Spathodea campanulata bud fluid reinforced mechanical, hydrophilicity and degradation studies of poly (vinyl alcohol) matrix

Naganagouda Goudar¹ · Vinayak N. Vanjeri¹ · Saraswati P. Masti² · Ravindra B. Chougale¹

Received: 28 January 2020 / Accepted: 2 March 2020 / Published online: 6 March 2020
© Springer Nature Switzerland AG 2020

Abstract

The present study aims to scrutinize the influence of *Spathodea campanulata* (SC) bud fluid on various properties of PVA. The different volumes of SC bud fluid doped PVA films were prepared by the solvent evaporation technique and analyzed by various characterization techniques. The molecular interactions of PVA with SC bud fluid was confirmed by FTIR studies. Tensile properties such as tensile strength and Young's modulus increased from 48 ± 4 to 59 ± 10 and 2069 ± 110 to 2537 ± 404 MPa respectively compared to PVA. The DSC measurements of prepared films depicted an increased glass transition temperature than PVA. The thermal stability of composite films was not affected compared to the pristine PVA. The XRD results of all the films represented a semi-crystalline material with a characteristic peak at $2\theta = 20^\circ$. The morphology studies revealed the homogeneous surface due to the good dispersion of SC bud fluid. The water contact angle findings shown the composite films were hydrophilic. Composite film solubility in water increased after the incorporation of SC bud fluid. The water vapor transmission rate of composite films were assessed. The Soil Burial tests indicated the degradation rate of PVA increased in composite films due to increased hydrophilicity.

Graphical abstract

Keywords *Spathodea campanulata* · Poly (vinyl alcohol) · Composite films · Hydrophilicity

✉ Ravindra B. Chougale, chougalerkud19@gmail.com | ¹P G Department of Studies in Chemistry, Karnatak University, Dharwad 580 003, India. ²Department of Chemistry, Karnatak Science College, Dharwad 580 001, India.

1 Introduction

The extreme usage of petroleum-based plastics which are non-biodegradable, severely damaged nature. About one-third of the total production of plastics is used in food packaging products. Researchers all over the world are trying to replace petroleum-based plastics by biodegradable plastics which are of less cost and efficient in terms of strength, flexibility, antioxidant, antimicrobial, chemical resistance and many more. To accomplish the requirements PVA was chosen as matrix because PVA is a synthetic, biodegradable, non-carcinogenic, water soluble and biocompatible polymer [1, 2]. PVA has excellent film forming ability, hydrophilicity and chemical stability. Therefore, it was blended with different synthetic and natural polymers [3–6]. It is used in various applications such as controlled drug delivery systems, recycling of polymers, film formation [7]. Although PVA is having reasonably good properties, efforts are being undertaken to improve some of the mechanical properties to make it comparable with other commodity plastics [8]. Natural extracts are the possible option to improve the biological activity and mechanical properties of the biodegradable polymers. To improve the properties of a biodegradable polymer there are various studies conducted using several natural extracts [9–14]. *Spathodea campanulata* P. Beauv (SC) is a flowering plant belonging to the Bignoniaceae family. In vitro antibacterial activity of leaf extracts of this plant against standard strains was evaluated [15]. Various phytochemical studies were performed with different parts of SC including stem barks, flowers, leaves and fruits. The stem barks contain Spathodic acid, steroids, saponins, ursolic acid, tomentosolic acid and pectic substances [16–19]. The molluscicidal activity was shown by flowers and stem bark extracts [20]. These are also used to treat diuretic and anti-inflammatory [21]. Banerjee & De showed the presence of anthocyanins in flowers of SC [22]. The stem bark preparations are used to treat fungal skin diseases, herpes, stomach aches and diarrhea [19], Hypoglycemic, anti-HIV and antimalarial activities were also observed in stem bark extracts [23, 24]. The flower bud is brown, horn-shaped, and contains the fluid which circulates in the vascular system of a plant, consisting chiefly of water with dissolved sugars and mineral salts. It contains large pinnate foliage and large showy orange or scarlet, curved flattened, tulip-like flowers arranged in terminal panicles or racemes above the foliage at the top of the trees [25]. The SC bud fluid contains mainly alkanes, terpenoids, aromatic hydrocarbons, alcohols, carbonyl compounds, esters, lactones and phenols. There is only one survey in the literature regarding the isolation and identification of SC bud fluid components

& around 35–60 compounds were isolated, identified by solvent extraction & GC–MS respectively [25]. The major components which might be responsible for hydrogen bonding are given in Fig. 1.

The flower bud exudate was used to treat eyes related diseases [25, 26]. These essential characteristics of *Spathodea campanulata* tree and bud fluid inspired us to select as a natural extract for our study. For the first time in the literature the SC bud fluid was doped into a polymer matrix and the consequences of SC bud fluid on multifunctional properties of PVA were studied exhaustively. There are no such reports available in the literature. The current study might be the basis for further exploration of SC bud fluid on different polymers in the coming days. The present investigation aims to optimize the SC bud fluid composition in the PVA matrix and to study the influence of SC bud fluid on the physicochemical properties of the PVA matrix.

2 Experimental

2.1 Materials

PVA (Molecular Weight 115,000, Degree of Hydrolysis 98–99 mol%) was procured from Loba Chemicals, Mumbai. Millipore water used throughout the experiment. All the chemicals were used as received without any purification.

