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THE EFFECT OF CHEMICAL MODIFICATION OF RICE HUSK WITH GLYCIDYL METHACRYLATE ON THE MECHANICAL AND PHYSICAL PROPERTIES OF RICE HUSK-POLYSTYRENE COMPOSITES

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ABSTRACT

Rice husks (RH) were chemically modified with glycidyl methacrylate (GMA). The chemical loading of the GMA (weight percent gain, WPG) increased as the reaction time was increased. The modification of RH with GMA gave enhanced flexural, tensile and impact properties of rice husk-polystyrene (RH-PS) composites. These may be due to the increased interaction at the interfacial region between the surface of GMA-modified RH and PS. The modification improved dimensional stability and reduced water absorption of the composites.

INTRODUCTION

Wood-plastic composites have received a lot of attention particularly on the use of lignocellulosics as a reinforcing component. The utilization of wood or other lignocellulosic materials, as a reinforcing component in polymer composites has become more attractive, particularly

in price driven/high volume applications.¹⁻¹⁰ This development has been attributed to several advantages offered by these types of filler over their inorganic counterparts, such as, lower density, greater deformability, less abrasiveness to expensive molds and mixing equipments, and lower cost.

There has been growing interests on the utilization of non-wood lignocellulosic. Various materials have been examined, to either replacing existing wood species in making conventional panel products¹¹ or producing plastics composites.¹²⁻¹⁶ The increasing trend in using these non-wood based materials has been induced by the growing demand for light weight, high performance materials coupled with diminishing natural fiber resources (wood in particular) and escalating costs of raw materials and energy.

One of the lignocellulosic materials which is of great relevance in Malaysia is the large quantity of biomass generated by the rice industries. The potential utilization of fillers derived from these industries, i.e., rice husks, or white and black rice husk ashes, for the production of thermoplastic composites have been reported by several workers.^{17,18} In general, it has been found that incorporation of fillers into thermoplastic matrix reduces both tensile and flexural strength of the resulting composites.¹⁹ Thus, it is the purpose of this work to study the effect of GMA-modification of RH on the mechanical as well as physical properties of the RH derived composites.

EXPERIMENTAL

Materials

Rice husks (RH), in the form of ground powder, was supplied by BERNAS Sdn. Bhd., Alor Setar, Kedah, Malaysia. The polystyrene used was of general purpose grade (density of 1.07 g/cm³), purchased from Keng Cheong Trading Co. Sdn. Bhd., Butterworth, Malaysia. Glycidyl methacrylate (GMA) was purchased from Fluka-Chemica.

Chemical modification of RH with GMA

RH in the form of powder (35-60 mesh (270-500 micron)) washed with acetone under reflux for 2 h. The powder was then dried in an oven at 105°C for approximately 17 h. Reaction was conducted by placing the dried RH in a GMA/Dimethylformamide (DMF) solution (1:9, volume). The mixture was heated at 90°C for several hours to obtain various levels of weight percent gain (WPG). The reacted RH was filtered, then refluxed in acetone for 3 h, to remove unreacted reagents. The GMA-modified RH was then dried in an oven at 105°C for 3 h.

Preparation of composite

The unmodified or GMA-modified RH and PS were mixed using a Haake Rheocord System which consisted of a Rheodrive 5000 drive unit and Rheomix 600 roller blade mixer. The RH, PS pellets and benzoyl peroxide (5% of RH weight) were hand-mixed prior to putting them in the

mixer. The mixing was carried out at 180°C at a rotor speed of 20 rpm for approximately 20 min. The mixture was then transferred to a mold with the dimensions of 160 X 160 X 3 mm. The mixture was preheated for 10 min at 180°C, followed by hot-pressing at the same temperature for another 10 min. Cooling was carried out for 5 min under pressure. Two levels of RH (unmodified and GMA-modified)-PS mixtures were employed, i.e., 40:60 and 60:40 by weight.

