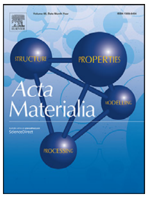
Acta Materialia 222 (2022) 117432

Contents lists available at ScienceDirect

Acta Materialia

journal homepage: www.elsevier.com/locate/actamat



Full length article

Hybrid machine learning/physics-based approach for predicting oxide glass-forming ability

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ARTICLE INFO

Article history:
Received 28 June 2021
Revised 4 October 2021
Accepted 20 October 2021
Available online 23 October 2021

Keywords:
Machine learning
Energy landscapes
Glass-forming ability
crystallization

ABSTRACT

Predicting the liquid compositions that will vitrify at experimentally accessible quench rates remains one of the grand challenges in the field of condensed matter physics. This glass-forming ability can be quantified as the critical quench rate needed to suppress crystallization. Knowledge of this critical quench rate also informs which glass composition could be used for new applications. There have been several physical and empirical models presented in the literature to predict the critical quench rate/glass forming ability. These models range from those theoretically derived to those quantified only through experimental characterization. In this work, we instead propose a new method to calculate the critical quench rate using the recently developed toy landscape model combined with machine learning. The toy landscape model accesses the underlying physics that control the vitrification behavior by directly simulating the liquid thermodynamics and kinetics. The results are discussed in terms of industrial impact, physical insights, and how the glass science community can develop improved predictions of glass-forming ability.

1. Introduction

Modeling of glass-forming ability (GFA) can be traced back to the seminal paper of Zachariasen in 1932 where he laid out structural rules that predict whether a glass could be formed based on a given oxide composition [1–3]. Although these rules are still widely cited today, their practical use is qualitative at best. Since the work of Zachariasen, there have been several attempts to understand and quantify GFA. Among these is topological constraint theory (TCT) which was first proposed by Gupta and Cooper [4] in oxide glasses (GC-TCT) but was followed closely with work done by Phillips and Thorpe [5] in chalcogenides (PT-TCT). Both theories conjectured that the optimal glass-forming ability occurred when the atomic degrees of freedom (f) was equal to zero. The atomic degrees of freedom, f, is

$$f = d - n_c, \tag{1}$$

where d is the dimensionality of the system (usually 3) and $n_c$is the number of rigid constraints per atom. GC-TCT uses a rigid polytope approach to calculate the degrees of freedom while PT-TCT uses

$$n_c = \frac{1}{N}\sum_{i=0}^{N}\left(\frac{r_i}{2} + 2r_i - 3\right). \tag{2}$$

N is the number of atoms and r_i is the connectivity of each atom, i. The PT-TCT approach was then expanded further by including temperature dependence by Mauro and Gupta (MG-TCT) [6,7]. MG-TCT incorporates a multiplicative factor for each individual constraint that accounts for the temperature dependence of the constraint rigidity. MG-TCT also relates the configurational entropy to the degrees of freedom ($S_c \propto f$). These two extensions allow for a quantitative prediction of properties, a list of which is available in multiple review articles [8–12]. One property that is of interest for this work is the glass transition temperature (T_g) which can be predicted as a function of the average number of rigid constraints per atom at the glass transition temperature ($n_c(T_g)$) and a constant (A) calculated from a known composition,

$$T_g = \frac{A}{d - n_c(T_g)}. \tag{3}$$

Another approach for predicting glass-forming ability comes from calorimetry, where multiple glass stability parameters (GSP)

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https://doi.org/10.1016/j.actamat.2021.117432

have been proposed. These are parameters that are intended to estimate the resistance of a glass to crystallization. This topic was reviewed most recently in a statistical analysis from Jiusti et al. [13,14] who investigated glass stability parameters as a function of T_g, T_l (the liquidus temperature), T_c (the peak crystallization DSC temperature), T_x (the onset crystallization temperatures), η (viscosity), and $\eta(T_l)$ (liquidus viscosity). Jiusti et al. found that some glass stability parameters can roughly estimate r_{crit} and showed that the following expressions reasonably predicted the critical quench rate when using a linear relationship (these were chosen as representative from the recommended parameters with J being the least accurate):

