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A low-cost effective media from starch kitchen waste for bacterial cellulose production and its application as simultaneous absorbance for methylene blue dye removal

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Abstract

Daily large quantities of kitchen wastes rich in starchy components are discarded into the environment. The current study documents the use of starchy kitchen wastes (SKW) as a low-cost effective substrate for bacterial cellulose (BC) production and loaded with carbonaceous compounds to remove cationic dyes from wastewater. Box-Behnken design (BBD) was applied to optimize the maximum production of reducing sugars by enzymatic hydrolysis of SKW and the hydrolysate at different concentrations used as precursors for BC biosynthesis using *Komagataeibacter hansenii* AS.5. SEM, FT-IR, and mechanical properties were performed for BC membranes characterization. The synthesized BC membranes were loaded with either charcoal or graphite and evaluated for removal of methylene blue (MB) as a representative cationic dye. The BBD showed that 148.0 g/l of reducing sugars (54.8% saccharification) can be produced using 24.3% of SKW (84.3 g/l/ dry) treated with 2-ml amylase (313 U/ml) at 50 °C for 5 h and 40 min. The optimum concentrations of SKW hydrolysate produced 2.11 g/l of BC membranes (0.3 g/l/day). The original and loaded membranes showed absorption capacities of 53, 98.7, and 100% removal of MB using BC, graphite-BC, and charcoal-BC membranes, respectively. Therefore, SKW can be used as alternative medium for BC production and applied successfully for dye removal.

Keywords Starch kitchen wastes · Box-Behnken design · Bacterial cellulose · Methylene blue dye removal

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1 Introduction

Cellulose, the most abundant polymer on earth, can be produced from many sources including plants and micro-organisms. Among microbial celluloses, BC is effective in terms of purity (free from lignin and hemicellulose) and homogeneity [1, 2]. An extracellular product with the same form of cellulose in nature and widely produced by acetic acid bacteria such as *Komagataeibacter hansenii* and *Komagataeibacter xylinus* [3], BC has unique structural advantages over PC (plant cellulose), such as high degree of polymerization, crystallinity and surface area, and nano-fibrillary network [4]. A major drawback in the widespread commercial application of BC is its relatively expensive production process. This could be attributed to the low productivity of known applied strains and the costly cultivation media requirements, (approximately 30% of the total BC production cost) [5], Therefore, medium cost reduction represents a key step towards extensive application of BC. To achieve this, a wide range of biomass wastes was surveyed including tobacco waste

extract [6], orange peel [7], citrus juices [8], beverage industrial waste [9], cotton-based waste textiles [10], and black tea [11]. Food wastes, originating from processing industries, restaurants, canteens, households, and markets, are important sources of biomass wastes. According to the Food and Agriculture Organization of the United Nations (FAO), 1.3 billion tons of food wastes are produced, which is around one-third of food produced for human consumption on a global scale [12]. In many countries, food waste is usually incinerated, composted or directly land filled with other wastes after simple recycling [13]. With growing environmental awareness, several studies have reported the application of food kitchen wastes towards production of bioelectricity [14], biogas [15, 16], volatile fatty acids and H_2 [17], enzymes [18], biofuels [19, 20], and pullulan [21]. Starch kitchen wastes (SKW) could be a potential candidate for BC production upon enzymatic hydrolysis. Amylase enzyme converts SKW to free glucose and reducing sugars which in turn can be fermented into BC. Optimization of the SKW hydrolysis process is crucial and directly affects the final product yield. Statistical designing of this optimization process reduces both time and effort compared to one variable at time method (OVAT) [22]. In addition, all studied variables are optimized at the same time where interaction between such variables could also be evaluated [23, 24]. Among statistical designs, BBD represents an efficient tool with a small manageable set of data and requirements for process parameter optimization of kitchen waste [25]. Based on the unique physical and chemical properties, BC is used in wound healing [26], oil separation [27], skin and bone tissue engineering [28], bioelectrode [29], cosmetics [30], heavy metal removal [31, 32], and dye removal [33]. MB, commonly used for dye removal in several works, and removal of MB itself from textile, dyeing, printing, and coating industries, has become one of the major environmental issues [34]. Due to the low efficiency of BC for dye removal, many attempts were dedicated to manipulating its structure including BC/polydopamine/TiO_2 [35], BC/attapulgite magnetic structure [32], and BC/polydopamine/reduced graphene oxide [33]. Charcoal (Ch) and graphite (Gr) are worldwide naturally occurring biomaterials characterized by their high porosity and efficient absorption for a long time. Ch has been used for water treatment, harmful gas elimination, antibacterial efficacy, blood purification, absorption, and electromagnetic shielding [36, 37]. Furthermore, Gr is a naturally occurring mineral, used for battery application, graphene production, and removal of dyes [38, 39]. In the present investigation, the SKW matrix from different sources was collected and submitted to BBD optimization in order to maximize the production of reducing sugars. The final optimized hydrolysate with high reducing sugars

at different concentrations was used as a carbon source for the production of BC. The obtained BC membranes were characterized by scanning electron microscope (SEM), Fourier-transform infrared spectroscopy (FT-IR), and other mechanical features. Finally, the BC gel-like matrix was applied for the removal of MB compared to Ch and Gr-loaded BC matrices.

2 Materials and methods

2.1 Collection and analysis of SKW

SKW is a mixture of house kitchen wastes including bread, rice, pasta, and noodles. The collected wet wastes were milled through a laboratory blender to obtain a well-homogenized mash with small particle size. The prepared mash was packaged and stored at 4 °C till use, to prevent acidification during the storage time. The SKW was analyzed for moisture, solid, and reducing sugar contents.

2.2 Amylase source and activity determination

Amylase used throughout this work was produced by the locally isolated *Bacillus methylotrophicus* SCJ4 [40]. The enzyme was prepared in a stock solution (0.1 mg/ml), where its units (313 U/ml) were assayed according to the iodine colorimetric method [41] with slight modification. The dried form of the enzyme was prepared through freeze drying using the culture supernatant as source of the enzyme.

