RESEARCH ARTICLE

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Banana fiber-reinforced biodegradable soy protein composites

Abstract Banana fiber, a waste product of banana cultivation, has been used to prepare banana fiber reinforced soy protein composites. Alkali modified banana fibers were characterized in terms of density, denier and crystallinity index. Fourier transformed infrared spectroscopy (FTIR), scanning electron microscopy (SEM) and thermogravimetric analysis (TGA) were also performed on the fibers. Soy protein composites were prepared by incorporating different volume fractions of alkali-treated and untreated fibers into soy protein isolate (SPI) with different amounts of glycerol (25%–50%) as plasticizer. Composites thus prepared were characterized in terms of mechanical properties, SEM and water resistance. The results indicate that at 0.3 volume fraction, tensile strength and modulus of alkali treated fiber reinforced soy protein composites increased to 82% and 963%, respectively, compared to soy protein film without fibers. Water resistance of the composites increased significantly with the addition of glutaraldehyde which acts as cross-linking agent. Biodegradability of the composites has also been tested in the contaminated environment and the composites were found to be 100% biodegradable.

Keywords Banana fibers, soy protein, composites, glycerol, water uptake

1 Introduction

The interest in using plant fibers in the preparation of polymer composites has increased substantially in the last dec-

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ade. The reason for this is that many natural fibers compete very well with glass fibers on specific strength and stiffness. This could be a sustainable alternative in many applications [1]. Plant fibers are renewable, biodegradable [2,3] and have low density. These fibers contain mainly α-cellulose, hemicellulose, lignin and some minor constituents such as fats, waxes, inorganic matters, etc. Considerable work has been done in the last decade to develop composites using natural fibers as reinforcement and synthetic thermosetting/thermoplastic resins [4–7] as matrices. Most of the synthetic thermosetting/thermoplastic resins are hydrophobic in nature and natural fibers are hydrophilic in nature and the composites fabricated thus far have poor interfacial properties. Several researchers have tried to increase the adhesion between fibers and matrix by chemical [8,9] and fungal [10] modification of fibers.

Soy protein isolate (SPI) is one of the natural matrices of interest because it is cheap, renewable, easily available, hydrophilic and is expected to have good compatibility with natural fibers. Adhesive and plastics based on soy protein [11,12] and natural fibers such as sisal-reinforced soy protein composites have been extensively investigated [13,14]. About 150% enhancement in tensile strength and modulus was observed for jute yarn-Biopol® composites prepared by the hot-press molding technique [15]. Otaigbe et al. [16], Paetau et al. [17], Liang et al. [18], Lodha et al [19] and Liu et al. [20] have used SPI as resin to make green composites using glycerol as a plasticizer. In recent years, hemp fibers have also been tested as reinforcement agents in bio-composites [21,22].

However, fewer reports are available on the use of banana fibers as reinforcing material for composites. It is to be noted that banana fiber is a waste product of banana cultivation. Banana fibers were chemically modified by alkali and characterized by density, denier, Fourier transformed infrared spectra (FTIR), scanning electron microscopy (SEM), thermogravimetric analysis (TGA) and wide angle X-ray diffraction (WXRD). Banana fibers reinforced composites were prepared using SPI as the matrix. Mechanical properties of the composites thus formed were evaluated as a function of fiber content and matrix nature, that is, plasticized soy protein. A study on the morphology, biodegradation and water resistance

of the banana fiber reinforced soy protein composites was also carried out.

2   Experimental

2.1   Materials and methods

SPI Supro 670 procured from Du Pont, India containing 92.5% protein on dry basis was used as protein source for preparing composites. Modified banana fibers were obtained from the Regional Research Laboratory, Jorhat, India. Glycerol, toluene, petroleum ether, sodium hydroxide and glutaraldehyde were all from Qualigens (India) and were used as received. Strains of *Aspergillus niger* (ATCC No. 2104) (filamentous fungi) were used for the microbial growth study on the fiber reinforced soy protein composites.

2.2   Separation and modification of fibers

A bundle of banana fibers was completely dipped in toluene for 24 h followed by washing with petroleum ether and then drying at 80°C for 24 h. Modification of the fibers was carried out by immersing the fibers in 5% or 10% NaOH solution at room temperature followed by washing with distilled water until neutral and then drying in an oven at 80°C for 24 h. The alkali modified fibers are designated as S5-BF and S10-BF where S5 and S10 represent fibers modified using 5% and 10% sodium hydroxide, respectively. BF stands for banana fiber. The unmodified fibers are represented as BF only.