2.2 Collection of *Spathodea campanulata* bud

The SC buds were collected from its plant at Karnatak University Campus, Dharwad, Karnataka, India, and it was then authenticated by Botany Professor, Karnatak University, Dharwad. The collected buds were washed with double distilled water. The light brown color fluid was withdrawn from the bud with the help of a micro syringe and treated with charcoal at 35 °C to remove coloring impurity. The

Fig. 1 The representative components of SC bud fluid

filtered solution was kept in a vial covered with carbon paper until further use.

2.3 Preparation of films

The films were prepared by solution casting and solvent evaporation method. To prepare pure PVA film, an exactly weighed amount of PVA was dissolved in double distilled water with stirring at 80–90 °C for an hour (h) and allowed it to attain RT. The different volume of SC bud fluid was added to the PVA solution (coded as PSC) with the help of a microsyringe and stirred for 20 h at RT.

The homogeneous solution was then casted onto clean and dry Petri dishes for solvent evaporation at RT. After 6–7 days the films were peeled off from the Petri dishes and stored in desiccators until further use. The detailed composition is given in Table 1.

3 Characterizations

3.1 Fourier Transform Infrared (FTIR) spectroscopy

The interaction among the components was successfully examined with an attenuated total reflection (ATR) method of IR spectrometer [Fourier Transform Infrared (FTIR) spectroscopy-attenuated total reflection, PerkinElmer Spectrum Version 10.5.4]. Measurements were conducted between 400 and 4000 cm^{-1} at 4 cm^{-1} resolution.

3.2 Mechanical properties

The thickness of the prepared films was measured using a digital micrometer (Mitutoyo Digital Micrometer, high accuracy S700 series, Japan). Several thickness measurements were taken at several locations of each film and mean value was reported. A Universal Testing Machine (UTM, DAK System Instrument, Mumbai), was used to determine the tensile properties such as tensile strength (T_g), Young's modulus (Y_m), and elongation at break (E_b) according to ASTM D882-91. Rectangular film samples of 2.5 cm × 10 cm was cut with a sharp knife from the prepared film samples and then placed in the extension grip

of the testing machine. The film samples were stretched at a crosshead speed 1 mm/min at RT in air.

3.3 Differential scanning calorimetry (DSC)

The glass transition (T_g) and melting temperature (T_m) of the prepared films were analysed using Differential Scanning Calorimetry (Instrument: DSC Q20 V24.10 Build 122 TA Instruments, Walters LLC New Castle, Delaware, USA). The film samples of weight 2–3 mg were sealed in an aluminium crucible and heated from 25 °C to 400 °C at 10 °C/min in an inert N_2 gas (50 mL/min).

3.4 Thermogravimetric analysis (TGA)

Thermogravimetric analysis (TGA) was carried out on an equipment SDT Q600 V20.9 TA instruments. The samples of 5–10 mg was taken and heated up until 600 °C at a heating rate of 10 °C/min under N_2 atmosphere (100 mL/min).

3.5 X-ray diffraction (XRD) study

The change in structural properties of PVA was investigated using Rigaku D/Max-IIA, X-ray diffraction (XRD) (Tokyo, Japan). The radiation was generated from Cu-K_β ($\lambda = 1.5406 \text{ \AA}$) source at a voltage 40 kV and 40 mA was used to scan the samples between $2\theta = 5^\circ$ – 80° with a scanning speed of 5°/min. The percentage of crystallinity was calculated by using the Eq. (1).

$$\text{Crystallinity}(\%) = \frac{\text{Total Area of Crystalline Peaks}}{\text{Total Area of all Peaks}} \times 100 \quad (1)$$

3.6 Scanning electron microscopy (SEM)

The morphology of the prepared films was examined using a JEOL JSM-6360 Scanning Electron Microscopy (SEM) at an acceleration voltage of 10 kV. All specimens were sputter-coated with a conductive layer of gold to avoid charging to the high electron beam. The film specimens were mounted on metal stubs using double-sided sticky carbon tape.

3.7 Atomic force microscopy (AFM)

Atomic Force Microscopy (AFM) (Nanosurf Easyscan 2, Switzerland), were used for investigating the surface topography and morphology of the composite films at ambient temperature. Topographic images were obtained by contact angle mode using aluminium coated cantilever and images were captured at different locations.

Table 1 Composition of PSC composite films

Sample code	PVA	SC bud fluid	Thickness (mm)
PVA	2 g	–	0.06 ± 0.01
PSC-1	2 g	0.2 mL	0.06 ± 0.01
PSC-2	2 g	0.4 mL	0.05 ± 0.01
PSC-3	2 g	0.6 mL	0.06 ± 0.01

3.8 Water contact angle (WCA) measurement

Contact angle measurements were carried out to acquire information on the hydrophilicity of the film surfaces. A contact angle analyzer, SEO Phoenix, was used to determine the contact angle of water. The measurements were conducted at RT and the size of the drop was 7 μ L with all measured probe liquids. At least five measurements at different locations per test point were performed. Contact angle calculations were done with associated software. The results are presented as average $\pm$ standard deviation.

3.9 Water solubility (WS)

To check out the stability of the prepared films in water, the solubility of films in water was carried out according to Shojaei-Aliabadi et al. [27]. In brief, initially, the prepared films were dried in an oven at 105 ± 2 °C to remove the moisture adhered to the surface. The dried samples of size 2×2 cm² were weighed and immersed in a beaker containing 40 mL of water and kept vigorous stirring for 6 h at RT. After 6 h the films were removed from the beaker and blotted with tissue paper and dried in an oven to get the final weight. The percentage of WS was calculated using the Eq. (2)

$$WS(\%) = \frac{W_i - W_f}{W_i} \times 100 \quad (2)$$

where W_i is the initial weight of samples, W_f is the final weight of samples.

