Electrochemical Destruction of Aromatic Organic Compounds for effective wastewater treatment

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Abstract. The research is devoted to the study of the destruction of aromatic organic compound – 1,3-dihydroxybenzene by electrochemical oxidation process. Impact assessment was carried out during the process of such parameters as the concentration 1,3-dihydroxybenzene, nature of medium (pH), electrolysis time, organic matter conversion coefficient, electrochemical cell configuration. Electrochemical destruction process was actualized in coaxial membraneless electrochemical reactor with coaxial electrode arrangement achieving the effective mass-transfer. The results of current density diversification and pH approved the optimal terms to be the current density 4 kA/m², pH = 11.0. Assessment of conversion coefficient of 1,3-dihydroxybenzene was estimated by spectrophotometric method. Conversion coefficient of 1,3-dihydroxybenzene was 95.78% during 3 hours of electrolysis.

1. Introduction
Nowadays there is the discussion of water treatment containing aromatic organic compounds. They are synthetic dyes, pharmaceuticals (antibiotics), phenol and its items, benzoic acid, hydrocarbons, halogenated sulfur and Carbon-nitrogen bond, pesticide, high-density metals and other organic complexes [1-2]. Such materials has the way of toxicity and biodegradability. The theme has been closely discussed abroad [3-4]. In Russia, it has been discussed lately [5-7]. The classical method of disposal of aromatic organic compounds is biological treatment but it has number of shortcomings. One of them is impossibility of the matters gross failure resulting from its structure persistency and thereby low treatment, toxic matters can inactivate microorganisms and make difficulties of biological processes [1, 2]. The developing, engineering and introduction of profitable engineering solutions with ecological and economical point of view has applicability.

Nowadays advanced method of the treatment is oxidative destruction (advanced oxidation processes) by chemical and electrochemical ways [8].

Advanced oxidation processes based on chemical and electrochemical generating of oxidative agents (hydroxyl radical, peroxide and hydroperoxide of radicals) being in a possession of oxidative activity and able further to their interaction with pollutants containing in wastewater [9]. The method is mostly often implemented for organic compounds of the aromatic series. Obtaining oxidizing agents is possible using hydrogen peroxide. It is pollution-free and has a high oxidizing ability, forms only water and oxygen in the process of degrading [10]. The oxidizing power of hydrogen peroxide can be increased by the generation of hydroxyl radicals in various promising oxidative processes [11]. Large-
scale production \( \text{H}_2\text{O}_2 \) is based on multi-staged anthraquinone oxidation method. However, it has disadvantages of high-energy consumption and waste generation [11]. For that reason researches are aimed at \( \text{H}_2\text{O}_2 \) in-situ in order to reduce energy costs and potential risks during transportation, storage and concentrated solutions processing of \( \text{H}_2\text{O}_2 \). There are two directions for the synthesis of \( \text{H}_2\text{O}_2 \). The first one – direct synthesis from \( \text{H}_2 \) and \( \text{O}_2 \) using catalysts (Pd, Au и/или Au-Pd). Nevertheless, it has undesirable reactions (equation 1 and 2) causing an explosive hazard [11].

\[
\begin{align*}
\text{H}_2 + 1/2\text{O}_2 & \rightarrow \text{H}_2\text{O} \\
\text{H}_2\text{O}_2 + \text{H}_2 & \rightarrow 2\text{H}_2\text{O}
\end{align*}
\]

(1) (2)

The second one is electrochemical synthesis through a two-electronic oxygen reduction reaction. In this way, only oxygen or air is required and no hazardous waste from synthesis products [15].

