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ORIGINAL ARTICLE

Design and fabrication of polysaccharide based excellent chemical resistant and UV barrier ternary blend films for green packaging applications

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Abstract The development of green materials for active packaging applications is a research hotspot due to setbacks of petrochemical derived plastics. Thus, the present study aims to develop ternary blend films by doping different wt% of Tragacanth gum (TG) to Poly(vinyl alcohol)/Chitosan (PC) blend using solvent evaporation technique. Further, their various physicochemical properties were evaluated systematically. Differential scanning calorimetry studies revealed excellent compatibility and thermal stability of PC blend was significantly reinforced with 15 wt% of TG. UV–visible spectroscopy study demonstrated the excellent shielding efficacy of UV radiation by ternary blend films. Moreover, overall migration results confirmed the limited release of film constituents into food simulants and swelling ratio analysis indicated the good swelling resistance at higher wt% of TG. The ternary films exhibited tremendous chemical resistance against extreme acidic and basic environments and these green biofilms could be considered for active packaging applications.

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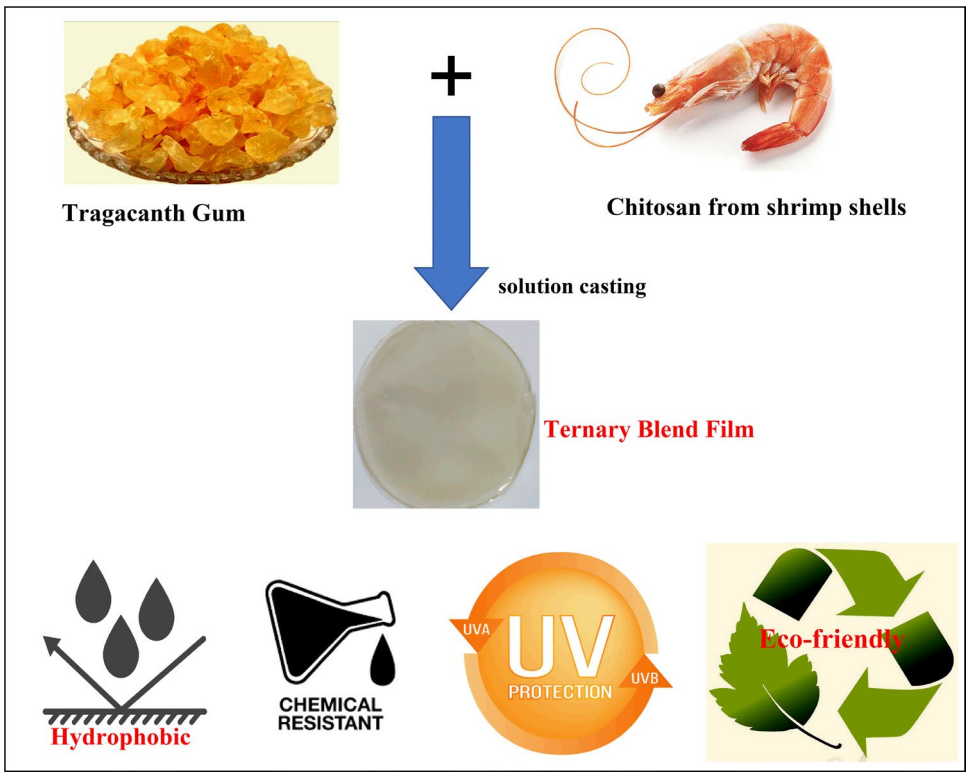
Graphical abstract



Keywords Chitosan · Tragacanth gum · Poly(vinyl alcohol) · Ternary blend films

Abbreviations

PVA	Poly(vinyl alcohol)
CS	Chitosan
PC	Poly(vinyl alcohol)/Chitosan
TG	Tragacanth gum
DSC	Differential scanning calorimetry
TGA	Thermogravimetric analysis

Introduction

Demand for consumer-friendly packaging material increased drastically as the pandemic shuts down restaurants and food-service outlets. The increase in demand from consumers is expected in the coming days for ready-to-eat food for which packaging material is needed. Thus, the development of consumer reliable and green packaging material is the need of the hour. The current commercially available petroleum-derived plastics are a serious threat to the global environment due to their non-biodegradability. So, the use of biodegradable polymer in the material formulation is gaining importance, Poly(vinyl alcohol) (PVA) is one such biodegradable biocompatible, hydrophilic, non-toxic and less expensive polymer (Yu et al. 2023; Das et al. 2019). But PVA lacks the biological activity by default and higher water vapor permeation and slow degradation rate limit its usage in the packaging area to some extent (Goudar et al. 2020a, b; Goudar et al. 2020a, b).

