

## Rubberwood-polymer Composites: The Effect of Chemical Impregnation on the Mechanical and Physical Properties

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**ABSTRACT:** Impregnation technique with suitable chemicals can be used to improve the dimensional stability and mechanical properties of wood. The impregnation can be carried out via in-situ polymerization either by using catalyst-heat treatment or irradiation technique. In this study, the rubberwood-polymer composites were prepared by impregnating the rubberwood with glycidyl methacrylate (GMA), diallyl phthalate (DAP) and the combination of GMA and DAP. The catalyst-heat treatment was employed for the polymerization process to take place. The weight percentage gain of impregnated samples increased as impregnation pressure increased for all monomers used. At the highest chemical loading, rubberwood-polymer composites impregnated with GMA exhibited the best dimensional stability and mechanical performance.

*Keywords:* Wood-polymer composites, impregnation technique, glycidyl methacrylate, diallyl phthalate, dimensional stability, mechanical properties.

### 1.0 INTRODUCTION

The dimensional stability and mechanical properties of wood can be improved by using impregnation technique with suitable chemicals that can react with a cell wall component [1]. Impregnation of wood for producing wood-polymer composites (WPC) can be carried out via in-situ polymerization either by using catalyst-heat treatment or irradiation technique [1].

The most common monomers used are styrene, methyl methacrylate (MMA), vinyl or acrylic monomers. Apart from difficulties encountered during machining of these types of WPC, such as clogging of abrasive grit along with saw dust, these types of WPC displayed a relatively low dimensional stability in water [2]. This may be due to the fact that most monomers used are confined only in the lumen, not in the cell wall [2]. If a monomer can enter and bulk the cell wall, the dimensional stability of the WPC will be improved. Thus, a system is sought to impart effective dimensional stability, as well as to reduce the difficulties during machining. The system should consist of a monomer that has the ability to penetrate into the cell wall and copolymerize with other monomers that can provide reactive sites for crosslinking. Since the crosslinkable compositions in the preparation of WPC will lead to superior performance with respect to high temperature resistance during machining and sanding, this study was carried out to produce WPC based on glycidyl methacrylate (GMA) and diallyl phthalate.

GMA is a difunctional monomer, which consists of a terminal C=C and an epoxy group. GMA was shown to be able to react with wood hydroxyl groups through its epoxy end [3]. Diallyl phthalate (DAP) is a tetrafunctional monomer and therefore provides ideal reactive

sites for crosslinking reactions. The purpose of this study is to improve the mechanical and dimensional stability of wood composites via monomer impregnating technique.

## 2.0 EXPERIMENTAL

### 2.1 Material

Rubberwood sample used in this study was supplied by Merbok MDF Sdn. Bhd., Sungai Petani, Kedah, Malaysia. The monomers, glycidyl methacrylate (GMA) and diallyl phthalate (DAP) and initiator benzoyl peroxide (BPO) were purchased from Komita (M) Sdn. Bhd. Penang, Malaysia. The monomer systems used in this study are listed in Table I.

Table 1 The Monomer System Used in Impregnation

Sample	Monomer System
GMA	100% glycidyl methacrylate + 2% benzoyl peroxide (by weight)
GMA/DAP	50% diallyl phthalate + 50% glycidyl methacrylate + 2% benzoyl peroxide (by weight)
DAP	100% diallyl phthalate + 2% benzoyl peroxide (by weight)

### 2.2 Method

#### 2.2.1 Preparation of samples

The rubberwood samples were sawn into specimens of 30 × 20 × 20 mm (length × width × thickness) for dimensional stability and compression tests. The dimensions of samples for bending test were 150 × 10 × 10 mm and for impact test was 50 × 10 × 10 mm. Six (6) replicates were used for each test.

#### 2.2.2 Extraction of samples

All samples were extracted in a solution mixture consisting of toluene, ethanol and acetone (4:1:1, vol./vol.) for 3 h. The extracted samples were oven dried at 105<sup>0</sup>C for 16 h.

#### 2.2.3 Samples treatment

All oven dried samples were measured for their dimensions and weights. The samples were then placed in an impregnation chamber, which was evacuated to three levels of vacuum pressure of about 5, 15 and 25 mm Hg and held for 5 min. The respective monomer system was introduced into the chamber as the vacuum pressure was released. The samples were kept immersed in the monomer solution for 4 h at ambient temperature and atmospheric pressure to obtain further impregnation. Samples were then removed from the chamber and wiped free of excess impregnate. Monomer intake was determined as follows (equation 1):

Samples were wrapped with aluminum foil and placed in an oven for 24 h at 105<sup>0</sup>C for polymerization to take place. The curing process was continued for another 24 h at 105<sup>0</sup>C for

24 h. The samples were then measured to determine volume increase after cure and weighed to determine the polymer loading.

$$\text{Weight Percentage Gain (WPG), (\%)} = \frac{W_1 - W_2}{W_2} \times 100\% \quad (1)$$

where  $W_1$  is the weight of wood samples after curing, and  $W_2$  is the weight wood samples (oven dry) before impregnation and curing.

