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Novel hybrid treatments of textile wastewater by membrane oxidation reactor: Performance investigations, optimizations and efficiency comparisons

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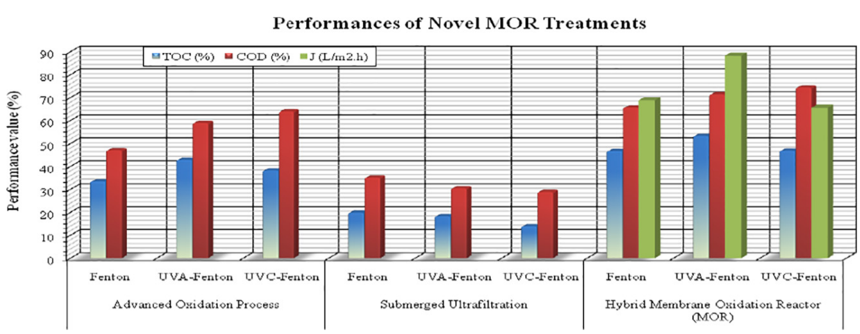
HIGHLIGHTS

- Novel membrane oxidation reactor was demonstrated for textile wastewater treatment.
- Hybrid Fenton/photo-Fenton treatment was performed accompanied by submerged UF.
- High level efficiencies were provided with reduced chemicals and processing time.
- Effluents produced by hybrid reactor could be directly discharged to sewer systems.
- Developed technologies seem very feasible for wastewater treatment and water recovery.

GRAPHICAL ABSTRACT



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ABSTRACT

Feasible reclamation of industrial wastewaters by consuming less resource and time requires researchers to develop advanced and sophisticated solutions to meet today's versatile needs. In this respect, novel technological applications of hybrid membrane oxidation reactor (MOR) comprising of the Fenton or photo-Fenton enhanced ultrafiltration (FEUF and pFEUF), was demonstrated for treating textile washing wastewater. Their comparative hybrid performances were explored based on response surface analyses of Taguchi experimental designs that were optimized for maximized responses at minimum oxidant and acid consumptions. From eleven specific variables, those affecting the hybrid treatment performances at significant levels were found as H_2O_2 amount, process time, membrane type, Fe^{2+} concentration and temperature. The pFEUF treatment showed better and faster organics removal efficiency than by FEUF, and the UF process was seen to be more affected from changing operational conditions in pFEUF. Organic pollutants were oxidized by 56.6 ± 8.7% degradation and 31.5 ± 3.2% mineralization, while UF allowed a synergistic contribution to the hybrid MOR performance by 38.1 ± 4.7% and 17.3 ± 3.1%, respectively. Compared to simultaneous MOR and external UF after Fenton, sequential MOR was found as the best solution by an efficiency of 84.5% COD, 70.5% TOC, and 155.6 $L/m^2 \cdot h$. The effluents could be readily produced with quality suitable for directly discharging to the sewage infrastructure system resulting in a complete treatment. This study proved that the developed MOR techniques are technologically favorable for the treatment

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of industrial organic wastewaters due to high treatment performances and less resource, time and land needs. It can be finally declared that they can be used as rather attractive solutions for not only wastewater reclamation but also water recovery by further handling of their effluents.

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1. Introduction

Increases in industrial activities lead to raises water stress of countries by triggering ascents in sectoral water demands and continue to widen industrial water pollution problems worldwide. This situation brings pressure on industrial wastewater treatment requirements and use of natural water resources and requires more stringent effluent discharges to receiving media (Aydıner et al., 2016; UN ESC, 2018). Thus, advancement of new treatment technologies that are proficiently used and broadly practicable in excessive water-consuming industries for instance textile, paper, steel, food and beverage shows itself as a reality to be crucially considered in developments of nations for a more sustainable and resilient ecosystem.

As leading one of these industries, the substantial water amount of 60 and 200 kg per kg fabric is consumed in dyeing and washing processes in a textile sector, and treatment of wastewaters produced require integrated applications of numerous techniques within a system for successful environmental protection, because they contain a great deal of contaminants such as dyes, salts and chemical additives (Chidambaram et al., 2015; Holkar et al., 2016). Miscellaneous reclamation methods such as coagulation-flocculation (Verma et al., 2012), activated sludge (Nawaz and Ahsan, 2014), membrane bioreactor (Jegatheesan et al., 2016), anaerobic reactor (Yurtsever et al., 2016), membrane filtration (Ellouze et al., 2012), advanced oxidation process (AOP) (Soares et al., 2017) and adsorption (Leal et al., 2018) are capably used in treatment of textile wastewaters. Although these techniques are extensively implemented from laboratory scale to commercial applications, they have some disadvantages due to some restrictive problems being encountered in practice. These drawbacks can be summarized as high chemical necessity and large amount of complex sludge for coagulation-flocculation (Nawaz and Ahsan, 2014), regeneration requirement and high cost of adsorbent for adsorption (Yagub et al., 2014), detrimental effects of some dyes on living microorganisms for biological processes (Yukseler et al., 2017), high investment cost and fouling and concentrate issues for membrane processes (Chidambaram et al., 2015), and in some specific cases inadequacy or expensiveness of well-known techniques against large variable composition of textile wastewaters (Soares et al., 2017). So, advanced solutions toward meeting versatile up-to-date needs in treatments of these wastewaters must be developed for the case of less resource and installation space in order to reduce capital and operating costs (Aouni et al., 2009). It is seen that recent progresses on the reclamations of waters and wastewaters make it important to develop innovative systems combining the AOP with other treatment processes (Blanco et al., 2014; Tijani et al., 2014; Diya'uddeen et al., 2015; Holkar et al., 2016; Lafi et al., 2018). This approach providing superiority regarding the effective elimination of contaminants from waters in a hybrid reactor improves the treatment efficiency by integrating the processes and overcoming the other one's drawbacks.

Superior diversity of hybrid reactors was recognized for not only industrial wastewater treatment of but also industrial water recovery with special purposes (Mozia, 2010; Ganiyu et al., 2015; Iglesias et al., 2016). As promising technological solutions, hybrid reactors where AOPs integrated with miscellaneous membranes seem to be too advantageous due to higher purification yield, lower energy and chemical expenditures and enhanced membrane flux with reduced fouling (Mozia, 2010; Mozia et al., 2014). The AOP hybridization with a submerged filtration in a photocatalytic membrane reactor (PMR) which is operated at low temperatures by sustaining catalyst efficiency has been noticeably actuated for the treatments of secondary effluents (Jiang et al., 2018), natural organics in water and seawater (Choo et al., 2008; Kim et al., 2010), and pharmaceuticals (Fernández et al., 2014), oily (Ong et al., 2014) and textile or dye wastewaters (Kertèsz et al., 2014; Lee et al., 2015; Su et al., 2016; Ahmad et al., 2017). Although the leading AOP techniques such as Fenton and photo-Fenton recently received a great interest in removal of organics from wastewaters (Babuponnusami and Muthukumar, 2014; Sreeja and Sosamony, 2016), limited number of researches were conducted for their combinations in a system as integrated with a sequencing batch reactor (Blanco et al., 2014), membrane bioreactors (Feng et al., 2010; Karaolia et al., 2017) and membrane technologies (Miralles-Cuevas et al., 2015). However, any research on the investigation of technological feasibility of Fenton and photo-Fenton operations hybridized by membrane processes for the reclamation of industrial organic wastewater was not achieved, despite the presence of two novel techniques developed as the photo-Fenton-like catalysis (Zhang et al., 2013) and the cathodic electro-Fenton (Liang et al., 2016). Based on the fact that the immersed membranes that were combined not only by catalytic or photocatalytic oxidation but also by every kind of AOP can be technically defined as membrane oxidation reactor (MOR), the Fenton or photo-Fenton oxidation supported by submerged membrane appears as worthwhile and potent technological MOR applications to be explored.

In current work, hybrid treatments of textile wastewater were conducted by the developed two MOR techniques, namely Fenton or photo-Fenton enhanced ultrafiltration (FEUF or pFEUF). Despite the fact that the first introductions of both MORs were documented with our previous work (Aydiner et al., 2016) and patent application (Aydıner and Imer, 2017), this paper is the first study to deeply investigate and exhaustively analyze the performances of the FEUF and pFEUF techniques in the treatment of textile washing wastewater. The main objectives of this study are to: *i*) reveal the importance and influences of all specific variables in each hybrid treatment; *ii*) determine the contributions of Fenton or photo-Fenton oxidation and membrane to overall performance; *iii*) optimize the reactor effectiveness by minimizing the chemical and time demands for obviously exposing industrial favorability of both hybrid applications; and *iv*) comparatively assess the achievements of bringing AOP and UF together as either hybrid or system processes for fully manifesting performance differences of all explored techniques.

In this regard, hybrid performances from experiments designed with Taguchi's L_{32} array were monitored for organics removal efficiencies and membrane water flux using response surface methodology (RSM) by considering all specific variables. Experimental plans and response analyses were executed for simultaneous MOR hybridization by using an experimental design and analysis software. Performance estimation models were individually built for responses in both MORs, and the optimized responses were verified by confirmation experiments to elicit the comparative performances of both processes and overall operations. In final, in order to reveal the technological and practical viability potentials of the developed hybrid techniques, the treatment efficiencies in the different assembling of UF to the AOP as a simultaneous or sequential MOR process were compared with each other by also considering their systematic arrangement, i.e., the integrated [AOP/UF] system.

2. Material and methods

2.1. Characterization of textile washing wastewater

Experimental studies were performed with wastewater samples supplied from washing baths of a textile industry in Sakarya, Turkey, of which characteristics are presented in Table 1.

2.2. Membranes and chemicals

In the UF experiments, four commercial membranes viz., UP005, UP020, UH050 and UV150 purchased from Microdyn-Nadir® (Germany) were used. The characteristic properties of the membranes were presented in Table S1 in the Supplementary material.

Iron (II) sulphate heptahydrate ($FeSO_4 \cdot 7H_2O$, Merck 99.5%) and hydrogen peroxide (H_2O_2, Merck 35%) were used to provide the desired molar concentrations in the Fenton reactions. Before experiments, pH of wastewater was adjusted to the desired values by using sulphuric acid solution (H_2SO_4, Merck 95–98%), and sodium hydroxide solution prepared from anhydrous pellets (NaOH, Merck 99.5%). The solutions were prepared using deionized water from a Millipore Milli-Q.

