

Effects of Sodium Sulfate and Sodium Chloride for Sonochemical Degradation on 1,4-benzoquinone and Hydroquinone in Aqueous Solution

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ABSTRACT

1,4-benzoquinone, with the molecular formula $C_6H_4O_2$, is generally known as a para-quinone. It is a six-member ring compound with an oxidized derivative known as 1,4-hydroquinone, which is a bright yellow crystal that has an irritating odour. On the other hand, hydroquinone, also known as benzene-1,4-diol, has the chemical formula $C_6H_4(OH)_2$. It looks like a white granular solid. Quinone is generally used as a precursor to hydroquinone. The skeletal muscle relaxant, ganglion blocking agent, benzoquinonium, is made from benzoquinone. It is utilized to suppress double-bond migrations during olefin metathesis reactions. 1,4-benzoquinone is also used in the synthesis of Bromodol, while hydroquinone is the main ingredient in black-and-white photographic developers such as film and paper developers, where it diminishes the silver halide to elemental silver. There are various other uses related to this diminishing power. As a polymerization inhibitor, hydroquinone prevents the polymerization of methyl methacrylate, acrylic acid, etc. Studies have demonstrated the various effects of Na_2SO_4 and $NaCl$ on the sonochemical degradation of 1,4-benzoquinone and hydroquinone using a 200-kHz sonicator. The highest degradation rate was obtained in the presence of 0.433 M Na_2SO_4 for 1,4-benzoquinone. After 30 minutes of ultrasonic irradiation, the total concentration of 1,4-benzoquinone decreased to 99% in the presence 0.433 M Na_2SO_4 . Without Na_2SO_4 , the sonochemical degradation rate of 1,4-benzoquinone was 4.5 times higher than that of hydroquinone, whereas in the presence of 0.433 M Na_2SO_4 under the same conditions the initial reaction rate of 1,4-benzoquinone was increased to become 10.6 times higher than that of hydroquinone. On the other hand, in the presence of $NaCl$, no effects were observed for the decomposition of hydroquinone but negative effects were clearly observed for the decomposition of 1,4-benzoquinone.

Keywords: 1,4-benzoquinone; Hydroquinone; Na_2SO_4 ; $NaCl$; Sonication

INTRODUCTION

For the last few decades, Advanced Oxidation Processes (AOPs) such as O_3 , UV with H_2O_2 , UV with O_3 , Fe^{2+} with H_2O_2 and UV with O_3 and H_2O_2 have become visible as undertaking technologies for the degradation of uncooperative organic hazardous wastes (Rosenfeldt et al. 2004; Spangord et al. 2000 & Anotai et al. 2006). AOPs produce $\cdot OH$ radicals which has redox potential 2.8 V vs. Normal Hydrogen Electrode (NHE) are strongly reactive for organic pollutants. At present, applications of new AOPs with $SO_4^{\cdot -}$ radicals which are formed via reaction of OH radicals with sulfate ions has been investigated because $SO_4^{\cdot -}$ radical anion ($E^0 = 2.5-3.1$ V vs. NHE) also possess high oxidation potential. There are various researches have been brought out by the use of $SO_4^{\cdot -}$ radicals for the decomposition of a variety of organic pollutants (Waldemer et al. 2007; Fernandez et al. 2004; Bandala et al. 2007 & Hori et al. 2005).

Sonochemical degradation method is a promising one because maximum reaction conditions including pyrolysis and radical reactions are generated via the formation of cavitation bubbles with high temperatures and pressures.

As for sonolysis in aqueous solutions, hydroxyl radicals are produced through the expeditious collapse of cavitation bubbles (Makino et al. 1983 & Riesz et al. 1985). So that, the sonochemical degradation method has raised a wide attention in environmental applications such as organic pollutants control (Mallakpour et al. 2014 & Saccani et al. 2014), biological hydrogen production (Guo et al. 2011 & Yang et al. 2012), and excess sludge reduction (He et al. 2011; Yang et al. 2013 & Guo et al. 2013). Sonochemical degradation of phenol and phenolic compounds has been investigated by several researchers and investigated some intermediates such as hydroquinone, benzoquinone, catechol (Lim et al. 2014; Jyothi et al. 2014; Rokhina et al. 2013; Wang et al. 2013; Verma et al. 2013; Sakakura & Mitsuo 2012; Joseph et al. 2011; Zhang et al. 2011; Torres-Palma et al. 2010; Berlan et al. 1994; Entezari et al. 2003; Petrier et al. 1994; Dükkanç et al. 2006 & Serpone et al. 1993). These compounds further decomposed into more hydrophilic compounds that is formic and oxalic acid (Dukkanc et al. 2006 & Yim et al. 2003).

