Title: Deep learning prediction of material properties

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**Abstract:** (121/125 w)

Knowledge of the tryptic relationship between chemical composition, molecular structure and macroscopic properties of materials like minerals or glasses is fundamental in many disciplines, including metallurgy, Earth and material sciences. Several avenues allow prediction of this relationship, but usually this task remains very challenging. Deep learning represents a new tool to perform such task, allowing one to link experimental results, models, and even different theories. Focusing on alkali aluminosilicate melts, of prime importance for volcanology and glass sciences, we show how a deep neural network leverages experimental data and theoretical knowledge for prediction of melt and glass properties, including viscosity, fragility, density, or even glass Raman signals. Such approach provides a new, exciting way to build models for various applications.

**One sentence summary** (96/125 characters): Deep learning bridges the gap between data and models for practical material property prediction

**Main Text:**

The intrinsic properties of materials are governed by their chemical composition, and the associated atomic/ionic structure. Understanding this connection is fundamental to material engineering and manufacturing, and also to addressing many key questions across physics, chemistry and the geosciences. Information may be drawn from several sources, particularly experimental observations and thermodynamic or molecular dynamics (MD) simulations, or a combination of these (e.g. *1*, *2*). Techniques such as CALPHAD calculations (*3*) and MD simulation (*4*) 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, like oxide glasses or minerals, within the pressure-temperature regimes relevant to industry or the Earth sciences. In these cases, thermodynamic models often offer an intermediate way of predicting macroscopic properties, but the experimentalist still has to link thermodynamic variables to data, and this often turns out to be challenging. In this publication, we show that machine learning can serve as the “middle man”, allowing scientists to make sense of their data and to put them in the context of different models and theories.

As a concrete example, we consider the links between the chemical composition, structure and physical properties of aluminosilicate melts and glasses (e.g., see the review of *5*). These materials are central to the Earth and material sciences: they form the liquid part of magmas, with melt viscosity directly determining whether volcanic eruptions have effusive or explosive character (*6*), and, as glasses, they are so important to nowadays societies that we may be living in The Glass Age (*7*). A number of different theories exist that attempt to connect the structure of these melts to their properties. These often involve variables that are either difficult or impossible to measure directly, such as the glass configurational entropy *Sconf*. In this publication, we employ a deep neural network to represent such latent variables within the physical theory, and use this to infer their relationship to composition. For simplicity, we focus on compositions in the K2O-Na2O-Al2O3-SiO2 system, as a fairly complete (albeit sparse) dataset is available (Fig. S1).

There is a vast landscape of theories (and their associated set of equations) describing the viscous flow of liquids, supported by different arguments depending on use case (e.g. *11*). Within each theory, viscosity is modeled as dependent on a certain subset of latent variables. For example, the Adam-Gibbs model (8) assumes that viscous movements of melts occurs through cooperative molecular re-arrangements. It represents the viscosity () as a function of temperature (*T*) and composition (*x*) as:

, (1)

with *Ae* a high-temperature viscous limit, *Be* a term proportional to the energy barriers opposed to the cooperative molecular re-arrangements, and *Sconf* and *Cpconf* the melt configurational entropy and heat capacity, respectively. *Tg* is the temperature at which melt is frozen-in into glass upon quench, a.k.a the glass transition temperature (in reality, this corresponds to a small temperature interval, but, for convenience, we adopt the empirical definition of *Tg* as equal to the temperature at which *η* = 1012 Pa·s). Alternatively, one might adopt the Free Volume model, which states that melts present liquid-like and solid-like molecular cells, their mobility being ensured by atomic diffusivity within/between liquid-like cells. In equation, we have

**,** (2)

with a high-temperature viscous limit, and , and latent variables linked to free volumes’ properties like percolation, size and numbers. Many other models may also be found in the literature, and a few are given in the Supplementary Material.

Each of these models is built upon its own set of assumptions, and for some applications it may be evident that one choice is more appropriate than others. Often, however, there is no clear rationale for adopting one model instead of another; one simply requires good predictive performance (i.e., the ability to accurately forecast the viscosity of a melt given its composition and temperature). Besides, models built on a specific theory and linking melt chemistry and molecular structure exist (*1*, *2*) but they usually are specific and only predict a limited subset of properties, and are based on strong assumptions (e.g. how chemistry determines silicate tetrahedral speciation in silicate glasses).

