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1 Oxidation of antipyrine by chlorine dioxide: reaction 2 kinetics and degradation pathway

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6 Abstract:

7 Antipyrine (ANT, phenazone), a widely used anti-inflammatory analgesic in medical
8 treatment, has been frequently detected in the aquatic environment. Chlorine disinfection process
9 is thought as an efficient way to remove ANT, however, the potential risks of chlorine disinfection
10 by-products (DBPs) such as trihalomethane (THMs) and haloacetic acids (HAAs) cannot be
11 ignored. Chlorine dioxide (ClO₂) has been adopted as an effective alternative disinfectant of
12 chlorine to reduce THMs and HAAs formation. In this work, the reaction kinetics and degradation
13 pathway of ANT with ClO₂ were studied to investigate the feasibility of using ClO₂ as oxidant to
14 degrade ANT. Experimental results demonstrated that ANT oxidation by ClO₂ followed
15 second-order kinetics, and the second-order rate constant (k_{app}) was determined to be 4.8×10^{-1}
16 $M^{-1}s^{-1}$ at neutral pH. Higher pH could accelerate the reaction when $pH < 9$, while strong alkaline
17 environment ($pH > 9$) might significantly slow down the oxidation process. Structural change
18 during the reaction was proposed with the assistance of fourier transform infrared spectroscopy
19 (FT-IR), C=C and C-N bond of ANT were vulnerable under electrophilic attack of ClO₂.
20 Degradation pathways of ANT with ClO₂ were suggested based on the main intermediate products.
21 ANT was firstly transformed into ANT-Cl through single-electron-transfer (SET) and substitution
22 reaction. Further oxidation of this intermediate product involved ring-opening reaction and
23 de-carbonyl reaction.

24 **Key words:** antipyrine; chlorine dioxide; reaction kinetics; degradation pathway

25 1. Introduction

26 Pharmaceuticals are receiving increasing attention as potential bioactive chemicals in aquatic

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