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DEGRADATION OF REFRACTORY COMPOUNDS USING SONO FENTON AND SONO ELECTRO FENTON METHODS

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ABSTRACT

In recent days the most challenging issue is the treatment of industrial waste water. The industries generate increasing amounts of wastewater, contaminated with toxic and hazardous organic compounds, which cause severe problems to the environment. The refractory substances are the one of the toxic organic compounds present in the waste water, that are resistant to biodegradation or degrade very slowly; they persist in water bodies for long time. Among the harmful refractory compounds, phenolic compounds have deserved more attention because of their toxicity and frequency of industrial wastewaters. The phenolic compounds in the wastewater stream mainly coming from oil refineries, coal conversion plants, petrochemicals and polymeric resins. The treatment technologies available for phenolic wastes are physical, chemical, biological and electrochemical processes. Several researches have investigated that the anaerobic biological processes is used for the removal of phenolic compounds but, the biological processes are cannot treat higher concentration associated with industrial waste water.The removal of harmful organic molecules using Advanced Oxidation Processes (AOPs) is an effective tool for pollution control and environmental protection. Out of various advanced oxidation processes we have selected sono fenton and sono electro fenton methods for our study. Ultrasound is a convenient and effective method of generating hydroxyl radicals which is the key oxidant in AOPs. In order to improve the degradation efficiency the sono fenton process is combined with electrochemical process. This study describes the use of ultrasound and associated chemical reactions, with hydrogen peroxide and ferrous sulfate, as a powerful means of remediating water contaminated with organic pollutants. In Sono fenton method, the ultrasonic waves of frequency 34kHz is supplied by transducer, which is used to produce hydroxyl radicals and also fenton reagent is externally added as a catalyst. Finally a comparison was made between Sono fenton and Sono electro fenton methods against the degradation of refractory compounds.

Keywords: Advanced oxidation process, ultrasonic waves, refractory compounds, pollutants, fenton reagents.

1.INTRODUCTION

At present, a large part of pollution in the public water system is caused by industries. In one particular case, which is important within these industrial effluents is phenol. The word "Phenols" includes phenol and its derivatives, i.e, those aromatic organic compounds that contain one or different hydroxyl groups. Although the phenol is a compound that appears most frequently in liquid effluents, the presence of some of its derivatives, to be precise, ortho, meta, and p-cresols. The phenol vapour and liquid are toxic and easily absorbed through the skin. Once inhaled the vapour its corrodes the respiratory tract and the lungs. Severe burns result from the liquid coming into contact with skin and eyes.

The harmful characteristics of phenol compounds lie in concentrations of parts per billion, which are inferior to toxic concentrations, and it contributes disagreeable smell and taste to chlorinated water. Normally the taste cannot be detected in concentrations inferior of 0.1 to 0.01 ppb. The phenolic compounds are considered toxic for some aquatic life, forms in concentrations superior to 50ppb and the ingestion of one gram of phenol can have fatal consequences in humans. Additional effect is the capacity of phenols to combine with existing chlorine in drinking water, giving rise to chlorophenol compounds that are even more toxic and difficult to eliminate.

Sonochemistry has been demonstrated as a promising method for the destructions of aqueous pollutants. The ultrasonic irradiation introduces cavitations bubbles that grows and subsequently collapse through compression rarefaction cycles (Gustavo Stoppa Garbellini, 2012). The

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temperature and pressure in the collapsing bubbles increased. Under Such extreme condition, water molecules undergo hydrolysis to yield hydroxyl radicals and hydrogen atoms. Other radicals can also be produced depending on the properties of dissolved gases and other solutes present in the medium. The mechanism proposed for the sonochemical degradation of aqueous pollutants includes oxidation by hydroxyl radical, pyrolytic decomposition, supercritical water oxidation and combustion (Mason & Lorimer, 2002; Flint & Suslick, 1991).

The combination of ultrasonic waves with an electrochemical oxidation and/or reduction can result in a powerful method for phenol degradation. The electro fenton method is an indirect electro oxidation technique with higher oxidation power than anodic oxidation for water remediation.

The propagation of acoustic wares in a liquid medium induces cavitation that is the formation and growth of bubbles and even their violent collapse at high acoustic pressure. Among the physical effects of these collapses of the high rates of micro mixing, the cleaning of the electrodes surfaces by dissolving or pitting the inhibiting layers, these effects result mainly in an enhancement of the solid – liquid mass transfer between the electrodes and the solution.

