

# Enhanced bioremoval of refractory compounds from dyeing wastewater using optimized sequential anaerobic/aerobic process

W. Bae<sup>1</sup> · D. Han<sup>1</sup> · E. Kim<sup>1</sup> · R. A. de Toledo<sup>2</sup> · K. Kwon<sup>1</sup> · H. Shim<sup>2</sup>

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**Abstract** The upflow anaerobic sludge blanket process followed by the biological aerated filter process was employed to improve the removal of color and recalcitrant compounds from real dyeing wastewater. The highest removal efficiency for color was observed in the anaerobic process, at 8-h hydraulic retention time, seeded with the sludge granule. In the subsequent aerobic process packed with the microbe-immobilized polyethylene glycol media, the removal efficiency for chemical oxygen demand increased significantly to 75 %, regardless of the empty bed contact time. The average influent non-biodegradable soluble chemical oxygen demand was 517 mg/L, and the average concentration in effluent from the anaerobic reactor was 363 mg/L, suggesting the removal of some recalcitrant matters together with the degradable ones. The average non-biodegradable soluble chemical oxygen demand in effluent from the aerobic reactor was 87, 93, and 118 mg/L, with the removal efficiency of 76, 74, and 67 %, at 24-, 12-, and 8-h empty bed contact time, respectively. The combined anaerobic sludge blanket and aerobic cell-entrapped process was effective to remove the refractory compounds from real dyeing wastewater as well as in reducing organic loading to meet the effluent discharge limits. This integrated process is considered an effective and economical treatment technology for dyeing wastewater.

**Keywords** Biological aerated filter · Color removal · Microbial immobilization · Recalcitrant compounds removal · Upflow anaerobic sludge blanket

## Introduction

Due to the growth of dyeing industries and the development of various textiles, dyestuffs, and dyeing processes, a significant amount of refractory compounds such as surfactants, polyvinyl alcohol (PVA), ethylene glycol (EG), and dyestuffs are released into the environment. The greater part of dyes, which can be toxic or mutagenic, in dyeing wastewater is poorly removed by the conventional wastewater treatment system (Lin and Lin 1993; Frijters et al. 2006). Different types of technologies have been developed to remove these refractory compounds. Physical and chemical treatment systems such as coagulation/sedimentation, Fenton oxidation, and ozone oxidation suffer from some drawbacks, including the requirement of intensive energy and supplementary chemicals, the high cost, and the formation of hazardous by-products (Hong et al. 2013; Fan et al. 2014). Biological treatment, in comparison, may be a good alternative due to the lower operational cost compared to most physicochemical technologies and the eco-friendly features (Popli and Patel 2015), despite such shortcomings as sludge bulking and low treatment efficiency for refractory compounds (Ahn et al. 1999). Li and Zhao (1997) reported that biological processes applied in the treatment of textile wastewater showed a practical limit in terms of the removal of organics, color, and other biodegradable substances in different activated sludge processes, with the elimination efficiencies of 90 % for biochemical oxygen demand (BOD), 40–50 % for chemical oxygen demand (COD), and

✉ H. Shim  
hjshim@umac.mo

<sup>1</sup> Department of Civil and Environmental Engineering, Hanyang University, Sa 1-Dong, Ansan, Gyunggi-Do 425-791, Korea

<sup>2</sup> Department of Civil and Environmental Engineering, Faculty of Science and Technology, University of Macau, Avenida da Universidade, Taipa, Macau SAR, China

20–30 % for color. A sequential anaerobic/aerobic treatment was developed to treat the cotton textile mill wastewater using an upflow anaerobic sludge blanket (UASB) followed by a continuously stirred tank reactor (CSTR) (Isik and Sponza 2004). The COD and color removal efficiencies of 40–85 and 39–81 % were reported, respectively, in real and artificially colored wastewaters at a total hydraulic retention time (HRT) of 5.75 days. The combination of physicochemical and biological treatment processes was also considered (Sudarjanto et al. 2006; Lucas et al. 2007; Lotito et al. 2014), but it was still not easy to meet the discharge limit for color. Zou (2015) recently developed a hybrid method combining ozonation, activated carbon, and biological aerated filter (BAF) for the advanced treatment of dyeing wastewater for reuse, despite the high cost for ozonation. In comparison, the biological approaches to treat dyeing wastewater were reported successful by employing anaerobic/aerobic (Brown and Hamburger 1987; Shaw et al. 2002; Amaral et al. 2014), facultative anaerobic (Li and Xi 2004), and anaerobic (Senthilkumar et al. 2011) processes to remove aromatic amines formed from the azo-bond dyestuff. The aerobic sludge granules highly concentrated with microorganisms (Kolekar et al. 2012), the redox mediator (Rios-Del Toro et al. 2013), and the PVA immobilization media (Chen et al. 2003) had been applied for the removal of color from dyeing wastewater. The degradation of PVA contained in dyeing wastewater by pure or mixed microorganisms had also been reported (Kim et al. 1997; Choi et al. 2004). To improve the efficiency of dyestuff biodegradation, such microbial immobilization technologies using polyvinylidene fluoride (Al-Zuhair and El-Nass 2011), PVA (Takei et al. 2011), alginate (Chen et al. 2010), and polyethylene glycol (PEG) (Kim et al. 2013) have been receiving considerable attention.