3.10 Water vapor transmission rate (WVTR) test

The Test was conducted using the method described by Muhammad Salman Sarwar et al., with some modifications [28]. The glass bottle of 17.5 mm diameter was taken as container and filled with 10 mL of distilled water. The films were wrapped on the mouth of container and tightened with the Teflon tape. The initial weight of glass bottles (W_1) were weighed and placed in an oven at 40 °C for 24 h. After 24 h the glass bottles were taken out of the oven and weighed again to get the final weight (W_2). The WVTR was calculated using the Eq. (3).

$$WVTR = \frac{W_1 - W_2}{A} \times T \text{ g/m}^2\text{h} \quad (3)$$

where A is the area of the circular mouth of the glass bottle and T is the time (24 h).

3.11 Soil burial test

The biodegradability of the prepared films was studied by soil burial test under laboratory conditions according

to the method described by Sa-Ad Riyajan et al. [29]. The fresh soil was collected from the Botanical garden, Karnataka University Campus, Dharwad, Karnataka, India. The samples were cut into 2×2 cm² and dried at 40 °C to get the initial dry weight (w_1). The samples were buried in the soil at depth of about 8–10 cm under the soil surface. The moisture of soil was maintained by spraying water on the surface of the soil. The change in weight was recorded after 20 days by removing the films from soil and washing followed by drying in an oven (w_2). The biodegradation was calculated using Eq. (4).

$$\text{Biodegradation (\%)} = \frac{w_1 - w_2}{w_1} \times 100 \quad (4)$$

4 Results and discussion

4.1 Fourier transform infrared (FTIR) spectroscopy

To affirm the interaction among the components in the given composition range FTIR was carried out. The FTIR spectra of PVA, PSC composites are shown in Fig. 2. The FTIR spectra of bare PVA showed a characteristic broad absorption peak at 3306 cm⁻¹ attributed to the presence of -OH group. The weak absorption peaks at 2920 cm⁻¹ and 2852 cm⁻¹ are due to the asymmetric and symmetric stretching vibrations of CH₂ group respectively [30, 31]. The weak absorption peak observed at 1327 cm⁻¹ is due to the C-OH bending vibration. The peak at 1732 cm⁻¹ is due to the presence of carbonyl (C=O) stretching of the

Fig. 2 FTIR spectra of neat PVA and PSC composite films

non-hydrolysed residual vinyl acetate group of PVA [32]. The FTIR spectra of PSC composite films revealed different features. The interaction between the PVA and SC bud fluid was confirmed by observing the changes in the spectra of PSC composite films. In the FTIR spectra of PSC composite films, the broad peaks at 3288, 3280, and 3294 are assigned to -OH stretching of PVA.

The peak shifted from 3306 to 3280 cm^{-1} (26 cm^{-1}) indicates the considerable interaction of -OH groups of PVA with the polar groups of SC bud fluid components. The -OH group peak intensity of pure PVA was decreased successively after incorporating SC bud fluid indicates the intermolecular interaction between the two components. The intensity of peak at 1732 cm^{-1} (C=O) was decreased and also shifted to lower frequency after doping with SC bud fluid, this may be due to the hydrogen bonding of carbonyl group with the polar groups of SC bud fluid. The remaining IR spectral data are presented in Table 2.

4.2 Tensile properties

The mechanical properties such as tensile strength (T_s), Young's modulus (Y_m) and Elongation at break (E_b) were assessed to know the strength and flexibility of the prepared films. The stress–strain curves are presented in Fig. 3. The PSC composite results are tabulated in Table 3. At lower volume of SC bud fluid, T_s of the pure PVA was increased from 47.9 ± 4 to 59 ± 10 MPa and Y_m was increased from 2069 ± 110 to 2537 ± 404 MPa. But there was a decrease in E_b which was decreased from $118.1 \pm 5\%$ to $43.9 \pm 4.2\%$, indicates the decrease in composite films flexibility, which is due to the restriction of the ductile flow of the polymer chains [33]. I. Korbag & S. Mohamed Saleh, who studied the influence of lignin on mechanical and biodegradability of PVA also reported the same observation and mentioned that the addition of lignin into PVA matrix slightly decreases E_b [34]. At lower content of SC bud fluid (0.2 mL), there was an increase in T_s and Y_m , which corresponds to the uniform distribution of SC bud fluid in the PVA matrix [35]. As the content of SC bud fluid increased from (0.4 mL to 0.6 mL), the T_s and Y_m were decreased. Which may be due to the weak molecular interaction between the polar groups of SC bud fluid and -OH group

Fig. 3 stress–strain curves of blank PVA and PSC composite films

of PVA. The decrease in T_s , Y_m , and E_b at higher loading of SC bud fluid attributed to the insufficient interaction and bad compatibility or non-homogeneous dispersion induced by the SC bud fluid [17, 36, 37]. A negative correlation existed between properties T_s , Y_m , E_b and content of SC bud fluid, that is an increase in the volume of SC bud fluid in the PVA matrix led to decreased T_s , Y_m , and E_b . The present findings of this study are in good agreement with the literature reported by other researchers [36, 38]. The results are relatable with the FTIR study.

