

Laboratory batch experiments of the combined effects of ultrasound and air stripping in removing CCl_4 and 1,1,1-TCA from water

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Abstract

Ultrasonic and air-stripping techniques for removal of carbon tetrachloride (CCl_4) and 1,1,1-trichloroethane (1,1,1-TCA) from water were studied in batch experiments. Ultrasound (US) is effective for destroying organic compounds in aqueous solutions whereas air stripping (AS) efficiently transfers volatile compounds from the liquid to the gas phase. In simultaneous US and AS experiments, synergistic effects were observed and attributed to the effect of US on the mass transfer process. Using a photographic method, ultrasonic break up of gas bubbles and changes in gas holdup ratios were examined. In the two different gas-sparging systems studied, ultrasonic waves did not break up gas bubbles. In contrast, bubbles from the smaller porous size diffuser were coalesced due to sonication. In addition, both photographic and gas holdup experiments demonstrated that ultrasonic irradiation increased the gas holdup ratio. The enhancement observed in the removal of the compounds appeared to be due to this greater ultrasonic gas holdup ratio.

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1. Introduction

Advanced oxidation processes (AOPs) have recently received greater attention for the degradation of organic compounds in aqueous solutions [1,2]. Ultrasound is an advanced oxidation process, which has been used for a variety of different applications including plastic welding, emulsification, medical imaging, and atomization of particles [3]. Chemical effects associated with ultrasound rely on acoustic cavitation. Intense sound waves traveling through a liquid during expansion cycles produce cavitation bubbles. During compression cycles of the sound waves, the cavitation bubbles are imploded violently yielding extremely high temperatures up to

5000 K and pressures in a range of 500–1000 atm [4]. Under these extreme conditions, compounds are degraded by direct pyrolysis and hydroxyl free radical oxidation mechanisms [5,6]. Most recent work has focused on optimizing ultrasonic irradiation of aqueous solutions adjusting several parameters and combining ultrasound with other advanced oxidation processes. Both acoustic power intensity and frequency are important parameters for ultrasonic decomposition of organic compounds [7–9]. In addition, ultrasound in combination with H_2O_2 , noble gases, Fenton's reagent, ozone (O_3), or ultraviolet light (UV) can lead to more effective environmental remediation processes due to the production of high concentrations of oxidizing species such as $\bullet\text{OH}$ and localized transient high temperatures and pressures [10–13]. For example, ultrasound combined with O_3 has been shown to be especially promising for degradation of organic compounds due to effective ultrasonic decomposition of O_3 molecules to hydroxyl radicals [14,15]. Previous studies of combined process involving ultrasound and ozone have

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Nomenclature

a	specific interfacial area (cm^{-1})
A	total bubble surface area in liquid (cm^2)
AS	air stripping
C_L	compound concentration in liquid (mg/cm^3)
C^*	non-existent liquid phase concentration of compound in equilibrium with its gas phase concentration (mg/cm^3)
CCl_4	carbon tetrachloride
CV	coefficient of variation
d	gas bubble diameter (mm)
d_{BM}	mean gas bubble diameter (mm)
E	compound removal efficiency (%)
E_{AD}	additive removal efficiency by US and AS (%)
E_{AS}	compound removal efficiency by AS (%)
E_{US}	compound removal efficiency by US (%)
GC	gas chromatograph
H_2O_2	hydrogen peroxide
k_{AD}	first-order removal rate constant from additive effect of US and AS (min^{-1})
k_{AS}	air stripping removal rate constant (min^{-1})
k_{US}	ultrasonic degradation rate constant (min^{-1})
k_{USAS}	combined ultrasonic and air stripping rate constant (min^{-1})
K_L	overall liquid phase mass transfer coefficient (cm/min)
N	total number of bubbles in liquid
O_3	ozone
s	standard deviation (mm)
t	time (min)
TCA	trichloroethane
US	ultrasound
UV	ultraviolet light
V_G	volume of gas in liquid (cm^3)
V_{GM}	mean gas flow (cm^3)
V_L	volume of liquid processed (cm^3)
VOCs	volatile organic compounds

Greek letters

α	t -test confidence level (%)
ε	gas holdup ratio (vol. gas/total vol. liquid)

reported enhanced ultrasonic mass transfer of O_3 , which contributes to greater degradation rates of organic substrates [16–18]. However, there have been no comprehensive studies conducted to demonstrate the relative contribution(s) of physical ultrasonic effects to the mass transfer.

