



Dye Removal Using Cellulose Nanocrystals as an Adsorbent Extracted from Indian Water Chestnut Shells

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Abstract:

The cationic dye adsorption using nanocellulose is being investigated, with various nanocellulose-based adsorbents exhibiting good adsorption capacities. The dye molecules exhibit intense electrostatic interactions with the modified cellulose surface, which primarily accomplishes the adsorption process. The paper shows experimental research outcomes of removing two cationic dyes (Brilliant Green and Crystal Violet) from an aqueous solution using cellulose nanocrystals extracted from the Indian water chestnut shells (IWCS) as an adsorbent. The maximum adsorptive capacity of Brilliant Green was 43.59 mg/g, and that of Crystal Violet was 48.25 mg/g. The experimental data showed that 87.89 % of Brilliant Green dye and 96.5 % of Crystal Violet dye were removed. The results specify that the cellulose nanocrystals obtained from the IWCS were suitable for removing Brilliant Green and Crystal Violet dyes from an aqueous solution.

Keywords: cationic dye removal; adsorption; cellulose nanocrystals; aqueous solution; Indian water chestnut shells

1. Introduction

The survival of human beings, plants, and animals depends on water [1]. Due to increased industrial activities, urbanization, and the uninhibited usage of chemicals for various domestic purposes, the resultant pollution of water bodies can pose serious problems across the world [1]. Food processing, textile, leather, dyeing, and cosmetics are some of the industrial sectors that cause water pollution due to the discharge of dye chemicals [1]. Paper, fertilizer, electroplating of metals, leather processing, mining, fabric production, photography, beauty products, and pharmaceutical industries are among the many other sectors that use dyes and heavy metals [2]. Colourants, oils, surfactants, lubricants, carbon-based (organic) solvents, petroleum-based compounds, and pharmaceuticals form the major components of wastewater pollutants and industries such as fabric production, food, beauty products, leather processing, paper, and pharmaceuticals use artificial dyes that are classified as highly dangerous organic pollutants [3]. Large quantities of water are needed to achieve proper cleaning and efficient disinfection in the household textile laundering process, and subsequently, the effluents from laundering, various impurities, dyes, waste detergents, and microorganisms, are generated in huge quantities [4]. Dyes are used by textile, leather, plastics, and other industries, necessitating the usage of large amounts of water and subsequently producing a substantial amount of coloured wastewater [5]. Fabric production, plastic, paper, printing, carpet, beauty products, and food industries utilize over 10,000 distinct dyes and pigments for infusing them with the required colour to the products, and about 15 % of the dyes that remain in industrial wastes are released into water bodies causing undesirable visual pollution, reduction in the re-oxygenation capacity of water, contributing to acute, chronic toxicities and posing challenges to the treatment of water by conventional techniques [6]. The first pollutant that is observed in wastewater is its colour, and it is commonly accepted that the general public normally acknowledges the colour of the water as an indication of the quality of the water [5]. Dyes, herbicides, pesticides, heavy metals, etc., are the various types of contaminants found in industrial effluents that are disastrous if they are not treated properly, cause various incurable diseases, and also are life-threatening [7]. Dyes in wastewater that find their way to the water bodies inhibit photosynthetic activity because the dyes do not allow light to infiltrate the natural water bodies [8]. Small amounts at very low concentrations of micropollutants like dyes and heavy metals, as a result of pollution caused by industries, can tremendously degrade the quality of water in the waterbodies and also reduce the ability to dissolve oxygen effectively [2]. Industrial liquid wastes affect the water quality in the water bodies and thereby can cause serious health risks for plants, animals, and human beings [3]. Methyl orange dye can pollute the water even at small concentrations, and industries tend to produce large amounts of methyl orange dye as waste [3]. The malachite green dye is extensively utilized in fabric production industries for dyeing purposes, and its post-use impact poses severe threats to ecosystems and human beings [7].

