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Improvement of COD and color removal from UASB treated poultry manure wastewater using Fenton's oxidation

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Abstract

The applicability of Fenton's oxidation as an advanced treatment for chemical oxygen demand (COD) and color removal from anaerobically treated poultry manure wastewater was investigated. The raw poultry manure wastewater, having a pH of 7.30 (±0.2) and a total COD of 12,100 (±910) mg/L was first treated in a 15.7 L of pilot-scale up-flow anaerobic sludge blanket (UASB) reactor. The UASB reactor was operated for 72 days at mesophilic conditions ($32 \pm 2\,^{\circ}$C) in a temperature-controlled environment with three different hydraulic retention times (HRT) of 15.7, 12 and 8.0 days, and with organic loading rates (OLR) between 0.650 and 1.783 kg COD/(m^3 day). Under 8.0 days of HRT, the UASB process showed a remarkable performance on total COD removal with a treatment efficiency of 90.7% at the day of 63. The anaerobically treated poultry manure wastewater was further treated by Fenton's oxidation process using Fe^{2+} and H_2O_2 solutions. Batch tests were conducted on the UASB effluent samples to determine the optimum operating conditions including initial pH, effects of H_2O_2 and Fe^{2+} dosages, and the ratio of H_2O_2/Fe^{2+}. Preliminary tests conducted with the dosages of 100 mg Fe^{2+}/L and 200 mg H_2O_2/L showed that optimal initial pH was 3.0 for both COD and color removal from the UASB effluent. On the basis of preliminary test results, effects of increasing dosages of Fe^{2+} and H_2O_2 were investigated. Under the condition of 400 mg Fe^{2+}/L and 200 mg H_2O_2/L, removal efficiencies of residual COD and color were 88.7% and 80.9%, respectively. Under the subsequent condition of 100 mg Fe^{2+}/L and 1200 mg H_2O_2/L, 95% of residual COD and 95.7% of residual color were removed from the UASB effluent. Results of this experimental study obviously indicated that nearly 99.3% of COD of raw poultry manure wastewater could be effectively removed by a UASB process followed by Fenton's oxidation technology used as a post-treatment unit.

Keywords: Poultry manure wastewater; Fenton's oxidation; pH; COD removal

1. Introduction

Poultry wastes are potential sources of many major environmental problems. The increasing trend of poultry production in both developed and developing countries results in large quantities of poultry wastes. The solid waste annually produced by poultry farm birds was estimated at millions of tonnes [1]. However, improperly managed poultry manure can result in severe consequences to environment such as odor problem, attraction of rodents, insects and other pests, release of animal pathogens, groundwater contamination, surface water runoff, deterioration of biological structure of the earth and catastrophic spills.

Anaerobic digestion is one of the beneficial and advantageous processes in manure treatment. Bacteria that function without oxygen degrade organic matter inherent in poultry waste. These microorganisms are both temperature and oxygen sensitive and thus design criteria for systems utilizing anaerobic processes will vary regionally.

Advances in the understanding of anaerobic system functions and reactor design, has led to evolution of a new generation of high-rate anaerobic processes [2]. These process configurations include anaerobic contact process, anaerobic filters (AFs), anaerobic expanded/fluidized bed, reactors and up-flow anaerobic sludge blanket reactor (UASB), etc. It is reported that AFs and UASB reactors have a wide-scale applicability for treating various types of wastewaters. These types of reactor configurations are frequently used for medium to high-strength wastewater having a wide COD range of 2000–20,000 mg/L [3].

In the anaerobic digestion of poultry wastes, a number of different reactor configurations have been reported [4]. The pre-treatment of the liquid fraction of hen manure in terms of its treatment efficiency on total COD reduction and methane

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production was investigated using two 2.6 L UASB reactors [5]. The feasibility of applying the UASB reactor for treatment of poultry wastewaters was examined [1]. The study was performed in a continuous flow UASB pilot-scale reactor of 3.5 L volume at 26–34 °C for 95 days to assess the treatability of poultry wastewater. An experimental study was conducted to investigate anaerobic treatability and biogas generation potential of brolier and cattle manure in seven sets of anaerobic batch reactors [6]. Finally, the anaerobic digestion of four types of agricultural wastes including poultry droppings, cow dung, corn stalk and mixed substrate was investigated [7]. In the study, a batch pilot-scale reactor having a diameter of 58 cm, a length of 106 cm and a total volume of 0.28 m^3 was operated for 40 days.

Interest in using anaerobic digestion for poultry manure management is rapidly growing as farmers and governments are faced with mounting economic and environmental concerns [8]. However, with environmental regulations becoming more stringent, regulatory compliance has become a matter of increasing concern to the poultry industries, and there is a need to install more effective subsequent waste treatment facilities. It is reported that Fenton's oxidation is an appropriate further alternative for the advanced treatment of wastewater effluents having non-biodegradable organic pollutant contents and dark color after an undergoing biological treatment. This technology is capable to remove almost all parts of the organics which consist of both soluble initial and microbial inert fractions of COD formed during the biological treatment [9].

Fenton's oxidation has been used to treat a variety of industrial wastes containing toxic organic compounds such as phenols, formaldehtde and dyestuffs, and may be applied to wastewaters, sludges, or contaminated soils, with the effects being organic pollutant destruction, toxicity reduction, biodegradability improvement, biological oxygen demand (BOD)/COD removal, and odor and color removal [10]. Because Fenton's oxidation process yields satisfactory final effluents, in recent years this technology has been applied to many environmental problems such as further treatment of organics in anaerobically treated leachate by Fenton coagulation [11], advanced treatment of opium alkaloid industry effluents using Fenton's oxidation [9], treatment of methyl *tertiary*-butyl ether (MTBE) contaminated wastewaters using Fenton's reagent [10], oxidation of aromatic groundwater contaminants [12], treatment of water-based paint wastewater with Fenton process [13], advanced oxidation of olive-oil mills wastewater [14], and removal of atrazine by step-wise Fenton's processes [15].

In the first step of this study, organics in raw poultry manure wastewater were degraded using a pilot-scale UASB reactor. Because the UASB effluent had a colloidal nature and higher levels than the acceptable local sewer system discharge standards for COD and color, Fenton's oxidation process was conducted to further remove organic residues in the UASB effluent. The overall objective of this study was to determine optimum conditions for COD and color removal from the anaerobically treated poultry manure wastewater effluent using Fenton's oxidation process. In addition, it was also aimed to demonstrate the applicability of a two-stage system for the effective treatment of poultry manure wastewater using an UASB process followed by Fenton's oxidation technology.

2. Materials and methods

2.1. Poultry manure source and feed preparation

Fresh poultry manure was collected from a moderate size commercial poultry farm (Hakan's Poultry Farm) located at Buyukkilicli Village in Silivri, Istanbul and stored in the refrigerator at 4 °C to minimize substrate decomposition before the experiment. Characteristics of fresh poultry manure used as feedstock during the experimental period are given in Table 1.

