UNIVERSITI PUTRA MALAYSIA

ARTIFICIAL NEURAL NETWORK ASSESSMENT FOR PREDICTING ELASTIC AND OPTICAL PROPERTIES OF BINARY TELLURITE AND BORATE GLASS SYSTEMS

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By

NURAIDAYANI BINTI EFFENDY

Thesis submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirement for the Degree of Doctor of Philosophy

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Chairman : Sidek Hj. Ab Aziz, PhD
Faculty : Science

The introduction of artificial neural networks (ANNs) in the glass field has greatly improved this area to further increase fabrication productivity. ANNs are the systems that help the glass expert to estimate a few parameters such as density, ultrasonic velocity, elastic moduli and optical band gap in the glass composition. In this present works, the ANNs system was implemented in a series of zinc-tellurite (ZnO-TeO_2), bismuth-tellurite (Bi_2O_3-TeO_2), zinc-borate (ZnO-B_2O_3) and bismuth-borate (Bi_2O_3-B_2O_3) glass systems which have been successfully fabricated using conventional melting and quenching methods with the configuration of $(ZnO)_m$ $(TeO_2)_{100-m}$ where m = 0, 5, 10, 15, 20, 25, 30 mol%, $(Bi_2O_3)_n$ $(TeO_2)_{100-n}$ where n = 0, 5, 7, 10, 13, 15 mol%, $(ZnO)_p$ $(B_2O_3)_{100-p}$ where p = 0, 40, 45, 50, 55, 60 mol% and $(Bi_2O_3)_q$ $(B_2O_3)_{100-q}$ where q = 0, 40, 45, 50, 55, 60 mol%, respectively. The experimental measurements have been investigated on the physical, structural, elastic and optical properties of binary tellurite and borate glass systems containing various amounts of ZnO and Bi_2O_3 concentrations. The experimental density measurement on the effect of ZnO substitution in both tellurite and borate glass systems showed the increment values as the amount of ZnO content increases with the highest density value of 5.283 g/cm^3 at 30 mol% of ZnO in tellurite glass systems. Meanwhile, the molar volume value displayed an opposite behavior with the glass density which the lowest value of the molar volume is 21.569 cm^3/mol at 60 mol% of ZnO in borate glass systems. The dropping value of the molar volume is attributed to the changes in the glass network connectivity. For the Bi_2O_3 substitution in both tellurite and borate glasses, the experimental density and molar volume exhibited a similar behavior which is increasing with the increase of Bi_2O_3 content. The highest density and molar volume values are 6.550 g/cm^3 and 46.935 cm^3/mol at 60 mol% of Bi_2O_3 in borate glass systems, respectively. The glassy state and amorphous nature of all glass samples have been confirmed through the presence of a broad hump peak in the XRD analysis. FTIR transmission and Raman absorption spectra have discovered the existence of

TeO_4, TeO_3, BO_4 and BO_3 structural units in the glass samples. The substitution of modifier ZnO and Bi_2O_3 into the tellurite and borate glass structure caused the glass structure to become more rigid and increase the elastic moduli values. This modification process affected the formation of bridging oxygen which leads to an increase in cross-link density and gives a better packing in the glass structure as calculated in bond compression and Makishima-Mackenzie theoretical model. The optical behavior revealed that the shifts of the absorption edge to the longer wavelength leading to the reduction in the optical band gap value. The minimum optical band gap value for the effect of ZnO and Bi_2O_3 substitution in both tellurite and borate glass systems is 2.557 eV at 30 mol% of ZnO in tellurite glass systems and 2.210 eV at 15 mol% of Bi_2O_3 in tellurite glass systems. Subsequently, the experimental values resulting from the composition of the glass series were compared with the values obtained from the prediction by ANNs. This study has concluded that the ANNs system was relevant to be used in the glass fields since the coefficient of R^2 values showed by the prediction against the experimental graph were between 0.9941 to 1.000 which is considered to be very satisfactory.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk Ijazah Doktor Falsasah

