

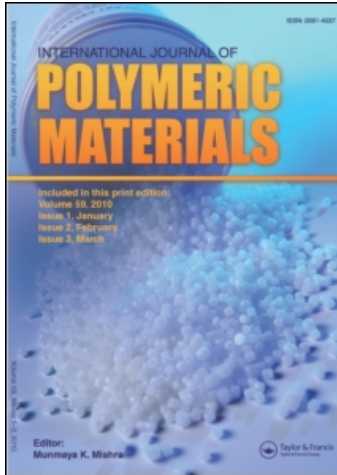
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## Moisture Absorption Behavior of Palm/Polypropylene Composites in Distilled Water and Sea Water

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*The objective of this study was to investigate the moisture absorption behavior of uncompatibilized and compatibilized palm fiber/polypropylene composites in distilled water and sea water. The amount of moisture absorbed at saturation by polypropylene during immersion in both environments increased significantly by introduction of palm fiber in the polymer and this amount was not affected by use of a coupling agent/compatibilizer (Epolene E-43). On the other hand, incorporation of the compatibilizer Epolene E-43 in the composites increased moisture diffusion rate significantly. Moisture diffusivity values obtained in distilled water and sea water were similar in the case of palm/PP composite specimens without any compatibilizer. However, the moisture diffusion rate was faster in distilled water than that in sea water in the case of using compatibilizer Epolene E-43 in the palm/PP composite.*

**Keywords:** cellulosic fiber, composite, diffusion, diffusivity, palm, polypropylene, sea, water, wood

### INTRODUCTION

Composite materials based on fibers of natural polymers, such as wood cellulose fibers, and thermoplastics recently attracted much attention due to the remarkable environmental and economical advantages. The advantages of using cellulosic fibers as reinforcements in thermoplastics

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include low densities, low cost, nonabrasive nature, possibility of high filling levels, low energy consumption, high specific properties, biodegradability, and worldwide availability [1–4]. In the building community, there is a growing demand for high-performance, low-maintenance, and low-cost building components. To meet this demand, natural fiber-thermoplastic composites are being used to produce such products as decking, window and door elements, panels, roofing, and siding. There has also been a rapid growth in the extrusion business with the use of wood-filled compounds for buildings and construction. The better provision for longevity, appearance, life-cycle cost, and value makes wood-plastics composites popular in use in the markets. Wood-plastics are resistant to insects, rotting and slipping, are attractive and paintable, are stiffer than plastics and can be used like wood. The extruded composites can eliminate sizing and calibration. This is because the stiff extrudate can hold its shape much better than unfilled plastics. Engineered wood materials, that is, wood-plastic composites, are also being investigated by the U.S. Navy for waterfront construction applications primarily because of their superior durability characteristics compared to wood [5].

Polypropylene/wood fiber composites have attracted special attention because of their wide applicability in automobile and panel manufacturing applications. These composites take advantage of the superior properties of polypropylene (PP) in comparison to other thermoplastics including easy processibility by all processing methods (molding, extrusion, film, and fiber manufacturing). In addition, polypropylene is far superior to polyethylene in terms of heat resistance and mechanical properties. Its low density makes it especially attractive in lightweight applications that require strength. Furthermore, PP composites can be used in electrical applications because of their excellent electrical properties.

There are, however, a few disadvantages of using cellulosic fibers in thermoplastics such as the high moisture absorption of the fibers [6–7], the low processing temperatures permissible [1,3,8], and the incompatibility of hydrophilic cellulose fibers and typical hydrophobic commodity thermoplastics, such as polyolefins [1,3,7–10].

The moisture absorbed by the composite and the corresponding dimensional changes can be reduced dramatically if the fibers are thoroughly encapsulated in the plastic and the adhesion between the fiber and the matrix is strong [3,7]. If necessary, moisture absorption of the fibers can be significantly reduced by the acetylation of the hydroxyl groups present in the fiber [11]. The shortcomings of the moisture absorption of the composite can also be minimized by selecting

applications where the high moisture absorption is not a major drawback. For example, polyamide and its composites absorb water, but applications are such that this deficiency is not of prime importance [3].

