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Efficiency of Attached-Growth Sequencing Batch Reactor in the Treatment of Recycled Paper Mill Wastewater

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Graphical abstract

Abstract

In this study, an attached-growth bioreactor was operated using granular activated carbon (GAC) with additional biomass; and evaluatedits performance in the treatment of real recycled paper mill effluent at chemical oxygen demand (COD) level in the range of 800-1300 mg/L, a fixed hydraulic retention time of 24 hours and COD:N:P ratio of about 100:5:1. A laboratory-scale aerobic sequencing batch reactor (SBR) was used. The efficiency of this biological treatment processwas studiedover a 300-day period, in order to evaluate their performance, especially for the removal of nitrogen compound and of biodegradable organic matter. It has been found that this process was able to remove organic matter (expressed as COD; 91-99%) and turbidity (89-99%) almost completely and simultaneously; the removal of nitrogen (expressed as NH_3-N; 70-94%), phosphorus (expressed as PO_4^{3}-P; 42-71%), suspended solid (81-99%) and colour (72-91%) were sufficiently achieved. The overall performance confirmed that an attached-growth SBR system using additional biomass on GAC is a promising configuration for wastewater treatment in terms of the performance efficiency and process stability under fluctuations of organic load.

Keywords: Attached-growth bioreactor; sequencing batch reactor; granular activated carbon; recycled paper wastewater treatment

Abstrak

Dalam kajian ini, satu bioreaktor filem tetap dioperasikan menggunakan granular karbon teraktif (GAC) dengan penambahan biojisim; dan prestasinya dinilai dalam merawat efluen sebenar dari kilang kertas kitar semula dengan kepekatan permintaan oksigen kimia (COD) dalam julat 800-1300 mg/L, masa tahanan hidraulik 24 jam dan nisbah COD:N:P 100:5:1. Satu reaktor sesekumpul berjujukan (SBR) berskala makmal yang beroperasi secara aerobik digunakan dalam kajian ini. Kecekapan proses rawatan biologi ini telah dikaji dalam tempoh 300 hari, dalam usaha untuk menilai prestasinya, terutamanya dalam penyingkiran bahan organik yang mudah terbiodegradasi dan sebatian nitrogen. Hasil kajian ini menunjukkan bahawa proses ini mampu menyingkirkan bahan organik (dinyatakan sebagai COD; 91-99%) dan kekeruhan (89-99%) secara serentak dan hampir sepenuhnya. Manakala, penyingkiran nitrogen (dinyatakan sebagai NH_3-N; 70-94%), fosforus (dinyatakan sebagai PO_4^{3}-P; 42-71%), pepejal terampai (81-99%) dan warna (72-91%) adalah memuaskan. Prestasi keseluruhan mengesahkan bahawa sistem SBR filem tetap menggunakan biojisim pada GAC adalah konfigurasi yang memberangsangkan dalam merawat air sisa berdasarkan kecekapan prestasi dan kestabilan proses dalam keadaan bebanan organik yang sentiasa berubah-ubah.

Kata kunci: Bioreaktor filem tetap; reaktor sesekumpul berjujukan; granular karbon teraktif; rawatan air sisa kertas kitar semula

1.0 INTRODUCTION

Nowadays, pulp and paper mills are facing a lot of challenges in treating and reusing industrial effluents due to the complexity of residual natural organic matter from the treated effluents. Many pulp and paper mills need to treat the effluent to required compliance levels, not only for conventional parameters such as chemical oxygen demand (COD), biochemical oxygen demand (BOD), suspended solid (SS), and colour, but also the residual organic matters consisting of potentially toxic chlorinated compounds.[1] About 500 chlorinated compounds, measured as adsorbable organic halides (AOX), have been identified, such as chloroform, chlorate, resin acids, chlorinated hydrocarbons, phenols, catechols, guaiacols, furans, dioxins, syringols, vanillins, etc.,[2] and their removal from effluents is highly significant from an environmental safety perspective. AOX may bioaccumulate in fish tissue, causing a variety of clastogenic, carcinogenic, endocrinic, and mutagenic effects, which may subsequently also pose problems to humans after consumption of the contaminated fish.[3]