Chitosan (CS) is a biodegradable, cationic polysaccharide that attracted researchers in recent years due to its easy availability, unique antimicrobial activity, film-forming ability, high permeability to gases and nontoxicity (Bonilla et al. 2013; Haghighi et al. 2019; Yuvaraja et al. 2017; Qin et al. 2019). CS films are very fragile due to their rigid glucosamine units and inter/intramolecular interactions. The blending of CS with PVA is a good option to solve the issues of PVA and vice-versa. Various studies were conducted in the literature on PVA/CS blends. Bonilla et al. studied the influence of chitosan in different ratios on the physical and antimicrobial properties of PVA (Bonilla et al. 2013). Wu et al. studied the potentiality of CS/PVA blend for the inhibition of biofilm formation against *P. aeruginosa* PAO1 (Wu et al. 2018). S.Y. Park et al. studied the various physical properties of CS/PVA blend by using different solvents (Park, et al.

2001). Despite the pros of PVA/CS blends, PVA/CS blends have some cons too, such as low elasticity, low thermal resistance, limited miscibility and moderate antimicrobial activity (Bonilla et al. 2013, Narasagoudr et al. 2020).

Therefore, researchers developing the PVA/CS blend by doping a filler, active components, or another biopolymer to meet the needs of the industrial applications. M. Shojaee Kang Sofla et al. studied the effect of glycerol/polyethylene glycol on various physicochemical properties of PVA/CS blends (Kang et al. 2020). Jiang et al. studied the synergistic plasticization effect of $AlCl_3{\cdot}6H_2O$ and glycerol on CS/PVA blend $AlCl_3{\cdot}6H_2O$ as a solvent to dissolve CS (Jiang et al. 2016). Haghighi et al. developed a biodegradable film based on CS/PVA enriched with Ethyl lauroyl arginate (LAE) and studied the potentiality of the film as a packaging material (Haghighi et al. 2019). Ghaderi et al., studied the effect of fish gelatin on multifunctional properties of CS/PVA of various ratios (Ghaderi et al.2019).

But there are very limited reports in the literature using a natural polysaccharide to improve the multifunctional properties of PVA/CS blends. Tripathi et al. conducted a study to develop a chitosan based antimicrobial packaging material using PVA and Pectin as other supporting materials (Tripathi et al. 2010). Chetouani et al., investigated the physicochemical properties of CS/oxidized Pectin/PVA blend films (Chetouani et al. 2017).

Hence, the present study uses a natural anionic polysaccharide Tragacanth Gum (TG) as a supporting matrix for the PVA/CS blends. TG is a complex polysaccharide of branched acidic hetero-polysaccharides containing D-galacturonic acid. TG is edible, biocompatible, biodegradable, non-toxic, colorless, and odorless along with showing a good barrier toward oxygen transfer (Janani et al. 2020; Goudar et al. 2020a, b). TG migration from the bioplastic film into food is not harmful (Zare et al. 2019).

The current study hypothesizes that the functional groups of PC blend interact with –OH and –COOH groups of TG and might form a miscible blend & reinforcement of PC blend could be observed in the given composition range. Moreover, cationic and anionic interactions between CS and TG will be key factors in getting homogeneous miscible films. The present study aims to investigate the influence of TG on the multifunctional properties of the PC blend.

Material and methods

Materials

PVA (molecular weight = 1,15,000 & degree of saponification 98–99 mol %) was supplied by Loba, India. CS (75% deacetylated) with viscosity of 200 centipoises was obtained from Loba Chemicals, Mumbai. Tragacanth gum (TG) was received from Central Drug House, India. Glacial acetic acid was procured from Spectrochem, India. NaOH was procured from Merck India. Analytical grade HNO_3, Ethanol and Millipore water was used as received.

