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The ozone mass transfer characteristics and ozonation of pentachlorophenol in a novel microchannel reactor

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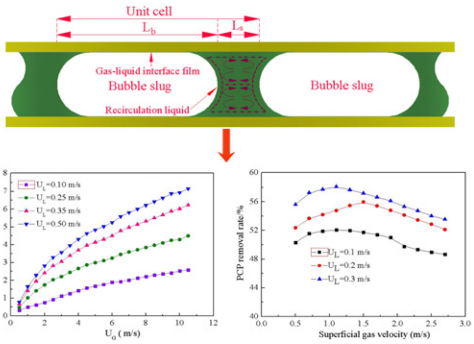
HIGHLIGHTS

- Correlations of $k_L\alpha$ with Sh_L, Re_G, Re_L, Sc for different flow patterns were proposed.
- Gas–liquid interfacial area was determined by chemical adsorption method.
- k_L at different U_G, U_L values were evaluated based on interfacial area determined.
- The reaction rate between PCP and O_3 fell in the instantaneous regime.
- Both mass transfer and residence time dominated PCP removal rate in microchannel.

GRAPHICAL ABSTRACT

The simplified geometry model of gas–liquid two-phase reaction in the microchannel reactor employed, in addition, the evolution of $k_L\alpha$ and PCP elimination rate with superficial liquid and gas velocities were shown above. The results revealed that the use of microchannel reactor could lead to noticeable enhancement in both mass transfer efficiency and the pollutant degradation efficiency.



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ABSTRACT

A "T" junction microchannel was employed to investigate the ozone mass transfer characteristics without chemical reaction taking place in the solution firstly. The correlations of the experimental $k_L\alpha$ values with dimensionless numbers: Sh_L, Re_t, Re_L, Sc for Taylor, slug–annular and churn flow regimes were proposed through a least square regression method. The gas–liquid interfacial area was determined by chemical adsorption method in the present work. The results indicated that the effect of superficial liquid velocity on the interfacial area was insignificant compared with that of superficial gas velocity. And on the basis of the interfacial area determined above, the liquid side ozone mass transfer coefficient under varied superficial gas and liquid velocities was calculated. The mass transfer rate was accelerated in the presence of PCP in solution. The reaction rate between PCP and O_3 was proved to fell in the instantaneous regime based on the experimental results of E_{O3} increasing with either liquid PCP concentration or the reciprocal interfacial ozone concentration, $(C_{O3,L,i})^{-1}$. In addition, further studies indicated that dissociation of PCP and the elevation of the average mass transfer driving forces caused by increasing pH and gaseous ozone concentration, respectively, availed the degradation of PCP in the microchannel.

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Nomenclature

H	main microchannel height (m)
B	main microchannel width (m)
L	main microchannel length (m)
U_G	superficial gas velocity (m/s)
Q_G	gas phase volumetric flow rate (m^3/s)
Q_L	liquid phase volumetric flow rate (m^3/s)
V_{mc}	volume of main microchannel (m^3)
U_L	superficial gas velocity (m/s)
U_b	the unit cell velocity (m/s)
$C_{O3,G}$	gaseous O_3 concentration (mol/L)
C	aqueous O_3 concentration of liquid inlet (mM)
$C_{A,L}$	ozone concentration in the liquid bulk (mM)
$C_{A,L,i}$	ozone concentration in the gas–liquid interface (mM)
C^*	equilibrium concentration of O_3 in the gas–liquid interface (mM)
k_L	liquid side mass transfer coefficient (m/s)
$k_{L,f}$	liquid side mass transfer coefficient for the liquid film (m/s)
$k_{L,c}$	liquid side mass transfer coefficient for the cap of the bubble slug (m/s)
k_d	self-decomposition rate constant of ozone (s^{-1})
$k_L\alpha$	liquid side volumetric mass transfer coefficient (s^{-1})
P_{O3}	O_3 partial pressure in the gas phase (atm)
He_{O3}	Henry's constant of O_3 kmol/(m^3/atm)
T	reaction temperature (K)
L_b	length of the bubble (m)
L_{uc}	length of the unit cell (m)
L_s	length of the liquid slug (m)
d_h	hydraulic diameter of the main channel (m)
d_b	hydraulic diameter of the gas bubble (m)
$D_{O3,L}$	diffusion coefficient of O_3 in water (m^2/s)
D_{CO2}	diffusion coefficient of CO_2 in water (m^2/s)
$D_{PCP,L}$	diffusion coefficient of PCP in water (m^2/s)
$k_{1,1}$	second order rate constant of O_3 with PCP (M^{-1} s^{-1})
K_{OH}^{-}	reaction rate constant between CO_2 with OH^- (M^{-1} s^{-1})
J_A	mass flux (g/m^2 s)
C_0	initial PCP concentration in solution (mol L^{-1})
SV	space velocity (s^{-1})

