

# Evaluation of the design of anaerobic-aerobic systems for treating azo dye-containing effluents

*Avaliação da concepção de sistemas anaeróbios-aeróbios para tratamento de efluentes contendo corantes*

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## ABSTRACT

This study investigated the treatment of a synthetic effluent containing the azo dye Reactive Black 5 (50 mg·L<sup>-1</sup>) in acidogenic reactors followed by different aerobic post-treatment units, namely, moving bed biofilm reactors (MBBRs) and trickling filters (TFs). The effect of the addition of the redox mediator anthraquinone-2-sulfonate (AQS) (50 μM) to the system, hydraulic retention time (HRT) (6 and 4 h) in the MBBRs, and type of support medium (polyurethane foam cubes and K1 biomedica) in the TFs was evaluated. The acidogenic reactors were mainly responsible for the decolorization of RB5 in the anaerobic-aerobic designs evaluated in this study, and AQS significantly improved their decolorization efficiency. The use of a shorter HRT (4 h) in the MBBRs negatively influenced the color and COD removal performance in the reactors. Polyurethane foam proved to be more viable as a support medium, as it is a more accessible and low-cost material. Finally, acidogenic reactors followed by TFs filled with polyurethane foam cubes seemed to be the most promising design, both in terms of removing color, COD, and ammonia, and concerning the energy demand for system aeration.

**Keywords:** acidogenic reactor; color removal; MBBR; redox mediator; trickling filter.

## RESUMO

Este estudo investigou o tratamento de um efluente sintético contendo o corante azo Reactive Black 5 (50 mg·L<sup>-1</sup>) em reatores acidogênicos seguidos por diferentes unidades de pós-tratamento aeróbio: reatores de leito móvel com biofilme (em inglês, *moving bed biofilm reactors*, MBBR) e filtros biológicos percoladores (FBP). Foi avaliado o efeito da adição do mediador redox antraquinona-2-sulfonato (AQS) (50 μM) ao sistema, do tempo de detenção hidráulica (TDH) (6 e 4 horas) nos MBBR e do tipo de meio suporte (cubos de espuma de poliuretano e biomídia K1) nos FBP. Os reatores acidogênicos foram os principais responsáveis pela descoloração do RB5 nas concepções anaeróbias-aeróbias avaliadas neste trabalho, e o AQS melhorou significativamente sua eficiência de descoloração. A utilização de um TDH mais curto (4 horas) nos MBBR influenciou negativamente o desempenho na remoção de cor e de DQO nos reatores. A espuma de poliuretano demonstrou ser mais viável como meio suporte, já que é um material mais acessível e de baixo custo. Finalmente, reatores acidogênicos seguidos por FBP preenchidos com cubos de espuma de poliuretano foram a concepção mais promissora, tanto em termos de remoção de cor, DQO e amônia quanto em relação à demanda energética para a aeração do sistema.

**Palavras-chave:** reator acidogênico; remoção de cor; MBBR; mediador redox; filtro biológico percolador.

## INTRODUCTION

In recent years, the rapid growth of the textile industry has raised environmental concerns due to the generation of a large amount of effluents, most of which are classified as hazardous (Xu *et al.*, 2019). Textile wastewater is characterized by high pH, turbidity, color values, complex composition, and low biodegradability. These effluents can have serious effects on the photosynthetic function of aquatic life due to low light penetration and oxygen consumption (Yang, B. *et al.*, 2018). Among the dye classes, azo dyes stand out because

they are currently responsible for more than 50% of global dye production (Brüschweiler; Merlot, 2017).

Currently, several non-biological technologies, such as advanced oxidation processes, coagulation/flocculation, adsorption, and electrocoagulation, are used in the treatment of dye-containing effluents. However, these methods require high doses of chemicals, increasing process costs, and they do not usually achieve complete contaminant mineralization. However, biological treatment is considered a safer, economically viable, and effective option for

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removing dyes and other pollutants found in textile effluents (Moghaddam; Moghaddam, 2016).

