Research Article

Assessing the performance of modified waste cotton cloth (MWCC) installed in a biological contact reactor as a biofilm carrier used for domestic wastewater treatment

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Abstract

The current study examined the performance of modified waste cotton cloth (MWCC) in the removal of organic pollutants from domestic wastewater as an emerging technique for waste textile resource management and reuse. The MWCC biofilm carrier was self-made with discarded cotton cloths cut into smaller strips with 66 cm long and average weight of 2.4 g each. The surface of the non-modified waste cotton cloth (NM-WCC) was modified through the hydrosulphuric acid (H_2SO_4) method to enhance the physical characteristics of the material such as hydrophilicity and biofilm attachment. The best filling rate of the carrier used for the experiment was 65% as determined by preliminary studies. Two self-made biological contact biofilm reactors: R1 (installed with NM-WCC) and R2 (fixed with MWCC), were used for the experiment. The experimental results showed that MWCC had higher removal efficacy of chemical oxygen demand, ammonia–nitrogen (NH_4^+–N) and total phosphorus (TP) at 98.34%, 85.44% and 60.20%, respectively. The hydraulic retention time decreased from 21 to 8 h on the 4th day. The scanning electron microscope was used to analyse the surface characteristics of the NM-WCC and MWCC. The water-holding capacity of the biofilm carriers was determined through the static immersion method and ASTM D1117-80 guiding principles. The water contact angle was estimated through the static process by adapting Young's equation. The findings of this research could significantly contribute to the discovering of alternative innovative technological prospects of utilizing H_2SO_4 MWCC as an effective biofilm carrier in domestic wastewater treatment.

Keywords Modified waste cotton cloth · Waste management · Biofilm carrier · Domestic wastewater treatment · Biological contact biofilm reactor · Organic pollutants

1 Introduction

In recent studies, many researchers have shown increasing concerns about environmental pollution emanating from solid wastes [1–3]. To ensure sustainable development while maintaining safe environment, there is the need to explore more innovative methodologies which are effective, cheap and environmentally friendly for wastewater treatment [4]. Cotton materials have been used widely for various domestic, commercial and industrial activities due to their increasing availability and special characteristics including low price, gas permeability, softness, environmental sustainability, biodegradability and hydrophilicity [5, 6]. The hydrophilic characteristic of the cotton fabric makes it high affinitive to water and other liquids, thereby increasing the chances of biofilm growth and development [7, 9]. Also, globalization, fashion and international business have made it easier for an increased demand

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and supply of cotton cloths worldwide [5, 10]. This can be attributed to low cost, increased incomes, population concentration, increasing urbanization and improved standards of living [11, 12]. Consequently, waste cotton cloths have extensive sources for recycling and reuse [13]. According to America Environmental Protection Agency (EPA) report published in 2018 for 2015 municipal solid waste (MSW) statistics, about 16.03 million tons of textiles were discarded of which greater portion was cotton textiles [14, 15]. Hamawand et al. [3] reported that there were over 25 million tons of cotton cloths which were produced and utilized globally, resulting in discarding of large quantity of WCC concurrently. It has also been reliably observed that the disposal of cotton cloths indefinitely leaves some pollution footprints such as release of methane by decomposing cloths [16], which is a harmful greenhouse gas and a significant contributor to global warming [8, 17]. Again, decomposing WCC in landfills contributes to leachate which is a potential source of contaminants for both surface and underground water [18]. Furthermore, improper disposal of WCC creates the room to host disease causing agents such as pathogens and germs which pose health threats to people and animals. In addition, ammonia emission into the environment has attracted great concern in recent studies. Decomposing WCC in landfills or other unapproved sites also releases ammonia which is toxic to both terrestrial and aquatic ecosystems [19, 20]. These among others pose serious environmental and occupational hazards to humans and the ecology as a whole [2, 21, 22]. One innovative technique to mitigate this menace is to reuse or recycle waste cotton cloth for biological treatment of municipal wastewater while concurrently ensuring proper waste management and environmental protection [4, 23, 25].

The modern studies that emphasize the extensive utilization of waste cotton cloth concentrate on the extraction of some chemical compounds, such as cellulose nanocrystals [17] and extraction of nanocellulose through a novel one-pot isolation system [26]. Furthermore, a study on production of single and mixture adsorption was conducted by Boudrahem et al. [27] and bacterial cellulose production by Feng et al. [8]. Also, waste cotton cloth has been used for tensile and fatigue characterization in the area of polypropylene matrix for composites applications such as the industries dealing with automotive [28]. In the field of wastewater treatment, waste cotton cloth has been used for quick heavy metal removal from industrial effluent [29], to reclaim uranium in radioactive waste filtrate [30] and immobilization of waste cotton cloth into a bioreactor [31]. Also, modification and recycling of waste cotton cloth have been studied for its effective utilization. Vats and Rissanen [32] investigated the elements responsible for improving the processes of up-cycling of waste cotton textiles. Their study was under pinned by the fact that cotton textiles discarded into the environment cause health and environmental hazards. Therefore, converting these waste products by adding value to them through up-cycling is more economical, feasible and safe. Their experiment mainly focused on the factors that contributed and enhanced up-cycling of cotton waste material. The results of their research illustrated that cotton waste textiles are unevenly degraded. Hence, it is difficult to ascertain and predict a particular mode of their decomposition. Furthermore, Pransilp et al. [33] modified the surface characteristics of cotton textile through the pretreatment of gas plasma. They found out that pretreatment of cotton fabric by sulphur hexafluoride, oxygen and nitrogen enhanced the hydrophilicity, adsorption of ink by inkjet printers and colour duplication. In addition, the potency of WCC as a composite building material has been conducted by Algin and Turgut [12]. Their experimental outcomes indicated that waste cotton as a composite material for making building blocks is highly efficient and does not break easily. There was also more energy savings, lightweight, bright and smooth surface compared with the conventional cement/concrete building blocks. Finally, their findings concluded that the material could serve as substitute to concrete blocks, wooden board, sound blockers and ceiling boards among others.

Previous studies provided an insight of the current potentials of waste cotton cloth and how it can be applied in different ways to mitigate their harmful environmental impacts such as global warming. Nonetheless, there is relatively less available literature on the extensive application of WCC to prepare a biofilm carrier used for domestic wastewater treatment. The present study aims to present relevant data, demonstrating that modification of waste cotton cloth (WCC) through H_2SO_4 utilization increases the removal efficiency of some organic pollutants from domestic wastewater treated in a BCBR. This study engineered two main operating ideologies concurrently: solid waste management and wastewater treatment. As a form of solid waste recycle and reuse, WCC was modified and utilized to make biofilm carrier installed in a BCBR. On the other hand, it has been simultaneously used in biological treatment of wastewater by using the attached biofilms on the WCC to degrade organic pollutants in domestic wastewater. This resulted in the creation of favourable microbiological aquatic environment for biofilm development. This helped to achieve high removal efficiency of the parameters analysed such as TP, COD and NH_4^+–N at acceptable standards in both reactors (R1 and R2). In this perspective, the COD was analysed through the potassium dichromate method, the spectrophotometry was utilized to monitor NH_4^+–N concentration, and TP was quantitatively checked by the ammonium molybdate spectrophotometric technique.