In order to meet the more stringent effluent limitations due to the severity of these toxic effects, various physical, chemical, and biological treatments have been employed for pulp and paper wastewater treatment,[4,5,6] among which biological treatments have become the focus of many studies because of their lower cost and potential to mineralise the contaminant. Until now, conventional wastewater treatment, such as activated sludge processes, are widely used for the treatment of pulp and paper wastewater. Conventional wastewater treatment of pulp and paper mill effluent using suspended growth biomass is sufficient for reuse; however, the persistence of recalcitrant organics limits reuse opportunities in paper production without further processing.[7] Also, the conventional adsorption technique using activated carbon (AC) source as a final treatment is indispensable for removal of these recalcitrant organics from pulp and paper mill wastewater. Essentially, the conventional adsorption technique has the disadvantages of inadequate exploitation of the adsorptive capacity, and a high cost of conventional thermal or chemical regeneration process for spent AC. In search for more efficient and economically practicable processes, a combination of attached-growth biomass (known as biofilm) and granular activated carbon (GAC) adsorption was studied by Muhamad *et al*.[8] The presence of GAC offers a highly porous and rough media, which is able to adsorb organic matter, as well as provide an appropriate surface onto which active biomass can be attached and grown. Furthermore, the GAC can be generated from various types of resources including those originating from agricultural waste, such as coconut shells, coconut husks, apricot shells, walnut shells and bamboo.[9] Hence, this method is ecofriendly, cheap and simple.

The aim of the present paper is to evaluate the effectiveness of the attached-growth systems with additional biomass on the removal of COD, SS, turbidity, ammoniacal nitrogen (NH_3-N), phosphate-phosphorus (PO_4^{3}-P) and colour from recycled paper wastewater. The biological treatment systems was carried out in a Sequencing Batch Reactor (SBR).

■2.0 MATERIALS AND METHODS

2.1 Wastewater Samples Collection and Composition

Wastewater samples were collected from Muda Paper Mills Sdn. Bhd., a recycled paper mill, situated in the western part of Malaysia, relies on 100% recycled fibres for the raw paper material. The effluent used in this study is the physicochemically treated effluent discharged from a dissolved air flotation (DAF) unit of an existing effluent treatment plant specifically designed and operated for the treatment of recycled paper wastewater. From January 16 until November 13, 2014, the effluent was collected once a month and kept in a refrigerator for further use. The typical characteristics of the recycled paper wastewater used as the feed in this experiment are presented in Table 1.

Table 1 Characterisation of the DAF unit effluent and the changes in the influent characteristics after nutrient addition (Please note that the standard deviation of pollutant concentration is indicated in the bracket)

Parameter	Units	Before nutrient addition	After nutrient addition
pH		6.4 (0.2)	7.6 (0.6)
COD	mg/L	1057 (150)	1057 (150)
SS	mg/L	149 (30)	149 (30)
Turbidity	NTU	73 (15)	73 (15)
NH_3-N	mg/L	4.1 (0.6)	54 (8)
PO_4^{3}-P	mg/L	0.03 (0.02)	11 (1)
Colour	Pt-Co	121 (3)	121 (3)

2.2 Seeding Sludge

Activated sludge obtained from the activated sludge treatment plant of the same recycled paper mill served as inoculums for the lab-scale bioreactor. The original seed sludge contained 6000 mg/L mixed liquor suspended solid (MLSS).

