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## Hybrid cavitation methods for water disinfection: simultaneous use of chemicals with cavitation

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### Abstract

This study brings out the potential efficacy of hybrid techniques for water disinfection. The techniques studied include, hydrodynamic cavitation, acoustic cavitation and treatment with chemicals such as ozone and hydrogen peroxide. The phenomena of cavitation which involves formation, growth and violent collapse of vapor bubbles in a liquid media is known to generate a high intensity pressure which affects the cell and microorganism viability. The hybrid technique which combines hydrodynamic cavitation, acoustic cavitation, hydrogen peroxide and/or ozone appears to be an attractive alternative to a single technique for the reduction in the heterotropic plate count bacteria as well as indicator microorganisms like the Total coliforms, Fecal coliforms and Fecal streptococci.

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## 1. Introduction

A variety of physical and chemical techniques are routinely used for potable water disinfection including chlorination [1], ozonation [2] and ultraviolet light [3]. Hybrid techniques employ the combination of various oxidation techniques, which can result in the generation of sufficient hydroxyl radicals and their oxidizing potential for water purification. These processes are known as the advanced oxidation processes (AOP) [4]. The OH radical is a powerful oxidizing radical, second only to fluorine [5] and is therefore appealing in its use for water treatment. Advanced oxidation systems generally combine ozone, hydrogen peroxide and ultraviolet radiation e.g.  $O_3$  and  $H_2O_2$ ,  $O_3$  and UV, and  $H_2O_2$  and UV.

A number of hybrid techniques have been reported in the literature which include the combination of UV radiation and ozonation for the treatment of humic acids [3] and low molecular weight organic compounds [4], combination of ultrasonication and ozonation for aromatic compound degradation [6], inactivation of microorganisms [7] and disinfection of water [1]. The advanced oxidation process (PEROXONE) which is a combination of ozone and hydrogen peroxide has been used for disinfection of water [8].

However, chemical disinfection techniques suffer from disadvantages like formation of possibly carcinogenic byproducts [6]. Therefore there is a need for developing additional disinfection processes, which could eliminate or reduce the use of these disinfecting chemicals.

In this article we investigate the viability of ultrasonication, hydrodynamic cavitation and hybrid cavitation processes involving the use of chemicals like hydrogen peroxide and ozone along with cavitation. Previous studies have indicated that these techniques can inactivate a wide range of microorganisms and ultrasonication [7] and hydrodynamic cavitation [8] which are essentially different means of generating cavitating conditions i.e. using sound and flow energies respectively, have been particularly useful for cell disruption. Cavitation is a phenomena of formation, growth and collapse of microbubbles within a liquid. If this phenomena occurs due to the passage of high frequency sound waves then it is called acoustic cavitation (ultrasonication) and if it occurs due to the pressure variations in the flowing liquid due to the change in the

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| Nomen                                  | clature   |  |  |
|--|---|--|--|
| HPC<br>CFU<br>dc/dt<br>k<br>C<br>$C_1$ | heterotropic plate count<br>colony forming units<br>rate of disinfection<br>rate constant<br>concentration of microorganism<br>initial concentration of microorganism | C <sub>2</sub><br>t<br>Rs.<br>HC<br>US | final concentration of microorganism<br>time of treatment<br>Rupees<br>hydrodynamic cavitation<br>ultrasonic |

geometry of the flowing system, it is called hydrodynamic cavitation. A detailed account of the mechanism of cavitation by ultrasonication and hydrodynamic cavitation has been published elsewhere [9].

## 2. Experimental

The disinfection study was carried out on bore well water, which had a bacterial population as indicated in Table 1. The bore well water was filtered to remove suspended particles and mud before subjecting it to the following disinfection treatment techniques.

#### 2.1. Hydrogen peroxide

Hydrogen peroxide (30% w/v) from S.D. fine chemicals was used in this study. All the experiments were performed on 1 l of bore well water using different doses (5–150 mg/l) of hydrogen peroxide on 100% basis (i.e. the strength of  $H_2O_2$  used is actually 30% w/v). The treatment was carried out for a period of 1–2 h depending on the nature of the experiment and the solution was kept well mixed with the aid of a magnetic stirrer.

#### 2.2. Ozone

The ozone generator used in this study was supplied by Arshad Electronics, India Ltd. It can be operated upto a maximum current of 1.2 A. The generator produces ozone according to the corona discharge method. Dry air was used as the feed gas. The air flow rate was 28 lps.

The ozone generator was operated at 1.2 A for all the experiments performed. At this current, it was found that the generator produces ozone at a rate of 0.2 g/h,

Table 1 Variation in bacterial population in bore well water

| Month HPC Total coli-<br>bacteria forms/100<br>(CFU/ml) ml | <ul> <li>Fecal coli-<br/>forms/100<br/>ml</li> </ul> | Fecal strep-<br>tococci/100<br>ml |
|--|--|-----------------------------------|
|  |  |                                   |
| Jan–Mar 2500–8000 50–200                                   | 20-100   | 30-200                            |
| Apr–Jun 3000–5000 60–150                                   | 30-80  | 40 - 70                           |
| Jul-Sept 1000-3000 25-60                                   | 18-30  | 20-50                             |
| Oct-Nov 2500-4000 125-170                                  | 20-60  | 30-80                             |
| Nov-Dec 6500-7500 150-200                                  | 80-100   | 180-200                           |

with a concentration of 50 mg/l of ozone in the exit air. The rate of ozone generation was found by bubbling ozone for 15 min through 400 ml of a 2% potassium iodide solution. Ozone concentration was then determined by titration with sodium thiosulphate by the iodometric titration procedure as described in the Standard Methods [10].

