# Anaerobic Treatment of Tetra-Methyl Ammonium Hydroxide (TMAH) Containing Wastewater

Kuan-Foo Chang, Show-Ying Yang, Huey-Song You, and Jill Ruhsing Pan

***Abstract*—Toxicity of tetra-methyl ammonium hydroxide (TMAH) was investigated in an upflow anaerobic sludge bed (UASB) by anaerobic toxicity assay test. System performance was determined by monitoring total organic carbon (TOC) and ion chromatograph (IC) analysis. The result indicates that TMAH concentration less than 10 000 mg/L is nontoxic to the anaerobic archaea. TMAH containing in the wastewater was completely degraded under anaerobic condition and was transferred to biogas ($CH_4$ and $CO_2$) and $NH_3$. The maximum volumetric loading of UASB is 6.95 kg TMAH/m$^3$-day (4.0 kg TOC/m$^3$-day) and 7.30 kg TMAH/m$^3$-day (4.1 kg TOC/m$^3$-day) for synthetic wastewater and wastewater from a full-scale thin-film transistor liquid crystal display manufacturing industry during the 240-day experiment, respectively. The removal efficiencies of THAM containing wastewater were both higher than 95% treated by UASB.**

***Index Terms*—Anaerobic, anaerobic toxicity assay (ATA) test, methanogen, tetra-methyl ammonium hydroxide (TMAH), thin-film transistor liquid crystal display (TFT-LCD), upflow anaerobic sludge bed (UASB).**

## I. Introduction

SEMICONDUCTOR and thin-film transistor liquid crystal display (TFT-LCD) are the most important industries in Taiwan. A large amount of developer is used in the photolithography processes which generate wastewater containing a high concentration of the developer. Tetra-methyl ammonium hydroxide (TMAH, $(CH_3)_4NOH$) is the major ingredient of the developer. It is a poisonous and corrosive alkaline solution. Depending on the level and duration of exposure, the symptoms of TMAH poisoning include: blurred or double vision; pinpoint pupils; changes in heart rate and blood pressure; abdominal cramping, nausea, and vomiting; diarrhea, excessive salivation sweating, or bronchial secretions; urinary incontinence; muscle twitching, tremors, or convulsions (MSDS database, http://www.msdssearch.com/DBLinksN.htm). This nitrogenous compound is also eutrophic to the water environment. For example, the $NH_3 - N$ equivalency of a six-generation TFT-LCD factory with TMAH concentration of 1000 mg/L and 30 000 cubic meter per day wastewater equals a 1 000 000 population city. Therefore, it is important to find a proper treatment for the TMAH-containing wastewater in Taiwan.

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The TMAH-containing wastewater can be treated by catalytic oxidation, aerobic biodegradation, and anaerobic biodegradation technologies. Catalytic oxidation technology has been developed by Hirano *et al.* [1] to treat TMAH-containing wastewater. The TMAH was first pyrolyzed to trimethylamine and methanol. Then, selective oxidation with a base metal series catalyst for nitrogenous compounds, TMA decomposes into $N_2$, $CO_2$, and $H_2O$. After the catalyst system, the 1% TMAH-containing wastewater was all decomposed. However, the disadvantage of this system is that it is too expensive for the end user.

Biodegradation is potentially the most cost-effective means for treating TMAH-containing wastewater. However, very little has been reported concerning TMAH degradation by microorganisms. Three tetra-methyl ammonium (TMA) degraders have been isolated from the activated sludge acclimated to TMA, which are the members of genus *Paracoccus* [2]. A previous study also found that the acclimation of the TMAH biodegradation can be shortened by seeding the isolated TMA degrader and the degradation of TMAH is superior [3]. However, some of the byproducts of this system are formaldehyde and acetaldehyde, which are toxic compounds for microorganism [1]. Thus, the aerobic treatment of TMAH wastewater requires huge systems that must be maintained under highly restrictive conditions. The process also generates $N_2O$ gas which contributes to the greenhouse effect [1], [4].

