Title: Deep learning prediction of **melt** properties

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**Abstract:** (140/125 w)

Knowledge of the links between chemistry, structure and the 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 yet allows revealing the molecular and thermodynamic origin being this. Here, we construct a deep learning model, linking a neural network to physical and thermodynamic equations, to predict alkali aluminosilicate melts and glasses properties, including viscosity, density, or even glass Raman signals. The model, which captured the links between different quantities and even theories, reveals, for instance, that profound changes in structure and configurational entropy drive the rheology of rhyolite magmas, and explains the effusive or explosive character of their eruptions. More generally, our approach provides a new, exciting way to build models 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 liquid move? How do they exchange heat with their media? How do they crystallize? Those fundamental questions underly many practical problems, like the dynamic of volcanic eruptions (*1*) , the formation of rocks (*2*), or the manufacturing of novel glass materials. Addressing them involves knowledge of liquid properties governed by their composition and associated atomic/ionic structure, such as viscosity, heat capacity and entropy, variables much needed for fluid dynamic and phase diagram calculations. At present, specific experimental observations and thermodynamic or molecular dynamics (MD) simulations, or a combination of these (e.g. *3*, *4*), is performed to gather the necessary information, usually for 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). However, they 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 they usually they only predict a limited subset of (or just one) properties on a restricted compositional range (e.g., see for viscosity *7*–*10*), are built making strong simplifications, using a given theory or without any theoretical backgrounds. In this publication, we show that a model combining deep learning with different theoretical equations solves such shortcomings, allowing addressing the questions previously raised through practical liquid property predictions and the 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, for which a fairly complete (albeit sparse) dataset is available (Fig. S1). This system is interesting for addressing problems like why enrichments in Al and K in rhyolitic magmas yield more violent volcanic eruptions (*13*). Viscosity is key to this problem (*13*), but the underlying structural and thermodynamic controls remain to be truly characterized. Beyond volcanology, better understanding and predicting properties of alkali aluminosilicate glasses can help designing technological glass materials like handheld device screens.

To address those problems, we need a practical model linking melt composition (*x*) and structure to its physical properties, and intensive variables like temperature (*T*). We construct a deep neural network that predicts, given composition *x*, glass properties such as density, refractive index and Raman spectra (Fig. 1a, Supplementary Materials). It further outputs the latent variables required to predict melt viscosity (**) within five theoretical and empirical frameworks: Adam-Gibbs, MYEGA, Avramov-Milchev, Tamman-Vogel-Fulcher and Free Volume (Supplementary Material). Indeed, there is a vast landscape of theories describing the viscous flow of liquids (e.g. *14*). For example, the Adam-Gibbs model (8) assumes that liquid movements occurs through cooperative molecular re-arrangements; it represents ** as

, (1)

with *Ae* representing a high-temperature limit, *Be* a term proportional to the energy barriers opposed to molecular re-arrangements, and *Sconf* and *Cpconf* the melt configurational entropy and heat capacity, respectively. *Tg* is the ‘glass transition temperature’, at which melt is frozen-in into glass upon quench. Alternatively, the Free Volume model states that ions diffuse in/between liquid-like and solid-like molecular cells in liquids. This takes the form

**,** (2)

with again representing the high-temperature limit, and , and latent variables related to free volumes’ properties like percolation, size and numbers. Other models may also be found in the literature, and a few are given in Supplementary Material. At present, there is no consensus toward a particular model for all liquids. In such case, we propose to use a trans-theoretical approach, combining the necessary physical and thermodynamic equations with a deep neural network (or several if necessary) for latent variable predictions (Fig. 1a). This deep learning (DL) model is trained using a suite of selected data, seeking best average performance across all theories. In particular, it should be noted that where quantities (e.g. ) appear in multiple frameworks, our approach provides one value that is optimal across all theories.

After training, the DL model allows accurate predictions of liquid and glass properties, and of structural features like Raman spectra. Trans-theoretical predictions of ** (Fig. 1b, Figure S3) are performed with good precision (σ** < 0.4 log Pa·s on unseen data, Table S3; for comparison, σ** > 0.6 log Pa·sfor the best empirical magma viscosity models (e.g. *7*)), as well as other melt and glass properties: 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 DL model thus embeds structural information, and allows estimation of structural parameters from Raman spectra including *RRaman* (Fig. 1c), the ratio of intra- and inter-tetrahedral aluminosilicate vibrations. This serves as a proxy for glass network topology (3D connectivity of the polyhedral SiO2-AlO2 network), and is linked to variations in melt properties (*15*).