The feed for UASB was prepared by diluting fresh poultry manure with tap water and then mixing it with a vertical stirrer (Makita HP1500) for 5–10 min to obtain a uniform environment in feeding material. The diluted manure was then filtered through a screen of 1.18 mm mesh size (Endecotts Ltd.) to reduce potential clogging of tubing and operational problems may be caused by broken egg shells, hair or feathers and inert bedding materials such as sand, sawdust and wood shavings existed in the fresh manure. Prior to feeding, stored feed was warmed to the reactor operating temperature using Chiltern Hotplate Magnetic Stirrer, HS31.

2.2. UASB setup and operation

Raw poultry manure wastewater was pretreated in a pilot-scale UASB reactor (diameter 12 cm, total height 160 cm, total tank capacity 19.85 L and made from 5.0-mm transparent plexiglas) having a working volume of 15.7 L for digestion. The reactor was provided with conical bottom of 20 cm length and a feed inlet pipe of 1.5 cm diameter to avoid chocking during operation. An outlet weir was provided at the top (1.51 m), which is connected to an outlet gutter and outlet pipe to the effluent collection tank. The reactor had ports for sampling, feeding, effluent and gas collecting. Gas was collected from the headspace on the top of the reactor and gas production was measured by the liquid displacement method. The gas collecting and measuring systems consisted of a gas–solid–liquid separator (made from an inverted plastic funnel of 11 cm diameter), a gas collecting pipe, a water trap, a graduated gas measuring tube and a water tank for keeping of the gas measuring tube.

The reactor system was operated for 72 days at mesophilic conditions (32 ± 2 °C) in a temperature-controlled environment maintained by two adjustable radiators with thermostat (Demirdokum DEYR 7B CM) after the start-up period.

Bacteria in the reactor break down volatile solids in the manure to produce methane. This length of time for this pro-

Table 1
Characteristics of fresh poultry manure used as feedstock

Constituent	Mean ± S.D.
Water content (%)	77.5 ± 0.59
Volatile solids (% of total solids)	64.5 ± 1.13
Density (kg/m^3)	1102.16 ± 114.5

cess to take place, the hydraulic retention time (HRT), takes from 3 to 20 days, depending upon the size of the digester, its type, and its operating temperature [16]. Therefore, HRTs were selected, with the option to increase or decrease the HRT by adjusting feed flow rates into the reactor. On the basis of the cross-sectional area of the reactor (95.03 cm^2) and applied feed flow rates from 1 to 2 L/day, hydraulic loading rates (L_H) were controlled between 0.105 and 0.21 $m^3/(m^2\ day)$.

The UASB system was conducted with three different HRTs of 15.7, 12.0 and 8.0 days, and with organic loading rates (OLR) between 0.650 and 1.783 kg COD/(m^3 day). The pH of feed to the reactor ranged from 6.96 to 7.82, with an average value of 7.3 (±0.2). Stability of the treatment process and components of wastewater samples were monitored in Environmental Engineering Laboratory at Yildiz Technical University in Istanbul, Turkey.

The UASB system was operated in a daily-continuous mode feeding by pumping of fresh feed into the reactor and collecting effluent samples daily. In feeding, different target HRTs were achieved using a peristaltic pump (FPU5-MT-220, OmegaFlex®). Feed wastewater samples were prepared daily and pumped to the reactor from the feeding tank with a stable up-flow velocity of about 0.70 m/h by operating the peristaltic pump in a feeding mode of 50 rpm (133 mL/min of flow rate) for 6 mm of tube size.

The UASB effluent was collected for the subsequent treatment of Fenton's oxidation. A detailed schematic diagram of the experimental setup is illustrated in Fig. 1.

2.2.1. *Seed sludge*

Seeding is strongly recommended in order to increase the efficiency of the digestion process. However, seeding with mature granules requires less time for start-up, compared to reactors started with flocculent seed (biomass from a conventional anaerobic digester) [17]. Because granular biomass has higher settling velocity and higher specific activity than flocculent biomass, the reactor was seeded with 4.5 L of actively digesting granular sludge (28.6% of the working volume) from an ongoing mesophilic UASB reactor of Pasabahce Distillery Inc. (Istanbul, Turkey). Then, the system was filled to its respective volume of 15.7 L with diluted poultry manure wastewater (79.1% of the total tank capacity). Prior to seed, the total solids (TS) content of the granular sludge was about 90.8 g TS/L. The volatile solids (VS) content of the sludge was found to be 82.3% of TS. During the study period, the 15.7 L reactor contained about 336.3 g of VS.

The UASB reactor had six sludge sampling ports, localized at 0.35, 0.50, 0.65, 0.80, 0.90 and 1.10 m from the bottom of the reactor. This arrangement was done to determine the sludge bed profile in the UASB reactor. The reactor contents were maintained at the respective temperatures (32 ± 2 °C) for a week to allow temperature equilibration and utilization of substrate contained in the seed.

Initial morphology of some sample granules is shown in Fig. 2. Images of granules were taken with a digital camera (Sony Cyber-shot DSC-N1) combined with a stereomicroscope (Prior, James Swift) prior to seed.

2.2.2. *Basal medium*

A nutrient solution/basal media containing all necessary micro- and macro-nutrients for an optimum anaerobic microbial growth was prepared with the following components, and added 1 mL/L of the daily fed subtrate [18]: 5 g/L $MgSO_4{\cdot}7H_2O$, 6 g/L $FeCl_2{\cdot}6H_2O$, 10 g/L $CoCl_2{\cdot}6H_2O$, 1 mg/L H_3BO_3, 1 mg/L $ZnSO_4{\cdot}7H_2O$, 1 mg/L $CuSO_4{\cdot}5H_2O$, 100 mg/L $MnCl_2{\cdot}6H_2O$,

