

REMEDICATION AND DISINFECTION OF WATER USING JET GENERATED CAVITATION

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ABSTRACT

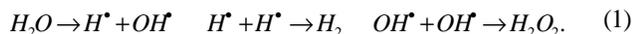
Cavitation generated by DYNAJET[®] cavitating jets can greatly enhance various chemical reactions, oxidize aqueous compounds, and disrupt microorganisms. Controlled laboratory experiments have established the ability of these jets to destroy microorganisms and oxidize a broad range of compounds including pesticides, Volatile Organic Compounds, arsenic, and reduce TOC (Total Organic Carbon).

Experiments were conducted using a variety of cavitating jet configurations and operating conditions. Histories of the contaminant concentration reductions and destruction of selected bacteria strains were obtained from periodic sampling. Multiple order of magnitude reduction in e. coli concentration has been demonstrated. Comparisons with ultrasonic experiments show that jet-induced cavitation can achieve oxidation with up to two orders of magnitude greater energy efficiency.

INTRODUCTION

Ultrasonic cavitation is known to produce sonochemically activated reactions in water resulting in the formation of highly effective oxidizing hydroxyl radicals [1-4]. Usually this is achieved using ultrasonic horns that send a high intensity acoustic beam into the solution and excite microcavities. Such systems have been found to promote a wide range of chemical reactions can be promoted [2] and dilute aqueous mixtures of organic compounds can be oxidized [5-10]. However, such devices essentially self limit the efficiency of the process by achieving cavitation only in a thin layer near the surface of the sonifier. We generate cavitation in a wide body of the liquid by an array of submerged cavitating jets [11-14]. This process can be made very efficient and benefits from the relatively high efficiencies of pumps.

When subjected to cavitation, water undergoes dissolution according to the following chemical reaction [3,4]:



The free hydroxyl radical OH^\bullet is one of the most powerful oxidizing agents. Oxidation of organic compounds results in various intermediate and end products depending on the compound. These include water vapor, carbon dioxide, inorganic ions and short chain inorganic acids (e.g., [2,5,15]). Intermediate products usually undergo subsequent oxidation.

It is generally accepted that water dissociates under intense insonification *due to the resulting growth and collapse of microscopic bubbles*. It has been estimated that the pressure may reach as high as *high as 1.2×10^4 atm, and the temperature could reach 10,000 °K* [16] in a small region of space where the bubble collapse occurs. Such conditions could explain the enhancement by cavitation of the chemical dissociation of the aqueous medium releasing hydroxyl radicals [17,18].

A number of recent studies have indicated the potential to destroy microorganisms in drinking water with ultrasonically generated cavitation, usually in conjunction with another microcide such as chlorine, ozone, or catalysts [19-21]. Ultrasound was found to reduce bacteria sufficiently to decrease the amount of chlorine required for disinfection [20] and to deactivate bacteria subjected to long duration and moderate levels of exposure [20,22-24].

The DYNAJET[®] cavitating water jet technology represents one successful attempt to harness and utilize the destructive power of cavitation. Various means and nozzle designs can be used to induce the explosive growth of microscopic cavities or bubbles within a liquid jet. Moving away from the orifice region, these bubbles encounter higher pressures and collapse [25,26]. In the STRATOJET[®] [25,26], we incorporate passive acoustic resonance in the flow, which structures the vortices in the turbulent jet [27]. This has proven to generate substantially more cavitation than the non-resonating cavitating jets in submerged cleaning and cutting applications. A strobe photograph of the ring shaped cavities generated by a STRATOJET[®] is provided on the left of Figure 1.

The swirling cavitating jet, DYNASWIRL[®] achieves early cavitation using a vortex flow. The cavitation starts inside the nozzle assembly and extends in front of the nozzle orifice (Figure 1). The flow enters a swirl chamber by means of tangential injection slots. The swirl produces a *central line vortex that cavitates*. As the feed pressure is increased, the filament along the jet axis attains a helicoidal shape with a vortex breakdown at the end of a rotating cork screw shape in some configurations. If the jet impacts on a nearby solid surface, the cavities so generated will collapse in the high pressure stagnation region of the jet. This jet has the additional advantage of generating a very large cavity volume and surface area.