2.3 Lab-Scale Bioreactor and Operating Schedule

The experiment was conducted in laboratory scale SBR with a total volume of 5 L, as depicted in Figure 1. Table 2 illustrates the reactor configuration details. The cylindrical Plexiglas reactor was operated in batch mode under aerobic condition for the recycled paper wastewater. The fixed bed material used in this research was the commercially available GAC which is a coconut shell-based activated carbon. The reactor configuration was packed with GAC (density: 1250 kg/m^3) of sizes in the range of 1-2 mm. This fixed-bed material roles as the adsorption medium and also to support the formation of biofilm. Fine air bubbles for aeration were supplied in the liquid phase over the GAC through a porous stone to maintain a dissolved oxygen concentration in the range of 2-3 mg/L.

Figure 1 Lab-scale attached-growth SBR with additional biomass

All reactors were filled with 2.5 L of recycled paper wastewater and operated with an optimal HRT of 24 hours,[8] consisting four consecutive phases: filling and reaction (aerated; length: 22 h), settling (non-aerated; length: 1 h), and drawing (non-aerated; length: 1 h). Since the nitrogen (N) and phosphorus (P) levels calculated for the DAF effluent were not high enough to provide an adequate supply for a biological treatment system, urea and trisodium phosphate were added to achieve a COD:N:P ratio of approximately 100:5:1.[8,10] Table 1 presents the changes in the influent characteristics after nutrient addition in this experiment.

Table 2 Reactor configuration details

Total volume (L)	5
Supported media	GAC
Volume occupied by the GAC (L)	1
Net void volume inside the reactor (L)	0.6
Net void ratio inside the reactor	1.5
Liquid (working) volume (L)	2.5
Inoculated biomass volume (L)	0.5
Initial MLSS conc. (inoculated biomass) (mg/L)	1000

2.4 Analytical Procedure

The wastewater was characterized for various physical and chemical properties. NH_3-N, PO_4^{3}-P and colour were measured on liquid samples after filtration with Whatman-type nitrate cellulose membrane filters (0.45 mm). COD, NH_3-N and PO_4^{3}-P and colour were measured by the reactor digestion method (Method: 8000), the Nessler method (Method: 8038), the PhosVer 3 (Ascorbic Acid) method (Method: 8048), and the Platinum-Cobalt standard method (Method: 8025) respectively, using a spectrophotometer (Hach DR3900, USA). COD concentration measured on unfiltered samples. The pH was not controlled during the experiment.Water pH was monitored by a pH meter (Metrohm 827, USA). Dissolved oxygen (DO) was measured with a DO instrument (YSI 550A, USA). Turbidity was analyzed with a turbidity meter (Hach 2100AN, USA). The suspended biomass concentration was determined as mixed liquid suspended solids (MLSS). MLSS and SS analysis were performed according to APHA standard methods.[11] The performance of the reactor was evaluated by estimating the substrate (COD, NH_3-N, PO_4^{3}-P, SS, turbidity or colour) removal efficiency of each parameter by comparing the initial substrate (COD, NH_3-N, PO_4^{3}-P, SS, turbidity or colour) concentration in the feed with the substrate (COD, NH_3-N, PO_4^{3}-P, SS, turbidity or colour) concentration in the reactor outlet.

■3.0 RESULTS AND DISCUSSION

The COD, SS, turbidity, NH_3-N and PO_4^{-3}-P removal efficiencies over a 300-day period are shown in Figures 2-6.Table 3gives the mean pollutant removal performances in the bioreactor and recycled paper mill effluent discharge guidelines in Malaysia. In this study, the real wastewater collected from recycled paper mill may exhibit large variations in the amounts of COD leading to unstable results obtained during the early period and periods of time required to reach the steady-state conditions. During the initial 7 days, the bioreactor displayed poor performance at high influent COD concentration of 1262 mg/L resulting in the low removal efficiencies. During this stage, most quality parameters of the treated effluent from the bioreactor not complied with the standard limits stated by the Malaysia legislation regarding industrial wastewater treatment.[12] During the following days, the removal efficiencies increased gradually and were found to be fluctuated at this stage as the system not fully acclimatised. The data listed in Table 3 covers all data recorded after NH_3-N removal was started to be stabilised on day 49 (except the colour data comprised between days 152-300). The discussion in this section relate to the data after the start-up period and comprised between days 49-300.