4.3 Differential scanning calorimetry (DSC)

The crystallization and melting behavior of polymer composites strongly influences the macroscopic properties of materials [33]. To examine these parameters and also to find out the miscibility between the components, Differential Scanning Calorimetry (DSC) technique was used. The thermograms of neat PVA, PSC composite are shown in Fig. 4 and the data are given in Table 4. The Appearance of a small endothermic peak in all composite films below 80°C is due to loss of moisture [37, 39]. The neat PVA shows two endothermic peaks at 52.4°C and 189°C which corresponds to the glass transition

Table 2 FTIR Spectral data of neat PVA and PSC composite films

Sample Code	-OH stretching (cm^{-1})	$\text{-CH}_2\text{-}$ asymmetric stretching (cm^{-1})	$\text{-CH}_2\text{-}$ symmetric stretching (cm^{-1})	-C-OH bending (cm^{-1})	-C-O- stretching (cm^{-1})
PVA	3306	2928	2852	1327	1090
PSC-1	3288	2928	2853	1329	1089
PSC-2	3280	2928	2852	1326	1088
PSC-3	3294	2928	2851	1328	1087

Table 3 Mechanical properties of PVA and PSC composite films

Sample code	Tensile strength (MPa)	Young's modulus (MPa)	Elongation at break (%)
PVA	47.9 ± 4.0	2069.5 ± 110	118.1 ± 5
PSC-1	59.0 ± 10.0	2537.5 ± 404	109.7 ± 20.1
PSC-2	45.0 ± 5.4	2210.4 ± 346.3	43.9 ± 4.2
PSC-3	43.0 ± 2.4	1661.7 ± 14.4	61.4 ± 8.1

Fig. 4 DSC thermograms of PVA and PSC composite films**Table 4** T_g , T_m and T_d of PVA and PSC composite films

Sample code	Glass transition temperature (T_g) (°C)	Melting temperature (T_m) (°C)	Decomposition temperature (T_d) (°C)
PVA	52.4	192.7	319.0
PSC-1	71.5	222.0	286.0
PSC-2	78.5	224.0	287.0
PSC-3	68.5	223.0	290.0

temperature (T_g), and melting temperature (T_m) respectively [40]. After incorporating with SC bud fluid, the T_g and T_m increased from 52.4 °C to 78.5 °C and 189 °C to 222 °C respectively, indicating the strong intermolecular interaction between the matrix and bud fluid components. In the present study increase in the T_g value for PSC composite films may be attributed to the hydrogen bonds formed between polyphenolic compounds such as benzyl alcohol, *trans*-verbenol, Eugenol, linoleic acid [41] of bud fluid and -OH groups of PVA. Similar results were reported by other authors working on different plant extracts [57]. A single endothermic peak between the temperature range of 50–120 °C viz a single T_g for all the films proves the miscibility of matrix and bud fluid over the given composition [35].

Table 5 Thermal properties of pure PVA and PSC composite films

Sample code	Weight loss (%) I	Weight loss (%) II	Weight loss (%) III
PVA	20.11% (39.6–93.9 °C)	69.92% (248.6–299.8 °C)	14.07% (407–461.7 °C)
PSC-1	20.99% (42.2–85.9 °C)	62.75% (243–296.3 °C)	15.34% (404–460 °C)
PSC-2	16.33% (43.4–108.4 °C)	72.82% (249–294 °C)	14.77% (405.6–462.3 °C)
PSC-3	15.55% (56.5–107 °C)	59.77% (251.8–293.4 °C)	11.45% (404.8–462.5 °C)

Fig. 5 TGA thermograms of PVA and PSC composite films

4.4 Thermogravimetric Analysis (TGA)

To study the influence of SC bud fluid on the thermal stability of PVA the TGA was carried out. The TGA thermograms of pristine PVA and PSC composite films are shown in Fig. 5. All the films including pure PVA have shown three stage degradation patterns. The neat PVA shows first degradation step at 39.6–93.9 °C (20.11%) which is due to the elimination of physisorbed water molecules [42]. The second major weight loss of PVA has occurred at 248–299 °C (69%) is ascribed to the elimination of acetate groups of PVA via chain scission reaction. The third weight loss occurred at 407–461 °C is attributed to decomposition of residual groups of PVA [43]. After incorporation of SC bud

fluid into the PVA matrix, the thermal stability of composite films was decreased by 4–5 °C as shown in Table 5. The neat PVA has exhibited a residue of 14.07% which is due to carbonaceous residue. The PSC-1 and PSC-2 has residue of 15.34% and 14.77% respectively, due to a minor increase in the resistance to thermal degradation process. But the PSC-3 has residue of 11.45% which is much lesser than PVA, PSC-1 and PSC-2 indicating the lesser thermal stability of PSC-3 compared to PSC-1 and PSC-2. As for as PSC-3 residue is concerned, the residue is mainly due to the carbonaceous residue of PVA and not due to the SC bud fluid. The components of SC bud fluid degrade in the first and second degradation step only. The decreased thermal stability of the composite films might be due to the lesser stability of SC bud fluid to the temperature. The similar results were reported by Kasai et al., who studied the influence of *Piper nigrum* leaves extract on physicochemical properties of CH/ PVA blend films [35]. The decreased thermal stability of the composite films is contrary to the FTIR, DSC and mechanical properties results.

The increased intermolecular interaction might accelerate the water molecule evaporation from the composite films ultimately reducing the thermal stability [44]. The detailed data of the thermal degradation process of composite films are presented in Table 5.