A dye can be broadly categorized as a cationic, anionic, direct, disperse, vat, solvent, sulphur, or reactive based on its composition, characteristics, and applications. In addition to these, there are other classes [9]. Cationic dyes are hazardous, non-biodegradable, toxic, and carcinogenic, causing skin irritation, allergic dermatitis, and mutation effects, and about 12 % of the cationic dye produced annually is wasted and contaminates the industrial water streams [10]. Cationic dyes are bright-coloured, basic dyes that are widely employed in several sectors. Crystal Violet ($C_{25}N_3H_{30}Cl$, molecular weight 407.97 g/mol), the basic dye, is a triaryl methane dye used to colour paper and ink for ballpoint pens and inkjet printers. Crystal Violet (CV) is used to colorize various items, including leather, antifreeze, detergents, and fertilizers [11,12]. Brilliant Green ($C_{27}H_{33}N_2.HO_4S$, molecular weight 482.62 g/mol) exhibited a peak absorbance of 625 nm. Brilliant Green (BG) is a fundamental cationic dye. BG is extensively utilized across uses like dyeing of textiles, leather treatment, formulations for making cosmetics, and for applications concerning paper printing. BG is utilized in the production of green ink and serves as a staining agent in bacteriological media. BG serves as a topical disinfectant and also functions as a bacteriostatic agent in medium concerned with tissue culture, even though it poses significant risks to both human and animal health [13].

Low-cost and high-efficiency technologies for treating wastewater have seen a rising demand as the human population and economy continue to rise worldwide [7]. Numerous techniques (ozone treatment, adsorption, precipitation, multistep coagulation, flocculation, filtration, sedimentation) have been developed over the last few decades due to material engineering technical innovation for treating wastewater and for providing safe, affordable drinking water, and to fight water pollution [7]. Activated carbons, widely studied as effective adsorbents in water treatments, have been employed in the filtration of colours, heavy metals, and many other types of chemical contaminants [7]. Almuslem et al. [5] reported on the various procedures for the removal of dye from wastewater, membranes, etc. A variety of non-conventional, economical, and sustainable adsorbents sourced from industrial/agricultural waste and natural materials have been utilized for dye removal from wastewater [4]. Typical adsorbents consist of activated carbons, agricultural-based solid wastes, industrial by-products, clay minerals, polymeric resins, biomass, etc [4]. Membrane separation, chemical and biological degradation, and electrochemical techniques are some of the related concepts of adsorption wastewater technologies, and physical adsorption has emerged as one of the most practical techniques for managing wastewater [7]. Adsorption, photodegradation, chemical oxidation, advanced oxidation process, and biological treatment approaches have been described for the treatment of water concerning textile dye treatment [2]. Various techniques, including ion exchange, membrane treatment, electrolysis, coagulation/flocculation, electrodialysis, and adsorption, were used to remove the non-biodegradable dyes. Among these, adsorption has emerged as the superior technique offering numerous advantages such as flexibility, lesser initial cost, ease of design, resistance to toxic pollutants, and simplicity in operation [6].

When compared with other systems for reusing water, the adsorption technique, which is a well-known equilibrium separation method, was found to be an excellent technique for water purification, and also concerning the initial cost, design simplicity, operational ease, simplicity of design, sensitivity to harmful contaminants, and flexibility [5]. Adsorption is widely regarded as the most efficient process for removing dyes or other forms of pollutants from water [6]. A diverse range of adsorbents utilized includes minerals, organic materials, activated carbons, clays, zeolites, silica beads, polymeric resins, and biomass [6]. Adsorption is a highly recommended technique for dye decontamination [14]. The adsorption process of dye removal is preferred as it helps in the removal of various types of colouring chemicals when compared with many other dye removal processes [5]. The adsorption method is cost-effective and time-effective [2].

