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

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**Abstract**

Aluminosilicate melts and glasses are of paramount importance for geo- and materials sciences. They include most magmas, and are used to product a wide variety of everyday materials, from windows to smartphone displays. Despite this importance, no general model with which to predict the molecular structural, thermodynamic and viscous properties of aluminosilicate melts exists. To address this, we introduce a deep learning framework, ‘i-Melt’, which combines a deep artificial neural network with thermodynamic equations. It is trained to predict 19 different latent and observed properties of melts and glasses in the K2O-Na2O-Al2O3-SiO2 system, including configurational entropy, viscosity, optical refractive index, density, and Raman signals. Viscosity can be predicted in the 100-1015 log Pa·s range using five different theoretical frameworks (Adam-Gibbs, Free Volume, MYEGA, TVF, Avramov-Milchev). The precision is better than 0.4 log Pa·s on unseen data. Density and optical refractive index (through the Sellmeier equation) can be predicted with errors of 0.05 and 0.005, respectively. Raman spectra for K2O-Na2O-Al2O3-SiO2 glasses are also predicted, with a mean error of ~ 15 %. This relatively high error is due to the limited number of examples available for training. Latent variables can also be predicted with good precisions. For example, the glass transition temperature, *Tg,*can be predicted to within 16 K, while the melt configurational entropy at the glass transition can be predicted to within 0.8 mol-1 K-1.

i-Melt can be used as a tool to explore systematically how structural and thermodynamic variables vary together with changes in melt/glass composition, and how different variables are correlated. We further use the model to help the understanding of the apparent links between changes in the molecular structure of silicic rhyolite lava and the dynamics of silicic volcanic eruptions, such as those at Yellowstone (U.S.A.), for example.

Our deep learning framework represents a new tool to help understand and link experimental data to theory, and to allow behaviours to be predicted across a variety of applications, from material sciences to geochemistry.

**1. Introduction**

How do molten silicates move? How do they exchange heat with other media? How do they crystallize? Questions such as these underpin many practical problems, ranging from the dynamics of volcanic eruptions (Dingwell, 1996) and the formation of igneous rocks (Bowen, 1928), to the manufacture of novel glass, glass-ceramic and ceramic materials, including the development of enhanced technological glass materials suitable for smartphone screens (Varshneya and Bihuniak, 2017). To address such questions, knowledge of melt and glass physical properties, such as viscosity, heat capacity and entropy, is necessary. These properties, in turn, ultimately are governed by the liquid’s composition and, therefore, atomic/ionic structure (see for reviews Le Losq et al., 2019b; Mysen and Richet, 2019). Some properties, such as silicate melt and glass heat capacity, can be reasonably predicted with existing models (e.g., see for heat capacity Stebbins et al., 1984; Richet and Bottinga, 1985; Richet, 1987; Tangeman and Lange, 1998; Webb, 2008; Giordano and Russell, 2017). However, other properties are more difficult to model. This includes viscosity, because of its complex dependence on temperature and melt composition. Silicate melt viscosity variations with temperature*, T*, are, in most cases, non-Arrhenian. At constant T, viscosity can exhibit large and non-linear variations with changing melt composition, particularly if T is in the supercooled temperature domain. This domain is located well below the liquidus, close to the glass transition temperature, which separates melts from glasses. Such a situation is unfortunate because viscosity is of great interest: it influences not only volcanic processes (Dingwell, 1996; Papale, 1999; Gonnermann and Manga, 2013; Gonnermann, 2015; Cassidy et al., 2018), but also glass-forming processes in the glass manufacturing industry.

Currently, predictions of a property such as viscosity rely on (i) empirical models, (ii) thermodynamic models, or (iii) molecular dynamics (MD) simulations. Empirical models are interpolative in nature, and bring no information about the links between structural, thermodynamic and dynamic properties of silicate melts. They can be sufficiently precise enough, and thus, can be very useful, to accurately predict some properties, such as the glass and melt heat capacities (Stebbins et al., 1984; Richet and Bottinga, 1985; Richet, 1987; Tangeman and Lange, 1998; Russell and Giordano, 2017). In the case of viscosity, empirical models rely on empirical equations (Bottinga and Weill, 1972; Shaw, 1972; Persikov, 1991; Hess and Dingwell, 1996; Hui and Zhang, 2007; Giordano et al., 2008; Duan, 2014), such as the Arrhenius or the Tammann-Vogel-Fulcher (TVF) laws. In term of precision, these empirical models usually claim errors of 0.6 log Pa·s or larger, but predictive uncertainties can reach much higher values in practice (e.g., Robert et al., 2013; Le Losq and Neuville, 2013; Sehlke and Whittington, 2016; Di Genova et al., 2017). They provide a practical way of making viscosity predictions, but their usefulness is restricted by the chemical composition and temperature range within which the models have been calibrated. In addition, these models do not provide any further information about the flow process.

Thermodynamic models circumvent the short-comings of empirical models, particularly. their interpolative nature and the lack of physical/thermodynamic background that prevents one to understand the processes underlying melt flow. Initially, thermodynamic models for viscosity were limited to mixtures of specific melts comprising only a few oxides (e.g., Richet, 1984; Hummel and Arndt, 1985; Neuville and Richet, 1991). Analogous attempts were made to model other properties, such as the model of Mysen (1995) which calculates the configurational heat capacity of silicate melts from their fractions of tetrahedral SiO4 *Qn* units (*Q* being a tetrahedral unit and *n* the number of bridging oxygen it carries; *4-n* thus gives the number of non-bridging oxygens). Recent works further leveraged the recent advances in our knowledge of links between melt structure, thermodynamic properties and viscosity, to construct more complex thermodynamic models of the properties of silicate melts, either directly from the melt chemical composition or based on knowledge of their structure. For example, Sehkle and Whittington (2016) proposed a model, based on the Adam-Gibbs theory of relaxation processes (Adam and Gibbs, 1965, see section 2.5.2), to predict the viscosity of tholeiitic melts with an average error of 0.13 log Pa·s, on the 100-1012 Pa·s range. Le Losq and Neuville (2017) also proposed a model based on the Adam and Gibbs theory of relaxation processes. Their model connects viscosity, heat capacity, configurational entropy, structure and chemical composition for melts in the Na2O-K2O-SiO2 system and allows viscosity predictions with an average errorlower than 0.2 log Pa·s on the 100-1012 Pa·s range. A third example is the model proposed by Starodub et al. (2019). It combines an associate solution model of melt structure with the Avramov-Milchev equation of viscous flow (Avramov and Milchev, 1988; Avramov, 2011) to predict melt viscosity on the 100-1012 Pa·s range in the Na2O-K2O-Al2O3-SiO2 system.

