Title: Deep learning prediction of melt properties

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**Abstract:** (123/125 w)

Knowledge of the links between chemistry, structure and properties of materials is critical to many disciplines, including metallurgy, geology and the glass industry. For instance, the composition of lavas strongly influences their rheology, and, hence, the dynamic of volcanic eruptions. No general model allows revealing the molecular and thermodynamic origin of these properties. Such a model is needed for practical application of experimentally-derived properties often in chemically simple model systems to natural magmatic processes. Here, a deep learning model embedding thermodynamic equations predicts alkali aluminosilicate melts and glasses properties, including viscosity, density, and glass Raman signals. The model captured the links between different quantities and theories. It reveals that the correlation between rhyolite magma composition and eruptive styles finds its roots in large changes in magma structure and configurational entropy. More generally, our approach provides a new way to build models of properties(?) for various applications.

**One sentence summary** (112/125 characters): Deep learning bridges the gap between data and models for practical material property prediction and exploration

**Main Text:**

How do liquids 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 (*1*) , the formation of rocks (*2*), and the manufacturing of novel glass 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. At present, this information is inferred from specific experimental observations and/or thermodynamic or molecular dynamics (MD) simulations (e.g. *3*, *4*); these are usually restricted to a small range of liquid chemical compositions. ~~CALPHAD calculations~~ ~~(~~*~~5~~*~~) and MD simulation~~ ~~(~~*~~6~~*~~) have proved successful for some simple materials (such as binary alloys) or under specific conditions (e.g. very high temperatures in MD, but are not yet able to effectively predict the properties of complex materials, such as oxide glasses, at conditions relevant to Earth and material sciences.~~ Thermodynamic or empirical models may provide an intermediate way of predicting macroscopic properties, but these are usually highly simplified, and limited to predicting a handful of properties in a restricted compositional range (e.g., for viscosity see *7*–*10*). Here, we show that these shortcomings can be resolved by combining multiple theoretical equations using deep learning. This allows prediction of liquid properties under a wide range of realistic circumstances, and enables investigation of the fundamental links between material composition, structure and properties.

As a concrete example, we consider aluminosilicate melts and glasses, materials central to the Earth and material sciences (*11*, *12*). We particularly focus on compositions in the K2O-Na2O-Al2O3-SiO2 system, central to understanding why rhyolitic magmas enriched in Al and in K yield more violent volcanic eruptions (*13*), and for which a fairly complete dataset is available (Fig. S1). Viscosity is key to this problem (*13*), 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.

To address any such problems, we require a model that links melt composition (*x*) and structure to physical properties and intensive variables such as temperature (*T*). We construct a deep neural network that takes an input composition, and predicts a wide range of directly-observable glass properties including density, refractive index and Raman spectra (Fig. 1a; Supplementary Material). In addition, it outputs the latent variables (such as configurational entropy, *Sconf(Tg)*, a thermodynamic image of the liquid structure) required to predict melt viscosity (**) within five theoretical and empirical frameworks: Adam-Gibbs, MYEGA, Avramov-Milchev, Tamman-Vogel-Fulcher and Free Volume Theory (Supplementary Material). These are some of the more commonly-encountered members of a vast landscape of theories describing viscous flow of liquids. At present, there appears to be no consensus towards any one model appropriate for all liquids. As a result, we propose a trans-theoretical approach, whereby our neural network simultaneously enables predictions across multiple theories, and is trained to provide best average performance across all five frameworks. Where quantities (such as the glass transition temperature, *Tg*) appear as latent variables within multiple frameworks, our approach further provides one value that is optimal across all theories.

After training, the model allows accurate predictions of liquid and glass properties, including structure-dependent properties such as Raman spectra. Trans-theoretical predictions of ** (Fig. 1b, Figure S3) are possible with good precision (σ**< 0.4 log Pa·s on unseen data, Table S3; for comparison, σ**0.6 log Pa·s**for the best empirical magma viscosity models (e.g. *7*)). Known viscous *Tg* and *Sconf(Tg)* are predicted within 16 K and 0.8 J mol-1 K-1, respectively, and glass density and refractive index are predicted within 0.009 g cm-3 and 0.005, respectively (Fig. S4). Despite a limited dataset (Fig. S1), global variations of Raman signals have been captured well (Fig. 1c, S4, S7) and can be predicted with a reasonable error (Supplementary Text). The model thus embeds structural information, and allows estimation of structural parameters from Raman spectra including the ratio of intra- and inter-tetrahedral aluminosilicate vibrations, *RRaman* (Fig. 1c). This serves as a proxy for glass network topology (the 3D connectivity of the polyhedral SiO2-AlO2 network), and is linked to variations in melt properties (*15*).

With the the model, we can explore why viscosity varies substantially according to the concentration of Al and K in Na2O-K2O-Al2O3-SiO2 melts. We observe the known division between topological and chemical effects (*16*) on different properties. Glass network topology broadly speaking correlates with the glass transition temperature (Fig. 2a), and to quantities proportional to energy barriers opposed to ionic mobility (Fig. 2b), or the glass refractive index (Supplementary Materials). The glass transition temperature exhibits detailed compositional dependence, in agreement with the well-known mixed alkali effect (MAE, *17*) resulting from metal cation interactions within the SiO44- polyhedral network (e.g. *9*). This chemical effect strongly affects properties influenced by cationic interactions and steric hindrance effects, such as the configurational entropy*(9*, *16*, *18*, *19*)*,* or the term *CFV* in the free volume theory that encompass local cationic influences on melt free volume (Fig. 2c,d, Supplementary Text). The model allows such effects to be seen, and its trans-theoretical character further allows observing relationships between latent variables from different theories(Supplementary Text).