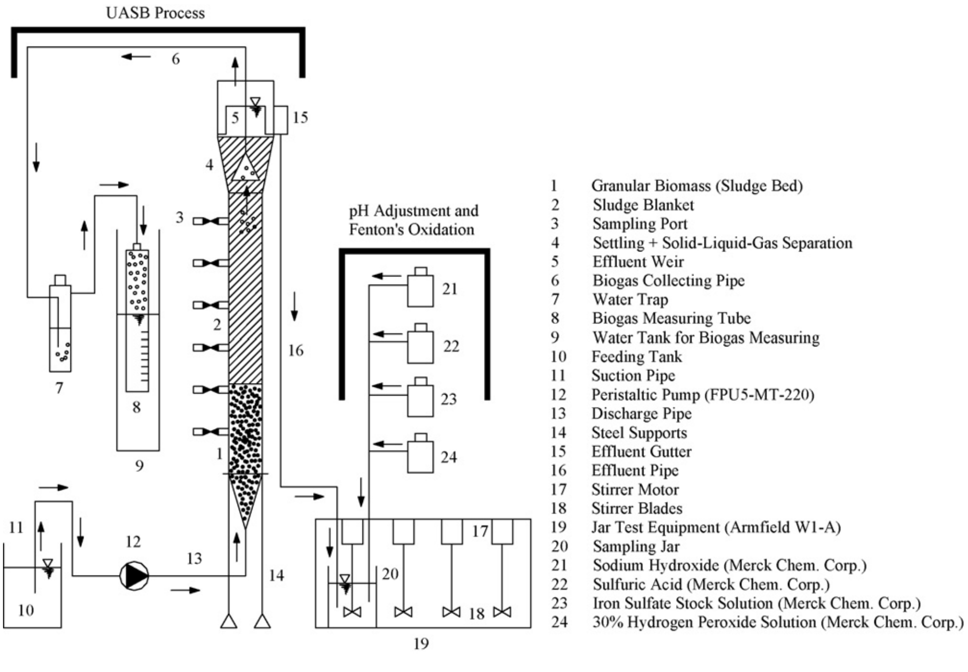

Fig. 1. Detailed schematic diagram of the experimental setup.

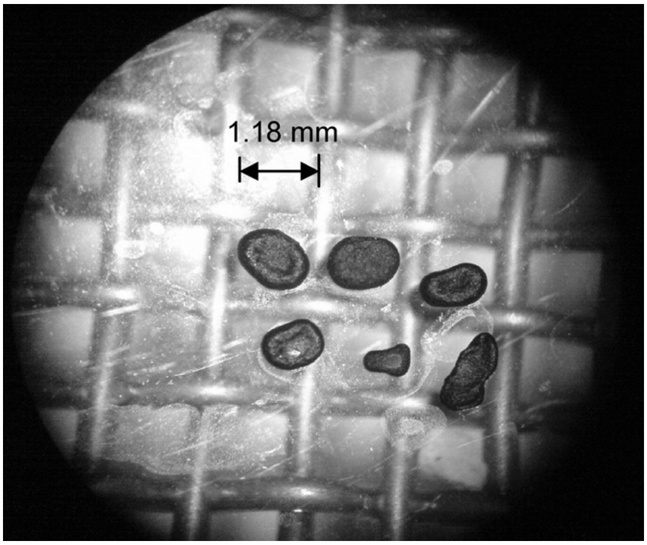

Fig. 2. Initial morphology of some sample granules on a screen of 1.18 mm mesh size.

1 mg/L $(NH_4)_6Mo_{24}\cdot4H_2O$, 585 mg/L $Al_2(SO_4)_3\cdot18H_2O$, and 1 g/L $Na_2SiO_3\cdot9H_2O$.

2.3. Fenton's oxidation

A stock solution of 10 g/L of Fe^{2+} was prepared by dissolving $FeSO_4\cdot7H_2O$ (Merck Chemical Corp.) in 0.2N H_2SO_4. In addition to iron sulfate reagent, 30% H_2O_2 solution (Merck Chemical Corp.) having a density of 1.11 kg/L was used in the oxidation process. In each oxidation test, 500 mL of anaerobically treated poultry wastewater sample was collected from the UASB effluent. In the first step of Fenton's oxidation process, the pH of the UASB effluent wastewater was adjusted to desired value by the addition of 1N H_2SO_4 and 1N NaOH. During the whole oxidation process, the pH of samples were also set at desired value by adding these reagents (1N H_2SO_4 and 1N NaOH) gradually in addition to pre-adjustment of the pH. The $FeSO_4\cdot7H_2O$ and H_2O_2 solutions were then added to the effluent sample and conducted for 5 min of rapid mixing at 100 rpm using a Jar Test Equipment (Armfield, W1-A). The effluent sample was then gently stirred at 10 rpm for 25 min. After the flocculation process, the sample transferred to a graduated settling column for 30 min of settling. About 100 mL of supernatant sample was then collected for COD and color analysis after the settling process. In order to prevent interferences in analytical measurements, the pH of collected supernatant sample was increased up to about 11 by adding 6N NaOH gradually to precipitate Fe^{2+} ions in the form of $Fe(OH)_3$. Finally, MnO_2 reagent was then added to remove the residual H_2O_2 from the collected supernatant [19–21].

2.4. Analytical methods

In the daily operation of UASB system, influent and effluent pH values were measured by a pH meter (Jenway 3040 Ion Analyser) and a pH probe (HI 1230, Hanna Instruments). Color of wastewater samples were measured with a Merck photometer (model: SQ 118) and determined as Hazen color unit according to method number 138. Soluble COD (SCOD) was determined by filtering sample through 0.45 μm filter paper. All other experimental analyses were performed according to standard methods [22]. These parameters were determined by the procedures described in method numbers of 5220 B (open reflux method for COD), 2540 B (total solids dried at 103–105 °C), 2540 D (total suspended solids dried at 103–105 °C), 2540 E (fixed and volatile solids ignited at 550 °C), 5210 B (5-day BOD test), 2320 B (titration method for alkalinity), 4500 NH_3-N E (titrimetric method for ammonia), 4500 N_{org} B (macro-Kjeldahl method for total Kjeldahl nitrogen), and 4500 P (persulfate digestion method for total phosphorus). Samples were ignited at 550 °C using an ashing furnace (Lenton) for volatile solids (VS) and volatile suspended solids (VSS) analyses. Absorbance values were recorded at 690 nm using a spectrophotometer (Pharmacia Biotech LKB Novaspec II) for total phosphorus (TP) analysis. Biogas composition was determined using a portable multi-channel environmental gas analyser (Gas Data LMSxi G3 Landfill Gas Analyser).

2.5. Statistical analysis

All standard deviations reported here were calculated using the statistical functions in Microsoft® Excel 2000 used as an ODBC (open database connectivity) data source. Data were entered in a Microsoft® Excel 2000 spreadsheet and means, ranges, number of data points, and standard deviations were calculated. In addition, polynominal regressions models were performed in Excel and the corresponding regression coefficients were determined for data sets of sludge bed profiles: SCOD, pH, and VS/TS ratio. Experimental results were reported as the mean value of each parameter ± standard deviation using Eq. (1):

$$E_R = \bar{x} \pm \sigma = \frac{1}{n}\sum_{i=1}^{n} x_i \pm \sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(x_i - \frac{1}{n}\sum_{i=1}^{n} x_i\right)^2} \tag{1}$$

where E_R is the experimental result, $\bar{x}$ the mean value, n the number of measurements, and x_i is the ith data point.

3. Results and discussion

3.1. UASB process

The UASB reactor was operated for 72 days after the acclimation period of granular biomass used as seed sludge. The effluent of the UASB process was collected for the subsequent treatment of Fenton's oxidation. Characteristics of the prepared poultry manure wastewater and the UASB effluent are given in Table 2.