Structure-thermodynamic models, like those discussed above, can be precise and provide important information about the links between composition, structure and properties. However, such models suffer from an important drawback: a good knowledge of melt structure is required. While this can be achieved for silicate melts through, for instance, Nuclear Magnetic Resonance (NMR) spectroscopy (e.g., Maekawa et al., 1991), such information is difficult to obtain for aluminosilicate compositions. For example, in their study of the repartition of Al between *Qn* tetrahedral units, Mysen et al. (2003) modeled the 29Si NMR spectra of aluminosilicate glasses. Their results highlight the challenges of such modeling: the numerous Al-Si interactions broaden the 29Si NMR signals. Such problems make it difficult to determine with confidence the *Qn* speciation in Al-bearing silicate melts. Raman spectroscopy may also be used (McMillan, 1984; Matson and Sharma, 1985; Merzbacher and White, 1991; Mysen, 1999), as testified by successful results for alkali silicate and aluminosilicate glasses (Mysen, 1990, 2007; Mysen and Frantz, 1992; Neuville and Mysen, 1996; Malfait et al., 2007; Zakaznova-Herzog et al., 2007; Koroleva et al., 2013; Nesbitt et al., 2021). However, the variations of the Raman peak cross sections, which are needed to convert Raman peak areas in *Qn* unit fractions, are not known as well as would be desired. Furthermore, the peak-fitting protocols can be subject to discussion, as shown by a lack of consensus visible when comparing different studies (e.g., compare the methods described in the studies of B. O. Mysen et al., 1982; Mysen, 1990; You et al., 2005; Malfait, 2009; Le Losq and Neuville, 2013; Le Losq et al., 2014; Bancroft et al., 2018; Nesbitt et al., 2019, 2021). In any case, models that only rely on *Qn* fractions are incomplete because many other structural details affect the properties of aluminosilicate melts. These include, but are not limited to, changes in Al coordination with composition, temperature and pressure (Stebbins et al., 2000, 2008; Toplis et al., 2000; Allwardt et al., 2005a, b; Kiczenski et al., 2005; Neuville et al., 2007; Massiot et al., 2008; D. R. Neuville et al., 2008; Le Losq et al., 2014; Morin et al., 2014; Drewitt et al., 2015), Al-Si ordering between tetrahedral units (Lee and Stebbins, 1999; Lee, 2005; Lee et al., 2016; Allu et al., 2018), or excess non-bridging oxygens (Stebbins and Xu, 1997; Stebbins et al., 1999; Oglesby et al., 2002; Iuga et al., 2005; Thompson and Stebbins, 2011, 2012, 2013; Xiang et al., 2013). From the above non-exhaustive depiction of the current state of our knowledge of melt structure, it appears that our incomplete knowledge of the complex polyhedral melt/glass structure currently limit the extension of thermodynamic models to multicomponent magmatic and industrial melt/glass compositions.

MD simulation (Rapaport, 2004) is another pathway that allows us to infer the structure and properties of complex melts. Such models bring important structural, dynamic and thermodynamic information by simulating atomic movements over picosecond timescales (Guillot and Sator, 2007; Vuilleumier et al., 2009; Bauchy et al., 2013; Wang et al., 2014; Dufils et al., 2017). They provide models that can be used to assess how melt behaves at temperatures typically higher than 2000 K, how atoms move and interact, and how this affects their physical properties such as viscosity and density. While predictions are informative about processes at super-liquidus conditions and can be useful for high-temperature applications (e.g., in glass furnace or in planetary magma oceans), they are less helpful at lower temperatures, for example in the 700 - 1300 °C temperature range typical of volcanic eruptions. Furthermore, MD simulation simulates the system on very short timescales, of tens of picoseconds at most for classical MD models, with even shorter timescales for *ab initio* calculations. This is very far from glass-making conditions, and extremely far from volcanological timescales which span a wide range, from minutes to several thousands of years, and sometimes beyond.

From the above discussion, there is no general framework that links variations of temperature (*T*), pressure (*P*), melt composition (*x*), structure and physical properties, in order to perform accurate predictions of the melt properties of interest for volcanology and material sciences. Here, we show that such a model can be derived by leveraging the use of physics-guided neural networks (PGNN). The PGNNs combine physical equations with artificial neural networks. This brings advantages in comparison to both traditional physical/thermodynamic models and “pure” machine learning models. PGNNs leverage our current knowledge of physical systems by integrating existing physical equations, but add the power of machine learning to infer connections between variables not yet understood theoretically – for instance, between glass chemical composition and configurational entropy. PGNNs have been successful in many applications (Willard et al., 2020), including the analysis of seismic waveforms (Ren et al., 2020) and lake temperature modeling (Karpatne et al., 2018). This has inspired recent efforts to model the viscosity of ionic liquids with neural networks (Paduszyński and Domańska, 2014; Beckner et al., 2018). For silicate melts, Cassar (2020) recently proposed the ViscNet model, a PGNN model that combines either the VFT or the MYEGA viscosity equations (see section 2.5.2) with a neural network to perform viscosity predictions of silicate and aluminosilicate melts. Such results are very encouraging and showcase the ability of PGNN to provide pragmatic, practical models for property predictions.

Here, we go a step further by presenting a PGNN model that combines several physical equations with a deep learning neural network (named i-Melt), and predict many different melt and glass properties of interest for geology and industry, including melt viscosity and configurational entropy, glass density, optical refractive index and Raman spectra. i-Melt is thus a PGNN “multitask” model, which has the ability to predict different features/properties of the same object: a silicate melt/glass. i-Melt was trained on melt and glass compositions in the K2O-Na2O-Al2O3-SiO2 system, for which a fairly complete, albeit sparse, experimental dataset is available. In this system, i-Melt allows systematic exploration of the links between melt/glass composition, structure and properties as it will be presented in the following.