For combined US and O_3 treatment processes, Sierka and Amy [16] and Olson and Barbier [17] reported that increased mass transfer of O_3 was due to mechanical effects induced by ultrasonic waves. Olson and Barbier [17] concluded that when O_3 and US were used simultaneously, the mass transfer

of O_3 increased by 69% relative to that obtained by O_3 and US in series. In contrast, Weavers and Hoffmann [18] reported that the enhancement they observed in the mass transfer of O_3 under an ultrasonic field was due to the greater concentration gradient. Weavers and Hoffmann also pointed out that when using O_3 and US treatments simultaneously, ultrasound degraded O_3 and decreased its concentration in the liquid. As a result, a decrease in the liquid concentration of O_3 increased the concentration gradient as well as the rate of the mass transfer, as shown by Eq. (1).

In a batch reactor system and in the absence of any other reaction in the reactor, the mass transfer of a compound between liquid and gas bubbles can be expressed [19]:

$$V_L \frac{dC_L}{dt} = AK_L(C_L^* - C_L) \quad (1)$$

In the above expression, V_L (cm^3), A (cm^2), K_L (cm/min), and C_L (mg/cm^3) are the volume of liquid, the total liquid–gas bubble surface area, the overall liquid phase mass transfer coefficient, and the concentration, respectively. C_L^* represents the hypothetical liquid phase concentration of a species in equilibrium with the gas phase concentration of that species. The ratio of A/V_L ($=a$) is the specific liquid–gas bubble interfacial area [20,21]. The volume of gas bubbles (V_G) divided by the total volume of liquid ($V_G + V_L$) is the gas holdup ratio (denoted as ε). If the volume of gas in a liquid column is sufficient, the gas holdup ratio can be experimentally determined by removing holdup water to a graduated cylinder [22]. Assuming spherical bubbles, the specific interfacial area can be also expressed as a function of bubble diameter (d) and the gas holdup ratio (ε) [23,24]:

$$a = \frac{6\varepsilon}{d} \quad (2)$$

According to Eqs. (1) and (2), at a constant gas flow rate the rate of mass transfer increases with increasing gas holdup ratio and decreasing bubble diameter. If high frequency ultrasound waves break up bubbles and increase the gas holdup ratio, the specific interfacial area as well as the mass transfer will increase.

The primary objective of this study was to examine the synergy of combined ultrasound (US) and air stripping (AS) treatment in the removal of CCl_4 and 1,1,1-TCA from aqueous solutions. As with the US/ O_3 system, combining US with AS can also yield beneficial mass transfer effects of acoustic waves contributing to greater removal rates of volatile organic compounds (VOCs) from the liquid. In a combined US/ O_3 system, O_3 is transferred from the gas to the liquid phase [16–18] whereas in a coupled US/AS system, an organic compound can be also transferred from the liquid to the gas bubbles. As with US/ O_3 treatments, gas bubbles are likely influenced by intense sound waves in an US/AS treatment. Therefore ultrasonic mass transfer effects such as a decrease in the mean bubble diameter and an increase in the gas holdup ratio should be similar in both processes.

