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Natural Gas Processing Wastewater Management

Fayza Aly Nasr, Ibrahim Abdelfattah*, Abdelrahman Shana
Water Pollution Research Department, National Research Centre, 33 EL Bohouth St., P.O. 12622, Dokki, Giza, Egypt.

THE aim of this research is to focus on adequate treatment technology for handling wastewater discharged from natural gas processing industries. An Egyptian natural gas company located at Alexandria was selected for the study. The study represents a useful reference for treatment of such kind of wastewater. Dark Fenton, biological treatment and the combination of them were carried out. The wastewater produced by subject industries has high concentration of chemical oxygen demand, (COD=52665 mg/L), low biological oxygen demand (BOD=8350 mg/L) and low total suspended solids (TSS=370 mg/L). The value of BOD/COD was low (0.16) which indicates that this wastewater inhibits the metabolic activity of bacterial seed because of their refractory properties causing biodegradability to be difficult. Fenton oxidation treatment raise the BOD/COD to more than 0.36. Biological treatment using both aerobic and anaerobic bioreactors for the treatment of mixture of Fenton pretreated gas processing wastewater and sewage gave satisfactory results. The subsequent anaerobic treatment using UASB reactor at optimum HRT [18h] qualifies the produced wastewater for discharge into the sewerage system. The additional aerobic treatment using the continuous-flow activated sludge at 12h HRT qualifies the treated wastewater for discharge into the sea.

Keywords: Refractory, Industrial, Wastewater, Treatment, Safe discharge.

Introduction

Natural gas production is a sophisticated cleaning process developed to purify the gas and remove non-methane hydrocarbons and fluids by separation from the incoming gas stream, producing the so-called dry natural gas for export. Natural gas is in the wet condition as extracted which enhances formation of methane hydrates in the transmission pipelines that downgrades the natural gas quality. To satisfy the recognized specs, the water content upper limit should be within 9.3×10^{-4} liters of water per cubic meter of gas. Dehydration of natural gas is performed using ethylene glycol which absorbs water vapor from the gas stream. The process relies on the contact interface between the wet gas and glycol solution whereby glycol particles absorb water from the gas and become heavier thus sink to the bottom of the reactor before being removed. The effluent wastewater from the dehydration unit generally contains organic and inorganic pollutants of high (COD). Ethylene glycol wastewater is toxic, carcinogenic and hardly biodegradable [1, 2]. The glycol solution, bearing all the water stripped from the natural gas, is disposed of by discharge into surface waters, underground injection, storage in open air pits, and road spreading. Subject options present significant risks to public health or the environment. It means that these organics should be removed from industrial wastewater before reaching water streams [3, 4].

Advanced oxidation processes [AOPs] has been widely used in removal of pollutants [5-7]. Furthermore, they considered as interesting alternative, for their possible use in combination with biological treatments for wastewater treatments, a pretreatment step, via partial oxidation it is improving the biodegradability,

*Corresponding author e-mail: iabdelhafiz@yahoo.com, Tel: 0020-12-74999938,
ORCID: 0000-0003-2567-8830
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or as a post treatment for persistent compounds degradation [1]. Turan [8] investigated the treatment of shale gas wastewater utilizing effective electro-Fenton (EF) system. The treatment efficiency depended on the H_2O_2/COD, initial pH and reaction time, the maximum COD, color and total phenol removal efficiencies were around 87.3%, 89.1% and 91.75% under the optimum conditions, respectively.

Nasr et al, (2016) studied pretreatment of gas processing wastewater using Fenton oxidations and determined the optimum operating conditions: H_2O_2 dose of 1.6 M, Fe^{2+} dose of 80 mM, reaction time of 30 minutes and at pH around 3. [1]. The treated effluent characteristics didn't satisfy the limits set by law for wastewater discharge into the sea but the biodegradability [BOD/COD] improved from 0.17 to 0.36 in 30 minutes. Hence, this facilitates the subsequence biological treatment.

Category wise, biological treatment systems are divided into two basic types: aerobic and anaerobic treatments which take place in the presence and absence of oxygen respectively. The aerobic processes are used more for their efficiency and operational simplicity. There are several kinds of aerobic processes but the most popular are activated sludge systems, aerated lagoons, trickling filters, and rotating biological contactors (RBC). The most important parameters for the activated sludge systems are sludge retention time, oxygen and nutrient requirements, nutrient-to-microorganism ratio, mixed liquor suspended solids (MLSS) and sludge production rate.