Under 8.0 days of HRT and an OLR of 0.76 kg COD/(m^3 day), the UASB process demonstrated an optimal performance on total COD removal with a treatment efficiency of 90.7% at the day of 63. During collection of the UASB effluent

Table 2
Characteristics of prepared poultry manure wastewater and the UASB effluent

Constituent	UASB influent (mean ± S.D.)	UASB effluent (mean ± S.D.)
Total chemical oxygen demand, TCOD (mg/L)	12,100 ± 910	1750 ± 200
Biological oxygen demand, BOD_5 (mg/L)	5,900 ± 390	420 ± 50
Soluble chemical oxygen demand, SCOD (mg/L)	2,090 ± 170	1120 ± 90
Total solids, TS (mg/L)	8,280 ± 700	1980 ± 200
Volatile solids, VS (mg/L)	5,370 ± 450	1380 ± 130
Total suspended solids, TSS (mg/L)	5,020 ± 380	1130 ± 70
Volatile suspended solids, VSS (mg/L)	4,020 ± 340	970 ± 130
Total Kjeldahl nitrogen, TKN (mg/L)	1,830 ± 130	1380 ± 120
Ammonia nitrogen, NH_3-N (mg/L)	990 ± 70	1180 ± 70
Total phosphorus, TP (mg/L)	450 ± 30	380 ± 20
pH	7.30 ± 0.2	8.28 ± 0.3
Alkalinity (mg $CaCO_3$/L)	3,210 ± 200	2690 ± 200

for the subsequent treatment of Fenton's oxidation, the UASB reactor on average removed 85.3 (±1.9)% of COD.

The observed SCOD, BOD_5, TS, TSS, VS, and VSS removal efficiencies averaged 46.3 (±6.5)%, 93 (±1.2)%, 75.8 (±3.6)%, 77.4 (±2.5)%, 74 (±3.7)%, 75.5 (±4.3)%, respectively. No striking reductions in both TKN and TP were observed. The TKN through the UASB was reduced by 23 (±10.1)% on average. TP removal was about 13.4 (±9.1)%. The removals in TP and also the loss of N in the UASB should be due to both new biomass production, as well as settling in the reactor [23]. Relatively low treatment efficiencies may be expected for TKN and TP, since anaerobic reactors are known to reduce negligible amounts of nutrients [8].

The NH_3-N concentration on average was increased by about 21 (±11.8)% after the UASB treatment because of the conversion of organic N into NH_3-N. This also resulted in an increase of pH, as given in Table 2. The increase in NH_3-N can be attributed to the anaerobic bioconversion of proteins contained in manure into amino acids and then to ammonia [8]. The alkalinity was reduced by 15.8 (±8.8)% on average. This reduction can be attributed to the buffering of volatile fatty acids during the digestion process.

Biogas production rates (Q_g) ranged from 4.2 to 13 L/day and averaged 6.87 (±2.46) L/day, depending on various operating conditions. High volumetric COD removal rates (R_V) ranging from 0.546 to 1.608 kg $COD_{removed}/(m^3\ day)$ were achieved, with an average value of 0.875 (±0.312) kg COD $_{removed}/(m^3\ day)$ (Table 3).

At steady state the daily mass of influent COD is equal to the daily mass of COD leaving the system as methane in the excess sludge produced, in the effluent and daily amount of COD oxidised [24]:

$$MS_i = MS_e + MS_x + MS_d + MS_o \quad (2)$$

where MS_i is the daily mass of influent COD, MS_e the daily mass of effluent COD, MS_x the daily mass of COD in discharged sludge, MS_d the daily mass of digested sludge, and MS_o is the daily mass of oxidised sludge. Normally, COD measurements for a reactor are calculated for the influent wastewater, the effluent wastewater and the gas production. In Eq. (2), MS_e and MS_x are contained in the effluent wastewater (COD_{out}) while the daily mass of oxidised sludge (MS_o) is incorporated into the biomass. For anaerobic bacteria, the growth rate is very slow that this amount is negligible. The daily mass of digested (MS_d) is released as methane $COD_{methane}$. The COD mass balance then consists of

$$COD_{in} \rightarrow COD_{out} + COD_{methane} \quad (3)$$

On the basis of the experimental data, COD mass balances were calculated for the influent, effluent and biogas fractions. COD_{in} and COD_{out} were determined for the data sets of influent and effluent COD concentrations, and daily feed flow rates, respectively. The mole of methane in biogas was calculated using the well-known ideal gas equation, and then theoretical COD of methane was determined for its oxygen equivalent. A plot of mass COD balance for the reactor is depicted in Fig. 3.

The COD mass balance revealed that 87.4 (±1.8)% of the COD taken in was accounted for. This indicates that the stability of the reactor on average was 87.4%. The rest that was not accounted for is the COD fraction that is incorporated into the

Table 3
Biogas production rates (Q_g) of UASB reactor at different operational periods of the study

	15.7[a] (0.105[b])		12[a] (0.138[b])		8.0[a] (0.210[b])	
	Range[c]	Average ± S.D.[c]	Range[c]	Average ± S.D.[c]	Range[c]	Average ± S.D.[c]
OLR (kg COD/(m^3 day))	0.65–0.853	0.73 ± 0.046	0.92–1.21	1.04 ± 0.065	1.44–1.783	1.6 ± 0.09
R_V (kg $COD_{removed}$/(m^3 day))	0.55–0.710	0.61 ± 0.04	0.79–1.04	0.89 ± 0.06	1.24–1.61	1.41 ± 0.09
Q_g (L/day)	4.2–5.6	4.83 ± 0.35	5.85–7.95	6.89 ± 0.50	9.50–13.0	11.1 ± 0.76

[a] HRT (day).
[b] L_H (m^3/(m^2 day)).
[c] Values.

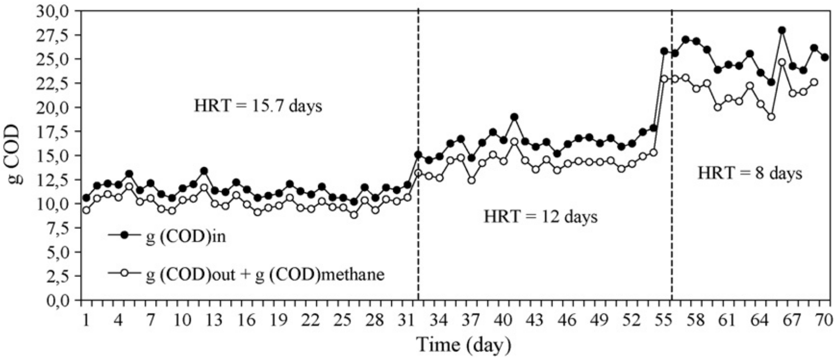

Fig. 3. Plot of the COD mass balances for the UASB reactor.

biomass as this is assumed to be negligible in the COD mass balance equation. The result of COD mass balance also showed that 72.7 (±2.1)% of influent organic matter on average was transformed to biogas with a methane content over 70%.

