Azo dye decolorization in an up-flow bioelectrochemical reactor with domestic wastewater as a cost-effective yet highly efficient electron donor source

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ABSTRACT

A major challenge of employing bioelectrochemical system (BES) for reductively degrading recalcitrant contaminants in industrial wastewater is lacking sufficient electron donors. In this work, domestic wastewater (DW) was demonstrated to efficiently drive BES for implementing the decolorization of azo dye, acid orange 7 (AO7). Side benefit was the simultaneous treatment of DW. Decolorization efficiency in BES fed with DW (R_{DW}) was found to be comparable with that either fed with glucose (R_{Glu}) or acetate (R_{Ac}). Much lower reductant usage ratio was observed in R_{DW} . As a result, when the ratio of electron donors to azo dye decreased to 4.4 mol COD mol⁻¹ AO7, DE of R_{DW} kept over 90% while DEs of R_{Ac} and R_{Glu} were significantly dropped due to the insufficient electrons donation. Besides serving as electron donor, DW was proved to also provide some conductivity and buffer capacity. Accordingly, DE of R_{DW} was less deteriorated when fully removing the external buffer slats. This study comprehensively revealed the feasibility and superiority of DW as a cost-effective electron donor source in BES and brings this technology closer to the practice.

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1. Introduction

Bioelectrochemical systems (BESs) have been suggested to be a promising technology in reductive degradation or transformation of diverse contaminants, including nitroaromatics (Mu et al., 2009b; Wang et al., 2011), azo dyes (Cui et al., 2016a, 2016c), halogenated compound (Kong et al., 2015; Liang et al., 2013), heavy metals (Huang et al., 2014; Luo et al., 2014), nitrate (Pous et al., 2014), etc. Although superior performance of BES compared to the traditional biological processes were extensively reported in these contaminants, this technology is still far away from the practice as most of the existed studies were performed at lab scale and under ideal conditions, such as strong buffered system, sufficient electron donors and excellent conductivity.

Azo dyes are one group of the most concerned contaminants in

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refractory wastewater treatment, because they account for more than half of the used dyestuffs in the textile industry which is highly water consuming (Cui et al., 2016b). Due to the low utilization efficiency, about 2–50% of these dyestuffs are unemployed and exist in the effluent (Ganesh et al., 1994), which is harmful attributed to the apparent color and the toxicity to the organisms (Selvam et al., 2003). The decolorization of azo dyes is a reductive process involved in four electrons transfer for breaking one azo bound (–N=N–). In BESs, electron donors (organic carbon) are oxidized by anode respiration bacteria (ARB) with electrons transfer to the anode and further going to the cathode for implementing the decolorization (Mu et al., 2009a). To date, a number of electron donors, including acetate, glucose, yeast extract, etc., have been demonstrated to drive the cathodic decolorization (Cardenas-Robles et al., 2013; Cui et al., 2014a, 2016a). However, these chemicals were external supplied, which could therefore cause the extra cost in operation. Compared to the addition of chemicals, using waste streams as electron donor source is obviously more cost effective and may gain the benefit of treating the waste streams simultaneously. Various kinds of waste streams (e.g. lignocellulose (Gregoire and Becker, 2012), digestate (Wang et al., 2014) and domestic wastewater), have been reported as the

Abbreviations

AN	1-amino-2-naphthol
AO7	Acid orange 7
ARB	Anode respiration bacteria
BES	Bioelectrochemical system
COD	Chemical oxygen demand
DE	Decolorization efficiency
DR	Decolorization rate
DW	Domestic wastewater
FID	Flame ionization detector
HRT	Hydraulic retention time
NaAc	Sodium acetate

NaCl	Sodium chloride
PBS	Phosphate buffer solution
R _{Ac}	BES fed with acetate
R _{DW}	BES fed with DW
R _{Glu}	BES fed with glucose
RSR	Reductant supply ratio
RUR	Reductant usage ratio
SA	Sulfanilic acid
SCE	Saturated calomel electrode
SS	Suspended solids
TCD	Thermal conductivity detector
VSS	Volatile suspended solids

efficient substrates for ARB in other types of BESs, such as microbial fuel cell for electricity generation and microbial electrolysis cell for hydrogen production (Heidrich et al., 2014; Kim et al., 2015). In case of driving azo dyes decolorization, domestic wastewater (DW) seems more adaptable because it is readily accessed. In addition, most of the textile factories were located close to cities or towns and their discharged wastewater is usually treated together with DW (Chen et al., 2011). Thus, to understand the performance of DW as electron donor source for decolorization in BESs is crucial in regard to practicing this technology.

