DOI: 10.5604/01.3001.0053.7661
Volume 118 • Issue 2 • June 2023
Journal of Achievements in Materials and Manufacturing Engineering
International Scientific Journal published monthly by the World Academy of Materials and Manufacturing Engineering

Studying the influence of waste glass and montmorillonite powders on the thermal conductivity and hardness of poly(methyl methacrylate) polymer matrix

A. Almansoori [a,*], R.A. Ghadban [a], M.H. Ali [a], M.M. Sabri [b]
[a] Technical Institute of Basra, Southern Technical University, Basra, Iraq
[b] Department of Physics, Faculty of Science and Health, Koya University, Koya KOY45, Kurdistan Region, Iraq
* Corresponding e-mail address: a.almansoori@stu.edu.iq
ORCID identifier: https://orcid.org/0000-0002-9757-7724 (A.A.)

ABSTRACT

Purpose: The present study aims to evaluate the effect of montmorillonite nanoclay (MMT) and waste glass powder (WGP) on the hardness and thermal conductivity of PMMA polymer composites. Thus, this study concentrates on the potential use of MMT and WGP as reinforcements, in different concentrations, in PMMA polymer matrix, with the expectation of improving the performance of PMMA polymer composites in various applications.

Design/methodology/approach: There is a growing demand for PMMA with increased mechanical properties and thermal stability for applications where inorganic glass would fail. Montmorillonite (MMT) clay and Waste Glass Powder (WGP) have physical and chemical properties compatible with PMMA. Therefore, they could potentially enhance PMMA's hardness and thermal conductivity. Silicon dioxide in glass silica and MMT and octahedral aluminium hydroxide sheet in MMT can strengthen both covalent and hydrogen bonding architecture in PMMA composite for better mechanical strength and thermal conductivity. Thus, PMMA composites were designed by combining MMT powder and WGP powder in different ratios before being incorporated into the PMMA polymer matrix and tested for hardness and thermal conductivity.

Findings: The present study measured Brinell Hardness (HB) and electrical conductivity values of four PMMA composites containing different proportions of MMT and WGP. MMT/WGP filler mix had optimal hardness (HB number = 74) when glass content was 1% (3MMT1G) or better still (HB number = 63) when an equal mix ratio was used (1MMT1G). PMMA composite with 3MMT1G also had the highest thermal conductivity (0.01899W/m.K^{-1}). However, the higher the glass content, the lower the thermal conductivity of the PMMA composite. Thus, the present study has demonstrated that 3MMT1G filler was the best for enhancing the thermal and mechanical properties of PMMA composite.

Research limitations/implications: The results of this study demonstrate the potential of this new composite material for a variety of applications. Further research is needed to explore the full potential of this material and to develop new and improved versions.

Practical implications: Reusing waste glass as filler materials in composites requires minimal processing and therefore has lower environmental impacts than synthetic options.

Originality/value: Experimental data from the present study has provided new insights into Glass/MMT mix design in PMMA composites. The PMMA composite containing 3MMT1G exhibited the best hardness and thermal conductivity characteristics. Thus, the present study has successfully optimised Glass/MMT mix design for PMMA composite for applications requiring these features.

Keywords: PMMA composites, Hardness, Thermal conductivity, Waste glass powder, MMT clay

Reference to this paper should be given in the following way:

A. Almansoori, R.A. Ghadban, M.H. Ali, M.M. Sabri, Studying the influence of waste glass and montmorillonite powders on the thermal conductivity and hardness of poly(methyl methacrylate) polymer matrix, Journal of Achievements in Materials and Manufacturing Engineering 118/2 (2023) 49-56. DOI: https://doi.org/10.5604/01.3001.0053.7661