Asakawa *et al.* has isolated a Methanogen (strain NaT1) that belongs to the family of *Methanosarcinaceae* and can grow on TMAH as the sole energy source [5]. They concluded that the growth yield was approximately 2.5 g/mol $CH_4$ with 30-mM TMAH as the sole carbon and energy source. The previous studies also found the pathway of TMA degradation as

$$\begin{aligned}
&(CH_3)_4N^+ + \text{H-S-CoM} \\
&\quad\rightarrow (CH_3)_3NH^+ + CH_3 - \text{S-CoM} \\
&(CH_3)_3NH^+ + \text{H-S-CoM} \\
&\quad\rightarrow (CH_3)_2NH_2^+ + CH_3 - \text{S-CoM} \\
&(CH_3)_2NH_2^+ + \text{H-S-CoM} \\
&\quad\rightarrow CH_3NH_3^+ + CH_3 - \text{S-CoM} \\
&CH_3NH_3^+ + \text{H-S-CoM} \\
&\quad\rightarrow NH_4^+ + CH_3 - \text{S-CoM}
\end{aligned}$$

where H-S-CoM is coenzyme M and $CH_3$-S-CoM is methyl-coenzyme M produced from H-S-CoM [6]. Tanaka

[7] also found that a methanogen can use TMA as a substrate and metabolizes TMA to 3 $CH_4$, 1 $CO_2$, and 1 $NH_3$.

Biological treatment is the most economical method for wastewater. And anaerobic technology is a low investment (land and construction) cost, low operation cost, and also has potential energy recovery compared to an aerobic system. Thus, this paper focuses on the anaerobic technology to treat TMAH-containing wastewater to understand the feasibility of this technique.

## II. Material and Methods

### A. Laboratory Reactor Setup

Two 10-L upflow anaerobic sludge bed (UASB) reactors with a three-phase separator were set up to treat TMAH-containing wastewater and a schematic diagram of the UASB can be seen in Fig. 1. One treated synthetic wastewater (UASB-1) and the other treated wastewater collected from LCD manufacturing factory (UASB-2). It was made by acrylic material with dimensions of 10 cm(L) $\times$ 10 cm(W) $\times$ 100 cm(H) for a reaction zone excepted three-phase separator. About 8 L of granular sludge was inoculated as seed biomass. Suspended solid (SS) and volatile suspended solid (VSS) concentration of the granular sludge was 36 950 and 28 250 mg/L, respectively. The granular sludge was collected from a full scale UASB reactor treating food industry (bread, milk, soybean sauce, and soft drink manufacturing) wastewater. As can be seen in Fig. 1, the sludge bed was formed by granular sludge which was heavier than suspended sludge. A sludge blanket was formed by suspended sludge. The sludge bed and blanket are both important for treating wastewater. Two peristaltic pumps (Masterflex, HV-07524-40 driver with an easy-load pump head, HV-075018-02) were applied for feeding and circulation. The feeding rate was from 2 to 20 mL/min for different hydraulic retention time (HRT) tests. The circulation rate was adjusted to 50 mL/min.

The synthetic wastewater was prepared by dilution of 25% TMAH solution which was made by Merck, Inc., Germany (S4311548 512) to a desired concentration. The prepared TMAH concentration range was between 500 to 3000 mg/L. After diluting to a desired concentration, the pH value was adjusted to 7.50 by concentrated HCl. Then, to every 10 L synthetic wastewater was added a 2.5-mL trace element solution [8] and 2 g $NaHCO_3$ for buffering.

The wastewater from a TFT-LCD manufacturing factory in north Taiwan was also tested. It is a fifth-generation TFT-LCD manufacturing factory and total organic carbon (TOC) concentration of its raw wastewater is in a range of 200–1600 mg/L with a pH value of about 11.5. Before feeding, the wastewater was also adjusted in pH value to 7.50. Then, to every 10 L wastewater was added a 2.5-mL trace element solution and 2 g $NaHCO_3$ for buffering.