A stock solution of ozone was prepared by passing ozone into 100 ml of sterile distilled water for a period of 24 min. The concentration of ozone in this solution was 50 mg/l. Appropriate amount of this ozone solution was used as the dosage for various experiments.

All the experiments were performed on 1 l of bore well water using different doses of ozone. 10, 20, 40, 60 and 80 ml of the ozone stock solution was used to achieve 0.5, 1, 2, 3 and 4 mg/l of ozone concentration in water used in the study. The treatment was carried out for a period of 15 min and the solution was kept well mixed with the aid of a magnetic stirrer.

## 2.3. Acoustic cavitation

Ultrasonication was carried out with an ultrasonic horn (Supersonics) which operated with a frequency of 22 kHz and an electrical power rating of 240 W. 100 ml of bore well water was subjected to Ultrasonication for a period of 15 min. The temperature was maintained at 35-37 °C with the aid of an ice bath.

The ultrasound bath used had a peak operating frequency of 20.5 kHz. It was also supplied by Supersonics. The bath had an internal surface made of stainless steel. The internal dimensions of the bath were  $145 \times 145 \times 150$  mm. The electrical power consumption of the bath was 120 W. 2 l of bore well water was subjected to sonication. To prevent the temperature from rising above 35–37 °C, the ultrasonic irradiation has been used intermittently, for the system to cool down during the quiet period.

#### 2.4. Hydrodynamic cavitation

The set-up used to induce hydrodynamic cavitation is shown in Fig. 1. The set-up essentially consisted of a closed loop circuit including a holding tank, a centri-



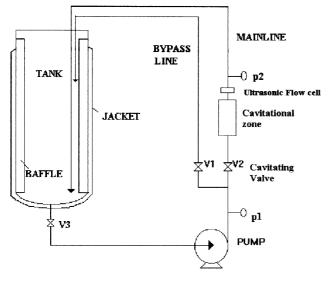


Fig. 1. Hydrodynamic cavitation set-up with ultrasonic flow cell—V1, V2, V3: control valves; p1, p2: pressure gauges.

fugal pump and a valve. The set-up used had a capacity of 80 1 and a power rating of 5.5 kW. Detailed description of the set-up used is discussed elsewhere [9].

The suction side of the pump is connected to the bottom of the tank. The discharge from the pump branches into two lines. The main line consists of a valve V2 (ball valve) which acts as a cavitating device due to its ability to throttle the flow. A hard glass tube next to this valve makes the visual observation easier. A by-pass line is provided to control the liquid flow through the main line. Control valves (V1 and V3) are provided at appropriate places to control the flow rate through the main line. The holding tank is provided with cooling jacket to control the temperature of the circulating liquid. Pressure gauges are provided to measure the inlet pressure (p1) and the fully recovered downstream pressure (p2) which in most of the cases was atmospheric.

During the experiment, the by-pass valve was left open till the pump reached its maximum speed and then partially or totally closed. The second valve was then throttled to obtain the required pump discharge pressure. Bore well water (75 l) was used in all the experiments. Experiments were carried out at 1.72, 3.44 and 5.17 bar pump discharge pressures for a period of 1-2 h depending on the nature of the experiment. Experiments were also carried out in the presence of a multiple hole orifice plate placed along the flow of liquid. The orifice plate had 33 holes of 1 mm diameter. The effective flow area was 25.92 mm<sup>2</sup>. This orifice plate changed the cavitating conditions by changing the flow pattern in the form of multiple liquid jets corresponding to each hole on the orifice plate. The change in the pattern of turbulence has been shown to affect the cavity dynamics significantly affecting cavity collapse pressures [11].

#### 2.5. Hybrid techniques

#### 2.5.1. Acoustic cavitation and hydrogen peroxide

150 mg/l of  $H_2O_2$  (for HPC bacteria) and 5 mg/l of  $H_2O_2$  (for indicator microorganisms) were added to bore well water before subjecting it to ultrasonication in the ultrasonic bath/horn as described in Section 2.3.

## 2.5.2. Hydrodynamic cavitation and hydrogen peroxide

 $H_2O_2$  (150 mg/l) was added to bore well water before subjecting it to hydrodynamic cavitation in the hydrodynamic cavitation set-up as described in Section 2.4 for the HPC bacteria and for the indicator microorganisms 5 mg/l  $H_2O_2$  was used.

#### 2.5.3. Acoustic cavitation and ozone

0.5, 1, 2, 3 and 4 mg/l of ozone was added to bore well water before subjecting it to ultrasonication in the ultrasonic horn and bath as described in Section 2.3.