**2. Methods**

***2.1 Experimental Design***

The development of the deep learning model requires the collection and compilation of viscosity, density, refractive index data, and Raman spectra for glasses and melts in the K2O-Na2O-Al2O3-SiO2 quaternary system (Fig. 1). The viscosity of supercooled melts for peralkaline compositions in this system is not well 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, by using the PyTorch library. All codes and data necessary to reproduce this study can be accessed using Jupyter Notebooks, available from Github at [https://github.com/charlesll/](https://github.com/charlesll/neuravi)i-melt.

***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. Validation of the accuracy of viscosity data across different studies is critical and was checked on melt compositions including Na2Si3O7, NaAlSi3O8 and NaAlSi2O6. We plotted all together the literature data for such compositions, and observed the mean trend of these data. Most data fall within 0.1 log Pa·s, forming a clear general trend. Published data with deviations larger than 0.1 log Pa·s compared to this general trend were discarded. Density and refractive index come from various publications, in particular from publications reviewed in Mazurin et al. (1987). Raman spectra were published data from the IPGP and Carnegie Institution for Science laboratories (see below for details). All the data and their sources are provided in the database available in the software repository. We thus have four different streams of data:

- *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). These data were, thus, used to train the artificial neural network with a “performance oriented” mindset, i.e. we want the model predictions to be as accurate as possible. *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, additional 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 protocol used in the Geomaterial laboratory at IPGP (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. All measured viscosities were Newtonian – no dependence on the strain rate was observed.

***2.4 Raman spectroscopy***

Raman spectra of silicate and aluminosilicate glasses acquired at IPGP (Paris, France) 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 that were excited with a laser power of 100-150 mW on the sample were acquired between 20 and 1500 cm-1 on pieces of glass from the starting materials.

Additional Raman spectra acquired at the Geophysical Laboratory on glasses along the Na2Si4O9-Na2(NaAl)4O9 and K2Si4O9-K2(KAl)4O9 joins, previously published in Mysen (1996, 1999), 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 for 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 the development of an intelligent model that links 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 such as melt viscosity through five theoretical and empirical equations commonly used for reproducing experimentally-observed variations of silicate melt viscosity with temperature: Adam-Gibbs, MYEGA, Avramov-Milchev, Tamman-Vogel-Fulcher and Free Volume Theory. In the next section, we will present the possibility of performing such *trans-theoretical* predictions, i.e. the ability to predict a given property using different theoretical/empirical frameworks but a single, common artificial neural network. The network predicts, given melt composition, the different parameters of the different theoretical/empirical equations, which, in turn, provide different values of the desired property. This allows one to predict melt viscosity using, for instance, the Adam-Gibbs or Free Volume equations depending on preference, and to compare final predictions as well as observe the connection and correlation between the different variables of the equations.

*2.5.2 Trans-theoretical predictions*

No consensus exists regarding a “best theory” to explain and reproduce the variations of melt viscosity with parameters such as temperature, pressure or composition. Despite this lack of consensus, 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. Here, we adopt the empirical definition of *Tg* as equal to the temperature for which *η* = 1012 Pa·s, and the associated melt relaxation time is of ~100 s. The *Tg* calculated from this definition agrees within 20-30 K with the *Tg* determined from calorimetric measurements (e.g., Russell and Giordano, 2017), which themselves depend on the cooling/heating rates during calorimetric measurements. The adopted *Tg* definition is thus coherent for viscosity modeling, as *Tg* derived from the viscosity data refers to the relaxed melt and its equilibrium structure.

Alternatively, one might adopt the Free Volume theory (Cohen and Grest, 1979, 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, a constant that depends on the molecular volume, a constant that has a dimension of temperature and that should be positive, and a constant identified as the temperature at which continuity of liquid-like cells is reached.

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 TTVF adjustable parameters. We can also cite the Avramov and Milchev (1988) (AM) model, or the MYEGA model (Mauro et al., 2009) that is derived from eq. 1. Both propose equations relating viscosity, temperature as well as *Tg(x)* and *m(x),* the glass transition temperature and the fragility (equal to the slope of the viscosity *versus* temperature curve at *Tg*) of a melt with composition *x*. The AM model is expressed as (Avramov, 2011):

, (4)

with AAM a pre-exponential terms proportional to . Similarly, the MYEGA equation 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 because unlike the Adam-Gibbs or Free Volume equations, these expressions are not expressed in terms of measurable physical quantities, such as heat capacity, for example. However, they do model the viscosity dependence on temperature very well.

While the equations presented above are popular for silicate melts, no over-riding consensus exists towards any one model appropriate for all liquids. 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 is trained to predict melt viscosity using all five theoretical/empirical frameworks (eqs. 1-5). It provides optimal common values for parameters that appear in multiple theories, such as the glass transition temperature *Tg*. As a result, it allows comparison of viscosity predictions between the different theories, and observation of how parameters from the different theories correlate with each other, potentially providing insight into physical inter-relationships.

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

If a model performs well when tested against the training dataset, but fails at making reliable predictions on new, unseen datasets, it is said to be ‘overfitting’. This is a common problem affecting machine learning models. Here, we deploy several strategies to mitigate it. First, we monitor the phenomenon: the available datasets were split into 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 to trigger early stopping. The latter method consists in stopping the training process when the Root-Mean-Square-Error (RMSE), measured between predictions and observations, on the validation data subset stops decreasing and starts diverging from that measured using the training data subset. This allows stopping the training process before the over-fitting phenomenon appears (Goodfellow et al., 2016). The final predictive abilities of the trained neural networks were then evaluated using the entirely 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 glass/melt 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 near unity and to 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 between 0 and 1. Viscosity, density and refractive index were not scaled, as scaling the outputs was not found to affect network convergence. However, when outputs are unscaled, it is essential to initialise the bias of the output layer of the neural network to match the expected range of the predictions to be made, as developed for (e.g.) Mixture Density Networks (Bishop, 2006). After pre-processing, the different scaled *training*, *validation* and *testing* data subsets were saved in Hierarchical Data Format HDF5 files for their future use.

