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Acetylcellulose Recovery from Waste Residual for Attenuating Reactive Dye from Aquaculture Waste as a Fascinating Synergistic Ecology Effect

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Abstract: Waste valorization is attracting not only the scientific world but also the world. Acetylcellulose wastes from cigarette filter residuals are signified as the largest global impact of solid waste. Acetylcellulose recycling for desired products is a promising way for environmental management. In this regard, the current investigation is dealing with the immersion of residual filters sequentially into aqueous solutions of alkali and ethanol before water washing, which converts them into a superhydrophobic acetylcellulose adsorbent material. The morphology and characteristics of the acetylcellulose fiber were characterized using a Scanning Electron Microscope (SEM) and Fourier-Transform infrared spectroscopy (FTIR). The adsorption tendency was checked for Levafix Blue dye compromised in an aqueous stream as a model textile polluted effluent. The experimental results exposed that the acetylcellulose fiber displayed a sensible textile dye elimination from the dying stream. Langmuir isotherm is well fitting the adsorption matrix and the reaction follows the 1st-order kinetic model. The so-obtained acetylcellulose fiber showed tremendous efficiency for dye removal from aqueous effluent. The attained maximum monolayer adsorption capacity was recorded as 4.8 mg/g at pH 3.0 and an adsorbent dose of 1 g/L through the isotherm time of 2 h. Also, temperature elevation could increase the adsorption capacity to 5.7 mg/g. Due to this excellent affinity to adsorb dye at an economic wise rate is shown as a promising candidate for textile dye elimination form aqueous effluent.

Keywords: acetylcellulose waste; levafix dye; adsorption; wastewater treatment; isotherm models

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1. Introduction

Since the twentieth century, there has been an ever-increasing alarm for the existence of solid waste that causes extreme toxicity to the environment [1]. To a large extent, the presence of such waste in the environment is causing ecological damage [2]. This necessitates appropriate adequate treatment and management before eliminating their disposal into the ecosystem. Hence, converting such waste-based materials for new ecologically benign substances is attaining the scientific community. In this concept there has been some very recent but limited progress in the search for greener materials from solid waste [2,3].

Worldwide, the most littered solid waste is signified as a cigarette filter [4–6]. Trillions of cigarette filters are signified as a hazard solid waste and is deposited per annum. Instead, 97% of such filters were compromised as a modified natural polymer that is so-called acetylcellulose fiber (ACF) [4]. Although this acetylcellulose waste is categorized as undervalued material, the scientists' and researchers' interest is seeking sustainable elucidations for tackling cigarette filter pollution concerns [5]. Till now, generally, solid waste pollution

management includes landfilling incineration, recycling, and degradation [7]. Numerous research works have been conducted to convert cigarette filter waste into reusable material for various applications to attain the circular economy concept [8]. For instance, it has previously been applied in the environmental engineering field, infrastructures and building materials, metallurgical engineering, and energy storage devices. However, according to the research cited [9] minimal application of cigarette filters in environmental engineering, compared to wide applications in buildings and structure fields.

An encouraging technique to control the environmental impacts of cigarette filters is to convert them into desired products in a circular economy concept. Acetylcellulose [10] which is an economically attractive and usable adsorbent material is the main constituent of cigarette filters. Furthermore, acetylcellulose fiber (ACF) was previously reported in the literature [11–15] as an excellent substance with great potential for adsorption capacity. That is a superior substance for the field of separation.

The contamination of dyes has developed into a significant environmental issue with the development of the textile industry. Due to their severe toxic effectiveness, and non-biodegradable and bioaccumulated characteristics, dyes in aqueous media pollution pose a serious threat to the environmental biosphere and public health [16]. Due to the consumption of massive amounts of water in textile dying processes, an ultimate amount of wastewater is generated. Such wastewater mainly contains unconsumed dyes and chemical activities that represent a great environmental concern worldwide [17,18]. The result is a direct threat to the aquatic system when these effluents are not treated before their final disposal [6,19–21]. However, the textile discharge is composed of toxic and stable substances that are difficult to remove. Textile industry discharges pose high loads of various dyes exhibiting vivid colors and posing significant challenges for degradation using conventional methods. These characteristics result in reduced water transparency, leading to the obstruction of solar radiation penetration into the ecosystem which is essential for photosynthetic processes. The result is a massive disturbance to the aquatic creatures [6].

Therefore, for the great significance of such aqueous waste, it must undergo treatment and management. Effluent discharged from textile industries is eliminated based on various techniques including physicochemical treatments such as adsorption [20], coagulation, sedimentation, and filtration. Also, biological treatments based on activated sludge are applied [21]. Such techniques unfortunately are unfavorable since they generate a large amount of sludge by-product and are not efficient at destroying or eliminating dye molecules from aqueous effluent [6]. Thus, the appropriate adsorption system offers a flexible design for efficient and economic operation concerning high superiority to reclaim water [17,20]. However, natural and waste materials have attracted interest as alternatives to synthetic adsorbents due to economic regard and chemical consumption [6,21–24]. It is noteworthy to mention that acetylcellulose is signified as an inert polymer, that is benign and odor-free. Hence, the recycling of smuggled cigarette filters for pollutant elimination is extremely attractive [25–28]. Notably, acetylcellulose could be derived from fruit branches and other naturally occurring substances [29–34]. However, using acetylcellulose from cigarette filter waste sources is significant since its presence in such waste material causes massive harm to the environment. Acetylcellulose is a photodegradable polymer that is not easily biodegradable therefore, its occurrence may persist in the environment for years [32–36].

The uptake of contaminants from aqueous effluents using waste valorized materials is vital not just for toxicity exclusion, but also for valued waste recovery. The application of the adsorption technique for the determination of different analyses in different matrices is a valuable alternative due to the availability of a large range of valuable adsorbents. Good adsorbent properties that involve high surface area, high removal efficiency, and reusability support them to be good candidates for environmental remediation [19,21]. In this sense, this current investigation aims to prepare an economic adsorbent from the waste cigarette filters for the adsorption of reactive dye in an aqueous medium. The capacity of the material for Levafix blue adsorption and the effects of different parameters, such as the

isotherm time, the aqueous media pH, temperature, adsorbent dose, and dye loading are investigated. Also, the adsorption kinetics and isotherm models are assessed to evaluate the adsorption system.

2. Experimental Investigation

2.1. Acetylcellulose Fibers Collection and Preparation

Acetylcellulose fibers derived from cigarette filters are used as an adsorbent material. Initially, the cigarette filters are collected from a Coffee shop station near the University of Menoufia, Egypt before subjection for cleaning and treatment. Initially, the smoked filters are de-warped from the covered paper and subjected to successive washing with distilled water. Then it is ultrasonically cleaned in ethanol alcohol to remove any remaining impurities. Subsequently, the cleaned fiber is removed from the solution by filtration. Afterward, the samples were subjected to oven drying at 105 °C to eliminate any moisture content before their utilization. Then, ACF samples were stored in a dry place at room temperature for further use.

2.2. Wastewater

Levafix Blue CA is signified as a kind of reactive dye and was delivered by DyStar Ltd., (Raunheim, Germany) where the aqueous effluent containing a synthetic textile dye was employed as a representative model. Levafix Blue dye is a dark blue powder, which is categorized as a bifunctional and combined anchor. The dye powder was applied for use with no further purification or treatment. The aqueous solution is arranged by preparing a stock solution of 1000 mg/L of the dye, which was then diluted as needed to the required concentrations.

2.3. Characterisation Study

The sludge samples that were prepared underwent examination and imaging using a Field-emission scanning electron microscope (FE-SEM, Quanta FEG 250). Fourier Transform Infrared (FTIR) spectra analysis of acetylcellulose (performed using Jasco, FT/IR-4100, type A) was conducted to determine the specific functional groups involved in the adsorption test. Additionally, the particle size distribution of the acetylcellulose material was determined using the IMAGEJ 1.48 V program.

2.4. Methodology and Analytical Determination

100-mL of Levafix Blue CA dye-containing wastewater was poured into a glass container to investigate the dye loading on its removal efficiency. The initial solution of the aqueous environment's pH was adjusted, when needed, to the desired values by using diluted H_2SO_4 (1:9) and/or 1.0 M NaOH solutions. The desired pH values were achieved by utilizing a digital pH meter (Model AD1030, Adwa instrument, Szeged, Hungary) to make necessary adjustments. Subsequently, ACF-material was added to the wastewater, and the reaction was conducted for dye reduction. Subsequently, after selected time intervals, samples were withdrawn periodically and subjected to spectrophotometric analysis after the samples were filtered using a micro-filter. Spectrophotometric performances were applied using Model Unico UV-2100, USA at the maximum absorbance peak of a wavelength of 610 nm which was monitored and applied to evaluate the amount of residual Levafix Blue CA dye remaining in the aqueous effluent after adsorption.

