

VARIATION IN THERMAL STRESS BEHAVIOUR OF COMPOSITE MATERIALS BY THERMOMECHANICAL TECHNIQUE (TMA)

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Received: 3 Oct., 2023 / Accepted: 12 Nov., 2023 / Published: 25 Jan., 2024.

<https://doi.org/10.25271/sjuoz.2024.12.1.1210>

ABSTRACT:

In this study, a polymeric-based composite material was made using 80% epoxy polyester 20% and unsaturated polyester resin. The polymeric composites were prepared by manual moulding method by adding a support material of silica powder to the mixture by weight (0%, 5%, 10%, and 15%) to examine one of the thermal properties which are thermal expansion. A comparison was made with the polymeric mixture before and after the reinforcement. The results of the study showed that the coefficient of thermal expansion increases with increasing temperatures, while it decreases at high temperatures. The results also showed that there is a decrease in the degree of glass transition with an increase in the concentration of the additive, and it was also noted that an increase in the percentage of silica at 15% decreases the value of the thermal expansion coefficient compared to other samples.

KEYWORDS: Epoxy resin ; Polymeric ; Silica powder ; Thermomechanical technique ; Thermal analysis ; Thermal expansion

1. INTRODUCTION

Polymeric blends are made by the physical mixing of two or more different polymers to produce a mixture with desirable mechanical properties. Polymeric blends have witnessed great interest and rapid growth compared to other polymeric materials systems because of their high performance in most applications and their low cost [1-3]. Thermal tests are one of the most important tests for researchers in polymer science because of their importance for testing the quality of polymers when used industrially, and thermal analysis methods are one of the methods used for examination. Thermal analysis devices were developed and produced in a variety of ways for D4 Pont-Elmer Perkin's thermal analysis [4, 5]. Thermal analyses are defined as a set of analyses through which physical and chemical changes are measured continuously as a function of temperature when the material is exposed in a controlled manner to heat differential thermodynamics and thermomechanical analysis [6, 7].

The method of thermomechanical analysis is one of the methods concerned with studying the change of thermal properties as a result of a change in temperature. The thermomechanical analysis method is used to measure the mechanical response of polymers when changing temperature and under specific conditions [8]. Through this method, some mechanical properties of polymeric compounds can be measured. The coefficient of thermal expansion of the polymeric material is measured, and the increase in volume is measured when the model is exposed to different temperatures. It is also possible through this method to determine the degree of glass transition of the material and the degree of softness of it, as well as the melting point [9]. In 2011, the researcher (Nabil) and his group studied the effect of polyester moulding and plasticization on the thermal expansion behaviour using the

thermomechanical decomposition TMA technique in order to study the possibility of using thermal variables from the longitudinal expansion coefficient and the change in length through the processes of return and recovery of the polymer to obtain a mixture or material. Overlapping is ideal for engineering applications [10]. In the year 2021, the two researchers (Aqeel and Mohammed) studied the improvement of the thermal specifications of the epoxy-talc power composite using the TMA technique, where the results showed that increasing the percentage of talc improved the thermal properties represented by the degree of glass transition, and the values of the thermal expansion coefficient showed a slight decline with an increase in temperatures [11].

The research aims to investigate the variation in the thermal expansion behaviour using thermomechanical analysis techniques and to improve the thermal properties of the composite material and the prepared samples used in this research in order to use them in suitable industrial applications.

2. METHODS

The coefficient of thermal longitudinal expansion is defined as the change in the length of the model in terms of temperature [12]. The thermal expansion coefficient is related to the change in the dimensions of the material when the temperature changes, so the longitudinal expansion coefficient is calculated according to the following relationship [13]:

$$\alpha = \frac{1}{L} \frac{\Delta L}{\Delta T} \quad (1)$$

where α is the longitudinal expansion coefficient, L is the original length, L is the new length, and T is the temperature. One of the important characteristics that determine the homogeneity of polymeric mixtures is the degree of glass transition, which is an average of a range of temperatures at which the polymer transforms from a strong, hard, glassy

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substance into a flexible one [14]. At a temperature less than T_g , the polymer is a brittle solid, as the particles have a small ability to move, and plastic deformation can occur when the temperature is higher than T_g [13, 15].

2.1 Materials Used

The materials used in the research include two main parts: the matrix and the reinforcing material. The polymeric substrate is divided into two types: epoxy resin and unsaturated polyester resin (UPE). As for the precipitant, silica was used as a support material in the preparation of the polymeric composite material.

