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A comparison between anaerobic and aerobic biological treatment for real wastewater containing high concentration of dimethylamine: case study of wastewater from artificial leather production

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ABSTRACT

In this work, a feasibility study into the treatment process of artificial leather wastewater using both activated and anaerobic sludge was performed. The effects of biomass concentration, different feed dilutions as well as the presence of a nutrient medium in both conditions were examined. The results indicated that as the mixed liquor suspended solids were increased from 800 to 3000 mg/L, chemical oxygen demand (COD) removal increased 41% and 10% in aerobic and anaerobic conditions respectively. Adding nutrient medium had no significant effect on COD removal in both conditions. Under aerobic conditions and at a lower hydraulic retention time of 3 d, COD removal was about 99% while in anaerobic conditions and in similar situations maximum COD removal was up to 60% during 7 d. The effects of amendment of two carbon sources (glucose and methanol) were investigated and results indicated that in comparison with methanol, using glucose as the carbon source could lead to higher COD removal, under anaerobic conditions (23%) while it has no significant effect at the aerobic condition. In conclusion, COD removal was higher under aerobic conditions than anaerobic conditions. Furthermore, dimethylamine concentration in treated wastewater under aerobic conditions was below the detected limit of the analyzer (<1 ppb).

Keywords: Activated sludge; Anaerobic sludge; Artificial leather; Biological treatment; Dimethylamine

1. Introduction

Iran is located in the arid and semi-arid region of planet earth that faces a water crisis. Therefore, researchers consider industrial wastewater treatment methods as an efficient solution for challenging this problem. Industrial wastewater is classified as hazardous because it often contains a wide range of toxic, resistant and bio-accumulative substances [1–3].

The artificial leather industry is known as a polluting industry in terms of high levels of dimethylformamide (DMF) in its discharge which is considered carcinogenic due to the presence of amine compounds. Therefore, its recovery from waste streams is of great importance. DMF is a colorless liquid that is miscible with water. It is a common solvent for chemical reactions and is a polar aprotic solvent with a high boiling point that is used as the solvent to dissolve polyurethane resins and coat them on the base fabric in the production of artificial leather.

Generally, the distillation process has been the most popular method applied to reduce the harmful effect of DMF on the environment [4]. In this process, dimethylamine (DMA) is an intermediate formed during the distillation

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process, had a high concentration in artificial leather [5]. Application of DMA and other aliphatic amines in numerous other chemical and pharmaceutical industries and thus their discharge into water sources has raised concerns due to the associated problems resulting from their toxicity and low biodegradability [6]. Furthermore, the presence of DMA may lead to hazardous material such as N-nitrosodimethylamine (NDMA), which is a potent carcinogen, being produced. The effective approach to achieve NDMA control is to remove its precursors such as DMA from waste streams being released into the environment that could help to reduce NDMA formation [7–11].

Various approaches have been proposed for removing these compounds from effluents, including physical, chemical and biological methods. Physical and chemical methods are not recommended for the elimination of amine compounds due to the high operational costs, low environmental compatibility and also the inability to completely eliminate the desired compounds [12]. However, biological methods are considered among the most promising candidates for the treatment of wastewater by researchers that could pave the way in reusing treated industrial effluents for agricultural purposes [13]. Biological treatment methods are divided into two categories including the use of pure microorganisms or mixed culture which is separated into aerobic and anaerobic methods. Previous studies have been focused on the bio-removal of amine compounds from synthetic effluent using pure strains [9]. Few reports have focused on the investigation of DMA biodegradation using a mixed culture of microorganisms [7–11], for example, Wang et al. [10] utilized a mixed culture for removal of 4 different precursors of NDMA such as DMA, dimethylaminobenzene, dimethylformamide, and trimethylamine (TMA) using synthetic wastewater.

The most common procedure among the biological treatment method is the use of activated sludge or anaerobic sludge. Except for dissolved oxygen, temperature and pH, there are several affecting parameters in biological treatment efficiency including the kind of substrate, hydraulic retention time (HRT), biomass and feed concentration and addition of nutrient media which could affect microbial communication and diversity.