PENILAIAN RANGKAIAN NEURAL ARTIFISIAL UNTUK MERAMAL SIFAT ELASTIK DAN OPTIK BAGI SISTEM BINARI KACA TELLURIT DAN BORAT

Oleh

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Pengenalan rangkaian neural artifisial (ANN) di dalam bidang kaca telah meningkatkan hasil produktiviti sektor ini. Rangkaian neural artifisial adalah suatu sistem yang membantu pakar kaca untuk meramal beberapa parameter seperti ketumpatan, halaju ultrasonik, moduli elastik dan jurang jalur optik bagi suatu komposisi kaca. Dalam kajian ini, sistem ANN telah dilaksanakan pada sistem kaca zink-telurit (ZnO-TeO_2), bismut-telurit (Bi_2O_3-TeO_2), zink-borat (ZnO-B_2O_3) and bismut-borat (Bi_2O_3-B_2O_3) di mana sistem kaca ini telah berjaya dihasilkan mengunakan kaedah lebur dan pelindapan secara konvensional dengan konfigurasi $(ZnO)_m$ $(TeO_2)_{100-m}$ di mana m = 0, 5, 10, 15, 20, 25, 30 mol%, $(Bi_2O_3)_n$ $(TeO_2)_{100-n}$ di mana n = 0, 5, 7, 10, 13, 15 mol%, $(ZnO)_p$ $(B_2O_3)_{100-p}$ di mana p = 0, 40, 45, 50, 55, 60 mol% dan $(Bi_2O_3)_q$ $(B_2O_3)_{100-q}$ di mana q = 0, 40, 45, 50, 55, 60 mol%. Pengukuran eksperimen telah dikaji pada sifat fizikal, struktur, elastik dan optik untuk sistem kaca telurit dan borat yang mengandungi pelbagai kandungan ZnO dan Bi_2O_3. Pengukuran ketumpatan eksperimen terhadap kesan penggantian ZnO pada kedua-dua sistem kaca telurit dan borat menunjukkan nilai kenaikan apabila jumlah kandungan ZnO meningkat dengan nilai ketumpatan tertinggi 5.283 g/cm^3 pada 30 mol% ZnO dalam sistem kaca telurit. Sementara itu, nilai isipadu molar menunjukkan sifat yang berlawanan dengan ketumpatan kaca di mana nilai terendah isipadu molar adalah disekitar 21.569 cm^3/mol pada 60 mol% ZnO dalam sistem kaca borat. Nilai penurunan isipadu molar ini adalah disebabkan oleh perubahan dalam sambungan rangkaian kaca. Untuk penggantian Bi_2O_3 dalam kaca telurit dan borat, ketumpatan dan isipadu molar yang diukur secara eksperimen menunjukkan tingkah laku yang serupa iaitu meningkat dengan peningkatan kandungan Bi_2O_3. Nilai tertinggi ketumpatan dan isipadu molar adalah masing-masing 6.550 g/cm^3 dan 46.935 cm^3/mol pada 60 mol% Bi_2O_3 dalam sistem kaca borat. Sifat berkaca dan amorfus untuk semua sampel kaca telah disahkan melalui kehadiran puncak yang luas dalam analisis XRD. Transmisi FTIR dan spektrum penyerapan Raman menemui keberadaan struktur TeO_4, TeO_3, BO_4 dan BO_3 dalam sampel kaca. Penggantian pengubah ZnO dan Bi_2O_3 ke dalam struktur kaca telurit dan

borat menyebabkan struktur kaca menjadi lebih kukuh dan meningkat nilai sifat elastik. Proses pengubahsuaian ini mempengaruhi pembentukan oksigen penghubung yang menyebabkan peningkatan kepadatan pautan silang dan memberikan ikatan yang lebih baik dalam struktur kaca seperti yang telah dihitung dalam teori pemampatan ikatan dan *Makishima-Mackenzie* model. Tingkah laku optik pula menunjukkan bahawa peralihan pinggir penyerapan ke bahagian gelombang yang lebih panjang menyebabkan tenaga jurang optik menjadi berkurang. Nilai minimum jurang jalur optik kesan daripada penggantian ZnO dan Bi_2O_3 di kedua-dua sistem kaca telurit dan borat adalah 2.557 eV pada 30 mol% ZnO dalam sistem kaca telurit dan 2.210 eV pada 15 mol% Bi_2O_3 dalam sistem kaca telurit. Selepas itu, nilai eksperimen yang dihasilkan dari komposisi siri kaca dibandingkan dengan nilai yang diperolehi dari ramalan oleh rangkaian neural artifisial. Kajian ini telah menyimpulkan bahawa sistem ANN ini relevan untuk digunakan di dalam bidang kaca kerana nilai pekali R^2 yang ditunjukkan adalah diantara 0.9941 hingga 1.000 yang dianggap sangat memuaskan.