#### 2.5.4. Hydrodynamic cavitation and ozone

The set-up used for this hybrid process is essentially similar to the hydrodynamic cavitation set-up described in Section 2.4 (Fig. 1) except for the following differences. The holding tank has a capacity of 10 1. The multistage centrifugal pump (KSB Pumps Ltd, India) has a power consumption of 1.5 kW and has a speed of 2800 rpm. The cavitating constriction is ball valve made of SS. 10 1 of bore well water was used in all the experiments. Experiments were carried out at 1.72, 3.44 and 5.17 bar pump discharge pressures for a period of 1– 2 h depending on the nature of the experiment and the samples were withdrawn at regular intervals. 0.5, 1 and 2 mg/l of ozone were added to bore well water before subjecting it to hydrodynamic cavitation as described above.

#### 2.5.5. Hydrodynamic cavitation and acoustic cavitation

An ultrasonic flow cell was installed in the hydrodynamic cavitation set-up on the discharge side of the pump such that the water after undergoing hydrodynamic cavitation is subjected to acoustic cavitation in the flow cell (Fig. 1). The flow cell could be operated at two frequencies viz. 25 and 40 kHz either individually or together, having a power rating of 120 W each. Bore well water (75 l) was subjected to acoustic and hydrodynamic cavitation in experiments conducted in a manner identical to only hydrodynamic cavitation set-up.

## 2.5.6. Hydrodynamic cavitation, acoustic cavitation and hydrogen peroxide

5 mg/l of  $H_2O_2$  in the case of the indicator microorganisms and 150 mg/l of  $H_2O_2$  for the HPC bacteria were added to 75 l of bore well water and then subjected to both acoustic and hydrodynamic cavitation as described in Section 2.5.5.

## 2.6. Hydrogen peroxide decomposition studies

150 mg/l hydrogen peroxide was added to 1 l of bore well water and treated as described in Section 2.1 for a period of 1 h. Its decomposition in the presence of hydrodynamic cavitation at different discharge pressures was also studied. 150 mg/l  $H_2O_2$  was added to 75 l of bore well water and treated for a period of 2 h at a pump discharge pressures of 1.72, 3.44 and 5.17 bar. Samples were withdrawn at different time intervals and the concentration of hydrogen peroxide was analysed by iodometric titration with 0.01 N sodium thiosulphate solution.

#### 2.7. Ozone decomposition studies

Aqueous ozone concentration was determined using an iodometric method [12]. This method has been reported in the literature for ozone determination [13–15]. The iodometric method is based on the liberation of free iodine from potassium iodide (KI) solutions by reaction with ozone [12]. The liberated iodine is titrated with a standard solution of sodium thiosulphate using starch as the indicator. The titration is performed at a pH of 3–4 since the reaction is not stoichiometric at a neutral pH due to the partial oxidation of thiosulfate to sulfate.

The iodometric method can be used to determine ozone concentrations above 1 mg/l. The procedure of the potassium iodide reaction and titration with sodium sulphate was performed as recorded in Standard Methods [12].

#### 2.7.1. Ozone decomposition in the absence of ultrasound

Ozone was bubbled through 100 ml of sterile distilled water for a period of 24 min to give initial ozone concentration of 50 mg/l. 25 ml of this solution was withdrawn at regular intervals and added to 400 ml of 2% KI, acidified with 0.1 N sulfuric acid and titrated with 0.005 M potassium thiosulphate. Thus the residual concentration of ozone remaining at different time intervals was calculated as stated in the Standard Methods [12].

## 2.7.2. Ozone decomposition in the presence of ultrasound

Ozone was bubbled through 100 ml of sterile distilled water for a period of 24 min. This solution was then subjected to ultrasonication by the ultrasonic horn. 25 ml of this solution was withdrawn at regular intervals and added to 400 ml of 2% KI, acidified with 0.1 N sulfuric acid and titrated with 0.005 M potassium thio-sulphate. Thus the concentration of ozone remaining at different time intervals was calculated as stated in the Standard Methods [12].

Similar experiment was carried out in the case of the ultrasonic bath except that the ozone solution was not subjected to ultrasonication directly but a beaker containing the ozone solution was placed in the ultrasonic bath.

#### 3. Method of analysis

The disinfection efficacy of the techniques described above was assessed by the number of microorganisms destroyed. Enumeration of the HPC bacteria, Total coliforms, Fecal coliforms and Fecal streptococci were done as recommended by the American Public Health Association [10] using HPC agar (Hi-media), M-Endo Agar LES, M-FC Agar Base and the K F Streptococcal Agar (Hi-media) respectively.

## 4. Results and discussions

## 4.1. Percentage disinfection achieved

#### 4.1.1. Hydrogen peroxide and ozone

When a known quantity of water is treated with a known amount of hydrogen peroxide or ozone, it is observed that as the time of treatment is increased, the number of microorganisms killed also increases (Table 2). This effect is due to the increase in the contact time between the organism and the chemical disinfectant as the time of treatment is increased.

