APPLICATION AND IMPLICATION OF ZERO-VALENT IRON FOR TRANSFORMATION OF DISINFECTION BY-PRODUCTS: A COMPARISON STUDY BETWEEN NANOSCALE PALLADIZED IRON AND MICROSCALE IRON

Hsing-Lung Lien[*]

Department of Civil and Environmental Engineering
National University of Kaohsiung
Kaohsiung 811, Taiwan

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ABSTRACT

The objectives of this study were aimed at (1) investigating the feasibility of using nanoscale Pd/Fe particles as an effective reagent for the degradation of disinfection by-products (DBPs), and (2) preliminarily evaluating the potential impact on public health risks when interaction between DBPs and cast iron pipe walls occurs in the water distribution systems. Transformation of trihalomethanes (THMs) including chloroform, bromoform, dichlorobromomethane and dibromochloromethane by nanoscale and microscale iron particles was examined. Transformation of dibromomethane was also studied. Complete dehalogenation of THMs by nanoscale Pd/Fe particles was achieved in less than one hour. Methane, accounting for 60-90% of the THMs lost, was the major product. The fast reaction rate offers a great potential for the use of nanoscale Pd/Fe particles in the treatment of DBPs. A repetitive experiment to study the long-term performance of nanoscale Pd/Fe particles indicated they lasted over 700 hours without exhausting their ability to dehalogenate THMs in a closed batch system. Because THMs readily react with iron, a significant change of levels and identity of THMs may occur in the water distribution systems comprised of cast iron pipes. Nevertheless, the preservation of drinking water with high dissolved oxygen concentrations is a practical method to stabilize the water quality during the transportation.

INTRODUCTION

Chlorine disinfection of drinking water provides a degree of public health reliability in drinking water safety and it is an essential part of drinking water treatment today. However, the disinfectants themselves can react with organic materials in the water to form unintended by-products (or so-called disinfection by-products) posing potential long-term health risks. The disinfection by-products (DBPs) currently regulated by the USEPA are two major classes: trihalomethanes (THMs) and haloacetic acids (HAAs). THMs include chloroform, bromoform, dichlorobromomethane and dibromochloromethane. HAAs, a family of 9 compounds, of which only 5 are being regulated, include chloroacetic acid, dichloroacetic acid, trichloroacetic acid, bromoacetic acid and dibromoacetic acid [1]. In Taiwan, the drinking water standards regulated only THMs and the maximum total THMs concentration was set at 100 μg/L [2].

In general, two approaches have been conducted to control the concentration of DBPs in drinking water. One is to remove chemical precursors (e.g., natural organic matter) before chlorination; the other is to remove DBPs after chlorination. Methods (e.g., advanced oxidation processes and filtration) that have been studied for the effective removal of the chemical precursors are limited [3-4], whereas advanced treatments of DBPs after chlorination have been of considerable interest including energy electron beam irradiation [5], activated carbon adsorption [5], potassium permanganate (KMnO$_4$) oxidation [6], and iron-mediated degradation [7].

The use of zero-valent iron (ZVI) for treatments of chlorinated organic contaminants represents a promising technology in groundwater remediation since Gillham and O'Hannesin revealed the granular iron metal can be used for in situ groundwater remediation [8-9]. The degradation processes involve the oxidative corrosion of iron and the subsequent reduction of the chlorinated organic compounds:

$$C_xH_yCl_z + zFe^0 + zH^+ \rightarrow C_xH_{y+z} + zCl^{-1} + zFe^{2+} \quad (1)$$
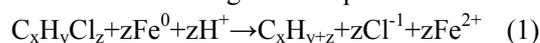
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[*] To whom all correspondence should be addressed.
　E-mail address: lien.sam@nuk.edu.tw

For example, studies have indicated that zerovalent iron readily degraded HAAs and chloroform through sequential hydrogenolysis [7, 10].

Iron nanoparticle technology, utilizing ZVI in the range of 1-100 nm as a reactive reagent for contaminant remediation, is an innovative extension of the pioneering work by Gillham et al. and many other investigators on the ZVI technology. Since 1995, work at Lehigh University has pioneered the research and development of iron nanoparticles for environmental remediation. The first synthesis of iron particles with the borohydride method was developed in 1997 [11]. Work has established that nanoscale (bimetallic) iron particles were effective reductants and catalysts for a wide variety of common environmental contaminants, especially for chlorinated organic compounds [e.g., 12-14]. Interests in the iron nanoparticle technology have been growing rapidly over last 3-4 years. Recent field tests have further demonstrated promising prospective for in situ remediation [15-16].