The schematic representation of the graphical experimental set-up and steps is illustrated in Figure 1. Adsorption capacity (q_e) is investigated by using initial (C_o) and final (C_t) Levafix Blue dye concentrations, Levafix Blue solution volume (V), and acetylcellulose adsorbent mass (m) as Equation (1):

$$q_e = (C_o - C_t)\frac{V}{m} \quad (1)$$

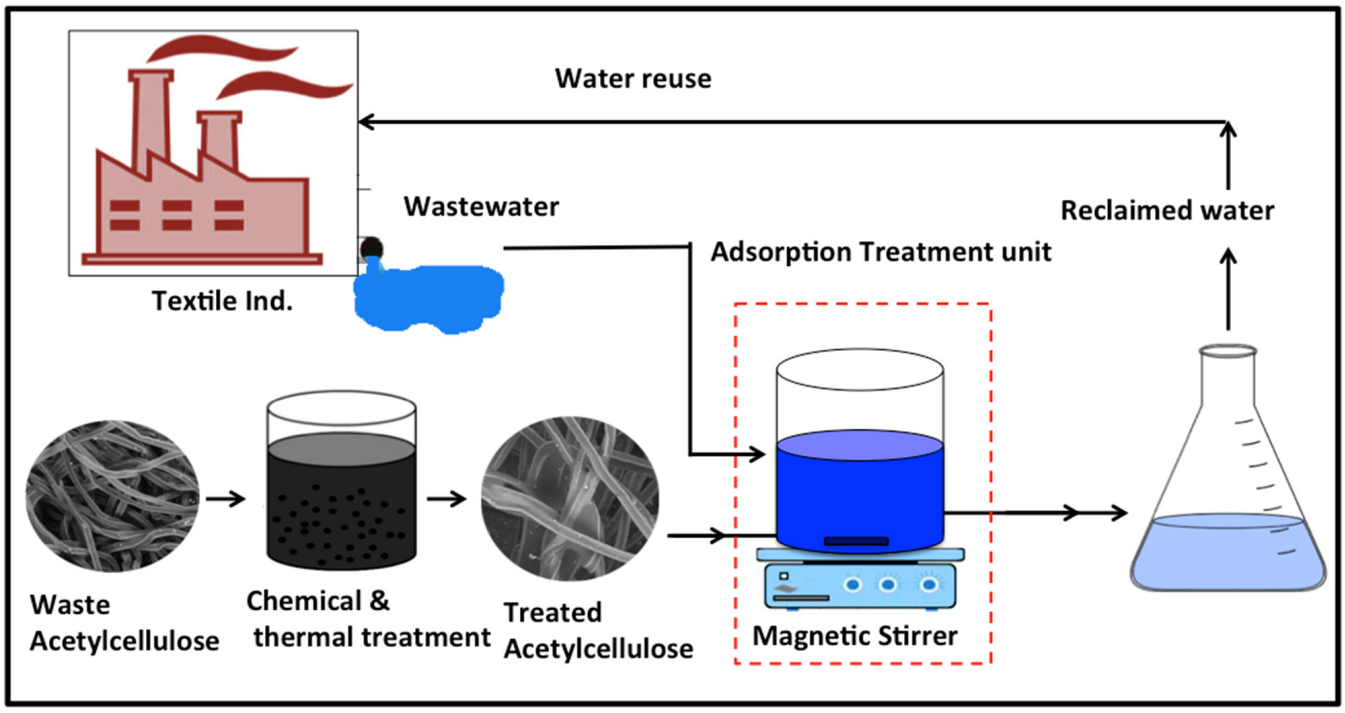

Figure 1. Overview of converting waste fibbers into valuable acetylcellulose adsorbent material.

3. Results and Discussions

3.1. Characterization of the Prepared Fiber

3.1.1. Acetylcellulose SEM Imaging

The morphological analysis of acetylcellulose fiber, ACF, is conducted by scanning electron microscopy (SEM) to examine the morphological changes that occur before and after cleaning. Scanning electron micrographs of fibers prior to and after cleaning are exhibited in Figure 2. Figure 2a,b,d, and e display the slightly organized straight fibers with narrow pores on their surfaces of ribbon-shaped fibers. Notably, after treatment (Figure 2d,e), the non-cellulosic materials are removed in the treatment process; hence, the values for fineness in the treated fibers are increased. It was observed that the volume of the constituent fibers prior to (Figure 2a–c) and after (Figure 2d,e) treatment declined by the cleaning process. Consequently, the surface area of the fibers and the amount of the available exposed cellulose on the fiber surface are increased which improves the adsorption matrix through interfacial bonding. Hence, this affinity might possess a positive impact on the adsorption process.

Also, the particle size distribution was calculated, The corresponding values are illustrated in Figure 2, specifically in panels (c) and (f) for acetylcellulose fibers prior to and after treatment, respectively. The acetylcellulose fiber diameter distribution exhibited a narrow change as illustrated in Figure 2c,f. It is notably exhibited that particle size is lesser in the case after treatment which means more sites are available since the surface area is smaller than the untreated acetylcellulose fibers ribbon.

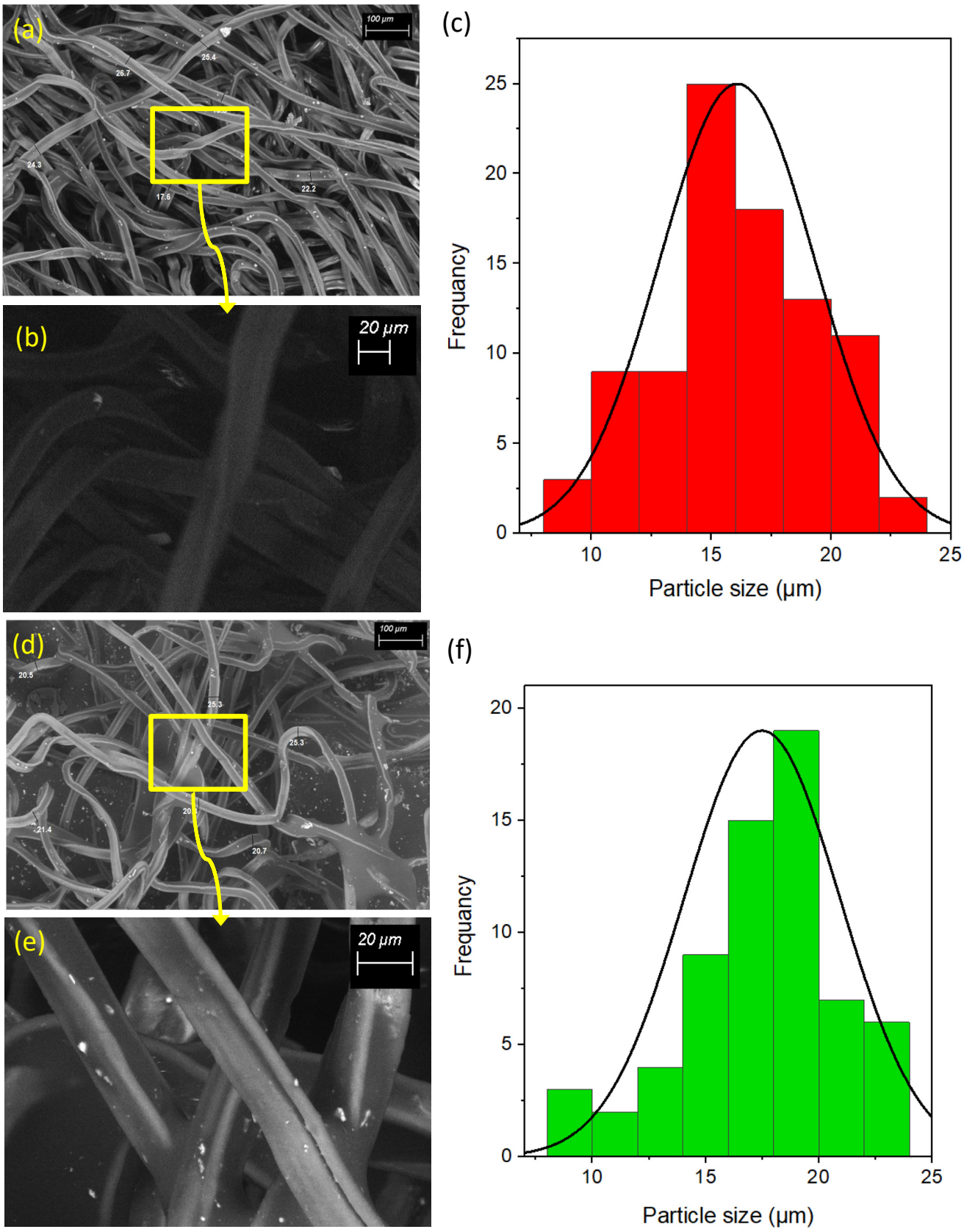

Figure 2. Ribbon-shaped SEM micrographs of acetylcellulose fibers, ACF: (**a**,**b**) used fibers, (**d**,**e**) cleaned fibers with different magnification; (**c**,**f**) are particle size distribution for used and cleaned acetylcellulose fibers.