Since the essence of promising oxidative processes is consisted in the generation of highly active oxidizing agents, Fenton system is distinguished from possible methods for their preparation (\( \text{H}_2\text{O}_2/\text{Fe}^{3+} \)) [12, 13]. The oxidative decomposition and organic matters transformation by Fenton system has been known for almost a century (Fenton, 1894) [14]. There is an advantage for this method - \( \text{H}_2\text{O}_2 \), used as an oxidizing agent is cheaper than others and of using iron as catalyst in particular. Various ionic forms of iron are in water form complexes with water and hydroxyl ions depending on \( \text{pH} \) and temperature. Fenton process is based on the use of Fenton reagent, i.e. mixtures of \( \text{Fe}^{2+} \) salt (catalyst) and \( \text{H}_2\text{O}_2 \) (oxidizing agent), which is a strong oxidizing agent of organic matters and contributing to the radicals generation of \( \text{HO}^- \) and \( \text{HO}_2^- \) at acidic values \( \text{pH} \) [15]. \( \text{HO}^- \) has a high standard oxidative potential (2.8 V) and demonstrate a high reaction rate compared to other conventional oxidizing agents like \( \text{Cl}_2, \text{O}_2, \text{O}_3, \text{H}_2\text{O}_2 \) and \( \text{KMnO}_4 \) [16]. The radical reacts with many organic and inorganic compounds with high rate constants [8].

The classical Fenton process [17] chemically using \( \text{Fe}^{3+} \) as a catalyst proceeding in acid medium, (\( \text{pH} 2.8-4.0 \)), is as follows (equation 3):

\[
\text{Fe}^{3+} + \text{H}_2\text{O}_2 \rightarrow \text{Fe}^{2+} + \text{HO}^- + \text{HO}_2^-
\]

(3)

Fenton process suggests [11, 18] hydroxyl radicals to be produced in accordance with equation (3), while the catalyst is reduced in accordance with equation (4) or from the reaction with \( \text{Fe}^{3+} \) with intermediate organic radicals (equation (5-7)).

\[
\begin{align*}
\text{Fe}^{3+} + \text{H}_2\text{O}_2 & \rightarrow \text{Fe}^{2+} + \text{HO}_2^- + \text{H}^+ \\
\text{RH} + \text{HO}_2^- & \rightarrow \text{R}^+ + \text{H}_2\text{O} \\
\text{R}^- + \text{Fe}^{3+} & \rightarrow \text{R}^+ + \text{Fe}^{2+} \\
\text{R}^- + \text{HO}_2^- & \rightarrow \text{R-OH}
\end{align*}
\]

(4) (5) (6) (7)

\( \text{R} \) – organic compound.

Anyway, the number of competitive reactions can also occur in accordance with chemical equation (8-11), affect the oxidation process:

\[
\begin{align*}
\text{Fe}^{2+} + \text{HO}^- & \rightarrow \text{Fe}^{3+} + \text{HO}^- \\
\text{H}_2\text{O}_2 + \text{HO}^- & \rightarrow \text{HO}_2^- + \text{H}_2\text{O} \\
\text{HO}_2^- + \text{HO}^- & \rightarrow \text{O}_2 + \text{H}_2\text{O} \\
\text{HO}^- + \text{HO}^- & \rightarrow \text{H}_2\text{O}_2
\end{align*}
\]

(8) (9) (10) (11)

The main parameters determining the course of Fenton process are \( \text{pH} \), the concentration of \( \text{Fe}^{2+} \) and \( \text{H}_2\text{O}_2 \) ions, the initial content of pollutants and associated ions. The optimal \( \text{pH} \) is from 2.0 to 4.0. At \( \text{pH} \) above 4.0, \( \text{Fe}^{2+} \) ions are oxidized forming complex compounds with hydroxyl ions. The oxidative activity of \( \text{H}_2\text{O}_2 \) also decreases in alkaline medium. In this regard, Fenton process requires adjustment of the \( \text{pH} \) and the corresponding consumption of reagents significantly increases operating costs [15, 19]. One of the ways to intensify the process and expand the \( \text{pH} \) range is to use additional chemicals.