PROPERTIES

1. Introduction

Polymethyl methacrylate (PMMA), popularly known as acrylic resin, is a synthetic methacrylic resin produced by the radical polymerisation of methyl methacrylate (MMA) monomer [1,2]. Physically, it is a transparent, rigid, lightweight thermoplastic exhibiting impact-, weather-, and chemical-resistant properties [3,4]. PMMA's mechanical and optical properties and its colour versatility make it a suitable substitute for inorganic glass in products such as shatterproof windows and displays, illuminated signages, skylight/sunroofs, motorcycle and automobiles windshield, and aircraft canopies [1,5]. It is also suitable for biomedical and medical applications, including bone cement/fillers, contact or intraocular lens, denture bases, lenses and spectacle glasses, PMMA-based microcapsules (for cosmetic, drug delivery, and self-healing) due to its biocompatibility (non-toxicity and inertness) [6,7].

The heavy application of PMMA in the automobile industry, the aviation field, and roofing systems have necessitated further research on further strengthening it for applications requiring even higher tensile strengths without significantly altering its thermal conductivity [8]. Material engineers have increasingly focused on finding compatible additives and admixtures to increase PMMA's hardness (modulus and tensile strength and hardness) and thermal stability (enhanced thermal conductivity). Montmorillonite (MMT) clay and inorganic glass waste have been proposed as candidate admixtures that can improve the properties of PMMA. Nanoclays, including MMT, have been immensely researched for their potentially excellent reinforcement characteristics [9-11]. MMT is a natural clay mineral with a nanolayered crystal structure. As shown in Figure 1, MMT structure comprises a single sheet of octahedral alumina sandwiched between two tetrahedral silica sheets [12-14].

The excellent reinforcement characteristics of MMT were demonstrated in an original study by Kojima et al. in 1993 [15]. Nylon 6 Clay-Hybrid (NCH) containing molecular composites of nylon 6 and MMT silicate layers exhibited superior modulus strength compared to plain nylon 6. MMT (4.7 wt. %) enhanced heat distortion temperature (HDT) in NCH to 152°C, which was about 1.7-fold higher than HDT in plain nylon 6 (87°C) [15]. MMT (2-8 wt. %) enhanced mechanical properties of MMT/vinylester and MMT/vinylester/glass composites, including tensile, interlaminar shear, flexural, and impact strength and durability (fatigue life) [16]. Several studies demonstrated that MMT nanoclay enhanced the mechanical properties, thermal stability, and durability of different types of polymers like, polyester [17], Nylon 6 [18], poly(methyl methacrylate-co-butyl acrylate) [19], and epoxy [20]. Enhanced mechanical strength and thermal properties have been reported in the hybrid composites of polypropylene/rice husk [21] and 3D carbon nanotube epoxy composites [22]. Thus, compelling experimental evidence suggests that MMT can also improve the mechanical and thermal properties of MMT/PMMA composite. Indeed, PMMA-MMT clay nanocomposites synthesised by ultrasonic mixing and magnetic stirring were, in a previous study, demonstrated to exhibit enhanced thermal stability [23]. However, the thermal stability increase in ultrasonically synthesized PMMA-MMT composite was about 30% more than in composite synthesised by magnetic stirring [23]. The observed thermal stability in MMT/PMMA composite is due to its negative thermal coefficient behaviour, which makes it useful for engineering applications [24], especially where inorganic glass would be unsuitable.

Waste glass powder (WGP) is a readily available material that can be incorporated into a polymer matrix. Inorganic glass is a mixture of mainly (75%) silica (SiO_2), sodium oxide as a fluxing agent and calcium carbonate. Glass is packed in an amorphous (noncrystalline) manner with strong covalent bonds linking the Si to the O atoms with central sodium atom [25]. This unique structure makes inorganic glass a suitable admixture material for PMMA.