In addition, other operating parameters such as DO, was checked with a DO probe (JPB-607) and pH was recorded with the pH probe (pHS-3B). Also, the surface characteristics of the WCCs were analysed with the scanning electron micrograph (SEM JSM-7001F). The water-holding capacity of the biofilm carriers was determined through the static immersion method and ASTM D1117-80 guiding principles. Also, water contact angle was estimated through the static process by adapting Young's equation.

The findings of this research could significantly contribute to the discovering of alternative innovative technological prospects of utilizing H_2SO_4 MWCC as a biofilm carrier in domestic wastewater treatment. The results of this study could also help to realize resource utilization of WCC and minimize the decomposing effects of it when discarded in landfills with their related environment hazards. Furthermore, the outcome of this research will provide a baseline data for future assessment of other alternative biofilm carriers for organic pollutant removal in wastewater. In addition, information from this study could serve as a guiding principle for wastewater treatment decision-making, policy formulation, development stakeholders, future research, government and non-governmental agencies.

2 Materials and methods

2.1 Reactor and operation

A schematic diagram of the self-made BCBR for the experimental setup is shown in Fig. 1. The reactor is a plastic container with 22 cm inner diameter, 42 cm total height and 32 cm working height, with a working volume of 12 L and cylindrical in shape. The reactor was packed with the rectangular strips of the WCC with 66 cm long and average weight of 2.4 g each. The filling ratios of the filler were 35%, 45% and 65% [34]. The WCC strip fillers were hanged in the biofilm reactor with the steadying bar on top of the reactor. Also, wastewater was pumped into the biofilm reactor from the feed tank by the use of water diffuser and an influent pump. A liquid flow meter was utilized to regulate the flow of influent into the bioreactor [31]. Also, oxygen was pumped into the biological reactor by air compressor, while its flow rate was accurately regulated by the air flow meter. Biofilm microbes in the wastewater were kept energetic through aeration [35]. In order to prevent the biomass washout, air diffuser was used in the biofilm reactor. Uniform aeration is helpful to protect the biofilm. The dissolved oxygen (DO) was monitored with a DO probe fixed at the top of the reactor [36]. ECO aquarium heater was used to control the temperature of the wastewater at 23.5 ± 2 °C. The treated wastewater then flowed into the sedimentation tank, while the supernatant solution flowed out of it from the top, but the suspended microbes were retained in the sedimentation tank. [37]. Lastly, the sludge settled and it was scooped from the bottom of the sedimentation tank.

2.2 Municipal sewage

Municipal wastewater was derived from the primary settling tank of a local sewage treatment plant in Zhenjiang, China, and it was used as influent for the experiment. The main characteristics of the municipal domestic wastewater are depicted by the parameters and their concentrations which include NH_4^+–N (14–31 mg/L), COD (155–500 mg/L), TP (1.2–5.6 mg/L), pH (6.6–8.2), TN (23–48 mg/L) and SS (54–110 mg/L) [36].

2.3 Analytical methods

The potassium dichromate method was used to calculate the COD [38, 39]. Also, spectrophotometry [40] was used to measure the concentration of NH_4^+–N by the use

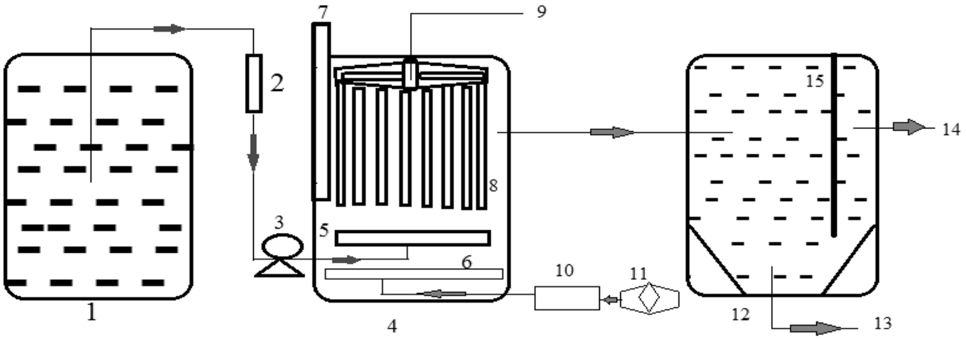

Fig. 1 Schematic diagram of the experimental setup showing the biological contact biofilm reactor (BCBR). (1) Feed tank, (2) liquid flow meter, (3) influent pump, (4) biofilm reactor, (5) water diffuser, (6) air diffuser, (7) ECO aquarium heater, (8) reticulated WCC filler, (9) DO probe, (10) air flow meter, (11) air compressor, (12) sedimentation tank, (13) sludge, (14) effluent, (15) clapboard *Source*: Adapted from Ru-Jin et al.[36]

of a multiparameter bench photometer (HI 83200, Hanna Instruments Inc., America). Ammonium molybdate spectrophotometric method was used to calculate the concentration of TP [23, 27, 41]. DO probe (JPB-607, Shanghai Jingsheng Scientific instrument Co. Ltd., China) was used to measure the level of DO with an online real time determination. A pH probe (pHS-3B, Shanghai Leici instrument Co. Ltd., China) was used to monitor the pH of effluent and influent in a systematic manner [36]. In addition, the scanning electron microscope (SEM, Field Emission Scanning Electron Microscope, JSM-7001F JEOL-Japan) was used to analyse the surface characteristics of the WCC. Also, the hydrophilicity of NM-WCC and MWCC biofilm carriers was estimated through the water absorption rate and water contact angle. The water-holding capacity of the NM-WCC and MWCC was determined through the static immersion method and ASTM D1117-80 guiding principles [7, 42, 43]. The process of water contact angle measurement used to characterize the surface hydrophilic properties of the WCCs was the static contact angle measurement method by the use of water contact angle meter, JY-82B through Young's equation [43].

2.4 Preparation of MWCC filler

2.4.1 Preparation of non-modified WCC filler

The waste cotton cloth used for the experiment contained 100% of cotton material obtained from a commercially available WCC market. Waste cotton cloth was measured and cut into thin strips with length 66 cm and 3.5 cm width with average weight of 2.4 ± 0.4 g each. Six pieces of the waste cotton strips were fixed on a thin plastic ring forming a bar. Three different sets of bars representing the three filling rates were used for the experiment. That is; 7 (35%), 9 (45%) and 13 (65%) bars of the WCC strips were installed in the bioreactor. Pebbles with average weight of 30.5 ± 10.1 g were tied at the bottom of each strip to ensure stable emersion in the bioreactor. The first reactor was installed with the non-modified waste cotton cloth (NM-WWC) and named R1, while the second reactor, R2, was fixed with modified waste cotton cloth (MWCC) to help compare and analyse the removal efficiencies of the two (2) reactors (Fig. 2).

2.4.2 Modification of WCC filler

After the NM-WCC filler was prepared, it was further modified to achieve the intended material. Sulphuric acid (H_2SO_4) [44] modification method was employed to transform the surface of the WCC strips. In the first step of the modification, 2600 ml of normal water was measure into a glass beaker with a total volume of 3000 ml. Afterwards, 20 ml of H_2SO_4 was added to the water and stirred continuously. Then, the WCC strips were fully submerged into the solution and allowed to soak for 1 h. Furthermore, the strips were removed from the solution after 60 min and allowed to dry at a room temperature. After drying, the materials were rinsed in normal water and allowed to dry again. At this moment, the surface of the WCC became modified [42, 45].