To tackle such problem, we propose to develop a trans-theoretical approach combining machine learning with physical/thermodynamic equations to predict various structural and physical properties. With melt composition as an input, the model uses a “core” deep neural network to predict latent variables of equations from multiple theories as well as observable physical and structural glass properties, in pursuit of better overall behavior (Fig. 1). The chosen latent variables are those required to predict melt viscosity within five theoretical and empirical frameworks: the Adam-Gibbs, MYEGA, Avramov-Milchev, Tamman-Vogel-Fulcher and Free Volume theories (see Supplementary Material). In addition, we predict three composition-dependent quantities that are readily observed experimentally: the quenched melt density, its Raman spectrum, and the parameters necessary to predict its refractive index at a given wavelength from the Sellmeier equation. We train the neural network using a suite of measurements of viscosity, density, Raman and optical spectra, seeking best average performance across all theories (Supplementary Materials). In particular, it should be noted that where quantities (like ) appear in multiple theoretical frameworks, our approach provides one value that is optimal across all models.

After training, the model allows systematic predictions of melt viscosity using theories like Adam-Gibbs or Free Volume, as shown in Figure 2a where model predictions and measurements are compared for melts with ~66 mol% SiO2 and varying Al/(Na+K) and K/(K+Na). Semi-empirical and empirical equations, like the famous Tamman-Vogel-Fulcher equation, yield a good precision too (Supplementary Text and Figure S3). In all cases, predictive errors are lower than 0.50 log Pa·s (Table S3). Interestingly, based on only numerical performance, the different viscosity models yield similar precisions and it may seem difficult to discriminate a better one. However, the Adam-Gibbs theory relates thermodynamic variables (*Sconf*, *Cpconf*...) to melt viscosity (*9*) and performs as well as, or better than others equations. Consequently, present results confirm that this theory seems well appropriate for aluminosilicate melts. Aside this and more generally, the obtained results clearly indicate that deep learning allows practical trans-theoretical modeling, a feature extremely interesting in many areas. Furthermore, the model offers the possibility to predict aluminosilicate 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).

A critical development is that the deep neural network links thermodynamic/dynamic information to structural information. Given a limited Raman dataset (48 compositions, Supplementary Text), we did not aim at having excellent predictions of Raman spectra, but we simply wanted the model to capture the general Raman signal variations as a function of glass composition. This goal is achieved (Figs. 2b, S7, Supplementary Text). In turn, the model allows systematic prediction of glass Raman spectra and extraction of structural parameters like *RRaman* (Fig. 2b), which is the ratio of intra- and inter-tetrahedral aluminosilicate vibrations in the glass that can be used as a proxy for glass network topology (3D connectivity of the covalent polyhedral SiO2/AlO2 units) and linked to variations in melt properties (*1*).

Using the model, we go on to systematically explore how chemical, structural and thermodynamic/dynamic variables correlate each other in the glass-forming region of the quaternary Na2O-K2O-Al2O3-SiO2 diagram (Fig. S1). High but non-linear correlations are detected between the structural parameter *Rraman* and melt/glass properties like *Tg*, *Be/Sconf(Tg),* *BCG*, or the 589 nm glass refractive index (Fig. 3). Such results agree well, for instance, with the well-known link between Tg and the glass polyhedral connectivity (REF?). However, the present model allows a systematic observation of details like a non-negligible chemical mapping (Fig. 3a) that can originates from various effects, like the mixed alkali effect where the mixing of alkali elements in Al-poor silicate glasses generates depletions in their Tg (*1*). Such subtleties are also visible in transport properties. *Be/Sconf(Tg)* and *BCG* are two terms in the Adam-Gibbs and Free Volume theory that are linked to “energy barriers” opposed to molecular movements; those “energy barriers” can be linked to either the intrisic entropies of molecules in the Adam-Gibbs theory or to available liquid-like free volumes in the Free Volume theory. We see that both quantities scale well with the network topology (Fig. 3b); some chemical mapping is present but not critical. This agrees with previous thermodynamic modeling of *Be* for instance, where *Be* variations in Na2O-K2O-SiO2 melts can be modeled as additive contributions from SiO2 tetrahedral units with different numbers of bridging oxygens (*1*). 3D network connectivity development is thus a critical parameter affecting the mobility of atoms in melts, largely affecting through this the temperature at which melts quench to glass, and even ultimately the glass properties like their optical refractive index (Fig. 3c).

