Synthesis and characterization of paddy straw chitosan nanocomposite as an efficient photocatalytic bio-adsorbent for the removal of rhodamine B and malachite green dye from aqueous solution

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Abstract
Current study deals with an agriculture waste paddy straw (PS) incorporated with chitosan (CS) to prepare photocatalytic paddy straw chitosan nanocomposite (PS-CSNC) with cross-linking agent sodium tripolyphosphate was investigated and catalysis-based photocatalytic adsorptions of Rhodamine B (RhB) and Malachite green (MG) in aqueous solution under UV irradiation were also examined. Moreover, this study examined that the factors influence the adsorption process, such as catalyst concentration, pH and temperature, using kinetic models. Additionally, the composite characterization was achieved by several analysis, such as UV–visible spectroscopy, Fourier-transform infrared spectroscopy, X-ray diffraction analysis, thermogravimetry/differential thermal analysis and field effect scanning electron microscopy. The study shown that the optimized concentration of PS-CSNC catalyst (3.0 g L⁻¹), below neutral pH (6.5) of the solution and temperature 60 °C achieved excellent removal of both dyes. Initial sorption process involved the removal mechanism of both RhB and MG followed by photocatalytic adsorption. The process of adsorption was well adapted with pseudo second-order kinetics. Overall results suggested that chitosan-based agriculture waste paddy straw nanocomposite could be an eco-friendly and cost-effective bio-adsorbent for removal of RhB and MG in wastewater treatment.

Keywords Chitosan · Paddy straw · Nanocomposite · Rhodamine B · Malachite green · UV irradiation

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Introduction

Water pollution is one of the most grave worldwide issues; however, industrial effluent is main source of water pollution (Ado et al. 2015). Day by day, enormous quantity of industrial effluent discharged into aqueous stream, and this seriously influences the strength of every single living life form and also ecological system. Several industries, namely cosmetics, food, leather, textile, plastics, printing, pharmaceutical, painting and photographic industries, correspond to the main sources of water pollution along with enormous quantities of dyes into the surroundings (Hassaan and Nemr 2017).

In 1990s, more than 10 million different dyes were synthesized commercially in 1990, worldwide. Roughly, industries discharged 8–20% of utilized dyes to aquatic environment, which includes toxic dyes also (Sharma et al. 2019). Because of its ecological impact, polluted wastewater along with dyes is supposed to be an ample management before discharging. Numerous treatments methods, namely flocculation, chemical oxidation, and membrane separation, have been used to decrease full amount of dyes in industrials wastewater. On the other hand, these technologies show more disadvantages when compared to biological treatment and are poorly reusable (Ghorai et al. 2014).

Rhodamine B and Malachite green are familiarly used in textile industries. These dyes are stable and degradation resistance due to their complex structural properties (Ali et al. 2015; Inyinbor et al. 2017) which exhibited carcinogenic and mutation properties (Ali et al. 2019). A textile industries wastewater effluent has harmful effects on fauna and flora in the aquatic eco-system. Removal of these deleterious dyes before discharging to water system is compulsory for the animal health and environmental protection.

Numerous techniques are employed for the treatment of dyes for example adsorption, photocatalysis, chemical precipitation, electrochemical technologies, ion exchange, and membrane filtration (Qasem et al. 2021; Shrestha et al. 2021; Xu et al. 2021). Adsorption followed by photocatalysis was initiated to be more hopeful due to their efficient adsorption, removal and degradation facilities in very less time intervals (Ahmed and Iqbal 2022). Among these techniques is a well-known method with several advantages over conventional methods is simple, eco-friendly and cost-effective under sunlight or UV irradiation. Diverse resources have been used as adsorbents for removal dyes stuffs, namely carbon-based composite, silica-based composite, cellulose composite and chitosan-based composite (Aslam et al. 2019). Diverse adsorbents have been employed for dye degradation from aqueous system, such as animal bone meal, sugarcane pulp and egg shell (Akar et al. 2013; El Haddad et al. 2012; Slimani et al. 2014).

Among the various sorbents used, chitosan would be the mainly suitable for producing adsorbents for dyes (Sharijazi et al. 2021).