3.1.2. Acetylcellulose FTIR

The Fourier transform infrared (FTIR) spectroscopy of both acetylcellulose prior to and after cleaning are investigated and are displayed in Figure 3. As shown in Figure 3, both curves (a and b) illustrate the bands at 1030, 1327, 1432, and 1756 cm^{-1} appearing in both ACF samples that reflect the presence of various groups. The acetylcellulose fiber exhibited similar band intensities at 1104 cm^{-1} are an indication of the presence of C-O. Furthermore, the distinctive peak associated with O=C bonding should be placed at wavenumber 1721 cm^{-1}. The band at 778 cm^{-1} corresponds to the CH bending mode, which appears in the cleaned sample. The newly formed bonds of CH, C-O, and O=C to the cleaned acetylcellulose, 778, 1104, and 1721 cm^{-1}, are not prominent. 778 cm^{-1} is corresponding to the CH group and 1104 cm^{-1} is signifying C-O bonding, however, O=C is signified by 1721 cm^{-1} band. Nevertheless, the band of 1049 cm^{-1} was attained in the used sample without cleaning and disappeared in the cleaned sample, which corresponds to C-O group. The spectral bands observed in the range of 3000 to 2900 cm^{-1} can be attributed to the symmetrical stretching of CH_3 groups. Additionally, the notable bands appearing around 3500 and 3400 cm^{-1} are indicative of OH stretching. [6,20]. These results indicate few differences are observed between ACF-used and cleaned ones. Also, the FTIR spectrum data reveals that ACF- contain cellulose acetate in their composition. Further, the treatment process did not promote the destruction of the cellulose fiber.

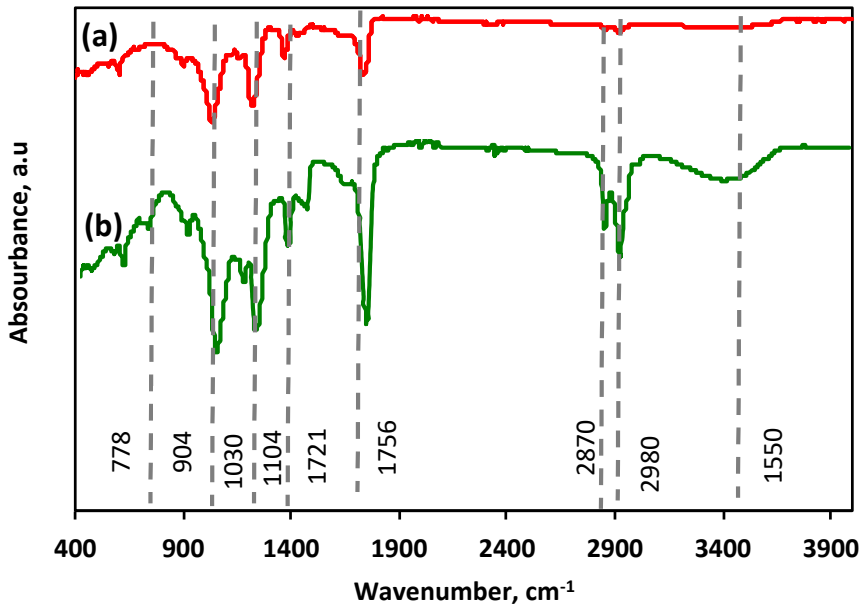

Figure 3. FTIR of acetylcellulose fibers, ACF: (**a**) used fibers, (**b**) cleaned fibers.

3.2. Application of Acetylcellulose Fiber for Levafix Blue CA Removal

3.2.1. Determination of Equilibrium Isotherm Time

Initially, prior to scheming the sorption matrix, it is essential to assess the isotherm equilibrium time profile. The time of Levafix Blue dye sorption through acetylcellulose ribbon fiber was explored at room temperature, and the corresponding results are displayed in Figure 4. Data compared the sorption capacity of used and cleaned cigarette filters as presented in Figure 4a, and b, respectively.

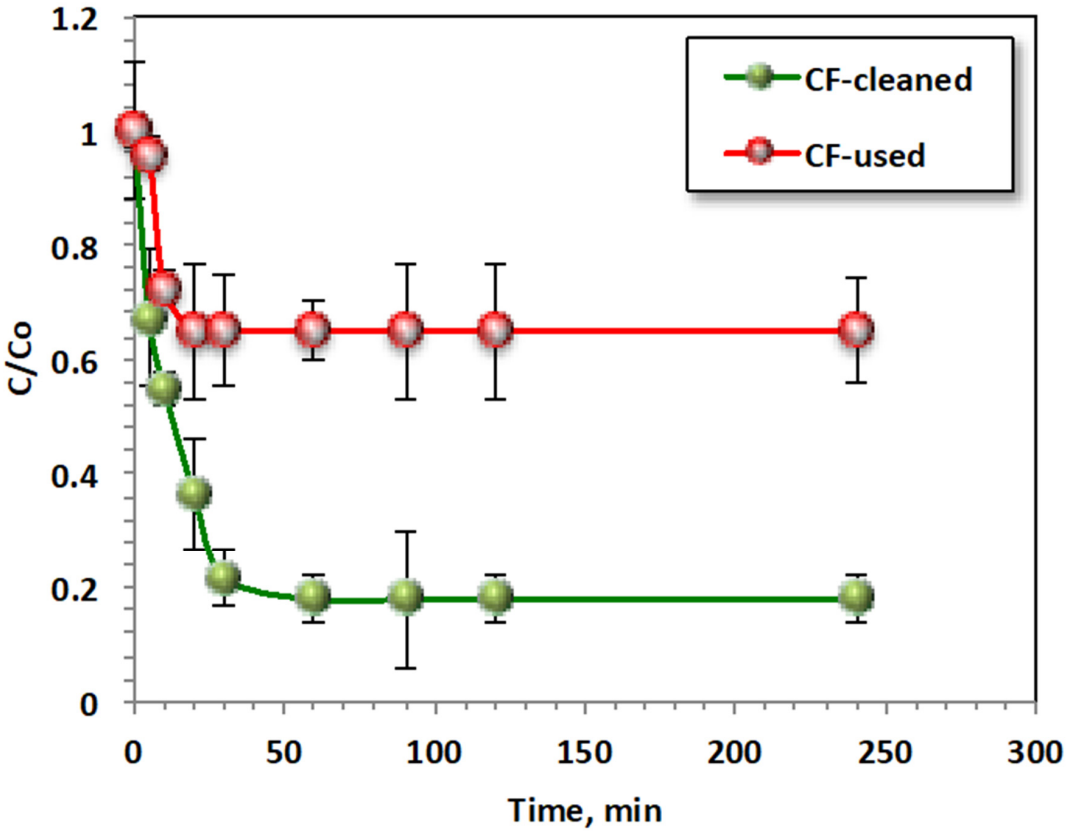

Figure 4. Adsorption isotherm time-profile on the Levafix Blue CA dye uptake with various adsorbents (Levafix Blue 25 mg/L; ACF ribbon 1 g/L; pH 6 and 298 K).