From the view point of water supply engineering, iron nanoparticle technology may be one of the feasible methods for effective treatments of DBPs, because DBPs are readily reduced. On the other hand, the interaction between DBPs and water pipe walls such as unlined cast iron pipes in water distribution systems has received attention [7]. According to Eq. (1), it is likely that DBPs may undergo metal-mediated reduction reactions at the pipe wall. Furthermore, because of the aging of materials, the interior surface of such pipes, typically exposed to an oxidizing environment, may result in the build-up of corrosion products mediating the reduction of DBPs. As a result, the quantity and quality of DBPs may vary in water distribution systems depending on the type and age of pipes, and retention time. The uncertainty over the identity and levels of DBPs may become an issue on the impact of drinking water quality on public health. As a consequence, there is a need to better understand the fate of these compounds when interacting with the iron pipe walls, and to develop new treatment systems for DBPs removal.

In this study, the objectives were aimed at (1) investigating the feasibility of using nanoscale Pd/Fe particles as an effective reagent for the degradation of DBPs, and (2) preliminarily examining the potential impact on public health risks when interaction between DBPs and the cast iron wall occurs in the water distribution systems. Trihalomethanes including chloroform, dichlorobromomethane, dibromochloromethane and bromoform, and dibromomethane were selected as model compounds.

EXPERIMENTAL METHODS

I. Materials and Chemicals

HPLC grade halogenated methanes were ob-tained from Aldrich. A standard gas mixture for GC analysis was obtained from Supelco. Single standard gases of 1.04% ethylene and 1.04% methane were acquired from Aldrich. Sodium borohydride ($NaBH_4$, 98%) and ferric chloride ($FeCl_3 \cdot 6H_2O$, 98%) were from Aldrich. Palladium acetate ($[Pd(C_2H_3O_2)_2]_3$) was from Alfa. Commercial grade iron with a typical particle diameter of 10 μm was obtained from Aldrich.

II. Synthesis of nanoscale Pd/Fe particles

Synthesis of nanoscale iron particles was achieved by adding 1:1 volume ratio of $NaBH_4$ (0.25 M) into $FeCl_3 \cdot 6H_2O$ (0.045 M) solution. Ferric iron was reduced by borohydride according to the following reaction [12]:

$$4Fe^{3+} + 3BH_4^- + 9H_2O \rightarrow 4Fe^0 \downarrow + 3H_2BO_3^-$$
$$+ 12H^+ + 6H_2 \uparrow \quad (2)$$
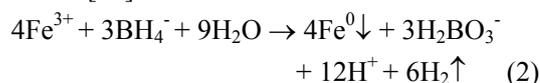
The suspension was mixed vigorously under room temperature (22±1°C). Palladized Fe particles were prepared by soaking the freshly prepared nanoscale iron particles with an ethanol solution containing 1 wt% of palladium acetate. This caused the reduction and subsequent deposition of Pd on Fe surface.

III. Batch Experiments

Five halogenated methanes including chloroform (CF), dichlorobromomethane (DCBM), dibromochloromethane (DBCM), bromoform (BF) and dibromomethane (DBM) were tested in the study. Batch experiments were conducted in 150 mL serum bottles. The metal loading of nanoscale Pd/Fe particles was 12.5 g/L in reaction with chloroform but it was reduced to 5 g/L in reaction with brominated methanes because of the high reactivity of nanoparticles. For a typical experiment, 20 μL methanol solution of a halogenated methane was spiked into an aqueous solution to a desired concentration. The serum bottles were then capped with Teflon Mininert valves and mixed on a rotary shaker (30 rpm) at room temperature (22±1°C).

IV. Analytic Methods

At selected time intervals, 20 μL headspace gas aliquot was withdrawn by a gastight syringe for GC analysis. Concentrations of halogenated compounds were measured by a HP5890 GC equipped with a DB-624 capillary column (J&W, 30 m × 0.32 mm) and an electron capture detector (ECD). Temperature conditions were programmed as follows: oven temperature at 50 °C for 5 minutes and then increased to 180 °C for 5 minutes with a rate of 20 °C/min; injection port temperature at 180 °C; and detector temperature at 300 °C. Carrier gas for GC was ultrapure nitrogen at a flow rate of 4.86 mL/min. Calibration curves for each

model compound were made initially and the variability was checked daily before analysis (<15%). Amounts of organic compounds dissolved in the aqueous phase were calculated using Henry's law.

