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REUSE OF PLASTIC WASTE AS BIO-CARRIERS FOR DOMESTIC WASTEWATER TREATMENT – AN APPROACH TOWARDS CIRCULAR ECONOMY

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Abstract

With the rapid growth of economic development, the circular economy has been raised in many international environmental and socio-economic conferences and forums nowadays. The recycling of plastic waste was one of those aspects. This paper studied the potential of directly reusing plastic bottle caps as bio-carriers in moving bed bioreactors without further waste processing. As the caps had lower surface areas than the commercial carriers, total bacteria communities on the commercial carriers (K3) were 3-4 times higher than those on the bottle caps at different counting magnitudes. Nevertheless, the bottle caps could accommodate similar nitrogen-based bacteria (ammonia-oxidizing bacteria - AOB and nitrite-oxidizing bacteria - NOB density), which promotes well for the nitrification and denitrification processes. The COD and N-NH4 removal efficiencies were 92% and 86% on average, respectively. The high pollutant removal capacity reveals the very promising potential of bottle caps as bio-carriers for domestic wastewater treatment, promoting waste reduction and treatment expenses and presenting an excellent example of economic circular.

Keywords: plastic waste; bio-carriers; microorganism; nutrient removal; wastewater treatment.

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1. Introduction

Rapid population growth and rising capita income have rapidly increased the demand for plastic products in recent decades. According to the Organisation for Economic Cooperation and Development (OECD) report (2022), in 2019, there were 79 million tons of plastic waste generated globally, with about 34 million tons of plastic waste buried in waste landfills, 26 million tons burned in landfills, open-air (including household waste burning activities and incineration at landfills) and about 7 million tons are thought to be lost to the environment [1]. The total volume of plastic waste collected and recycled globally is about 55 million tons. Developed countries in Europe, Japan, and Korea have high rates of solid waste collection and recycling due to mature policies and infrastructure and extended producer responsibility policies (EPR - Extended Producer Responsibility), which help promote recycling for plastic products and packaging [2].

In Vietnam, the volume of plastic waste has increased recently (approximately 2.7 million tons in 2018; about 2.93 million tons in 2021), especially in the Central Highlands, North Central, and Mekong Delta regions. The amount for recycling was 0.89 million tons, corresponding to about 30% of the generated plastic waste. Still, the volume of plastic waste recycled in 2021 was 0.77 million

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tons/ year [2]. Of these, more than 60% are nylon and disposable plastic bags. The massive amount of plastic waste is a big challenge for recycling because these plastics have low value, contain many impurities, are challenging to clean, and quickly cause environmental pollution during the recycling process [2]. In addition, the lack of standardised and reliable methods be applied worldwide due to local conditions of the waste management system is a constraint for plastic waste management and recycling [3]. According to statistics from the Vietnam Plastics Association, in 2020, there were more than 2,000 plastic enterprises nationwide with a capacity of about 7.6 million tons of plastic products per year. Since domestic raw plastics can only meet about 20 - 23% of demand, the industry must import 5.5 - 6.6 million tons of plastic materials of all kinds, such as PE, PP, ABS, PC, and PS, every year [4].

According to the Vietnam National Strategy on Green Growth for the period 2021 - 2030, Vision 2050, three critical goals of reducing emissions, greening economic sectors, and Greening lifestyles have been set clearly. Specifically, it is necessary to focus on applying the circular economy model through the economical and efficient exploitation and use of natural resources and energy, waste reuse, and recycling to minimize adverse environmental impacts. In Vietnam, encouraging waste reuse has been mentioned in the Law on Environmental Protection (2020) and Decree 08/2022/ND-CP. Waste reuse is also an essential part of circular economic development. Remarkably, the recycling rate of plastic waste needs to improve to meet the requirements of domestic raw materials.

Plastic waste can be reused and recycled in many applications, including the packing industry [4], construction and building materials (i.e., flooring, fencing, pavement, floor coverings, and insulation materials) [5], automotive industry, agriculture, and gardening sector (irrigation systems, greenhouse coverings, ground coverings, and garden furniture), healthcare sector, sports equipment (sports gear, bicycle parts), textile products, electronic devices, fuel production, and wastewater treatment [6]. In wastewater treatment, plastic wastes were used to synthesize membranes [7] and carbon-based adsorbent materials [8–10] or processes as bio-carriers [11–14]. Nevertheless, no research has yet to use bottle caps as bio-carriers in wastewater treatment.

