# TENSILE, SWELLING AND MORPHOLOGICAL PROPERTIES OF BENTONITE FILLED ETHYLENE-PROPYLENE-DIENE MONOMER (EPDM) COMPOSITES

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*Bentonite (Bt) was used as reinforcement in EPDM and the effect of Bt loading on tensile, swelling and morphological properties of EPDM/Bt composites were studied. The composites were prepared using a laboratory scaled two-roll mill with a varying Bt loading of 0 to 70 parts per hundred rubber (phr). Tensile strength, elongation at break ($E_b$), tensile modulus at 100% elongation (M100) and swelling properties of EPDM/Bt composites were all improved in comparison to the pristine EPDM. The optimum tensile and swelling properties of EPDM/Bt composites were obtained with incorporations of 50 phr Bt. The morphology of tensile fractured surface of EPDM/Bt composites observed under scanning electron microscope (SEM) reveals the good dispersion of Bt particles in EPDM matrix as well as good adhesion between Bt and EPDM. Greater interaction between Bt and EPDM were observed with addition of 50 phr Bt whereas agglomerations of Bt particles were observed with addition of 70 phr Bt into EPDM matrix.*

**Keywords:** EPDM, Bentonite, Tensile Properties, Scanning Electron Microscope

## INTRODUCTION

Ethylene propylene diene monomer (EPDM) is a type of synthetic polyolefin rubber developed by Dupont in the 1960s [1]. Demand for EPDM was raised, both in industrial and specialty applications due to its wide range of properties such as excellent resistance to heat, aging and oxidation as well as good mechanical, dynamic and electrical properties. Besides, EPDM also has a great resistance to water, ozone, radiation, weathering and several chemicals (brake fluids and glycol) and also low temperature flexibility, high resilience and the ability to accept high loading of reinforcing materials, fillers and plasticizers. [2-5].

Generally, fillers or reinforcement materials such as carbon black (CB), silica or clay minerals are used in rubber composites to improve the mechanical and thermal properties as well as to reduce cost and sometimes the weight of the rubber compounds. However due to several issues such as pollution, dark colour and usage of petroleum feedstock in synthesis of CB causes researchers to focus on the development of "white" fillers. Several studies have been reported on the substitution of "white" fillers for CB such as silica, calcium carbonate and clay [6-11].

But yet again, researchers faced some other complications with the usage of white fillers in EPDM composites. Silica which is the major "white" filler used as reinforcement in rubber composites is incompatible with EPDM and requires the addition of compatibilizers or coupling agents which complicates the processing method and subsequently increases the production cost. Meanwhile the use of clay minerals (layered silicate) gives rise to deterioration of mechanical properties due to difference in polarity between clay and EPDM [12-14]. Hence, the search for new filler material would be the alternative to maintain the low cost, properties and to eliminate organophilization reaction during the manufacturing of EPDM composites.

In this research work, bentonite (Bt) which is a type of clay has been introduced as a new type of filler in EPDM composite. Bt has a unique atomic structure which attributes to its high adsorption capacity for polymers and are capable for exchanging ions on the silicate layers with reactive hydroxyl (-OH) groups on the surface. Besides, the irregularly shaped, micro-size and the presence of surface pores increase the specific surface area to promote the interaction between Bt particles and EPDM [15-19]. The aim of this work is to study the potential of bentonite as a new type of reinforcement material for EPDM and to study the effect of bentonite loading on the tensile, swelling and morphological properties of EPDM composite.

## MATERIALS AND METHODS

### *Materials*

EPDM, 778Z with ethylene content of 67%, ENB of 4.3% and $M_L$ (1+4) 125°C of 63 MM was supplied by Keltan DSM Elastomers. Density of EPDM, measured by a Precisa (XT220A), was 0.823 $gcm^{-3}$. Bentonite (Bt) was supplied by Ipoh Ceramics (M) Sdn. Bhd. The average particle size and specific surface area of Bt is 5.31 μm and 0.975 sqm/g,

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respectively as measured using a Malvern Mastersizer. After a 24 hours drying at 80°C in an oven, the density of Bt was found to be 2.654 gcm$^{-3}$, measured by a Micromeritices Accupyc 1330 (gas pycnometer). Other ingredients such as zinc oxide (ZnO), stearic acid (SA), sulphur, tetramethyl thiuram disulphide (TMTD) and 2-mercapto benzothiazole (MBT) were all obtained from Bayer (M) Ltd and used as received.