$$K = \frac{T_c - T_g}{T_l}, \tag{4}$$

$$\gamma = \frac{T_c}{T_g + T_l}, \tag{5}$$

and

$$J = \frac{\eta(T_l)}{{T_l}^2}. \tag{6}$$

$K$is the stability parameter based on the work of Weinberg [15] and γcomes from the work of Lu and Liu [16]. It is worth noting that all the expressions above are functions of the glass transition temperature (a viscosity parameter), the liquidus temperature, and the rate of the kinetics (either through viscosity or an explicit measurement of peak crystallization kinetics). Although these expressions provide a correlation to the critical quench rate, there is an implicit flaw in Eqs. (4) and (5): the glass must be first synthesized to measure T_c. Synthesizing glasses is not practical for liquids that strongly favor crystallization, and as such, these parameters are not sufficient to predict new glass-forming systems. Although the J stability parameter is the least accurate of the three GSP discussed above, Eq. (6) is of interest since all of the parameters can be calculated and measured in the liquid state. Additionally, it captures the notion that a high liquidus viscosity is key to a good glass forming system [3,17] and it was tested more extensively in the work by Jiusti et al. [14,18,19]. The liquidus viscosity is expressed using the Mauro-Yue-Ellison-Gupta-Allan (MYEGA) equation [20] and the universal high-temperature limit of viscosity from Zheng et al. [21,22] as

$$\eta(T_l) = -2.93 + 14.93\frac{T_g}{T_l}\exp\left[\left(\frac{T_g}{T_l} - 1\right)\left(\frac{m}{14.93} - 1\right)\right]. \tag{7}$$

m is the liquid fragility index. All parameters in the above equation are also measurable using DSC in the liquid state [23,24].

When searching for new glass-forming compositions, yet another method for predicting glass forming ability is to search for a deep eutectic in the phase diagram, i.e., a local minimum in liquidus temperature [25]. This agrees with Eqs. (4), (6), and (7). Despite these various models, the role of the liquidus temperature, the glass transition temperature, the crystallization temperature, and the fragility index as they relate to GFA, have remained largely qualitative or have required extensive experimental characterization. Feller et al. [14,26] have presented an approach to predicting glass-forming ability based on covalency of the network and intermediate range structure; however, this requires extensive NMR measurements and remains impracticable for designing new glass-forming compositions.

According to the kinetic theory of glass formation [3], GFA can be most accurately quantified by the critical rate required to cool the liquid from above the liquidus temperature to the glass transition temperature while keeping the crystal volume fraction below under a certain threshold, typically defined by the detectability limit of x-ray diffractometry (XRD) [27]. The critical quench rate (r_{crit}) [13] is defined as the cooling rate at which the resulting volume fraction of crystals exactly equals this threshold limit. As such, a good glass-forming system has a very low critical quench rate. It is often assumed that there are some background nuclei always present in the system. r_{crit} is then primarily controlled by the rate of crystal growth (U) at the temperature with the fastest rate of growth (T_{max}). This corresponds to the critical quench rate as given by [13,28],

$$r_{crit} = \frac{T_l - T_{\max}}{\sqrt{\frac{X_s}{\pi N_s [U(T_{max})]^2}}}. \tag{8}$$

In this expression, X_s is the detectability limit, and N_s the number density of nuclei in the material. Though this gives a physical origin to the GFA, it is not trivial to measure and as such, limits the applicability of the expression.

The challenge of calculating the critical quench rate is largely due to an untenable calculation of the crystal growth rate due to multiple unknowns in the expression for crystal growth (from Wilson-Frenkel theory [29]) which is a function of the free energy difference between the crystal and liquid (ΔG), the diffusion co-efficient (often written with the Stokes-Einstein relationship, $D = kT/6\pi a\eta$ [30]), and the jump distance (a):

$$U(T) = \frac{kT}{6\pi\,\eta(T)a^2}\left(1 - \exp\left[-\frac{\Delta G(T)}{kT}\right]\right), \tag{9}$$

where k is Boltzmann's constant. Throughout this work a is held to 2.5 Å.

