Flammability, Thermal and Mechanical Properties of Polybutylene Terephthalate/Dolomite Composites

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ABSTRACT: The objective of this study is to determine the effects of dolomite content on flammability, thermal and mechanical properties of polybutylene terephthalate (PBT)/dolomite composites. PBT and dolomite were blended together at different dolomite contents (5 wt%, 10 wt% and 15 wt%) via twin screw extruder prior to injection moulding. Limiting oxygen index (LOI) test was used to determine the flammability of the PBT composites. Degree of crystallisation ($X_c$), glass transition ($T_g$), melting ($T_m$) and crystallisation ($T_c$) temperatures were obtained using differential scanning calorimetry (DSC). Three types of mechanical tests were carried out, i.e., flexural, tensile and Izod impact. Morphological study was characterised using scanning electron microscope (SEM). LOI analysis showed that the incorporation of dolomite reduced the flammability of PBT composites. DSC results showed that dolomite has marginal effect on $T_g$ and $T_m$ of the PBT composites. On the other hand, $T_c$ and $X_c$ increased with increasing dolomite content which indicates that it can be used as a nucleating agent. The presence of dolomite increased the flexural properties, tensile strength and Young’s modulus of PBT composites. However, the elongation at break and impact strength were significantly decreased due to poor adhesion between dolomite and PBT. PBT/D-15 composite was found to have the optimum mechanical properties compared with other PBT/dolomite composites. SEM micrograph revealed that dolomite is uniformly distributed at lower dolomite content but agglomerated at higher content.

Keywords: Dolomite, polybutylene terephthalate, flammability, thermal properties, mechanical properties

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1. INTRODUCTION

Polybutylene terephthalate (PBT) is one of the important commercial plastics due to its high toughness, high stress-cracking resistance, high resistance to fuels, oils, fats and many solvents, low water absorption, good friction and wear characteristics. Furthermore, PBT is more favourable than its counterpart, polyethylene terephthalate, because of its fast crystallisation and easy processing. PBT can be formed into desired performance-based products, various functional components and structural parts commonly made from metal or thermosets, such as electrical, automotive, telecommunication, machine components, food and medical applications.

In order for PBT to be competitive with other engineering plastics and to be used in a wider range of applications, the properties of PBT need to be further improved. One of the ways that the properties can be improved is through the incorporation of fillers which can increase the stiffness of the composites. A study on the effect of glass fibre on PBT showed that the Young’s modulus increased with increasing glass fibre content. It is previously reported that the addition of inorganic filler can also improve the crystallinity of PBT. Deshmukh et al. studied the effect of different types of filler, i.e., calcium carbonate (CaCO₃), talc and wollastonite, on the non-isothermal crystallisation kinetics of PBT. They found that the crystallinity of the PBT/filler composites increased compared with PBT.

Flame retardancy is another property which needs to be improved since polymers are generally flammable. This improvement can be achieved using flame retardants. However, in many cases, the amount of flame retardant used is large which may affect the ductility of the composites. In a recent paper, it was reported that the addition of an ammonium polyphosphate, which is a non-halogen flame retardant has improved the flammability but at the expense of impact strength.

Dolomite with the chemical structure of CaMg(CO₃)₂ is one of the important minerals used in ferroalloys, glass, alloy steels and flooring tiles. In chemical industry, dolomite is used in making magnesium salts including magnesia (MgO), which are used in pharmaceuticals. In agriculture industry, dolomite is used to decrease the acidity of the soil and to adjust magnesium concentration in soil. Besides that, it is also used as an aggregate in construction and building applications.

Several researchers have reported studies on the effect of dolomite on mechanical properties of polymers. Ali et al. reported that dolomite increased the compressive strength of polyester-based polyurethane composites. Another study reported that dolomite enhanced the modulus and hardness of polyester/
Nik Adik et al. investigated the effects of stearic acid on tensile properties of polypropylene (PP)/dolomite composites at various dolomite contents. The study shows a slight improvement on tensile strength and modulus with the incorporation of stearic acid-modified dolomite (sa-dolomite). Interestingly, unmodified dolomite composites, at all contents, are higher than neat PP and PP/sa-dolomite composites. However, the tensile properties decreased with increasing dolomite contents due to agglomeration of dolomite particles.

Another interesting property of dolomite is that it can be used as flame retardant for polymers since it releases carbon dioxide (CO$_2$) gas at elevated temperature. A few researchers have reported the effect of dolomite on the flammability of polymer/dolomite composites. Amiri et al. reported that polyurethane (PU)/dolomite composite produced longer burning time and lower heat release rate compared to PU/clay composites based on cone calorimeter data. Although many research works have been reported on the incorporation of fillers into PBT, to the authors’ knowledge, no study has been reported on PBT/dolomite composites especially on flammability property. Therefore, the objective of this study is to investigate the effects of dolomite content on flammability, thermal and mechanical properties of PBT/dolomite composites.