Although there are many reports for sonochemical degradation of phenol and phenolic compounds (Lim et al. 2014; Jyothi et al. 2014; Rokhina et al. 2013; Wang et al.

2013; Verma et al. 2013; Sakakura & Mitsuo 2012; Joseph et al. 2011; Zhang et al. 2011, and Torres-Palma et al. 2010), to the best of our knowledge, sonochemical decomposition of 1,4-benzoquinone and hydroquinone under argon gas pressure is not reported.

In this study, we performed the sonochemical degradation of 1,4-benzoquinone and hydroquinone in an aqueous solution at a frequency of 200 kHz. The lack of the research is we have performed 30 minutes of experiment and used only two inorganic salts. 1,4-benzoquinone provide an influence to both powerful mineral acids and alkali, which phenomenon condensation and decomposition of the composite. Its acute toxicity (oral LD50: 130 mg/kg body weight for rats) (Patnaik 2007). On the other hand acute toxicity of hydroquinone (oral LD50 value for both sexes combined was >375 mg/kg) (Topping et al. 2007). As electron acceptor benzoquinones perform a vital role in the breathing organisms (Okamura et al. 2000 & Kawamukai 2002). Consequent to UV irradiation of benzoquinone by-products in aqueous media, hydroxyquinone and oxygen are produced (Leighton et al. 1929; Lente et al. 2004 & Joschek et al. 1966). The oxidation of diverse quinones to hydroxyquinones was expressed in the research (Spyroudis 2000).

On the other hand, hydroquinone is considered the main potential molecular messengers for semiquinone-type natives in the ignition of complicated polymeric and oligomeric arrangements accommodated in biomass components. Hydroquinone and its outgrowths are strong registered products of the ignition and pyrolysis of frequent types of biomass, as well as tobacco (Visser et al. 1985 and Lee et al. 1999). Consumption of hydroquinone has been displayed to induce choking, oxidizing impression, affected breathing in humans over and above reduced bone marrow and corneal damage in mice (Bilimoria et al. 1975 and Leanderson & Christer 1992). For all that, the discarding interest with hydroquinone in ignition and pyrolysis is its deterioration to produce persistent, semiquinone-type free radicals and other toxic outgrowths. It is acknowledged that pyrolysis of hydroquinone edges to the construction of p-benzoquinone and phenol (Sakai & Masayuki 1976 and Khachatryan et al. 2006) over and above a number of other aromatic and polycyclic aromatic hydrocarbon products (Ledesma et al. 2002 and Marsh et al. 2004). In this paper, we analyzed the effects of Na_2SO_4 and NaCl on the sonochemical decomposition of 1,4-benzoquinone and hydroquinone to enhance the rate of degradation. In addition, we also suggested that the degradation mechanism of 1,4-benzoquinone and hydroquinone was different from that of other phenolic compounds. Na_2SO_4 and NaCl also known as degrading agent in AOPS. Uddin et al. (2016) published on sonochemical decomposition in presence of inorganic salts corresponded of no effect or slight negative or positive effects.

EXPERIMENTAL SECTION

1,4-benzoquinone purity 98%, hydroquinone, sodium sulfate (anhydrous) both purity 99%, and sodium chloride purity 99.5% was purchased from Wako Pure Chemical Industries, Ltd. Japan. All the chemicals were reagent grade and used after received. Decontaminated water (18.2 M Ω cm resistivity) was prepared from a Millipore Milli-Q Gradient water purification system and was used to prepare all aqueous solutions. Argon (99.999% purity) was purchased from Osaka Sanso, Japan.

SONOLYSIS

Ultrasonic generator (Kaijo 4021, Lot no. 1033, MFG. no. 34C3) and a oscillator of 65 mm inner diameter were used for sonochemical degradation and were control at 200 kHz with an input power of 200 W. The glass vessel with a gross volume of 60 ml sample solution was used for ultrasonic irradiation under argon atmosphere. The vessel had a side arm with a silicon rubber septum for argon gas bubbling and sample extracting (every 0, 5, 10, 20 and 30 min) by the glass syringe (1 ml) without exposing the sample to air.