A new approach called toy landscapes modeling (TLM) has recently been shown to accurately predict the crystal growth rate of glass-forming melts with minimal assumptions [31]. The TLM method is a physics-based approach which creates a simplistic enthalpy landscape of the glass-forming liquid. It has been shown to accurately model structural relaxation, viscosity, and crystallization behavior [31]. It is based on creating a 'toy' landscape that closely mimics the distributions of states found in real landscapes. It uses a log-normal distribution of states and is parameterized through knowledge of the viscosity. When parametrizing TLM for crystal growth, it was found that the model was able to provide accurate predictions for growth with a root-mean-square-error (RMSE) of 0.333 orders of magnitude (when lead based compounds were omitted) [31]. As such, it is possible to use TLM to obtain the crystal growth parameter needed to calculate the critical quench rate given the T_g, T_l, and the liquid fragility index (m). This method allows for a direct evaluation of GFA with a completely quantitative and physics-based methodology that can be applied to any system.

In this article, we will present a novel method that combines a machine learning (ML) approach with the physics-based TLM model to offer quantitatively accurate predictions of glass-forming ability. This combined method enables prediction of the critical quench rate for common systems while simultaneously testing several common approaches for finding glasses with optimum GFA. The four approaches evaluated are:

a. Select a composition at a eutectic (thermodynamic approach)
b. Increase the ratio between the number of network formers to the modifiers (structural approach)
c. Find a glass where $n_c(T_g) \approx 3$(topological approach)
d. Maximize the liquidus viscosity (kinetic approach)

The reader should note that GFA is purely a representation of the rate at which the liquid must be quenched to reach the glass transition without reaching a detectable volume fraction of crystals. The GFA does not provide information about the degree of liquid-liquid immiscibility which may occur. Further work would be needed to account for phase-separation and as such it will not be addressed in this manuscript.

2. Methods

The ‘Toy Landscape Method’ was recently proposed by Wilkinson and Mauro [31] and uses an approximation of the energy landscape of a material. The landscape consists of a log-normal distribution of sites with equal probability spacing to ensure an equivalent degeneracy of each site. The distribution is then fit such that the slope of the configurational entropy vs temperature matches that which is predicted by the MYEGA model. This gives a set of states that are representative (though simplified) of the overall material behavior and can give kinetic and thermodynamic insights. The states can be used (along with a Boltzmann distribution) to calculate the free energy,

$$G=\sum_{i=0}^{N}\frac{\exp\left[-\frac{H_i}{kT}\right]}{\sum_{j=0}^{N}\exp\left[-\frac{H_j}{kT}\right]}H_i+kT\frac{\exp\left[-\frac{H_i}{kT}\right]}{\sum_{j=0}^{N}\exp\left[-\frac{H_j}{kT}\right]}\ln\left[\frac{\exp\left[-\frac{H_i}{kT}\right]}{\sum_{j=0}^{N}\exp\left[-\frac{H_j}{kT}\right]}\right], \tag{10}$$

in which $N$is the number of basins and H_i is the enthalpy of a particular basin. In this equation the second term is equivalent to $TS_c(T)$. The viscosity can then be given by,

$$\log_{10}\eta=\log_{10}\eta_\infty+\frac{T_gS_c(T_g)[\log_{10}\eta(T_g)-\log_{10}\eta_\infty]}{TS_c(T)}. \tag{11}$$

It is worthwhile to emphasize that TLM is not capable of capturing all underlying physics nor is it the only way to construct a landscape. The crystal free energy could then be approximated using 2 constants, H_{cry} and T_l (if assuming that the enthalpy of the crystal is constant),

$$G_{cry}=H_{cry}-k\frac{T}{T_l}\left[G_{liq}(T_l)-H_{cry}\right] \tag{12}$$

This gives all the parameters needed for Eq. (9) from a few experimentally accessible parameters. In the paper of Wilkinson and Mauro [31] they showed that using this simple model, one can predict the critical quench rate within an order of magnitude. Further details on TLM are available elsewhere [31].

In order to make predictions of the growth curve using TLM, the inputs required are T_g, m, T_l, H_{cry}, and $\log_{10}\eta_\infty$. The value of T_g comes from the neural network trained by Cassar et al. [32], the heat of fusion (H_{cry}) was shown to have minor effects on the prediction of the growth rate and as such, was held constant at -0.1 eV which is in line with our previous work [31], and $\log_{10}\eta_\infty$ can be approximated with $\log_{10}\eta_\infty=-2.93$ based on the work of Zheng et al. [21]. Additionally, consistent with Jiusti et al., [13] we set $X_s/N_s=10^{-5}$. To approximate the liquidus temperature and account for predictive error, we used a random forest method. The random forest approach works by constructing a large number of decision trees and then averaging results from each one to get a prediction. At the same time, polling the decision trees give a standard deviation (δ) which will be used as the error metric in this work.