This study aimed to evaluate the performance of bottle caps that were reused directly (without any processing) as potential bio-carriers for application in Moving Bed Biofilm Reactor (MBBR) for domestic wastewater treatment. The highlight of this study was to evaluate the density of AOB and NOB bacterial groups on different carrier material samples (bottle caps-studied material and commercial material K3) to understand the ability of bottle caps on "carrying" bacteria. Nitrifying bacteria play a significant role in wastewater treatment systems. Nitrifying bacteria includes two main groups: (1) AOB bacteria group (represented by *Nitrosomonas species*, *Nitrosococcus species*, *Nitrosospira species*, *Nitrosolobus species*, and *Nitrosovibrio species*) carries out the metabolism process of transforming ammonium to nitrite, and (2) The NOB bacterial group (represented by the *Nitrobacter species*, *Nitrococcus species*, *Nitrospira species*, and *Nitrospina species*) performs the conversion of nitrite to nitrate. The growth of nitrifying bacteria in the treatment system depends on the nutrient source and environmental factors such as temperature, pH, and dissolved oxygen content. The more these bacteria are in the bioreactor, the more influential the nitrogen removal process is [15]. In reality, domestic wastewater has dozens or hundreds of bacterial species and groups competing for organic matter and nutrient compounds in the wastewater [15]. Previously, the comparison of the effectiveness or density of microorganisms of bio-carriers from different studies was only partially accurate due to different testing conditions (type of wastewater, experimental running conditions). Not many evaluated from the same testing conditions. Therefore, this study will overcome this drawback by comparing two kinds of bio-carriers based on the same testing and environmental conditions.

2. Material and Methods

2.1. Materials

The raw water used for research was domestic wastewater from the first chamber of the septic tank that collected wastewater from the H2 building and student canteen, Hanoi University of Civil Engineering. The experiment was conducted between March 2021 to May 2022.

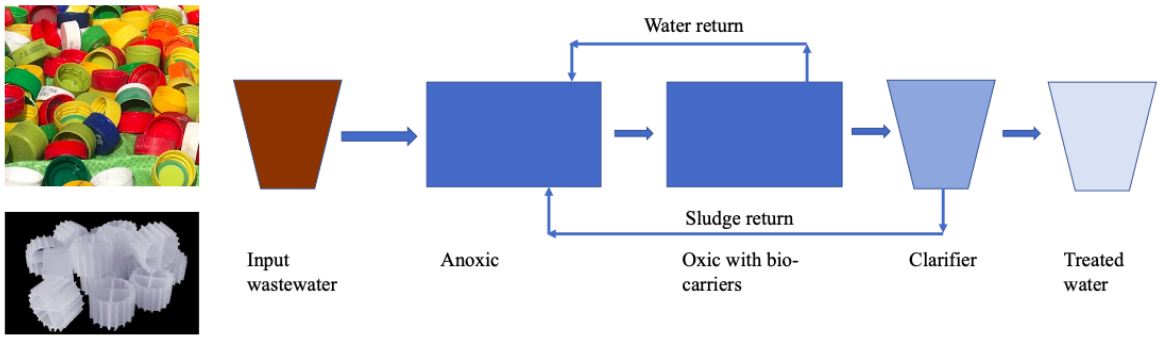


Figure 1. Bottle caps and K3 as bio-carriers (left) and the lab-scale experimental set-up (right)

The bio-carriers were bottle caps collected from plastic recycling manufacturers in Phan Boi village, My Hao commune, Hung Yen province (Fig. 1). These bio-carriers are made of High-Density Polyethylene (HDPE) which can be floated and durable in water.