2.2 Matrix Material

In this study, two types of resins were used as a base material, namely: Epoxy resin, which is a thermosetting polymer and is in the form of a viscous liquid with certain specifications, including high adhesion ability and low shrinkage upon drying. In this study, epoxy resin (Quickmast 105) was used, manufactured by Ayla Construction Chemicals and licensed by DCP English, with a density of $1.04g / cm^3$, and the epoxy resin is converted to a solid state after adding a hardener of the type Metaphenylene Diamine (MPDA), which is a liquid substance with a transparent color, in a ratio of (1:3) so that a reaction occurs between them at room temperature. It begins with a gelatinous material, and after a certain period of time, we find that the material hardens at room temperature. After that, a curing process is performed for a number of hours in order to reduce the percentage of contractions and increase the bonding between molecules. The weakness of this polymer is its high brittleness (Britten's). But the advantage of epoxy is that it has intertwined chains.

Unsaturated polyester resin, which is manufactured by SIR Saudi Arabia, is a thermosetting polymer that is in the form of a transparent thermosetting liquid (with a density of $1.2 g/cm^3$), and this resin can be cured to a solid state after adding to it a hardener of the type ethyl methyl ketone peroxide (MEKP) as a hardener of 2g per 100g of resin to turn into a solid at room temperature.

2.3 Reinforcing Material

Silica material was used as a reinforcement material with a density of $2.32 g/cm^3$ and a grain size of $35\mu m$ and it was obtained through the use of special sieves (shaker sieves) in order to separate the required size in this work. Table 1 shows the internal structure of silica by using (XRF) x-ray fluorescence system.

Table 1: shows the chemical analysis of the elements that are included in the composition of silica.

Compounds%	Chemicals Elements
96.5-98 %	SiO ₂
0.5-1.0 %	Al ₂ O ₃
0.07-0.5 %	Fe ₂ O ₃
0.1-0.4 %	CaO
0.1-0.15 %	MgO
>0.1 %	Na ₂ O

2.4 Preparation Technique

To prepare the mixture sample (80% EP + 20% UPE); a weight ratio was taken for each resin according to the mixing

amounts with the hardener using a sensitive scale. For epoxy, the mixing ratio is 1:3. As for unsaturated polyester, the mixing ratio is 2 per 100 g. Mix the mixture well with an electric mixer for 10 minutes. The liquid was poured in the form of a torrent in the middle of the mould so that it flows to all areas of the mould in a continuous and regular manner until the mould is filled, and then we leave the casting for 24 hours at room temperature. We put the samples in a drying oven at a temperature of 50 °C for 5 hours to obtain the best crosslinking and reduce the internal stresses formed during shrinkage. The special samples were cut to perform the thermomechanical analysis examination, with a length of 20 mm and a diameter of 5 mm.

Table 2: shows the mixture with the percentage of concentration used in the research.

Composite Material	Type of Material Blend 80%EP/20% UPE
Blend	80%EP/20% UPE
Blend+ silica particles	Matrix Blend Reinforced with 5% by silica 35 μm
	Matrix Blend Reinforced with 10% by silica 35 μm
	Matrix Blend Reinforced with 15% by silica 35 μm



Figure 1: Photograph of the samples.

2.5 Device System (TMA)

The system was used in conducting the thermomechanical analysis test in the Department of Physics, Anbar University. The system consists of a model room, a model stand, sensors, an oven, a controller, a data output unit, a calculator, and a printer. Figure 2 shows the principle of operation of a thermomechanical analysis system (TMA) is that when the sample is placed in the place designated for it in the heart of the convection oven, which contains thermal sensors, the system

begins to work automatically by recording the readings according to the temperature range that was determined and prepared previously, where the sample begins to expand or shrink according to the nature of the settings placed, and the thermal sensors begin to record the change that occurs to the model along with it. The sample is very accurate and sends electronic signals to the computer system, which in turn converts these signals into understandable data and forms by drawing a shape for the changes that occurred in the sample in terms of the change in length, temperature, and time.



Figure 2: The thermomechanical analysis (TMA) system.

3. RESULTS AND DISCUSSION

The study measured the change in sample length from a polymeric mixture containing silica, focusing on the thermal range (25-200 °C). Results showed a rise in thermal energy of particles, causing plastic changes and an increase in sample basic length. The results of the TMA analysis gave values for the degree of glass transition T_g , where the mixture (80% EP/20% UPE) was 99.2 °C. We note from Figure 3 that the value of T_g for the blends Blend +5% silica is 95.7 °C, Blend +10% silica is 53.9 °C, Blend +15% silica is 42.5 °C, and this explains that the addition of silica powder to the mixture works to complicate the bonding between the polymeric chains. Thus, it hinders the polymeric chains and restricts their movement, which makes the polymeric chains need a higher amount of heat to reach the appropriate degree of freedom of movement and to move the molecular aggregates [16].