#### 4.1.2. Acoustic cavitation

In the case of both the ultrasonic equipments, the bath and the horn, there was an increase in the percentage disinfection with the treatment time for all the microorganisms (Table 2). This is because increasing the time of exposure to ultrasound increases the probability of a cell or a microorganism coming into contact with a collapsing cavity, which would lyse it. The results obtained here are consistent with our earlier work [9].

#### 4.1.3. Hydrodynamic cavitation

It was observed that, when the pump discharge pressure was increased, the disinfection efficiency also improved. At very high pump discharge pressures (5.17 bar) the time required to achieve a certain level of disinfection is less as compared to that at lower pressures. Thus the percentage disinfection is greater at higher discharge pressures for the same time of treatment. Also, it was observed that the presence of a multiple hole orifice plate in the flow path of the liquid increased the percentage disinfection obtained for all the microorganisms. Thus, highest percentage disinfection was obtained at the discharge pressure of 5.17 bar with a multiple hole orifice plate and this value for all the microorganisms have been reported in Table 2.

#### 4.1.4. Acoustic cavitation and hydrogen peroxidelozone

Wolfe et al. [4] believed that the hydrogen peroxide molecule itself was not responsible for the disinfection action but, rather, that the free hydroxyl radical (HO–) that it produced on decomposition, was the specific in-

Table 2 Percentage disinfection obtained for various techniques

| No. | Disinfection technique  | % Reduction in Total coliforms |        | % Reduction in Fecal coliforms |        | % Reduction in Fecal streptococci |        |
|-----|---|--------------------------------|--------|--------------------------------|--------|-----------------------------------|--------|
|     |   | 15 min                         | 60 min | 15 min                         | 60 min | 15 min                            | 60 min |
| 1   | 5 mg/l H <sub>2</sub> O <sub>2</sub>  | 13                             | 28     | 9                              | 21     | 9                                 | 20     |
| 2   | 2 mg/l O <sub>3</sub>   | 60                             | 94     | 78                             | 100    | 74                                | 97     |
| 3   | US-horn   | 55                             | _      | 47                             | _      | 50                                | _      |
| 4   | US-bath   | 75                             | _      | 89                             | _      | 80                                | _      |
| 5   | US-horn + 5 mg/l $H_2O_2$   | 65                             | _      | 90                             | _      | 84                                | _      |
| 6   | US-horn + 2 mg/l $O_3$  | 99.6                           | _      | 99.3                           | _      | 98.4                              | _      |
| 7   | US-bath + 5 mg/l $H_2O_2$   | 95                             | _      | 96                             | _      | 88                                | _      |
| 8   | US-bath + 2 mg/l $O_3$  | 98.3                           | _      | 97                             | _      | 97                                | _      |
| 9   | HC (75 l set-up) at 5.17 bar with multiple hole orifice plate                   | 58                             | 85     | 38                             | 92     | 45                                | 85     |
| 10  | HC (75 l set-up) at 5.17 bar with multiple hole orifice plate + 5 mg/l $H_2O_2$ | 75                             | 90     | 60                             | 96     | 57                                | 89     |
| 11  | HC (75 l set-up) at $1.72$ bar + US flow cell (40 kHz)                          | 85                             | 96     | 60                             | 80     | 57                                | 79     |
| 12  | HC (75 l set-up) at 1.72 bar + US flow cell (40 kHz) + 5 mg/l $H_2O_2$          | 92                             | 97     | 75                             | 90     | 70                                | 92     |
| 13  | HC (10 1 set-up) at 5.17 bar  | 66                             | 83     | 57                             | 76     | 40                                | 65     |
| 14  | HC (10 l set-up) at 5.17 bar + 2 mg/l $O_3$                                     | 80                             | 94     | 88                             | 100    | 74                                | 97     |

- Data not available.

activating agent. Similarly, ozone acts by virtue of the oxygen radical generated by the decomposition of the ozone molecule. This oxygen radical can attack organic compounds in the cell membranes of the microorganisms, which result in the rupture of the membranes, affecting the cell viability and thus disinfection is achieved.

The process of generation of hydroxyl radicals/oxygen radical can be enhanced if the decomposition rate of the hydrogen peroxide/ozone can be accelerated. This can be achieved by cavitation and an attempt has been made, to do the same. Ultrasound and hydrogen peroxide act synergistically in two ways: (1) ultrasound facilitates the transport of hydrogen peroxide into the cell membranes of the microbes and thereby enhances disinfection; (2) ultrasound causes the rupture of the cells which causes the release of enzymes like peroxidase. Peroxidase reacts with the hydrogen peroxide decomposing it to water and oxygen radical. This oxygen radical then causes further disinfection by acting on the microbial cell.

It was observed that the disinfection efficiency of acoustic cavitation was increased when hydrogen peroxide or ozone was added. From Table 2, it can be observed that the results obtained with respect to time of treatment were similar to the results of only acoustic cavitation.

# 4.1.5. Hydrodynamic cavitation and hydrogen peroxidel ozone

The percentage disinfection obtained when either hydrogen peroxide or ozone is combined with hydrodynamic cavitation is higher than that obtained when hydrodynamic cavitation is used alone. Thus there is a significant synergistic effect due to the presence of hydrogen peroxide or ozone (Table 2). It was also observed that the overall disinfection rates obtained for the combination of hydrodynamic cavitation and hydrogen peroxide/ozone was more than additive (addition of the rates obtained due to individual treatment) showing the possible synergism.