4.5 X-ray diffraction (XRD) study

Figure 6. shows the X-ray diffractograms of the blank PVA and PSC composite films. The characteristic diffraction peaks of neat PVA occurred at $2\theta = 19.6^\circ$ and 40.7° which is attributed to the semicrystalline nature of PVA [9, 45–47]. The small hump at 40° was also characteristic for PVA [48]. The percentage of crystallinity of PVA was found to be 29. After incorporating the SC bud fluid into PVA matrix, there are no much changes in the diffraction peaks of PVA but there is an increase in the percentage of crystallinity from 36.6 to 54.5%, indicating the considerable interaction between the –OH groups of PVA and the active groups of SC fluid. The intensity of the broad peak at $2\theta = 19.6$ was decreased after blending it with SC bud fluid. It was observed that the addition of SC bud fluid does not affect the crystal type of the film, which means that the SC bud fluid has been uniformly distributed inside the host polymer [49]. The diffractograms of PSC composite films represented a typical property of partially crystalline materials with a characteristic peak at $2\theta = 20^\circ$ and are similar to the results reported by other authors [50, 51]. The percentage of crystallinity calculated from diffracted intensity data of PSC composite films are presented in Table 6. The percentage of crystallinity of all PSC composite films was increased compared to bare PVA film leading

Table 6 Percentage of crystallinity of PVA and PSC composite films

Sample code	2 Theta (degree)	Crystallinity (%)
PVA	19.6	25 ± 4
PSC-1	19.6	29 ± 9
PSC-2	19.6	28 ± 6
PSC-3	19.6	31 ± 4

Fig. 6 X-ray diffraction pattern of neat PVA and PSC composite films

to less flexible and more strengthened films as witnessed by mechanical properties.

4.6 Scanning electron microscopy (SEM)

The consequence of SC bud fluid on microstructure of PVA was analysed with SEM. The SEM micrographs of neat PVA and PSC composite films are shown in Fig. 7. The control film (PVA) exhibits smooth and homogeneous compact microstructure without any visible depressions. With the addition of SC bud fluid into the PVA matrix, the change in morphology of composite films was observed based on the composition of bud fluid. At lower content of SC bud fluid, the films were homogeneous with some visible agglomeration of bud fluid components. The patches were observed after the addition of SC bud fluid into the PVA matrix which might be due to the nonhomogeneous distribution of bud fluid over the surface. The PSC-1 and PSC-2 composite films exhibited such type of morphology, in PSC-2 it was more clearly visible. The possible reason for

Fig. 7 SEM micrographs prepared composite films.(Magnification: 25.00 K)

this observation is that the PVA contains numerous –OH groups and SC bud fluid also contains the –OH groups that has resulted in the crosslinked network. Another reason for the current observation might be due to the coalescence phenomenon during the solvent evaporation process. At higher content of SC bud fluid, the surface was uneven, coarse and fluctuant due to lack of compatibility between the PVA matrix and bud fluid components. The lesser compatibility between the PVA matrix and bud fluid components could be due to the agglomeration of bud fluid components at a higher content. The current scenario is well cited in the literature [35, 36, 52–54].

4.7 Atomic force microscopy (AFM)

Atomic Force Microscopy was carried out to obtain the qualitative and quantitative surface parameters of the prepared films. The AFM images and respective 3D images of neat PVA and different composite films are shown in Fig. 8. The Parameters such as line roughness (Rq) and area

roughness (Sq) are obtained with the associated software and summarized in Table 7. The surface morphology of neat PVA film is smooth and homogeneous. The surface morphology of PSC composite films varied after incorporation of bud fluid. At lower content of SC bud fluid, the composite films are less rough than higher content loaded composite films. Indicating the good homogeneity and compatibility at lower content (0.2 mL) of SC bud fluid. As the volume of SC bud fluid increased, (from 0.2 mL to 0.6 mL) the line roughness (Rq) increased from 10.5 nm to 21.1 nm and also area roughness (Sq) increased from 9.3 nm to 20.1 nm. This indicates that at higher volume of bud fluid, the composite films are rougher. The PSC-3 composite film exhibited more heterogeneity. The present observation is in good agreement with the previous research works [27, 52, 55].

Fig. 8 AFM micrographs of neat PVA and prepared composite films

Table 7 Line roughness (Rq) and area roughness (Sq) of neat PVA and PSC composite films

Sample code	Line roughness (Rq) (nm)	Area roughness (Sq) (nm)
PVA	9.00 ± 1.1	10.2 ± 1.2
PSC-1	10.5 ± 1.2	9.3 ± 0.8
PSC-2	16.6 ± 1.0	20.1 ± 1.3
PSC-3	21.1 ± 0.6	16.1 ± 5.2

4.8 Water contact angle (WCA) measurement

To understand the surface wettability, all the films were analyzed with water contact angle analyzer using sessile drop method. The surface hydrophobicity is crucial, as it determines the water resistance of a packaging material, which helps to maintain the quality of food products [55]. This contact angle is a measure of the non-covalent forces between the ultrapure water and the first monolayer (surface) of the films and is an indicator of their superficial hydrophilic/hydrophobic properties [56]. The contact angle of the neat PVA and PSC composite films were presented Fig. 9. The contact angle of pristine PVA film was found to be 55° indicating the hydrophilic nature of PVA. The PSC composite films show drastic changes in the contact angle. At lower volume of SC bud fluid, there is an increase in the hydrophilicity, which is attributed to

Fig. 9 Water contact angle images of neat PVA and PSC composite films

the strong intermolecular hydrogen bonding between the $-OH$ groups of PVA and the polar groups of SC bud fluid. At this content of SC bud fluid, the composite films were more hydrophilic than the neat PVA film. The present findings are in good agreement with the FTIR and mechanical properties of composite films. The increased wettability of the composite films may be due to the hydrophilicity of added SC bud fluid [57].