Hydrocarbon products in the headspace were qualitatively identified with a Shimadzu QP5000 GC-MS and further quantified with GC analysis. A quadrupole mass spectrometer was set to scan from 10 to 150 m/z with data collection every 0.1 seconds. Oven temperature was set at 30 °C, injection port temperature at 150 °C and detector temperature at 230 °C. Identification of individual compounds was made by comparing the resulting mass spectral pattern with those of the National Testing and Information Service spectral library and verified with standards as well as the GC retention time. The GC equipped with a flame ionization detector (FID) and an AT-Q column (Alltech, 30 m × 0.32 mm) was conducted to quantify the hydrocarbons. Oven temperature was set at 30 °C, injection port temperature at 250 °C and detector temperature at 300 °C. The GC retention time of the sample should be within 0.1 minute of the reference standard.

V. Kinetic Analysis

Reaction rates of dehalogenation in a batch reactor are described by pseudo-first-order rate law [17]:

$$\frac{dC}{dt} = -k_a C = -k_{SA} a_s \rho_m C \tag{3}$$

Where C is the concentration of organic compound in the aqueous phase (mg/L); k_a is the measured rate constant (h^{-1}); k_{SA} is the surface-area normalized rate constant (L/h/m^2); a_s is the specific surface area of metal (m^2/g); ρ_m is the mass concentration of metal (g/L); and t is time (h). For a specific system, k_{SA}, a_s and ρ_m are constants. BET analysis gave a specific surface area of approximately 33.5 m^2/g [13].

RESULTS AND DISCUSSION

I. Transformation of Halogenated Methanes with Nanoscale Pd/Fe Particles

The results of bromoform (BF) and dibromomethane (DBM) in reaction with nanoscale Pd/Fe particles are shown in Figs. 1 and 2, respectively. The initial concentration of BF and DBM were 30 mg/L and the metal particle loading was 0.25 g in a 50 mL aqueous solution. As shown in Fig. 1, complete degradation of BF was accomplished within 0.5 hour with many end-products. Two brominated intermediates (DBM and bromomethane (BM)) and two major hydrocarbons (methane and ethane) were detected within six hours. Trace amounts of long-chain hydrocarbons such as propene and butanes were also observed. At
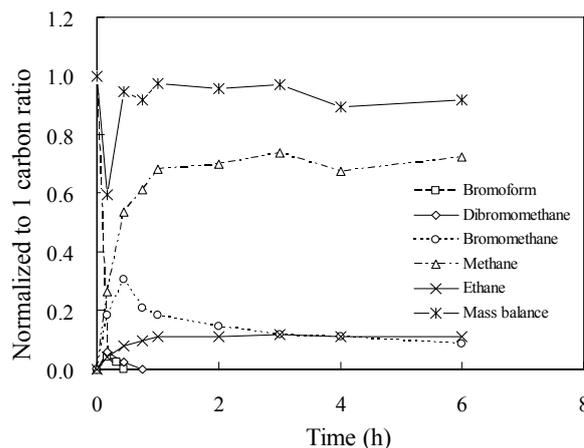

Fig. 1. Transformation of 30 mg/L of bromoform with nanoscale Pd/Fe particles. Metal to solution ratio was 5 g/L.
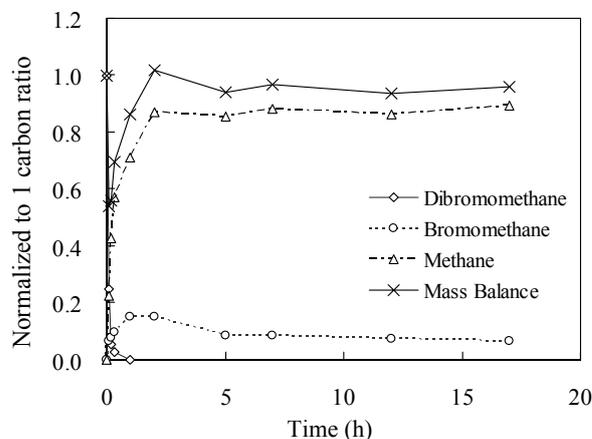