#### 4.1.6. Hydrodynamic cavitation and acoustic cavitation

From Table 2 it can be observed that the percentage disinfection obtained for this hybrid process was much higher than that obtained for the individual techniques for all the microorganisms studied. It has been proved that hydrodynamic cavitation is energy efficient in cavitation generation whereas acoustic cavitation results in a more violent collapse of cavities and the combination of the two processes (hydrodynamic and acoustic cavitation) has been shown to be beneficial for the rate enhancement of chemical reactions [16]. Similarly there was an enhancement in the disinfection rates obtained with the combination of hydrodynamic cavitation and acoustic cavitation.

## 4.1.7. Hydrodynamic cavitation, acoustic cavitation and hydrogen peroxide

The addition of hydrogen peroxide was found to increase the percentage disinfection obtained for the hybrid process of acoustic and hydrodynamic cavitation (Table 2).

## 4.2. Hydrogen peroxide decomposition studies

Practically no decomposition of hydrogen peroxide was observed when used alone as well as with cavitation.

### 4.3. Ozone decomposition studies

## 4.3.1. Ozone decomposition studies in the absence of ultrasound

When the ozone stock solution was studied for decomposition, it was observed that ozone decomposes gradually over a period of 20 min. 55% decomposition was observed at the end of 15 min (Fig. 2).

## 4.3.2. Ozone decomposition studies in the presence of ultrasound

When the ozone stock solution was subjected to ultrasonication in the ultrasonic bath/horn, it was observed that ozone decomposes faster in the presence of ultrasound. Thus only 55% decomposition of ozone was observed at the end of 15 min when ozone stock solution was allowed to stand in the absence of ultrasound. However when the ozone stock solution was subjected to ultrasonication, then 75% decomposition and 80% decomposition of ozone was observed in the case of the ultrasonic horn and the ultrasonic bath respectively (Fig. 2).

This clearly points out the fact that acoustic cavitation accelerates the decomposition of ozone which accounts for the higher disinfection efficacy of the hybrid techniques discussed above. The cavitation could also increase the permeability of the oxygen radical through the microbial cell wall and increase the disinfection efficiency of the ozone.

## 4.4. Rate of disinfection

When the microbial count vs treatment time was plotted, it was observed that there was an exponential decrease in the number of HPC bacteria as well as indicator microorganisms.

Rate = No. of CFU killed/s,

160

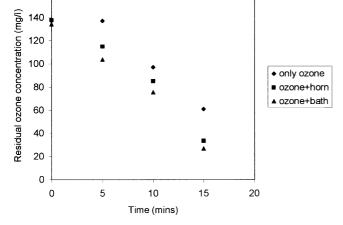


Fig. 2. Decomposition of ozone with and without sonication.

$$-\frac{\mathrm{d}c}{\mathrm{d}t} = kC$$
 (assuming first order dependence [9])

Therefore

$$-\int_{C1}^{C2} \frac{\mathrm{d}c}{C} = k \int_0^t \mathrm{d}a$$

Therefore

$$\ln \frac{C_1}{C_2} = kt$$

The rate constant was estimated by plotting  $\ln C_1/C_2$  vs time. A straight line passing through origin was obtained, the slope of which was the rate constant,  $k (\min^{-1})$ . Based on this, the results obtained for various techniques studied in this work have been discussed. Table 3 gives the disinfection rate constants for the indicator microorganisms for all the techniques employed in this study.

When different doses of hydrogen peroxide were used, it was observed, that as the hydrogen peroxide dose was increased, the rate constant also increased. It is known that hydrogen peroxide is a powerful oxidant. This oxidizing property is because of the hydroxyl radicals that are formed during the decomposition of hydrogen peroxide. As the concentration of  $H_2O_2$  increases, the hydroxyl radicals formed also increases which in turn increases the rate of disinfection [4]. It has also been shown in the literature that  $H_2O_2$  is formed in aqueous solutions under cavitating conditions, basically as a result of the combination of OH- radicals formed in the absence of their consumption by the reactive species present in the body of water [17]. The highest rate of disinfection was obtained with 150 mg/l H<sub>2</sub>O<sub>2</sub> for HPC bacteria and 40 mg/l H<sub>2</sub>O<sub>2</sub> for the indicator microorganisms. When concentrations higher than  $40 \text{ mg/l H}_2\text{O}_2$ was used for disinfection of indicator microorganisms, it was observed that all the microorganisms got destroyed instantly. In order to study the rate of disinfection, it was necessary to use lower concentrations of hydrogen peroxide. Thus, 5 mg/l H<sub>2</sub>O<sub>2</sub> was chosen as the concentration to be used for the hybrid studies and hence for the sake of comparison, the results obtained for only this value of 5 mg/l  $H_2O_2$  has been shown in Table 3.