Figure 2 shows the COD removal for the bioreactor. As shown in Figure 2 and Table 3, the organic matter was almost removed. This removal was very high in the bioreactors studied, as can be observed in Table 3 through the parameter COD. The removal percentage of COD for the bioreactor had value of 95 ± 2%, as shown in Table 3. Muhamad *et al*.[8] obtained similar percentage of COD removal (95%) in the pilot-scale attached-growth SBR using GAC, known as GAC-SBBR with similar operating conditions to those used in this research. The values of COD for the effluent of the bioreactor always obey the Malaysia standard limits, as can be observed in Figure 2 and Table 3.During days 126-146, the bioreactor was fed with high carbon source of 1319 mg COD/L. As shown in Figure 2, no significant difference in the removal efficiency of COD was observed during this period (days 126-146), even the influent COD concentration higher than 1000 mg/L.

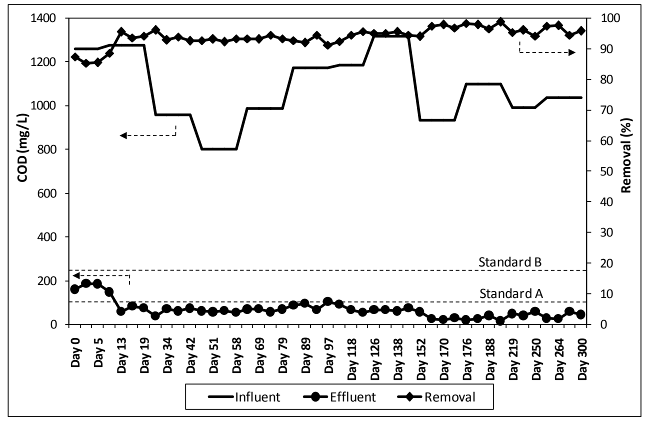

Figure 2 Variation of influent and effluent COD and its removal efficiency for attached growth SBR with additional biomass (Please note that the dashed lines with arrow represent the influent, effluent and removal)

Figure 3 demonstrates the SS removal for the bioreactor. As can be observed from Figure 3, during bioreactor start-up (days 0-34), the removal of SS was inconsistent from day to day as the system was not fully acclimatised. A significant improvement in SS removal was observed after 34 days operation and the SS removal stabilised in the range of 81-99% (Figure 3). As shown in Table 3, the value of SS for the effluent of the bioreactor was 12 ± 6 mg/L. The performance of the bioreactor was excellent with respect to the removal of SS, with value of 92 ± 5%. In accordance with the legislation requirements, the concentrations of SS of the treated effluent from attached-growth SBR with additional biomass were always lower than 20 mg/L (standard A) and 50 mg/L (standard B), as shown in Figure 3 and Table 3. The high removal of SS together with the high reduction percentages of COD indicate that the GAC employed together with additional biomass had excellent role as adsorbent, filtration unit and biofilm attachment, so the system was able to accept, with the same volume, a higher organic load in the influent. Figure 4 describes the turbidity removal for the bioreactor.The efficiency of the bioreactor in the reduction of turbidity is impressive, even at high influent COD, as shown in Figure 4. The percentage reduction of more than 95% can be achieved at influent turbidity of 93 NTU. As shown in Table 3, the levels of turbidity for the effluent of the bioreactor showed a positive relationship, meaning that a decrease in SS concentrations results in a decrease in turbidity levels. Turbidity could provide a good estimate of the concentration of SS in a water sample, even though turbidity is not a direct measure of suspended particles in water.[13]