4.9 Water solubility (WS)

Water Solubility is a crucial property in food packaging applications because potential applications could require partial water solubility to enhance food integrity and water resistance [58, 59]. The results of solubility test of PVA and composite films are presented in Fig. 10. The neat PVA has shown 32% of water solubility and PSC composite films shows an increased solubility compared to pure PVA which is in good agreement with WCA results. The high solubility of composite films is due to the increased hydrogen bonding and presence of highly polar components of SC bud fluid.

4.10 Water vapor transmission rate (WVTR)

The WVTR is a key feature for a food packaging material and it must be low. WVTR was studied to check out the potentiality of prepared films for food packaging applications. The influence of SC bud fluid on WVTR of PVA matrix was studied. The obtained results are presented in Fig. 10. The neat PVA has shown WVTR of 39 g/m².day. After the incorporation of SC bud fluid into the matrix, the WVTR increased to a maximum of 43 g/m².day which might be due to the high hydrophilicity of composite films. The WVTR findings of composite films are relatable to WCA and WS results.

4.11 Soil burial test

The influence of SC bud fluid on the soil degradation rate of PVA was studied in laboratory conditions. The neat PVA requires around 90 days for 30.2% degradation [60]. The results of soil burial tests are presented in Fig. 11. After

Fig. 11 Soil Degradation rate of PVA and PSC composite films

20 days the PSC composite film's appearance become shrank by the action of microorganisms, indicating the degradation. The neat PVA has presented 15.6% degradation in the soil after 20 days of burial. The incorporation of SC bud fluid into the PVA matrix accelerated the soil degradation process, which might be due to the highly hydrophilic nature and early degradation of small molecules of plant extract. The composite film's weight loss varied from 18 to 28%. The Soil degradation rate of PVA was greatly enhanced by the incorporation of SC bud fluid.

Fig. 10 Solubility and WVTR of PVA and PSC composite films

5 Conclusion

In this present contribution, SC bud fluid incorporated PVA composite films were successfully prepared by the solvent evaporation technique and characterized. The interaction among the components was corroborated by FTIR studies. The T_g and Y_m of PSC composite films were greatly improved. The incorporation of SC bud fluid into the matrix has increased the T_g at a lower volume. The thermal stability of PSC composite films was not affected by the incorporation of SC bud fluid. The composite films exhibited semicrystalline property. The morphology studies revealed homogeneity at a lower amount of SC bud fluid. The prepared PSC composite films were hydrophilic. The WS results suggested the high solubility of composite films in water. The WVTR of PSC composite films were increased due to the hydrophilicity of the films, the soil degradation rate of PVA was improved in composite films. Based on the above results, the optimal content of SC bud fluid to dope into the PVA matrix is 0.2 mL. There is a chance for further exploration of the current study to reach the desired application level.

Acknowledgements One of the authors Mr. Naganagouda Goudar would like to thank the Council of Scientific and Industrial Research (CSIR) New Delhi, India, for awarding the Junior Research Fellowship (JRF). Authors thank the University Science Instrumentation Centre (USIC), DST-SAIF and DST PURSE-Phase II programme, Karnatak University, Dharwad, Karnatak, for providing the instrumentation facility. Authors would like to thank Department of Chemistry, Karnatak Science College, Dharwad—580001, for providing the Universal Testing Machine (UTM) facility to study the tensile properties of PVA based films.