Mechanical Testing

The sheet was cut into 4 types of test samples; i.e. flexural, tensile, impact, and water immersion tests. Tensile tests were carried out according to ASTM D618 on samples with the dimensions of 15 X 1.9 X 0.3 cm (length X width X thickness), using a Universal Testing machine Model STM-10 at a cross-head speed of 5 mm/min. Flexural tests were conducted on the same machine according to ASTM D790, i.e. a three-point bending system. The samples with dimensions of 15 X 1.5 X 0.3 cm, were tested at a cross-head speed of 2.0 mm/min. The Izod impact test was carried out according to ASTM D256 on unnotched samples with dimensions of 6.5 X 1.5 X 0.3 cm, using a Zwick Impact Pendulum Tester Model 5101. A minimum of 7 samples were tested in each case.

Water absorption and thickness swelling test

Samples were immersed in distilled water at 30°C. The water

absorption were determined by weighing the specimens after immersing in water for a specific time. A Mettler balance type AJ150 was used, with a precision of 1 mg. The water absorption at any time t , M_t , was calculated by;

$$M_t = \frac{W_w - W_d}{W_d}$$

where, W_d and W_w are original dry weight and weight after exposure for a specified period, respectively. Thickness swelling (T_t) is calculated according to the formula given below;

$$T_t = \frac{T_w - T_d}{T_d}$$

where, T_d and T_w are original dry weight and weight after exposure for a specified period, respectively.

RESULTS AND DISCUSSION

Figure 1 shows the result of the reaction between GMA and RH. The results clearly shows that the chemical loading, in terms of weight percent gain (WPG), increases with longer reaction time. This indicates that more reactions occur as the reaction time is prolonged. Since the GMA-reacted RH was exhaustively washed by refluxing with acetone for 3 hours, the results indicate that GMA is chemically bonded to the RH. The bonding likely occurred through reaction of epoxide groups of GMA and RH hydroxyl groups. As shown by several studies,²⁰⁻²⁴ GMA is sufficiently

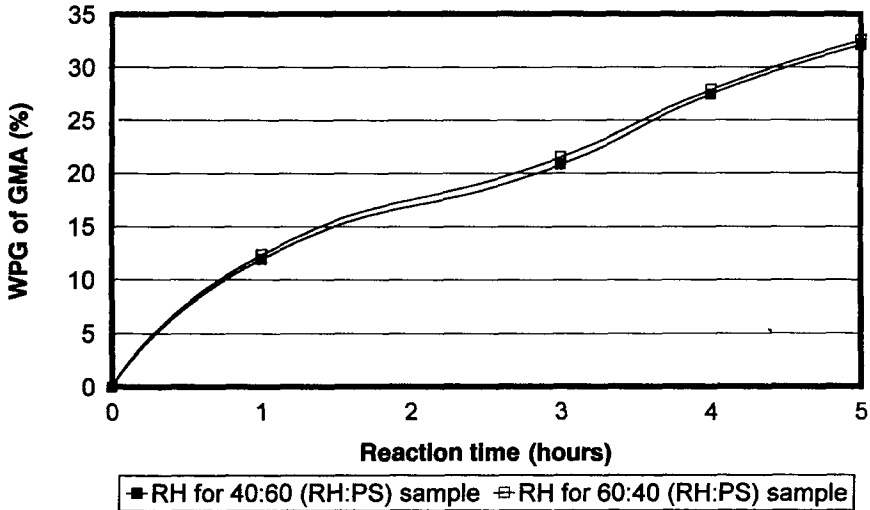


FIGURE 1 The reaction profile of GMA with RH

reactive to react with lignocellulosic hydroxyl groups through nucleophilic reaction. The conditions employed in this work favor the reaction of the epoxide groups, thus leaving the terminal C=C groups of GMA unreacted. Since, the GMA-modified RH is mixed with PS together with benzoyl peroxide during mixing, prior to forming the board samples, there is a strong possibility that further reactions could occur through this thermally labile C=C bonds. Previous studies²⁰⁻²³, which were carried out in the presence of free-radical initiator and the application of heat, showed evidence for this kind of reaction. The proposed chemical reaction of GMA and RH is shown in Figure 2.