2.3   Preparation of composites

Soy protein composites were prepared in two stages. In the first stage, 1.5 g of glycerol was mixed for about 1 h with 3.0 g of SPI in 30 mL water having pH of 9.0–9.5 using a magnetic stirrer. Different volume fractions of fibers of 0.5 cm length were then added to SPI/glycerol mixture. The fibers and the matrix (SPI/glycerol mixture) were mixed to coat the fibers uniformly. The resulting mixture was then poured onto the glass plate to make a film by the solution casting method. The film was peeled off after drying.

In the second stage, the peeled-off film was placed in an aluminum mold (110 mm × 90 mm × 0.25 mm). The composite was prepared by pressing the peeled-off film using a Carver hydraulic system at a pressure of 10 tons at 155°C for 7 min followed by cooling. A neat SPI film was also prepared under similar conditions. Composites based on SPI and banana fibers have been designated as CBF followed by a numerical suffix indicating the volume fraction of fibers multiplied by 10. For example, soy protein composite having 0.2 and 0.3 volume

fraction of the fibers have been designated as CBF-2 and CBF-3, respectively.

The effect of glycerol content and volume fraction of fibers on the properties of composites were also evaluated. The composites thus formed were designated as G25CBF-3 and G50CBF-3 where G25 and G50 represent the weight percent of glycerol in the SPI matrix and all other designations remained the same. Specimens were also prepared by mixing different amounts of glutaraldehyde with G25CBF-3 in order to investigate the effect of additives on the water resistance of the composites.

2.4   Water absorption

Water uptake in banana fibers and banana fibers reinforced soy protein composites was measured according to ASTM D570-81. The specimens were pre-conditioned by drying in an air oven at 80°C for 26 h or until constant weight was obtained. The weighed samples were then submerged in distilled water at room temperature for 26 h. The specimens were removed from the water and surface water was removed with tissue paper before weighing.

2.5   Microbial study

About 3.9 g of potato dextrose agar (PDA) was dissolved in 100 mL of distilled water and the solution was autoclaved at 115°C for 20 min. The autoclaved media solution was poured into a sterile Petri dish under sterile conditions to prepare solid media for fungal growth (*Aspergillus niger*). In order to observe the fungal growth, soy protein composites after autoclaving (115°C for 20 min) were used. Part of the autoclaved soy protein composites were put on the PDA media and rest of the composites were used as a nutrient source for fungal growth. Inoculation was done by streaking the inocula on the protein composites with the use of a needle. The inocula were prepared by growing the fungi on potato dextrose agar media for 4–5 days at 28°C.

2.6   Characterization of fibers and composites

The denier of the fibers was determined by weighing accurately 20 fibers of 5 cm length. Four such measurements were done and the average weight of 100 cm long fibers (5 × 20 cm) was obtained. The denier, which is the weight of 9000 meter of fibers, was then computed. The density of soy protein film in the presence of glycerol, untreated fibers and fibers modified chemically by treatment with 5% or 10% NaOH was determined at room temperature using a density gradient column prepared using xylene and CCl_4 having density of 0.865 and 1.59 g/cm^3, respectively. A small tightly packed bundle of fibers was dropped in the column and the samples were left overnight for equilibration. The height was then noted. The column was calibrated using standard floats.

The density was then determined from the calibration plot.

One or two fibers were taken from the bundle of fibers for SEM analysis. The fiber was mounted on special stubs and silver coated under argon atmosphere for a coating thickness of approximately 200 Å before scanning. A Cambridge stereo scan model S4-10 was used for studying the morphology of fibers and composites at an accelerating voltage of 25 KV. For structural characterization, the FTIR spectra of treated and untreated banana fibers (as KBr pellets) were recorded using a Nicolet Protege 460 E. S.D. FTIR spectrophotometer. Powdered samples were used to record FTIR spectra of treated and untreated banana fibers as KBr pellets. WXRD was used to determine the structure of untreated and chemically modified fibers. Fiber in powder form was subjected to Ni-filtered Cu $K\alpha$ radiation using a Bruker D8 advanced diffractometer. The sample crystallinity was determined by means of the equation given below using the height of the 200 peak (I_{200} = 22.5°) and the minimum between the 200 and 110 peaks (I_{AM} = 18°). I_{200} represents both crystalline and amorphous material while I_{AM} represents amorphous material only.

$$X_{cr} = \frac{I_{200} - I_{AM}}{I_{AM}} \qquad (1)$$

The expression requires that the amorphous material diffracts with the same intensity at 18° and 22.5° and that the crystalline cellulose does not contribute to the intensity at 18° [23].