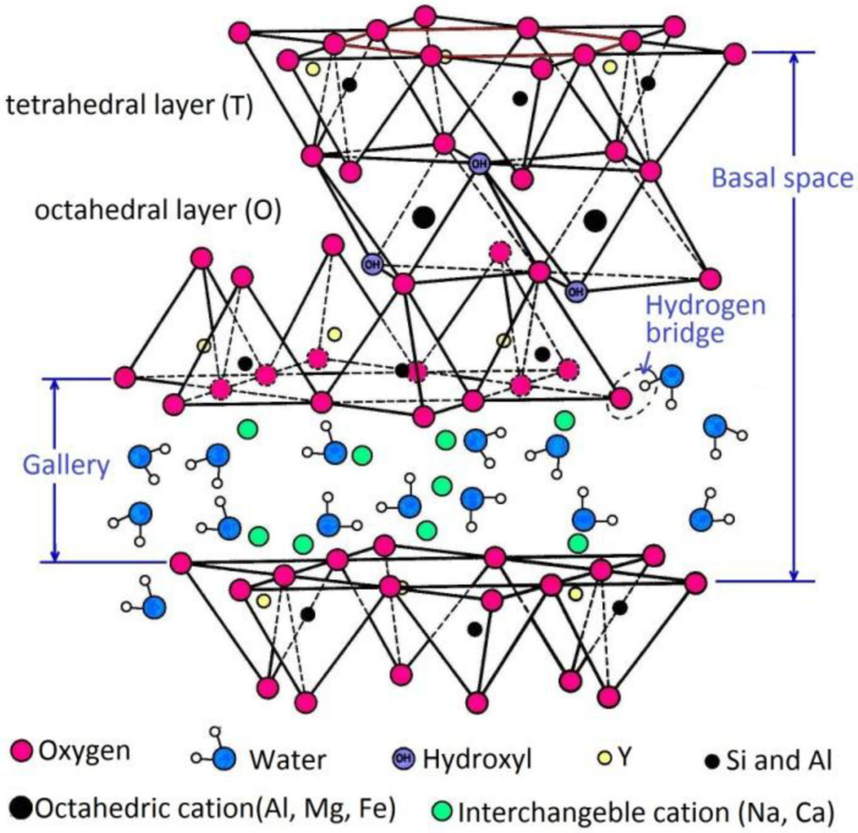

Fig. 1. Schematic representation of the Montmorillonite (MMT) clay structure. The side view shows 2:1 (tetrahedral: octahedral) layered alumino-silicate sheets assembled by weak van der Waals interactions and electrostatic forces, while the top view of MMT showing a hexagonal structure formed by oxygen and hydroxyl ligands from the sandwiched octahedral layer [26]

A previous study by Jang and Han in 1999 evaluated the effect of silica-based (~50–60% SiO_2) Glass-Fibre (GF) content (10 to 40% by volume) on the mechanical properties of GF/PMMA composite [27]. The study demonstrated that the flexural modulus strength of the GF/PMMA composites increased almost linearly with the GF content [27]. Another similar study shown also enhanced resistance to bending in GF/PMMA composite. The composite also exhibited increased resilience or resistance to impact and crack propagation (>30 kJ/m^2), which correlated with measured Vikers hardness of >12 kgf/mm^2 [16]. The addition of GF is also likely to increase the thermal conductivity of the GF/PMMA composite. A previous study by Elimat et al. (2008) demonstrated that adding 2wt% $CaCO_3$ in PMMA increased its thermal conductivity from 0.168 W/mK (for neat PMMA) to 0.175 W/m.K [28]. Adding glass containing $CaCO_3$ would transform the glass/PMMA composite to conduct heat better than neat PMMA, hence the desired thermal stability [29].