During the startup period of the UASB-1, the feeding rate of the simulated wastewater was 2 mL/min with the TMAH concentration of about 300 mg/L as TOC (loading of the UASB-1 was about 0.08 kg TOC/$m^3$-day) for sludge acclimation. When the TOC removal efficiency is consistently more than 80%, the loading of UASB is raised. During operation, inlet and outlet TOC concentration were measured every one or two days a time. The TMAH concentration was analyzed once a week to compare with the TOC data. The fed wastewater concentration was from 200 to 1600 mg/L as TOC (loading of the UASB-2 was in a range of 0.21 to 4.1 kg TOC/$m^3$-day).

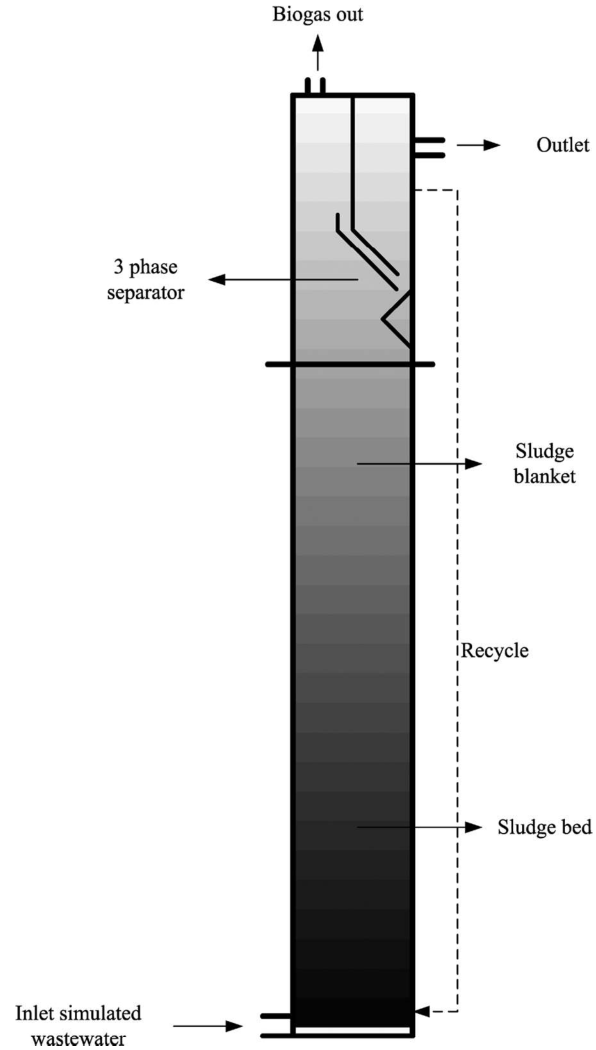

Fig. 1. Schematic diagram of UASB for TMAH-containing wastewater treatment.

### B. Anaerobic Toxicity Assay (ATA) Test for TMAH

An ATA test was conducted at 37 °C in a 90-r/min shaker using 250-mL serum bottles as described by Owen et al. [9]. Serum bottles were filled with 150-mL solution containing a suitable mineral medium [10], 560 mg/L of glucose-chemical oxygen demand (COD), and with 0–10 000 mg/L TMAH for the test. Furthermore, 5000 mg/L $NaHCO_3$ was added for maintaining the neutral pH. The granular sludge (50 mL with SS and VSS concentration of 36 950 mg/L and 28 250 mg/L, respectively) was the same as the UASB that was used for the ATA test. A glucose COD level of 560 mg/L ensured the presence of nonlimiting substrate (TMAH) conditions in the serum bottles. Methane production of each assay bottle was determined during a subsequent 6 h of incubation period. Maximum specific methanogenic activity was calculated from the methane