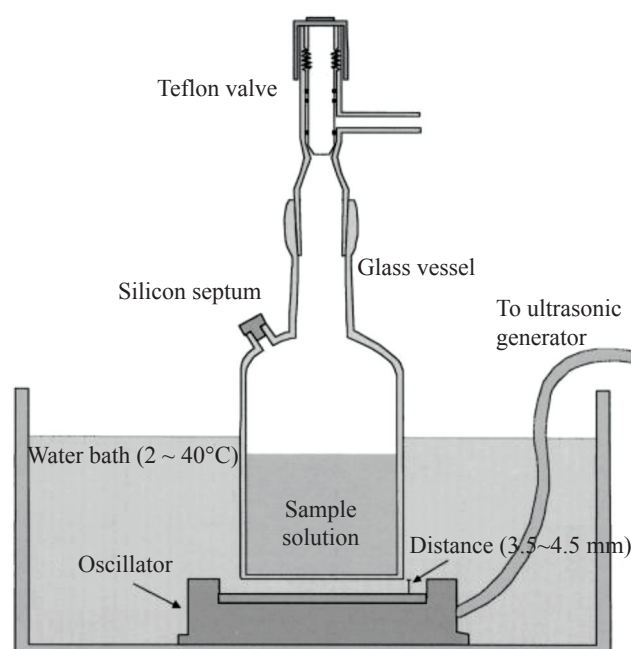


FIGURE 1. The schematic diagram of experimental setup for sonolysis (Okitsu et al. 2002).

The schematic diagram of the experimental setup as shown in Figure 1 was described in the literature (Okitsu et al. 2002). The glass vessel was flat bottomed and 1 mm thick. The vessel was climb up at a constant position (4 mm from the oscillator). The sonicated solution concentrations were determined with a HPLC.

CALORIMETRICALLY POWER MEASUREMENT

When a wave experiences any inequality in the belongings of the channel in which it is propagating, its way of behaving is disturbed. Moderately changes in the way extending over many wavelengths conduct mostly to alter in wave speed and generate direction-the phenomenon of refraction. Sonochemical power usually indicated as the electrical input or output power to and from the generator. Several procedures are available to evaluate the amount of ultrasonic power entered into a sonochemical reaction (Mason 1991). Calorimetric method, that involves computation of the initial rate of a temperature increase produced when a system is illuminated by power ultrasound. This is established on the supposition that almost all the mechanical energy fabricates heat and thus the output power can be procured via calorimetry. In the present experiment, acoustic energy was measured by the calorimetric method.

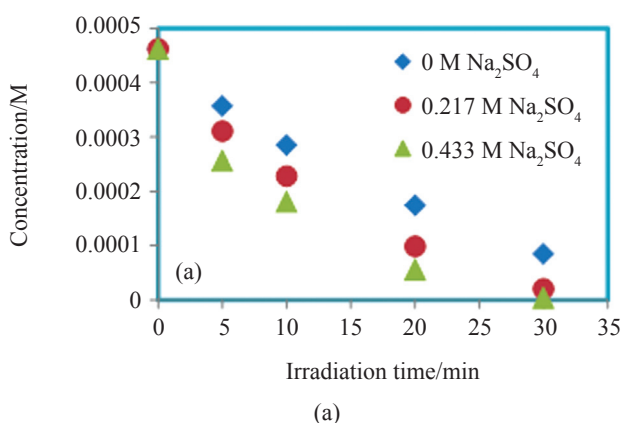
For one and all system the temperature (T) in the reaction cell was recorded against time (t) at 10s, intervals, using a thermocouple placed in the reaction vessel. From the T versus t data, total acoustic power can be calculated using the Equation (1).

$$P_w = m C_p (\Delta T / \Delta t) \quad (1)$$

Where,

P_w = power (Watt),

m = mass of water used (100 g),



C_p = heat capacity of water ($4.18 \text{ J}\cdot\text{g}^{-1}\cdot\text{K}^{-1}$),
 ΔT = temperature rise [$(28.2-27.1) = 1.1^\circ\text{C}$],
 Δt = interval of time (30s).

Before and after ultrasonic irradiation, the inside reactor cell temperature of water was thermostated at room temperature. Calorimetric power quantifications were carried out three times under the same conditions, and the volume of solution in the reaction vessel was 60 g. The calorimetric power was input in the cell was around 15 W.

RESULTS AND DISCUSSION

Throughout the sonolysis of water, it is popular that acoustic cavitation originates highly reactive primary radicals such as OH and H due to the thermal decomposition of water as shown in Equation (2) (Ashokkumar et al. 2008 & 2004). A number of recombination and other reactions (Equations (3)-(4)) occur. The OH radical is a nonselective oxidant with a high redox potential (2.8 V), having the power to oxidize most organic pollutants.



where “sonication” refers to sonication.