To create a database for fragility for building a machine learning-based model, raw viscosity data was taken from GlassPy by Cassar [33]. The data are fit using the MYEGA model, and if the fit resulted in a RMSE of less 0.1 in log units, the data was accepted into the database. If a higher threshold was used, a large distribution of fragilities was found while if a lower threshold was used, the dataset was reduced greatly. The resulting database for fragility is then used as an input for Gaussian process regression, random forest, and linear regression. Both the random forest and Gaussian process regression hyperparameters were optimized through a grid search using Scikit-Learn [34] along with a 70% training set for cross-validation. The species considered in this model were O, B, Si, Al, Ca, Li, and Na. Lithium was only included to compare to previous work by Jiusti et al. [13].The RMSE of the fragility model using Gaussian Process Regression was 5.0 and the RMSE of the liquidus temperature using random forest was 60 K. Random forest, linear regression, and gaussian process regression were all used for liquidus temperature and fragility. However, we only used the model that reproduced the test set with minimal error. Neural networks were not considered as they offer no straightforward way to quantify uncertainty. The software and data used for training is available in the supplemental information.

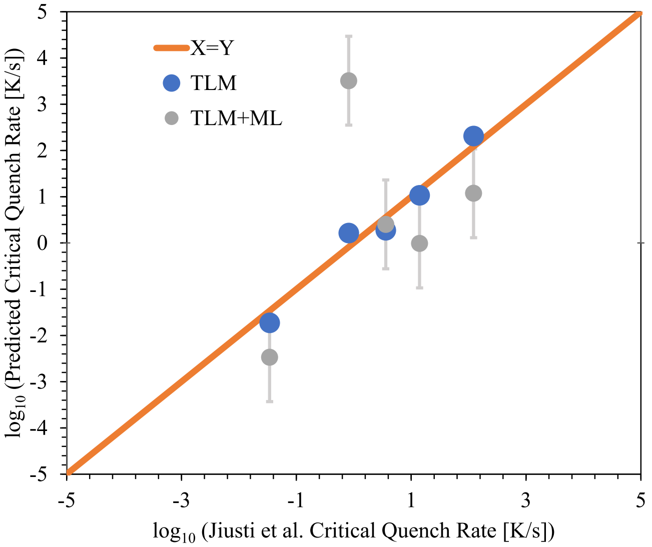

Fig. 1. Testing the predictive power of the ML + TLM vs just TLM showing less accurate results but with the error bars being within reasonable error. The TLM model gives a root-mean-square error of the log values of 0.25 log(K/s) while conversely the TLM+ML model shows an error of 1.8 log(K/s) but also has an average log error bar of 1.35 log(K/s). The highest point comes from a large error in the liquidus in GlassPy (citing Kaplun and Meshalkin [35]) which is reported as 1230 K while Jiusi et al. report a value of 1016 K [18,36]. Data for the liquidus temperatures in GlassPy can be found in the supplemental information.

The error bars for the critical quench rate were calculated by running the TLM two additional times, once where $m=m+\delta(m)$ and $T_l=T_l-\delta(T_l)$, and then once where $m=m-\delta(m)$ and $T_l=T_l+\delta(T_l)$. This gives the maximum and minimum quench rate from the average critical quench rate based on standard deviation of the predicted variables of interest. It is also important to keep in mind that TLM has an error associated with it (though significantly smaller than that of the machine learning methods).

3. Results

To first confirm the validity of the approach, we ran the model on five well-studied systems with experimental critical quench rate parameters available. These systems were chosen as they represent all atoms used in this study and only required one additional elemental parameterization (Li); they are: Li_2O $2B_2O_3$, Li_2O $2SiO_2$, Na_2O $2B_2O_3$, Na_2O $2SiO_2$, and CaO Al_2O_3 $2SiO_2$. All of these systems have well known parameters as reported by Jiusti et al. The results of this are shown in Fig. 1.