The main objective of current study was to evaluate the sequential anaerobic/aerobic process for its potential to effectively remove recalcitrant contaminants from real dyeing wastewater. The applied combined process consisted of the UASB reactor seeded with the sludge granule followed by the BAF filled with microbes-immobilized PEG media. This would be the first time the sequential UASB/BAF with cell-immobilized PEG media was used to treat real dyeing wastewater. The optimal operational factors for this combined process were also investigated to further improve the removal efficiency for color. This study was carried out at Department of Civil and Environmental Engineering, Hanyang University, Republic of Korea, during the period between 2013 and 2014.

## Materials and methods

### Reactor setup and operational conditions

The UASB reactor (working volume, 10 L) was made of a tapered acrylic cylinder and the inside water temperature was maintained at 30 °C using a water jacket (Fig. 1). The reactor was seeded with the sludge granule dominated by mesophilic microorganisms, obtained from the anaerobic wastewater treatment facility at a brewery industry located in Choongchungbuk-Do, Korea (Fig. 2a). Eighty percent of the reactor volume was filled with the sludge granule, and the biomass concentration was maintained at approximately 35,000 mg volatile suspended solids (VSS)/L. The BAF reactor consisted of a 2.0 m high acrylic cylinder (inner diameter, 0.10 m) and was filled with the cell-entrapped PEG media (packing, 75 %). A steel sieve was installed at both upper and lower parts of the reactor to prevent the media washout. The BAF was operated in an upflow feeding mode using a peristaltic pump (Masterflex, USA) at the empty bed contact time (EBCT) of 8 h. Backwashing was performed using a diaphragm pump (backwashing flow rate, 5 L/min). The dissolved oxygen (DO) concentration inside reactor was above 3 mg/L (air flow rate, 0.5 L/min). The vessel used to feed dyeing wastewater into reactor was maintained at 4 °C using a cooler to reduce the microbial contamination.

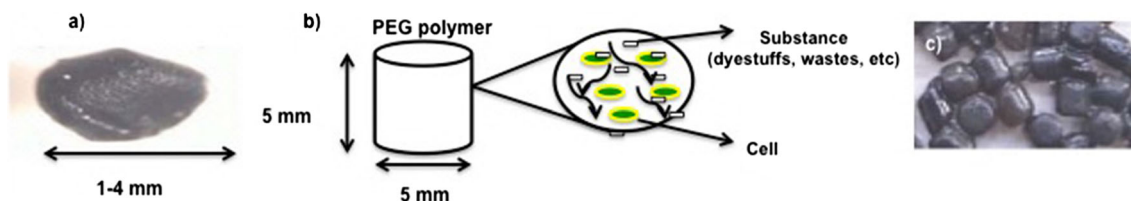
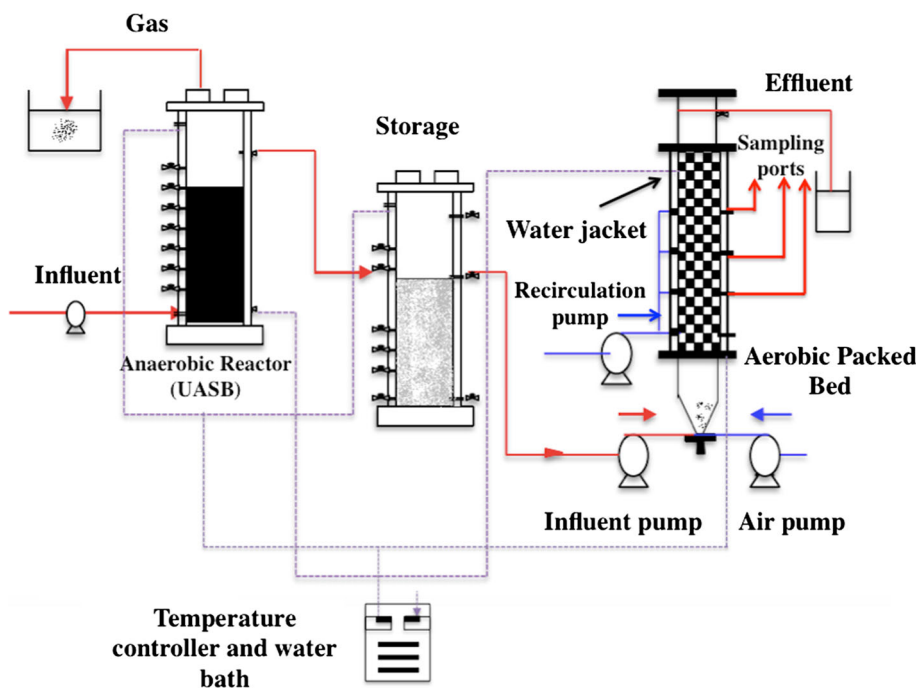
### Characteristics of dyeing wastewater