Fig. 2 Images of WCC strips used to make the biofilm carrier showing WCC strips (right), the biofilm carrier unit (middle) and those hanged in the reactor (left)

The optimum filling rate for a stable operationalization of the reactors was determined at 65% by preliminary experiment. To begin the experiment, the activated sludge was inoculated at room temperature into the bioreactors. After installation of the fillers into the activated sludge, the mechanical operations of the reactors were made to freely run for three (3) days consistently to culture the biofilms [46]. The sludge had a very good contact with the filler owing to the favourable biological and operational conditions. Afterwards, the sludge was fully cleared and then the biofilms were allowed to mature for another 3 days. At this stage, everything was set for the treatment of the municipal wastewater to begin [36]. The municipal sewage was consistently and intermittently fed into the biofilm reactors (R1 and R2). During the experiment, the hydraulic retention time (HRT), DO and COD among other structural parameters were measured [35, 47, 48].

2.5 SEM image

Figure 3 shows the scanning electron microscope (SEM, Field Emission Scanning Electron Microscope, JSM-7001F JEOL-Japan) images of the NM-WCC and MWCC which displayed the surface morphology of the materials. The threads within the cloth were partly closely packed, partly loosed and scattered. NM-WCC is represented by 'A and C' and MWCC is represented by 'B and D'. The porosity of the NM-WCC and MWCC was determined through the width of four major pore spaces depicted on each image (A, B, C and D). The ratios of measurement are as follows; AB (0.6 cm: 100 μm) and CD (1.5 cm: 100 μm). The average width of the

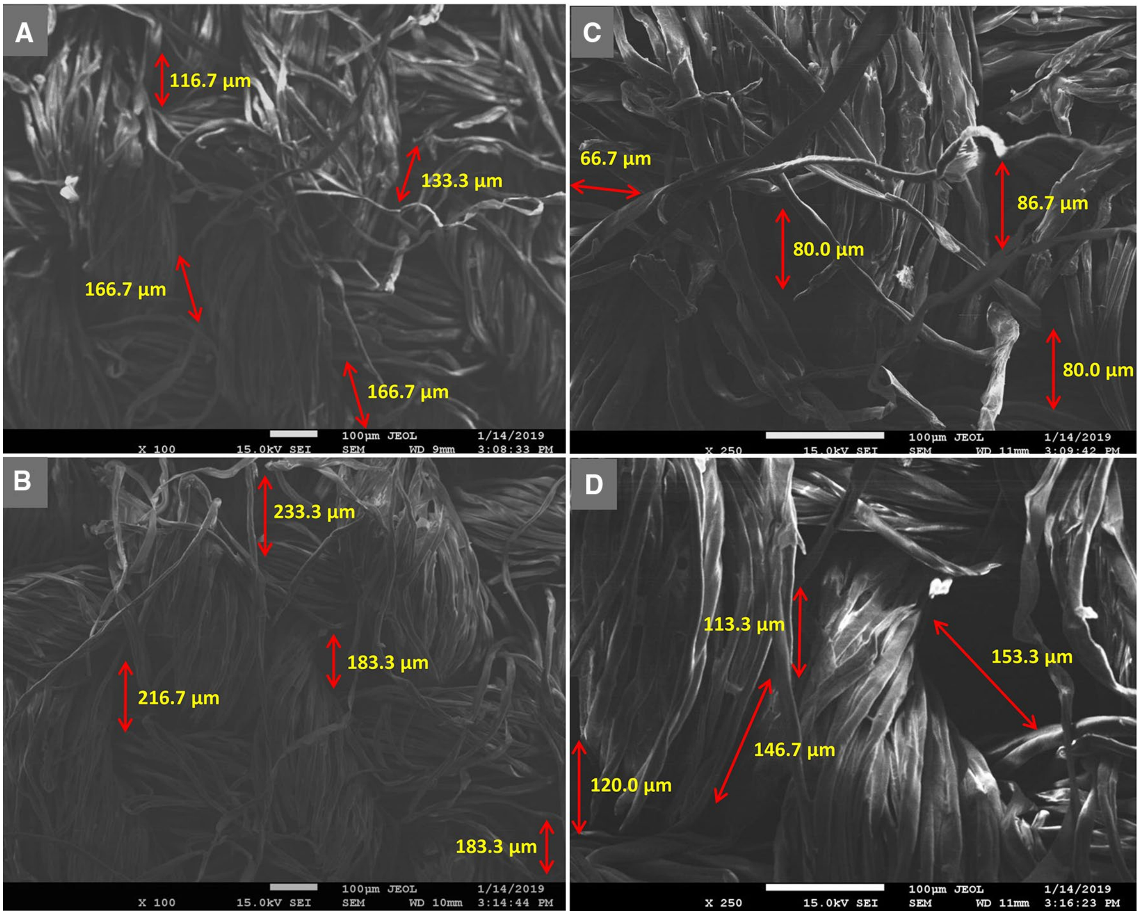

Fig. 3 Scanning electron micrograph (SEM) of NM-WCC (**A**, **C**) and MWCC (**B**, **D**)

pore spaces in A, B, C and D is 145.8 ± 25 µm, 204.2 ± 25 µm, 78.3 ± 8.4 µm and 133.3 ± 19.6 µm, respectively.

2.6 Water absorption rate (%)

The water-holding capacity of the NM-WCC and MWCC was determined through the static immersion method and ASTM D1117-80 guiding principles [7, 42, 43]. Dried WCC samples were weighed and submerged into distilled water of 20 °C at the depth of 5 cm, contained in a 500-ml glass beaker. The cloth samples were removed after 5 h of soaking and weighed again after 10 s of drainage. The samples were allowed to drain for extra 2 h and weighed once again to compare the water-holding capacities of NM-WCC and MWCC. The mass of the sample NM-WCC and MWCC was 2.39 g and 2.35 g, respectively. The water absorption rate was computed from the water absorbed between the two different periods which is given as the percentage of the original weight of the cotton cloth [42, 49].

$$\text{Water absorption rate }(\%) = \frac{(X_1 - X_0)}{X_0} \times 100$$

where X_0 is the WCC strip weight before immersion into water (g), X_1 WCC strip weight after immersion into water (g).

3 Results and discussion

3.1 SEM analysis

The SEM micrographs in Fig. 3 revealed the pore spaces within the WCCs. The porosity of the cloth created the necessary conditions for proper hydrophilicity and aeration which promoted micro-environmental conditions for biofilm growth and attachment [34]. The porosity of the NM-WCC and MWCC was determined through the width of four major pore spaces depicted on each image (A, B, C and D). The ratios of measurement are as follows: AB (0.6 cm: 100 µm) and CD (1.5 cm: 100 µm). The average width of the pore spaces in A, B, C and D is 145.8 ± 25 µm, 204.2 ± 25 µm, 78.3 ± 8.4 µm and 133.3 ± 19.6 µm, respectively. Given the average pore sizes of 'A' and 'B', with scale of 0.6 cm: 100 µm, B—which is the MWCC, had higher porosity of 204.2 ± 25 µm above 'A' which is the NM-WCC. Also, for 'C' and 'D' with the scale of 1.5 cm: 100 µm, the average porosity is 78.3 ± 8.4 µm and 133.3 ± 19.6 µm, respectively. This obviously shows that at different magnifications, MWCC had higher porosity over NM-WCC. This could be attributed to the surface modification of MWCC which expanded the cotton cloth threads, increased the surface roughness and porosity.