Biological treatment of effluent can be performed by anaerobic and aerobic treatment techniques [9]. Conventional activated sludge system still the most widely used aerobic treatment technology for domestic and industrial wastewater treatment. Sequencing batch reactor (SBR) is modified activated sludge system in which all the process cycles are carried in one basin. The aerobic treatment using sequencing batch reactor (SBR) is successfully utilized worldwide for the treatment of several industrial wastewaters such as; textile, paper pulping and petroleum refineries in addition to domestic wastewater [10, 11]. Diya'uddeen et al. [12] treated the high-strength wastewater with (BOD5)/(COD) = 0.14, using both Fenton and SBR. The effect of Fenton treatment on improving biodegradability was studied.

The anaerobic treatment systems are selected as preferable options for treating municipal and industrial effluents based on many advantages, such as: economic construction costs, limited land requirements, low sludge production, simple operation and maintenance, energy generation in the form of biogas [13, 14]. The up-flow anaerobic sludge blanket (UASB) process is classified in the forefront of other high rate anaerobic digestion processes for its diverse applications for all types of wastes [15]. Anaerobic treatment of heavy oil refinery wastewater was studied anaerobically by Wang et al. revealed an average removal efficiency of COD and total oil of 70% and 72%, respectively, at an organic loading rate (OLR) of 3.44 kg COD/m^3d [16]. Rastegar et al. investigated the treatment of petroleum refinery wastewater using UASB, they found that (COD) removal was 81% at a constant organic loading rate (OLR) of 0.4 kg/m^3.d and an (HRT) of 48 h [17].

The main aim of this study is to investigate the efficiency of the treatment processes for the treatment of hazardous refractory wastewater from natural gas processing industry. Specific objectives are: studying different treatment processes including combination of biological and dark Fenton treatments, assessment of the treatment technology to achieve the optimum operating conditions.

Experimental

Natural gas processing wastewater representing hazardous refractory wastewater were selected for this study. The factory is located at Alexandria, Egypt and discharges its wastewater into the sea. Different treatment options were selected for the save disposal of these wastewater such as: biological treatment, dark Fenton and combination of both of them.

Fenton process

The use of Fenton reagent (Fe^{+2} / H_2O_2) at pH near 3 as catalytic oxidation was carried out. All experiments were performed at room temperature in a Jar Test apparatus with stirring of the wastewater. Needed dose of Ferrous sulfate catalyst was added into the reaction vessel to start the experiments. The pH was adjusted at the desired value and kept stable during the reaction. The required amount of H_2O_2 [30%] was fed under continuous stirring of the wastewater using the Jar test method. This method was preceded with rapid mixing of the wastewater at 200 rpm for the desired time. After the desired time is reached, the reaction was stopped by removing the residual hydrogen peroxide through the addition of 10 M NaOH to raise the pH to 12. Under alkaline conditions, the iron got precipitated and was then removed by sedimentation. After 30 min settling time, the supernatant was withdrawn and neutralized to around pH 8.0 for chemical analysis and subsequent biological treatment methods.

Up-flow Anaerobic Sludge Bed Reactor (UASB)

The experiments were conducted using an up-flow anaerobic sludge bed reactor (UASB). A schematic diagram of the UASB reactor is given in Fig. 1. The reactor is made of Perspex cylinder, to allow observation of the interior. The reactor is 10 cm internal diameter 40 cm height, a total net volume of 3.1 liter and 2.9 liters effective volume, excluding the gas collector. The experiments were performed at a temperature ranged from 25 to 35C° with a detention time ranged from 8h to 24h hours and organic loads ranged from 4 to 12 kg COD/ m^3/d. The reactor was fed by anaerobic sewage sludge collected from wastewater treatment plant located at Gabal El Asfar, Cairo.

Sequencing Batch Reactor (SBR)

Batch-type laboratory experiments were carried out using activated sludge process. Two liters Plexiglas laboratory columns were used. The columns were inoculated with activated sludge Mixed Liquor Suspended Solids (MLSS) from a municipal wastewater treatment plant. The MLSS in the columns were fed with raw wastewater on daily basis under continuous aeration. Every day, aeration was stopped for 30 minutes to remove the treated effluent from the active biomass and add new wastewater feed. This process was continued till considerable amount of adapted sludge was produced. To study the effect of Hydraulic Retention Time (HRT) on the treatment process, several experiments were conducted. A fixed amount of sludge (3-4 g/l) was transferred to a different column to which the pre-treated wastewater was added. Dissolved oxygen concentration was adjusted to maintain a minimum concentration of 2 mgO_2/L. Samples of MLSS were collected at 0, 2, 3, 6, 8, 10, 12, 14, 16 and 24 hours of reaction time. The samples were left for 60 minutes to settle and clear supernatants were subjected for analysis of COD and TSS.