Mechanical properties of fibers were evaluated using a Zwick tensile testing machine (model Z010). For each sample, 30 fibers were tested under the following conditions.

Gauge length = 2.5 cm; Crosshead speed = 5 mm/min; Load cell = 1 kN

Mechanical properties of the composites were determined according to ASTM D-882 using a Zwick-Z010 test machine. Five specimens were tested for each formulation and the average value was reported.

TA 2100 thermal analyzer having a 910 DSC module and 951 TG module was used for the thermal characterization of banana fibers. Thermal stability was determined by recording TG/DTG traces in nitrogen atmosphere (flow rate of 60 cm^3/min) using powdered samples. A 10 $\pm$ 1 mg sample was heated at a rate of 10°C/min in each experiment.

3   Results and discussion

3.1   Characterization of banana fibers

Table 1 shows the properties of native and alkali modified fibers. The density was found to be 1.4 g/cm³. Treatment of fibers with varying concentrations of NaOH resulted in a marginal increase in the density of fibers which may be due to the depletion of lignin in fibers. Denier of the fibers decreased upon treatment with alkali. A weight loss of 20.3% and 25.4% was also observed upon treatment with 5% and 10% NaOH, respectively. Tensile strength and modulus of the fibers increased from 270 to 292 MPa and 10855 to 13696 MPa, respectively, upon treatment with 10% sodium hydroxide. Percentage elongation at breakage of the fibers decreased from 3.1 to 2.0 upon treatment with sodium hydroxide. It can be seen that alkali treatment of fibers resulted in increase in mechanical properties.

Table 1   Density and mechanical properties of treated and untreated fibers

properties	fibers designation		
	BF	S5-BF	S10-BF
density/g/cm⁻³	1.40	1.45	1.48
denier/g	122.8	108.9	90.0
maximum stress (σ)/MPa	270 $\pm$ 8.7	277 $\pm$ 10	292 $\pm$ 5.8
elongation/%	3.1 $\pm$ 1.1	2.1 $\pm$ 0.6	2.0 $\pm$ 1.1
modulus (E)/MPa	10855 $\pm$ 1510	13199 $\pm$ 2094	13696 $\pm$ 2525

Figure 1 shows the SEM micrographs of untreated and alkali treated fibers. The banana fibers presented a clean and smooth surface (Fig. 1a) whereas unevenness of the surface was observed after alkali treatment (Fig. 1b). Roughness in the fibers increased due to rupture of alkali sensitive bonds as the concentration of NaOH was increased from 5 to 10%. Moreover, the chemically modified fibers was not well separated but rather looked bundled. The diameter of the fibers also appeared to decrease upon treatment with higher concentrations of alkali (Fig. 1c).
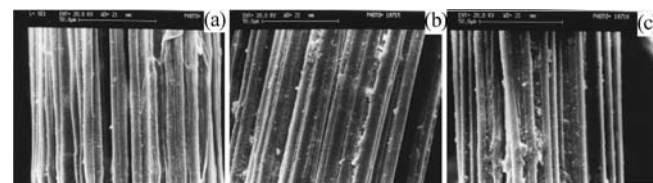

Fig. 1   SEM micrographs of banana fibers before and after alkali treatment (a) BF(b) S5-BF(c) S10-BF