Among the biological processes, sequential anaerobic–aerobic treatment is one of the most effective methods for degrading dyes. At the anaerobic stage, azo dyes are reduced to aromatic amines, with acidogenesis playing the most important role in this process, and, at the aerobic stage, these byproducts are easily mineralized (da Silva; Firmino; dos Santos, 2012; Thung *et al.*, 2018). However, even in anaerobic environments, some dyes present slow decolorization kinetics, requiring anaerobic reactors with long hydraulic retention times (HRTs). Thus, the use of redox mediators may be an alternative to reduce this time since they can transport reducing equivalents from an electron donor to the dye, increasing the reaction kinetics and, consequently, the decolorization rates (Field *et al.*, 2000; da Silva; Firmino; dos Santos, 2012).

Among the aerobic technologies used, attached growth systems stand out for presenting a high biomass retention capacity and, consequently, lower solid content in the final effluent, resulting in a smaller reactor volume. Thus, moving bed biofilm reactors (MBBRs) have been considered a promising technology in the treatment of recalcitrant contaminants (Yang, X. *et al.*, 2018). Similarly, trickling filters (TFs), although little used in the treatment of textile effluents, represent an interesting alternative as a post-treatment option, considering that their high solid retention capacity provides high removal of organic matter and nutrients, in addition to being referred to as a lower-cost technology, especially due to the absence of mechanical aeration (Kubota *et al.*, 2014; Hatamoto *et al.*, 2018; Watari *et al.*, 2021).

However, few studies have focused on aerobic attached growth systems, such as MBBRs and TFs, as post-treatment units for anaerobically treated textile effluents. Furthermore, most anaerobic pre-treatment units are methanogenic reactors (HRT > 6 h) (van der Zee; Villaverde, 2005; Nguyen *et al.*, 2020; Kozak; Cirik; Başak, 2021; Watari *et al.*, 2021; Bogale; Teffera; Aragaw, 2024). Therefore, the present study investigated different designs of anaerobic–aerobic systems for treating azo dye-containing effluents, i.e., acidogenic reactors followed by MBBRs or TFs. The effect of the addition of the redox mediator anthraquinone-2-sulfonate (AQS) (50  $\mu\text{M}$ ) to the system, HRT (6 and 4 h) in the MBBRs, and type of support medium (polyurethane foam cubes and K1 biomedica) in the TFs were evaluated.

## METHOD

### Experimental setup

In this study, two designs of anaerobic–aerobic systems for treating azo dye-containing effluents were evaluated: acidogenic reactors followed by MBBRs and acidogenic reactors followed by TFs.

Two lab-scale acidogenic anaerobic reactors ( $\text{AR}_1$  and  $\text{AR}_2$ ) were made of PVC and had a diameter of 100 mm, a height of 40 cm, and a working volume of 2.8 L.

Two lab-scale MBBRs ( $\text{MBBR}_1$  and  $\text{MBBR}_2$ ) were made of PVC and had a diameter of 200 mm, a height of 40 cm, and a working volume of 6 L. Ocean Tech's Kaldnes K1-type polyethylene biomedica (specific surface area of  $\sim 500 \text{ m}^2\text{-m}^{-3}$ ) was used as a support medium at a filling ratio of 40%. The aeration was carried out by a Maxxi Pro-6000 air compressor to maintain the minimum concentration of dissolved oxygen (DO) between 3 and 4  $\text{mg}\cdot\text{L}^{-1}$ . Figure 1 shows the schematic of the acidogenic reactor, followed by the MBBR.