Good performance of the UASB process may be explained by the contribution of the good quality of the seed sludge. The initial average diameter of the granules was found to be about 1.18 mm. The density of the granular sludge was measured to be 1075 kg/m^3. The mean settling velocity was determined using the well-known force balance equation as follows:

$$u_\mathrm{t}=\sqrt{\frac{4gd_\mathrm{p}(\rho_\mathrm{p}-\rho_\mathrm{w})}{3\xi\rho_\mathrm{w}}} \tag{4}$$

where u_t is the mean settling velocity (m/s), d_p the average diameter of the granules (m), g the acceleration of gravity (9.81 m/s^2), ρ_p the density of the granular sludge (1075 kg/m^3), ρ_w the density of water (1000 kg/m^3), and ξ is the drag coefficient. In determination of u_t, the drag coefficient (ξ) being a function of Reynolds number at terminal settling velocity was obtained using Perry's and Green's equation from the following equation:

$$\xi = 18.5\,Re_\mathrm{t}^{-0.6} \tag{5}$$

where Re_t is the Reynolds number at terminal settling velocity. The value of Re_t was calculated from the following equation:

$$Re_\mathrm{t}=\frac{\rho_\mathrm{w}d_\mathrm{p}u_\mathrm{t}}{\mu_\mathrm{w}} \tag{6}$$

where μ_w is the viscosity of water at room temperature (10^{-3} kg m/s or Pa s). Therefore, u_t was determined to be 0.0206 m/s (74.16 m/h) from the following steps:

$$u_\mathrm{t}=\sqrt{\frac{4gd_\mathrm{p}(\rho_\mathrm{p}-\rho_\mathrm{w})}{3\left(18.5[(\rho_\mathrm{w}d_\mathrm{p}u_\mathrm{t})/\mu_\mathrm{w}]^{-0.6}\right)\rho_\mathrm{w}}} \tag{7}$$

$$u_\mathrm{t}=\sqrt{\frac{4(9.81)(1.18\times10^{-3})(1075-1000)}{3\left(18.5[((1000)(1.18\times10^{-3})u_\mathrm{t})/10^{-3}]^{-0.6}\right)(1000)}} \tag{8}$$

$$u_\mathrm{t}=\sqrt{\frac{3.47274}{55500(1180u_\mathrm{t})^{-0.6}}} \tag{9}$$

Following the determination of u_t, the value of Re_t was determined and verified as follows:

$$Re_\mathrm{t}=\frac{\rho_\mathrm{w}d_\mathrm{p}u_\mathrm{t}}{\mu_\mathrm{w}}=\frac{(1000)(1.18\times10^{-3})(0.0206)}{(10^{-3})}\cong 24.31 \tag{10}$$

Results were found to be in accordance with the range of the granule diameters considered in a simulation analysis of the settling velocity model [25].

The UASB influent, having a BOD$_5$/TCOD ratio of about 0.50, was readily biodegradable. However, the UASB effluent, having a BOD$_5$/TCOD ratio of about 0.24, showed a low biodegradability index, which was recalcitrant to a possible further biodegradation. Because subsequent conventional biological wastewater techniques may fail to meet the discharging standards, the anaerobically treated poultry manure wastewater was further treated by Fenton's oxidation process using Fe^{2+} and H_2O_2 solutions.

3.2. Sludge bed profiles

Fig. 4 shows the sludge bed profiles taken along the length for SCOD, pH and VS/TS ratio, respectively. Fig. 4(a) illustrates that the soluble COD shows a decrease in the lower part of the sludge bed from the influent concentration of 2200 mg SCOD/L to about 1344 mg SCOD/L at Port 1, and thereafter decreases slowly about to 1030 mg SCOD/L throughout the rest of the sludge blanket. Similar pH and COD profiles were observed in UASB treatment of grain distillation wastewaters containing high suspended solids [26], and in the validation of an integrated mathematical model with results from an experimental study on treatment of high strength cheese whey in a UASB reactor [27], respectively. The SCOD profile revealed that the digestion process was nearly completed in lower parts of the reactor. The SCOD decreased only slowly over the sludge bed, and the removal rate in upper parts was not so significant as compared in the lower parts.

Fig. 4(b) depicts that the pH profile exhibits a gradual increase from the lower part of the sludge bed to the effluent. The increase

