Study of the Water Uptake and Internal Defects of Jute-Reinforced Polymer Composites with a Digital Neutron Radiography Technique

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ABSTRACT: The water-uptake behavior of jute fabric and its composites with polycarbonate and polypropylene was monitor by a digital neutron radiography technique. The thermal neutron radiography facility of neutron transmission radiography (NEUTRA) at spallation source (SINQ) of the Paul Scherrer Institute was used for this work. The internal defects, such as voids, cracks, and inhomogeneity, of the composites were studied. The water-uptake behavior of the jute fiber was also studied with a digital neutron radiography technique. The natural jute fiber showed higher water absorption than the composites. No voids or inclusions in these composites were observed. Moreover, both the jute and polymer were uniformly distributed and well mixed with the polymer matrix. © 2007 Wiley Periodicals, Inc. J Appl Polym Sci 105: 1958–1963, 2007

Key words: adsorption; reinforcement; composite

INTRODUCTION

The Paul Scherrer Institute operates the strongest continuously running spallation neutron source (SINQ).[1] The main features and properties of the facility were described by Lehmann and Pleinert.[2] The thermal neutron radiography facility NEUTRA[3] has been designed with a large diameter at the exit of the neutron flight tube (40 cm), which allows the inspection of large objects from industry and all other sectors. Usually, the option for neutron radiography inspection is chosen in cases for which X-ray or other nondestructive testing methods have failed. The utilization of a charge-coupled device (CCD) camera has other advantages, such as a low acquisition time, high frame rates, reproducibility, and a high dynamic range. This detector is applied to investigations of time-dependent processes, distribution analysis, and quantitative studies.[4] One of the main features of this method is the direct availability of the investigated results in a digitized form as matrix $\mathbf{P}(x,y)$. Each value of $\mathbf{P}(x_i,y_j)$ corresponds to the time- and energy-averaged reaction rate in the primary detector induced by the incident radiation. Some important properties of this method are introduced in ref. 5. Many complex structures, such as tubes, car doors, interior paneling, and sandwich plates, are made of composites.[6]

The radiography station is installed inside a concrete bunker guaranteeing satisfactory shielding conditions and adequate space for the detector and sample positioning. The spallation neutron source (SINQ) possesses some important advantageous properties, such as a low γ background, a large beam size (100–400 cm^2) and very flat beam profile, a high length/diameter ratio, a reasonable neutron flux level[5] ($\approx 3 \times 10^6$ cm^{-2} s^{-1}), good linearity, and good reproducibility, which are very useful for its future utilization. The characteristics of the NEUTRA facility are described in ref. 7.

SINQ is driven by a 590-MeV proton beam[1] generated by a cyclotron with a beam current of about 850 μA. To obtain a 16-bit conventional radiographic image, one has to integrate the scintillator light on the CCD chip. The exact exposure time, of course, is determined by the sample thickness, the sample materials, and the neutron flux provided from the source. Additionally, the readout time, depending on the dynamic range and the pixel number of the CCD, puts a lower limit on the highest reachable frame rate.[8]

Because of increasing environmental consciousness and the demands of traditional composite structures, which are usually made of glass, carbon, or aramid fibers embedded in epoxy, unsaturated polyester resins and phenolics are considered critical. Through the embedding of natural reinforcing