## 2.3 Testing

### 2.3.1 Mechanical test

Bending and compression tests (ASTM D790) were performed on the cured and control samples using a Universal Testing Machine Model STM 10 at a crosshead speed of 2 mm/min. The impact tests were done using a Zwick Impact Tester according to ASTM D256.

### 2.3.2 Dimensional stability

Dimensional stability and water absorption test were determined by soaking the cured samples in a water bath at room temperature for 24 h for each cycle until 7 days. The weight and dimension of the samples were measured before and after soaking. The water absorption (equation 2), anti shrink efficiency (ASE) (eq. 3) were calculated as follows:

$$\text{Water absorption (\%)} = \frac{W - W_0}{W_0} \times 100\% \quad (2)$$

where  $W$  is the sample weight after soaking, and  $W_0$  is weight of sample prior to soaking.

$$\text{Anti shrink efficiency (ASE) (\%)} = \frac{S - S_0}{S_0} \times 100\% \quad (3)$$

where  $S$  is the volumetric swelling coefficient for treated wood, and  $S_0$  is the volumetric swelling coefficient for untreated wood. The volumetric swelling coefficient ( $S$ ) is calculated follows (eq. 4):

$$S = \frac{V - V_0}{V_0} \times 100\% \quad (4)$$

where  $V$  is the volume of wood after soaking, and  $V_0$  is the volume of wood before soaking.

## 2.0 RESULTS AND DISCUSSION

The results of bending test are shown in Figure 1, 2 and 3. The weight percentage gain (WPG) of impregnated samples increase as the impregnation pressure is increased (Fig.

4) for all monomers used. Figure 1 shows that in general the moduli of rupture (MOR) of the impregnated samples are higher than the untreated samples. This is possibly due to the formation of covalent bonding between wood OH groups and epoxy groups of GMA. This may contribute to the enhancement of stress transfer in the system (see Fig. 5 for a possible reaction of GMA and rubberwood samples). Samples impregnated with GMA demonstrate higher MOR than samples impregnated with 50% GMA/50%DAP and DAP for all impregnated pressures. As shown in Figure 2, stiffness or moduli of elasticity (MOE) of impregnated samples are generally higher than those without impregnation. MOE of the samples increases with the increase in pressure loading. The increasing of pressure may result in more loading of monomers in the lumen. The stiffness of the polymer formed may contribute to the increase of stiffness of the impregnated wood. The results of toughness show similar trends as MOR and MOE (Figure 3). It is also found that the samples impregnated with DAP and GMA/DAP at 5 and 15 mmHg of impregnation pressure showed lower toughness than untreated samples. It is understandable because DAP cannot react with OH groups of wood but rather will react with itself to produce homopolymer in the cell wall. It is assumed that the amount of homopolymer formed for 5 and 15 mmHg impregnation pressure is not sufficient to impact any increase in toughness. On the other hand it may disrupt the continuity of the cell wall structure. GMA impregnated samples show the highest toughness followed by GMA/DAP and DAP samples at 25 mmHg impregnation pressure. Since toughness corresponds to the amount of energy needed to break the samples, this shows that GMA impregnated samples absorb more energy than GMA/DAP and DAP samples. However, at 5 and 15 mmHg, GMA/DAP samples show higher toughness than the rest of the samples. This may indicate the possibility of copolymerization between methacrylic double bond of GMA which is attached to wood and DAP [4].