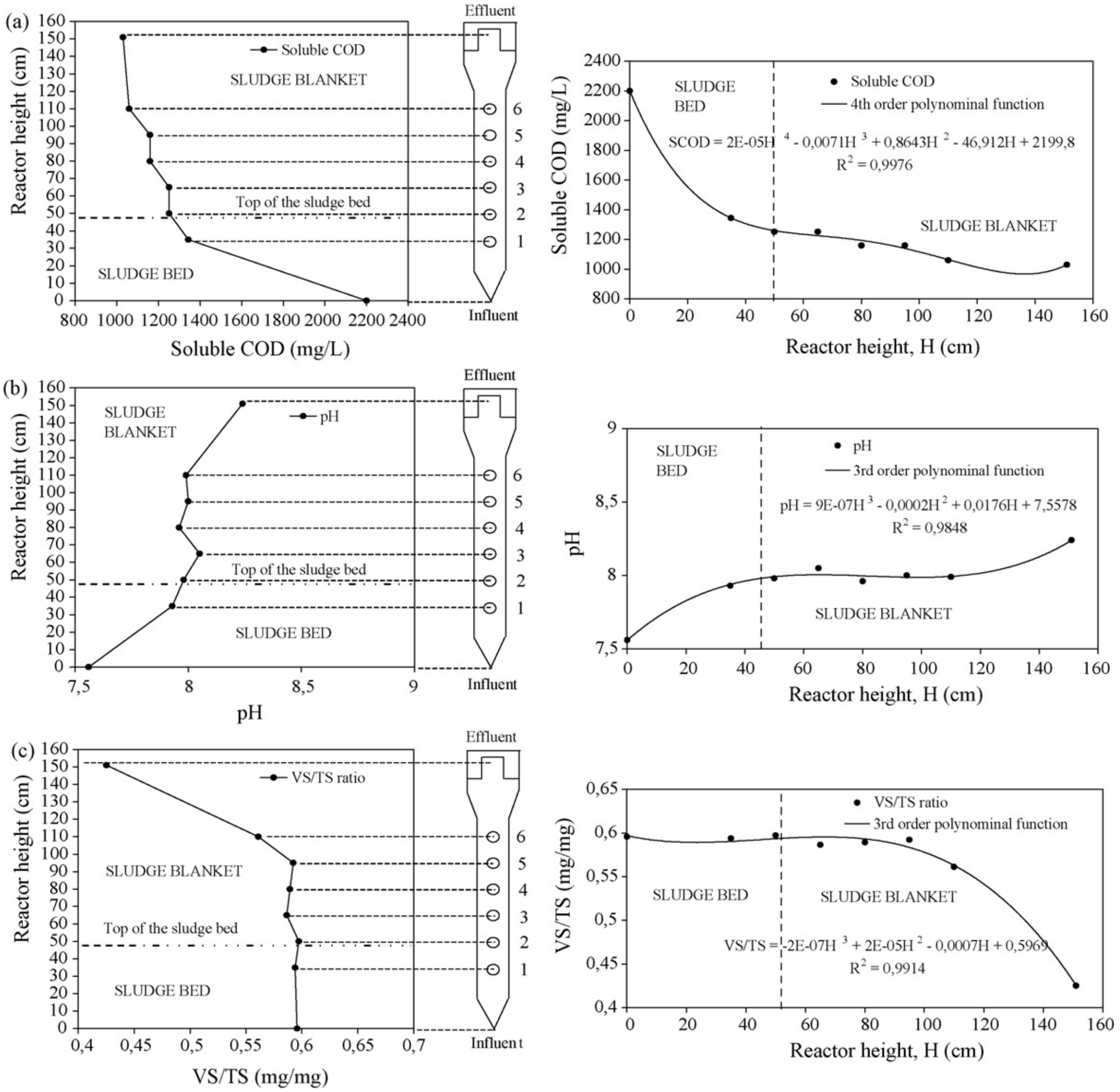

Fig. 4. (a–c) Profiles of pH, SCOD and VS/TS ratio with corresponding regression functions along the reactor height.

in the pH can be attributed to the anaerobic bio-convertion of proteins contained in manure into amino acids and then to ammonia as mentioned before.

Fig. 4(c) shows that VS/TS ratio over the sludge bed. The nearly constant ratio indicated that the sludge was equally stabilised over the bed. The relationship between VS and TS in the sampling zone was 58.7 (±1.3)% on the average. The relatively high VS/TS ratio indicated that low amount of inert solids were accumulated in the sludge bed. The low amount of inert solids in the sludge bed can be attributed to the removal of broken egg shells, hair or feathers and inert bedding materials such as sand, sawdust and wood shavings by filtering of the raw wastewater through a screen before feeding into the reactor. Similar VS/TS profiles were obtained in the experimental studies on anaerobic pre-treatment of sewage in an integrated UASB-digester system [28], and domestic sewage treatment in a full-scale UASB reactor [29].

As shown in Fig. 4, sludge bed profiles were depicted with corresponding regression functions along the reactor height. A fourth-order polynominal regression model was fitted to the SCOD data, with a correlation coefficient of 0.9976. Moreover, third-order polynominal regression models were fitted to data sets of pH and VS/TS ratio, with correlation coefficients of 0.9848 and 0.9914, respectively. By using highly correlated regression models, values at different heights of the reactor can be satisfactorily estimated for the experimental data.

3.3. Fenton's oxidation

3.3.1. Effect of the initial pH

A series of preliminary batch experiments using different Fe^{2+} and H_2O_2 concentrations was conducted at a pH ranging from 2.0 to 7.0 to determine the optimal condition for the initial pH. Findings of preliminary batch experiments showed that pH 3.0 was the optimal initial pH at the dosages of 100 mg Fe^{2+}/L and 200 mg H_2O_2/L for both COD and color removal in Fenton's oxidation of the UASB effluent. At pH 3.0, removal efficiencies

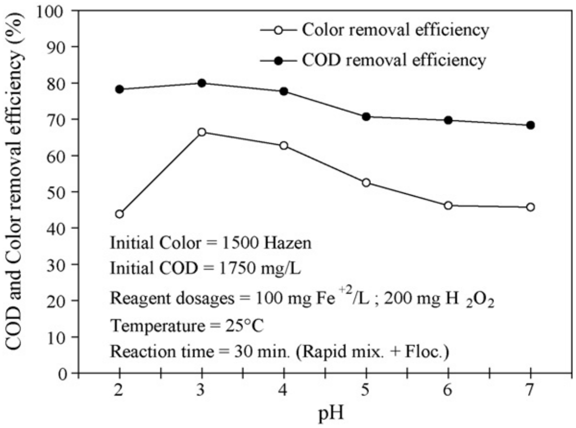

Fig. 5. Effect of initial pH on both COD and color removal efficiencies in Fenton's oxidation tests for reagent dosages of 100 mg/L Fe^{2+} and 200 mg/L H_2O_2.

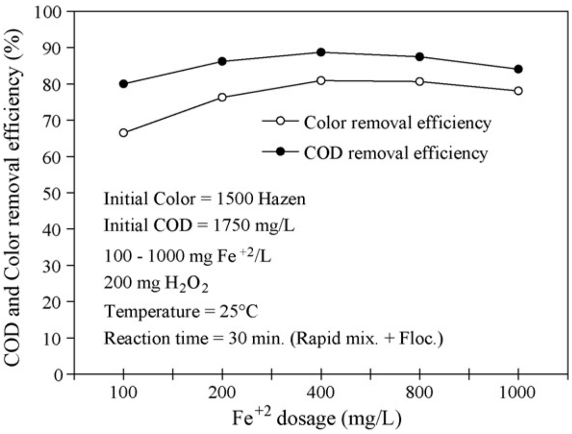

Fig. 6. Effect of Fe^{2+} dosage on both COD and color removal efficiencies in Fenton's oxidation tests for 200 mg of H_2O_2/L and initial pH of 3.0.

of residual COD and residual color in the UASB effluent were about 80% and 66.5%, respectively. At pH 5.0–7.0, both COD and color reductions were smaller, compared to results of pH 3.0. This could be due to decrease in the synergistic effect of H_2O_2 and Fe^{2+} at pH >5.0 [30].

Hence, pH 3.0 was found as the initial pH for the further batch experiments investigating the effects of Fe^{2+} and H_2O_2 dosages on both COD and color removals from the UASB effluent. Fig. 5 shows the effect of initial pH on COD and color removal efficiencies using the dosages of 100 mg Fe^{2+}/L and 200 mg H_2O_2/L, respectively. In the next step, effects of increasing dosages of Fe^{2+} and H_2O_2 were investigated on the basis of preliminary test results.