To the authors' knowledge, there has not yet been any report on the biological treatment of actual wastewater containing DMA, particularly regarding synthetic leather. Thus, the aim of the present study is to investigate the feasibility of the treatment process of artificial leather wastewater using both activated and anaerobic sludge and to make necessary comparisons. In addition, the effect of adding glucose and methanol, as the sources of carbon, on cell growth and biodegradation were evaluated.

2. Material and methods

2.1. Mixed culture

2.1.1. Aerobic mixed culture

The aerobic mixed culture was obtained from the sludge recycle stream of the sedimentation tank in a biological wastewater treatment plant of a drug manufacturing company (CinnaGen, Karaj, Iran). Initially mixed liquor suspended solids (MLSS) of the sludge was 2,300 ± 100 mg/L.

2.1.2. Anaerobic mixed culture

The anaerobic sludge was obtained from the anaerobic lagoon of a drug manufacturing company (Osveh Company, Tehran, Iran). The anaerobic sludge was refrigerated at 4°C prior to being used in the experiments. The initial MLSS of sludge was 26,000 ± 200 mg/L.

2.2. Wastewater

The wastewater of artificial leather production was sampled from an artificial manufacturing factory located in Tehran, Iran. This wastewater was collected from condensed water vapors coming from the top of the distillation column of the DMF recovery plant of this factory. The main characteristics of the artificial leather wastewater are presented in Table 1.

2.3. Adaptation of activated sludge to the wastewater

Activated sludge was acclimated to wastewater according to the method described by Yang et al. [11] in a period of 45 d. The activated sludge was fed into a sequencing batch reactor (SBR) of 5 L every 48 h containing 1 g glucose and 10% volume fraction of real wastewater. The temperature of the reactor was kept constant at 30°C [14]. The sludge retention time was maintained in the SBR reactor for 24 h, at a HRT of 15 d, and batch tests were carried out afterward. Then, every 10 d, as the concentration of glucose was reduced, and the concentration of effluent in the feed was increased up to a certain point so that the concentration of the wastewater would reach 100%. During the acclimation period of the activated sludge, the nutrient media fed to the reactor had the following composition (g/L): $MgSO_4 \cdot 7H_2O$ (0.2), $CaCl_2 \cdot 2H_2O$ (0.02), KH_2PO_4 (1.5), K_2HPO_4 (1.5), and 10 mL of the trace element was used in the solution. The trace element solution included (mg/L): $FeSO_4 \cdot 7H_2O$ (300), $MgCl_2 \cdot 4H_2O$ (180), $CoCl_2 \cdot 6H_2O$ (106), $Na_2MoO_4 \cdot 2H_2O$ (34) [11]. The pH was adjusted to 7–8 [14,15]. The activated sludge was aerated with a pump using a flow rate of 4.5 L/min and the rate of aeration was adjusted to ensure that the dissolved oxygen concentration in the tank was always kept above 3 mg/L [14]. In this project, one factor at a time method was used instead of an experimental design for optimization [16,17].

2.4. Biodegradation experiments

2.4.1. Aerobic biodegradation

Batch experiments were conducted in 100 mL serum bottles. The working volume containing wastewater,

Table 1
Wastewater characteristics

Parameter	Value
Chemical oxygen demand	1,500 mg/L
Total organic carbon	552 mg/L
Dimethylamine	991 mg/L
pH	11

nutrient media and activated sludge was 20 mL. The initial MLSS was set at 3,000 mg/L. Samples were shaken on a rotary shaker at 170 ± 2 rpm and 30°C ± 2°C. During days 0, 1, and 3 of the batch experiments, samples were taken from serum bottles, for chemical oxygen demand (COD) measurement. To investigate the effect of co-substrate as the carbon source, 1g glucose or methanol with COD of around 1,000 mg/L was added to the serum bottle. The amount of glucose was measured using test kits at 0, 12, and 24 h under aerobic conditions [18,19].

To assess the kinetics of the pollutant removal, batch experiments were performed with 1g glucose and wastewater as the carbon source.

The sorption experiments of wastewater were performed on sterilized sludge using an autoclave. After being in a shaker incubator for 72 h, measurements and control were conducted based on the reduction of COD. All the biodegradation and sorption experiments were conducted in triplicate.