*2.5.4 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 the SiO2, Al2O3, Na2O and K2O components. 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). By changing the number of activation units between different hidden layers in a single network, training convergence or final predictive abilities were not improved. For simplicity, we, thus, chose to keep thesamenumber of activation units in the different hidden layers. 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 are 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 Sellmeier 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*, *TFV*, *TTVF*, the melt fragility *m*, and the glass density *d*.

The use of the logarithm in the latter case was inspired by a similar 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 the use of logarithms to aid rapid convergence during training. Other terms such *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 predict of melt viscosity. The neural network also directly provides different observables such as 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)*

The artificial neural network allows us, therefore, 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 integral to the neural network. During network training, the database of observed glass properties was used to optimize these parameters, seeking values that enable good average predictive performance.

*2.5.4 Training i-Melt*

During training, we monitored the least-square deviations between measurements and predictions for viscosity from eqs. 1 to 5 as well as density, optical refractive index and Raman spectra. Loss functions were also added for known viscous *Tg* and *Sconf(Tg)* values in the dataset *Dviscosity*. This allowed better constraints on the estimates of *Sconf(Tg),* a parameter that is difficult to evaluate because strong correlations between *Be* and *Sconf(Tg)* prevent equation 1 from having a non-ambiguous solution. This correlation originates from the involvement 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, 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, which consists of stopping the training process when the first signs of over-fitting are detected (Goodfellow et al., 2016), was used to avoid overfitting: when the global loss function on the *validation* data subset ceased to decrease for more than 50 epochs, training was halted and the network exhibiting the best validation loss was saved. Dropout (Srivastava et al., 2014), a method that entails randomly turning off a given fraction of activation units at each training iteration, was also applied. This method is known to promote generalization and reduce overfitting.

**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 generalization 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). How the number of hidden layers and that of hidden activation units affected the neural network overall performance was monitored by training 3000 artificial neural networks under the same conditions on the same datasets. During this process, the dropout parameter, *p,* was also varied. This parameterrepresents the percent of neurons per layer turned off at each training iteration (Srivastava et al., 2014). In parallel, training datasets with different numbers of samples were also generated. This allowed observing how the number of samples affects the training of a given neural network architecture. The neural network performances were documented using the RMSE between viscosity data predictions and measurements. 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 examined here, data from at least 70 different chemical compositions were necessary to train the model efficiently, i.e. to get a model providing good predictions while avoiding overfitting.

Regarding the neural network architecture (Fig. 3b,c,d), 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. Therefore, best performance was, in general, reached with more than 1000 neurons in total in the hidden layers (Fig. 3b). Those results indicate that moderately deep neural network generalizes better than shallow ones on this problem with small datasets. The dropout method helps slightly in 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 difference of the RMSE between the training, testing and validation data subsets are lowest for *p* < 0.1.

From this random search, the 10 best neural networks with the lowest error on the validation data subset were selected. Given the results presented in Fig. 3, overfitting by each network should be very limited but may be still present. To limit this issue further, all reported predictions by i-Melt were 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. This, combined with the developed training protocol (see above), allowed i-Melt to provide good generalization abilities. Furthermore, multi-task learning is performed here, as the neural network is trained to predict different features (properties like density or *Tg*, observables like Raman spectra...) from the same objects. This helped further limiting overfitting because 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). We observed this by performing a few tests, training a few neural networks to only predict viscosity. Those resulted in RMSE of ~ 0.5 – 0.6 log Pa·s, higher than those of neural networks trained to predict multiple properties (lower than 0.4 log Pa·s, see below).

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

This overall training strategy results in i-Melt providing good predictions for unseen samples from the testing data subsets not included in the training, despite our small experimental datasets. Trans-theoretical predictions of *η* (Figs. 4, 5) are possible with good precision. The RMSE valuesare lower than 0.4 log Pa·s on the testing data subset (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). Eqs. 1 to 5 all yield similar values (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, such bias is not observed in the other validation and testing datasets. Overall, the most consistent predictions appear to come from the Adam-Gibbs equation along with the related MYEGA and the Avramov-Milchev models, with no systematic outliers visible in the RMSE histograms (Fig. 5a,c,d, iv).

The melt/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 (Fig. 6a,b). The glass density and refractive index are also predicted to within 0.05 g cm-3 and 0.005, respectively (Fig. 6c,d, Table 3). For the two latter properties, a few outliers are visible and correspond to extreme compositions along the SiO2-Al2O3 join (Figs. 1c,d, 6c,d) for which only a few data points are available. This is therefore unsurprising, particularly considering that there is large variations in glass and melt properties along this join (e.g., Okuno et al., 2005; Ando et al., 2018).

***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 Raman spectra of glass. Considering the very small experimental Raman dataset (Fig. 1b), global variations of Raman signals have been well-captured (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 integrals of Raman intensities *IR* observed at the Raman shifts ω, *IRω,* in two regions: between 0 and 670 cm-1, and between 800 and 1300 cm-1. In other terms, we calculate two areas under the curve, *ALW* (integrated intensity in the 0-670 cm-1 range) and *AHW* (integrated intensity in the 800-1300 cm-1 range), and obtain *RRaman* from their ratio:

. (7)