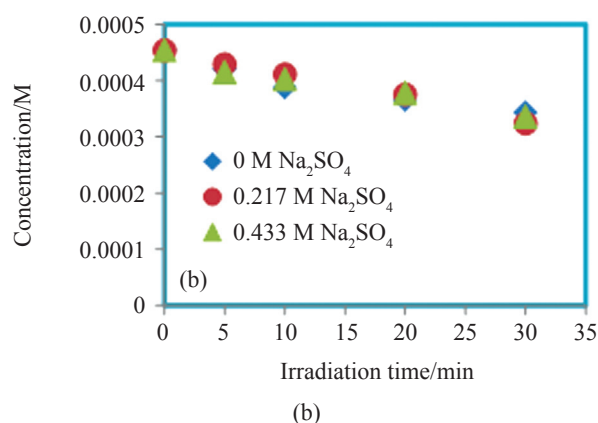


FIGURE 2. Effects of sonochemical degradation of a) 1,4-benzoquinone and b) hydroquinone in absence and presence of Na₂SO₄.

Sonochemical degradation of 1,4-benzoquinone and hydroquinone was inquired into the absence and presence of Na₂SO₄ respectively using UV-vis spectrophotometer. Figure 2(a) and (b) show the effects of sonochemical degradation of 1,4-benzoquinone and hydroquinone in the absence and presence of various concentrations of Na₂SO₄. Figure 1(a) shows, the initial rate of sonochemical decomposition of 1,4-benzoquinone increased 2.8 times in presence of 0.433 M Na₂SO₄ than in absence of Na₂SO₄. In contrast, at same concentration of Na₂SO₄ the sonochemical degradation of hydroquinone was not increased conspicuously as shown in Figure 2(b).

Therefore, to understand the sonochemical degradation mechanism of 1,4-benzoquinone and hydroquinone research was performed in presence of different electrolytes such as Na₂SO₄ and NaCl using HPLC. Figure 2 also shows change in the concentration of phenolic compounds throughout sonication under Ar atmosphere. From Figure 2, it was observed that the degradation rates of 1,4-benzoquinone was strongly affected by the addition of Na₂SO₄, on the other hand hydroquinone was slightly affected by the addition of Na₂SO₄. After 30 min ultrasonic irradiation reaction, the total concentration of 1,4-benzoquinone was decreased 99% in the presence of 0.433 M Na₂SO₄. The initial rate of

1,4-benzoquinone (in presence of 0.433 M Na_2SO_4) was increased 2.6 times than in the absence of Na_2SO_4 . On the other study, in the presence of same concentration of Na_2SO_4 , during sonochemical degradation of hydroquinone the decomposition rate slightly increased. In the presence of 0.433 M Na_2SO_4 , after 30 min sonication the total concentration of hydroquinone decreased only 26%, whereas in the absence of

Na_2SO_4 it was 24%. From Figure 3 experimentally observed in absence and presence of Na_2SO_4 the rates of sonochemical decomposition of 1,4-benzoquinone and hydroquinone were different. Figure 2 shows the comparison on time dependence of sonochemical degradation of 1,4-benzoquinone and hydroquinone in absence and presence of Na_2SO_4 .

