Degradation of benzene present in wastewater using hydrodynamic cavitation in combination with air

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A R T I C L E  I N F O

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A B S T R A C T

The degradation of benzene present in wastewater using hydrodynamic cavitation (HC) alone as well as in combination with air has been studied using nozzles as cavitating device of HC reactor. Initially, the energy efficiency of the HC reactor operated at different inlet pressures was determined using the calorimetric studies. Maximum energy efficiency of 53.4% was obtained at an inlet pressure of 3.9 bar. The treatment processes were compared under adiabatic as well as isothermal conditions and it was observed that under the adiabatic condition, the extent of degradation is higher as compared to isothermal condition. Studies related to the understanding the effect of inlet pressure (range of 1.8–3.9 bar) revealed that the maximum degradation as 98.9% was obtained at 2.4 bar pressure using the individual operation of HC under adiabatic conditions and in 70 min of treatment. The combination of HC with air was investigated at different air flow rates with best results for maximum degradation of benzene achieved at air flow rate of 60 mL/sec. A novel approach of using cavitation for a limited fraction of total treatment time was also demonstrated to be beneficial in terms of the extent of degradation as well as energy requirements and cost of operation. Based on the cavitational intensity, the resonant radius of aggregates of cavitation bubbles was also determined for distilled water as well as for aqueous solution of benzene. Overall, significant benefits of using HC combined with air have been demonstrated for degradation of benzene along with fundamental understanding into cavitation effects.

1. Introduction

Aromatic compounds such as benzene, toluene, naphthalene and xylenes (BTNX) are persistent pollutants and major causes of environmental concern. The existence of these pollutants in the environment is mainly due to release through underground storage tanks in the refineries, oil wells as well as leakage during transportation [1,2] though industrial and commercial usage also is a major contributing factor. Among the different aromatic compounds, benzene is a widely used aromatic hydrocarbon to make plastics, resins, synthetic fibers, dyes, detergents, cosmetics as well as used as an industrial solvent. In the case of benzene, industrial exposure and auto exhaust accounts for about 20% of the total exposure and in addition, tobacco smoke, which is the byproduct on the combustion of tobacco, is another important source of exposure to the environment. Benzene has a significant ability to remain in the environment due to the strong chemical stability. Benzene, being toxic compound, causes many hazardous health effects such as cancer of blood forming organs, skin irritation, and redness, defects in the immune system which increases infection chances [3–5] etc.

Considering the high chances of occurrence and hazardous effects, it is important to develop an efficient approach for the complete removal of benzene.

An advanced technology called hydrodynamic cavitation (HC) is considered in the present work for the treatment of wastewater contaminated with benzene based on earlier reports for successful treatment of various aromatic compounds [6–9]. In HC, cavitation generation is based on the local variations in the pressure and kinetic energy in the flowing liquid due to geometric constrictions such as nozzles, venturi or orifice [10,11]. During cavitation, hot spots are generated with an extremely high temperature and pressure of about 5000 K and 1000 atm respectively [12–14]. The generated hot spots along with intense turbulence can intensify the oxidation of pollutants. A literature analysis is now presented to demonstrate that HC has not been applied for the degradation of benzene in wastewater highlighting the novelty of the current work.

Various processes for degradation of benzene reported in the literature include aerobic-anaerobic degradation [15], photocatalysis based on TiO₂ [16] or the nano composites [17], two-phase extraction
2.3. Methods

2.1. Materials

Benzene of 99.9% purity was obtained from Deza, A.S. Czech Republic Company. The distilled water required for making different solutions in present work was obtained freshly from the laboratory distillation unit.

2.2. Experimental set up

Fig. 1 A and B depicts the schematic and photographic representation of HC reactor used in the work. The set up mainly consists of feed tank, a reciprocating pump, cavitation chamber involving nozzle and control valves to adjust the flow through the cavitating device. The cavitation chamber is made up of quartz glass in the form of 3 nozzles, each of diameter 2 mm. The angle between the axis of the nozzles lies between 160 and 165°. The temperature of the solution was monitored by a thermometer. The air circulation in the reaction system was provided using air compressor and air flow was controlled by a rheometer.

2.3. Methods

The benzene solution of concentration as $9.35 \times 10^{-3}$ M was prepared using distilled water and taken in feed tank. Total reaction volume was kept constant as 15 L and treatment time as 70 min. Initially, the solution was stirred for 10–15 min in order to maintain the temperature of the solution around 20 °C (ambient condition). The inlet pressure was adjusted using control valves. The different experiments were performed under adiabatic and isothermal conditions. The isothermal condition was maintained using heat exchanger coil which was placed in the feed tank. The temperature was practically constant i.e. temperature fluctuation did not exceed beyond 0.5 °C–1 °C, in the case of isothermal operation.