However, contrary to a general school of thinking in development lately, not all melt and glass properties are explain by changes in the topology and connectivity of the aluminosilicate network. For instance, the variation in melt configurational entropy, due to the atomic disorder in the melt, can be separated in topological and chemical contributions (*1*–*4*). While the topological part is linked to the 3D network topology, the chemical part arises from entropic excess induced by cationic and molecular mixing within melt structure. Such mixing effect can be random (*1*) or not (*1*, *2*), and is thus difficult to predict. This translates into a complex mapping between the glass *Sconf* and its network topology (Fig. 4a), resulting from the addition of topological and complex chemical contributions arising from Na-K or Al-Si mixing for instance (Supplementary Text). As a result, glass *Sconf* cannot be calculated from only the knowledge of its topology. This implies that it is not possible to only use the information from topological-sensitive methods like Raman spectroscopy to link melt chemistry, structure and viscosity. This unfortunately questions the recent hopes that appeared in the literature of modeling magma viscosity from Raman spectra (e.g. *1*). Other properties like fragility, which defines the slope at *Tg* of the viscosity versus *Tg/T* curve and thus the fragile/strong character of liquids (*1*), or the *CCG* parameter that can be linked to the compositional and pressure effects on free volumes, vary in a complex way with glass structure and chemical composition (Fig. 4b,c). Such complexity explains why no general model of glass *Sconf* or fragility has been proposed to date, despite the importance of such parameters. This further stresses the need for new research efforts focused on understanding the behavior of non-network forming elements (not involved in the topological glass network), like alkali and alkaline-earth cations, as such knowledge could become critical for better modeling material properties in the future. For now, the present work show that the use of machine learning allows us to circumvent such problems, even reavealing them, and ultimately allows either practical property predictions or a more fundamental exploration of the links between the composition, structure and properties of liquids. As such, combined with experimental data and theories, the use of machine learning can represent an important step toward solving long-standing problems in many disciplines, probably including the theory of the nature of glass and of the glass transition that remains currently elusive as shown by the recent work of Bapst et al. (*1*).

References and Notes. (30 citations max main text)

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*Supplementary Material References*

**To be done prior to submission**

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**Author contributions:** CLL designed the study, collected the data, performed Raman and viscosity experiments, developed the computer code and drafted the manuscript. DN and BOM helped in the design of the neural network. CLL, BOM and DN performed Raman measurements. 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).

