Municipal wastewater treatment using a novel granular activated carbon based integrated fixed-film activated sludge sequencing batch reactor (IFAS-SBR)

by

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Abstract

Integrated fixed-film activated sludge (IFAS) systems incorporate benefits provided by both attached and suspended growth systems. It is an advanced technology in wastewater treatment with high treatment efficiency and low capital and operational maintenance costs. This study used granular activated carbon (GAC) as a new carrier in plastic bio-ball carriers. Two laboratory integrated fixed-film activated sludge sequencing batch reactors (IFAS-SBR) – one with conventional plastic biofilm carriers (plastic-IFAS) and one with GAC encaged carriers (GAC-IFAS) – were operated for 120 days to treat synthetic municipal wastewater. Reactor treatment performance and biofilm formation mechanism were investigated in this paper. The GAC-IFAS sequencing batch reactor showed better and more stable performance compared to the plastic-IFAS sequencing batch reactor. GAC is a promising bio-carrier alternative due to its adsorption capacity and its porous structure, which provides a larger surface area for the biofilm growth. The results demonstrated that the concentration of attached biomass was 1.12 – 1.78 times higher than that of suspended flocs in GAC-IFAS sequencing batch reactor using HRTs of 12 h to 4 h. Moreover, adsorption and biodegradation could be achieved on the outer and inner layers of its surface concurrently. The assessment of adsorption kinetics was also performed by batch tests. Utilization of the newly developed GAC bio-carrier greatly improved the bioreactor performance for wastewater treatment.

Keywords: IFAS-SBR; primary effluent; carrier type; HRT; biodegradation.

Preface

For this manuscript, X. Yang was responsible for experimental design, laboratory experiments, data collection and analysis as well as the manuscript composition. Y. Zhou and L. Zhang assisted with the experimental design and data collection. Y. Liu planned and supervised the study. All authors contributed to the manuscript preparation.

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Introduction

1.1 Background

Energy efficient treatment of municipal wastewater, especially the reclamation of primary effluent, is an emerging topic due to limited water and energy resources. Insufficient wastewater treatment can result in excess organic pollutants, nitrogen, and phosphorus being discharged into the environment, leading to issues like eutrophication (Kadiya et al., 2012). Immobilization of biomass in the form of biofilm is an efficient method to retain slow-growing microorganisms, such as nitrifiers, which contributes to better nutrient removal in continuous flow reactors (Kermani et al.,2008). In biofilm formation process, larger carrier surface area can provide more sites for microorganisms to adsorb and to grow. Furthermore, the attached growth system is normally considered less sensitive to toxic influents and variations in environmental conditions (Wang et al., 2005).

Generally, biofilm carriers are made of distinct materials with different sizes and structures, with plastic media being the most popular choice (e.g. Kaldnes K1, K2, K3 and K5, etc.) (Hoang et al., 2017). Other common choices are polyurethane foam, activated carbon (granular and powdered), modified carriers (e.g. bioplastic-based moving bed biofilm carrier, polyvinyl alcohol-gel carrier, etc.), naturally occurring materials (e.g. sand, zeolite, light expanded clay aggregate, etc.), ceramic carriers, and wood chips (Deng et al., 2016; Nair et al., 2019).

Granular activated carbon (GAC) is a promising bio-carrier material due to its high adsorption and biofilm self-generation capacity. In addition, GAC's porous structure means it contains a higher internal surface area compared to plastic carrier, which provides microorganisms with many more sites to adsorb and grow (Huang et al., 2017). The biofilm mechanism in GAC media involves physical adsorption, biosorption, and biodegradation, which greatly improves the efficiency of wastewater treatment (Martín-Pascual et al., 2012). Studies have demonstrated the success of using GAC in bioreactors (Martín-Pascual et al., 2012). Using a filling ratio of 50%, this study showed that the maximum removal efficiency of SCOD was 56.97%, 58.92% and 46.13%, under hydraulic retention times (HRTs) of 15,10 and 5h respectively. However, due to its relatively high densities, GAC based reactors require high upflow velocities – as seen in integrated fixed-film activated sludge (IFAS) systems – and high mixing requirements. Modified bio-carrier utilizing GAC are a promising alternative media due to its low cost, rapid and stable attachment of biofilms, and strong tolerances for organic loading.

In this study, a new form of a biofilm carrier IFAS reactor – the bioball with GAC encaged – was developed (named GAC-IFAS) and applied to primary effluent treatment. Each bioball (with a diameter of 16 mm) was modified by filling with 6 cylindrical GACs (each with a diameter of 4 mm and a height of 7 mm).

1.2 Specific Objectives

The application of GAC based bio-carrier in IFAS system is a potentially promising approach to improve the rate of nutrient removal. However, the application of GAC as bio-carrier in the treatment of wastewater in sequencing batch reactor has been rarely reported. Based on the aforementioned research gap, the overall objective of this thesis is to evaluate and compare the long-term treatment performances and stabilities of two integrated fixed-film activated sludge reactors – one using a conventional plastic carrier and the other using a GAC engaged biocarrier. GAC-IFAS and plastic-IFAS reactors were initially incubated within two weeks at an HRT of 12 h. Meanwhile, a long-term performance assessment (i.e. organic matter removal, nutrient degradation rate, and system stability) of both reactors was conducted at HRTs decreasing from 12 to 4 hours, and OLRs increasing from 0.95 to 3.04 g COD/L-d. These experiments were established to provide information and recommendations to GAC enhanced IFAS design.

1.3 Thesis organization

This thesis recorded the performance of GAC-IFAS and plastic-IFAS reactors in treating synthetic primary effluents at different HRTs ranging from 12 to 4 hours. The organization of this thesis is as follows: Chapter 2 provides a literature review on the current status of activated sludge studies on wastewater treatment. The literature review points out the significance of using the IFAS reactor for wastewater treatment. Various applications of carriers in different systems for wastewater treatment are also presented, compared, and discussed in this chapter. Chapter 3 details the reactor configuration, experimental design, analytical methods, and calculations used throughout the thesis. Chapter 4 presents the results and discussions of the experimental work and data. The synthetic primary effluent treatment performance of the GAC-IFAS and plastic-IFAS reactors in the areas of organic matter removal, nutrient degradation rate, biomass concentration, and system stability are presented and explained in this section. Finally, Chapter 5 summarizes the conclusions generated from the long-term operation of the GAC-IFAS and plastic-IFAS reactors. Major performance parameters are pointed out as references for engineering practice. Recommendations for future work are also proposed in this chapter.

Chapter 2

Literature Review

2.1 Significance of primary effluent treatment

Municipal wastewater is becoming a global environmental concern due to increasing populations and is exacerbated by limited water resources and the rapid development of urban areas and industries (Trapani et al., 2010; Rosso et al., 2011). Excess ammonium and carbon compounds discharged into water streams contribute to eutrophication and depletion of dissolved oxygen due to the activity of activated bacteria in water bodies. Therefore, removing such substances from municipal wastewater to reduce their harmful environmental impacts is of great importance (Zinatizadeh et al., 2015).

Current technologies like chemically enhanced treatment (involving coagulation and flocculation), membrane processes, and biological units are used to treat wastewater so they can meet regulations and re-use standards. In particular, membrane bioreactors (MBR) show clear advantages when removing nutrient and micro-pollutants from urban wastewater, and also have simpler construction and decreased energy costs (Haandel et al., 2012). However, three major concerns still exist in these conventional treatment methods including membrane fouling, large tank requirements for sludge settling and biomass recycling, and limited efficiency of nitrogen and phosphorus removal (Qi et al., 2018; Xuan et al., 2020; Wang et al., 2006). In order to avoid biological process shortages and to maximize the recovery of resources, this thesis proposes the implementation of integrated fixed-film activated sludge (IFAS) systems to satisfy the needs of organics and nitrogen removal.

The IFAS process was first introduced as an improvement to moving bed biofilm technology, and incorporates the benefits provided by both attached and suspended growth systems (He et al, 1997; Mannina et al., 2017). IFAS is considered a cutting-edge process, offering advantages over the moving bed biofilm process and conventional activated sludge process such as enhanced nutrient removal, reduced footprint, and longer solids retention time (Wang et al., 2006; Waqas et al., 2020). IFAS has proven effective, for it removed up to 90% combined chemical oxygen demand and ammonia with nutrients from an effluent stream at optimum conditions, provided sufficient retention time (Arias et al., 2018; Yan et al., 2015).

IFAS can be regarded as an aerobic, anoxic, and anaerobic process which allows immersion and movement of carriers. In the IFAS system, biomass can either be attached on the surface of or inside of the small carriers, which have lower density than water and therefore can move freely and continuously in the tank (Kadiya et al., 2012; Regmi et al., 2011). The regulation of biofilm is important for optimizing the treatment efficiency in the system (Yan et al., 2015; Church et al., 2018).

IFAS has been recognized as attractive option for simultaneous nitrification and denitrification (SND) be achieved in one single reactor (Zhu et al., 2015). SND has several advantages: (1) there is no need for two separate tanks, therefore, continuous effluent output can be achieved with a smaller footprint (Hu et al., 2011; Javid et al., 2013); (2) the utilization of carbon is lower, which can subsequently decrease sludge yield by 30% (kermani et al., 2009); (3) it needs less alkalinity as the alkalinity is produced during denitrification, and (4) it consumes less energy because less aeration is required (Calderon et al., 2012).

2.2 Integrated fixed-film activated sludge (IFAS) system operating parameters

In IFAS systems, the critical factors affecting the overall process performance are liquid flow diffusion and nutrient penetration, which is affected by biofilm thickness (Azimi et al., 2007). It can be seen from Fig. 1 that the hydrolysis of solutes and diffusion through the loose biofilm and liquid portion provide the dense biofilm with the main compounds required for vital metabolism and growth. The end products are then released from the dense biofilm into the loosely bound biofilm, and then into the liquid phase (Xavier et al., 2005; Kadiya et al., 2012).

High shear stress generated by mixing and/or aeration outside the hydrodynamic boundary layer promotes: (1) a more compact biofilm with higher density (Li et al., 2016); (2) a more stable and stronger biofilm (Liu and Tay., 2001); (3) a thinner biofilm due to biofilm sloughing.

The critical factors which influence the biofilm characteristics and stability will be discussed in the following sections.

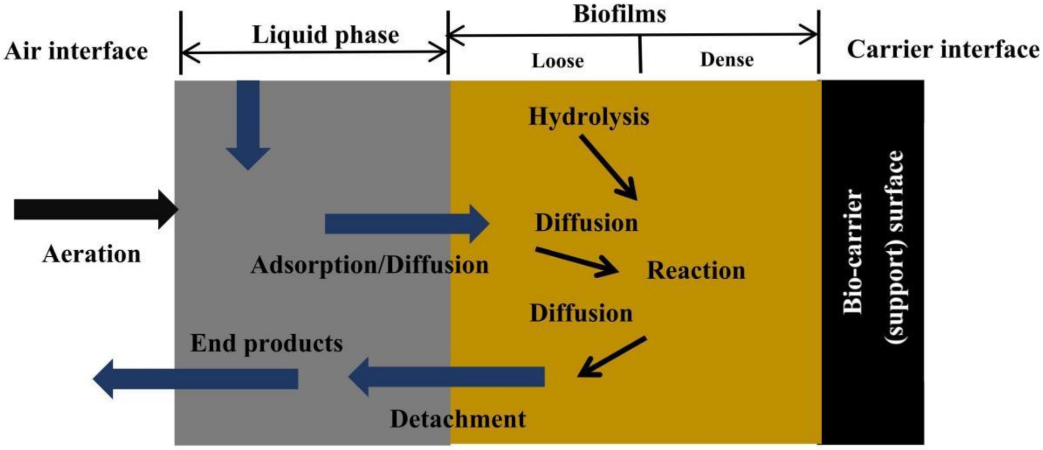


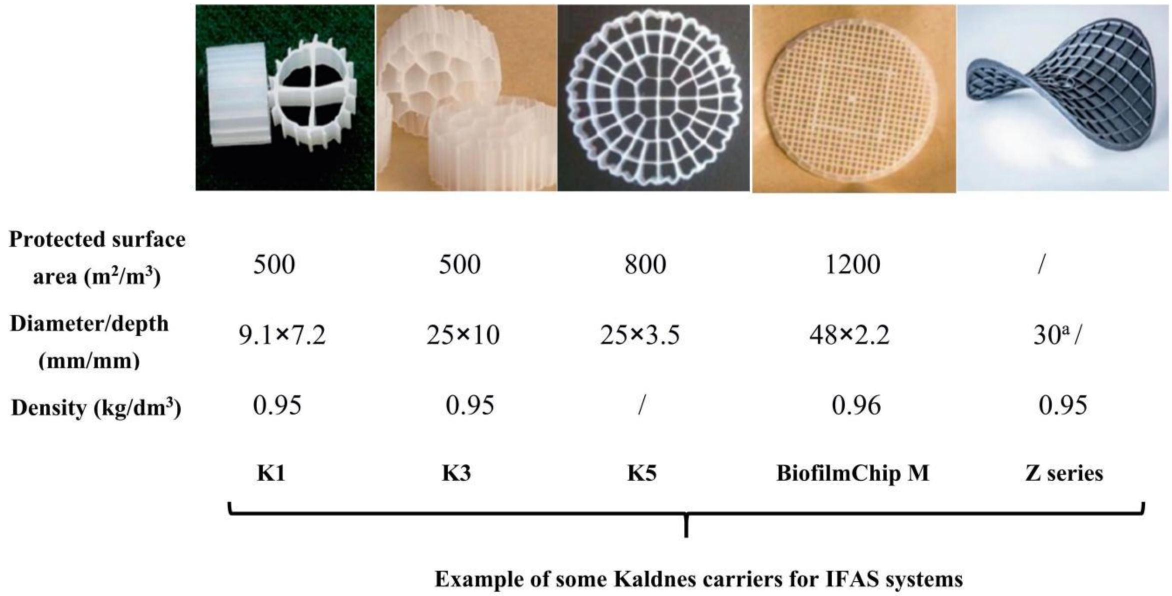
Fig. 1. A schematic of transport and fate of main constituents in the biofilm.