Methods

Preparation of films

To prepare ternary blend films, the different wt% of TG was dissolved in millipore water and stirred for 6 h, then filtered. Then TG in different wt% was doped into previously prepared PC blend solution and stirred for 12 h at room temperature (RT, 27 ± 3 °C). The clear and viscous solution was then cast onto Petri dishes for solvent evaporation at RT. The control PC blend was prepared by dissolving definite quantities of PVA and CS in millipore water and 2% acetic acid respectively and stirred at room temperature for 10 h. Further, homogeneous solution was cast onto cleaned and dried Petri dishes. After 5–6 days, the films were peeled off from the Petri dishes and stored in a desiccator until further use. The Thickness of films was measured using a digital micrometer (Mitutoyo, Japan). Three measurements were taken for each film and the value was expressed as average ± standard deviation.

Characterization techniques

Differential scanning calorimetry (DSC)

The effect of TG on glass transition temperature of PC blend films was analyzed using Differential Scanning Calorimetry (Instrument: DSC Q20 V24.10 Build 122 TA Instruments, USA). The film samples were sealed in an aluminium crucible and heated between 25 and 400 °C at 10 °C per minute in an inert atmosphere of N_2 gas (50 mL/min).

Thermogravimetric analysis (TGA)

To study the thermal stability of prepared films, TGA was carried using SDT Q600 V20.9 TA instruments. 5–10 mg of film was taken for analysis and heated up until 600 °C at a heating rate of 10 °C per minute under an inert gas (N_2) atmosphere (100 mL/min).

UV–visible spectroscopy

UV–visible spectroscopy was performed using PerkinElmer Lambda 365 spectrophotometer, UK, in the wavelength range of 200–800 nm. Further, 3 cm × 1 cm size of film was used for analysis taking air as reference. Equation (1) was