Chitosan (CS) is one of the hydrocolloid and biopolymeric substances and it has potential application as heavy metals and dyes adsorbent (Liu et al. 2019). CS (Fig. 1B) has substantial quantities of –NH2 and –OH groups owing for wastewater treatment. Biopolymer is a linear positively charged polysaccharide and its amino groups are responsible for its adsorption capacity and act as adsorbent (Samuel et al. 2020; Sathiyanival et al. 2020; Scialla et al. 2019). On the other hand, it has some disadvantages of using CS alone for adsorption and are solubility in acids, ability to undergo clumping, weak properties, and difficult to separate, whereas, chitosan is combined with other materials, such as chitosan-based material gels, flakes, and powders, was also used (da Silva Alves et al. 2021; Wan Ngah et al. 2011; Wu et al. 2010).

Paddy straw (PS) is composed of 30–40% cellulose, 20–35% hemicelluloses and 15–25% lignin (Ruiz et al. 2013). Yearly, billions of tons of grain straws have been produced in the world mainly in the developing countries, but only a small part of them are used as animal feed, biogas production, biofuel, hemicellulose and lignin extraction, and isolation of cellulose, while most of them are burned (Hassan et al. 2011). Nanocomposites (NC) are formed by the mixture of two or more resources that have somewhat dissimilar properties. We hypothesized that a composite with a pore structure, on the surface, and strength of the materials can be synthesized from paddy straw and chitosan via the dissolution process. Paddy straw rich in cellulose, makes strength of the composite, while chitosan offers the dye-adsorbing sites. Therefore, in this study, the agriculture waste paddy straw and chitosan are the two different resources that work mutually to give the sole property of the composite. Moreover, paddy straw chitosan nanocomposite (PS-CSNC) has good thermal stability (Hassan et al. 2012).

Hence, in the present study, it has been aimed for the synthesis of industrially important chitosan-based paddy straw nanocomposite as catalytic material, which could be offering for the removal of RhB and MO dyes. Parameter optimizations were examined by catalyst concentration, solution pH and temperature for photocatalytic adsorption of dye. The percentages of dye removal were also calculated and reported.

Materials and methods

Chemicals required

Chitosan and sodium tripolyphosphate were supplied by Sigma Aldrich, while rhodamine B and malachite green.
were supplied by HiMedia Laboratories. All the chemicals are analytical grade, which were used for in this study.

**Collection and processing of paddy straw**

PS was collected from local paddy field at Tirunelveli, Tamil Nadu, India. Collected paddy straws were rinsed thoroughly by single-distilled water followed by double-distilled water to eliminate unwanted dust particles, chopped into small pieces and shade-dried at room temperature (Fig. 1A). Above 50 g of dried paddy straw was powdered.

**Preparation of paddy straw biomass**

Powdered PS (10 g) was added into the 100 ml of double-distilled water under vigorous stirring for 4 h. The mixture was mixed by ultra-sonication for 20 min at room temperature. The solution filtered through filter paper and stored in 4 °C and used within a week (Li et al. 2017).

**Preparation of paddy straw chitosan nanocomposite**

CS (1 g) was dissolved in acetic acid (1%). PS biomass (10 mL) was added slowly into CS precursor solution under stirring, followed by sodium tripolyphosphate (TPP) (8 mL; 0.1%) added into the reaction mixture drop-wise. The suspension solution incubated at 70 °C for 4 h in water bath. Finally, PS-CSNC was separated from the reaction solution by centrifugation at 7000 rpm for 20 min and allowed to dry overnight to attain chitosan paddy straw nanocomposite.

**Preparation of adsorbate**

Commercial quality RhB and MG utilized in this present study (without further purification). Structure of adsorbate RhB and MG is illustrated in Fig. 1C, D, respectively. A 500 mg L⁻¹ stock solution was prepared for both adsorbates; furthermore, it was diluted at different concentrations prepared with constant volume of double-distilled water to required concentration.

**Characterization of chitosan nanocomposite**

**UV–visible spectroscopy (UV–visible) analysis**

UV–visible spectroscopy is an analytical instrument to find the formation of nanocomposite. Spectra of the chitosan nanocomposite were recorded from 200 to 500 nm wavelength using UV–visible spectroscopy (UV–Visible Perkin Elmer Lambda).
Fourier-transform infrared spectroscopy (FTIR) analysis

Fourier-transform infrared spectroscopy (FT-IR Perkin Elmer) is an essential instrument to identify the functional groups in the composite, which is done with KBr pellets. FTIR was performed for CS and PS-CSNC and the spectra were normalized and ranging from 400 to 4000 cm\(^{-1}\) wavelength.