2.4. Analysis

Concentration of benzene in the solution was analyzed using UV–visible spectrophotometer (Specord 40 M) at regular intervals of time. For any analysis, fixed quantity of sample was taken and the absorbance measured at wavelength of 254 nm. Firstly, standard calibration curve was determined with known concentration of benzene and then the equation was used to find the concentration of unknown sample based on the measured absorbance.

The reproducibility of results has been checked by repeating experiments performed in the present work at least twice. The observed errors were within ± 4% of the reported values. Similarly, reproducibility of absorbance values obtained from spectrophotometer has been also checked with errors within ± 2% of reported average.

3. Results and discussion

3.1. Calibration study

The calorimetric study was performed to determine the energy efficiency of HC reactor. The rise in temperature per unit time observed for the case of fixed water quantity of 17.5 L was measured at different inlet pressures (range of 1.5–3.9 bar). The temperature rise was monitored after every 5 min and obtained data for temperature have been depicted in Fig. 2. The rated power output of the pump was 1100 W, whereas the actual power dissipated was calculated using the following equation

$$\text{Power (W)} = m \times C_p \times \left(\frac{dT}{dt}\right)$$  \hspace{1cm} (1)

where, $m$ is the mass of fluid circulating in the system, kg; $C_p$ is heat capacity, J kg$^{-1}$ K$^{-1}$; $dT/dt$ is the rate of change of the temperature with respect to time.

Energy efficiency (expressed as %) was calculated as follows

$$\text{Energy efficiency} = \frac{\text{Actual power dissipated in the system}}{\text{Supplied electric power}} \times 100$$  \hspace{1cm} (2)

The energy efficiency was calculated when the pump operated at different inlet pressures and obtained data has been represented in Table 1. From Table 1, it can be seen that the maximum energy efficiency of 53.4% was obtained at 3.9 bar as inlet pressure to the system.

Energy efficiency expresses the actual utilization of energy introduced into the system for the generation of cavities [24]. The sample calculation of the energy efficiency has been represented in Appendix 1. It is important to note that higher energy efficiency does not necessarily mean higher net effects in terms of degradation but based on the energy efficiency data, still higher pressures beyond 3.9 bar were not investigated in the present work.

3.2. Degradation of benzene using HC reactor operated under adiabatic condition

3.2.1. Effect of inlet pressure

Experiments were performed at different inlet pressures ranging from 1.8 to 3.9 bar by adjusting the control valves in order to check the effect of pressure on the extent of degradation of benzene. The obtained results are depicted in Fig. 3 and it is seen that the extent of degradation...
of benzene was favored initially but decreased subsequently beyond the pressure value of 2.4 bar, which can be established as optimum. It is also important to note that even in the adiabatic operation, the increase in the temperature was not beyond 45 °C, which means that there will not be much losses of benzene into the surroundings. During the operation as well, no smell was noticed confirming no release of any vapors into the surroundings. The first order rate constants were determined using integral analysis as shown in Fig. 4. The maximum degradation of 98.9% with rate constant of 0.078 min⁻¹ was achieved at an inlet pressure of 2.4 bar. Subsequently, at inlet pressure of 3.9 bar,
much lower degradation of benzene as 58% with rate constant as $0.019 \text{ min}^{-1}$ was achieved. The observed trend is attributed to the fact that, beyond the optimum value of inlet pressure (in this case 2.4 bar), cavitational intensity reduces due to cavity cloud formation [25,26]. There have been many literature reports showing the dependence of optimum on the type of pollutant as well as reactor configuration. Raut-Jadhav et al. [27] reported an optimum value as 5 bar for the degradation of methomyl using combination of HC and H$_2$O$_2$. Innocenzi et al. [9] studied the decolorization of methyl orange using HC combined with advanced Fenton process. The maximum decolorization of 31.57% was reported at optimized value of 4 bar whereas, beyond the optimum pressure (5 bar) the extent of decolorization decreased to 21.58%. Badmus et al. [28] investigated the decolorization of dyes present in textile wastewater using jet loop based HC system. The maximum decolorization as 69.37% was reported at best conditions of inlet pressure as 4 bar with further increase in the inlet pressure to 5 bar resulting in much lower decolorization as 9.3%. Saharan et al. [29] studied the degradation of orange-G dye using HC reactor operated using different geometric constrictions. It was reported that the optimum inlet pressure for slit venturi was 3 bar whereas, for circular venturi and orifice plate, the optimum value was 5 bar. The literature analysis and the results presented in the current work confirmed the necessity of detailed study related to the effect of inlet pressure as it is specific for the geometric constriction as well as type of the pollutant.