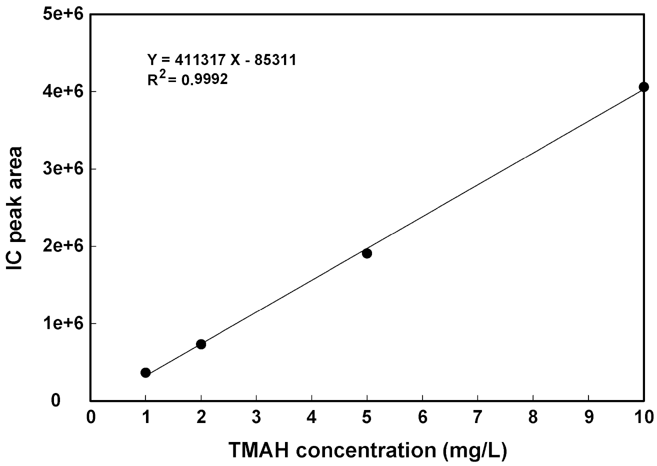


Fig. 2. Regression curve of IC peak area and TMAH concentration.

production through this period. Methanogenic activities of the samples containing TMAH and its metabolites were compared to the blank sample (without TMAH) to determine the degree of inhibition effect on glucose utilization. Inhibition was defined as a decrease in cumulative methane production compared to the blank sample.

### C. Analysis

An ion chromatograph (IC, Dionex DX-100) with CS-15 column was used to analyze the concentration of TMAH and ammonium ion. The elute was 11 mM $H_2SO_4$ at the flow rate 1 mL/min. The R square of the calibration curve for TMAH concentration versus IC peak area was higher than 0.999 with TMAH concentrations of 1.0, 2.0, 5.0, and 10.0 mg/L (Fig. 2). The method detection limit (MDL) of the TMAH was 0.45 mg/L. The recovery of TMAH analysis was 92%.

COD and biological oxygen demand ($BOD_5$) analyses of wastewater were followed by the standard method [11]. TOC concentration was analyzed by TOC (Seivers 800 TOC analyzer). The first step of a Seivers 800 TOC analyzer is oxidation of organic carbon in water by a UV reactor to $CO_2$. Then, the produced $CO_2$ passes through a selective membrane and is analyzed by a detector. Finally, the $CO_2$ concentration is transferred to TOC concentration.

## III. Results and Discussion

### A. Analytical Characteristics of TMAH-Containing Wastewater

Listed in Table I, analytical characteristics of synthetic TMAH-containing wastewater including TMAH concentration (measured by IC), TOC, COD, and $BOD_5$ concentrations as well as calculated data including theoretical oxygen demand (ThOD) and theoretical TOC. ThOD is calculated by assuming 1 TMAH compound is transferred completely to 4 $CO_2$ and 1 $NO_3^-$ compounds (totally needs 11 oxygen atoms for reaction). Thus, 212 mg (2.38 mmol) TMAH needs 26.2-mmol oxygen and its equals 420 mg. Theoretical TOC is calculated by 1 TMAH compound contains 4 carbon atoms. Thus, 212 mg (2.38 mmol) TMAH contains 9.52 mmol carbon and its equals 114 mg theoretical TOC. A 212 mg/L TMAH solution also measured by TOC, COD, and $BOD_5$ and the results listed in Table I were 115, 2.0, and 343 mg/L, respectively.

TABLE I
Theoretical Oxygen Demand (ThOD), COD, $BOD_5$, and TOC Concentration of TMAH-Containing Wastewater

| Analytical item | Concentration (mg/L) |
|---|---|
| Measured concentration by IC | 212 |
| ThOD | 420 |
| COD | 2.0 |
| Theoretical TOC | 114 |
| Measured TOC | 115 |
| $BOD_5$ | 343 |