X-ray diffraction (XRD) analysis

X-ray diffraction analysis is used to identify the crystallinity of CS and PS-CSNC. Spectrum patterns of the XRD were recorded using D5000 X-ray diffractometer, Siemens with Cu Kα radiation (\(\lambda = 1.5406 \, \text{Å}\)) in the 2θ ranging from 10° to 80°.

Thermo-gravimetric/differential thermal analysis (TG/DTA) analysis

Thermo-gravimetric/differential thermal (Perkin-Elmer TGA 7-thermal analyzer) analysis is used for studying thermal stability of PS-CSNC. The heating ranged from 40 to 700 °C at rate of 10 °C min\(^{-1}\) under nitrogen environment (flow rate: 20 mL min\(^{-1}\)).

Field effect scanning electron microscopy (FESEM) analysis

Field emission SEM (Hitachi FESEM SU6600) is an instrument used for investigating the morphological pattern of PS-CSNC. The morphological pattern is scanned by increasing magnification and adapted with 500 nm.

Photocatalytic reaction

The photocatalytic reactor was planned on fluidized bed idea, with three-stage contact design and furthermore maximizing UV illumination area. The progress of water and air treatment system is dependent on heterogeneous photocatalysis and eco-friendly (Ray and Beenackers 1998). The three-stage fluidized bed reactor consists of 8 W lamp (4 No.) placed on the two each side. From feed tank through pump transports the feeds to the reactor. Four outlets from feed tank were placed on behind feed tank. The aeration tubes are connected with header and placed on lower compartment. The reaction process was done under room temperature for 2 h with different concentrations of feed and catalyst. Biosynthesized PS-CSNC is used for rhodamine B (RhB) and malachite green (MO) dye removal. Supernatant was collected from aliquot and filtered, furthermore estimated the residual dye by UV–visible spectrophotometer at 554 and 620 nm, respectively. The photocatalytic removal percentage was calculated using Eq. (1) (Alshabanat and Al-Anazy 2018):

\[
\text{Dye removal} (\%) = \frac{D_0 - D_e}{D_0} \times 100, \tag{1}
\]

where, \(D_0\) is initial dye concentration (\(t=0\)); \(D_e\) is final dye concentration after the treatment at specific time interval.

Results and discussion

Characterization of chitosan nanocomposite

UV–visible spectroscopy (UV–visible) analysis

UV–visible spectrum of PS-CSNC is shown in Fig. 2. Exactly, PS-CSNC exhibited peak at 258 nm. Accordingly, UV–visible spectrum of CS exhibited peak at 226 nm (AbdElhady 2012). It conforms to formation of chitosan nanocomposite. Attachment of CS with PS showed bathochromic shift. The shift exhibited formation of PS-CSNC in nano-size (Suneetha 2018).

Fourier-transform infrared spectroscopy (FTIR) analysis

FTIR spectrum of chitosan and paddy straw-mediated synthesized chitosan nanocomposite is displayed in Fig. 3. FTIR was used to identify amine linkage between paddy straw and chitosan. In the spectrum of chitosan (Fig. 3A), were found leading peaks at 3305, 2185 and 1645 cm\(^{-1}\). The peak at 3305 cm\(^{-1}\) in chitosan corresponds to combine –NH\(_2\).
and –OH group stretching. The –CONH$_2$ group existed and peaked at 1645 cm$^{-1}$.

The peak at 2185 cm$^{-1}$ corresponds to symmetric –CH$_2$ stretching. In the spectrum (Fig. 3B) of paddy straw, chitosan nanocomposite existing peaks were at 3360, 1719, 1605, 1315, 1038 and 809 cm$^{-1}$. The peak 1719 cm$^{-1}$ found to be attributed to stretching vibrations of C–N and N–H. The leading peaks existed at 1605 and 1038 cm$^{-1}$. These peaks have stretch vibration form –NH bending of NH$_2$ and the C=O of –NHCONH–, respectively. The peak responsible for both –OH and –NH$_2$ stretching at 3305 cm$^{-1}$ in chitosan is larger and the peak 3360 cm$^{-1}$ in chitosan nanocomposite displayed narrow peak, which indicates interaction of intermolecular hydrogen bonding between paddy straw and chitosan molecules (Qiu et al. 2019). Similar findings are in good relation with our studies that the peaks at 1038 cm$^{-1}$, which are attributed to –CO stretching in 4-glycosidic linkage, found in chitosan nanocomposite (Zeng et al. 2015).