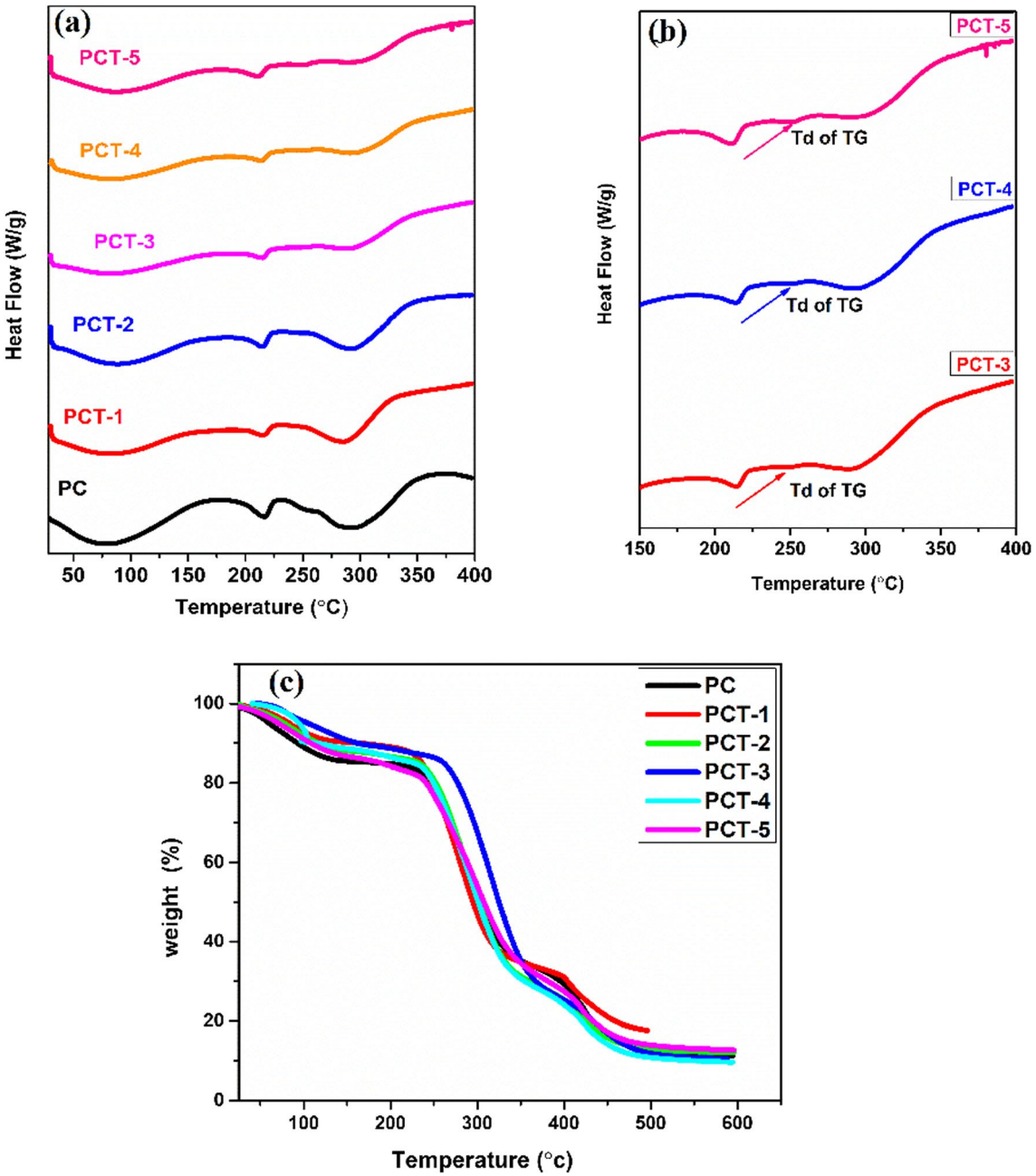


Fig. 1 **a** DSC thermograms of PC and PCT blend films. **b** DSC thermograms of PCT-3, PCT-4 & PCT-5 between the range 150–400 °C. **c** TGA thermograms of PC and PCT blend films

used to determine the opacity (at 600 nm) of films using absorbance data.

$$\text{opacity} = \textit{absorbance at } 600\ nm/b \quad (1)$$

where b is the average thickness of the film (mm).

Transparency was calculated using Eq. (2)

$$\text{transparency} = -log\%T/b \quad (2)$$

where, %T is the percentage of transmittance at 600 nm and b is the average thickness of the film (mm).

Swelling ratio

The Swelling index of prepared films in water was determined gravimetrically. Briefly, 2×2 cm^2 size of the samples was oven dried at 60 °C and weight was determined as W_1. Afterward, the samples were soaked in respective beaker

Table 1 Composition, thickness T_g, T_m, T_d and melting enthalpy (ΔH_m) of PC blend & PCT blends

Sample code	PVA (wt%)	CS (wt%)	TG (wt%)	Thickness (mm)	Glass transition temperature (T_g) (°C)	Melting temperature (T_m) (°C)	Decomposition temperature (T_d) (°C)	Melting enthalpy (ΔH_m) (J/g)
PC	75	25	–	0.06	53	216	292	35.2
PCT-1	75	25	5	0.07	57.5	216	286	44.2
PCT-2	75	25	10	0.06	60.5	215	292	38.1
PCT-3	75	25	15	0.08	58.5	214	290	33.9
PCT-4	75	25	20	0.08	57	214	290	29.7
PCT-5	75	25	25	0.07	57	211	290	33.6

Table 2 Thermal stability of PC and PCT blend films

Sample code	Weight loss (%) I	Weight loss (%) II	Weight loss (%) III
PC	13.7% (45–116 °C)	51.7% (246–320 °C)	22.1% (404–449 °C)
PCT-1	9.3% (60–112 °C)	56.8% (240–312 °C)	15.7 (399–442 °C)
PCT-2	12.1% (70–124 °C)	59.6% (250–325 °C)	15.8% (403–455 °C)
PCT-3	10.5% (68–121 °C)	64.3% (274–350 °C)	14.8% (422–474 °C)
PCT-4	11.3% (94–106 °C)	61.1% (248–326 °C)	17.6% (407–455 °C)
PCT-5	13.3% (54–124 °C)	56.2% (248–332 °C)	17.1% (411–439 °C)

with 40 mL of Millipore water. After at different time intervals (30, 60, 90, 120 and 180 min) samples were removed from the beaker, removed water drops using tissue paper and weight was determined as W_2. The swelling index (%) was determined using Eq. (3).

$$\text{swelling index (\%)} = \left(W_2 - W_1/W_2\right) \times 100 \quad (3)$$

Overall migration study

The release of TG from the PC matrix in 3 food simulants namely distilled water, 50% ethanol and 3% acetic acid was evaluated according to Indian Standard IS:9845 (1998 II revision). Distilled water mimics the aqueous food, 50% ethanol represents the alcoholic beverages whereas 3% acetic acid represents the acidic foods (pH < 5). Concisely, the pre-weighed samples were immersed in 40 mL of respective food simulant and stored for 10 days at 40 °C in a hot air oven. After 10 days the films were removed from food simulant and dried. The change in weight was determined and the amount of extractive was calculated using Eq. (4).

$$\text{Amount of extract (mg/kg or mg/l or ppm)} = M/V \times 1000 \quad (4)$$

where M is the weight change (mg) and V is the total volume of simulant (mL) used in each replicate.