*ALW* 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 *AHW* those assigned to stretching of Al-O and Si-O bonds in *Qn* units (Brawer and White, 1975, 1977; Virgo et al., 1980; Furukawa et al., 1981; Mysen et al., 1982; McMillan, 1984). The integration boundaries were selected by observing all the spectra plotted together. They correspond to common limits that delimitate the frequencies of the inter-tetrahedral *Qn*-*Qn* and intra-tetrahedral *Qn* vibrational regions. While some small changes could be made 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 to silica glass is accompanied by increases in the fractions of depolymerized *Qn* units (like Q2 and Q3 units) and, in parallel, by an increase of the number of non-bridging oxygens per tetrahedral unit (NBO/T, see Mysen et al., 1982). This results in a large decrease in the ratio *RRaman* (Giordano and Russell, 2018). *RRaman* thus can serve as a measure of the glass SiO2-AlO2 network connectivity and topology: 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, as confirmed by the study of Giordano and Russell (2018). i-Melt can predict *Rraman* within 15 %. There is a strong data gap for *Rraman*  between ~2 and ~4 (value corresponding to that of silica glass) because there are very few Raman spectra in the dataset at SiO2 concentrations above ~ 90 mol%, and, above such concentration, *Rraman* varies strongly with silica content. Consequently, only one spectrum (that of silica) constrains the model at very high *RRaman* values. This data gap originates from the difficulty to obtaining samples above ~ 90 mol% SiO2. At such high silica concentrations, unmixing can happen during quench for Na2O-SiO2 compositions (e.g., Jarry and Richet, 2001). In addition, high liquidus temperatures make the synthesis of glasses difficult for Al-bearing compositions (Schairer and Bowen, 1955, 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,* permits further testing of the internal consistency of i-Melt. Indeed, experimental data indicate that a direct correlation between *m* and the ratio of the configurational heat capacity at *Tg* over the configurational entropy at the glass transition*, Cpconf(Tg)/Sconf(Tg),* exists (Webb, 2008; Russell and Giordano, 2017). This is predicted by the Adam and Gibbs theory because (Toplis et al., 1997a):

. (8)

i-Melt predicts this correlation (Fig. 7). The model is, thus, internally consistent because it respects the correlation expected from eq. 8. The model predictions fall between the trends found by the experimental studies of Russell and Giordano (2017) and Webb (2008). The results of those studies predicted a good correlation between *m* and *Cpconf(Tg)/Sconf(Tg).* Here, some scatter is visible. It most probably arises from the propagation of the uncertainties affecting the different predicted values. The *m* and *Sconf(Tg)* predictions are affected by uncertainties, as well as the melt and glass *Cp* calculations. 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)*  calculations are also affected by uncertainties. The combination and propagation of all those sources of uncertainties thus probably explains the greater scatter observed in Figure 7 compared to experimental studies (Webb, 2008; Russell and Giordano, 2017). Finally, some outliers are also visible, and correspond to compositions mostly on the SiO2-Al2O3 join. Along this binary, no supercooled viscosity data are available to constrain the melt fragility, and melt/glass *Cp* predictions probably are affected by important errors. The combination of those two problems probably explains the occurrence of the observed outliers.

**4. Discussion**

The deep learning framework offers opportunities to explore objectively the links between chemical, structural, thermodynamic and dynamic parameters of materials. Beyond this, the framework allows systematic predictions of melt and glass properties that can be of use to address problems in volcanology, such as the link between lava composition and the dynamics of eruptions, or other fields. Below, we will develop a few examples showcasing the information that can be obtained from the deep learning framework.

***4.1 Exploration of composition-structure-property links***

i-Melt allows a systematic exploration of the links between different observed and latent variables. This allows investigating the contributions of 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) to different properties. For example, *RRaman*, and, therefore, the glass network topology correlates with the glass transition temperature (Fig. 8a). It also correlates 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 result agrees very well with the general knowledge of the influence of the topology/connectivity of the aluminosilicate network on melt transport properties (e.g., Bockris et al., 1955; Mysen et al., 1980; Mysen, 1991).

However, the correlation between *RRaman* and glass transition temperature *Tg* is not perfect. Some influence of the glass composition on the *RRaman* *versus* *Tg* relationship is visible in figure 8a. It probably originates from metal cation chemical mixing effects that can affect *Tg* (Isard, 1969; Day, 1976)*.* Indeed, while cationic mixing effects influences only slightly properties directly linked to the aluminosilicate network connectivity (Le Losq and Neuville, 2017), they strongly affect properties such as *Sconf(Tg)* that is influenced by cationic / molecular interactions and steric hindrance effects (Richet, 1984; Hummel and Arndt, 1985; Neuville and Richet, 1991; Neuville and Mysen, 1996; Maehara et al., 2005; Goldstein, 2011). This agrees with predictions for *Sconf(Tg)*. The results in figure 8c show a clear effect of melt composition on the *Sconf(Tg) versus RRaman* relationship*.* Such chemical effects also affect *CFV* (eq. 2, Fig. 8d). This latter term encompasses local cationic influences on melt free volumes in the Free Volume theory (Cohen and Grest, 1979). As a result, it can be expected that mixing different cations will affect this term, explaining the observation made in figure 8d. Results actually suggest a link between *CFV* and *Sconf(Tg)*, as corroborated by a Spearman correlation coefficient of 0.93 between the two variables.

Other properties show interesting correlations. Figure 9 shows the Spearman correlation coefficients, *rs*, between the different variables/properties predicted by the deep learning framework. We observe high correlations between the parameters of the Free Volume and the TVF equations. For example, *BFV* and *BTVF* are highly correlated (rs = 0.995). While *Be* is not strongly correlated with the latter variables, the ratio *Be/Sconf(Tg)* is (rs = 0.996 and 0.999 with *BFV* and *BTVF*, respectively). *BFV* and *BTVF* play the role of some kind of activation energies in eqs. 2 and 3. They thus are related to the energy barriers opposed to the molecular movements at the root of viscous flow. The ratio *Be/Sconf(Tg)* also is related to those energy barriers (see below, eqs. 9, 10). The strong correlations between *RRaman* and *Be/Sconf(Tg), BFV and BTVF* (Fig. 9) thus indicates that the SiO2-Al2O3 aluminosilicate network connectivity and topology mostly controls those energy barriers. This may explain the correlation between the network topology as quantified by *RRaman* and the glass transition temperature of alkali aluminosilicate melts, discussed previously (Figs. 8, 9).