2.2. Bio-carrier's analysis

a. Surface area

The average surface area of each bottle cap is measured by calculating the surface areas of all faces of the cap. It is 0.00336 m^2.

b. Specific surface area

A one-liter glass volumetric beaker was filled with the bottle caps and packed as tightly as possible. Then, the number of bottle caps used to fill the volumetric beaker (N caps) was counted. Finally, the specific surface area of the bottle cap was calculated based on the following:

$$\text{SSA} = \frac{0.00336 \times \text{N}}{\text{V}} \tag{1}$$

where SSA is specific surface area; 0.00336 is average surface area of one cap (m^2); N is number of caps; V is volume of beaker (L).

c. Porosity

In addition, pour water into the volumetric beaker with fully packed bottle caps.

$$\text{Porosity}\,(\%) = \frac{\text{V}_1 \times 100}{\text{V}_2} \tag{2}$$

where V_1 is volume of water containing bottle caps (mL), V_2 is volume of water without bottle caps (mL).

d. Density

First, determine the weight of the beaker (A_1, g) when it's empty. Then, pack the beaker full with bottle caps, and weigh the beaker with fully-packed bottle caps (A_2, g).

$$\text{Density}\left(\text{g/cm}^3\right) = \frac{\text{A}_2 - \text{A}_1}{\text{V}} \tag{3}$$

where A_1 is weight of the empty beaker (g); A_2 is weight of beaker with fully-packed bottle caps (g); V is volume of beaker (L).

e. Surface examination

For better understanding of the surface of bio-carriers, we used the Scanning electron microscopy (SEM) method to produce detailed, magnified images of an object by scanning its surface with a focused beam of electrons. This was implemented using the Tabletop Microscope (TM4000plus, Hitachi, Japan).

2.3. Experimental setup and testing procedure

A laboratory-scale experimental set-up (Q = 20 L/day) was installed in the lab of the Water Supply and Sanitation Division, Hanoi University of Civil Engineering. The setup included 05 small plastic tanks: feed tank (D = 390 mm, H = 420 mm), Anoxic tank (B × L × H = 385 × 230 × 285 mm), Oxic tank (B × L × H = 385 × 230 × 285 mm), clarifier (D = 200 mm, H = 500 mm) and the treated water tank (D = 200 mm, H = 500 mm). Two peristaltic pumps (MP-2000, EYELA, USA) were used, in which one was used to pump water from the feed tank to the anoxic tank and the other was used to pump the recycled water (NO_3 rich flow) from an oxic tank to an anoxic tank for denitrification. The concentration of dissolved oxygen in the oxic and anoxic tanks was maintained at 4 – 6mg O_2/l and 0.5 - 1 mg/L O_2/l, respectively. The experimental setup was seeded with activated sludge from the Yen So wastewater treatment plant with a mixed liquor suspended solid (MLSS) of 2.5 g/l to accelerate the microorganism growth or MLSS increase.

The plastic carriers were put in the oxic tank with 30% in volume. Even though the carriers were floating, their movement within was limited as the caps were bigger in size and heavier than the commercial carriers.

The experiment was conducted at two input COD concentrations: 135 ± 41 mg/L (or 1.07 ± 0.16 kgCOD/m^3/day volumetric loading rate) and 400 ± 35 mg/L (3.17 ± 0.28 kgCOD/m^3/day loading rate) to evaluate the performance of the bottle caps as bio-carriers at different loading rates.

The final test, conducted in four weeks, involved evaluating the "microorganism accommodation" ability of the bottle caps compared to one type of commercial carrier. The selected commercial material was high-density polyethylene K3 (Jiangxi, China), which has a diameter of 2.5 cm (1.0 cm thickness), density of 0.94 - 0.97 g/m^3, surface area of 500 m^2/m^3 [16] (see Fig. 1). Both carriers were put in the oxic tank with 15% in volume per each type, under the same testing conditions (same operation conditions and experimental system). At the end of the test, ten pieces of each bio-carrier were taken out and sent to Microlab for microorganic analysis.

2.4. Sample analysis

Samples were taken for analysis with the sampling frequency and analytical parameters as in Table 1. pH was measured by a portable pH analytical device (1100, LAQUA Horiba, Japan), while dissolved oxygen (DO) was analyzed by a portable DO meter (HI9142, HANNA, Italy).