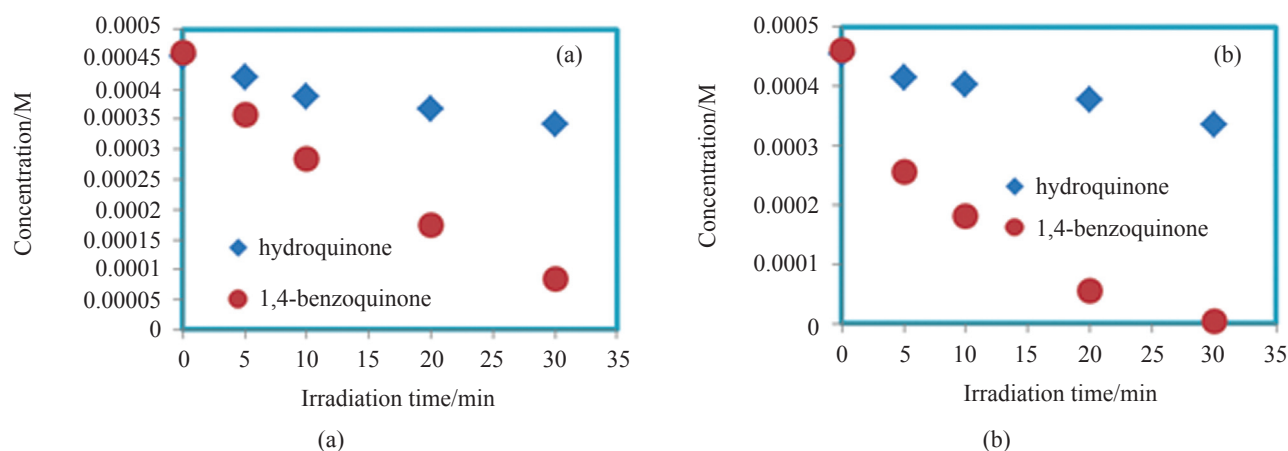


FIGURE 3. Comparison on time dependence of sonochemical decomposition of 1,4-benzoquinone and hydroquinone in the a) absence, b) presence of Na_2SO_4 .

Figure 3 clearly shows that, the sonochemical degradation rate of 1,4-benzoquinone in the absence of Na_2SO_4 was 4.5 times higher than hydroquinone, whereas in presence of 0.433 M Na_2SO_4 at same condition the 1,4-benzoquinone reaction rate was increased 10.6 times higher than hydroquinone. The pKa values of 1,4-benzoquinone and hydroquinone were 4.2 and 4.0 respectively. In the particular case of 1,4-benzoquinone that reported in a cyclic voltammetric study of the aqueous electrochemistry system. The common shape of the potential-pH diagrams for the other quinones were similar to that of 1,4-benzoquinone, but not hydroxyquinone (Bailey et al. 1985).

We also explored the effects of NaCl on the rate of sonochemical decomposition 1,4-benzoquinone and hydroquinone. Even in the case of NaCl, no effect was observed for hydroquinone but clearly observed negative effect for 1,4-benzoquinone sonication as seen in Figure 4.

In the absence of salt, the initial rates of sonochemical degradation 1,4-benzoquinone were faster than those of hydroquinone. It is well known that the hydrophobicity of an organic solute is one of the most important parameters for sonochemical decomposition, because highly hydrophobic compounds tend to concentrate at the gas liquid interface where the concentration of OH radicals is very high (Henglein et al. 1985). To confirm hydrophobicity of solutes, we measured log P of these compounds and found that 1,4-benzoquinone (log P = 0.16*, 0.19 (Moret et al. 1996) and hydroquinone (log P = 0.58*, 0.59 (Veith et al. 1979), here, * symbol indicates experimental value.

Nanzai et al. mentioned that aromatic compounds decomposition rate increased with increasing value of log P

(Nanzai et al. 2008) but present results were contradictory with Nanzai et al. The addition of Na_2SO_4 significantly affects the rates of decomposition for 1,4-benzoquinone, but addition of NaCl does not clearly affect the decomposition rates for 1,4-benzoquinone. Cheng et al. (2010) observed, NaHCO_3 and Na_2SO_4 are found to reduce the sonochemical rate constants for PFOX (X = S or A; perfluorooctane sulfonate or perfluorooctanoate respectively).

The results of present research were reverse with Hofmeister series. Paterova' et al. (2013) observed completely reversed Hofmeister series and correlative with present results. In presence of Na_2SO_4 , sonolysis of 1,4-benzoquinone significantly enhanced the rate of decomposition, on the other hand, at same condition sonolysis of hydroquinone was slightly/not enhanced the rate of degradation as shown in Figure 2. In presence of NaCl, sonolysis of 1,4-benzoquinone reduced the rate of degradation reaction that is negative effect observed. On the other hand at the same condition, sonolysis of hydroquinone no effect was found (as shown in Figure 4).

Hydroquinone is a reducing agent that is reversibly oxidizable to quinone. The oxidation potential of hydroquinone at 20°C and pH 7.03 is 0.2982 volts. Quinones are suggested to be a dominant redox-active moiety within natural organic matter (Nurmi & Paul 2002) and humic substances (Scott et al. 1998). Electron transfer to quinones can be expected to lead to an increase in semiquinone radical intermediates as well as hydroquinone Figure 6. Figure 5 shows the time dependence of 1,4-benzoquinone sonication in the absence and presence of different concentration of Na_2SO_4 . It was observed that 1,4-benzoquinone was degraded under argon