Compliance with ethical standards

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

References

- Kim D, Jung J, Park S, Seo J (2015) Preparation and characterization of LDPE/PVA blend films filled with glycerin-plasticized polyvinyl alcohol. *J Appl Polym Sci* 41985:1–8
- Ahmed A, Niazi MBK, Jahan Z et al (2019) Enhancing the thermal, mechanical and swelling properties of PVA/starch nanocomposite membranes incorporating g-C₃N₄. *J Polym Environ* 28(1):100–115
- Yan J, Tian H, Zhang Y, Xiang A (2015) Effect of urea and formamide plasticizers on starch/PVA bioblend sheets. *J Appl Polym Sci* 42311:1–8
- Shawky HA (2009) Synthesis of ion-imprinting chitosan/PVA crosslinked membrane for selective removal of Ag (I). *J Appl Polym Sci* 114:2608–2615
- Sonker AK, Rathore K, Teotia AK et al (2018) Rapid synthesis of high strength cellulose—poly (vinyl alcohol) (PVA) biocompatible composite films via microwave crosslinking. *J Appl Polym Sci* 47393:27–30
- Jung G, Kim H (2014) Synthesis and Photocatalytic Performance of PVA/TiO₂/graphene-MWCNT nanocomposites for dye removal. *J Appl Polym Sci* 40715:1–7
- Tripathi S, Mehrotra GK, Dutta PK (2010) Preparation and physicochemical evaluation of chitosan/poly(vinyl alcohol)/pectin ternary film for food-packaging applications. *Carbohydr Polym* 79:711–716
- Kumar SV, George J, Sajeevkumar VA (2018) PVA based ternary nanocomposites with enhanced properties prepared by using a combination of rice starch nanocrystals and silver nanoparticles. *J Polym Environ* 26:3117–3127
- Ahmadi R, Ghanbarzadeh B, Ayaseh A et al (2019) The antimicrobial bio-nanocomposite containing non-hydrolyzed cellulose nanofiber (CNF) and Miswak (*Salvadora persica* L.) extract. *Carbohydr Polym* 214:15–25
- Akhtar MJ, Jacquot M, Jasniowski J et al (2012) Antioxidant capacity and light-aging study of HPMC films functionalized with natural plant extract. *Carbohydr Polym* 89:1150–1158
- Estevez-areco S, Goyanes S (2019) Potato starch-based biocomposites with enhanced thermal, mechanical and barrier properties comprising water-resistant electrospun poly (vinyl alcohol) fibers and yerba mate extract. *Carbohydr Polym* 215:377–387
- Chranioti C, Nikoloudaki A, Tzia C (2015) Saffron and beetroot extracts encapsulated in maltodextrin, gum Arabic, modified starch and chitosan: Incorporation in a chewing gum system. *Carbohydr Polym* 127:252–263
- Medina C, Gutiérrez TJ, Goyanes S et al (2016) Biodegradability and plasticizing effect of yerba mate extract on cassava starch edible films. *Carbohydr Polym* 151:150–159
- Medina-jaramillo C, Ochoa-yepes O, Bernal C, Famá L (2017) Active and smart biodegradable packaging based on starch and natural extracts. *Carbohydr Polym* 176:187–194
- Kowti R, Gulzar Ahmed M, Gowda TS et al (2010) Antimicrobial activity of ethanol extract of leaf and flower of *Spathodea campanulata* P. Beauv. *RJPBCS* 1:691
- Ilodigwe EE, Akah PA, Nworu CS (2010) Anticonvulsant activity of ethanol leaf extract of *Spathodea campanulata* P. Beauv (Bignoniaceae). *J Med Food* 13:827–833
- Ngouela S, Nyasse B, Tsamo E et al (1990) Spathodic acid: a triterpene acid from the stem bark of *Spathodea campanulata*. *Phytochemistry* 29:3959–3961
- Ngouela S, Tsamo E, Sondengam B (2007) Extractives from bignoniaceae: constituents of the stem bark of *Spathodea campanulata*. *Planta Med* 54:476–476
- Silva C, Torres MD, Chenlo F, Moreira R (2017) Calyx-water consumption by blue-and-yellow macaws in *Spathodea campanulata* (Bignoniaceae) floral buds. *Ornitologia Neotropical* 26:201–206
- Mendes NM, Souza CP, Araújo N, Pereira JP, Katz N (1986) Atividade moluscicida de alguns produtos naturais sobre *Biomphalaria glabrata*. *Mem Inst Oswaldo Cruz Rio de Janeiro* 81:87
- Boniface PK, Verma S, Shukla A et al (2014) Membrane stabilisation: a possible anti-inflammatory mechanism for the extracts and compounds from *Spathodea campanulata*. *Nat Prod Res* 28:2203–2207
- Banerjee A, De B (2001) Anthocyanins in some flowers of West Bengal. *J Med Arom Plant Sci* 23:600–604
- Makinde J, Amusan O, Adesogan E (1987) The antimalarial activity of *Spathodea campanulata* Stem Bark Extract on *Plasmodium berghei* berghei in Mice. *Planta Med* 54:122–125
- Niyonzima G, Laekeman G, Witvrouw M et al (1999) Hypoglycemic, anticomplement and anti-HIV activities of *Spathodea campanulata* stem bark. *Phytomedicine* 6:45–49