When different doses of ozone were used, it was observed, that as the ozone dose was increased, the overall rate of disinfection also increased. It is known that ozone is a powerful oxidant. This oxidizing property is because of the oxygen radical that is formed during the decomposition of ozone. This oxygen radical can attack organic compounds in the cell membranes of the microorganisms, which result in the rupture of the membranes, affecting the cell viability and thus disinfection is achieved [18]. As the concentration of  $O_3$  increases, the oxygen radical formed also increases which in turn increases the rate of disinfection. For the HPC

Table 3 Disinfection rate constant, k (min<sup>-1</sup>) for various techniques

| No. | Disinfection technique   | Total coliforms | Fecal coliforms | Fecal streptococci |
|-----|--|-----------------|-----------------|--------------------|
| 1   | 5 mg/l H <sub>2</sub> O <sub>2</sub>   | 0.0062          | 0.0048          | 0.004              |
| 2   | 2 mg/l O <sub>3</sub>  | 0.0448          | 0.0869          | 0.0644             |
| 3   | US-horn  | 0.0565          | 0.1685          | 0.1249             |
| 4   | US-bath  | 0.0723          | 0.0381          | 0.0468             |
| 5   | US-horn + 5 mg/l $H_2O_2$  | 0.0724          | 0.1854          | 0.131              |
| 6   | US-horn + 2 mg/l $O_3$   | 0.4337          | 0.3899          | 0.3148             |
| 7   | US-bath + 5 mg/l $H_2O_2$  | 0.1997          | 0.1525          | 0.0998             |
| 8   | US-bath + 2 mg/l $O_3$   | 0.2816          | 0.2247          | 0.242              |
| 9   | HC (75 l set-up) at 5.17 bar with multiple hole orifice plate                      | 0.00347         | 0.026           | 0.0236             |
| 10  | HC (75 l set-up) at 5.17 bar with multiple<br>hole orifice plate + 5 mg/l $H_2O_2$ | 0.045           | 0.0527          | 0.0395             |
| 11  | HC (751 set-up) at 1.72 bar + US flow cell (40 kHz)                                | 0.0648          | 0.031           | 0.0302             |
| 12  | HC (75 l set-up) at 1.72 bar + US flow cell (40 kHz) + 5 mg/l $H_2O_2$             | 0.0734          | 0.0439          | 0.0473             |
| 13  | HC (10 l set-up) at 5.17 bar   | 0.0355          | 0.0329          | 0.02               |
| 14  | HC (10 l set-up) at 5.17 bar + 2 mg/l $O_3$  | 0.0555          | 0.0956          | 0.0618             |

bacteria and the indicator microorganisms, disinfection studies were carried out using 0.5, 1, 2, 3 and 4 mg/l  $O_3$ and highest rate of disinfection was obtained with 4 mg/l  $O_3$ . However, 2 mg/l  $O_3$  was chosen as the concentration to be used for the hybrid studies and hence for the sake of comparison, the results obtained for only this value of 2 mg/l  $O_3$  has been shown in Table 3.

From Table 3 it can be observed that the rate constant in the case of the ultrasonic bath is more than the ultrasonic horn for all the microorganisms. This is because the cavitational zone in the case of the ultrasonic horn is restricted to the tip of the horn and hence only those microorganisms present around the tip get killed. As compared to the area surrounding the tip of the horn, the cavitationally active area in the bath is much more and the cavities are relatively well distributed in the case of the bath increasing the probability of interaction between the microbe and the collapsing cavity. On the other hand, the cavities will remain concentrated around the tip of the horn and unless good external agitation is provided the probability of interaction will remain low. The difference in the results can only be attributed to the spatial distribution of the cavitational events.

When hydrogen peroxide is added to water before subjecting it to ultrasonication in the bath or the horn, the rate constant obtained is higher than that obtained when only the horn or the bath is used. This can be explained as follows:

- 1. One hypothesis is that ultrasound ruptures the chemical bonds between molecular components in the cell membranes of the microorganisms, which leads to an increase in the permeability to chemical substances like hydrogen peroxide [18].
- 2. The increase in the efficacy of this hybrid process may also be due to an increase in the hydrogen peroxide decomposition and the increased activities of free

radicals in water by the ultrasonic treatment [18], though no specific evidence of increased  $H_2O_2$  decomposition rate has been observed.

When ozone is added to water before subjecting it to ultrasonication in the bath or the horn, the overall disinfection rate obtained is higher than that obtained when only the horn or the bath is used. This can be explained as follows:

- 1. The most common explanation for the influence of ultrasonics is the theory of the diseggregation of flocs of microorganisms. Microbes tend to be present in the form of clumps protecting inner microbes, if these clumps are broken then better disinfection can be achieved, as the exposure of the inner microbes to the disinfectant increases. This concept has been accepted by various authors [5,18].
- 2. Another hypothesis is that of Kryszczuk who reports a transient rupture of chemical bonds between molecular components of cellular membranes which results in an increase in permeability of substances in general [18].
- 3. Boucher et al. [19] assume ultrasonic acceleration of diffusion allowing more rapid penetration of the toxic gas molecule into the microorganism.
- 4. Dahi [18] states that the disinfectant and oxidant of ozonation are the free radicals which are produced when ozone decomposes. Ultrasonic treatment increases the ozone decomposition and the activity of free radicals in water. When some threshold activity of free radicals is attained, a very rapid inactivation of bacteria is observed.