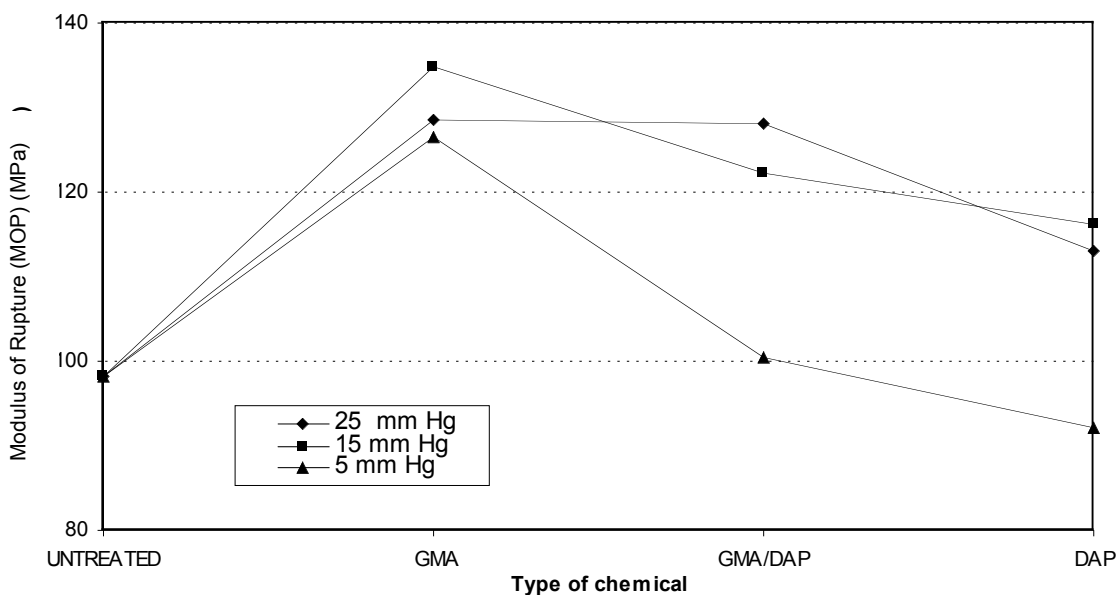


Figure 1: Modulus of rupture (MOR) of samples impregnated with various WPG at various pressures

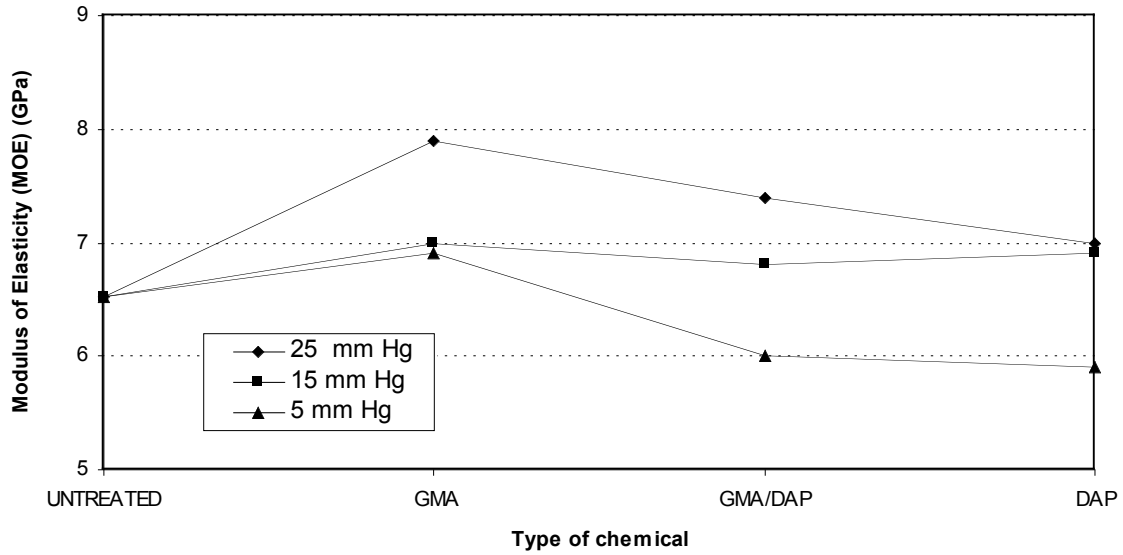


Figure 2: modulus of elasticity (MOE) of samples impregnated with various WPG at various pressures