2.2.1 Carrier types

The material of carrier media onto which the biofilm attached is critical and affects the microbial ecosystem. Specific surface area (SSA) is the main factor influencing biomass formation on solid surface, and contributes to nutrient removal in IFAS systems. It is desirable to utilize a carrier with a high internal surface area and with a similar density to water to facilitate free-floating (Leiknes and Ødegaard, 2007).

A variety of materials have been used as carrier media, including activated charcoal, stones, sand, plastic sheets, metals, and foams (Lariyah et al., 2016; Zhang et al., 2016). As an example, the K-series AnoxKaldnes™ carriers, as shown in Fig. 2, are mainly composed of polyethylene and have a density of 0.95 g cm^{-3} (Ødegaard, 1999). They have distinctive differences in dimension (diameter, height and shape), effective surface area for biofilm formation, and transections (i.e. openings) (Paulina et al., 2019).

The impact of carrier type and filling ratio on the efficiency of COD degradation in treated wastewater was estimated by Paulina Śliz. et al. (2019). For example, significant impacts on COD removal were found only at low hydraulic load conditions. The tested model of the bioreactor consisted of five independent chambers with diameter D = 0.14 m and height H = 2.0 m, which were filled with biomass carriers (K1, K2, K3, K4 and K5) at 0%, 20%, 40%, 60%, and 70% of active volume. The highest COD reduction efficiency (70.46–72.59 %) was obtained in reactors packed with K3 and K1 with filling of carriers from 40% to 70%.



	K1	K3	K5	BiofilmChip M	Z series
Protected surface area (m^2/m^3)	500	500	800	1200	/
Diameter/depth (mm/mm)	9.1×7.2	25×10	25×3.5	48×2.2	30^a /
Density (kg/dm^3)	0.95	0.95	/	0.96	0.95

Example of some Kaldnes carriers for IFAS systems

Fig.2. Main characteristics of some AnoxKaldnes™ carriers.

Ye et al. (2010) studied the biofilm performance of high surface area density vertical flow (VF) structured sheet media, demonstrating that VF media combined with proprietary distribution media is capable of achieving complete nitrification (tertiary ammonia and pre-denitrification rates of 1.4 g NH_4^+-N/m^2-day, and 1.0-2.0 g NO_3^--N/m^2-day), as well as high COD removal rates (SCOD removal rate of 30 g SCOD/m^2-day) in IFAS systems. The utilization of this media not only maximized the oxygen and nutrient distribution over the entire surface area of the media (Pham et al., 2008; Tang et al., 2016), but also optimized mixing and biomass control. This ultimately promoted intimate contact between the thin biofilm and substrates/oxygen provided by the aeration associated with dedicated media towers (Waqas et al., 2020).

2.2.2 Packing ratio

The packing ratio is important for controlling microbial activity. It is a function of feed composition and can range from 30 to 70 % (Barwal and Chaudhary, 2014; Rodgers and Zhan, 2003). Several studies have demonstrated that the highest contaminant removal efficiency was achieved with carriers filled in the range of 40 – 60 %. (Kamani et al., 2008; Kadiya et al.,2012). Quanetal et al. (2012) evaluated the effects of the packing ratio (20%, 30%, and 40%) of polyurethane foam (PUF) and HRT (5, 7 h) on the performance of an MBBR treating urban synthetic wastewater. In this study, the packing ratio showed little influence on the COD removal efficiency. Nearly 96.3% ammonium removal efficiency was obtained at an HRT of 5 h with a 40% packing ratio, while only 37.4% removal was realized with a 20% packing ratio.

Packing filling ratio is also associated with the selection of HRT, organic loading rate (OLR) and sludge retention time (SRT). Higher packing ratio lowers the amount of floc and free volume, which eventually lowers the HRT (Wang et al., 2005). IFAS can accomplish high nutrient removals due to higher SRT and higher biofilm amount under a high packing ratio. However, the industrial IFAS operates at a lower packing ratio mainly to decrease the energy input (for aeration) and operational expenses (Lariyah et al., 2016; Martín-Pascual et al., 2015). The filling ratio of the domestic IFAS generally varies between 8 – 60 %, while the industrial IFAS varies between 15 – 60 %. However, Martín-Pascual et al. (2015) showed that using 20% carrier media was enough to achieve high organic removal efficiency.

2.2.3 Hydraulic retention time (HRT)

Hydraulic loading is a critical factor which directly determines reactor HRT and further influences the reactor volume and IFAS efficiency, and therefore must be optimized (Kim et al., 2010). Low strength wastewater generally requires low HRT (3 – 12 h), while longer HRT generally facilitates COD removal, particularly when treating persistent chemicals. Extension of HRT from 6 to 12 h shows only minor improvement in COD removal for low strength wastewaters with influent COD values in the range of 170 – 310 mg/L.

Zhang et al. (2016) operated lab-scale IFAS at HRTs of 6, 8 and 12 h to evaluate the effect of HRT on COD removal for an anaerobic anoxic oxic-biological contact oxidation (AAO-BCO) process. Their results showed that increasing HRT slightly affects COD removal (80–84 %). Nevertheless, ammonium removal decreased by 3% with the increase in hydraulic loading

(0.39–1.56 kg/m^3-d). The minor effect of HRT on COD removal suggested that the AAO-BCO process was quite robust.

Zinatizade et al. (2015) investigated the effects of two numerical variables, both at three levels – 4, 8 and 12 h for HRT and 2, 3 and 4 mg L^{-1} for DO, and the effect of a single categorical variable at two levels (Ring form and Kaldnes-3). Maximum COD removal efficiency was found to be 85% and 88% for the system with Ring form and Kaldnes-3 at HRT of 12 h and DO of 4 mg/L respectively. The maximum denitrification rates for Ring form and Kaldnes-3 were obtained at 90 and 70 mg N/L-d, respectively at DO of 3 mg L^{-1} and HRT of 8 h.

2.2.4 Dissolved oxygen concentration

In MBBR and IFAS systems, aeration provides dissolved oxygen (DO) to the biomass, while also creating a cross-flow velocity to scour the carrier media surface. Wang et al. (2006) recommended that the dissolved oxygen (DO) concentration in the reactor be kept higher than 2 mg L^{-1} for efficient COD removal. They also observed that by decreasing the DO from 2 to 1 mg L^{-1}, COD removal was improved by 13%, indicating that DO was a limited factor. However, COD removal efficiency only increased by 5.8% when DO concentrations were increased from 2 to 6 mg L^{-1}. It was concluded that simultaneous nitrification and denitrification could be achieved in a single bioreactor with a limited DO concentration and a HRT of 6 h.

It was apparent that nitrification depends on DO because DO diffusion through the biofilm was the rate-determining step for media nitrification (Lazarova et al., 1995). The highest

N-removal efficiency (89.1% on average) was obtained when the DO was kept at 2 mg L^{-1} (Liang et al., 2010). At lower DO concentrations (<1 mg L^{-1}), anoxic conditions occurred and ammonia concentration in the effluent became higher.

2.2.5 Mixing conditions

Adequate turbulence is ideal for efficient system performance (Nof et al., 2013). The development of a very thin, evenly distributed, and smooth biofilm on the carrier media is required so that transportation of substrates and oxygen to the biofilm surface is efficient (Leiknes and Ødegaard., 2001). Due to the potential of biocarrier stagnancy in the early stages of biofilm formation, mixing in IFAS systems are challenging (Eslami et al., 2018; Torre et al., 2013). As microbial populations start to attach and develop on the surface area, they become heavier (e.g. greater density than water) and therefore mixing capabilities can be improved (Rusten et al., 2006).

It is not recommended to detach biomass on carriers under high speed mixing conditions. Indeed, biofilm detachment from the outer surface of the carrier can easily result from collision and attrition of bio-carriers in the reactor (Rafiei et al., 2014). It was demonstrated that a superficial air velocity below a threshold of 5 m h^{-1} (Wang et al., 2018) decreased TAN removal in small (<0.8 L) and medium scale (0.8-200 L) reactors. It was reported that TAN removal could be promoted through intense mixing using a vortex mixer at small scale, even at low TAN concentrations (Yuan et al., 2008).

2.2.6 Other factors

Other physico-chemical characteristics of wastewater such as pH, nutrient levels, ionic strength (Marques et al., 2008), and temperature also play an important role in microbial growth.

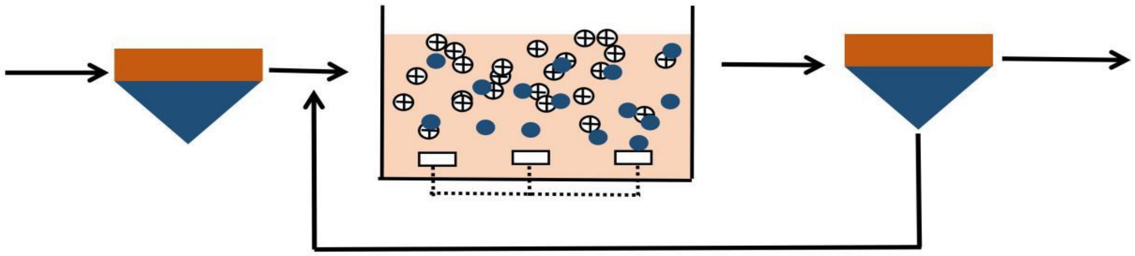
2.3 IFAS reactors application for municipal wastewater treatment

There are currently five main types of integrated fixed-film activated sludge (IFAS) reactor systems (see Fig. 3) used in municipal wastewater treatment: namely the IFAS continuous flow reactor (IFAS-CFR), integrated fixed-film activated sludge reactor in sequencing batch reactor mode (IFAS-SBR), integrated fixed-film activated sludge membrane bioreactor (IFAS-MBR); microalgae-IFAS system (MAIFAS) and up-flow anaerobic sludge blanket-IFAS system (UASB-IFAS). The performances of these various IFAS systems are summarized in Table 1.

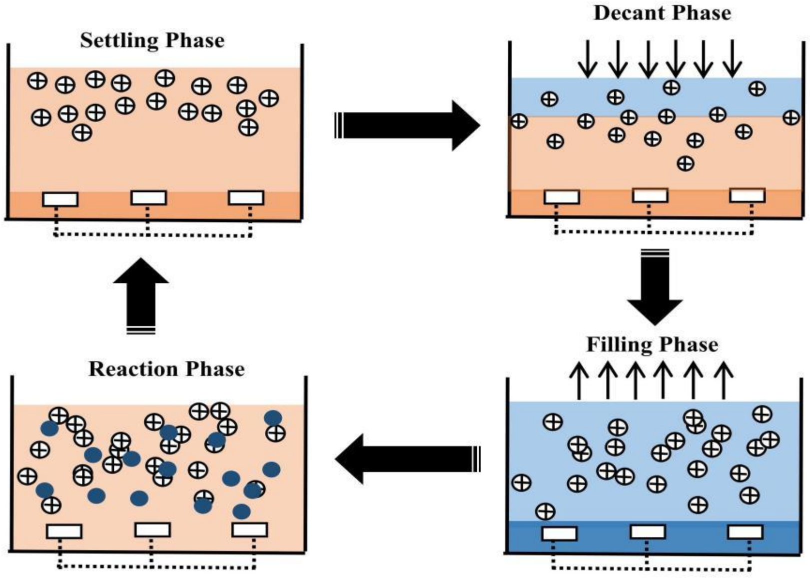
2.3.1 IFAS continuous flow reactor (IFAS-CFR)

An integrated fixed-film activated sludge (IFAS) reactor is a cost-effective solution to upgrade wastewater treatment plants (WWTP), with addition of free-floating carrier media which supports biofilm growth (Kim et al., 2020). Six full-scale IFAS systems were used to study trace organic contaminant (TrOC) removal for municipal wastewater treatment (Brennan et al. 2019). The results showed that all IFAS-WWTPs performed well in terms of ammonia, COD, and TSS reduction. However, removal of total nitrogen (TN) varied with nitrate concentration. TrOC removal was recorded at over 90%, but other forms of TrOC had more variable removal efficiencies. Phanwilai et al. (2020) investigated nitrogen removal efficiency in municipal wastewater using integrated fixed-film activated sludge-continuous flow reactor (IFAS-CFR) with high influent COD:N ratios ranging from 8:1 to 15:1. Removal efficiency of COD and BOD all achieved a high level (96– 98 %), while NH_4^+-N and TN removal efficiencies were less, varying at 72–98 % and 64–77 %, respectively.