Formations of paddy straw-mediated synthesized chitosan nanocomposite were confirmed by FTIR analysis. Ye et al. (2014), reported that boarder peak existed at 3309 cm$^{-1}$ in chitosan; similarly in our study, chitosan showed peaks at 3305 cm$^{-1}$, which is attributed to –NH$_2$ and –OH group stretching in chitosan.

**X-ray diffraction (XRD) analysis**

XRD diffractogram patterns of chitosan and paddy straw-mediated synthesized chitosan nanoparticles are shown in Fig. 4. The physical properties, such as crystalline structure and compatibility of each material component in blended films, are described by XRD analysis (Su et al. 2012). Average crystalline sizes of the PS-CSNC were found from 120 to 220 nm by Scherrer’s calculation. Chitosan (Fig. 4A) exhibited at $2\theta = 18.55^\circ$, 20.95$^\circ$ and 26.07$^\circ$. The XRD spectrum pattern of board peak was at $2\theta = 31.88^\circ$. The chitosan showed its diffraction peak at $2\theta = 20.95^\circ$ (curve A) corresponding to plane (100) of an orthorhombic crystal (Banik et al. 2012). Similar XRD diffractogram pattern was stated by Fukuoka et al. (1993).

Paddy straw-mediated synthesized chitosan nanocomposite (Fig. 4B) XRD diffractogram pattern exhibited leading peaks at $2\theta = 28.33^\circ$, 44.65, 47.47 and 56.65$^\circ$. The XRD diffractogram peaks at $2\theta = 18.55^\circ$, 20.95$^\circ$ and 26.07$^\circ$ (curve A) of chitosan were disappeared to stretch at $2\theta = 28.33^\circ$. 
(curve B) in nanocomposite during the formation of chitosan nanocomposite. The XRD diffractogram peaks at $2\theta = 28.38^\circ$ and $44.65^\circ$ in chitosan nanocomposite found that they insensitively and sharply descend. Therefore, crystallinity of the blended polymer had much increased (Gokila et al. 2017). The results confirm the biosynthesized chitosan nanocomposite and it can be used for wastewater remediation due to presence of high thermal stable crosslinking agent TPP. Additionally, it suggested molecular level dispersion of paddy straw and amorphous crystalline nature of paddy straw chitosan nanocomposite (Banik et al. 2012).

Thermo-gravimetric/differential thermal analysis (TG/DTA) analysis

Thermo-gravimetric analysis (TGA) is used to measure the mass changes in nanocomposite, which is associated with decomposition and oxidation as a result of mass gain and loss. TG analysis (Fig. 5A) of the chitosan nanocomposite was measured by simultaneous thermal analysis. Chitosan nanocomposite degradation starts at 200 °C and it has broad degradation temperature at 650 °C with maximum char yield. Chitosan nanocomposite degradation showed three stages of mass loss (Fig. 5A).

First- and second-stage thermal degradation of chitosan nanocomposite exhibited ranging from 200 to 250 °C and 300 to 330 °C, which corresponded to evaporation of moisture and decomposition of polymer, respectively (Unnikrishnan et al. 2011). The temperature increased from 200 to 250 °C, TGA curve displayed 10% (1.72 µg) weight loss of PS-CSNC due to evaporation of moisture content, 50% (8.7 µg) weight loss was observed at 300 to 330 °C due to decomposition of polymer. As increased temperature from 450 to 500 °C in third stage, 70% (13.7 µg) weight losses were obtained from PS-CSNC due to char formation, as displayed in Fig. 5A (Rana et al. 2010). In third stage, weight loss of PS-CSNC is 93% (16.213.7 µg), indicating presence of PS component.

Differential thermal analysis (DTA) curves for PS-CSNC are displayed in Fig. 5B. In PS-CSNC DTA curve, two endothermic peaks were appeared at 105 and 217.1 °C. Not only that, DTA curve showed short exothermic peak at 261.6 °C and broader exothermic peak at 444.1 °C, which is probably corresponding to PS-CS nanocomposite, as displayed in Fig. 5B (Anitha et al. 2011). However, PS-CSNC absorbent meets the thermal qualification as an excellent absorbent for RhB and MG dye removal from wastewater because the wastewater treatment was carried out under room temperature.