To understand the role of each parameter, we have explored three common glass-forming systems: soda lime silicate, sodium borosilicate, and calcium aluminosilicate. These glasses have well known structures (as discussed below) and comprise most industrial glasses in the commercial market today. It is not unreasonable to think that new compositions in the future can be derived from these families. As such, they remain some of the most important systems to understand for future glass compositions. The most important of these properties is the glass-forming ability. These three systems have been calculated in Fig. 2 using a combination of machine learning and TLM. This approach combines the empirical predictive power of ML with TLM to enable predictions of kinetic properties that have historically been inaccessible. Also, in Fig. 2, the liquidus temperature is shown for discussion of the thermodynamic and kinetic approaches to GFA. Fig. 2 is the first prediction of GFA based solely on composition and is a promising step towards decoding the glass genome [37–40].

Soda lime silicates (SLS) are the most commonly used glasses on earth. SLS consist of mostly silicon tetrahedra with 3 bridging oxygen (Q^3) with silicon being the only network former in the system. In Fig. 2A, the critical quench rate and liquidus temperature are shown for soda lime silicates with different modifier ratios and amounts of SiO_2. It is important to note that the SLS system shows a mild mixed modifier effect and that the critical quench rate slightly deviates from linearity with respect to $[Na_2O]/[Na_2O+CaO]$ [10,41].

Sodium borosilicates (NBS) serve as the base to many of the well-recognized commercial systems such as Pyrex, Duran, and Schott NBK-7 [3]. The structure is a mixture of the binary sodium borate and sodium silicate systems in which a fraction of boron species changes from three-fold to tetrahedral coordination. This leads to an increased connectivity of the network where nonbridging oxygens do not begin to form until a ratio of ~0.5 alkali to boron is present, though this value can change depending on cooling rate [42,43]. As the ratio continues to increase, the introduction of nonbridging oxygens near silicon tetrahedra results in a less connected network. The calculated critical quench rate and liquid temperatures for this system are shown in Fig. 2B. The effect of the conversion of boron units resulting in a non-monotonic progression of the critical quench rate can be clearly seen in Figure 2B (at $Na_2O/[Na_2O+SiO_2]$ ratio of approximately 0.35). The reader should note that, in Figure 2A and 2C, silica is held constant while in 2B the borate content is held constant. This is done so that it is easier to observe the non-linear evolution.

Calcium aluminosilicate glasses (CAS) are ubiquitous in geology and in industry. Understanding the formation of CAS systems can be used to gain greater control of applications of commercial applications, as well as a more detailed understanding of lava flows. The structure for this glass system is also anomalous to the SLS system due to the dependence on the ratio of calcium to aluminum. Since alumina tetrahedra are negatively charged, a charge balancing mechanism is needed. When the amount of calcium is greater than the amount of aluminum, the negative charge can be accommodated by calcium ions. Excess calcium will then create nonbridging oxygens that modify the underlying SiO_2 network. However, when insufficient calcium exists to charge balance the Al tetrahedron, which occurs when the ratio of Al_2O_3 to CaO is equal to or greater than one, it is debated whether three-coordinated oxygens or five-coordinated Al will form [44]. It remains unclear which mechanism takes place, but both would result in a higher network connectivity with fewer nonbridging oxygens. As such, it can be difficult to fabricate glasses in this range since they would require high quench rates. In Fig. 2C the estimates for the critical quench rate and liquidus temperature of the CAS system with varying percentages of SiO_2 can be found. In Fig 2C, progression of the calcium is seen to dominate the critical quench rate, which is intuitive since the calcium controls the structure of the aluminum units at all compositions. For further discussion of aluminum speciation on GFA see the work of Zheng et al. [45].

4. Discussion

Before discussing the results, it is worth noting the limitations and consequences of the model presented here. Since industrial processes have an upper limit to possible quench rates, this method allows for large compositional spaces to be removed from consideration and allows for the conversion of simple, accessible properties into the critical quench rate, making a much-needed step to decode the glass genome. Additionally, this method can be used to investigate the validity of the predictive methods discussed before. However, it is limited by the fact that there is an innate error in TLM (RMSE 0.333 orders of magnitude) when considering crystallization rates.