In recent years, cellulosic polymers have gained attention as promising adsorbents for eliminating dyes and heavy metals from water [2]. These polymers offer several advantages, including biodegradability, hydrophilicity, recyclability, and non-toxicity [2]. Modified cellulose, sunflower stalk residue, sawdust, and fabric wastes of cellulose have been investigated for dye extraction from aqueous solution [7]. Chitosan, a cationic biopolymer with natural antibacterial activity, has been widely used after cellulose [7]. Good chemical stability, together with ease of chemical modification, is possible concerning cellulosic polymers, as the structural features of the cellulosic polymers are well studied [2]. The abundance of availability, organic nature, inexpensive, renewability, biocompatibility, biodegradability, high aspect ratio, reactive surface of -OH groups, excellent mechanical strength, and huge specific surface area make cellulose a perfect material for dye removal [6].

The distinctive properties of cellulose nanofibers resulted in the advancement of cellulose nanofibers in the nanotechnology domain [1]. The growing demand for efficient dye removal has led to the increased use of nanomaterials due to their high adsorption efficiency, expansive surface area, and the presence of an abundance of dynamic spots for pollutant interaction [6]. The requirement for strong surface interactions between adsorbents and adsorbates, along with many factors like large surface area, numerous adsorption sites, high adsorption capacity and rate of adsorption, and reduced reaction time, makes nanomaterials a preferred choice in advanced water treatment technologies [6]. Cellulose Nanowhiskers (CNW), Cellulose Nanofibers (CNF), and Cellulose Nanocrystals (CNC) are found to have various types of surface functional groups, particularly the ionic groups [15]. The ionic surface functional groups, including sulfates, are introduced in the surface of CNW, CNF, and CNC due to the acid hydrolysis mediated with the help of sulphuric acid [15].

Nanocellulose is extracted using multi-step procedures that require a considerable amount of time [7]. Nanocellulose has shown its potential to be an effective natural adsorbent, making it a capable solution for removing harmful wastewater contaminants [5]. Nanocellulose can be successfully used as a nano dye adsorbent

concerning wastewater treatment [16]. For low-dosage applications concerning industrial wastewater treatment reactors, nano adsorbent from bio-waste can be used for effective dye removal, thereby proving itself to be an economically feasible option [16]. Nanocellulose adsorbents exhibit good potential for eco-friendly wastewater dye removal due to mechanisms like enhanced dye adsorption through electrostatic forces, physisorption, and hydrogen bonding [17].

Magnetic nanocellulose prepared from raw sugarcane [18], nanocellulose based filter paper made from *Neolamarckia cadamba* [1,19], cellulose nanocrystals prepared from sugarcane bagasse [5], cellulose nanocrystals prepared from the residue of bioethanol production from wood [20], cellulose nanofibers obtained from Norwegian spruce wood [3], porous bio-based polyurethane hybrid composites reinforced with modified nanocellulose developed from grape seedoils and chaulmoogra in addition with *moringa olifera* sea pods [21], nanocellulose fibers derived from coconut (*cocos mucifera*) coir [16], amino-functionalized nanocrystalline cellulose from fully bleached hardwood kraft pulp [6], nanocellulose chitosan composite beads [7], cellulose nanocrystals extracted from wastepaper and the subsequent chitosan based fabricated nanocomposite membrane [22], bacterial nanocellulose manufactured using stillage of rice beer [23], nanocellulose films, derived from sugarcane bagasse, enhanced with manganese oxide [24], nanocellulose hydrogel prepared using soy hull [25], water resistant biomass aerogel composed of nanocellulose and gelatin [26], nanocellulose prepared from corn cob and subsequent fabrication of electrospun biocomposite made along with chitosan together in an poly(hydroxyalkanoate) matrix [27], chemically treated nanocellulose nanocrystals extracted from sugarcane bagasse [28], nanocellulose isolated from sweet lime peel [29], *Komagataeibacter saccharivorans* bacterial nanocellulose/graphene oxide nano-biocomposites [30], nanocellulose prepared from sugarcane bagasse agricultural waste [14], nanocellulose from pinecones wastes [31], nanocellulose obtained from durian husk [32], cellulosic nanoadsorbent prepared from the fruit coat of Kendu (*Diospyros melanoxylon*) [33], magnetic nanocellulose based on olive waste [34], nanocellulose composites made from oil palm empty fruit bunch [35], nanocellulose/chitosan nanocomposites synthesized from palm leaves [36], nanofibrillated cellulose filter paper made using *Shorea roxburghii* [37], nanocellulose crystals from rice straw and the subsequent preparation of magnetic chitosan/nanocellulose aerogels [38], cinnamon nanocellulose [39], wet-resistant cellulose nanofibril-based aerogel from sugarcane bagasse [40], treated sugarcane bagasse nanocomposite prepared from sugarcane bagasse agricultural waste [10] were some of the nanocellulose materials used as an adsorbent for dye removal. Different plant-based nanocelluloses are being extracted, and their properties are studied to understand them better for use in various applications, including dye removal. The potential of CNC derived from the IWCS for removing CV and BG dyes from water remains largely unexplored. Hence, the CNC extracted from the IWCS was investigated to understand its performance in eliminating BG and CV dyes from an aqueous solution.