Using the DL model, we explore why viscosity changes largely with Al and K concentrations in Na2O-K2O-Al2O3-SiO2 melts. We observe the known division between topological and chemical effects (*16*) on different properties: glass network topology largely correlates with their *Tg* (Fig. 2a), as well as terms proportional to energy barriers opposed to ionic mobility (Fig. 2b), or the glass refractive index (Supplementary Materials). In details, a fine compositional dependence can be visible for properties like *Tg*, in agreement with the well-known mixed alkali effect (MAE, *17*) on *Tg* that results from metal cation interactions within the SiO2 polyhedral network (e.g. *9*). This chemical effect strongly affects properties influenced by cationic interactions and steric hindrance effects, like *Sconf(Tg)* *(9*, *16*, *18*, *19*)*,* and *CCG* that encompass local cationic influences on melt free volume (Fig. 2c,d). The DL model allows observing such effects, and its trans-theoretical character further allows observing relationships between latent variables from different theories(Supplementary Text).

From the above observations, chemical mixing effects largely affect macroscopic variables like *Sconf(Tg)*. They can be random (*16*, *19*) or not (*20*, *21*), and are usually difficult to predict. The DL model solves this problem because it allows systematic observations of such mixing effects (Fig. 3). For our problem, increasing [Al] leads to decreasing *Sconf(Tg)* (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). Besides, K-rich and Al-rich melts show higher local order (*4*), this resulting in small *Sconf(Tg)* values (Fig. 3b,f).

We now can understand why eruptions of rhyolite magmas (essentially alkali aluminosilicate melts with some impurities of Fe, Ca, Mg, and volatiles) may tend to be more explosive as magmas are rich in K and Al. Among many parameters like degassing and nano-cristallization (*13*, *24*), rhyolite magmas erupted explosively and effusively show a clear chemical distinction (*13*). The DL model shows that this relates to how melt structure (Fig. 4a) controls *Sconf(Tg)* (Fig. 4b). Increasing [Al] and [K] leads to increase network connectivity and nano-structuration (Figs. 3, 4a), and to reduce *Sconf(Tg)* (Fig. 4b) down to a threshold beyond which magmas become mostly associated with explosive eruptions.

This application highlights the usefulness of our model for practical property predictions in Earth sciences. Beyond this, it reveals the fine structural and thermodynamic controls on magma viscosity, itself related to the dynamic of volcanic eruptions. The DL model needs to include more elements for its use in different domains, but the present results demonstrate that combining neural networks and physical/thermodynamic models represents an important step toward solving long-standing problems in many disciplines. Given the trans-theoretical insights broudht 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*).

References and Notes. (30 citations max main text)

1. D. B. Dingwell, Volcanic Dilemma - Flow or Blow? *Science*. **273**, 1054–1055 (1996).

2. N. L. Bowen, *The evolution of igneous rocks* (Dover Publications, 1956).

3. Y. Wang, T. Sakamaki, L. B. Skinner, Z. Jing, T. Yu, Y. Kono, C. Park, G. Shen, M. L. Rivers, S. R. Sutton, Atomistic insight into viscosity and density of silicate melts under pressure. *Nat. Commun.* **5** (2014), doi:10.1038/ncomms4241.

4. C. Le Losq, D. R. Neuville, W. Chen, P. Florian, D. Massiot, Z. Zhou, G. N. Greaves, Percolation channels: a universal idea to describe the atomic structure and dynamics of glasses and melts. *Sci. Rep.* **7**, 16490 (2017).

5. N. Saunders, A. Miodownik, *CALPHAD (calculation of phase diagrams): a comprehensive guide* (Elsevier., 1998).

6. D. C. Rapaport, *The art of molecular dynamics simulation* (Cambridge University Press, Cambridge, U.K., ed. 2nd, 2004).

