

PREDICTION OF ULTRASONIC PARAMETERS AT LOW TEMPERATURES FOR TELLURITE GLASSES USING ANN

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The deformation potential in $x \text{Nb}_2\text{O}_5 - (1-x) \text{TeO}_2$, $0.1 \text{PbO} - x \text{Nb}_2\text{O}_5 - (0.9-x) \text{TeO}_2$ and $0.2 \text{PbO} - x \text{Nb}_2\text{O}_5 - (0.8-x) \text{TeO}_2$ tellurite glass systems, was calculated using the Central Force Theory (CFT). Analysis of the compositional dependence of the experimental deformation potential in the three glass systems has been achieved. The quantitative analysis was based on the number of oxygen atoms that vibrate in the double well potential, the deformation potential and the mutual potential energy according to (CFT) with longitudinal elongation. Comparisons between the previous measured values and the predicted values using the Artificial Neural Network (ANN) model for 11 different tellurite glass compositions have been implemented which showed good agreement between them.

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1. Introduction

Tellurite-based glasses have been the subject of both academic and technological interest due to the unique properties [1-11]. The longitudinal ultrasonic attenuation in ternary $80 \text{TeO}_2 - (20-x) \text{WO}_3 - x \text{K}_2\text{O}$ tellurite glass systems has been measured [12] using pulse echo technique at ultrasonic frequencies 2, 4, 6 and 8 MHz in the temperature range from 160 to 300 K. The absorption spectra showed the presence of well-defined broad peaks at various temperatures depending upon the glass network structure and the operating frequency. ANN technique has been used to simulate and predict important parameters such as density, longitudinal and shear ultrasonic velocities and elastic moduli for more than 30 glass compositions has been reported [13] and the predicted results were found to be in good agreement with those experimentally measured parameters.

To continue our previous works with tellurite glasses, three different glass series $x\text{Nb}_2\text{O}_5 - (1-x) \text{TeO}_2$, $0.1 \text{PbO} - x\text{Nb}_2\text{O}_5 - (0.9-x) \text{TeO}_2$ and $0.2 \text{PbO} - x\text{Nb}_2\text{O}_5 - (0.8-x) \text{TeO}_2$ with different Nb_2O_5 contents and constant PbO contents were investigated. Quantitative analysis based on (CFT) with longitudinal elongation, was performed to investigate the effect of Nb_2O_5 as a network modifier on the structure of the three glass systems. The (ANN) model was designed to predict activation energy and deformation potential.

2. Theoretical Considerations

Bridge and Patel (1986) [14] presented a model of the magnitude of the two-well barrier heights according to the (CFT) and deformation potentials that would occur based on the phenomenological theory.

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Considering a linear arrangement of three atoms consisting of an anion in the middle of two cations (or vice versa) separated by a distance R and assuming that the potential energy of the system is given by a superposition of two potentials of the form in equation (1),

$$U = \left(\frac{-a}{r}\right) + \left(\frac{b}{r^m}\right) + \left[\left(\frac{-a}{(R-r)}\right) + \left(\frac{b}{(R-r)^m}\right)\right] \quad (1)$$

Deformation potential was calculated theoretically according to equation (2) given by Bridge and Patel [11] as,

$$D_{th} = \frac{q}{2} \left(\frac{M}{n_b N_A \rho} \right) \frac{\delta y}{\delta x} \quad (2)$$

where q is longitudinal modulus, M is the mass of one kilo mole, N_A is Avogadro's number, ρ is the density, n_b is the number of A-O-A units per formula unit (4 for TeO_2 , 6 for Nb_2O_5 , and 4 for PbO), δx is the bond length times 2 and δy is the separation of minima in the two-well potential.

3. Analysis and Discussions

In the present study, several neural network architectures are designed and tested to finally find and assign the best network model to simulate the acoustic properties of tellurite glasses based on minimizing the Root Mean Square Error (RMS-Error). The experimental acoustic properties of some tellurite glasses are used in this study for training the developed ANN model. Then the properties of pure tellurite glass will be predicted using this developed and trained model without the need to make new experiments. Different configurations of hidden neurons have been checked to find the best number of neurons in the hidden layer. It was found that 12 neurons in the hidden layer produce the best results. This model enables the direct simulation of such parameters listed before without melting raw material oxides (as in some cases, they are of high costs), depending on some theoretical input parameter such as molecular weight, dissociation energy of oxides to give the predicted outputs.