Four lab-scale TFs were made of PVC and had a diameter of 100 mm and a height of 80 cm. In two TFs ( $\text{TF}_{1\text{-PF}}$  and  $\text{TF}_{2\text{-PF}}$ ), polyurethane foam cubes of

approximately  $8 \text{ cm}^3$  (specific surface area of  $\sim 200 \text{ m}^2\text{-m}^{-3}$ ) were used as a support medium. In the other two TFs ( $\text{TF}_{1\text{-K1}}$  and  $\text{TF}_{2\text{-K1}}$ ), Ocean Tech's Kaldnes K1-type polyethylene biomedica (specific surface area of  $\sim 500 \text{ m}^2\text{-m}^{-3}$ ) was used as a support medium. The aeration was carried out naturally through openings in the walls of the filters. Figure 2 shows the schematic of the acidogenic reactor, followed by the TFs.

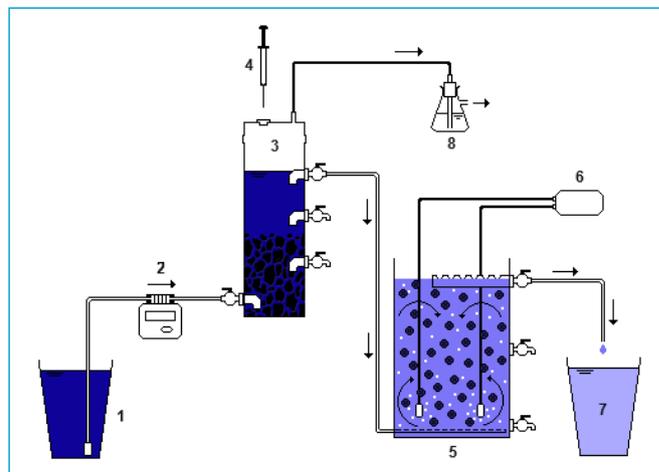
### Feeding of the anaerobic-aerobic systems

The anaerobic–aerobic systems evaluated in this study were fed with synthetic textile wastewater containing  $50 \text{ mg}\cdot\text{L}^{-1}$  of the azo dye Reactive Black 5 (RB5) (50% purity, Sigma-Aldrich),  $1.5 \text{ g}\cdot\text{COD}\cdot\text{L}^{-1}$  of glucose as an organic substrate, some macro and micronutrients as specified by Firmino *et al.* (2010), and  $1 \text{ g}\cdot\text{L}^{-1}$  of sodium bicarbonate as a buffer. The influent of  $\text{AR}_1$  was supplemented with  $50 \mu\text{M}$  of AQS (Sigma-Aldrich) as a redox mediator. All reactors were operated at a room temperature of  $\sim 28^\circ\text{C}$ .

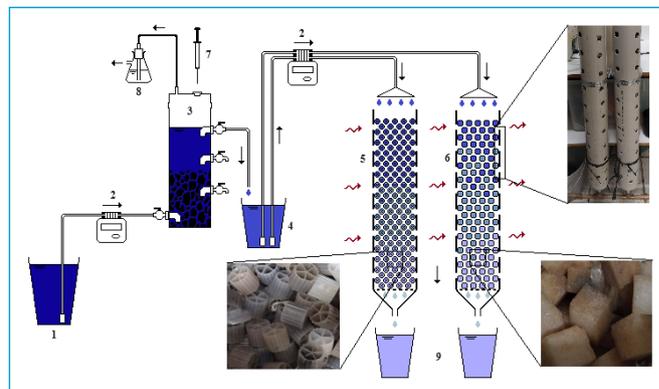
## Experimental design

### Acidogenic reactor + MBBR

Two anaerobic–aerobic treatment systems, each consisting of an acidogenic reactor followed by an MBBR ( $\text{AR}_1\text{-MBBR}_1$  and  $\text{AR}_2\text{-MBBR}_2$ ), were operated



**Figure 1** - Schematic of the two-stage systems: acidogenic reactor + MBBR. 1, influent; 2, pump; 3, acidogenic reactor; 4, biogas sampling port; 5, MBBR; 6, air compressor; 7, effluent; 8, water seal.