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fibers, such as jute, flax, hemp, and ramie, into a biopolymeric matrix made of derivatives from cellulose, starch, lactic acid, and so forth, new fiber-reinforced materials called biocomposites have been created and are still being developed.[9,10] As far as the mechanical properties are concerned, biocomposites are comparable to the well-known glass-fiber-reinforced plastics. One of the most important drawbacks of cellulose fibers is that they are hydrophilic in nature. The elementary unit of a cellulose macromolecule is a hydro-D-glucose, which contains three hydroxyl (—OH) groups. These hydroxyl groups form intramolecular hydrogen bonds inside the molecule and intermolecular hydrogen bonds with other cellulose molecules as well as hydroxyl groups of moist air. Therefore, wood is hydrophilic in nature, and its moisture certainly reaches up to 12–13%.[11] This is the most important drawback of a natural fiber for composite fabrication. The groupings of long-chain cellulose molecules in the cell wall contain crystalline and amorphous regions. In the crystalline regions, it is believed that —OH groups of adjacent cellulose molecules are mutually bonded or crosslinked. Therefore, there are no sites to hold water within the crystalline regions. Within the amorphous or disordered regions, however, the hydroxyl groups are accessible for the absorption of water.[12] Natural-fiber composites combine good mechanical properties with a low specific mass. However, their high level of moisture absorption, poor wettability, and insufficient adhesion between untreated fibers and the polymer matrix lead to debonding with age.[13] That said, moisture absorption can have some secondary benefits, such as reducing static electricity in the final plastic object. The water absorption and specific gravity of the lignocellulose fiber composite are important characteristics that determine the applications of these materials. Water absorption could lead to a decrease in some of the properties and should be considered when applications are selected. To improve the properties of the composites, the natural reinforcing fibers can be modified by physical and chemical methods. The surface of jute fiber is one of the best instances of hydrophilic behavior induced by the predominance of —OH groups. Recently, the physical and mechanical properties of jute fabric/Biopol composites have also been reported.[14,15] The water absorption behavior of a wood plastic composite was also studied by Khan et al.[16] The aim of this work was to study the water absorption behavior and internal defects of jute-reinforced polymer composites [e.g., polycarbonate (PC) and polypropylene (PP) composites] with a digital neutron radiography technique, and they were also studied with the neutron attenuation coefficients of these composites.

EXPERIMENTAL

Materials

PC (8040, FDA-grade) sheets of different thicknesses and PP films were purchased from GE Co. (United States) and Vestolen GmbH (Germany), respectively. The coupling agents 2-hydroxethyl methacrylate and 2-ethyl hexylacrylate were obtained from Merck (Germany). Dicumyl peroxide (Merck) was used as a thermal catalyst. Bleached Hessian cloth (HC) was supplied by the Bangladesh Jute Research Institute (Dhaka, Bangladesh). The sizes of the PC and PP composites were 88 × 80 × 1.6 mm^3 and 79 × 59 × 2.3 mm^3, respectively. Both specimens were rectangular.

Methods

Surface treatment of jute

HC was immersed in a solution of 3% 2-hydroxethyl methacrylate and 1% dicumyl peroxide in methanol for 5 min. Then, HC was dried at the ambient temperature.

Composite fabrication

The composites were prepared through compression molding, three layers of jute fabric/HC being sandwiched between four layers of PC or PP sheets. These layers of jute fabric and PC or PP sheets were pressed in a Carver model 2518 laboratory press at 5 tons and 180°C, and the pressing continued for 5 min. The jute concentration in the composites was about 32%.

Obtained images

The thermal neutron radiography facility at SINQ of the Paul Scherrer Institute was used for this work. Highly light-sensitive Andor DV434-BV CCD camera detectors (cooled at −45° by a peltier element in most cases), looking onto the weak light emission from a neutron sensitive scintillator [Li_6F/ZnS (Ag)], were usually used in the experiments.

The PC and PP composite samples were dried at 75°C in an oven. Neutron radiographs (Fig. 1) for the dry samples were then taken at an exposure time of 50 s. No measurable additional activity was induced in the samples by this short neutron bombardment. Then, these composites were fully immersed in water, and a series of radiographs (Fig. 1) of the wet samples were taken for 2, 6, 12, 20, and 30 h of water absorption. Before the radiographs were taken, these samples were waved with soft tissue paper to remove the additional water, and then their weights were measured by a six-digit elec-