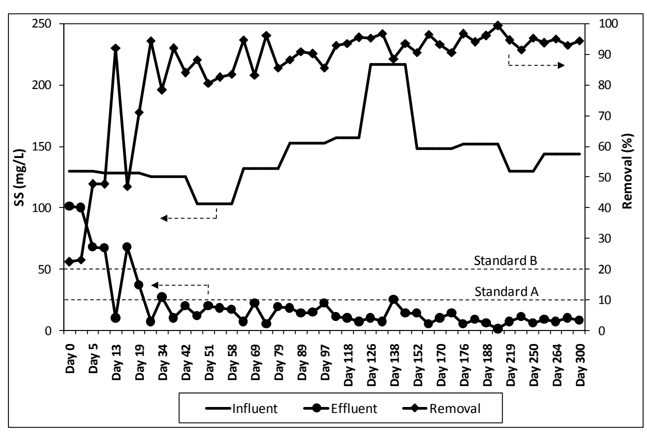

Figure 3 Variation of influent and effluent SS and its removal efficiency for attached growth SBR with additional biomass (Please note that the dashed lines with arrow represent the influent, effluent and removal)

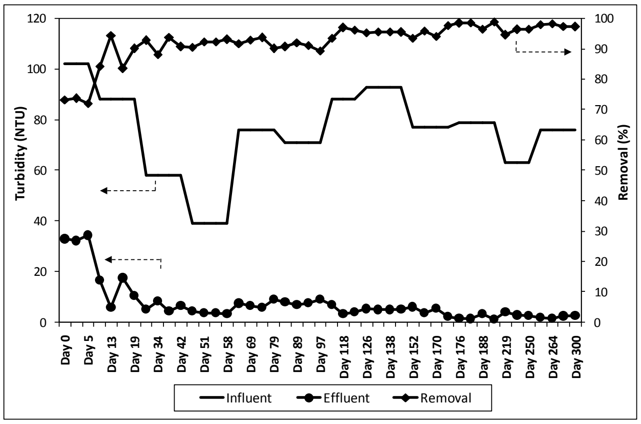

Figure 4 Variation of influent and effluent turbidity and its removal efficiency for attached growth SBR with additional biomass (Please note that the dashed lines with arrow represent the influent, effluent and removal)

The emissions of nitrogen and phosphorus from the pulp and paper industry are not very specific since the wastewater contain limited amounts of nutrients. The nutrients are crucial for microbial growth and thus for the BOD reduction, the addition of external nitrogen and phosphorus to the effluents is necessary in order to keep the level in appropriate range.[14] In this study, the average concentrations of NH_3-N and PO_4^{3}-P in the influent and the effluent of the bioreactor are indicated in Table 3. There, the respective removal percentages of these nutrient can also be seen. The variation of influent and effluent NH_3-N and its removal efficiency is shown in Figure 5. The bioreactor had a percentage of nitrogen removal of 86 ± 6%. As shown in Table 3, the average concentration of NH_3-N in the effluent of the bioreactor was lower than the Malaysia standard limits stated by the legislation regarding industrial wastewater treatment (10 mg NH_3-N/L for standard A and 20 mg NH_3-N/L for standard B). Nevertheless, the standard limit of A could not be obeyed if the concentration of NH_3-N in the effluent achieved maximum NH_3-N concentration of each bioreactors, as can be observed in Table 3 and Figure 5. In order to achieve the nutrient-spike with a COD:N:P ratio of 100:5:1, the reactor was supplied with 63-72 mg/L of nitrogen source between days 85-146. During this period (days 85-146), the NH_3-N effluent quality was hardly enough to meet the discharge standard A (10 mg NH_3-N/L), with a percentage of NH_3-N removal of 81 ± 3%. During the following days, the bioreactor was fed with 49-54 mg/L of nitrogen source, in which the concentrations of NH_3-N in the effluent always obey the Malaysia standard limits. These results showed that the NH_3-N removal decreased with increasing nitrogen load. In this case, only 50 mg/L of nitrogen source in the bioreactor is necessary to obtain sufficient biodegradation if the concentration of COD in the influent is between 1000-1300 mg/L.