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This thesis was submitted to the Senate of the Universiti Putra Malaysia and has been accepted as fulfillment of the requirement for the degree of Doctor of Philosophy. The members of the Supervisory Committee were as follows:

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LIST OF ABBREVIATIONS

TeO_2	Tellurium oxide
B_2O_3	Boron oxide
ZnO	Zinc oxide
Bi_2O_3	Bismuth oxide
XRD	X-ray diffraction
FTIR	Fourier transform infrared
NBO	Non-bridging oxygen
BO	Bridging oxygen
TM	Transition metal
UV-Vis	Ultra-violet Visible
E_{opt}	Optical band gap
V_m	Molar volume
ρ	Density
V_L	Longitudinal velocity
V_S	Shear velocity
L	Longitudinal modulus
G	Shear modulus
K	Bulk modulus
E	Youngs modulus
H	Microhardness
σ	Poisson's ratio
d	Fractal bond connectivity
ANN	Artificial neural networks

CHAPTER 1

INTRODUCTION

1.1 Background of study

The rapid development in the field of the glass industry nowadays requires a new technology that can predict some important properties in a glass system before melting the composition of the glass materials. An artificial neural network (ANN) also simply called a neural network (NN) is a computing system inspired by the biological neural network of human brains (Yao et al., 2019). ANNs are known as one of the mathematical tools for modeling that can be used to predict linear, non-linear and complex problems in material properties. The application of ANNs greatly facilitated experimental works in designing the research materials (Zhang and Friedrich, 2003; Junior et al., 2005; Zhou et al., 2009). This model uses a list of neurons, which interconnected each other, carrying information that functions like the human brain. The neural networks have the ability to solve very complex problems after being trained by a database and during this training process, the network creates a relationship between the input data and the output data. Therefore, the technology provided by ANNs is believed to help further increase the production of the glass industry. In looking at the effectiveness of new technology brought by ANNs, four types of glass series namely zinc-tellurite (ZnO-TeO_2), bismuth-tellurite (Bi_2O_3-TeO_2), zinc-borate (ZnO-B_2O_3) and bismuth-borate (Bi_2O_3-B_2O_3) glass systems have been tested using the ANNs systems.

Borate and tellurite base glass networks are believed to have very attractive properties which lead to the considerations in both academic and technological fields. Glass based materials are being studied intensively nowadays due to the properties of easy to fabricate, no grain boundary, affordable, excellent in corrosion resistance and has a strong enough hardness (Joseph et al., 2002; Pal et al., 2011; Bootjomchai, 2015). In fact, both borate and tellurite glasses for instance has very unique characteristics such as low melting point, high refractive index, high dielectric constant, high mechanical strength, good chemical durability and good transmission in visible and infrared wavelength (Rajendran et al., 2003; Babu et al., 2009; Honma et al., 2010) which makes these base glasses suitable for laser host materials, optical fiber, optical switching devices and Raman amplifier (Nasu et al., 1990; Tanabe et al., 1990). In addition, borate base glasses also can be used in the non-linear devices for frequency conversion in the ultraviolet region, opto-acoustical electronics and piezoelectric actuator (Saddeek et al., 2007; El-Falaky et al., 2012).

The borate and tellurite glasses when combined with other modifier oxides such as zinc oxide (ZnO) and bismuth oxide (Bi_2O_3) will be easier to produce the glassy phase with various compositions. This incorporation will further enhance the physical, structural, elastic and optical properties which are greatly being

advantageous in glass technology applications. The materials of ZnO and Bi_2O_3 are relatively stable, affordable, non-toxic and environmentally friendly (Du and Ilegbusi, 2004; Klink and Crouch, 2009; Sumalatha et al., 2013). Moreover, Bi_2O_3 can be classified as one of the heaviest elements, a non-radiative nucleus and have good properties such as high density, high refractive index and high infrared transparency (Stone et al., 2000; Pan and Ghosh, 2000). However, Bi_2O_3 is not counted as a network former due to bismuth ions (Bi^{3+}) has a small field strength. In contrast, zinc ions (Zn^{2+}) can enter the base glass structure as a network modifier or form as ZnO_4 tetrahedral unit (link with oxygen ions) when acts as a network former (Eraiah, 2006; Sindhu et al., 2007; Chagraoui et al., 2009; Rao et al., 2011; Stefan et al., 2012).

