

# EFFECT OF METHYL-METHACRYLATE-GRAFTED-NATURAL RUBBER-49 ON MECHANICAL PROPERTIES OF FILLED CALCIUM CARBONATE UNPLASTICISED PVC

J.W.Lim<sup>1</sup>, Azman Hassan<sup>1</sup>, Aznizam Abu Bakar<sup>1</sup>

<sup>1</sup>Polymer Engineering Department, Faculty of Chemical and Natural Resources Engineering, University Technology Malaysia, 81300 UTM Skudai, Johor Bahru.

**Abstract.** *Unmodified unplasticised PVC (PVC-U) has the disadvantages of being prone to occasional brittleness and is notch sensitive. Various types of rubber are being used in PVC-U as impact modifier which formed multi-phase systems. The main objective of this research is to study on possibility of methyl-methacrylate-grafted-natural rubber-49 (MG49) to be used as impact modifier in filled calcium carbonate unplasticised PVC. MG 49 is a modified NR which is made by polymerizing methyl methacrylate monomer in latex so that polymer chains become attached to the rubber molecule. In the sample preparation, dry blending was carried out with laboratory blender and milled into sheets using two-roll mill. Test specimens were molded using a hot press before carrying out the various mechanical testing that is impact, flexural and tensile testing. The results show that the impact strength increases as the impact modifier MG 49 content increases in the filled calcium carbonate PVC-U samples. However, similar increase in the impact modifier content resulted in a decrease in flexural modulus and yield stress. The formulation containing 8 phr impact modifier MG 49 and 10 phr calcium carbonate was found to be most optimum formulation in term of impact strength, flexural modulus and yield strength.*

**Keywords:** *PVC-U; MG 49; impact modifier; mechanical properties; calcium carbonate*

## 1. Introduction

Polymer blending is a useful technique for designing materials with a variety of properties. The polymer blends often exhibit properties that are superior compared to the properties of each individual component polymer. The main advantages of the blend system are simplicity of preparation and ease of control of physical properties by compositional change. In this study, blending of PVC with impact modifier will be discussed. Unmodified unplasticized PVC (PVC-U) has the disadvantage of being prone to occasional brittleness and is notch sensitive. To increase the impact resistance, multi-phase system consisting of PVC and a rubber component, as known as impact modifiers, were developed and are being used. Research using methyl methacrylate-grafted-natural rubber (MG 49) as impact modifier in PVC is still limited. In order to fulfill this requirement this study has been carried out as part of the objective of this research.

MG rubber is a modified natural rubber (NR) which is made by polymerizing methyl methacrylate monomer in latex so that polymer chains become attached to the rubber molecule. Three grades of Heveaplus MG are commercially available: MG 30 based on a nominal 30 % methyl methacrylate content, MG 40 based on 40%, and MG 49 based on 49%. Heveaplus MG rubber giving compounds of high tensile modulus, hardness, good electrical properties and fine finish. It also acts as a specific adhesive for bonding PVC to NR. According to Bevan and Bloomfield (1963), quoted from "Handbook of Elastomers"[2], there is a substantial increase in bond strength obtained between NR and PVC once the level of poly(methyl methacrylate) is excess of 35%. Rezaifard and others [1] have done studies on evaluation of MG30 and MG50 as toughening agents for epoxy resin and concluded that both MG rubbers are good toughening agents for epoxy resin. Previous

and concluded that both MG rubbers are good toughening agents for epoxy resin. Previous researches have also show that adding rubber material as impact modifiers will decrease the modulus and yield stress of the plastic-rubber blend [9,10]. Filler is one of the most important additives used in this research that is precipitated calcium carbonate (PCC) with diameter 1  $\mu\text{m}$ . PCC was added into formulation sample to overcome the problem of the modulus reduction.

## 2. Experimental

### 2.1 Materials

The PVC used in this study is a suspension type homopolymer PVC with solution viscosity K-value 66, MH-66, supplied by IRM (Industrial Resins Malaysia Sdn. Bhd.). It is a medium molecular weight resins designed for general purpose, rigid and flexible applications. The impact modifier MG 49 used in this research supplied by Green HPSP (M) Sdn. Bhd. It is produced by polymerizing 50 phr monomer MMA into 50 phr natural rubber poliisoprene. Others additives using in this research were list Table 1.