The processing temperature of the cellulosic fibers in thermoplastics is limited due to the potential fiber degradation at higher temperatures. This just limits their application to the plastics with low melting temperatures. However, it has been reported that no deterioration of properties is observed when processing temperatures are maintained below about 200°C [3]. It has also been noted that if the composite compositions are treated with sodium borate, boric acid, or phenolic resin, the chance of burning of the compositions during processing can be decreased [8,12].

The inherent polar and hydrophilic nature of the cellulosic fibers and the nonpolar characteristics of polyolefins create difficulties in compounding and result in inefficient composites. However, it has been shown that the use of compatibilizing and coupling agents for treating fibers prior to, or as an addition in, the compounding step enhances the compatibility and adhesion between the fibers and the matrix and the fiber dispersion in the matrix improving the mechanical properties [13–18]. The most common method for compatibilization is grafting matrix compatible components onto cellulose or grafting cellulose compatible species to the thermoplastic molecules. Grafting can be achieved using plasma treatment, ionizing radiation, or reactive chemical additives. Another compatibilizing method is the use of coupling agents, which are materials that are compatible with cellulose and matrix polymer, such as silanes and stearic acid [19].

Maleated (maleic-anhydride-modified) polypropylene has been particularly successful as a coupling agent in cellulose-polypropylene composites improving mechanical properties as a consequence of enhanced interfacial adhesion [1,3,9,20].

Rozman et al. [21] prepared polypropylene (PP) hybrid composites using oil palm empty fruit bunch and glass fibers as reinforcing agents in PP matrix. The incorporation of both fibers into PP matrix has resulted in the reduction of flexural and tensile strengths. Both flexural and tensile modulus have been improved with the increasing level of overall fiber content loading. Three types of coupling agents, that is, maleic-anhydride-modified PP (commercial name Epolene, E-43), polymethylenepolyphenyl isocyanate (PMPPIC) and 3-bi(trimethoxysilyl)-propylmethacrylate (TPM), were utilized. Overall, E-43 and TPM had imparted considerable improvements in the flexural and tensile properties. However, only slight improvements in some cases were shown for those composites treated with PMPPIC. All composites with coupling agents showed lower water absorption [22].

Palm trees are abundantly available in Saudi Arabia and polypropylene is locally produced by SABIC. In a recent study [23–24], the proponents investigated the feasibility of utilizing wood fibers obtained from waste palm tree branches in reinforcing polypropylene. They investigated the influence of processing parameters (fiber loading, degree of mixing, mixing temperature, use of compatibilizers) on characteristics of polypropylene-cellulose fiber composites. It was shown that it is feasible to produce composites with up to 40% fiber content that have strength comparable to that of the pure polymer using simple processing equipment and with minimum pretreatment of fibers.

The objective of this study was to investigate the water sorption characteristics of the palm fiber–polypropylene matrix composite and the effect of using a compatibilizer on that important material property. Water sorption is a serious problem that is expected to take place in cellulose–palm fiber composites. Diffusion of liquid in polymer composites can be visualized as a series of jumps in which Brownian motion of chain segments of the polymer produces transient voids in the vicinity of the liquid penetrant, enabling it to move within the polymer. The temperature dependence of diffusivity is described by the Arrhenius law, and the activation energy is related to the amount of energy required for the process of diffusion [25].

Assessment of material behavior in real service environment involves exposure of the material to a simulated environment for a certain period, and evaluation of the exposed specimen for changes in properties. Water- and fluid-immersion tests have been widely utilized to evaluate changes in the properties of polymeric materials [26–27]. The emphasis here was placed upon determining the moisture absorption behavior of palm fiber/polypropylene matrix composites under complete immersion in distilled water and sea water. The moisture intake of the composite was measured as a function of the specimen type and exposure time.