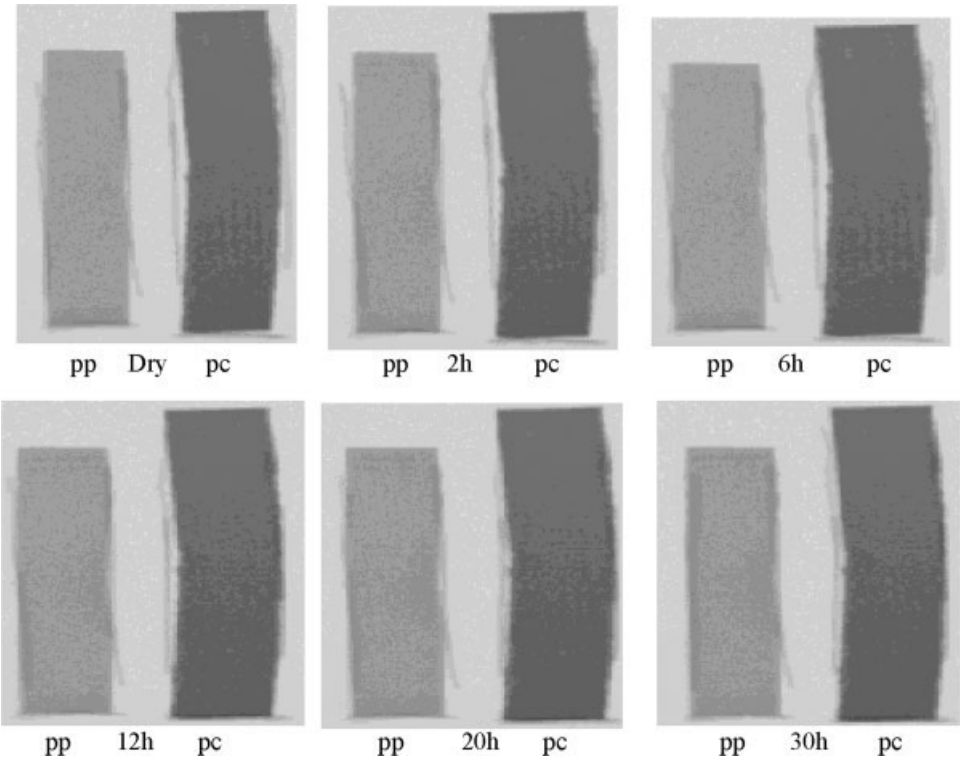

Figure 1 Radiographs of the dry and wet PC and PP composites.

tronic balance. These samples were placed on an aluminum plate with the help of aluminum adhesive tape. These samples were put in almost the same position after they were taken off the sample holder for wetting and for measuring their weight. The aim was to put each sample in exactly the same place in the field of view of the CCD camera. At each time step (2, 6, 12, 20, and 30 h), the ratio images of the wet radiograph to the dry radiograph for each pixel (image point) were calculated.

On the other hand, to study the water-uptake/absorption behavior of jute fiber, it was fixed on a thin aluminum sheet by aluminum adhesive tape. Then, it was put into an aluminum container. On the front side of the container, a thick cadmium sheet was fixed to reduce the neutron scattering from the water in the container. At first, the radiograph (Fig. 2) of the jute fiber was taken under the dry condition. Thereafter, the container was filled with water, and the radiographs (Fig. 2) of the water uptake of the jute fiber were recorded by a CCD camera based digital neutron radiography technique at different water absorption times. In both the dry and wet conditions, the exposure time was 50 s (ca. 3-s readout times for the CCD camera). In that case, the water absorption times were 1, 5, 10, 20, 40, and 60 min.

During the experiment, the neutron transmission through a sample was recorded in a neutron radiographic measurement as a two-dimensional digital image. Here the digital images consisted of square pixels of 0.067 mm. Additionally, the information from an image of the open beam profile recorded in the absence of the sample was used. This open beam data were used as input for signal transfer functions,