2.3 Statistical optimization for enzymatic hydrolysis of SKW

For SKW hydrolysis optimization, BBD was applied using three variables, namely SKW concentration, enzyme concentration, and hydrolysis time. As indicated in Table 1, each variable was studied at three different levels coded − 1, 0, and 1 representing the low, middle, and high values of each variable. The BBD matrix implies 12 different combinations (trials) and 3 central trials to detect handling errors (Table 1), and each row and column represent an experiment and a variable used, respectively. All trials were incubated at 50 °C, pH 6, according to the optimum conditions for enzyme activity [42]. The amount of released reducing sugars was measured as the response (Y) and culture conditions were taken as independent variables (X). Each trial was conducted in triplicate and mean values were represented as responses. The experimental results were fitted by regression to a predictive quadratic polynomial model that

gave reducing sugars production (Y) as a function of culture conditions (X):

$$Y = B0 + B1X1 + B2X2 + B3X3 + B12X1X2 + B13X1X3$$
$$+ B23X2X3 + B11(X1)2 + B22(X2)2 + B33(X3)2 \quad (1)$$

where Y is the expected response; B_0 is the model constant; X_1, X_2, and, X_3 are the variables under study; B_1, B_2, and B_3 are the linear coefficients; B_{12}, B_{13}, and B_{23} are the cross-interacted coefficient; and B_{11}, B_{22}, and B_{33} are the quadratic coefficients. The results of BBD were represented in the form of surface plots, where each plot was developed by two independent variables on X and Y axes with the dependent variable (response) on the Z axis, while the third variable was set at its middle value.

2.4 Assay of SKW hydrolysate

SKW hydrolysis progress was evaluated by measuring the reducing sugars released during the course of hydrolysis. The released reducing sugars were determined with the dinitrosalicylic acid (DNS) method [43], wherein brief, 1 ml of sample was mixed with 1.5 ml of freshly prepared DNS solution and boiled for 10 min. The resulting color was measured at 575 nm with a standard curve of glucose. Percentage of saccharification was calculated according to [44] as = amount of reducing sugar $\times 0.9 \times 100$/initial

amount of SKW, where the 0.9 is the molar mass conversion of starch to sugars.

2.5 Model accuracy and verification

The accuracy of BBD results was verified by comparing the expected yield at model level with the real measured yield, upon applying the model conditions in the laboratory.

2.6 Data analysis and 3D graphs representation

Regression analysis and analysis of variance (*ANOVA* test) were conducted through the statistical analyses on Microsoft Excel 2010, where the 3D graphs representing the effect of each two factors on waste hydrolysis were created through the STATISTICA 7 program.

2.7 Microorganism and preinoculum

In the present investigation, *Komagataeibacter hansenii* AS.5 (*K. hansenii* AS.5) was used as the BC-producing strain, as described in an earlier study [45]. Preinoculum was prepared by using Hestrin and Schramm (HS) medium, with the following composition (g/l): 20 D-glucose, 5 peptone, 5 yeast extract, 2.7 Na_2HPO_4, and 1.15 citric acid [46]. The medium was inoculated with a single colony of *K. hansenii* AS.5 and incubated at 30 °C, 200 rpm for 48 h, to activate

Table 1 BBD matrix representing the coded and real values of three independent variables with measured and predicted reducing sugars as response

Trial	Variables*			Response	
	X_1 SKW conc	X_2 Enzyme volume	X_3 Hydrolysis time	Estimated reducing sugar (g/l)	Predicted reducing sugar (g/l)
1	0(20)	0(1)	0(4)	114 ± 5.2	118.5 ± 5.3
2	0(20)	− 1(0.5)	− 1(2)	34.38 ± 2.7	32.44 ± 2.5
3	− 1(10)	0(1)	1(8)	69.09 ± 3.5	63.01 ± 3.1
4	− 1(10)	1(2)	0(4)	84.90 ± 4.9	89.05 ± 5
5	0(20)	1(2)	1(8)	135.1 ± 6.2	137.04 ± 6.4
6	1(30)	0(1)	− 1(2)	53.80 ± 3.1	59.88 ± 3.2
7	1(30)	1(2)	0(4)	135.03 ± 6.1	135.8 ± 6.2
8	− 1(10)	− 1(0.5)	0(4)	35.10 ± 2.5	34.34 ± 2.4
9	0(20)	1(2)	− 1(2)	121.1 ± 5.9	114.2 ± 5.5
10	0(20)	0(1)	0(4)	108.7 ± 5.1	118.5 ± 5
11	− 1(10)	0(1)	− 1(2)	48.35 ± 2.9	51.05 ± 3.1
12	0(20)	− 1(0.5)	1(8)	59.05 ± 3.3	65.89 ± 3.8
13	1(30)	− 1(0.5)	0(4)	41.66 ± 2.7	37.52 ± 2.5
14	1(30)	0(1)	1(8)	106.8 ± 5	104.1 ± 5.8
15	0(20)	0(1)	0(4)	132.7 ± 6	118.5 ± 8

Variables*: X_1–X_3 (selected variable to optimize their levels); the coded low, middle, and high levels are presented as − 1, 0, and 1, respectively. Variable levels are presented between brackets expressed as % for X_1; ml for X_2; and h for X_3

the producer strain. The bacterial strain was maintained on HS agar slants at 4 °C for further use.

2.8 Production of BC using SKW hydrolysate

The enzymatic hydrolysis condition of SKW with the highest reducing sugar yield was used for BC production. Under optimum conditions for SKW enzymatic hydrolysis, the hydrolysate was centrifuged at 5000× for 15 min to remove sediments and the supernatant was collected. Glucose-free HS media were used as a basal medium for BC production from SKW. As an alternative carbon source, the glucose-free HS medium was supplemented with 10% SKW hydrolysate supernatant. Medium pH was adjusted to 6.0, inoculated with 10% (v/v) of *K. hansenii* AS.5 preinoculum, and incubated for 7 days at 30 °C in static conditions. By the end of the incubation time, the BC pellicle (gel) at the air–liquid interface was observed. The flasks containing media were sterilized by autoclave at 121 °C for 15 min.

2.9 Effect of SKW hydrolysate concentrations on BC production

Different concentrations of SKW hydrolysate (20, 40, 60, 80, and 100%) were added to glucose-free HS media as carbon sources. Erlenmeyer flasks (250 ml) were inoculated with 10% (v/v) preinoculum and cultivated statically at 30 °C for 7 days. Each culture was performed in triplicate and the mean values for BC are given as g/l.

