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Rubber glove wastewater treatment by integrated fixed-film activated sludge using polyvinyl alcohol gel beads: effect of hydraulic retention time and percent of biomass carriers

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ABSTRACT

In the present study, an integrated fixed-film activated sludge (IFAS) process using polyvinyl alcohol (PVA) gel beads as biomass carrier was applied to treat the wastewater from the rubber glove industry. Chemical oxygen demand (COD), ammonium nitrogen, nitrate, orthophosphate, pH and dissolved oxygen were analyzed prior to the IFAS process. Two systems comprising IFAS/biofilm (PVA gel beads only) and IFAS/hybrid (PVA gel beads + activated sludge) were investigated. The COD removal efficiency was studied through manipulating the hydraulic retention time (HRT: 2, 4, 6, 8 h) and biomass carriers percent filling (5%, 10% and 20%). The present results showed that increases in HRT and biomass carrier's percent filling improved COD removal significantly. The highest COD removal efficiency achieved was 87% at HRT of 6 h using the IFAS/hybrid method. At lower HRT of 2 and 4 h, the IFAS/biofilm system showed higher COD removal efficiency of (58% ~ 61%) compared to the IFAS/hybrid (38%–46%) and vice versa at higher HRT of 6 and 8 h. This was probably due to rapid absorption of soluble compound into PVA gel beads before degradation. In conclusion, the IFAS system using PVA gel beads could be a promising effective alternative treatment method to replace the conventional activated sludge system for the removal of COD in rubber glove wastewater.

Keywords: Integrated fixed-film activated sludge (IFAS); Chemical oxygen demand (COD); Hydraulic retention time (HRT); Polyvinyl alcohol (PVA) gel; Rubber glove wastewater

1. Introduction

The rubber glove industry is one of the major manufacturing industries in Malaysia that generates large amount of wastewater. The sources of wastewater mainly come from the cleaning process of compounding latex storage tank, former cleaning tank and process leaching tank which contains a large amount of fine suspended particles and dissolved organic substances. This contributes to high chemical oxygen demand (COD), biochemical oxygen demand (BOD), total suspended solids (TSS), total nitrogen (TN) and trace heavy metals in the untreated wastewater (Table 1). Chemical methods such as coagulation with dissolved air floatation (DAF) and activated carbon filtration have been adopted for treatment of low strength rubber glove wastewater. For medium strength or high strength wastewater, existing chemical methods are incorporated with biological treatment systems such as aerobic activated sludge, trickling

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filter or facultative ponding system to further improve the efficiency of rubber glove wastewater treatment [1].

Various studies on treatment of wastewater from the rubber glove industry using biological methods as secondary treatment have been carried out. For example, a batch aeration system has been used to treat rubber glove wastewater with organic loading rate (OLR) varied from 0.9–1.28 kg BOD_5 m^{-3} d^{-1} [4]. The findings showed that the optimum F/M ratio of 0.4 d^{-1} and 12 h hydraulic retention time (HRT) resulted in highest BOD_5 and COD removal efficiencies of 98.6% and 89.3%, respectively. In addition, a sequencing batch reactor operated under aerobic condition had successfully been used to treat rubber glove wastewater to meet the permissible regulatory discharge limits [2]. At a high MLVSS of 9,600 mg L^{-1} and a subsequent 20 d HRT and 24 h cycle period, COD and BOD removal efficiencies of 81.24% and 93.73%, respectively could be achieved at OLR of 0.0467 kg COD m^{-3} d^{-1}. Furthermore, a cultivated aerobic granular sludge has been applied to treat rubber processing wastewater and the process obtained good COD removal efficiency of 96.5% at OLR of 3.7 kg COD m^{-3} d^{-1} after 90 d of operation [5]. In another study, a combination of flocculation with DAF system followed by the activated sludge system proved to be an effective approach in treating rubber glove wastewater [1]. In the study, the wastewater treatment plant operated at higher F/M ratio of 0.1–0.2 d^{-1} for extended aeration system performed well and managed to meet regulatory discharge limits.