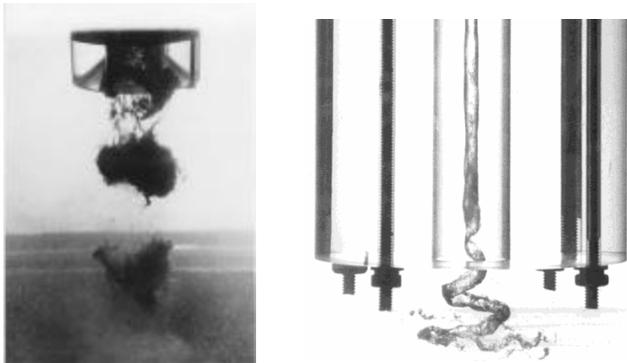


Figure 1. Strobe Visualizations of two classes of DYNAJETs[®] cavitating jets. Left: large scale STRATOJET[®] with ring shaped cavities (dark areas) downstream of the nozzle. Right: Plexiglas walled DYNASWIRL[®] swirling cavitating jet nozzle with central line vortex cavity.

The dimensionless parameter characterizing cavitation is the cavitation number, σ ,

$$s = \frac{P_{amb} - P_v}{rV^*^2/2} = \frac{P_{amb} - P_v}{\Delta P} \quad (2)$$

where r is the liquid density, V^* is the mean jet velocity, and ΔP is the pressure drop across the nozzle. The particular value at which cavitation is incipient is defined as

$$s_i = \left[\frac{P_{amb} - P_v}{\Delta P} \right]_{\text{at inception}} \quad (3)$$

Thus if the operating conditions for a submerged jet are such that $s/s_i < 1$, cavitation will occur, and as s/s_i continues to decrease below unity the amount of cavitation will increase.

Our DYNASWIRL[®] cavitating jets exhibit an increase in s_i over conventional jets by a factor of up to 10 with current designs. This means that at a given ambient pressure, *cavitation can be generated with only 1/10 as large a ΔP . For higher pressures, cavitation will intensify and the amount of fluid subject to cavitation will increase.* This is due to the presence of a strong vortex of circulation Γ and a viscous core of radius r_c . The value of the minimum pressure on the vortex axis, P_{min} , is estimated as [26]:

$$P_{min} = P_a - \frac{r \Gamma^2}{4\rho^2 r_c^2} \quad (4)$$

In order to compare the decreases in chemical or microorganism concentration of systems of different volumes and flow rates, we non-dimensionalize the time by the mean residence time of a particle of fluid in one cycle of the loop such that the non-dimensional time, t^* , is:

$$t^* = tQ / V = N_{cycles} \quad (5)$$

Here, V is the system volume and Q is the flow rate through the loop. The non-dimensional time t^* can be viewed as the number of cycles through the loop of exposure to cavitation that a fluid particle has undergone (“*cavitation cycles*”).

EXPERIMENTAL SETUP

Controlled laboratory experiments were conducted using various jet oxidation and microorganism removal flow loops. All are fabricated primarily of stainless steel with stainless steel or Plexiglas reaction chambers and are driven by centrifugal or positive displacement pumps at moderate pressures and flow rates through the DYNAJET[®] nozzles specially designed to cavitate. Both classes of cavitating jet nozzles shown in Figure 1 were employed. The STRATOJET[®] style of nozzle orifice was utilized in a multi-orifice configuration. Figure 2 provides a sketch of a loop utilized for microorganism destruction experiments.

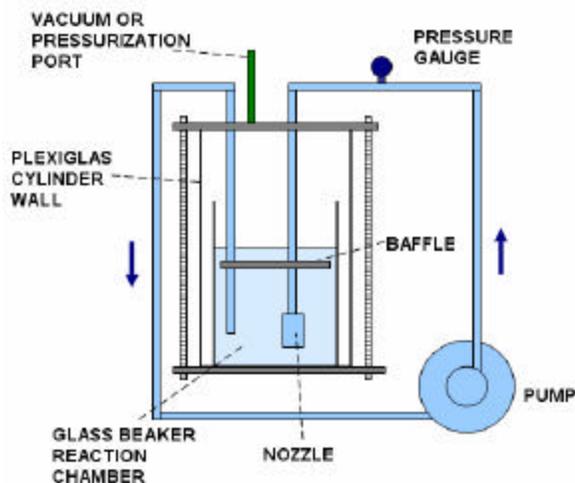


Figure 2. Sketch of laboratory flow loop designed for microorganism destruction investigations. All components except the pump can be rapidly disassembled to be disinfected in an autoclave.

In each loop, the liquid flows into a cavitation reaction chamber into which various DYNAJET[®] jet configurations are inserted. The pressure of the flow entering the chamber to form the jet(s) and the ambient pressure at the jet exit are monitored and controlled. Temperature is maintained at the desired value by use of a cooling loop when required. The pH is monitored and controlled when desired. Sampling ports are located in the lid or exit pipe of each reaction chamber.

For microorganism destruction investigations we have specially designed a small volume (~1.5 liters) recirculating test loop. The volume has been kept small to facilitate thorough disinfection between runs. A sketch is provided in Figure 2. All components except the pump can be disassembled and placed in

an Autoclave for disinfection after each test. The pump was disinfected with chlorine bleach solution and then flushed repeatedly with distilled water. Disinfection of the system was confirmed by plating samples of the rinse water to assure that there were no measurable viable colonies. Sufficient flushing of the bleach was confirmed by introducing bacteria into the system following flushing and monitoring their growth in comparison with a control group.