From the above observations, cationic mixing affects macroscopic variables such as *Sconf(Tg)*. Cationic mixing can be random (*16*, *19*) or not (*20*, *21*), and is usually difficult to predict. The model solves this problem by enabling systematic quantification of such mixing effects (Fig. 3). For the system analyzed here, increasing [Al] leads to used here(Fig. 3a,b). Furthermore, Al-to-alkali ratio largely affects the MAE: without Al, the MAE results in an entropy excess (Fig. 3c) and, hence, in large decreases in melt viscosity (*η ∝* *1/Sconf(Tg)*, *18*). Increasing [Al]/[Na+K] leads to changing the role of alkali metals in the network (*22*). Na and K segregate in different molecular nano-environments (*9*, *23*), inducing less and less excess entropy of mixing as [Al]/[Na+K] increases (Fig. 3d,e,f). In addition, K-rich and Al-rich melts show higher local order (*4*) resulting in small *Sconf(Tg)* values (Fig. 3b,f).

We are now in a position to understand why eruptions of rhyolite magmas (essentially alkali aluminosilicate melts with some impurities of Fe, Ca, Mg, and volatiles) tend to be more explosive if the magmas are rich in K and Al. Amongst other features such as degassing and nano-cristallization (*13*, *24*), rhyolite magmas that erupt explosively and effusively show a clear chemical distinction (*13*). The model demonstrates that this relates to how melt composition drives their structure (Fig. 4a) and configurational entropy (Fig. 4b). Increasing [Al] and [K] increases network connectivity and nano-structuration (Figs. 3, 4a), reduces *Sconf(Tg)* (Fig. 4b) to a threshold beyond which magmas become too viscous, explaining their association with explosive eruptions.

This application highlights how our model can be used for practical property predictions in the Earth sciences. It reveals the fine structural and thermodynamic controls on magma viscosity, which, in turn, govern the dynamics of volcanic eruptions. The model can readily be extended to include quantities of interest across a range of domains and applications, with the present results demonstrating that a combination of neural networks and physical/thermodynamic models can offer new perspectives on long-standing problems in many disciplines. Given the trans-theoretical insights brought by deep learning models, such approach helps addressing fundamental questions like how melts flow (Supplementary Materials), and could be applied to better understand the nature of glass and of the glass transition that remains currently elusive as shown by the recent work of (*25*).

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**Acknowledgments:** CLL thanks Malcolm Sambridge (Seismology & Mathematical Geophysics, RSES, Australian National University), Lexing Xie and Cheng Soon Ong (CECS, Australian National University), and Sung Keun Lee (Seoul National University) for various discussions regarding optimization, machine learning, and melt and glass properties.

**Funding:** CLL acknowledges funding from a “Chaire d’Excellence” from the Université de Paris, from the Australian Research Council Laureate Fellowship FL1600000 to Prof. Hugh O’Neill (ANU-RSES) as well as from the Postdoctoral Fellowship of the Carnegie Institution for Science during the realisation of this project.

**Author contributions:** CLL designed the study, collected the data, performed Raman and viscosity experiments, developed the 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.

**Data and materials availability:** 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).

**Supplementary Materials:**

Materials and Methods

Supplementary Text

Figures S1-S7

Tables S1-S3

**Fig. 1.** **Schematic of deep leaning model and property and structure prediction examples.** **(a)** A neural network is constructed to take melt composition as its input. The outputs are taken to be various melt and glass properties, plus Raman vibrational properties (see text). Once trained, relationships between chemistry, structure and properties of melts and glasses can be systematically explored. **(b)** Melt viscosity can be derived from these outputs 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. **(c)** Structural investigation are made through Raman spectra predictions, which compare well with experimental data for albite and sodium trisilicate glasses for instance. R*Raman*, the ratio of intertetrahedral (below 670 cm-1) to intratetrahedral (above 870 cm-1) network vibrations, can be calculated and used for structural investigations. It increases as the connectivity and organisation of the polyhedral SiO2-Al2O3 network develops in the glass. It is thus very sensitive to 3D glass network connectivity and rigidity.

**Figure 2: Melt and glass properties vary in a complex way with glass network topology**. **(a)** The model allows observing that parameters such as the viscous glass transition temperature *Tg* **(a)** and *BCG,*a free volume activation energy term **(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 *CCG* **(d).** Each symbol represents the calculation for a randomly generated composition in the glass-forming domain of studied system.

**Figure 3: Variations of the configurational entropy at *Tg* in K2O-Na2O-Al2O3-SiO2 melts.** *Sconf(Tg)* vary non-linearly with oxide contents in the ternary diagrams Na2O-Al2O3-SiO2 and K2O-Al2O3-SiO2 in **(a)** and **(b)**. 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 4: Chemical, structural and entropic mapping of rhyolite eruptions.** The rheological agpaitic index is calculated as (Na2O + K2O + CaO + MgO + FeO)/(Al2O3 + Fe2O3). From (*13*), Al-rich and K-rich rhyolite magmas, like those of Yellowstone (U.S.A.) are generally associated with explosive eruptions. The rheological tipping point proposed by (*13*) find roots in how increasing Al and K contents promote 3D network connectivity (a) and, thus reduces melt entropy (b) down to a particular threshold here identified to be around 9 J mol-1 K-1.