### 3.2.2. Effect of air injection in combination with HC

The effect of using air was studied by introducing air into the feed tank using compressor at varying flow rate between 40 and 120 mL/min at fixed inlet pressure of 3.9 bar. The trends for the extent of degradation and rate constants obtained at different air flow rates have been depicted in Fig. 5 and Fig. 6. From the obtained results, it is seen that higher degradation is observed with an increase in the air flow rate from 40 to 60 mL/min however, further increase in the air flow rate to 80 mL/min reduced the extent of degradation. The maximum degradation of benzene as 86.7% with rate constant being $0.029 \text{ min}^{-1}$ was achieved at air flow rate of 60 mL/min within 70 min of treatment. At the same inlet pressure, only HC resulted in lower degradation as 58%. The observed effects initially i.e. below the optimum flow rate can be attributed to the fact that gaseous species present in air such as nitrogen, oxygen, and other gases promotes OH radical formation [9] which ultimately enhances the extent of degradation of benzene. Also, the presence of air provides additional nuclei giving higher cavitational intensity that further helps in the degradation of pollutant [12]. Beyond the optimum flow rate of air, too much cavitation events lead to reduced cavitational intensity based on cushioned collapse giving lower extents of degradation. In the present work, although only HC resulted in comparatively higher extent of degradation at best condition of pressure of 2.4 bar (98.9%) than that obtained when operated using the combination of HC with air, the formation of some intermediates were observed, meaning the mineralization extent is lower. However, the combination of HC with air resulted in no formation of intermediates, which means that the mineralization extent would be higher compared to only operation of HC.
Fig. 3. Effect of inlet pressure on the extent of degradation of benzene under adiabatic conditions.

Fig. 4. Kinetic data fitting for the degradation of benzene at different inlet pressures under adiabatic conditions.
Fig. 5. Effect of combination of HC with air on the extent of degradation of benzene under adiabatic conditions.

Fig. 6. Kinetic data fitting for the combination of HC with air on the extent of degradation of benzene under adiabatic conditions.
Joshi and Gogate [30] studied the combination of HC with air applied for industrial wastewater treatment by injecting air into the main line at pressure of 85 psi. The combination of HC with air was reported to give 15.5% of COD reduction, which was higher than the individual treatment approaches of only air and only HC. Rajoriya et al. [12,31] studied the application of combined approach of HC with air for the decolorization of reactive blue 13 (RB 13) as well as the treatment of effluent obtained from textile dying industry respectively. It was reported that higher extent of decolorization as 53% was obtained at air flow rate of 1.5 L/min [12]. Similarly in the other study, with the same air flow rate (1.5 L/min), the combination of HC with air resulted in TOC, COD, and color reduction as 23.8%, 17.39%, and 28.18% respectively for the treatment of textile industry wastewater, which was again higher compared to individual operations [31]. Though the reported trends are similar, the quantitative results are different for each system marking the necessity of detailed investigations presented in the current work.

### 3.3. Degradation of benzene using HC reactor operated under isothermal condition

Series of experiments were also performed under isothermal conditions at different inlet pressures ranging from 1.8 to 3.9 bar. Fig. 7 and Fig. 8 depict the results obtained for the benzene degradation and fitting of kinetic rate constants. The maximum degradation of 98% was achieved at pressure of 2.4 bar, which decreased to 73.6% with a further increase in pressure to 3.9 bar (Table 2). The similar trend of increase in the inlet pressure increasing the extent of degradation till the optimum value is reported by many research scientists [8,32], though with different optimum as explained earlier for the case of adiabatic operations. Under isothermal condition, the combination of HC with air resulted in 83.5% of degradation at inlet pressure of 3.9 bar when air was injected at flow rate of 60 mL/min whereas, the combination of HC with air resulted in 86.7% under adiabatic condition at the same inlet pressure as well as air flow rate. Also, under isothermal condition, the extent of degradation of 89% was achieved at inlet pressure of 3 bar when air was injected at flow rate of 80 mL/sec whereas, under adiabatic condition, the combination of HC with air resulted in 98.8% of degradation at same inlet pressure as well as air flow rate (Table 3). The presence of air (oxygen) also contributes to the oxidation of benzene under adiabatic conditions due to an increase in the temperature which ultimately enhances reaction rate. This is particularly observed within 30–40 min of the treatment time. However, under isothermal conditions, the role of oxygen decreases as the temperature is practically constant.