Continuous flow activated sludge system (CFAS)

The treatment unit is made of prespex material and designed in rectangular cross section at the top end. The bottom is conveyed and sloped upwards whereas special support retains the unit in the upright position. Influent is passed through a 2.7 liter volume aeration compartment with internal baffle to maximize the aeration duration. Settling takes place in the settling compartment of 3.5 liter volume. Sludge collection port close to the bottom side is used for sludge sampling purpose. Effluent collector at the top right hand side directs the effluent to the exit as shown in Fig. 2.

Wastewater sampling and analysis

Composite samples were collected over the working hours. Physical–chemical, analyses were carried out according to the standard method for examination of water and wastewater [18].

Results and Discussion

Raw wastewater characteristics

Figure 3 indicates sources of the natural gas wastewater. The wastewater produced from glycol dehydration unit is contaminated with high organic and inorganic pollutants which are hardly biodegradable.

This study on industrial wastewater showed COD, BOD, TSS and total phenols values of 52665, 8350, 370 and 0.9 mg/L respectively (Table 1). The pH recorded between 4.5 and 5 suggests acidity of wastewater nature while high amount of nitrogen with an average value of 39.6 mg N/L and limited amount of phosphorus with an average value of 12.7 mg P/L were recorded. It should be noted that limits declared by local law for industrial wastewater discharge into the sea are 100, 60, 60, and 0.015 mg/L for COD, BOD, TSS and total phenols respectively. COD, BOD, TSS and phenols values in Table 1 are above the set limits therefore, treatment is necessary to lower the organic load. Since the BOD/COD ratio in wastewater is an agreed-on tool to express the biodegradability of the wastewater, a better biodegradability of wastewater is possible when BOD/COD ratio is greater than 0.3 [19, 20]. As a matter of fact, bacterial seed's metabolic activity is suppressed when the BOD/COD ratio is less than 0.3 since wastewater contains toxic and/or refractory compounds difficult to be biodegraded. The natural gas wastewater BOD/COD ratio is less than 0.16 as achieved by our study and illustrated in Table 1. This triggered the need for an effective wastewater pretreatment which was conducted using dark Fenton treatment process for enhancing biodegradability [21].