To understand the validity of glass forming theories in the literature, we can compare the results predicted by TLM to the four different approaches outlined in the Introduction. The thermodynamic approach (in which a eutectic composition is selected) works well for the soda lime silicate and sodium borosilicate systems; however, in the CAS system, there is no correlation between the liquidus temperature and the glass forming ability. Instead, the critical quench rate increases monotonically (corresponding to a decrease of the glass-forming ability) with respect to compositions despite a eutectic in Fig. 1C around $[CaO]/[CaO + Al_2O_3] = 0.5$.

The second approach utilizes Zachariasen's theory of glass formation [2]. In this approach, each component is classified as a network former or modifier. Since network formers, such as SiO_2, B_2O_3, and Al_2O_3, provide a connected framework for the glass system while modifiers act to break up this network, the GFA can be optimized by having a large amount of network formers and a small number of modifiers (intermediates are dealt with on a case-by-case basis). At first glance only the CAS system tends to follow this trend. However, NBS also follows this trend since these glasses undergo the boron anomaly. When examining the SLS data the glass forming ability undergoes a monatomic change as one modifier is switch to another keeping the mean connectivity of the network the same. This fact shows that Zachariasen's method cannot distinguish between types of modifiers which is a fundamental limitation in the method when designing glass compositions.

The third approach, which uses topological constraint theory (TCT), is the most recent and is based on a more nuanced structural understanding. TCT states that the optimal glass forming ability occurs when the atomistic degrees of freedom match the dimensionality of the system. Mauro and Gupta showed that the fragility [6] was related to the derivative of the number of rigid constraints with respect to temperature. This implies that the fragility cannot be predicted merely by the absolute value of the constraints alone. TCT is also unable to predict the liquidus, which creates two free parameters in total (i.e., fragility and liquidus temperature). Therefore, it is difficult to determine whether the number of constraints can describe glass forming ability (GFA) since the glass transition temperature is the only value that can be accurately predicted from the absolute number of constraints. In Fig. 3A, the GFA is plotted vs the number of constraints while holding the liquidus temperature at 1600 K, the fragility constant, and $A = 1500K$. Although this could be explored through a specific system this approach allows for a generalized approach allowing us to understand the nature of these materials. In Fig 3A, it is possible to see that the critical quench rate decreases as the number of constraints increase (approaching 3). The same trend holds true regardless of which fragility is chosen. However, as observed in the SLS system, the critical quench rate decreases as calcium is exchanged for sodium (decreasing the number of rigid constraints). This is due to the rapid depression of the liquidus temperature over the same range implying that the TCT approach is only valid when there are small changes in the liquidus temperature.

The failure of the constraint approach for quantifying the glass-forming ability in systems where the liquidus temperature is rapidly depressed means that a predictive model needs to incorporate the glass transition temperature, the fragility, *and* the liquidus temperature. This results in stability expressions that must be de-