Fig. 2. Transformation of 34 mg/L of dibromomethane with nanoscale Pd/Fe particles. Metal to solution ratio was 5 g/L.

its peak concentration, BM accounted for about 30% of the BF disappearance at 30 minutes while a relatively small amount of DBM (less than 10%) was also detected. Methane was the major product, which accounted for approximately 70% of the BF lost. Ethane was minor hydrocarbon product and its concentration remained relatively constant with a yield of about 15%. Overall, the mass balance of carbon was between 85% and 100% except during the initial reaction period where the mass recovery was only 60%. This may be due to initial sorption of BF to the metal surface.

Dehalogenation of DBM with nanoscale Pd/Fe particles is shown in Fig. 2. Similar to BF, the degradation of DBM was completed in less than one hour. Methane was the major product accounting for about 80% of DBM disappearance. Only trace amounts of ethane and ethylene were detected (not shown in Fig. 2). Bromomethane was the only detectable brominated intermediate with a yield of about 15%. A similar car-

Table 1. Product distribution from the transformation of halogenated methanes with nanoscale Pd/Fe particles.

Compounds	Products
$CHBr_3$ (BF)	CH_3Br (8.2%), CH_4 (71%), C_2H_6 (11%)
CH_2Br_2 (DBM)	CH_3Br (6.2%), CH_4 (90%), C_2H_6 (1%)
$CHBr_2Cl$ (DBCM)	CH_3Br (10%), CH_4 (75%), CH_3Cl (5%), C_2H_6 (3%), CH_2BrCl (1%)
$CHBrCl_2$ (DCBM)	CH_2Cl_2 (7%), CH_4 (63%), C_2H_6 (4%), CH_2BrCl (2%)

bon mass balance as described above was observed in this case where a deficiency of carbon in the aqueous phase was found during the initial phase.

An extensive study of dehalogenation was also conducted to more halogenated methanes including dichlorobromomethane (DCBM) and dibromochloromethane (DBCM). A summary of product distributions from BF, DBM, DBCM and DCBM in reactions with nanoscale Pd/Fe particles is given in Table 1. Methane was found to be the major product with a minimum yield of 60%. Ethane appeared as a minor hydrocarbon product, which accounted from a few percent to about 12% of its parent compound disappearance. Bromomethane (BM) was the primary halogenated intermediate from reactions except for the degradation of DCBM. It accounted for about 10 to 15% of its parent compound disappearance. Formation of long-chain hydrocarbons (longer than parent compounds) such as ethane and C3 to C5 alkanes (e.g., propane, hexane) in the dehalogenation of halogenated aliphatic compounds using zero-valent iron and palladium have been widely reported [13, 18, 19]. It is believed that hydrocarbon formation is likely the result of surface-mediated reactions similar to the well-known Fischer-Tropsch process [19].

It is interesting in comparison of the product distribution between DBCM and DCBM where the halogenated intermediates may provide insights into dehalogenation reactions. As shown in Table 1, the dehalogenation of DBCM led to produce a noticeable amount of chloromethane, which is competitive to the yield of BM. On the other hand, the dehalogenation of DCBM resulted in producing noticeable amounts of dichloromethane but no further reduction to chloromethane was found. It has been demonstrated that iron has nearly no reactivity to DCM [13, 20]. The formation of chloromethane in the degradation of DBCM underwent the removal of two bromine atoms from DBCM. This suggested that the removal of one bromine atom is easier than the removal of one chlorine atom in the dehalogenation process. Consequently, it is reasonable to observe the production of chloromethane and dichloromethane from DBCM and DCBM, respectively (Eqs. (4) and (5)).