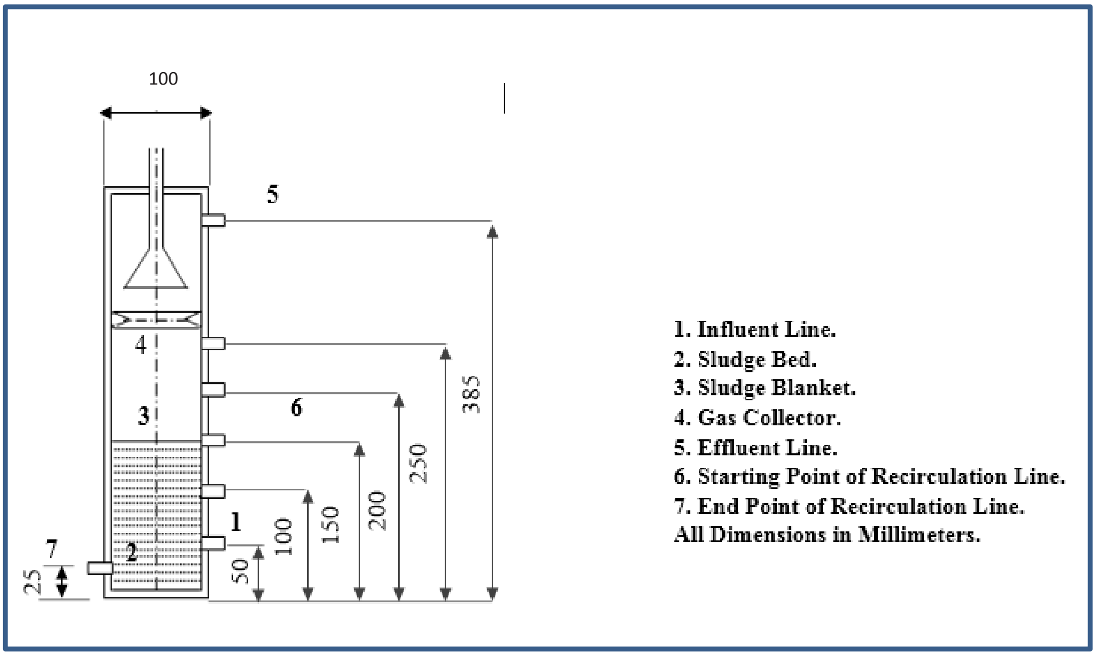

Fig. 1 Schematic diagram of the UASB Reactor

Combined dark Fenton with biological treatment

The study will concentrate on both aerobic and anaerobic bioreactors for the bioremediation of Fenton pretreated gas processing wastewater. The treated wastewater by Fenton process is denoted by high COD concentration. It was, therefore, diluted by domestic wastewater produced by the company to reduce the COD to one third. This action is essential to enable treatment biologically. The COD: N: P ratio of the Fenton pre-treated wastewater was not within the accepted range for biological treatment, it is usually recognized that the minimum nutrient requirement for anaerobic biomass growth represented by COD: N: P ratio is 600: 7: 1 [22] and for aerobic COD: N: P ratio is 300: 5: 1 [23], which was maintained by adding $Na(NH_4)_2PO_4$ and urea.

Anaerobic treatment using up-flow anaerobic sludge bed reactor (UASB)

Up flow anaerobic sludge blanket (UASB) reactor has been identified as a robust and potential among the different anaerobic reactors [24, 25]. Hydraulic loading rate and volumetric organic loading rate represent most important parameters of the design, operation and treatment performance of the UASB reactor. The UASB reactor can sustain a range of volumetric organic loading rate up to 7-11 kg COD/m3.day with average COD removal efficiency of 85% at 30 °C. Also up flow velocity is one of the important design parameters of the UASB reactor and should be within range of 0.5-0.7 m/h which is enough to provide good mixing conditions without sludge washout. The up flow velocity is determined after estimating the volume of the reactor by adjusting the ratio between the area of the reactor and its height. Keeping the up flow velocity in this range enhance TSS removal in the reactor [26-29].

The COD, BOD and TSS percentage removal as per the recorded data output in Table 1 are found to be higher than those achieved by El-Gohary et al. [30] who recorded 53%, 51% and 68 % removal efficiencies for COD, BOD and TSS respectively, when treating Fenton - oxidized olive wastewater using up-flow anaerobic sludge bed at 24h. Similarly, the results are higher than those achieved by Nasr et al. [31] who recorded average removal efficiencies of 61% for COD of flavor industry wastewater being treated using UASB.

Comparison is apparent between the achieved results and those obtained by Enitan et al. [32] who used the UASB for the treatment of Brewery wastewater [77% COD]. On the other hand, the results are less than those obtained by Wang et

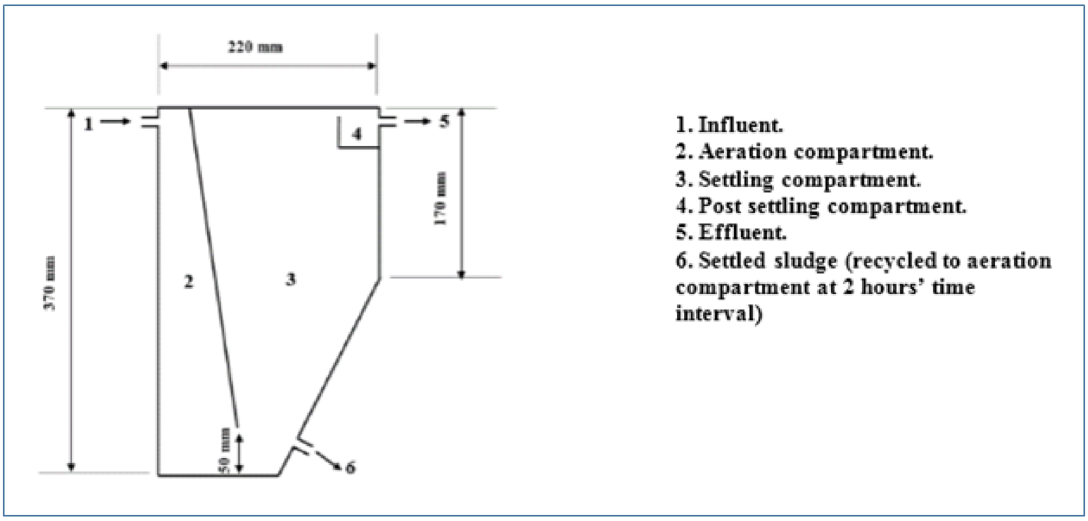

Fig. 2. Schematic drawing of the Continuous Flow Activated Sludge System (CFAS)