Figure 6 shows the performance of the bioreactor regarding PO_4^{3}-P removal. Unlike NH_3-N, the bioreactor had the lowest percentage of PO_4^{3}-P removal, with a value of 60 ± 8%, as indicated in Table 3.As shown in Figure 6, the PO_4^{3}-P removal showed a wider range of efficiency (42-71%) between days 49-300.Mino *et al.*[15] noted that a high phosphorus removal efficiency can be achieved by wasting excess sludge with high phosphorus content. Although Malaysia did not address phosphorus discharge limit in the industrial wastewater treatment, discharging treated effluent with high phosphorus should be monitored and controlled. Excessive amounts of nutrients (e.g., phosphorus) cause a negative effect on rivers or lakes into which treated effluents are discharged by promoting eutrophication.[16]In China, phosphorus limit for existing pulp mills, paper mills and integrated pulp and paper mills is 0.8 mg/L.[17]To achieve the mandatory phosphorus target, manipulating of nutrient ratio especially phosphorus concentration is necessary for this study since the PO_4^{3}-P effluent quality was hardly enough to meet the effluent discharge standards of China.

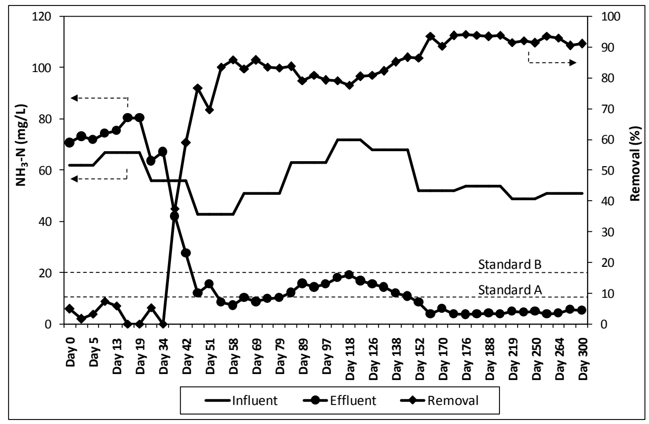

Figure 5 Variation of influent and effluent NH_3-N and its removal efficiency for attached growth SBR with additional biomass (Please note that the dashed lines with arrow represent the influent, effluent and removal)

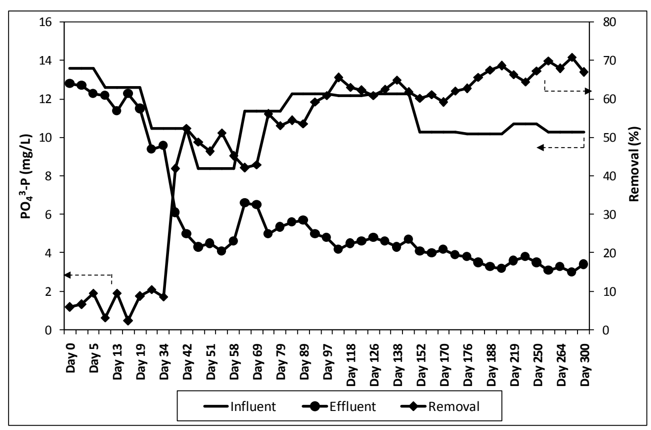

Figure 6 Variation of influent and effluent PO_4^{3}-P and its removal efficiency for attached growth SBR with additional biomass (Please note that the dashed lines with arrow represent the influent, effluent and removal)

Table 3 Mean pollutant removal performances of the bioreactor. Please note that the standard deviation of pollutant concentration is indicated in the bracket

Parameter	Unit	Influent concentration[a]	Attached-growth SBR with additional biomass		Industrial effluent guidelines in Malaysia (2014)	
			Effluent conc.	Efficiency (%)	Standard A	Standard B
pH		7.9 (0.6)	8.0 (0.4)	-	6.0-9.0	5.5-9.0
COD	mg/L	1057 (150)	53 (23)	95 (2)	80	250
SS	mg/L	149 (30)	12 (6)	92 (5)	20	50
Turbidity	NTU	73 (15)	4 (2)	95 (3)	-	-
NH_3-N	mg/L	54 (8)	8 (4)	86 (6)	10	20
PO_4^{3}-P	mg/L	11 (1)	4 (1)	60 (8)	-	-
Colour	Pt-Co	121 (3)	21 (8)	82 (6)	100[b]	200[b]