In recent years, the rubber glove manufacturing in Malaysia has expanded tremendously due to the high demand for medical gloves. This results in the rise of organic loading in the wastewater, which is the main cause of operational failure in many existing wastewater treatment plants. Additionally, the public concern regarding environment protection and health risk problems as well as managing effluent discharge limits are among the driving factors for wastewater treatment plant upgrade. However, decision-makers face the real challenge of space constraint to rehabilitate existing wastewater treatment plant capacity. On the other hand, one of the common problems frequently faced by plant operators is the solid separation due to bulking sludge from the excessive filamentous growth. Solid carryover results in high turbid effluent and leads to poor wastewater treatment performance. Hence, this limitation can be overcome by using an integrated fixed-film activated sludge (IFAS) system which combines conventional activated sludge with those of the biofilm system in a single tank.

IFAS system incorporates fixed or free biomass carriers to an activated sludge reactor, in which the biomass carriers provide more surface area for the growth of microorganisms, thus able to increase the biomass population. In other words, the IFAS system enhances the treatment plant capacity in the same tank volume by accomplishing biodegradation through both suspended as well as attached biomass. In addition, the presence of the biomass carriers not only reduces solid loading on the final clarifier since it is retained within the reactor, but it also provides development of diverse microbial population which improves plant performance due to both organic and hydraulic shock loading [6].

Recently, there has been increasing interest in the development of polyvinyl alcohol (PVA) gel beads based IFAS system for municipal and industrial wastewater treatment. PVA gel beads are suitable to be used as biomass carrier as they enable large numbers of bacteria to grow, great fluidity in water, and require relatively low energy for mixing [7]. In addition, PVA gel beads also provide a higher hydraulic capacity in IFAS system as they are normally used at filling ratio around 5%–20%, against a much higher ratio of 30%–70% for other non-porous types of biomass carriers [8]. In India, treatment of municipal waste using 10% filling ratio of PVA gel beads as biomass carrier in IFAS system showed mean removal of COD and BOD of ~91% and 92%, respectively when the process was stabilized [7]. In another study [9] treated aquaculture wastewater with an expanded granular sludge bed reactor using PVA gel beads, the process yields up to 92% COD removal and over 90% total suspended solids removal efficiencies. Likewise, the study that treats phenol-containing wastewater showed the reactors supplied with PVA gel beads have significantly lower concentration of phenol in their effluent [10].

To the authors' best knowledge, there are limited studies on the application of IFAS in the treatment of rubber glove wastewater. In this context, the crucial factor for the IFAS tank sizing includes the pollutant removal efficiency and the associated percent filling ratio of the chosen biomass carrier based on the pollutant loads. Furthermore, there is also a lack of comprehensive understanding about the hydraulic retention time (HRT) on the IFAS behavior. Thus, the purpose of this study is to investigate the feasibility of utilizing the PVA gel beads based IFAS system as secondary treatment strategy to remove organic matter in rubber glove wastewater. While determining the optimal HRT and biomass carrier's percent filling, their effect on COD removal is also discussed. The outcome of this study could serve as a

Table 1
Characteristic of standard rubber glove wastewater in Malaysia

	Rubber glove wastewater [1]	Rubber glove wastewater [2]	Mean [3]
Location	Negeri Sembilan	Perak	Malaysia
pH	5.1–8.4	6.4	6.8
COD (mg L^{-1})	300–1,490	933	770
BOD (mg L^{-1})	85–600	542	276
TSS (mg L^{-1})	184–1,306	191	236
TN (mg L^{-1})	NA	–	37
Zn (mg L^{-1})	0.48–11.6	23	12.5

preliminary study to examine the technical feasibility of IFAS system in the rehabilitation of an existing overloaded conventional activated sludge wastewater treatment system.

2. Methodology

2.1. Material

The synthetic wastewater mimics the biologically treated rubber glove effluent was prepared followed the recommendation in a published reference [11]. Table 2 summarizes the composition of the synthetic wastewater used in this study. The prepared synthetic wastewater has COD:N:P ratio of 100:5:1. All chemicals used were obtained from R&M Chemicals.

Fresh synthetic wastewater was prepared to be fed into IFAS reactor and maintained to meet the targeted concentrations of COD, N–NH_4^+ and PO_4^{3-} concentration at 976 ± 50 mg L^{-1}, 42 ± 10 mg L^{-1} and 10 ± 5 mg L^{-1}, respectively. The biomass carrier PVA gel beads were supplied by Kuraray Co., Tokyo, Japan. The PVA gel beads are porous hydrogel, hydrophilic in nature and suitable for immobilization of microorganisms. Table 3 presents the PVA gel beads with their specifications.