2.3. Hybrid MOR system

The operating of hybrid MOR system was carried out by 3 L borosilicate glass reactor mixed completely from bottom by air diffuser in that UF module was fully immersed to the vessel. The reactor was equipped by a temperature-controlled water bath connected to a coolant flow channel inside the reactor to maintain the desired temperatures along the treatment operations. All metal parts of the reactor including flow connections were made of stainless-steel except for UV lamps and membrane module. Two different lamp sources were used in photo-Fenton oxidations, UVC (254 nm) and UVA (365 nm), through a total irradiation power of 40 watts. The reactor reservoir was employed with four 10-watt lamps shielded by quartz glass exactly submersible to the reactor for the UVC enhanced photo-Fenton, whereas for the UVA illumination, five 8-watt actinic lamps clad outside its transparent glass wall were placed. All lamps were positioned symmetrically to tetragonal and pentagonal hypothetic corners on the reactor's cross-section for the UVC and UVA illuminations, respectively, that allowed to the highest light intensity over its cylindrical geometry of 13.3-cm diameter and 30.0-cm height. In addition to the light layouts, the module was embedded in the reactor so as to maximize intensity of light on the membrane surface by measurements made at various projectional locations over the diameter plane (Aydıner and Imer, 2017). By this means, it was intended to expose organics retained on the surface to further oxidation. The lab-scale experimental set-up of hybrid MOR system operated as either FEUF or pFEUF was schematically shown in Fig. 1(a) (Aydiner et al., 2016).

Both processes of the hybrid reactor can be employed complementarily with each other in a planned schedule that are conjugated at necessary oxidant contact or depletion period with simultaneously or sequentially operating of the submerged UF to operational AOP treatment. The membrane module made of Delrin® acetal resin (DuPont) was immersed into the reactor by stainless-steel plate frame. It was fixed within the reactor by the top squeezing connection with the purpose of sealing of the membrane during vacuum filtration. Its effective membrane area was 39.4 cm^2 with a dimension of 14.6 cm × 2.7 cm. The air diffuser was placed at the bottom of the reactor in such a way that bubbles were exactly stripping the membrane surface. Thanks to this functionality gained to the reactor, both particles and organics fouling over the membrane could be diminished via effective bubble stripping of the surface by air diffuser of which aeration rate was adjusted by a valve-regulated flow meter (Aydıner and Imer, 2017). The filtrate was withdrawn from the membrane under constant vacuum using peristaltic pump (PLP 380, Behr Labor-Technik, Germany). The reactor was also equipped with digital scale (AND EJ-6100) to determine water flux. The water volume passing across the membrane was instantaneously monitored and recorded on a computer. So, the permeated water flux was determined from the time-dependent changes of filtrate volume by the RsKey software (A&D Company, Japan).

The principal superiorities of both technologies developed are to: *i*) handle the wastewater by combining two processes in the same reactor for efficient removals of organic pollutants; *ii*) reduce the effluent organics to discharge levels at acceptable inorganic content and without producing any membrane concentrate; *iii*) produce the effluent by submerged membrane able to be employed by relative high water flux under its synergistic contribution to all treatment; and *iv*) pave the way for the recurrence of catalyst reusability in oxidation reactions.

2.4. Integrated [AOP/UF] system

The integrated AOP/UF treatment was performed by Fenton oxidation in the reactor and then membrane filtration at the outside of the reactor. As the first step, Fenton process was applied within the reactor shown in Fig. 1(a) without handling the submerged filtration. Thereafter, a dead-end membrane filtration system (HP4750, Sterlitech®) operable at high pressure was utilized for the external filtration of wastewaters treated by AOP (Fig. 1(b)). Its stainless-steel module with an internal diameter of 49 mm and an effective membrane area of 14.6 cm^2 had a liquid feed capacity of 300 mL. The module having stirring cell was operated at concentration mode of feed stream with stirring speed of 300 rpm. Trans-membrane pressure was supplied by directly feeding nitrogen gas into the module. The permeated water was collected in glass beaker, and its weight was measured by digital scale for the flux analysis as applied in the UF of the hybrid MOR system.

2.5. Performance calculations

Membrane performance was determined for the permeate water flux and rejection parameters. The water flux described by Darcy's law was calculated versus time by Eq. (1).

$$J = \frac{1}{A}\frac{dV}{dt} \quad (1)$$

where A is the membrane area, V the total permeate volume, and t the filtration time. At the AOP or UF operations, removal efficiency or rejection, R (%) was calculated separately from variations of chemical oxygen demand (COD) and total organic carbon (TOC) concentrations of water

Table 1
Characteristic properties of textile washing wastewaters.

Parameter	Unit	Value
pH	–	6.22 ± 0.04
Temperature	°C	25.4 ± 1.2
Conductivity	µS/cm	2633 ± 321
TDS	mg/L	1343 ± 170
COD	mg/L	2830 ± 605
TOC	mg/L	1030 ± 148
UV_{254}[a]	1/cm	1.863 ± 0.046
Suspended solids	mg/L	21.7 ± 12.7
Turbidity	NTU	1.0 ± 0.1
Color	A @436–620 nm	0.209 ± 0.080
Sulphate	mg/L	710 ± 369
Chloride	mg/L	35.4 ± 12.7
Phenol	mg/L	14.4 ± 3.33
Toxicity[b]	TU	19.5 ± 2.7

[a] Was analyzed by 25-fold dilution to provide that its value corresponds to the measurement range in the standard method.
[b] Toxicity is evaluated in TU (toxicity unit) as follows: (0) non-toxic, (<1) low toxic, (1–10) toxic, (11–100) high toxic, (>100) excessive toxic.

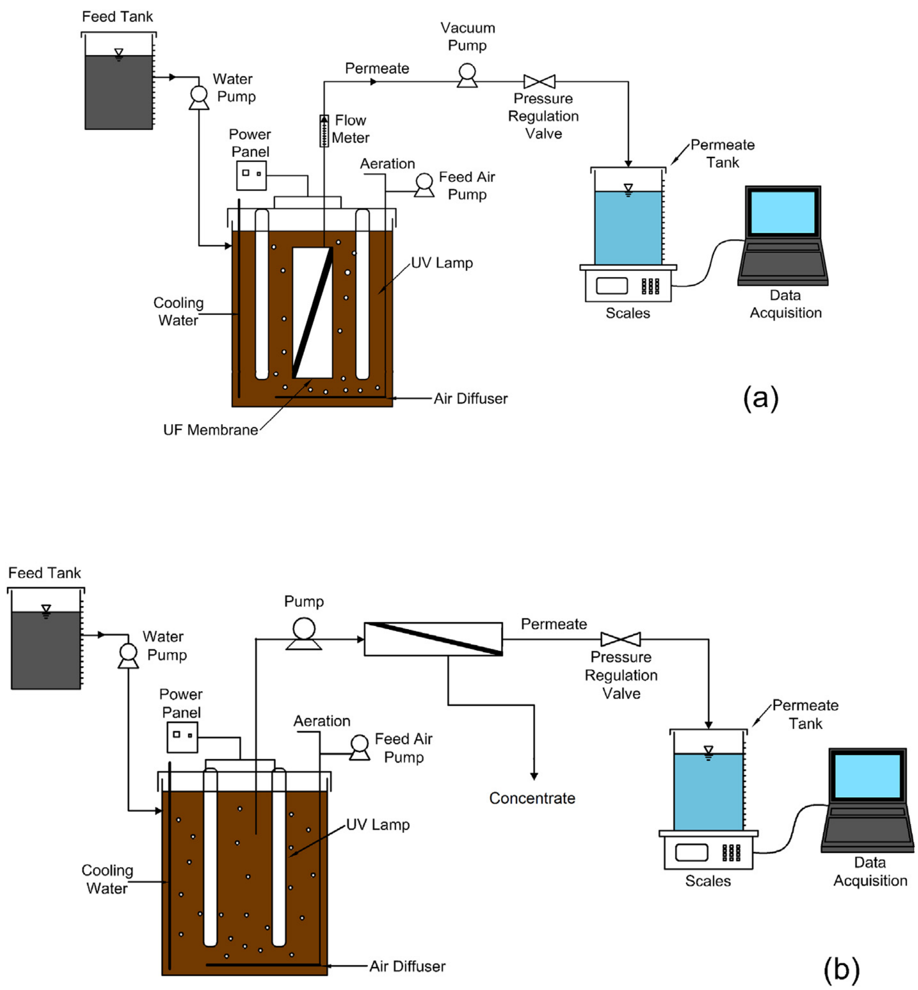


Fig. 1. Schematic representations of the experimental set-ups of hybrid MOR system (a) and [AOP/UF] integrated system (b).

samples using Eq. (2).

$$R(\%) = \left(1 - \frac{C_a}{C_b}\right) \times 100 \tag{2}$$

where C_a is the concentration after oxidation or filtration while C_b is the concentration before.

2.6. Analytical procedures

The analytical procedures specified by Standard Methods of the American Public Health Association (APHA) were utilized for the determination of each parametric value in the untreated and treated wastewater (APHA et al., 2012). The temperature, conductivity, total dissolved solids (TDS) and pH were analyzed by Hach HQ40d multi-parameter device (Hach Co., USA). Turbidity measurement was carried out by 2310-B Nephelometric method using Hach 2100Q portable turbidimeter. The Hach DR6000 UV VIS spectrophotometer was used for the analyses of color, sulphate, chloride, UV_{254} and phenol. The SO_4^{2-} and Cl^- parameters were measured by 4500-D turbidity and 4500-E potentiometric methods, respectively. The phenol concentration was determined by 5530-C chloroform extraction method. The color as average absorbance at 436–620 nm wavelength was analyzed with regard to 2120-C spectrophotometric method. The analysis of UV-absorbing organic matters at 254 nm was done by 5910-B ultraviolet absorption method. Total suspended solids (TSS) of samples were examined by drying at about 103–105 °C in accordance with 2540-D gravimetric method.

Toxicity was quantified with the ISO 11348-3 method by using of freeze-dried bacteria (EN ISO 11348-3, 2007). As indicators for organic removals, the COD and TOC parameters were analyzed by 5220-C closed reflux titration method, and 5310-B high temperature catalytic oxidation method using Teledyne Tekmar analyzer, respectively. The analyses were duplicated by the measurements confirmed with reproducibility of ±3%. The interference of residual hydrogen peroxide on the COD analyses was removed using the COD-H_2O_2 relation in the range of 0–5000 mg H_2O_2 per L of distilled water (Mert et al., 2018).

$$[COD] = (0.4054) \times [H_2O_2] \; R^2 = 0.9938 \tag{3}$$

Moreover, the light intensity of UV irradiations inside the hybrid AOP/submerged UF reactor was measured by means of UVA-365 and UVC-254 light radiometers (Lutron Co., Taiwan). The results of light measurements made at the center of the reactor's circular cross-section are given in Fig. 2 against varying lamp numbers of both light sources.