2. Materials and methods

Indian water chestnut shells (locally known as singhara) obtained from a local market in Kanpur, Uttar Pradesh, India, are used as raw material to extract CNC. The extracted CNC was used as an adsorbent to investigate the removal of two cationic dyes, i.e., BG and CV, from an aqueous solution.

2.1 Extraction of cellulose nanocrystals from the Indian water chestnut shells

The cellulose was extracted using a method suggested by Hemmati et al. [41]. from the dried powdered IWCS with chemical treatment processes such as the dewaxing process using a benzene/methanol mixture, alkaline treatment using a NaOH solution, and the bleaching process using sodium chlorite. After extracting cellulose, acidic hydrolysis using concentrated sulphuric acid was performed to extract the CNC using a method suggested by Hemmati et al. [41]. The size of the CNC was obtained using particle size analysis. The crystallinity index was found with the help of the methodology adopted by Segal et al. [42] and Nam et al. [43] using X-ray diffraction analysis. The methodology adopted for the extraction of CNC from the IWCS (agricultural waste) and their characterization was given by Tiwari and Sanjog [44].

2.2 Dye removal using a batch adsorption study

The adsorption of BG dye and CV dye using CNC from IWCS was implemented using the methodology given by Poornachandhra et al. [45] and Paul et al. [46]. A stock solution of 1000 ppm of BG and CV dye was prepared. An amount of 0.1 g of each dye was dissolved in 100 mL of distilled water.

The adsorption experiment was performed in a shaker at 100 rpm for 120 min at room temperature (25 °C) to obtain a better-optimized output. The five different concentrations (20, 40, 60, 80, and 100 ppm) of BG and CV dyes were prepared using a stock solution. 25 mL of each concentration was taken in a conical flask, and the absorbance of all five concentrations was evaluated with the help of a UV-visible spectrophotometer at a maximum wavelength of 624 nm and 585 nm for BG and CV. The pH for the BG dye solution was maintained at 7, and CV at 8, as this pH results in maximum adsorption efficiency.

For the adsorption, 0.05 g of the CNC adsorbent was used in 25 mL of the dye solution. The concentration was 100 ppm. After the adsorption, the residual solution was filtered and tested for absorbance using a UV-visible spectrophotometer. The formulae used to calculate the maximum adsorptive capacity and percentage of removal are as follows.

$$Q_e = (C_0 - C_e) V$$

$$\% \text{ Dye removal} = \frac{W}{C_0} (C_0 - C_e) * 100$$

where Q_e refers to the maximum adsorptive capacity in mg/g, C_0 is the pre-adsorption concentration in mg/L, C_e is the post-adsorption concentration in mg/L (calculated by dividing absorbance by the slope of the curve), V is the volume of adsorbate solution in L, and W is the weight of adsorbent in grams. The weight (W) of the adsorbent was 0.05 g, and the volume (V) of the adsorbate was 1000 mL.

