MLL213 Term Paper Report

PREDICTION OF GLASS TRANSITION TEMPERATURE USING ARTIFICIAL NEURAL NETWORKS(ANN)

Submitted To:

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Abstract

We have used an ANN to predict the glass transition temperatures of various multicomponent oxide glasses with as high as 65 elements. The inputs were the composition vectors of various oxides and the model predicted the Tg for various oxides quite accurately (The author achieved an R^2 score of 0.998, and our model achieved an R^2 score of 0.925).

Introduction

The glass transition temperature is a temperature below which a material exists in a glassy state and above which the material exists in a rubbery state. Figure 1.A represents the variation of modulus of elasticity variation with temperature, thus highlighting the key aspect of the glass transition temperature. One can clearly note the change in the property after Tg is attained. Another aspect to note about Tg is that this transition is a second-order transition, which is characterized by a sudden jump of any thermodynamic property like gibbs free energy, Volume, etc. at Tg, this aspect is highlighted at Figure 1.B . A clear spike in Volume can be observed at Tg and above Tg compared to temperatures below Tg. If anyone is interested to find the melting point of the material from Tg, we can simply approximate it to Tg+(30-40 celsius/kelvin), of course if would depend on the material in use but in general this approximation works well with most of the materials that we use. It is interesting to note that there are 10^{52} compositions possible for the glasses, but the studied compositions to date are only 10^5, this fact motivated us to work on this field of glasses and to possibly predict the Tg of various oxide glasses which is by far one of the most important categories of commercial glasses by using Artificial neural Networks technique, the accuracy of which was quite good. Another thing to note is that the oxide glasses that we used had as high as 65 elements, but when training the model we had put several constraints so that we get accurate results. Another fact that motivated us to work on this paper was that there were various applications of Tg in various fields like crystallization, thermal tempering, and annealing. Tg had vivid applications in these fields which motivated us to predict the Tg of various oxides glasses.

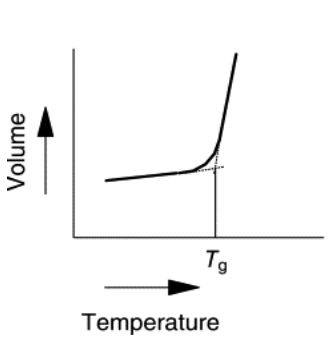


Fig 1.A

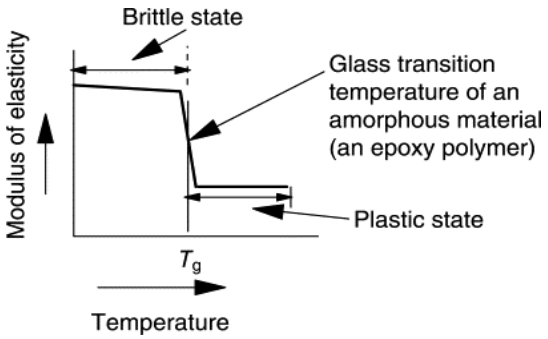


Fig 1.B

Methods Implemented & Data Collection

Data Collection

To get the Tg data for our term paper, we used the Sciglass 7.12 database, which was built using academic research and numerous patents.In order to train our data appropriately we need to place some limitations, which are as follows -

1)We kept the atomic fraction of oxygen to be at least 10% in each oxide so as to restrict our dataset to oxide glasses because they are the most relevant and practical in terms of commerce.

2)The presence of each chemical element in at least 1% of the dataset entries is the second restriction. It's necessary so that every atom in the analysis has at least some representation during training.

3)We only take into account molecules that contain oxygen and at least two other chemical components. Because unavoidable (and unaccounted for) impurities typically have a significant impact on the Tg of single oxides (such as SiO_2, B_2O_3, GeO_2, TeO_2 etc.), affecting our analysis and making it subject to significant uncertainty.

After applying these constraints, we obtained a dataset with 55,150 examples out of which nearly 10000 were used for testing and around 45000 were used for training We considered total 65 elements including most importantly oxide glass forming ions i.e. silicon, boron, phosphorous, tellurium, germanium, arsenic, and antimony. The majority of oxide glasses that we consider are fabricated using 4 different components (including oxygen). And just 10% of the dataset's compositions include 11 or more distinct elements in them.

Although the readings range from 300 K to 1450 K, but most of the examined glasses had a Tg close to 750 K.

Methods Implemented

In this study, the glass transition temperature of multicomponent oxide glasses was predicted using ANNs (Multilayer perceptron model). Glass composition (in at%) was utilized as a fingerprint to predict the glass transition temperature.