The dyeing wastewater used was obtained from the equalizer after the screening followed by the pH adjustment at a dyeing wastewater treatment plant in Ansan, Korea. The characteristics of dyeing wastewater were as follows: plant design treatment capacity, 100,000 m<sup>3</sup>/day; influent pH, 7.5 ± 0.5; oxidation reduction potential (ORP), 60 ± 28 mV; total chemical oxygen demand (TCOD), 869 ± 67 mg/L; soluble COD (SCOD), 720 ± 68 mg/L; total biochemical oxygen demand (TBOD), 286 ± 59 mg/L; soluble BOD (SBOD), 216 ± 44 mg/L; and color, 1288 ± 196 ADMI (American Dye Manufacturers Institute) units.

### Microbial immobilization

Microorganisms capable of degrading refractory compounds, isolated from the above mentioned dyeing wastewater treatment plant, were first suspended in a PEG solution containing additive (inorganic compound), cross-linker (D-sorbitol), and promotor (*N,N,N,N'*-tetramethylethylenediamine), and mixed thoroughly with an

**Fig. 1** Schematic of the sequential anaerobic/aerobic reactor process



**Fig. 2** Pictures of **a** anaerobic sludge granule ( $\times 100$ ), **b** concept of cell-immobilized media, and **c** cell-immobilized media

initiator (potassium persulfate). The composition of immobilizing materials was 18 % (w/v) PEG prepolymer, 40 % (w/v) additive, 0.9 % (w/v) cross-linker, 0.5 % (w/v) promoter, 0.25 % (w/v) initiator, and 5 % (w/v) mixed bacterial culture capable of degrading refractory compounds. The polymerization method used was a modification from Sumino et al. (1992). After the addition of initiator, the mixture was immediately passed through a polyvinyl chloride (PVC) tube (inner diameter, 4 mm) and left at room temperature for about 10 min. The subsequently obtained elastic gel containing microorganisms was extruded from the PVC tube and cut at the length of 4 mm, producing pellets of uniform shape (porosity,  $\leq 0.5 \mu\text{m}$ ) (Fig. 2b). The pellets were washed thoroughly in water before use. All the chemicals used for polymerization were purchased from the Sigma-Aldrich Chemical Co. (St. Louis, MO, USA).

**Evaluation of color removal at different HRTs**

To investigate the effect of different hydraulic retention time (HRT: 24, 8, and 6 h) on the decolorization, the dyeing wastewater was fed to the UASB reactor followed by the BAF reactor using peristaltic pumps at various flow rates. At the end of reactor operation, the cell-immobilized media in BAF were taken out to remove the biomass grown/attached on the reactor wall and settled at the reactor outlet. After washing the media, the reactor was fed with tap water to the full capacity, followed by a new operational run at another HRT. The experimental runs were operated with a decreasing HRT from 24 to 6 h, corresponding to an increase in the loading rate.

### Evaluation of COD and color removal along the height of UASB reactor

To evaluate the effect of changes in sludge granule at different reactor heights, samples were taken from four different heights (up to 2, 4, 6, and 8 L working volume from the reactor bottom), and the changes in color, SCOD, pH, and ORP were measured.

### Evaluation of COD and color removal at different heights and EBCTs in BAF reactor

After the BAF reactor was operated using the effluent discharged from the preceding UASB reactor as feeding, samples were taken from four different heights (up to 20, 60, 100, and 140 cm from the reactor bottom) to investigate the characteristics of color and COD removals at different EBCTs (24, 12, and 8 h).