$$CHBr_2Cl\,(DBCM) \xrightarrow{H^+ + 2e^-} CH_2BrCl + Br^-$$

$$\xrightarrow{H^+ + 2e^-} CH_3Cl + 2Br^- \quad (4)$$

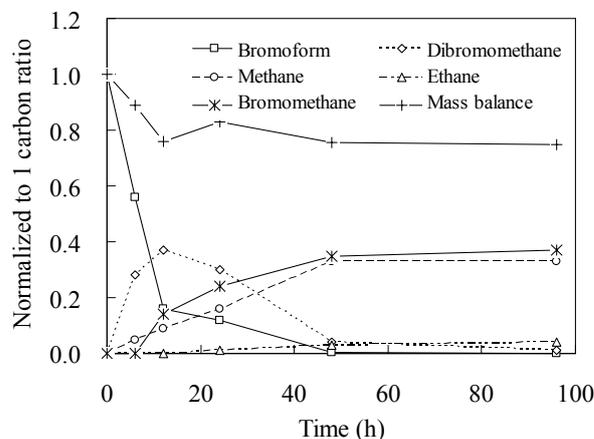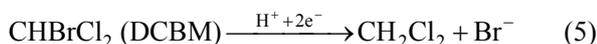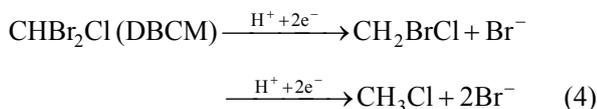$$CHBrCl_2\,(DCBM) \xrightarrow{H^+ + 2e^-} CH_2Cl_2 + Br^- \quad (5)$$

Fig. 3. Transformation of 30 mg/L of bromoform with Aldrich iron particles. Metal to solution ratio was 500 g/L.

II. Transformation of Halogenated Methanes with Microscale Iron Particles (Aldrich)

A parallel test for the degradation of BF with microscale iron particles was conducted to compare the results with the nanoscale Pd/Fe particles. Initial concentration was 30 mg/L of BF as described above. Batch experiments were carried out in a 20 mL aqueous solution in which a much higher loading of iron particles (10 g of Aldrich iron) was employed.

The results are shown in Fig. 3. In comparison with the nanoscale Pd/Fe particles, the microscale iron particles exhibit a very low dehalogenation rate. For example, complete degradation of BF was achieved within 0.5 hour using nanoscale Pd/Fe particles; however, more than 50 hours of the contact period were required to degrade BF using microscale iron particles. Moreover, dehalogenation by the Aldrich iron was incomplete, generating large amounts of lesser brominated end-products. More than 40% of BF was converted to DBM and slowly decreased to below its detection limit after 100 hours. Subsequently, a significant amount of BM (~40%) was produced. A steady concentration of BM was observed in the course of reaction. Methane was the major hydrocarbon product accounting for about 40% of the BF lost while ethane was detected as a minor product (< 10%).

This study revealed two fundamental differences between nanoscale Pd/Fe particles and commercial grade iron, including the product distribution and reaction rates. The product distributions of halogenated
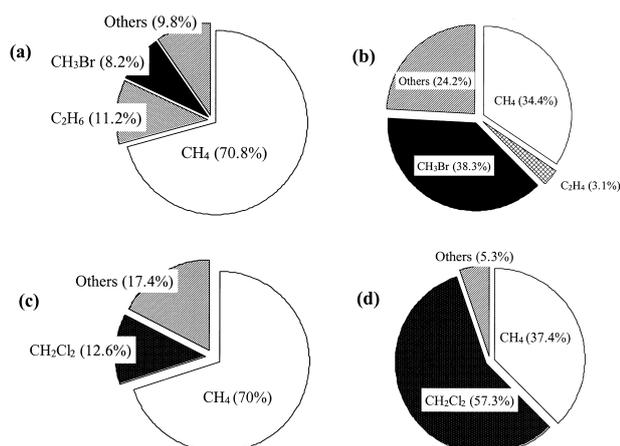
Fig. 4. Product distributions from the transformation of bromoform and chloroform with different iron. (a) Bromoform with nanoscale Pd/Fe, (b) bromoform with Aldrich iron, (c) chloroform with nanoscale Pd/Fe, and (d) chloroform with Aldrich iron.

Fig. 5. First-order plots of various THMs reaction with (a) nanoscale Pd/Fe particles and (b) Aldrich iron particles.

methanes reacting with nanoscale Pd/Fe and Aldrich Fe particles are shown in Fig. 4. Figures 4 (a) and (b) are the product distributions of BF reacting with nanoscale Pd/Fe and Aldrich Fe particles, respectively. Methane was the major product in both studies whereas the formation of BM was significantly higher in the Aldrich iron case. The production of BM increased from about 10% in the presence of nanoscale Pd/Fe particles to about 40% in the study with Aldrich iron. Moreover, instead of ethane, an incomplete dehalogenated product, ethylene, was formed by Aldrich iron. Similar results of chloroform dehalogenation are also observed as shown in Figs. 4 (c) and (d). Again, methane appeared as the major product in both studies; however, DCM production tended to overwhelm methane production in the Aldrich iron study. Because it has been demonstrated that the reactivity of DCM is very slow, significant DCM production means it accumulates in the system. This indicates that the use of nanoscale Pd/Fe particles is more beneficial than commercial grade iron particles for the treatment of THMs.