In this study low cost and environmentally safe reagents are used to degrade the phenolic compounds. The parameters such as pH, current intensity, hydrogen peroxide concentration, ferrous sulfate concentration and the electrolyte volume was investigated.

2. MATERIALS AND METHODS

Preparation of phenol solution

In the present study the phenol solution was prepared synthetically, for the preparation of 1ppm concentration of phenol; 1mg of phenol was dissolved in 1lt of water. Likewise for this experiment, 1000ppm concentration of phenol was prepared by dissolving 1gm of phenol in 1lt of water.

Preparation of electrolyte

Sodium sulfate is taken as an electrolyte for this study. To prepare the electrolyte dissolve 35.5gms of sodium salt in 1lt of the distilled water to get 0.25M sodium sulfate.

Sono fenton process:

The experiment was conducted in batch mode. The sono reactor having the size of 200x150x125mm. The volume of phenol taken for this study was 1000ml. The experiment was conducted at room temperature. A fenton reagents (hydrogen peroxide and ferrous sulfate solutions) of known quantity was added to the reactor and the solution was well mixed to achieve sonication at low frequency (34 kHz) using a bath type ultrasonic generator. The sample was periodically collected at specified intervals. The parameters involved in the Sono fenton method are pH, hydrogen peroxide concentration and ferrous sulfate concentration.

Sonoelectro fenton process

The experiment was conducted in batch mode; stainless steel is used as electrodes for both anode and cathode. Both cathode and anode was dipped in aqueous solution containing phenol and hydrogen peroxide. The support electrolyte was sodium sulfate. Sono electrochemical oxidation was carried out in the galvanostatic mode at current intensities not exceeding 500 mA.

Analytical methods:

The quantitative estimation of organic compounds plays an important role in the analytical laboratory. The quantitative estimation is based on the standard chemical reactions and generally involves volumetric methods. Phenol on bromination reaction in aqueous solution gives 2, 4, and 6-tribromo phenol. Instead of using a standard solution of bromine water, a bromate-bromide mixture, which is readily, liberates bromine in presence of an acid. The aqueous solution of phenol was treated with an excess of brominating agent, the unreacted bromine was back- titrated against standard sodium thio sulfate solution using starch is an indicator.

$$\text{Degradation efficiency} = \frac{d-e}{d} \times 100 \qquad (2..1)$$

d = Initial concentration of phenol in mg/lt
e = Final concentration of phenol in mg/lt

3. RESULTS AND DISCUSSION

In Sono fenton method the fenton reaction rate was strongly increased by irradiation with ultrasound during the reaction of Fe^{3+} ions are accumulated in the system. After the Fe^{2+} ions are consumed and the reaction was practically stops. The mechanisms involved in fenton reagent are as follows,

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + OH^{\cdot} \qquad (1)$$

The influential parameters such as ferrous sulfate concentration, hydrogen peroxide concentration and pH were taken for this study. The optimized conditions of ferrous sulfate concentration, hydrogen Peroxide concentration and the solution p^H was 40, 500 and 3.The Phenol degradation efficiency of 65% at optimum condition was observed within 30min.

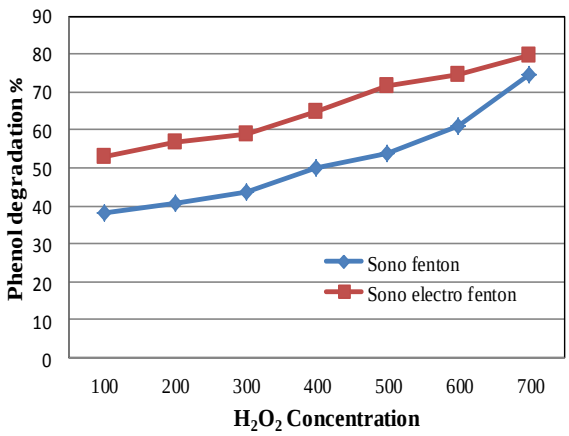

Fig 1 Degradation efficiencies of phenol with different hydrogen peroxide concentrations.

The investigator praveena juliya et al reported that the degradation of phenol using fenton reagents producing the efficiency was 77.6% at pH 5. In comparison with our work slight variation in pH values but the degradation efficiency was low. In future work we have to increase the pH values to obtain the maximum degradation efficiency.