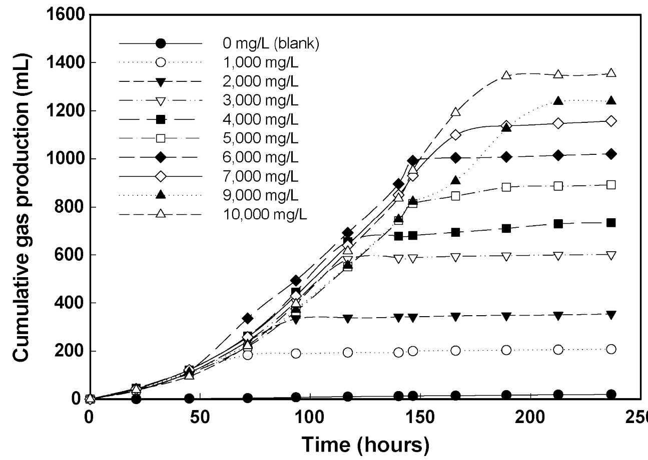


Fig. 3. Cumulative gas production of ATA test for 0–10 000 mg/L TMAH.

It is very strange that TMAH solution cannot be analyzed by a COD method. Only 2 mg/L COD can be analyzed for 212 mg/L TMAH solution. It is due to the fact that TMAH cannot be oxidized by $K_2Cr_2O_7$. This phenomenon also can be found in the wastewater treatment plant of Southern Taiwan Science Park, where more than 80% industries are LFT-LCD manufacturing. The COD/BOD ratio of this plant is less than 1 due to containing lots of TMAH wastewater. This paper suggests that the TMAH-containing wastewater should be measured by TOC. Because of that, the ratio of theoretical and measured TOC of 212 mg/L TMAH solution is 0.99. The other reason is that TOC analysis is more easily and conveniently analyzed than $BOD_5$, although a TMAH solution can be analyzed by $BOD_5$ method. Thus, the followed uses TOC concentration to represent the TMAH wastewater.

### B. ATA Test for TMAH

Total ATA test experimental time of TMAH was about 240 hours. The result of cumulative gas production of different concentration of TMAH is shown in Fig. 3. Biogas was produced in every concentration of TMAH excepted of blank. The cumulative gas production increased with the increasing TMAH concentration. For example, the cumulative gas production for 1000, 5000, and 10 000 mg/L were 180, 880, and 1340 mL, respectively. As can be seen in Fig. 3, the trends of all curves were similar. It indicates that TMAH is

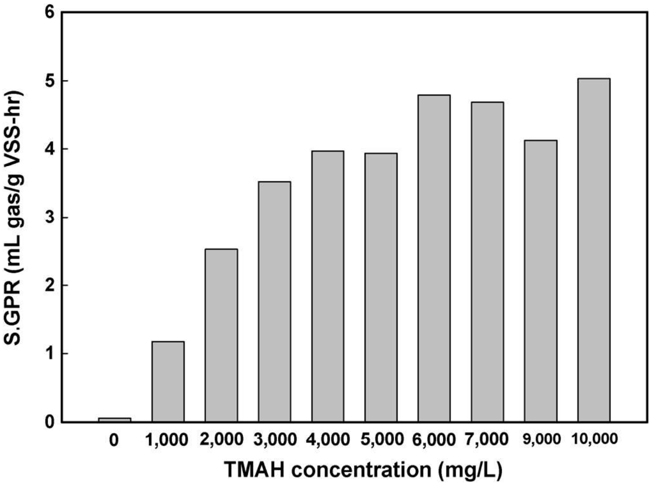


Fig. 4. Specific gas production rate (SGPR) in different TMAH concentration.

not only biodegradable by anaerobic archaea but nontoxic to methanogens under 10 000 mg/L.

The average VSS of each ATA test was about 7000 mg/L (about 1.43 g per test). The specific gas production rate (SGPR) can be defined as gas production rate per gram VSS per hour. Fig. 4 shows the SGPR versus different TMAH concentrations. The result indicates that the SGPR was raised from 0.06 to 4.79 mL gas/g VSS-hour as the higher TMAH concentration from 0 to 6000 mg/L. However, when the TMAH concentration was higher than 6000 mg/L, the slope of the SGPR became more even (from 4.79 to 5.03 mL gas/g VSS-hour). It indicates that the SGPR reached the maximum rate when TMAH had a concentration higher than 6000 mg/L.