2.7. Taguchi design of experiments (DoE)

In order to acquire optimum performances of the hybrid MOR operations, a holistic Taguchi approach was implemented under the DoE structure where experimental layouts of AOP and UF processes and overall system were placed on the same orthogonal array of the design. The experimentations were applied jointly for the FEUF and pFEUF operations of the MOR at the predefined levels of input variables inherent to both processes. While UV light source was taken account at two levels

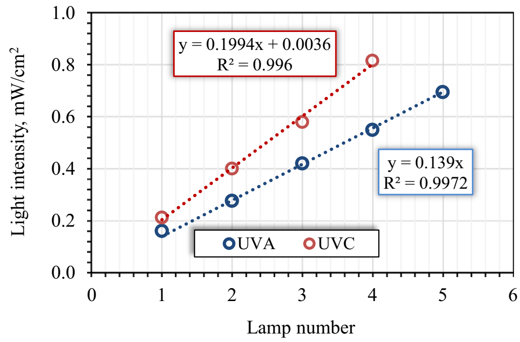


Fig. 2. Changes of the intensity of light versus lamp number in the MOR.

at 254 and 365 nm, all the rest variables were considered as four levels. Eight variables namely process time, temperature, pH, H_2O_2/TOC, H_2O_2/Fe^{2+}, aeration rate, vacuum rate and membrane type were included to the DoE planning of every treatment study. The influences of light source and intensity were additionally examined in the photo-Fenton experiments in addition to the abovementioned variables.

Taguchi DoE plans formed in an overarching frame are given in Table 2 for the design levels and actual values of input variables. By taking into account that the full DoE designs at four levels-eight factors (4^8) for FEUF, and at two level-one plus four level-nine factors ($2^1 \times 4^9$) for pFEUF necessitate excessive number of experiments, Taguchi L_{32} orthogonal arrays were decided up as adequate experimental matrices for our variable screening aims that were comprehended by broad literature survey and constituted by preliminary tests relied on our past experiences. The experimental matrices for statistically evaluation of the MOR efficacies are presented at Table S2 for the actual operating values of the variables.

2.8. Statistical analyses of performances

Based on the successful responses of RSM analyses at optimizing Taguchi designs that were accompanied for solving environmental issues as well as other problematical topics (Wang et al., 2013; Yang et al., 2015; Aggarwal et al., 2018), the DoE performances of both MORs were statistically analyzed with the RSM tool of the software Design Expert®11.0 (Stat-Ease Inc., USA). Three response parameters, namely TOC and COD removals and UF water flux were used for the evaluation of treatment performances. In the analyses, the influences of all input variables on each response were first put forward by multiple linear regression (MLR) models involving interactive variables, since their correlations between experimental results and model predictions resulted in better than those of the MLR models composed of single inputs. Accordingly, before RSM analyses, the model equations of every response were built separately by the software according to the analysis of variance (ANOVA) used for complete evaluation of the Taguchi DoE results (Huang et al., 2016). So, the importance levels of the constructed models and the variables' effects on responses were obviously unfolded.

The significances of the models and variables were ascertained at a confidence level of 95% at the basis of the *F*-test calculations of the software (Yang et al., 2015). The probability, *P* was utilized in order to evaluate the ANOVA model results given at Tables S2 and S3. The *P* values <0.05 meant model terms are significant but higher than 0.10 are not. The first-order empirical equation presented below was used for the statistical analyses of the DoE consequences (Aydiner et al., 2009).

$$\Upsilon = z_0 + \sum_{i}^{k}(z_i \cdot \chi_i) + \sum_{i,j}^{k}\left(z_{ij} \cdot \chi_i \chi_j\right) \tag{4}$$

where Υ is the dependent response parameter, χ_i and χ_j the independent input variables, *k* the total variable number, *i* and *j* the variable numbers, and z_0, z_i and z_{ij} the intercept, linear, and interaction coefficients, respectively. The linear models that had correlations of 0.622 to 0.806 for the responses were retrieved to unique association compliance without deviation between data and predictions by the addition of interactive variables to the polynomial fitting process. To check whether models have high prediction capability at solution points of variable levels to be obtained by confirmation experiments aiming to fulfill the optimal conditions of MOR, coefficients are severally indicated for responses at Table S5.

2.9. Performance optimizations and model confirmations

The optimizations of each MOR operation were jointly executed for their responses using the RSM optimization module of the software enabled to determine the most appropriate levels of input factors accompanied with different operational constraints convinced for each response (Arslan-Alaton et al., 2009). In this regard, the validations of experimental performances of hybrid MOR applications (FEUF, $pFEUF_{UVA}$ and $pFEUF_{UVC}$) were enforced with individual confirmation experiments conducted at the optimized conditions. The optimal circumstances explored for the studied ranges of the input variables except for H_2O_2 were elucidated by the direct use of the software at minimum oxidant and acid consumptions.

Considering that Fenton can be employed continuously to the submerged filtration in practice, the H_2O_2 expenditure or H_2O_2/TOC ratio was minimized as the main aim to reduce operation cost alongside minimizing the acid spend for adjusting initial pH. By carefully taking account the best performance of each MOR treatment, the major target values for the responses R_{TOC}, R_{COD} and *J* were aimed to reach 55, 75 and 80 in the FEUF and 65, 85 and 100 in the pFEUF. The appropriateness of the optimization procedure was approved by an acceptable desirability exceeding 0.8 together with also supplying the most

Table 2
Design levels and actual values of input variables planned for Taguchi DoE arrays.

Process variables				Design levels and actual values			
Code	Symbol	Remark	Unit	Lowest (1)	Low (2)	High (3)	Highest (4)
A	t	Process time	min	15	30	45	60
B	T	Temperature	°C	25	30	35	40
C	pH	pH	–	3	4	5	6
D	H_2O_2/TOC	H_2O_2/TOC ratio	g/g	6	14	22	30
E	H_2O_2/Fe^{2+}	H_2O_2/Fe^{2+} ratio	g/g	3	7	11	15
F	ν_A	Aeration rate	L/min	1	2	3	4
G	ν_W	Vacuum rate	rpm	55	70	85	100
H	M_T	Membrane type	–	UP005	UP020	UH050	UV150
I[a]	I_L	Intensity of light	Lamp number	1	2	3	4
K[a]	L_T	Light type	–	–	UVC-254	UVA-365	–

[a] These two variables, coded as I and K are the additional parameters being only valid for photo-Fenton, so that each one of Taguchi experimental designs applied for Fenton and photo-Fenton enhanced hybrid membrane processes involves all the rests individually.

approximate scores of the major targets. Because light intensity in the reactor could only be controlled by addition or removal of lamp, the optimal pFEUF solutions were searched individually for the numbers of the placed lamps. Thus, lamp number provided the highest score in total per light intensity was selected as the most preferable solution for the reactor lighting. After completion of optimization procedure, the optimized performances were verified with confirmation experiments by also revealing the prediction capabilities of MLR models derived from the ANOVA analyses. Consequently, the competency of the RSM optimizations of the developed MOR treatments which involved UF simultaneously with the Fenton or photo-Fenton oxidations has been clarified in terms of effective and economical treatment of textile wastewater.

3. Results and discussion

3.1. DoE performances of hybrid MORs

According to the DoE matrices of 4^8 for FEUF and of $2^1 \times 4^9$ for pFEUF dedicated at Table S2, all examinations under this title were grounded on the joint statistical evaluations of outcomes obtained from individual AOP and submerged UF and overall hybrid MOR system (Table 3). The DoE results pointed out that the oxidation offered better treatment efficiency than the UF, when they have combined in a reactor. The AOP performances in the FEUF and pFEUF were better with the averages of 2.1–2.6 times in TOC and 1.8–2.2 times in COD. Almost the same average flux of 36 L/m^2·h was observed in the UF of both hybrids. Besides, the water fluxes arrived at the maximum of 79.2 and 119.2 L/m^2·h in the FEUF and pFEUF, respectively. In a general perspective, the UF was more influenced from changing operational conditions in the pFEUF reactor and more effective on the whole system's performance than that in the FEUF.

3.1.1. Fenton enhanced UF (FEUF)

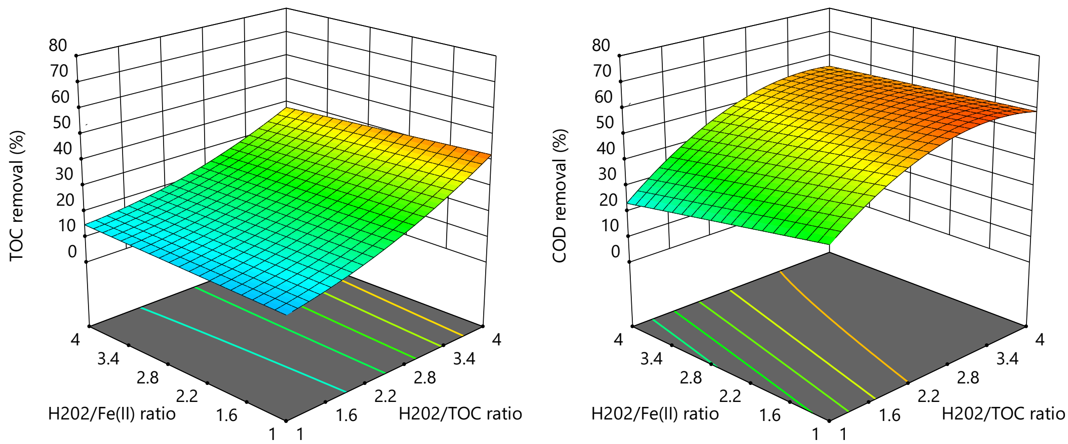
In the FEUF treatment, the organic removal efficiencies of the AOP oxidation and the MOR are indicated at Fig. 3 for the foremost Fenton parameters, i.e., H_2O_2/Fe^{2+} and H_2O_2/TOC. The results in Fig. 3 are independent of UF membrane type and the influences of membrane type on the organics rejections and the UF flux are indicated at Fig. S1. Also, the flux variations of the best UH050 membrane for the UF operating parameters except for the process time are shown in Fig. 4.

From the ANOVA results of the FEUF system (Table S3), all regression models were seen to be too significant for whole responses with *P*-values lower than 0.0001. But the statistical roles of influential variables on the performances to some extent differentiated for responses. While the membrane type and H_2O_2 and Fe^{2+} concentrations prominently affected water flux, the H_2O_2 amount essentially dominated the overall organic removal efficiency. Hence, it was figured out that the H_2O_2/Fe^{2+} ratio has been main adjusting parameter for not only organic treatment but also membrane flux in the Fenton-enhanced submerged UF.