The author had used a TPE(Tree based Parzen Estimator) to choose the best hyperparameters:

Hyperparameter	Lower search space limit	Upper search space limit	Best topology
1st hidden layer			
Nº of neurons	60	500	350
Dropout (%)	0	25	6
2nd hidden layer			
Nº of neurons	50	500	500
Dropout (%)	0	50	11
Batch (2^n)	$n = 5$	$n = 10$	$n = 7$
Patience	7	15	15

We used the same architecture and hyperparameter values as the author.

Results

1.The author got an R^2 value of 0.998 which is a very good prediction (Fig 1). The network predicted 95% of the data with a relative deviation ϕ lower than ±9%, whereas 90% of the data were predicted with a ϕ lower than ±6%. This is an important finding because the scattering in the reported Tg in our dataset is of the same order.

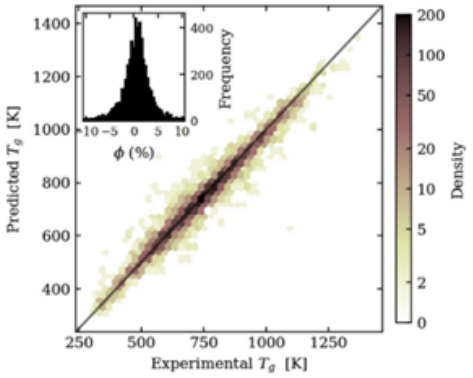


Fig. 1 Predicted Tg by the trained network versus reported values for the test dataset containing 5515 oxide glasses.

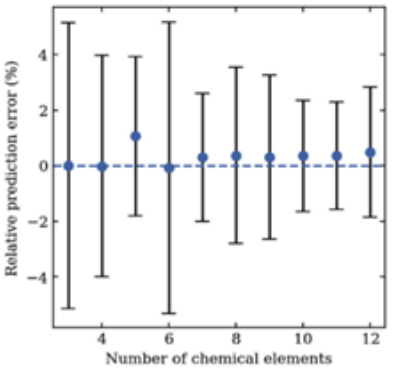


Fig. 2

2. The author has randomly drawn 50 different examples from the test set for each given number of chemical elements, ranging from 3-13. It is seen from the graph that the mean and standard deviation of the relative prediction error in Tg does not depend on the number of elements in the glass (Fig 2). This is a very significant result for the development of new multicomponent glasses composed of a large number of chemical elements.

3. Moving further, we saw in the data collection part that the author didn't consider single oxide glasses but the trained network worked well and extrapolated the behavior of complex glasses to a reasonable value of Tg for single oxide glass-formers, except Silica (SiO2), for which the predicted value was lower than expected (see Table 1). This result, however, is in agreement with what we observed with glasses having Tg values above 1250 K, that the network tends to underestimate the glass transition temperature in these cases.

4. Finally, 15 glass compositions published in a specialized journal (Journal of Non-Crystalline Solids) between 2016 and 2018, which were not part of the original dataset, were tested. Overall relative deviation, ϕ, came out to be very reasonable (see Table 2), and supports the adoption of ANN for our main goal, that is to aid in the development of new glass compositions.

Composition	T_g (mean and std)	Min. T_g	Max. T_g	Predicted T_g
SiO_2	1440 (60)	1293	1495	1210
B_2O_3	540 (20)	483	580	534
GeO_2	800 (30)	743	883	753
V_2O_5	484 (3)	482	492	497
TeO_2	580 (40)	498	658	573
P_2O_5	610 (50)	536	673	609
Sb_2O_3	550 (50)	500	618	547
As_2O_3	441 (7)	433	453	438

Table 1. Test of the Tg prediction capability of the induced network for glass compositions containing only two chemical elements. All values in Kelvin.