The structural characterization of fibers before and after alkali treatment was done using FTIR and is shown in Fig. 2. The absorbance peaks at 3432 $\pm$ 10, 2922 $\pm$ 1 and 1640 $\pm$ 5 cm^{-1} were attributed to –OH, C–H and C=O stretching and vibration, respectively. Absorbances between 1000–1550 cm^{-1} represent the lignin and hemicellulose. Absorbance peak at 1023 $\pm$ 1 cm^{-1} is indicative of C–O stretching present in alcohol of hemicellulose. Treatment with caustic soda resulted in the loss of lignin from the banana fibers. The intensity of the peak at 1548 $\pm$ 1 cm^{-1} attributed to C=C stretching of aromatic

ring decreased when the fibers was treated with 5% NaOH (S5-BF) (Fig. 2). After treatment with 10% NaOH (S-10-BF), no absorbance peak was observed at 1548 cm^{-1} thereby indicating the removal of lignin. Other absorbance peaks did not show any changes in intensity.
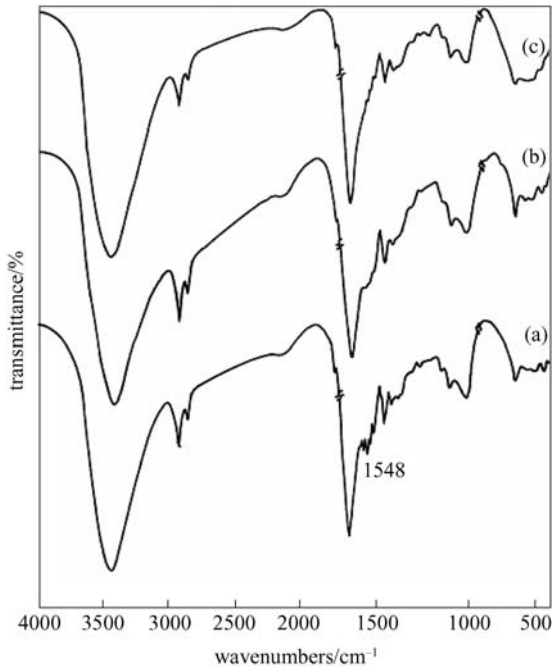

Fig. 2 FTIR spectra of banana fibers before and after treatment (a) BF (b) S5-BF (c) S10-BF

WXRD patterns for BF, S5-BF, S10-BF fibers are shown in Fig. 3. Crystallites of cellulose present in natural fiber are very small resulting in considerable peak broadening and overlap [24]. In the X-ray diffraction pattern of banana fibers, a main diffraction peak at 2θ–22.5°, associated with the diffraction plane of (002) of cellulose I was observed. A broad unresolved peak in the range of 13°–18° (2θ) corresponds to (101) and ($\overline{101}$) crystallographic planes of cellulose. The crystallinity index as calculated by the Segal method for BF was 68.0. Treatment of banana fibers with alkali results in reduced crystallinity. The crystallinity index decreased from 44.4 for S5-BF to 22.7 for S10-BF. The intensity of both the peaks also decreased after treatment with NaOH as shown in Fig. 3.

Figure 4 shows TG/DTG traces of banana fibers before and after treatment recorded in nitrogen atmosphere at a heating rate of 10°C/min. The traces were characterized by noting the initial decomposition temperature (IDT), final decomposition temperature (T_f) and associated mass loss (%). The temperature of maximum rate of mass loss was determined from the derivative thermogravimetric (DTG) trace. These results are summarized in Table 2. Single step degradation was observed in the treated (S10-BF) and untreated fibers (BF). About 9% mass loss was observed
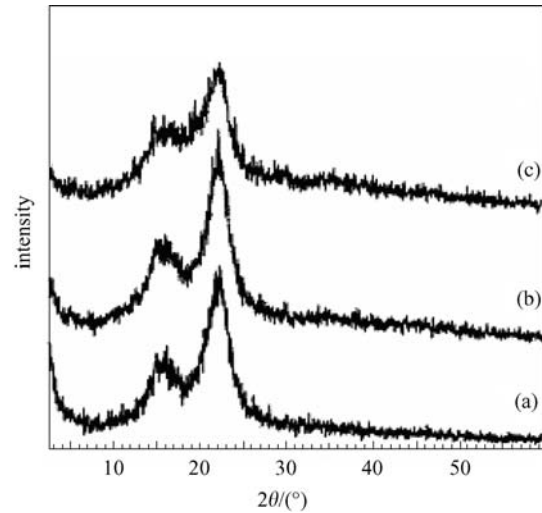