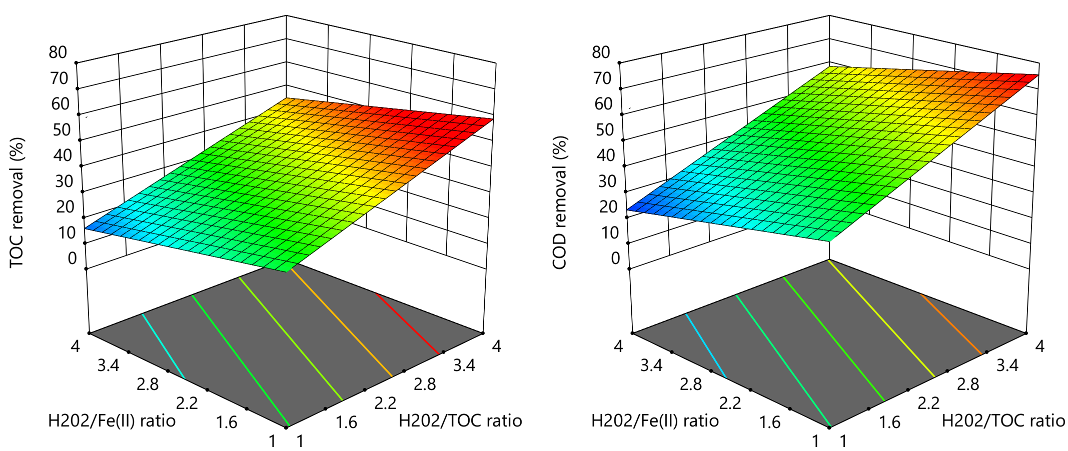
According to Fig. 3, the overall treatment went under dominancy of AOP whose performance was controlled in line with change in oxidant amount. In other words, the organics removals occurred depending mainly on the concentration of H_2O_2 added to the hybrid reactor, and the concentration of Fe^{2+} added provided relatively a lower contribution to the AOP performance. Feng et al. stated that the increased Fe^{2+} amount did not help to improve TOC disruption rate, but instead

Table 3
Taguchi DoE results of AOP, UF and MOR (FEUF and pFEUF) operations.

Test no	FEUF							pFEUF						
	AOP		Submerged UF		MOR			AOP		Submerged UF		MOR		
	R_{TOC} (%)	R_{COD} (%)	R_{TOC} (%)	R_{COD} (%)	R_{TOC} (%)	R_{COD} (%)	J (L/m^2·h)	R_{TOC} (%)	R_{COD} (%)	R_{TOC} (%)	R_{COD} (%)	R_{TOC} (%)	R_{COD} (%)	J (L/m^2·h)
1	31.7	36.0	11.1	16.1	39.2	46.3	79.2	42.0	58.8	19.6	41.0	53.4	75.7	35.4
2	42.7	46.2	8.4	32.9	47.5	63.9	54.8	47.6	61.2	12.9	37.8	54.3	75.9	35.0
3	31.6	59.4	6.8	12.1	36.3	64.3	12.2	35.1	53.5	11.4	25.5	42.5	65.4	9.5
4	22.4	31.8	0.3	10.4	22.6	38.8	61.7	37.0	57.4	12.1	34.7	44.6	72.2	56.7
5	7.3	17.7	8.5	18.8	15.1	33.2	17.9	31.3	56.6	3.4	13.4	33.7	62.4	18.7
6	22.0	30.4	14.7	20.7	33.5	44.8	37.3	30.6	50.2	10.0	13.9	37.6	57.1	34.3
7	35.2	54.0	0.7	5.9	35.7	56.7	17.9	39.5	59.7	15.5	25.3	48.8	69.9	12.9
8	11.6	33.4	14.8	22.7	24.7	48.5	13.3	19.9	35.4	5.4	10.4	24.2	42.1	12.9
9	27.3	58.2	3.3	9.8	29.7	62.3	15.2	39.7	64.7	1.3	6.5	40.4	67.0	6.9
10	42.9	50.1	9.5	15.3	48.3	57.7	40.7	55.0	75.3	13.7	23.9	61.2	81.2	25.5
11	45.4	61.1	7.0	25.2	49.2	70.9	24.4	44.4	57.6	21.0	38.4	56.1	73.9	11.8
12	33.1	61.1	11.7	23.5	41.0	70.2	12.6	29.4	43.3	12.9	24.4	38.6	57.1	15.2
13	4.7	7.5	7.6	14.6	12.0	21.1	60.9	7.0	12.5	11.5	24.0	17.6	33.5	11.8
14	39.5	57.9	18.9	29.1	51.0	70.2	60.2	52.2	63.6	23.3	34.2	63.3	76.0	40.0
15	35.7	44.9	11.5	21.1	43.1	56.5	58.6	35.2	50.0	18.9	36.3	47.4	68.1	45.7
16	34.8	45.4	13.4	22.7	43.5	57.8	29.3	47.5	60.8	12.0	22.3	53.8	69.5	21.7
17	42.8	62.8	16.2	28.6	52.1	73.5	20.2	47.3	60.9	15.0	25.7	55.2	70.9	9.9
18	44.0	55.0	9.8	16.7	49.5	62.5	25.1	50.3	62.4	20.3	42.5	60.4	78.4	57.1
19	46.6	65.8	5.3	9.8	49.4	69.2	16.4	49.6	67.4	9.6	16.5	54.4	72.8	19.3
20	39.2	50.1	11.6	21.7	46.2	60.9	43.0	43.9	69.6	12.6	15.3	51.0	74.3	88.7
21	28.8	37.0	17.8	23.4	41.5	51.7	65.9	34.0	47.8	43.7	53.8	62.8	75.9	119.2
22	40.1	57.5	20.2	35.9	52.2	72.8	7.2	43.6	58.3	15.9	30.1	52.5	70.8	27.8
23	26.6	50.3	11.5	17.0	35.1	58.7	25.1	35.9	62.1	15.3	21.1	45.8	70.1	24.4
24	35.6	62.5	5.6	10.4	39.2	66.4	17.9	42.6	68.2	14.7	23.0	51.0	75.5	15.2
25	14.1	34.6	17.4	25.2	29.1	51.1	28.4	20.8	42.1	24.5	46.1	40.2	68.8	62.8
26	11.3	20.0	15.5	22.3	25.0	37.8	8.4	14.4	23.6	20.9	33.3	32.3	49.1	30.1
27	13.2	29.8	23.6	26.2	33.6	48.2	54.8	16.0	27.8	22.8	33.7	35.1	52.2	49.9
28	13.0	17.8	11.2	17.8	22.7	32.5	74.2	5.7	8.0	28.8	45.0	32.8	49.4	91.4
29	27.7	38.2	13.6	32.1	37.5	58.1	60.9	33.4	55.2	18.5	31.6	45.8	69.4	46.4
30	40.0	58.6	9.2	16.7	45.5	65.6	13.3	35.2	48.9	14.5	24.5	44.6	61.4	16.0
31	35.8	52.8	17.8	23.0	47.2	63.7	18.3	36.5	53.4	28.2	48.6	54.4	76.1	28.2
32	31.1	42.4	7.6	13.7	36.4	50.3	65.0	33.1	49.3	22.8	33.3	48.3	66.2	73.1
Min.	*4.7*	*7.5*	*0.3*	*5.9*	*12.0*	*21.1*	*7.2*	*5.7*	*8.0*	*1.3*	*6.5*	*17.6*	*33.5*	*6.9*
Max.	*46.6*	*65.8*	*23.6*	*35.9*	*52.2*	*73.5*	*79.2*	*55.0*	*75.3*	*43.7*	*53.8*	*63.3*	*81.2*	*119.2*
Aver.	*29.9*	*44.7*	*11.3*	*20.0*	*38.0*	*55.8*	*35.6*	*35.5*	*52.1*	*16.7*	*29.3*	*46.4*	*66.5*	*36.0*
Std. dev.	*12.1*	*15.4*	*5.5*	*7.3*	*10.9*	*13.0*	*22.3*	*12.6*	*15.8*	*8.1*	*11.6*	*11.0*	*11.2*	*27.3*



(*a*) AOP of FEUF



(*b*) FEUF

Fig. 3. Organic removal efficiencies vs. H_2O_2/Fe^{2+} and H_2O_2/TOC in AOP of FEUF (a) and FEUF (b), (all of remaining variables except for H_2O_2/Fe^{2+} and H_2O_2/TOC are at 2.5 DoE coded value).

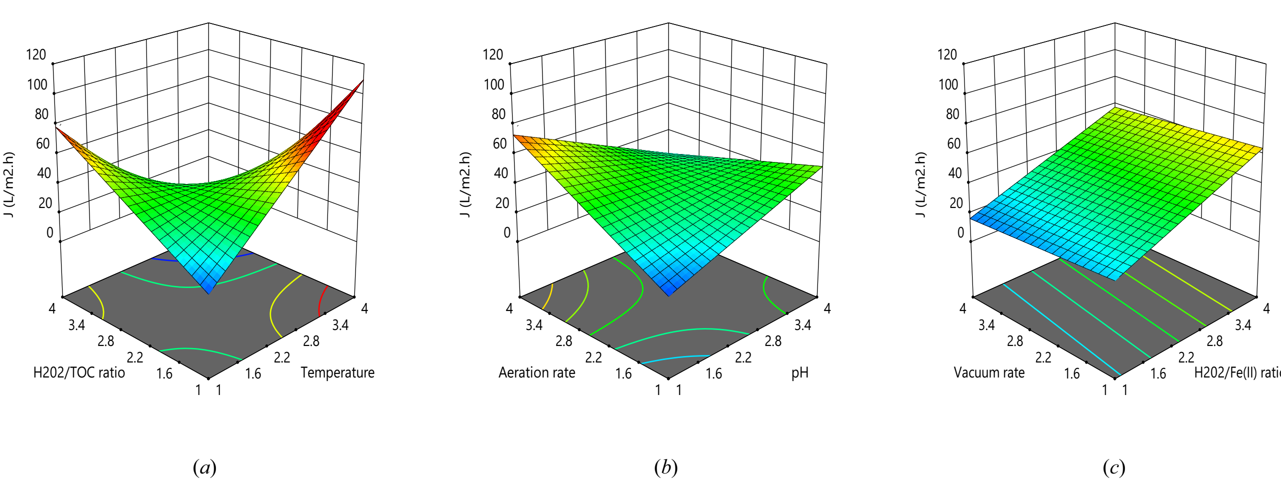


Fig. 4. Water flux performance of UH050 membrane in FEUF against H_2O_2/TOC and T (a), ν_A and pH (b), and ν_W and H_2O_2/Fe^{2+} (c), (all the rest variables except those shown have a DoE coded value of 2.5).

noted caused a slight reduction at TOC removal in high Fe^{2+} amount, although not encountered at the studied ranges of the variables in the hybrid FEUF system (Feng et al., 2010). They pointed out that as inherent to the Fenton, the overdosing of ferrous ions led to the self-destruction of •OH and thereby reduce the disruption of organic substances (Eq. (5)).

$$^{\bullet}OH + Fe^{2+} \rightarrow Fe^{3+} + OH^{-} \quad (5)$$

Fig. 3 indicated that both removal parameters represented the exponential and linear changes in AOP and FEUF, respectively, being a little more in FEUF by contribution of UF process. The system's COD efficiency vs. the H_2O_2 amount increased faster and greater than the TOC. As the H_2O_2 and Fe^{2+} concentrations increased (H_2O_2/Fe^{2+} decreased), as became in the AOP, organic rejection improved for COD more notably. The differences between results regarding the H_2O_2 and Fe^{2+}consumptions were due to the fact that organic substances decomposed to intermediate products, as understood, did not turn fully to end products such as CO_2, H_2O etc. The best efficacies for the TOC and COD removals were found at the condition of maximum H_2O_2 and minimum Fe^{2+} with 42% and 58% by the AOP and 58% and 74% by the FEUF. The effectiveness corresponding to the differences between relevant components in Fig. 3 showed that the UF not only had a performance lower than the AOP, but also contributed somewhat to the overall treatment efficiency. The partial low organic rejections by each one of membranes immersed to the reactor were also affirmed from its performance graphs visualized separately for all membranes (Fig. S1). Nevertheless, performance changes appeared at trends specific to every response, as independent of membrane kind. Despite different MWCOs and materials, the flux considerably declined linearly at the similar rates in all membranes when the amounts of both H_2O_2 and Fe^{2+} rose to their limit operating values. UH050 was determined as the most contributing membrane to the overall degradation efficiency. In addition to UP020, it allowed obtaining comparatively higher rejections with a performance of 16% TOC and 35% COD at maximum H_2O_2 and minimum Fe^{2+}, while the best fluxes were attained as about 80.0 $L/m^2 \cdot h$ at minimum amounts of H_2O_2 and Fe^{2+}.