**Figure 2** - Schematic of the two-stage systems: acidogenic reactor + TFs. 1, influent; 2, pump; 3, acidogenic reactor; 4, acidogenic effluent; 5, TF with K1 biomedica; 6, TF with polyurethane foam cubes; 7, biogas sampling port; 8, water seal; 9, TF effluent.

simultaneously over two periods. In the first period, the MBBRs were operated at an HRT of 6 h, and, in the second period, the HRT was reduced to 4 h. The HRT of the acidogenic reactors was maintained at 2 h. To investigate the influence of the redox mediator AQS on color removal, 50  $\mu\text{M}$  of AQS was added to the influent of AR<sub>1</sub> throughout the experiment.

### Acidogenic reactor + TF

Two anaerobic-aerobic treatment systems, each consisting of an acidogenic reactor followed by two TFs in parallel (AR<sub>1</sub>-TF<sub>1-PF</sub>/TF<sub>1-K1</sub> and AR<sub>2</sub>-TF<sub>2-PF</sub>/TF<sub>2-K1</sub>), were operated simultaneously. The TFs were operated in parallel to assess the influence of two types of supporting media: polyurethane foam cubes (in TF<sub>1-PF</sub> and TF<sub>2-PF</sub>) and K1 biomedica (in TF<sub>1-K1</sub> and TF<sub>2-K1</sub>). The acidogenic reactors were operated at an HRT of 2 h, and the TFs at a hydraulic loading rate of approximately 0.1 m<sup>3</sup>·m<sup>-2</sup>·day<sup>-1</sup>. To investigate the influence of the redox mediator AQS on color removal, 50  $\mu\text{M}$  of AQS was added to the influent of AR<sub>1</sub> throughout the experiment.

### Physicochemical analyses

The analyses for COD, pH, ammonia, and volatile suspended solids (VSS) were carried out according to APHA (2012). The nitrogen fractions (ammonium, nitrite, and nitrate) and phosphate were determined using Dionex™ ICS-1100 ion chromatography (Thermo Scientific®) (Rolleberg *et al.*, 2018). Color analysis was performed using UV-Vis spectrophotometry (DR6000, Hach, United States) at a wavelength of 598 nm (maximum absorbance for RB5) (Xavier *et al.*, 2023).

## RESULTS AND DISCUSSION

### Acidogenic reactor + MBBR

#### Color removal

In the first period, an average color removal of around 67% was observed in the acidogenic reactor supplemented with AQS (AR<sub>1</sub>), while in the control

acidogenic reactor (AR<sub>2</sub>), without AQS, the average color removal was only 55% (Table 1). Therefore, the addition of this quinone-based redox mediator significantly improved the color removal efficiency of the acidogenic reactor, as previously reported (da Silva; Firmino; dos Santos, 2012).

The decolorization of azo dyes under anaerobic conditions results from a combination of biological and chemical mechanisms. The substrates, once oxidized by microorganisms, serve as electron donor sources. Then, these electrons are transferred to the dye, which acts as a final electron acceptor, resulting in the cleavage of its chromophore groups (azo bonds). Furthermore, in the presence of redox mediators, the reductive decolorization kinetics of azo dyes can increase considerably due to electrochemical interactions between the primary electron donors, mediators, and dyes (dos Santos *et al.*, 2005).