The initial and final conditions of TMAH-containing wastewater were also analyzed and the result was listed in Table II. The initial TMAH concentration was 1682 mg/L and the $NH_3-N$ concentration was under detection limit. After 4 minutes reaction, the TMAH and TOC concentrations were decreased slightly to 1635 and 2150 mg/L, respectively. Meanwhile, the $NH_3-N$ concentration and gas production were raised to 12 mg/L and 26 mL, respectively. At the end of the reaction (146 minutes), the TMAH and TOC concentrations were decreased to 235 and 257 mg/L, respectively, as well as the $NH_3-N$ concentration and gas production were increased to 237 mg/L and 1542 mL. This result indicates that the TMAH can be decomposed by anaerobic archaea and produced biogas ($CH_4$ and $CO_2$) and $NH_3$.

The TMAH concentration is transferred to N equivalent and equals 259 mg/L TMAH-N in the initial time. At the end of the experiment, the TMAH-N plus $NH_3-N$ concentration equals 273 mg/L. The little difference between these two data is due to the acceptable analytical error. It indicates that the TMAH is indeed transferred to $NH_3$ in the test. For the biogas, the composition of the biogas was not analyzed; however, it can be estimated roughly by gas production. Generally, 1 g TMAH equals about 2.1 g ThOD (Table I) and 1 g ThOD can produce about 500 mL biogas (350 mL $CH_4$ and 150 mL $CO_2$). The consumed TMAH is 1.45 g in this test and equals 3.1 g ThOD. Thus, gas production in this test should be 1550 mL and is very close to the measured data 1542 mL. This result indicates that the methyl of TMAH can be transferred into biogas by anaerobic archaea successfully.

TABLE II
ANAEROBIC TREATMENT TEST OF TMAH-CONTAINING WASTEWATER

| Time (min) | Gas production (mL) | TOC (mg/L) | TMAH (mg/L) | $NH_3$-N (mg/L) |
|---|---|---|---|---|
| 0 | 0 | 2,200 | 1,682 | 0 |
| 4 | 26 | 2,150 | 1,635 | 12 |
| 8 | 46 | 1,800 | 1,613 | 13 |
| 17 | 126 | 1,460 | 1,528 | 13 |
| 25 | 187 | 1,350 | 1,492 | 35 |
| 42 | 346 | 1,400 | 1,456 | 55 |
| 66 | 592 | 1,245 | 1,387 | 96 |
| 94 | 876 | 1,025 | 1,083 | 92 |
| 114 | 1,096 | 875 | 852 | 164 |
| 123 | 1,216 | 589 | 550 | 209 |
| 137 | 1,444 | 459 | 413 | 260 |
| 146 | 1,542 | 257 | 235 | 237 |

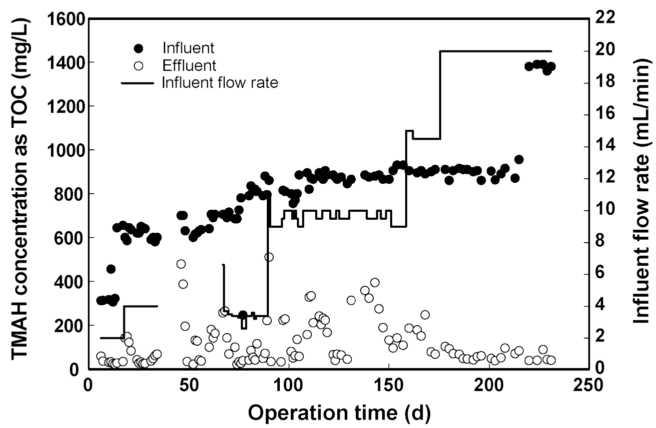


Fig. 5. Performance of the upflow anaerobic sludge bed (UASB-1).

