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Bicarbonate activation of hydrogen peroxide: A new emerging technology for wastewater treatment



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ABSTRACT

The serious limitations of available technologies for decontamination of wastewater have compelled researchers to search for alternative solutions. Catalytic treatment with hydrogen peroxide, which appears to be one of the most efficient treatment systems, is able to degrade various organics with the help of powerful $\bullet\text{OH}$ radicals. This review focuses on recent progress in the use of bicarbonate activated hydrogen peroxide for wastewater treatment. The introduction of bicarbonate to pollutant treatment has led to appreciable improvements, not only in process efficiency, but also in process stability. This review describes in detail the applications of this process in homogeneous and heterogeneous systems. The enhanced degradation, limited or lack of leaching during heterogeneous degradation, and prolonged catalysts stability during degradation are salient features of this system. This review provides readers with new knowledge regarding bicarbonate, including the fact that it does not always harm pollutant degradation, and can significantly benefit degradation under some conditions.

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1. Introduction

The discharge of untreated water into aquatic natural resources has resulted in widespread contamination by various pollutants, including organic dyes, endocrine-disrupting chemicals, agrochemicals, halogenated aliphatic and aromatic compounds, and sulfur and nitrogen containing organic compounds [1–6]. The increasing accumulation of these toxins requires efficient, practical and cost effective technologies for treatment of wastewater. Traditional treatment systems such as physico-chemical (flocculation, chlorination and ozonation), adsorption or thermal treatment generally require post-treatments to achieve complete de-contamination [7–8]. However, these treatments are not efficient, especially in the case of highly polluted water, and some of these methods result

in the production of even more toxic compounds. Alternatively, advanced oxidation processes (AOPs) are suitable for efficient removal of pollutants in wastewater. With versatile oxidants, these AOPs can generate different reactive oxygen species (ROS) such as hydroxyl radicals ($\bullet\text{OH}$), singlet oxygen ($^1\text{O}_2$) and superoxide anion ($\bullet\text{O}_2^-$) for complete removal of toxic pollutants [9–11]. It is well known that use of ozone or molecular oxygen as an oxidant is subject to gas/liquid solubility problems, which result in high energy consumption, while treatment with H_2O_2 appears to be a feasible protocol among AOPs [12]. The lack of a gas–liquid boundary makes use of H_2O_2 as an oxidant greatly attractive, and the degradation can be conducted under mild conditions, demonstrating its advantages when compared with wet air oxidation (WAOs). In previous studies, Fenton's reagent (Fe^{2+}) or other transition

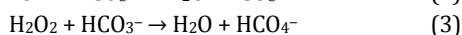
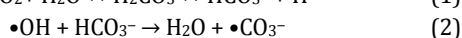
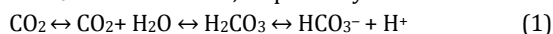
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metals such as Fe³⁺, Cu²⁺, Mn²⁺ and Co²⁺ have been widely applied as homogeneous catalysts during the degradation of pollutants [13–15]. However, the limited range of pH (2–4) and the recovery of homogeneous catalyst are serious issues in their practical application. The strategies of immobilization of transition metals over different supports have been extensively explored to minimize the catalyst loss and heavy metal pollution [16–21]. However, during advanced oxidation treatment of wastewater, the solution pH may gradually drop, encouraging catalyst leaching and deactivation. As shown in Table 1, the solution pHs of different systems clearly demonstrate their influence on catalyst leaching [22–29]. Therefore, a constant pH is important for process stability and reduced environmental damage during wastewater treatment.

Bicarbonate is highly soluble in water, relatively non-toxic and widely distributed in nature (50–200 ppm in natural water), as well as in biological systems (14.7–25 mmol/L), and exists in different forms constituting the main biological buffer Eq. (1) [30]. Normally, bicarbonate is more soluble at pH 7.0–9.0, while at above or below this range it changes to less soluble Na₂CO₃ or carbonic acid, respectively.