III. Rates of Dehalogenation Reactions

The rate of transformation of halogenated organic compounds in a batch system can be described by pseudo-first-order kinetics. Experimental results obtained from the product analysis were used to fit Eq. (3). Figure 5 shows the results of the best fits of the experimental data presented in Figs. 1, 2, and 3. The results of measured rate constants for the transformation of THMs using nanoscale Pd/Fe particles and Aldrich Fe particles are illustrated in Figs. 5 (a) and (b), respectively. The slopes calculated from each straight line in Fig. 5 were the measured rate constants (k_a).
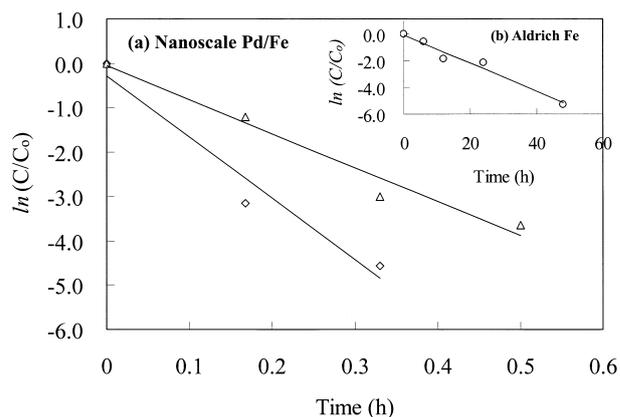
The surface-area normalized rate constants (k_{SA}) can subsequently be estimated by substituting the k_a value into Eq. (3).

The results of the kinetic analysis including the k_{SA}, k_a and half-lives are summarized in Table 2. The results of chloroform obtained from my published studies are also incorporated into Table 2 [13]. The coefficient of determination (r^2) was in the range from 0.95 to 0.99 in all experiments, indicating that the experiments were operated under consistent conditions. In general, the nanoscale Pd/Fe particles show excellent performance in the reactions with THMs. A high surface reactivity of nanoscale Pd/Fe particles was observed. The k_{SA} value of nanoscale Pd/Fe particles in the transformation of BF was over 300 times greater than that of Aldrich Fe (Table 2). Because the k_{SA} value has already taken into account the effect of surface areas, the increase of the reactivity for nanoscale Pd/Fe particles can therefore be attributed to the catalytic function of palladium as it has been found in many dehalogenation reactions [14, 18, 20].

With the high reactivity, the nanoscale Pd/Fe particles may offer a great opportunity for water treatment. A critical characteristic of water treatments is a relatively short hydraulic retention time, which typically ranges from minutes to a few hours. For example, the typical hydraulic retention times of the rapid mixing and flocculation units are about one minute and one hour, respectively [21]. The short half-life (from a few minutes to less than one hour) of dehalogenation using nanoscale Pd/Fe particles suggests that the use of nanoscale Pd/Fe particles in the removal of THMs may be a promising technology.

IV. Durability Test

This study indicats that the nanoscale Pd/Fe particles have excellent performance in terms of their high reactivity and stability in a relatively short experimental period (within a few minutes). The long-

Table 2. Dehalogenation rate constants of THMs.