Chemical contaminant removal was measured by periodic sampling and measurement of concentrations resulting in curves of concentration vs. time. A given contaminant will, in general, be initially degraded into intermediate compounds, prior to complete oxidation of the constituents to carbon dioxide, water vapor, and various simple inorganic compounds. We employed several means of evaluation of contaminant destruction. Oxidation of organic constituents and measurement of TOC (Total Organic Carbon) were determined utilizing reagent kits and a UV-Vis Spectrophotometer.

For the microorganism destruction experiments, we utilized a non-pathogenic strain of *E. coli* (MC4100). Sterile media is inoculated with the *E. coli*, which was then grown as a batch culture until reaching the desired concentration at which time it was introduced into the test loop. A portion of the inoculated media was maintained outside the test loop during the test to serve as a control.

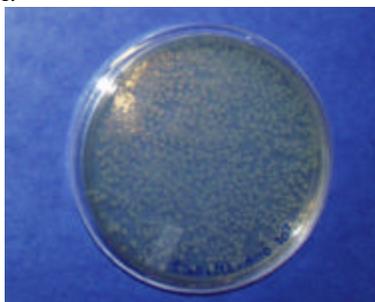


Figure 3. Photograph of viable *E. Coli* (MC4100) colonies obtained by plating (here after a 10^4 dilution).

The liquid was then circulated through the test loop and the cavitating jet nozzle. Initial and periodic samples are taken and the bacteria concentration measured by plating on agar with successive dilutions and then incubating overnight. Figure 3 provides an example photograph of such a plate. The resulting colonies formed are then counted. Duplicate plates were used at each sample time. Curves of bacterial concentration versus time were then created and compared with control and initial values.

CHEMICAL CONTAMINANT RESULTS & DISCUSSION

Oxidation experiments have been conducted on aqueous solutions of a wide variety of compounds. A selected illustrative subset is presented here.

Influence of Jet Pressure

Results of oxidation of the herbicide 2,4-D obtained from a commercially available pesticide product are presented in Figure 4. Solutions of approximately 400 ppb 2,4-D initial concentration were subjected to cavitation generated by DYNASWIRL[®]

cavitating jets with oxygenation at two different pressures (while other variables were held constant) are presented. Both concentrations normalized on the initial concentration versus dimensionless time and the mass of 2,4-D removed as a function of energy expended are presented. The multiple data points at the same time for a given case represent the results of multiple measurements for a given experiment and can be used as a measure of measurement uncertainty.

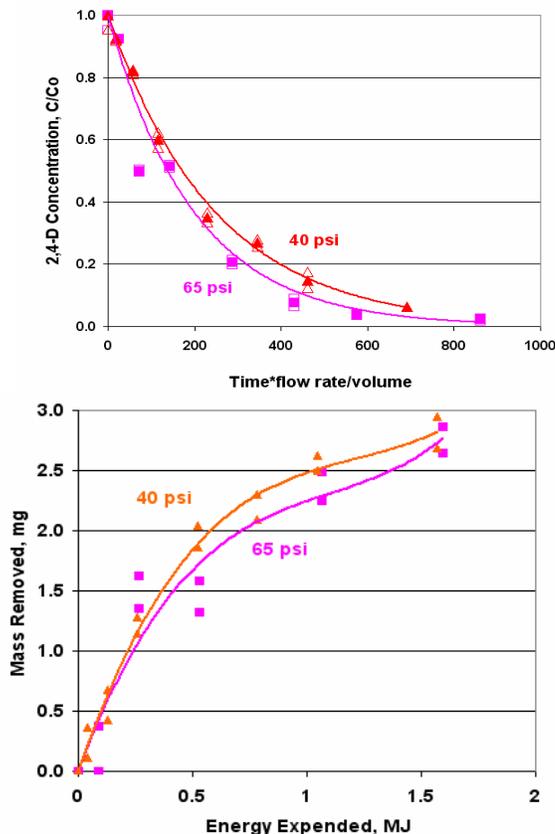


Figure 4. Oxidation of 2,4-D: comparison of effects of different nozzle pressure conditions. Top: Normalized concentration histories. Bottom: Mass of 2,4-D removed versus energy expended.

Substantial reductions in 2,4-D concentrations can be seen for both cases with an order of magnitude reduction achieved after about 450 cavitation cycles ($t^*=450$). Comparison of the two cases operated at different pressures (40 psi and 65 psi) shows a faster reduction of C/C_0 at the higher pressure. However, the efficiency of operation appears to favor the lower pressure as can be seen in the lower plot of Figure 4.