Up to now, DW as electron donor source for azo dye decolorization in BESs has not been systemically studied. Although the feasibility of DW as electron donor source has been reported (Kim et al., 2015), the performance was not compared with other extensively used electron donors under identical conditions and consequently a fair evaluation of the efficiency is difficult. Additionally, unlike the solid organics as electron donor, DW would dilute the original azo dye containing wastewater. The decrease of conductivity and buffer capacity may influence the decolorization efficiency. This issue is also worth to be clarified before DW is capable of serving as an eligible electron donor source in BESs.

According to the concerns as above, here, the decolorization performance with DW as the electron donor source in BES was systemically investigated. Conventional external carbon sources, acetate and glucose, were employed as comparisons. The effects of azo dye loading rate, electron donor concentration and the external buffer salts on decolorization efficiencies were studied under these three different electron donor source conditions. The perspective of using DW as electron donor source for driving BES in practice was discussed as well.

2. Material and methods

2.1. Reactor configuration

Three identical cylindrical reactors were manufactured with plexiglass (schematic representation of the reactors were shown in Supporting Information (SI), Fig. S1). Each one had a working volume approximately 1.25 L with the inner diameter of 8 cm and a height of 25 cm. Two pairs of electrodes were installed into the reactor, from the bottom to the top, which were arranged as down-cathode, down-anode, up-cathode and up-anode with the distance of 2.5 cm between each electrode. Both anodes and cathodes were constructed by granular graphite (diameter from 3 to 6 mm, Linzhang county Deyuan carbon co., LTD, Handan China) with an apparent size of 8 cm in diameter and 4 cm in height, giving the total electrode volume of 200 cm³. Before using, the granular

graphite was washed by soaking in 32% HCl for four times to remove the foreign materials (Mu et al., 2009a). Graphite rod ($\Phi = 4$ mm) penetrated into the electrode zone and worked as an electron collector. A saturated calomel electrode (SCE, +247 mV vs. standard hydrogen electrode, model-217, Shanghai Precise Sci. Instru. Co., Ltd. China) was used as the reference electrode. During all of the experiments, voltage of 0.5 V was supplied between anode and cathode by a DC power supply (FDPS-180, Fudan Tianxin Scientific and Educational Instruments Co., Ltd, Shanghai, China). A 10 Ω resistor was connected in series into the electric circuit. The voltages cross this resistor were recorded every 10 min by a data acquisition system (Keithley 2700, Keithley Co. Ltd., U.S.), which were automatically converted to current according to Ohm law.

2.2. Chemicals

Acid orange 7 (AO7) was used as the model azo dye (purity > 95%, Shanghai Sangon Biotech Co., Ltd., China). The products of AO7 reduction were sulfanilic acid (SA) and 1-amino-2-naphthol (AN) (Mu et al., 2009a). In this study, SA (primary reagent, Aladdin Industrial Corporation) was used to calculate the product formation efficiency. All other chemicals were analytical reagent.

2.3. Startup and operational conditions

All of the three reactors were startup in batch mode. Effluent that collected from a long-term operated single chamber BES was amended with sodium acetate (NaAc, 1000 mg l⁻¹) and AO7 (200 mg l⁻¹) as the inoculum solution. In the first batch operation cycle, 50 mL anaerobic activated sludge (SS of 35.57 g l⁻¹ and VSS of 17.11 g l⁻¹) was also added into the reactors to strengthen the biomass. After that, the inoculum solution was replaced every two days. When a stable current output was observed, the reactors were considered to be successfully started up.