2.10 BC purification and quantification

By the end of the fermentation course, the observed BC at the air–liquid interface was collected, transferred to another container, and purified in several steps. First, BC was washed three times with distilled water (DW) to remove the remaining components of the culture medium. Subsequently, the BC was treated 2–3 times with 0.5% NaOH solution at 90 °C for 30 min to remove the remaining bacteria and other impurities adsorbed on the BC, and then washed 3–4 times with DW until a neutral pH bright-white BC was obtained. Finally, the purified BC was dried at 70 °C overnight to get a constant weight. BC production has been reported as gram dry weight of cellulose per liter of the medium (g/l) [47].

2.11 Characterization of BC membrane

SEM (JEOL JSM 6360 LA, Japan) was used to investigate the surface structure of the BC samples. The SEM experiment was performed at a magnification of 5000–15,000×.

The functional groups and chemical bonds of BC samples were performed using FT-IR (JASCO FTIR-4100 E, Japan), operated in absorption mode with the wavenumber range from 4000 to 500 cm^{-1}, using a resolution of 1 cm^{-1}. A universal tensile testing machine (Universal Testing Machine, model: AG-I/50 N-10 KN, Japan) was used to determine the mechanical properties of dried BC membranes. The BC membrane was cut into rectangular strips (10 cm long and 1 cm wide) for measurement, with a gauge length of 20 mm. BC membrane thickness was determined by an electronic digital micrometer before the examination. Each test was performed using 3 specimens and the average of the results was recorded.

2.12 Preparation of BC gels loaded with Ch or Gr

BC gels were obtained from *K. hansenii* AS.5 using SKW hydrolysate as a carbon source. BC gel (~ 4 g wet, 5×5 cm diameter, and 0.5 cm thickness) was loaded by Ch and Gr through the swelling diffusion method [48]. About 0.5 g of Ch or Gr was stirred separately in 100-ml DW for 1 h, and homogenized through ultrasonication (750 wt., 20 KH, pulse 45, Amp 1) for 15 min to obtain a homogenous dispersion. Once a homogenous aqueous dispersion of Ch or Gr was obtained, the original BC gels were immersed in an Erlenmeyer flask containing Ch or Gr dispersion for 24 h, 200 rpm at room temperature, to ensure complete absorption of BC gels with Ch or Gr. In post-incubation, the BC gels were removed and then immersed briefly in DW for 30 s and wiped lightly once with tissue to remove non-absorbed Ch or Gr from BC gel surface. Water-soaked BC gels were taken as control. Finally, the original BC and BC-loaded gels were placed in a Petri dish and kept at 4 °C in the refrigerator until use [48].

2.13 Application of BC-Ch- and BC-Gr-amended gels for MB dye removal

The three prepared gels (original BC, BC-Ch, and BC-Gr) were tested individually for their ability to remove 10 mg/l of MB using a pressure-free system. The system consists of two chambers connected by a medium junction that contains one of the tested membranes (Fig. 1). Fifty milliliters of 10 mg/l dye solution was added to the upper chamber and allowed to pass through the membrane to the lower chamber. Optical densities (OD) of the upper and lower liquids were measured at 655 nm, and the percentage of dye removal by each membrane was calculated. The reusability of BC-Ch- and BC-Gr-amended gels was also evaluated to determine the full capacity and durability of the two membranes in successive dye removal processes. For this, the used gels in the previous dye removal cycle were washed in 50-ml PBS
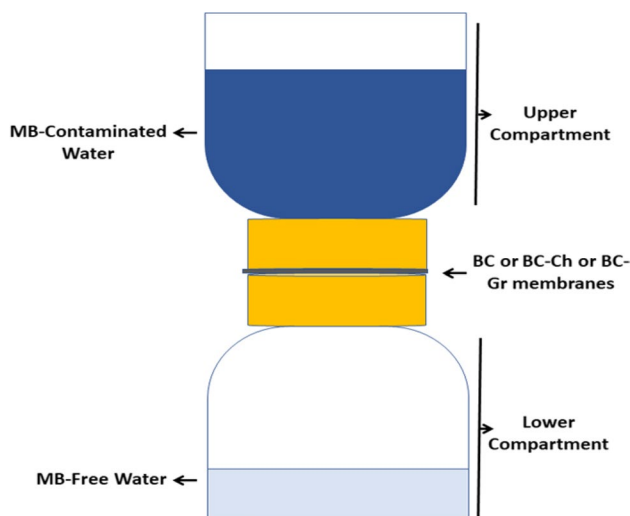
Fig. 1 Design of the system used for the treatment of MB-contaminated wastewater using BC gels

buffer (pH 7.0) and the released dye was determined at different time intervals at 655 nm.

2.13.1 Dye retention capacity

To evaluate the dye retention capacity for original BC, BC-Ch, and BC-Gr gels, the three gels used in the previous dye removal experiment were washed in 50-ml PBS buffer (pH 7.0) for 2 h and the released dye was quantitated over different time intervals at 655 nm compared to 10 mg/l MB dye solution as control (100%).

3 Results and discussion

3.1 BBD for enzymatic hydrolysis of SKW

The high starch content in the kitchen wastes qualified them for several biotechnological applications [49]. In this study, the used SKW was a mixture of collected house kitchen wastes with estimated moisture to solid contents of 65.3% and 34.7%, respectively, with an initial reducing sugar concentration of 25.8 g/l. The SKW was hydrolyzed by *Bacillus methylotrophicus* SCJ4 amylase, where the released fermentable sugars were used as the main substrate for BC production. During enzymatic hydrolysis of the SKW, BBD was applied to evaluate the optimum conditions for maximum production of the reducing sugars. BBD was also widely applied for various optimization processes including enzyme production [50], waste degradation process [51, 52], and waste composting [25]. In the present investigation, three variables were evaluated for their effects on SKW hydrolysis at three levels. These variables include SKW concentration, enzyme volume, and hydrolysis time. The matrix design was

built from 12 different trial combinations, with three central trials, to evade the handling errors. The data (Table 1) indicated variation in the amount of reduced sugars resulting in different trials ranging from 34.38 to 135.1 g/l with 15.5 and 60.8% scarification, respectively. The variation thus had a direct impact of the optimization process on the hydrolysis results. The significance F value represents the statistical significance of the used variables on the experiment's response (reducing sugars), where values below 0.05 indicate significance while that below 0.01 indicates high significance [53]. In this experiment, the value of significance F (0.001958) indicated a highly significant effect of the applied variables upon the estimated response. BBD depends upon a second-order polynomial model in predicting the optimum experimental conditions for the desired response (maximum reducing sugars). From the regression analysis, the applied polynomial model (Eq. 1) can be written as:

$$
\begin{aligned}
Y_{(reducing\ sugar)} = {} & 118.5 + 12.5X_1 + 38.2X_2 + 14.1X_3 \\
& + 10.89X_1X_2 + 8.07X_1X_3 - 2.67X_2X_3 \\
& - 31.1(X_1)^2 - 13.2(X_2)^2 - 17.87(X_3)^2
\end{aligned} \tag{2}
$$

where X_1, X_2, and X_3 are variables under study.