In addition, the bottle caps (carriers) and commercial carriers (K3, polyethylene) were sent to Microlab for microorganic analysis in terms of ammonia-oxidizing bacteria (AOB) and nitrite-oxidizing bacteria (NOB) and total microorganism density. A selective medium for the growth of AOB included Na_2HPO_4, KH_2PO_4, $MgSO_4$, $MgCl_2$, $FeCl_3$, $CaCl_2$, $(NH_4)_2SO_4$ (called DSMZ medium) and red phenol (0.5%) solutions. A similar medium was used for NOB cultivation except for replacing $(NH_4)_2SO_4$ and red phenol with $NaNO_2$. The growth medium for the total microorganism comprised high protein, pepton, and NaCl solution. Analytical procedure for AOB and NOB involved the preparation of a series of 3 penicillin vials containing 4.5 ml of DSMZ medium suitable for the growth of AOB and

Table 1. Sampling frequency and Analytical method

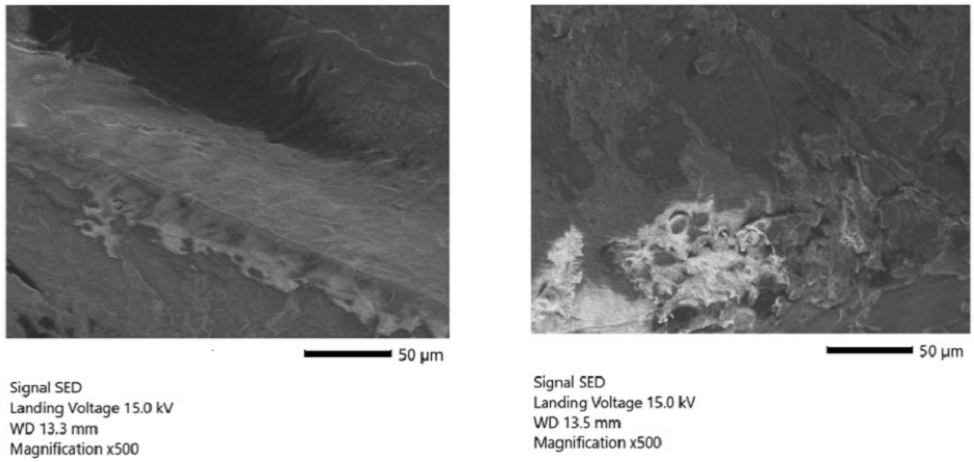
No	Analytical parameters	Sampling frequency	Analytical method
1	pH	Daily	TCVN 6492 : 2011 [15]
2	Temperature	Daily	TCVN 13088: 2020 [17]
3	Flowrate	Daily	
4	DO	Daily	TCVN 7325 : 2016 [18]
5	TN	Twice per week	TCVN 6638: 2000 [19]
6	MLSS	Once per week	2540E, US standard methods
7	Suspended solid (SS)	Twice per week	TCVN 6625: 2000 [20]
8	COD	Twice per week	TCVN 6491: 1999 [21]
9	$N-NH_4^+$	Twice per week	TCVN 6179-1:1996 [22]

NOB bacteria. A sterile needle was used to suck 0.5 ml of the sample to be analyzed into penicillin vial No. 1, then shake well. After that, another sterile needle was used to suck 1 ml from tube 1 to tube 2, shake well. This process can be repeated several times with the addition of penicillin vials containing the fluid medium. The above vials of penicillin were cultured at 28-30 °C for 15 days. The growth of AOB bacteria was observed through changing the environment from red to yellow. For NOB bacteria, their growth was determined through the presence of NO_3^- (using difenilamine reagent) in the culture fluid. Record positive vials and look up the Man 1983 table to determine the number of nitrogen-metabolizing bacteria in the sample to be analyzed. To determine the presence of NO_3^- in the culture fluid, 2 drops of concentrated H_2SO_4 and 2 drops of difenilamine reagent were added into the culture fluid. If the culture fluid appears blue, NO_3^- is present in the culture fluid.

3. Result and discussions

3.1. Characteristics of bio-carriers

The bottle caps were a mix of durable, floatable PP and HDPE plastic with average diameters of 2.2 ± 0.34 cm, porosity of 81.6% ± 2.78, density of 0.92 - 0.97 g/m^3, and specific surface area of 255.4 ± 5.3 m^2/m^3.