Table 1
Composition of artificial groundwater

Components	Concentration (mg/L)
Ca ²⁺	25.65
CaSO ₄ (aq)	4.12
Mg ²⁺	5.69
Na ¹⁺	9.85
K ¹⁺	7.72
H ₂ CO ₃ * (aq)	22.99
HCO ₃ ⁻	3.52
SO ₄ ²⁻	33.51
Cl ⁻	57.65

Final pH ≅ 5.5

2. Experimental

2.1. Materials and reagents

Carbon tetrachloride (99.9+%) and 1,1,1-trichloroethane (99+%) were used as received from Mallinckrodt, Inc. For all experiments, a 20 L solution of artificial groundwater was prepared by adding the following amount of reagents to deionized water: 1.20 g of MgSO₄(H₂O)₇; 0.4 g of KNO₃; 0.72 g of NaHCO₃; 0.70 g of CaCl₂; 0.70 g of Ca(NO₃)₂; 0.50 g CaSO₄(H₂O)₂; 0.62 g of NaH₂PO₄; and 7 mL of concentrated HCL. Equilibrium concentrations of species were calculated using MINTEQA2 (Table 1) and assuming the storage container of the artificial groundwater was a closed system.

2.2. Experimental apparatus

The batch ultrasonic–air stripping apparatus included a constant-temperature water bath, an ultrasonic power supply with a generator/transducer, a metallic probe, wire-mesh and fritted glass gas diffusers, and a 1 L liter glass reactor. Initial tests demonstrated that the temperature did not change during air stripping and combined ultrasonic–air stripping experiments, so the temperature control bath was not used. The 600 W ultrasonic power supply (Sonics&materials Vibra-Cell, Model VC 600 – 120 V, 10 A, 50/60 Hz) has a 20 kHz acoustic frequency, which was transmitted to the piezoelectric transducer within the converter and changed to mechanical vibrations. A 1.9 cm diameter metal probe was connected to the transducer; it intensified the mechanical vibrations to produce acoustic pressure. The probe was immersed about 2.5–3.0 cm below the liquid surface. The power intensity used in the experiments was 35 W/cm². For air stripping and the concurrent ultrasonic/air stripping experiments, a wire-mesh or fritted-glass gas diffuser was connected to lab-supplied air in a ventilation hood. Using a bubble flow meter, air-flow rates were adjusted to 500 mL/min for all experiments (Fig. 1). The glass reactor has five ports, one for the ultrasonic probe, two for inlet and outlet gas flows, one for sampling, and one for acoustic pressure control. In addition, the reactor was made airtight with an O-ring seal in the threaded Teflon

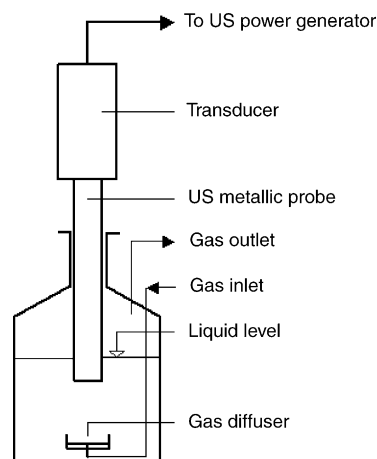


Fig. 1. Ultrasonic–air stripping cell.

collar connecting the glass cell to the stainless steel probe. All reactor ports including sampling, gas inlet, and gas outlet were sealed with Teflon valves and covered with rubber septa. The gas outlet was connected to a granular activated carbon column to absorb organic compounds in the exhaust gas from the reactor.