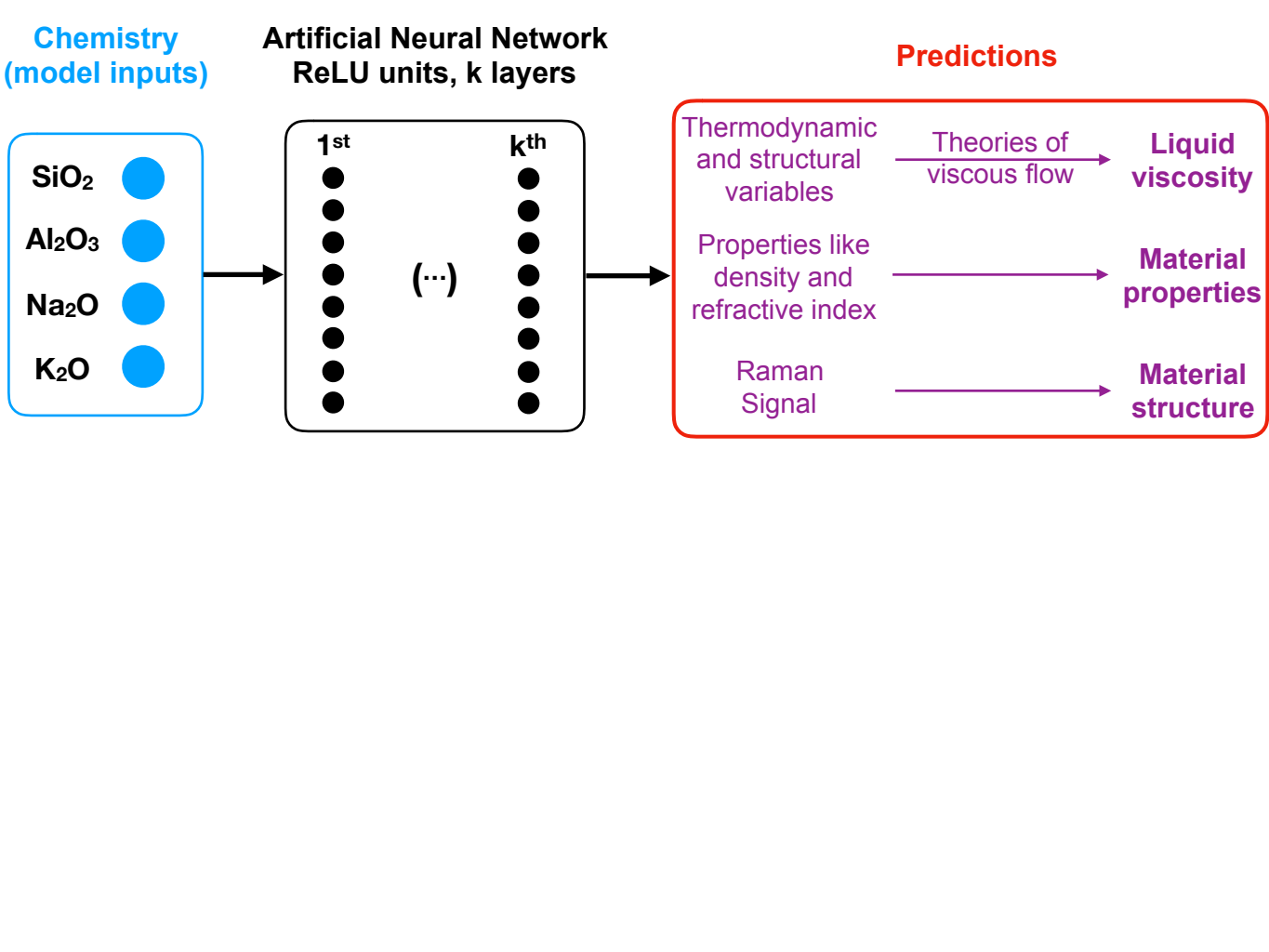
**Supplementary Materials:**

Materials and Methods

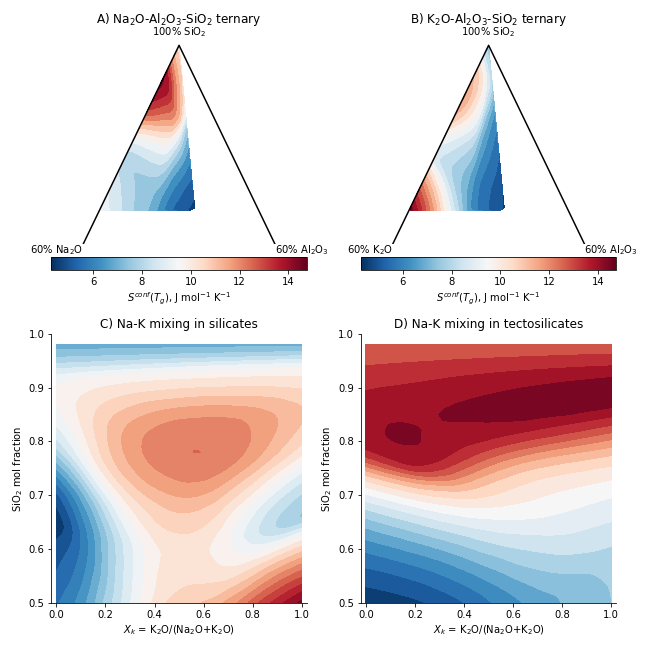
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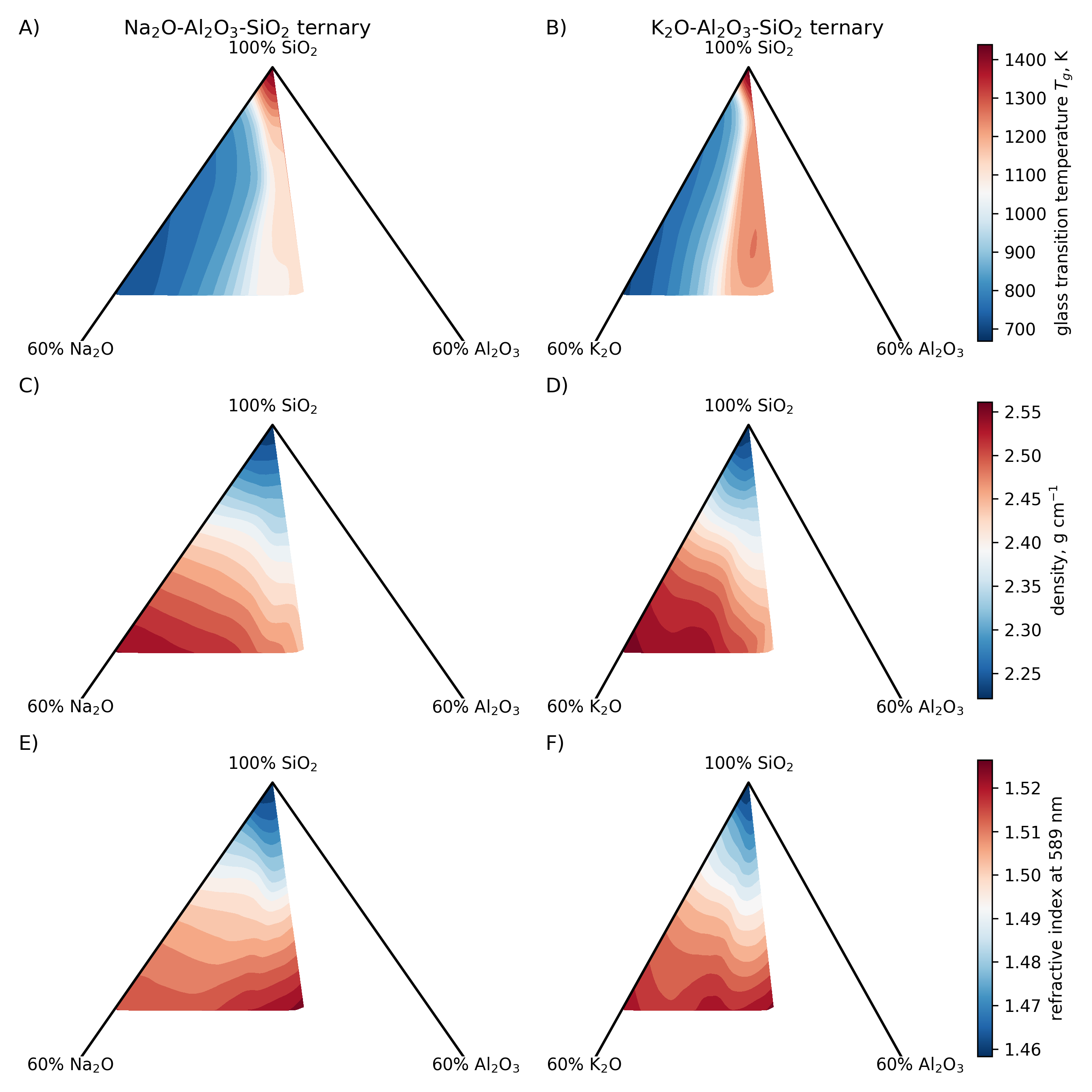
Figures S1-S7