Fig. 3 X-ray diffraction patterns of banana fibers before and after treatment (a) BF (b) S5-BF (c) S10-BF

in the temperature range of 50°C–200°C, which can be attributed to desorption of physically adsorbed water. Major mass loss (69%–70%) was observed in the temperature range of 300°C–400°C leaving behind a char yield of 21%–22%.
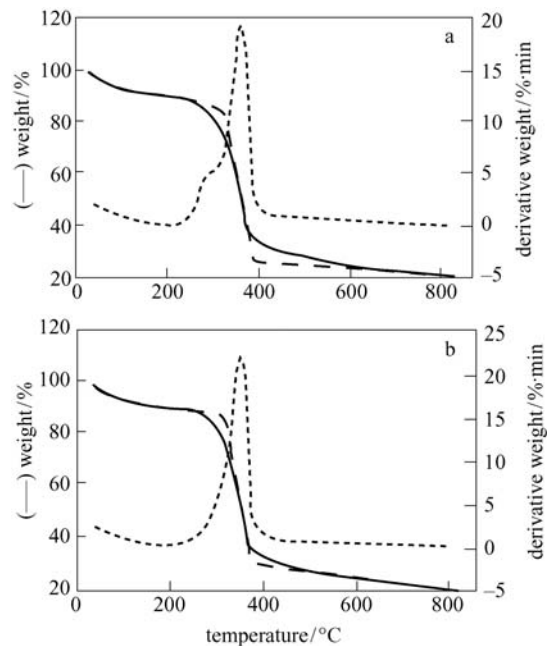

Fig. 4 TG/DTG traces of (a) BF (b) S10-BF

Table 2 Thermal characterization of untreated fibers and chemically modified banana fibers

sample designation	IDT /°C	T_{max} /°C	T_f /°C	Mass loss/%	Y_c at 700°C/%
BF	322	360	390	68.9	22.1
S10-BF	315	348	373	69.9	21.1

3.2 Mechanical properties of banana fibers reinforced soy protein composites

The properties of the composites are found to be dependent on the volume fraction of the fibers and on the nature of the matrix. In the following section, the mechanical properties of the banana fibers reinforced composites are discussed as a function of these parameters.

3.2.1 Fiber content

The mechanical properties, that is, tensile strength and modulus of SPI composites with S10-BF and BF fibers are summarized in Table 3. The soy protein composites prepared using varying volume fractions of S10-BF and BF fibers showed an increase in the tensile strength and modulus of up to 0.3 volume fractions of fibers followed by decrease in tensile strength. Soy protein composites prepared using alkali treated banana fibers (S10-BF) gave higher tensile strength and modulus compared to composites prepared using untreated fibers (BF). This could be due to better interaction between the matrix and the fibers after modification by sodium hydroxide. This has been confirmed by the increase in roughness as observed in the SEM photograph of alkali modified fibers.

3.2.2 Effect of plasticizer content in matrix

Table 4 shows the mechanical properties of the composites with varying length of the fibers and varying content of plasticizer at optimum weight fraction. For this purpose, composite samples were prepared using fibers of 0.5 and 11 cm length while the volume fraction of the fiber content was kept at 0.3. The plasticizer content in soy protein matrix used for making composites was varied from 25% to 50% (W/W). The tensile strength and modulus was highest with SPI matrix having 25% (W/W) glycerol. Further increase in the concentration of glycerol resulted in a decrease in the tensile strength and modulus of the composites. As for the 11-cm long treated fibres, a significant reinforcement effect in SPI composites was observed for all the samples regardless of plasticizer content. The direction of the measurement of mechanical properties for these composites was along the fibers' orientation. Tensile strength and modulus of the CBF-3.0 composite having 0.5 cm length of the fibers and 50% of glycerol content was 3.2 and 420 MPa, respectively, while composites having 11 cm length of fibers have tensile strength and modulus of 3.7 and 480 MPa, respectively. Composites having long fibers had higher mechanical properties regardless of the plasticizer content. Longer fibers will have less fiber ends and there would be less flaws or low stress bearing points which is the reason for the increased tensile strength and modulus for these composites.

3.3 Dispersion of banana fibers in the composites

The dispersion of banana fibers in the matrix is shown in Figure 5. Since the surface of the fibers became rough and the diameter of the fibers decreased after alkali treatment, the dispersion of alkali treated banana fibers in soy protein was better. The surface morphology of short fibers-reinforced soy protein composites samples was homogeneous (Fig. 5a) with no pores. The fractured surface of the random fibers-reinforced soy protein composite showed few fibers which were short in length. These were seen protruding suggesting brittle failure (Fig. 5b).