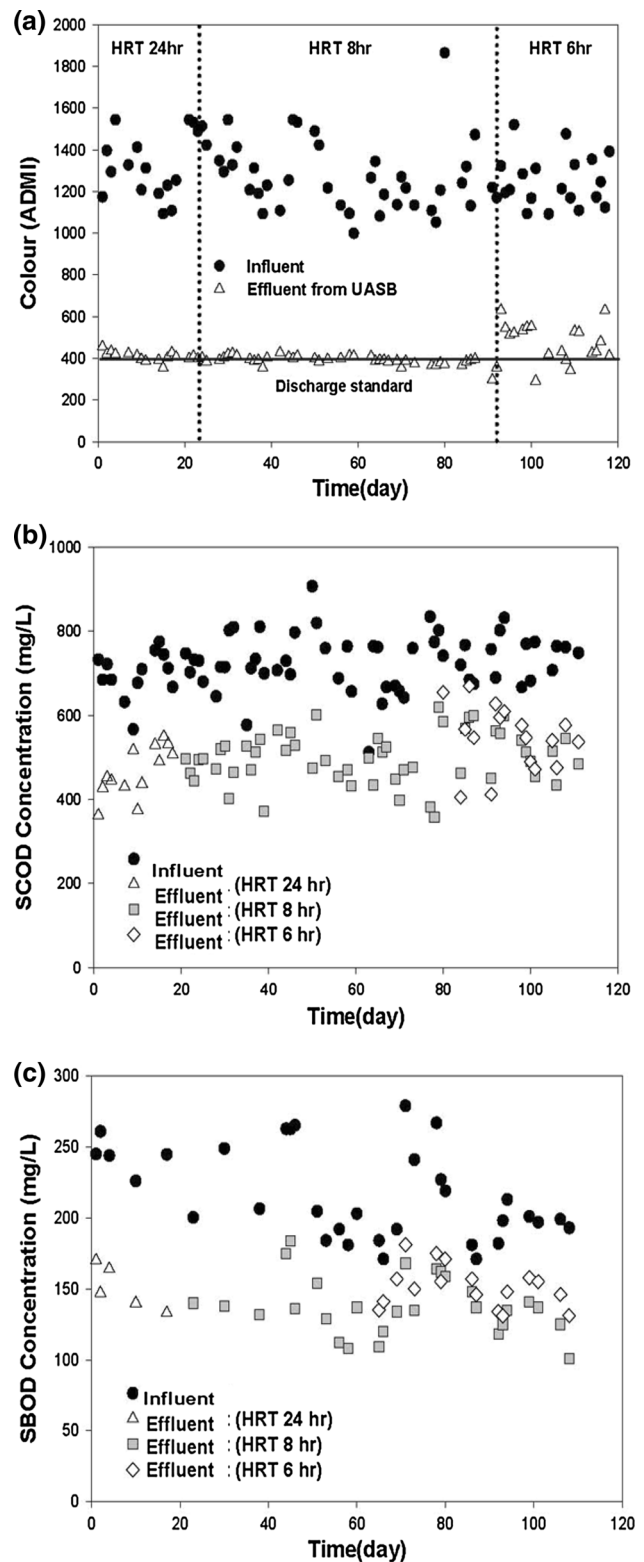
### Analysis

Samples were first filtered through a glass-fiber filter (Whatman, GF/C). BOD, COD,  $\text{NH}_4^+\text{-N}$ ,  $\text{NO}_2^-\text{-N}$ ,  $\text{NO}_3^-\text{-N}$ , total phosphorus (TP), SS (suspended solids), and color were measured following Standard Methods (APHA/AWWA/WEF 2005). The method and measurement accuracy was evaluated by blank (deionized water) and replicate analyses.

## Results and discussion

### Effect of HRT on refractory compounds removal in UASB reactor

Figure 3a shows the removal efficiency for color at different HRTs in the UASB reactor. The average effluent color was 397 ADMI at the early operational period (HRT = 24 h), with the removal efficiency of 69 %. Even though the HRT was reduced to 8 h after 20 days, the removal efficiency was still stable at 70 % (effluent color at 392 ADMI). When the reactor was operated with 6-h HRT, a relatively lower average removal efficiency of 63 % was obtained (effluent color at 473 ADMI). The obtained results are in accordance with Oh et al. (2004), where the UASB coupled with an activated sludge reactor was used to treat real textile wastewater (color, 500–1400 ADMI) at different HRTs (6, 12, and 18 h), with 65 % color removal at 18-h HRT. The decolorization of recalcitrant dyes in



**Fig. 3** Effect of HRT on **a** color, **b** SCOD, and **c** SBOD removal in the UASB reactor

anaerobic reactors is generally a slow process, which requires long HRTs to reach a satisfactory extent of color removal (Li et al. 2009). Keskinan and Lugal Gökso (2007) also stated that the variation in HRT is an important factor in dye treatment and the treatment performance increased with the increasing HRT in their study. Rodrigues da Silva et al. (2012) also observed that a short HRT (3 h) was not sufficient to allow a higher decolorization, suggesting that a short HRT was not long enough to allow the dye reduction satisfactory. According to the legal discharge limit for color intensity in Korea (400 ADMI; Oh et al. 2004), the developed biological methodology in current study can be potentially used to treat real dyeing wastewater. On the other hand, even though the anaerobic dye reduction could be achieved using different microorganisms, no single microbial strain has been reported able to decolorize a broad range of dyes. Therefore, the use of mixed cultures such as anaerobic granular sludge would be a more rational alternative (Shah 2015).