Subsequently, the operation of reactors was changed to continuous flow mode. Domestic wastewater (DW) collected from a local sewage well was filtrated by a 400-mesh sieve to remove particles that might block the interspace of electrodes. COD of the filtrated DW was detected as 309 ± 18 mg l⁻¹ in all experiment, and other key factors of the DW are listed in SI, Table S1. DW was then amended with AO7 (200 mg l⁻¹) as the influent of one reactor (R_{DW}). As the controls, the other two reactors (R_{Ac} and R_{Glu}) were fed with the synthetic wastewater with acetate and glucose as the electron donor source, respectively, which contributed the same COD as that in DW. Besides the electron donor sources, the synthetic wastewater was composed of AO7 (200 mg l⁻¹) and other minerals as reported previously (Cheng et al., 2015).

The performances of three reactors were evaluated under various hydraulic retention times (HRTs), concentrations of electron donor source and buffer capacities of the wastewater, which are summarized as in Table 1. In the Stage 1, the influents of all reactors were buffered with 50 mM phosphate buffer solution (PBS, pH ~7.2). The concentration of electron donor source was set at ~300 mg COD l⁻¹. HRTs were fixed at 24 h, 12 h, 8 h and 6 h, respectively. In the Stage 2, the influents were buffered with PBS as well and fed into the reactors with a constant HRT of 6 h. The concentrations of electron donor source were varied at 240, 180, 120 and 80 mg l⁻¹. In the Stage 3, HRT and the concentration of electron donor source of the influents were respectively constant at 6 h and 300 mg l⁻¹. Effect of PBS on the AO7 decolorization was conducted by operating reactor with 50 mM and without PBS, another experiment was set to distinguish the effect of buffer capacity and conductivity by replacing the PBS with sodium chloride (NaCl), in which the dosing of NaCl provided the conductivity equal to 50 mM PBS. All of the experiments mentioned above were carried out at ambient temperature (23 ± 2 °C). For each condition, the reactors were operated for at least 12 HRTs. Data analysis was based on at least 5 samples from different HRTs in each steady state.

2.4. Analytical methods and calculations

Liquid samples taken from reactors were immediately filtered through the 0.45 µm filters (Tianjin Jinteng Experiment Equipment Co., Ltd., China). AO7 concentration was quantified by a UV–Vis spectrophotometer (UV-1800, Shanghai Meipuda instrument Co., Ltd., China) at a wavelength of 484 nm (Mu et al., 2009a). SA was measured by a high performance liquid chromatography (HPLC, e2695, Waters Co., U.S.) equipped with a UV–Vis detector (model 2489, Waters Co., U.S.) and a C18 column (5 µm; 4.6 mm × 250 mm, Symmetry, Waters Co., Ltd., U.S.). To prevent the auto-oxidation of the products from AO7 reduction, 1000 mg l⁻¹ Na₂SO₃ solution was added to dilute liquid sample by 1:1. Gas composition was analyzed by a gas chromatograph (GC, 7890A, Agilent, Inc., US) equipped with a flame ionization detector (FID) and a thermal conductivity detector (TCD). Chemical oxygen demand (COD) determination by using the HACH vials. pH and conductivity were measured with FE20-FiveEasy™ pH meter (Mettler Toledo, China) and Bante 950 conductivity meter (Bante, China), respectively. Calculations including AO7 decolorization efficiency (DE, %), decolorization rate (DR, g m⁻³ d⁻¹), reductant usage ratio (RUR), reductant supply ratio (RSR) and decolorization product (SA) recovery efficiency (η_{SA}, %), are shown in SI.

3. Results and discussion

3.1. Decolorization performance with domestic wastewater as electron donor source

With the decrease of HRT, AO7 loading rates increased from 200 to 800 g m⁻³ d⁻¹ as shown in Fig. 1. Decolorization efficiencies (DEs) of all three reactors were always over 98%, resulting in the increase of decolorization rates (DRs) almost linear with the AO7 loading rate ($R^2 > 0.998$), and finally reached to the range between

Fig. 1. Effect of electron donor source types (domestic wastewater, NaAc and glucose) on AO7 decolorization efficiency at different loading rates.