Table 1: Types of additives

Additives	Types
Fillers	PCC (1 $\mu\text{m}$ )
Impact Modifiers	MG 49
Stabiliser	Organo-Tin
External lubricants	Cast
Internal lubricants	Hst
Processing aids	Acrylic Polymers
Pigment	TiO <sub>2</sub>

### 2.2 Sample preparation

Before processing the PVC, all the additives shown in Table 1 were blended according to the formulation in Table 2. High-speed laboratory mixer was used in the dry blending of the resin and additives. To produce molded sample, the dry blends of PVC powder and additives were firstly sheeted on a two roll mill with temperature  $158 \pm 0.5$  °C. The sheeted PVC was cut and filled into the moulds before placing in a hot press for molding at 185 °C.

### 2.3 Measurements of impact properties

Impact test was performed according to ASTM standard D256 on an Izod impact tester at impact force 7.5 J, impact speed 3.8 m/s and operation condition at 23°C.

### 2.4 Measurements of flexural properties

Flexural test was performed according to ASTM standard D790 on an Instron 5567 machine at crosshead speed of 3mm/min, operation condition at 23°C and 50 % humidity. Length span support was 51mm.

### 2.5 Measurements of tensile properties

Tensile test was performed according to ASTM standard D638 on an Instron 5567 machine at crosshead speed of 50mm/min, operation condition at 23°C and 50 % humidity.

Table 2: Blend Formulation

	Formulation (phr)							
	S0	S1	S2	S3	S4	S5	S6	S8
PVC	100	100	100	100	100	100	100	100
MG49	0	0	4	8	12	16	20	8
PCC	0	10	10	10	10	10	10	0
Cast	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Hst	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6
PA20	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
TiO2	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0
Tin	2	2	2	2	2	2	2	2

### 3. Results and Discussion

#### 3.1 Impact test

As shown in Figure 1, the impact strength of PVC-U sample increases with increasing MG 49 from 0 to 12 phr. However, in the range 16 to 20 phr MG 49 the value of impact strength sample decreases slightly. It is interesting to observe a significant increase (around 34% increment) in impact strength values as the impact modifier loading increases from 4 to 8 phr. This may suggest that a loading of 8 phr MG 49 impact modifier content in 10 phr calcium carbonate filled PVC-U were most effective in term of impact strength enhancement. The entire sample failed in brittle mode with stress whitening being observed at fracture surface of the samples. These results are in agreement with the findings of Lutz [15] who stated that the impact strength of impact modified PVC-U is improved as the impact modifier concentration increases from 2 to about 20-25 phr. However the impact strength begins to drop off from a maximum above 20-25 phr.

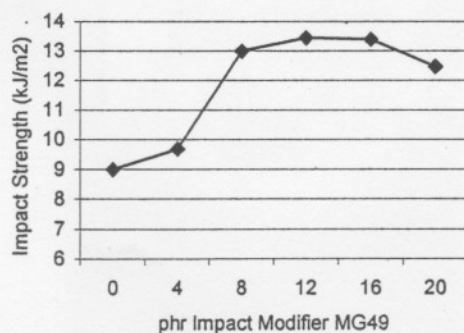


Fig. 1 Effect of MG 49 Content (phr) on Impact Strength

Figure 2 show the impact strength sample with content of precipitated calcium carbonate. As shown in figure 2, the impact strength of PVC-U sample increases (around 7% increment) with adding 10 phr of calcium carbonate. Normally calcium carbonate is group to extender fillers, but in this research it be came functional fillers to increases the impact strength. These result is in agreement with the review by Mathur [12] who stated that a gray areas where overlap and poor definition exist, like same of the extender fillers, when reduced particle size/surface-treated, would be reclassified as functional fillers.