Cleaned filters showed higher sorption capacity for Levafix Blue than the unclean one, most of the dye was adsorbed within the first two hours. The acetlycellulose ribbon which possesses available clean pores is more pronounced for adsorption instead of the non-clean ribbon. Due to acetlycellulose's special characteristics of hydrophobicity behaviors towards water and excluding water, the ACF ribbon adsorbed dye molecules. Thus, the cleaning process might provide more specific sites. The importance of acetylcellulose in enhancing the adsorption capacity aligns with the findings reported by Yang et al. [10], as stated in previous published studies. Their results showed high significant adsorption uptake of Au(III) onto acetlycellulose fiber. The surface of acetylcellulose demonstrates efficiency in adsorbing dye molecules through hydrophobic interactions. These findings provide evidence for the crucial role of acetylcellulose's hydrophobic nature, enabling higher dye uptake. Moreover, as depicted in Figure 4, both adsorbents exhibit additional dye uptake beyond the initial 2 h period, primarily due to the occupation of available active sites. However, once the acetylcellulose fibers become saturated, there is no further dye uptake observed.

3.2.2. Effect of Various Adsorption Parameters

For the object of maximizing the adsorption uptake, the effect of the adsorption parameters was investigated and the results are shown in Figure 5. Initially, the adsorbent dose plays a significant role in the degree of adsorption uptake. In this regard, to estimate the effect of adsorbent dose, adsorption experiments were conducted using various adsorbent doses of ACF ribbon in the range of 0.25 to 2 g/L, whereas other adsorption parameters are kept constant (Levafix Blue concentration of 25 mg/L, temperature pH 6.0 and 298 K). Results illustrated in Figure 5a estimate the adsorption capacity of Levafix Blue enhanced with the elevation in ACF ribbon Sorbents amounting up to 1 g/L since the upsurge in the

available adsorption sites. However, further elevation in the ACF ribbon does result in a deduction in Levafix Blue uptake.

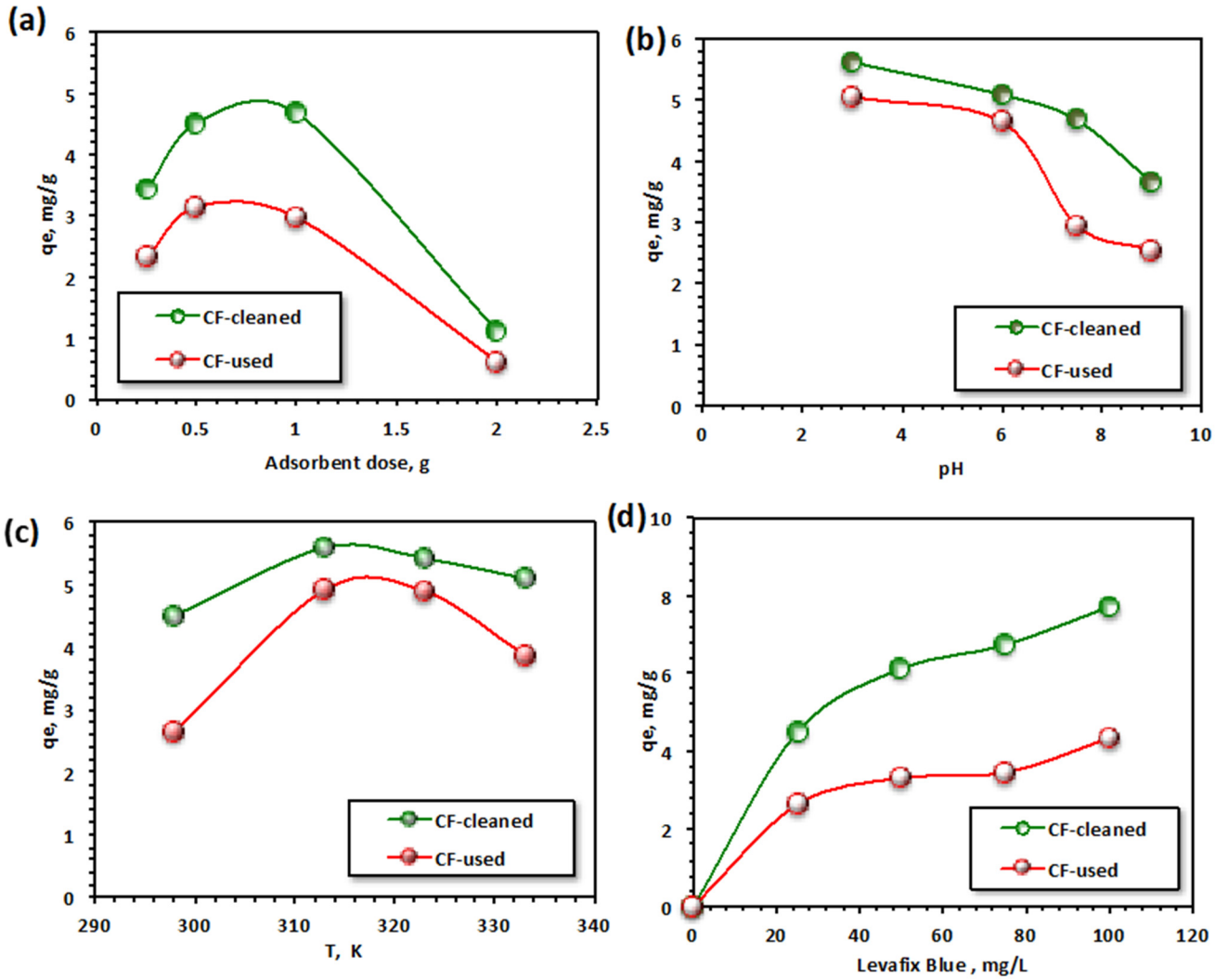

Figure 5. Equilibrium adsorption uptakes are obtainable at various (**a**) adsorbent dose; (**b**) pH; (**c**) temperature and (**d**) different initial dye concentrations.

The data in Figure 5a might be associated with the upsurge of adsorbent concentration at a constant adsorbate loading that results in the unsaturation of sorption sites. Also, the extra ACF ribbon that is present in excess could trigger the aggregation and ribbon contact with each other. Hence, the result was a deduction in the overall surface area of the available ACF ribbon. Thereby, the ACF ribbon dose is proportional to Levafix Blue uptake. To add up, the Levafix Blue removal drop could be a result of the restabilization of the dye particles. Such results are in agreement with the previous reports of Chu [22] for dye molecule adsorption with different adsorbent materials.

To evaluate the pH effect and maximize the adsorption capacity, the pH effect on the adsorption capacity was assessed at different pH values (3, 6, 8, and 10) whereas all other operational parameters are fixed. The data presented in Figure 5b indicates that, for all ACF ribbon samples, the maximum adsorption uptake is achieved at a pH of 5.6 and 5.04 for the cleaned and used ACF fibers, respectively. However, a further increase in pH value leads to a decline in the adsorption capacity, particularly at alkaline pH levels. This phenomenon can be attributed to the potential damage to the active sites on the ACF ribbon adsorbent material. Additionally, the pH value influences the surface charge of the ACF material. As a result, the adsorption of Levafix Blue dye is closely related to the surface charge of the ACF

ribbon, rather than the pH of the solution. Also, the increase in the solution's pH weakened the attraction between the ACF ribbon and the Levafix Blue aqueous solution. Chu [22] reported the pH value might affect the removal of dye using alum sludge adsorbent.

Figure 5c shows the temperature effect on Levafix Blue adsorption capacity since the industrial discharge may be disposed of at different temperatures. From this concept, the aqueous solution temperature was adjusted over the range of 298 to 333 K. The data displayed in Figure 5c exhibits that the Levafix Blue CA sorption capacity is increased with temperature increase. However, the reaction is further diminished with the further temperature upsurge up to 313 K. Such results might demonstrate the improvement of the Levafix Blue CA desorption stage in the adsorption process verifying the reaction is exothermic at a high-temperature degree [21]. Moreover, the temperature elevation deteriorates the sorptive forces between the ACF ribbon active sites and the Levafix molecules, and between adjacent Levafix Blue dye molecules on the sorbed phase. A comparable reaction trend was investigated and recorded by Parker et al. [23].

The impact of the initial Levafix Blue loading is exhibited in Figure 5d. Data represents the influence of the initial Levafix Blue dye concentration that is in the range of (25–100 mg/L) at 298 K on the adsorption capacity using both adsorbents cleaned and non-cleaned ACF ribbon. Generally, for all the applied materials, a noticeable enhancement in the adsorption uptake is attained with an increase in the dye concentration. This is due to the high Levafix dose resulting in an interaction between the Levafix molecules and ACF ribbon adsorbent material. The cleaned ACF ribbon showed higher adsorption capacities overall since the available pores are more than the used and non-cleaned ones.