7. D. Giordano, J. K. Russell, D. B. Dingwell, Viscosity of magmatic liquids: A model. *Earth Planet. Sci. Lett.* **271**, 123–134 (2008).

8. J. C. Mauro, A. J. Ellison, D. C. Allan, M. M. Smedskjaer, Topological Model for the Viscosity of Multicomponent Glass-Forming Liquids. *Int. J. Appl. Glass Sci.* **4**, 408–413 (2013).

9. C. Le Losq, D. R. Neuville, Molecular structure, configurational entropy and viscosity of silicate melts: Link through the Adam and Gibbs theory of viscous flow. *J. Non-Cryst. Solids*. **463**, 175–188 (2017).

10. K. Starodub, G. Wu, E. Yazhenskikh, M. Müller, A. Khvan, A. Kondratiev, An Avramov-based viscosity model for the SiO2-Al2O3-Na2O-K2O system in a wide temperature range. *Ceram. Int.* **45**, 12169–12181 (2019).

11. B. O. Mysen, P. Richet, *Silicate Glasses and Melts* (Elsevier, ed. 2nd, 2019).

12. C. Le Losq, M. R. Cicconi, G. N. Greaves, D. R. Neuville, in *Handbook of Glass* (Springer, 2019; https://www.springer.com/us/book/9783319937267).

13. D. Di Genova, S. Kolzenburg, S. Wiesmaier, E. Dallanave, D. R. Neuville, K. U. Hess, D. B. Dingwell, A compositional tipping point governing the mobilization and eruption style of rhyolitic magma. *Nature*. **552**, 235 (2017).

14. Y. Bottinga, P. Richet, A. Sipp, Viscosity regimes of homogeneous silicate melts. *Am. Mineral.* **80**, 305–318 (1995).

15. D. Giordano, J. K. Russell, Towards a structural model for the viscosity of geological melts. *Earth Planet. Sci. Lett.* **501**, 202–212 (2018).

16. D. R. Neuville, P. Richet, Viscosity and mixing in molten (Ca, Mg) pyroxenes and garnets. *Geochim. Cosmochim. Acta*. **55**, 1011–1019 (1991).

17. J. O. Isard, The mixed alkali effect in glass. *J. Non-Cryst. Solids*. **1**, 235–261 (1969).

18. P. Richet, Viscosity and configurational entropy of silicate melts. *Geochim. Cosmochim. Acta*. **48**, 471–483 (1984).

19. D. R. Neuville, B. O. Mysen, Role of aluminium in the silicate network: In situ, high-temperature study of glasses and melts on the join SiO2-NaAlO2. *Geochim. Cosmochim. Acta*. **60**, 1727–1737 (1996).

20. D. R. Neuville, Viscosity, structure and mixing in (Ca, Na) silicate melts. *Chem. Geol.* **229**, 28–41 (2006).

21. S. K. Lee, Structure and the extent of disorder in quaternary (Ca-Mg and Ca-Na) aluminosilicate glasses and melts. *Am. Mineral.* **90**, 1393–1401 (2005).

22. C. Le Losq, D. R. Neuville, P. Florian, G. S. Henderson, D. Massiot, The role of Al3+ on rheology and structural changes of sodium silicate and aluminosilicate glasses and melts. *Geochim. Cosmochim. Acta*. **126**, 495–517 (2014).

23. C. Le Losq, D. R. Neuville, Effect of the Na/K mixing on the structure and the rheology of tectosilicate silica-rich melts. *Chem. Geol.* **346**, 57–71 (2013).

24. F. Cáceres, F. B. Wadsworth, B. Scheu, M. Colombier, C. Madonna, C. Cimarelli, K.-U. Hess, M. Kaliwoda, B. Ruthensteiner, D. B. Dingwell, Can nanolites enhance eruption explosivity? *Geology* (2020), doi:10.1130/G47317.1.

25. V. Bapst, T. Keck, A. Grabska-Barwińska, C. Donner, E. D. Cubuk, S. S. Schoenholz, A. Obika, A. W. R. Nelson, T. Back, D. Hassabis, P. Kohli, Unveiling the predictive power of static structure in glassy systems. *Nat. Phys.* **16**, 448–454 (2020).

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**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 DL 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.