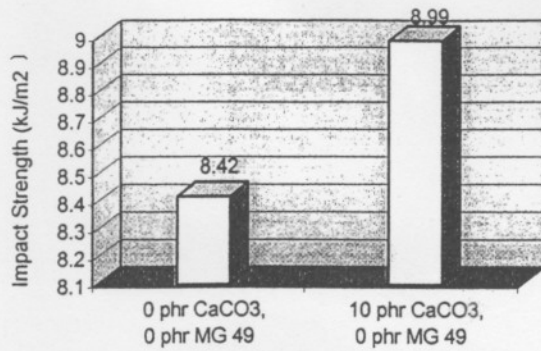


Fig. 2 Effect of Calcium Carbonate Filler on Impact Strength

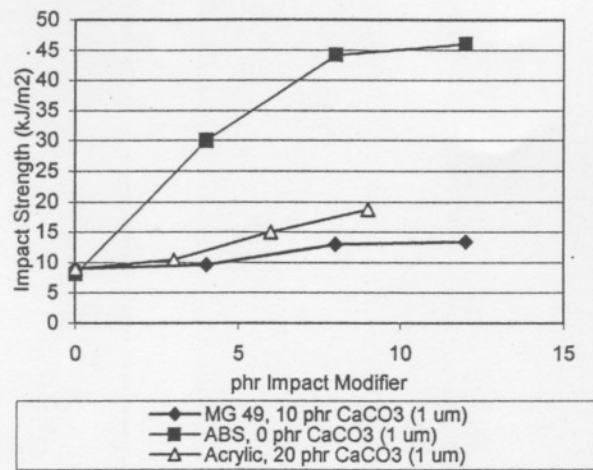


Fig. 3 Comparison Performance between difference types of impact modifier on impact strength.

The results impact test of this research were compared with previous investigate conducted in UTM[9,10]. Fig. 3 shown that performance of MG 49 is very much smaller (>210%) than ABS impact modifier. However it were similar impact strength with acrylic polymer impact modifier where the difference between them only around 28%.

### 3.2 Flexural Test

In flexural test, flexural modulus was measured which it was one of the basic properties of polymer. Fig. 4 shown that flexural modulus sample PVC-U decreases with the content of impact modifier MG 49. It observed that flexural modulus decreases only slightly (around 4%) from 4 to 8 phr MG 49. These result is in agreement with the research results of Yee Joon Wee and Sivaneswaran[9 ,10] where the flexural modulus sample PVC-U decreases with the content of acrylic polymer and ABS impact modifier. Flexural modulus affected by interfacial adhesion between polymer and fillers. Flexural modulus properties decrease when the interfacial adhesion was weak.

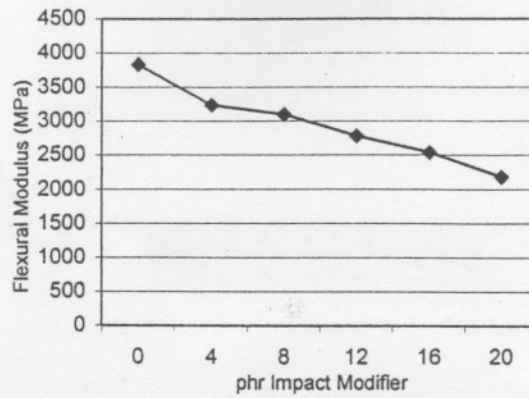


Fig. 4 Effect of MG 49 Content on Flexural modulus

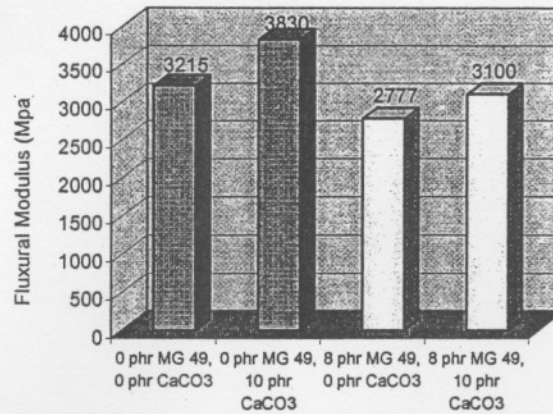


Fig. 5 Effect of Calcium Carbonate on Flexural Modulus

Fig. 5 shown that flexural modulus sample PVC-U increases for every formulation when 10 phr precipitated calcium carbonate were added. These results were agreement with other research like Nielson (1965) reported that stiffness is a direct effect of flexural modulus and increase with fillers volume fraction if calcium carbonate filler is harder than matrix. Flexural modulus sample increases with content of filler, this can explain by certain zones around the filler particles, the polymeric phase differs in structure and properties from bulk polymer matrix. The polymer segments attached to fillers surfaces by primary or secondary valence bonds in turn cause a certain immobilization of the adjacent segments. These increases the glass transition temperature thus causes a stiffening of the material as a whole.