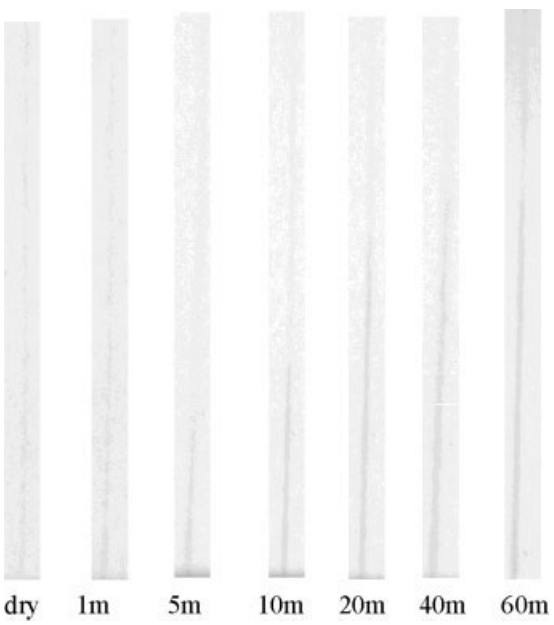

Figure 2 Radiographs of the dry and wet jute fibers for different water absorption times (1, 5, 10, 20, 40, and 60 min).

which transformed it into output that simulated the image recorded with the sample.

RESULTS AND DISCUSSION

Dry samples

For the investigation of digital images, secondary effects disturbing the interpretation of the measurements (multiple scattering and fission neutrons) can be avoided when the distance between the samples and the detector plane is more than 10 cm. Image Pro-Plus image software was used for image processing, quantification of the image data, and automated pattern recognition. The digital image data represent the attenuation of the thermal neutron beam due to the jute fiber and jute-reinforced polymer composites. The attenuation of the thermal neutron beam emerging from the neutron source is mainly due to scattering and absorption interactions of the neutrons with the atomic nuclei of the sample. The attenuated response for dry samples can be written as follows:[17]

$$I = I_0 \exp(-\mu_d x_d) \quad (1)$$

where I is the intensity of the attenuated neutron beam, I_0 is the intensity of the incident neutron beam, μ_d is the linear neutron attenuation coefficient of the sample, and x_d is the thickness of the sample. The exponential law of neutron beam attenuation neglects the contribution of scattered neutrons recorded by the neutron area detector.

The composite samples were made by jute embedded in plastic materials. Figure 3 shows the linear neutron attenuation coefficient values of the PC and PP composites. This value was obtained by the solution of eq. (1) for μ_d. It becomes obvious that one composite sample has less neutron attenuation than the other composite. This effect can be explained by the contents of the samples achieved during the manufacturing process. It also shows that the value of the neutron attenuation of the PP composite was larger than that of the PC composite. With the expansion of the small region of the neutron radiograph (Fig. 1, dry), no voids, cracks, or cavities were found. Figure 4 shows the neutron transmission profiles at different positions of the radiographic image of the composites (dry condition). These profile data were measured with the help of Image Pro-Plus software by the drawing of the horizontal line profile on the radiographic image of the composites. From these transmission profiles, it was observed that the neutron transmissions at all points of each horizontal position were equal, and no discontinuity of the neutron transmission through these composites was observed. Hence, it is concluded that the constituents of these composites were uniformly distributed and reinforced polymer composites were well manufactured.

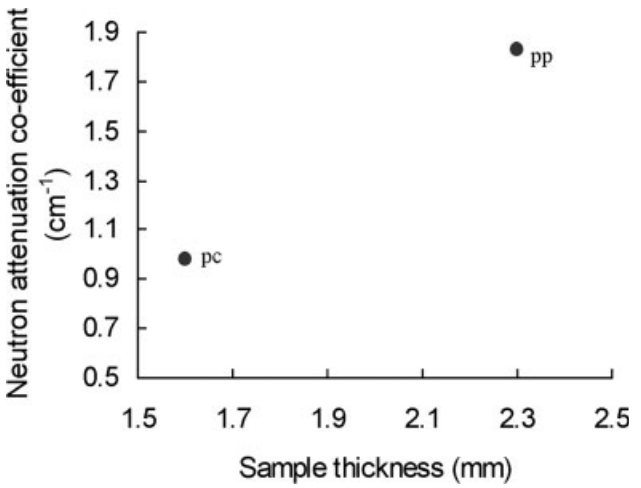

Figure 3 Neutron attenuation coefficients for the PC and PP composites.