795.05 ± 60.78 and 840.96 ± 49.99 g m⁻³ d⁻¹ (SI, Fig. S2). In order to confirm the decolorization of AO7 was due to the cleavage of the azo bound, one of the reduced products, sulfanilic acid (SA), was measured in the effluents (Yemashova et al., 2004). As shown in SI, Fig. S3, SA accumulations waved in a narrow range (86–98 mg l⁻¹), accounting to over 87% of the removed AO7 in all tested conditions. Current generation was continuously monitored during the decolorization. The currents of the two pairs of electrodes in each reactor were summed and presented in Fig. 2. As the DEs were always closed to 100% despite of the AO7 loading rates, the current generation was likely controlled by the availability of AO7 as the electron acceptor which showed an increase trend by shorting HRT. In each HRT condition, the types of electron donor source revealed non-significant influence on AO7 decolorization in terms of DR and product accumulation. The decolorization performance of R_{DW} presented here is also comparable to other reported BES fed with simple electron donor sources (NaAc and glucose) at similar azo

Fig. 2. Currents of domestic wastewater, NaAc and glucose feeding BESs at different loading rates.

Table 1

Operational conditions of hydraulic retention time (HRT), electron donor source concentration and buffer system in this experiment.

Stage	HRT (h)	Electron donor source concentration (mg l ⁻¹)	Buffer system
1	24, 12, 8, 6	~300	50 mM PBS
2	6	240, 180, 120, 80	50 mM PBS
3	6	~300	0 PBS & NaCl (conductivity = 50 mM PBS)

dye loading rate (Cui et al., 2014b). These results clearly indicated that those complicates organics contained in DW were capable of efficiently drive BES to reductively decolorize azo dye as compared to the conventionally used simple organics.

Unlike to the fate of AO7, the changes of COD in reactors fed with different electron donor sources were shown significantly different (Fig. 3). Because the DEs of all reactors were almost the same and the reduced products of AO7 are stable under anaerobic condition (Yemashova et al., 2004), the variety of COD removal was most likely the result of the different consumption of the electron donor sources. The removed COD in R_{DW} was shown as the lowest, which was followed by the R_{Glu} and then by the R_{Ac} . Although the COD removal in R_{DW} was not the highest, yet it clearly demonstrated the simultaneous treatment of DW. The RURs of R_{DW} were estimated from 9.31 ± 0.86 to 6.00 ± 0.94 mol COD mol⁻¹ AO7 with the loading rates of AO7 increasing from 200 to 800 g m⁻³ d⁻¹. In case of glucose as electron donor source, more reductant was consumed with the RURs various from 10.29 ± 1.67 to 11.12 ± 0.28 mol COD mol⁻¹ AO7. In R_{Ac} , the RURs further increased to the range between 16.03 ± 0.96 and 11.37 ± 1.01 mol COD mol⁻¹ AO7. Theoretically, the reduction of 1 mol AO7 requires 4 mol electron equivalents, corresponding to the RUR of 1 mol COD mol⁻¹ AO7. In despite of the types of electron donor sources, the obtained RURs were higher than this theoretical value, indicating not all of the electrons of the electron donor sources flowed to the AO7 but lost through certain parallel routes, such as the biomass production and the unwanted methanogenesis (van der Zee and Villaverde, 2005).