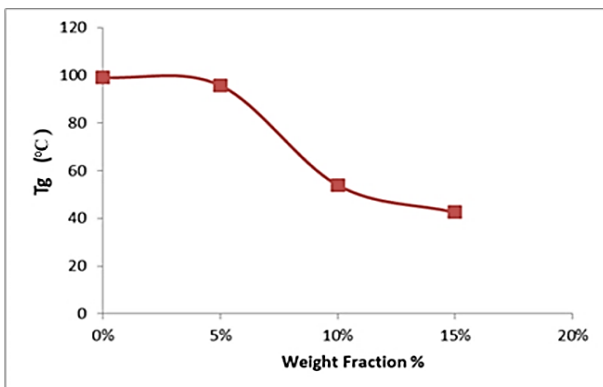


Figure 3: The glass transition temperature T_g , as a function of the weight ratios of silica powder.

The presence of the support material inside the mixture works to weaken the forces between the molecules, which reduces the energy required to break the bonds. As the increase in temperature leads to an increase in the movement of the polymeric chains and the relaxation of the bonds between them, the transition to the plastic state takes place [17]. Thus, T_g decreases with increasing the weight fraction of silica powder as the material changes from less elastic to more elastic [18]. In Figure 4, the effect of the thermal expansion coefficient on the

temperature of the prepared superimposed material is shown, and the rise in the temperature of the superimposed material above the limits of 100 °C affects the value of the thermal expansion coefficient. These results are similar to those of previous research and studies [21, 22].

The expansion coefficient increases with the temperature of all samples. The increase is up to 100° C then it gradually decreases, where the material shows low longitudinal expansion behaviour. This is due to the change in the physical structure of the material that causes the material to collapse and the material becoming unresponsive to expansion at high temperatures [19, 20]. It is proven that the 5% mixture has appropriate values for the coefficient of thermal expansion with temperature.

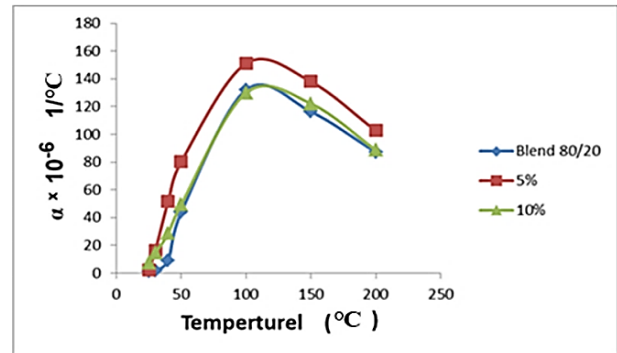


Figure 4: thermal expansion coefficients (α) as a function of temperature.

As Figure 5 shows, the decrease in the values of the thermal expansion coefficient of the mixture with the increase in the weight ratios of silica powder is due to the thermal stability of the basic nature of the materials used for the mixture, which are well characterized by thermal stability [17, 19].

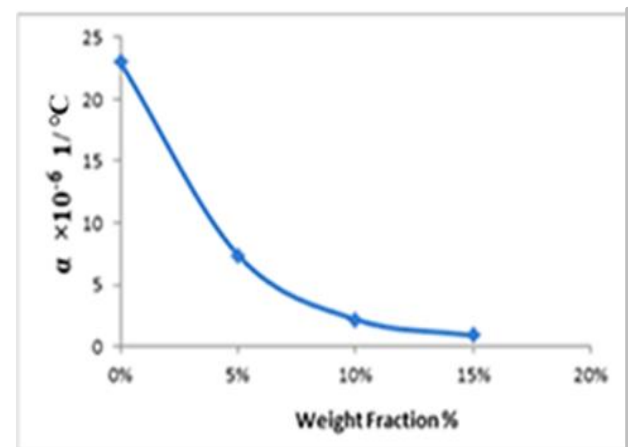


Figure 5: coefficient of thermal expansion (α) as a function of the weight ratios of silica powder at a temperature of 25 °C.

CONCLUSION

During this research, the most important conclusions were reached, and they are summarized as follows:

1. The decrease in T_g values occurred when the proportion of silica powder in the mixture was increased. An increase in the concentration of the material added to the mixture led to a decrease in the thermal expansion coefficient, which contributed to improving the material's resistance to thermal expansion.
2. Variation in the values of thermal expansion with the rise in temperature, where the values of the expansion coefficient change after the temperature of 100 °C and the material

becomes unresponsive to the increase in temperature, which indicates that the material will be damaged at high temperatures. Through this method, materials with good thermal properties were obtained at a low cost.