2. EXPERIMENTAL

2.1 Materials

Dolomite mineral used in this study was collected from selected dolomite quarries in Sungai Siput, Perak, Malaysia. The original samples were in the form of rocks and were ground into small particles (less than 200 µm) by using a sand crusher. PBT (grade 1100-211M) was manufactured by Chang Chun Chemical (Jiangsu) Co. Ltd. China which is in white pellet form.

2.2 Preparation of PBT/Dolomite Composites

The PBT/dolomite composites were prepared according to the formulation in Table 1. PBT and dolomite were dried for 24 h in an oven at 80°C and were physically mixed prior to compounding process. The composites were compounded via a Brabender Plasticorder (PL 2000) twin screw extruder. The resulting extrudates were then passed through a pelletiser and dried again before being injection moulded (JSW 100Ton) into flexural, tensile and impact samples. Temperature setting for both twin screw extruder and injection moulding is 220°C–240°C.
Table 1: Formulation of PBT/dolomite composites.

| Sample designations | PBT (wt%) | Dolomite (wt%) |
|---------------------|-----------|---------------|
| PBT                 | 100       | –             |
| PBT/D-5             | 95        | 5             |
| PBT/D-10            | 90        | 10            |
| PBT/D-15            | 85        | 15            |

2.3 Limiting Oxygen Index

Limiting oxygen index (LOI) test was conducted according to ASTM D2863-17a. The LOI apparatus consists of a vertical chimney with 75 mm to 100 mm in diameter and 450 mm to 500 mm in height. The specimens with dimension of 70 mm × 6 mm were supported inside the chimney. A gas mixture of oxygen and nitrogen were supplied from the bottom of the chimney for about 30 s. After that, a source of flame was applied to the top of the specimen to ignite it and burned into specimens downward. The aim was to determine the minimum concentration of oxygen that can sustain combustion for 3 min.

2.4 Differential Scanning Calorimetry

Differential scanning calorimetry (DSC) was performed on the Mettler Toledo STAR system to obtain glass transition temperature (T\text{g}), crystallisation temperature (T\text{c}) and melting temperature (T\text{m}). The samples were weighed about 5–10 mg and sealed in an aluminium pan. The heating and cooling rate were 10°C min\textsuperscript{−1} within temperature range of 30°C to 260°C. The characterisation was conducted under nitrogen atmosphere with flow rate 50 ml min\textsuperscript{−1}.

2.5 Flexural Test

Flexural strength is the ability of composite material to withstand bending forces applied perpendicular to its longitudinal axis. To determine the value of flexural strength and flexural modulus, three-point bend testing was performed according to the ASTM D790 at room temperature via Lloyd EZ 20kN universal tensile machine. The span and crosshead speed were set to 50 mm and 3 mm min\textsuperscript{−1} respectively. At least five specimens were tested for each formulation and the average values were taken.
2.6 Tensile Test

Tensile tests were conducted according to ASTM D 638 to obtain tensile strength, Young’s modulus and elongation at break. The specimens were prepared via injection moulding and tested on a Llyord EZ 20kN universal tensile machine at crosshead speed of 5 mm min\(^{-1}\). An average value of at least five specimens per formulation were taken and recorded.

2.7 Impact Test

To measure the toughness, i.e., the ability of polymer composites material to absorb energy when subject to impact, V-notched samples were notched by Atlas automatic notching machine and tested using a Toyosekei Izod pendulum. The specimens were tested at room temperature according to ASTM D256. For each test, five specimens of each formulations were used, and the average value was calculated.

2.8 Scanning Electron Microscopy

The morphological properties of the composites were characterised from the tensile fractured surface of specimens via scanning electron microscopy (SEM). Samples were coated with a layer of platinum/palladium to produce a conductive surface and to improve the imaging of samples. Conductive surfaces enable the electrons to flow evenly and to avoid the electrons from discharging while being scanned. A TM3000 Hitachi electron microscope was used and the accelerated voltage was 15 kV. The magnification was set to 400X.