Chemical resistance study

Chemical resistance of ternary blend films was studied using ASTM: D543-06 standard employing HNO_3 and NaOH as the model chemicals. Briefly, pre-weighed samples were immersed in respective chemicals for 24 h. Then they were removed from chemicals, washed with water and dried in an oven. Further, the weight change was determined and the percentage of weight loss/gain was evaluated.

Results and discussion

Differential scanning calorimetry (DSC)

The effect of insertion of TG into PC blend's crystalline behavior and melting phenomenon was evaluated using DSC and obtained thermograms are presented in Fig. 1a.

The glass transition temperature (T_g) and melting temperature (T_m) was determined and presented in Table 1. The PC blend has shown a single T_g at 53 °C and T_m at 216 °C which is comparable with previous research works (Bonilla et al. 2013; Vanjeri et al. 2019). For ternary system also there was only one T_g in the thermograms, assuring the miscibility of the system. The TG insertion has resulted in increase in T_g of PC blend from 53 to 57.5 °C. The increased T_g could be attributed to the enhanced interactions by insertion of TG. The hydrogen bonding between the hydroxyl, amine groups of PC blend and hydroxyl, carboxyl groups of TG lead to the increase in T_g of PCT blend films. The blending of TG with PC has decreased the T_m from 216 to 211 °C. The depression in Tm of neat blend is an evidence for the miscibility among the components (Naveen et al. 2010). Further, decrease in the melting