Flexural strength of the RH-PS composites are depicted in Figure 3. It can be seen that for both composition of RH:PS, the strength increases

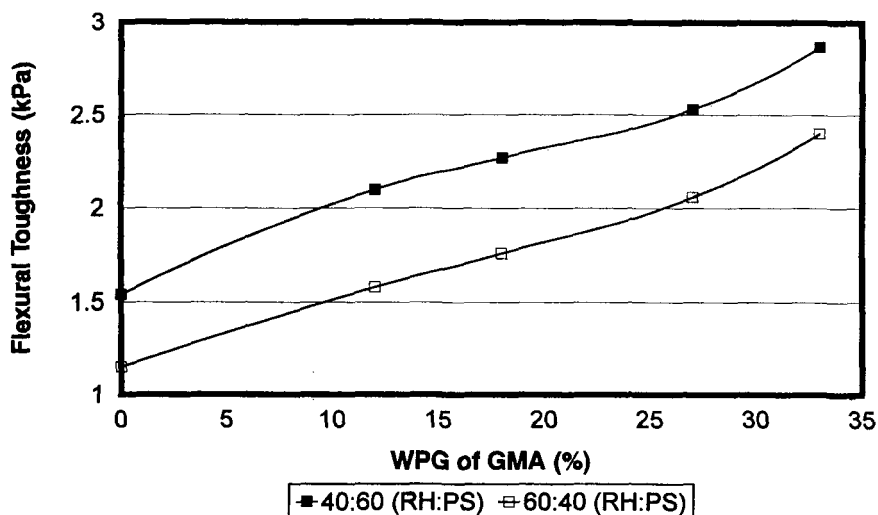


FIGURE 4 The effect of GMA modification on the flexural toughness

as the WPG of GMA is increased. The phenomena can be explained by two possibilities; (i) the increase of hydrophobicity of the RH surface by the introduction of GMA residue on the RH surface (ii) the reaction of C=C bonds through free radical reactions to form some form of polymerization or crosslinking. These two factors are believed to be predominant factors in improving the compatibility between RH and PS, in terms of producing better interaction at the interfacial region. Several studies²⁵⁻²⁶ show that increasing the hydrophobicity of the lignocellulosic surface significantly improves compatibility of the surfaces between polar lignocellulosic and non-polar polymer matrix. These arguments are further supported by the flexural toughness results of the composites (Figure 4), which show the