Field effect scanning electron microscopy (FESEM) analysis

Morphological structure of PS-CSNC was visualized under the field emission scanning electron microscope (Fig. 6). The composite sample was diluted by deionized water before the analysis. Structural analysis of PS-CSNC
discerned as spherical shape and average size of 100 nm. Similar results are also found in chitosan nanoparticles at early research (Hassan et al. 2018). The dissimilar morphological structures of PS-CSNC were formed by cross-linking agent TPP. The effective assimilation of PS into CS exhibited spherical-shaped and distributed in single population without any aggregation, which is similar to the previous study investigated by Ariff et al. (2017).

**Photocatalytic removal of dye**

Photocatalytic adsorption activity of PS-CSNC was investigated by RhB and MG removal. The photocatalytic removal action was carried out under UV irradiation. The study revealed 96.04% of RhB and 98.15% of MG removal under the UV irradiation at the end of 1 h as in Fig. 7. The results shown from 10.0 mg L\(^{-1}\) of RhB and MG at 3 g L\(^{-1}\) PS-CSNC were used, the reaction was done in photocatalytic chamber at 60 °C. While delayed removal reaction was observed at low temperature. Maximum removal efficiency (82.4%) was observed in methyl orange with treatment of 0.5 g L\(^{-1}\) TiO\(_2\)/MgO/Chitosan hydrogel concentration at 30 min (Arikal and Kallingal 2019). Similar investigation showed highest percentage of dye removal (99.1%) due to increase the catalytic concentration, which contributed maximum number of photons adsorbed and maximum number of dye molecules (Chiou and Li 2003).

**Effect of bio-catalyst concentration**

The amount of catalyst was optimized based on de-colorization as showed in Fig. 8. Concentration of catalyst amount on removal in RhB and MG by varying from 1.0 to 3.0 g L\(^{-1}\) was studied. It displayed that the percentage of removal increased with increase amount of PS-CSNC concentration (Fig. 8). When RhB and MG dye molecules are absorbed by PS-CSNC, no rate of removal was observed at above the level of concentration. Maximum level of RhB dye removal (95.34%) was obtained for 3.0 g L\(^{-1}\) of PS-CSNC treatment at 120 min. Additionally, minimum level of removal (93.01%) was found 1.0 g L\(^{-1}\) of catalyst. The accessibility
of surface amine groups on PS-CSNC increased its removal of action (Sadiq et al. 2020). Similarly, CS/MgO composites for RhB removal being 0.14 g L\(^{-1}\) were reported previously (Nga et al. 2020). Initial concentration delivered driving force to overcome mass transfer resistance of RhB between liquid and solid phase (Inyinbor et al. 2017).

Meanwhile, 97.75% of MG was found with 2.0 g L\(^{-1}\) of PS-CSNC at 120 min treatment. Obtained results showed the lesser amount of catalyst is needed for MG removal. Therefore, this amount of catalyst is fixed as optimum amount that is used for further studies. Furthermore, crosslinking CS composite has good mechanical strength, heat tolerance and chemical stability (Szeto et al. 2017). However, there is no considerable incremental removal with increasing amount and can be attributed to dye molecules between the solution and catalyst (Arumugam et al. 2019).

**Effect of pH**

Significantly, pH affects the removal performance of photocatalytic adsorption (Szeto et al. 2014). Percentage of increment was observed by different pH of 5.8, 6.2 and 7.8 with RhB-PS-CSNC and MG-PS-CSNC system as in Fig. 9. Maximum RhB (93.01%; 3.0 g L\(^{-1}\); 100 min) and MG (96.82%; 3.0 g L\(^{-1}\); 10 min) removal percentages were recorded at pH 6.2. While, after RhB, removal percentage dropped significantly. However, for both RhB and MG PS-CSNC system, optimum adsorption was found at pH 6.2, while the percentage of removal is decreased for RhB at 100 min after (Fig. 9A) and saturated for MG after 120 min photocatalytic removal (Fig. 9B). Whereas, 2,3-dihydroxy-benzaldehyde functionalized chitosan composite can be able to be adsorbed at pH 8 for RhB (Alakhras et al. 2021) and CS/MgO composite can be able to be adsorbed at pH 7 for MG at 120 min (Nga et al. 2020).

At minimum pH, catalyst surface is highly positive. An abhorrence between cationic dye molecules (RhB) and positive charged catalyst surface expressed low adsorption (Inyinbor et al. 2017). As the pH (< pH 6.2) increased, the level of removal is decreased due lack of OH formation (Hasmath Farzana and Meenakshi 2014). RhB ions are monomeric molecular from at below neutral pH, which could be simply diffused into PS-CSNC. Whereas, previous studies reported not-significant high-removal capacity of MG at pH 8 and pH 9 for choline chloride–urea and choline chloride–glycerol, respectively (Sadiq et al. 2020).