(a) Before testing (b) After testing

Figure 2. SEM images of bottle caps

Direct observation by bare eyes showed that the surface of the bottle cap material was relatively smooth, with slight roughness. The inside surface of the caps had spiral-shaped edges, and the outside had small raised spikes. Therefore, it can be hypothesized that the surface roughness inside and outside the circle creates a suitable environment for microorganisms to adhere to grow.

The SEM image of the material (Fig. 2(a)) showed that the surface had uneven roughness. Some places were rough and convex (bright image), and others were concave surfaces (dark image). Therefore, it was predicted that some micro-organisms accumulated on the surface of bottle caps, and their density may not be uniform. As shown in Fig. 2(b), which presents the carriers after the test, there was lots of micro-organism accumulation, filling all the concave surfaces of the caps as carriers.

3.2. Characteristics of feed water

The feed water had the typical properties of domestic wastewater in the condition of Vietnamese combined sewerage, including temperature = 28.8 ± 3.6, pH = 7.5 ± 0.7, TSS of 81.5 ± 2.67 mg/L, $N-NH_4^+$ = 55.8 ± 21.8 mg/L, TN = 66.52 ± 28.3 mg/L. In comparison, COD varied from 120 - 430 mg/L, in the range of low and medium organic loading rates [23]. During the testing period, temperature, pH, and DO were fluctuated but still within the acceptable range (Figs. 3 and 4). For instance, DO in the anoxic tank was 1.04 ± 0.44 mg/L to ensure good condition for the denitrification process. At the same time, it was 5.55 ± 1.02 mg/L in the oxic tank, mainly in the range of 2 - 6 (mg/l), ensuring dissolved oxygen in the tank at a sufficient level for aerobic microorganisms [24].

The temperature is relatively stable, fluctuating between 25 - 40 °C. The lowest value is 21.3 °C, and the highest value is 35.6 °C, due to the influence of seasonal weather (Fig. 3). It should be noted that the optimal temperature values for ammonia-oxidizing bacteria (AOB) and nitrite-oxidizing bacteria (NOB) are 30 - 35 °C and 35 - 40 °C, respectively [25].

The longitudinal pH ranged from 6.5 - 8.5, as shown in Fig. 3, satisfied the requirement of the National Technical standard for domestic wastewater, column A- QCVN 14:2008/BTNMT (pH = 5 - 9) [26]. It is relatively stable and suitable for microbial activity. The pH values indicated an alkaline environment, although there was a slight increase during treatment. In pollutant decomposition, organic nitrogen is converted into ammonium nitrogen and nitrate NO_3^- sequentially. In anoxic conditions, denitrification occurs, reduces NO_3^- ions to N_2, and consumes H^+ cation, thus increasing the alkalinity of wastewater but not inhibiting microorganisms [27].