2.2. Acclimatization of microorganism in IFAS reactors

A 140 mm (D) × 205 mm (H) polyethylene cylindrical tank with working volume of 2 L was used as a bench scale batch IFAS reactor as presented in Fig. 1. Prior to acclimatization, the activated sludge obtained from a rubber glove wastewater plant in Taiping, Perak was used for microorganism seeding. During the acclimatization process, synthetic wastewater, activated sludge and PVA gel beads in the ratio of 7:1:2 were fed into the reactor. The reactor was then allowed to run continuously in order for biofilm to develop on the PVA gel beads. Air pumps with air diffusers were placed at the bottom of the reactor to provide aeration to the growth of bacteria and enable them to run their activities. COD, NH_4^+–N and PO_4^{3-} in the reactor were analyzed and the synthetic wastewater was replaced by fresh batch of solution when the COD value dropped to half of its initial value. During this period, the dissolved oxygen was controlled above 3 ppm to maintain aerobic condition in the reactor, while pH was maintained at 6.5–8 at temperature 25°C. An acclimation period of 15 d [10] was allowed for PVA gel beads and activated sludge to adapt to the corresponding experimental conditions.

Table 2
Composition of synthetic wastewater

Chemicals	Concentration (mg L^{-1})
Glucose	1,000
$CaCl_2$	12
$MgSO_4$	12
$(NH_4)_2SO_4$	250
$NH_4H_2PO_4$	40

Table 3
Characteristics of biomass carriers

Characteristic of media	Value
Appearance	
Shape	Spherical
Diameter (mm)	4
Effective surface area (m^2/m^3)	2,500
Specific surface area (m^2/m^3)	1,000
Specific gravity	1.025

2.3. Batch IFAS reactor operation

After the acclimatization stage, two different sets of experiments were carried out in batch mode, (i) IFAS/biofilm reactor (only PVA gel beads) and (ii) IFAS/hybrid reactor (contains both activated sludge and PVA gel beads). In the IFAS/biofilm reactor, 200 mL of PVA gel beads, which was equivalent to biomass carrier percent filling of 10% (v/v) was added to synthetic wastewater and ran at 2 h HRT. Aerobic condition was maintained in the reactor and pH was maintained at 6.5–8. At the end of the experiment, sludge sedimentation took place for 30 min by turning off the air pumps. Subsequently, 300 mL of the supernatant water was withdrawn from the reactor for quantitative analysis in COD, NH_4^+–N and PO_4^{3-}. Then, 300 ml of fresh synthetic wastewater was added and the operation cycle was repeated at different HRT (4 h, 6 h and 8 h). In another experiment to study the effect of different biomass carrier's percent filling, the same batch IFAS system operation cycle was performed for biomass carrier percent filling of 5% (v/v), 10% (v/v) and 20% (v/v), respectively at 4 h HRT. While in the IFAS/hybrid reactor, 400 mL of activated

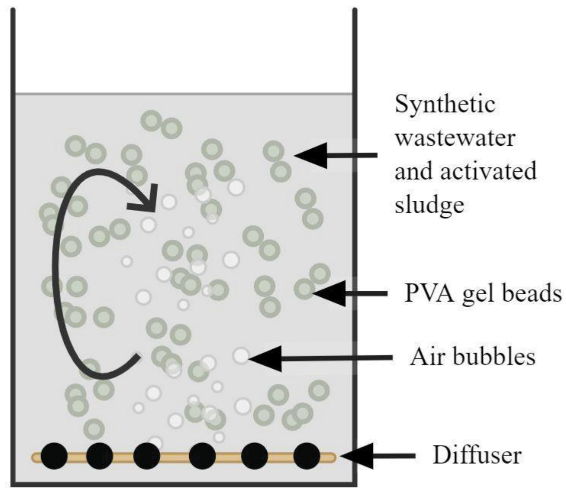

Fig. 1. Experimental setup of the batch IFAS reactor with PVA gel beads.

sludge was added to the mixture of synthetic wastewater and PVA gel beads. Similar operating conditions and procedures applied for IFAS/biofilm reactor were used in the case of the IFAS/hybrid reactor.