## C. Performance of UASB System

The performance of the UASB-1 is shown in Fig. 5. There are two ways to raise the volumetric loading, one is to increase the TMAH concentration and the other to increase the influent flow rate. In the startup period of the UASB-1, the inlet TMAH concentration was about 300 mg/L as TOC and the influent flow rate was 2 mL/min (HRT $= 3.47$ d). The effluent TOC concentrations in this period were 58 mg/L and then decreased to about 20 mg/L. The startup period ended at the 13th operation day and the TMAH concentration was doubled to about 600 mg/L. The effluent TOC concentration was raised from 20 mg/L to about 150 mg/L in three days of operation, and then the TOC decreased slowly to a level of 25 mg/L. The 600-mg/L TOC feeding was continued for about two months of operation, but the flow rate of simulated wastewater was raised from 2 mL/min, to 4 mL/min, and finally 6 mL/min (HRT $= 3.47$, 1.74, and 1.15 d, respectively). The effluent TOC concentration raised when the inlet flow rate was added and then also decreased to about 30 mg/L in this period. After the 600 mg/L TOC feeding, the TMAH concentration and inlet

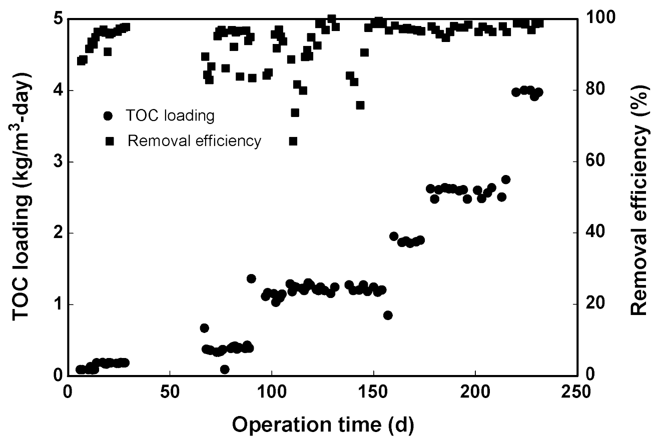

Fig. 6. TOC loading and removal efficiency of the UASB-1.

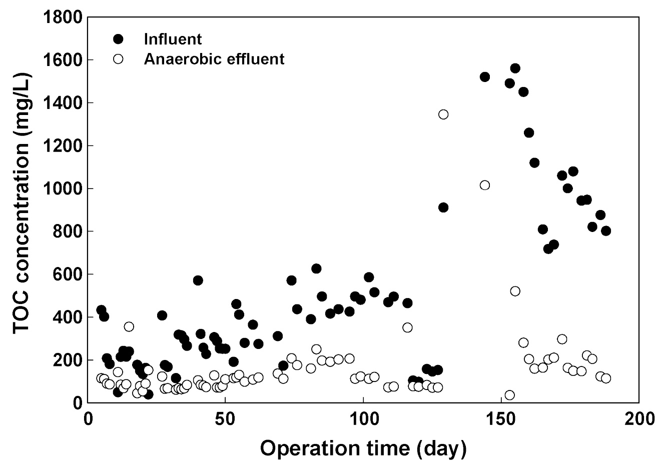

Fig. 7. Inlet and outlet TOC concentration for a full-scaled optoelectric factory wastewater treated by UASB-2.

flow rate of the USAB system were raised again step by step. At the end of the experiment, the TMAH concentration was about 1400 mg/L as TOC and the inlet flow rate was 20 mL/min ($HRT = 0.34$ d). The effluent TOC concentration was between 38.2 and 42.9 mg/L in this time.

The loading of the UASB system and treatment efficiency is displayed in Fig. 6. As can be seen, the TOC loading of the UASB system was raised step by step from 0.1 to 4.0 kg/m$^3$-day (0.17 to 6.95 kg TMAH/m$^3$-day). The removal efficiencies were all over 80% except three data. The removal efficiency was not stable during the experiment period of 50–150 days. This was due to the cold temperature of winter. This system was not equipped with a temperature controller and 10 °C–15 °C water caused the unstable removal efficiency. However, the removal efficiencies were all over 95% at the end of experiment. This result indicates that the TMAH-containing wastewater can be treated by anaerobic biotechnology, such as the UASB, successfully.