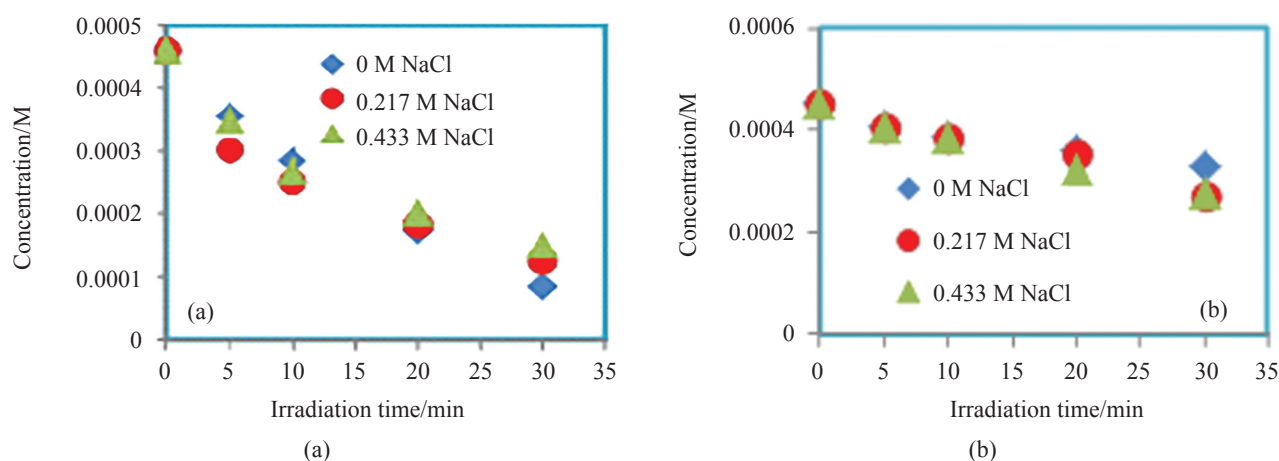


FIGURE 4. Sonochemical degradation of a) 1,4-benzoquinone and b) hydroquinone in absence and presence of NaCl.

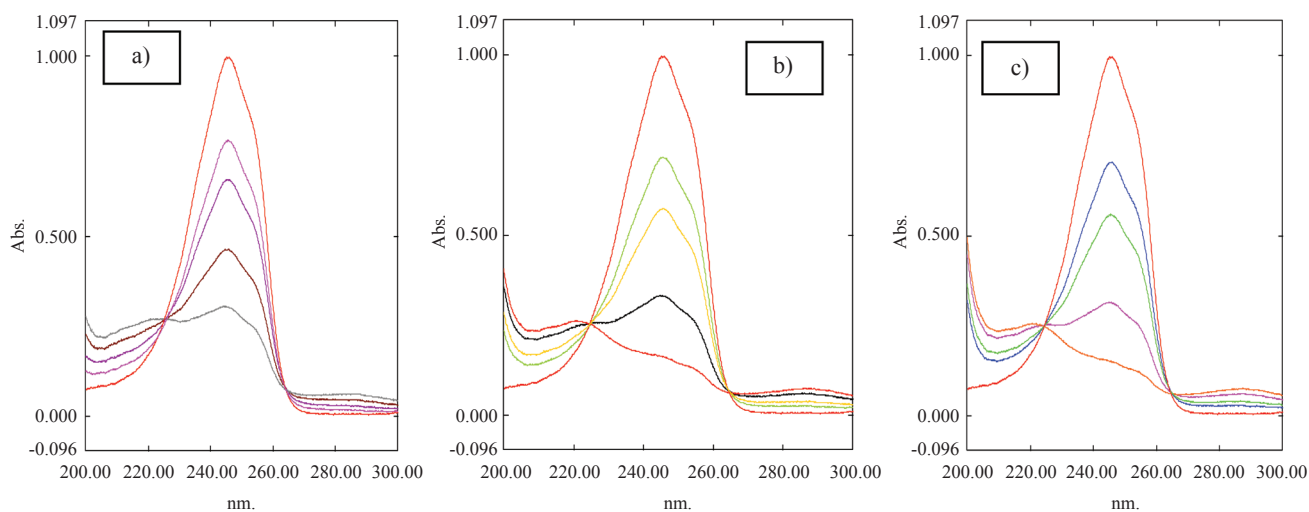


Figure 5. Time dependence of 1,4-benzoquinone sonication, isosbestic points were observed in the absence a), in the presence of 0.217 M b) and 0.433 M c) Na_2SO_4 .

atmosphere due to influence of sonolysis. It can be seen that the absorption peaks corresponding to 1,4-benzoquinone at around 245.7 nm was gradually decreased. In Figure 5, two isosbestic points were also observed at 225.6 nm and 264.5 nm, indicating that 1,4-benzoquinone certainly reduced to form hydroquinone and other compounds.