It was also observed that the rate constants obtained for the combination of ultrasonication and hydrogen peroxide or ultrasonication and ozone was more than additive for all the microorganisms studied, indicating the possible synergism.

Again in the case of hydrodynamic cavitation also, the rate of disinfection is increased when hydrogen peroxide or ozone is added. Similar explanation as given for ultrasonication holds good here also. Here also it was observed that the rate constants obtained for the combination of hydrodynamic cavitation and hydrogen peroxide or ozone was more than additive in the case of all the microorganisms, indicating the similarity of cavitating action generated acoustically or hydrodynamically.

Similarly, when the ultrasonic flow cell is used along with hydrodynamic cavitation and hydrogen peroxide, the rate of disinfection is further enhanced as the intensity of cavitation in the hydrodynamic cavitation setup is increased by the presence of the ultrasonic flow cell which in turn accelerated the decomposition of the hydrogen peroxide or increased the probability of cell wall rupture, coupled with enhanced  $H_2O_2$  penetration leading to an increase in the death rate of the microorganisms present in the bore well water. In addition to the generation of the free radicals, are the individual effects of hydrogen peroxide, ultrasonic cavitation and hydrodynamic cavitation.

From Table 3 it can be seen that the values of the rate constants differ slightly for each of the microorganism studied even though the trend remains the same for all. This can be attributed to the difference in the cell wall structure of these microorganisms, which in turn affects their susceptibility to the disinfection techniques studied in this work.

#### 4.5. Energy efficiency and cost of treatment

Table 4 gives the energy efficiency and the cost of the disinfection techniques for the destruction of Total

coliforms. According to the United States Public Health Services (USPHS), the raw water supply containing coliforms not in excess of 5000/100 ml can, with modern water treatment processes, produce potable water meeting the bacterial standards. Drinking water thus produced should not contain more than 1 coliform/100 ml. According to the EEC Guidelines; 1975, the maximum permissible limit for drinking water is 1000 count/ 100 ml at 37 °C. Total coliforms/100 ml, Fecal coliforms/100 ml and Fecal streptococci/100 ml should be zero. 99% disinfection was selected as the criteria for calculation. A sample calculation is shown in Appendix A.

From Table 4 it can be seen that the ultrasonic bath is more energy efficient as compared to the ultrasonic horn. Addition of hydrogen peroxide or ozone increases the energy efficiency in the case of the bath and the horn because the rate of disinfection is increased i.e. more number of microorganisms are killed for the same amount of energy utilized. Again it can be seen from Table 4 that the ultrasonic bath is the most energy efficient in terms of power consumed among the various physical techniques used in this study and the hydrodynamic cavitation set-up operating at 1.72 bar in combination with the ultrasonic flow cell and hydrogen peroxide appears to be the second best. The method of estimation of the energy is based only on the electrical energy consumption (column 2 of Table 4) and the equipment's electrical to mechanical efficiency has not been considered. However in terms of actual energy dissipated (column 3 of Table 4) which accounts for the energy dissipation efficiency of the concerned equipment (transformation of electrical to mechanical) is considered then the order of efficacy of a method could be different. Thus when large scale physical water treatment is desired, hybrid techniques involving hydrodynamic cavitation will be an economical choice in terms of

Table 4

Energy efficiency and cost of treatment for the destruction of Total coliforms (Appendix A)

| No. | Disinfection technique                                  | Energy efficiency based on electrical energy consumption (J) | Energy efficiency based on actual energy dissipated (J) | Cost (Rs./l) |
|-----|---|--|---|--------------|
| 1   | 5 mg/l H <sub>2</sub> O <sub>2</sub>                    | _  | _   | 0.000125     |
| 2   | 2 mg/l O <sub>3</sub>                                   | _  | _   | 0.00004      |
| 3   | US-horn   | $1.17 	imes 10^{6}$  | $3.57 \times 10^{4}$                                    | 9.75         |
| 4   | US-horn + 5 mg/l $H_2O_2$                               | $9.15 \times 10^{5}$   | $2.78 	imes 10^4$                                       | 7.62*        |
| 5   | US-horn + 2 mg/l $O_3$                                  | $2.3 	imes 10^{-3}$  | $7.9 	imes 10^{-2}$                                     | 0.36*        |
| 6   | US-bath   | $4.58 \times 10^{5}$   | $1.78 \times 10^{5}$                                    | 0.19         |
| 7   | US-bath + 5 mg/l $H_2O_2$                               | $1.66 \times 10^{5}$   | $6.45 \times 10^{4}$                                    | 0.07*        |
| 8   | US-bath + 2 mg/l $O_3$                                  | $2.1 	imes 10^{-2}$  | $7.2 	imes 10^{-2}$                                     | 0.039*       |
| 9   | HC (5.17 bar) with plate                                | $4.37 \times 10^{7}$   | $1.52 \times 10^{7}$                                    | 0.48         |
| 10  | HC (5.17 bar) with plate + 5 mg/l $H_2O_2$              | $3.37 \times 10^{7}$   | $1.17 \times 10^{7}$                                    | 0.37*        |
| 11  | HC (1.72 bar) + US flow cell (40 kHz)                   | $2.34 \times 10^7 + 5.11 \times 10^5$                        | $1.05 \times 10^{6} + 1.33 \times 10^{5}$               | 0.26         |
| 12  | HC (1.72 bar) + US flow cell (40 kHz) + 5 mg/l $H_2O_2$ | $2.07 \times 10^7 + 4.51 \times 10^5$                        | $9.29 \times 10^5 + 1.18 \times 10^5$                   | 0.23*        |
| 13  | HC (5.17 bar) 10 l set-up                               | $2.9 	imes 10^{-3}$  | $3.2 \times 10^{-3}$                                    | 0.28         |
| 14  | HC (5.17 bar) 10 l set-up + 2 mg/l $O_3$                | $5.9 	imes 10^{-3}$  | $6.4 	imes 10^{-3}$                                     | 0.14*        |