3.2 Hydrophilicity

One of the famous surface science terminologies is hydrophobicity which is generally the water-holding capacity of a material. Among the indices used to estimate hydrophilicity of solid surfaces are water absorption rate and water contact angle [45].

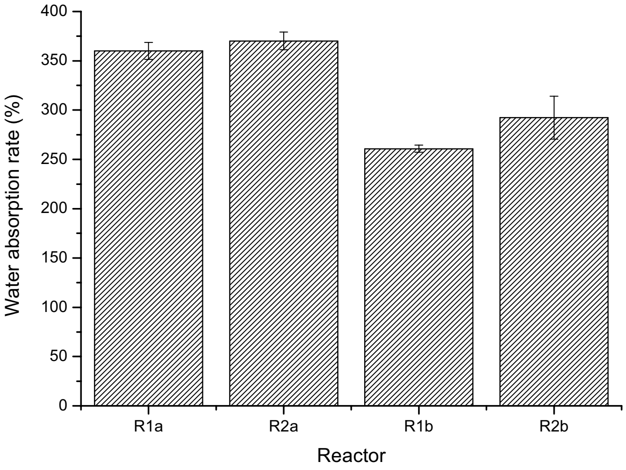

Fig. 4 Water absorption rate of NM-WCC and MWCC checked after 5 h of immersion (R1a, R2a) and 2 h of drainage (R1b, R2b) through the static immersion test method and ASTM D1117-80 guiding principles

3.2.1 Water absorption rate

The hydrophilicity of the NM-WCC and MWCC checked after 5 h of immersion into water (R1a, R2a) and 2 h of drainage (R1b, R2b) is shown in Fig. 4. The MWCC (R2a) after 5 h of soaking had the highest hydrophilicity determined at absorption rate of 370.07%. The water-holding capacity reduced from 370.07 to 292.30% (R2b) after 2 h of drainage. This may be because, the surface of the MWCC has been transformed to enhance its wettability. On the other hand, the rate of water absorption checked after 5 h of immersion for NM-WCC (R1a) was 295.05%. Its absorption rate also reduced to 258.83% (R1b) after 2 h of drainage. This indicates that the MWCC absorbs more water (hydrophilic) than the NM-WCC. This could probably be attributed to the surface modification of the MWCC which increased the porosity, roughness and water-holding capacity of the material.

3.2.2 Water contact angle

Figure 5 shows the water contact angles of NM-WCC and MWCC. The water contact angles of NM-WCC and MWCC are 139.6° and 120.51°, respectively. This implies that the modification of waste cotton cloth made the material surface more affinitive (hydrophilic) to water than the original WCC [7, 45]. In this study, the porosity of the MWCC was depicted in the SEM image. Also, the roughness of the MWCC surface was determined through texture and the surface image of the material. In Fig. 3, the porosity and roughness of MWCC increased obviously. So the improvement of hydrophilicity could be attributed to the increased surface roughness and porosity of MWCC which is essential for biofilm formation.

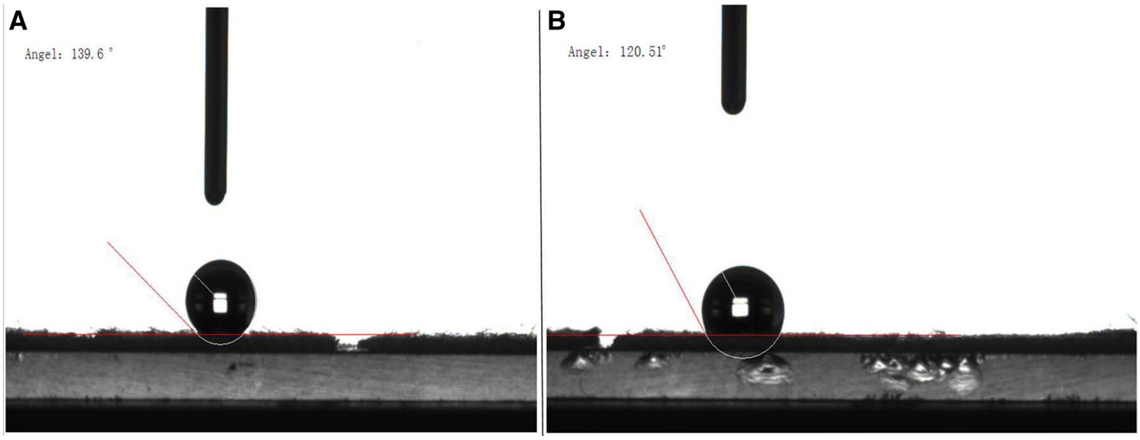

Fig. 5 Images of water contact angles of NM-WCC (**A**) and MWCC (**B**)

3.3 The biofilm formation

The growth characteristics of biofilms attached on the surface of NM-WCC and MWCC as biofilm carriers were determined during the maturation period. Theoretically, the biofilms are considered matured when they exhibit dark-yellowish-brownish colour on the surface of the filler, while simultaneously, COD and NH_4^+–N removal efficiencies reach more than 80%. The growth characteristics of biofilm on NM-WCC and MWCC biofilm carriers are shown in Table 1, which indicated that the biofilms in the reactors matured on the 13th and 11th days, respectively. At that time, the thickness of matured biofilm was 1.8 ± 0.2 mm and 2.5 ± 0.3 mm in R1 and R2, respectively. The relative density of matured biofilm of R1 and R2 was 47.33 ± 1.2 mg/cm^3 and 54.67 ± 1.8 mg/cm^3 accordingly. The maturation of the biofilms was enhanced throughout the stable operation period till the end of the experiment. R2 exhibited higher, density of biofilms growth and attachment on the MWCC. This could be attributed to the improved surface of MWCC.

3.4 Performance of biofilm reactor

At the period when the biofilms were considered matured, the two (2) biological contact reactors were stably kept operating for 30 days with relatively steady conditions of the basic parameters which are DO (4.6 ± 1.2 mg/L), pH (7.8 ± 0.3), temperature (23.5 ± 2 °C) and HRT (8 h). Figures 6 and 7, respectively, show the DO and temperature ranges of the two biofilm reactors (R1 and R2).

Afterwards, the potency of pollutant removal by the biofilm reactors installed with NM-WCC and MWCC fillers was measured, NH_4^+–N, TP and COD as the three basic operation indices for monitoring the water quality were also studied. The three parameters were observed within the intervals of every 2 days, while the biofilm reactors run steadily.