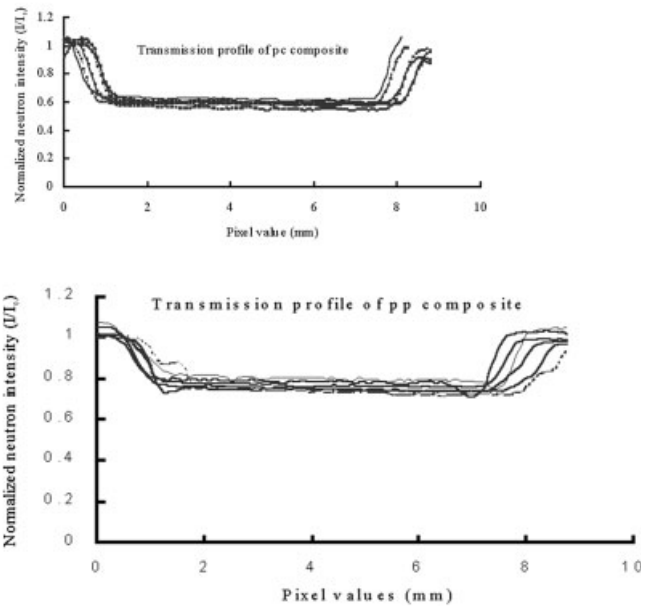

Figure 4 Neutron transmission profiles for the PC and PP composites.

Wet samples

For the quantitative analysis of the water absorption, the area of interest (AOI) was considered on each sample image, and this excluded the pixels of the composites in the edge region. The AOI was about 95% of the original images of these composites. The water absorption inside the AOI through the edge of these samples was measured with the help of eq. (1)

and the following equations:

$$\text{Dry condition: } I_d = I_0 \exp(-\mu_d x_d) \quad (2)$$

$$\text{Wet condition: } I_w = I_0 \exp[-(\mu_d x_d + \mu_w x_w)] \quad (3)$$

$$\text{Ratio of eqs. (2) and (3) : } I_w/I_d = \exp(-\mu_w x_w) \quad (4)$$

where I_d is the intensity of the attenuated neutron beam of the dry sample, I_w is the intensity of the attenuated neutron beam of the wet sample, μ_w is the linear neutron attenuation coefficient for the wet sample, and x_w is the absorbed water. Therefore,

$$\text{Total absorption of water inside the AOI} = \Big(\sum_{\text{all pixels}} \mu_w x_w\Big)/(\text{Number of pixels inside the AOI}) \quad (5)$$

The total absorption of water inside AOI can be found by the solution of eq. (5). On the other hand, the measured values (Table I, column 4) for the whole area of the composites were obtained on a dry weight basis with a six-digit electronic balance and the following formula: Water absorption (%) = [(Weight of the wet sample − Weight of the dry sample)/Weight of the dry sample] × 100. The calculated values (Table I, column 3) of the AOI were obtained with eq. (5). Figure 5 shows the net water absorption inside the AOI of the samples. At each time step, the amount of water absorption in the AOI by the PC composite was much smaller than that by the PP composite (Table I, column 3). Figure 6 presents the relation between the absorption of water and the water absorption time of the composites. The extent of the absorption of water by the PC composite was constant during 2–20 h of immersion, but after 20 h, some water absorption occurred. On the other hand, for the PP composite, water absorption increased slowly with increasing time from the beginning, and after a certain time, it became saturated. Figure 7 shows the water-uptake behavior of HC at different time steps, such as 1, 5, 10, 20, 40, and 60 min. Figures 6 and 7 show that the absorption of water of these composites was very small compared with that of the natural jute fiber/HC. This minor water absorption took place through the edges of these composites, at which the fiber ends were present as natural fibers. The absorption of moisture by the fiber was minimized in the composites because of encapsulation by the polymer and good fiber–matrix bonding. Good adhesion decreases the rate and amount of water absorbed in the interface region of a composite. It is difficult to entirely eliminate the absorption of moisture without the use of expensive surface barriers on the composite sur-