All operating parameters in the influent and effluent of the reactor were measured using a HACH DR 2400 Spectrophotometer. COD was measured by reaction digestion method (Method 8000), ammonium nitrogen (NH_4^+–N) by the Nessler method (Method 8038), nitrate nitrogen (NO_3^-–N) by the chromotropic acid method (Method 10020) and orthophosphate (PO_4^{3-}) by the ascorbic acid method (Method 8048) respectively, with reagent kits from Hach Company, Loveland, Colorado. The performance of IFAS system was indicated by COD removal efficiency, which was calculated based on:

$$\text{COD removal efficiency}\left(\%\right)=\left(\frac{\text{COD}_e-\text{COD}_i}{\text{COD}_i}\right)\times 100\% \quad (1)$$

where COD_e and COD_i represent COD of effluent and influent, respectively.

3. Results and discussion

3.1. Effect of HRT

The COD removal efficiency at different HRTs for constant COD of 976 ± 50 mg L^{-1} was investigated in order to find the optimum HRT. Fig. 2 illustrates the total COD removal efficiency at different HRTs for both IFAS/biofilm and IFAS/hybrid reactors. At 10% biomass carrier filling percentage, both reactors demonstrated good removal efficiencies of organic matter at higher HRT of 6 h and 8 h, where COD effluent reached the permissible limit for treated wastewater for discharge (i.e., below 200 mg L^{-1}). It can be seen that in the treatment of rubber glove wastewater using the IFAS approach, HRT of 8 h and 6 h were found to be the optimal HRT for IFAS/biofilm and IFAS/hybrid, respectively. This is because the IFAS system could achieve highest COD removal efficiency at the specified HRT. However, due to the marginal improvement in COD removal efficiency (5% difference) for HRT at 8 h, compared with 6 h using IFAS/biofilm, HRT of 6 h (at OLR = 3.87 kg COD m^{-3} d^{-1}) could be recommended as design criteria for dimensioning IFAS tank in achieving adequate organic removal from rubber glove wastewater. Similarly, this recommendation is also supported by the fact that COD is insignificantly improved by increasing the HRT [12]. Previously, Zhang et al. [13] studied the effect of HRT (6–12 h) on COD removal using a laboratory scale IFAS on anaerobic anoxic oxic-biological contact oxidation (AAO-BCO) process at 170–310 mg L^{-1} influent COD. Their results demonstrated that increasing HRT slightly affects COD removal (78%–83%) even when the HRT is as short as 6 h.

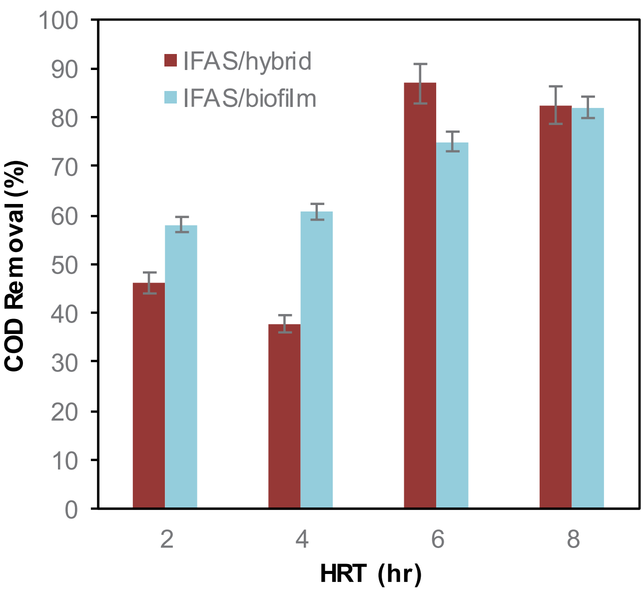

Fig. 2. COD removal efficiency at different HRT for IFAS/hybrid and IFAS/biofilm reactor with 10% biomass carriers filling.

In IFAS/biofilm reactor, HRT and COD removal efficiency have a proportional relationship in which increase in HRT results in higher COD removal efficiency. Similar observations were reported by Musa and Idrus [14], Majid and Mahna [15], and Shafie et al. [16]. The increment in COD removal efficiency could be justified by the fact that the increase of HRT in the IFAS/biofilm reactor provides greater contact time to facilitate the microbial adaption to the environment and enhance the degradation of organic matters by microbial populations in the PVA gel beads [12]. In biofilm process, every PVA gel bead serves as active surface area sustaining the microorganism and protects them from predation in the micro-scale pores of the PVA gel beads [17]. Removal of organic matters takes place when the substrates are transported from wastewater to the biofilm, diffuse through a mass transfer resistance layer and are consumed by the fast-growing heterotrophic bacteria. Comparable COD removal efficiency was observed at HRT of 2 h and 4 h, which indicated rapid bio-sorption of soluble COD into PVA gel beads but insufficient time for bacteria to degrade the organic matter. At higher HRT, the presence of adequate contact time and increasing population of heterotrophic bacteria in the superficial layers of the biofilm may quickly convert the organic matter to useful by-products.