In order to get insight into the possible electron loss, electron balance was analyzed at the AO7 loading rate of 800 g m⁻³ d⁻¹ (Table 2). Because of the similar decolorization efficiencies, electrons that flowed to azo dye were close in all reactors. Gas production and the composition analysis were conducted as well. In all reactors, the gas phases were mainly composed by CH₄ with infinitesimal H₂ and CO₂ (two orders of magnitude lower than CH₄). The rate of CH₄ production in R_{DW} was 0.29 ± 0.06 mmol l⁻¹ influent (equal to 2.32 mmol electrons l⁻¹ influent), which was much lower than that in R_{Glu} (0.50 ± 0.07 mmol l⁻¹ influent or 4.00 mmol electrons l⁻¹ influent) and R_{Ac} (1.26 ± 0.02 mmol l⁻¹ influent or 10.08 mmol electrons l⁻¹ influent). The dissolved CH₄ (Table 2) were estimated according to the Henry's law based on the CH₄ volume fraction in gas phases (Sander, 2015; Yeo et al., 2015) (details can be found in SI). In addition to methanogenesis, electrons can be also used to supporting bacteria growth. Electrons sunk in assimilation were estimated according to the substrate based yield coefficients and shown in Table 2 (Batstone et al., 2002; Filipe et al. 1998; Ren and Wang, 2004). By comparing the

Table 2

Electron balance analysis in domestic wastewater, NaAc and glucose feeding BESs at AO7 loading rate of 800 g m⁻³ d⁻¹.

	R_{DW}	R_{Ac}	R_{Glu}
Electron consumption			
AO7 decolorization ^a	2.27	2.24	2.27
CH ₄ collected in gas phase ^b	2.32	10.08	4.00
CH ₄ dissolved in aqueous phase ^c	4.06	7.72	5.36
Microbial growth ^d	0.06	1.37	2.41
Electron supply			
Removed COD ^e	13.61	27.31	24.12
Electron recovery efficiency			
η (%)	64.00	78.40	58.21

Unit (exclude electron recovery efficiency) is mmol electrons l⁻¹ influent.

^a 1 mol AO7 decolorization consumed 4 mol electrons.

^b 1 mol CH₄ contributed 8 mol electrons.

^c According to Henry's law as shown in SI (Sander, 2015; Yeo et al., 2015).

^d Microbial yield, $Y_{DW} = 0.0044$, $Y_{Ac} = 0.05$, $Y_{Glu} = 0.1$, biomass as COD/consumed COD (Batstone et al., 2002; Filipe et al. 1998; Ren and Wang, 2004).

^e 1 mol COD released 4 mol electrons.

consumed electron equivalents (COD), electrons that sunk in decolorization, methanogenesis and assimilation could account for 78.40%, 58.21% and 64.00% of those released from COD removal in, R_{Ac} , R_{Glu} and R_{DW} , respectively. Here, the relative poor electron balance observed in R_{Glu} and R_{DW} is likely because that the fermentation of DW and glucose is not taken into account. As shown in Table 2, R_{Ac} has the highest CH₄ production rate as expected because acetate is one of the few substrates catered to the methanogens (Chen et al., 2011; Khan et al., 2014). Compared to R_{DW} , the more produced CH₄ in R_{Ac} could explain over 80% of its more consumed COD. R_{Glu} also produced more CH₄ than R_{DW} . When taking into account its higher biomass yield, these two aspects could explain around half of the more consumed COD. These results clearly indicated that the greater RURs in R_{Ac} and R_{Glu} compared to R_{DW} were attributed to more electrons lost into the side reactions.

3.2. Decolorization performance with decreasing electron donor source concentrations

DEs kept above 90% regardless of the types of electron donor sources till the concentration (as COD) dropped to about 120 mg l⁻¹ (Fig. 4). At the lower COD of the electron donor sources (80 mg l⁻¹), DE of R_{DW} was almost not change, while 9.33% and 19.92% decrease of DEs were observed in R_{Glu} and R_{Ac} , respectively. Similar to that the electron donor source concentration was 300 mg COD l⁻¹, R_{DW}

Fig. 3. COD consumptions and reductant usage ratios (RURs) of domestic wastewater, NaAc and glucose feeding BESs at different loading rates.