Meanwhile, pH is higher, zwitterion of MG is formed in water. The aggregation of zwitterion from is because of electrostatic interaction between carboxyl and xanthene group of monomer (Lin et al. 1999). Below the natural pH, the preponderance of hydroxyl group generates a competition between \(-\text{N}^+\) and \(-\text{COO}^-\) and decreases the aggregation of MG. Therefore, adsorption of MG ions was increased in carbon surface (López Arbeloa and Ruiz Ojeda 1982). In all the pH treatment conditions, PS-CSNC showed maximum rate of adsorption of MG at pH 6.2. Therefore, the result concluded the optimum level of pH for further studies.

**Effect of temperature**

The photocatalytic removal reaction is very sensitive to temperature. The effect of temperature with UV irradiation is in the range from 30 to 60 °C as illustrated in Fig. 10. The result shows that the maximum temperature was...
conforming to optimal catalytic removal rate of reactive dyes. The maximum thermal energy existed in dye molecules for sufficient removal. Maximum rate of removal was observed on RhB (96.04%; catalyst = 3.0 g L⁻¹) and MG (98.15%; catalyst = 3.0 g L⁻¹) at 60 °C in 120 min reaction. Instead, RhB rates of removal were decreased with *Irvingia gabonensis* endocarp-coated chitosan composite from 80.00 to 26.57 mg g⁻¹ at 27–60 °C. This is due to redaction in binding force between catalyst and adsorbate (Inyinbor et al. 2017). Adsorption of MG with coir pith activated carbon, chitosan and sodium dodecyl sulfate composite showed decreased rate of removal due to increasing temperature accredited to weakening of bond between MG and PS-CSNC (Chowdhury and Saha 2010). Moreover, the solubility of reactive dyes was increased with increase of temperature. Therefore, reaction speed was increased with increase of temperature. From the result, optimum temperature was found at 60 °C, which is fixed for further removal studies.

**Adsorption kinetics**

Pseudo-first-order and pseudo-second-order kinetic modals were applied for their effect on RhB and MG adsorption rate on PS-CSNC; meanwhile, catalytic concentration, solution pH and temperature were investigated by Lagergren’s pseudo-first-order and pseudo-second-order modal. Pseudo-first-orders described adsorption rate based on the capacity of adsorption (Ho 2004).

Linearized form of pseudo-first-order is described Eq. (2):

\[
\log(q_e - q_t) = \log(q_e) - \frac{k_1 t}{2.303},
\]

where, \(k_1\) is the adsorption rate constant of pseudo-first-order, \(q_e\) is the amount of dye absorbed on to PS-CSNC at equilibrium (mg g⁻¹) and \(q_t\) is the amount of dye adsorbed on to PS-CSNC at specific time \(t\) (mg g⁻¹).

Linearized form of pseudo-second-order is described Eq. (3):

\[
\frac{T}{q} = k_2 q_e^2 + \left( \frac{1}{q_e} \right) t,
\]

where, \(k_2\) is the adsorption rate constant of pseudo-second-order, \(q_e\) is the amount of dye absorbed on to PS-CSNC at equilibrium (mg g⁻¹) and \(q_e^2\) is pseudo-second-order adsorption rate constant (g mg⁻¹ min⁻¹).

The first-order values \(k_1\) are listed in Table 1 and obtained from plot \(\log(q_e - q_t) vs. t\) for PS-CSNC with concentration of catalyst, pH and temperature as illustrated in Figs. 11, 12 and 13. Similarly, second-order rate constant \(k_2\) is listed in Table 2 and degradation rate constant obtained from plot \(t/q vs. t\) for PS-CSNC with concentration of catalyst, pH and temperature as also illustrated in Figs. 14, 15 and 16.