Table 1 collected predicted values of density, attempt frequency, activation energy, deformation potential and number of loss centers per unit volume of the present investigated tellurite glass series. Values of TeO_2 glass [15] were used for the training model. Fig.1 represented the mutual potential energy for different elongations for binary and ternary tellurite glasses according to (CFT) with the equilibrium interatomic distance for binary $(100-x)\text{TeO}_2 - x\text{Nb}_2\text{O}_5$, ternary $(90-x)\text{TeO}_2 - x\text{Nb}_2\text{O}_5 - 0.1\text{PbO}$ and $(80-x)\text{TeO}_2 - x\text{Nb}_2\text{O}_5 - 0.2\text{PbO}$ glasses. For the binary tellurite glass system $(100-x)\text{TeO}_2 - x\text{Nb}_2\text{O}_5$, the oxygen density increases from 4.299×10^{28} to 4.539×10^{28} (m^{-3}), the mutual potential energy increases from 0.306 (eV) to 0.351 (eV) with increasing the equilibrium interatomic separation from 0.200 (nm) to 0.204 (nm) with increasing the modifier percentage from 5 mol.% to 20 mol.% for the elongation factor $e \approx 74$ %. For the ternary tellurite glass systems $(90-x)\text{TeO}_2 - x\text{Nb}_2\text{O}_5 - 0.1\text{PbO}$ and $(80-x)\text{TeO}_2 - x\text{Nb}_2\text{O}_5 - 0.2\text{PbO}$, the oxygen density increases from 4.030×10^{28} to 4.447×10^{28} and decrease from 4.579×10^{28} to 4.409×10^{28} (m^{-3}), respectively, the mutual potential energy increases from 0.291 (eV) to 0.352 (eV) with increasing the equilibrium interatomic separation from 0.1997 (nm) to 0.2047 (nm) and 0.2004 (nm) to 0.2054 (nm), respectively, with increasing the modifier percentage from 0 mol.% to 20 mol.% for the elongation factor $e \approx 74$ %.

Table 1: Experimental and predicted densities $\rho (M)$, $\rho (ANN)$, attempt frequencies $f_o (M)$, $f_o (ANN)$, activation energies $E_p (M)$, $E_p (ANN)$, deformation potentials $D (M)$, $D (ANN)$ and number of loss centers per unit volume $n (M)$, $n (ANN)$.

Glass composition mol %			ρ (kg/m ³)	$\rho_{(ANN)}$ (g/m ³)	$F_o \times 10^{11}$ (s ⁻¹)	$F_{o(ANN)} \times 10^{11}$ (s ⁻¹)	E_p (eV)	$E_{p(ANN)}$ (eV)	D (eV)	$D_{(ANN)}$ (eV)	$n \times 10^{27}$ (m ⁻³)	$n_{(ANN)} \times 10^{27}$ (m ⁻³)
TeO ₂	Nb ₂ O ₅	PbO										
1.00			5.101	5.03	-	0.0263	0.070	0.070	0.255	0.255	1.780	1.779
0.96	0.04		5.632	5.56	7.6500	7.6001	0.196	0.200	0.507	0.507	1.800	1.800
0.9	0.1		5.506	5.43	3.7400	3.7419	0.193	0.190	0.501	0.501	1.400	1.401
0.85	0.15		5.417	5.34	2.4000	2.3981	0.184	0.185	0.486	0.486	1.300	1.299
0.775	0.225		5.290	5.22	1.2700	1.2709	0.175	0.173	0.468	0.468	0.920	0.920
0.9	0	0.1	5.845	5.855	4.3300	4.3200	0.184	0.187	0.485	0.484	1.740	1.741
0.85	0.05	0.1	5.784	5.780	2.4400	2.4410	0.177	0.175	0.472	0.474	1.458	1.460
0.8	0.1	0.1	5.723	5.715	1.6700	1.6702	0.172	0.170	0.464	0.464	1.247	1.245
0.7	0.2	0.1	5.529	5.512	0.6100	0.6060	0.161	0.160	0.444	0.441	1.006	1.000
0.8	0	0.2	7.278	7.266	2.9300	2.9200	0.180	0.180	0.477	0.477	1.753	1.750
0.7	0.1	0.2	6.025	6.017	2.0800	2.0796	0.175	0.174	0.469	0.470	1.599	1.600
0.6	0.2	0.2	5.904	5.892	1.1500	1.1470	0.168	0.166	0.457	0.455	1.450	1.444