If diffusion is restricted to one dimension, such as is the case presented by a thin film of thickness,  $l$ , adsorbing a fluid according to Fick's law, where diffusion into the edges of the film can be ignored, the amount of diffusant,  $M_t$ , taken up by the sheet in a time,  $t$ , can be given by Reference [28]:

$$\frac{M_t}{M_\infty} = 4 \left( \frac{Dt}{l^2} \right)^{\frac{1}{2}} \left( \frac{1}{\pi^{\frac{1}{2}}} + 2 \sum_{n=0}^{\infty} (-1)^n \operatorname{ierfc} \frac{nl}{2(Dt)^{\frac{1}{2}}} \right) \quad (1)$$

The uptake is considered to be a diffusion process controlled by a constant diffusion coefficient,  $D$ , and  $M_\infty$  is the equilibrium sorption

attained theoretically after infinite time. Eq. 1, with suitable interpretation of  $M_t$ , and  $M_\infty$ , also describes desorption from the same sheet, initially conditioned to a uniform concentration, whose surface concentrations are instantaneously brought to some lower value or zero at  $t = 0$ . The value of  $D$  can be deduced from an observation of the initial gradient of a graph of  $M_t/M_\infty$  as a function of  $(t/l^2)^{1/2}$ . This observation is made easier by the fact that, for a constant diffusion coefficient, the graph for a sorption experiment is a straight line, to within the normal limits of experimental error, for  $M_t/M_\infty$  as much as about 50 percent. That is, at short times, where  $M_t/M_\infty$  is less than 0.5, Eq. 1 can be approximated by the following [28]. Recently, some doubt has been cast on this figure with 15 percent deemed more appropriate.

$$\frac{M_t}{M_\infty} = 4 \left( \frac{Dt}{\pi l^2} \right)^{\frac{1}{2}} \quad (2)$$

Although analytical methods for obtaining expressions for the sorption rate from Fick's equations are not possible when  $D$  is a function of the concentration, Eq. 2 can be used as an initial sorption law but with  $D$  substituted by  $\bar{D}$  where  $\bar{D}$  represents some kind of average diffusion coefficient. It turns out, however, that Eq. 2 holds up to higher values of  $M_t/M_\infty$  when the diffusion coefficient increases with concentration, whereas for  $D$  decreasing with increasing concentration,  $M_t/M_\infty$  is only proportional to  $t^{1/2}$  over the very initial region of the sorption [28].

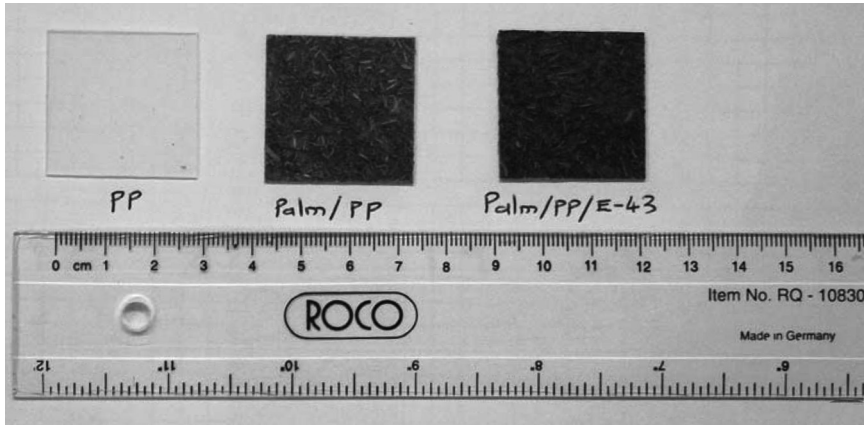
## EXPERIMENTAL

### Materials

Materials utilized in this investigation include date palm fibers, polypropylene matrix (obtained from SABIC), and the coupling agent, maleated polypropylene (Epolene E-43), from Eastman Chemicals Company.

The palm branches were initially shredded into small pieces using a mechanical shredder. The shredded fibers were sieved to remove fines and nonfibrous materials present in the branches.

Compounding of the wood fibers and thermoplastics was performed using a twin screw extruder that accomplishes the tasks of defibrillation of palm wood and mixing of cellulose with the thermoplastic. Heat was applied to keep the blend at a temperature of about 185°C. A coupling agent, maleated polypropylene (Epolene E-43) was used. It was shown to produce well compatibilized composites [23]. The extrudate was pelletized into small pellets.