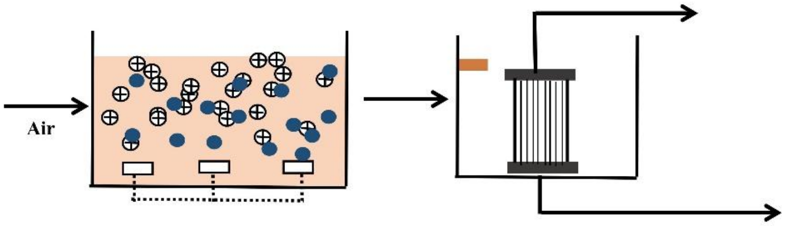
(a) IFAS continuous flow reactor (IFAS-CFR)



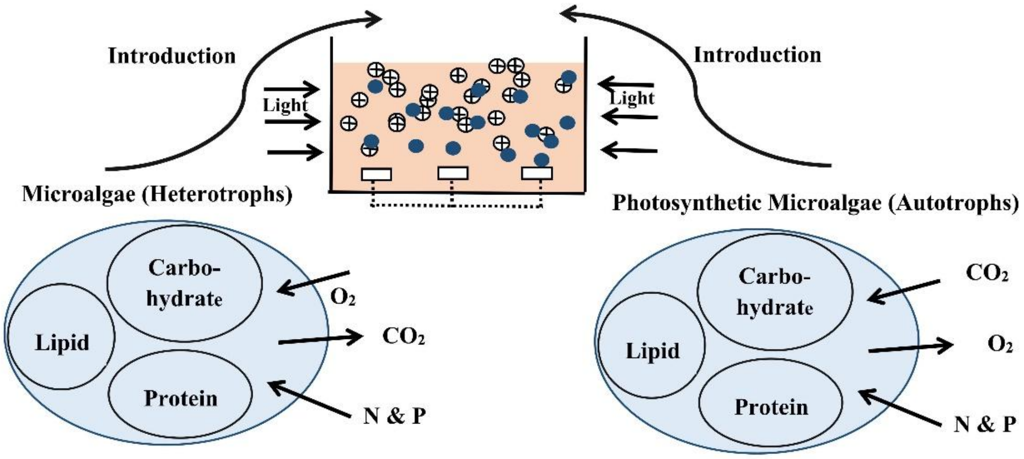
(b) IFAS in sequencing batch reactor mode (IFAS-SBR)



(c) IFAS-MBR



(d) Microalgae-IFAS system (MAIFAS)



(e) Up-flow anaerobic sludge blanket-IFAS system ((UASB-IFAS)

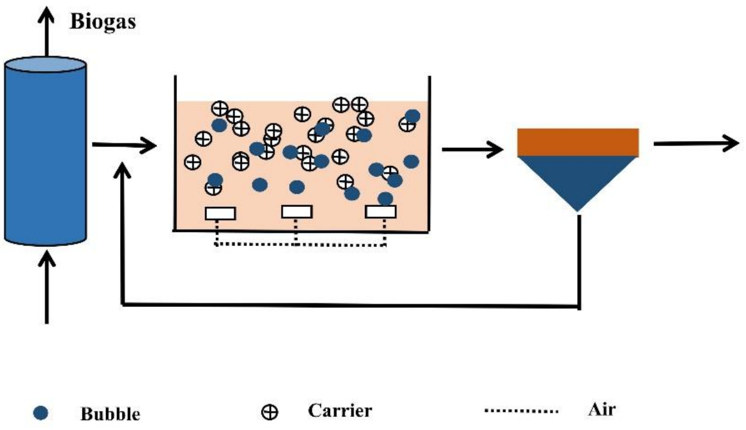


Fig. 3. Schematic diagrams of five applications of IFAS technology studied for nutrient removal in municipal wastewater:(a) IFAS continuous flow reactor (IFAS-CFR) ;(b) IFAS in SBR mode (IFAS-SBR); (c) IFAS-MBR reactor; (d) Microalgae-IFAS system (MAIFAS); (e) Up-flow anaerobic sludge blanket-IFAS system ((UASB-IFAS).

2.3.2 IFAS-SBR reactor

The concept of one stage integrated fixed-film activated sludge (IFAS) sequencing batch reactors for lagoon supernatant nitritation–anammox treatment has been developed using significant packing ratios of carrier media ranging between 20-42 % and HRT variations of 1.2 to 2.5 days (Yang et al., 2019). It was shown that NH_4^+-N and TN removal efficiencies increased from 81.8% and 74.8% to 95.2 % and 87.4 % respectively, with increased biocarrier filling ratio (from 35% to 55%). However, removal efficiencies dropped from 91.8% and 82.1% to 89.6% and 78.8% when HRT was lowered from 1.5 d to 1.2 d, respectively.

Zhou et al. (2019) addressed the effects of silver nanoparticles (AgNPs) on reactor performance and microbial community analysis in integrated fixed-film activated sludge-sequencing batch reactors (IFAS-SBRs). A minor effect of AgNPs addition on nutrient removal was observed, achieving overall removals of 99.9% ammonia, 98.8% phosphorus and 96.6% COD. However, the microbial community structure was revealed to be different on the phylum level following long-period exposures to silver nanoparticles, whereas the important functional genera remained at high populations.

Recently, concerns have been raised regarding the CAS process due to its high energy consumption and waste sludge production, which leads to issues of environmental sustainability. To tackle the challenging situation, Gu et al. (2017) developed a novel A-B process. The A-stage was an anaerobic moving bed biofilm reactor (AMBBR) used for COD capture, and the B-stage was an integrated fixed-biofilm and activated sludge sequencing batch reactor (IFAS-SBR) used for biological nitrogen removal. Results showed that 85% COD removal in synthetic wastewater was achieved at steady-state AMBBR with an energy production rate of 0.28 kWh/m^3 wastewater treated, while 85% of nitrogen was removed in the IFAS-SBR. Compared to the CAS process, the waste sludge production was reduced by about 75% in the proposed A-B process due to the efficient COD capture at the A-stage, leading to significant energy savings because energy required for aeration of COD was reduced.

2.3.3 IFAS-MBR

Conventional membrane bioreactors (MBR) are based on the separation of activated sludge with biomass by applying ultrafiltration membranes with pore sizes of ~40 μm for mixed liquor suspended solids (MLSS) ranging from 7 to 10 g MLSS m^{-3}, and membrane fluxes of around 20 L m^{-2} h^{-1} (Mannina, 2017). The combination of integrated fixed-film activated sludge (IFAS) with MBR in series (MBBR-MBR) is a recent advance for improving MBR performance due to its potential to reduce membrane fouling, to prolong filtration duration, and to promote microbial degradation of certain organic compounds (Luo et al., 2015; Vergine et al., 2018). Previous research by Mannina et al. (2020) also concluded that changes in the C/N ratio (within 2-10) had positive effects on removal rates in a lab-scale IFAS-MBR, while also achieving high nitrification under low SRT.

Wang et al. (2021) employed a novel IFAS-MBR with low aeration for the treatment of real municipal wastewater. The removal rate of COD, total nitrogen, and phosphorus were 95.3±1.3 %, 78.1±7.2 % and 93.7±5.8 %, respectively. Moreover, concentrations of COD decreased from 31.9±3.7 (sludge supernatant) to 12.7±1.6 mg/L (permeate) after membrane filtration. This illustrates that the novel IFAS-MBR system provides an energy-efficient alternative due to its highly efficient performance and low operating costs, enabled by less aeration requirements and absence of external carbon sources.

A good balance in organics and nutrients removal also has been shown by IFAS-MBR. Mannina et al. (2017) examined organic removal ability using a pilot-scale hybrid domestic IFAS-MBR treating synthetic wastewater characterized with a COD of 607 mg/L, total nitrogen (TN) of 65 mg/L, total phosphorous (TP) of 11 mg/L, and COD/TN/TP ratio of 100/10.7/1.8. The plant was operated with AnoxKaldnes K1 carrier media (filling ratio of 15-40 %) under the condition of HRT of 20 h and DO level of 5.33 mg/L. Maximum removals of COD (98%), ammonia (98%) and TP (40.4%) were observed in this study despite feed composition variations. The biofilm contributed to 98% of nitrification and almost complete conversion of ammonium.

2.3.4 Microalgae-IFAS system (MAIFAS)

The novel symbiotic microalgae-based IFAS (MAIFAS) technology has been introduced as a promising strategy for nutrient removal and energy consumption reduction. Microalgae-IFAS (MAIFAS), a new type of hybridization process, integrating microalgae and bacteria for nutrient assimilation and organics oxidation. This developed MAIFAS process can improve existing IFAS process by uncoupling photo-aeration between suspended solids for P removal and biofilms for nitrification, offering low energy input and higher effluent quality (Church et al., 2018).

Church et al.(2008) investigated the removal of N and P from synthetic wastewater in a novel MAIFAS sequencing batch reactor over 150 days. The result showed MAIFAS could achieve greater than 99% ammonium and 51% phosphorous removals, and the removal rates were much higher than the conventional microalgae photobioreactor which typically removed 57% ammonium and 49% phosphorous (Bilad et al., 2014). Moreover, MAIFAS biofilm analysis indicated an abundance of AOB (1.5% Nitrosomonadaceae) compared to NOB (0.2% Nitrospira). Moreover, the addition of microalgae to the IFAS systemcan could produce biomass for other purposes, promoting significant changes in the bacterial community structure and its metabolic activity.

2.3.5 Up-flow anaerobic sludge blanket-IFAS system (UASB-IFAS)

An innovative process based on the incorporation of a UASB reactor and an IFAS system is proposed in order to promote high microbial diversity and organic micropollutants (OMPs) removals. Arias et al. (2018) conducted IFAS-UASB and achieved high removals of COD (93%) and TN (44%), as well as up to 85% methane yield by heterotrophic denitrifiers and aerobic methanotrophs. A high removal of micropollutants was obtained in this hybrid system (> 80%), illustrating UASB-IFAS system could enhance the biological removal of OMPs as well as to achieve the abatement of nitrogen by using the dissolved methane as an inexpensive electron donor.

Dohdoh et al.(2021) evaluated the effect of carrier-filling media on the performance of a classical integrated UASB-AS system and hybrid UASB-IFAS modified system. The comparison between the conventional UASB-AS integrated system and the hybrid UASB-IFAS modified reactors demonstrated that both systems have comparably high efficiencies in organic matter removal (>95%). It's indicated that the carrier-filling media improved the treatment efficiency significantly. An additional advantage of carrier-filling media on UASB-IFAS system was its high stability when changing the hydraulic loading. For both examined systems, the results indicated that most of the organic matter was removed in the anaerobic reactors, and little additional removal occurred in the aerobic units. As for the ammonium, residuals values all met effluent standards for both systems at all HRTs except 3 h. Also, the results showed that nitrogen removal was very low or absent in the anaerobic units.

Table 1. Overview of IFAS performances on municipal wastewater treatment

IFAS Process	HRT / Carrier media	Treatment performances	References
Lab-scale IFAS-SBR	6.5 h / cubic sponge	COD removal 85% TKN removal 85%	Gu et al. (2017)
Pilot-scale IFAS-MBR	20 h / AK, K1	COD removal 98% Ammonium removal 98% Total P-removal 40.4%	Mannina et al., 2017
Lab-scale IFAS-SBR	12 h / Ak, K1	COD removal 95% Ammonium removal 57% Total P-removal 49%	Church et al., 2018
Lab-scale IFAS-SFD-MBR	8 h / PE carrier	COD removal > 90% Ammonium removal 95% TKN removal 42%	Vergine et al., 2018
Pilot scale	4.5-8.1 h / BMX1	TCOD removal > 90% Ammonium removal > 99% TKN removal > 90%	Moretti et al., 2015
Full scale	NA / AK, K3	Ammonium removal > 90% TKN removal 75.1%	Regmi et al., 2011
Lab-scale IFAS-SBR	8 h / synthetic with silver nanoparticle	COD removal 96.8% Ammonium removal 99.3% PO_4^{3-}- P removal 98.5%	Zhou et al., 2019
Pilot scale hybrid IFAS	3.47 h /AK, K1	Ammonium removal 81.19% COD removal 100%	Di Trapani et al., 2013
Lab-scale IFAS-SBR	1.5 d / AK. K5	Ammonium removal 91.8% TKN removal 82.1%	Yang et al., 2019

2.4 Summary

To date, only a limited number of researchers have evaluated the application of modified bio-carrier IFAS systems for municipal wastewater treatment.

The objective of the IFAS system is to achieve biomass growth as a biofilm on small moving carriers in the aeration tank during operation (Delnavaz et al., 2010). Factors such as temperature, pH, dissolved oxygen, and nutrition concentration, as well as carrier type and surface characteristics influence bioreactor performance. The properties of biofilms have been widely investigated (Ansari et al., 2012; Barwal and Chaudhary, 2014; Bassin et al., 2012). It is necessary to investigate the biofilm variations and nutrient removal rates at each stage to understand its formation and development.