The wastewater from a TFT-LCD manufacturing process of an optoelectric company was also tested by the UASB-2 and the results were shown in Fig. 7. The TOC concentration of raw wastewater was about 200–1600 mg/L with pH value of 11.5. The raw water TOC concentration form the TFT-LCD company was fluctuated. The effluent TOC concentration ranged between 70–150 mg/L with removal efficiency of 50%–97%. The removal TOC loading of the system was between 0.21 and 4.1 kg TOC/m$^3$-day (0.36 to 7.3 kg TMAH/m$^3$-day). The efficiency was not as good as the simulated wastewater. This is probably due to the fact that the raw water of TFT-LCD company contained not only TMAH, but also some organic carbons and these unknown organic carbons may be nonbiodegradable by anaerobic microorganisms. However, the results of the pilot test suggested that the TMAH-containing wastewater can be treated by the UASB and the design parameter such as organic loading and HRT, described in Figs. 5–7, of a full scale UASB reactor also can be found in this paper. For instance, the influent flow rate and TOC concentration of a TMAH wastewater are 500 $m^3/day$ and 2000 mg/L, respectively. Consequently, the total organic loading is 1000 Kg TOC/day. If the UASB is used to treat the TMAH wastewater and the design parameter of organic loading is 4 Kg TOC/m$^3$-day, the 250 $m^3$ of the anaerobic tank volume could be calculated and the corresponding HRT is 0.5 day. According to this designation, higher than 90% TOC removal efficiency as well as all TMAH are decomposed to $NH_3$ and can be estimated (Fig. 5).

## IV. Conclusion

In this paper, an ATA test was conducted to understand the biodegradability of TMAH-containing wastewater and simulated TMAH-containing wastewater as well as wastewater from a full-scale TFT-LCD manufacturing company that were treated by two pilot UASB. TMAH is widely used as a developer in TFT-LCD manufacturing. This organic nitrogen compound is poisonous, corrosive, and eutrophic to the water environment. The ATA test results indicate that TMAH less than 10 000 mg/L was nontoxic and proceeded through anaerobic treatment. After anaerobic reaction, TMAH was decomposed to $CH_4$, $CO_2$ was released to the atmosphere, and $NH_3$ remained in liquid phase. The simulated TMAH-containing wastewater and wastewater from full-scale TFT-LCD manufacturing industry were treated by two pilot UASB. The volumetric loadings of these two kinds of wastewater were 6.95 kg TMAH/m$^3$-d (4.0 kg TOC/m$^3$-d) and 7.30 kg TMAH/m$^3$-d (4.1 kg TOC/m$^3$-d), respectively, as well as the removal efficiency of the two pilots was higher than 95% after 240-day operation.

The findings of this paper is important to environmental engineering since TMAH is a common widespread contaminant of water environment in TFT-LCD manufacturing countries such as Taiwan, Japan, Korea, and China. Anaerobic treatment technique such as UASB has a competitive advantage in land-limiting countries due to its high volumetric loadings. Anaerobic treatment also provides nearly complete biodegradation of TMAH, and this can prove to be effective an economic treatment option for wastewater from the TFT-LCD manufacturing industry.

TFT-LCD manufacturing is a novel industry in last decade. Chemicals used in TFT-LCD manufacturing such as TMAH are different from traditional chemical industries. COD and $BOD_5$ are two important indexes to regulate wastewater discharge in Taiwan. However, TMAH cannot be analyzed by COD method.

The result of the COD/$BOD_5$ ratio of TMAH-containing wastewater would confuse the related department of environmental protection. To prevent illegal discharge, legislative discharge limits should add a TOC limit in Taiwan for TMAH-containing wastewater.

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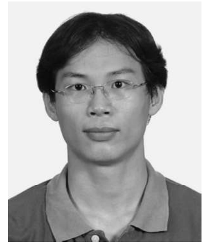

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