The obtained findings suggest that MMT/WGP filler mix is optimal when glass content is 1% (3MMT1G) or, better still, when an equal mix ratio is used (1MMT1G). The glass contains strong O-Si-O covalent bonds with a central sodium atom [25], while MMT clay contains layers of O-Al(Mg)-O and O-Si-O sheets [12,13]. It has been shown that PMMA and silica interact through hydrogen bonding [30]. It means that excess silica may weaken the hydrogen bonding and the entire bonding architecture in the PMMA composite. Indeed, MMT clay already contains silicon dioxide (in O-Si-O sheets) also in the glass. Thus, a higher glass content in an MMT/WGP filler mix results in excess silica content supplied by both MMT and WGP. Excess silica could potentially interfere with the strength of the overall bonding architecture in PMMA composite, hence, the observed decrease in hardness. A previous experimental study by Chladek et al. (2015) demonstrated that the hardness of PMMA/silica composite ball increased with silica-based nanofiller content by up to 10% [31]. Higher contents were not tested, suggesting that 10% of silica-based fillers was optimal for the PMMA/silica composite mix design. Interestingly, Boulerba and Zoukel (2021) demonstrated that the strength of hydrogen bonding between the PMMA and silica was influenced by the solvent type used, with chloroform showing optimal bonding [30].

Experimental evidence from the literature indicates that both MMT and WGP are potential good admixtures for enhancing the mechanical and thermal properties of PMMA. Therefore, the present study aims to evaluate the effect of nanoclay MMT and WGP on the hardness and thermal conductivity of PMMA polymer composite.

2. Experimental work

2.1. Materials and methods

This section provides a detailed explanation of the materials, methods and experimental techniques were used to obtain the data.

Polymers

Poly(methyl methacrylate) polymer (PMMA) purchased from the Petrochemical Industries Company in Basra.

Filling materials

Waste glass and nanoclay (montmorillonite) according to the ratios shown in Table 1. Waste glass powder was obtained by crushing ordinary home glass waste. Domestic waste transparent glass like jars and bottles collected from household waste was cleaned and washed first before they were dried in an oven at a temperature of 70°C for 6 hours to remove any biological contaminates, impurities and moisture remains. Then, the clean and dried glass was crushed and ground in a mixer grinder. The coarse powder was then ground into a fine powder using a ball mill, and a sieve screening was used to ensure uniform powder consistency and desired size.

Table 1.
Polymer and filling materials per cent loadings

Polymer	MMT ratio	Glass ratio	Symbol
PMMA	1%	1%	1MMT1G
PMMA	2%	3%	2MMT3G
PMMA	3%	2%	3MMT2G
PMMA	3%	1%	3MMT1G

Montmorillonite (MMT) nanoclay was purchased from Fluorochem Ltd (UK) with the chemical formula $[Al_{1.67} Mg_{0.33} (Na_{0.33})] Si_4O_{10} (OH)_2$. This nanoclay belongs to smectite group with tetra-octa-tetra hedral structure, as shown in Figure 1. However, this is a prestine nanoclay with no chemical surfactant that was used to avoid the potential chemical hazards of some organic surfactants.

Mixing method

The nanoclay (MMT) and glass powders (WGP) (with the ratios shown in Table 1) were mixed in a distilled water at room temperature and stirred in a magnetic stirrer for 2 hours and followed by a sonication for 30 minutes before being left the mixture to dry in a vacuum oven for 24 hours at a temperature of 70°C. These mixtures were stored in desiccators to ensure their physical and chemical stability by keeping them from moisture and other contaminants. It is important for the successful use of the mixture as fillers with the PMMA polymer matrix.

2.2. Fabrication and testing

Fabrication of composites

The resulting mixtures of the selected fillers were then added to the polymer matrix in the proportions mentioned in Table 1. The mix composites of the PMMA polymer matrix and mixtures of fillers (MMT and WGP) were mixed via high-speed mixture for 1 hour, followed by sonication for 30 minutes to improve the dispersion of fillers into the polymer matrix. The composites were placed in a steel mould with dimensions of (L=7.5 cm, W=7.5 cm, and t=3 mm). They were then heated for 30 minutes at 70°C (below its glass transition temperature) before pressure was applied using a hydraulic-driven hot press. Applying pressure helps create a strong bond between the polymer matrix and the fillers, resulting in a strong and durable composite material. After heating, the composite was cooled to room temperature and removed from the mould, and the resulting products were machined to the desired shape. The resulting composite was tested for its mechanical and physical properties, i.e., hardness and thermal conductivity.