2.4.2. Anaerobic biodegradation

800 mL serum bottles were utilized to carry out the anaerobic experiments. 500 mL of the serum bottles were filled with anaerobic sludge and wastewater. The serum bottles were shaken on a rotary shaker at 170 ± 2 rpm. The temperature of the liquor was adjusted at 35°C ± 2°C. The samples were flushed with nitrogen gas for 20 min to make the anaerobic condition. The effect of different values of MLSS on removal efficiencies was investigated. The serum bottles were fed every 7 d using initial wastewater, then the sludge was given after 1h to sediment before the top liquor being taken. This procedure was repeated 3 times. The test occurred every 7 d, on days 7, 14 and 21, using glucose as the co-substrate and in two CODs, that is, 1,984 ± 80 and 1,016 ± 65 mg/L, respectively. In addition, methanol was investigated as the co-substrate in two CODs, that is, 1,126 ± 65 and 792 ± 50 mg/L, respectively. All the biodegradation experiments were carried out in triplicate under anaerobic conditions.

2.5. Analytical methods

The MLSS content of sludge was measured according to the Standard Methods [20]. The growth of microorganisms was measured by monitoring optical density (OD) at 600 nm using a UV-Vis spectrophotometer (JASCO-Japan, V550). Then the biomass concentration was estimated applying a calibration curve of OD_{600} vs. dry weight (per liter). The COD was measured according to 5220D Standard Methods (Association and Association, 1989). To determine the COD values, samples were centrifuged at 8,000 rpm for 30 min [11]. DMA was measured by means of a gas chromatography instrument (GC) (Yang Lin, ACME-6100) equipped with a helium ionization detector and a capillary column TRACSIL TRB-5 (30 m × 0.53 mm × 3.0 μm) [21]. Before injection, 0.1 mL of 2.5 M NaOH was added to 1 mL of sample, then serum bottles were sealed using rubber septa and aluminum crimp cap. To reach vapor-liquid equilibrium, samples were maintained at 35°C for 24 h then the concentration of DMA of the gas phase was determined via injecting headspace gas into the GC instrument. The temperature of both injector and detector ports was 200°C. The oven temperature was initially set at 35°C, which was then raised to 70°C at a rate of 10°C min^{-1} and then was finally maintained at 70°C for 1 min. Helium at 20 mL/min was applied as the carrier gas in gas chromatography. Formaldehyde was measured by means of a spectrophotometer according to the method described by Eiroa [22]. To measure the amount of COD that was adsorbed by the microorganisms, firstly, the microorganisms were sterilized in autoclaved and they were inactivated, then a solution with specified COD was added to the solution containing inactivated microorganisms. Since they were inactivated, COD was only adsorbed on the surface of the microorganisms and there was no COD reduction from the biodegradation process. Adsorbed COD was obtained by deducing of final COD from the initial COD.

3. Results and discussion

3.1. Effect of biomass concentration on removal efficiency of COD

The effect of MLSS on the removal efficiency under aerobic and anaerobic conditions is shown in Table 2. According to Table 2, when MLSS was raised from 800 to 3,000 mg/L, the removal efficiency of COD grew by almost 30% and 10% in aerobic and anaerobic conditions respectively. Indeed, increasing MLSS concentration lead to an increase in the removal efficiency of toxic wastewater due to the increase of $S_{threshold}$ and the reduction of inhibitory effect. According to previous outcomes, the kinetic model of biodegradation changes from Haldane to Monod because of the increasing floc's size, the decrease of F/M and increasing resistance to toxic materials [23].

Under both aerobic and anaerobic conditions, due to the fixed initial concentration of the COD values, as the MLSS grew the F/M ratio showed a drop from 1.7 to 0.4 which was reported as the suitable levels of F/M ratio [24].