The regression analysis results (R square = 0.97) showed high accuracy of the applied model, where the closer the R value to 1 indicates a strong correlation between measured and predicted values at the model level [22]. The result of correction factor adjusted R square (0.926) also revealed a strong correlation between estimated and predicted response values, where only less than 7.4% of the response could not be fitted within the applied model. The ANOVA summary of the BBD model represents the degrees of freedom (*df*), sum-of-squares (*SS*), mean square (MS), F-ratio (*F*), and significance F values as presented in Table 2. The interaction consequences of the independent variables on the measured response were visualized with surface plots (Fig. 2). The plot indicated that waste concentration implied positive interaction with enzyme volume and hydrolysis time in contrary to enzyme concentration that interacted negatively with hydrolysis time. The *Solver* function of MS Excel was applied to determine the exact parameter values at the used model. The results implied that using SKW concentration of 24.3 g/100 ml (84.3 g/l/dry) with enzyme volume of 2 ml (0.1 mg/ml), for 5 h and 40 min

Table 2 ANOVA for BBD analysis to maximize reducing sugars from SKW

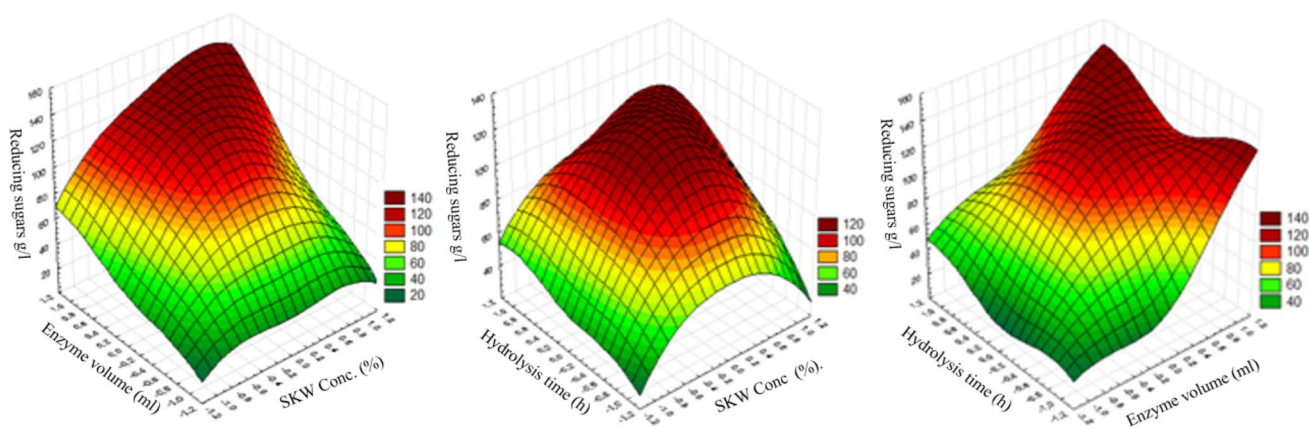|	df	SS	MS	F	Significance F
Regression	9	20,107.20	2234.13	20.56	0.001958
Residual	5	543.37	108.67		
Total	14	20,650.58			

Fig. 2 3D surface plots representing the possible interactions of each of the two separate variables and their impacts on the reducing sugars production

hydrolysis time, resulted in maximum production of simple sugars of 151 g/l (56% saccharification).

3.1.1 Model verification

To evaluate the applicability and accuracy of the quadratic polynomial model, a verification experiment was conducted under the predicted optimal conditions as following: SKW concentration of 24.3 g/100 ml, with enzyme volume of 2 ml for hydrolysis time of 5 h and 40 min at 50 °C and 200 rpm. The estimated reducing sugar was 148.0 g/l (54.8% saccharification), while the expected was 151 g/l (56% saccharification). Thus, a high degree of accuracy (98.0%) is evidence of model validation. Kim and his colleagues applied response surface methodology (RSM) based on the central composite design for optimization of food waste hydrolysis to obtain the predicted and estimated reducing sugar concentrations as 117.0 and 120.1 g/l, respectively [54]. Another study reported maximum reducing sugar as 100 g/l upon enzymatic hydrolysis for food wastes using a cellulolytic enzyme [55]. The hydrolysis time of 5 h and 40 min in the current study is in agreement with Bhatt and his colleagues who reported an optimum time of 6 h for kitchen wastes hydrolysis by amylase [18].

3.2 Evaluation of the effect of SKW hydrolysate upon BC production

BC production cost is one of the main obstacles towards its wide commercial applications. Most of this high cost could be attributed to carbon sources and hence, the reduction in this cost is widely desirable [49]. Using SKW as an alternative source for BC production represents a dual solution for both environmental and economical point of view. Initially, 10% of optimized hydrolysate was supplemented to glucose-free HS media to achieve 0.32 g/l BC after 7 days. In order to

investigate the effect of SKW hydrolysate concentration on BC production, SKW hydrolysate was added exogenously to the glucose-free HS media at varying concentrations from 20 to 100% and the BC productions were compared after 7 days of static cultivation. The results revealed that the SKW hydrolysate had a positive effect on BC production at 20, 40, 60, 80, and 100% concentrations to produce 0.82, 1.26, 1.48, 1.64, and 2.11 of BC g/l respectively. This suggests that the gradual increase in BC production was proportional to the increase in the concentration of the SKW hydrolysate supplemented to the medium with a maximum production yield at 100%. BC production at 100% (2.11 g/l) (0.3 g/l/day) represented a 2.5-fold increase in the BC production compared to that at 20% SKW concentration (0.82 g/l) (0.1 g/l/day). These results are consistent with other studies that reported maximum BC production between 2.0 and 2.25 g/l using SKW hydrolysate by *Acetobacter xylinum* [56, 57]. The addition of different substrate concentrations to production media may enhance the production process as reported by [6], in which the maximum BC production was 2.2-fold the initial production. The yield of BC from food wastes differs based on factors such as pretreatment of food waste [58], type of food waste applied [59], and production conditions [60]. Collectively, the results indicated that extra concentration of SKW may be a useful strategy to improve BC production as a culture medium.