Table 1 The physical growth characteristics of the biofilms

Reactor code Reactor name	R1 NM-WCC	R2 MWCC
Biofilm maturation time (days)	13	11
Thickness of matured biofilm (mm)	1.8 ± 0.2	2.5 ± 0.3
Density of matured biofilm (mg/cm^3)	47.33 ± 1.2	54.67 ± 1.8

R1 reactor 1, *R2* reactor 2, *NM-WCC* non-modified waste cotton cloth, *MWCC* modified waste cotton cloth

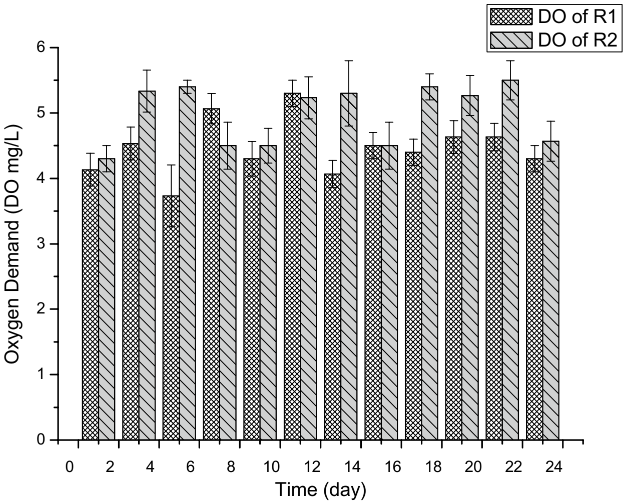

Fig. 6 Dissolved oxygen demand (DO) level of R1 and R2

3.4.1 COD removal

The monitored removal efficiency of COD varied with time during the operational period of the biofilm reactors as shown in Fig. 8. When the treatment time increased from 1 to 8 days, the removal efficiency of COD in R1 rapidly increased from 60.1 to 88.2% while R2 also rapidly increased from 63.3 to 95.8%. Moreover, when the treatment time further increased from 8 to 30 days, the removal efficiency of COD in R1 slowly increased and fluctuated a little from 88.2 to 93.4%, while R2 removal rate similarly increased slowly and fluctuated a little from 95.8 98.1%. This may be due to the fact that when biofilm microbe's growth period was below 10 days, the biofilm thickness and density were low which could be due to low reproduction rate. The microbial cells within this period are not matured enough to absorb, assimilate and degrade

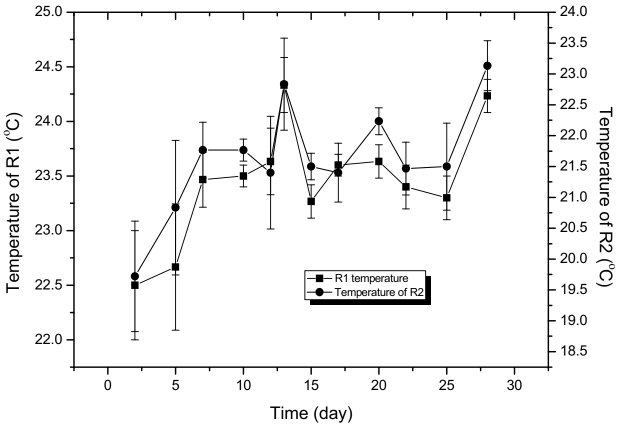

Fig. 7 Temperature range of R1 and R2

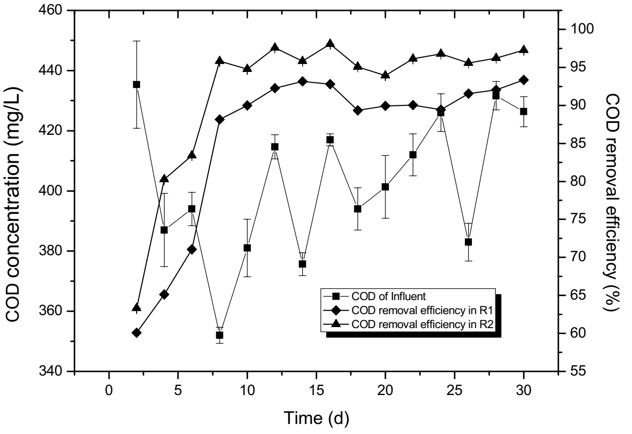

Fig. 8 COD removal efficiency of biofilm reactors installed with NM-WCC and MWCC

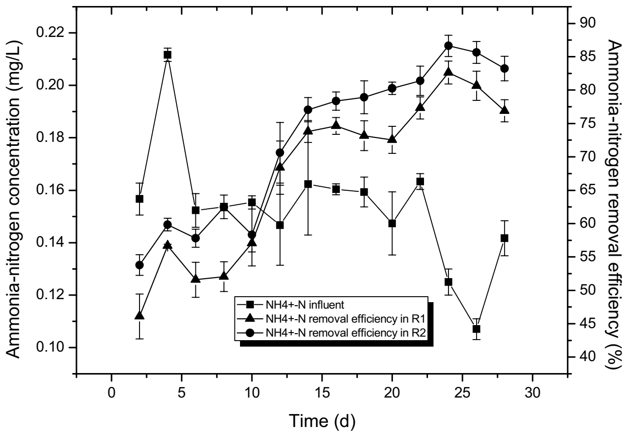

Fig. 9 Ammonia–nitrogen (NH_4^+–N) removal efficiency of R1 and R2

more organic pollutants which will lead to relatively low removal efficiency of COD. However, as the treatment time increases, the biofilms get enough time for their bioactivities which gives them the opportunity to ingest and decompose more organic pollutant leading to high removal efficiency of COD. During the stable operational period, the average removal efficacy of COD in R1, average COD of effluent concentration and average COD of influent were 85.9%, 56.8 mg/L and 402.1 mg/L, respectively, which are in concordance with the result of biofilm formation test. On the other hand, the average removal efficacy of COD and average COD of effluent concentration in R2 were 92% and 32.53 mg/L, respectively. The difference of the average COD removal efficiency between R2 and R1 was 6.1%. This could be attributed to the fact that MWCC enhanced more biofilm formation and pollutant degradation.

3.4.2 NH_4^+–N removal

Figure 9 illustrates the removal efficiency of NH_4^+–N with time variations in the biological contact biofilm reactor. As it can be seen from the above figure, when the treatment time increased from 1 to 12 days, the removal efficiency of NH_4^+–N in R1 quickly increased from 46.08 to 68.4%, while R2's NH_4^+–N removal rate also increased rapidly from 53.8 to 70.6%. Additionally, when the treatment time further increased from 12 to 30 days, the removal efficiency of NH_4^+–N in R1 slowly increased and fluctuated between 68.4 and 82.0%. Meanwhile, the R2 removal efficiency of NH_4^+–N similarly increased slowly and fluctuated a little between 70.6 and 86.6%. This may be as a result of the immature cells of the autotrophic nitrifying bacteria (AOB) and nitrite-oxidizing bacteria (NOB) within this stage; therefore, the ability to degrade more NH_4^+–N in the domestic wastewater is inhibited causing relatively low removal efficiency of ammonia–nitrogen. Hence, as the wastewater treatment time increases from 12 to 30 days, the nitrification bacteria become more matured and energetic. This then enchases more breakdown of ammonia–nitrogen by the nitrification bacteria leading high removal efficiency