Table 2
Effect of MLSS on removal efficiency of COD in aerobic and anaerobic condition

Process	Initial COD (mg/L)	Final COD (mg/L)	Initial MLSS (mg/L)	Initial F/M	Removal efficiency %
Aerobic	1,335 ± 100	605 ± 10	800 ± 90	1.7	55
	1,249 ± 90	54 ± 2	3,000 ± 200	0.4	96
Anaerobic	1,321 ± 60	971 ± 20	800 ± 90	1.7	27
	1,321 ± 60	835 ± 15	3,000 ± 200	0.4	37

According to Table 2, as F/M decreased, the removal efficiency of COD grew. Also, since the concentrations of initial feed and also MLSS concentration are two influential factors that can vary F/M, the effect of different feed at fixed MLSS concentration was needed to be studied. These experiments were performed. As presented in Table 3, under aerobic conditions, increasing the COD of the feed caused no obvious change in the removal efficiency whereas, under anaerobic conditions, a decrease of 21% was observed.

The result of Table 2 also shows that the removal efficiency in aerobic conditions was found to be 96%, at a maximum concentration of biomass and after 72 h. However, the results achieved for the anaerobic system indicated a lower removal efficiency of 37% after 7 d.

Previous reports show contradictory results about the removal efficiency of wastewater containing aromatic amines in aerobic and anaerobic conditions. For example, results of Meiberg and Harder [25] showed that the removal efficiencies of DMA and TMA from synthetic effluent in aerobic condition were higher than anaerobic; However, Wang et al. [10] indicated that the anaerobic process with 95% of efficiency in comparison with 99% of the aerobic process using different low concentrations of DMA.

In this study, by two reasons of adaptation of sludge and the effect of toxicity on methanogenic microorganisms, the anaerobic condition has a lower efficiency than aerobic condition. Adaptation of anaerobic sludge was not done because the sample of sludge was taken from a wastewater treatment plant containing aromatic amines materials, while the activated sludge was acclimated for a period of 45 d by means of the stepwise method. The investigation conducted by Arabjafari et al. [23] proved that the duration of adaptation affects the efficiency of elimination, in biological treatment. Methanogen bacteria are considered highly sensitive to toxicity. Hence, the non-compatibility of sludge to the wastewater leads to a decrease in the total efficiency of the process [26]. The result of Wang et al. [10] that shows similar efficiency between aerobic and anaerobic conditions was due to low concentration of these compounds (low than μg/L).

To investigate the effect of acclimation, the study has been performed using 3 passages in which different CODs have been applied in order to investigate the influence of consecutive passages on the effectiveness of the anaerobic process (Table 4).

As it is illustrated in Table 4, at constant values of the initial COD, moving from each passage to the next one, the removal efficiency grew successively which could indicate the adaptation of the sludge to the input wastewater.

3.2. Effects of adding nutrient media to actual wastewater on removal efficiency

As seen in Table 5, at almost equal values of the initial COD, initial MLSS of 3,000 mg/L and adding nutrient media under aerobic and anaerobic conditions had no significant effect on the removal efficiency when the wastewater was used as the sole source of carbon.

Table 5
Effect of adding nutrients on COD removal efficiency

Process	Initial COD (mg/L)	Final COD (mg/L)	Removal efficiency %	Nutrient
Aerobic	1,493 ± 100	99 ± 5	93	+
	1,249 ± 90	54 ± 2	97	–
Anaerobic	1,147 ± 50	744 ± 20	35	+
	1,321 ± 60	835 ± 15	37	–

Table 3
Effect of different initial COD on removal efficiency

Process	Initial COD (mg/L)	Final COD (mg/L)	Initial MLSS (mg/L)	Initial F/M	Removal efficiency %
Aerobic	709 ± 30	36 ± 2	3,000 ± 200	0.2	95
	1,249 ± 70	54 ± 5	3,000 ± 200	0.4	97
	1,493 ± 60	99 ± 7	3,000 ± 200	0.5	93
Anaerobic	988 ± 20	607 ± 5	3,000 ± 200	0.3	39
	1,321 ± 60	835 ± 20	3,000 ± 200	0.4	37
	1,466 ± 70	589 ± 60	3,000 ± 200	0.5	60

Table 4
Variation of output COD during different days in the presence of glucose and methanol under anaerobic condition

Initial COD (mg/L)	Removal efficiency at 1st period (%)	Removal efficiency at 2nd period (%)	Removal efficiency at 3rd period (%)
1,984 ± 80	53	63	82
1,016 ± 65	20	65	82
1,126 ± 65	31	37	62
792 ± 50	12	44	57