***Preparation EPDM/Bt Composites***

Bt was dried in a vacuum oven at 80°C for 24 hours prior to the compounding to expel the moisture. All the ingredients were prepared according to the compounding formulation as shown at Table 1. The compounding was done using a laboratory scaled two-roll mill, model XK-160 at room temperature with constant mixing time of 20 minutes. The curing properties of the compounds were studied using a Monsanto Moving Die Rheometer (MDR 2000) at temperature 150°C and the compounds were compression moulded into 2mm sheets using a hot press based on the curing time obtained.

**Table 1. Compounding Formulation.**

| Material | C1 | C2 | C3 | C4 | C5 | C6 | C7 |
|---|---|---|---|---|---|---|---|
| EPDM | 100 | 100 | 100 | 100 | 100 | 100 | 100 |
| Bt | 0 | 5 | 10 | 15 | 30 | 50 | 70 |
| ZnO | 5 | 5 | 5 | 5 | 5 | 5 | 5 |
| SA | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 |
| MBT | 0.8 | 0.8 | 0.8 | 0.8 | 0.8 | 0.8 | 0.8 |
| TMTD | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 |
| Sulphur | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 |

* All weight in part per hundred rubbers (phr)

***Tensile Properties***

Dumb-bell shaped specimens were punched out from the compression moulded sheets after 24 hours of storage using a tensile specimen cutter. Tensile strength, elongation at break ($E_b$) and the tensile modulus at 100% elongation (M100) were measured in accordance to ASTM D 412-51 using a Universal tensile testing machine Instron 3366 at room temperature with the crosshead speed of 500 mm/min.

***Swelling Properties***

Swelling test was done in toluene in accordance to ASTM D 471-79. Cured test specimens of the compounds of dimension 30 x 5 x 2 mm were weighed using an electronic balance. The test specimens were then immersed in toluene for 72 hours and the specimens were weighed again to calculate the mass increase due to toluene uptake in term of swelling percentage as shown in Eq. (1).

$$\text{Swelling Percentage} = [(M_2-M_1)/M_1] \times 100 \quad (1)$$

Where $M_1$ is the initial mass (g) of specimen and $M_2$ is the mass of specimen (g) after immersion in toluene.

***Scanning electron microscopy (SEM) Observations***

The morphologies of Bt particles and the tensile fractured surfaces of EPDM/Bt composites were analyzed using a Supra-35VP field emission scanning electron microscope. Test specimens were placed in an aluminium mount with a double sided sticky tape and a thin layer of Pd-Au was coated to prevent the electrostatic charging effect during the observations. This study is intended to observe the microstructure of Bt as well as to analyze the dispersion and adhesion of Bt particles in EPDM matrix.

## RESULTS AND DISCUSSION

***Microstructure of Bentonite***

Fig. 1 is the SEM micrograph of Bt particle. Typically, Bt particles are made up of several entities which is clay minerals, macro-grains, water, air and etc. Bt particles are irregular in shape with many surface pores/voids and the diameters of particles ranges from 5 – 100 μm. Generally, Bt particles have almost similar particle shape as that of most clay minerals but the unique feature of Bt is the presence of surface voids/pores as shown in Fig. 1., which increases the specific surface area for interaction with polymer as well as the possibility for physical interactions via interlocking and/or penetration of matrix at the Bt surface.

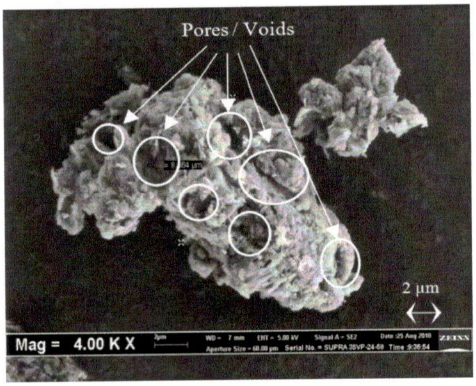


**Fig. 1. SEM micrograph of Bt particle.**

***Tensile Properties***

Fig. 2 shows the tensile strength of EPDM/Bt composites as a function of Bt loading. Tensile strength was increased with the incorporation of Bt compared to the pristine EPDM which gradually increased to an optimum value of about 7.6 MPa with 50 phr Bt loading and decreased slightly at further Bt loading of 70 phr. The raise in tensile strength up to 50 phr Bt loading was due to several factors particularly the good dispersion of Bt particles in EPDM matrix which promotes the good interfacial interaction as well as some possible physical interaction via EPDM chain penetration and/or interlocking at the Bt surface pores. Ismail et al., has reported that stronger interfacial interaction between filler particles and rubber contribute to a more efficient load transfer which subsequently enhances the mechanical performance of the composite system [6]. However at high Bt loading i.e. 70 phr, Bt particles tend to agglomerate due to the presence of large amount of Bt in a limited space promotes higher filler-filler interaction which results in slight decrease in tensile strength.