Table 1 listed the coefficient values of first-order rate constant, when compared with different dosages of composite with RhB, at maximum catalyst concentration (PS-CSNC = 3.0 g L⁻¹) showing higher \(r^2\) value (0.999) than other catalyst concentrations. Similarly, higher absorption rate constant \((r^2 = 0.996)\) is shown in maximum catalyst concentration with MG. Thus, rate constant adsorption is
more linear in maximum catalyst concentration for RhB and MG than other minimum catalyst concentration as illustrated in Fig. 11.

pH-dependent catalytic adsorption of rate constant is illustrated in Fig. 12A, B for RhB and MG, respectively. Figure 13A for RhB and Fig. 13B for MG represented that temperature-dependent rate constant of PS-CSCN. Both Figs. 12 and 13 assisted to know rate constant and capacity of adsorption equilibrium by pseudo-first-order kinetic. These results recommended that adsorption of RhB and MG on PS-CSNC, cannot be described by pseudo-first-order kinetic model due to coefficient \( r^2 \) value is less than 0.999 (Daifullah et al. 2007).

### Table 1 Pseudo-first-order kinetic rate constant adsorption of RhB and MG using PS-CSNC

| Parameter | RhB | | MG | |
|------------|-----|---|---|---|
| Dosages (g/L) | Intercept | Slope | \( q_e \) (mg/g) | \( k_1 \)  (min\(^{-1}\)) | \( r^2 \) | Intercept | Slope | \( q_e \) (mg/g) | \( k_1 \)  (min\(^{-1}\)) | \( r^2 \) |
| 1.0 | 1.21962 | -0.01748 | 3.385901 | -0.00029 | 0.98723 | 1.19036 | -0.01696 | 3.288265 | -0.00028 | 0.97933 |
| 2.0 | 1.3007 | -0.01901 | 3.671866 | -0.00032 | 0.99846 | 1.57637 | -0.02504 | 4.837364 | -0.00042 | 0.95106 |
| 3.0 | 1.32549 | -0.01948 | 3.764029 | -0.00032 | 0.99964 | 1.27495 | -0.01853 | 3.578522 | -0.00031 | 0.99622 |
| pH | 5.8 | 0.96223 | -0.01121 | 2.617527 | -0.00019 | 0.8276 | 0.90545 | -0.01416 | 2.473045 | -0.00024 | 0.93975 |
| 6.2 | 1.21962 | -0.01748 | 3.385901 | -0.00029 | 0.98723 | 1.27351 | -0.02007 | 3.573373 | -0.00033 | 0.93986 |
| 7.8 | 1.10769 | -0.01391 | 3.027357 | -0.00023 | 0.96503 | 1.06426 | -0.01675 | 2.898693 | -0.00028 | 0.95122 |
| Temperature (°C) | 30 | 1.1586 | -0.01634 | 3.18547 | -0.00027 | 0.96865 | 1.14196 | -0.01822 | 3.132903 | -0.0003 | 0.98029 |
| 45 | 1.42887 | -0.01909 | 4.17398 | -0.00032 | 0.95916 | 1.46355 | -0.02116 | 4.321273 | -0.00035 | 0.94616 |
| 60 | 1.41133 | -0.0193 | 4.101407 | -0.00032 | 0.98441 | 1.51194 | -0.02428 | 4.535521 | -0.0004 | 0.98225 |

**Fig. 11** Pseudo-first-order kinetic linearized plot for dosage-dependent PS-CSNC; A and B corresponding for RhB and MG, respectively

**Pseudo second-order kinetics**

The second-order kinetics was also used to represent the kinetics adsorption of RhB and MG by PS-CSNC catalyst. The second-order kinetic rate constant values \( (k_2) \) were presented in Table 2 and Figs. 14, 15 and 16.

The degradation rate constants were increasing with the increasing initial dye concentration in RhB and showed maximum value for MG at 3 g L\(^{-1}\) (Table 2). This is due to the increasing dye concentration increases the adsorption of H\(_2\)O molecules on the surface of the catalyst. This mechanism allows the amount of hydroxyl radicals formed on the surface of the catalyst (Hu et al. 2013; Subramanian...
et al. 2003). Hence, it is clear that the second-order kinetic data clearly indicate the increased reaction rate with increase in concentration of dye with RhB and up to certain limits with MG.

The maximum rate constant value was for RhB with pH 6.2; whereas, increasing pH increased the rate constant value in MG and the maximum value was found with the pH 7.8. On the other hand, the temperature showed maximum rate constant value for the RhB at 60 °C, whereas MG showed maximum rate constant value at 30 °C. Hence, it is clear that the catalyst alone not only influenced the rate of the reaction type of substrate, but also influenced the rate of the reaction. The charge of catalyst and size of the catalyst can be affected by pH and temperature which directly influence the photocatalytic activities of catalyst (Chong et al. 2010). When compared to the first-order reaction, the reaction rate constants were high and fit well in the pseudo second-order reaction and hence the adsorption of RhB and MG follows the pseudo second-order reaction in the present study.