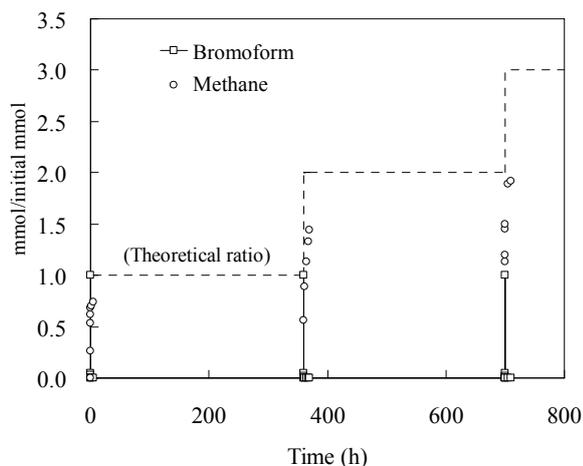|		$CHBr_3$	CH_2Br_2	$CHBr_2Cl$	$CHCl_3$
Nanoscale Pd/Fe					
	k_a (h^{-1})	13.84	7.766	9.750	2.275
	k_{SA} (L/h/m^2)	0.079±0.02	0.044	0.056±0.02	0.0065
	$t_{1/2}$ (h)	0.05	0.089	0.071	0.30
	r^2	0.951	0.966	0.989	0.958
Microscale Fe (Aldrich)					
	k_a (h^{-1})	0.107			
	k_{SA} (L/h/m^2)	2.38×10^{-4}			
	$t_{1/2}$ (h)	6.48			
	r^2	0.974			
Ratio (Nano. Pd/Fe / Aldrich Fe)		332			

Fig. 6. Repetitive additions of bromoform in reaction with nanoscale Pd/Fe particles in a closed batch system.

term performance of nanoscale Pd/Fe particles was examined by using repetitive experiments in the batch system lasting hundreds of hours in this study. A three-cycle experiment using BF as a model compound was conducted for more than 700 hours as shown in Fig. 6. No apparent decrease of reactivity was observed in the experiment. The same by-products as shown in Fig. 1 were detected, namely, dibromomethane, bromomethane, methane and ethane. Because the experiment was established under a closed batch system, the accumulation of by-products was significant after the repetitive addition of the parent compound. For example, accumulation of methane met 200% of one spiked amount of BF (molar ratio) after three cycles. The overall mass balance was about 75% of the total amount of BF added. The deficiency of mass balance may be largely attributed to the leakage of chemicals from the headspace over a long experimental time. This was further confirmed by the observation of incomplete mass recovery of BF in the blank sample where approximately 20% deficiency of mass balance was observed. It should be pointed out that although the leakage occurring under the long-term experiment was somewhat significant, it was

negligible for the experiment operated under a short time period such as the experiments for the determination of reactivity.

V. Public Health Implication of Iron Reduction with DBPs

In Taiwan, most of the water distribution systems were comprised of cast iron pipes [22]. Most of them have been used for more than decades. For example, the total service length of water distribution systems in Taipei was approximately 3242 km where cast iron pipes accounted for about 85.65% of the total length of water distribution systems [22]. Although the study did not test the reactivity for real cast iron pipes, kinetic analysis indicates that zero-valent iron readily reduces DBPs in water. For example, the half-life of BF reduction with Aldrich iron was about 6.5 hours. Because Aldrich iron is in the form of a fine powder, its reactivity is likely higher than that of cast iron pipe walls. However, the potential impact of public health resulted from the change of levels and identity of DBPs in water distribution should not be ruled out. Further investigation on the issue is needed.

Dissolved oxygen (DO) is an important parameter for water quality where the purified water should have a high DO concentration. To evaluate the effect of DO on the DBP degradation rates, experiments were conducted in which the solution was saturated with excessive oxygen by purging the solution with pure oxygen for 5 minutes. The initial BF concentration was 35 mg/L and the metal loading of nanoscale Pd/Fe particles was 0.25 g in a 50 mL aqueous solution. A slow rate of BF degradation was observed under oxygen-excessive conditions as shown in Fig. 7. More than 25% of BF still remained in the system after 60 hours. The surface-area normalized rate constant was calculated to be 8×10^{-5} L/h/m^2 under oxygen-excessive conditions. However, a rapid dehalogenation of BF, which had a k_{SA} value of 0.079 L/h/m^2, was achieved in the presence of nanoscale Pd/Fe particles under ambient conditions in the closed batch system. In comparison with the ambient conditions,
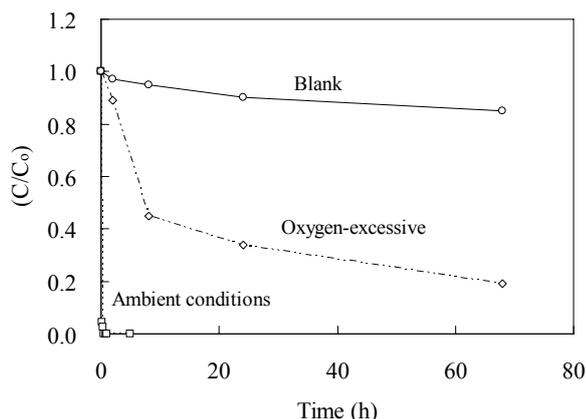
Fig. 7. Inhibition effect of dissolved oxygen in the transformation of bromoform with nanoscale Pd/Fe particles.