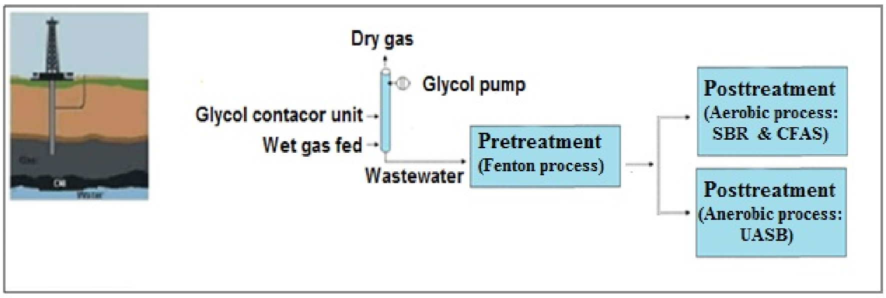

Fig. 3. The natural gas wastewater sources and the suggested treatment technology

al. [16] who obtained 86% COD removal using the UASB for the treatment of heavy oil refinery wastewater and less than those achieved by Hafez et al. [33] who used the UASB for treatment of flax retting Refinery wastewater and obtained 90 % COD removal. The average residual TKN and phosphorus values were in line with the permissible limits given by MD 44/2000 (Table 1).

The results showed that operating the UASB reactor at HRT of 18 h and 24 h gave close results at the two HRT, an indication of feasible selection of the 18 h HRT and the applied OLR is 5.5 kg COD/ m^3.d. for optimal performance and operation for economic advantage. The physical-chemical and biological characteristics of treated wastewater proved to be satisfactory and in agreement with the permissible standards given by the National law for the discharge of wastewater into the sewerage system as shown in Table 1 and Fig. 4.

Law 4/1994: National permissible limits for wastewater discharge into the sea

MD 44/2000**:** National permissible limits for wastewater discharge into sewerage system

*Average of five samples

Aerobic treatment using sequencing batch reactor (SBR)

Aerobic treatment using SBR was conducted to conclude the optimum detention time for CFAS which is the most applied technology in the field. Aerobic digestion of mixture of Fenton pretreated gas processing wastewater and sewage using a Plexiglas batch-type laboratory experiments were conducted using activated sludge process. Two liters Plexiglas laboratory columns were used. The Samples of MLSS were collected at 0, 2, 3, 6, 8, 10, 12, 14, 16 and 24 hours of reaction time. The samples were left for 60 minutes to settle and clear supernatants were subjected for analysis of COD and TSS. From the results it could be seen

TABLE 1. Characteristics of raw wastewater, Fenton treated and UASB effluents at 18h HRT

Parameter*	Unit	Fenton			UASB			Law 4/1994	MD 44/2000
		Raw	Treated	%R	Raw	Treated	%R		
pH	--	4.8±0.3	10±0.1	--	7.8±0.2	7.6±0.1	--	6-9	6-9.5
COD	mg/L	52665±27	11223±19	80	4083±24	850±36	79.2	100	1100
BOD_5	mg/L	8350±11	4084±12	57.2	1432±12	448±13	68.7	60	600
TSS	mg/L	370±52	62±8	87	165±42	51±11	69	60	800
TKN	mg/L	39.6±3.1	6.7±1.6	83.7	354±	2±19	45.2	10	100
Oil and Grease	mg/L	67±5	4±1	93.3	54±8	5±2	90.7	15	100
TP	mg/L	12.7±2.1	1.2±0.4	87.3	10±٣	6±٠,٣	40	2	25
Phenol	mg/L	0.9±0.08	ND	100	ND	ND	--	0.015	0.05

TABLE 2. Physicochemical characterization of Fenton pre-treated wastewater followed by SBR at 12h HRT

Parameter*	Unit	Fenton			SBR		
		Raw	Treated	%R	Raw	Treated	%R
pH	--	4.8±0.3	10±0.1	--	7.8±0.2	--	
COD	mg/L	52665±27	11223±19	80	4057±124	94±7.9	97.7
BOD_5	mg/L	8350±11	4084±12	57.2	1595±154	57.2±9	96.4
TSS	mg/L	370±52	62±8	87	175±31	17.9±4.3	89.8
TKN	mg/L	39.6±3.1	6.7±1.6	83.7	603.2±	7.8±٢,٢	87.0
Oil and Grease	mg/L	67±5	4±1	93.3	56±5	ND	100
TP	mg/L	12.7±2.1	1.2±0.4	87.3	131.5±	1.8±٠,٤	86
Phenol	mg/L	0.9±0.08	ND	100	ND	ND	--

that a gradual increase in the COD degradation rates from 29.8 % to 96.4 % occurs by raising the detention time from 1 to 12 hours and no significant increase in the COD degradation till 24 hours (Fig. 5). Therefore, the optimum detention time is 12 hours. The characteristics of the treated wastewater at the optimum HRT explained as COD, BOD, TSS and oil & grease were 94 mgO_2/L, 57.2 mgO_2/L, 17.9 mg/L and ND, respectively (Table 2). The treated wastewater quality produced in terms of physical-chemical and biological characteristics proved to be in compliance with the permissible standards given by the National law for discharging wastewater into the sea water.