Testing

The hardness test of the sample PMMA composites was performed using the Brinell hardness tester in the inspection laboratory in the State Company of Southern Fertilizers in Basra, according to DIN 50351:1985-02 (an indenter metallic ball with a diameter of 10 mm and a maximum load of 50 N were used). While the thermal conductivity was performed using Lee's Disk Apparatus in the College of Engineering, University of Basra, in accordance with the standards set by ASTM-E1225-04 [32]. The Lee's Disk method is a method used to measure the thermal conductivity of materials. It consists of two bronze plates connected to an electrical heater and a DC power supply. An infrared thermometer is then used to measure the temperature of the plates. The specimen was placed between the two plates, which were then tightened to hold the specimen between the plates. After the heater was switched on, the thermal conductivity K can be calculated by measuring the temperature difference between the two plates. The equation below calculates the material's thermal conductivity based on the temperature difference between the two plates [33]. This equation was successfully used to determine the thermal conductivity of low-density polyethylene/MMT composite [32].

$$K = K' \frac{d(T_2 - T_0)(T_1{}' - T_2{}')A'}{d'(T_2{}' - T_0{}')(T_1 - T_2)A} \quad (1)$$

where:
K = thermal conductivity of the studied specimens equals to W/m.K;
K' = thermal conductivity of the standard specimen equal to 0.23 W/m.K;
d = thickness of studied specimens equals to 3 mm;
d' = thickness of standard specimens equals to 6 mm;
T_o = ambient temperature equals to 34.5°C;
T'_o = ambient temperature of the standard specimens equals to 28°C;
T_1-T_2 = the temperature difference across the studied specimen;
T'_1-T'_2 = the temperature difference across the standard specimen = (100-60)°C, respectively;
A = the surface area of the studied specimen;
A' = the surface area of the standard specimen.

3. Results and discussion

Until this study, no study has evaluated the effect of different mix ratios of MMT to Waste Glass Power (WGP) for PMMA composites. The present study performed Brinell Hardness (HB) test on PMMA composites containing different proportions of MMT and WGP. Table 2 shows the hardness test results of four PMMA composite samples. The highest hardness number (74) was noted in the PMMA composite containing 3% of MMT and 1% of WGP (3MMT1G). The lowest hardness number (50) was noted in the PMMA composite containing 3 % MMT and 2% WGP (3MMT2G). This data suggests that when the proportion of the MMT is held constant, increasing glass content decreases the hardness of the PMMA composite. Conversely, a markedly higher hardness number (63) was noted when equal proportions of MMT and glass (1MMT1G) were used to produce PMMA composites. When a lower proportion of MMT than WGP was used (2MMT3G), a slight decrease in hardness number (54) was noted in the resulting PMMA composite. The hardness number decreased to (50) when a higher proportion of MMT than WGP was used (3MMT2G).

Table 2.
Brinell Hardness (HB) test results of PMMA composites containing different proportions of MMT and (WGP) powders

Sample	Glass/filler content, %	Hardness, HB
1MMT1G	50	63
2MMT3G	60	54
3MMT2G	40	50
3MMT1G	25	74

The current study's experimental data confirm that MMT and WGP have a strengthening effect on PMMA composite. However, there is an apparent difference in the effects these admixtures have on the PMMA composite. Table 3 shows the thermal conductivity results of PMMA composites containing different proportions of MMT and WGP. Thermal conductivity was optimal (0.01899 W/m.K) in PMMA composite containing 3% MMT and 1% WGP (3MMT1G). The lowest thermal conductivity was noted in the PMMA composites with 2MMT3G mix design (0.013106 W/m.K).