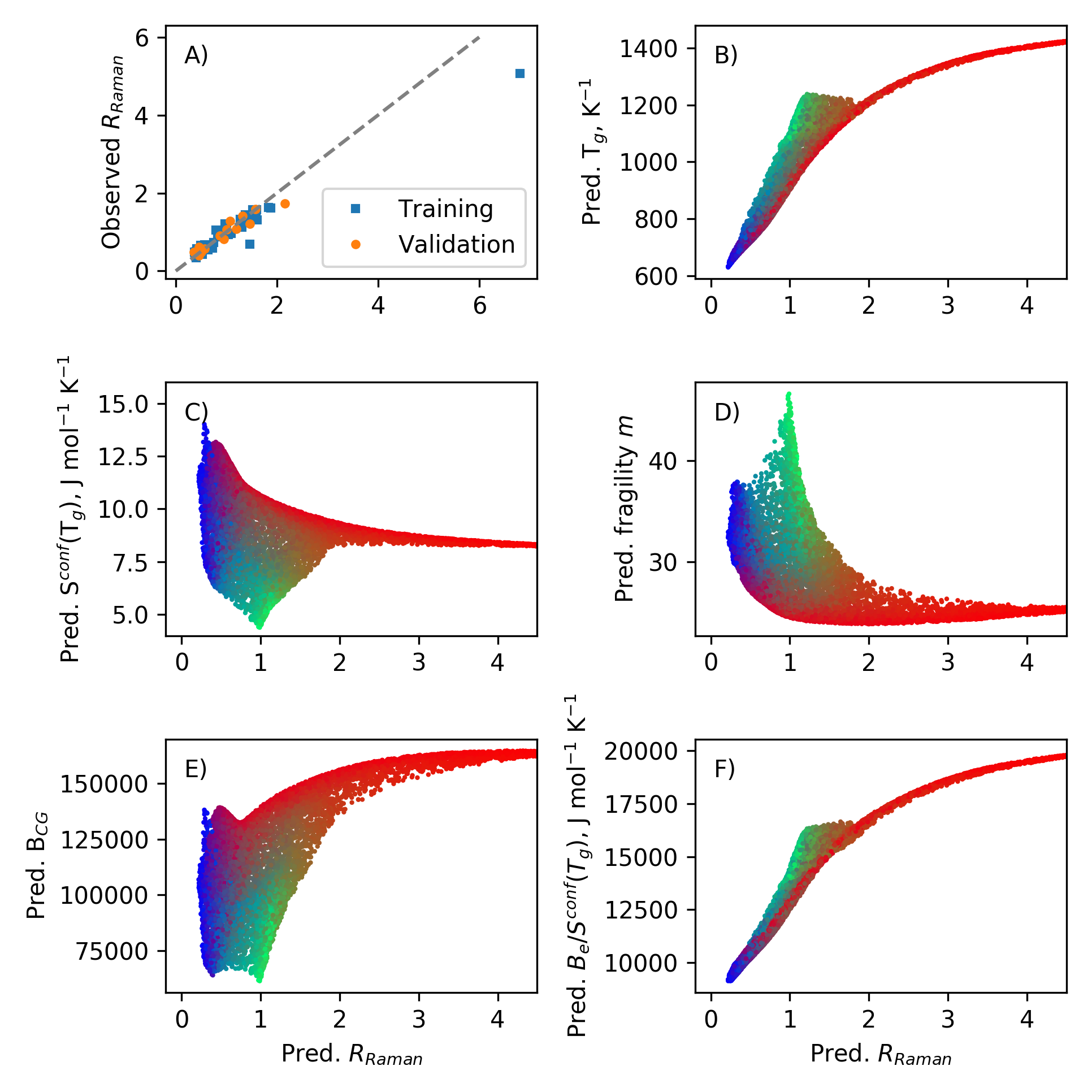
Tables S1-S3: Composition of the synthesized glasses.