### 3.3 Tensile Test

Yield stress is the value of stress when the plastics begin unrecovery deformation or when  $\sigma_y$ , slope of curve was zero. Fig. 6 shown that yield stress of sample PVC-U decreases with content of impact modifier MG 49. Tensile properties polymer are effect by secondary bonding strength between polymer chains like Van der waals bonding, hydrogen bonding or dipolar bonding. PVC-U consists one atom chlorine each repeat unit. So that the bonding strength between PVC-U molecular chain are strong. But the particles of impact modifier disturb and weakness the bonding. So that, yields stress sample decreases.

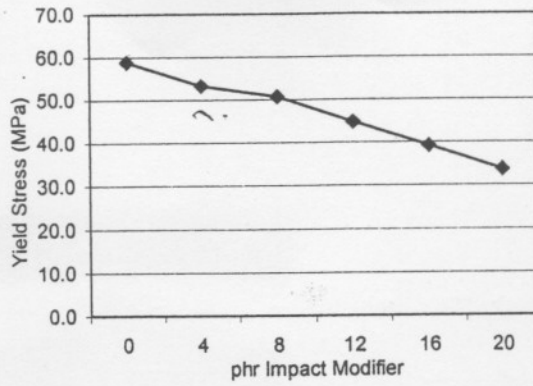


Fig. 6 Effect of MG 49 Content on Yield Stress

Fig. 7 shown that the effect of calcium carbonate content to yield stress are same with the flexural modulus where yield stress decreases with content of impact modifier. Particles calcium carbonate added into PVC-U matrix were distributed and located between the space of chains and it will stop the slippage of polymer chains when the tensile force applied. Thus, the yield stress will be increase.

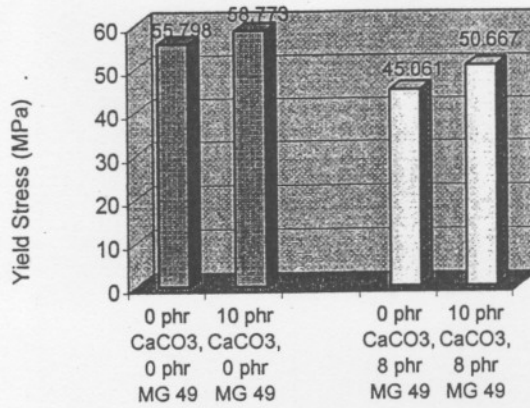


Fig. 7 Effect of Calcium Carbonate on Yield Stress

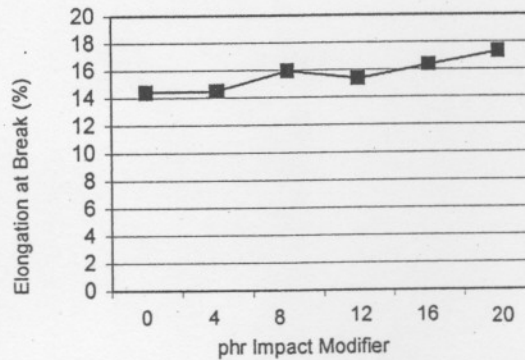


Fig. 8 Effect of MG 49 Content on Elongation At Break

Elongation at break is the value of strain when the samples break up. Fig. 8 shown that elongation at break for sample PVC-U increases with inconsistent where elongation at break decreases when content of MG 49 at range 8 to 12 phr. But, overall Fig.8 has shown that elongation at break for PVC-U sample were increases with content of impact modifier.

## Conclusion

From the study, PVC-U blended with MG 49 as an impact modifier has higher impact strength than the unmodified PVC. The use of PCC also increases the impact strength PVC-U. However performance of MG 49 impact modifier is very much smaller than ABS impact modifier but it is similar with the performance of acrylic polymer. MG 49 was also found to decrease the flexural modulus and yield stress. The use of 10 phr precipitated calcium carbonate can overcome this detrimental effect of MG 49 on flexural modulus and yield stress. From the overall analysis, it is found that formulation S3 (8 phr MG 49 impact modifier, 10 phr calcium carbonate) was the optimum composition in terms of impact, flexural and tensile.

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