3.4 Water resistance

For this purpose, pre-weighed samples of banana fibers-reinforced soy protein composites were immersed in

Table 3 Tensile strength (σ) and modulus (E) of soy protein/glycerol (50% W/W) banana fibers composites: Effect of fiber content

sample designation	mechanical properties using BF		mechanical properties using S10-BF	
	σ/MPa	E/MPa	σ/MPa	E/MPa
CBF-0	1.78 ± 0.10	39.5 ± 3.0	1.78 ± 0.10	39.5 ± 3.0
CBF-2.0	1.05 ± 0.20	145 ± 5.0	2.45 ± 0.15	301 ± 8.5
CBF-2.5	1.45 ± 0.20	225 ± 5.5	2.53 ± 0.10	355 ± 9.1
CBF-3.0	1.62 ± 0.15	256 ± 7.9	3.24 ± 0.12	420 ± 5.4
CBF-3.5	1.55 ± 0.10	279 ± 8.1	2.54 ± 0.13	432 ± 5.1

Table 4 Tensile strength (σ) and modulus (E) of soy protein composite with S-10-BF: Effect of glycerol content and fiber length

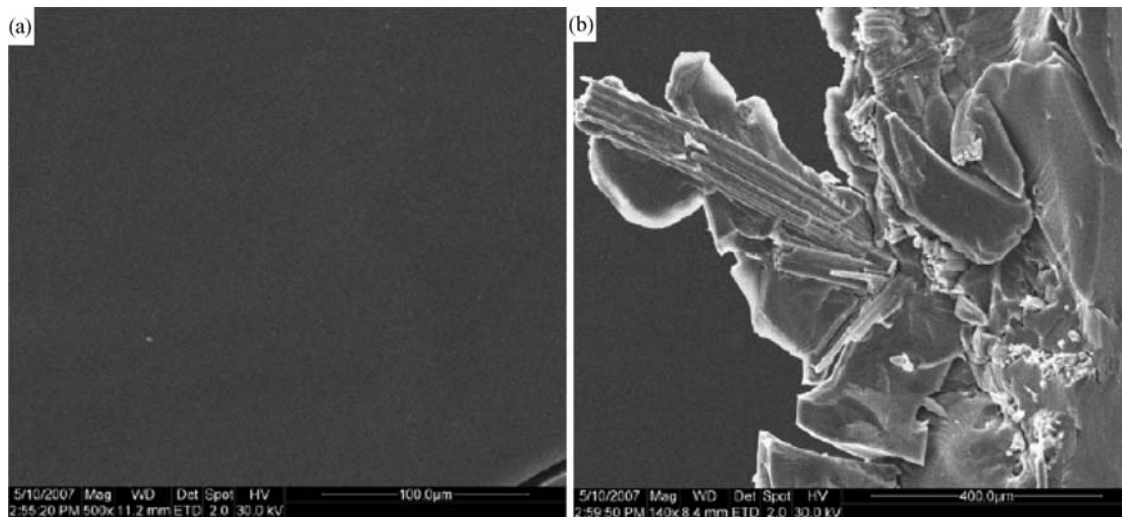| sample designation	0.5 cm (S10-BF)		11 cm (S10-BF)	
	σ/MPa	E/MPa	σ/MPa	E/MPa
G25CBF-3.0	4.55 ± 0.15	968 ± 22.5	5.9 ± 0.22	1074 ± 20
G37.5CBF-3.0	3.20 ± 0.10	530 ± 12.0	4.9 ± 0.17	742 ± 22.1
G50CBF-3.0	3.24 ± 0.12	420 ± 5.4	3.7 ± 0.13	480 ± 5.9