3. RESULTS AND DISCUSSIONS

3.1 Limiting Oxygen Index

The flammability of PBT/dolomite composites were characterised via LOI in which the minimum oxygen concentration required to sustain the burning of samples for 3 min. Highly flammable polymer materials showed low LOI values compared to less flammable polymer materials which have high LOI values. The LOI results which is presented in Table 2 showed that PBT has the lowest LOI value of 20 which is below the atmospheric oxygen concentration (21%), indicating that it is flammable.
It can be observed that LOI increased with increasing dolomite content indicating that the flammability of PBT/dolomite composites decreased with increasing dolomite content. This is due to dolomite releasing CO$_2$ at elevated temperature. According to Valverde et al., dolomite decomposes via single stage reaction at low CO$_2$ partial pressure, while Olszak-Humienik and Jablonski concluded that the reaction is in equilibrium at temperature approximately above 315°C as illustrated in the following equation:\textsuperscript{15,16}

\[ \text{CaMg(CO}_3\text{)}_2 \rightarrow \text{CaO} + \text{MgO} + 2\text{CO}_2 \]

The decomposition of dolomite is an endothermic reaction, in which the reaction absorbs heat energy by producing metal oxides (CaO and MgO) and releasing CO$_2$, hence slowing the spread of fire.\textsuperscript{16,17} The release of CO$_2$ also dilutes the oxygen concentration in the atmosphere and further slows the spread of fire.\textsuperscript{17,18}

The small improvement is considered good since the amount of dolomite is also small, i.e., up to 15 wt%. According to Mouritz and Gibson, most polymers required a high content of filler to show improvement in flammability effectively.\textsuperscript{19} The minimum content is commonly 20 wt% and the average content is usually 50 wt% to 60 wt%.

### 3.2 Differential Scanning Calorimetry

The transition temperatures ($T_g$, $T_m$ and $T_c$) and degree of crystallinity ($X_c$) of PBT/dolomite composites are presented in Table 3. It can be seen that the incorporation of dolomite has marginal effect on $T_g$ and $T_m$ of the PBT composites. The results obtained are similar to the work reported by Khashkhozheva et al. where the incorporation of montmorillonite into PBT did not significantly increase $T_m$ of the composites.\textsuperscript{20}
Table 3: DSC data of PBT/dolomite composites.

| Sample    | $T_x$ (°C) | $T_m$ (°C) | $T_c$ (°C) | $X_c$ (%) |
|-----------|------------|------------|------------|-----------|
| PBT       | 58.8       | 221        | 194        | 16.7      |
| PBT/D-5   | 60.1       | 222        | 198        | 21.1      |
| PBT/D-10  | 59.2       | 222        | 199        | 29.3      |
| PBT/D-15  | 59.3       | 220        | 199        | 32.3      |

On the other hand, there is a small shift in $T_c$ of the PBT composites which indirectly indicate faster crystallisation rate compared to PBT. Based on the work reported by Deshmukh et al., the incorporation of nano calcium carbonate into PBT increased the $T_c$ and the calculated crystallisation rate were also increased.\(^{21}\) Khashkhozheva et al. also reported similar results.\(^{20}\)

It is also worth noting that the $X_c$ increased with increasing dolomite content. This is expected since $X_c$ is directly correlate with $T_c$ and crystallisation rate. High $T_c$ and crystallisation rate will result in higher $X_c$. These results are in tandem with the research reported by Khashkhozheva et al. and Deshmukh et al.\(^{20,21}\)

### 3.3 Flexural Properties

Flexural strength of PBT composites at different dolomite contents is shown in Figure 1. It can be seen that the incorporation of dolomite significantly increases the flexural strength up to 10 wt% dolomite contents. This increment is due to good dispersion of dolomite particles in PBT matrix and able to transfer stress effectively. At a higher dolomite content (15 wt%), the flexural strength decreased but it is still higher than PBT by approximately 4%. The decrease was probably caused by agglomeration of dolomite particles. A similar finding published by Narkhede et al. on PBT/modified organoclay nanocomposites found that the flexural strength increased by 9% at 0.5 phr.\(^{22}\) The study also reported that the flexural strength started to decrease at organoclay content of 1 phr and higher.
The influence of dolomite on flexural modulus of PBT composites is illustrated in Figure 2. It is worth noting that the incorporation of dolomite significantly improved the flexural modulus. PBT/D-15 has the highest modulus of 2764 MPa compared to others. This increment was due to rigid dolomite particles that restrict the chain mobility and increase the stiffness of the composites. Narkhede and Shertukde reported similar result where the addition of organoclays, treated with different intercalating agents into PBT has significantly improved the flexural modulus from approximately 1800 MPa to 3000 MPa.\textsuperscript{23}
3.4 Tensile Properties

Figure 3 shows the tensile strength of PBT composites increased up to 10 wt% dolomite content. This indicates that dolomite particles are uniformly distributed as justified by SEM micrographs and provide a uniform stress transfer throughout the PBT matrix. The strength decreased as more dolomite was added to the composites due to agglomeration of dolomite particles. This result is similar to previous study by Nik Adik et al. who reported that the tensile strength of PP/dolomite composites increased up to 5 wt% and decreased with increasing dolomite content. Another study by Bakar et al. on recycled PP (rPP)/dolomite composites found that the addition of dolomite into rPP increased the tensile strength of PP composites up to 30 wt%.