This investigation recorded the enhancing adsorption uptake with the elevating adsorbate concentration which was previously recorded by Mittal and Gupta [24] when applying oiled soya ash for dye removal. Additionally, Geng et al. [25] reported similar results in using clay minerals in dye adsorption from an aqueous waste stream. Therefore, such data confirms the improvement in the mass transfer rate between the adsorbate and the surface of the adsorbent substance [25].

3.2.3. Acetylcellulose Regeneration

The recyclability of the acetylcellulose is investigated and the data is recorded in Figure 6. Initially, the ribbon was regenerated after use. The fiber is regenerated by subjecting it to hot water cleaning to remove any due saturation and attached to the acetylcellulose fiber. Then, the fiber is tested for 6 cycles of adsorption–desorption. As displayed in Figure 6, Levafix Blue dye removal efficiency onto the acetylcellulose fiber was maintained well since the removal capacity is still above 74% after six cycles of adsorption. This verifies that acetylcellulose is an ideal candidate for Levafix Blue dye water as a textile-simulated aqueous media discharge treatment application.

The adsorbent surface was examined after it was loaded with dye molecules as shown in Figure 7a. The loaded SEM image displays the Levafix Blue dye adsorption on the acetylcellulose ribbon. The depicted surface after adsorption (Figure 7a) is clearly different in shape and size compared to the unsaturated with dye ribbon and changed to an uneven and rougher surface (Figure 2b). This means the clean unused ribbon possesses a large number of active sites available and is responsible for facile Levafix Blue adsorption. After adsorption, (Figure 7a) Levafix Blue fills the surface of the adsorbent and becomes saturated, covered by the dye molecule. Also, as seen in the image, the blockage of pores was recognized which verifies the successful Levafix Blue adsorption process. This is due to the Levafix Blue dye molecules covering the surface of acetylcellulose fiber in a layered fashion, which displays a different morphology from the original acetylcellulose ribbon.

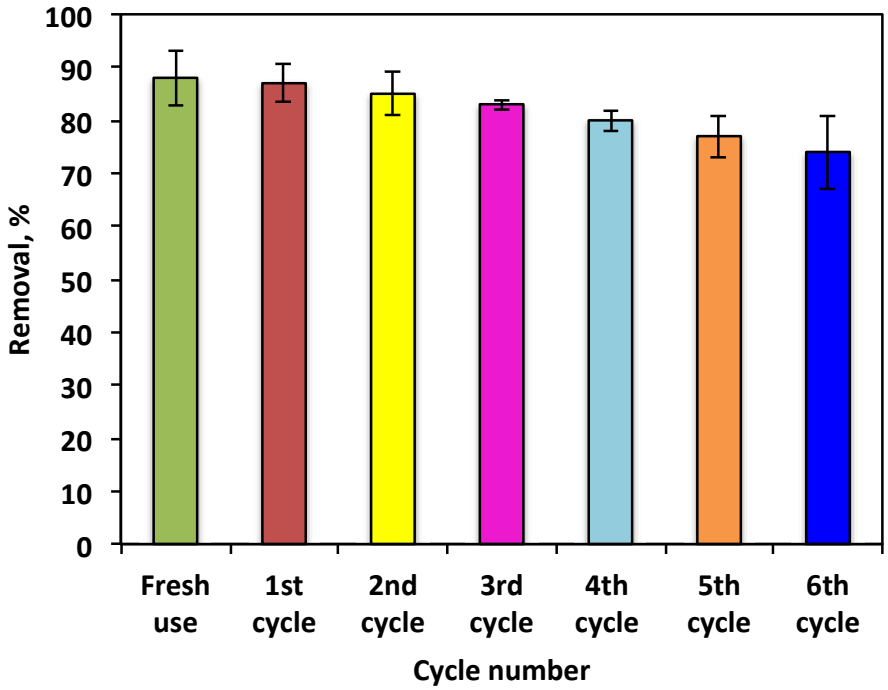

Figure 6. Regeneration and successive use of acetylcellulose ribbon.

Figure 7b illustrates the surface of the acetylcellulose ribbon after cyclic use and cleaning. The shape of the ribbon's fibrous structure was changed, and the surface was disturbed due to successive use and washing. Also, a smoother surface is attained compared to the first use which is saturated with dye. Coarse patches were still appearing, and some ribbon parts were still undiscerned. However, it is noteworthy to mention that even if the ribbon shape is changed acetylcellulose fiber is still able to dye sorption for successive cycles which valorizes the tendency of acetylcellulose derived from waste residuals to be a sustainable candidate for dual waste elimination.

This study is leading to useful results in the environmental engineering field. This treatment method not only eliminates the negative environmental externalities impact of cigarette residuals but also introduces an economic and costless method for wastewater treatment. Thus, cigarette residuals that are signified as a source of acetlycellulose are an appropriate industrial ecology adsorbent type.

To investigate and verify the importance of the current study compared to the literature, comparative data is achieved. A comparison of different types of Levafix dyes adsorption performance using the acetylcellulose ribbon derived from waste residuals in the current study with various adsorbents reported in the literature has been provided in Table 1. It is clear that acetylcellulose has a superlative ability to remove Levafix dye with a high adsorption capacity. Acetylcellulose could adsorb the dye as high as 87% when tested at room temperature as simulated textile wastewater. Thus, the bottom line is that acetylcellulose adsorbent substance acquires a better ability to eliminate textile residuals at simple technique tendency. Although the other studies also revealed a high removal efficiency and reached complete removal, the treatment is adopted from adsorbent material based on chemical uses [26–29]. It is important to mention that the current investigation of adsorbents is derived from waste residuals. Furthermore, previous studies have focused on adsorption-based treatments or the combination of adsorption and oxidation reactions, unlike the present study which indicates that the treatment is not solely reliant on adsorption [26–28]. This implies that such techniques may incur additional treatment costs, thus increasing the overall economic burden. In contrast, the utilization of acetylcellulose waste materials as affordable substances for eliminating textile pollutants from aqueous streams offers various advantageous aspects. These include their potential to reduce waste disposal costs and contribute to environmental safety.