The extent of degradation obtained under adiabatic condition and isothermal conditions was also dependent on the pressure (Table 2). At lower pressures (till 3 bar), adiabatic operation was found to be favorable possibly due to the fact an increase in the temperature favored the kinetics for degradation and hydroxyl radical formation. At higher pressure, however, the extent of degradation was low (58%) for adiabatic conditions than that obtained under isothermal condition (73.6%) at the same inlet pressure of 3.9 bar. This can be attributed to the fact that under the adiabatic condition at inlet pressure of 3.9 bar, the reaction temperature reached to 50 °C which leads to generation of vaporous cavities collapsing less violently and ultimately reduces the extent of degradation. Typically it is observed that an increase in the solution temperature increases the extent of degradation only till an optimum value. Beyond the optimum value, the generation of vaporous cavities is responsible for lowering the cavitation intensity and hence, the extent of degradation [6,33]. Under isothermal condition, the solution temperature was constant at 20–22 °C which resulted in an enhanced extent of degradation as compared to that obtained under adiabatic condition at higher pressures. Wang and Zhang [6] also observed the similar trend of increase in the extent of degradation of alachlor with an increase in the temperature till the optimum value of 40 °C, however, further increase in the temperature to 60 °C resulted in the lower rates of degradation of alachlor. The current work has clearly

![Fig. 7. Effect of inlet pressure on the extent of degradation of benzene under isothermal conditions.](image)
illustrated the role of temperature and pressure both in deciding the extent of degradation.

3.4. Effect of using cavitation for limited period of time followed by only circulation

In the approach, for the initial 30 min of treatment time, the system was operated under cavitation mode followed by operation under only circulation mode (without cavitation) for 40 min with mixing provided using an overhead stirrer. Such an approach would give lower cost of treatment as energy requirement for without cavitation is lower and also too many cavities leading to cushioning effects are not expected especially at higher pressures. Same operational conditions have been maintained at different pressures varied from 2.5 to 3.9 bar with continuous air circulation at fixed flow rate of 80 mL/min for the total treatment time of 70 min. From the results depicted in Fig. 9, it is observed that the maximum extent of degradation of benzene as 92.8% was achieved at an inlet pressure of 3.9 bar. The energy released during cavitation limited for initial 30 min is absorbed by dissolved gases initially which helps in giving sufficient oxidizing species to initiate the benzene degradation. Degradation continues in only circulation mode as well ultimately enhancing the extent of degradation at lower energy requirements. The treatment using cavitation limited for initial 30 min of total treatment of time followed by circulation for 40 min resulted in a significantly higher degradation (92.8%) as compared to the treatment using cavitation (58.5%) for a treatment of 70 min under the same conditions as pressure of 3.9 bar and continuous air circulation at flow rate of 80 mL/min. When cavitation is applied for entire treatment time over conditions of inlet pressure as 3.9 bar and air flow rate of 80 mL/min, too much cavitation leads to cushioning effects and hence lower extents of degradation. In the case of HC reactor, pump requires large amount of energy to circulate reaction solution for longer period of time, say 70 min. However, cavitation limited for initial 30 min reduces the power consumption of pump required for total treatment time which reduces the total treatment cost. Hence, the use of cavitation for limited treatment time can be considered as the cost effective approach for degradation of benzene. However, a prerequisite for the degradation of benzene in this mode is the presence of oxygen in the reaction system.

3.5. Analysis of cavitation phenomena

The cavitational intensity obtained in water and aqueous solution of benzene have been studied using a spherical hydrophone (8105) attached to computer for measurements. The frequency of hydrophone

| Inlet pressure (bar) | Extent of degradation under adiabatic conditions (%) | Extent of degradation under isothermal conditions (%) |
|---------------------|-----------------------------------------------------|-----------------------------------------------------|
| 1.8                 | 90.2                                                | 88.3                                                |
| 2.4                 | 98.9                                                | 98                                                  |
| 3                   | 96                                                  | 92.3                                                |
| 3.9                 | 58                                                  | 73.6                                                |

Table 2
Comparison of final extent of degradation under adiabatic and isothermal conditions using HC alone.

Table 3
Comparison of final extent of degradation obtained for the combined approach of HC + air under adiabatic and isothermal conditions.

| Conditions                                      | Extent of degradation under adiabatic conditions (%) | Extent of degradation under isothermal conditions (%) |
|------------------------------------------------|-----------------------------------------------------|-----------------------------------------------------|
| HC + air (Inlet pressure 3.9 bar and air flow rate 60 mL/min) | 86.7                                                | 83.5                                                |
| HC + air (Inlet pressure 3 bar and air flow rate 80 mL/min)   | 98.8                                                | 89                                                  |
lies within $0.1 \times 10^5$ to $1.6 \times 10^5$ Hz. The obtained results for frequency and intensity spectra were analyzed through software (adobe audition 1.5). The different experiments performed using water and benzene solution at inlet pressure of 3.9 bar were analyzed using hydrophone and the obtained results have been depicted in Fig. 10A and B.