The influence of the jet pressure is further illustrated in Figure 5. Here, solutions containing Methyl Orange, a compound similar to various detergents, were subjected to cavitation by the swirling cavitating jet at pressure drops across the nozzle varying between 15 and 80 psi. Presented in this figure are the plots of concentration versus time and energy efficiency versus time. The efficiency is calculated by simply taking the ratio of the cumulative mass of Methyl Orange removed to the cumulative input hydraulic energy. As can be seen, the continuously

increases with decreasing pressure drop with the highest efficiencies obtained with a pressure drop of only 15 psi, while the most rapid removal is found at a pressure of 60 psi.

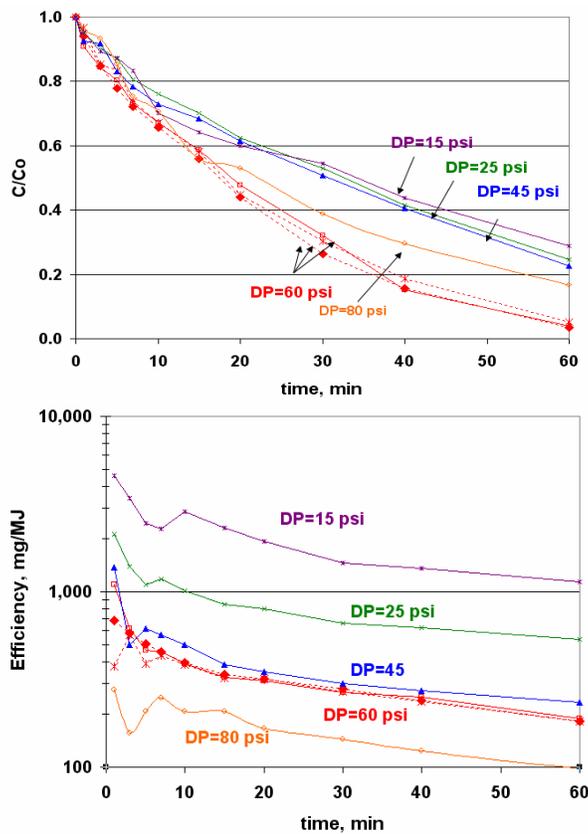


Figure 5. Influence of nozzle pressure drop on oxidation of Methyl Orange with DYNASWIRL[®] cavitating jet. Top: concentration histories. Bottom: Efficiencies (mass removed/hydraulic energy input).

Influence of Batch Volume

As part of a parametric study on jet oxidation performance, we have investigated the influence of the total batch volume in the loop on the energy required to achieve a given level of reduction. This is illustrated in Figure 6 in which the total batch volume in the test loop was systematically varied between 1.8 and 22.7 liters – a factor of approximately 12 – for Methyl Orange recirculated through an array of 8 STRATOJET[®] type orifices. All other conditions were held constant. Figure 6 presents the cumulative hydraulic energy input required to achieve an on order of magnitude reduction in the concentration of Methyl Orange for different volumes. This energy is normalized on that for the smallest volume (1.8 liters). As can be seen, the required energy continuously drops until reaching a broad minimum between 10 and 15 liters, and then begins to rapidly rise. There is a factor of 4 variation between the minimum and maximum required energies – a significant amount.

This variation is probably related to time constants associated with the bubble nuclei, the mixing, and the reacting

species. The optimum occurs when the cycle time matches this time constant.

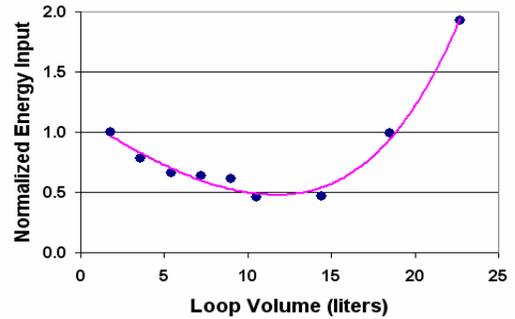


Figure 6. Influence of loop batch volume on oxidation of Methyl Orange with an array of STRATOJET[®] cavitating jet orifices. Shown are the energies required to achieve $C/C_0 = 0.1$ normalized on that for the smallest total volume (1.8 liters).