Kim et al. [21] observed similar results using TMA as the carbon and nitrogen source. Their results also proved that under aerobic and anaerobic conditions, the nutrient medium had a negligible effect on the removal efficiency. Khalili and Bonakdarpour [27] reported that by increasing the level of nutrient material in the activated sludge which was used anaerobically, a fall in the removal efficiency of dye occurred. While Azizollahzade et al. [28] showed that increasing nutrient media in synthetic effluents could lead to a rise in removal efficiency of the anaerobic sludge. The used wastewater in these researches was synthetic, but their outcomes are contradictory. But in real wastewater due to the presence of macronutrients such as nitrogen and phosphorous, the addition of phosphorous and micronutrient media has no notable influence on removal efficiencies. It is noticeable that the concentration of nitrogen and phosphors inside the wastewater before supplementing external nutrients was 15.6 and 0 mg/L respectively.

3.3. Effects of co-substrate on removal efficiency of COD

Under both aerobic and anaerobic conditions, the effects of glucose and methanol as the carbon sources were studied. The results are reported in Table 6. As shown, by adding glucose, the removal efficiency under aerobic conditions was increased from 96% to 99%; whereas, adding methanol caused the removal efficiency to drop from 96% to 65%.

On the other hand, under anaerobic conditions, by adding glucose, the removal efficiency improved from 37% to 57%; while, adding methanol made no change in the removal efficiency (37% to 37%).

According to literature, adding external carbon source in both aerobic and anaerobic conditions lead to the enriching of cultures by which the efficiency of COD removal increase.

3.4. COD, total organic carbon, DMA and microbial growth variation vs. time under aerobic condition

According to Table 6, it was found that under aerobic conditions, by adding glucose with MLSS = 3,000 ± 200 mg/L and F/M = 0.7 the maximum removal efficiency was achieved. Therefore, the investigation of COD, total organic carbon (TOC), DMA and microbial growth variation with time was performed under these conditions.

The results of COD variation vs. time (during 72 h) are shown in Fig. 1. As shown, the highest removal efficiency of COD was observed during the first 24 h.

The kinetic model of the COD removal efficiency under aerobic condition can be represented by the following equation [29,30]:

$$\frac{-dC}{dt} = kC^n \tag{1}$$

where C is the concentration of COD (mg/L), t is time (h) and n is the reaction order. Integration of Eq. (1) results in Eq. (2) when $n = 1$ and in Eq. (3) when $n \neq 1$:

$$\ln C = -kt + \ln C_0 \tag{2}$$

$$\left(\frac{C}{C_0}\right)^{(1-n)} = 1 - \left[\frac{(1-n)kt}{(C_0)^{(1-n)}}\right] \tag{3}$$

For $n = 2$ Eq. (3) is simplified to the following equation:

$$C = \left[\frac{C_0}{1 + KC_0 t}\right] \tag{4}$$

where C_0 is initial concentration of COD (mg/L) at $t = 0$. In order to elucidate the order of reaction, data were replotted as $(1/C)$ vs. t (Eq. (4)) or $\ln C$ vs. t (Eq. (2)). The R^2 obtained from the variation of the COD concentration is quite compatible with the second-order equation ($R^2 = 0.99$).

Also, TOC variation vs. time is reported in Fig. 2. Same as COD, the maximum TOC removal was obtained in early 24 h. The total removal efficiency of TOC was up to 96%.

The TOC removal efficiency can also be obtained using Eqs. (1)–(3) by setting the concentration of TOC in the equation instead of the COD. It was noticed that the TOC removal efficiency is quite compatible with the second-order equation as $R^2 = 1$ was obtained while applying the first-order equation led to a $R^2 = 0.7$ which means little compatibility. Also, the second-order equation can be considered as the kinetic model of the TOC removal efficiency.