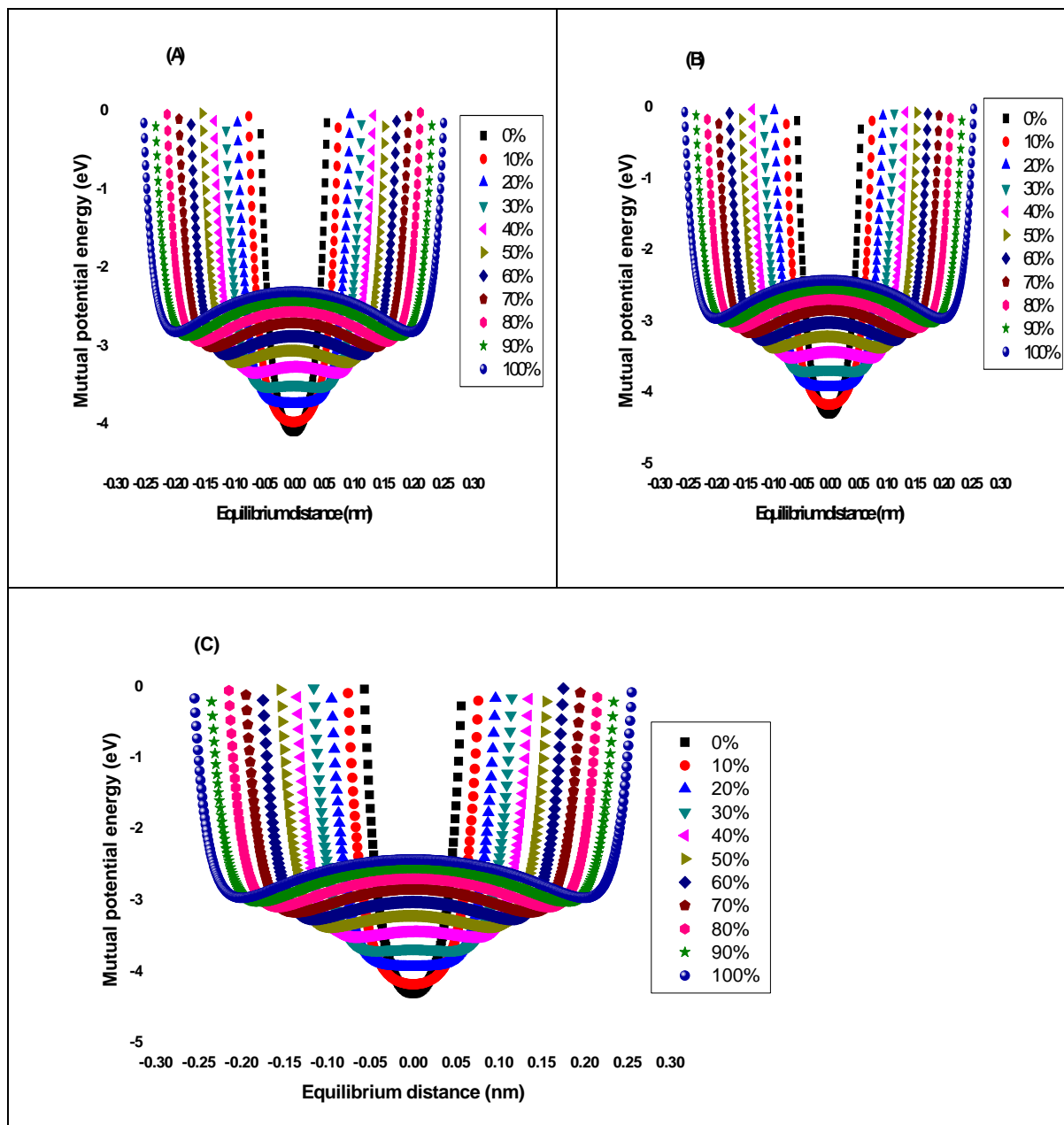


Fig. 1: Potential wells for longitudinal motion of oxygen atoms at elongations from 0 % to 100 % for; (a) $0.95 \text{ TeO}_2 - 0.05 \text{ Nb}_2\text{O}_5$, (b) $0.8 \text{ TeO}_2 - 0.1 \text{ Nb}_2\text{O}_5 - 0.1 \text{ PbO}$, (c) $0.7 \text{ TeO}_2 - 0.1 \text{ Nb}_2\text{O}_5 - 0.2 \text{ PbO}$.