Zhao et al. (2010) studied on enhanced oxidation of 4-chlorophenol using sulfate radicals initiated from zero-valent and peroxydisulfate at ambient temperature and found sulfate radical anion as the dominant active species was responsible for the oxidation of 4-chlorophenol in the ZVI-PDS system, mechanism adopted as Equation (5).



INTERMEDIATES

In sonolysis of 1,4-benzoquinone and hydroquinone intermediate was detected at retention time (R.T.) 3.4 min in absence and presence of different concentrations of Na_2SO_4 and NaCl. Another intermediate was detected at retention time 10 min only for hydroquinone sonolysis.

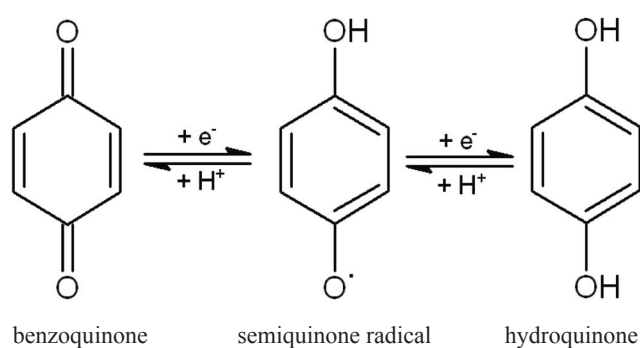


FIGURE 6. Reduction of 1,4-benzoquinone to the intermediate semiquinone radical and further reduction to hydroquinone.

Figure 7 shows the formation of intermediate at R.T. = 10.00 min for sonication of hydroquinone in absence and presence of different concentration of Na_2SO_4 . The highest amount was produced in absence of Na_2SO_4 and lowest was in presence of 0.433 M Na_2SO_4 . It may 1,4-benzoquinone and quantitatively measurement as shown in Figure 8. The maximum 1,4-benzoquinone was produced 15.73 μM in the absence of Na_2SO_4 .

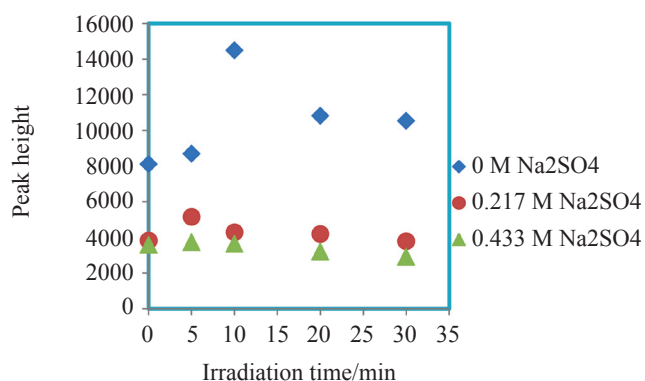


FIGURE 7. Formation of intermediate at R.T. = 10.00 for sonication of hydroquinone in presence of different concentration of Na₂SO₄.

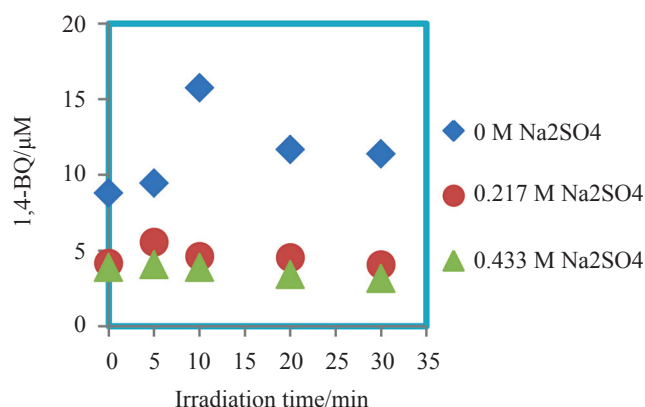


FIGURE 8. Formation of 1,4-benzoquinone in hydroquinone solution at different concentration of Na₂SO₄ (Peak height).

Figure 9 shows the formation of intermediate at R.T. = 3.4 ≈ 3.7 for sonication of a) 1,4-benzoquinone, and b) hydroquinone in the absence and presence of different concentration of Na₂SO₄. The highest amount of intermediate was produced in the presence of 0.217 M Na₂SO₄. Also,

experimentally same intermediates were detected in the presence of different concentrations of NaCl. Figure 10 is indicating the reaction mechanism of 1,4-benzoquinone and hydroquinone.

