of iron in aluminosilicate melts (Le Losq et al., 2020). Indeed, an increase in the Al/(Na+K) ratio of aluminosilicate melts, in addition to strongly affecting melt polymerisation and properties (Fig. 12), promotes iron reduction (Dickenson and Hess, 1982). Such phenomenon could promote the rapid apparition of iron nanolites, and hence acts as an accelerator in the increase of magma viscosity and promote the explosivity of the eruption. (Cáceres et al., 2020; Di Genova et al., 2020).

In any case, those results demonstrate that a more complete version of i-MELT, embedding the effects of Ca, Mg, Fe and water, would provide the ability to explore and quantify even further the links between magma composition, structure, properties, and volcanic eruptive styles.

**5. Conclusion**

In conclusion, the results presented here highlight how a deep learning framework like i-MELT can be used for practical property predictions in the Earth sciences. Here, it reveals the fine structural and thermodynamic controls on magma viscosity, which, in turn, affects the dynamics of volcanic eruptions. The presented model further has the ability to predict glass properties, and can be used for the design of new glass materials. i-MELT can readily be extended to include quantities of interest across a range of domains and applications, like glass toughness and hardness. More generally, the present results demonstrate that the approach we adopted, based on the combination of machine learning with physical/thermodynamic models, can offer new perspectives on long-standing problems in many disciplines.

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**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/neuravi> (open access will be provided upon acceptance). Correspondence can be addressed to the corresponding author.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| **Glass name** |  | **%SiO2** | **%Al2O3** | **%K2O** | **%Na2O** | **Density, g cm-1** |
| KA80.05 | nom. mol% | 80.00 | 5.00 | 15.00 | 0.00 |  |
|  | nom. wt% | 71.40 | 7.60 | 21.00 | 0.00 |  |
|  | an. wt% | 74.8(4) | 7.6(1) | 15.1(2) | 0.00(4) | 2.320(1) |
| KA72.07 | nom. mol% | 72.00 | 7.00 | 21.00 | 0.00 |  |
|  | nom. wt% | 61.60 | 10.20 | 28.20 | 0.00 |  |
|  | an. wt% | 61.4(3) | 10.2(2) | 27.4(3) | 0.00(2) | 2.408(1) |
| KA65.09 | nom. mol% | 65.00 | 8.75 | 26.25 | 0.00 |  |
|  | nom. wt% | 53.70 | 12.30 | 34.00 | 0.00 |  |
|  | an. wt% | 53.3(5) | 12.5(4) | 31.7(3) | 0.00(3) | 2.451(9) |
| NA65.09 | nom. mol% | 65.00 | 8.75 | 0.00 | 26.25 |  |
|  | nom. wt% | 60.79 | 13.89 | 0.00 | 25.32 |  |
|  | an. wt% | 61.7(4) | 13.7(2) | 0.03(2) | 24.5(7) | 2.472(4) |
| NA58.10 | nom. mol% | 58.00 | 10.50 | 0.00 | 31.50 |  |
|  | nom. wt% | 53.55 | 16.45 | 0.00 | 30.00 |  |
|  | an. wt% | 54.6(3) | 16.4(2) | 0.05(2) | 28.9(4) | 2.502(5) |

**Table 1.** Composition of the synthesized glasses. Nominal (nom.) and analyzed (an.) compositions are reported. Standard deviations on measured values on 10 different spots (for EPMA measurements) or glass chips (for density measurements) are given in parenthesis (1σ confidence interval).

|  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **T, K** | **KA80.05** | **T, K** | **KA72.07** | **T, K** | **KA65.09** | **T, K** | **NA65.09** | **T, K** | **NA58.10** |
| 1013.1 | 9.10 | 921.5 | 9.37 | 941.3 | 9.55 | 834.0 | 9.01 | 827.3 | 10.10 |
| 1001.8 | 9.32 | 891.0 | 10.17 | 935.1 | 9.71 | 829.0 | 9.18 | 836.9 | 9.73 |
| 989.6 | 9.51 | 872.0 | 10.75 | 919.4 | 10.13 | 813.8 | 9.61 | 817.6 | 10.51 |
| 981.6 | 9.78 | 852.0 | 11.40 | 913.6 | 10.32 | 803.2 | 9.94 | 796.2 | 11.42 |
| 967.6 | 10.05 |  |  | 898.7 | 10.78 | 798.6 | 10.09 | 805.7 | 10.97 |
| 949.7 | 10.50 |  |  | 892.0 | 10.96 | 787.5 | 10.50 | 847.1 | 9.36 |
| 940.3 | 10.83 |  |  | 882.1 | 11.28 | 779.9 | 10.81 | 855.9 | 9.09 |
| 928.2 | 11.05 |  |  | 867.3 | 11.86 | 773.7 | 11.07 | 828.3 | 10.01 |
| 918.4 | 11.32 |  |  | 855.4 | 12.29 | 772.9 | 11.07 | 834.4 | 9.81 |
| 905.1 | 11.63 |  |  |  |  | 762.6 | 11.52 | 787.4 | 11.80 |
| 896.6 | 11.92 |  |  |  |  | 756.3 | 11.74 | 777.4 | 12.36 |
|  |  |  |  |  |  | 752.1 | 11.95 |  |  |