Table 3.
Thermal conductivity results of PMMA composites

Sample	Glass/filler content, %	Thermal conductivity (K), W/m.K
1MMT1G	50	0.01488
2MMT3G	60	0.013106
3MMT2G	40	0.016601
3MMT1G	25	0.01899

Experimental data in the present study demonstrated that thermal conductivity increased with decreasing glass/MMT ratio with optimal thermal conductivity at 25% glass/filler content ratio, the lowest ratio tested in the current study. Data from the literature show that thermal conductivity of neat PMMA is 0.168 W/m.K [28], which is markedly lower than 0.8-0.99 W/m.K for ordinary glass [34]. However, slightly higher values were recorded in the previous study when different method was used [35,36]. Thus, the thermal conductivity of PMMA needs to be enhanced to be superior to that of glass. On the other hand, MMT has a superior thermal conductivity of 1.232 W/m.K [37], and therefore, MMT is the best component to improve the thermal conductivity of PMMA composite. In the present study, glass/MMT resulted in PMMA composites with markedly decreased thermal conductivity from neat PMMA of 0.168 W/m.K (from the literature) to 0.013106-0.01899 W/m.K. While it was anticipated that glass and MMT (with higher thermal conductivity values) would enhance that of the PMMA composite, differing findings were observed in the present study. However, in general, a higher proportion of MMT resulted in a higher thermal conductivity of PMMA composite (3MMT1G). It suggests that glass reduces PMMA's thermal conductivity, and therefore, the thermal conductivity of PMMA composites correlated negatively with Glass/MMT ratio. Thus, the higher the glass content, the lower the thermal conductivity of the PMMA composite.

The present study has successfully optimized glass/MMT mix design for PMMA composite. The PMMA composite containing 3MMT1G exhibited the best hardness and thermal conductivity characteristics. Experimental data from the present study has provided new insights into glass/MMT mix design in PMMA composites. First, glass is a negatively limiting component since the higher the glass content in glass/MMT mix design, the lower the hardness and thermal conductivity of the PMMA composite. Conversely, MMT is a positively limiting component; therefore, increasing its proportion in glass/MMT increased the mechanical and thermal properties of the resulting PMMA composite. The observed behaviour could be attributed to relatively lower covalent bond strength in glass's silicon dioxide than MMT. However, further studies are warranted to measure covalent bond strengths in PMMA composites containing different glass/MMT mix designs. Thus, The combination of these properties makes PMMA composites ideal for a wide range of uses, enabling them to be suitable for use in a wide range of industrial and consumer applications such as window frames, automotive parts, sporting goods, and medical implants [3].

4. Conclusions

PMMA is an attractive substitute for glass in industrial applications requiring more mechanically resilient and thermostable material. From the reviewed literature on material properties, MMT clay and Waste Glass Powder (WGP) have excellent properties. Several experimental studies reviewed in the present study have demonstrated that MMT clay and inorganic glass, which act as a filler provide a reinforcing effect by enhancing the mechanical strength and thermal stability of various polymer composites. The obtained polymer composites, therefore, become stronger with better thermal properties. In addition, using waste inorganic glass and MMT clay with polymer composites can also reduce the production cost and improve the composite material's overall performance.

The present study demonstrated that MMT/WGP filler mix achieved optimal hardness (HB number = 74) in PMMA composite when glass content was 1% WGP (3MMT1G) or better still (HB number = 63) when an equal mix ratio was used (1MMT1G). PMMA composite containing a greater proportion of MMT (3MMT1G) also exhibited the highest thermal conductivity value (0.01899 W/m.K). Findings from the present study demonstrated that glass was a negatively limiting component. The higher the glass content, the lower the hardness and thermal conductivity of the PMMA composite. While MMT was a positively limiting component, such that the higher its proportion in MMT/WGP, the higher the mechanical and thermal properties of the resulting PMMA composite, indicating their potential for use in a wide range of applications.

In summary, the results of the current study demonstrate that PMMA composites combined with MMT and WGP in appropriate amounts exhibit improved thermal and mechanical properties, enabling them to be suitable for various applications. Although the combination of the fillers has improved the properties of PMMA composites, higher amounts of the MMT content results in a better performance of the composite.

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