Granular activated carbon (GAC) is a promising bio-carrier material due to its adsorption capacity and capability to support biofilm formation. In addition, the existence of pores in GAC means that it contains a larger surface area when compared with other common plastic carriers, so it can provide more sites both inside or on its surface for the adsorption and growth of microorganisms. More significant information on BGAC application and its removal mechanisms in IFAS system is provided throughout this study, which will help people understand it and enable them to utilize it more efficiently in future research.

Chapter 3

Methodology

3.1 Reactor configuration

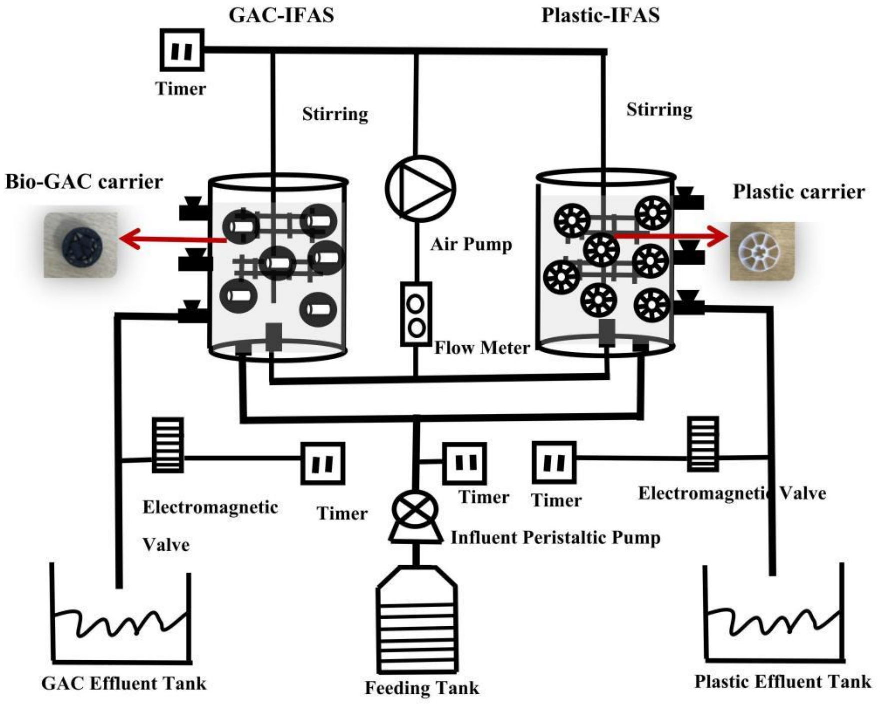


Fig. 4. Experimental set up of lab-scale GAC-IFAS and Plastic-IFAS reactors.

A lab-scale integrated fixed-film activated sludge reactor (IFAS) system was conducted in cylindrical tank with working volume of 0.875 L. The experiment was operated as sequencing batch mode by the control of timer and electromagnetic valve, as shown in Fig. 4. About 82 pieces of GAC engaged bioball, and 115 pieces of conventional plastic carrier were filled into two reactors, respectively. The utilization of modified stirring system (bared wire) promote carriers mixing well in reactor, thus corrosion and flotation.

3.2 Inoculation and enrichment

The GAC-IFAS and plastic-IFAS reactors were initially carried at HRT of 12h, with the organic loading rate (OLR) of 0.97 g COD/L-d, representing start-up phase. Within this stage, microbial culture and biofilm formation on carrier was done by inoculation with 400mL aerobic activated sludge from local WWTP in Edmonton, which had a total suspend solids (TSS) and volatile suspended solids (VSS) of 2.65 ± 0.19 g/L and 1.98 ± 0.21 g/L, respectively.

For enrichment of functional biofilm, each reactor was fed with about 1350mL synthetic municipal wastewater in sequencing batch mode within first two weeks. The composition and characteristics of the synthetic wastewater were provided in Table 2 and Table 3, respectively. The reactor was purged air (0.9±0.15 L/min) and incubated at room temperature (20±0.5 ℃).

3.3 Experimental set up

3.3.1 Synthetic primary effluent collection and characterization

All Numerous synthetic formulations have been made to replicate wastewater. Majority of them tailored towards providing suitable substrates for biological activity (O’Flaherty and Gray., 2013). Hence, the IFAS reactor was fed with synthetic mediums utilized by previous study (Nopens et al., 2001, Ho et al., 2010), with addition of 5 mL inactive primary sludge to adjust the particulate COD to actually wastewater. The stock was prepared as a 30 times concentrated liquid, stored at 4 ℃ in fridge within one week and diluted with deionized water before using.

Table 2. Chemical composition of synthetic primary effluent

Component	Unit	Concentration (Value/L)
Soluble starch	mg	130
$NaCH_3COO$	mg	80
yeast extract	mg	20
NH_4Cl	mg	76
Urea	mg	43
KH_2PO_4	mg	44
$KHCO_3$	mg	600
Milk powder	mg	150
Ferrous chloride ($FeCI_2$ $4H_20$)	mg	3.58
Zinc chloride ($ZnCI_2$)	mg	0.18
Nickel chloride ($NiCI_2$ $6H_20$)	mg	0.17
Cobalt chloride ($CoCI_2$ $6H_20$)	mg	0.20
Manganese chloride ($MnCl_2$ $4H_2O$)	mg	0.19
Primary sludge	mL/L	5

Table 3. Characteristics of synthetic primary effluent

Parameters	Unit	Number
Total Solids (TS)	mg/L	1130.6
Total COD (TCOD)	mg/L	511.5
Soluble COD (SCOD)	mg/L	238
NH_4^+- N	mg/L	35.5
DO	mg/L	2.20
PO_4-P	mg/L	13.1
Total Nitrogen (TN)	mg/L	59.4
pH	/	7.26

3.3.2 Carrier type

In this research two types of carrier were used to study the influence of carrier type on wastewater treatment. These carriers are basically different in material, shape and size. One kind of carrier is a cylindrical high-density polyethylene ring (specific surface area of 450 m^2/m^3) (diameter of 20 mm and height of 10 mm, respectively) with a inner cross-shaped cut out. The other kind of carrier is a promising alternative, named granular activated carbon (GAC) carrier, which can be modified by removing porous sponge inside spherical bio-ball (diameter of 16 mm) and filling in 6 cylindrical granular activated carbons (specific surface area > 1000 m^2/g) (diameter of 4 mm and height of 7 mm, respectively) in each bio-ball. In order to keep carrier suspension and higher nutrient removal, packing ratio of bio-carrier in this study was 18% ($V_{support}$ / $V_{reactor}$).

3.3.3 Reactor set up and operation

To ensure the adequate air diffusion and homogeneity of mixed liquor, as well as substrate transportation in reactors, aeration and mixing systems were necessary. Generally, dissolved oxygen concentration is a critical parameter for carbon and nitrogen removal, and several studies has pointed that the minimum DO concentration for organics removal should be more than 2 mg/L.

The GAC-IFAS and plastic-IFAS reactors were operated in sequencing-batch mode at 21±1 ℃ as follows: for instance, at HRT of 6 h, the time cycle of 3 h for whole process including influent feeding of 5 min, mixing time of 2.5 h, settling time of 20 min and effluent discharging of 5 min, so as to keep exchange ratio of 50%. The stirrer and aeration were stopped during settling period. From day 0 to 32 (start up, stage I), the reactors were operated at HRT of 12 hours with an average OLR of 0.97 g COD/L-d. From day 32 to 64 (stage II), day 64 to 92 (stage III) and day 92 to 120 (stage IV), the OLR was increased in step-wise to 1.52, 2.05 and 3.04 g COD/L-d, respectively, by reducing the HRT to 8 h, 6 h and 4 h. The mixing speed was kept at 115 rpm for both reactors with aeration rate of 0.9±0.15 L/min. The DO concentration and pH were maintained at 1.71± 0.56 mg/L and 6.5-8.0, respectively.

3.4 Batch experiment and activity assessment

Batch tests were performed to measure the specific activity of ammonium oxidizing bacteria (AOB) and heterotrophic bacteria and nitrogen conversion contribution in attached biofilm and suspended flocs under steady state. The batch test contained seven parallel conditions: control (no attached biofilm and suspended flocs); only attached biofilm (5 carriers from GAC-IFAS and plastic-IFAS reactors, respectively); only suspended flocs (30 mL mixed liquor from GAC-IFAS and plastic-IFAS reactors, respectively); both attached biofilm and suspended flocs (5 carriers and 30 mL mixed liquor from GAC-IFAS and plastic-IFAS reactors, respectively). The substrate contained 250 mg L^{-1} COD (as soluble chemical oxygen demand) and 35 mg L^{-1} NH_4^+-N (as ammonium chloride).

All samples were collected at the end of each operation stage, and carriers were washed by phosphate buffered saline (PBS) solution three times before activity test. Assays were operated in triplicate in 150 mL bottles in a shaker (200 rpm) at room temperature (20-22 °C). pH and air flow rate were maintained at 6.68-7.42 and 0.8-1.0 L min^{-1}, respectively. Collected samples were passed through 0.45 μm filters and measured for NH_4^+-N, SCOD concentrations. The specific activity of ammonium oxidizing bacteria (AOB) and heterotrophic bacteria were determined by the maximum substrate removal rate divided by concentration of volatile suspended solid (VSS) in biofilm and suspended flocs, respectively. Therefore the nitrogen conversion contribution were estimated by the maximum removal in either biofilm or flocs divided by that in both biofilm and flocs.

3.5 Batch kinetic experiment

The preliminary batch experiments were carried out to determinate the effects of contact time and contribution of sorption and biodegradation on the efficiency of pollutant removal (in terms of SCOD). The experiment was conducted on three groups of batch reactors containing two types of media: BGAC， InBGAC， as well as control group with no media (Sharaf et al., 2021). The BGAC group was defined as the biologically activated carbon, supporting combined sorption of GAC and biofilm and biodegradation. While InBGAC was defined as inhibited BAC, activing sorption, including GAC adsorption and biofilm absorption with 0.1% sodium azide PBS solution addition. In particular, appropriate amount of BGAC carrier (22.7 g/L) taken from reactor (at stage III) was added to 100 mL of synthetic primary effluent. The experiments were performed for triplicates at 21 ± 1 °C on a horizontal shaker (120 rpm) within 24 h. Supernatant samples were collected at time intervals of 0, 50, 100, 200, 300, 400, 500, 800, 1200, 1440 min and analysis for the soluble chemical oxygen demand (SCOD) (Guistra et al., 2017). Moreover, before carrying the adsorption test, selected granular activated carbons were washed by deionized water and then let it dry.

3.6 Cycle test for nutrient removal

Cycle tests were performed to evaluate the degradation rate of carbon and nitrogen in GAC-IFAS and plastic-IFAS reactors during each experimental stages. All samples were collected from the feeding of each cycle, then collect samples with 30 min interval, and the number of samples depending on the HRT variation. Assays were operated in triplicate at room temperature (20-22 °C). Moreover, pH and air flow rate were maintained at 6.5-7.5 and 0.8-1.0 L min^{-1}, respectively. Collected samples were all passed through the 0.45 μm filters and measured for NH_4^+-N, soluble COD concentrations.

3.7 Chemical and statistical analysis

Water quality parameters, including total chemical oxygen demand (TCOD), soluble oxygen demand (SCOD), ammonium (NH_4^+-N), total nitrogen (TN), phosphate (PO_4^{3-}-P) and alkalinity concentrations, were measured with Hach reagent kits (Hach Company, Loveland, Colorado) after the liquid samples were filtered by 0.45 μm filters (fisherSci, Canada). Dissolved oxygen concentration and pH value were determined by DO meter and pH meter (Mettler Toledo,USA), respectively. TSS and VSS were determined according to a standard method described in Federation and Association (2005). Moreover, all effluent samples were taken once two days and kept at 4℃ in fridge. Statistical analysis was performed using one-way ANOVA model, and correlations were considered statistically significant when P-value < 0.05.

The amount of SCOD sorped per amount onto adsorbent at each time (q_t; mg g^{-1}), was calculated using the following formula:

$$q_t = \frac{(C_0 - C_t)\,V_L}{M_A} \qquad \text{Eq. (1)}$$

Where C_0 is the initial concentration of SCOD in synthetic primary effluent (mg L^{-1}); C_t is the SCOD concentration of substrate at time t (mg L^{-1}); and V_L is the liquid volume (L); M_A is the mass of adsorbent (g). The removal efficiency ($Removal\%$) of SCOD from synthetic primary effluent was calculated using the following equation:

$$Removal\,\% = \frac{(C_0 - C_t) * 100}{C_0} \qquad \text{Eq. (2)}$$

Different mathematical expressions based on adsorption kinetics study of SCOD mainly include the law of pseudo-first order (PFO) and pseudo-second order (PSO) according to Safwat et al.(2018). (As shown in Table 4). The slopes and intercepts of the linear trend-lines were plotted and then estimated R2 and q_e values.