2.3. Experimental procedure

2.3.1. Ultrasonic and air stripping experiments

Contaminated liquid solution (500 mL) was prepared by mixing an appropriate amount of saturated CCl₄ or TCA solution with the artificial groundwater resulting in a nominal 50 mg/L initial concentration. Before an experiment was conducted, the ultrasonic power supply system was warmed up for about 20 min. The transducer of the system was tuned before each experiment to adjust the reading on the power supply monitor to the minimum value. At each 2 min sampling interval, about 1 mL of ultrasonically treated liquid sample was withdrawn from the reactor using a sampling syringe equipped with a 20 cm needle. The sample was injected in a 10 mL headspace vial, which had been sealed with crimped aluminum caps with a 10 mm Teflon-faced septum. Samples were subsequently analyzed using a Hewlett Packard gas chromatograph, Model 5890, Series II. The Hewlett Packard gas chromatograph (GC) system includes a DB-624 model capillary column with a length of 30 m. This capillary column was chosen based on its selectivity for analysis of chlorinated organic compounds. The GC was equipped with flame ionization and electron capture detectors. The flame ionization detector (FID) is easily maintained and provides sensitive detection of chlorinated hydrocarbons. Calibration curves were prepared daily before performing the experiments. The limit of detection of TCA and CCl₄ using the GC was 1.0 mg/L.

2.3.2. Photographic experiments

A Canon “Power Shot S230” model digital camera was used for photographing bubbles in a reactor containing 1 L of solution. The camera had a 0.001 s shutter speed capable

of identifying small moving objects, and was adjustable to a maximum of 2048×1536 pixel resolution to obtain high quality pictures. Before photographing bubbles in the reactor, the optical distance between the reactor and the camera was adjusted to obtain clear images of bubbles as well as to minimize optical errors. In this experimental study, optical distances in a range of 50–60 cm provided the highest quality of pictures. Maximum optical errors were estimated from photographs by measuring the difference from centimeter scales attached to the front and the back of the reactor. In the worst case, assuming all bubbles were located at the back of the cylindrical reactor with a 10 cm diameter, and the bubble sizes were measured relative to the front scale, the maximum optical error for the 50–60 cm optical distance was approximately 5%. This assessment, however, is oversimplified relative to the real experimental condition of this study, because all bubbles were mixed in the liquid rather than located at the back of the reactor. Therefore, the real optical error was less than 5%. After the optical distance between the reactor and camera was adjusted to 50–60 cm, the liquid in the glass reactor was bubbled at an air injection rate of 500 mL/min. Gas diffusers with pore size openings of approximately 1.0 mm (wire-mesh) and 0.3 mm (fritted-glass) were used in these experiments. Bubbles in the reactor were photographed before and while applying ultrasonic irradiation. Gas injection rates to the reactor were held constant at 500 mL/min. Ultrasonic power intensity used in the experiments was 35 W/cm^2 . Photographs taken of bubbles in the liquid were labeled and transferred to a computer as JPEG files.

The next steps used in photographic measurements of bubbles were to assume an appropriate shape for the bubbles and to estimate their sizes. Photographs of bubbles in the liquid reactor showed that the bubbles were generally elliptical in shape. Because a photograph shows bubbles in two dimensions, an assumption about the third dimension was still necessary to determine their sizes. Previous work suggests that bubbles in liquid can be represented by oblate spheroids [25,26]. For an oblate ellipsoidal bubble, the third dimension on the equatorial z -axis that cannot be seen from a photograph is equal to the longest dimension on the major equatorial x -axis [27]. The bubbles were scaled on photographs using the graphical design software program "Microstation SE 1997". Approximately 35–100 bubbles were measured from air stripping or air stripping-ultrasound bubble experiments when the wire-mesh gas diffuser was used to aerate the liquid. About 200–300 gas bubbles for each picture were measured when the fritted-glass gas diffuser was employed in the experiments.

2.3.3. Gas holdup experiments

Gas holdup experiments were performed in a cylindrical reactor with a 10 cm diameter and a height of 30 cm. The fritted glass diffuser was placed at the bottom of the reactor. To obtain sufficient gas holdup, 2 L of water was used in the experiments. The water height in the reactor was adjusted to

a small hole opened on the reactor surface where extra water due to gas holdup poured out. Before and during applying ultrasonic irradiation at 35 W/cm^2 , the water column was aerated at 500 mL/min. The additional water due to gas holdup was collected and measured in a graduated cylinder.