As the COD value determined by glucose was 1,000 mg/L and the COD of wastewater was assumed 1,461 mg/L, glucose was measured using test kits, and the results showed that it was consumed within the first 12 h and afterward

Table 6
Effect of co-substrates on COD removal efficiency in aerobic and anaerobic conditions

Process	Initial COD (mg/L)	Final COD (mg/L)	Removal efficiency %	Co-substrate
Aerobic	1,249 ± 70	54 ± 2	96	–
	2,391 ± 80	29 ± 2	99	Glucose (1 g/L)
	1,910 ± 40	670 ± 15	65	Methanol (1 g/L)
Anaerobic	1,321 ± 60	835 ± 20	37	–
	2,711.5 ± 80	1,111 ± 70	59	Glucose (1 g/L)
	2,143 ± 80	1,350 ± 20	37	Methanol (1 g/L)

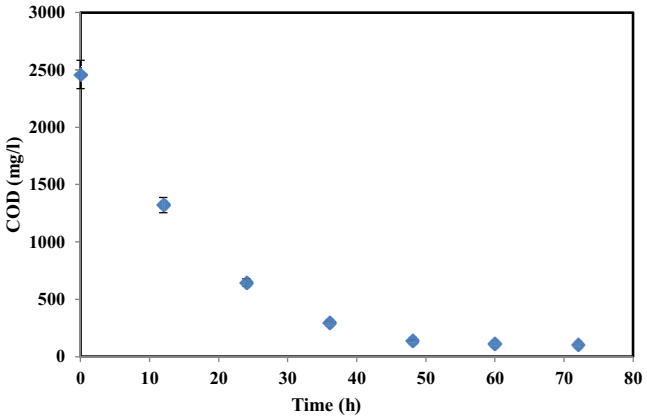

Fig. 1. Variation of COD during 72 h.

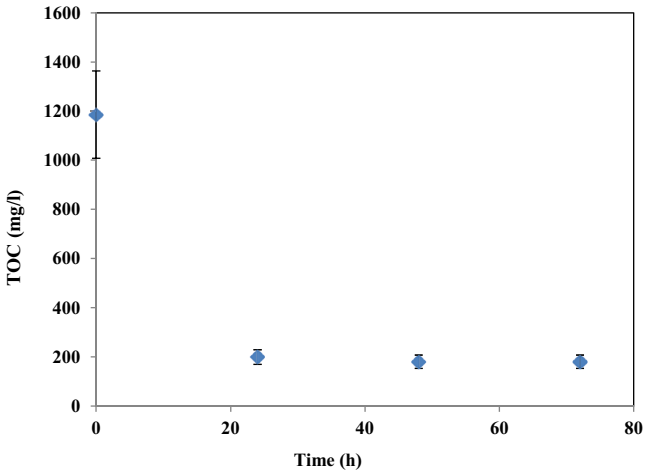

Fig. 2. Variation of TOC vs. time.

DMA was eliminated within 3 d. The complete removal of DMA can be seen in Figs. 3a–c, which is related to the days 0, 1 and 3 respectively that the concentration of DMA was 1,000, 800 and 0 ppm (Fig. 3). Also, the TOC data confirm the complete removal of DMA.

As the COD value determined by glucose was 1,000 mg/L and the COD of wastewater was assumed 1,461 mg/L, glucose was measured using test kits, and the results showed that it was consumed within the first 12 h and afterward DMA was eliminated within 3 d. Due to the result of formaldehyde and Fig. 3c show that DMA was removed. As a calibration curve at this COD, DMA was measured at 1,000 ppm. The removal efficiency of COD showed that DMA was removed.

According to Fig. 3, the peak of DMA was not observed, because the concentration of DMA was lower than the detection limit of the analyzer (<1 ppb).

Fig. 4 shows the changes of formaldehyde concentration during 72 h under aerobic conditions.

Under the aerobic conditions, COD removal occurred in 3 d because of that, formaldehyde was investigated in 3 d. During the first 24 h, it was observed that the initial formaldehyde and glucose were removed successfully. Then, at t = 60 h, formaldehyde was re-produced which could refer to the biodegradation of DMA (Wang et al. [10]). After that, at t = 72 h, the produced formaldehyde started to disappear. It should be noted that 2.2 mg/L of formaldehyde was not eliminated which could be a justification for the final COD in the system, according to the pathway presented by Fournier et al. [7] formaldehyde.