Adsorption was performed on a 100 ppm concentration of dye solution. Adsorbent (0.05 g) was added to the dye solution. The sample is kept for 120 min in an orbital shaker. After 120 min, the solution becomes clear, indicating the successful removal of the dye. The solution is filtered and tested for absorbance using a UV-visible spectrophotometer. The percentage of removal and maximum adsorptive capacity have been calculated from the observed values mentioned in the tables below.

3. Results and discussion

The IWCS was used as the raw material to obtain the CNC, which was utilized as an adsorbent to remove BG and CV dyes from aqueous solution. The different concentrations, absorbance before and after adsorption, maximum adsorptive capacity, and the percentage of dye removal were investigated. The CNC exhibited an average size of 265 nm in diameter and 83 nm in width. The crystallinity index of the CNC was 65 %.

Table 1 shows the different concentrations and absorbance of BG dye. Table 2 shows that the absorbance before adsorption was 100 mg/L, and after adsorption using 0.05 g of adsorbent was 12.82 mg/L. The maximum adsorptive capacity is 43.59 mg/g. This shows that 43.59 mg of dye will be removed per gram of dye from the aqueous solution. 87.89 % of the BG dye was removed.

Table 1: The absorbance of different dye concentrations of BG dye.

Brilliant Green	
Concentration (C_0)	Absorbance
20	0.93
40	1.07
60	1.12
80	1.17
100	1.21
Slope	0.0156

Table 2: Brilliant Green dye after adsorption.

Concentration (C_0) (mg/L)	Absorbance	Post Adsorption Concentration (C_e) (mg/L)	Maximum Adsorptive Capacity (Q_e) (mg/g)	Dye Removal (%)
100	0.2	12.82	43.59	87.89

The graph showing the absorbance versus the concentration for the BG dye is shown in Fig. 1. The values based on the curve shown in the graph were used to calculate the slope. The slope is used to calculate the value of post-adsorption concentration.

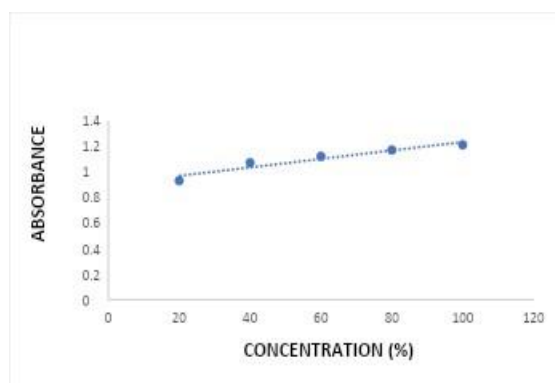


Fig. 1: Absorbance versus concentration graph for Brilliant Green dye.

The different concentrations and absorbance of the CV dye are shown in Table 3.

Table 3: The observation table of Crystal Violet for different concentrations and absorbance.

Crystal Violet	
Concentration (C ₀)	Absorbance
20	0.034
40	0.045
60	0.058
80	0.068
100	0.092
Slope	0.0007

Table 4 shows the value of absorbance before and after adsorption. Before adsorption, the concentration is 100 mg/L, while after adsorption, it is 3.5 mg/L. The maximum adsorptive capacity is 48.25 mg/g. The result shows the best adsorption of the adsorbent on CV dye, removing 96.5 % of the dye from the aqueous solution.

Table 4: Crystal Violet dye after adsorption.

Concentration (C ₀) (mg/L)	Absorbance	Post-Absorption Concentration (C _e) (mg/L)	Maximum Adsorptive Capacity (Q _e) (mg/g)	Dye Removal (%)
100	0.00245	3.5	48.25	96.50

The graph showing the absorbance versus the concentration for the CV dye is shown in Fig. 2. The values based on the curve shown in the graph were used to calculate the slope. The slope is used to calculate the value of post-adsorption concentration.