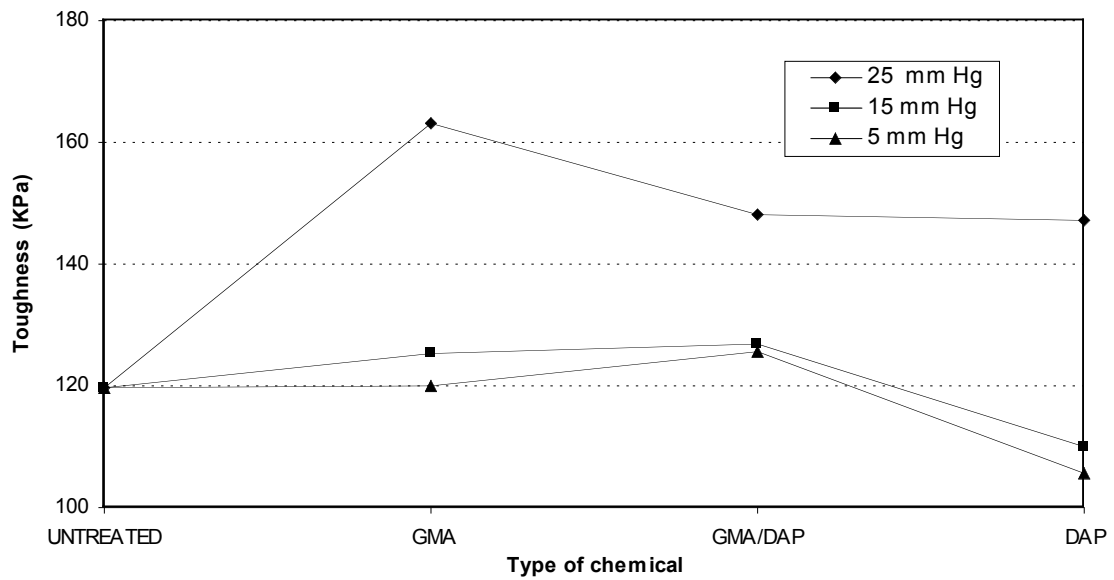


Figure 3: Toughness of samples impregnated with various WPG at various pressures

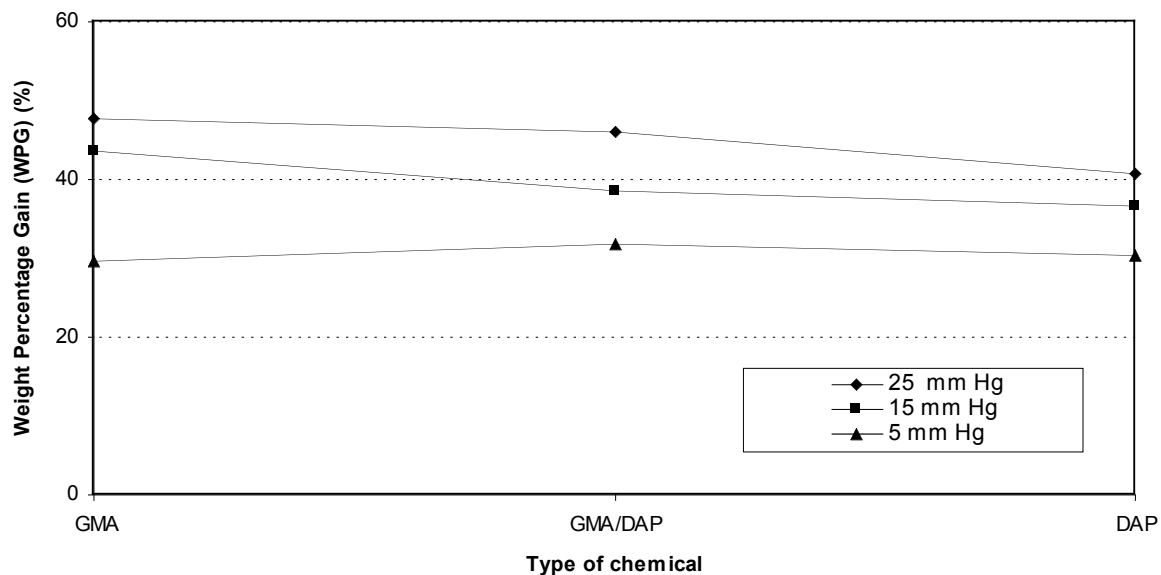


Figure 4: The weight percentage gain (WPG) of bending test samples at various pressures