3. Results

Ultrasonic (US), air stripping (AS), and combined ultrasonic–air stripping (USAS) treatment of CCl_4 and 1,1,1-TCA from the liquid was evaluated based on the removal efficiency (E) and the pseudo-first-order rate constant (k). The combined use of US and AS could be additive, antagonistic, or synergistic. The additive effect of US and AS was defined by the following equation:

$$k_{\text{AD}} = k_{\text{US}} + k_{\text{AS}} \quad (3)$$

where k_{AD} is the additive rate constant; k_{US} and k_{AS} are the rate constants from individual US and AS experiments, respectively. In this study, if k_{USAS} is the removal rate constant during concurrent use of US and AS, the combined effects are described as follows: If $k_{\text{USAS}} = k_{\text{AD}}$, the combined effect is additive; If $k_{\text{USAS}} < k_{\text{AD}}$, the combined effect is antagonistic; If $k_{\text{USAS}} > k_{\text{AD}}$, the combined effect is synergistic.

Figs. 2 and 3 show the removal of CCl_4 and 1,1,1-TCA from liquid by US and AS. Error bars in the figures represent two standard deviations of the mean fractional remaining concentrations from three replicate experiments. At 35 W/cm^2 , about 40–45% of initial CCl_4 and 1,1,1-TCA mineralization was achieved within 10 min during sonication. For the experiments made using the wire-mesh diffuser, after 10 min approximately 70% of the initial CCl_4 or 1,1,1-TCA was removed from the liquid by air stripping at 500 mL/min (Fig. 2). For the experiments made using the fritted-glass gas diffuser, the removal of either compound from water by air stripping at 500 mL/min was greater than 90% (Fig. 3). These results

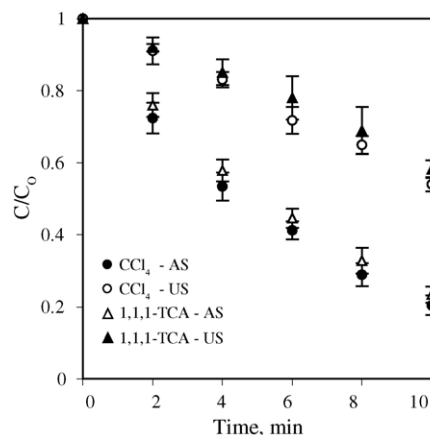


Fig. 2. Removal of CCl_4 and 1,1,1-TCA from water by ultrasound (US) and air stripping (AS) for wire-mesh gas diffuser (average bubble diameter = 3.35 mm).

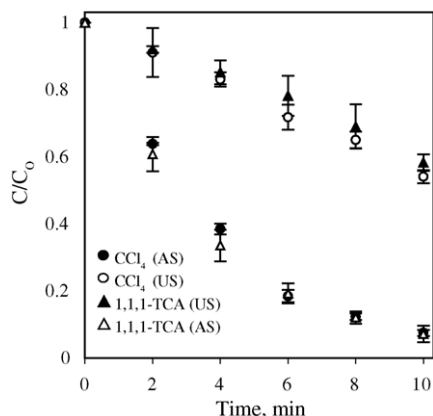


Fig. 3. Removal of CCl_4 and 1,1,1-TCA from water by ultrasound (US) and air stripping (AS) for fritted-glass gas diffuser (average bubble diameter = 1.10 mm).

suggest that the removal of volatile organic compounds from liquid increases with decreasing diffuser pore size. At a given constant airflow rate and liquid volume, the number and total interfacial area of bubbles leaving the fritted-glass diffuser with pore openings of 0.3 mm are much greater than those of bubbles leaving the wire-mesh diffuser with 1 mm pore sizes. The larger the interfacial area, the greater is the mass transfer rate between liquid and air [20,21].

