



Sulfate radical-based technology for the removal of 2-methylisoborneol and 2-methylisoborneol-producing algae in drinking water sources

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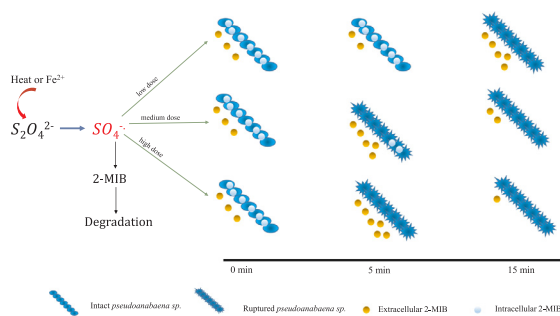
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HIGHLIGHTS

- Key parameters including temperature, PS/Fe²⁺ dose, pH and ions were studied.
- A kinetic model for 2-MIB degradation by SO₄^{•-} was developed and validated.
- SO₄^{•-} caused algae cell lysis, release and further degradation of intracellular 2-MIB.
- KMnO₄, and NaClO caused cell lysis and 2-MIB release but not able to degrade it.

GRAPHICAL ABSTRACT



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ABSTRACT

The most common odor substance found in surface water is 2-methylisoborneol (2-MIB), and this substance cannot be effectively removed by conventional water treatment processes. Persulfate (S₂O₈²⁻, PS)-based oxidation has recently received wide attention due to its promising ability to remove refractory pollutants. In this research, the degradation of 2-MIB by PS activated with iron (II) (PS-Fe²⁺) or heat (PS-Heat) was investigated. Kinetic analysis indicated that 2-MIB degradation in both of the systems (PS-Heat and PS-Fe²⁺) followed a pseudo-first-order reaction. For the PS-Heat system, a high temperature, a high PS initial concentration and weakly acidic conditions benefit the degradation of 2-MIB. Coexisting anions inhibit 2-MIB removal in the following order: CO₃²⁻ > HCO₃⁻ > Cl⁻. For the PS-Fe²⁺ system, the optimum molar ratio of PS to Fe²⁺ ([PS]: [Fe²⁺]) was 1:1. The 2-MIB degradation rate increased under acidic conditions and with the addition of citrate; however, the rate decreased with an excess Fe²⁺ dosage. In addition, this study was the first to examine the behavior pattern of algal photosynthesis activity as well as the total and extracellular 2-MIB of *Pseudoanabaena* sp. (a typical 2-MIB-producing cyanobacterium) in the PS-Fe²⁺ system. Moreover, a comparison study of the removal of 2-MIB in actual algae-containing water under the oxidation of PS-Fe²⁺, potassium permanganate (KMnO₄), and sodium hypochlorite (NaClO) was firstly carried out. The results provide useful theoretical and engineering information for treating algae-loaded water.

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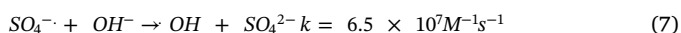
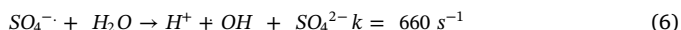
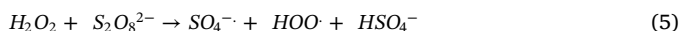
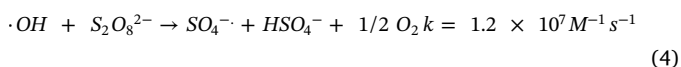
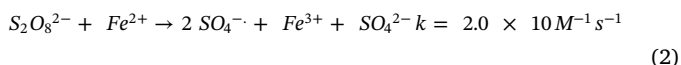
1. Introduction

Addressing taste and odor problems in drinking water is a significant challenge for water authorities worldwide [1]. 2-Methylisoborneol (2-MIB), as one of the most representative and significant odorant compounds in drinking water sources, exhibits a musty-earthly smell and is mainly produced by cyanobacteria (blue-green algae) and actinobacteria (also called actinomycetes from their former classification as fungi) [2,3]. Normally, due to 2-MIB extremely low odor threshold concentration in the nanogram per liter range, it is difficult to remove it effectively using conventional water treatment procedures alone [4].

The combination of conventional treatment with advanced oxidation processes (AOPs), such as ozone (O³) oxidation and granular activated carbon adsorption, is one of the most applied alternatives. However, this approach is not always sufficient to guarantee that the treated water concentration achieves the drinking water standard for 2-MIB (10 ng/L) [5,6]. Chlorine, chlorine dioxide, and other forms of chemical oxidation (i.e., permanganate) have also been used for taste and odor control, but these methods cannot remove 2-MIB at the nanogram per liter concentration [7,8].