3.3.2. Effect of Fe^{2+} dosage

The effect of Fe^{2+} dosage on the removal of residual COD and color in the UASB effluent was investigated by conducting a series of batch experiments. Batch experiments were conducted by dosing different Fe^{2+} dosages varying from 100 to 1000 mg/L for a fixed dosage of 200 mg H_2O_2/L at initial pH 3.0. Both COD and color removal were increased with Fe^{2+} dosage. However, further addition of Fe^{2+} over 400 mg/L did not increase the removal efficiency in these parameters, due to triggering of disproportionation of the oxidant. Under the condition of 400 mg Fe^{2+}/L and 200 mg H_2O_2/L, removal efficiencies of residual COD and color were obtained to be 88.7% and 80.9%, respectively. Fig. 6 depicts the effect of Fe^{2+} dosage on the removal of residual COD and color in the UASB effluent at initial pH 3.0.

For the increasing dosage of Fe^{2+}, the most effective oxidation was achieved using Fenton's reagent with a 1:2 ratio of H_2O_2:Fe^{2+} at 25 °C. Fenton's oxidation removed 1552 mg/L of COD from the UASB effluent with the dosages of 400 mg Fe^{2+}/L and 200 mg H_2O_2/L at initial pH 3.0 for a total reaction time of 30 min. Therefore, to remove 1 g of COD in the UASB effluent, only 0.26 g of Fe^{2+} and 0.13 g of H_2O_2 were consumed, respectively.

3.3.3. Effect of H_2O_2 dosage

A series of batch experiments was conducted by dosing different H_2O_2 dosages varying from 200 to 1200 mg/L for a fixed dosage of 100 mg Fe^{2+}/L at initial pH 3.0. Results in Fig. 7 illustrate that further addition of H_2O_2, up to 1200 mg/L, gave good results on both COD and color removal. No sludge flotation was observed during the reaction under these conditions. Optimal COD and color removals were obtained at the dosage of 100 mg Fe^{2+}/L and 1200 mg H_2O_2/L. Under this condition, 95% of residual COD and 95.7% of residual color were removed from the UASB effluent. Fig. 7 illustrates the effect of H_2O_2 dosage on the removal of residual COD and color in the UASB effluent at initial pH 3.0.

For the increasing dosage of H_2O_2, the most effective oxidation was achieved using Fenton's reagent with a 12:1 ratio of H_2O_2:Fe^{2+} at 25 °C. Fenton's oxidation removed 1662 mg/L of COD from the UASB effluent with the dosages of 100 mg Fe^{2+}/L and 1200 mg H_2O_2/L at initial pH 3.0 for a total reaction time of 30 min. Therefore, to remove 1 g of COD in the UASB effluent, only 0.06 g of Fe^{2+} and 0.72 g of H_2O_2 were consumed, respectively.

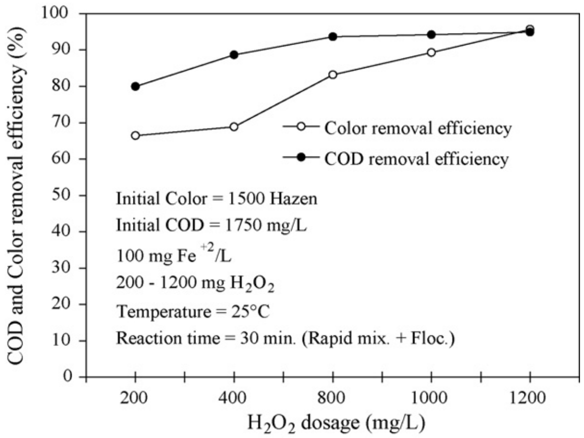

Fig. 7. Effect of H_2O_2 dosage on both COD and color removal efficiencies in Fenton's oxidation tests for 100 mg of Fe^{2+}/L and initial pH of 3.0.

3.4. *Comparisons with literature data*

3.4.1. *Anaerobic processing of poultry manure*

Table 4 summarizes performance data concerning the comparison of different process typologies on anaerobic processing of poultry manure. The performance data figures out that a wide scale range of different reactor volumes varying from batch to full scale implementations were conducted in anaerobic processing of poultry manure. Biogas yields were achieved between 180 mL/g COD_{added} and 74 m^3/day for a wide scale range of different reactor configurations. Most of studies, including the present study, are carried out at mesophilic conditions maintained between 25 and 35 °C. Table 5 shows that total COD removals range from 32% to 78%, depending on other operational conditions. On the basis of total COD removals, the present study shows a more effective COD removal than those reported by others. This is followed by a 78% of COD reduction with an OLR of 2.9 kg COD/(m^3 day) achieved in a laboratory scale (3.5 L) continuous flow UASB reactor conducted by Atuanya and Aigbirior [1], and 73.3–75% of total COD reductions with HRTs of 0.87–1.81 days achieved in two laboratory scale (2.6 L) UASB reactors operated by Kalyuzhnyi et al. [5], respectively. Low COD removals may be attributed to relatively high initial OLR (or COD loading) and/or low HRTs conducted by others. Differences in performances may also be attributed to the different bacterial populations used as seed sludge in the reactors. Since no other studies reported the removals of SCOD, TS, TSS, VSS, VS, TKN, TP, and BOD_5 in anaerobic processing of poultry manure, there are no comparable values for those parameters measured in this study.

3.4.2. *Fenton's oxidation*

Table 5 summarizes performance data concerning the comparison of different process typologies on Fenton's oxidation. The performance data shows that optimum initial pH is found to be 3.0–4.0 in most of studies including the present study. COD and color removals obtained by Aydin and Sarikaya [9] and Badawy and Ali [33] are comparable with the results obtained in this study. Differences in performances may be attributed to the characteristics of wastewaters, reagent dosages, initial values of pH and COD, and also reaction times.

3.5. *Economical discussion*

Biological treatment of wastewater, groundwater, and aqueous hazardous wastes is often the most economical alternative when compared with other treatment options. Although many organic molecules are readily degraded, many other synthetic and naturally occurring organic molecules are biorecalcitrant. It is well known that advanced oxidation processes (AOPs) are very promising methods for the remediation of contaminated ground, surface, and wastewaters containing non-biodegradable organic pollutants. However, costs associated with chemical oxidation alone can often be prohibitive for wastewater treatment. A potentially viable solution is the integration of chemical and biological treatment processes as an economical means for treating biorecalcitrant organic compounds in wastewater [35]. With the

Table 4
Comparison of different process typologies on anaerobic processing of poultry manure