**Table 2.** Viscosity measurements in log10 Pas. Errors on viscosity are lower or equal to 0.03 log10 Pas.

|  |  |  |  |
| --- | --- | --- | --- |
| **Data subset:** | **Training** | **Validation** | **Testing** |
| **Adam-Gibbs (eq. 1, log Pas)** | 0.34 | 0.22 | 0.35 |
| **Free Volume (eq. 2****, log Pas)** | 0.49 | 0.24 | 0.35 |
| **TVF (eq. 3, log Pas)** | 0.40 | 0.25 | 0.38 |
| **MYEGA (eq. 5, log Pas)** | 0.38 | 0.26 | 0.36 |
| **Avramov-Milchev (eq. 4, log Pas)** | 0.35 | 0.24 | 0.35 |
| **Density (g cm-1)** | 0.04 | 0.05 | 0.05 |
| **Raman spectra (%, LAD)** | 18 | 14 | - |
| **Refractive index** | 0.003 | 0.003 | 0.005 |

**Table 3: Root-mean-square errors (RMSE) of the deep learning framework.** RMSE calculated between measured and predicted melt viscosity, density, refractive index, except for Raman spectra where a different metric is used (median least absolute deviation LAD).

**Figure 1.** Melt viscosity **(a)**, glass Raman spectroscopy **(b)**, glass density **(C)** and glass refractive index **(d)** datasets used in this publication.Each symbol corresponds to a sample. The glass-forming domain at usual laboratory cooling rates is indicated in grey.

**Figure 2.** Schematic of i-MELT. An artificial neural network takes input melt composition, and outputs various melt and glass properties. Once trained, relationships between chemistry, structure and properties of melts and glasses can be systematically explored.

**Figure 3.** Neural network architecture and dropout influence on predictive performance. Predictive performance was documented using the RMSE between viscosity predictions (from eq. S1) and measurements in training, validation and testing data subsets. 3,000 neural networks with randomly selected architectures were selected and trained to obtain those results. The effects of the numbers of **(a)** compositions in the training data subset, **(b)** hidden activation units, **(c)** hidden layers, **(d)** number of activation units per layer, and of the dropout probability **(e)** were explored. Subplot (c) is a violin plot with extreme values showed. Subplots (b), (d) and (e) are scatter plots in which each slightly transparent symbol corresponds to a given neural network; less transparence is directly indicative of a higher number of models for a given X-Y value.

**Figure 4.** Prediction examples: **(a)** Melt viscosity can be predicted using various theories with a great accuracy, as shown by examples highlighting the good match between measurements (symbols) and model (curves) predictions from the Adam-Gibbs and Free Volume theories (Supplementary Materials); **(b)** Structural investigation are made through Raman spectra predictions, which compare well with experimental data for albite and sodium trisilicate glasses for instance.

**Figure 5:** Comparison between predicted and measured viscosity in the Na2O-K2O-Al2O3-SiO2 system. Predictions can be made using theories like Adam-Gibbs **(a**, eq. 1) and free volume **(b**, eq. 2), or empirical equations like MYEGA **(c**, eq. 5), Avramov-Milchev **(d**, eq. 4), and Tamman-Vogel-Fulcher **(e**, eq. 3). See table 3 for RMSE.

**Figure 6:** Comparison between predictions and observed viscous glass transition temperature **(a)**, *Sconf(Tg)* (values from Le Losq et al., 2014; Le Losq and Neuville, 2017; Le Losq et al., 2017)values from (Richet, 1984; Neuville and Mysen, 1996; Neuville, 2006; Le Losq and Neuville, 2013; Le Losq et al., 2014; Le Losq and Neuville, 2017; Le Losq et al., 2017) **(b)**, density **(c)**, refractive index **(d)**, and R*Raman* **(e)**. See table 3 for RMSE.

**Figure 7:** 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. The back dotted line is the relationship observed by Webb (2008) using experimental heat capacity data. Except two extreme outliers that corresponds to Al2O3-SiO2 melts with more than 30 mol% Al2O3, a general good agreement is observed.

**Figure 8:** Melt and glass properties vary in a complex way with glass network topology. **(a)** i-MELT reveals that parameters such as the viscous glass transition temperature *Tg* **(a)** and *BFV,*an activation energy term in the Free Volume theory (eq. 2) **(b),** strongly vary depending on 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 in the glass-forming domain of studied system.

**Figure 9:** Configurational entropy at *Tg* of K2O-Na2O-Al2O3-SiO2 melts. *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)**. Al enrichment generally lead to decreasing *Sconf(Tg)*, and minimizing the MAE. Al- and K-rich melts thus present low *Sconf(Tg)*.

**Figure 10:** Melt fragility variations with composition; fragility is represented in the glass forming domains 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)**. No MAE is observed on melt fragility, which depends largely on melt silica content.

**Figure 11:** 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 ternary Na (left) and K (right) aluminosilicate diagrams.

**Figure 12: (a)** structural *RRaman* and **(b)** 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 and explosive eruptions from Di Genova et al. (2017) are represented. Some scatter in the maps is visible during transitions, it originates from some model noise and the fact that the color transitions are numerically sharp.