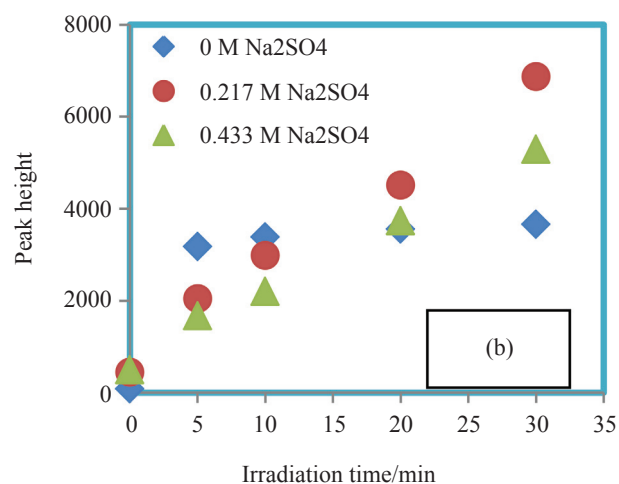
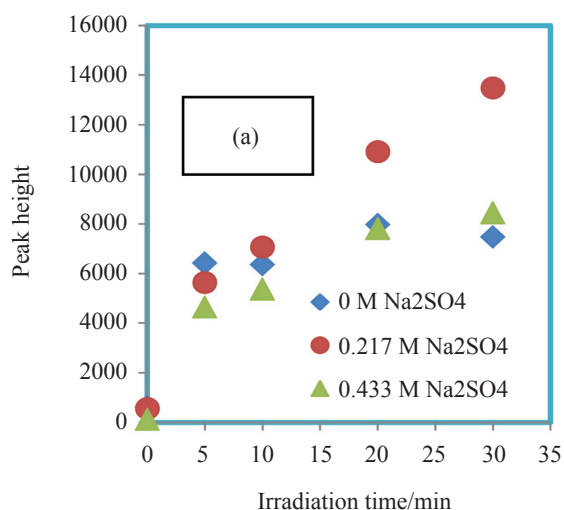


FIGURE 9. Formation of intermediate at R.T. = 3.4 ≈ 3.7 for sonication of a) 1,4-benzoquinone and b) hydroquinone in the absence and presence of different concentration of Na₂SO₄.

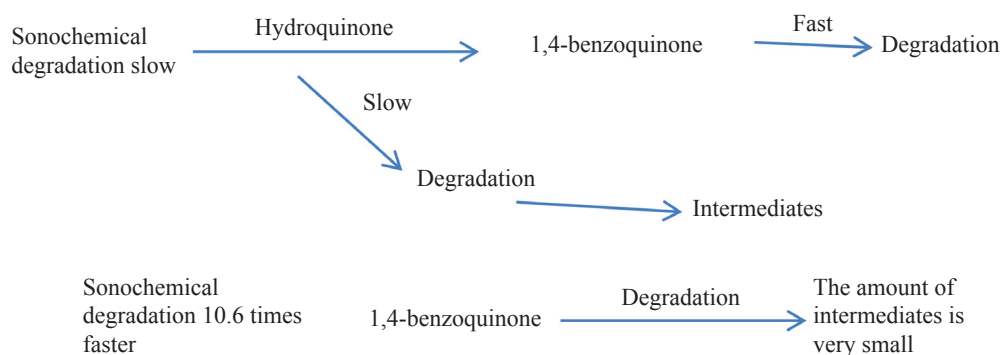


FIGURE 10. Reaction mechanism of 1,4-benzoquinone and hydroquinone

CONCLUSIONS

The effects of Na₂SO₄ and NaCl on the sonochemical degradation of 1,4-benzoquinone and hydroquinone were investigated by using 200 kHz sonicator. In absence and presence of Na₂SO₄ initial rates of sonochemical degradation were significantly increased in the order 1,4-benzoquinone > hydroquinone. Based on the experimental results, in absence of Na₂SO₄ 1,4-benzoquinone sonochemical degradation rate was 4.5 times higher than hydroquinone, whereas in presence of 0.433 M Na₂SO₄ at same condition 1,4-benzoquinone reaction rate was increased 10.6 times higher than hydroquinone. On the other hand, in presence of NaCl initial rate of sonochemical degradation of 1,4-benzoquinone and hydroquinone was different. In the presence of different concentrations of NaCl the initial rate of sonochemical degradation of 1,4-benzoquinone was reduced. Also, in presence of same concentration of NaCl the initial rate of degradation of hydroquinone was no/very little effect.

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