Chemical Engineering Science 60 (2005) 6360–6370

Chemical Engineering Science

www.elsevier.com/locate/ces

Modeling and scale-up simulation of U-tube ozone oxidation reactor for treating drinking water

Katsuhiko Muroyama[a,*], Masahiro Yamasaki[a], Mamoru Shimizu[a], Eiji Shibutani[a], Takeshi Tsuji[b]

[a]*Department of Chemical Engineering, Kansai University, Osaka 564-8680, Japan*
[b]*Aqua Technology Research Dept., JFE Engineering Corporation, Kawasaki City, Kanagawa 210-0855, Japan*

Received 1 November 2004; received in revised form 20 April 2005; accepted 20 April 2005
Available online 20 June 2005

Abstract

In the present study, we developed a novel simulation model of the U-tube reactor for treating drinking water, which is composed of a coaxial inner tube serving as an efficient concurrent down-flow ozone dissolver and an outer column carrying out reactions between ozone and organic substances including odorous materials (2-methylisoborneol: 2-MIB) dissolved in the raw water. We assume that the U-tube is composed of a plug flow section (inner tube) followed by a tanks-in-series section (outer bubble column) and take into account the effect of the hydrostatic pressurization on the flow and absorption equilibrium for the gaseous components including ozone and other inactive species in developing the mass balance models. An algorithm is constructed of the differential multiple mass balance equations for the inner tube sections and multiple difference mass balance equations in the series tanks in the outer column section to enable the scale-up from a pilot plant to a full-scale plant. The gas holdup and gas–liquid mass transfer coefficient were measured in a model reactor and correlated for the use of the simulation calculation. Available literature data and correlations on the rates of reactions between ozone and organic substances including odorous material 2-MIB, gas–liquid equilibrium for active and inactive gases and axial fluid mixing properties are also incorporated in the simulation calculation. The simulation results well explained the available data of the ozone absorption efficiency and the removal efficiency of the odorous material in a pilot U-tube reactor. The simulation procedure was also successfully extended to verify the performance of a full-scale U-tube reactor. It is shown that the ozone absorption is practically a single function of the gas/liquid ratio while the removal efficiency of the odorous material is a single function of the ozone dose for a specified U-tube configuration.

Keywords: Drinking water; Ozone treatment; U-tube reactor; Simulation; Scale-up; Volumetric gas–liquid mass transfer coefficient; Gas holdup

1. Introduction

Due to utrophication the increased inclusion of nitric compounds and humic substances occurs in the river water, inevitably resulting in an excess chlorination which in turn generates unwanted chlorine compounds such as trihalomethanes in the treated water. In an advanced drinking water treatment, the employment of an ozone treatment followed by an activated carbon adsorption is becoming popular in Japan (Somiya, 1989). The ozone treatment can reduce the odorous materials such as geosmin and 2-methylisoborneol (2-MIB) and the precursors to the trihalomethanes, and compensate for the chlorination treatment, thus reducing the occurrence of the total chlorinated organic compounds. Note that ozone is an environmental risk free substance because the dissolved ozone in water readily self-decomposes and reduces to oxygen (Sato, 1992).

U-tube ozonation reactor is a novel bubble column constructed of a concentric downflow tube and a outer column; in the downflow tube ozonated air or ozonated oxygen is mixed with fresh river water and an efficient dissolution of ozone is achieved under hydrostatical pressurization, while in the outer column the dissolved ozone reacts with the precursors to trihalomethanes and the odorous materials such as geosmin and 2-MIB for a sufficient residence time to reduce the concentrations of these target materials within

* Corresponding author. Tel.: +81 6 6368 0945; fax: +81 6 6388 8869.
E-mail address: muroyama@kansai-u.ac.jp (K. Muroyama).

0009-2509/$ - see front matter © 2005 Elsevier Ltd. All rights reserved.
doi:10.1016/j.ces.2005.04.064

an allowable level. Hydrodynamics of the U-tube reactor was studied on the flow pattern and the gas holdup in the concurrent downflow tube was measured and correlated by Roustan et al. (1990, 1992a,b). The liquid mixing properties in a full scale U-tube reactor were also studied by Roustan et al. (1993).