[a]after nutrient addition, [b]ADMI units

Colour reduction was also considered in this study. Colour of effluent is among the major environmental issues for the pulp and paper industry especially in the Asia countries as the conventional biological treatment systems currently used to control the effluent have little effect on colour. Regulations controlling colour effluent are becoming increasingly stringent.Nowadays, the pulp and paper industry is under increasing pressure to identify suitable solutions to the colour impacts which are currently considered tobe unacceptable. Based on regulation in some countries taken from literature study, Canada, USA and China have colour regulation specifically for pulp and paper industry.[17,18]In Malaysia, the colour regulation is used for general industry as stipulated in the Environmental Quality Regulation 2009 (Industry Effluents) under the Malaysia Environmental Quality Act 1974 (Act 127).[12] The colour unit used in the Malaysia legislation regarding industrial wastewater treatment is ADMI unit, as shown Table 3. The Pt-Co unit used in this study has almost similar values with ADMI. As shown in Table 3, a significant reduction of colour was observed during the experimental study, with value of 82 ± 6%. Figure 7 presents the colour difference between influent and treated effluent from the bioreactor and also final effluent from the existing treatment plant of the recycled paper mill. As seen from Figure 7, significant colour differences were observed between the influent and treated effluents collected from the bioreactors, with value of 13Pt-Co, compared with the influent (116 Pt-Co). According to Sierra and Lettinga,[19] the brownish colour results from the presence oflignin and its derivatives, which are difficult to degrade naturally. Compared with the conventional biological treatment processes used in the real recycled paper mill, the performance of the bioreactor used in this study for colour removal from pulp and paper mill wastewater can be considered satisfactory. As shown in Figure 7, a significant colour difference can be observed between treated effluent of the bioreactor and final effluent from the existing treatment plant of the recycled paper mill, with value of 13 Pt-Co and 89 Pt-Co, respectively. A study by Diezet al.[16] found that a slightly colour removal (13%) was obtained by the activated sludge system than that achieved in this study. In this study, the main colour removal mechanism was probably by physical adsorption since the GAC has high adsorption capacity.

In term of overall performance stability based on all removal efficiencies, the bioreactor showed a stable performance after 2-4 weeks operation, as can be seen in Figures 2–6. This was probably due to the role of the employed GAC and biofilm from the additional biomass. This results establish the potential application of attached-growth SBR with additional biomass, to provide effective treatment of recycled paper mill effluents under fluctuated loadings. In term of HRT, the optimal HRT of 24 hours used throughout this study is the best condition to treat real wastewater from a paper mill whereby the average removal efficiency for the COD, SS and turbidity can be achieved as high as 95%, 92% and 95%, respectively. A similar study was performed by Wan-Osman *et al*.[20] to treat real paper mill wastewater. From the study, they have found that COD removal of 92% was achieved in 48 hours of operation.

Figure 7 Photograph of (a) influent, (b) treated effluent collected from the bioreactor, and (c) final effluent from the existing treatment plant of the recycled paper mill

4.0 CONCLUSIONS

In this study, the evaluation of the attached-growth SBR with additional biomass treating real recycled paper wastewater showed that the bioreactor showed good performance in terms of the efficiency and process stability under fluctuations of organic load. The COD, SS, turbidity, NH_3-N, PO_4^{3}-P and colour reduction for this process had values of 91-99%, 81-99%, 89-99%, 70-94%, 42-71% and 72-91%, respectively. Further studies on kinetic, isotherms and mechanism of the toxic phenolic compounds removal using this process are being conducted.

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