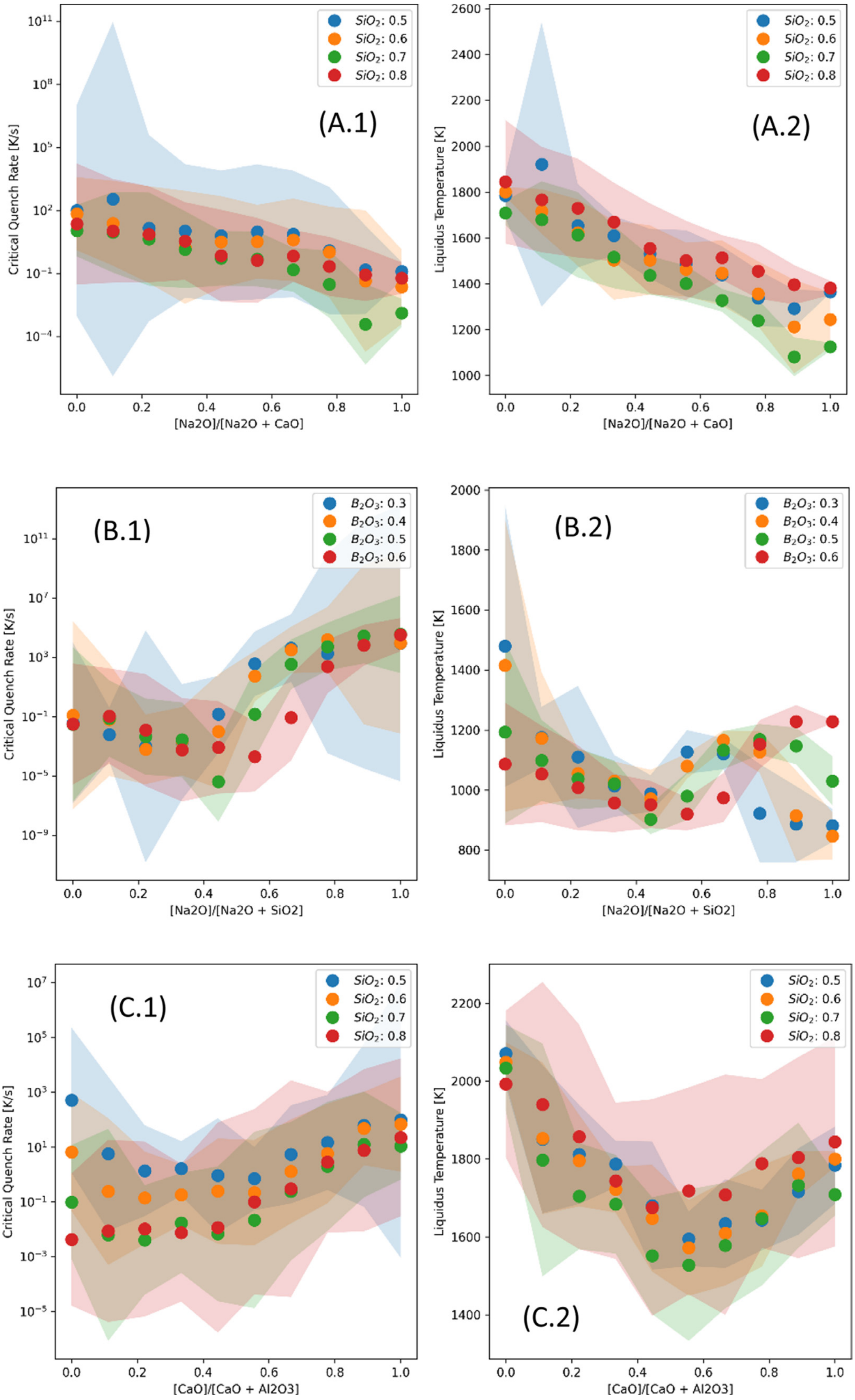

Fig. 2. (A) SLS, (B) SBS, and (C) CAS systems showing the critical quench rate (Panel 1) and the liquidus temperature (Panel 2). The points show the absolute value and shading shows the error bar calculations. The reader should note that the error is derived from the machine learning approaches and not from any other source.