The initial Hydrogen peroxide Concentrations was taken as 100ppm and it was increased up to 500ppm and the effects were calculated .It was found that degradation of phenol increases with increasing the hydrogen peroxide concentration as shown in fig 1**.** The initial concentration of Fe^{2+} was fixed and hydrogen peroxide (30% w/v) concentration was varied and the variations of phenol concentration up to 30min were measured.

The Solution pH has significant effect in sono fenton method. The hydrogen ion concentration in the solution is the contributing factor for the degradation of phenol in waste water.

The fig 2 shows that the degradation of phenol was high at p^H 3, and also it was observed that the degradation products are acidic compounds because the pH values reduced during the experiment.

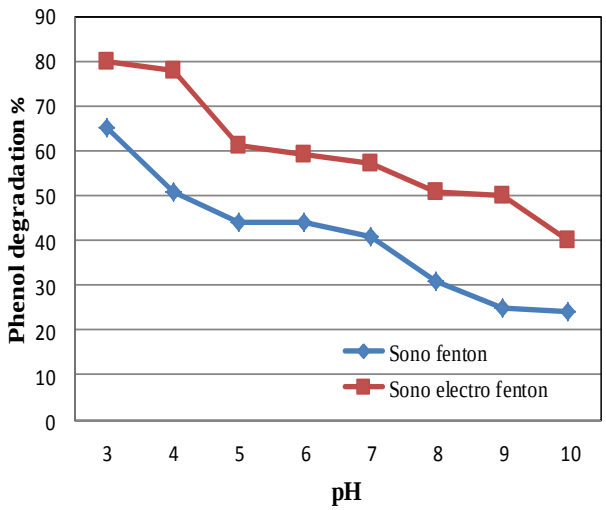

Fig 2 Effect of pH on phenol Degradation

In sono electro fenton method electrolyte is a substance which liberates ions and allows electric current to passes through, thereby improving anodic and cathodic reactions. In the present study sodium sulfate solution of 0.25M was used as an electrolyte. The concentration of phenol at various time intervals was noted with respect to various current intensities.

Trabelsi.F,Ait Lyazid have investigated in ultrasonic reactors by means of electrochemical method with a 20kHz sonication, the electrochemical oxidation of phenol in Nacl media allows the conversion of 75% of initial phenol with in 10 min of treatment. At 500 KHz a conversion of 95% of the initial phenol was obtained within the same treatment time and final product degradation were acetic and chloroacrylic acid. In our work the electrochemical method with 34 kHz sonication in presence of fenton reagents, the degradation of phenol was 85% within 30 min. The work performed by the above researchers the degradation efficiency was 95% at 500 kHz frequency but we obtained 85% efficiency at a minimum frequency level and minimum time intervals. The degradation was increased on addition of Fe^{2+} ions.

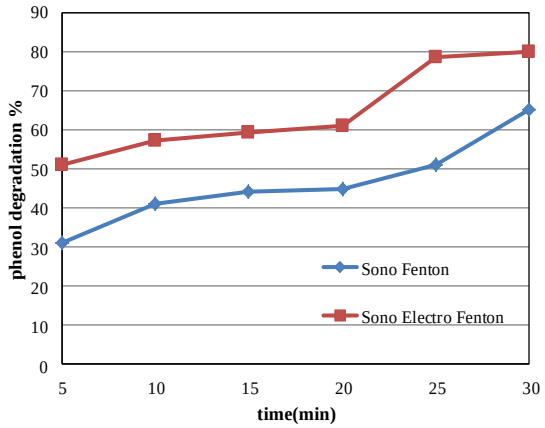

Fig 3.3 comparison of degradation efficiency of phenol by sonofenton and sonoelectrofenton methods

Sono fenton method the degradation efficiency at optimum condition was 65% within 30min. In Sono electro fenton method the optimized conditions were, hydrogen peroxide concentration 500ppm, electrolyte volume 300ml, current intensity 0.5amps and the solution p^H was 3. The degradation efficiency at optimum condition was 80% within 30min.

4. CONCLUSION

Sonication combined with electrochemical process in presence of fenton reagents is a promising method for the degradation of toxic organic pollutants and treatment of industrial wastewater. Sonoelectrofenton gives better degradation efficiency than sonofenton method but power consumption and cost is too high. Therefore, working at low power ultrasound with different fenton reagents should be planned in order to overcome this drawback. . From this way the present study concluded that sono fenton method with hydrogen peroxide and ferrous sulphate reagents attains the degradation efficiency of 65% is the most economical method for phenol degradation. In future we have to introduce some other fenton reagents for better degradation results.

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