The removal rate of either compounds from the solution by AS was greater than those by sonication alone. Because both CCl_4 and 1,1,1-TCA are highly volatile compounds, air stripping promotes their removal from water. It should be noted, however, that both CCl_4 and 1,1,1-TCA are degraded by US while using AS alone, they are merely transferred from the liquid into the gas bubbles. Because AS is not a destruction process, additional treatment for the off-gas will be required to prevent chlorinated hydrocarbons polluting the atmosphere and ultimately dissolving into rainwater and resulting in surface and groundwater contamination.

Results of the ultrasonic and air stripping treatments of CCl_4 were similar to those of 1,1,1-TCA. This similarity is expected because the physical and chemical properties of these compounds (such as their vapor pressure, liquid and gas phase diffusivities) are similar.

Experiments were also conducted using US and AS treatments simultaneously. Pseudo-first-order rate constants from the concurrent use of US and AS (k_{USAS}) were compared with those by additive results obtained from individual US and AS tests (k_{AD}). The k_{AD} values were equal to the summation of the first-order rate constants ($k_{\text{US}} + k_{\text{AS}}$) from US and AS experiments. The k_{US} and k_{AS} values are pseudo-first-order rate constants from US and AS tests, respectively. The ultrasonic degradation of compounds does not follow first-order rate law, because the rate changes with initial concentration. However, most studies employ pseudo-first-order rate constants to describe experimental data [10,28]. Combining ultrasound with air stripping enhanced the removal of CCl_4 and 1,1,1-TCA beyond the additive effect of US and

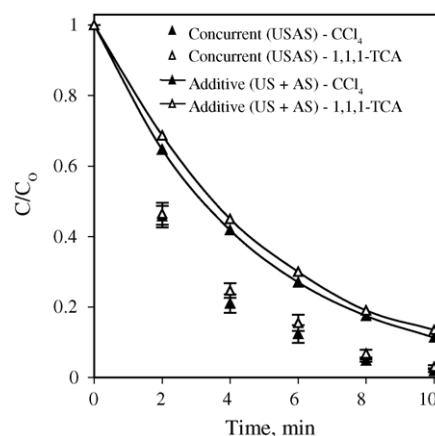


Fig. 4. Removal of CCl_4 and 1,1,1-TCA from water by combined ultrasound (US) and air stripping (AS) for wire-mesh gas diffuser (average bubble size = 3.50 mm).

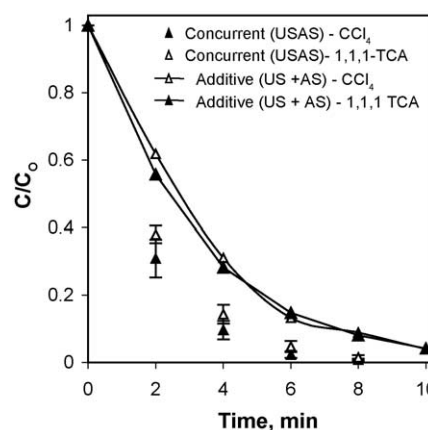


Fig. 5. Removal of CCl_4 and 1,1,1-TCA from water by combined ultrasound (US) and air stripping (AS) for fritted-glass gas diffuser (average bubble size = 1.25 mm).

AS (Figs. 4 and 5). For both diffusers used in the experiments, at 500 mL/min and 35 W/cm² the apparent enhancement in the removal rate constants relative to the additive values (k_{AD}) was greater than 80%. Experimental observations in this study indicated that the concurrent use of US and AS operates in a synergistic manner; the rate constant for the combined method was greater than the sum of the individual rate constants for US and AS separately.

4. Discussion

Photographic and gas holdup tests were performed to examine apparent synergistic effects of the combined US and AS process on the removal of CCl_4 or 1,1,1-TCA from the water. The wire-mesh and fritted-glass diffusers employed for these tests were the same diffusers used in the previous experiments.