Developments in the glass industry are usually made from empirical selections by an educated guess and guided using a look and try approach. This approach is quite expensive and time-consuming because if an experimental work does not meet certain criteria, then it will be repeated many times using other compositions which makes this approach less ideal to support the rapidly evolving demands of new glass technology. This fact by then served as a key encouragement for the increment in the ability to predict the main glass properties and others materials with reasonable accuracy. Therefore, it is time to move from the traditional "try and error" or "luckiness" into the new era in predicting some of the main glass properties that can lead to the production of the most important and useful glass combinations. There are several tools that might be used based on the scientific theoretical concept for the empirical pure components such as the topological constraint theory, density functional theory, molecular dynamics simulation, big data analysis and evolutionary algorithm (Cassar et al., 2018; Mauro, 2018). However, these theoretical models are very limited to simple materials that may only involve less than five components. Therefore, the demand for very high technology such as ANNs is very flexible to be used not just only for simple materials, but can also be applied to the materials that have a very complex composition. ANNs are believed to be able to help many industries in this world not only in the fields of engineering, medicine and construction but also in the field of material science too.

In this study, the four different types of binary glasses have been prepared using conventional melting and quenching methods and characterized on the physical, structural, elastic and optical behaviors in order to study comprehensively the effect of ZnO and Bi_2O_3 substitution in both tellurite and borate host glasses. The structural and optical properties of tellurite and borate glasses added with ZnO and Bi_2O_3 have been discovered by using X-ray diffraction (XRD), Fourier transforms infrared (FTIR), Raman and UV-Visible spectroscopy. Meanwhile, the elastic properties of the glass systems have been revealed through Ultrasonic Data Acquisition System and also calculated using bond compression and Makishima-Mackenzie theories. The quantitative parameters that have been measured experimentally such as density, molar volume, ultrasonic velocity, elastic moduli and optical band gap will then be tested and compared with the theoretical values from ANNs systems.

1.2 Problem statement

Compared with other conventional glass systems, tellurite and borate host glasses have attracted the attention of many researchers because of their superior glass-forming nature, high thermal stability and low melting temperature (Zamyatina et al., 2020; Abouhaswa et al., 2021). Tellurite and borate glasses belong to unique and special inorganic vitreous systems that exhibit different coordinated tellurite and borate groups which are TeO_3, TeO_4, BO_3 and BO_4 having completely different positions or sites in IR vibration bands. Development of tellurite and borate glass research added with alkali or alkali earth oxides especially ZnO and Bi_2O_3 possess great chemical stability in addition to having interesting physical and optical properties which make these types of glasses can be applied in many specific optical purposes (Ouis and Marzouk, 2020; Zamyatina et al., 2020). Normally, the conventional melting and quenching method to produce commercial inorganic phosphor and laser materials are completely established in the glass industry. This technique is referring to the pure starting materials such as TeO_2, B_2O_3, ZnO and Bi_2O_3 are mixed and melted until the correct and suitable glass composition is found for certain applications. This method will increase the production and energy cost as well as take a long time to achieve a target. To overcome this problem, ANN systems can be introduced and developed in the glass industry to predict some important parameters such as density, molar volume, ultrasonic velocity, elastic moduli and optical band gap values of the glass networks in order to minimize the cost and time-consuming.

In recent years, interesting developments have taken place in the field of materials science when artificial intelligence (AI) application was introduced as one of the novel techniques to improve certain properties (Merayo et al., 2019). ANNs are one of the most important tools in the AI evolutions in which this system can perform tasks by learning from the dataset given as an input without any specific directions like the human brain. ANNs have revealed their capability in materials science for predicting glass transition temperature, density and mechanical properties on some of the inorganic glass compositions, tellurite glass system, polymer and glass-forming alloy (Cassar et al., 2018; Ahmmad et al., 2021). Several tools might be used based on the scientific theoretical concept but these theoretical models are very limited to simple materials which may only involve less than five components. Therefore, ANN systems are very flexible to be used not just only for simple materials, but can also be applied to materials that have a very complex composition. Most of the theoretical models do not show a very high accuracy value. So that, the effectiveness of ANN systems has been approved by the goodness of fit R^2 value to indicate the accuracy of the predicted data. Thus, the aims of this research are to study comprehensively the optimum preparation or fabrication of some tellurite and borate glass structures added with ZnO and Bi_2O_3 as well as the implementation of ANNs in predicting some important parameters of glass compositions. This research also quantitatively compared the data obtained from the theoretical calculations and prediction with the experimental measurement.