Recently, a variety of AOPs based on the sulfate radical (SO₄^{•-}, E⁰ = 2.6 V) called SR-AOPs have gained much attention and are considered an innovative alternative to typical AOPs based on the hydroxyl radical (•OH, E⁰ = 2.8 V) [9–11]. Moreover, SR-AOPs can effectively oxidize a wide range of organic compounds and SO₄^{•-} has a better ability and higher selectivity than •OH to degrade several types of pollutants [10,11].

Precursor molecules such as peroxymonosulfate (PMS, HSO₅⁻) and persulfate (PS, S₂O₈²⁻) anions can be activated to generate SO₄^{•-}. The described activation methods include heat (Eq. (1)), iron (II) (Eq. (2)), UV light (Eq. (3)), and ultrasound [12–15]. Moreover, it has been shown that hydrogen peroxide (H₂O₂) can activate PS to form SO₄^{•-} and that SO₄^{•-} and •OH can promote each other's production (Eqs. (4)–(7)) [16].



Studies have been conducted on the degradation of 2-MIB by SO₄^{•-}. Xie et al. [17] found that 2-MIB could be removed effectively using the UV/persulfate process with an initial 2-MIB concentration of 40 µg/L. Bu et al. [18] reported that 2-MIB could be degraded efficiently by *in situ* generated persulfate during electrochemical oxidation. However, additional methods and the corresponding mechanisms for PS activation have not been reported as they relate to the degradation of 2-MIB. In this study, the effects of the experimental conditions on the degradation of 2-MIB by PS activated with ferrous ion (PS-Fe²⁺) and heat (PS-Heat) were systematically investigated.

In drinking water sources, 2-MIB is highly associated with the metabolism of cyanobacteria. For instance, a common cyanobacterial genus, *Pseudoanabaena*, can produce 2-MIB within the cell and partly release it into water especially after the cell's death [19]. Therefore, 2-MIB produced by *Pseudoanabaena* could be categorized into intracellular 2-MIB and extracellular 2-MIB, both of which pose critical challenges to drinking water treatment. However, there have been limited studies on the effect of SO₄^{•-} on the inactivation and rupture of cyanobacteria and the corresponding degradation of intracellular and extracellular 2-MIB. Recently, Pulse-Amplitude-Modulation (PAM) fluorometry is widely used to represent the activity of cyanobacteria by detecting the changes in fluorescence parameters (e.g. quantum yield, Φ) [20,21]. Our previous study showed that Φ provided a good explanation of the immediate and long-term impacts of oxidant on photosynthetic viability of cyanobacteria [22]. This paper systematically studied the effect of SO₄^{•-} on the inactivation and rupture of *Pseudoanabaena* sp. and the degradation of 2-MIB (including total and extracellular 2-MIB). Moreover, the removal of 2-MIB in actual algae-containing water under the oxidation of KMnO₄, NaClO and PS-Fe²⁺ was comparatively investigated.

The theoretical and engineering value of this article includes: (1) providing a solution for water treatment plants in the treatment of water containing taste and odor (T&O) compounds, (2) describing the 2-MIB concentration change pattern when using PS-Fe²⁺ to treat algae-containing water and (3) comparing the removal of 2-MIB in actual algae-containing water under different oxidants.

2. Materials and methods

2.1. Materials

All the chemicals used were at least of analytical grade, except as noted. 2-MIB (HPLC grade, ≥99.9%) and 2-isobutyl-3-methoxypyrazine (IB, HPLC grade, ≥99.9%) were purchased from Sigma-Aldrich. Sodium persulfate (Na₂S₂O₈), sodium dihydrogen phosphate (NaH₂PO₄), sodium dibasic phosphate (Na₂HPO₄), sulfuric acid (H₂SO₄), sodium hydroxide (NaOH), sodium chloride (NaCl), sodium carbonate (Na₂CO₃), sodium bicarbonate (Na₂HCO₃), ferrous sulfate

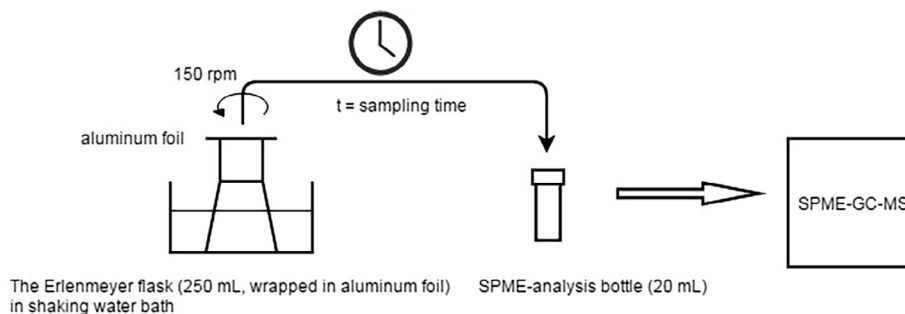


Fig. 1. The schematic diagram of experimental setup of the study as in Section 3.1.

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