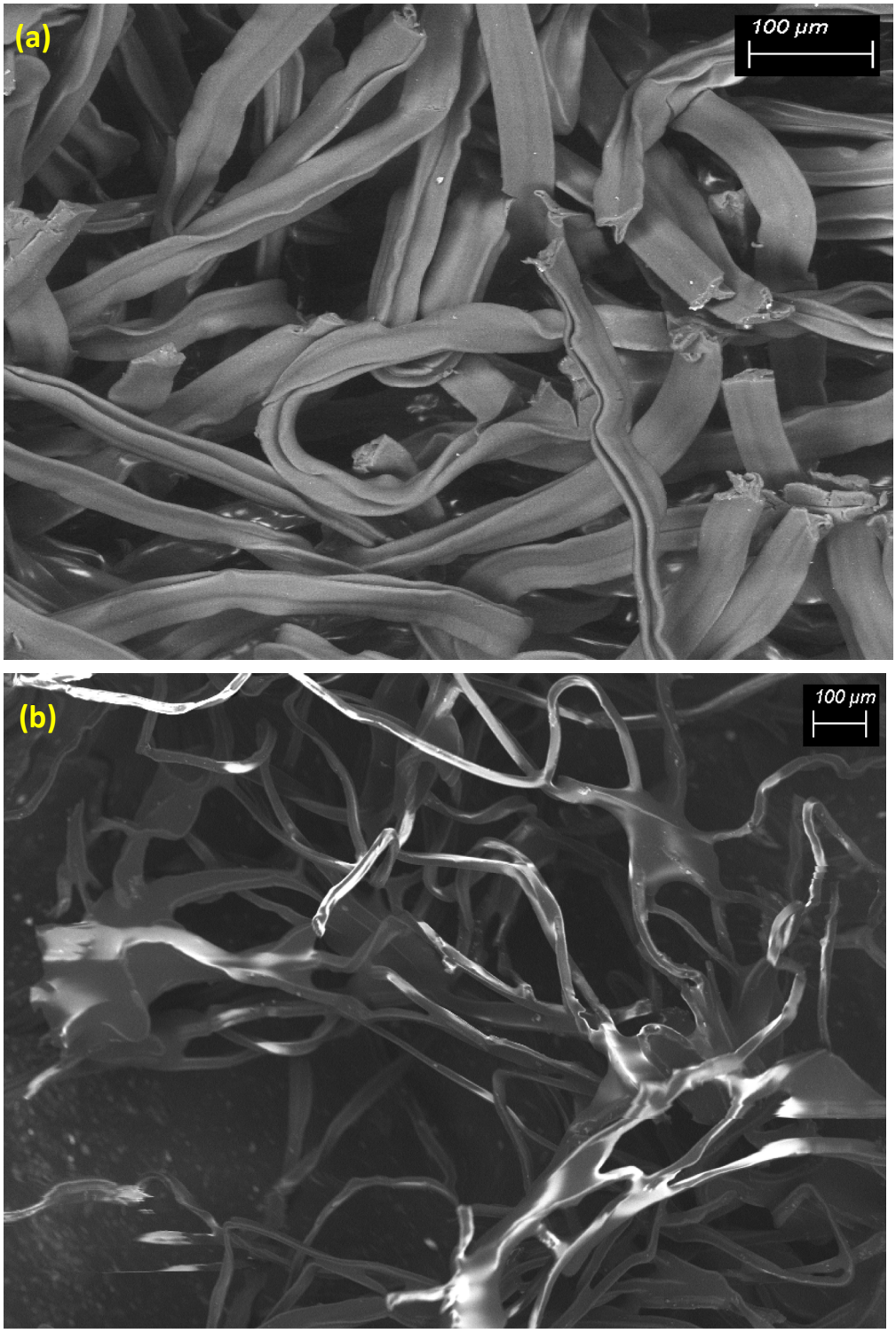

Figure 7. SEM image of acetylcellulose ribbon (**a**) before adsorption (**b**) after cyclic use of Levafix Blue dye adsorption and cleaning.

3.2.4. Adsorption Isotherm Moelds

To investigate the adsorption mechanism of Levafix Blue dye on acetylcellulose, the experimental results were examined to assess the performance of various isotherm models (Langmuir, Freundlich, and D-R), as depicted in Figure 8. The constant parameters of the linearized equations corresponding to these isotherm models were determined and presented in Table 2.

Table 1. Comparative adsorption capacities for Levafix dye by numerous adsorbents with the current investigation.

Adsorbent	Levafix Dye Name	Adsorption Efficiency	Isotherm Time	Operating Conditions	Ref.
Acetylcellulose fiber	Levafix Blue	87%	120 min	pH 3.0, T 298 K	Current work
Magnetite/sludge	Levafix Fast Red	81%	30 min	System augmented with UV	[26]
Magnetite/sludge	Levafix Amber	66%	30 min	System augmented with UV	[27]
Aluminum based sludge/Fe_3O_4	Levafix Blue	100%	30	System augmented with UV, pH 3.0, T 298 K	[28]
$MgCl_2$ particles	Levafix Brilliant Blue		150 min	~7.0, 476 mg/L	[14]
MgO nanoparticles	Levafix Fast Red CA	99%	45 min	6.0, 5 × 102	[29]
Fuller earth	Levafix Blue CA	99.9%	50 min	~3.0	[19]
Silica	Levafix Amber CA	68.6%	150 min	5.0, 10 mg/30 mL dye solution	
Magnetite nanoparticles	Levafix Blue CA	8%	110 min	pH 6.5, 40 mg/L magnetite	[18]
Dolomitic	Levafix Brilliant Red	99%	10 min	pH 10.0	[16]

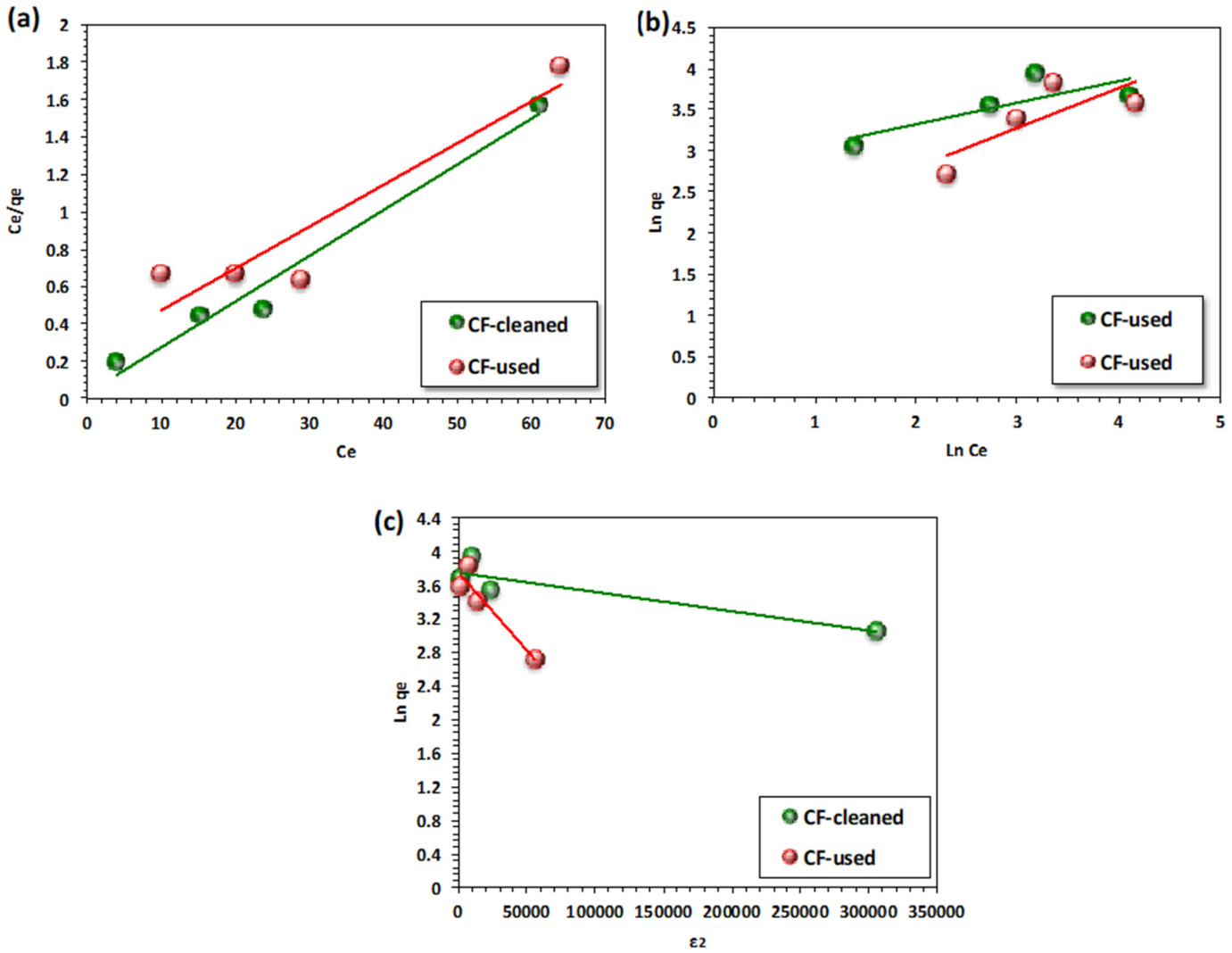

Figure 8. Linearized isotherms models for acetylcellulose adsorbent (**a**) Langmuir, (**b**) Freundlich, (**c**) D-R (Levafix Blue 25 mg/L; adsorbent dose 1 g/L; pH 3.0 and 298 K).

Table 2. Isotherm Parameters for Levafix Blue CA dye Adsorption on Acetylcellulose.

	Isotherm Parameters	ACF-Cleaned	ACF-Used
Langmuir	a_L (L/mg)	0.83	0.090
	K_L	34.01	4.034
	Qo (mg/g)	40.98	44.64
	R^2	0.97	0.91
Freundlich	K_F	16.43	6.22
	n	3.82	2.06
	R^2	0.63	0.61
D-R	qm (mol/g)	36.13	42.36
	K′ (mol^2/J^2)	2.0×10^{-6}	2.0×10^{-5}
	E (kJ/mol)	5.0×10^2	1.5×10^2
	R^2	0.82	0.88

The purposes of such fitting are to assess the maximum adsorption capacity and describe the equilibrium between Levafix Blue and acetylcellulose. This provides different binding sites and affinities in the adsorbent. The assessment of the acetylcellulose adsorption process was based on the comparison of the correlation coefficients, R^2 values, of those models (Table 2). The experimental data was assessed through their linearized form. The linearized form of the Langmuir model was applied in this study [30]:

$$\frac{C_e}{q_e} = \frac{1}{K_L} + \frac{a_L}{K_L} C_e \tag{2}$$

$$Q_o = \frac{K_L}{a_L} \tag{3}$$

where C_e is the Levafix Blue concentration at equilibrium (mg/L), q_e is the equilibrium adsorption capacity, and a_L and K_L are Langmuir adsorption constants. Q_o is the monolayer adsorption capacity of the solid adsorbent (mg/g).