3.3 Purification and characterization of BC membrane

The raw BC membranes collected from the liquid–air interface of the production media were purified and cleaned from remaining medium components and bacterial debris with 0.5% Sodium hydroxide (Fig. 3). The impact of different concentrations of SKW hydrolysate (20–100%) on the properties and characteristics of the purified BC membranes was investigated with different analytical methods, including surface

structure by SEM and functional and chemical ponds with FT-IR spectroscopy along with their mechanical properties.

3.3.1 SEM analysis

The SEM results clearly indicated that the purified BC membranes obtained from different concentrations of SKW hydrolysate are porous and homogeneous 3-dimensional network-like structures of randomly arranged cellulosic fibers (Fig. 3). The scanning results of all membranes (20–100%) also indicated nearly similar morphological structures with no significant difference. These results are concurrent with data from other studies that reported the BC production with nearly identical membrane morphological structure, using different media and/or varied substrate concentrations [60, 61].

3.3.2 FT-IR spectroscopy

FT-IR spectroscopy was used to determine the chemical structure of the BC membranes including the functional groups and molecular bonding properties using the bond vibration frequencies [62]. The FT-IR spectra of all BC membranes exhibited several clear absorption peaks from approximately 3336.2 to 869.7 cm^{-1} with all typical bands of the BC in the five samples as shown in Fig. 4. From the data obtained (Table 3), we can conclude that all the BC samples share no obvious difference in FTIR spectra, indicating that the obtained BC membranes in all concentrations of SKW hydrolysate have the same chemical composition with no influence on their functional groups. The similarity in the

FT-IR for BC membrane from SKW in the current study and other studies [61, 63] indicated that the SKW is a good candidate for BC production and implied no contamination or by-products in the formed membranes. Moreover, FT-IR data analysis indicated the presence of crystalline regions and confirming the purity of all produced BC membranes [64]. Our results are similar to those in earlier studies, where the similarity in FT-IR results of produced BC membranes at different concentrations of the same media as well as at different media compositions had been reported [6, 65–67].

3.3.3 Mechanical properties

Different mechanical properties of the produced BC membranes from SKW hydrolysate at 20, 40, 60, 80, and 100% concentrations were performed to include: maximum load (0.49, 0.98, 1.80, 2.35, and 2.53 N, respectively), tensile strength (74.3 78.1, 93.3, 129.6, and 162.5 MPa, respectively), elongation at peak (0.48, 0.51, 0.55, 0.84, and 1.25%, respectively), Young's modulus (74.3, 80.4, 131, 195, and 216 MPa, respectively), and thickness (0.10, 0.15, 0.22, 0.27, and 0.31 mm, respectively) of the dried BC membranes. The results indicated that an increase in the membrane thickness was proportional to the increase in SKW hydrolysate concentrations, which is in agreement with other reports [68]. This increase directly improved the mechanical properties with maximum values at 100% concentration (0.31 mm). Several studies reported the optimization of various parameters that enhance the mechanical properties of produced BC
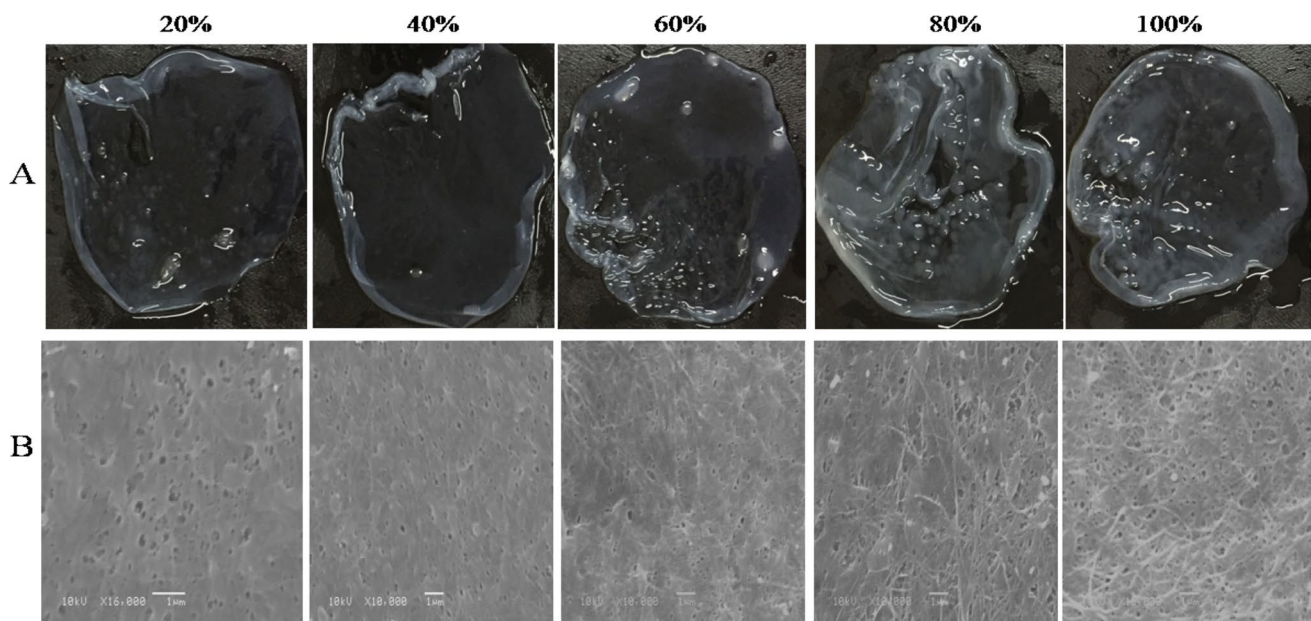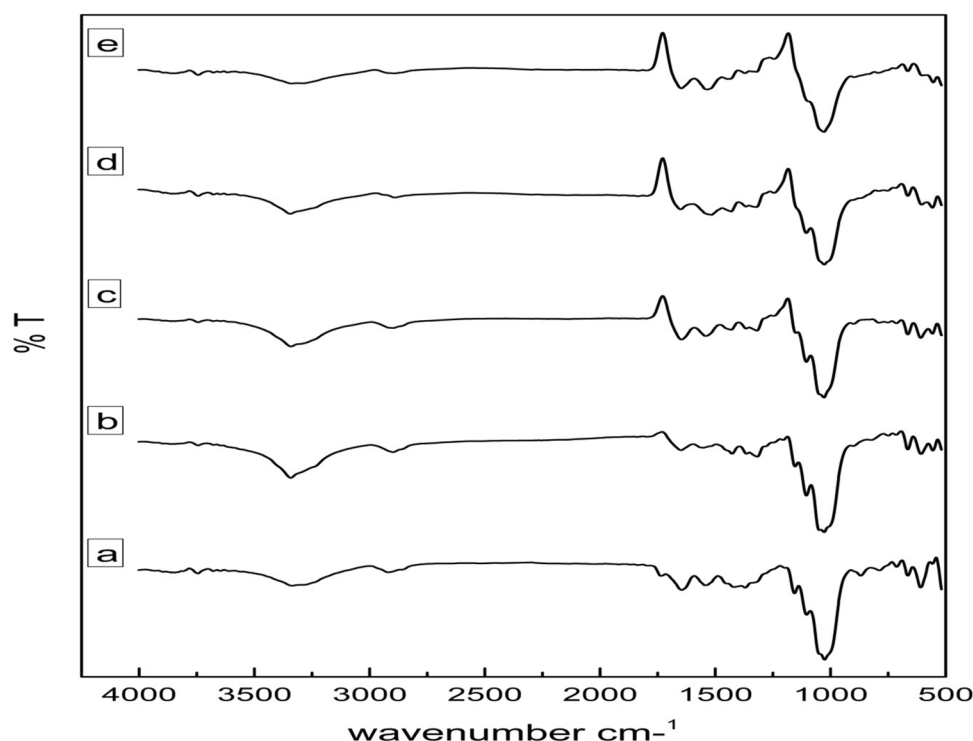