In the order of significance of effects, the important variables on the UF flux were membrane, H_2O_2/Fe^{2+} and vacuum rate, whereas aeration rate and temperature were at partial importance (Table S4). Even so being higher markedly for temperature and H_2O_2, the flux increased with parametric values increasing to the DoE limits. The flux declined specifically by the decreases in the variables' values, except for vacuum rate. It was undoubted along the hybrid treatment that organic contaminants in the reactor were withdrawn to the membrane surface by vacuum, water passed through the membrane but organic molecules from small to large were partially retained over the surface. As a matter of fact, a reddish-brown layer formed by accumulation of ferric particles along the entire membrane surface was noticed after the FEUF experiments. It was perceived that when Fe^{2+} increased (H_2O_2/Fe^{2+} decreased), the flux chiefly decreased by both the presence of secondary membrane layer and the decreasing of water permeability or porosity of this layer. On the other hand, the UF water flux that differed sharply regarding H_2O_2/TOC and temperature effects proved that the water permeability of the membrane was indirectly dependent on organic degradation efficiency of the Fenton oxidation in the reactor. It can be accordingly claimed that the formation of concentration polarization layer including macromolecules and low molecular weighted solutes extremely likely at immediate boundary of the surface particle layer toward reactor liquor. In that circumstance, the rejected molecules diffused back to bulk solution as consequence of concentration gradient just over the surface, leading also to higher fouling resistance by penetration of them into the membrane and hence lower flux (Mozia et al., 2006; Aydiner, 2010; Shi et al., 2014). Another evidence proving the polarization's presence has been the severe decline in the flux occurred by the reduction of the airflow which was applied for diminishing the membrane's surface fouling.

3.1.2. Photo-Fenton enhanced ultrafiltration (pFEUF)

In the pFEUF system, the organic treatment efficiencies of AOP oxidation and overall MOR were displayed for the essential photo-Fenton parameters in Figs. 5 and 6 in views of variable groups of H_2O_2 and Fe^{2+}, and H_2O_2 and intensity of light, respectively. The results presented in the figures are independent of the UF membrane and UV light types, and the membrane effect graphs on the UF responses were shown in Figs. S2 and S3 for the abovementioned order of variable couples. Apart from these, the flux performances of the UH050 membrane that performed the best at quite high organic efficacy (DoE test no: 21) were exhibited in Fig. 7 together for all system parameters.

The ANOVA analyses of the pFEUF system (Table S4) indicated that regression models were significant ($P < 0.05$) for responses, but significance levels relatively lessened for COD and *J*. The statistical results of variables effective to responses did not differ heavily with each other. With inevitable activity that resulted in better degradation than mineralization, the operational performances of AOP and pFEUF came along with similar variation trends in TOC and COD for the specific ranges of the variables. H_2O_2 and time came into prominence as the adjudging variables that undertook the whole performance of hybrid reactor as well as temperature and membrane that were found as the other principal factors for TOC and water flux, respectively. Unlike FEUF, the contribution of the added Fe^{2+} to the overall performance was determined at a trivial level. This pointed out to the fact that Fe^{3+} generated from Fe^{2+} by Fenton reactions repeatedly reduced to Fe^{2+} ions with not only the UV light but also the oxidation of organic-radicals (•R_i) according to Eqs. (6)–(10) (Moraes et al., 2004; Bacardit et al., 2007; Arai et al., 2008; Karthikeyan et al., 2011). The recurrence of Fe^{2+} triggered by irradiation appeared to be more pronounced than by organic decomposition because the photo-Fenton performed better and faster oxidation activity than Fenton alone in hybrid reactor. At this frame, it can be stated that organics more effectively degraded by photo-oxidative Fenton reactions by which the performance affected mainly from the principal photo-Fenton variables, viz., H_2O_2 and intensity of light (Figs. 5 and 6).

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^{-} + {}^{\bullet}OH \quad (6)$$

$$Fe^{2+} + {}^{\bullet}OH \rightarrow Fe^{3+} + OH^{-} \quad (7)$$

$$^{\bullet}R_i + Fe^{2+} + H^{+} \rightarrow Fe^{3+} + R_iH \quad (8)$$

$$Fe^{3+} + H_2O + h\nu \rightarrow Fe^{2+} + H^{+} + {}^{\bullet}OH \quad (9)$$

$$^{\bullet}R_i + Fe^{3+} \rightarrow Fe^{2+} + R_i^{+} \quad (10)$$

Using UH050, the highest COD removal vs. the H_2O_2 and Fe^{2+} additions was 72% by AOP at the maximum amounts, while reached 78% by pFEUF at maximum H_2O_2 and minimum Fe^{2+}. Mineralization of organic matters was brought to around 54% by the photo-assisted Fenton. The increase in H_2O_2 remarkably raised the removal efficiency of each type of light source, especially at its low amounts. As the light intensity and oxidant amount rose, mineralization to end-products and degradation to intermediate species yielded 47 and 62% in photo-Fenton and 59 and 80% in hybrid MOR, respectively. In the presence of H_2O_2, the noticeable rises in performances by more addition of Fe^{2+} have emerged in neither AOP nor overall system. The differences between the results of AOP and pFEUF demonstrated a clear reduction in the UF efficiency compared to FEUF as the result of the fact that organic matters more degraded to intermediate molecules at lower sizes that weakened the membrane's sieving mechanism. From opposite perspective, it was comprehended that the membrane behaved as a barrier that capable

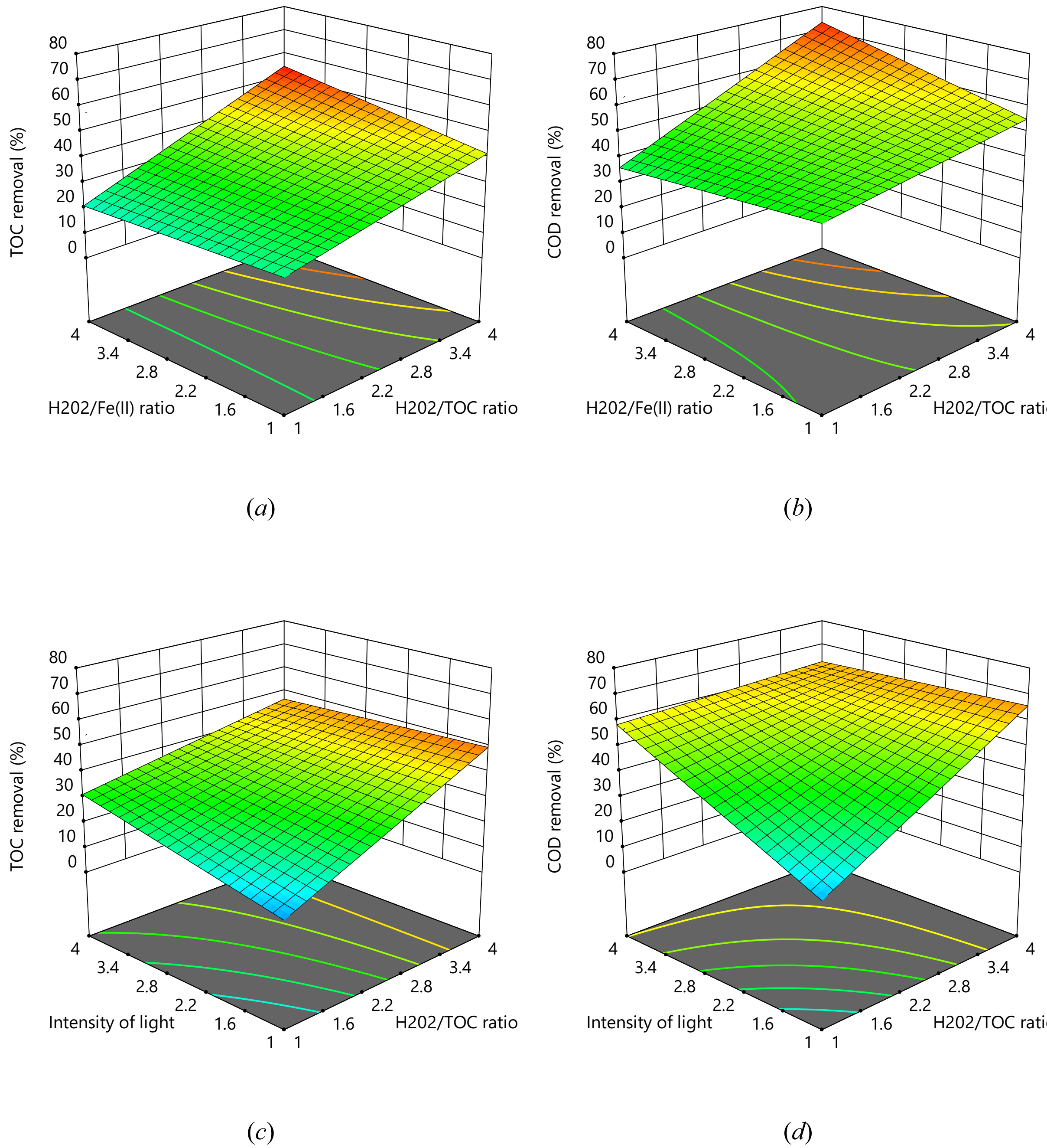


Fig. 5. Organic removal efficiencies in AOP of pFEUF versus H_2O_2/Fe^{2+} and H_2O_2/TOC ((a) and (b)) and intensity of light and H_2O_2/TOC ((c) and (d)) (all of remaining variables except for those shown are at 2.5 DoE coded value).

of rejecting far more organic species depending on relative larger molecular sizes composed due to weak structural fractionation under descending effect of hydrolysis at low H_2O_2 expenses (Figs. 5 and 6).