The resonant radius of the cavitation bubbles ($R_{\text{res}}$) was determined by the following formula:

$$R_{\text{res}} = \frac{3}{16} \times \frac{\omega_{\text{res}}}{\pi}$$

where, $\omega_{\text{res}}$ – resonant pulsation frequency of cavitation bubble, Hz.

The different cavitation bubbles aggregate radius were obtained under adiabatic condition, for the approach of HC with and without air at inlet pressure of 0.39 MPa, as depicted in Table 4. The data depicted for bubble radius reveals that there is not much difference due to the introduction of air into the tank, which can be attributed to the large difference in the macro sizes of the air bubbles and the micro sizes of the cavitating bubbles locally. From Fig. 10A, it can be also seen that there are two resonant frequencies obtained for water as 1700 and 5200 Hz in which aggregates of bubble radius were 1.9 and 0.6 mm respectively. From Fig. 10B, it can be seen that the resonant frequency of 5200 Hz and bubble radius of 0.6 mm were also obtained for aqueous benzene solution. The polydisperse (bubbles of different sizes) system of aggregates of bubbles was obtained when cavitation is generated in water whereas, when cavitation is generated in benzene solution, the monodisperse system was obtained.

Some literature reports are available based on the use of hydrophone for mapping the cavitational effects inside the sonochemical reactor [34,35]. The pressure pulses obtained from hydrophone have been used in order to check the maximum bubble size reached during cavitation with the help of theoretical simulations of the bubble dynamics equations which ultimately gives an idea about type of cavitation. Since not much information has been reported for hydrodynamic cavitation, the present study highlighting the fundamental information would be useful in better understanding of the wastewater treatment using HC.

4. Conclusions

The present work has clearly established the effectiveness of the HC alone as well as the combined treatment of HC with air for the degradation of benzene. Initially, calorimetric efficiency studies of HC reactor at different inlet pressures revealed that the maximum energy efficiency was obtained at inlet pressure of 3.9 bar. However, the maximum extent of degradation of benzene as 98.9% was achieved at 2.4 bar using HC operated under adiabatic condition, confirming that dissipating too much energy is also not a good idea. The intensified approach of HC with air was demonstrated for the degradation of benzene under adiabatic and isothermal conditions, though the efficacy was dependent on the operating pressure. It has been also observed that the adiabatic condition is beneficial in terms of the enhanced degradation as compared to the degradation obtained under isothermal conditions; again the efficacy was dependent on pressure. It was also observed that the use of cavitation for initial 30 min followed by only circulation of 40 min resulted in 92.8% of degradation at an inlet pressure of 3.9 bar in presence of air circulated at flow rate of 80 mL/min. The approach clearly demonstrated the effective use of cavitation for limited time period so as to reduce the cost of treatment. The present work also demonstrated the cavitational intensity measurements in terms of determination of resonant radius of aggregated of cavitation bubbles. Overall, from the obtained results, it can be established that the combined approach of HC with air under adiabatic conditions and optimized pressure is the best treatment approach for near complete removal of benzene from aqueous solution.

CRediT authorship contribution statement

Pooja Thanekar: Methodology, Investigation, Writing - original
draft. Parag R. Gogate: Writing - review & editing, Funding acquisition, Project administration. Z. Znak: Conceptualization, Supervision, Writing - review & editing, Funding acquisition, Project administration. Yu. Sukhatskiy: Methodology, Investigation, Writing - review & editing. R. Mnykh: Methodology, Investigation, Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix 1

Sample calculation of Energy efficiency at inlet pressure of 1.5 bar.
The actual power dissipated into system, \( W \) is

\[
Power\ (W) = m \times C_p \times \left( \frac{dT}{dt} \right)
\]
here, \( m \) = mass of water circulating in the system = 17.5 Kg; \( C_p \) = heat capacity of water = 4184 J.Kg\(^{-1}\).K\(^{-1}\); observed \( \frac{dT}{dt} = 0.08 \) °C/sec.

Actual dissipated power = \( 17.5 \times 4184 \times 0.08 \) = 98 W.

Energy efficiency (%) = \( \frac{\text{Actual power dissipated in the system}}{\text{Supplied electric power}} \times 100 \) = \( \frac{98}{1100} \times 100 = 8.9\% \).

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