As shown in Fig. 3b, the influent SCOD was 723 mg/L and the effluent after anaerobic treatment was 462 mg/L (36 % removal) and 492 mg/L (32 % removal) at 24- and 8-h HRT, respectively. In comparison, at 6-h HRT, the removal efficiency decreased significantly to 24 % (547 mg/L in effluent). This lower COD removal efficiency also observed in others (Oh et al. 2004; Amaral et al. 2014) and can be associated with the presence of hardly biodegradable compounds as well as the possible toxic effects of dyestuffs and sodium chloride in real dyeing wastewater on the microbial activity. Wang et al. (2008) showed the degradation of some complicated pollutants present in printing and dyeing wastewaters was better using the UASB reactor (COD removal, 35 %) compared to the anaerobic acidogenic unit (COD removal, 4 %), at the same HRT (15 h). In their study, although the COD removal efficiency was only 35 %, the categories of alkanes were reduced by 75 % and consequently the UASB reactor was proven to be effective to degrade refractory compounds, resulting in the generation of intermediates (phenol, at 31 µg/L) more readily degraded in the subsequent anaerobic step. These findings are in accordance with the current study where the anaerobic unit was used as a pretreatment to improve the biodegradability of dyeing wastewater.

The removal efficiencies for SBOD were 39, 36, and 28 % at the HRT of 24, 8, and 6 h, respectively, despite the fluctuations of organic loading in influent (Fig. 3c). According to Frijters et al. (2006), it was almost impossible to quantify the dye concentrations and know its exact

composition at a given time due to the diversity of organic compounds present. However, since the largest group (60–70 %) of synthetic colorants is considered azo dyes (Isik and Sponza 2006; Popli and Patel 2015), it is still possible that such chromophores as azo group ( $-N=N-$ ) can be broken down into more easily biodegradable compounds during the anaerobic step, then to be further mineralized in the subsequent aerobic step. The exact mechanism for the dye reduction, whether occurring intracellularly or extracellularly, is still a subject of investigation (Shah 2015). In the current study, after color and COD removals were compared, the 8-h HRT was found suitable for the UASB reactor to reduce the energy cost and treat a larger amount of wastewater per unit time.

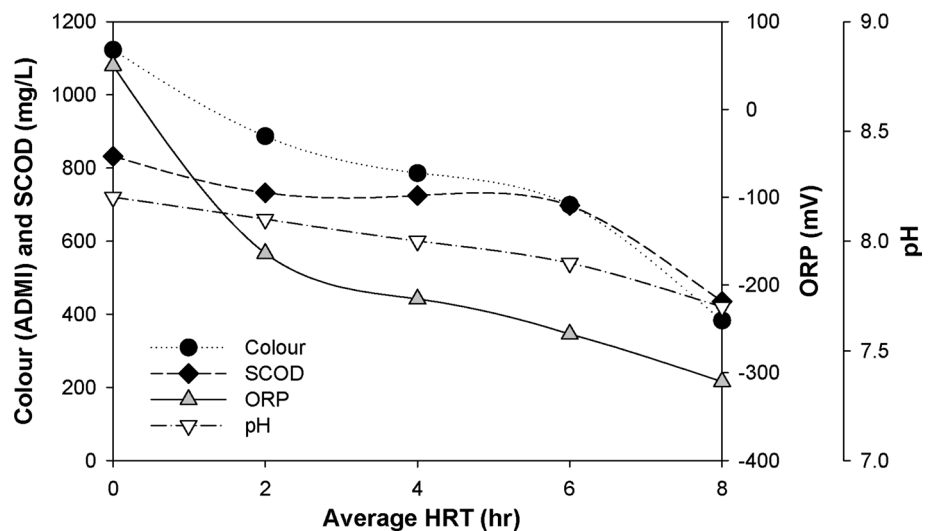
### Removal efficiency along the UASB reactor height

The changes in color, SCOD, pH, and ORP at different heights in the anaerobic reactor are shown in Fig. 4. The reactor was operated at 8-h HRT, and samples were taken from four different heights (up to 2, 4, 6, and 8 L from the reactor bottom), representing the average HRT passed from the bottom. The removal efficiency for color was approximately 20 % at the 2-h HRT location and increased continuously to 80 % as the HRT increased. The ORP decreased from  $-260$  to  $-310$  mV at the highest location (corresponding to the 8-h HRT), and the redox potential is in a good agreement with the standard redox potentials for different azo dyes, generally between  $-430$  and  $-180$  mV (Dubin and Wright 1975). The SCOD levels did not change much up to the 6-h HRT height, but a higher organic removal was observed at the 8-h HRT location. This result implies that the color formers in dyestuff were broken down at the 6- to 8-h HRT locations, and the hardly biodegradable compound could be converted to the easily biodegradable ones, resulting in the higher organic removal (Panswad et al. 2001). There was no significant pH variation during the organics removal along the UASB reactor height.