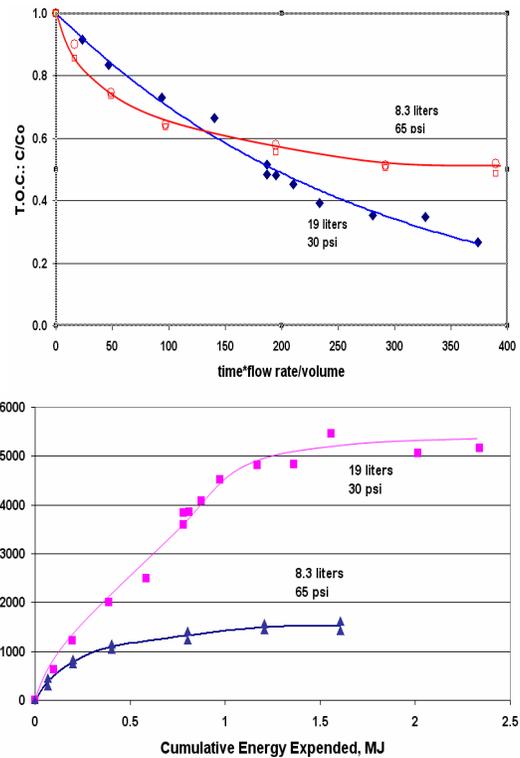


Figure 7. Oxidation of Malathion (as indicated by TOC measurements) with two different DYNAJET[®] swirling cavitating jet operating pressures and loop volumes.

This volume effect was also observed in oxidation of the insecticide Malathion as can be seen in Figure 7 in which Malathion removal is indicated by TOC reduction. Aqueous solutions of Malathion were prepared as described above using a commercial product as the source of Malathion.

Results are presented for two different experimental cases with the dual orifice swirling cavitating jet. One experiment was conducted at a pressure of 30 psi in a loop with a total volume of 19 liters, while the second was conducted at 65 psi and a smaller loop volume of 8.3 liters. The TOC level is cut by approximately a

factor of 4 for the 30 psi/19 liter case and a by factor of two for the 65psi/8.3 liter case within $t^* \sim 380$. This effect is more readily seen in the plot of cumulative TOC mass reduction versus hydraulic energy expended. As can be seen, despite operation at a lower pressure, the larger volume case has a total TOC mass reduction approximately three times larger than the smaller volume case.

Oxidation of Other Substances

A variety of different substances of varying chemical compositions and classes have been successfully oxidized with the DYNAJET® cavitating jets. A brief summary of some of these data are presented here.

Aqueous solutions of the herbicide Alachlor were prepared using a commercial product. Figure 8 presents the results of an experiment utilizing the swirling cavitating jet at 65 psi. A 2/3 reduction in concentration is achieved at a nondimensional time $t^* = 500$.

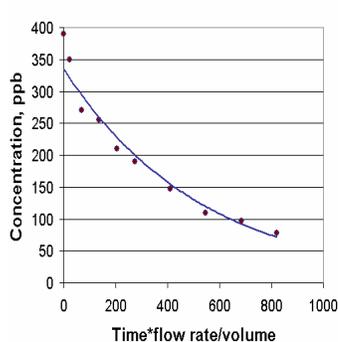


Figure 8. Oxidation of Alachlor with DYNAJET swirling cavitating jet at 65 psi, pH=3, Volume=8.3 liters

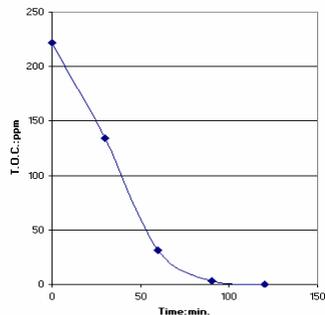


Figure 9. TOC reduction in algae infested pool water with a DynaJet swirling cavitating jet at 30 psi, no pH control, no oxygenation.

Samples of swimming pool water that had been left untreated over the winter and had developed a significant amount of algae were exposed to the swirling jet nozzle. Figure 9 presents results that show a reduction in TOC from 220 ppm to 30 ppm in 1 hour and to less than 1 ppm in an additional hour..

Figure 10 presents results of oxidation of an aqueous solution of approximately 10 ppm Chlorhexidine, a biocide. (This work was done in conjunction with Dr. Gregory Moller of the University of Idaho. In a cooperative effort, his laboratory prepared samples of water containing chlorhexidine and evaluated the results by Liquid and Gas Chromatography and Mass Spectroscopy. This mixture was exposed to the swirling cavitating jet at 30 psi. The results in Figure 10 show a decrease from 11 ppm to 5 ppm in 15 minutes and a further reduction to 2 ppm in a total of one hour.

Figure 11 presents the results of oxidation of Chloroform, a Volatile Organic Compound (VOC) that is a groundwater contamination concern. Experiments were at a jet pressure of 60 psi run with initial concentrations ranging from 40 to 150 ppm of

TOC. No influence of concentration when normalized on initial concentration is seen. All cases are seen to rapidly reduce the TOC concentration.