Table 4. Mathematical expressions used to study the adsorption kinetics of SCOD

Model	Mathematical formula	Parameter	
		Definition	Unit
Pseudo-first order kinetics	$q_t = q_e\,(1 - e^{-k_1 t})$	q_t: the amount of sorped SCOD per mass of adsorbent at any time	mg g^{-1}
		q_e: the amount of sorped adsorbate per mass of adsorbent at any time	mg g^{-1}
		k_1: pseudo-first order rate constant	h^{-1}
		t: time	h
Pseudo-second order kinetics	$q_t = q_e \frac{q_e k_2 t}{1 + q_e k_2 t}$	k_2: pseudo-second order rate constant	mg g^{-1} h^{-1}

Chapter 4

Results and discussions

4.1 Organics removal during operational stages

The GAC-IFAS and plastic-IFAS reactors were initially carried at hydraulic retention time (HRT) of 12 h, representing start-up phase. The averaged total COD concentration of influent varied at 504.8 ± 23.3, 510.5 ± 17.7, 505.6 ± 28.1 mg L^{-1} for Stage II to IV during operation, respectively. Ammonia concentration varied at 33.5 ± 2.4, 35.8 ± 4.1, 35.1 ± 1.8 mg L^{-1} for Stage II to IV, respectively. Moreover, DO concentration was determined similarly about 2 mg/L, which provide realistic aerobic/anoxic condition for both organic and nutrient removal in single reactor (Joana et al., 2018) (Table 5).

Table 5. Test conditions of IFAS reactors during different operational stages

Stage	Influent COD (mg L^{-1})	Influent NH_4^+-N (mg L^{-1})	Volumetric organic loading rate OLR (kg COD m^{-3} d^{-1})	Volumetric ammonia loading rate (kg NH_4^+-N m^{-3} d^{-1})	DO (mg L^{-1})	HRT (h^{-1})	Day (d^{-1})
I	554.6 ± 27.1	36.2 ± 3.4	1.10 ± 0.054	0.072 ± 0.007	3.1 ± 0.5	12	0-32
II	504.8 ± 23.3	33.5 ± 2.4	1. 52 ± 0.070	0.101± 0.007	2.3 ± 0.2	8	32-64
III	510.5 ± 17.7	35.8 ± 4.1	2.05 ± 0.071	0.144 ± 0.016	2.2 ± 0.2	6	64-92
IV	505.6 ± 28.1	35.1 ± 1.8	3.04 ± 0.169	0.211 ± 0.011	1.8 ± 0.3	4	92-120

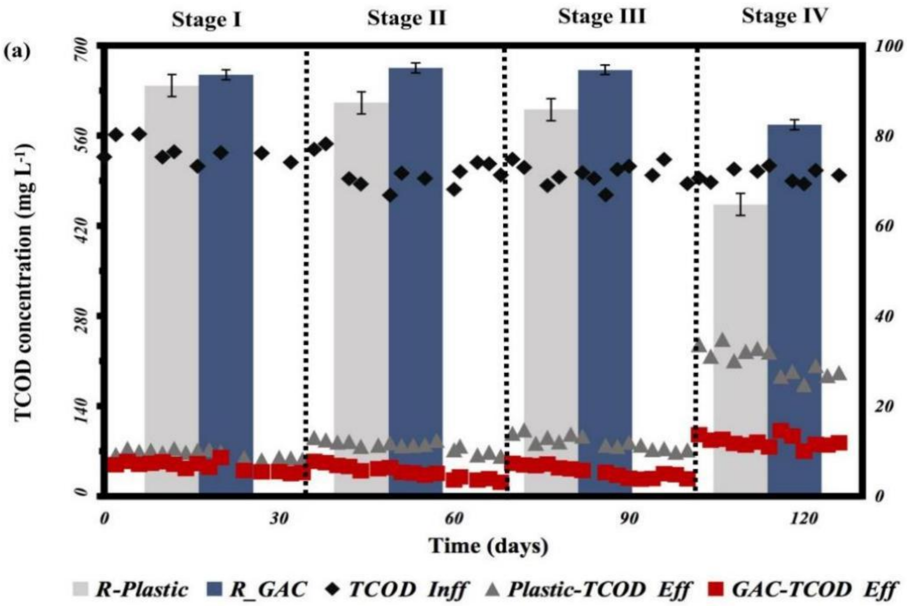
[a] Run I and II-IV are the start-up stage and experimental stage for IFAS reactors.

During start up phase (stage I), it was indicated that no obvious difference in TCOD removal between both IFAS reactors, due to biofilm acclimatization at first. The averaged TCOD removal efficiency was 93.5% and 91.1% for GAC-IFAS and plastic-IFAS reactors, respectively, suggesting both reactors could perform well under the low organic loading conditions. As seen in Fig.5 , averaged removal of TCOD and SCOD in GAC-IFAS reactor were 95.02% and 87.9%, respectively, in Stage II. After HRT was reduced from 8 h to 6 h, the TCOD removal efficiency was remained in GAC-IFAS reactor, while slightly reduced to 85.8% ($p>0.05$) in Plastic-IFAS reactor,. In Stage IV, the averaged removal efficiency of SCOD significantly reduced in both reactors, and which was 77.9% and 52.5% ($p<0.05$), respectively, for GAC-IFAS and plastic-IFAS reactors.

Therefore, the optimal HRT for organic treatment in both reactors was suggested to be 6 h, with an organic loading rate of 2.05 ± 0.071 kg COD m^{-3} d^{-1}. However, GAC-IFAS reactor showed better treatment performance due to relatively more stable and abundant heterotrophs biofilm formation. The reason for different treatment results may attribute to the diverse microbial community and biomass concentration developed on GAC and plastic carriers and in suspended flocs of two reactors (Bradley et al., 2016).

Based on studying the variation between organic treatment rate and HRT, cycle tests under each stage were performed. According to Fig. 5(c), the SCOD removal in both IFAS reactors was fast within 5.5 ± 0.34 h at different HRT, and then followed by steady state. The averaged SCOD removal rates for GAC-IFAS reactor (37.3 ± 3.5, 46.9 ± 5.2 mg L^{-1} h^{-1} for stage II and III, respectively) were higher than those for plastic-IFAS reactor (28.4 ± 4.0, 33.1± 2.5 mg L^{-1} h^{-} for stage II and III, respectively) ($p<0.05$). Nevertheless, The settling ability of the biomass leaving reactor decreases with increasing organic loading. It's noticed that GAC-IFAS reactor had obvious advantage for organic treatment with loading variation.

In this context, significant difference on carbon removal was observed under experimental conditions employed. It's inferred to consider that result was possibly related to the distinct biomass distribution pattern over different shape, material, size and surface characteristics. For GAC, the nutrients would be adhered inside and outside on supports providing more active sites, hence greatly leading to microorganisms growth. Under such conditions, the free passage of nutrients and oxygen through attached biomass layer was restricted, thus contributing to the thick biomass formation (Huang et al., 2017).



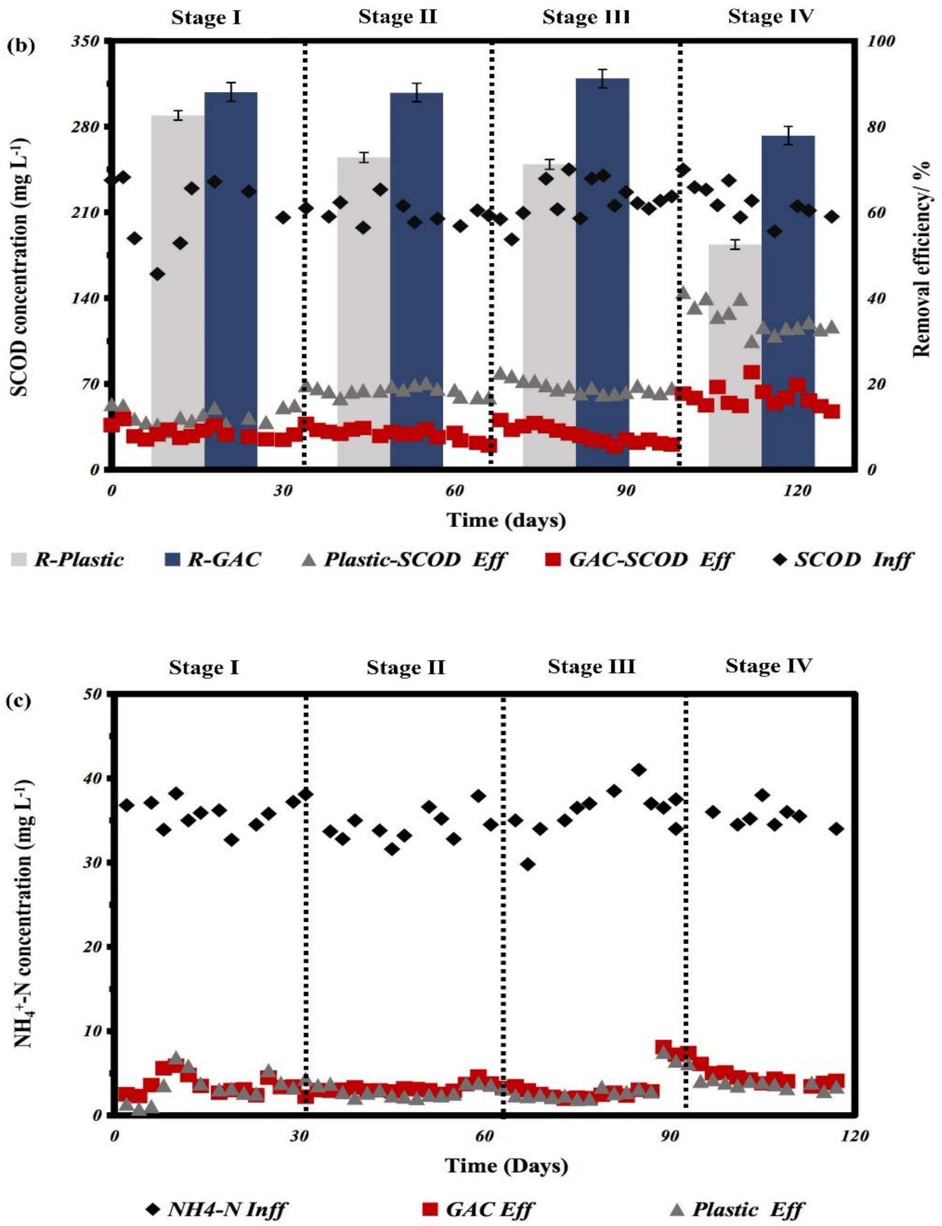


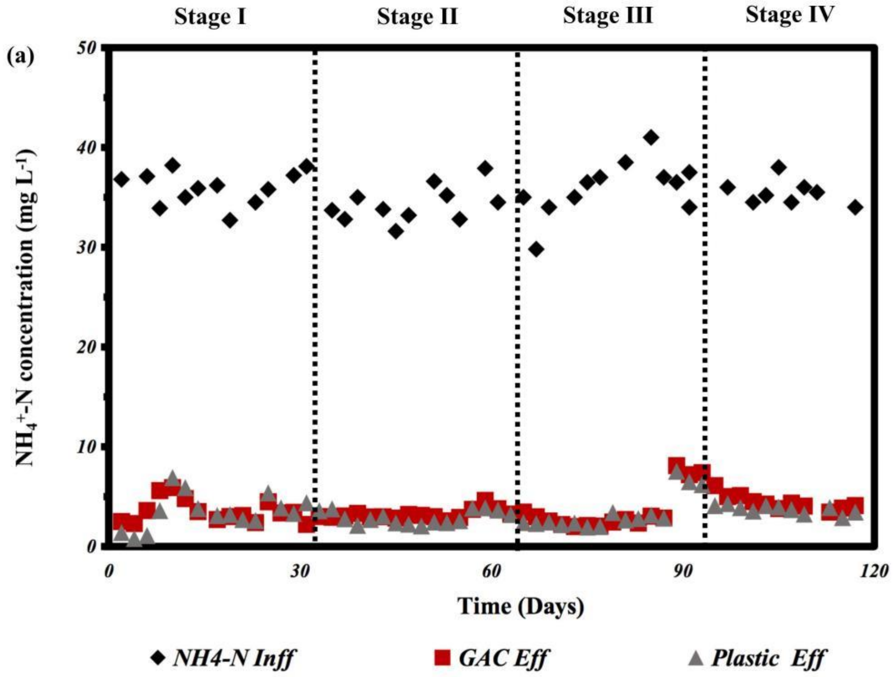
Fig. 5. Carbon removal performance during four reactor operation phases treating synthetic primary effluent (PE) in IFAS reactors. The influent and effluent water quality along time. Total chemical oxygen demand (a) and Soluble chemical oxygen demand (b) concentrations in the reactor feed in different phases, as well as their removal efficiency (c) cycle test of SCOD degradation. Each phase is divided by dash lines, and Error bars represent standard deviations from different samples.(the same for the following figures)

4.2 Nitrogen removal performance

Throughout first 2 weeks of operation, the NH_4^+-N removal efficiency for both IFAS reactors were lower, with the value of nearly 77.6%. While the ammonium removal in stage II for GAC-IFAS and plastic-IFAS reactors averaged at 93.51 ± 3.97 % and 93.16 ± 4.45 %, respectively, showing GAC carriers could result in better NH_4^+-N removal ($p>0.05$). According to Fig. 6(c), the effluent concentration of ammonium in both reactors were all below 3 mg L^{-1} in stage IV, achieving over 85% reduction. Ammonium removal rate in stage III and IV were 6.18 ± 0.68 , 7.40 ± 1.02 NH_4^+-N mg L^{-1} h^{-1} for GAC-IFAS reactor and 5.91 ± 0.93 , 7.51 ± 0.64 NH_4^+-N mg L^{-1} h^{-1} for plastic-IFAS reactor, respectively ($p=0.06$, Fig.6.(c)). The possible explanation for similar response of ammonium removal to HRT variation could be the adsorption of COD onto GAC, increasing the localized COD concentration, which is the fundamental mechanism for GAC impacts.