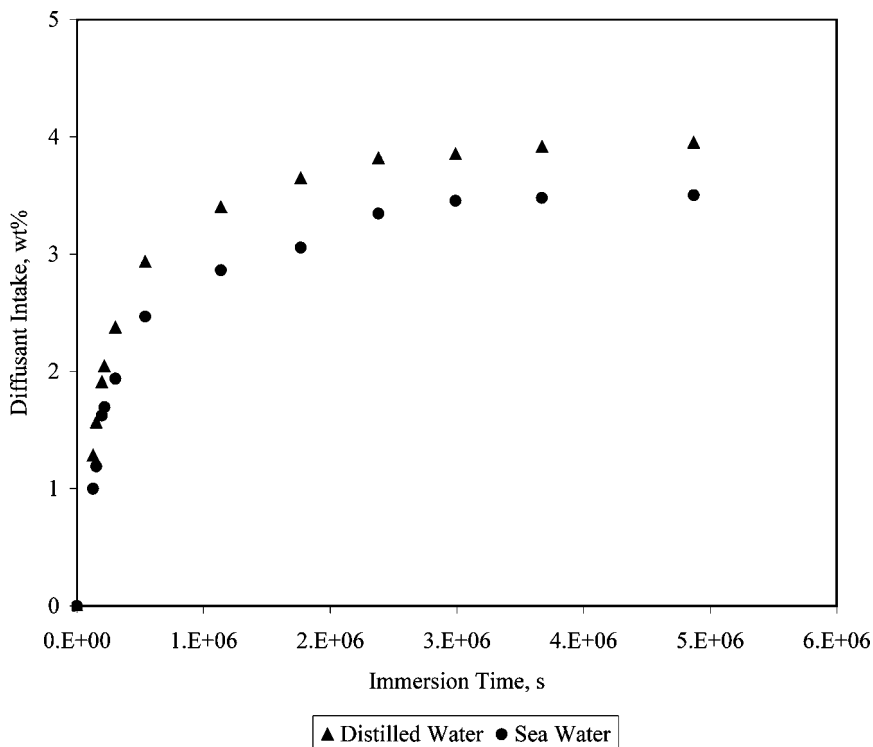
**FIGURE 1** A sample photograph of the specimens used in water diffusion study.

### Diffusion Tests

Relatively thin composite sheets (for example  $30 \times 30 \times 1$  mm) (as shown in Figure 1) for the moisture diffusion tests were molded from the prepared pellets utilizing a compression molder. Three pieces of each particular material (polypropylene, untreated palm fiber reinforced polypropylene (at 30 wt% fiber content) and compatibilized palm/polypropylene composite (at 30 wt% fiber and 4 wt% compatibilizer content)) were immersed in distilled water and sea water for several months at room temperature. At various time intervals, test specimens were removed from the test liquid, dried, and weighed by an analytical balance with good precision.

## RESULTS AND DISCUSSION

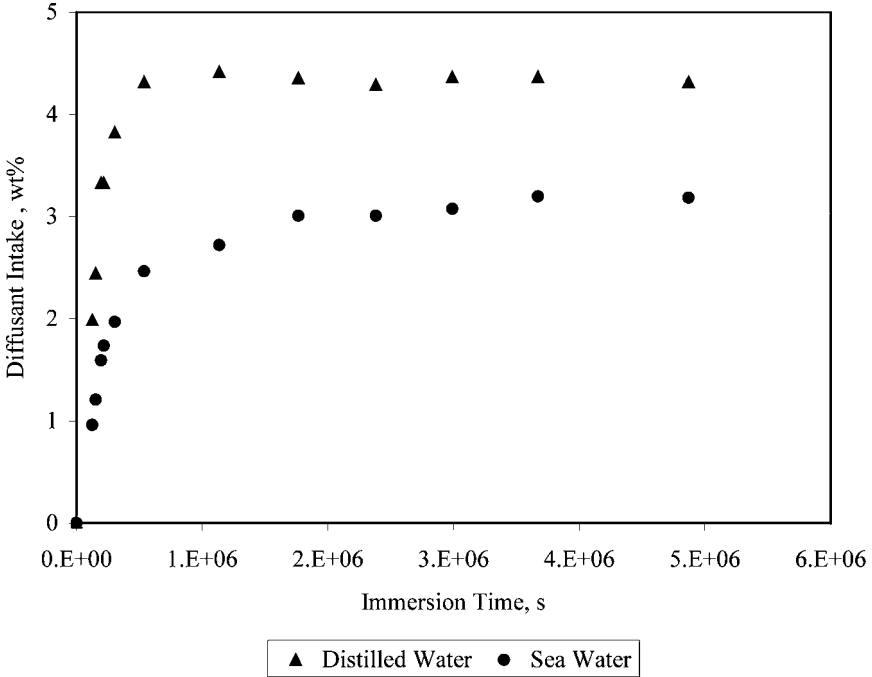
Moisture diffusion in the palm fiber/polypropylene composite specimens reached equilibrium in about  $6 \times 10^6$  s (70 days). Plots of moisture intake versus immersion time in distilled water and sea water for untreated palm fiber reinforced polypropylene (at 30 wt% fiber content) and compatibilized palm/polypropylene composite (at 30 wt% fiber and 4 wt% compatibilizer content) are presented in Figures 2 and 3. The amount of moisture intake by polypropylene without any fiber content was negligible and the experimental data were not significant enough to include in the figures.