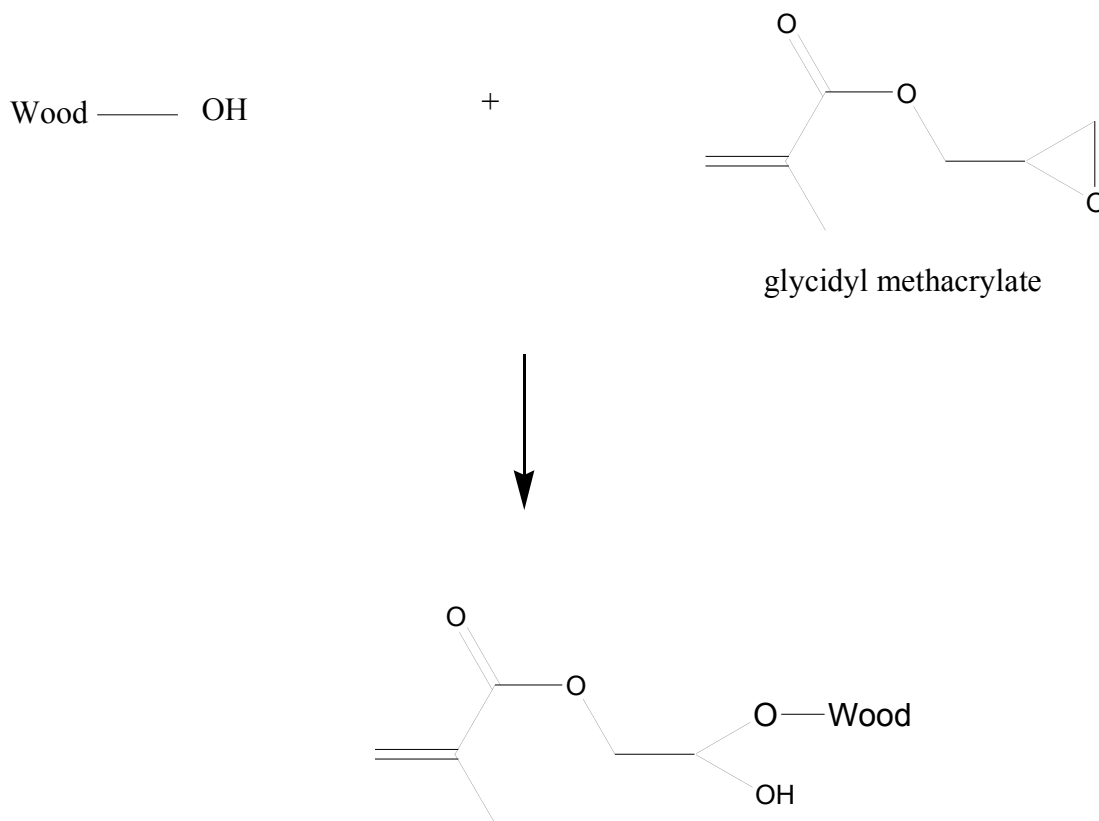


Figure 5: Possible reaction between GMA and Wood

Figure 6 shows the WPG results for the compressive strength samples. The copolymerization may produce a continuous interphase between wood and polymer in the cell wall. Inadvertently this would result in better stress transfer in the cell wall. Figure 7

shows that all impregnated samples display higher compression strength than those of unimpregnated. Unimpregnated sample may fail in compression because of the buckling of relatively thin cell walls due to a long column type of instability. The presence of polymer of the cell wall enhances the lateral stability [5]. The GMA impregnated samples show the highest compressive strength than the other type's monomer-impregnated samples at all pressure loading especially at 25 mmHg. It is expected because GMA has the ability to react with the cell wall and thus forming a polymer coating on its surface. This would enhance the lateral stability of the cell wall.

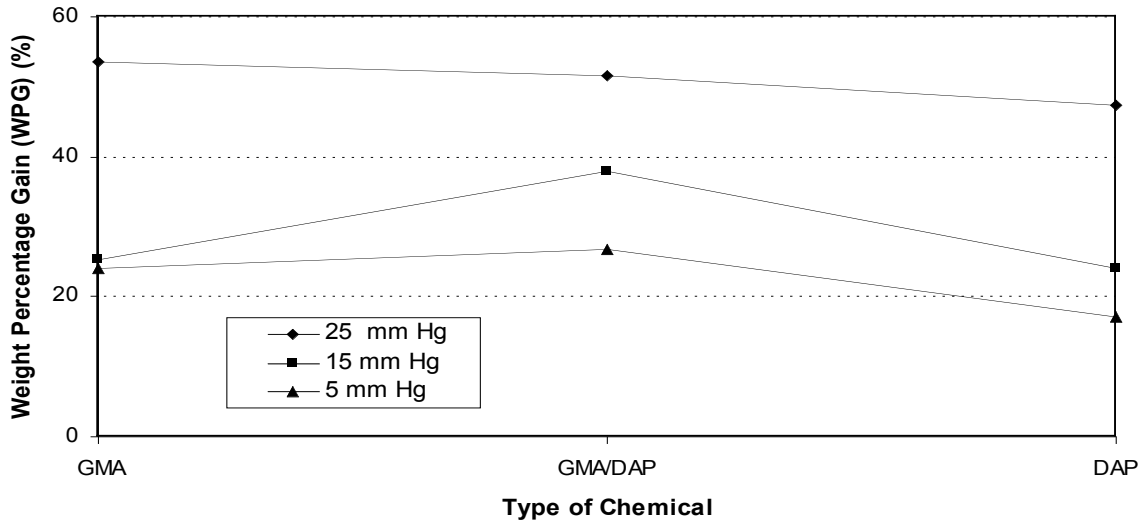


Figure 6: The weight percentage gain (WPG) of compression test samples at various pressures

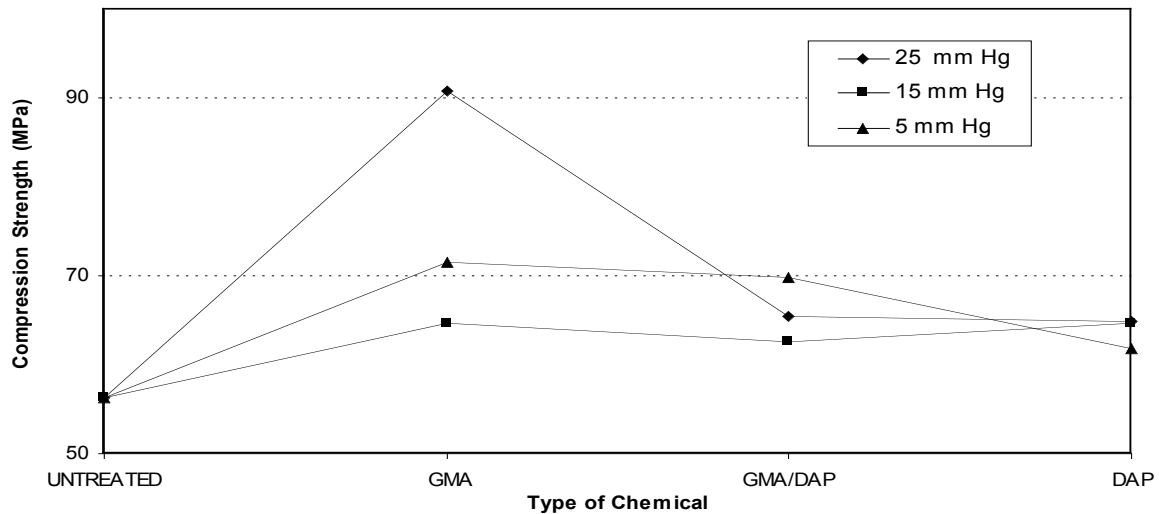


Figure 7: Compression strength of samples impregnated with various WPG at various pressures