Fig. 5 SEM micrographs of banana fibers reinforced composites (a) surface (b) cross section

water and changes were observed after 26 h. After immersion in water, disintegration of G25CBF-3.0 sample was observed. Fibers were seen totally separated from the matrix. Fiber-reinforced composites prepared using GSPI-25 matrix modified by treatment with 0.4% (*W*/*W*) glutaraldehyde also resulted in separation of fibers from the matrix. Above 0.4% (w/w) of glutaraldehyde, however, composites remained intact after immersing in water for 26 h. Water absorption in the composites after 26 h decreased from 72% to 62% as the concentration of crosslinker in G25CBF-3.0 increased from 0.8% to 2.4 % (Fig. 6). The high water absorbing tendency of the composites may be due to the hydrophilic nature of both fibers (288% $\pm$ 5%) and matrix (78% $\pm$ 3%).
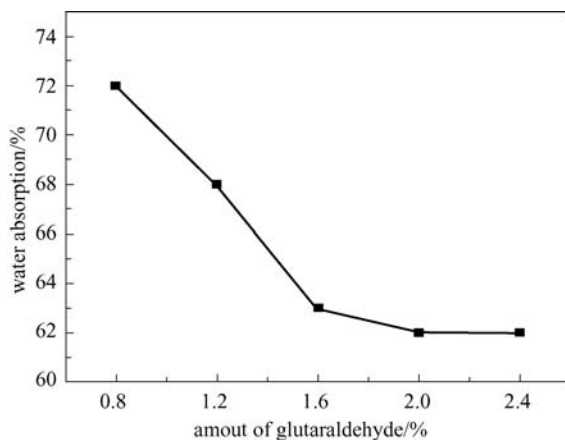

Fig. 6 Effect of glutaraldehyde on water absorption of banana fibers reinforced soy protein composites

The mechanism of reaction between soy protein and glutaraldehyde is given in Figure 7. Soy protein contains 18 different amino acids out of 20 amino acids present in natural proteins. Amino acids such as asparagine, glutamine, lysine and arginine contain more than one –NH$_2$ group that facilitates the reaction with glutaraldehyde. To show the crosslinking reaction, a short amino acid sequence, KVRRF (representing lysine, valine, arginine, arginine and phenylalanine), of α/β subunits of 11S globulin is reacted with glutaraldehyde.

3.5 Microbial growth

Figure 8 shows the growth of microorganism on the composites. No growth of microorganisms was observed even after 6 months when autoclaved G50CBF-3.0 composite was used as the nutrient for *A. niger* (Fig. 8a) growth. This showed excellent stability of the soy protein composites against fungal growth. However, when autoclaved G50CBF-3.0 films were mounted on solid PDA media, growth of *A. niger* was observed after 48 h. Figure 8b shows the growth of *A. niger on* G50CBF-3.0 films mounted on PDA media. Maximum growth was observed after 72 h.