Fig. 3 Images of BC membranes produced at different concentrations of SKW hydrolysates. **A** Photograph for eye visualized purified BC membranes, and **B** SEM micrograph for BC membranes

Fig. 4 FT-IR of BC membranes from SKW hydrolysates at 20, 40, 60, 80, and 100% (a, b, c, d, and e, respectively)

membranes including production time [67], sodium fluoride concentrations [66], static conditions [75], and BC-

Due to the environmental aspects and high cost of the most reported dye removal techniques [77], an ecofriendly eco-

Table 3 Functional and chemical bonds of BC membranes from different SKW hydrolysate concentrations

Assignment	Band position at cm^{-1}					Data are in agreement with
	20%	40%	60%	80%	100%	
OH	3336.2	3342	3342	3343.9	3338.1	[68]
CH$_2$	2919.7	2898.4	2900.2	2890.2	2894.6	[69]
COOH	1644.9	1648.8	1644.9	1648.8	1646.9	[70]
Asymmetric C-H	1419.3	1427	1430.9	1430.9	1446.3	[71]
Symmetric C-H	1369.2	1356.3	1365.3	1367.2	1371.1	[71]
C–O–C	1157	1155.1	1151.2	1157.4	1159.2	[72]
C–OH	1103	1105	1105	1105	1108	[73]
C–C-OH	1025.9	1027.8	1027.8	1027.8	1029.8	[74]
C-H	869.7	900.5	898.6	892.6	896.7	[6]

producing strain [63].

3.4 Removal of MB dye by the prepared gels

The increased application of synthetic dyes in several industries implies a huge amount of the discharged dyes in wastewater samples, representing a direct environmental and human health threat [76]. MB, a cationic dye, is widely applied in many industries and has been reported to be a good model for dye removal in multiple studies [77, 78].

nomic method for the removal of dyes from wastewater is highly demanded. BC gels obtained from 100% SKW hydrolysate loaded with Ch (BC-Ch) or Gr (BC-Gr) (Fig. 5) were tested for their ability to remove MB dye (10 mg/l) compared with plain BC gel. As shown in Fig. 6, the three gels revealed a varied capacity to remove the MB dye. However, this dye removal capacity was dramatically reduced with repeated application. The plain BC gel was able to absorb 53% of the dye in the first application round. The dye removal capacity dropped to 36.3 and 30.3% in the second and third cycles,
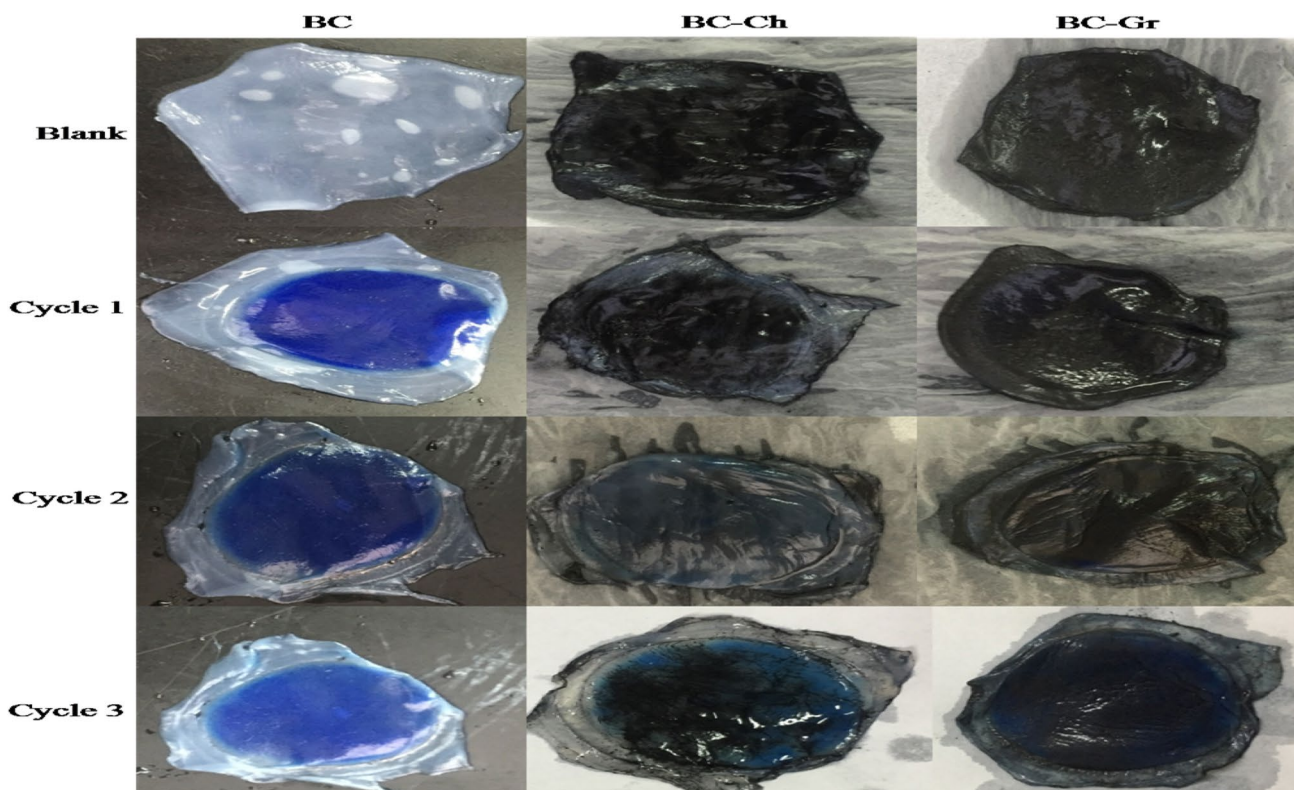
Fig. 5 The prepared three samples of BC, BC-Ch, and BC-Gr gels and their reusability for three successive cycles of MB removal