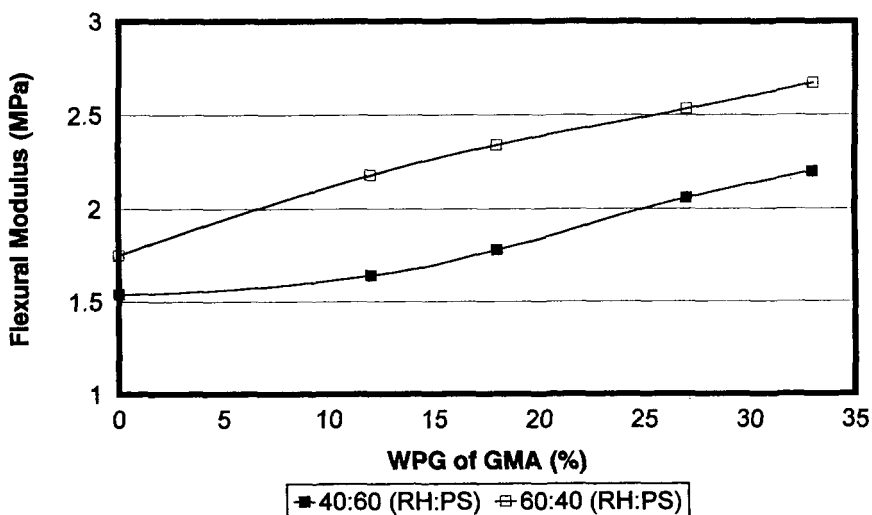


FIGURE 5 The effect of GMA modification on the flexural modulus

same trend as in the strength. The result may reflect that more energy is needed to break or bring into failure the GMA-modified sample as compared to the unmodified ones. It indicates that the stress from the PS matrix is transferred more efficiently to the GMA-modified RH than the unmodified. The strength of composites decreased with higher RH contents.

Figure 5 shows that the flexural modulus is improved as more GMA is introduced on the RH surface. Figures 6, 7 and 8 show the effect of GMA-modification on the tensile properties of the RH-PS composites. It is clearly seen that the modification has resulted in the increase in the tensile strength, toughness and modulus of the composites.