Figure 8 also shows that for impact strength of treated samples the WPG increases as the pressure increased. Overall, the impact strength of treated samples increases with

increasing pressure (Figure 9). Crack initiation and crack propagation in impregnated samples was sufficiently modified by the presence of polymer in the cell wall as well as in lumen which improves the impact strength [5]. Of the three monomer systems used, the GMA samples show the highest impact strength as compared to GMA/DAP and DAP systems. This may be due to the smaller molecular size of GMA which allows more penetration of monomer into the cell wall, thus improving impact strength [5].

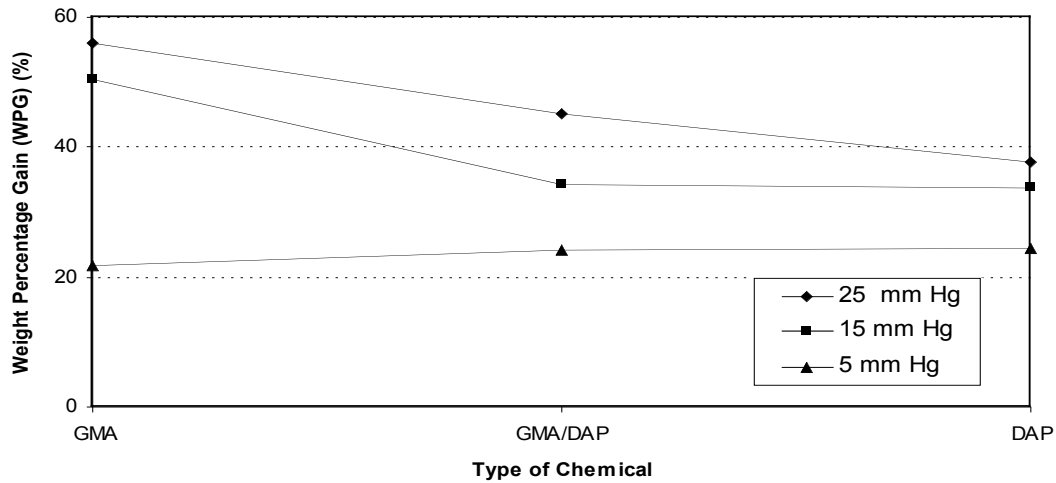


Figure 8: The weight percentage gain (WPG) of impact test samples at various pressures

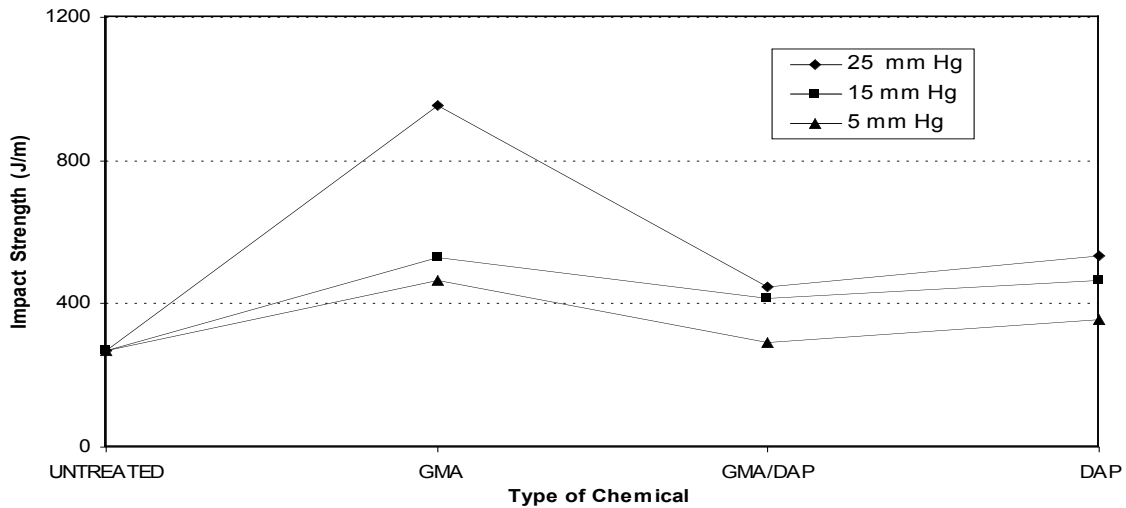


Figure 9: Impact strength of samples impregnated with various WPG at various Pressures