Notably, carbonate/bicarbonate ions are believed to change the redox properties of metals ions in ocean water, providing an opportunity to organisms to selectively take up metals [31]. Moreover, bicarbonate is reportedly involved in the structure and function of water oxidizing complex (a part of the photosystem II protein complex that oxidizes water to dioxygen), and increases the rate of electron transfer from Mn²⁺ to apo-PS II by forming ternary complex [31–33]. Apart from numerous functions in biological systems, the role of bicarbonate activation has also been studied extensively in organic reactions [34–39]. However, the application of this reagent in combination with certain AOPs for wastewater treatment is a newly emerging topic. In fact, the roles of bicarbonate in AOPs during wastewater treatment are controversial among researchers because enough data are available to support as the view of its promoting or scavenging AOPs during wastewater degradation. Nevertheless, bicarbonate still possesses some encouraging properties for its application in wastewater. For example, its buffer capacity (pH 8.0–9.0), low cost, high solubility and particularly its capability to activate H₂O₂ are attractive properties

for developing a sustainable treatment system [30,34–39]. As in AOPs, various reactive oxygen species (ROS) such as •OH, •O₂⁻ and ¹O₂ mainly controlled the degradation of pollutants. The generation of even more reactive species (HCO₄⁻, •CO₃⁻, HCO₃•) along with the aforementioned ROS was proposed in bicarbonate activated hydrogen peroxide (BAP) systems in different studies [40–45]. Although the •CO₃⁻ radical produced through (Eq. 2) is less reactive (E_o +1.78 V, pH 7) than •OH radical (E_o +2.3 V, pH 7), its occurrence in high concentration (10⁻¹³–10⁻¹⁵ mol/L) and long lifetime in reaction medium may provide it with sufficient opportunity for pollutant degradation [46]. Moreover, •CO₃⁻ radical is a very strong single electron oxidant that can react by both electron transfer and hydrogen abstraction. For example, it was reported that •CO₃⁻ radical can react rapidly with organic pesticides, sulfur containing compounds and aromatic compounds [39,46]. Similarly, peroxy-monocarbonate (HCO₄⁻) produced through Eq. (3) is a well-known two-electron oxidant that has been investigated since its discovery in 1984 [47]. HCO₄⁻ anion is considered to be more reactive than H₂O₂ (100–500 times) in different reactions depending on the substrate [47]. In view of various active oxidizing reagents generated from BAP systems and their important roles in other fields, the addition of the BAP system to wastewater treatment is attractive. Although this is a newly emerging method and many issues still need to be addressed, summarizing its progress will help researchers to explore new technologies for the future development of a sustainable and green solution for wastewater treatment.

2. BAP as a newly emerging wastewater treatment system

Different attempts have been made to solve the serious issues occurring during pollutant degradation when treating wastewater [48–57]. However, the challenges associated with these available technologies still require the development of new efficient, cost effective and sustainable treatment systems. In the recently developed BAP system, the operation conditions are generally mild, with a pH around neutral or weakly alkaline, unlike the strongly alkaline conditions commonly used during peroxide bleaching [58]. Similarly, the components of the BAP system are economical, environmentally friendly and have the potential for treatment of chemical warfare agents such as HD [bis(2-chloroethyl) sulfide], GB [pinacolyl methylphosphono-

Table 1

Leaching of supported catalysts during treatment of organic pollutants.

Catalyst	Conversion (%)	pH	Conditions	Leaching	Ref.
CuO/C	100	Not controlled	Phenol 1 g/L, H ₂ O ₂ 0.1 mol/L, catalyst 2 g/L, 180 min, 80 °C	Not reported	[22]
Cu/ZSM-5	36	Not controlled	Phenol 0.01 mol/L, H ₂ O ₂ 0.1 mol, catalyst 0.1 g/L, 180 min, 50 °C	3.2%	[23]
Cu/α-Al ₂ O ₃	100	Not controlled	Phenol 0.01 mol, H ₂ O ₂ 0.1 mol, catalyst 5 g/L, 180 min, 90 °C	65%	[24]
Al-Cu modified clays	Not reported	Buffered pH 4–5	Phenol 0.05 mmol/L, H ₂ O ₂ , 0.5–0.1 N, catalyst 0.5 g, 20 h, 20 °C	2 ppm	[25]
Cu-silicate	49.2	Not controlled	Phenol 1 g/L, H ₂ O ₂ , 0.1 mol, catalyst 0.6 g/L, 120 min, 70 °C	63.5%	[26]
Cu-Y-5	50	Not controlled	Phenol 0.01 mol, H ₂ O ₂ , 0.03 mol, catalyst 0.1 g/L, 180 min, 50 °C	4.80%	[27]
Co-diatomite	100	pH 8.2 buffered with bicarbonate	MB 60 mmol/L, H ₂ O ₂ 80 mmol/L, NaHCO ₃ 50 mmol/L, catalyst 11.4 g, treatment time 180 h	0.20 ppm	[29]
Co/RM-T	100	Not controlled	Catalyst 0.2 g, oxone 1g, phenol 25 ppm, 25 °C, 90 min	7.6%	[28]
Co/RM-NT	100	Not controlled	Catalyst 0.2 g, oxone 1 g, phenol 25 ppm, 25 °C, 90 min	7.4%	[28]

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