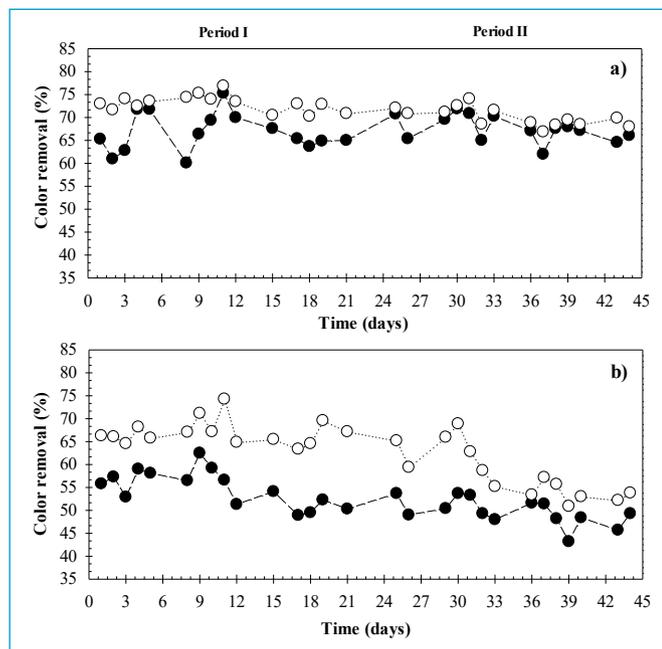
The MBBR<sub>1</sub> post-treatment had a minor impact on the total color removal in the AQS-supplemented system. After this aerobic stage, the efficiency increased only ~6%, reaching an average total value of 73%. However, the impact of MBBR<sub>2</sub> on the control system without the mediator proved to be more promising, showing an increase of approximately 12%, leading to an average total color removal of approximately 67% (Table 1). Therefore, even though AQS improved color removal in the acidogenic reactor, after the MBBR, the total color removal in the control system was only 6% lower.

In the second period, AR<sub>1</sub> managed to maintain an average color removal efficiency close to 68%. In contrast, AR<sub>2</sub> showed an average color removal efficiency of approximately 50% (Table 1). In practice, the control acidogenic reactor did not have its removal reduced in the second period, as, already in the first period, especially from the 12th day of operation, it indicated that, under stable conditions, it would tend to color removals close to 50% (Figure 3). Therefore, this suggests that the performance of the acidogenic reactor in decolorizing RB5 increased by 18% in the presence of AQS, and this strategy seems interesting for the operation of anaerobic reactors. With the reduction of the MBBR HRT to 4 h, the system with AQS showed a drop in the decolorization efficiency of only 3%. However, in the control system, the average color removal was significantly reduced to 58% (a 9% decrease) (Table 1).

**Table 1** – Operational performance of the acidogenic reactors and MBBRs (at HRTs of 6 and 4 h) with and without AQS (50  $\mu\text{M}$ ).

Parameter	HRT <sub>MBBR</sub> = 6 h		HRT <sub>MBBR</sub> = 4 h	
	AQS	No AQS	AQS	No AQS
Acidogenic color removal (%)	66.7 (4.2)	55.0 (4.0)	67.6 (2.9)	49.7 (3.0)
Total color removal (%)	73.1 (1.8)	67.1 (2.8)	70.1 (2.1)	58.0 (5.7)
Acidogenic COD removal (%)	37.7 (4.2)	34.5 (8.3)	38.0 (4.2)	27.4 (5.2)
Total COD removal (%)	77.7 (12.4)	75.0 (13.0)	71.2 (5.1)	69.3 (6.7)
Acidogenic N-NH <sub>4</sub> <sup>+</sup> removal (%)	21.4 (6.2)	17.6 (4.4)	19.9 (4.9)	14.2 (6.4)
Total N-NH <sub>4</sub> <sup>+</sup> removal (%)	86.9 (3.7)	85.0 (6.4)	89.2 (5.4)	93.8 (3.5)
Total inorganic N removal (%)	44.4 (8.0)	62.6 (11.4)	48.6 (17.9)	81.7 (6.8)
Acidogenic P-PO <sub>4</sub> <sup>3-</sup> removal (%)	6.6 (3.1)	9.6 (2.0)	5.5 (4.5)	7.9 (2.7)
Total P-PO <sub>4</sub> <sup>3-</sup> removal (%)	15.0 (5.6)	14.3 (6.0)	13.7 (4.9)	11.7 (6.8)
pH in AR	6.1 (0.2)	5.7 (0.2)	5.5 (0.4)	5.3 (0.3)
pH in MBBR	7.9 (0.5)	8.0 (0.3)	7.9 (0.2)	8.1 (0.4)
Effluent VSS (g·L <sup>-1</sup> )	0.2 (0.1)	0.2 (0.2)	0.3 (0.2)	0.4 (0.2)

The standard deviation is shown in parentheses.