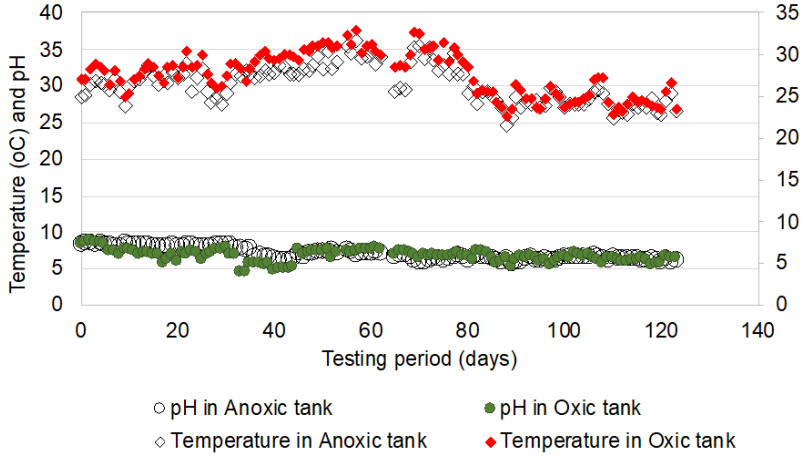


Figure 3. pH and temperature in the bio-reactors

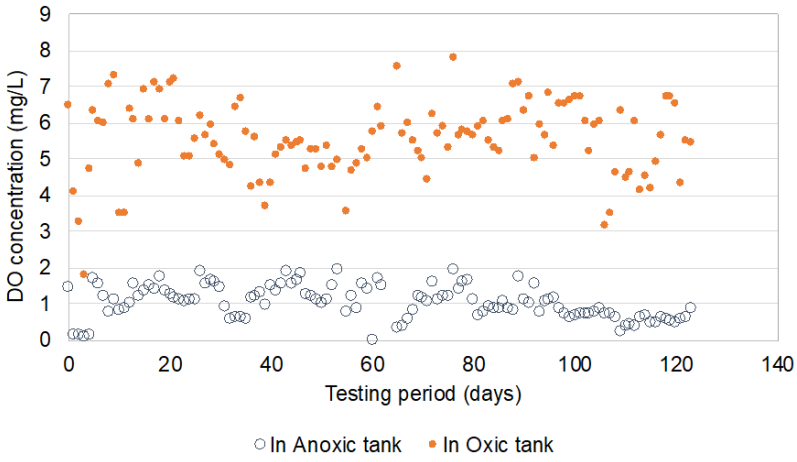


Figure 4. DO concentration in the bioreactors

3.3. Evaluation of microorganism density on bio-carriers

a. AOB bacteria and NOB bacterial group

The number of nitrifying bacteria in the two analyzed samples is shown in Table 2.

Table 2. AOB and NOB density

Sample	AOB density (MPN/ml)	NOB density (MPN/ml)
Bottle caps	10^4	10^4
K3 carriers	10^3	10^3

The above results show that the analyzed samples all contained AOB and NOB bacteria groups with a relatively high number of 10^3 - 10^4 (MPN/ml). Interestingly, the number of NOB bacteria was almost the same as that of AOB bacteria because the product of the oxidation of ammonium to nitrite (performed by AOB bacteria) would be further oxidized by NOB bacteria to nitrate. It should be noted that the surface area of the K3 carrier is 500 m^2/m^3, while that of the bottle cap is 255 m^2/m^3. The caps had rougher surfaces but fewer angles and edges than those of K3 by direct observation. Normally, higher surface area or rougher surface would create more housing for bacteria. Even though the commercial K3 has higher surface area, but the surface is not as rough as bottle cap, the bacteria density was therefore similar.

b. Total microorganism density

Microorganisms are diverse and can be classified into beneficial or harmful groups. The beneficial groups are those participating in the metabolism of environmental pollutant removal, such as nitrogen-oxidizing ones (i.e., AOB, NOB), sulfur-oxidizing ones, or difficult-to-decompose ones. As for the harmful group, they are often disease-dispreading pathogenic microorganisms such as *Pseudomonas aeruginosa, Bacillus anthracis, Burkholderia species*, etc. Therefore, the total microorganisms, including both types of microorganisms in the testing samples, were also of concern.

It can be seen from Table 3 and Fig. 5 that the total bacteria communities on the commercial carriers (K3) were 3-4 times higher than those on the bottle caps at different counting magnitudes. Meanwhile, the concentrations of beneficial groups, such as nitrogen oxidizing ones (i.e., AOB, NOB), were similar. Therefore, it can be expected that many of the “unwanted” pathogenic species for nitrification were present in those commercial carriers.

Table 3. Total microorganism density

	10^3 (MPN/ml)	10^5 (MPN/ml)	10^7 (MPN/ml)
Bottle caps (VL1)	110	42	10
K3 carriers (VL2)	> 500	135	46