the reactivity of nanoscale Pd/Fe particles declined by three orders of magnitude in the oxygen-excessive conditions indicated that the excess oxygen severely inhibited the dehalogenation of THMs. The inhibitory effect of oxygen has also been observed in the study of dehalogenation of chlorinated methanes by microscale iron and palladized iron [20]. Reaction rates were reduced by at least a factor of three in the aqueous solution with oxygen. This suggests that preservation of drinking water with high DO concentrations is a practical method to stabilize the water quality during the transportation. In addition, the effect of other oxidants such chlorine on the fate and degradation rates of DBPs is worthy of further investigation.

CONCLUSIONS

In this study, the fate and reaction kinetics of DBPs in reaction with both nanoscale Pd/Fe and microscale Fe particles were investigated. An excellent performance of nanoscale Pd/Fe particles in the degradation of DBPs suggests they can serve as effective remediate reagents for DBP removal. However, a significant interaction between DBPs and microscale Fe particles implies that a potential change of levels and identity of DBPs may occur in water distribution systems if they are comprised of cast iron pipes. In particular, the following conclusions and suggestions can be drawn:
- The complete dehalogenation of DBPs by nanoscale Pd/Fe particles has been observed. It generally took less than one hour to achieve complete dehalogenation for most THMs. Methane, accounting for 60-90% of the TMHs lost, was the major product.
- Compared to nanoscale Pd/Fe particles, microscale Aldrich iron particles showed a slow reaction rate in transformation of bromoform by a factor of 300 times. Incomplete dehalogenation leading to pro-

duce less-halogenated intermediates is often observed in the microscale iron system.
- The durability test indicated nanoscale Pd/Fe particles lasted over 700 hours without exhaustion of their capacity in a closed batch system. However, excessive oxygen deteriorated the performance of nanoscale Pd/Fe particles.
- The maintenance of a high dissolved oxygen concentration in drinking water is a practical method to stabilize the water quality during the transportation where reduction of DBPs with cast iron pipe walls may be inhibited.

Although this study demonstrated the feasibility for the use of nanoscale Pd/Fe particles to effectively degrade THMs, further studies are needed before they can be applied to the treatment of drinking water. They include (i) development of a method to immobilize nanoparticles onto a supporter, (ii) evaluation of the long-term economical feasibility for nanoscale Pd/Fe particles, (iii) investigation of the fate and kinetics on other DBPs such as haloacetic acids in reaction with nanoscale Pd/Fe particles.

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零價鐵在轉換消毒副產物之應用與影響：比較奈米級鈀鐵複合金屬與微米級零價鐵之研究

連興隆[*]

國立高雄大學土木與環境工程學系

關鍵詞：消毒副產物、環境奈米技術、鐵、水處理

摘　　要

　　本研究之目的是：(1)探討利用奈米級鈀/鐵複合金屬顆粒做為降解消毒副產物之可行性，以及(2)初步評估在自來水配水系統中，當消毒副產物與鑄鐵管管壁發生反應時，對大眾健康的潛在風險衝擊。三鹵甲烷包括氯仿、溴仿、二氯一溴甲烷、二溴一氯甲烷等四種消毒副產物及二溴甲烷，被用來與奈米級和微米級鐵金屬反應並測試其轉換情形。三鹵甲烷可在 1 個小時內，被奈米級鈀/鐵複合金屬完全降解，其中 60-90%被轉換成主要產物甲烷。此一快速的反應速率，使奈米級鈀/鐵複合金屬具有處理消毒副產物之潛力。利用重覆添加進行長效性測試顯示，奈米級鈀/鐵複合金屬可在批次反應器中，持續 700 小時而不失去其降解三鹵甲烷之反應力。因為三鹵甲烷容易與零價鐵反應，當在以鑄鐵管為主之配水系統中，可能會對水中三鹵甲烷的種類與含量造成改變。然而，維持自來水中高的溶氧量，將有助於維持自來水輸送過程中水質的穩定。