The average efficiency of NH_4^+–N removal for R2 installed with MWCC was 75.74% with corresponding 0.043 mg/L of average effluent concentration and average influent concentration of 0.159 mg/L. This exhibited effective performance of R2 over R1. This can be deduced from the individual removal capacities of the two reactors. During the stable operational period, the best removal efficiency of R2 was 86.63%, whereas maximum NH_4^+–N removal efficiency of R1 was determined at 82.63% with difference of 4% between the two reactors. This could be due to the enhanced surface of MWCC which created more favourable conditions for biofilm growth and attachment. NH_4^+–N was primarily removed by two groups of autotrophic nitrifying bacteria (AOB) and oxidation of nitrite to nitrate by nitrite-oxidizing bacteria (NOB). At first, AOB oxidizes ammonia to nitrite, and then NOB oxidizes nitrite to nitrate (Watson et al. 1981) according to the following equations:

$$NH_3 + O_2 \rightarrow NO_2^- + 3H^+ + 2e^-$$

$$NO_2^- + H_2O \rightarrow NO_3^- + 2H^+ + 2e^-$$

There was also a positive relationship between R1 and R2 in terms of changes in the pollutant removal rates. On the 12th day of the experiment when both reactors begun stabilizing, removal efficiencies in R2 and R1 were 70.33 and 62.07%, respectively. Moreover, when the experiment progressed successfully, removal efficiency of R2 increased to 86.63% and R1 also increased to 82.63% before the end of the experiment. This indicated that increasing days gave more periods for biofilm maturation and growth which improved the removal efficiencies in both reactors. NH_4^+–N influent concentration also decreased with time in both reactors. On the 2nd day, with 0.157 mg/L of NH_4^+–N influent concentration in both reactors, R2 effluent was 0.723 mg/L and R1 effluent was also recorded at 0.843 mg/L. However, on the 26th day, the NH_4^+–N influent concentration in both reactors decreased to the 0.107 mg/L with decreased R2 effluent concentration to 0.015 mg/L, while R1 effluent also decreased to 0.021 mg/L. This could be attributed to the active reactions of the nitrifying bacteria.

3.4.3 Total phosphorus (TP) removal

Figure 10 shows the TP removal efficiency in the biofilm reactors which varied with treatment time (day). When the treatment time increased from 1 to 14 days, the removal efficiency of TP in R1 quickly increased from 26.5 to 56.8%, while TP removal rate in R2 also increased rapidly from 36.5 to 57.2%. Furthermore, when the treatment time increased from 14 to 30 days, the removal efficiency of TP in R1 fluctuated between 47.8 and 56.8%. Meanwhile, the R2 removal efficiency of TP increased slowly and fluctuated a little between 57.2 and 61.9%. This phenomenon may have occurred because when treatment time is below 14 days, the attachment, growth and reproduction of biofilms will be under development stage; this limits the ability of polyphosphate bacteria to absorb and break down TP at a high rate. Moreover, as the polyphosphate bacterial stabilizes and matures, the TP removal efficiency also improves and maintain at relatively high rate. Nonetheless, the TP removal efficiency is comparatively lower than the other pollutants checked (COD and NH_4^+–N). This could be because aside biofilm formation time, other factors such as HRT, DO, pH and temperature also influence the removal rate of TP.

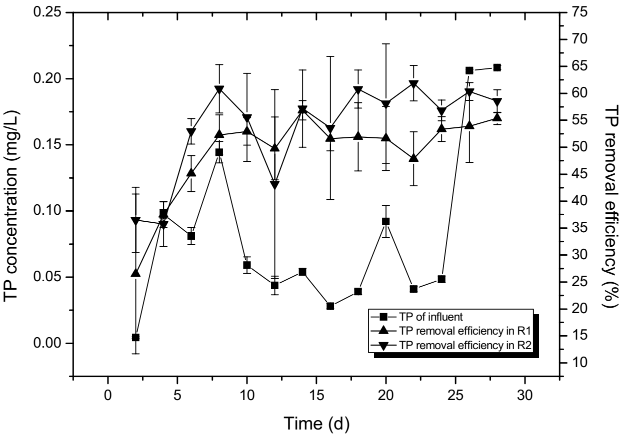

Fig. 10 TP removal efficiency

The average removal efficiency of TP in R2 and R1 was 53.72% and 49.04% with corresponding average effluents of 0.036 mg/L and 0.040 mg/L, respectively. The total removal efficiency of TP in R2 exhibited higher efficacy than R1. TP removal efficiency in R2 reached 60.79%, while R1 was lower at 55.36%. This could be as a result of the improved surface of the MWCC which improved biofilm growth and metabolism of TP. It was observed that during the stable operational period of the biofilm reactor, the influent concentration of TP in both reactors was 0.039 mg/L and below. Thus, it was found that the removal efficiency of TP was best at lower concentrations of organic pollutants. The phosphorus removal in the system was mainly attributed to the discharge of phosphate by polyphosphate bacteria in an anaerobic system with too much phosphorus intake in aerobic space while discharging phosphorus-rich sludge.

3.5 Modern recycling technologies of utilizing WCC

Most frequently used methods of WCC utilization and its characteristics as well as disadvantages are shown in Table 2.

WCC is abundant in many countries as the fashion industry keeps developing, while the demand and consumption of textile products keep rising. The abundance of WCC makes its price very low for easy affordability. In addition, the process of designing and manufacturing the WCC biofilm carrier is simple and easy. During the experimental period, MWCC proved feasible and effective for municipal sewage organic pollutant degradation in the biofilm reactor as it created an adaptive microenvironment for biofilm growth and attachment. Hence, the modern technique for optimum utilization of WCC lays in its usage as biofilm carrier for municipal sewage treatment. The future prospect of utilizing MWCC as a biofilm carrier for treating domestic sewage has tremendous advantages over the conventional and other recent methods of recycling the material [31]. This can be linked to the fact that, large volume of contaminated water is released from domestic, industrial, commercial and other sources which are released directly into the environment causing several health and ecological threats [50, 51]. Hence, given the limited quantity of potable water supply for all developmental purposes, wastewater treatment is vital for all countries gearing towards sustainable development. In addition to the abundant availability of the WCC in almost all countries with comparatively cheaper price to other recyclables, the intensive application of this technology could help overcome the secondary pollution