**FIGURE 2** Diffusant intake (relative to the original weight of the specimen) vs. immersion time in distilled water and sea water for untreated palm fiber/polypropylene composite specimen.

The amount of moisture absorbed at saturation by polypropylene increased significantly (from about zero to over 3 wt%) by introduction of palm fiber in it. It took about  $6 \times 10^6$  s for the composite specimens to become saturated with moisture. Comparison of the two figures indicates that the moisture intake at saturation does not decrease by use of the compatibilizer Epolene E-43 in palm/PP composites. This result appears to contradict the belief that use of a compatibilizer would improve the moisture absorption resistance of the cellulosic fiber reinforced polymers. The explanation might be that during immersion in distilled water or sea water, moisture would be absorbed mainly by the wood fibers in the composite and the presence of a little amount of compatibilizer would not affect the maximum moisture intake in the composite significantly as long as the fiber content is kept unchanged.

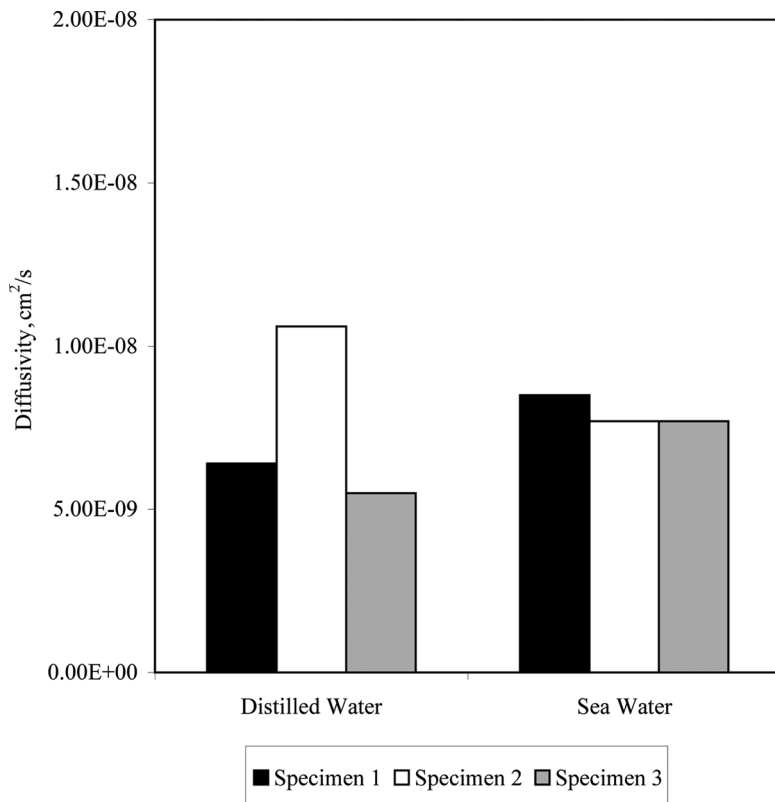




**FIGURE 3** Diffusant intake (relative to the original weight of the specimen) vs. immersion time in distilled water and sea water for compatibilized palm fiber/polypropylene composite specimen.