*Average of five samples

Aerobic treatment using continuous flow activated sludge system (CFAS)

Aerobic digestion of mixture of Fenton pretreated gas processing wastewater and sewage using a Plexiglas continuous aerobic reactor CFAS inoculated with sludge from the batch reactor was evaluated. The operating parameters were pH 7.5-7.8, temperature 25-30 C°, 3 g/l MLSS and COD initial concentration of 4132±205 mg/L, F:M ratio of 0.49 kg BOD/kg MLVSS (Mixed Liquor Volatile Suspended Solids) and the optimum detention time determined from the batch reactor [12 h] and average organic load of 8.4 kg COD/m^3day. The CFAS reactor performance represented as organic matter removal in terms of residual COD ranged from 113 mgO_2/L to 84 mgO_2/L. The corresponding average values are 94±2.9 mgO_2/l (Fig. 6). The average percentage removal value was 97.7% (Table 3).

These results are higher than those recorded by Gilbert et al. [34] who achieved average removal efficiencies of 92 % for COD when treating wastewater using CFAS tank. These results are comparable to those achieved by Santo et al. [35] who used the CFAS for treatment of petroleum refinery wastewater. The residual BOD ranged

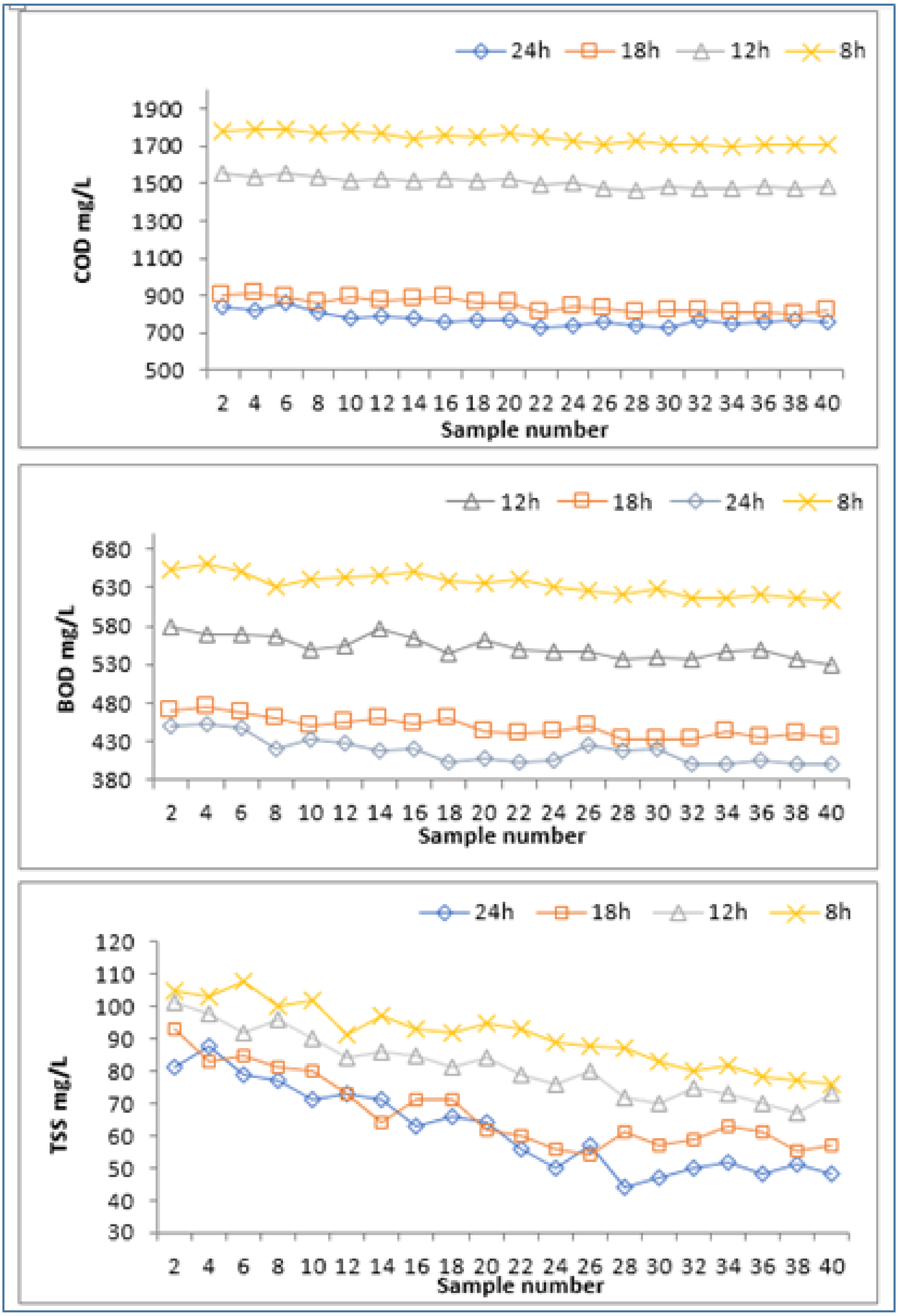

Fig. 4. Variation of COD, BOD, TSS concentration in UASB reactor effluent at HRT of 8, 12, 18 and 24 h

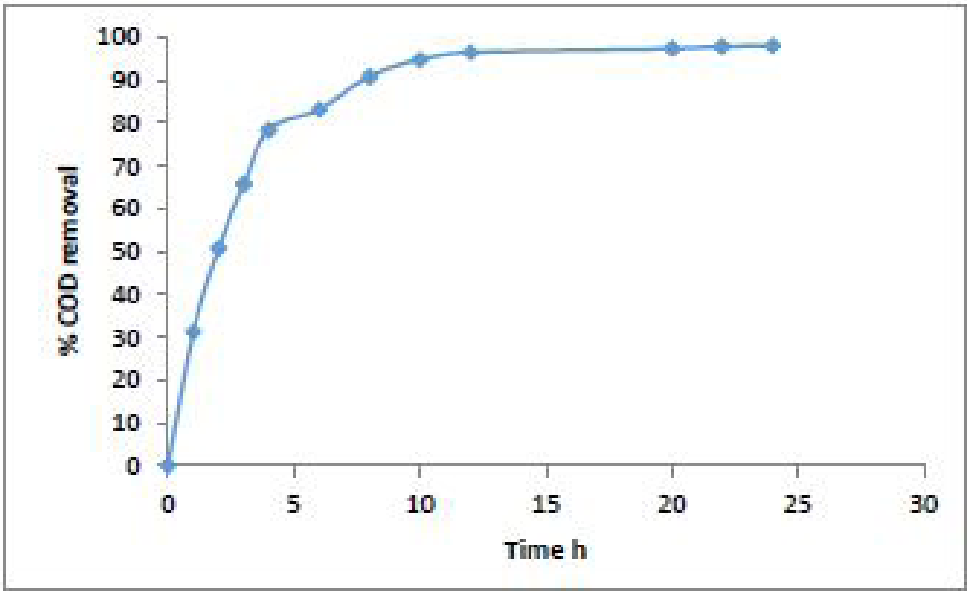

Fig. 5. Effect of aeration time on the biological activated sludge treatment for Fenton pre-treated natural gas wastewater

TABLE 3. Characteristic of raw wastewater, Fenton treatment and CFAS effluents at 12h HRT

Parameter*	Unit	Fenton			CFAS			Law 4/1994
		Raw	Treated	%R	Raw	Treated	%R	
pH	--	4.8±0.3	10±0.1	--	7.8±0.2	7.6±0.1	--	6-9
COD	mg/L	52665±27	11223±19	80	4132±205	94±2.9	97.7	100
BOD_5	mg/L	8350±11	4084±12	57.2	1521±117	٨±٤٥	97.0	60
TSS	mg/L	370±52	62±8	87	183±24	11±5	94.0	60
TKN	mg/L	39.6±3.1	6.7±1.6	83.7	704.5±	7.4±1.2	89.4	10
Oil and Grease	mg/L	67±5	4±1	93.3	62±7	ND	100	15
TP	mg/L	12.7±2.1	1.2±0.4	87.3	151.6±	1.6±0.4	89.3	2
Phenol	mg/L	0.9±0.08	ND	100	ND	ND	--	0.015