Table 2 Common recycling and reuse methods of WCC

Notable frequently utilization methods of WCC	Characteristics	Disadvantages
Extraction of some chemical compounds	Complex process	Raises environmental concern
Extraction of nanocellulose materials	Involves technicalities	Synthetic chemical material generally takes longer time to decay in the environment
Production of adsorptions	Complex process	Involves chemical use
Bacterial cellulose production	Complex process	Involves chemical use
Tensile and fatigue characterization in the area of polypropylene matrix for composites applications such the industries dealing with automotive	Complex process	High cost
Heavy metal removal	Complex process	High cost
Composite building and construction material	Involves simple process	low added value
Energy and antibacterial characterization	Complex process	High cost
Burning	Air pollution	Causes greenhouse gases emissions
Storage and conservation of microalgae	Simple process	Need much experience in the study area
Geotextile application	Low added value	Certain standards may limit WCC use in geotextiles

caused by other methods of utilizing the material [13, 22, 52]. Several researchers have identified the negative impacts of chemically related methods associated with some forms of recycling such as the production of adsorption, and extraction of nanocellulose constituents among others which could generate other environmental issues as well [1, 24]. Therefore, the modification of WCC exhibits more advantages as the process of production does not involve intensive chemical extraction or application, making it more environmentally friendly [5]. In addition, this process of utilization and reuse of waste material as a resource to mitigate water pollution menace is a sustainable way towards green technology and cost-effective for both poor and developed countries. Municipal wastewater treatment plants could easily apply this green technology without high cost or extensive technological know-how for better effluent [13, 53]. Unlike other methods of recycling WCC with many technicalities, the MWCC biofilm carrier can easily be designed and manufactured without hassles. This will make wastewater treatment very easy and ensure large scale application by both technical and non-technical environmental scientists. Furthermore, the results of the experiment conformed with that of previous studies [35, 36, 50]. The optimum removal efficiencies of COD, NH_4^+–N and TP in both reactors ranged between 90.33–98.34%, 80.58–85.44% and 56.26–60.20%, respectively, which are consistent with general acceptable water quality standards of treated domestic wastewater [19, 48, 54]. In biological treatment of wastewater, the attachment, dispersion, growth and organic pollutant decomposition by the microbes on the biofilm carrier are essential points of consideration. The study results confirmed the suitability of MWCC in enhancing more favourable microenvironment for biofilm formation and maturation than the original NM-WCC. This is similar to the research conducted by [29, 31] in using WCC to treat wastewater. Figure 3 and Table 1 illustrate the surface morphology of the WCCs and the growth characteristics of the biofilms on the materials, respectively. The biofilm grew and matured faster in R2 as a result of the surface electrophilic change due to the modification as it was proven by the removal efficiency of the organic pollutants, biofilm formation analysis and hydrophilia nature of the WCC which was also depicted on the SEM image. In addition, the technique of using MWCC biofilm carrier in a BCBR and comparing the result with the NM-WCC for domestic wastewater treatment and solid waste management is novel to this experiment. This technique is cheap, feasible, environmentally friendly and involves simple production process making it high prospective green technology for WCC recycle and utilization, especially in developing countries.

4 Durability analysis

At the end of the experiment, NM-WCC and MWCC were physically assessed to check the durability of each. The MWCC exhibited higher strength than NM-WCC. Table 3 and Fig. 11 show the physical characteristics and images of the two (2) biofilm carriers after the experiment, respectively.

MWCC exhibited higher mechanical strength, degradation and stress resistance than NM-MWCC.

Table 4 summarizes the characteristics and pollutant removal efficiencies of different modified and non-modified biofilm carriers used for wastewater treatment. The results indicate that MWCC has higher organic pollutant (COD, TP and NH_4^+–N) removal efficacy than other alternative biofilm carriers. Also, magnetism plays a significant role in enhancing the surface modification of some

Table 3 Durability analysis of NM-WCC and MWCC as biofilm carriers

	Characteristic	NM-WCC	MWCC
1	Weakness	Very weak at the end of the experiment	Still strong till the end of the experiment
2	Colour	Exhibited yellowish colour	Exhibited creamy colour
3	Stress perseverance	Many pebbles tightened at the bottom of the strips fell off due to the weakness of the material	Less pebbles tightened at the bottom of the strips fell off due its resilience
4	Resilience	Easily get torn when stretched	Highly vigorous when stretched
5	Durability	Less durable	More durable

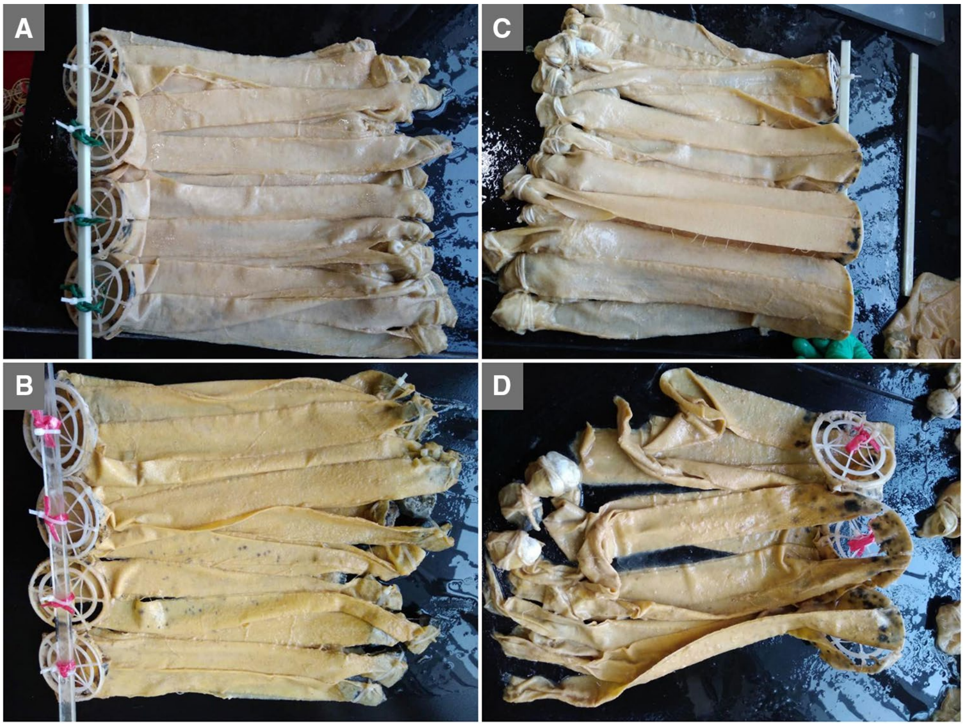

Fig. 11 Images of NM-WW (**b**, **d**) and MWCC (**a**, **c**) after the experiment

other biofilm carriers as reported by [58]. Recently, there has been increasing research emphasis on the influence of magnetism on bacterial activities as a result of corresponding changes in permeability of biofilm cell membrane which results in organic pollutant decomposition and promotion of microbial growth, reduced hydraulic retention time (HRT) and prevention of biomass washout [38, 53, 58]. As reported by Yanli et al. [59], homoeostasis of a cell membrane depends on how the fluidity, membrane integrity and permeability are regulated. Also, the improvement in the cellular properties increases the cell growth and metabolism of biofilm microbes. The utilization of magnetism can change the charge distribution on the surface of the cell membrane and influence the electrical characteristics of cell membrane channel. Under the action of magnetism, the permeability of biofilm cell membrane is likely to be improved [47, 57, 58]. Application of magnetic field is expected to enhance the performance of the biofilm as it was reported to enhance the microbial growth and physiology which results in higher wastewater treatment efficiency [60, 61]. The biofilms used as the consortium method to degrade pollutants are based on the ideology of microorganism in the wastewater using the organic or other dissolved pollutants as sources of their feed for growth [58]. The biofilm maturation/formation period of various carriers also differs with time (d). The MWCC achieved faster maturation period of 11 days which is higher than magnetic and non-magnetic PET [46] and plastic non-magnetic filler [36]. This could be attributed to the increased porosity, roughness and hydrophilicity which created very good microenvironment for biofilm attachment and growth.