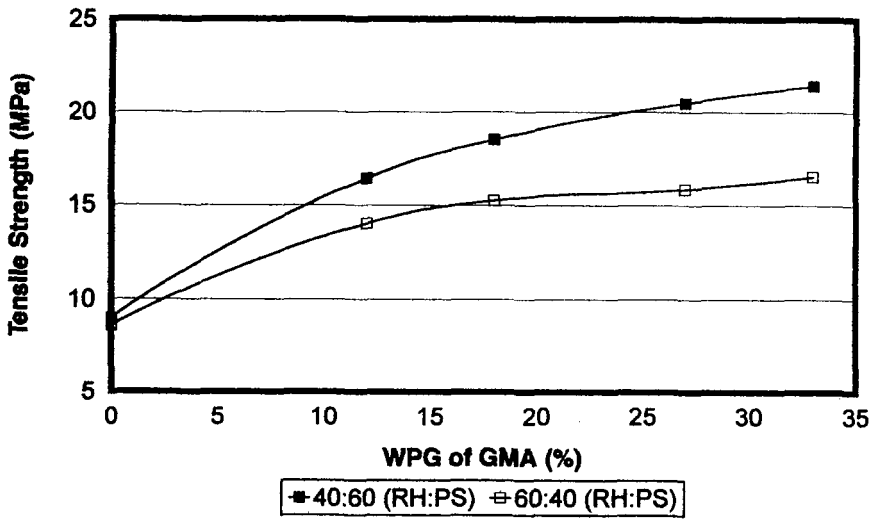


FIGURE 6 The effect of GMA modification on the tensile strength

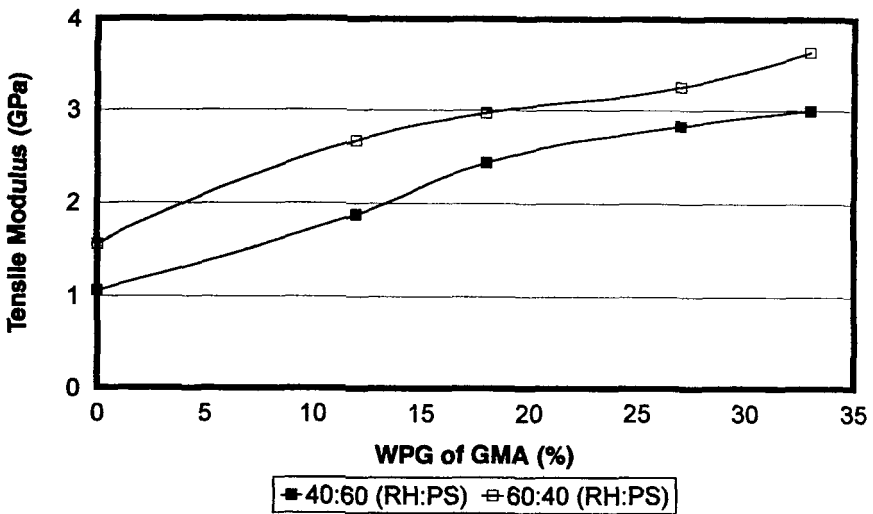


FIGURE 7 The effect of GMA modification on the tensile modulus

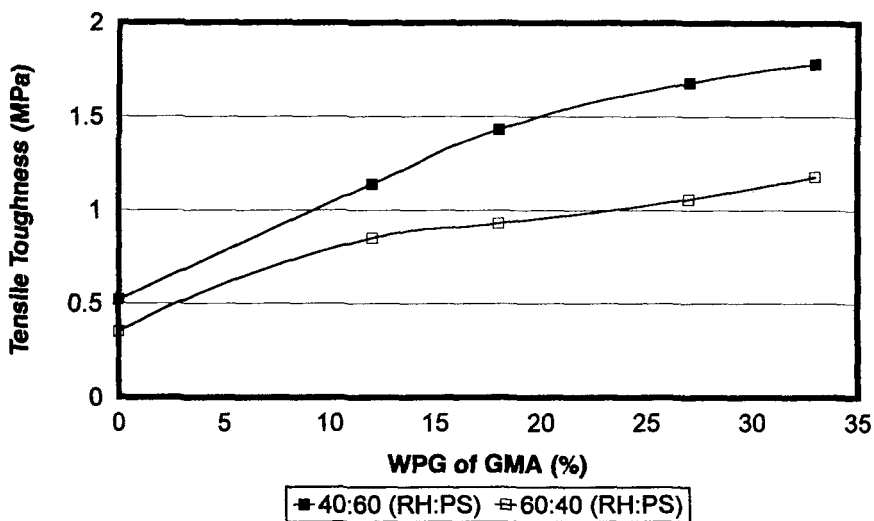


FIGURE 8 The effect of GMA modification on the tensile toughness

Impact strength also improved as the RH was modified with GMA (Figure 9). Thus, the GMA-modified RH composites exhibited an improved capability to resist crack propagation during the fracture process as compared with the unmodified RH.

Water absorption properties are depicted in Figures 10 and 11 for the first 5 h and overall 14 days water immersion, respectively. It is obvious that the unmodified composites show the highest rate of water absorption for the first hour as compared to the GMA-modified composites. The composites with greater loading of unmodified RH display higher absorption as compared to those with lower loading. This is expected, since if more unmodified RH is incorporated, more sites will be available