The contribution of UF to the overall reactor efficiency was more considerable in the pFEUF than that in the FEUF, as affirmed by the visual graphs of four membranes (Figs. S2 and S3). Although the UV150 membrane had the best flux with 100.0 L/m^2·h, the UH050 membrane possessed the most favor rejections by 26% TOC and 60% COD at maximum H_2O_2 and Fe^{2+}. The water flux of 70.0 L/m^2·h at the minimums of reagents declined in a linear behavior for membranes mostly with the H_2O_2 increment, except for the UP020 which behaved in reverse. Despite a successful operation of the membrane supported by photo-Fenton, it is perceived that the flux decline became via the formation of much smaller forms of organic constituents. In that sense, it is understood that the water flux deteriorated based on the membrane fouling that occurred due to retaining of the disrupted smaller molecules not only on the surface and but also in the pores. All variables seemed to have obvious effects on flux changes of UH050 (Fig. 7). As long as the temperature, aeration rate, intensity of light and processing time were elevated, the permeate flux was objectively improved, vice versa for H_2O_2, pH and Fe^{2+}, and as independent of the vacuum rate in all. Hermosilla et al. (2012) reported that the variation in temperature from 25 to 45 °C did not appreciably affect the photo-oxidative degradations. However, it was ascertained for the photo-Fenton supported treatment that the influence of temperature in the corresponded operating range was highly important for the UF water flux. A flux decline as about half as that of FEUF was seen by increasing of Fe^{2+} to maximum (Figs. 3(c) and 7(c)). This meant that more Fe^{2+} involved in the secondary membrane layer through a reduction of $Fe^{3+}$to Fe^{2+} by UV light, which in turn concluded with Fenton reactions enhanced on the surface

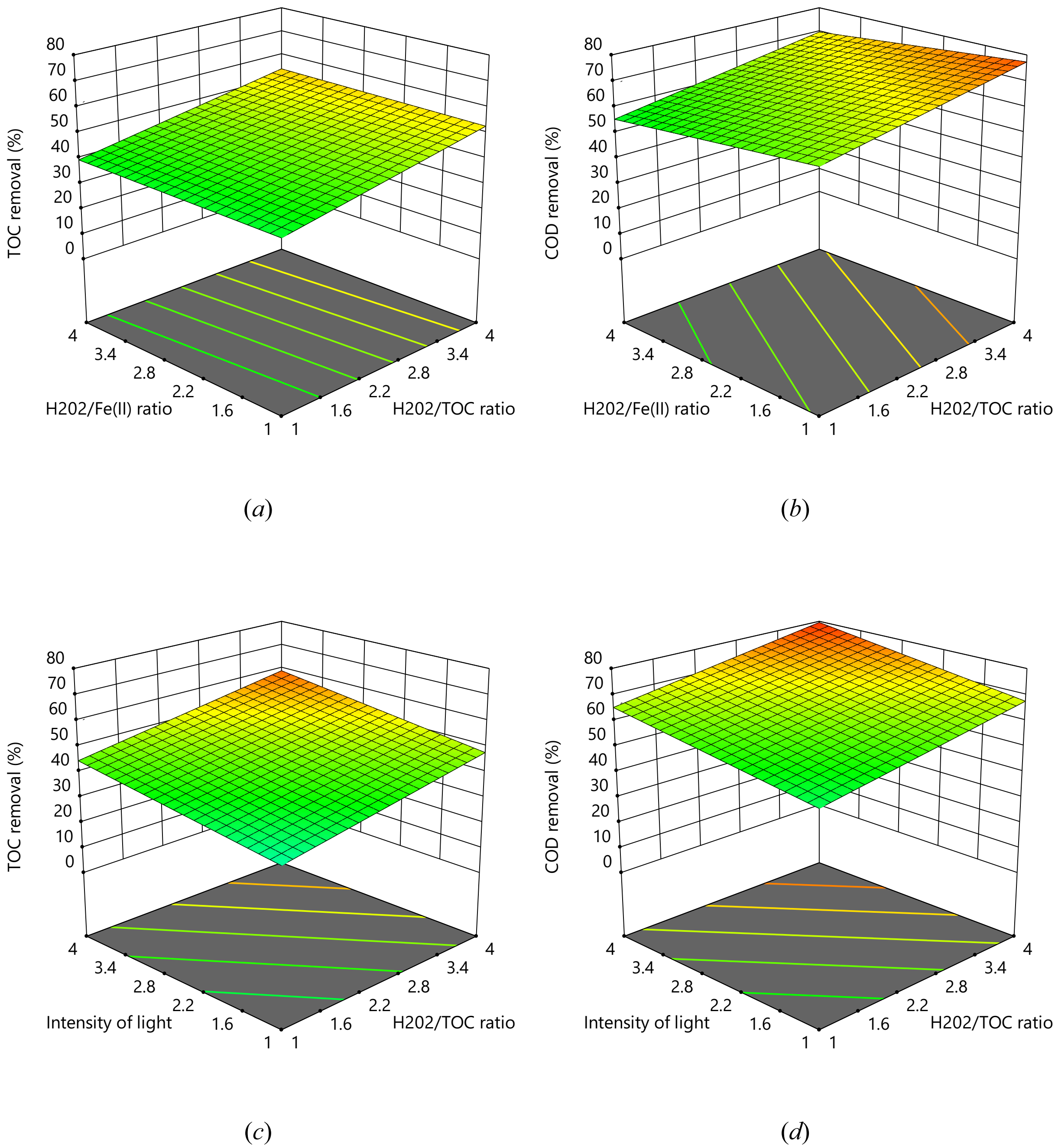


Fig. 6. Organic removal efficiencies in pFEUF in view of H_2O_2/Fe^{2+} and H_2O_2/TOC ((a) and (b)) and intensity of light and H_2O_2/TOC ((c) and (d)) (all of remaining variables except for those shown are at 2.5 DoE coded value).

that improved impressively the organics rejection of the membrane. Moreover, the most likely reason for the flux decline vs. pH increment is that the membrane's pores clogged intensely by much more organic molecules and ferric ions penetrated from the surface to the membrane due to the slowing hydrolysis reactions within the reactor.

3.2. Optimized performances of hybrid MORs

The optimum operating conditions were determined as the most preferable solutions by RSM optimization and are introduced in Table 4 individually for each MOR treatment applications. The desirability values of the solutions optimized by use of the software were estimated to be 0.9190, 0.9248 and 0.8289 for the FEUF, $pFEUF_{UVA}$ and $pFEUF_{UVC}$ operations, respectively. The experimental results optimized severally for the whole of the hybrid reactor supported by Fenton or photo-Fenton are shown in Fig. 8 by the time elapsed until the residual H_2O_2 in the effluent was zero. The quality analyses of effluents produced from confirmation experiments are presented in Fig. 9.

Since the amounts of the reagents that yielded the less oxidation at lower amounts in Fenton optimally increased in photo-Fenton, better MOR performances were observed by the UV irradiation which improved the oxidation activities of organic substances. The H_2O_2 and Fe^{2+} expenses in Fenton oxidation were optimized to values of 6.0 and 0.4 per unit TOC in optimal temperature of 26.5 °C, respectively. The expenditures optimized for a temperature of 40.0 °C in both of photo-Fenton that used three lamps reached up their optimums of 7.75 and 0.72 at 24 W UVA and 9.88 and 1.36 at 30 W UVC. The optimal durations were estimated as 60, 60 and 41 min for FEUF, $pFEUF_{UVA}$ and $pFEUF_{UVC}$, respectively. The optimized pH of wastewater was elevated from 3.73 to 4.0 and final 4.44 at the mentioned order of operations

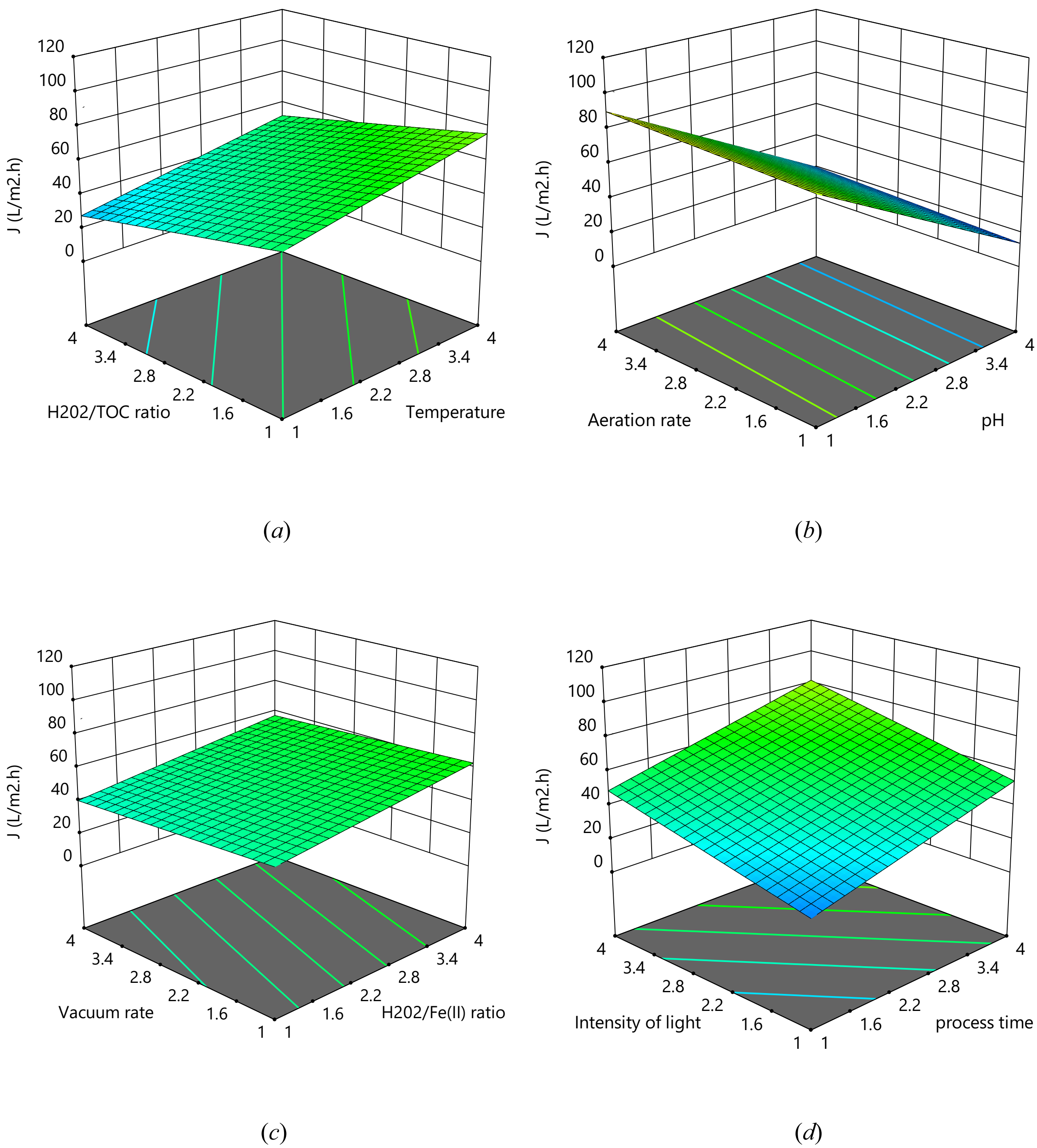


Fig. 7. Water flux performance of UH050 membrane in pFEUF against H_2O_2/TOC and T (a), ν_A and pH (b), ν_W and H_2O_2/Fe^{2+} (c), and I_L and t (d) (all the rest variables except those shown have a DoE coded value of 2.5).