In general, materials used as biofilm carriers are either organic or inorganic in nature.

Organic biofilm carrier is known to have large surf ace area; however, they are challenged with poor biofilm

Table 4 The analysis of characteristics and pollutant removal efficiencies of modified and non-modified biofilm carriers used for wastewater treatment. *Source*: adapted from [58]

No.	Type of biofilm carrier/filler	Modification substance	Biofilm characteristics and their measure		Main pollutant	Pollutant removal rate (%)	Reference(s)
1	MWCC	H_2SO_4	Biofilm-forming time(d) Thickness of mature biofilm (mm) Biofilm density (mg/cm^3)	11 2.5 ± 0.3 54.67 ± 1.8	COD NH_4^+–N TP	98.08 86.63 60.79	This study
2	NM-WCC	No	Biofilm-forming time(d) Thickness of mature biofilm (mm) Biofilm density (mg/cm^3)	13 1.8 ± 0.2 47.33 ± 1.2	COD NH_4^+–N TP	93.35 82.63 55.36	This study
3	Plastic magnetic filler	Air plasma modification	Biofilm-forming time(d) Thickness of mature biofilm (mm) Biofilm density (mg/cm^3)	10 2.1 41.32	COD NH_4^+–N	92.4 70.2,	[36]
4	Plastic non-magnetic filler	No	Biofilm-forming time(d) Thickness of mature biofilm (mm) Biofilm density (mg/cm^3)	14 1.4 32.59	COD NH_4^+–N	83.2 70.2,	
5	Non-magnetic PET	No	Attached biomass Time required for biofilm formation	10.8 g/L BV 25 days	NH_4^+–N and NO_2–N	N/R	[46]
6	Magnetic ceramsite	Fe_3O_4	Biomass (g) MLSS (g/L) SV (%) Zoogloea	35.34 28.36 64 Copious and Large	COD NH_4^+–N	10–20 20–30	[37]
7	Magnetic ceramsite	Fe_3O_4	Biomass (g) MLSS (g/L) SV (%)	33.86 28.72 60	Cr(VI)	5–10	[55]
8	Modified poly(ethylene terephthalate) fibre cloth (AQS-PETFC)	Anthraquinone-2-sulphonic acid (AQS)	N/R	N/R	Nitrobenzene	60	[56]
9	Non-modified HDPE	No	Biomass (g)	Y: 0.457 X: 2030	COD NH_4^+–N TN	93 51 49	[47]
10	Modified HDPE	PQAS-10	Biomass (g)	Y: 0.747 X: 2350	COD NH_4^+–N TN	93 92 72	
11	Non-modified PE	No	Biomass	TSS on carriers: 179 g/m2	COD NH_4^+–N TN	81.3 92.9 77.6	[57]
12	Modified PE	PQAS-10 and Fe2O3	Biomass	TSS on carriers: 192 g/m2	COD NH_4^+–N TN	83.8 93.3 80.2	

BR bioreactor, *N/R* not reported, *MAS* magnetic air stone, *AS* air stone, *COD* chemical oxygen demand, *NH_4^+–N* ammonia–nitrogen, *TP* total phosphorus, *SV* sludge volume, *PET* polyethylene terephthalate, *Y* biomass yield (mgVSS/mgCOD), *X* attached biomass (mg/L), *PQAS-10* polyquaternium-10; *CPAM* cationic polyacrylamides, *HDPE* high-density polyethylene, *NM-WCC* Non-modified waste cotton cloth, *MWCC* modified waste cotton cloth attachment (bio-affinity) due to their relative smooth surfaces [57]. Also, inorganic materials-based biofilm carriers possess high mechanical strength but limited with mass transfer as a result of poor porosity. For practical reasons, the modification of biofilm carrier surface whether electrophilic or hydrophilic is very vital for bacteria growth and attachment. Biofilm carriers modified to improve surface characteristics such as hydrophilicity and water contact angles have proven to exhibit high biofilm growth rate, attachment and pollutant removal efficiency [34, 45, 57].

Furthermore, aside hydrophilicity, another factor which affects formation, growth and attachments of biofilm to a carrier surface is electrophilicity [58]. As a result of phosphoric acid and carboxylic acid groups in the cell membrane of microorganisms, they have negative charges [30]. Again, as already indicated, some materials used to make biofilm carrier such as HDPE, PET and PQAS-10 have negatively charged surface which make them have similar charge with microbes. Therefore, the repulsion will affect biofilm growth and attachment on the surface of the carrier [47]. In order to mitigate this menace, the surface electronegativity of biofilm carrier is modified or altered to produce positively charged surface [26, 47, 57]. Further studies have also shown that modified biofilm carrier surfaces which are positively charged have lower contact angle than the unmodified biofilm carriers [47, 57].

5 Conclusions

The current study explored the feasibility and effectiveness of WCC as a biofilm carrier in biological treatment of domestic wastewater. The MWCC biofilm carrier was prepared by modifying the original WCC through the hydro-sulphuric acid (H_2SO_4) method to enhance the physical characteristics of the material such as hydrophilicity, roughness and biofilm attachment which created a good microenvironment and enhanced the degradation of organic pollutants in domestic sewage. The modification also altered the surface electronegativity of MWCC to produce positively charged surface which increased the affinity and attachment of biofilms on the carrier. Also, the surface modification of WCC reduced the period needed for biofilm maturation as compared to the original material. The SEM images showed morphology of the NM-WCC and MWCC surface characteristics. The hydrophilicity of the two biofilm carriers was checked through the water absorption rate and water contact angle. The analysis of both results indicated that MWCC was more hydrophilic than NM-WCC which could be attributed to the surface modification of MWCC. The results of the domestic wastewater treatment showed that MWCC was more viable and suitable for pollutant degradation than NM-WCC. The MWCC installed BCBR (R2) had higher removal efficiency of COD, NH_4^+–N and TP in municipal sewage treatment than NM-WWC installed in BCBR (R1). Surface modification also increased the chemical functional groups such as carbonyl and hydroxyl. Thus, this study presented relevant data demonstrating that modification of waste cotton cloth (WCC) through H_2SO_4 utilization increases the removal efficiency of some organic pollutants from domestic wastewater treated in a BCBR. This method of wastewater treatment is an innovative technique which strategically resolves solid and liquid waste challenges concurrently. This method is not only effective, feasible, inexpensive, energy saving, reduction of wastewater treatment cost and environmentally friendly, but it also serves as a source of resource utilization and reuse. Furthermore, it provides alternative textiles waste recycling for both developing and developed countries thereby reducing their environmental and health impacts. Furthermore, it provides insights for the prospects of WCC as biofilm carrier for domestic wastewater treatment which could also be applied in other forms of water treatment with low cost and high efficiency. The collection, sorting and recycling of WCC could create employment and contribute to solid waste management, cleaner water and environmental protection. Future studies can explore further into details the length, arrangement and shape of WCC for proper biofilm attachment. In addition, the effects of various parameters, such as temperature, DO and filling rate on pollutant removal rate can be further studied.

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Compliance with ethical standards

Conflicts of interest We declare no conflict of interest pertaining to this work

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