According to the pathway presented by Fournier et al. [7], DMF is degraded to formaldehyde and methylamine. Formaldehyde concentration was measured during 3 d of the aerobic experiment. As shown in Fig. 4, at the beginning of the experiment 6 mg/L of formaldehyde is present in the real wastewater which could be owing to the conversion of the amine compounds due to hydrolysis or the variation of pH of the wastewater. During the first 24 h, it was observed that the initial formaldehyde was removed successfully. Then, at t = 60 h, formaldehyde was re-produced which could refer to the biodegradation of DMA [10]. After that, at t = 72 h, the produced formaldehyde started to disappear.

The growth rate of microorganisms was also examined at different hours and the results are presented in Fig. 5.

According to Fig. 5, microorganisms undergo a log phase during the first 16 h in the presence of glucose. In this time, 20% of DMA was removed while glucose was removed completely because the microorganisms grew well in the presence of glucose. By increase of microorganism population and their tolerance towards new conditions, the rate of DMA removal increased and the maximum DMA removal took place at the stationary phase. Finally, by complete consumption of nutrients, the death phase had occurred.

4. Conclusion

In the present work, a feasibility study into the treatment process of artificial leather wastewater using both activated and anaerobic sludge was performed. The effect of biomass concentration, adding the external carbon sources, nutrient media on COD removal was taken into the investigation as well.

The results illustrated that increasing the biomass concentration from 800 to 3,000 mg/L resulted in removal efficiencies of approximately 21% and 10% in aerobic and anaerobic states, respectively. By increasing F in constant biomass concentrations, to the extent in which F/M remains less than 0.5, removal efficiencies under both aerobic and anaerobic conditions were not changed significantly. Adding nutrient media had no significant effect on the removal efficiency. The results show that the addition of glucose enhanced COD removal efficiency in both conditions especially in anaerobic conditions while the negative effect of methanol addition was observed in aerobic conditions. The removal efficiency under both aerobic and anaerobic conditions, after the compatibility period, in the presence of glucose as the carbon source, was estimated to be 96% in 3 d and 82% in 7 d. The removal efficiency of COD, TOC, formaldehyde, as well as DMA was investigated during 3 d in an aerobic run. Removal kinetic of DMA fitted with second-order and it was found that the kinetic model of TOC removal efficiency was of the same order.