respectively. In this study, BC-Ch-amended gel revealed the maximum dye removal activity (100%) at its first application round (Fig. 7), with 96 and 94% dye removal capacity after the second and third cycles of application, respectively.
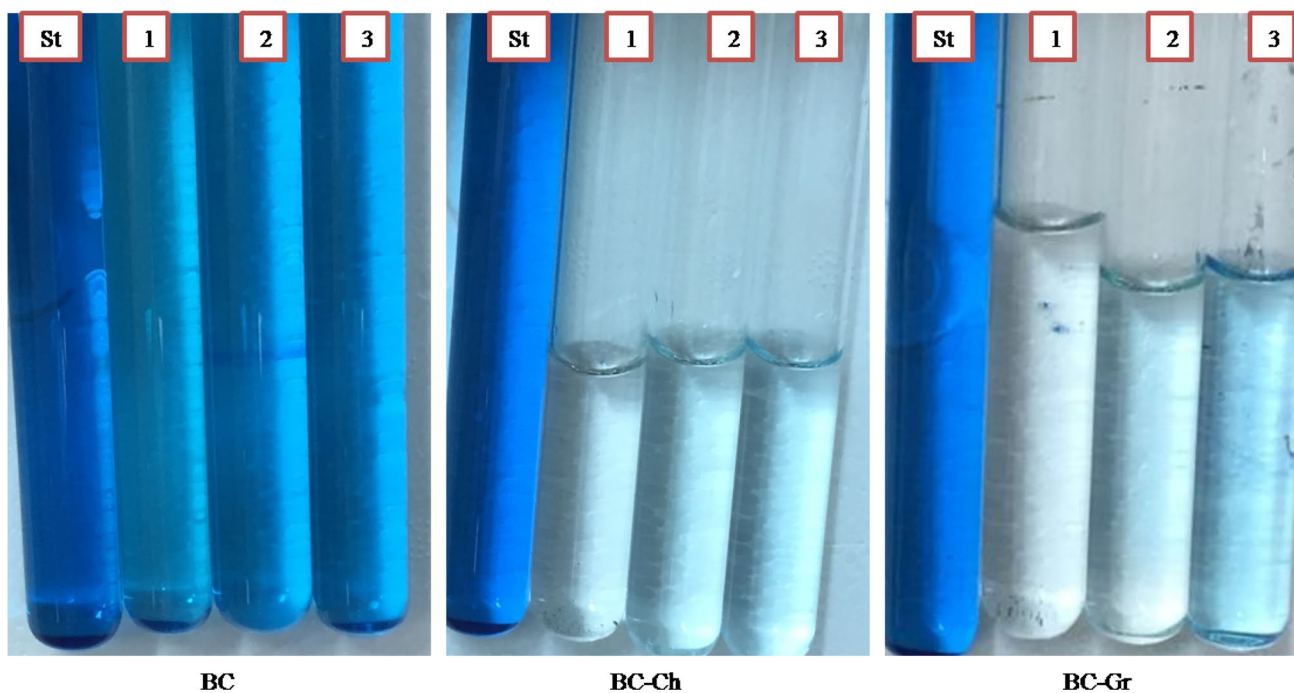

Fig. 6 Absorbance of MB removal using BC, BC-Ch, and BC-Gr gels after cycles 1, 2, and 3 compared with standard solution of the untreated MB dye (St)
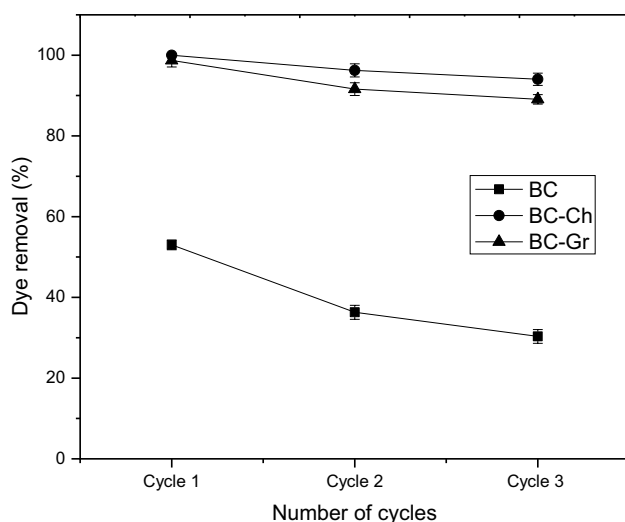
Fig. 7 Percentages of MB dye removal using BC, BC-Cr, and BC-Gr gels during three consecutive cycles

Carbon-based materials such as activated carbon have been previously used for the efficient removal of dyes from aqueous solutions at moderate temperature and neutral pH [79]. Some researchers preferred use of activated charcoal loaded by titanate nanotubes for the removal of different dyes such as MB, cationic rhodamine B, and anionic methyl orange [80]. Other materials such as clays [81, 82], zeolites [83], and siliceous materials [84] have been also been used for the adsorption of dyes from solutions. On the other hand, the BC-Gr-amended gel reported a moderate remediation ability of 98.7% dye removal in the first round, with 91.6 and 89% of the dye removal capacity after three successive cycles of remediation, respectively. The results are supported by other studies that reported the application of graphene in dye and

heavy metal remediation [39, 85]. Our results indicated that BC has the potential capacity for trapping MB dye (53%), and this bio-sorption capacity could be improved through manipulation of the plain BC with Ch (100% dye trapping) or Gr (98.7%). The reusability results also indicated the superiority of Ch-amended BC gel with only 6% loss in dye bio-sorption capacity, compared to BC-Gr and BC that lost about 11% and 69.6% of their dye bio-sorption activity after the third round of reusing. The mean flow rate of the dye-containing water through the three gels was 1.19, 0.49, and 0.63 ml/min for BC, BC-Ch, and BC-Gr gels, respectively.