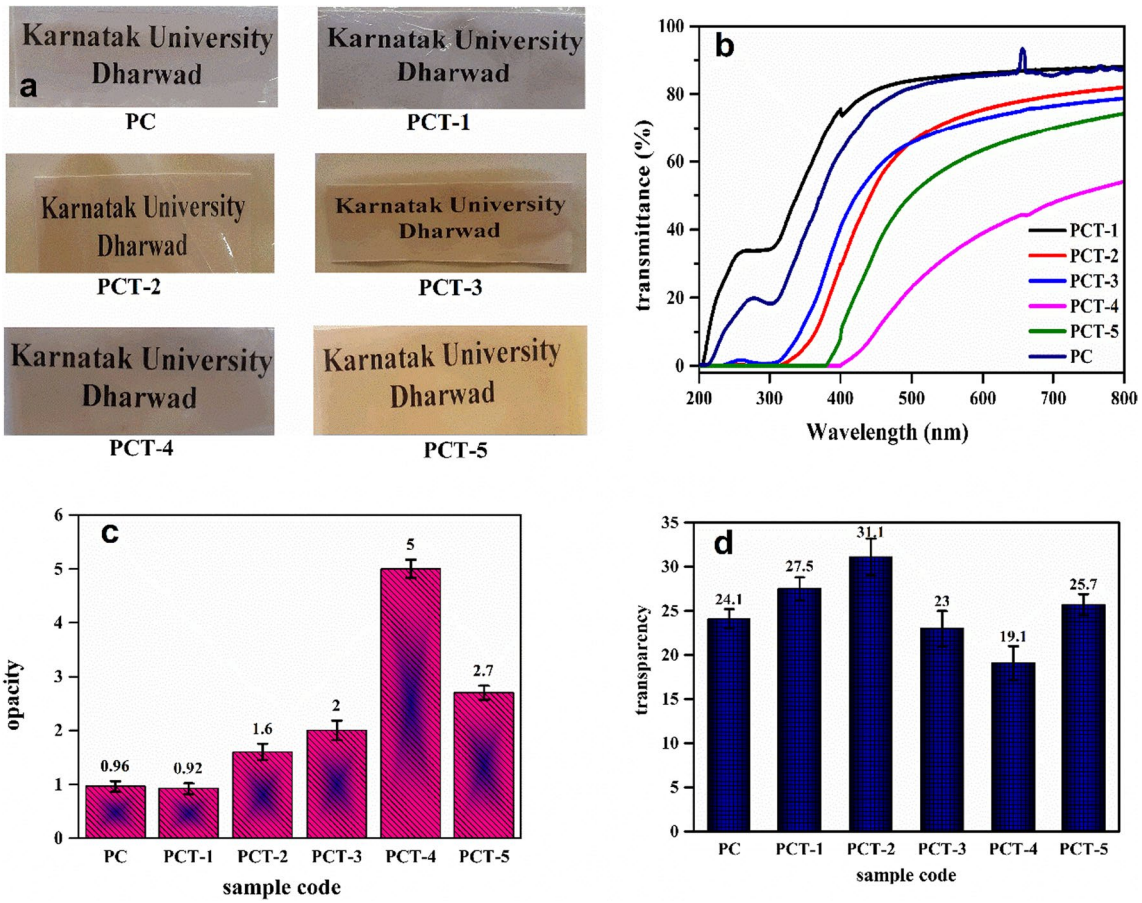


Fig. 2 **a** Visual appearance **b** Transmittance **c** Opacity and **d** Transparency of the prepared films

enthalpy (ΔH_m) of PC blend from 35.2 to 29.7 J/g on adding higher wt% of TG could be attributed to the destruction of crystalline arrangement in the matrix (Guirguis et al. 2012). In the DSC thermograms of PCT-3, PCT-4 & PCT-5, as shown in Fig. 1b, there was a peculiar peak around 250 °C, which is attributed to the decomposition of TG in matrix (Zarekhalili et al. 2017). This finding supports the heterogeneity in the matrix at higher wt% of TG. There were no significant changes in the decomposition temperature.

Thermogravimetric analysis (TGA)

The consequences of TG doping on the thermal stability of PC blend films were studied using TGA and obtained TGA thermograms are presented in Fig. 1c. Moreover, the detailed degradation values are given in Table 2. All the prepared films have exhibited a three step degradation process. The PC blend system has shown its first degradation pattern from 45 to 116 °C is attributed to the loss of moisture and physisorbed water molecules (Vanjeri et al. 2019).

The PCT blend also exhibited this behavior in the same range of temperature. The PC blend depicted its second degradation step from 246 to 320 °C which is due to the dehydroxylation, elimination of acetate groups of PVA and structural disintegration of CS (Kasai et al. 2019; Yang et al. 2016). The third decomposition temperature ranges from 404 to 449 °C is due to the degradation of polyene residues of PVA formed in the second step and gradual oxidative degradation of the carbonaceous residue of CS (Kasai et al. 2018; Lewandowska et al. 2009). The ternary blend film with 15 wt% of TG (PCT-3), its second degradation step onset temperature raised to 274 °C indicating better thermal stability. The offset temperature of the second degradation step increased from 320 to 350 °C indicating the thermal resistance of films to the degradation process. The increased thermal stability might be due to the increased physical interactions between the active groups of PVA, CS and TG. Overall, the added TG has a positive effect on the thermal stability of PC blend films at lower 15 wt% only.