REFERENCES

- Bhatnagar, M. S. (2004) *A Textbook of Polymer Chemistry*. S. Chand Publishing, (2004).
- Utracki, Leszek A., and B. D. Favis. (1989) Polymer alloys and blends. *Handbook of polymer science and technology* 4, 121-185.
- Karvanis, K.; Rusnáková, S.; Krejčí, O.; Žaludek, M. (2020) Preparation, Thermal Analysis, and Mechanical Properties of Basalt Fiber/Epoxy Composites. *Polymers*, 12, 1785.
- Yoon, P. J., T. D. Fornes, and D. R. Paul. (2002) Thermal expansion behavior of nylon nanocomposites. *Polymer* 43, 6727-6741.
- Jose James, (2017) Chapter 7-Thermomechanical Analysis and Its Applications, Editor(s): Sabu Thomas, Raju Thomas, Ajesh K. Zachariah, Raghendra Kumar Mishra, In *Micro and Nano Technologies, Thermal and Rheological Measurement Techniques for Nanomaterials Characterization*, Elsevier, Pages 159-171, ISBN 9780323461399.
- Preghenella, Michele, Alessandro Pegoretti, and Claudio Migliaresi. (2006) Atomic force acoustic microscopy analysis of epoxy-silica nanocomposites. *Polymer testing* 25, 443-45.
- Study the contrast of thermal expansion behavior for PMMA denture base, single and hybrid reinforced using the thermomechanical analysis technique (TMA). *AIP Conf. Proc.* 2769, 020027 (2023).
- Yang, L., Thomason, J.L. (2013). The thermal behaviour of glass fiber investigated by thermomechanical analysis. *J Mater Sci* 48, 5768–5775.
- Saba, N., and M. Jawaid. (2018) A review on thermomechanical properties of polymers and fibers reinforced polymer composites. *Journal of industrial and engineering chemistry* 67, 1-11.
- Rammo, Nabil N., Khalid H. Harbee, and Hind Abdulmajeed Mahdi. (2011) TMA and DSC study of molded and annealed polyester: a thermal expansion and crystallization evaluation. *Al-Mustansiriyah Journal of Science* 22, 7.
- Ali, Aqeel Mohsin, Mohammed A. Jaber, and Nooraldeen A. Toama. (2021) Thermal Properties of Polyester/Epoxy Blend. *Iraqi Journal of Science* 62, 1128-1134.
- Al Arbash, A., Z. Ahmad, F. Al-Sagheer, and A. A. M. Ali. (2006) Microstructure and thermomechanical properties of polyimide silic nanocomposites. *Journal of Nanomaterials* 1-9.
- Tognana, S., W. Salgueiro, A. Somoza, J. A. Pomarico, and H. F. Ranea-Sandoval. (2009) Influence of the filler content on the thermal expansion behavior of an epoxy matrix particulate composite. *Materials Science and Engineering: B* 157, 26-31.
- Biron, Michel. (2003) *Thermosets and composites*. Elsevier.
- [Mohammed, Awatif Adab, Sinaa Ibrahim Hussein, and Inaam Hussein Ali. (2007) Study of temperature effect on the Hardness for epoxy resin and unsaturated polyester. *Baghdad Science Journal* 4, (2007).
- [Wunderlich, Bernhard. (2005) *Basics of thermal analysis. Thermal Analysis of Polymeric Materials* 71-188.
- Al-Mosawi, Ali I. (2009) Study of some mechanical properties for polymeric composite material reinforced by fibers. *Al-Qadisiyah Journal for Engineering Sciences* 2, 1.
- Michels, Julien, Robert Widmann, Christoph Czaderski, Reza Allahvirdizadeh, and Masoud Motavalli. (2015) Glass transition evaluation of commercially available epoxy resins used for civil engineering applications." *Composites Part B: Engineering* 77 484-493.
- Pugazhenth, N., and P. Anand. (2021) Mechanical and thermal behavior of hybrid composite medium density fiberboard reinforced with phenol formaldehyde. *Heliyon* 7, 12.
- Junid, Ramli, Januar Parlaungan Siregar, Nor Azam Endot, Jeefferie Abd Razak, and Arthur N. Wilkinson. (2021) Optimization of Glass Transition Temperature and Pot Life of Epoxy Blends Using Response Surface Methodology (RSM). *Polymers* 13, no. 19-3304.
-] Liu, Tuan, Cheng Hao, Shuai Zhang, Xiaoning Yang, Liwei Wang, Jiarui Han, Yuzhan Li, Junna Xin, and Jinwen Zhang. (2018) A self-healable high glass transition temperature bioepoxy material based on vitrimer chemistry. *Macromolecules* 51, no. 15-5577-5585.
- [Startsev, O. V., Yu M. Vapirov, M. P. Lebedev, and A. K. Kychkin. (2020) Comparison of glass-transition temperatures for epoxy polymers obtained by methods of thermal analysis. *Mechanics of Composite Materials* 56, no. 2 :227-240.