Substrate used	Reactor type and volume	OLR or initial feeding value	HRT or operation time	Temperature	Efficiency (COD, BOD_5, TS or VS removal)	Biogas or methane yield	Reference and region
Poultry manure	Pilot-scale UASB, 15.7 L	0.650–1.783 kg COD/(m^3 day)	15.7, 12, and 8.0 days	32 ± 2 °C	85.3 (±1.9)% COD, 93 (±1.2)% BOD_5, 75.8 (±3.6)% TS, 74 (±3.7)% VS	4.2–13 L/day, 6.87 (±2.46) L biogas/day	Present study, Turkey
Poultry manure	Full scale anaerobic digester, 95 m^3	1.6–2.0 kg VS/(m^3 day)	30–52 days	35 °C	NS	55–74 m^3 biogas/day	Converse et al. [31], USA
Poultry manure	NS	4% and 1% influent and 2.53% VS conc.	29–12 and 30 days	37 °C	NS	0.245–0.372 and 0.627 m^3 biogas/kg VS	Webb and Hawkes [32], UK
Liquid fraction of hen manure	Two laboratory scale UASB, 2.6 L	11.05–12.07 g COD/L day	0.87–1.81 days	35 °C	73.3–75% COD	3.51–3.59 L biogas/L day	Kalyuzhnyi et al. [5], Russia
Poultry wastewater	Continuous flow UASB, 3.5 L	2.9 kg COD/(m^3 day)	13.2 h	26–34 °C	78% COD	0.26 m^3 CH_4/kg COD	Atuanya and Aigbirior [1], Nigeria
Broiler manure, cattle manure, and their mixtures	Seven sets of anaerobic batch reactors, 100 mL	12,000 and 53,500 mg COD/L	27–91 days of operation	35 °C and ambient temperature	32–43.3% and 37.9–50% total COD	180–270 and 223–368 mL biogas/g COD added	Gungor-Demirci and Demirer [6], Turkey
Poultry droppings, and agricultural wastes	Batch pilot-scale digester, 0.28 m^3	38.49 kg of substrate (wet weigh) in the ratio of 1:1 (substrate:water)	40 days	25–29 °C	NS	137.16 L biogas/day (poultry droppings)	Anozie et al. [7], Nigeria

OLR, organic loading rate; HRT, hydraulic retention time; COD, chemical oxygen demand; BOD_5, 5-day biological oxygen demand; TS, total solids; VS, volatile solids; UASB, up-flow anaerobic sludge blanket; NS, not specified.

combination of biological treatment and AOPs, investment and operating costs are calculated to be much lower for a biological process than a chemical one: investments costs for biological processes range from 5 to 20 times less than chemical ones such as ozone or hydrogen peroxide, while treatment costs range from 3 to 10 times less [36,37].

To meet strict laws on environmental protection, the COD in effluent discharged from poultry industries must be reduced to a significant extent, and there is a need to install a proper post-treatment (polishing) unit after an undergoing UASB reactor. Recently, many of AOPs, being a post-treatment unit, have been often conducted to reduce organic load or toxicity of biologically pre-treated wastewaters [11,9,38–41]. The AOPs, which generate hydroxyl free radicals with a high electrochemical oxidant potential in sufficient quantity to affect water constituents. They could be formed using classical oxidants (hydrogen peroxide, ozone, etc.) and UV radiation or catalyst. One common feature of such systems is high demand on electrical energy for devices such as ozonizers, UV lamps, ultrasounds and this result in higher treatment costs from the economic point of view. However, the only exception is Fenton process, where under acidic conditions, a Fe^{2+}/H_2O_2 mixture produces hydroxide radicals in a very cost-effective manner [42]. Similarly, it was reported that Fenton's oxidation was found to have less operating cost for color removal from wastewater per cubic meter than the cost for other AOPs such as ozone and ozone/hydrogen peroxide applications [40]. However, in practical applications, a certain amount of iron hydroxide sludge is produced by Fenton's method, and therefore this leads to the problem of disposing the sludge. The cost of ferrous ions and sludge treatment is about 1/4–1/2 of total operational cost. Conventionally, the produced iron hydroxide sludge is separated from wastewater by using sedimentation or flotation techniques [43]. Hence, from the economical point of view, different process modifications for the disposal of produced sludge by Fenton's oxidation were conducted in some investigations [43–45].

Table 5
Comparison of different process typologies on Fenton's oxidation

	Reference and region					
	Lau et al. [11], Hong Kong	Aydin and Sarikaya [9], Turkey	Birgul and Akal-Solmaz [19], Turkey	Park et al. [34], Korea	Badawy and Ali [33], Egypt	Present study, Turkey
Wastewater	UASB pretreated leachate	UASB + ASBR preatreated opium alkaloid industry effluent	Textile effluent	Livestock wastewater	Combined industrial and domestic wastewater	UASB treated poultry manure wastewater
Fe^{2+} dosage (mg/L or M)	300 mg/L	120 mg/L	30 mg/L	0.066 M	400	400 and 100
H_2O_2 dosage (mg/L or M)	200 mg/L	200 mg/L	150 mg/L	0.2 M	550	200 and 1200
H_2O_2/Fe^{2+} ratio	0.67	1.67	5.0	3.03	1.375	0.5 and 12
Initial COD (mg/L)	1500	700	820	NS	1750–3323	1750
Initial pH	6.0	4.0	3.0	4.0	3.0	3.0
Reaction time	RMT = 30 s; FT = 10 min; ST = 30 min	RMT = 5 min; FT = 25 min	RMT = 2 min; FT = 20 min; ST = 2 h	RMT + FT = 60 min	NS	RMT = 5 min; FT = 25 min; ST = 30 min
COD removal (%)	70	90	52	70	90	88.7 and 95
Color removal (%)	NS	95	96	70	Up to 100	80.9 and 95.7

UASB, up-flow anaerobic sludge blanket; ASBR, aerobic sequencing batch reactor; COD, chemical oxygen demand; RMT, rapid mixing time; FT, flocculation time; ST, settling time; NS, not specified.

4. Conclusions

With 8.0 days of HRT and an OLR of 0.76 kg COD/(m^3 day), the UASB process showed an optimal performance on total COD removal with a treatment efficiency of 90.7% at the day of 63. During collection of the UASB effluent for the subsequent treatment of Fenton's oxidation, the UASB process on average removed 85.3 (±1.9)% of COD in the raw poultry manure wastewater, which contained an average COD concentration of 12,100 (±910) mg/L. Preliminary batch experiments showed that optimal initial pH was found to be 3.0 for the further COD and color removal from the anaerobically treated poultry manure wastewater using Fenton's oxidation. About 89% of residual COD and 81% of residual color were further removed from the UASB effluent using 400 mg Fe^{2+}/L and 200 mg H_2O_2/L at an optimal initial pH of 3.0. Furthermore, about 95% of residual COD and 96% of residual color were succesfully removed from the UASB effluent with the dosages of 100 mg Fe^{2+}/L and 1200 mg H_2O_2/L. For both conditions of increasing dosages of Fe^{2+} and H_2O_2, final effluents after Fenton's oxidation had

COD concentrations, which were fairly lower than the acceptable sewer system discharge level of the present regulations of Istanbul Water and Wastewater Administration (ISKI), Turkey. Results of this experimental study clearly indicated that removal of COD from the raw poultry manure wastewater could be effectively improved up to about 99.3% with the further contribution of Fenton's oxidation technology used as a post-treatment unit.

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