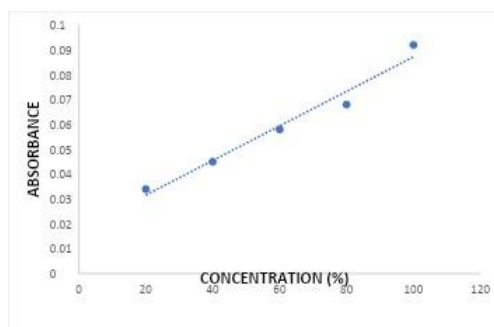


Fig. 2: Absorbance versus concentration graph for Crystal Violet dye.

CNC demonstrated its effectiveness for removing Methylene Blue (MB) from a pH 9 solution. At 25 °C, CNC achieved a peak adsorption capacity of 118 mg dye/g CNC [47]. Polyvinyl alcohol, chitin, and nanocellulose-based reinforced biopolymer composites cross-linked using Maleic acid were utilized for the MB cationic dye removal, and the peak adsorption capacity was 467.5 mg/g with an adsorption percentage of 83.67 ± 1.08 % [48]. High adsorption, like 3429.23 mg/g for Congo Red using cellulose-based aerogel and 2282.3 mg/g adsorption capacity for removal of Malachite Green employing nanocellulose-based magnetic adsorbents, has been achieved [17]. A summary of the nanocellulose-based adsorbents for removing different dyes (Congo Red, Methylene Blue, Acid Orange 7, Acid Red, Methyl Orange, Malachite Green, Reactive Black 5, Direct Blue 78, and Safranin-O) has been tabulated by [17], wherein, it has been observed that the dye adsorption rates varied from 17.12 mg/g to 3429.23 mg/g. The values of maximum adsorption capacity of various vegetable biomass from various plant parts like leaf, leaf powder, stem, fruit, seed, and seed powder concerning the retention of CV dye, have been tabulated by Mosoarca et al. [49], wherein, it has been observed that the dye adsorption rates varied from 4.14 mg/g to 322.58 mg/g. The adsorption capacity of the royal palm leaf sheath subjected to the ball-milling process was 454.5455 mg/g of CV dye from synthetic wastewater [50]. The CV dye exhibited adsorption of about 88 % for the ball-milled royal palm leaf sheath and 67 % for the royal palm leaf sheath at pH 6 levels and for the contact time of 60 minutes [51]. Crystalline cellulose extracted from sugarcane bagasse was able to absorb CV dye in an aqueous medium and demonstrated the peak adsorption of 8.857 mg/g [51]. Treated sugarcane bagasse demonstrated a peak adsorption of 107.5 mg/g and 112.9 mg/g for CV and MB dyes, respectively [10]. The removal percentages of the CV and MB dyes using treated sugarcane bagasse were 96.4 % and 96.0 % respectively [10]. For sugarcane bagasse, the removal percentages were 64.8 % for CV dye and 63.2 % for MB dye, respectively [10]. The properties (molecular structure) of the dyes, environmental conditions (temperature, pH, contact time, and mixing speed), and nanocellulose characteristics (dosage, surface chemistry, and surface) considerably impact the adsorption characteristics of nanocellulose-based adsorbents [17].

The best adsorption and removal percentage was seen on the CV dye when compared to the BG dye. The results, when compared with the results given by other researchers, indicate that the CNC derived from the IWCS is effective, efficient, and comparable with other similar cellulosic materials for the removal of cationic dyes like BG and CV. However, there is ample scope for further research, taking into consideration the variations in parameters like temperature, mixing rate, concentration, contact time, pH values, etc.

4. Conclusion

The research shows that the cellulose nanocrystals extracted from the IWCS are an effective adsorbent for removing BG dye as well as CV dye. The result shows that 0.05 g of the adsorbent is capable of removing 87.89 % BG dye and 96.5 % CV dye. The peak adsorption capacity of the BG dye was 43.59 mg/g at pH 7, and that of CV was 48.25 mg/g at pH 8. Cellulose nanocrystals as adsorbents may be more suitable for removing many other similar cationic dyes that are very harmful to the environment, water bodies, and human life.

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