1.3 Scope of study

This study will emphasize the fabrication of four different types of glasses which are zinc-tellurite (ZnO-TeO_2), zinc-borate (ZnO-B_2O_3), bismuth-tellurite (Bi_2O_3-TeO_2) and bismuth-borate (Bi_2O_3-B_2O_3) glass systems using conventional melting and quenching methods. The structure of each glass sample will be tested using the X-ray diffraction technique to confirm the amorphous phase of the glasses. The detailed structure and internetwork bonding of the glass sample will perform using Fourier transform infrared spectroscopy (FTIR) and Raman spectroscopy devices. The elastic behavior of the glass sample has been measured using ultrasonic measurement and this experimental value will be supported through the calculation from bond compression and Makishima-Mackenzie theoretical model. The optical properties of the glass samples will be measured by using ultraviolet-visible spectroscopy. The quantitative of the experimental measurement such as density, molar volume, ultrasonic velocity, elastic moduli and optical band gap then will be tested and compared with the artificial neural network systems.

1.4 Objectives of study

The main objective of this study is to utilize an artificial intelligence system through an artificial neural network which used to predict some important physical parameters of glasses that may be applied before melting any glass composition. The objectives throughout this study can be listed as follows:

1. To fabricate the optimum binary tellurite and borate glasses added with ZnO and Bi_2O_3 using the melting and quenching technique.
2. To simulate some physical, elastic and optical properties of binary tellurite and borate glasses added with ZnO and Bi_2O_3 using artificial neural networks models.
3. To evaluate the elastic properties of binary tellurite and borate glasses added with ZnO and Bi_2O_3 using bond compression and Makishima-Mackenzie model.
4. To compare and quantitatively analyse the experimental data from physical, elastic and optical behavior with the prediction of artificial neural networks models.

1.5 Hypothesis of study

By referring to the objective of this study**,** the hypothesis can be listed as follows:

1. The tellurite and borate glass composition added with ZnO and Bi_2O_3 are believed to be successfully prepared using conventional melting and quenching techniques where the glass bulk produced free from air bubbles, non-hygroscopic and stable.

2. The incorporation of ZnO and Bi_2O_3 into the tellurite and borate glass systems may alter some of the basic characterizations of tellurite and borate-based glass networks such as the increase in density value and the changes in the elastic behavior which leads to an increase in the rigidity of the glass system.
3. Simulation from artificial neural networks provides the predictive value of the physical, elastic and optical properties of tellurite and borate glass composition when substituted with ZnO and Bi_2O_3. The predictive value of artificial neural networks is expected to show similar behavior with the obtained value from the experimental works.
4. The calculation of bond compression and Makishima-Mackenzie model is believed to show an equivalent trend and support the value observed by the experimental measurement.
5. The substitution of ZnO and Bi_2O_3 into the tellurite and borate glass systems is expected to be able to improve the optical absorption and contribute to the reduction in the optical band gap values.
6. The analytical comparison between theoretical calculation and prediction with experimental measurement are estimated to show a good relationship so that the theoretical model can be applied in the future in the glass field.

1.6 Thesis outline

This thesis contains six chapters. Chapter 1 briefly describes the general introduction of artificial neural network model, the tellurite and borate glass systems and the objective of the study. Chapter 2 recounts the previous studies on the borate and tellurite glass synthesis, the theory of glass preparation and its application in the glass field. The theory of artificial neural networks, bond compression and Makishima-Mackenzie model has been explained in Chapter 3. Chapter 4 focuses on the borate and tellurite glass preparation and characterization. The results concerning the effect of ZnO and Bi_2O_3 added in both tellurite and borate glasses have been discussed in Chapter 5. This chapter also elaborates on the values predicted by artificial neural networks and the comparisons with the experiment measurements. Finally, Chapter 6 concludes the research findings and suggestions for further research.

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