The obtained COD removal efficiency of 82% at OLR of 3.87 kg COD m^{-3} d^{-1} at HRT of 8 h in our study is consistent with the studies conducted by other researchers. Gani et al. [18] reported COD removal efficiency >80% using PVA gel beads containing MBBR in treating domestic wastewater at OLR of 1.69 to 7.82 kg COD m^{-3} d^{-1} at 20°C. In another study, Chen et al. [19] used PVA immobilized organisms to treat synthetic municipal wastewater at HRT of 2–10 h. They reported COD removal efficiency of more than 90% at COD loading rate lower than 2 kg COD m^{-3} d^{-1} and at highest OLR of 4.22 kg COD m^{-3} d^{-1} at 25°C, the COD removal efficiency still surpassed more than 80%.

In IFAS/hybrid reactor, COD removal efficiency fluctuated between 38%–87% when HRT gradually increased from 2 to 8 h. In the IFAS/hybrid reactor, the two major bacterial aggregates are the initial activated sludge biomass (suspended flocs) and the attached biofilm formed on the surface of the PVA gel beads. The mixed liquor suspended solids (MLSS) and volatile suspended solids (MLVSS) in IFAS/hybrid is 653 ± 55 mg L^{-1} and 397 ± 52 mg L^{-1}, respectively. The coexistence of these biomass in IFAS/hybrid reactor subsequently affects the mixing efficiency, distribution of dissolved oxygen and effective oxygen transfer rate in the reactor. Accordingly, these changes in operating conditions greatly influence the absorption of soluble COD into PVA gel beads and microbial activity in the suspended flocs, thus give rise to less efficiency for organic removal of reactor at lower HRT. However, substantial COD removal efficiency of 87% was recorded at HRT of 6 h, which is comparable with the study conducted by other researchers [20–22] using synthetic and real domestic wastewater. The high COD removal efficiency could be attributed to the simultaneous effect in the suspended flocs and attached biofilm of different heterotrophic microorganisms, which may assimilate readily and slowly biodegradable substrates. The slight decrease in COD removal efficiency to 83% at HRT of 8 h could be explained by a lower bacteria activity that could be due to death by substrate-decomposing bacteria [23].

3.2. Effect of percent biomass carrier

Generally, percentage of biomass carrier is another important parameter that affects HRT and solid retention time in an IFAS reactor. Too low filling ratio may cause insufficient bioactivity to degrade organic matters. In contrast, too high percent of biomass carrier may cause high mixing and aeration requirements and will affect biofilm stability due to substrate loading.

According to Sfaelou et al. [24], the surface organic groups such as –COOH, –OH, and $-NH_2$ on PVA gel beads increase its hydrophilic character. It is postulated that the microbial cells are able to bind tightly with polar groups on the PVA gel beads. This promotes the likelihood of adhesion and biofilm growth and results in a higher organic removal efficiency. From Fig. 3 it is clearly seen that the percent of PVA gel beads contributed positive impact to organic matter removal in the IFAS reactor, with steady increase in COD removal efficiency of 46%, 61% and 66%, respectively in the IFAS/biofilm system, which corresponding to percentage of biomass carrier of 5%, 10%, and 15%. At higher percentage of biomass carrier, greater accomplishment of organic removal is due to the higher solid retention time and amount of biofilm present in IFAS/biofilm reactor. This result is in agreement with the study conducted by Quang and Thủy [25] using the same type of biomass carrier to treat seafood processing wastewater after anaerobic digestion. Similar findings were also reported by other researchers using other porous or non-porous type biomass carriers [26,27], which improved the quality of treated wastewater.

On the other hand, IFAS/hybrid reactor showed a different COD removal pattern with increasing percentage of biomass carrier. It is known that suspended flocs and attached biofilms make different contributions to the overall performance in IFAS/hybrid system [28]. Suspended flocs possessed higher negative charge and became more hydrophobic than the attached biofilm [27]. The fast growing heterotrophic bacteria are more hydrophobic and tend to floc rather than forming biofilm on the biomass carrier. Thus, the hydrophobicity and surface charges of both biofilm and floc could potentially contribute to the different sorption and removal mechanism of dissolved organics and colloidal.