### Effect of EBCT on refractory compounds removal in BAF reactor

The BAF reactor was operated at different EBCTs (24, 12, and 8 h) to investigate the optimal EBCT for the effective removal of refractory compounds from dyeing wastewater. The changes of SCOD in influent (i.e., effluent from the UASB reactor) and effluent from the BAF reactor at each EBCT are shown in Fig. 5a. After the reactor reached a

**Fig. 4** Changes in color, SCOD, ORP, and pH at different heights in the UASB reactor



steady-state in about 20 days after the operation, the average influent SCOD concentration was 493 mg/L, while the average effluent concentrations at 24- and 12-h EBCT were 85 and 88 mg/L, respectively. The removal efficiencies were similar (83 and 82 %) despite the organic loading doubled. In comparison, the removal efficiency at 8-h EBCT decreased to 75 %. On the other hand, while the average SBOD concentration in effluent from the UASB reactor was 125 mg/L, the SBOD concentration in effluent from the BAF reactor was 8, 7, and 11 mg/L at 24-, 12-, and 8-h EBCT, respectively, indicating high removal efficiencies ( $\geq 90$  %) for the degradable compounds (Fig. 5b).

According to Ali et al. (2009), SS are important repositories for toxic heavy metals and dyes in the receiving water bodies, and for this reason, the impact of SS on water color was also evaluated. Figure 5c shows color and SS amounts in effluent from the BAF reactor at different EBCTs. Despite the significant fluctuations in influent, a significant amount of color was removed in the preceding UASB reactor (Fig. 3a), while just some color removal in the subsequent BAF reactor was observed. The color amount in effluent from the BAF reactor was almost constant regardless of increase or decrease of the effluent SCOD or SS concentrations. Therefore, color was considered not directly affected by the SS ( $\geq 1$   $\mu\text{m}$ ) as well as the change of SCOD. In comparison, when the effluent from the BAF reactor was coagulated with  $\text{FeCl}_3$ , color was further decreased to 300 ADMI, suggesting that color was more related to the colloidal particulate matters. These results are also in accordance with others (O'Neill et al. 1999; Oh et al. 2004; Hakimelahi et al. 2012) where the main reduction of dyes occurred in the anaerobic phase and

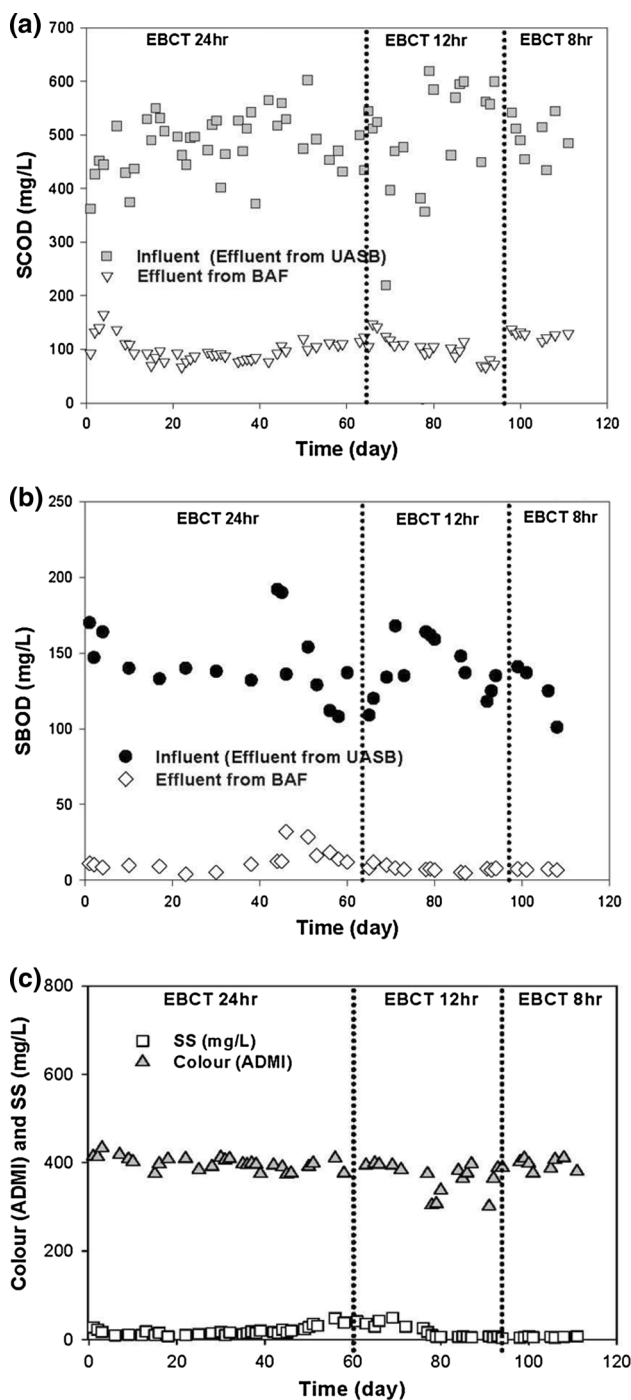
the contribution of aerobic phase to decolorization was negligible. The aerobic unit enhanced the SCOD and SBOD removals and was crucial to improve the effluent water quality. Kapdan and Oztekin (2006) also observed the aerobic phase provided improvement in COD removal and acted as a polishing step after the anaerobic color removal.