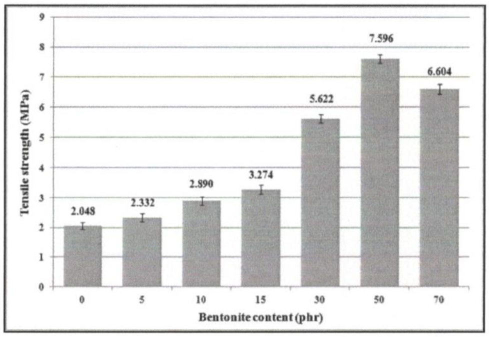


**Fig. 2. Tensile strength of EPDM/Bt composites.**

Fig. 3 illustrates the elongation at break ($E_b$) of the EPDM/Bt composites with the increasing Bt loading. $E_b$ shows similar trend to that of tensile strength which increased to an optimum value of 916% with addition of 50 phr Bt and reduced slightly at 70 phr Bt. As discussed earlier, good dispersion, interfacial interaction and physical interaction between Bt and EPDM attribute to the ability of the composite to withstand higher elongation before the fracture. An effective load transfer from EPDM matrix to Bt particles enhances the ability of EPDM/Bt composite to experience more elongation. At 70 phr Bt loading, Bt agglomeration restricts the mobility of EPDM chains and reduce the effective load transfer from EPDM to Bt. The composite become stiffer and rigid which lowers its ability to elongate with the applied load and gives a lower $E_b$ value.

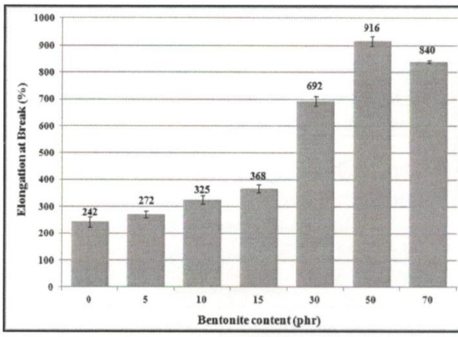


**Fig. 3. Elongation at break of EPDM/Bt composites**

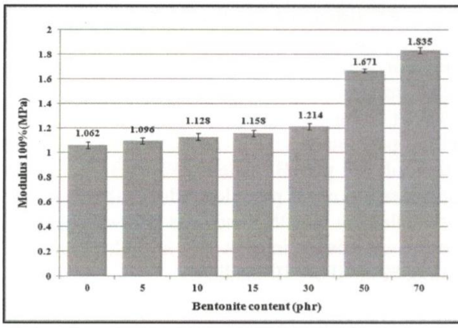


**Fig. 4. *M*100 of EPDM/Bt composites**

Fig. 4 illustrates the tensile modulus at 100% elongation (*M*100) of EPDM/Bt composites as a function of Bt loading. *M*100 was gradually increased with increasing Bt loading. Introduction of Bt onto EPDM reduces the free volume for chain mobility and results in a rigid rubber network. The stiffening effect of rubber network becomes more significant at higher Bt loading. Besides, the increasing interfacial interaction between Bt particles and EPDM chains acts as a part of filler network which also increases the rigidity of the rubber network to give a higher M100 with the increasing Bt loading [20-22].

***Swelling Properties***

Fig. 5 signifies the effect of Bt loading on the swelling percentage of EPDM/Bt which indicate the swelling resistance of the composites as the function of toluene uptake. Results show that the swelling % of EPDM was gradually decreased with increased Bt loading up to 50 phr and increased slightly at 70 phr Bt. This results is in a good agreement with the tensile properties obtained. The improvement of swelling resistance of EPDM/Bt composites was due to the good interfacial interaction between Bt and EPDM as well as the good dispersion of Bt in EPDM matrix. Both this factors allows the EPDM matrix to form a layer surrounding the Bt particles and prevents the solvent diffusion onto Bt.

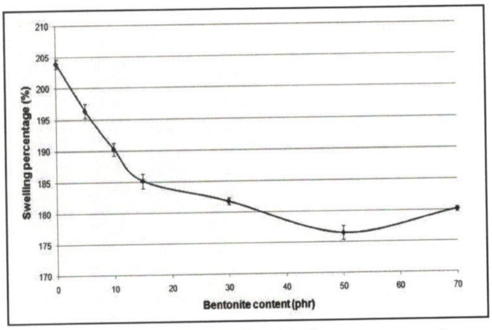


**Fig. 5. Swelling percentage of EPDM/Bt composites**

As discussed at the earlier sections, the presence of possible physical interaction via EPDM penetration and/or interlocking at the surface pores of Bt also influences the reduced toluene uptake by the EPDM/Bt composites. At 70 phr Bt loading, the swelling percentage was increased slightly compared to that of 50 phr Bt filled EPDM/Bt composite. Agglomeration of Bt particles reduces the wetting of Bt particles by EPDM and this allows the direct diffusion of solvent into Bt and a slight increase of swelling % at 70 phr Bt loading.