25. Eid HH, Shehab NG, El Zalabani SM (2014) GC-MS profile and cytotoxicity of the hydrodistilled and extracted volatiles of the buds and flowers of *Spathodea campanulata* P. Beauv. J Biol Act Prod from Nat 4:196–208
26. Adio Gbemisola I, Faluyi JO, Osoniyi O (2014) Evaluation of the effect of *Spathodea campanulata* flower bud exudate on cataractogenesis in rat lenses. Afr J Tradit Complement Altern Med 11:83–91
27. Shojaei-Aliabadi S, Hosseini H, Mohammadifar MA et al (2013) Characterization of antioxidant-antimicrobial κ -carrageenan films containing *Satureja hortensis* essential oil. Int J Biol Macromol 52:116–124
28. Sarwar MS, Bilal M, Niazi K et al (2018) Preparation and characterization of PVA/nanocellulose/Ag nanocomposite films for antimicrobial food packaging. Carbohydr Polym 184:453–464
29. Riyajan S-A (2019) Environmentally friendly novel maleated poly(vinyl alcohol) grafted 1,4-butanediol modified with biopolymer for encapsulation of capsaicin. J Polym Environ 27:2637–2649
30. Choo K, Ching YC, Chuah CH et al (2016) Preparation and characterization of polyvinyl alcohol-chitosan composite films reinforced with cellulose nanofiber. Materials (Basel) 9:644
31. Guzman-Puyol S, Ceseracciu L, Heredia-Guerrero JA et al (2015) Effect of trifluoroacetic acid on the properties of polyvinyl alcohol and polyvinyl alcohol-cellulose composites. Chem Eng J 277:242–251
32. Lee SV, Halim NA, Arof AK, Abidin ZHZ (2013) Characterisation of poly(vinyl alcohol) coating mixed with anthocyanin dye extracted from roselle flower with different nitrate salt. Pigment Resin Technol 42:146–151
33. Díez-Pascual AM, Xu C, Luque R (2014) Development and characterization of novel poly(ether ether ketone)/ZnO bionanocomposites. J Mater Chem B 2:3065–3078
34. Korbag I, Mohamed Saleh S (2016) Studies on mechanical and biodegradability properties of PVA/lignin blend films. Int J Environ Stud 73:18–24
35. Kasai D, Chougale R, Masti S et al (2019) An investigation into the influence of filler *Piper nigrum* leaves extract on physicochemical and antimicrobial properties of chitosan/poly (vinyl alcohol) blend films. J Polym Environ 27:472–488
36. Kasai D, Chougale R, Masti S et al (2018) Influence of *Syzygium cumini* leaves extract on morphological, thermal, mechanical, and antimicrobial properties of PVA and PVA/chitosan blend films. J Appl Polym Sci 135:1–17
37. Li JH, Hong RY, Li MY et al (2009) Effects of ZnO nanoparticles on the mechanical and antibacterial properties of polyurethane coatings. Prog Org Coatings 64:504–509
38. El-Hefian EA, Nasef MM, Yahaya AH (2012) Mechanical, thermal and surface investigations of chitosan/agar/PVA ternary blended films. E J Chem 8:S105–S112
39. Ahmadizadegan H (2016) Synthesis and gas transport properties of novel functional polyimide/ZnO nanocomposite thin film membranes. RSC Adv 6:106778–106789
40. Luzi F, Fortunati E, Di Michele A et al (2018) Nanostructured starch combined with hydroxytyrosol in poly(vinyl alcohol) based ternary films as active packaging system. Carbohydr Polym 193:239–248
41. Asad M, Saba N, Asiri AM et al (2018) Preparation and characterization of nanocomposite films from oil palm pulp nanocellulose/poly (Vinyl alcohol) by casting method. Carbohydr Polym 191:103–111
42. Wu Y, Ying Y, Liu Y et al (2018) Preparation of chitosan/poly vinyl alcohol films and their inhibition of biofilm formation against *Pseudomonas aeruginosa* PAO1. Int J Biol Macromol 118:2131–2137
43. Fortunati E, Puglia D, Luzi F et al (2013) Binary PVA bio-nanocomposites containing cellulose nanocrystals extracted from different natural sources: Part I. Carbohydr Polym 97:825–836
44. Zhang X, Liu Y, Yong H et al (2019) Development of multifunctional food packaging films based on chitosan, TiO₂ nanoparticles and anthocyanin-rich black plum peel extract. Food Hydrocoll 94:80–92
45. Sadek EM, El-Nashar DE, Ward AA, Ahmed SM (2018) Study on the properties of multi-walled carbon nanotubes reinforced poly (vinyl alcohol) composites. J Polym Res 25:249
46. Sabarish R, Unnikrishnan G (2018) Polyvinyl alcohol/carboxymethyl cellulose/ZSM-5 zeolite biocomposite membranes for dye adsorption applications. Carbohydr Polym 199:129–140
47. Wang T, Li Y, Geng S et al (2015) Preparation of flexible reduced graphene oxide/poly(vinyl alcohol) film with superior microwave absorption properties. RSC Adv 5:88958–88964
48. Das K, Ray D, Bandyopadhyay NR et al (2010) Preparation and characterization of cross-linked starch/poly(vinyl alcohol) green films with low moisture absorption. Ind Eng Chem Res 49:2176–2185
49. Lotfy S, Fawzy YHA (2014) Characterization and enhancement of the electrical performance of radiation modified poly (vinyl) alcohol/gelatin copolymer films doped with carotene. J Radiat Res Appl Sci 7:338–345
50. Gaikwad KK, Lee JY, Lee YS (2016) Development of polyvinyl alcohol and apple pomace bio-composite film with antioxidant properties for active food packaging application. J Food Sci Technol 53:1608–1619
51. Kaczmarek H, Podgórski A (2007) The effect of UV-irradiation on poly(vinyl alcohol) composites with montmorillonite. J Photochem Photobiol A Chem 191:209–215
52. Fabra MJ, Talens P, Chiralt A (2009) Microstructure and optical properties of sodium caseinate films containing oleic acid-beeswax mixtures. Food Hydrocoll 23:676–683
53. Liu J, Wang H, Guo M et al (2019) Extract from *Lycium ruthenicum* Murr. incorporating κ -carrageenan colorimetric film with a wide pH-sensing range for food freshness monitoring. Food Hydrocoll 94:1–10
54. Qin Y, Liu Y, Yuan L et al (2019) Preparation and characterization of antioxidant, antimicrobial and pH-sensitive films based on chitosan, silver nanoparticles and purple corn extract. Food Hydrocoll 96:102–111
55. Nur Hanani ZA, Aelma Husna AB, Nurul Syahida S et al (2018) Effect of different fruit peels on the functional properties of gelatin/polyethylene bilayer films for active packaging. Food Packag Shelf Life 18:201–211
56. Ferreira AS, Nunes C, Castro A et al (2014) Influence of grape pomace extract incorporation on chitosan films properties. Carbohydr Polym 113:490–499
57. Kaya M, Ravikumar P, Ilk S et al (2018) Production and characterization of chitosan based edible films from *Berberis crataegina*'s fruit extract and seed oil. Innov Food Sci Emerg Technol 45:287–297
58. Madera-Santana TJ, Freile-Pelegrín Y, Azamar-Barrios JA (2014) Physicochemical and morphological properties of plasticized poly(vinyl alcohol)-agar biodegradable films. Int J Biol Macromol 69:176–184
59. Nogueira D, Martins VG (2019) Use of different proteins to produce biodegradable films and blends. J Polym Environ 27:2027–2039
60. Mathew S, Jayakumar A, Kumar VP et al (2019) One-step synthesis of eco-friendly boiled rice starch blended polyvinyl alcohol bionanocomposite films decorated with in situ generated silver nanoparticles for food packaging purpose. Int J Biol Macromol 139:475–485

Publisher's Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.