3.3. Removal of ammonium nitrogen, orthophosphate and nitrate nitrogen in IFAS reactor

The performance of the IFAS reactor on removal of ammonium nitrogen, orthophosphate and nitrate nitrogen was determined at optimum HRT of 6 h and 10% (v/v) of biomass carrier. Under aerobic conditions, heterotrophic microorganisms consume soluble organic compounds and nutrients such as orthophosphate and ammonium nitrogen in the wastewater for cell maintenance (i.e., oxidation) and microbial cell growth (i.e., assimilation). Alternatively, ammonium nitrogen could also be oxidized to nitrate by autotropic bacteria in aerobic condition in the nitrification process. From Table 4, the treatment effect for ammonium nitrogen in both the IFAS/hybrid and IFAS/biofilm reactors was most likely due to cell synthesis rather than

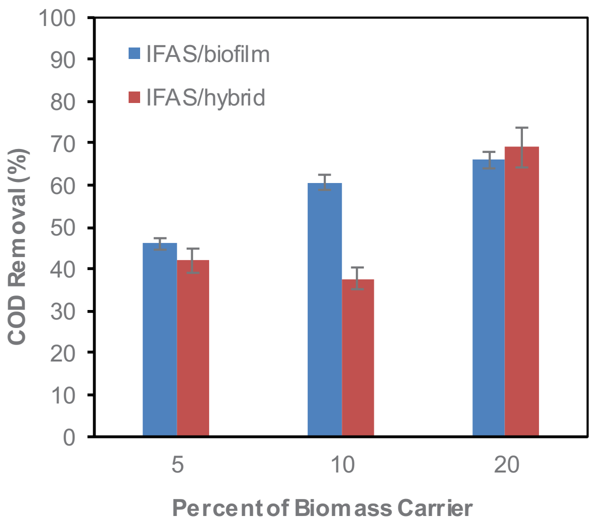

Fig. 3. COD removal efficiency at different percentage of biomass carrier for IFAS/hybrid and IFAS/biofilm reactor at HRT of 4 h.

Table 4
Percentage of removal for $N-NH_4^+$, PO_4^{3-} and $N-NO_3^-$

	OLR (kg COD m^{-3} d^{-1})	% removal of NH_4^+–N	% removal of PO_4^{3-}	% removal of NO_3^-–N
IFAS/hybrid	2.44	6.15	2.5%	1.4
IFAS/biofilm	2.36	17.40	3.8%	2.4

the nitrification process. This is because nitrification generally takes place at lower C:N ratio (5:1) and high HRT (up to few days) as autotropic bacteria normally grows slower than the heterotrophic bacteria, in which these suitable operating conditions were not present in the current study. This explanation is also supported by the insignificant accumulation of nitrate nitrogen in the effluent. On the other hand, the slight increase of orthophosphate in the IFAS system could be due to the death by substrate-decomposing bacteria.

4. Conclusions

In the present study, the investigation of IFAS performance on organic matters removal from synthetic rubber glove wastewater using COD as performance indicator was successfully carried out. PVA gel beads with porous matrix were found to provide extra surface area for the heterotrophic bacteria to attach and grow. The HRT of 6 h is recommended for the design of the IFAS system, at OLR of 2.4 kg COD m^{-3} d^{-1} with COD removal efficiency in the range of 77% to 87%. Overall, the increase in HRT and percent of biomass carrier showed positive effect on the COD removal efficiency, depending on the presence of suspended biomass in the reactor. The IFAS/hybrid reactor showed a better COD removal efficiency at HRT of 6 h and 20% biomass carrier filling ratio, compared to the IFAS/biofilm reactor. The low to insignificant removal of ammonium nitrogen, nitrate and orthophosphate in the current study could be due to insufficiency of nitrifying bacteria population and a high C:N ratio. Therefore, further study is required to investigate the effect of C:N ratio and the effect of intermittent aerobic and anoxic conditions for greater nutrient removal efficiency. In conclusion, this study showed that IFAS using PVA gel beads could be a promising effective alternative treatment method to replace the conventional activated sludge system for the removal of COD in rubber glove wastewater.

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