#### Removal efficiency along the BAF reactor height

As shown in Fig. 6a, at 20 cm height from the BAF reactor bottom, 65 % of total organics were removed at 24- and 12-h EBCTs, while 40 % removed at 8-h EBCT. The removal efficiencies generally increased as the EBCT increased, and 75 % removal efficiencies for SCOD were achieved from the reactor height of 20 cm, except for the 8-h EBCT. The SBOD concentration along the reactor height at the 8-h EBCT was below 10 mg/L from the 20 cm reactor height (Fig. 6b). Therefore, the removal of degradable organic compounds mainly occurred as soon as influent (effluent from the UASB reactor) was added to the BAF reactor.

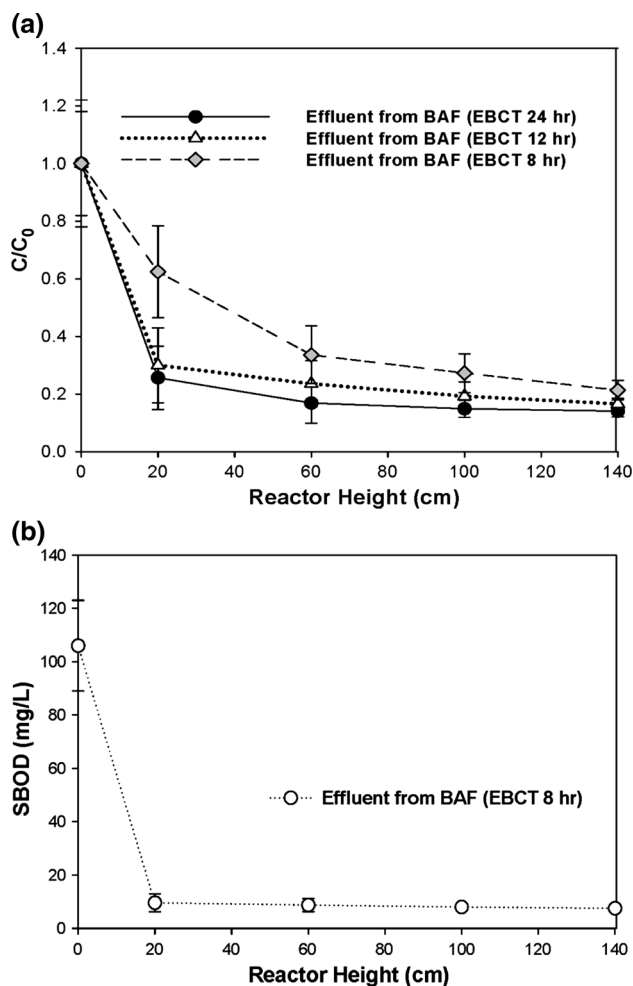
#### Evaluation of removal potential for phosphorus and nitrogen

The average TP concentration in effluent from the BAF reactor was 4 mg/L, while the average TN concentration was 28, 29, and 15 mg/L in influent, effluent from the anaerobic reactor, and effluent from the aerobic reactor, respectively. The higher  $\text{NH}_4^+\text{-N}$  concentration in effluent from the UASB reactor was probably due to the biological reduction of chromophores groups ( $-\text{N}=\text{N}-$ ) generating