The Freundlich model is appropriate for the highly heterogeneous surface and it is classified by the heterogeneity constant $1/n$. The linearized form of the model is according to Equation (4) [21]:

$$\ln(q_e) = lnK_F + \frac{1}{n} lnC_e \tag{4}$$

where K_F is the Freundlich constant that relates to the adsorption capacity (L/g). $1/n$ is the heterogeneity constant that measures the adsorption intensity and presents a significance of how the adsorption is favorable when n is superior to unity, characterizing a favorable sorption system.

Dubinin−Radushkevich, D-R isotherm is realistic to assess the nature of the adsorption system and its linearized form according to the following:

$$lnq_e = lnq_m - K_{DR}\varepsilon^2 \tag{5}$$

$$\varepsilon^2 = RTln\left(1 + \frac{1}{C_e}\right) \tag{6}$$

In Equation (7), qm represents the monolayer saturation capacity (L/g), and KDR denotes the D-R isotherm constant for adsorption energy. This constant provides insights into the mean free energy (E) of sorption per molecule of the sorbate. The calculated value of E indicates the nature of the adsorption process, whether it is a chemical or physical adsorption process.

$$E = \frac{1}{\sqrt{2K_{DR}}} \tag{7}$$

The D-R isotherm simulates that adsorption is limited to a monolayer and can be used to assess the adsorption energy [21,30].

According to the correlation coefficient values, R^2, Frendlich isotherm or D-R isotherms are not as adequate as the Langmuir isotherm model. The equilibrium data of acetylcellulose sorbent material demonstrated a strong correlation with the Langmuir isotherm model, as indicated in Table 2. This suggests that the adsorption process can be categorized as homogeneous surface adsorption. and the adsorbed Levafix molecules are sorbed onto the monolayer coverage till the acetylcellulose's active sites are saturated. Also, when the n value ranges from 1 < n < 10, the sorption is signified as a favorable one (Table 2, n is more than unity).

3.2.5. Kinetics of Levafix Blue Sorption Characteristics

For the objective of evaluating the magnitude and mechanism of Levafix Blue CA dye sorption on the acetylcellulose material, kinetic models were applied. Lagergren's pseudo-first order, pseudo-second order, and Bangham's equation were used to assess the sorption parameters for each kinetic model. Lagergren's pseudo-first order Equation (7) indicates that the sorption of the dye onto the acetylcellulose substance is a solo sorption dye molecule [21].

$$\log(q_e - q_t) = \frac{K_1}{2.303} t + \log(q_e) \quad (8)$$

The pseudo-second-order kinetic model simulates the sorption sites as proportional to the square number of vacant sites as displayed in (Equation (9)) [31].

$$\frac{\mathrm{t}}{\mathrm{q_e}} = \frac{1}{\mathrm{K_2 q_e^2}} + \frac{1}{q_e}\mathrm{t} \quad (9)$$

where K_2 expressed the adsorption pseudo-second-order rate constant.

Further, Banghams's model was applied to signify if the pore-diffusion is solely limiting the rate of the adsorption system according to the following equation:

$$\log\log\left(\frac{C_o}{C_o - q_t m}\right) = \log\left(\frac{K_B m}{2.303 V}\right) + \propto \log(t) \quad (10)$$

where K_B and α are Banghams's model constants and ($\alpha < 1$), m, and V are the mass and volume of acetylcellulose, respectively [32].

Comparing correlation coefficient values, R^2, for the three models (data computed in Table 3) signifies pseudo-second-order model is well-fitted in comparison to the models that possess the highest correlation coefficient ever. For both sorbent materials based on cleaned and used acetylcellulose, the excellent R^2 is 0.99. Furthermore, the calculated equilibrium capacity for the dye is closer to the results attained from the experimental data.

Table 3. List of parameters obtained from kinetic models for Levafix Blue CA dye Adsorption on Acetylcellulose.

kinetic Model Parameters	CF-Cleaned	CF-Used
Lagergren's Pseudo-first-order		
q_e (mg g^{-1}) × 10^{-2}	115.0	98.88
K_1 (min^{-1}) × 10^{-2}	6.44	11.05
R^2	0.99	0.99
Pseudo-second-order		
q_e (mg g^{-1})	27.39	16.72
K_2 (g mg^{-1} min^{-1}) × 10^{-2}	0.12	0.22
R^2	0.90	0.96
Bangham's		
α	0.655	0.704
K_B (g) × 10^{-4}	0.407	0.407
R^2	0.94	0.92

4. Conclusions

Acetylcellulose was extracted and prepared from augmented cigarette filter waste in this study. The fiber was extracted by treatment and washing to be available as adsorbent material. The material is examined and analyzed using SEM and FTIR before and after cleaning. The attained acetylcellulose fiber was estimated to be an efficient adsorbent, which revealed excellent dye removal efficacy. The adsorption affinity reached 4.8 mg/g at the optimized conditions of pH 6.0 and adsorbent dose 1 g through the isotherm time of 2 h. Isotherm modeling and kinetic data demonstrated the reaction follows 1st-order kinetics and the reaction follows Langmuir isotherm. Not only does such an investigation delivers a potential method for dye elimination, but also specifies a way to realize the resource utilization of waste acetylcellulose by-product from smoking residuals to satisfy the goal of the industrial ecology approach. Therefore, a complimentary effort is still essential in the future study to apply such technique in real polluted effluents.

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Nomenclature

ACF	acetylcellulose fiber
SEM	Scanning Electron Microscope
FTIR	Fourier-Transform infrared spectroscopy
Ce	Levafix Blue CA dye concentration at equilibrium (mg/L)
qe	equilibrium adsorption capacity
a_L	Langmuir adsorption constant
K_L	Langmuir adsorption constant
Q_o	monolayer adsorption capacity (mg/g)
K_F	Freundlich constant relate to the adsorption capacity (L/g)
$1/n$	heterogeneity constant of Freundlich model
q_m	monolayer saturation capacity (L/g)
KDR	D-R isotherm constant of adsorption energy
E	mean free energy
R^2	correlation coefficient values
K_B	Banghams's model constant
A	Banghams's model constants ($\alpha < 1$)
M	mass of acetylcellulose
V	volume of acetylcellulose

References

1. Guan, X.-H.; Chen, G.-H.; Shang, C. Re-use of water treatment works sludge to enhance particulate pollutant removal from sewage. *Water Res.* **2005**, *39*, 3433–3440. [CrossRef] [PubMed]
2. Al, M.F.; Mo'ayyad, S.; Ahmad, S.; Mohammad, A.-S. Impact of Fenton and ozone on oxidation of wastewater containing nitroaromatic compounds. *J. Environ. Sci.* **2008**, *20*, 675–682.
3. Abdou, K.A.; Mohammed, A.N.; Moselhy, W.; Farghali, A.-A. Assessment of modified rice husk and sawdust as bio-adsorbent for heavy metals removal using nano particles in fish farm. *Asian J. Anim. Vet. Adv.* **2018**, *13*, 180–188. [CrossRef]
4. Pourali, P.; Behzad, M.; Arfaeinia, H.; Ahmadfazeli, A.; Afshin, S.; Poureshgh, Y.; Rashtbari, Y. Removal of acid blue 113 from aqueous solutions using low-cost adsorbent: Adsorption isotherms, thermodynamics, kinetics and regeneration studies. *Sep. Sci. Technol.* **2021**, *56*, 3079–3091. [CrossRef]