![Figure 3: Tensile strength of PBT and PBT/dolomite composites.](image)

Young’s modulus of the PBT composites are presented in Figure 4 which showed similar trends as the flexural modulus. Similarly, PBT/D-15 has the highest modulus at 1483 MPa, 58% higher than PBT. This is due to the stiffness of dolomite that restricts the chain mobility of PBT matrix. This result is also in tandem with finding reported by Nik Adik et al. where the addition of dolomite has increased the Young’s modulus up to 25 wt%. Adesakin et al. also reported similar result.
Figure 4: Young’s modulus of PBT and PBT/dolomite composites.

The effect of dolomite on elongation at break of PBT composites is shown in Figure 5. It can be observed that the incorporation of dolomite has detrimental effect on elongation at break. The lowest elongation at break would be PBT/D-15 where it is 56% lower compared to PBT. This is expected since dolomite has poor adhesion towards the PBT matrix and tends to agglomerate. This has caused dolomite to act as stress concentrator points resulting in the composites to fail in a brittle manner. Similar results were reported by previous researchers working on polymer/dolomite composites, whereby the elongation of break decreased with increasing dolomite content.\textsuperscript{9,12}

Figure 5: Elongation at break of PBT and PBT/dolomite composites.
3.5 Impact Strength

Impact strength is the measure of one’s ability to absorb energy on collision. Figure 6 shows the effect of dolomite on impact strength of PBT composites. It can be seen that the impact strength decreased with increasing dolomite contents with PBT/D-15 having the lowest impact strength 20.0 MPa, 67% lower than PBT. This could be attributed to the presence of dolomite particles which acted as stress concentrators and initiated crack propagations in the PBT matrix. At higher dolomite content, the agglomeration became more significant and thus, further lowering the impact strength. This result is similar to a study reported by Nirukhe and Shertukde on the effect of surface treatments and content of montmorillonite (MMT) on PBT.\textsuperscript{25} They revealed that the addition of MMT significantly reduced the impact strength of the PBT/MMT composites, regardless the surface treatment of MMT, from approximately 52 MPa to 10 MPa for untreated MMT and to 20–30 MPa for treated MMT.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{impact_strength_graph.png}
\caption{Impact strength of PBT and PBT/dolomite composites.}
\end{figure}

It is also worth noting that the impact strength has similar trend to elongation at break where the addition of dolomite has detrimental effect on the composites. This is expected since elongation at break has indirect correlation with impact strength. The low elongation at break shows that area-under-the-curve value in stress-strain curve is small indicating that the composites has small capability to absorb energy. This also indicates that the results are in agreement with each other.
3.6 Overall Mechanical Properties

Stiffness and toughness are two important properties for structural applications. Stiffness is important for load supporting function while toughness offers durability. Figure 7 illustrates the properties of PBT composites in terms of toughness and stiffness. It can be seen that PBT has good toughness but not as stiff as PBT/D-15. On the other hand, PBT/D-10 has good stiffness and toughness, in between PBT/D-5 and PBT/D-15. It can be concluded that PBT/D-10 with flexural modulus of 2510 MPa and impact strength of 32 J/m has the optimum properties in terms of stiffness and toughness.

![Figure 7: Overall mechanical properties of PBT composites in terms of toughness and stiffness.](image)

3.7 Scanning Electron Microscopy

The morphology of tensile fractured PBT composites, shown in Figure 8 illustrates that the dolomite particles were evenly distributed throughout the PBT matrix. According to Zhang et al. and Hulugappa et al., evenly distributed filler can effectively transfer stress across the matrix and reinforce the composites.\textsuperscript{26,27}

It is also worth noting that dolomite particles loosely adhere to the matrix where gaps can be observed surrounding the dolomite particles. These gaps tend to hinder effective stress transfer at high strain rate test e.g. impact test and reduce the mechanical properties. However, these gaps did not affect the tensile and flexural strength due to slow strain rate during the test, hence the polymer chains have time to rearrange to accommodate the gaps.
4. CONCLUSION

The PBT/dolomite composites were successfully prepared via extrusion and injection moulding. The addition of dolomite into PBT has improved flammability and thermal properties of the PBT/dolomite composites. LOI test showed that PBT/dolomite composites with 10% and 15% dolomite content have LOI values above 21 which indicate good flame retardancy. Based on DSC thermogram, dolomite has acted as a nucleating agent for PBT by increasing the degree of crystallinity of the composites. Incorporation of dolomite also enhanced the flexural properties, tensile strength and Young’s modulus of PBT/dolomite composites. Both elongation at break and impact strength decreased with increasing dolomite contents, up to 15 wt% due to agglomeration of dolomite particles as evidenced by the SEM micrograph. From the overall mechanical properties, it is concluded that PBT/D-10 composite has the optimum properties in terms of toughness and stiffness.
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