***SEM Morphological Observation***

Fig. 6 shows the tensile fracture surface of unfilled, pristine EPDM. As can be seen, the fracture surface of pristine is flat and smooth with almost no voids. This indicates the low resistance of EPDM to fracture which breaks at lower tensile strength with lowest $E_b$.

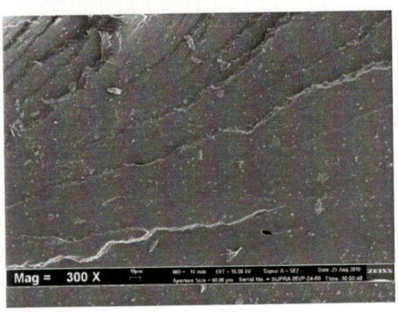


**Fig. 6. SEM micrographs of tensile fracture surface of unfilled, pristine EPDM.**

Fig. 7 (a) to (d) represents the fracture surface of tensile specimen of EPDM/Bt composites with different Bt loading at high magnification (300x). At 10 and 30 phr Bt loading, the fracture surface exhibit minimum voids due to Bt pullout as shown at Fig. 7 (a) and (b) (white circles). Meanwhile, at 50 phr Bt loading, good filler-rubber interactions and less Bt particle pull out were observed at fracture surface as shown by Fig. 7 (c) which shows a better interaction between Bt particles and EPDM. Finally the fracture surface of EPDM/Bt with 70 phr Bt loading shows a different morphology where Bt agglomerates were observed. The Bt particles stacked to form agglomerates as shown is Fig. 7 (d) (white circles), which increases the filler-filler interactions and consequently reduces the Bt-EPDM interactions.

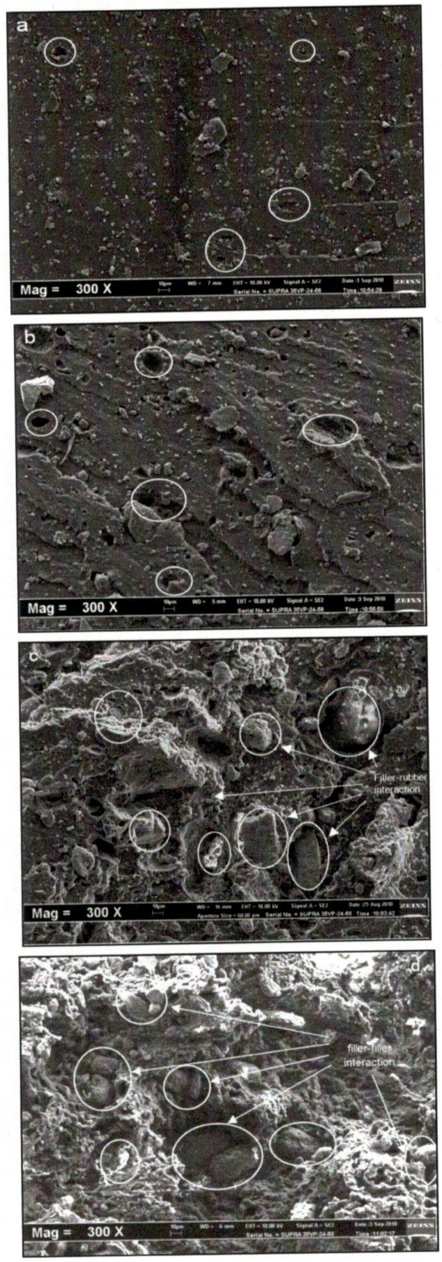


**Fig. 7. SEM micrographs of tensile fracture surface of EPDM/Bt composites with (a) 10 phr, (b) 30 phr, (c) 50 phr and (d) 70 phr Bt loading.**

## CONCLUSION

Bentonite has a high potential to be used as reinforcement filler in EPDM. The tensile strength, $E_b$ and swelling resistance of EPDM was enhanced with the incorporations of 50 phr Bt. Tensile modulus (M100) were gradually increased with the addition of Bt. SEM micrographs shows the prove of increasing interfacial interaction between Bt and EPDM with increasing Bt loading up to 50 phr and greater interaction in between Bt particles at 70 phr Bt which leads to the agglomeration.

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