Properties at the denominator in eqs. 1 to 5 show more complex correlations among themselves, and with other variables (Fig. 9). A generally strong correlation is observed between variables at the denominator of eqs. 1 to 5 and the pre-exponential terms reflecting high temperature viscosity limits, namely *ATVF*, *AAM*, *AFV* and *Ae* (Fig. 9). For example, *r*s = -0.93 for the correlation between *Sconf(Tg)* and *Ae*, *r*s = -0.981 for the correlation between *CTVF* and *ATVF*, and *rs* = 0.94 for that between *AAM* and the fragility, *m.* This reflects a numerical correlation between the pre-exponential terms and the denominators of viscosity equations 1-5. A way to avoid such correlations, which can bias calculations, is to set the *ATVF*, *AAM*, *AFV* and *Ae* pre-exponential terms to composition-independent values. Such practice agrees with the general idea that there is a common high temperature viscosity limit (Persikov, 1991; Russell et al., 2003; Giordano et al., 2008; Russell and Giordano, 2017). However, this can be questioned for alkali aluminosilicate melts. Indeed, the study of Robert et al. (2019) suggests that, for alkali tectosilicate melts, *Ae* could vary as a function of the melt Al/Si ratio. This agrees with earlier findings of Toplis (1998), who showed that *Ae* actually varies as a function of the ratio Be/(Al+Si) for various alkali and alkaline earth melt compositions. In the present model, *ATVF*, *Ae, AFV* or *AAM* are allowed to vary with melt composition, such that we can check if the model corroborate the findings of Robert et al. (2019) and Toplis (1998). In Figure 10a, we observe that, for compositions covering a wide compositional field of the glass forming domain (see inset in Fig. 10b), values of *Ae* range between ~ -1.0 and ~ -2.5 log Pa·s, those of *AFV* between ~ -2.0 and ~ -3.5 log Pa·s, those of *AAM* between ~ -0.2 and ~ -1.0 log Pa·s, and those of *ATVF* between ~ -6.0 and -4.0 log Pa·s. The distributions of those parameters are asymmetric and complex. *Ae* and *ATVF* clearly are trimodal, while *AFV* and *AAM* distributions are asymmetric and present sharp terminations on one of their side. This suggests the existence of complex compositional effects. Fig. 10b corroborates this idea, and actually the findings of Robert et al. (2019). There is a general effect of the melt Al/Si ratio on the value of *Ae.* *AFV* also shown variations that correlate, albeit in a complex manner, with Al/Si, while *AAM* do not show systematic variations with Al/Si (not shown). Those results thus corroborate the suggestion that for melts in ternary and quaternary systems, the pre-exponential terms in eqs. 1 and 2 may slightly depend on compositions, and particularly on the Al and Si concentrations and ratios.

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

The above analysis highlighted important correlations between variables from different theories (Fig. 9). The trans-theoretical character of i-Melt allows us to go further: it allows systematic inference for a given property using different theories, and observation of the relationship between the latent variables of these theories. For example, one might explore the links between the Adam-Gibbs and Free Volume theories, and, thus, test the proposition of Hodge (1994) of building a Free Volume version of the Adam-Gibbs theory.

In the Free Volume theory, solid-like and liquid-like molecular cells are distinguished and separated by a critical volume, *v\*.* 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 common philosophical underpinnings, including the important assumption that viscous flow occurs via cooperative movements of molecular entities in the melt. This relationship can be recognized upon consideration of the parameters of eqs. 1 and 2. Indeed, *BFV*  embeds some structural information because it depends on *v\**:

*BFV = v\* zo, (9)*

where *zo* is an adjustable parameter. Similarly, the ratio *Be/Sconf(Tg)* embeds molecular subunit length-scale information as (Toplis, 1998)

*Be/Sconf(Tg) = [ Δµ z\*(Tg) ] / R, (10)*

with *Δµ* the energy barriers opposed to the rearrangement of molecular subunits of size *z\*(Tg)*, and R the perfect gas constant. We can consider *v\** and *z\** as structural parameters embedding information about the volume or length-scale of the cooperative molecular regions. Therefore, these parameters should both depend on melt or glass structure. This is confirmed by the fact that both *BFV* and *Be/Sconf(Tg)* correlate very well with *RRaman* (*r*s = 0.991 and 0.988, respectively, see details in section 4.1 and also Fig. 9). This finding supports the idea that it should be possible to develop a Free Volume version of the Adam-Gibbs theory (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 indirectly observed through Raman signals (Fig. 8b) and strongly influences the glass transition temperature *Tg* (Fig. 8a).

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

In section 4.1, it was highlighted that cationic mixing results in excess entropy and influences, therefore, variables such as *Sconf(Tg)*, leading to its complex dependence on melt composition and structure (Fig. 8c). Mixing between two cations in silicate and aluminosilicate melts can be random (Neuville and Richet, 1991; Neuville and Mysen, 1996) or not (Seifert et al., 1982; Lee, 2005; Neuville, 2006; Le Losq and Neuville, 2013, 2017; Robert et al., 2019). It can occur 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 the electrical charge deficit of AlO4- units in aluminosilicate compositions (e.g., Neuville and Richet, 1991; Robert et al., 2019). Such mixing effects usually are difficult to predict, and subject to interpretation. i-Melt helps solving this problem by enabling systematic quantification and visualization of such phenomena. For example, as seen in Figure 11, *Sconf(Tg)* displays systematic variations as a function of the chemical composition of alkali aluminosilicate melts. Increasing Al concentration leads to decreasing *Sconf(Tg)* (Fig. 11a,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. 11c) and, hence, in decreases in melt viscosity because viscosity is proportional to the inverse of *Sconf(Tg)* (eq. 1). This pattern changes with increasing Al/(Na+K), because as Al is introduced into the glass network, the role of alkali metals changes (see chapters 4 and 8 of Mysen and Richet, 2019). In presence of Al, Na and K are present in different structural environments (McKeown et al., 1985; Jackson et al., 1987; Le Losq and Neuville, 2017), inducing less and less excess entropy of mixing as Al/(Na+K) increases (Le Losq et al., 2017; Robert et al., 2019). As a result, *Sconf(Tg)* varies more and more linearly upon mixing Na and K (Fig. 11d,e,f). Finally, i-Melt predicts small *Sconf(Tg)* values for K-rich and Al-rich melts (Fig. 11b,f), in agreement experimental findings (Richet and Bottinga, 1984; Le Losq and Neuville, 2013; Le Losq et al., 2017; Robert et al., 2019). This is explained by Al and K respectively promoting the polymerization of the melt network (decrease in NBO/T) and the formation of larger cooperative molecular domains involved in the melt viscous flow (e.g., Riebling, 1966; Taylor and Rindone, 1970; Rammensee and Fraser, 1982; Mysen, 1988; Toplis et al., 1997b; Mysen and Toplis, 2007; Xiang et al., 2013; Le Losq et al., 2017). The variations in *Sconf(Tg)* with the composition of aluminosilicate melts, thus, depend mostly on (i) how metal cations interact together, and (ii) on how those interactions are affected by the presence of Al, and by Si-Al interactions.