The difference between mean bubble diameters from AS and those from combined US and AS (USAS) tests was not

Table 2
Comparisons of mean bubble sizes from AS with those from USAS bubble experiments for the wire-mesh gas diffuser

Exp. no.	Air stripping (AS)			Combined ultrasonic–air stripping (USAS)			<i>t</i> -Test (%) significance levels (α) for comparing d_{BM} by AS with these by USAS
	Number of bubbles measured (<i>N</i>)	Mean bubble size (d_{BM}) (mm)	Standard deviation (<i>s</i>) (mm)	Number of bubbles measured (<i>N</i>)	Mean bubble size (d_{BM}) (mm)	Standard deviation (<i>s</i>) (mm)	
1	37	3.39	1.03	72	3.54	1.08	>25
2	49	3.28	1.18	76	3.59	1.25	>5
3	35	3.38	1.20	100	3.57	1.25	>25
4	32	3.25	1.26	79	3.51	1.20	>15

Note: AS air flow = 500 mL/min; US power intensity = 35 W/cm²; volume of liquid = 1.0 L.

significant at the 5% confidence level when the wire-mesh diffuser was employed for the experiments (Table 2). On the contrary, results of experiments made using the fritted-glass diffuser show that mean bubble diameters from combined US and AS tests are significantly greater than these from AS tests beyond the 0.5% confidence level (Table 3). The size of diffuser openings seems to have a profound effect on the coalescence of bubbles during ultrasonic irradiation. The diameters of bubbles from the fritted-glass diffuser were the only ones to change significantly during sonication. Relative to the number of bubbles from the wire-mesh diffuser the number of bubbles leaving the fritted-glass diffuser was almost five times higher (Tables 2 and 3). As a result, chances of ultrasonic coalescing in liquid for bubbles leaving the fritted-glass diffuser should be much higher than those for bubbles leaving the wire-mesh diffuser.

Results from the bubble experiments cannot not confirm the synergistic removal of CCl₄ and 1,1,1-TCA from the water by coupled US and AS. In fact, relative to the mean bubble diameters obtained from the experiments made using the fritted-glass diffuser, there should be an antagonistic effect with the combined US and AS process. This antagonism results because the ultrasonic irradiation increases the mean gas bubble diameter. According to Eq. (2) and the mean diameters of bubbles from both AS and USAS tests, the specific interfacial area should decrease by 10% as a result of the ultrasonic bubble coalescence. For wire-mesh diffuser bubbles, no significant change in the specific interfacial area should be observed during sonication since mean bubble diameters from AS and USAS tests are similar. These conclusions, however, are only based on the effect of ultrasound on bubble sizes; they do not account for its influence on the gas holdup ratio.

Table 3
Comparisons of mean bubble sizes from AS with those from USAS bubble experiments for the fritted-glass gas diffuser

Exp. no.	Air stripping (AS)			Combined ultrasonic–air stripping (USAS)			<i>t</i> -Test (%) significance level (α) for comparing d_{BM} by AS with these by USAS
	Number of bubbles measured (<i>N</i>)	Mean bubble size (d_{BM}) (mm)	Standard deviation (<i>s</i>) (mm)	Number of bubbles measured (<i>N</i>)	Mean bubble size (d_{BM}) (mm)	Standard deviation (<i>s</i>) (mm)	
1	200	1.134	0.293	278	1.273	0.363	<0.05
2	182	1.140	0.299	181	1.230	0.327	<0.5
3	179	1.081	0.265	161	1.184	0.330	<0.5

Note: AS air flow rate = 500 mL/min; US power intensity = 35 W/cm²; volume of liquid = 1.0 L.

Table 4
Comparison of gas holdup ratios (ϵ) from AS and USAS tests made using the fritted-glass diffuser

	Air stripping (AS)	Combined ultrasonic and air stripping (USAS)
Number of experiments	5	5
Mean ϵ values	0.0046	0.0083
Standard deviation	0.00025	0.00035
CV (%)	5.43	4.22

Airflow rate = 500 mL/min, power intensity = 35 W/cm², volume of liquid = 2.0 L.