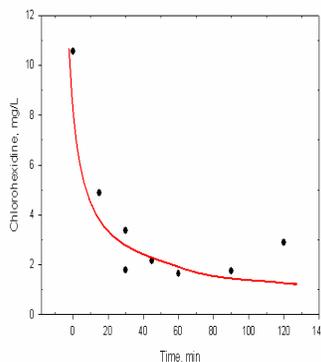


Figure 10. Oxidation of Chlorhexidine with a DYNAJET swirling cavitating jet measured by Gas Chromatography/Mass Spectrometry.

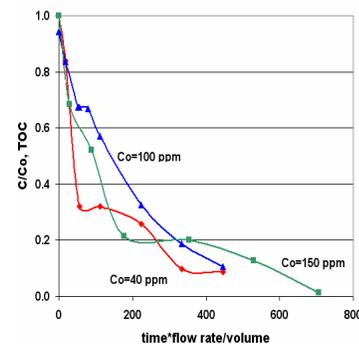


Figure 11. Oxidation of Chloroform in water with multi-orifice STRATOJET and swirling cavitating jets at 60 psi.

Alachlor and 2,4-D Mixtures Oxidation Experiments

Experiments were conducted to determine the effect of having mixtures of compounds. A mixture of Alachlor and 2,4-D was prepared utilizing the commercial products noted above with nominal initial concentrations of 400 ppb Alachlor and 500 ppb 2,4-D. This mixture was subjected to cavitation utilizing the swirling jet at 65 psi. Figure 12 presents the results for this experiment and compares these to the results of experiments run under identical conditions for solutions of each of the compounds Alachlor and 2,4-D alone.

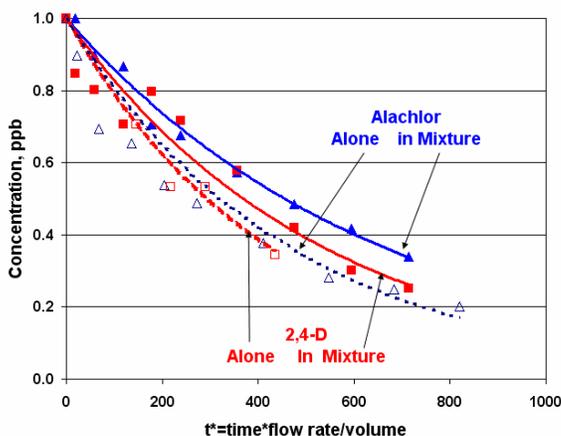


Figure 12. Oxidation of mixture of Alachlor (400 ppb) and 2,4-D (500 ppb) subjected to swirling cavitating jet at 65 psi compared to oxidation of solutions of each individual compound.

The data clearly show that the concentration reduction of each compound in the mixture is very comparable to that of the individual compound alone. The alachlor does exhibit about 1/3

less concentration reduction for a given nondimensional time in the mixture than when it is by itself. The 2,4-D concentration also decays somewhat more rapidly when by itself than when in the mixture. However, the differences are not substantial. These data strongly indicate that the presence of each compound is interfering very little with the oxidation of the other compound with each behaving nearly as if the other were not present. This suggests that, at least at these relatively dilute concentrations, the process reaction rate is being determined less by the supply of hydroxyl (or other) oxidizing radicals than by the transport and mixing of the oxidizing radicals and the target contaminants. Note also that these results show that the total mass of contaminant removed (the sum of the masses removed for Alachlor and 2,4-D) for the mixture is about *twice that for either compound run individually*.