Fig. 10 shows the WPG for dimensional stability test samples. Figure 11 demonstrated the results of water absorption tests. From the figure, the untreated samples exhibited higher percentages of water absorption than the treated samples. The results show that the untreated samples exhibits higher water absorption than the treated samples. It is expected because cell wall with OH groups will absorb water to its surface through the

formation of hydrogen bondings. From the results, it can be seen that those with GMA display the lowest absorption. This is expected since GMA through its epoxy group has the ability to react with OH groups of the cell wall and renders it less hydrophilic. Whilst, for DAP, the homopolymers formed in the cell wall as well as lumen may only retard the absorption of water on the cell wall surface, since DAP has no ability to react with the OH groups of the cell wall. The samples with GMA/DAP show the mixture of effect of these two monomers. The same results are consistent with other investigations [5, 6]. On the other hand, the GMA/DAP system (see Figure 12 for a possible reaction between GMA/DAP) was obviously undergone copolymerization for a higher cohesive strength and performed good physical properties [5].

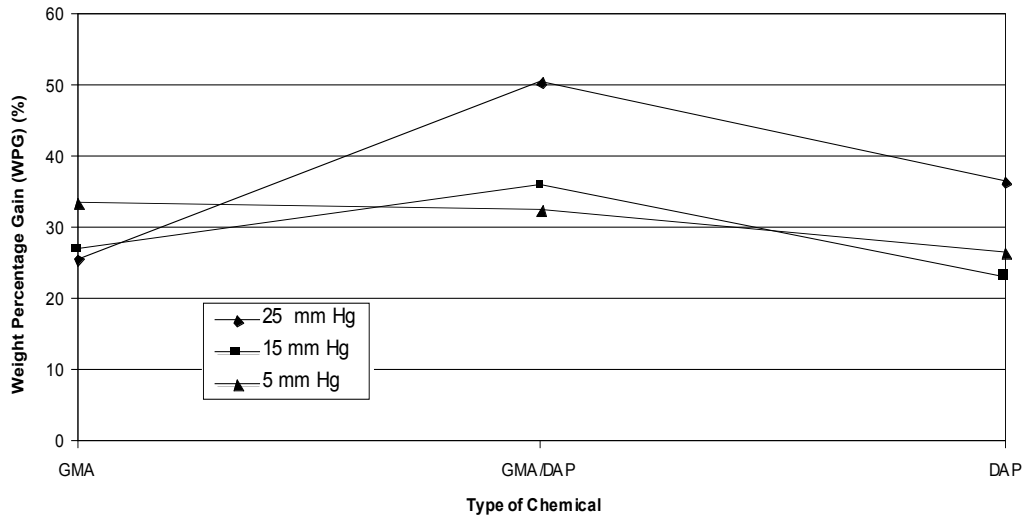


Figure 10: The weight percentage Gain (WPG) of dimensional stability test samples at various pressures