The influence of ultrasound on gas holdup ratio was initially determined from photographs taken of AS and USAS bubble tests. If ultrasound had no effect on the bubbles leaving the wire-mesh diffuser, their number from both AS and USAS tests at the constant airflow rate would be similar. However, the number of gas bubbles in the liquid is significantly increased during sonication at 35 W/cm² (Table 2). The gas holdup ratio should be also increased by the ultrasonic effect, because the mean bubble diameters from AS tests do not significantly differ from those from USAS tests.

Although the photographic method was useful for showing an ultrasonic effect on gas holdup for the bubbles of wire-mesh diffuser, its use was limited during analysis of bubbles from the fritted-glass diffuser. The higher number of bubbles in the liquid increases the chances of blocking the view of many others remaining at the back of the reactor. Therefore, it was impossible to make an estimation of the gas holdup by the photographic method when the fritted-glass diffuser was used in the experiments. The gas holdup ratios were determined by simply removing airlifted water into a graduated cylinder (Table 4). At 500 mL/min and 35 W/cm²,

the ultrasonic irradiation of the liquid increased the gas holdup by a factor of 2, which is quite similar with that predicted from the photographs taken of wire-mesh diffuser bubbles. An explanation for the ultrasonic effect on gas holdup ratios might be that when US and AS processes are operated in a countercurrent mode, gas bubbles are oscillated under the field of intense sound waves. As a result, they have longer retention times in the reactor. The relationship between bubble retention time and gas holdup volume can be described by the following equation [20–23]:

$$\tau = \frac{V_G}{Q_G} \quad (5)$$

where τ , V_G , and Q_G are bubble retention time, gas holdup volume, and gas flow rate, respectively. From Eq. (5), at a given constant gas flow rate and liquid volume; gas holdup volume increases with increasing bubble retention time.

With about 10% and 100% increases in the mean equivalent bubble diameter and the gas holdup ratio, respectively, by ultrasonic effects, Eq. (2) suggests a 90% overall increase in the specific bubble interfacial area for the fritted-glass diffuser. This ultrasonic enhancement in the specific interfacial area confirms the synergistic effect of combined US and AS treatment on the removal of CCl_4 and 1,1,1-TCA from the liquid.

5. Conclusions

Simultaneous combination of ultrasound (US) and air stripping (AS) behaves in a synergistic manner providing greater removal rates of CCl_4 and 1,1,1-TCA from the liquid than by either process alone. In the coupled US and AS system, pseudo-first-order rate constants for the removal rate constants of both compounds from water increase by more than 80% relative to the overall removal rate constants by individual US and AS experiments.

The combined process involves both antagonistic and synergistic mass transfer effects. An antagonistic effect of combined US and AS on the mass transfer process can occur due to the coalescence of gas bubbles during sonication. For example, when bubbles of the fritted-glass diffuser are ultrasonically coalesced, their total surface area relatively decreases leading to an antagonistic effect on the ultrasonic–air stripping removal of organic compounds from water. Photographs taken during experiments with the wire-mesh diffuser show no evidence of the ultrasonic break up or coalescence of gas bubbles.

The effect of ultrasound on gas holdup ratio was also studied to confirm the synergistic effect of combined US and AS method on the mass transfer. Gas holdup ratios were increased during sonication when either diffuser was used. The synergistic removal of the compounds from the water by combined US and AS can be attributed to this greater gas holdup ratio. For both diffusers used in the experiments, the influence of ultrasound on gas holdup ratio appears to be similar.

Because ultrasonic irradiation does not break up gas bubbles, an effective way for improving removal efficiencies of organic compounds by coupled US and AS treatment process is to increase the gas holdup ratio. This increase might be even achieved more effectively by using higher ultrasonic power intensities. Further experimental work should explore the efficiency of the combined US and AS treatment at higher ultrasonic power intensities.

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