In terms of oxygen atoms vibrating between static heavier atoms, the total number of acoustically active two well systems will be proportional to oxygen density. For the systems under investigation, the presence of different types of bonds; (Te-O-Te), (Te-O-Nb), (Te-O-Pb), (Nb-O-Nb), (Nb-O-Pb) and (Pb-O-Pb) means that there will be a spread of atomic ring sizes in the glasses. Of course, the average ring size in a given tellurite glass will be larger than the ring sizes occurring in the nearest equivalent crystalline tellurite, although there will be some rings smaller than the crystalline one. Thus, one can expect the number of distorted cation-cation spacing and the average degree of elongation increase with average ring size. For a given ring size, variations in bond strength do not affect the total number of distorted cation-cation spacing or the degree of distortion. As the A-O-A bond force constant increases, so also does the energy required to produce a given degree of bond angle or length distortion.

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Addition of niobium atom with six coordination number instead of tellurium atom with four coordination number, will increase the oxygen density and increases the equilibrium interatomic separation and increasing the mutual binding energy, i.e. the structure going into have strong connections as the more niobium atoms with higher coordination number enter the tellurite network. This is added to the stated before for the strengthen or weakening of the structure by the different additives. The preceding scene will be elucidated in the two well systems, in which the [O] atom is considered to move while the A- atom is infinitely heavy.

In comparing the glass systems, the strengthen structure of binary system has a wide range of elongation and consequently potential energy height which indicates that as the equilibrium interatomic distance increase, elongation increases which comes from the reverse relationship between the elongation factor and the interatomic distance. By other words, as a structure strengthen more, a more elasticity (i.e., elastic moduli) will result and leads to an increasing elongation percentage. This explains the less elongation percentage and less height of potential barrier in the ternary systems than the binary system.

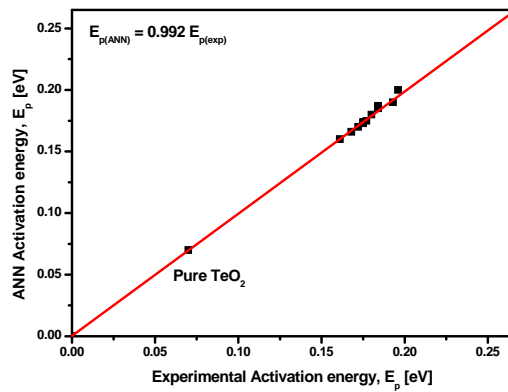


Fig. 2: Plot of the experimentally and predicted activation energies E_p and $E_{p(ANN)}$.

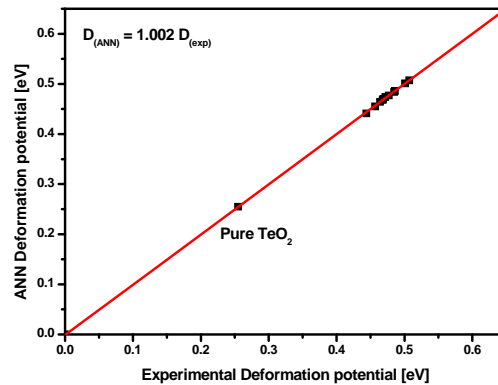


Fig.3: Plot of the experimentally and predicted deformation potential (D) and $D_{(ANN)}$.

Figs.2-3 show the plots of the experimentally obtained values of activation energy and deformation potential against those predicted using our model. The solid straight lines represent slopes which are near to unity indicating the good agreement between the experimentally obtained results and those predicted. The deviations between those the predicted and experimentally obtained values are in most cases lower than 0.5 % and never greater than 5 % in the investigated glasses, which gives a good approach for predicting our results revealing that our model is valid. It is very interested to gather the present analysis with other physical properties [16-20] to understand this interesting glass. It is very interested to gather the present analysis with other physical properties [16-20] to understand this interesting glass.

4. Conclusions

It can be concluded that:

- A model have been developed using ANN for direct estimation of the density, number of loss centers per unit volume, activation energy and deformation potential according to CFT of oxide glasses from their compositions,
- Comparison between the measured values and the predicted values for over 11 different tellurite glass compositions, showed good agreement between them,
- Addition of Nb₂O₅ to tellurite glass network will strengthen the structure through the increase in the mutual potential energy and the decrease in the experimental acoustic deformation potential.

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