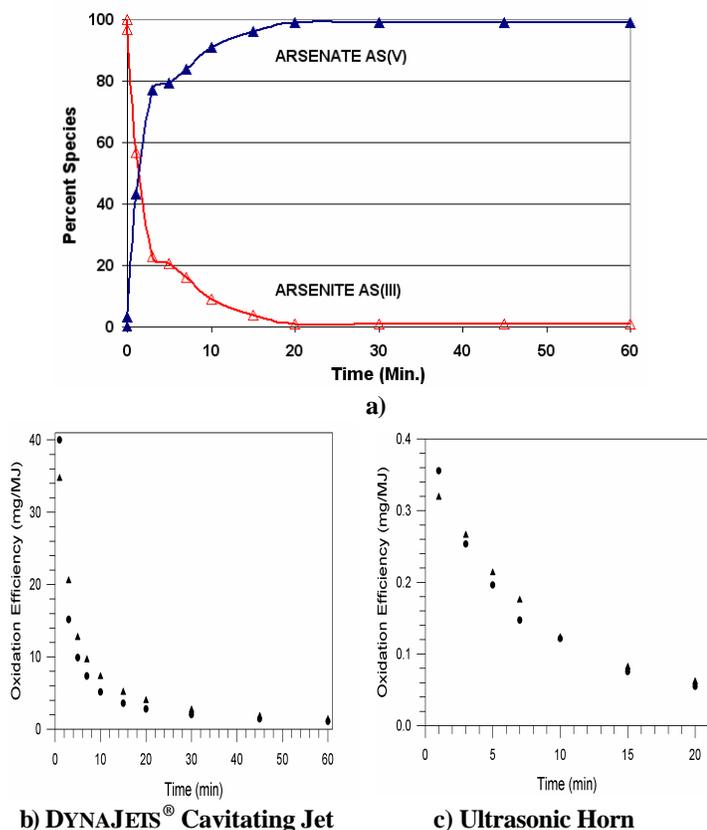


Figure 13. Results of DYNAJET® oxidation tests of arsenic in water. Initial concentration: 100 ppb AS(III). a) Speciation evaluation for different exposure times. b) & c) Efficiencies of the DYNAJET® and a 20 kHz ultrasonic horn showing two order of magnitude advantage for the DYNAJET® jet generated cavitation.

Oxidation of Arsenic

Samples of water containing 100 ppb of arsenic AS(III) were obtained from the U. of Idaho and were subjected to DYNAJET® cavitation. Figure 13a shows the rapid oxidation of arsenite, AS(III), to arsenate, AS(V). Significant amounts of oxidation are

found to be produced: 44% of the AS(III) was oxidized to AS(V) in 1 minute of operation, 79% in 5 minutes, and 99% in 20 minutes. The chemical analysis was performed at the U. of Idaho. For the speciation work a Dionex DX-120 Ion Chromatograph was used to separate the species on a PRP-X100 column with 50 mM PO4-/PO4-2 eluent, utilizing the HP 4500+ ICP-MS as the detector [28].

Figures 13b and c compare the oxidation efficiencies of the DYNAJET® data with those from experiments conducted at the U. Idaho utilizing a 20 kHz ultrasonic horn [28]. The oxidation efficiencies are expressed as the mass of arsenic oxidized per unit energy utilized. As can be seen, oxidation utilizing DYNAJETS® was *two orders of magnitude more efficient* than the ultrasonic horn in terms of energy required for oxidation of As. The drop in the efficiency with time for both systems is attributed to the decrease in amount of As(III) available for oxidation in a batch reactor.

MICROCIDAL RESULTS & DISCUSSION

A number of experiments were conducted with E. coli in M9 at an initial concentration between 5×10^8 and 2×10^9 CFU/ml. Such a concentration is near saturation for the E. coli and presents a worst case scenario of the most such bacteria that could be encountered.

Figure 14 summarizes the results of these experiments. As can be seen in this figure, up to 5 \log_{10} reductions in bacteria concentrations were achieved with the majority of these reductions ($\sim 3 \log_{10}$) found to occur in the first 20 to 40 min. The bottom plot of Figure 14 presents the normalized concentration reduction versus the hydraulic energy expended.

The data presented in Figure 14 are from experiments conducted in two different pressure ranges: 60-75 psi and 150 psi. All cases are seen to produce rapid decrease in bacteria concentrations. Operation at 60-75 psi can be seen to be more energy efficient than operation at the higher pressure for achievement of a 3 \log_{10} reduction. However, operation at 150 psi, though initially less efficient, is found to result in a greater reduction in bacteria concentration, 5 \log_{10} .

To assure that the observed bacteria destruction was due to exposure to the jet cavitation, experiments were performed with the jet nozzle removed. Figure 15 presents results of such an experiment run without an orifice (and thus without the source of cavitation) which exhibits only a minor reduction in bacteria $\sim 0.6 \log_{10}$ (perhaps due to shear in the pump). This is compared to experiments run in the same loop with two different cavitating jet nozzles producing three order reduction in bacteria concentration. This enables us to conclude that *the bacteria reductions observed in the cavitating jet experiments are due to the cavitation generated*. Also shown on this figure are the results of a control sample taken from the same batch utilized for the flow loop, but kept in the shaker water bath. As can be seen, the bacteria concentration actually increases moderately in the control sample, which would be expected as nothing is impeding the growth of bacteria.