Composition	Reported T_g	Predicted T_g	ϕ
$Ag_{14.3}V_{14.3}Te_{9.5}O_{61.9}$	474	475	−0.2
$Pb_{9.7}Ti_{1.7}Te_{16.9}B_{19}O_{61.7}$	563[b]	613	−8.9
$Mo_{11.4}W_{2.9}Te_{14.3}O_{71.4}$	589	626	−6.2
$Bi_{11.1}Mo_{10.3}Te_{10.3}O_{68.3}$	610	619	−1.4
$Cu_{2.3}Na_{17.1}P_{17.1}O_{58.5}$	623	612	1.8
$Bi_{14.3}Ti_{2.4}Te_{7.1}B_{14.3}O_{61.9}$	683	695	−1.7
$Ga_{9.7}Zn_{4.9}P_{18.9}O_{66.5}$	762	716	6.1
$Ba_{4.7}Cu_{0.9}K_{5.2}Zn_{13}B_{30.5}O_{56.2}$	775	757	2.4
$Cu_{12.8}Ti_{3.8}P_{17.9}O_{65.5}$	777	811	−4.3
$Ca_{2.9}Fe_{2.1}Mg_{42}Na_{0.4}P_{16.8}B_{6.4}O_{85.2}$	839	836	0.3
$Ca_{5.1}Fe_{0.1}K_{4.1}Mg_{3.9}Mn_{0.3}Na_{1.2}Al_{0.4}P_{1.4}Si_{19.8}O_{59.7}$	866	904	−4.4
$Ca_{15.8}Na_{2.4}Nb_{2.6}P_{15.8}O_{63.2}$	885	836	5.5
$Li_{1.9}Mg_{0.3}Zn_{0.6}Zr_{0.5}Ti_{0.5}Al_{10.2}P_{1.3}B_{2.7}Si_{15.8}O_{64}$	890	951	−6.9
$Ca_{1.9}Mg_{2.5}Sr_{1.9}Al_{6.8}B_5Si_{19.2}O_{62.6}$	940	984	−4.7
$La_{7.3}Al_{16.6}P_{6.7}Si_{5.7}O_{63.7}$	1118	1062	5.0

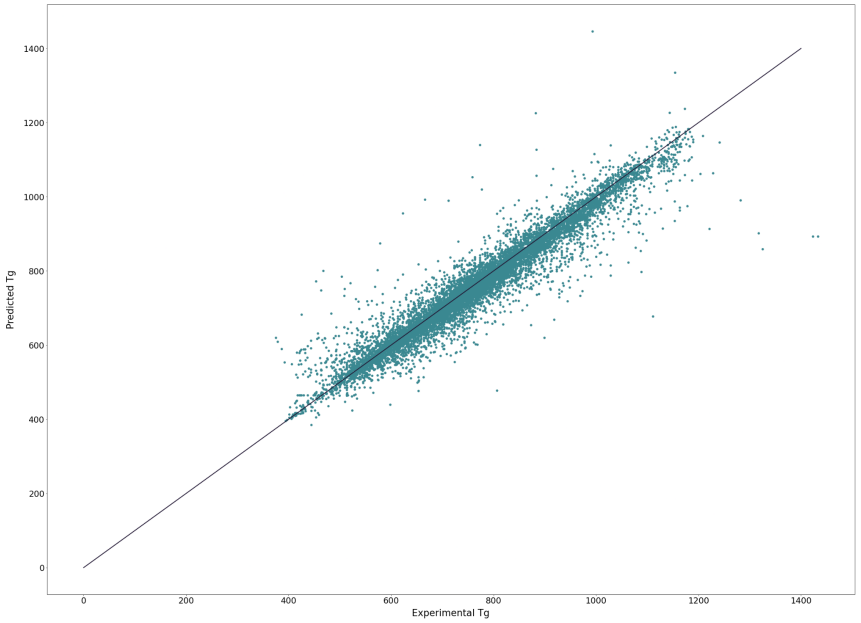
Table 2. Test of the Tg predicting capability of the induced network for 15 glass compositions that were not included in the original dataset. Tg is in Kelvin and ϕ is in %.

Concluding Remarks:

The majority of the Tg values for oxide glasses that had been collected in the past were incorporated into this investigation. For the purposes of training and validating the network, we compiled data on approximately 55,000 distinct glass compositions and the temperatures associated with each glass transition. Only three to twenty-one of the 45 chemical components under consideration can be found in these mixtures. Then, we deliberated, programmed, trained, and tested an artificial neural network that can predict the Tg of oxide glasses with an acceptable amount of error, roughly equivalent to the scatter present in the original dataset. Glasses with a Tg that is extremely high (over 1250 K) have a larger prediction uncertainty, but this uncertainty is not related to the amount of components that are present in the glass. This is the first time that we are aware of anyone attempting to predict the Tg of multicomponent glasses by using an updated method for ANN hyperparameter optimization. To the best of our knowledge.
In addition, the methodology that was developed here is very adaptable, so it can be easily modified to predict a wide range of different glass qualities. As a result of this, ANNs have the potential to be utilised in innovative ways to select and construct glasses that possess desirable properties for use in cutting-edge applications.

Our Implementation:

Reference Research Paper, Code & Dataset: https://github.com/aniket3301/MLL213-Term-Paper
Scatter Plot:



R2 Score: 0.925