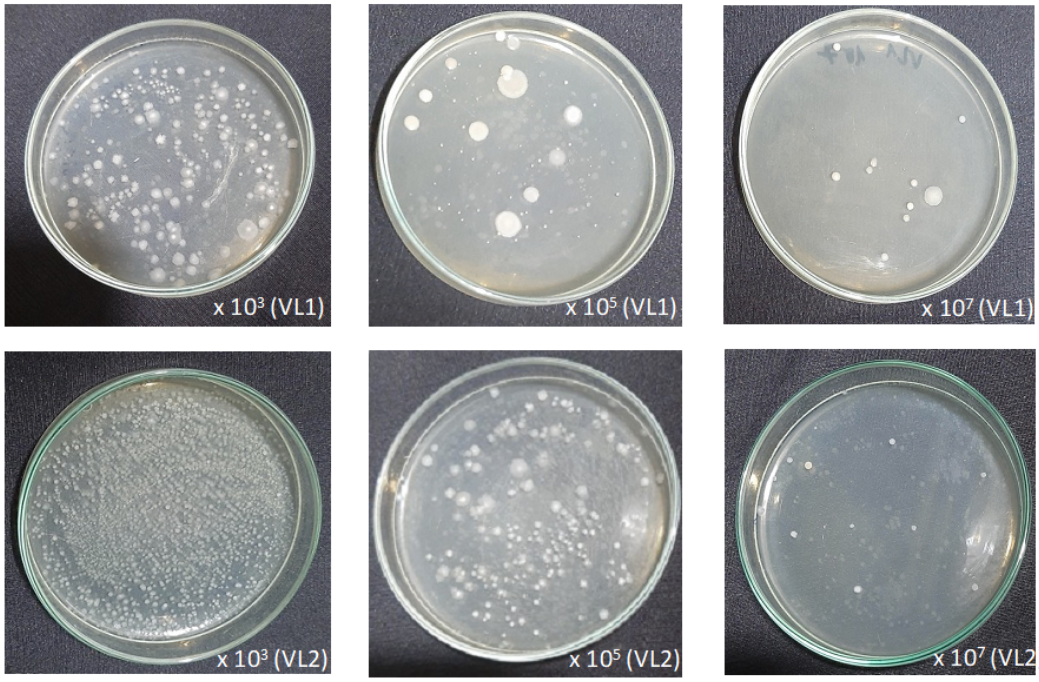


Figure 5. Images of total bacteria community growth on petri disks

3.4. Organic and nutrient removal efficiency

During the test, the SVI and MLSS of in the MBBR were 68.8 ± 8.6, 4.5 ± 1.07 g/L, respectively, which are favourable conditions for biological treatment. The $N-NH_4^+$ content in the input wastewater ranged from 25 - 112 mg/l, and the content in the treated output ranged from 3 - 40 mg/l, as seen in Fig. 6. Most of these reach lower values than the allowable value of column B - QCVN 14:2008/BTNMT [26]. When the input $N-NH_4^+$ content increased above 70 mg/l, the output wastewater did not meet standards (> 10 mg/L). Processing efficiency was from 70% to 92%, with an average efficiency of 86%. The ammonium treatment efficiency of the MBBR with reusable plastic bottle cap media was quite similar to Libing Chu's research results with the MBBR model - PCL and PU media [28] or another research with glass stone media [29], slightly lower than the research results with commercial S20-4 plastic substrates in the study of Le Hoang Viet and Nguyen Vo Chau Ngan [30].

To evaluate the capacity of these plastic carriers with shocks of organic loads, the organic loading rate in the feed was increased from 1.07 kg COD/m^3/day to 3.17 kg COD/m^3/day on average for six months. It was found that the efficiency was reduced slightly on the day of increasing the organic loading rate (in November 2021). It was hypothesized that the caps created suitable accommodation and protected microorganisms from being destroyed under environmental changes such as pH, salinity, or organic loading rates. Previous studies have shown that biofilms on carriers have longer survivability rates, and they were much more resilient than (i.e., suspended sludge), at least in the case of survivability [31].