Nevertheless this adsorption could contribute to improve treatment performance of COD, while for ammonium, no adequate adsorption sites supported, so that the impacts were not significant. In spite of lower HRT obtained in stage IV, higher removal rate was found for both IFAS reactors, demonstrating good performance on nitrogen removal under lower HRT. It was clear that the nitrogen reduction was relatively lower in start-up phase (stage I, 28.6 ± 2.59 %), indicating heterotrophs bacteria competed with nitrifying bacteria (including ammonia oxidizing bacteria and nitrite oxidizing bacteria) in the presence of relatively high concentration of organics in the third cell, which slowing nitrifier biofilm formation (Joana et al., 2018).

As shown in Fig. 6(b), total nitrogen removal all remained lower than 30 % for both reactors during start-up phase (stage I). This is due to that the oxic and the anoxic micro-zones could be formed at the outer layer and the inner layer of the biofilm, which was ascribed to DO concentration gradient within the biofilm of media owing to limited oxygen diffusion (Chu et al., 2011). Therefore, it took time for developing biofilm, especially the anoxic inner layer of biofim, and then denitrification process could be activated.



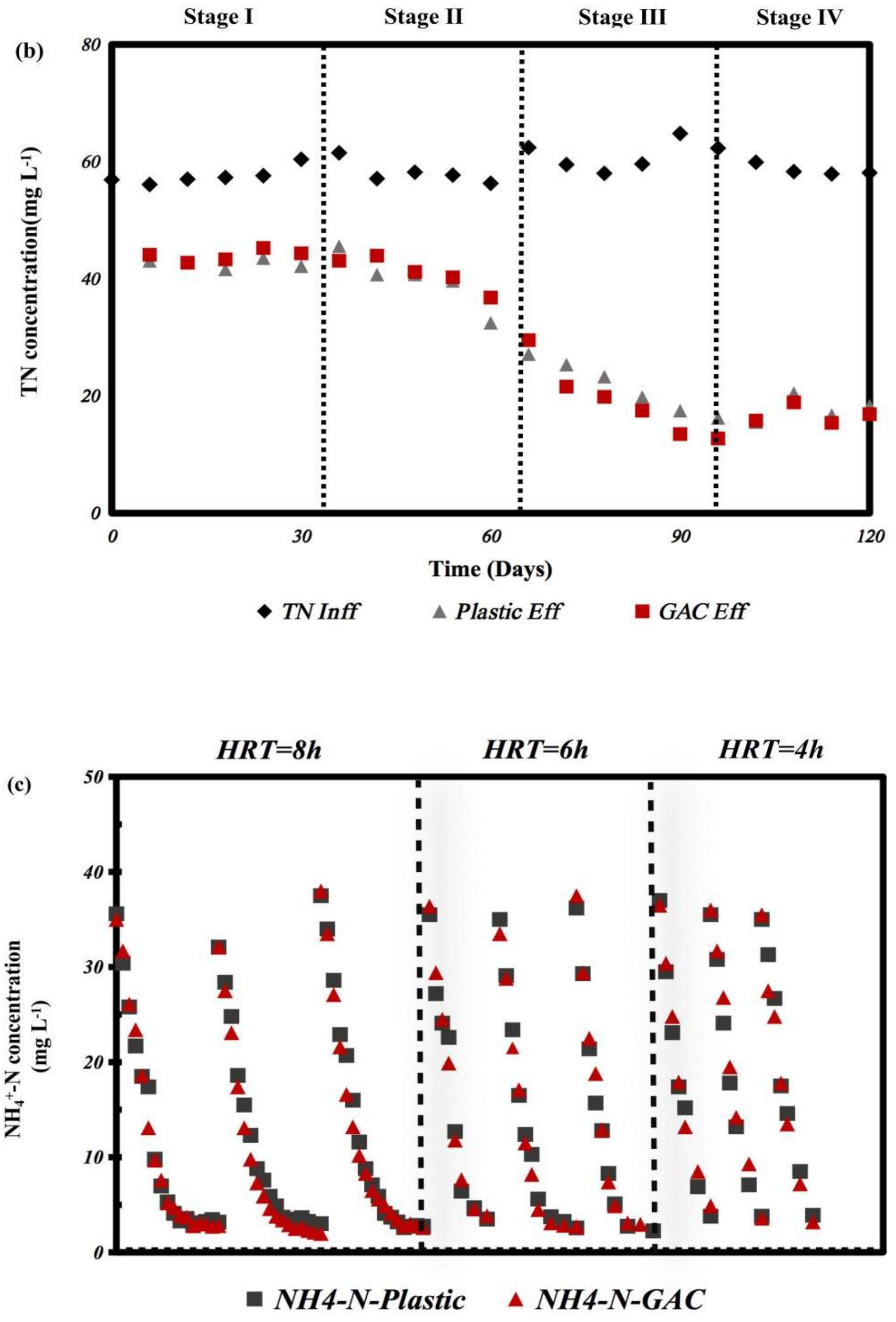


Fig. 6. Dynamics of (a) NH_4^+-N, (b) TN and (c) cycle test of ammonium removal during four reactor operation phases treating primary effluent (PE) in IFAS reactors.

Specifically, DO concentration was a significant parameter for carbon and nitrogen removal, especially for total nitrogen removal, which affected the formation of aerobic and anoxic zone onto outer layer and inner layer of biofilm, further the process of nitrification and reduction of nitrate. By decreasing DO concentration from 3.5 to 2 mg L^{-1} and HRT from 12 h to 8 h, respectively, the response increased. In this condition, oxidation ability of ammonia to nitrate was intensified and anoxic condition for denitrification was developed at high feeding. Additionally, further decreasing HRT to 6 h and DO concenration to 1.5-2.0 mg L^{-1}, respectively, TN reduction achieved averagely at 67.46 ± 5.98 % in stage III of GAC-IFAS reactor, which was higher than that in stage II (37.4 ± 3.85 %) ($p<0.01$).

Leyva-Díaz et al.(2013) once observed similar WWTP treatment performance with respect to nitrogen removal (67.34 ± 11.22 %), while the value of HRT (20 h) was higher than that used in this study (6 h). It's obvious that IFAS reactors demonstrated good performance by controlling DO concentration within 1.5-2.4 mg L^{-1}, even though under low HRT in this study. Compared with previous studies, the total nitrogen removal efficiency of 72% was reported for the systems with sponge modified plastic carrier at DO of 5.0-6.0 mg L^{-1} and HRT of 6 h in Deng et al., 2016. Moreover, in the similar research work, maximum total nitrogen removal was obtained to be 50%, packed with ring form carrier at the initial nitrogen concentration of 30-50 mg L^{-1}, HRT of 5.5-7.5 h and DO of 2.5-3 mg L^{-1} (Zinatizadeh et al., 2015). In this study, maximum 70.5% total nitrogen elimination achieved in the GAC-IFAS reactor also implied that simultaneous nitrification and denitrification (SND) process took place, although DO in both MBBRs was maintained at relatively low levels of 1.5–2.4 mg L^{-1}. It is noted that the decreasing effect of GAC-IFAS reactor was relatively more for the plastic-IFAS reactor, illustrating the anoxic condition was favored with GAC carrier, which can be because of its physical structure and biomass formation. Therefore, GAC modified bio-carrier, as DO reducing along with inner depth of adsorption, also preferred anoxic zone formation, and then thick biofilm would be easily formed on GAC to enhance SND process.

4.3 Biofilm formation and activity assessment

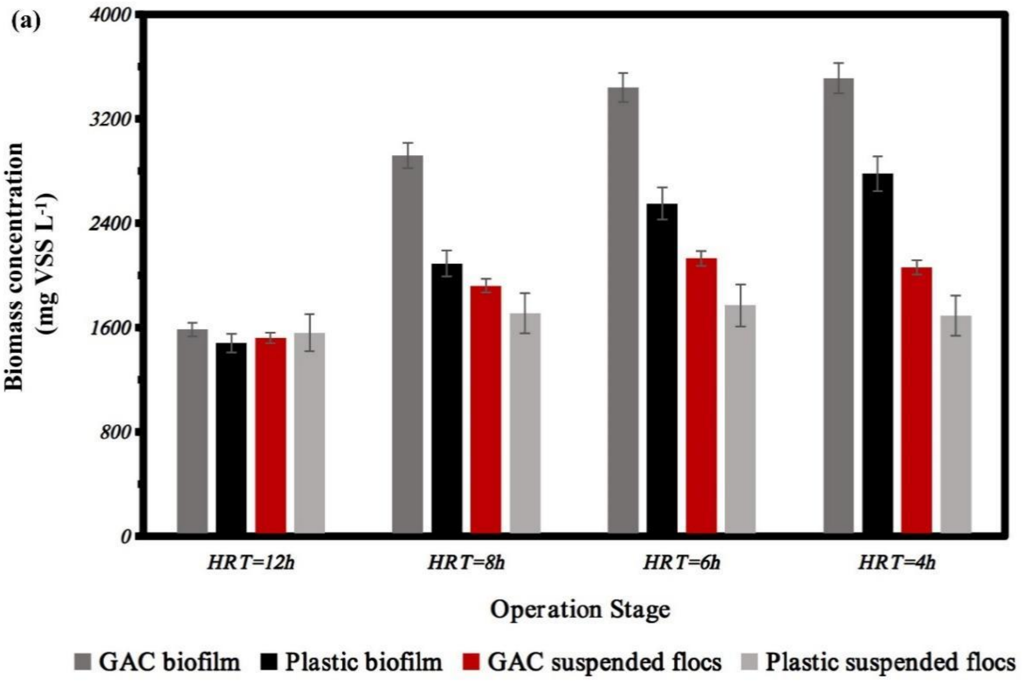
Biomass concentration in the IFAS reactors indicated treatment capacity of the system removing C and N. As shown in Fig. 7(a), the biomass concentration was a function of HRT and packing media. By reducing HRT from 12 h to 4 h in GAC-IFAS, the response of attached biomass concentration was significantly increasing from averaged 1.5863 g VSS L^{-1} to 3.5100 g VSS L^{-1} ($p<0.01$), more than suspended flocs from 1.5208 g VSS L^{-1} to 2.2645 g VSS L^{-1} ($p<0.05$). In both systems, a decrease of HRT caused an increase in the response as a result of high organic load. It is noted that the increasing effect of attached biomass on GAC (1.9237 g VSS L^{-1}) was more for the conventional plastic media (1.1508 g VSS L^{-1}) from start up phase to stage IV. The attached biomass on carrier and suspended flocs also reached steady state in the Stage III. The biomass of bioflim and suspended flocs in GAC-IFAS reactor was more (3.4405 ±

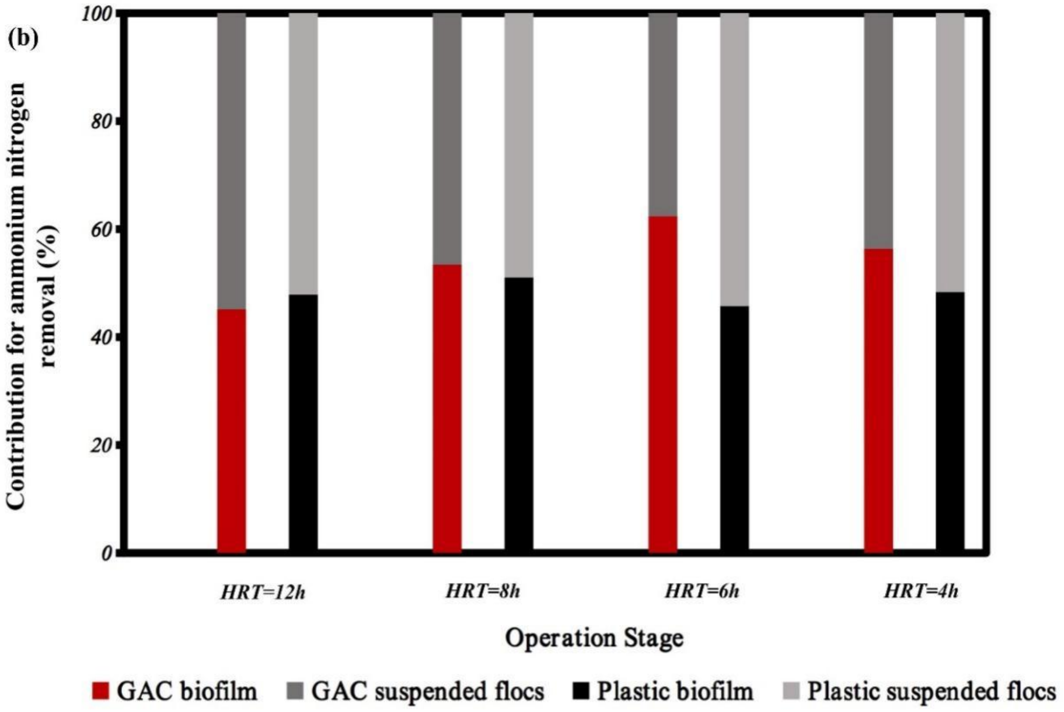
0.1455 g VSS L^{-1} and 2.1307 ± 0.2063 g VSS L^{-1}, respectively) than those for the plastic-IFAS reactor (2.5561 ± 0.2714 g VSS L^{-1} and 1.7731 ± 0.3050 g VSS L^{-1}, respectively).