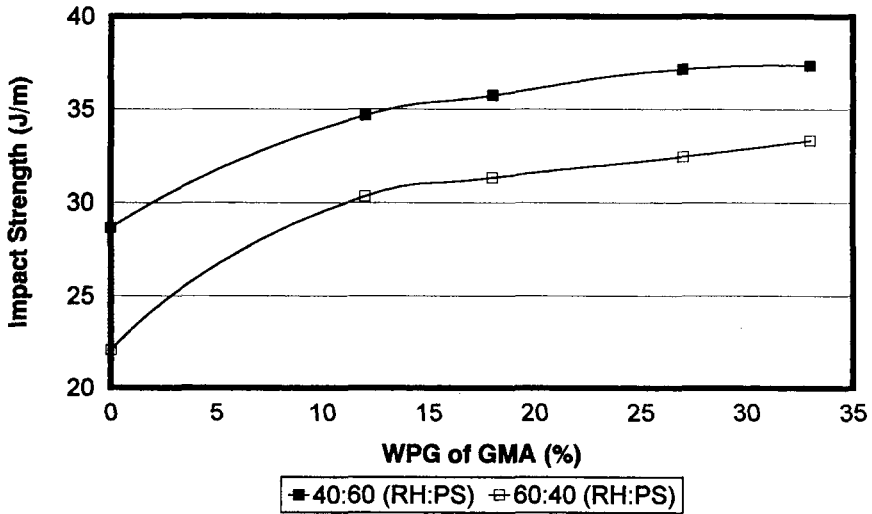


FIGURE 9 The effect of GMA modification on the impact strength

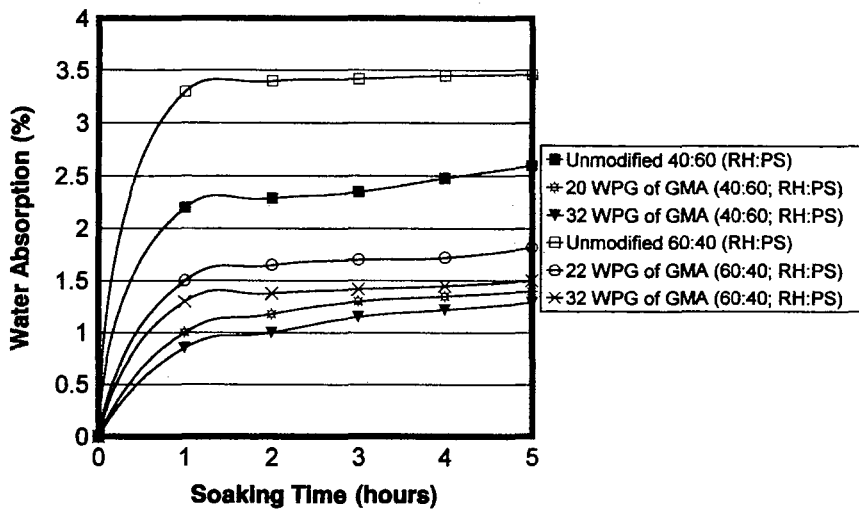


FIGURE 10 The effect of GMA modification and RH loading on the water absorption (for the first 5 hours soaking)

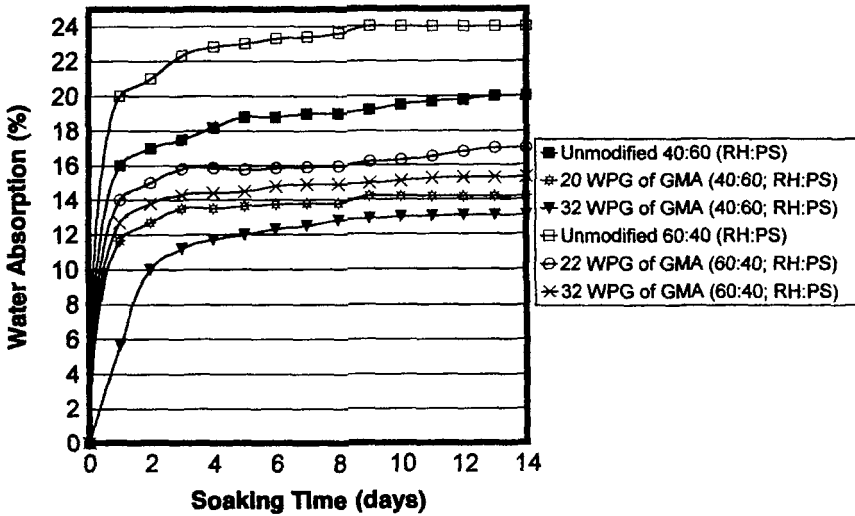


FIGURE 11 The effect of GMA modification and RH loading on the water absorption (14 days soaking)

for the hydrogen bonding to occur between hydroxyl groups of the RH and water.²⁷ It is clearly seen that the composites with GMA-modified RH have less water absorption for the first 5 hours of immersion. This is not surprising, since, modification with GMA may replace hydrophilic hydroxyl with more hydrophobic methacrylic groups.²¹⁻²³ The results also show that at the same level of GMA-modification, composites with higher loading of RH exhibit higher absorption than the lower ones. This again may be contributed by the greater availability of the hydroxyl groups of the RH. The extent of the water absorption for 14 days immersion follow the same trend as the first 5 hours soaking.