Fenton process (\( \text{H}_2\text{O}_2/\text{Fe}^{2+} \)) can be implemented both chemically and electrochemically. Depending on possibility of implementing the generation of oxidizing agents by both chemical and electrochemical, the preference is for the preference of electrochemical. As the chemical method has a significant drawback - a large consumption of \( \text{H}_2\text{O}_2 \) solutions and \( \text{Fe}^{2+} \) salts is required, the electro-Fenton process is an acceptable method [20]. In addition, the cost of the electrochemical production of
H₂O₂ is significantly lower compared to its chemical synthesis. Electrochemical advanced oxidative processes with the production of H₂O₂, HO⁻ and HO₂⁻ radicals should be classified as the most environmentally friendly methods. This is explained by the lack of the chemical reagents use, and, therefore, simplifies their production by electrochemical means. A number of factors causes the use of advanced oxidative electrochemical processes for the organic compounds destruction: the simplicity of directly organizing the process of electro-generation of oxidizing reagents, lower energy consumption and high efficiency of current density, low potential, effective destruction of pollutants and versatility. Electrochemical advanced oxidative processes [21] consist in the electrochemical generation of hydrogen peroxide in the cathode cell by two-electronic oxygen reduction (equation 12).

\[
O_2 + 2H^+ + 2e \rightarrow H_2O_2
\]  

(12)

Electrochemical production H₂O₂ in situ is based on the efficient supply of oxygen. The efficiency of H₂O₂ electric generation in the process of electro-Fenton mainly depends on the type of cathode material, oxygen supply rate and pH [19]. Table 1 presents possible methods for generating H₂O₂ [22].

| Method                                                                 | Process                                    | equations of reaction                  |
|-----------------------------------------------------------------------|--------------------------------------------|----------------------------------------|
| Electrogeneration H₂O₂ (two-electron O₂ reduction)                     | Cathode surface and inside                 | O₂(r) + 2H⁺ + 2e → H₂O₂                |
| Disproportion of H₂O₂                                                 | Cathode surface and inside                 | 2H₂O → O₂(r) + 2H₂O                   |
| Cathodic reduction of H₂O₂                                            | Cathode surface and inside                 | H₂O₂ + 2H⁺ + 2e → 2H₂O                |
| Anodic oxidation of H₂O₂                                              | Anode surface                              | H₂O₂ → HO₂⁻ + H⁺ + e                  |
| Disproportion of H₂O₂                                                 | Bulk electrolyte                           | 2H₂O → O₂(r) + H⁺ + e                 |

It is reported that carbonaceous materials such as graphite felt, reticulated vitreous carbon, activated carbon, activated carbon fiber, carbonic sponge, carbon/graphite felt, carbon/carbon nanotube with polytetrafluoroethylene composite, mercury, graphite, carbon felt, gas-diffusion electrode, and diamond doped boron can be used as cathode materials in the H₂O₂ power generation [23-26]. Gas-diffusion electrode-carbon material/polytetrafluoroethylene capable of high electrogeneration of H₂O₂ [23]. Such carbon materials are the most used cathode materials for the production of H₂O₂ by two-electronic oxygen reduction. Bubbling oxygen or air is also required to improve H₂O₂ production. The oxygen utilization efficiency is not great and amounts to about 0.1%. Therefore, the H₂O₂ power generation system with or without low oxygen content is the most economical and promising for use [23]. Although bubbling air is feasible for this process, nevertheless, this process requires certain difficulties in the operation of the installation. In this case, to obtain H₂O₂, oxygen can be supplied by passive diffusion using gas-diffusion electrodes [15]. The reduction of oxygen to hydrogen peroxide is effective with the use of carbonaceous materials in alkaline mediums [27]. Perez etc. have developed a cell includes aerator based on Ventury for supplying atmospheric oxygen to the carbon cathode without additional energy consumption [28].

The electro-Fenton process on a modified carbon felt with external aeration allows the generation of H₂O₂ with a concentration of 472.9 mg·l⁻¹ [23]. In the work [29] electrosynthesis H₂O₂ in-situ is researched with activated graphite felt. According to the research finding, the generation efficiency of H₂O₂ was 75.3% (H₂O₂ concentration = 343,0 mg·l⁻¹, pH = 7.0), 72.3% (H₂O₂ concentration = 385,2 mg·l⁻¹, pH = 13,0), 68% (H₂O₂ concentration = 472,9 mg·l⁻¹, pH = 1.0). The generation efficiency of H₂O₂ on carbon fiber, modified graphite, and a carbon nanotube was 52; 80.0 and 32.0%, respectively [30].