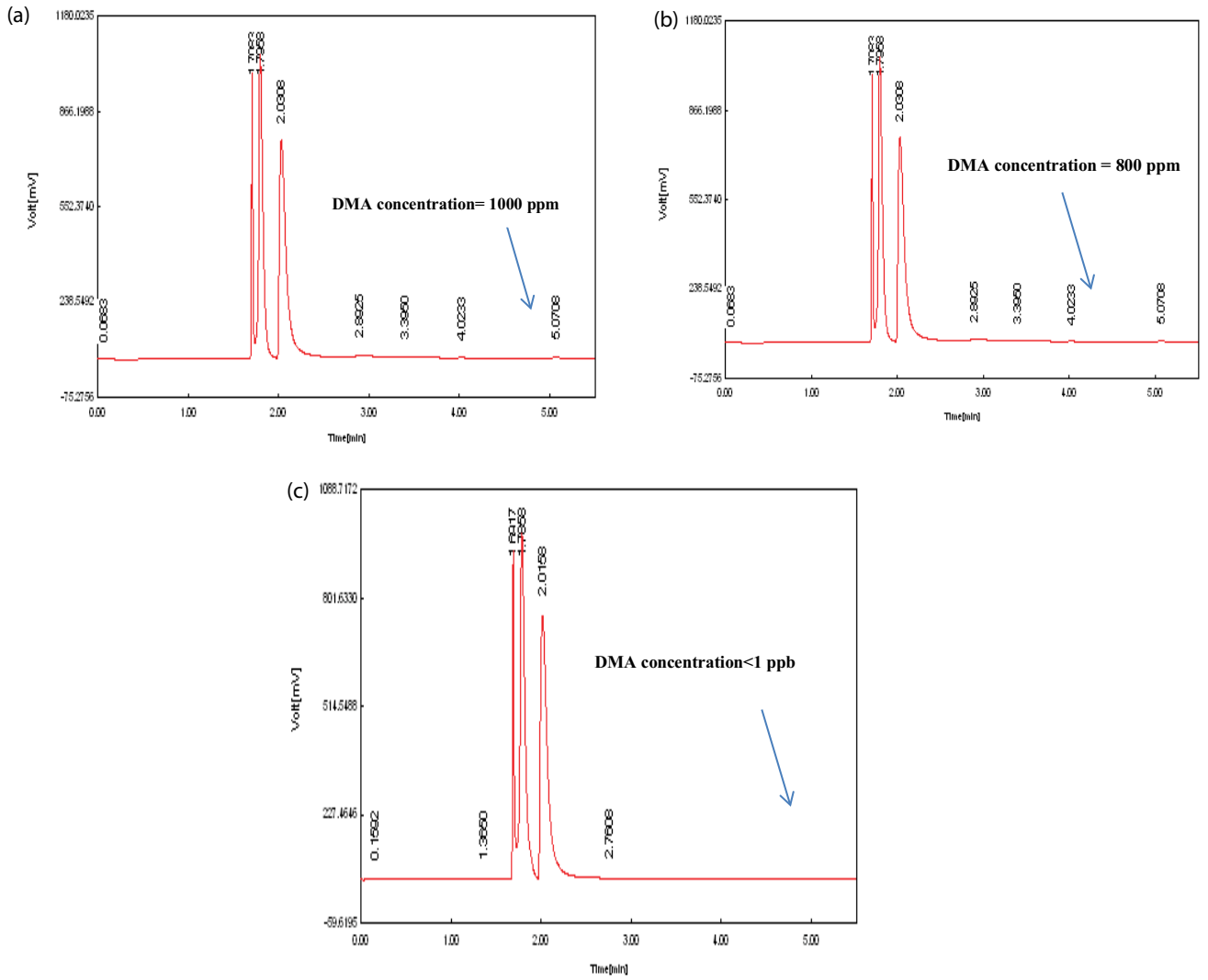

Fig. 3. Dimethylamine variation during day of (a) 0th, (b) 1th and (c) 3th.

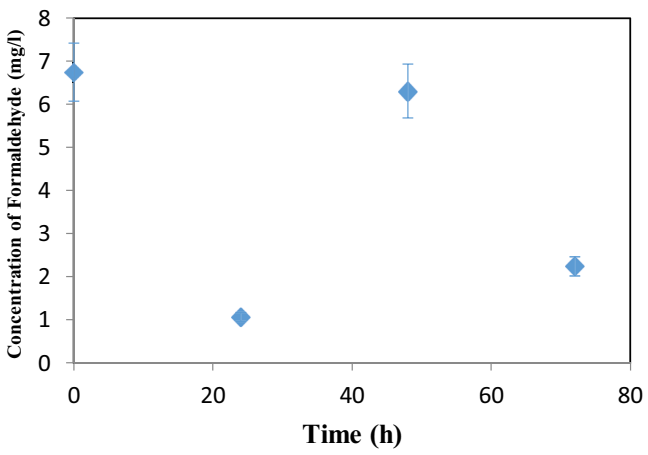

Fig. 4. Variation of formaldehyde during 72 h.

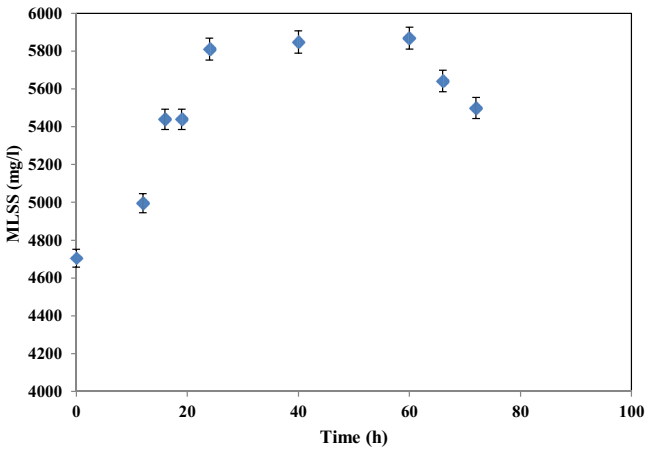

Fig. 5. Variation of cell growth during 72 h.

The analysis of formaldehyde during a run indicated that DMA was changed to formaldehyde and then mineralized to CO_2 as supported by gas chromatography result and TOC test.

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