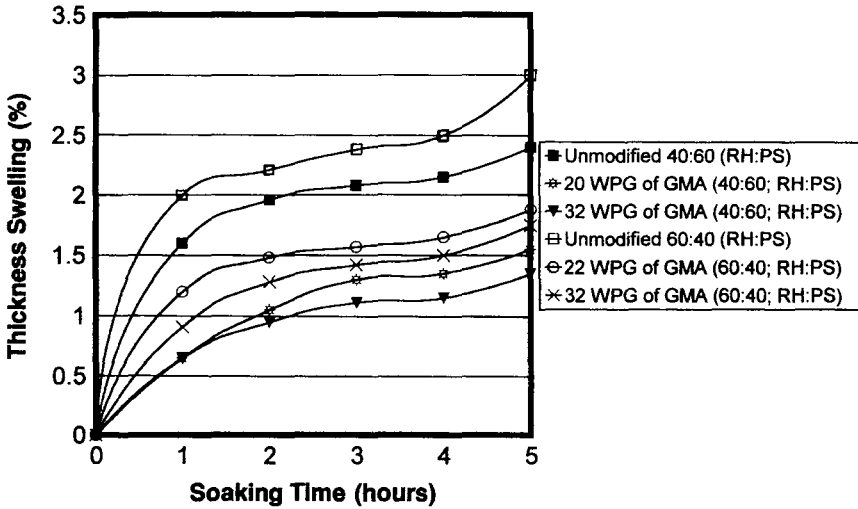


FIGURE 12 The effect of GMA modification and RH loading on the thickness swelling (for the first 5 hours soaking)

It is commonly known that any absorbed water will swell the cell wall. Thus, this phenomena will be reflected in the changes in the thickness of the sample. The thickness swelling of the composites which are shown in Figures 12 and 13, for the first 5 hours and overall 14 days water immersion, respectively, are similar in trend with the water absorption results. The composites with unmodified RH display higher swelling than the GMA-modified ones, as anticipated from the water absorption data.

CONCLUSIONS

The studies showed that rice husks reacted with glycidyl methacrylate under the conditions employed. The chemical loading of the

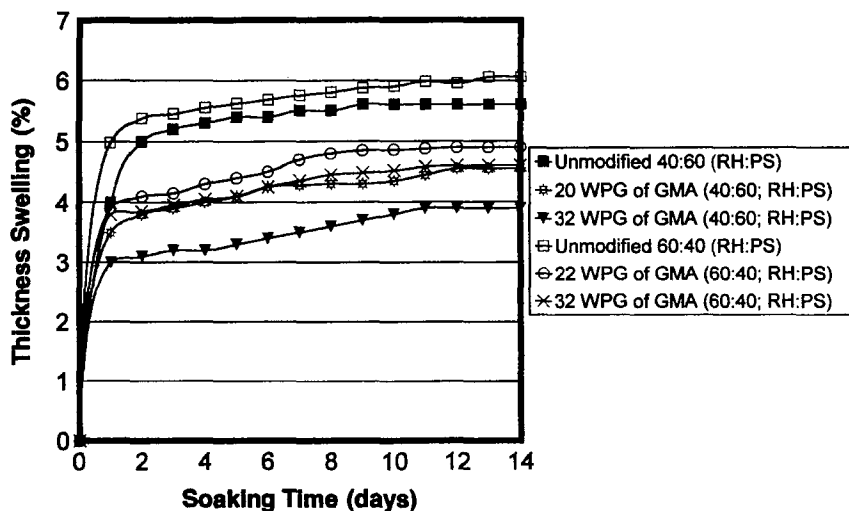


FIGURE 13 The effect of GMA modification and RH loading on the thickness swelling (14 days soaking)

GMA (weight percent gain, WPG) increased as the reaction time was increased. Chemical modification of RH with GMA enhanced the flexural, tensile and impact properties. These may be due to the increased interaction at the interfacial region between GMA-modified RH surface and PS. The modification also improved the dimensional stability of the composites.

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