Statistical analysis of the obtained results was performed using SPSS (version 23) program. As shown in Table 4 part A, the mean difference among all cycles of the membranes BC, BC-Ch, and BC-Gr was greater than 0.05, indicating insignificance. On the other hand, as shown in Table 4 part B, the mean differences among the membranes BC and BC-Ch, BC and BC-Gr, BC-Ch and BC, and BC-Gr and BC-Ch were all ≤ 0.05 indicating significance.

3.4.1 Dye retention capacity

Compared to plain BC membranes, the ability of the prepared gels (BC-Ch, and BC-Gr) to retain the trapped dye was evaluated. For this, the used gels in the previous dye removal cycle were washed with 50-ml PBS buffer (pH 7.0) for 2 h, and the released dye % was determined at different time intervals at 655 nm. As shown in Table 5, the three gels exhibited a distinct affinity for the trapped dye. The maximum released dye was 45.7% from plain BC after 1 h. BC-Ch gel had the highest affinity for trapping dye where the maximum released dye was 4.5% after 1 h upon three cycles of dye removal. However, BC-Gr gel showed a maximum release of 11.7% MB dye after

Table 4 Statistical analysis of the results obtained after using BC, BC-Cr, and BC-Gr gels for the removal of MB for three successive cycles

(I)x	(J)x	Mean difference (I–J)	Std. Error	Sig	95% confidence interval	
					Lower bound	Upper bound
A						
LSD						
Cycle 1	Cycle 2	9.2666	26.154	0.735	− 54.730	73.263
	Cycle 3	12.800	26.154	0.642	− 51.197	76.797
Cycle 2	Cycle 1	− 9.2666	26.154	0.735	− 73.263	54.730
	Cycle 3	3.5333	26.154	0.897	− 60.463	67.530
Cycle 3	Cycle 1	− 12.800	26.154	0.642	− 76.797	51.197
	Cycle 2	− 3.5333	26.154	0.897	− 67.530	60.463
B						
LSD						
BC	BC-Ch	− 56.8000	6.1986	0.000	− 71.967	− 41.632
	BC-Gr	− 53.2333	6.1986	0.000	− 68.401	− 38.066
BC-Ch	BC	56.8000	6.1986	0.000	41.632	71.967
	BC-Gr	3.5666	6.1986	0.586	− 11.601	18.734
BC-Gr	BC	53.2333	6.1986	0.000	38.065	68.401
	BC-Ch	− 3.5666	6.1986	0.586	− 18.734	11.601

Table 5 Percentages of the dye released from BC, BC-Ch, and BC-Gr gels after each dye removal cycle

Time	Original BC			BC-Ch			BC-Gr		
	Cycle 1	Cycle 2	Cycle 3	Cycle 1	Cycle 2	Cycle 3	Cycle 1	Cycle 2	Cycle 3
10 min	2.3 ± 0.5	6 ± 0.9	24 ± 2.5	0.07 ± 0.01	2.7 ± 0.5	3.8 ± 0.6	0.4 ± 0.06	1.2 ± 0.02	8.4 ± 1.2
30 min	2.8 ± 0.7	7.5 ± 1	35 ± 3.2	0.07 ± 0.01	3.1 ± 0.8	4.2 ± 0.9	0.7 ± 0.08	2.7 ± 0.08	9.8 ± 1.5
1 h	3.7 ± 0.7	9.4 ± 1.1	45.2 ± 4.1	0.2 ± 0.05	3.2 ± 0.7	4.5 ± 0.1	0.9 ± 0.1	3.8 ± 0.8	10.8 ± 1.7
2 h	4.8 ± 0.8	15.8 ± 1.7	45.7 ± 4.3	2 ± 0.05	3.4 ± 0.8	4.5 ± 0.1	1 ± 0.1	4.5 ± 0.9	11.7 ± 1.9

2 h for cycle 3. The limited dye released amounts from BC-Ch- and BC-Gr-amended gels, compared to plain BC gel, over time indicated a strong adsorption status between the dye and Ch- and Gr-amended membranes, and reflecting the effect of Ch in enhancing the BC activity towards synthetic dye removal. This limited absorption of cationic dyes to BC could be attributed to the weak anion nature of most natural cellulosic fibers [86], and hence modification of its abundant (OH) surface groups enhanced their dye removal activity [87].

4 Conclusion

The enzymatic hydrolysis optimization of SKW resulted in 148.0 g/l of reducing sugars (54.8% saccharification) through applying BBD optimum conditions as following 243 g/l of wet SKW (84.3 g/l/dry) treated by 2 ml amylase (313 U/ml) at 50 °C for 5.66 h. The SKW hydrolysate has been used as a raw material for BC production after the optimization of its enzymatic hydrolysis using BBD. The maximum BC weight of 2.11 g/l (0.3 g/l/day) was obtained after the microbial fermentation of 100% of the optimum SKW hydrolysate. Both SEM and FTIR analyses showed the formation of a porous and homogeneous 3-dimensional network of the cellulose fibers and the representative functional groups, respectively. The mechanical properties of BC membranes were also affected by the concentration of SKW hydrolysate. Loading the biosynthesized BC membrane with graphite or charcoal showed maximum percentages of MB removal of 98.7 and 100%, respectively. Our study thus suggests that charcoal-loaded BC originating from starchy kitchen waste can be effectively used for the treatment of dye-contaminated wastewater.

Author contribution Ahmed K. Saleh: methodology, investigation, formal analysis, experimental details, writing original draft. Hamada El-Gendi: methodology, investigation, formal analysis, writing original draft. Tarek H. Taha: conceptualization, investigation, writing (review and editing). Julie Basu Ray: conceptualization, final review, and editing.

Declarations

Conflict of interest The authors declare no competing interests.

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