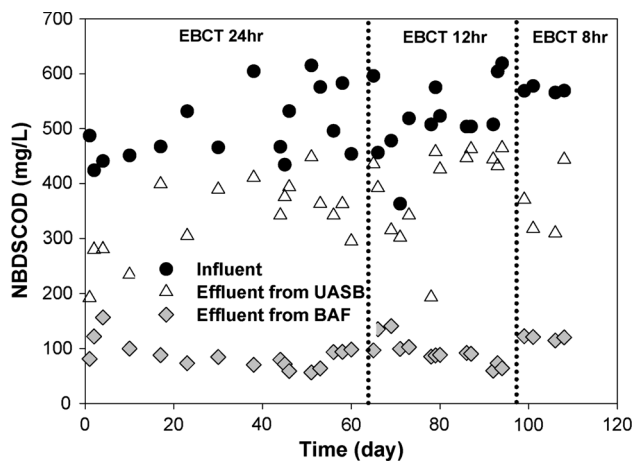


**Fig. 5** Effect of EBCT on **a** SCOD, **b** SBOD, and **c** color and SS removal in the BAF reactor

aromatic amines as most  $\text{NH}_4^+-\text{N}$  was measured as TN. The  $\text{NO}_3^--\text{N}$  concentration, on the other hand, was about 10 mg/L in influent, while it was not detected in effluent from the UASB reactor but was about 7.5 mg/L in effluent



**Fig. 6** Removal of **a** SCOD and **b** SBOD at different heights in the BAF reactor



**Fig. 7** Effect of EBCT on non-biodegradable (NBD) SCOD concentrations in the sequential anaerobic/aerobic reactor

from the BAF reactor, further implying the occurrence of nitrification in the aerobic process.

### Behavior of non-biodegradable SCOD

In order to evaluate the removal efficiency for recalcitrant matters, the non-biodegradable (NBD) SCOD was measured for influent, effluent from the UASB reactor, and effluent from the BAF reactor, at different EBCTs, and the results are shown in Fig. 7. The NBDSCOD was calculated as soluble ultimate BOD ( $SBOD_u$ ) subtracted from SCOD, and the  $SBOD_u$  value was assumed 1.5 times of SBOD. The average influent NBDSCOD concentration was 517 mg/L and the average concentration in effluent from the anaerobic reactor operated at 8-h HRT was 363 mg/L, suggesting the removal of some recalcitrant matters together with degradable matters. The average NBDSCOD concentration in effluent from the aerobic reactor was 87, 93, and 118 mg/L, with the removal efficiency of 76, 74, and 67 %, at 24-, 12-, and 8-h EBCT, respectively (Fig. 7). Despite the loading of recalcitrant matters gradually increased as the EBCT decreased, the aerobic reactor still showed a high removal efficiency ( $\geq 65$  %) for the NBDSCOD, regardless of the EBCT applied, and this was considered due to the superior effect of media immobilized with microorganisms in degrading recalcitrant matters present in dyeing wastewater. Compared to the suspended microbial technology, cell immobilization shows many advantages such as high biomass, high metabolic activity, and strong resistance to toxic chemicals, and could be cost effective since the immobilized microbes can be used several times without significant loss of activity (Martins et al. 2013). The immobilization and maintenance of high concentration of biomass are main factors that improve the bacterial tolerance to the toxic substances present in dyeing wastewater (Pagga and Brown 1986). In the current study, the immobilized microbes were essential to successfully reduce COD and toxicity of the UASB effluent. The developed sequential anaerobic/aerobic treatment system can be effective in decolorizing textile wastewater as well as in reducing organic loading to meet the effluent discharge limits due to its cost effective and environmentally friendly nature.

### Conclusion

The anaerobic UASB process seeded with the sludge granule, followed by the aerobic BAF process packed with the cell-immobilized PEG media in sequence was

employed to treat real dyeing wastewater and the removal efficiencies for refractory compounds were evaluated. The optimal HRT for the stable color removal in the UASB reactor was 8 h and the highest color removal occurred at the 6-h HRT location corresponding to the respective reactor height. The removal efficiencies for SCOD and SBOD in the BAF reactor was  $\geq 75$  %, occurred at the lower part of reactor, regardless of EBCTs. The refractory compounds present in dyeing wastewater could be effectively removed using the sequential continuous anaerobic/aerobic process. Most color was removed during the anaerobic process, while refractory compounds were effectively removed during the aerobic process. The UASB/BAF filled with cell-immobilized PEG media was used for the first time to treat real dyeing wastewater and showed the high removal efficiency for non-biodegradable SCOD. This integrated anaerobic/aerobic reactor process is considered an effective and feasible treatment technology for dyeing wastewater.

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### Compliance with ethical standards

**Conflict of interest** The authors declare that they have no conflict of interest.

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