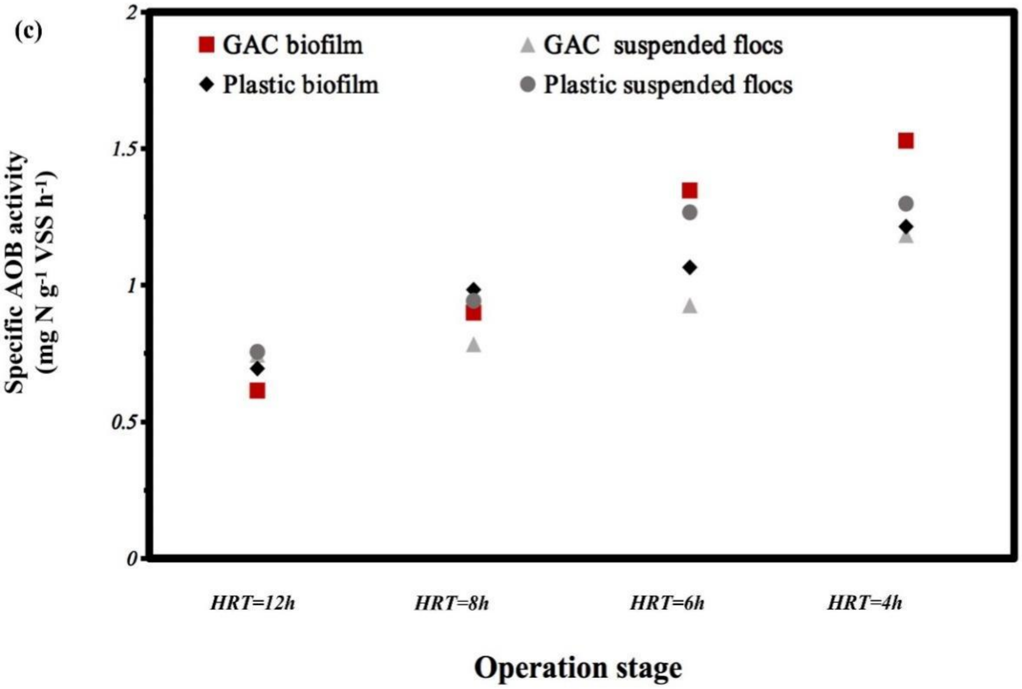
However, further reducing the HRT to 4 h, suspended flocs in GAC-IFAS and plastic-IFAS reactors slightly declined to 2.0611 ± 0.1542 g VSS L^{-1} and 1.6935 ± 0.0745 g VSS L^{-1}, respectively ($p>0.05$). For plastic carrier, the biofilm attachment and formation mainly maintained on the outer surface of plastic, which made biofilm could be less stable than that developed on GAC. As fresh GAC possesses large amount of pores as well as adsorption capacity, microorganisms can be entrapped into the pores and developed on both outer and inner surfaces of GAC (Guo et al., 2010). Hence, larger amount of biomass was attached onto the GAC modified bio-ball carrier as compared to that on conventional plastic carrier.

The batch tests were operated to observe the influence of the increasing organic loads on the maximum biomass specific ammonium oxidizing bacteria (AOB) activity and specific heterotrophic bacteria activity. Biomass enhancement of reactors under the increased volumetric organic loading (0.92-3.04 kg COD m^{-3} d^{-1} and 0.068-0.211 kg NH_4^+-N m^{-3} d^{-1}, respectively) was primarily associated with the stimulated growth of AOB bacteria and heterotrophic bacteria, which in turn contributing to nitrogen removal in reactors. As observed in Fig. 7(d), decreasing the HRT from 12 h to 4 h, specific heterotrophic bacteria activity in the biofilm achieved averagely 3.7286 to 8.4736 mg COD g^{-1} VSS h^{-1} for GAC carrier and 4.3254 to 6.6492 mg COD g^{-1} VSS h^{-1} for plastic carrier, respectively. It’s noticed that specific heterotrophic bacteria activity of attached biomass from stage II to stage IV in GAC-IFAS reactor was significantly higher ($p<0.05$) than that in plastic-IFAS reactor, showing in accordance with carbon removal efficiency in both reactors. It’s also noticed that the averaged heterotrophic activity on attached biomass was 1.22 mg COD g^{-1} VSS h^{-1} and 1.61mg COD g^{-1} VSS h^{-1} higher than that in suspended flocs for GAC-IFAS and plastic-IFAS reactors, respectively, illustrating heterotrophic bacteria may prefer to aggregate in attached biofilm to obtain the long retention time.

On the contrary, the specific activity of AOB bacteria, was significantly higher in suspended flocs than in biofilm in stage III and IV in plastic-IFAS reactor ($p<0.05$), due to the fact that the permeable and loose structures of suspension provide better access of NH_4^+-N and O_2 to nitrifying bacteria. Specific AOB activity of suspended flocs increased to 0.9128 mg NH_4^+-N g^{-1} VSS h^{-1} in Stage I and further increased to 1.3285 mg NH_4^+-N mg^{-1} VS h^{-1} in Stage III ($p<0.05$), which was associated with the increased volumetric ammonia loading rate (ALR).




(b)
Contribution for ammonium nitrogen removal (%)
100
80
60
40
20
0
HRT=12h
HRT=8h
HRT=6h
HRT=4h
Operation Stage
GAC biofilm
GAC suspended flocs
Plastic biofilm
Plastic suspended flocs


(c)
GAC biofilm
GAC suspended flocs
Plastic biofilm
Plastic suspended flocs
Specific AOB activity (mg N g-1 VSS h-1)
2
1.5
1
0.5
0
HRT=12h
HRT=8h
HRT=6h
HRT=4h
Operation stage

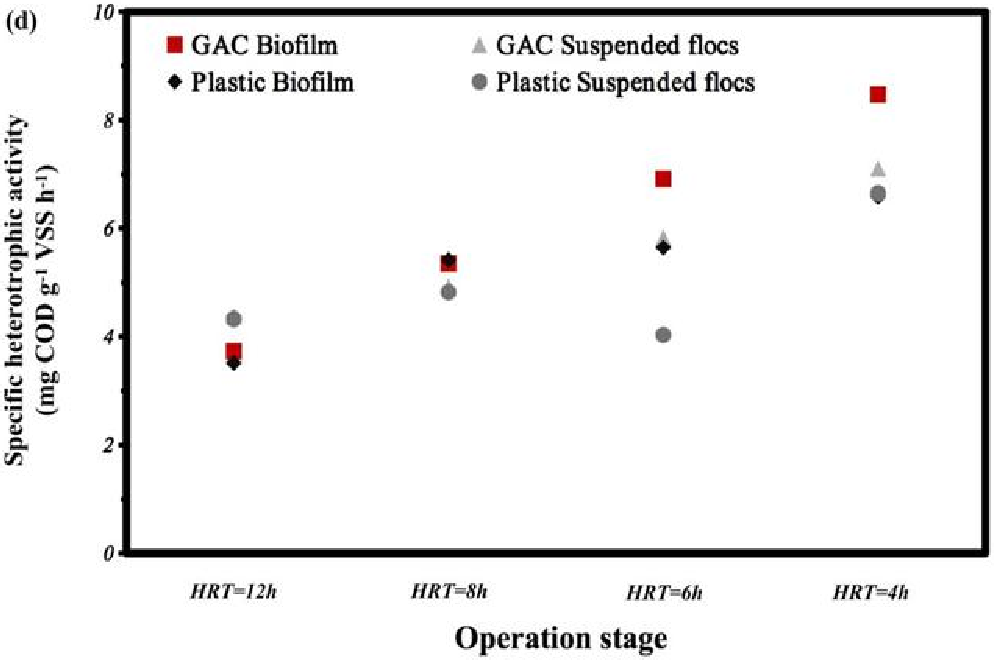


Figure.7. Biomass concentration(a), contribution for ammonium nitrogen removal (b), Specific activities of ammonium oxidizing bacteria (c) and heterotrophic bacteria (d) in both biofilms and suspended flocs configurations under different stage operations.

Interestingly, on basis of the activity batch tests results, it could be inferred that the contribution of the attached biomass to the total nitrifying activity was very pronounced of 62.41% in GAC-IFAS reactor at stage III. Furthermore, the specific nitrifying and heterotrophic activity of attached biomass (1.5289 mg NH4+-N g-1 VSS h-1 and 8.4736 mg COD g-1 VSS h-1) on GAC was estimated significantly higher than that on Plastic (1.2144 mg NH4+-N g-1 VS h-1 and 6.5423 mg COD g-1 VSS h-1) at Stage IV ($p<0.05$).

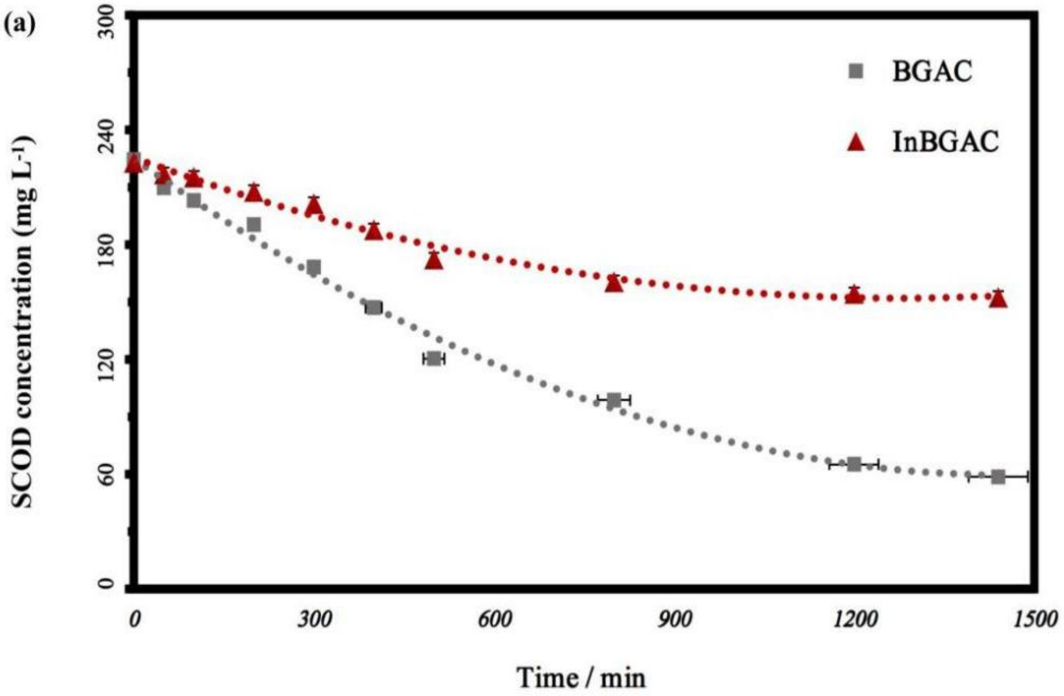
On one hand, the GAC was a kind of porous spherical-shaped carrier which have a protected surface area in its inner part which is not subjected to direct collision with other particles, favoring the accumulation of biofilm. Furthermore, this media configuration also could facilitate entrapment of the biomass in biofilm matrix (Bassin et al., 2016), while conventional plastic carrier not. Another point which should be considered is that the surface area of the carrier may change over time due to the overgrowth of biofilm and consequent media clogging (Forrest et al., 2014). These results indicated that the behavior of biomass in IFAS system differs, where attached biomass was relatively concentrated and stable compared with suspended flocs when reducing HRT.

4.4 BGAC carrier adsorption and biodegradation

SCOD concentration in primary effluent as a function of time for the BGAC and InBGAC groups was shown in Fig. 8(a). Obviously, SCOD removal efficiency and removal rate of BGAC group were further higher than that of InBGAC group ($p<0.05$). SCOD concentration were reduced to 168 and 201.4 mg L-1 after 6 h in the BGAC and InBGAC treatment groups, respectively, with removal efficiency of 35.8% and 10.2%. As previously mentioned, BGAC represented the combination of physical sorption , biofilm sorption and biodegradation, while InBGAC represented the combination process except biodegradation.