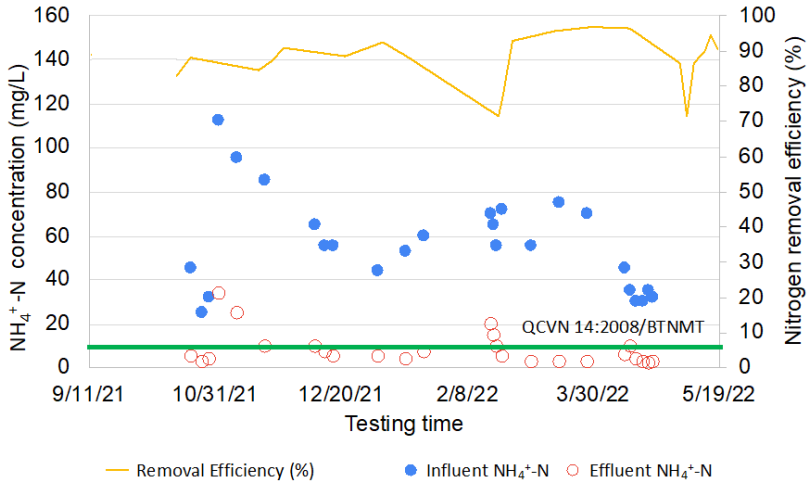


Figure 6. Evaluation of nitrogen control efficiency

Fig. 7 shows the removal efficiency was relatively high (from 70 to 97%). The effluent COD always met the national technical standard QCVN 14:2008/BTNMT (column A) [18]. The control of COD concentration in wastewater was due to the consumption by microorganisms for energy and cell synthesis. Besides, the suspended solid settling process also contributed to controlling organic matter. The high COD removal efficiency proved that the bottle caps had comparable organic handling efficiency to other commercial carriers, from 80-90% [32].

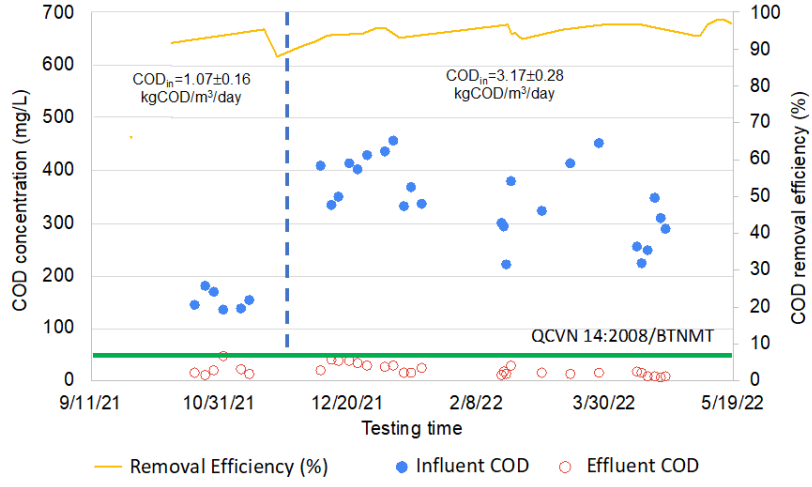


Figure 7. Evaluation of COD removal efficiency

Interestingly, it was the first time the bottle caps were used as bio-carriers in MBBR for wastewater treatment. Previous to our research, they were once used as support media in submerged aerated filters in the study of de Oliveira et al. [33]. The study achieved an average removal of COD equal to 78% by applying an organic loading rate below 2.6 kg COD/m^3/d and the removal efficiency of $N-NH_4^+$ greater than 76%. The dominance of AOB and NOB was also found when organic loading rates decreased, in which the *Nitrospira* bacteria population (AOB) was more significant than *Nitrobacter* (NOB) in all their experiment. With those good removal efficiencies, the bottle caps were shown to be a potential alternative material as a new type of support medium.

4. Conclusions

Using MBBR technology to treat wastewater from HDPE plastic materials shows stable and effective treatment performance. MBBR membrane filter system provides high and stable treatment efficiency of organic substances (COD: 70 - 97%) and nutrients ($N-NH_4^+$: 70 - 92%). With a circulation rate of 100%, the system's output wastewater meets National technical regulations on domestic wastewater QCVN 14:2008/BTNMT (column A).

Much effort has been made to improve plastic materials' recycling and reuse rates. Wastewater treatment by using recycled plastic materials as adsorbents and biofilm materials is still limited. Therefore, it is essential now to promote the reuse of plastic materials in wastewater treatment to bring added value to plastic waste, improve wastewater treatment efficiency, and reduce costs. The research opens up a sustainable and environmentally friendly way to treat wastewater and waste, thus decreasing solid waste emissions and contributing to thoroughly solving environmental problems, taking advantage of recycled plastic materials. Future research shall involve other types of plastic waste that could be reused as bio-carriers besides the bottle caps and test at other organic loading rates to confirm the feasibility of this application. Based on that, the formation and development of a circular economy can enhance in all aspects of life.

Acknowledgment

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