Law 4/1994: National permissible limits for wastewater discharge into sea

*Average of five samples

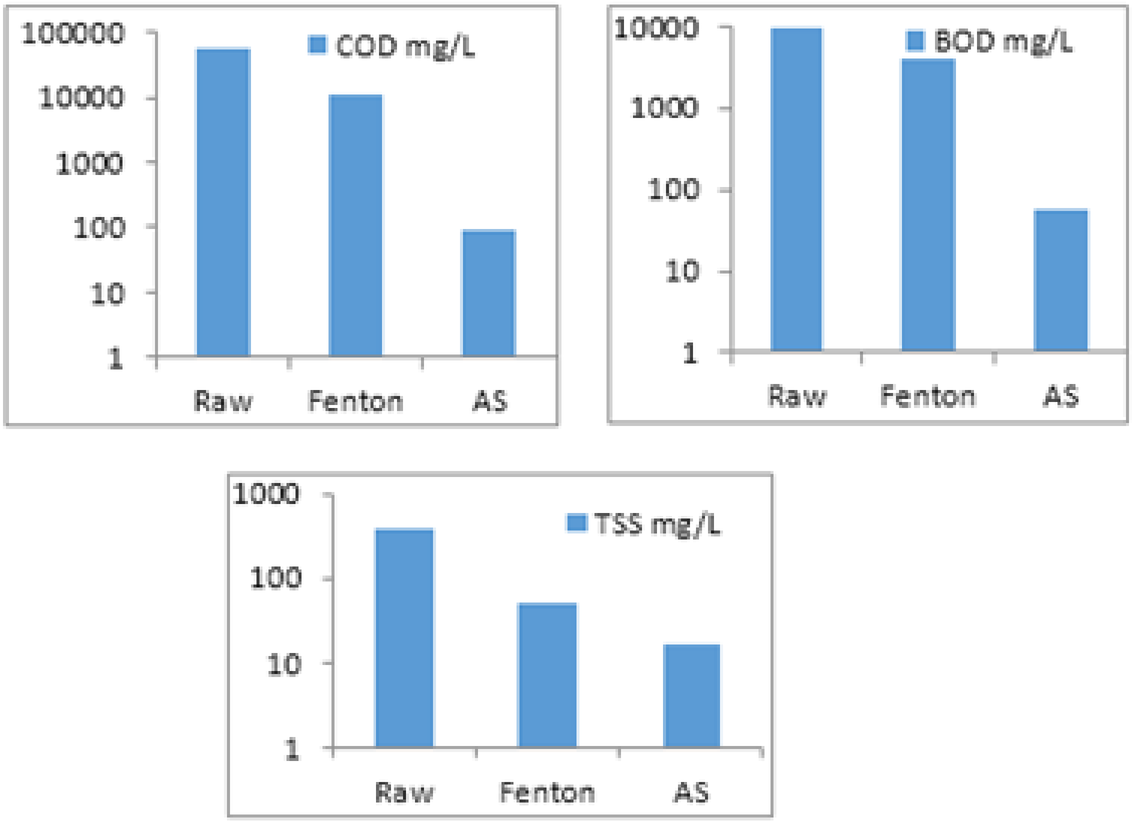

Fig. 6. COD, BOD, TSS concentration in raw, Fenton pretreated wastewater and CFAS effluent.

from 75 mgO_2/L to 45 mgO_2/L, the corresponding average values were 45±8 mg O_2/L (Fig. 6). The percentage removal value was 97%. Subject results are comparable to those obtained by Santo et al. [35] who used the CFAS for treatment of petroleum refinery wastewater. The residual TSS ranged between 26 mg/L and 13 mg/L, the corresponding average values are 11±5 mg/L. The average percentage removal value was 94%. These results are lower than those obtained by Santo et al. [35] who obtained average removal efficiency of 99 % for TSS when treating wastewater using AS tank. The treated wastewater quality produced represented as COD, BOD, TSS, TKN and TP were 94±2.9 mg/L, 45±8 mg/L, 11±5 mg/L, 7.4±1.2 mg/L and 1.6±0.4 mg/L. These results indicated that operating the CFAS reactor at HRT of 12 h gave satisfactory results (Table 3). Quality of the treated wastewater in terms of physical-chemical and biological characteristics proved to be in compliance with the permissible standards given by the Egyptian law for discharging wastewater into the sea water.

Conclusion

The wastewater produced by the natural gas processing industries is featured by high concentration of COD and low BOD values. The BOD/COD is less than 0.16 which indicates that this wastewater inhibits bacterial seeds metabolic activity. Biological treatment of a mixture of Fenton pretreated gas processing wastewater and sewage gave satisfactory results. The subsequent anaerobic treatment using UASB reactor at optimum HRT [18h] removed more than 79% of COD and qualifies the treated wastewater for discharge into the sewerage system. The subsequent aerobic treatment using the continuous-flow activated sludge at 12h HRT eliminated more than 97% of COD to qualify the treated effluent for discharging into the sea.

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