4 Conclusions

Alkali treatment of the banana fibers decreased the lignin component, crystallinity and increased the roughness of the surface and mechanical properties of the fibers. Mechanical properties of the fiber reinforced composites were strongly dependent on the volume fraction of the banana fibers and the amount of plasticizer used. It can also be concluded that alkali treatment of the fibers is necessary to get composites with moderate mechanical properties as well as better adhesion between fibers and matrix.
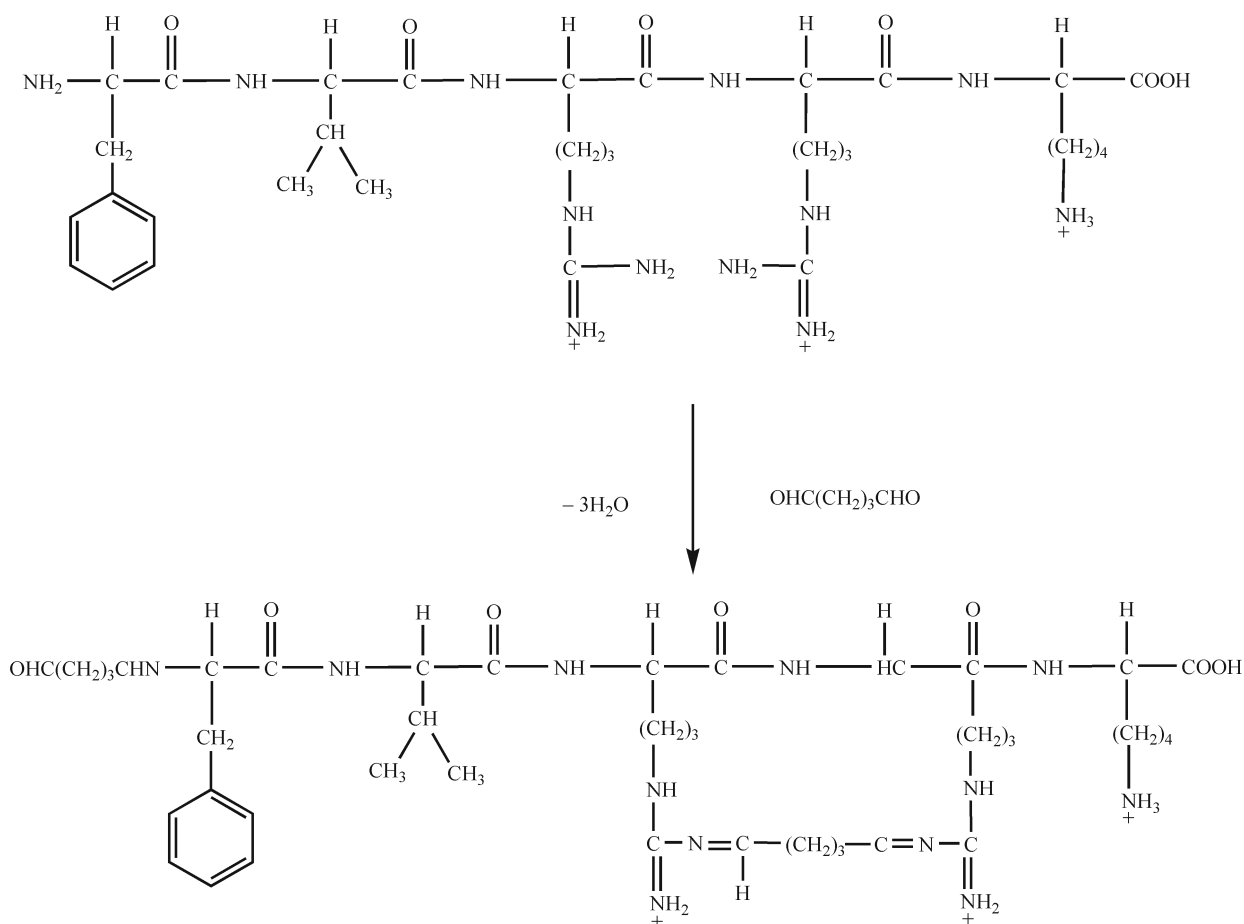
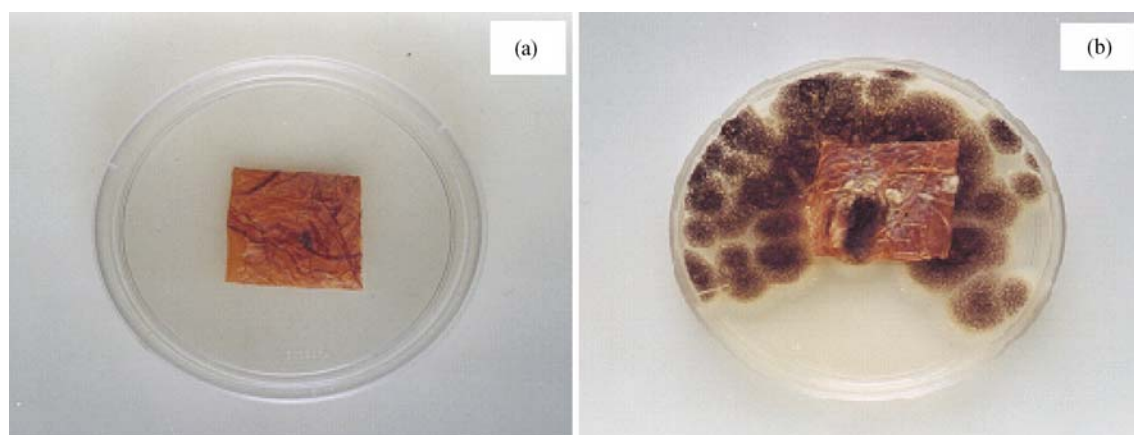**Fig. 7**   Crosslinking reaction of soy protein with glutaraldehyde

Fig. 8   (a) No growth of *Aspergillus niger* on autoclaved G50CBF-3.0 and (b) growth on autoclaved G50CBF-3.0 film mounted on PDA media after 48 h

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