**Fig. 1.** **Presentation of the model.** The melt composition is given in input to a deep neural network that predicts various melt and glass properties as well as their Raman vibrational properties (see text for details). Viscosity can then be calculated via different theories, and the relationships between chemistry, structure and properties of the melt and glasses systematically explored using this framework.

**Figure 2: Example of viscosity and Raman predictions. (a)** Measurements (symbols) and model predictions from the Adam-Gibbs and Free Volume theories are reported as a function of inverse temperature for different melt covering a broad range of composition, from silica to polymerised aluminosilicates and depolymerised silicates. (b) two example of Raman model predictions compared to data for albite and sodium trisilicate glasses. The model allows prediction of the melt viscosity within

**Figure 3: Maps of configurational entropy at the glass transition temperature.** *Sconf(Tg)*, is shown in the A) sodium and B) potassium aluminosilicate systems as well as during Na-K mixing in C) silicates (no Al) and D) tectosilicates (Al/(Na+K) = 1) compositions. Addition of aluminum generally result in decreasing *Sconf(Tg)*, which reach minimal values close to the *nepheline-orthoclase* composition. During Na-K mixing, excess entropy appears when those elements are network modifiers (in silicate, C) but is not detected in Al-rich compositions (e.g. in aluminosilicates, D).

**Figure 4: Maps of glass transition temperature, glass density and glass refractive index**. Glass transition temperature varies strongly with the (Na+K)/Al ratio and silica content in both the sodium (A) and potassium (B) aluminosilicate systems, while glass density (C, D) or optical refractive index depend mostly on the glass silica content (E, F).

**Figure 5: Relationship between glass Raman spectra and glass properties**. The ratio of the intratetrahedral to intertetrahedral vibrations, *RRaman* (see text for explanation), is a proxy for network topology. It is well reproduced by the present model despite a limited training set (A); the model thus captured well how glass composition is linked to their Raman signal and thus structure. *RRaman* correlates well with *Tg* (B) or *Be/Sconf(Tg)* (F), but variations with *Sconf(Tg)*, *m* or *BCG* are of a more complex nature (C, D, E). Each point is part of 10,000 randomly generated compositions in the quaternary diagram. Symbol colors are in RGB format and embed compositional information as red is scaled mol fraction of SiO2, green is scaled mol fraction of Al2O3, and blue is scaled mol fraction of K2O+Na2O. This allows to observe a clear compositional mapping in variations of *Sconf(Tg)*, *m* or *BCG*.