Dimensionless numbers

Re_L	Reynolds number of liquid phase, $Re_L = d_h U_L \rho_L/\mu_L$
Re_G	Reynolds number of gas phase, $Re_G = d_h U_G \rho_G/\mu_G$
Sh_L	liquid Sherwood number, $Sh_L = k_L d_h/D_{O3(L)}$
Sc_L	liquid Schmidt number, $Sc_L = \mu_L/\rho_L D_{O3(L)}$
Ca	Capillary number of the microchannel, $Ca = \mu_L U_B/\sigma$
Ha	Hatta number defined by $(D_{O3}k_{1,1}C)^{1/2}/k_L$
E_A	mass transfer enhancement factor
E_∞	infinite mass transfer enhancement factor
Fo	Fourier number
D^*	recalled Damköhler number

Greek letters

δ	liquid film thickness (m)
α	specific interfacial area for the liquid film (m^2/m^3)
υ	stoichiometric constant
τ_c	the gas–liquid contact time (s)
τ_s	the minimum liquid saturation time (s)
ρ_L	density of liquid phase (kg/m^3)
ρ_G	density of gas phase (kg/m^3)
μ_L	viscosity of liquid phase (Pa s)
σ	Surface tension (N/m)

Subscripts

in	microchannel inlet
out	microchannel outlet
G	gas phase
L	liquid phase
b	bubble
h	hydraulic
exp	experimental measured

1. Introduction

Pentachlorophenol (PCP), the highest chlorine-substituted species of chlorophenols, an ionizable hydrophobic organic contaminant, is widely used as fungicide, bactericide, herbicide, insecticide, molluscide, biocide, and wood preservative for a long period. Owing to the slather of PCP, it was found extensively present in air, both surface and ground waters, soil and even in the blood, urine, breast milk and other adipose tissue of human beings [1]. Due to its acute toxicity to living organisms including human beings, PCP ranked among the priority pollutant list of US Environmental Protection Agency in 1978, and was also classified as a probable human carcinogen by the US EPA [2]. Because of the toxic and recalcitrant nature of PCP, the degradation of PCP by traditional biological treatment processes is not satisfactory. Thus it is highly recommended to conduct more efficient methods for the removal of PCP from contaminated sites or degradation of it into less harmful intermediates and even complete mineralization. Among all the effective processes, ozonation process, one of Advanced Oxidation Processes (AOPs), employed for the disinfection of drinking water [3] as well as the degradation of many kinds of refractory organic compounds, was viewed as the most representative and efficient method owing to less harmful and toxic decomposition intermediates generation. Thus considerable studies involved of the degradation of organic pollutants by ozone have been conducted [4–7]. In addition, the degradation of pollutants with O_3 is a typical gas–liquid biphasic reaction, based on such point, efficient transfer of ozone from gas phase to liquid phase is of critical to the degradation of organic compounds in solution. To achieve such target, most gas diffusers and gas–liquid contactors have been introduced. The PVDF hollow fiber membrane contactor was employed by Jansen et al. [8] to investigate the reaction kinetics of the ozonation of humic substances in demineralized water. The results indicated the O_3 mass transfer resistance in the presence of humic substances was mainly dependent on the liquid phase while the resistance caused by membrane and gas phase was not significant. Besides of that, the initial ozonation reaction between O_3 and humic substances was viewed as instantaneous compared with the O_3 mass transfer. Gao et al. [9] evaluated the feasibility of the Karman contactor as ozonation unit, which was equipped with an ejector and a static Karman mixer. The results indicated that the Karman was an excellent ozonation contactor owing to its high liquid volumetric mass transfer coefficient value in a wide operating range. Mitani et al. [10] constructed an ozone reactor with tubular microporous gas diffuser for the noticeable reduction of bubble size so as to improve the overall mass transfer coefficient. And the reactor achieved one of the highest mass transfer rates compared with other bubble diffuser reactors reported in literatures. Furthermore, other gas–liquid mass transfer intensification techniques such as ejector, spray, gas-inducing microbubble, electrostatic spraying process were also employed for the enhancement of

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