The compatibilizer is, however, expected to affect the moisture diffusion rate into the composite significantly because the moisture gets into the fibers through the fiber–matrix interphase, which is significantly altered by the compatibilizer. To determine this effect, diffusivities of moisture in palm fiber reinforced polypropylene specimens (with and without compatibilizer) in distilled water and sea water were determined by use of Eq. 2, that is  $M_t/M_\infty$  plotted against  $4(t/\pi l^2)^{1/2}$  and the diffusivity is determined from the initial slope of the plot (slope is  $D^{1/2}$ ). The diffusivity values determined are presented in Figures 4 and 5 for untreated and compatibilized palm/PP composite specimens in distilled water and sea water.

As seen in the figures, moisture diffusivity into the palm/PP composite in distilled water and sea water increased significantly by use of the compatibilizer Epolene E-43 (from an average value of  $7.5 \times 10^{-9} \text{ cm}^2/\text{s}$  to an average value of  $1.82 \times 10^{-8} \text{ cm}^2/\text{s}$  in distilled water and from an average value of  $8.0 \times 10^{-9} \text{ cm}^2/\text{s}$  to an average



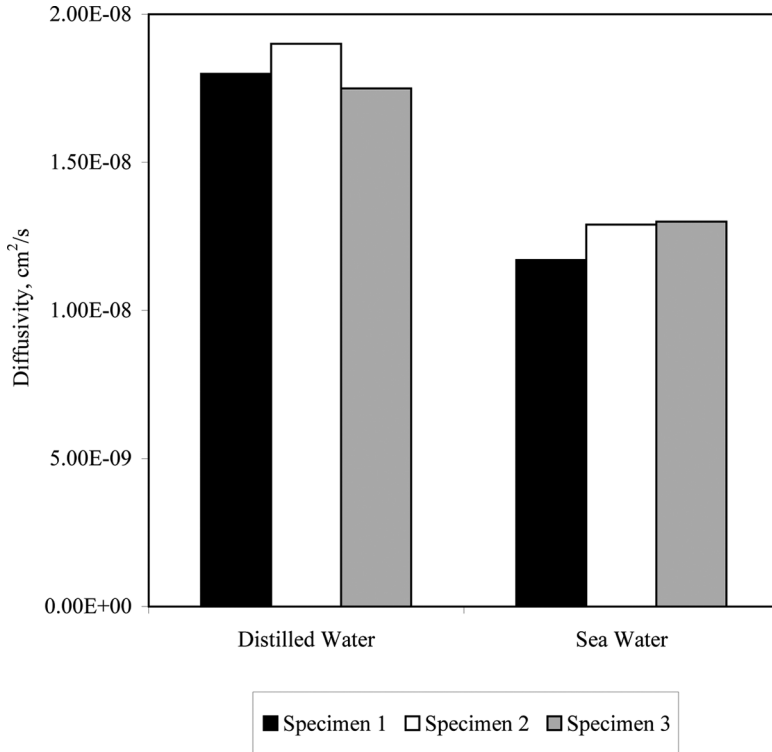
**FIGURE 4** Mass diffusivity in distilled water and sea water for palm/PP composite with no compatibilizer.

value of  $1.25 \times 10^{-8} \text{ cm}^2/\text{s}$  in sea water). Altering of the fiber–matrix interphase by the compatibilizer may have decreased its moisture resistance, instead of increasing it.

Diffusivity values were similar in distilled water and sea water for the untreated palm/PP composite. However, the moisture diffusion rate was about 50% higher in distilled water than that in sea water for the compatibilized palm/PP composite. High concentration of salts in the sea water, compared to distilled water, might be the reason for lower mass diffusivity.

## CONCLUSIONS

The results of this investigation disprove the belief that use of a compatibilizer would improve the moisture absorption resistance of the



**FIGURE 5** Mass diffusivity in distilled water and sea water for palm/PP composite with compatibilizer Epolene E-43.

cellulosic fiber-reinforced polymers. Use of Epolene E-43 as the compatibilizer in the palm/PP composite did not have any significant effect on the amount of moisture intake into the composite at saturation in distilled water and sea water. Moreover, the compatibilizer increased the moisture diffusion rate into the palm/PP composite in both environments, whereas the increase being less in sea water than that in distilled water.

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