TABLE I
Water Absorption of HC and Reinforced Composites (PC and PP) with Time

Composite	Water absorption time	Calculated water absorption in the AOI (g)	Measured water absorption in the entire composite (%)
PC	2 h	0.0027	1.1764
	6 h	0.0041	1.1764
	12 h	0.0046	1.1764
	20 h	0.0081	1.1764
	30 h	0.0094	1.7647
PP	2 h	0.0087	1.6129
	6 h	0.0149	2.4193
	12 h	0.0265	3.2258
	20 h	0.0275	4.4354
	30 h	0.0304	4.4354
HC	1 min	0.15	80
	5 min	0.27	137
	10 min	0.39	170
	20 min	0.43	177
	40 min	0.44	177
	60 min	0.46	177

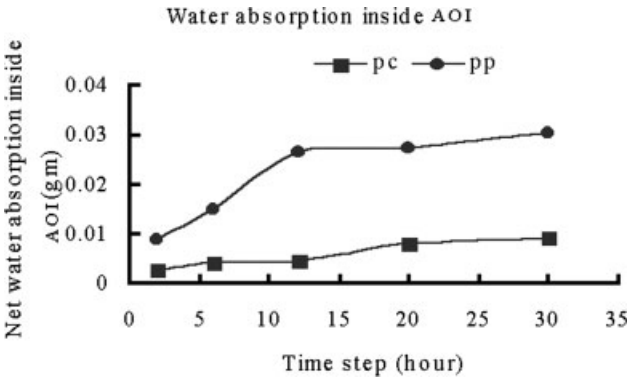

Figure 5 Net water absorption inside the AOI of the PC and PP composites.

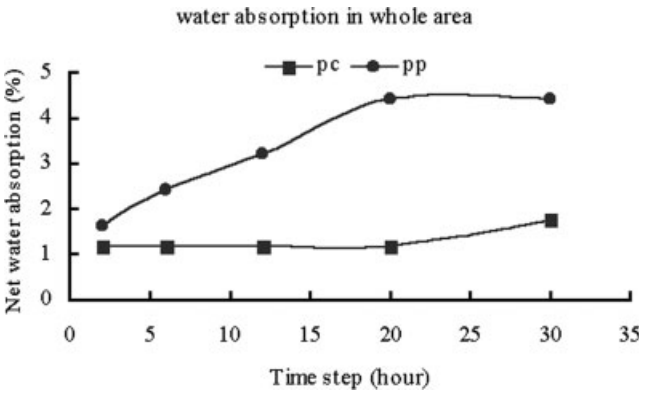

Figure 6 Absorption of water by the entire volume of the PC and PP composites at different water absorption times (2, 6, 12, 20, and 30 h).

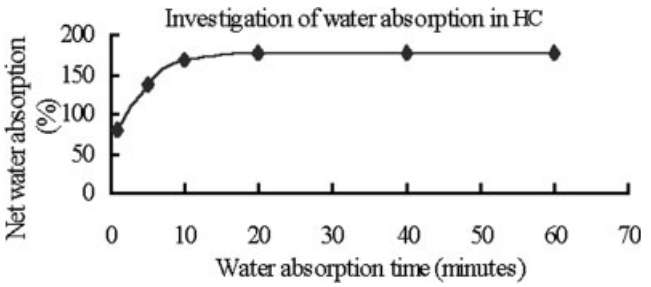

Figure 7 Water-uptake behavior of HC at different times (1, 5, 10, 20, 40, and 60 min).

face. On the basis of this investigation, it has been concluded that the quality of these composites is very good for protecting against water absorption versus natural jute fiber/HC. The small amount of water absorbed into the composites through the edges can be investigated only by a digital neutron radiography technique directly. It has also been concluded that no internal defects, such as voids, cracks, or inhomogeneity, of the composite contents can be observed, even though all defects, which are hidden in the composite, can be found easily with this method.

CONCLUSIONS

It is a more sophisticated, fundamental, and informative technique as well as conclusive method by which one can easily calculate the minor water absorption in a resulting sample versus other conventional methods. It is also concluded that water absorption in the composite is much less than that of natural jute fiber/HC, and no internal defects or inhomogeneity of the composite materials has been observed in the composite.

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