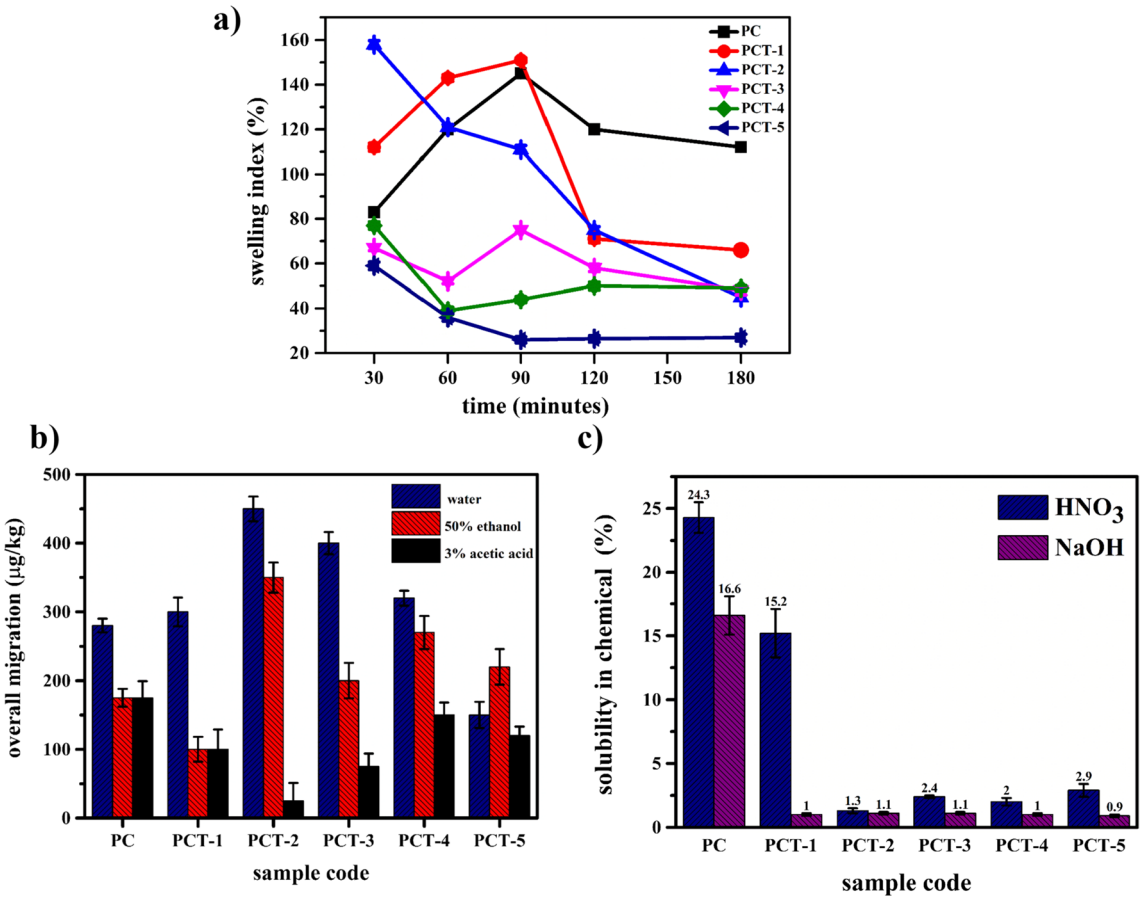


Fig. 3 **a** Swelling index (%) of the prepared films at different time intervals **b** Overall migration rate and **c** Chemical resistance of the prepared films

UV–visible spectroscopy

The effect of TG on UV rays blocking ability, transmittance, opacity and transparency of PC blend was evaluated and obtained results are presented in Fig. 2a–d). The visual appearance of the developed films is given in Fig. 2a. As the wt% of TG increased in the matrix, the color of the film transformed to light brown color. Further the transmittance (%) in the visible range for neat PC and PCT-1 was above 80% indicating high transmission of visible light through the matrix. But the UV blocking ability of these two films was not satisfactory considering the end application of the prepared materials. As the wt% of TG increased in the matrix, the propagation of visible light through the matrix was diminished indicating the increased opaqueness compared to PC blend. Moreover, UV blocking efficacy of the TG loaded films improved significantly except for PCT-1, specially PCT-4 exhibited 100% UV shielding efficiency in the UV region (200–400 nm). Apparently, the reinforcement in UV shielding ability of PCT-4 and PCT-5 could be attributed to the absorption of UV light by carbonyl groups (C=O) of TG leading to enhancement of $n \rightarrow \pi^*$ absorption (Li et al. 2014). Unfortunately, PCT-4 has sacrificed its transmittance (%) to a great extent compared to the PC blend.

Opacity

Opacity measurements were carried out at 600 nm and obtained findings are presented in Fig. 2c. The incorporation of TG has increased the opacity significantly reducing visible light transmission except for PCT-1. Since, PCT-1 has shown more transmittance and less opacity affirms the homogeneous distribution of TG at 5 wt% in matrix. Further, increased opacity of all other films might be due to rigid compact morphology and contraction of the films.

Transparency

The impact of TG insertion into PC matrix was evaluated systematically at selected wavelength (600 nm) and

results are presented in Fig. 2d. At lower wt% of TG, the prepared ternary films depicted more transparency than pure blend, but at higher wt% of TG transparency was decreased. The decreased transparency at higher wt% of TG could be attributed to the heterogeneity by aggregation of excess TG in the matrix thus reducing the interchain spacing eventually hindering the passing of visible light through the matrix. Further the current findings are in line with other researchers (Janani et al. 2020). Surprisingly, the visual appearance of some of the films are contradictory to the transparency findings because of thickness variation from film to film.