Fig. 4. Effect of domestic wastewater, NaAc and glucose concentrations on AO7 decolorization efficiency at AO7 loading rate of 800 g m⁻³ d⁻¹.

consumed less COD compared to R_{Glu} and R_{Ac} in case of decreasing the dosage of electron donor source (Fig. 5). The estimated RURs of all three reactors were declined rapidly by reducing the electron donor source concentration from 240 to 80 mg COD $\cdot$ l $^{-1}$ and then were respectively stable around 3.4, 5.6 and 5.8 mol COD mol $^{-1}$ AO7 for R_{DW} , R_{Glu} and R_{Ac} , which suggested the minimum required electron equivalents for AO7 decolorization under different electron donor source conditions. Compared to the reductants supply ratios (RSRs) that were theoretically calculated according to the concentrations of AO7 and the electron donor sources, RURs of all reactors were lower than the corresponding RSRs when the concentration of electron donor sources was set at or higher than 120 mg l $^{-1}$, indicating the provided electron equivalents was more than that required. When the concentration of electron donor source was further reduced to 80 mg l $^{-1}$, RSR decreased to 4.4 mol COD mol $^{-1}$ AO7 which became lower than the RURs obtained in R_{Glu} and R_{Ac} but was still higher than that in R_{DW} . This result clearly indicated that the decline of DEs in R_{Glu} and R_{Ac} was due to the insufficiency of electron donor sources.

3.3. Effect of buffer systems on the decolorization performance

PBS is the commonly used buffer system in bioelectrochemical studies, which could provide a desirable conductivity and buffer capacity for the systems. However, any external additions of buffer salts would increase the operation cost and may be even worse by introducing secondary pollutions. Based on this concern, all of the reactors were further tested by feeding synthetic wastewater without PBS. As shown in Fig. 6A, DE of R_{DW} was slight decreased by 5.94% to $93.52 \pm 0.74\%$, while DE of R_{Ac} and R_{Glu} were dramatically deteriorated to around 80%.

In absence of PBS, the effluent of R_{Ac} basified with pH increasing to 7.83 ± 0.09 , which was likely the result of the consumption of H^{+} in methanogenesis that cannot be supplemented through the buffer effect (Fig. 6B). Similarly, when reactor was fed by glucose, fatty acids can be produced by fermentation and therefore caused the decrease of effluent pH to 5.68 ± 0.13 . Unlike to R_{Ac} and R_{Glu} , effluent pH of R_{DW} remained stable without adding buffer salts. This pH stability was found to attribute to the natural buffer capacity of DW. As shown in SI, Fig. S4, the buffer capacity was defined as the pH change along with adding 1 mL 0.1 M HCl/NaOH into 100 mL synthetic wastewater. The pH of DW composited influent only changed 1.04/0.91 units with HCl/NaOH adding, which was much lower than that of the acetate (2.00/3.21 units)

and glucose composited (3.82/3.13 units) ones.

In addition to the alteration of pH, the conductivities of influents decreased a lot due to the absence of PBS (Fig. 6C). Nevertheless, the conductivity of the influent with DW as electron donor source was 1.4 and 2.6 times higher than that with acetate and glucose, respectively. The higher conductivity in DW here is likely due to its more complicate composition compared to the defined synthetic wastewater, especially those inorganic salts and VFAs in DW could considerably contribute the conductivity (Cook et al., 2015; Scott et al., 2007).

In order to distinguish the effects of pH and conductivity on the decolorization performances, additional experiment was conducted by replacing PBS using NaCl which provided the same conductivity as 50 mM PBS. As shown in Fig. 6B, pHs in all effluents were almost not change after replacing by NaCl, confirming the buffer capacities was not improved. Over 90% recoveries of decolorization efficiencies as well as the currents (Fig. 6D) were observed in R_{DW} and R_{Ac} , clear indicating their decolorization deterioration in absence of external salts owed to the limited bioelectrochemical process because of the low conductivity other than the pH change. In R_{Glu} , although the conductivity was almost fully recovered after adding NaCl, the corresponding DE and current were just recovered by 86% and 77%, respectively. This result suggested the limitation of bioelectrochemical decolorization in R_{Glu} fed with PBS free influent attributed both to the variation of conductivity and pH. Because of the natural higher conductivity and buffer capacity of DW, the current drop of R_{DW} was lower than that of R_{Ac} and R_{Glu} under external buffer salts free condition, and consequently explained the corresponding higher decolorization efficiency.