5. Laib, S.; Rezzaz-Yazid, H.; Yatmaz, H.C.; Sadaoui, Z. Low cost effective heterogeneous photo-Fenton catalyst from drinking water treatment residuals for reactive blue 19 degradation: Preparation and characterization. *Water Environ. Res.* **2021**, *93*, 1097–1106. [CrossRef] [PubMed]
6. Glugoski, L.P.; Cubas, P.; Fujiwara, S.T. Reactive Black 5 dye degradation using filters of smuggled cigarette modified with Fe3+. *Environ. Sci. Pollut. Res.* **2017**, *24*, 6143–6150. [CrossRef]
7. Lei, W.; He, P.; Zhang, S.; Dong, F.; Ma, Y. One-step triple-phase interfacial synthesis of polyaniline-coated polypyrrole composite and its application as electrode materials for supercapacitors. *J. Power Sources* **2014**, *266*, 347–352. [CrossRef]
8. El-Desoky, H.S.; Ghoneim, M.M.; El-Sheikh, R.; Zidan, N.-M. Oxidation of Levafix, C.-A.reactive azo-dyes in industrial wastewater of textile dyeing by electro-generated Fenton's reagent. *J. Hazard. Mater.* **2010**, *175*, 858–865. [CrossRef] [PubMed]
9. Doyan, A.; Leong, C.L.; Bilad, M.R.; Kurnia, K.A.; Susilawati, S.; Prayogi, S.; Narkkun, T.; Faungnawakij, K. Cigarette Butt Waste as Material for Phase Inverted Membrane Fabrication Used for Oil/Water Emulsion Separation. *Polymers* **2021**, *13*, 1907. [CrossRef]
10. Yang, J.; Kubota, F.; Baba, Y.; Kamiya, N.; Goto, M. Application of cellulose acetate to the selective adsorption and recovery of Au(III). *Carbohydr. Polym.* **2014**, *111*, 768–774. [CrossRef]
11. Goveia, D.; Lobo, F.A.; Rodrigues, U.P.; Dias, N.L.; Fraceto, L.F.; Rosa, A.H. Preparation of the orgamomodified cellulose acetate membranes for adsorption of the ions Cu(II), Cd(II), Mn(II) AND Ni(II). *Quim. Nova* **2010**, *33*, 1135–1140. [CrossRef]
12. Magosso, H.A.; Fattori, N.; Arenas, L.T.; Gushikem, Y. New promising composite materials useful in the adsorption of Cu(II) in ethanol based on cellulose and cellulose acetate. *Cellulose* **2012**, *19*, 913–923. [CrossRef]
13. Ju, J.; Duan, Z.; Zhao, W.; Liu, Y.; Zhang, L.; Li, Y. Improvement of bilirubin adsorption capacity of cellulose acetate/polyethyleneimine membrane using sodium deoxycholate. *Biochem. Eng. J.* **2013**, *79*, 144–152. [CrossRef]
14. Bhalara, P.D.; Punetha, D.; Balasubramanian, K. Kinetic and isotherm analysis for selective thorium(IV) retrieval from aqueous environment using eco-friendly cellulose composite. *Int. J. Environ. Sci. Technol.* **2015**, *12*, 3095–3106. [CrossRef]
15. Wu, Y.B.; Bi, J.; Lou, T.; Song, T.B.; Yu, H.Q. Preparation of a novel PAN/cellulose acetate-Ag based activated carbon nanofiber and its adsorption performance for low-concentration SO_2. *Int. J. Miner. Metall. Mater.* **2015**, *22*, 437–445. [CrossRef]
16. Bouyakoub, A.Z.; Lartiges, B.S.; Ouhib, R.; Kacha, S.; El Samrani, A.G.; Ghanbaja, J.; Barres, O. $MnCl_2$ and $MgCl_2$ for the removal of reactive dye Levafix Brilliant Blue EBRA from synthetic textile wastewaters: An adsorption/aggregation mechanism. *J. Hazard. Mater.* **2011**, *187*, 264–273. [CrossRef]
17. Halkikar, A.; Gernhart, Z.C.; Cheung, C.L. Recyclable magnetite nanoparticle catalyst for one-pot conversion of cellobiose to 5-hydroxymethylfurfural in water. *J. Nanomater.* **2015**, *16*, 315.
18. Aygun, A.; Yilmaz, T.; Nas, B.; Berktay, A. Effect of temperature on fenton oxidation of young landfill leachate: Kinetic assessment and sludge properties. *Glob. Nest J.* **2012**, *14*, 487–495.
19. Węgrzyn, A.; Radko, M.; Majda, D.; Stawiński, W.; Skiba, M.; Cież, D. Interaction between adsorbed molecules and tailor made large chelating ligands grafted on SBA-15 studied by means of thermoporometry. *Microporous Mesoporous Mater.* **2018**, *268*, 31–38. [CrossRef]
20. Sudiarti, T.; Wahyuningrum, D.; Bundjali, B.; Arcana, I.M. Mechanical strength and ionic conductivity of polymer electrolyte membranes prepared from cellulose acetate-lithium perchlorate. *IOP Conf. Ser. Mater. Sci. Eng.* **2017**, *223*, 012052. [CrossRef]
21. Parker, H.L.; Budarin, V.L.; Clark, J.H.; Hunt, A.-J. Use of Starbon for the Adsorption and Desorption of Phenols. *ACS Sus. Chem. Eng.* **2013**, *1*, 1311–1318. [CrossRef]
22. Chu, W. Dye Removal from Textile Dye Wastewater Using Recycled Alum Sludge. *Water Res.* **2001**, *35*, 3147–3152. [CrossRef]
23. Parker, H.L.; Hunt, A.J.; Budarin, V.L.; Shuttleworth, P.S.; Miller, K.L.; Clark, J.-H. The importance of being porous: Polysaccharide-derived mesoporous materials for use in dye adsorption. *RSC Adv.* **2012**, *2*, 8992–8997. [CrossRef]
24. Mittal, A.; Gupta, V. Adsorptive removal and recovery of the azo dye Eriochrome Black T. *Toxicol. Environ. Chem.* **2010**, *92*, 1813–1823. [CrossRef]
25. Geng, Y.; Zhang, J.; Zhou, J.; Le, J. Study on adsorption of methylene blue by a novel composite material of TiO_2 and alum sludge. *RSC Adv.* **2018**, *8*, 32799–32807. [CrossRef] [PubMed]
26. Rahma, T.H.; Mai, K.F.; El Sherbiny, S.A.; Tony, M.A. Identifying optimized conditions for developing dewatered alum sludge-based photocatalyst to immobilize a wide range of dye contamination. *Appl. Water Sci.* **2022**, *12*, 210. [CrossRef]
27. Thabet, R.; Fouad, M.K.; El Sherbiny, S.A.; Tony, M.A. Zero-Waste Approach: Assessment of Aluminum-Based Waste as a Photocatalyst for Industrial Wastewater Treatment Ecology. *Int. J. Environ. Res.* **2022**, *16*, 36. [CrossRef]
28. Rahma, T.H.; Mai, K.F.; Shakinaz, A.; El Sherbiny, S.A.; Tony, M.A. A circular economy use of waste drinking water treatment plant sludge for magnetic photocatalyst composite production into wastewater treatment. *Int. J. Environ. Anal. Chem.* **2022**, 1–20. [CrossRef]
29. Georgiou, D.; Melidis, P.; Aivasidis, A.; Gimouhopoulos, K. Degradation of azo-reactive dyes by ultraviolet radiation in the presence of hydrogen peroxide. *Dye. Pigment.* **2002**, *52*, 69–78. [CrossRef]
30. Kim, D.-H.; Yang, J.-S.; Baek, K. Adsorption Characteristics of As(III) and As(V) on Alum Sludge from Water Purification Facilities. *Sep. Sci. Technol.* **2012**, *47*, 2211–2217.
31. Dawodu, M.O.; Akpomie, K.-G. Evaluating the potential of a Nigerian soil as an adsorbent for tartrazine dye: Isotherm, kinetic and thermodynamic studies. *Alex. Eng. J.* **2016**, *55*, 3211–3218. [CrossRef]

32. Srivastava, V.C.; Swamy, M.M.; Mall, I.D.; Prasad, B.; Mishra, I.-M. Adsorptive removal of phenol by bagasse fly ash and activated carbon: Equilibrium, kinetics and thermodynamics. *Colloids Surf. A* **2006**, *272*, 89–104. [CrossRef]
33. Abu-Danso, E.; Bagheri, A.; Bhatnagar, A. Facile functionalization of cellulose from discarded cigarette butts for the removal of diclofenac pharmaceutical from water. *Carbohydr. Polym.* **2019**, *219*, 46–55. [CrossRef] [PubMed]
34. Hasima, S.; Kamila, N. Cellulose acetate from palm oil bunch waste for forward osmosis membrane in desalination of brackish water. *Results Eng.* **2022**, *15*, 100611.
35. Huang, M.R.; Li, X.G. Thermal Degradation of Cellulose and Cellulose Esters. *J. Appl. Polym. Sci.* **1998**, *68*, 293–304. [CrossRef]
36. Puls, J.; Wilson, S.A.; Holter, D. Degradation of Cellulose Acetate-Based Materials: A Review. *J. Polym. Environ.* **2011**, *19*, 152–165. [CrossRef]