In our previous study (Muroyama et al., 1999), we proposed the design model of a U-tube ozone oxidation reactor for treating drinking water, assuming that in the inner tube, the flows of gas and liquid are both in plug flow mode and in the outer tube, the flow of the gas is still in plug flow mode while the flow of the liquid can be modeled by tanks in series. The proposed model well simulated a pilot U-tube reactor, verifying that the calculated results well predicted the data on the ozone absorption efficiency and the removal efficiency of odorous material 2-MIB.

In the present study, we developed a novel design model for the U-tube reactor considering the effect of hydrostatical pressurization on the volumetric flow and the gas–liquid equilibriums for gaseous components including ozone and other inactive species. The ozone absorption, the reaction between dissolved ozone and dissolved odorous material 2-MIB in the liquid phase, the flow and mixing of both gas and liquid phases are combined to set up the multiple differential mass balance equations in the inner tube and the multiple mass balance difference equations based on the tanks in series model in the outer column. The physical absorption of inactive gases is also considered in the mass balance equations. An algorithm is constructed of the multiple mass balance equations with appropriate boundary conditions in the two reactor sections and solved to simulate the reactor performance characteristics including the ozone absorption efficiency and the decomposition efficiency of the odorous material for the U-tube ozonation reactor treating drinking water. Available data on the reaction kinetics, gas–liquid equilibrium, absorption rates of gases and fluid mixing reported in the literature or experimentally obtained by ourselves are also incorporated in the simulation calculations. The reactor performance characteristics evaluated for the pilot plant are well predicted by the simulation calculations. The simulation model is also successfully extended to verify the reactor performance for a full-scale U-tube reactor treating drinking water.

2. Experimental

2.1. Experimental apparatus

Fig. 1 shows the experimental U-tube apparatus for measuring the hydrodynamic properties and the gas–liquid mass transfer characteristics. The dimensions of the experimental column are as follows; the outer column is 454 mm in I.D. and 3550 mm in height and the inner tube is 75 mm in I.D. and its end is opened at 100 mm above the outer column bottom. The inner tube is fitted coaxially in the outer column and the length of its straight portion well exceeds the length of the outer column. The experimental conditions for both phase superficial velocities are listed in Table 1.

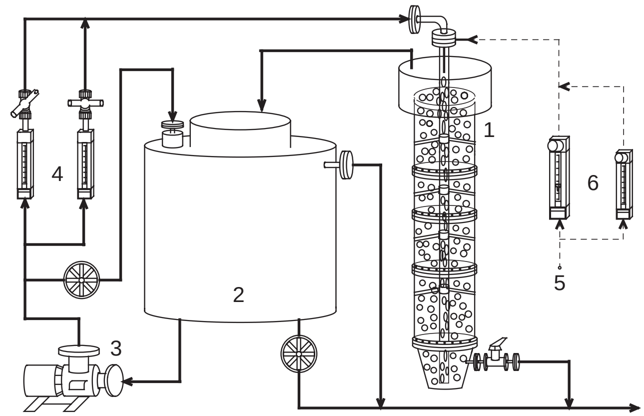

Fig. 1. Experimental apparatus for measuring hydrodynamic and mass transfer characteristics.

Table 1
Operating conditions for experimental system for measuring hydrodynamics and gas–liquid mass transfer characteristics

Inner tube	Superficial liquid velocity (m/s)	0.565–1.885
	Superficial gas velocity (m/s)	0.0113–0.339
Outer column	Superficial liquid velocity (m/s)	0.0212–0.0739
	Superficial gas velocity (m/s)	0.00443–0.0133

Two types of gas distributor were used; the first one is a single pipe of 6 mm I.D. stainless steel pipe and the second one is a four-way porous-tips nozzle, welded with sintered porous tips with average pore diameter of 40 μm.

2.2. Gas holdup measurement

The axial distribution of gas holdup in the inner tube was measured using pairs of electro conductivity cells of thin 25 mm × 60 mm square stainless steel plates which were mounted facing each other on the opposite sides of the inner wall surface at seven axial positions. After a steady state condition for the gas–liquid downflow was established in the inner tube, the inter-cell impedance was measured with 2 kHz AC by using a LCR meter (KC-547, KOKUYO Electric). In advance a correlation between the liquid holdup and the ratio of the inter-cell impedance for the single liquid flow to that for the gas–liquid flow or for the liquid–solid fluidization was obtained. The gas holdup was measured also in the outer column using a static pressure gradient method.