3.4. Implications

Decolorization of textile wastewater was suggested to be limited owing to the lack of efficient electron donor (Şen and Demirel, 2003). Compared to external adding electron donors, such as glucose and VFAs, DW is obviously more cost-effective. However, RURs in traditional anaerobic decolorization process are usually at the value from tens to hundreds, while the COD as electron donor source in DW is low. For a typical azo dye wastewater treatment plant, the scale is usually at 10,000 tons per day with the concentration of azo compound at 1 mM. To meet the average RUR (50 mol COD mol $^{-1}$ azo dye) in conventional anaerobic process, the required DW (COD is assumed as 300 mg l $^{-1}$) would be 5.3 times in amount higher than the textile wastewater. According to the typical personal wastewater discharge constant (180 l d $^{-1}$), the required DW equals to that contributed by a population of 294,000. This huge requirement of DW reduces the accessibility of DW because of the population limitation and the high cost of DW transportation. In case of BES, the RUR for decolorization has been reported to be much lower than conventional anaerobic process when using simple organics as electron donor (as shown in SI, Table S2). Here, we further demonstrated this excellent property of BES remained in DW as electron donor source. Based on the assumption as mentioned above, the mixture ratio of DW and textile wastewater can be as low as to 0.36 according to the experimental data in this work (3.4 mol COD mol $^{-1}$ azo dye), which is 14.7 times lower than that calculated according to conventional anaerobic process and therefore considerably facilitate DW as the electron donor source. In this case, DW capacity of at least 3600 tons per day can be simultaneously treated by serving as electron donor source for driving azo dye wastewater decolorization.

In practical, the concentration of azo dyes in textile waste and COD in DW could be varied and therefore may lead a loading shock to the BES. In the present work, high DE remained even the RSR decreased to 4.4 mol COD mol $^{-1}$ azo dye in R_{DW} , which provide an

Fig. 5. COD variations, reductant usage ratios (RURs) and reductants supply ratios (RSRs) at different concentration of domestic wastewater, NaAc and glucose at AO7 loading rate of 800 g m $^{-3}$ d $^{-1}$.

Fig. 6. Effect of buffer systems on: (A) Decolorization efficiency; (B) pH; (C) Influent conductivity; (D) Current at A07 loading rate of $800 \text{ g m}^{-3} \text{ d}^{-1}$.

endurable bottom line of RSR for operating BES efficiently. This bottom line can be used as an alert in case of loading shock or guide the mixing ratio of DW and azo dye containing wastewater into an acceptable range.

In regard to a BES, conductivity of the wastewater is often considered to significantly influence the performance (Nam et al., 2010). Unlike to adding solid electron donors, DW as electron donor source would dilute the original azo dye containing wastewater and consequently can decrease the conductivity in mixed wastewater, which could ostensibly make the DW as electrode donor source doubtful. In the present work, we did observe the deterioration of decolorization performance under PBS free condition. However, as DW naturally has certain conductivity, the decolorization efficiency just decreased a little and remained over 90%. This result further strengthens the feasibility of using DW to drive the azo dye decolorization in BES.

4. Conclusion

This work demonstrated that domestic wastewater (DW) could efficiently drive azo dye decolorization in BES. Compared to the conventional added sodium acetate and glucose, DW as electron donor source showed comparable decolorization performance under high RSR condition yet is obviously much more cost-effective and could gain side benefit of the simultaneous treatment of DW. In addition, BES fed with DW consumed less reduced equivalent, which can favor the decolorization under low RSR condition. Moreover, DW can also provide some buffer capacity and conductivity, enabling less deteriorated when fully removing the external buffer slats. These findings comprehensively revealed the feasibility and superiority of DW as an electron donor source in BES and illuminate the practical potential of this technology in wastewater treatment.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.watres.2016.09.027>.

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