The electrogeneration of H₂O₂ was researched [15] for the electro-Fenton process through the oxygen reduction reaction using polyacrylonitrile carbon fiber. The electro-Fenton process with cathodic regeneration of Fe²⁺ is able to ensure the continuity of the formation of H₂O₂ in an acidic medium. In this case, the yield of H₂O₂ directly depends on the cathode material. Despite the fact that the two-electronic oxygen reduction at carbon cathodes in highly alkaline medium is a well-known
reaction, however, the efficiency of using a cathode for the electrical generation of $H_2O_2$ by the Fenton reaction is required in acidic mediums. Hydrogen peroxide is formed at the cathode when oxygen or air is passed through the solution through the reaction (equation 12), and $Fe^{3+}$ ions in the solution can be reduced at the cathode according to the reaction (equation 13):

$$Fe^{3+} + e^- \rightarrow Fe^{2+}$$ (13)

The production of free hydroxyl radicals in the volume proceeds according to the reaction (equations 3 and 14) [24]:

$$Fe^{2+} + H_2O_2 + H^+ \rightarrow Fe^{3+} + H_2O + H^+ \cdot OH$$ (14)

Both processes contribute to an increase in the concentration of hydroxyl radicals $OH$ in the system.

Another variety is the Fenton anode process proceeds with the addition of $H_2O_2$ to the preparation. This process is carried out in an electrolyser with a consumable iron anode, divided into anode and cathode spaces. Direct and indirect oxidation are distinguished among the electrochemical processes of the organic compounds destruction based on electrochemical processes. It is possible to realize these processes in reactors of various configurations: diaphragm or membraneless. The indirect electrochemical oxidation approach is an alternative. In this case, reactive oxygen species in the form of intermediates are generated during the process in situ – $HO\cdot$, $HO_2\cdot$, $HO_2^-$ [31]. Today, this process can be considered acceptable due to its compactness, ease of automation and control, lack of precipitation and additional pollution. In this case, the deep conversion of complex organic molecules under the action of metastable oxidizing agents with the formation of simple compounds is characteristic that makes the process more efficient [31].

2. Methods
To realize the electrochemical destruction of aromatic compounds, a membraneless electrochemical reactor with a working chamber capacity of 0,4 m$^3$ and a coaxial arrangement of electrodes was used (Figure 1). X18H10T steel was used as the cathode, and a mesh oxide-ruthenium titanium electrode (ORTA) as the anode material. Preliminary preparation of the electrodes was carried out before electrolysis: the cathode was degreased with sodium carbonate and washed with distilled water. The experiments were carried out on model solutions of an organic compound belonging to the class of phenols: 1,3-dihydroxybenzene.

![Figure 1](image1.png)

**Figure 1.** Coaxial membraneless electrochemical reactor: (a) 1 – reaction vessel, 2 – blind cap, 3 – anode (ORTA), 4 – cathode (steel Chrom18Nickel110Titanium), 5 – current lead. (b) Model set (model setup) coaxial membraneless electrochemical reactor.

The process of electrochemical destruction was carried out under various conditions, varying the cathodic current density, electrolysis time, the concentration of 1,3-dihydroxybenzene, as well as pH.
The cathodic current density was varied in the range of 4–20 kA/m², the concentration of 1,3-dihydroxybenzene in the range of 0.025–1 mmol·l⁻¹, and the pH in the range of 9–14.

3. Results

Methods of indirect electrochemical oxidation consist of electrochemical generation at the cathode of H₂O₂ (equation 12) or metastable radicals as a result of an electrode reaction with oxygen according to equation 12 and subsequent chemical reactions of intermediates (HO⁺, HO₂⁻) with organic substrates in the electrolyte volume during anodic decomposition of intermediates (equations 15 and 16):

\[
\begin{align*}
H_2O_2 & \rightarrow HO_2^- + H^+ + e^- \\
HO_2^- & \rightarrow O_2 + H^+ + e^-
\end{align*}
\]

When implementing the electrochemical process of generating oxidizing agents in the case of indirect electrolysis, the choice of the configuration of the electrolyser is essential. Ways to intensify the processes of electrolysis can be implemented in electrolyser, the construction designs provide efficient mass transfer.