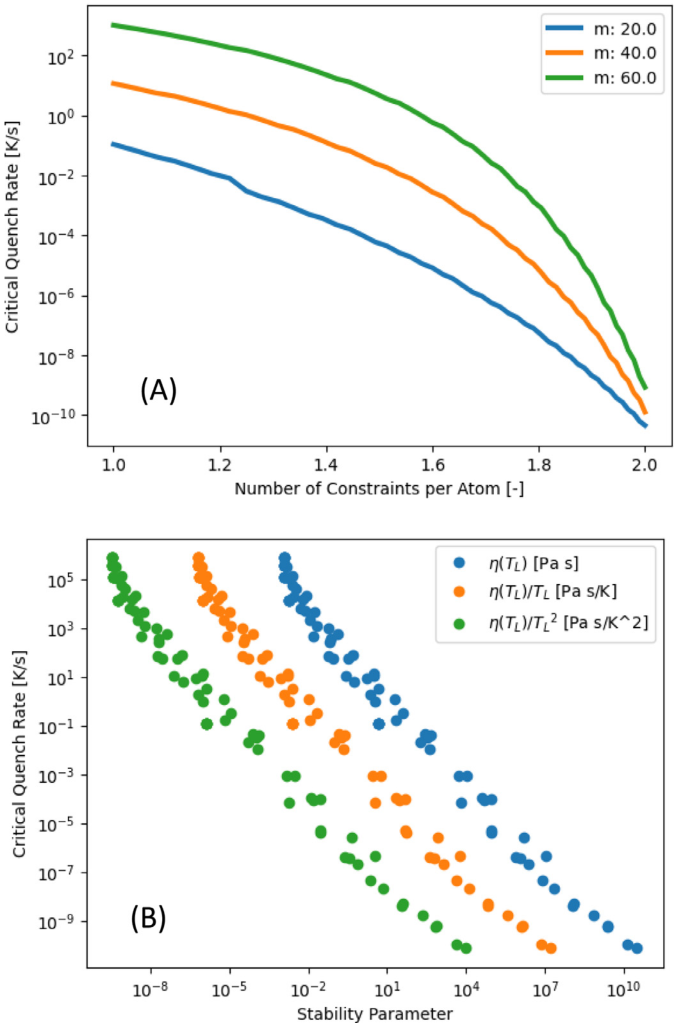

Fig. 3. (A) The critical quench rate as a number of constraints while the liquidus temperature is held constant at 1600 K. The constant for the glass transition temperature calculation was held at 1500. (B) The critical quench rate vs the two liquidus viscosity parameters. The data points were generated by doing a grid simulation with the glass transition temperature ranging from 500–1500 K, the fragility ranging from 20–100, and the liquidus temperature ranging from the glass transition temperature + 100–2000 K. When evaluating the correlation between the log of the stability parameter and the log of the critical quench rate an R^2 of 0.98 was calculated for each tested metric.

pendent on thermodynamics and kinetics, such as liquidus viscosity. In Fig. 3B, the clear relationship between the liquidus viscosity and the glass-forming ability is shown. This high correlation between the critical quench rate and both glass stability parameters (the liquidus viscosity while normalized and not normalized by the liquidus temperature) indicates that this is a strong metric when trying to predict which systems will form a glass. It is interesting that Jiusti et al. [13] had previously reported that the liquidus viscosity was not as important as other terms such as $T_c$when determining GFA. This indicates that T_c (the DSC peak crystallization temperature) seems to be a function of viscosity itself and is related to the peak crystallization temperature. Despite this, T_c remains impractical for the purpose of designing glass compositions since it requires a synthesized sample and differential scanning calorimeter measurements.

To recap we cannot employ Zachariasen's theory for **designing** glass compositions as it cannot distinguish modifiers. Also, we are unable to use most glass forming parameters as aids in designing glasses since they require synthesized glasses with specific markers from DSC or other experimental characterization. TCT theoretically can guide the design but only in systems where fragility and the liquidus temperature varies little. The eutectic approach works in some systems but not all. This leads to the testing of the liquidus viscosity. The liquidus viscosity is the only metric that required the same parameters as the physically based model. It then resulted in a linear relationship (R^2=0.98) between the critical quench rate and the value of interest, indicating this as the optimal glass forming metric.

The methods presented here allow for a quantifiable prediction of the critical quench rate using a new physics and machine learning hybrid approach. This predictive model improves our understanding of GFA more than previously reported methods in the literature. However, it is worth noting that if the TLM is not available, the liquidus viscosity gives a close approximation to the critical quench rate due to the strong linear relationship between the two. Other methods such as using eutectic compositions and pure structural approaches to predicting GFA were found to fail in some circumstances considered here. The liquidus viscosity also explains the origin of glass forming ability because the glass transition temperature and fragility are related to the structure of the glass and the liquidus temperature is purely driven by the thermodynamics of the liquid and crystal. This implies that the glass structure, liquid thermodynamics, and crystalline dynamics must be understood to understand glass forming ability.

5. Conclusions

Despite decades of research, we do not have a universal parameter for glass-forming ability that is quantitatively predictive from modeling. In this work, we used a combination of a physics-based model (viz., the toy landscape model) and machine learning to create a powerful tool that enables a quantitative prediction of the compositional dependence of critical quench rates. We then used this approach to validate other common approaches to choosing glass-forming liquids. We found that the Zachariasen approach and the eutectic approach both failed in some circumstances. The topological approach worked successfully with the caveat that the liquidus temperature could not vary dramatically. The best performing metric for glass-forming ability is simply the liquidus viscosity, where a higher viscosity at the liquidus temperature enables easier vitrification. This result is intuitive since it was the only metric that accounted for ***both*** thermodynamic and kinetic effects.

Declaration of Competing Interest

The authors declare no conflict of interests.

Acknowledgements

Work was supported by the National Science Foundation under grant number NSF-DMR 1746230.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.actamat.2021.117432.

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