Swelling ratio

Water holding capacity of the prepared blend films was determined by calculating swelling ratio of the films at different time intervals (30, 60, 90, 120 & 180 min). Further, the influence of TG on binary PC blend was scrutinized and obtained results are presented in Fig. 3a. The pristine PC blend has exhibited a linear swelling ratio with time up to 90 min, and thereafter the blend reached the equilibrium state. The PC has shown highest swelling ratio of 145% at 90 min indicating the blend has significant ability of water holding. The incorporation of hydrophilic natural polysaccharide into PC matrix enhanced the swelling ability to some extent at lower wt% of TG (5 & 10 wt%). Further, 15, 20 & 25 wt% of TG doped blend films exhibited decrease in swelling ratio compared to neat PC blend this might be attributed to their compact network type microstructure. Further the filling of voids of PC blend by TG, decreases the free volume between interchain, eventually decreasing the water holding capacity. To conclude, added TG at lower wt% has reinforcing effect on swelling behavior of PC blend.

Overall migration

The ideal packaging material in contact with food should be inert and should not release the constituents into food in unacceptable quantities (Narasagoudr et al. 2019). The release of constituents from the matrix was studied gravimetrically and obtained results are presented in Fig. 3b. According to the Indian standard (IS: 9845, 1998s revision), the overall migration of constituents from packaging material to food in contact should not exceed 60 mg/kg. From the figure, it is evident that overall migration in water is much higher than 50% ethanol and 3% acetic acid, this might be attributed to the readily water soluble nature of PVA and TG. Further, PCT-2 has a maximum overall migration in water (450 µg/kg) and in 50% ethanol (350 µg/kg) indicating the increased migration of constituents from matrix compared to blend. Moreover, the PC blend has presented high overall migration in 3% acetic acid compared to ternary films indicating the migration of CS from the matrix. But ternary films exhibited comparatively fewer migration in 3% acetic acid, this is corresponding to the compact microstructure that inhibits the release of constituents from the matrix. In 3% acetic acid, the developed films exhibited less migration values compared to water and 50% ethanol indicating the excellent acid resistance ability. Though the ternary films exhibited increased overall migration of constituents compared to blend, they are well within the range described for a food packaging material by Indian standard and European Union standards (60 mg/kg).

Chemical resistance study

The influence of different wt% of TG on the chemical resistance of PC blend was examined and obtained results are presented in Fig. 3c. The resistance of the prepared biofilms in HNO_3 and NaOH was studied to evaluate the stability in acidic and basic environments respectively. From the figure, it is evident that the neat PC blend has demonstrated $24.3 \pm 1.2\%$ and $16.6 \pm 1.5\%$ solubility in HNO_3 and NaOH depicting the moderate stability to acidic and basic environmental conditions. Further, PCT-1 has presented $15.2 \pm 1.9\%$ solubility in HNO_3 indicating moderate resistance against the acidic conditions. But in NaOH it presented only $1 \pm 0.1\%$ solubility, exhibiting excellent resistance to basic conditions. Moreover, as the TG wt% increase in the matrix, the resistance towards acidic environment increases, this might be attributed to the compact microstructure of films and excellent chemical resistance of TG (Zare et al. 2019). In basic conditions also the TG incorporated films were maintained the structural integrity without any visible deformation. To conclude, the incorporation of TG in different wt% into the PC blend has shown tremendous enhancement in chemical resistance.

Conclusion

The successful fabrication of different wt% of TG included PC blend films was carried out and their multifunctional properties were systematically evaluated utilizing various characterization techniques. DSC measurements depicted excellent miscibility by exhibiting a single glass transition temperature in the given composition range. Further, the thermal stability of PC blend film was significantly elevated by doping 15 wt% of TG. Moreover, the fabricated films were efficient UV blockers without losing significant transparency at 15 and 25 wt% of TG. The ternary blend films were more swellable at 5–10 wt% of TG and less swellable at 15, 20 and 25 wt% of TG. The overall migration results

in 3 food simulants were well within the limit framed by Indian standards for food packaging materials. The prepared green biofilms exhibited excellent chemical resistivity against extreme acidic and basic conditions. Thus, we strongly endorse the ternary films with 15, 20 and 25 wt% of TG could be considered for active packaging of alcoholic and acidic foods in extreme acidic and basic environments.

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Author contribution All the authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by NG, VDH, OJD, JPP, SPM, RBC. The first draft of the manuscript was written by NG and all the authors commented on previous version of the manuscript. All authors read and approved the final manuscript.

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Data availability All data generated or analyzed during this study are included in the article.

Code availability Not applicable.

Declarations

Conflict interest The authors have no relevant financial or non-financial interest to disclose.

Ethics approval Not applicable.

Consent to participate Not applicable.

Consent for publication Not applicable.

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