Therefore, the removal difference between two treatment groups could be estimated as the contribution of biodegradation. As shown in Fig. 8(b), After about 24 h of treatment, the SCOD concentration in BGAC decreased by 82.2%, which is higher than that in InBGAC with 29.5% reduction. It's estimated that equivalent to 52.7% removal efficiency attributed by biodegradation after 24 h, which was 27.1% more than that after 6 h. Hence, biodegradation plays significant role for organics removal in this study.

In this context, according to Giustra (2017), solute (in terms of COD) uptake on adsorbents (Granular activated carbon) involves two processes. The first one is the transport of SCOD, from solution to particle solution interface, and the second is the adsorption on the accessible surface site of adsorbent particle or intraparticle diffusion. This can allow biodegradation and adsorption simultaneously be taken.



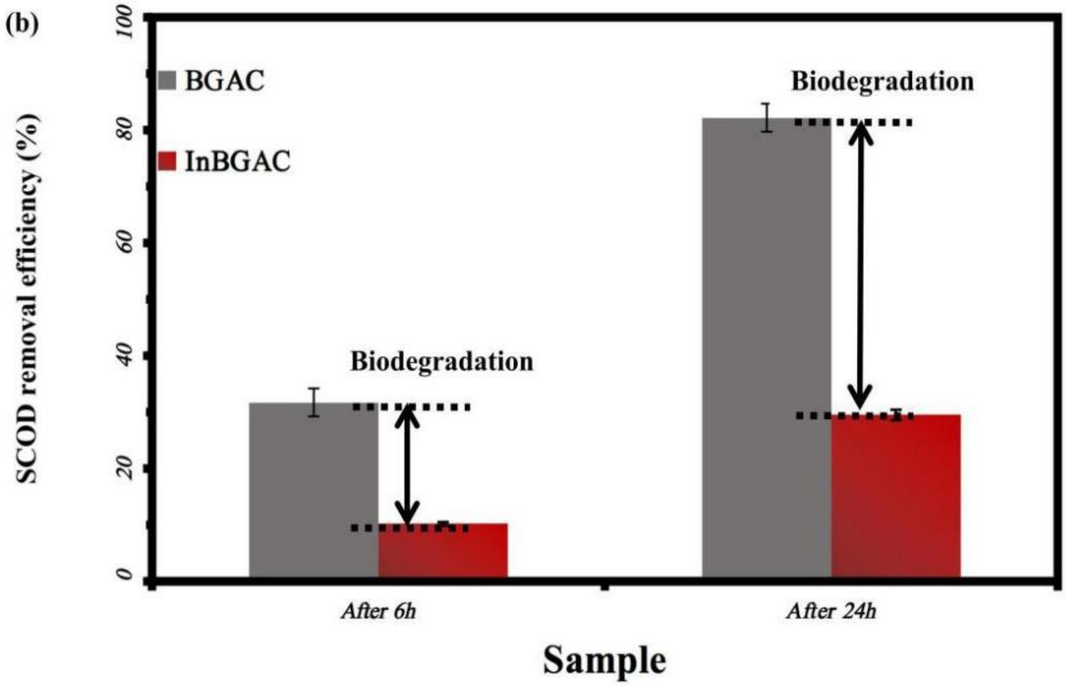


Fig. 8. (a) SCOD concentration (mg L-1) as a function of time (h) for the BGAC and InBGAC treating primary effluent. (b) SCOD removal efficiency (%) of the BGAC and InBGAC treatment groups after 6 and 24 h. (The gaps between arrows represent contribution of the biodegradation mechanism to the overall SCOD removal efficiency (%))**.**

Two types of models were used to describe the kinetics of SCOD adsorption onto the GAC media: the pseudo-first order and pseudo-second order models (Safwat et al., 2018). Fig.9 plotted the linear relationship of PFO and PSO expression for BGAC and InBGAC, respectively. For each case, the corresponding correlation coefficients (R^2) were estimated.

The experimental data of PSO (R^2=0.9913) was much higher than that of PFO (R^2=0.792) for BGAC, suggesting that the adsorption of SCOD onto granular activated carbon best fit the PSO kinetic model. Such a finding is also in good agreement with InBGAC. In addition, K_2 and qe were found to be 8.24×10^{-3} g mg^{-1} min^{-1} and 7.051 mg g^{-1}, respectively, for BGAC and 2.75×10^{-3} g mg^{-1} min^{-1} and 2.290 mg g^{-1}, respectively, for InBGAC.

Moreover, possible changes in the GAC bed structure, such as porosity and surface area, caused by attrition or by macro-molecules deposition (Mohammad et al., 2018; Naidu et al., 2019). The hydrolysis of synthetic substrates, could attribute to multiple biodegradable and non-adsorbable constituents, for instance, the formation of amino acids, small peptides, rich B vitamins, glutathione and nucleotide substances in yeast hydrolysates.

Compared with previous studies, results varied on different operational conditions (i.e carrier type, DO concentration and organic loading rate). Shorter hydraulic retention time and lower DO concentration was required in this study, differing from previous literature mostly between 8-20 hours and 3-8 mg/L, respectively, and notably, a higher OLR of 3.04 g COD/L-d was achieved in GAC developed bioreactor without any process addition.

Table 6. Performance comparation with previous study

Parameters	**This study**				**Lab-scale IFAS-SBR (Church et al., 2018)**	**Pilot-scale IFAS-MBR (Mannina et al., 2017)**	**Lab-scale IFAS-SFD-MBR (Vergine et al., 2018)**	**Pilot scale (Moretti et al., 2015)**	**Lab-scale IFAS-SBR (Zhou et al., 2019)**	**Pilot scale hybrid IFAS (Di Trapani et al., 2013)**	**Lab-scale IFAS-SBR (Gu et al. (2017)**
DO (mg/L)	1.5-2.1				7-8	5.33	2-4	2-5.7	-	2.16-3.25	-
Carrier	GAC				AK, K1	AK, K1	PE carrier	BMX1	synthetic with silver nanoparticle	AK, K1	cubic sponge
Average influent TCOD concentration (mg/L)	520				150	607	411.9-435.6	433	454.4-494.8	190.25-238.75	400
HRT (h)	12	8	6	4	12	20	8	4.5-8.1	8	3.47	6.5
OLR (g COD/L-d)	1.09	1.52	2.05	3.04	0.3	0.73	1.24-1.31	1.28.-2.31	1.36-1.49	1.32-1.65	1.48
TCOD removal (%)	93.5	95.02	95.47	78	95	98	90	90	96.8	100	85
Ammonium removal (%)	91.4	97.5	95.1	85	57	98	95	99	99.3	81.19	-
TN removal (%)	27.6	38.2	67.5	66.0	-	51.5	42	90	-	-	85
MLVSS (g/L)	1.58	2.63	3.44	3.51	1.8-2.0	-	5.4-6.4	1.5-2.5	-	3.3	-

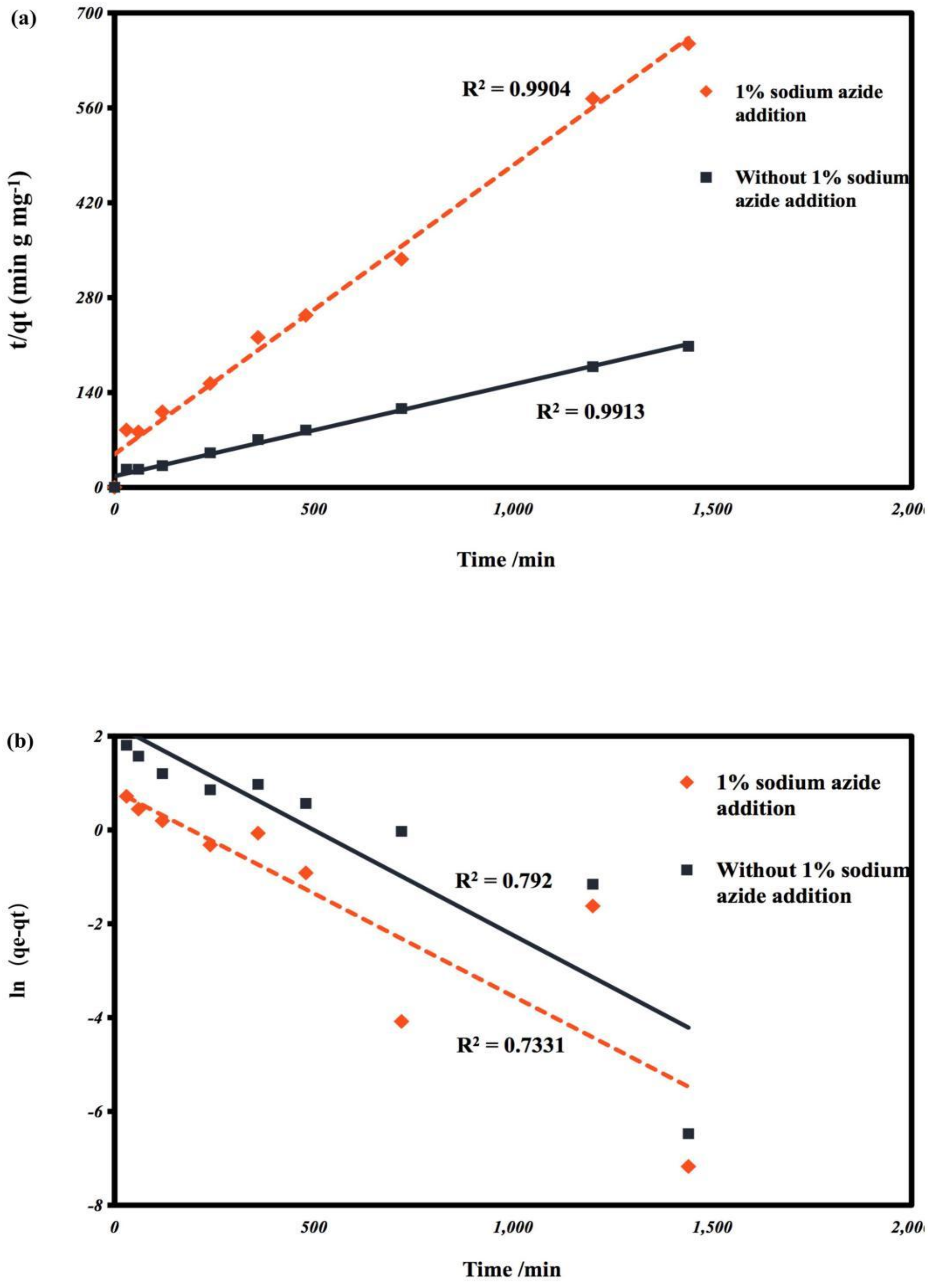


Fig. 9. Pseudo-second order (a) and pseudo-first order (b) rate as a function of time (h) for the SCOD removal in BGAC (without sodium azide addition) and InBGAC treatment groups.

Chapter 5

Conclusions and Recommendations

5.1 Conclusions

A list of the major findings from this study is summarized below:

- The performance of GAC-IFAS reactor varied depending on different HRTs. Highest COD and ammonia removal (91.2% and 93.5%, respectively) was achieved at low HRT of 6 h. Even so, higher nutrient removal (>81.8%) was obtained over the entire operation period in GAC-IFAS reactor, compared to plastic-IFAS reactor. Moreover, the total nitrogen removal also achieved nearly 70% as increasing the organic loading and biofilm formation.
- The GAC enhanced integrated fixed-film activated sludge (IFAS) reactor could provide an efficient performance in terms of nutrient removal and system stability at ambient temperature. Notably, a high OLR of 3.14 g COD/L-d was achieved in this reactor without any process instability, compared to mostly used OLR of 0.5-2.4 g COD/L-d in previous studies.
- Overall, the performance of GAC-IFAS reactor was better and relatively more stable than plastic-IFAS reactor, which attributed to the characteristics of GAC encaged carrier itself. This newly developed bio-carrier could provide larger surface area and lots of adsorption sites to support nutrient attachment and biofilm growth, so that organics and nutrients can be efficiently removed onto the outer layer and inner layer of GAC. Therefore, GAC modified

bio-carrier could be a promising solution to enhance treatment performance of municipal wastewater.

5.2 Recommendations

The application of carbon materials like activated carbon appears as a promising technology to improve the treatment efficiency in IFAS system, however, knowledge on this research topic is still limited. The recommendations for future work are as follows:

- Understanding the microbial community structure related to nutrient removal from municipal is a key scientific question for the potential application of granular activated carbon for wastewater treatment efficiency improvement.
- Exploration on how to further improve the performance of GAC enhanced integrated fixed-film activated sludge (IFAS) reactor is required:

i. Since the retained biomass on GAC could contribute to improving efficiency in terms of organic removal. The specific surface areas of the granular carbon can be increased to provide more attached area for adsorption, thus promoting biofilm formation and suspended solid removal.

ii. This study offered a novel and transformative solution for enhancement of nutrient removal, with modified granular activated carbon as one of the promising carriers for wastewater treatment. Therefore, feasibility of incorporating granular activated carbon into the integrated fixed-film activated sludge system is an interesting and promising approach to further enhance the performance.

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