One approach to significantly altering the ratio and parameters of electrode processes (cathodic and anodic) is manipulating the magnitude of the current density. Geometric characteristics changing of the electrodes surfaces and the ratio of the latter allows to control the specific speed of the electrochemical processes on the electrodes up to an insignificant value (reversible electrode), implement different mechanisms of electro- and mass transfer [32]. The initial premises in this way: a significant difference in the cathode and the anode areas and the cell filling with a background electrolyte; the electrolysis current is close to the limiting currents \(i_{\text{lim}}\) of mass transfer. It is possible to neglect the migration in an electric field in the presence of a background electrolyte; the ions are present in excess compared with substances involved in electrode reactions. Obviously mass transfer will be determined by convective diffusion [33].

In traditional electrochemical reactors with a porous diaphragm and plane-parallel vertical electrodes during the electrosynthesis of an oxidizing agent, oxygen generated at the anode is transferred to the cathode associated with tangible energy and material losses. Moreover, the pores of the diaphragm are clogged with suspended substances of various origins in the process of electrolysis that subsequently leads to an increase in resistance and energy costs. In this regard, it is advisable to implement the process in a coaxial membraneless electrolyser with cylindrical electrodes [34]. The choice of this configuration of the electrolyser (Figure 1) is explained by the more symmetric distribution of the electric field with a significant difference in the cathode and anode areas. Besides, based on an analysis of the data of [34, 35], it is postulated the speed of the process at the central electrode - cathode will be much higher than the speed of the process at the peripheral electrode. According to literature data, a significant yield of hydroxyl radicals can be achieved in an alkaline medium. In this regard, the process of electrogeneration of oxidizing agents was carried out followed by destruction of 1,3-dihydroxybenzene in NaOH solutions with various concentrations of the latter. The results of the experiment can be seen in Table 2.

| Concentration NaOH, mol·l⁻¹ | Conversion coefficient 1,3-dihydroxybenzene, % |
|----------------------------|---------------------------------------------|
| 0,001                      | 97,12                                       |
| 0,01                       | 95,78                                       |
| 0,1                        | 66,56                                       |
| 1,0                        | 60,03                                       |

Nature of media impact assessment on conversion degree of organic matter was under consideration (Table 3). It is possible to conclude according to Table 3 the alkaline nature of the medium contributes to a deeper conversion of 1,3-dihydroxybenzene. Therefore all subsequent experiments were carried out at pH=11.
Table 3. Effect of the nature of the medium on the conversion coefficient of 1,3-dihydroxybenzene.

| pH | COD before electrolysis, mgO₂·l⁻¹ | COD after electrolysis, mgO₂·l⁻¹ | Fractional conversion, % |
|----|----------------------------------|----------------------------------|---------------------------|
| 7  | 286.92                           | 95.43                            | 66.74                     |
| 11 | 297.88                           | 91.42                            | 69.31                     |

To assess the destruction of 1,3-dihydroxybenzene, the effect of time was evaluated (Table 4).

Table 4. Conversion coefficient of 1,3-dihydroxybenzene during degradation.

| Organic compounds | Fractional conversion, % |
|-------------------|--------------------------|
|                   | 1 hour | 2 hour | 3 hour | 5 hour |
| 1,3-dihydroxybenzene+ NaOH | 42.58  | 51.26  | 60.04  | 61.46  |
| 1,3-dihydroxybenzene + NaOH  | 75.75  | 82.98  | 95.78  | -      |

As can be seen from the Table 4, the optimal time with a maximum degree of conversion of organic matter is 3 hours. While a subsequent increase in the duration of the process does not lead to a high degree of conversion of the substance.

4. Conclusion
The possibility of electrochemical destruction of 1,3-dihydroxybenzene in a coaxial membraneless electrochemical reactor was demonstrated for the first time. It has been established the hydrodynamic mode of operation of the membraneless coaxial electrochemical reactor contributes to the enrichment of the reactor volume by products of electrochemical generation with metastable oxidizing agents - hydroxyl and hydroperoxide radicals are capable of destroying molecules of aromatic organic compounds. The effect of reducing the concentration of organic compounds is achieved by indirect oxidation.

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