Cost of H<sub>2</sub>O<sub>2</sub> (approx Rs. 0.000125/-) and cost of ozone (approx Rs. 0.00004/-) used has not been considered as it is negligible.

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energy consumption compared to ultrasonication alone which appears to be suitable on a relatively small scale.

Cost plays a vital role in the selection of a suitable disinfection technique, which in turn would affect the overall economics of a water treatment scheme. An ideal disinfection technique is the one, which is able to bring down the bacterial population to the desired level, and is also economical. Hybrid methods like the use of hydrodynamic cavitation, acoustic cavitation and hydrogen peroxide appear to be one such technique. However the cost of treatment is considerably more as compared to the use of hydrogen peroxide or ozone alone (Table 4). Costing for all the equipments studied in this paper is done on the basis of actual electrical energy, considering cost of electricity as Rupees (Rs.) 3/kWh (\$1.00  $\cong$  Rs. 45). Cost of hydrogen peroxide has been calculated by considering the cost for 30% w/v hydrogen peroxide as Rs. 25/kg and the dosage as 5 mg/l and cost of ozone has been calculated by considering the cost as Rs. 20/kg and the dosage as 2 mg/l. Chemical disinfection techniques i.e. treatment with hydrogen peroxide is cheaper by one or two orders of magnitude than the hybrid methods described in this paper. However, the disadvantages associated with chemical treatment such as the formation of toxic by-products like trihalomethanes (THM) could be reduced or altogether eliminated by these hybrid methods.

Many a time water is available at the treatment plant at considerable hydrostatic heads or pressures, which is then reduced using elaborate pressure reduction station to make it suitable for the chemical treatment such as chlorination or ozonation, etc. The design of these pressure reduction stations can be changed so as to make them work in a hydrodynamic cavitation mode, without the supply of any additional energy. This, is likely to reduce the treatment cost and also the quantity and the cost of the chemicals used in the treatment as is evident from the reduced use of  $H_2O_2$  (5 mg/l as against 150 mg/l) for the same level of disinfection as found out in this study. Thus, hydrodynamic cavitation if used in a hybrid mode, shows a considerable promise.

## 5. Conclusions

- 1. From the studies carried out in this work, it can be observed that hybrid techniques are far superior for treating water as compared to any individual physical treatment technique. Thus the combination of hydrodynamic cavitation, acoustic cavitation and hydrogen peroxide proved to be an efficient method of water disinfection.
- 2. The hybrid technique described in this paper not only reduces the HPC bacteria (CFU/ml) but also reduces

the Total coliforms, Fecal coliforms and Fecal streptococci, which are considered as the indicators of pollution in drinking water.

- 3. In terms of energy efficiency, which is an important criteria for large-scale water disinfection, the abovementioned synergistic process appears to be very attractive.
- 4. Employing such hybrid techniques can considerably reduce the toxic by-product formation, which is a serious concern for the water authorities today.

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## Appendix A

Calculation of energy efficiency and cost of treatment of total coliforms by ultrasonic horn as reported in Table 4.

Energy efficiency based on electrical energy consumption:

Electrical power consumption = 240 J/s

Considering initial microbial count

= 100 Total coliforms/100 ml

Time required to achieve 99% disinfection (t)

 $= (\ln 1/100)/k$  (where k is the rate constant)

 $= (\ln 0.01)/0.0565 = 81.50 \text{ min} = 4890.44 \text{ s}$ 

Energy efficiency =  $t \times$  electrical power consumption

 $= 4890.44 \times 240 = 1.17 \times 10^{6} \text{ J}$ 

Cost of treatment:

To reduce Total coliforms from 100/100 to 1/100 ml (99% disinfection)

Energy required =  $1.17 \times 10^6$  J/100 ml

= 
$$1.17 \times 10^7 \text{ J/l}$$
  
=  $1.17 \times 10^7 \times 2.7778 \times 10^{-7} \text{ kWh/l}$ 

Considering 1 kWh

= Rs. 3/-  
= 
$$1.17 \times 10^7 \times 2.7778 \times 10^{-7} \times 3$$
 Rs./l  
=  $9.75$  Rs./l

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