***4.4 Melt fragility and model extrapolations***

Two of the selected viscosity equations (eqs. 4, 5) share melt fragility as a common parameter in their expressions. Melt fragility is the slope of the viscosity *versus* T curve at *Tg*, and scales with the ratio between *Cpconf* and *Sconf* at *Tg* (eq. 8). In the investigated system, melt fragility varies smoothly with the SiO2 and Al2O3 concentrations (Fig. 12a,b). Increasing melt SiO2 content leads to largely decrease melt fragility, an observation that agrees with previous ones in alkali (e.g., Toplis et al., 1997a) and even alkaline-earth (e.g., Bechgaard et al., 2017) aluminosilicate compositions. Changing the K/(K+Na) ratio does not lead to large changes in fragility (Fig. 12c,d,e,f), in agreement with observations of Robert et al. (2019). At constant silica concentration, depolymerized alkali silicate melts are slightly more fragile than polymerized tectosilicate melts. i-Melt predicts that peraluminous Al-rich melts generally are more and more fragile with increasing Al concentration. However, at ratios of Al/(Al+Na+K) higher than ~ 0.6, the model extrapolates. This observation could indicate that (i) extrapolations are not fully robust and should be considered with care, or (ii) high Al concentrations indeed lead to high melt fragility.

The first question is important to consider, because as “intelligent” as they can be, machine learning algorithms still are interpolative in nature. Therefore, it is not necessarily wise to ask them to perform extrapolations. Here, we tested how the model generally behaves when performing extrapolations beyond the chemical composition range that the training dataset spans by (i) removing some density and optical refractive index data along the SiO2-Al2O3 join and (ii) trying to predict a value for a composition very far from those included in its training dataset, the *Tg of Al2O3*. Regarding (i), the model tends to provide a constant value for density or optical refractive index when asking to make predictions for compositions beyond those included in its training set. This situation is both good and bad. It is good because it indicates that the model does not “explode”, i.e. starts to predict small or large values very different from the mean ones when extrapolating (this is what happens traditionally with polynomial functions, for instance). It obviously is bad because it means that i-Melt will remains mostly interpolative in nature for some parameters like fragility, at least for the moment. Turning to the second test, we first estimated the Al2O3 *Tg* from the data of Secrist and MacKenzie (1965) and Urbain et al. (1982). Secrist and MacKenzie (1965) estimated a viscosity of 4x1010 Pa·s at 1173 K from the rate of crystallization of vapor-formed Al2O3 amorphous films. Fitting the high temperature viscosity data of Urbain et al. (1982) for Al2O3 and this point with equation 3, we have for Al2O3 melt *ATVF* = -2.8 ± 0.1, *BTVF* = 1842 ± 176 and *CTVF* = 1035 ± 19, and *Tg* = 1160 ± 23 K. i-Melt predicts a *Tg* of 963 K for Al2O3. This *Tg* estimation is realistic (the model did not ‘explode’) but significantly below the value calculated from viscosity data. This highlights that extrapolations with models like i-Melt may produce apparently realistic results, but still may be far from the true value.

With limited data, it is inevitable that i-Melt has to extrapolate for some predictions. Following the above discussion, caution is needed. Fortunately, for some properties, results appear to be broadly plausible. This is the case for *Sconf(Tg).* For instance, the model predicts a continuous decrease of *Sconf(Tg)* with increasing Al2O3 in the peraluminous field (Fig. 11a,b), a prediction that agrees with experimental data in sodium peraluminous melts (Le Losq et al., 2014). To conclude, this discussion highlights that, if necessary, machine learning model extrapolations can be performed but should be considered with care, making sure that predictions are realistic compared to the known trends of material properties.

***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. 13a,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. 10a). The model displays the well known decrease of *Tg* with addition of alkali metals, and increase with addition of SiO2, and, on a lesser extent, Al2O3 (Fig. 13a,b). In the potassic aluminosilicate system, there is a local *Tg*maximum near the KAlSi2O6 composition on the SiO2-KAl2O4 binary, at ~ 66 mol% SiO2 (Fig. 13b). This maximum correlates with a maximum in liquidus temperatures (*Tliquidus*). That of leucite KAlSi2O6 is of 1693 °C (Schairer and Bowen, 1955). It corresponds to a local maximum along the SiO2-KAl2O4 binary that correlates with the *Tg* maximum observed in figure 13b. No such maximum in *Tg* is observed on the sodic SiO2-NaAl2O4 binary, in agreement with the absence of a *Tliquidus* maximum along this binary (Schairer and Bowen, 1956). This agrees with the general correlation between *Tg* and *Tliquidus*(e.g., see Sakka and MacKenzie, 1971 and references therein), leading us to suggest that a model such as i-Melt could also predict *Tliquidus*.

Compared to *Tg*, glass density or optical refractive index display simpler variations with glass composition (Fig. 13c,d,e,f). As it is well known, glass density mostly depends on the concentrations of Na2O and Al2O3 added to SiO2. i-Melt reproduces this dependence well. The glass optical refractive index variations show a different pattern, the addition of Al2O3 having a greater effect than that of Na2O at comparable molar contents. The optical refractive index actually does not correlate strongly with any of the thermodynamic/dynamic variables. This is expected because the optical refractive index is mostly controlled by the electronic properties of the atoms present in the glass. Interestingly, the glass density correlates with fragility (rs = 0.908). Similar variations are thus visible when comparing melt fragility and glass density in the ternary sodium and potassium aluminosilicate diagrams (Figs. 12, 13).