**Figure 3** - Color removal of the acidogenic reactors and MBBRs (at HRTs of 6 and 4 h) with (a) and without AQS (b). Filled circles represent acidogenic efficiency, and empty circles represent total system efficiency.

The results also show that the acidogenic reactors were responsible for 91–97% of the total removal of RB5 in the AQS-supplemented system and 82–86% of it in the control system, confirming the importance of the anaerobic stage, particularly acidogenesis, in color removal. These findings corroborate the results of Firmino *et al.* (2010), whose acidogenic reactor (HRT of 4 h) was responsible for approximately 94% of the total decolorization (98.5%) of the azo dye Congo Red ( $\sim 210 \text{ mg}\cdot\text{L}^{-1}$ ) in a two-stage acidogenic-methanogenic anaerobic system (total HRT of 24 h).

The present study showed that the presence of AQS considerably improved the color removal performance of the acidogenic reactor (HRT of 2 h). However, after the aerobic stage (MBBR), especially with HRT equal to or greater than 6 h, its effect was not very relevant, and, probably, this strategy would not be the most interesting one.

### C, N, and P removal

During the two experimental periods, the AQS-supplemented acidogenic reactor was slightly superior to the control reactor, presenting average organic matter removal efficiencies of 38%. The control reactor showed a performance of 34% and 27% in the first and second periods, respectively (Table 1). The best performance in  $\text{AR}_1$  is possibly related to the presence of AQS, as it acts by increasing the electron transfer capacity and, consequently, the oxidative and reducing potential of the medium (Wolf *et al.*, 2009). For instance, Li *et al.* (2022) investigated the role of quinones in wastewater as redox mediators for the synergistic removal of an azo dye in microbial fuel cells. The authors observed that, in addition to improving the color removal performance of the azo dye Reactive Red 2 from 79% to 92%, the removal of organic matter (COD) also increased by approximately 10%.

In the first period, the total COD removal efficiencies remained close to 77% in the AQS-supplemented system ( $\text{AR}_1$ -MBBR<sub>1</sub>) and 75% in the control

system ( $\text{AR}_2$ -MBBR<sub>2</sub>). In the second period, with the reduction in the HRT of the MBBRs to 4 h,  $\text{AR}_1$ -MBBR<sub>1</sub> and  $\text{AR}_2$ -MBBR<sub>2</sub> presented average total COD removal efficiencies of approximately 71% and 69%, respectively, i.e., a 6% decrease in both systems (Table 1). This COD removal was below what was expected for MBBRs ( $\sim 85\%$ ). One hypothesis for this performance may be related to the short HRT adopted ( $\sim 6$  h), which is lower than the range used in previous studies (16–72 h) (Koupaie; Moghaddam; Hashemi, 2012; Sonwani *et al.*, 2019; Ong; Lee; Chang, 2020).

The removal of ammonia nitrogen ( $\text{N-NH}_4^+$ ) in the systems remained above 85%, even with the reduction in the HRT of the MBBRs. Therefore, no major variations in reactor performance were observed, reaching removals close to 93% in the control system during the second stage. These results are in accordance with the study by Shore *et al.* (2012), who used MBBRs with BioPortz™ biomedica (filling ratio of 50%) to treat secondary effluents. They observed that more than 90% of  $\text{N-NH}_4^+$  was removed from synthetic and industrial wastewater by the MBBRs.

Concerning total inorganic nitrogen, in the first period, the system with AQS showed an average removal efficiency close to 44%, and, in the control system, the removal efficiency was 18% higher. In the second period, there was practically no variation in the efficiency of  $\text{AR}_1$ -MBBR<sub>1</sub>. However, unexpectedly, even with the reduction in the HRT, the control system showed an increase of approximately 20% compared to the previous period (Table 1).