without being encountered a reduction in the organic disruption efficiency in spite of the self-destruction of hydroxyl radicals that is known to accelerate at high pH values (Ebrahiem et al., 2017). Torrades and García-Montaño (2014) found the optimal conditions of Fenton and photo-Fenton as pH 3.0, 0.16 Fe^{2+}/TOC (100 mg Fe^{2+}/L) and 4.03 H_2O_2/TOC (2500 mg H_2O_2/L) to degrade organic matters in real dye wastewater at optimized duration of 120 min. Arslan-Alaton et al. (2009) optimized the reagent spending to 1.17 Fe^{3+}/TOC (84 mg Fe^{3+}/L) and 16.53 H_2O_2/TOC (1190 mg H_2O_2/L) at optimum reaction time of 45 min in the treatment of synthetic azo dye production wastewaters by the photo-Fenton-like advanced oxidation. Sreeja and Sosamony (2016) designated the optimum catalyst and oxidant dosages for the treatment of synthetic textile wastewater by photo-Fenton as 0.014 Fe^{2+}/COD (10 mg Fe^{2+}/L) and 7.14 H_2O_2/COD (5100 mg H_2O_2/L) at the optimal conditions of pH 3.0 and 16 W power. In light of the optimum AOP performances abovementioned, it can be stated that the cost minimization goal conducted specifically to the optimal organic treatment of textile washing wastewater was accomplished by means of the reduction of acid and oxidant consumptions. It can be also expressed that the novel reactor could be applied for the treatment of different industrial wastewaters as talented at reasonable or shorter operating periods by performance optimization of UV enhanced Fenton oxidations. In this stage, it is recommended to conduct a detailed economic analyze that fully reveal the real economic performances of the developed hybrid treatments, as to be comparable to other MOR techniques such as PMR.

UH050 and UV150 were recognized as the best membrane solutions for optimal treatments. Besides, slower oxidation rate in the Fenton

Table 4
Optimized operating conditions of FEUF and pFEUF treatments in MOR.

Process variables	Unit	FEUF		pFEUF			
				UVA		UVC	
		Level	Value	Level	Value	Level	Value
t	min	4.00	60.0	4.00	60.0	2.71	41.0
T	°C	1.30	26.5	4.00	40.0	4.00	40.0
pH	–	1.73	3.73	2.00	4.0	2.44	4.44
H_2O_2/TOC	g/g	1.00	6.00	1.22	7.75	1.49	9.88
H_2O_2/Fe^{2+}	g/g	4.00	15.0	2.94	10.75	2.07	7.27
ν_A	L/min	1.79	1.79	1.00	1.00	2.89	2.89
ν_W	rpm	2.65	80.0	2.07	71.1	1.65	64.7
M_T	–	3	UH050	4	UV150	3	UH050
I_L	Lamp number	–	–	3 lamps		3 lamps	
L_T	–	–	–	3	UVA	2	UVC

concluded with residual H_2O_2 of 480 mg per L of effluent at optimal processing time, but zero and 8 mg/L in the UVA and UVC photo-Fenton. For specific pH need varying from 2 to 5 at Fenton reactions (Xu et al., 2017), optimal FEUF performances concluded that the variables H_2O_2, pH and Fe^{2+} had substantial influence on the organics eliminations. The higher extent of organic degradations by photo-Fenton was taken place only up to a certain limit by the increasing oxidant amount by means of extra production of •OH, supported by Ay et al. (2009). In the presence of negligible ferrous amounts of 3–10 mg/L remained in the MOR after 30 min reaction, the degradation and mineralization rates were reduced proportionally by the decreasing of H_2O_2 depletion rate in each hybrid run that induced reaching of variations in COD or TOC virtually to a constant stability. In the optimal durations, the COD and TOC eliminations were provided by 65.6 and 46.6% for FEUF, 72.1 and 54.2% for $pFEUF_{UVA}$, and 74.4 and 46.8% for $pFEUF_{UVC}$, respectively. The decreasing of residual H_2O_2 in the reactor by the time has been obvious evidence that the immersed membrane can be effectively employed without being any loss of oxidant during the oxidation reactions by starting up the membrane only after 100 min for Fenton or 40 min for photo-Fenton. In case of complete depletion of oxidant that required an extra time of 3.0 times by UVC and 4.4 times by UVA, the FEUF's achievement arrived at nearly the same treatment levels as those in pFEUF of 73.1% COD and 53.1% TOC. By considering the optimal oxidation conditions of textile wastewaters mentioned in literature, the overall MOR performances can be assessed at reasonable levels for TOC-COD efficacy in Fenton and photo-Fenton that were reported respectively as 58.1–62.9% and 70.4–76.3% by Torrades and García-Montaño (2014), and as 59.0–78.0% in photo-Fenton-like by Arslan-Alaton et al. (2009), while as 47.0% only for the COD in the photo-Fenton by Sreeja and Sosamony (2016).

From a general viewpoint upon the optimal performances of treatment techniques developed, it was deduced that they could be operated as having substantial advantages of photocatalytic membrane reactors (PMR) that capable of less operating time minimized by the maximized synergistic effects from the complement processes. Furthermore, iron particles retained in the interior of the reactor by the membrane can be efficiently utilized again for a continuous cycle of treatment via the recurrence of ferrous ions under UV lighting which diminish also to some extent the membrane's surface fouling, as for TiO_2 particles in the PMR (Iglesias et al., 2016). Therefore, the utilization of newly developed MORs in reclaiming industrial wastewaters are regarded as more favorable than the use of traditional coagulation-sedimentation due to its ability preventing the disposal of slurry to be formed in the reactor.

The treated industrial wastewaters in Istanbul-Turkey are discharged in accordance with the discharge limits described in the regulation of Istanbul Water and Sewerage Administration (IWSA) (2013). For industrial discharges to the wastewater infrastructure facilities of which sewage systems are resulted in a complete treatment, relevant parametric values of effluents are limited to pH 6–12, 1000 mg COD/L and 1700 mg SO_4^{2-}/L, Accordingly, the pH can be easily increased to

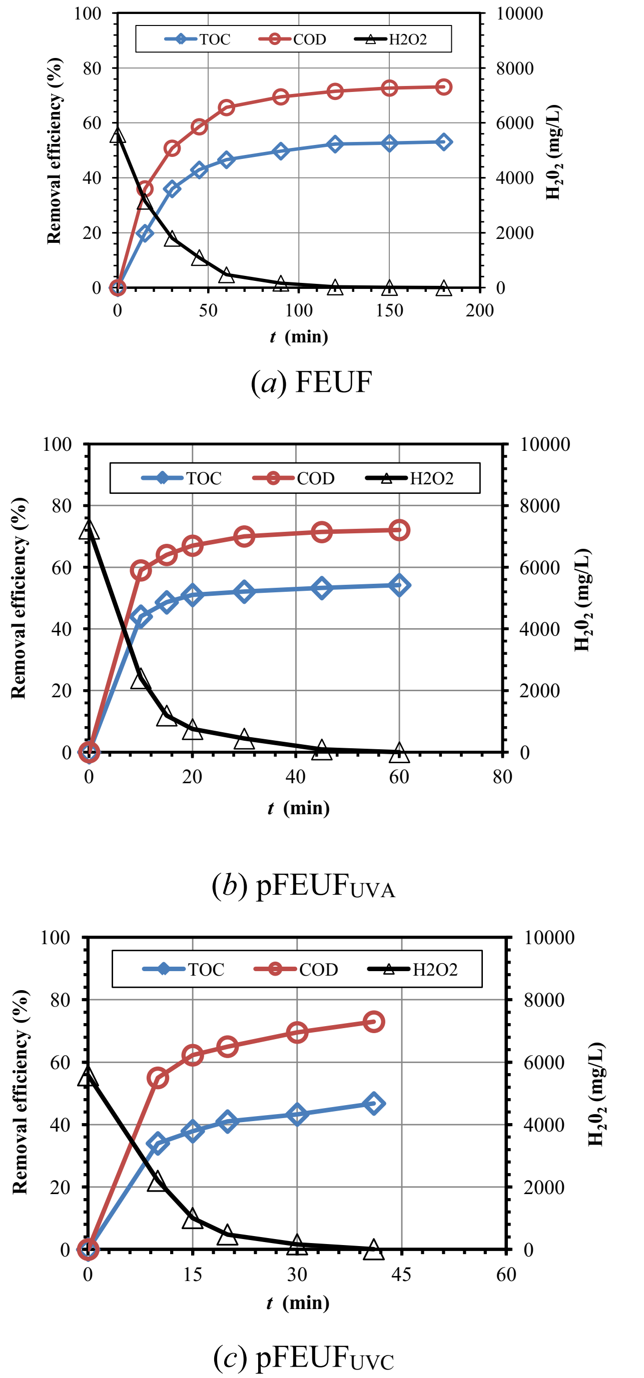
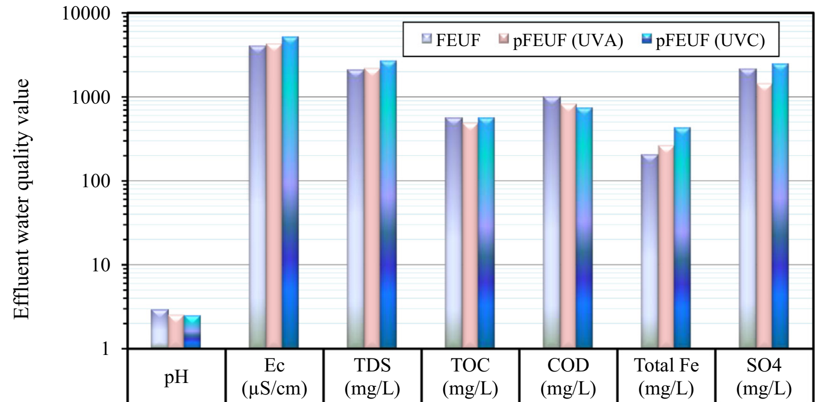


Fig. 8. Time variations of R_{TOC} (%) and R_{COD} (%) and residual H_20_2 at optimal conditions of MOR treatments by confirmation experiments ((a)-FEUF, (b)-$pFEUF_{UVA}$ and (c)-$pFEUF_{UVC}$).

discharge levels in the reduced need of a base such as NaOH. Since the use of H_2SO_4 for pH adjustment before oxidation was the main reason why sulphate was over its limits in effluents, the MOR-treated waters can be readily released to sewer systems due to their reduced organic contents by direct use of HCl in place of H_2SO_4 as to meeting a limit of 15 g Cl^-/L. Although heavy metals are not considered at the scope of this study in terms of the content of the wastewater used, other parametric values of discharge standards are 500 mg SS/L and 10 mg phenol/L with no limited value for color, toxicity and total Fe. The effluents produced by the FEUF and pFEUF treatments had the qualities of 40 and 70 mg SS/L, 6.9 and 4.2 mg phenol/L, 0.248 and 0.027 absorbance at 436–620 nm, 5 and 11 mg Cl^-/L, and 201 and



	pH	Ec (µS/cm)	TDS (mg/L)	TOC (mg/L)	COD (mg/L)	Total Fe (mg/L)	SO4 (mg/L)
FEUF	2.85	4030	2072	550	974	202	2140
pFEUF (UVA)	2.56	4200	2140	481	809	261	1422
pFEUF (UVC)	2.46	5060	2620	548	724	416	2450

Fig. 9. Water quality analyses results of effluents produced optimally by MOR.