Fig. 12 Pseudo-first-order kinetic linearized plot for solution pH; A and B corresponding for RhB and MG, respectively

Fig. 13 Pseudo-first-order kinetic linearized plot for solution temperature; A and B corresponding for RhB and MG, respectively
Conclusion

Paddy straw-associated chitosan nanocomposite photocatalytic bio-adsorbent was used for removal of rhodamine B and malachite green dyes under UV radiation. UV–visible spectrum absorbance confirmed the formation of composite. Synthesized PS-CS nanocomposites were characterized by FTIR and XRD analysis. Composite morphological patterns, such as average crystalline size (120–220 nm) and structure (spherical shape heterogenous population), were showed by FESEM. High thermal stability and minimal thermal degradative ability composite exhibited high level of porosity and degradation. Optimization of catalyst concentration, reaction solution pH and temperature showed the highest-level activity of PS-CSNC, enhanced in reaction solution because of electrostatic interaction between both RhB and MG dyes. Process of adsorption pseudo-second-order kinetics has shown better $r^2$ (correlation coefficient) value compared to pseudo-first-order kinetics. Finally, the results declared that the non-toxic, eco-friendly and low-cost PS-CSNC catalyst material could be a hopeful material for the adsorption and photocatalytic removal of rhodamine B and malachite green dyes from aqueous solutions.

Table 2 Pseudo second-order kinetic rate constant adsorption of RhB and MG using PS-CSNC

| Parameter       | RhB         | MG          |
|-----------------|-------------|-------------|
|                 | Intercept   | Slope       | $q_e$ (mg/g) | $k_2$ (min$^{-1}$) | $r^2$ | Intercept   | Slope       | $q_e$ (mg/g) | $k_2$ (min$^{-1}$) | $r^2$ |
| Dosages (g/L)   |             |             |             |                 |       |             |             |             |                 |       |
| 1.0             | 2.33333     | 0.09333     | 10.71467    | 0.003733        | 0.92857 | 3.2089      | 0.05706     | 17.52541     | 0.001015        | 0.98786 |
| 2.0             | 1.10118     | 0.07129     | 14.02721    | 0.004615        | 0.97106 | 0.6983      | 0.0557      | 17.95332     | 0.004443        | 0.99709 |
| 3.0             | 0.63919     | 0.0572      | 17.48252    | 0.005119        | 0.98445 | 1.19158     | 0.05434     | 18.40265     | 0.002478        | 0.99071 |
| pH              |             |             |             |                 |       |             |             |             |                 |       |
| 5.8             | 11.57333    | 0.084       | 11.90476    | 0.00061         | 0.77895 | 6.53852     | 0.11394     | 8.776549     | 0.001986        | 0.92446 |
| 6.2             | 2.33333     | 0.09333     | 10.71467    | 0.003733        | 0.92857 | 2.33155     | 0.07559     | 13.22926     | 0.002451        | 0.98522 |
| 7.8             | 4.37333     | 0.09317     | 10.73307    | 0.001985        | 0.91773 | 3.3851      | 0.10243     | 9.762765     | 0.003099        | 0.9679  |
| Temperature (°C)|             |             |             |                 |       |             |             |             |                 |       |
| 30              | 2.33333     | 0.09333     | 10.71467    | 0.003733        | 0.92857 | 1.95476     | 0.08588     | 11.64415     | 0.003773        | 0.98526 |
| 45              | 0.82586     | 0.05466     | 18.29491    | 0.003618        | 0.99199 | 0.82586     | 0.05466     | 18.29491     | 0.003618        | 0.99199 |
| 60              | 0.54152     | 0.04595     | 21.76279    | 0.003899        | 0.9952  | 0.55347     | 0.04416     | 22.64493     | 0.003523        | 0.9977  |

Fig. 14 Pseudo-second-order kinetic linearized plot for dosage-dependent PS-CSNC; A and B corresponding for RhB and MG, respectively.
Fig. 15  Pseudo second-order kinetic linearized plot for solution pH; A and B corresponding for RhB and MG, respectively

Fig. 16  Pseudo second-order kinetic linearized plot for solution temperature; A and B corresponding for RhB and MG, respectively

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Declarations

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