In both systems, phosphorus removals were similar, but low. The average values obtained were approximately 15%. No influence of the redox mediator on phosphorus removal in the MBBRs was observed. The effluent VSS of the MBBRs varied between 0.2 and 0.4  $\text{g}\cdot\text{L}^{-1}$ , demonstrating good biomass retention.

### Acidogenic reactor + TF

#### Color removal

The AQS-supplemented acidogenic reactor reached an average of 66% color removal, while the control acidogenic reactor obtained only 49% removal (Table 2). Therefore, once again, AQS improved the capacity of the acidogenic reactor to decolorize the wastewater (a 17% increase). After the TFs used polyurethane foam cubes as a support medium, the total color removal efficiency was approximately 72% in the AQS-supplemented system and 70% in the control system. As for the TFs that used K1 biomedica as a support medium (AQS-supplemented and control), the total efficiencies were similar ( $\sim 68\%$ ) (Table 2). Therefore, regarding color removal, the system containing TFs filled with polyurethane foam cubes presents a slight operational advantage, as, in addition to being slightly more efficient, it is a low-cost material and can promote fixation and fast and stable growth of microorganisms in the support medium due to its high porosity (Watari *et al.*, 2021).

The acidogenic reactors were responsible for more than 90% of the total removal in the AQS-supplemented systems and more than 70% in the control systems. Therefore, these results are important from the perspective of operating anaerobic systems with short HRTs ( $\sim 2$  h), making them more compact and less expensive.

Overall, the performance of the anaerobic phase in color removal agrees with most previous works. For example, Choerudin *et al.* (2021) evaluated the performance of a system consisting of an anaerobic membrane reactor (AnMBR) (HRT of 12 and 24 h) combined with a downflow hanging sponge (DHS) reactor (HRT of 1.4 and 2.8 h) in the treatment of a synthetic textile

**Table 2** – Operational performance of the acidogenic reactors and TFs (with polyurethane foam cubes and K1 biomedica as a support medium) with and without AQS (50 µM).

Parameter	AQS		No AQS	
	Polyurethane foam cubes	K1 biomedica	Polyurethane foam cubes	K1 biomedica
Acidogenic color removal (%)	65.6 (2.7)		48.8 (5.9)	
Total color removal (%)	71.4 (1.1)	68.8 (2.0)	70.7 (2.3)	68.3 (1.9)
Acidogenic COD removal (%)	40.6 (11.6)		26.6 (13.4)	
Total COD removal (%)	69.8 (5.8)	68.4 (7.0)	67.9 (4.2)	66.5 (9.2)
Acidogenic N-NH <sub>4</sub> <sup>+</sup> removal (%)	13.1 (7.6)		11.9 (7.8)	
Total N-NH <sub>4</sub> <sup>+</sup> removal (%)	96.9 (4.5)	79.3 (8.0)	96.0 (5.3)	86.2 (9.9)
Total inorganic N removal (%)	41.1 (6.4)	42.8 (10.2)	43.3 (13.2)	55.0 (11.5)
Acidogenic P-PO <sub>4</sub> <sup>3-</sup> removal (%)	8.8 (3.6)		10.3 (4.2)	
Total P-PO <sub>4</sub> <sup>3-</sup> removal (%)	-	18.5 (5.7)	-	19.2 (9.7)
pH in AR	5.3 (0.4)		6.1 (0.3)	
pH in TF	7.8 (0.3)	7.7 (0.5)	7.9 (0.4)	7.6 (0.2)
Effluent VSS (g·L <sup>-1</sup> )	0.2 (0.2)	0.2 (0.1)	0.1 (0.1)	0.2 (0.1)

The standard deviation is shown in parentheses.

wastewater containing 50 mg·L<sup>-1</sup> of the hydrolyzed azo dye Reactive Black 5. The combination reduced effluent color by approximately 92% for both HRTs. However, most of the color removal occurred in the AnMBR. The same behavior was observed by Nguyen *et al.* (2020), who evaluated the color removal of the azo dye Hellozol Reactive Black HSR (20 mg·L<sup>-1</sup>) in a combined system consisting of an anaerobic baffled reactor (HRT of 18.6 h) and a DHS reactor (HRT of 4.6 h). Most of the removal was observed in the anaerobic system (~90%), highlighting the importance of the anaerobic stage for dye reduction.