406 mg total Fe/L, respectively. This study clearly proves that the effluents produced by the developed MOR techniques can be directly discharged to sewage infrastructure ending up with a centralized wastewater treatment plant.

3.3. Efficiency comparisons of hybrid MORs and integrated [AOP/UF]

The developed MOR applications were operationally compared with each other based on their optimized performances. The efficiency

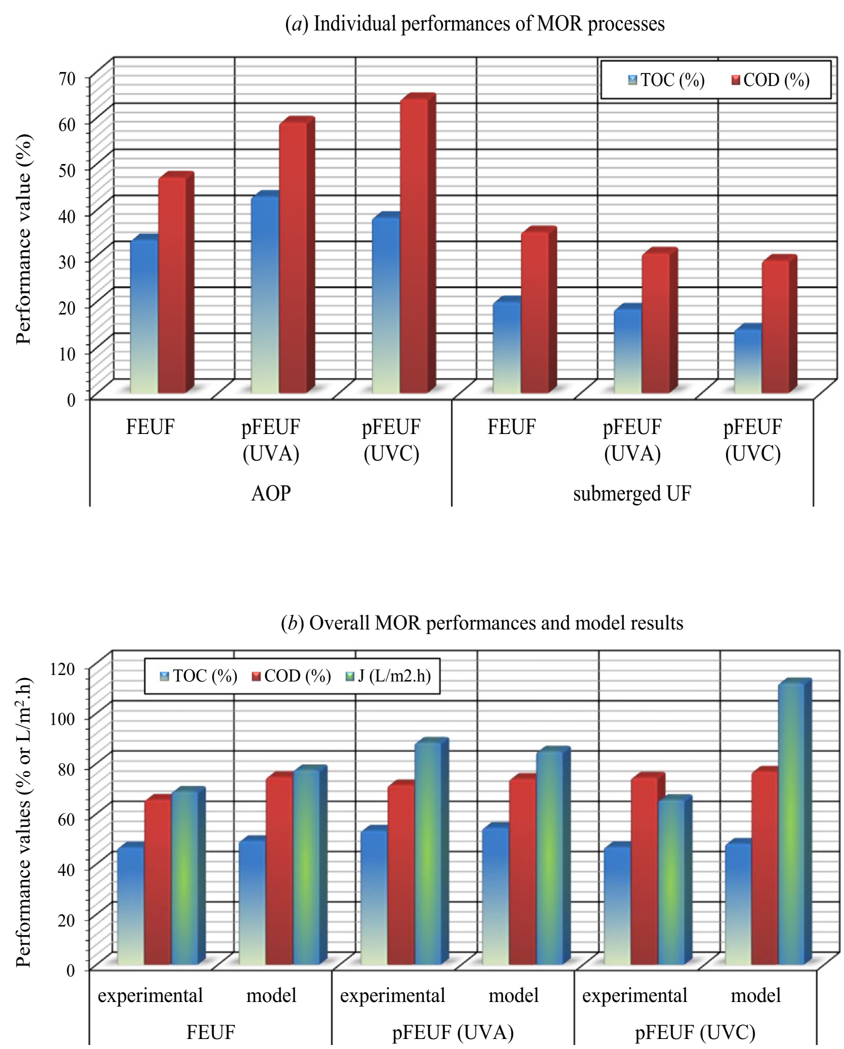


Fig. 10. Performance comparisons of hybrid FEUF, $pFEUF_{UVA}$ and $pFEUF_{UVC}$ treatments ((a)-process performances and (b)-MOR performances and model results).

comparisons of each process and system applications are illustrated in Fig. 10 in conjunction with the prediction outcomes of the model equations. According to a usual treatment comprising of AOP followed by external UF, i.e., [AOP/UF], overall MOR efficiency was evaluated through considering diversities to coming together of AOP and UF technologically. For this, the effectiveness of the distinctive systems constituted by different assembling of UF to the AOP which were operated at optimal Fenton were shown in Table 5 for the whole performances of novel MOR and integrated [AOP/UF].

As opposite to UF, the removal tendency of organics increased from Fenton to photo-Fenton. Because one third less time was executed for the AOP, mineralization to final products stayed somewhat lower in the UVC than the UVA. The disruption of dye molecules to smaller forms decreased the membrane's rejection efficiency depending on the increasing yield of oxidation. Although the MOR has confronted by a dilemma regarding effective operations of processes, it is noteworthy that overall efficacy was improved with a synergistic effect that resulted from operating them in together. The AOP and UF processes were individually run by the optimal percentages of 38.1 ± 4.7 TOC and 56.6 ± 8.7 COD, and of 17.3 ± 3.1 TOC and 31.5 ± 3.2 COD, respectively. The optimized water flux was obtained by representative average of 74.2 ± 12.3% $L/m^2 \cdot h$ in spite of the studied oxidation varieties of MOR.

The confirmative results were at satisfactory levels for all models except the $pFEUF_{UVC}$ flux. The estimation results of the FEUF models were verified with absolute errors below 13.7%. Very good agreements were obtained for $pFEUF_{UVA}$ by acceptable errors not exceeding 4.0%. Despite agreeable errors of about 7% for the efficiencies, a downward deviation of 71% arose in the UF flux of the UVC-Fenton reactor. This has been attributed to the insufficiency of the flux model used for the optimized performance verification due to the low desirability of 0.83. In fact, the main reason underlying of this discrepancy in a viewpoint of transport dynamics is that dye molecules which disrupted by oxidation to smaller organic structures tended to excite more severe fouling of the membrane as a consequence of their quite much participations to its interconnected pore voids as well as to the membrane's surface.

Compared to the integrated [AOP/UF] system of 47.9% TOC, 79.0% COD and 60.9 $L/m^2 \cdot h$, simultaneous or sequential UF-hybridized Fenton deducted in more attractive performances of 74.5%, 85.9% and 99.4 $L/m^2 \cdot h$, and 70.5%, 84.5% and 155.6 $L/m^2 \cdot h$, respectively. It was determined that the submerged inclusion of UF to AOP in a reactor ensured better efficiencies and flux than the external integrated system without notable inorganic increase in effluents. Thus, it was concluded that the Fenton or photo-Fenton enhanced hybrid UF operations can be applied at field with financial success by means of reducing the consumptions of reagents via the ongoing catalyst activity of Fe^{2+} at a cyclic treatment schedule under the influences of especially UV lighting and partially dye decomposition, as well as the prevention of oxidant escape out of the reactor by sequential operations. For economically realization of both hybrid MORs based on reducing the capital and operating costs in industrial wastewater treatment, another factor that is as crucial as the minimization of amounts and losses of H_2O_2 and Fe^{2+} is interoperability with AOP at high water flux of the membrane immersed to the reactor. In this respect, further studies on trade-off between fouling and flux behaviors are suggested in terms of demonstrating effective and economic uses of promising membrane processes to be able to be applied in the direct MOR or post-MOR treatments of domestic, municipal, other industrial organic wastewaters and organic membrane concentrates.

4. Conclusions

Novel membrane oxidation reactors built by hybridizations of Fenton and photo-Fenton to submerged UF were successfully demonstrated for treatment of textile washing wastewater. Thanks to the statistical RSM optimizations of Taguchi experiments designed for each hybrid, high purification efficiencies were achieved with reduced chemicals and oxidation time in that the photo-Fenton enhanced hybrid guaranteed better organic elimination than the Fenton. The most important operating factors influenced overall MOR performances were found as the H_2O_2 amount, process time, membrane type, Fe^{2+} concentration and wastewater temperature. The contribution of UF to organic removal efficiency of the hybrid reactor was determined as 38.1 ± 4.7% COD and 17.3 ± 3.1% TOC, while Fenton and photo-Fenton oxidations provided a more pronounced contribution by degradation of 56.6 ± 8.7% and mineralization of 31.5 ± 3.2%. The expenditures of H_2O_2 and Fe^{2+} per unit TOC were minimized, respectively, to 6.0 and 0.4 in Fenton of 26.5 °C, 60 min and 3.73 pH; to 7.75 and 0.72 in the UVA-Fenton of 40.0 °C, 60 min and 4.0 pH; and to 9.88 and 1.36 in the UVC-Fenton of 40.0 °C, 41 min and 4.44 pH. Different assembling strategies of UF to Fenton, i.e., simultaneously or sequentially hybrid MOR and externally integrated [AOP/UF] proved that effluents able to be discharged direct to sewerage systems resulting in a complete treatment could be readily produced with suitable organic and inorganic content by each assembling. But, sequentially MOR hybridization was determined as the most preferable solution by the highest efficiency of 84.5% COD, 70.5% TOC and 155.6 $L/m^2 \cdot h$ water flux. This study indicated that the developed MOR techniques are technologically favorable for high purification of industrial organic wastewaters and seem to have ability to be successfully testable at field scale because of their less resource, time and installation space requirements. Further works on distinct MOR variations that would be relied on the hybridizations of AOPs to the promising desalination technologies are suggested in order to develop other new solutions that are technically superior and economically viable in solving various water and wastewater issues.

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Table 5
Overall treatment performances and effluent qualities results of hybrid MOR and integrated [AOP/UF] systems.

Parameter	Unit	Raw wastewater	Submerged UF in MOR (FEUF)				[a]External UF after AOP in [AOP/UF]	
			Simultaneous to AOP		Sequential to AOP			
			Effluent	*R* (%)	Effluent	*R* (%)	Effluent	*R* (%)
pH	–	3.73	2.62	–	2.51	–	2.23	–
Conductivity	μS/cm	1218	2171	−78.2	2580	−111.8	2690	38.7
TDS	mg/L	602	1100	−82.7	1313	−118.1	1373	40.0
TOC	mg/L	400	102	74.5	118	70.5	279	47.9
COD	mg/L	1132	175	85.9	175	84.5	329	79.0
J	$L/m^2 \cdot h$	–	99.4		155.6		60.9	

[a] Volume reduction factor as the ratio of the starting feed volume to the final concentrate volume was applied as 5.0 during external UF treatment.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2019.05.248.

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