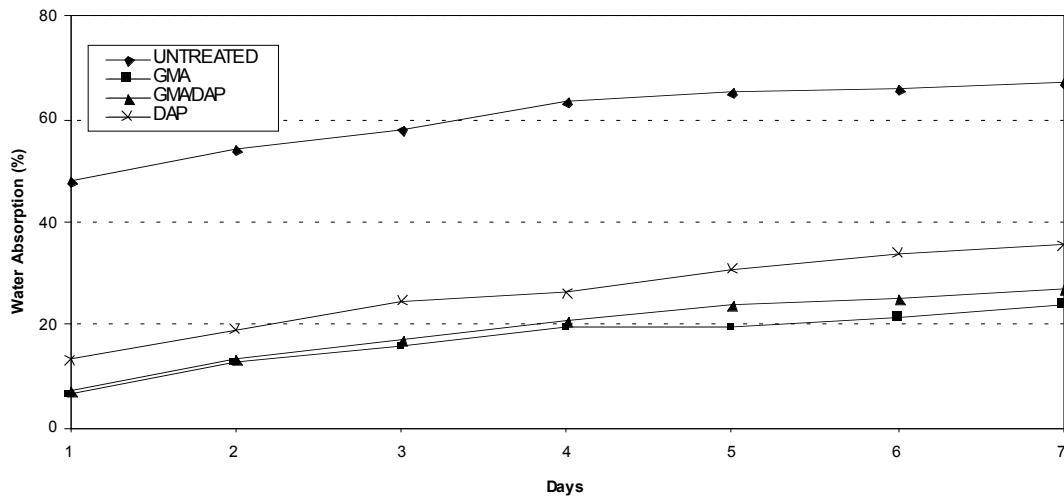


Figure 11: Water absorption of samples impregnated with various WPG at 25 mm Hg

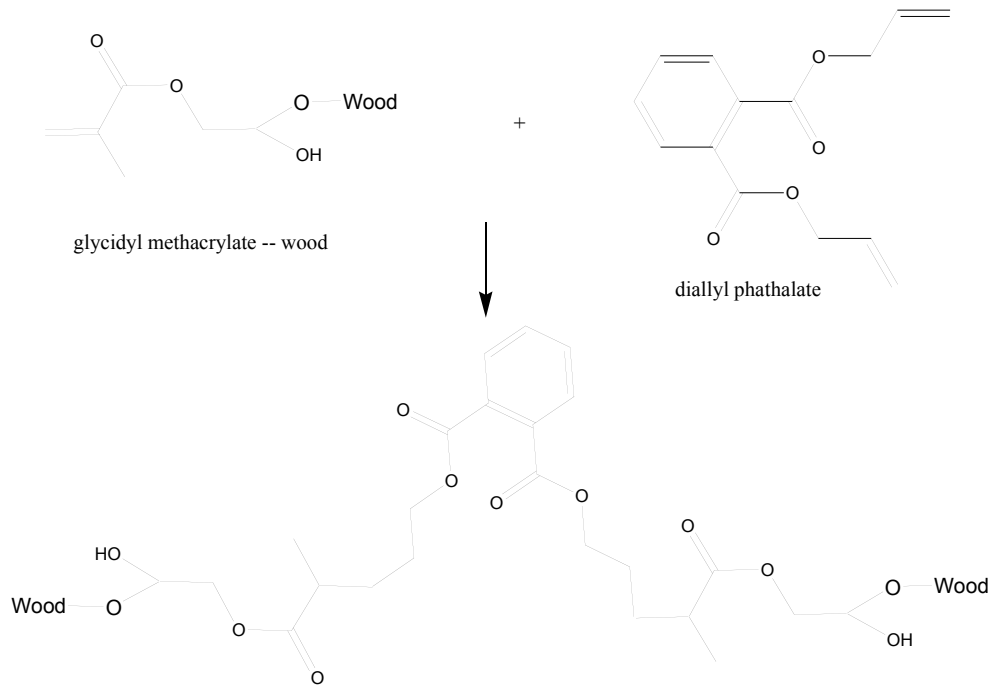


Figure 12: Possible reaction between GMA-Wood and DAP

The result of anti shrink efficiency (ASE) is displayed in Figure 13. The ASE of samples impregnated with GMA is higher than those of GMA/DAP and DAP. In general, the impregnated samples show a decrease in the ASE after soaking in water for 7 days. The results also demonstrated that those with GMA show more stable ASE than the rest. This is due to the ability of the GMA to react with OH groups of the cell wall, as mentioned earlier. The same explanation goes for those with DAP, as previously mentioned.

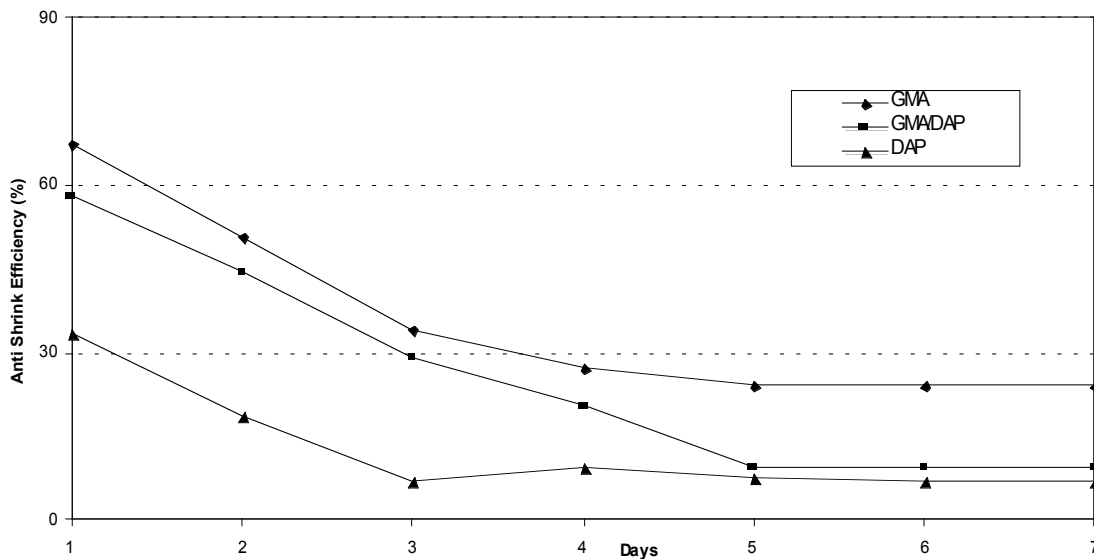


Figure13: Anti shrink efficiency of samples impregnated with various WPG at 25 mm Hg

#### **4.0 CONCLUSIONS**

As conclusion, the samples impregnated with GMA exhibited better mechanical and dimensional stability than those with DAP alone. The better mechanical and dimensional stability properties were be due to the better penetration of GMA into the cell wall and the reaction with OH groups in the wood.

#### **5.0 ACKNOWLEDGEMENT**

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