Even though AQS significantly improved color removal in the anaerobic stage, when combined with TFs, the total removals did not differ, and, therefore, for these studied designs, the use of the redox mediator seems to be dispensable, and polyurethane foam cubes are more suitable than K1 biomedica. In addition, the results of this study also emphasize the relevance of using TFs compared to MBBRs as a post-treatment unit for acidogenic reactors in dye removal, as they presented equivalent performance and constitute systems with low capital and operational expenditures due to the use of natural aeration. In fact, it is reported that the energy demand for TFs is approximately 75% lower than that required in the conventional activated sludge process (Nurmiyanto; Ohashi, 2019). Thus, the use of TFs is a possibility to be evaluated mainly in developing countries.

### C, N, and P removal

The average COD removal in the control acidogenic reactor was 27%. However, in the AQS-supplemented acidogenic reactor, the average COD removal was 14% higher. Therefore, the results reinforce that the presence of AQS also accelerates COD removal in acidogenic reactors. After the post-treatment stage with TFs, the total efficiencies of the systems reached values above 65%. No significant differences were observed in the COD removal efficiency between the two types of support medium used in the experiment (Table 2).

The removal of ammonia nitrogen (N-NH<sub>4</sub><sup>+</sup>) in the systems remained above 79% throughout the experiment, and no relationship was observed in the N-NH<sub>4</sub><sup>+</sup> removal performance with the presence of AQS. As for the removal of total inorganic nitrogen, the efficiencies varied between 41% and 55% in the systems, and it was not possible to identify the influence of the redox mediator

on the performance of the systems either. It is important to highlight the high levels of nitrogen removal achieved with the TFs studied, mainly because these systems use natural oxygenation and, therefore, are considered low-cost units.

Phosphorus removals in both systems were similar but low. Maximum average values reached ~19%. The TFs filled with polyurethane foam cubes did not show significant phosphorus removals. The effluent VSS values of the TFs varied between 0.1 and 0.3 g·L<sup>-1</sup>, demonstrating good biomass retention.

## CONCLUSIONS

The acidogenic reactors were mainly responsible for the decolorization of RB5 in the anaerobic-aerobic designs evaluated in this study. Therefore, the use of these compact reactors (due to their short HRTs) as a pre-treatment may be an interesting strategy for the decolorization of azo dye-containing effluents.

The presence of the redox mediator AQS significantly improved color removal in the anaerobic stage, increasing the potential of acidogenic reactors.

The use of MBBRs as a post-treatment for azo dye-containing effluents may be viable, but shorter HRTs may negatively influence their color and COD removal performance.

The two types of support media evaluated in the TFs presented satisfactory and equivalent performances. Therefore, being a more accessible and low-cost material, polyurethane foam is more viable for being used as a support medium.

Finally, between the two designs assessed, acidogenic reactors followed by TFs filled with polyurethane foam cubes seemed to be the more promising ones, both in removing color, COD, and ammonia and concerning the energy demand for system aeration.

## AUTHORS' CONTRIBUTIONS

Barbosa, P.T.: Formal analysis, Investigation, Writing – original draft. da Silva, M.E.R.: Conceptualization, Funding acquisition, Project administration, Supervision, Writing – review & editing. Firmino, P.I.M.: Conceptualization, Funding acquisition, Project administration, Supervision, Writing – review & editing.

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