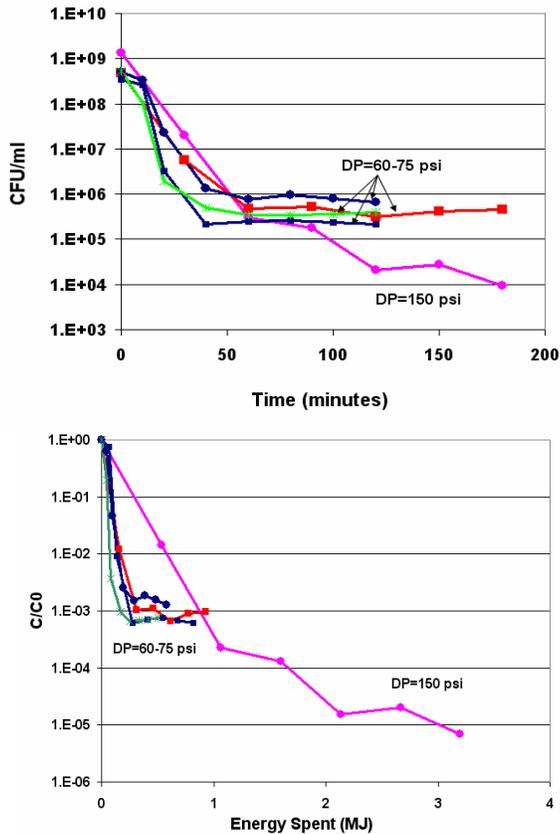


Figure 14. Summary of results of cavitating jet reduction of E. coli in M9 starting at high concentrations (~ 10^9 CFU/ml). Top: Raw data of bacteria count vs. time. Bottom: Normalized bacteria concentration vs. hydraulic energy expended.

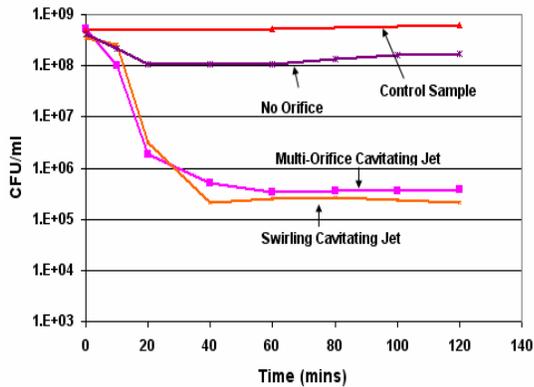


Figure 15. Results of microorganism reduction experiments with E. coli MC4100 in M9 media subjected to cavitation generated by DYNAJETS cavitating jets operating at 60 psi compared with operation in the absence of the jet and with a control sample.

We also conducted experiments at lower initial concentration of 10^7 CFU/ml. In addition to perhaps being a more representative quantity of bacteria, it was thought that the bacteria could behave differently near saturation, which could

affect the ease or difficulty in killing them. Figure 16 presents results of experiments conducted with this lower initial concentration. As can be seen, a much more rapid reduction is obtained at the moderate concentration with a 3 \log_{10} reduction achieved in 20 min. and a 5 \log_{10} reduction achieved in 30 min.

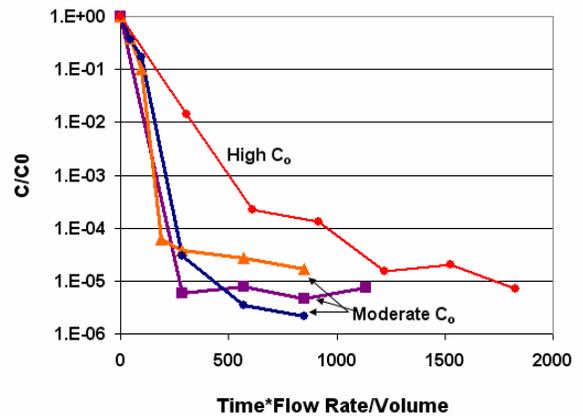


Figure 16. Results of E. coli reduction at moderate initial concentrations compared to those at high initial concentration using 8-orifice plate configuration.

CONCLUSIONS

DYNAJETS[®] cavitating water jets were shown to be able to remove a wide range of chemical contaminants from water when operated at modest pump pressures. A wide range of compounds including pesticides such as malathion, 2,4-D, and alachlor; the biocide chlorhexidine, and the VOC chloroform were found to be effectively and efficiently removed. TOC reduction of algae contaminated water as well as these compounds was demonstrated. Mixtures of compounds were also found to be efficiently removed. For mixtures of two pesticides, the removal rates of the individual compounds were found to be comparable in the mixture or in individual solutions. The cavitating jets were also found to be able to rapidly oxidize arsenic from its AS(III) form to AS(V), a form much more readily removed by conventional methods such as precipitation and filtration. This was done at two orders of magnitude greater energy efficiency than that found needed for oxidation by an ultrasonic horn. An influence of batch volume was found with variation in the efficiency of oxidation by as much as a factor of four.

The DYNAJETS[®] were also found to be able to rapidly reduce concentrations of E. Coli from initial concentrations of 10^7 to 10^9 CFU/ml. Repeatable reductions in excess of five orders of magnitude (5 \log_{10}) were rapidly achieved.

ACKNOWLEDGMENTS

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