***4.6 Volcanological implications***

Using i-Melt, we can address a variety of problems, including volcanological or geochemical questions. As an example, we can look at why eruptions of silicic volcanoes may be more explosive if the magma is rich in K and Al, an observation reported by Di Genova et al. (2017). Indeed, those authors reported a correlation between the explosivity of silicic eruptions and the contents of Al and K in the involved magmas. According to them, this correlation seems to stand regardless of the other many critical parameters driving the dynamic of volcanic eruptions, such as pre-eruptive volatile content, degassing path and nanolite content (e.g., Villemant and Boudon, 1998; Andújar and Scaillet, 2012; Di Genova et al., 2017, 2020; Moitra et al., 2018; Cáceres et al., 2020). In figure 14, the compositions of silicic lavas, compiled by Di Genova et al. (2017), are reported in term of agpaitic index (= [Fe3+ + Al3+] / [Na+ + K+ + Ca2+ + Mg2+ + Fe2+]) and K/(K+Na) ratios. Two clusters of data points are visible. They are associated to effusive and explosive eruptions. In their publication, Di Genova et al. (2017) show that this clustering is primarily controlled by the agpaitic index and the K/(K+Na) ratio of the samples. They observed that water and crystal contents of the melt also play an important role, but those parameters alone cannot explain the observed correlation. The eruptive style of rhyolite eruptions thus seems mainly influenced by small variations in magma composition, linked to the influence of potassium and trivalent cations (Al3+ mainly, but also Fe3+) on the silicate melt rheology.

Here, we can try to explore the thermodynamic origin of this correlation by using i-Melt. Most of the lavas emitted at silicic volcanic systems, such as Yellowstone or Long Valley (U.S.A.), contain more than 95 % of Na2O, K2O, Al2O3 and SiO2. Therefore, melts in the Na2O-K2O-Al2O3-SiO2 system may be considered as simplified analogues of the lavas involved in silicic volcanic eruptions, and i-Melt can be used to glimpse the links between eruptive dynamics and the composition, structure, and properties of magmas. Of course, this will not take the effect of volatile elements into account, but, as highlighted previously, the correlation reported by Di Genova et al. (2017) is actually for dry compositions and, according to them, apparently independent of melt water content. In any case, the following should be considered with care and is here used primarily as an example of use of i-Melt. In the future, more complete versions of models such as i-Melt will allow more robust exploration of the links between eruptive dynamics and the composition, structure, and properties of magmas.

The two data point clusters observed in figure 14 are associated with different *Sconf(Tg)* and *RRaman* values. The data point cluster associated with explosive eruptions present *Sconf(Tg)* values typically below *~ 9* J mol-1 K-1 and *RRaman* above ~ 1.5; the other cluster, associated to effusive eruptions, present values in *Sconf(Tg)* and *RRaman* respectively above and below those values. Decreasing the agpaitic index and increasing K/(K+Na) thus leads to an increase in network polymerization and structuration, hence in fewer available configurations available for viscous flow molecular movements. This results in increasing the melt viscosity, explaining potentially the volcanic eruptive style chemical clustering observed in figure 14. It should be emphasized that (i) the structural and associated entropic transition is actually smooth, and (ii) the role of aluminium is more important than that of potassium: most of the variance in figure 14 is actually associated with variations in the agpaitic index. The first of those two observations actually raise 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 polymerization and properties (Figs. 12a,b, 13a,b), promotes iron reduction (Dickenson and Hess, 1982). Such a phenomenon could promote the rapid appearance of iron-bearing nanolites, and hence act as an accelerator in the increase of magma viscosity to promote the explosivity of the eruption (Cáceres et al., 2020; Di Genova et al., 2020). This will be further promoted by the strong influence of Al on the properties of melt involved in explosive eruptions. Indeed, the explosive/effusive clusters visible in figure 14 mostly are separated by an agpaitic index of 1. Below 1, most metal cations charge balance Al3+ and Fe3+ in tetrahedral coordination, and the mobility of the melt at supercooled conditions is much lower than that of peralkaline melts (e.g., Taylor and Rindone, 1970). Therefore, the correlation observed in figure 14 is not surprising because rhyolite magmas can be peraluminous, with agpaitic index lower than 1. Considering this, the fine chemical control observed in figure 14 will probably not hold for other magma compositions, like andesites or phonolites, that have agpaitic index typically above 1.

In any case, those results showcase the potential of i-Melt. Extending this model to embed the effects of Ca, Mg, Fe and water would provide the ability to explore and quantify further the links between magma composition, structure, properties, and volcanic eruptive styles.

**5. Conclusion**

The results presented here highlight that a deep learning framework like i-Melt can be used for practical property predictions in the Earth and material sciences. The model has the ability to predict many different glass and melt properties. It achieves good precision in the glass forming domain. Extrapolations are possible but should be done with care. We see models like i-Melt, therefore, as a pragmatic compromise between informative, accurate but limited theoretical models and mono-task empirical models. i-Melt can readily be extended to include quantities of interest across a range of domains and applications, including glass toughness and hardness. The inclusion of properties that depend on a system including melt/glass plus other components, like liquidus, seems more challenging at the moment. This could be due to an intrinsic theoretical inconsistency as the model was trained to predict latent and observed variables of a “melt/glass” object, not of a more complex system. However, this should be tested further before any definitive conclusion. In any case, our results demonstrate that the approach of combining machine learning with physical and thermodynamic models offers exciting new perspectives.

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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, 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/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 **(a)** predicted glass transition temperatures and those calculated from undercooled viscosity data, when available, **(b)** *Sconf(Tg)* values from (Richet, 1984; Neuville and Mysen, 1996; Neuville, 2006; Le Losq and Neuville, 2013, 2017; Le Losq et al., 2014, 2017), **(c)** density,**(d)** refractive index, and **(e)** R*Raman*. 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 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). 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. 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)**,** 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 (n=10,000) in the glass-forming domain of the Na2O-K2O-Al2O3-SiO2 system (Fig. 1).

**Figure 9:** 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. Correlations were calculated from the predictions for the 10,000 randomly generated composition in the glass-forming domain of the Na2O-K2O-Al2O3-SiO2 system (Fig. 1).

**Figure 10:** High temperature viscosity limit. **(a)** histograms of the high temperature viscosity limits *ATVF*, *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 11:** 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)**. Al enrichment generally lead to decreasing *Sconf(Tg)*, and minimizing the MAE. Al- and K-rich melts thus present low *Sconf(Tg)*.

**Figure 12:** 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)**. No MAE is observed on melt fragility, which depends largely on melt silica content.

**Figure 13:** 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 14: (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 (red circles) and explosive (black squares) 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.