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On single bubble mass transfer in a volatile liquid

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ABSTRACT

We consider single bubble mass transfer of an non-condensible gas into a volatile liquid phase in industrial conditions, as observed for example in hydrocarbons liquid phase oxidation processes. Instantaneous bubble size, shape and velocity are measured using image processing with a particle tracking method. The mass transfer rate nitrogen into hot and pressurized liquid cyclohexane is deduced from the bubble volume decrease rate and is compared to literature correlations valid under isothermal conditions. Experiments are performed in a pressurized reactor for $P = 20$ bar, $30^\circ \leqslant T \leqslant 150\,^\circ$C and bubble Reynolds number $Re = O(10–100)$. The analysis of bubble rise dynamics shows that the gas-liquid system studied can be considered as a clean system. The mass transfer results are found to follow isothermal correlations predictions excepted for ambient temperature for which liquid evaporation in bubbles is shown to be coupled with mass transfer. This phenomena seems to be a consequence of having a high Lewis number.

1. Introduction

Bubbly flows are commonly used in chemical industries for the production of basic chemicals from hydrocarbons liquid phase oxidation such as oxidation of p-xylene (dimethyl ester), cumene (phenol, acetone), toluene (benzoic acid) or cyclohexane (cyclohexanone, cyclohexanol). In that kind of applications, usually performed in bubble columns or airlifts, gaseous oxygen transfers from bubbles to the liquid bulk where the hydrocarbon oxidation takes place at a temperature large enough to enable the chemical reaction ($T = 100–200\,^\circ$C). Such elevated operating temperature condition requires to operate in pressurized bubble column to reduce evaporation of the liquid phase in the reactors. As a result, hydrocarbons liquid phase oxidation is usually conducted under moderate operating pressures ($P = 10–100$ bar). In addition, to perform mass transfer and generate efficient species mixing in the liquid phase, reactors are often operated at very large gas flow rate with gas volume fraction often larger than 20%.

As pointed out recently by the detailed review of Rollbusch et al. [1], the amount of studies on bubbly flow *under pressurized conditions* is in fact to a small percentage of the whole literature on bubbly flow. In the last thirty years, the effect of increasing operating pressure on hydrodynamic and mass transfer of bubbly flows has been mainly investigated at the scale of a bubble column. For an air-water bubbly flow in a bubble column, [2–4] found that the increase of operating pressure results in an increase of gas volume fraction for the same gas superficial velocity. The same behavior is found by [5] for a nitrogen-nheptane bubbly flow or by [6] for an air-water/ethanol system. In the same time, [2] (air-water) and [7] (nitrogen-water and nitrogen-cyclohexane) observed a decrease of the bubble size when increasing operating pressure. Mass transfer in pressurized reactor has been investigated at the scale of a bubble column using the sampling method in [3] or using optical oxygen probes in [4]. For similar gas superficial velocities, these works report an increase of the global mass transfer coefficient ($k_L a_I$) that results in both cases from the increase of gas volume fraction with operating pressure. The effect of increasing operating temperature on mass transfer in bubble column has been investigated by [8] for various organic liquids or by [9] for an air-water system. Both works measured an increase of the global mass transfer coefficient that is attributed to the liquid phase properties variation with temperature and in particular to the increase of the mass diffusion coefficient of the solute. In those previous works, the study of the mass transfer was limited to the global mass trans-

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Nomenclature

Roman symbols

a	minor semi-axis of the ellipsoid, m
b	major semi-axis of the ellipsoid, m
c_{pL}	heat capacity of liquid phase at constant pressure, J kg^{-1} K^{-1}
C	nitrogen mass concentration in the liquid phase k, mg L^{-1}
C^I	saturation mass concentration, g L^{-1}
C_D	bubble drag coefficient
C_M	bubble added-mass coefficient
C_L	bubble lift coefficient
d	bubble diameter, m
D	diffusion coefficient of nitrogen in liquid cyclohexane, m^2 s^{-1}
g	gravity constant, 9.81 m s^{-2}
H	bubble column height, m
He	Henry's constant for oxygen in water, Pa
J	surface average mass flux, kg s^{-1} m^{-2}
M_i	molar mass of species i, kg mol^{-1}
$m^G_{N_2}$	mass of nitrogen inside bubble, kg
P	operating pressure, Pa
P_G	pressure in gas phase, Pa
P^{sat}	saturation pressure of cyclohexane in gas phase, Pa
Pe	Péclet number based on the equivalent diameter and U ($Pe = ReSc$)
Pe_{max}	Péclet number based on the equivalent diameter and U_{max} ($Pe_{max} = (U_{max}/U)Pe$)
Sh	Sherwood number based on the equivalent diameter
r	bubble radius, m
Re	Reynolds number based on the equivalent diameter
S_b	bubble surface, m^2
Sc	Schmidt number ($Sc = \nu_L/D_L$)
T	operating temperature, °C
T^I	bubble surface average temperature, °C
t	time, s
U	bubble relative rise velocity, m s^{-1}
u_{max}	maximum liquid velocity at the bubble surface, m s^{-1}
V_b	bubble volume, m^3
x_i^k	molar fraction of specie i in phase k
y_i^k	mass fraction of specie i in phase k

Greek symbols

λ_k	thermal conductivity of phase k, W m^{-1} K^{-1}
μ_k	dynamic viscosity of phase k, Pa s
ν_k	kinematic viscosity of phase k, m^2 s^{-1}
ρ_k	density of phase k, kg m^{-3}
σ	surface tension, N m^{-1}
χ	aspect ratio ($\chi = b/a$)

Subscripts

G	gas phase
L	liquid phase

fer coefficient ($k_L a_I$) because the bubble populations were usually polydispersed in size and non homogeneously distributed in the bubble column volume.

Recently, for an isothermal flow, the works of Colombet et al. [10,11] and Roghair et al. [12] on monodispersed dense homogeneous bubble swarms have shown that in the limit of high bubble Péclet number, the mass transfer in a dense bubbly flow can be described by using literature correlations dedicated to single bubble for bubbly flows up to 30% of gas volume fraction. This particular behavior at high Péclet number is mainly due to a mass diffusion layer around bubbles much smaller than other characteristic length scales of the flow at such Reynolds number.

Since the early 60s, it is known that bubble dynamics and mass transfer rate can be drastically influenced by the solutal Marangoni effect resulting from surface-active contaminants accumulation at gas-liquid bubble surface. For a clean system with no impurities or surfactant at the bubble surface, the terminal rise velocity of a spherical bubble can be accurately predicted by using the relation of Mei et al. [13]. For a fully contaminated surface, the terminal rise velocity of a spherical bubble can be determined with the correlation established by Schiller and Nauman [14] for solid particle. As for mass transfer, with a clean interface system, mass transfer rate is usually based on the Higbie's penetration theory [15] with a contact time defined as the ratio of the bubble diameter to the bubble rise velocity which is also known as the Boussinesq solution for a single spherical bubble [16]. Numerical simulations [17,18] have shown that this analytical solution appears to be very accurate to describe interfacial mass transfer for a single clean spherical bubble rising in a still liquid, at large bubble Reynolds and Péclet numbers. Some corrections based on results for a single bubble have been introduced by Winnikow [19], Takemura and Yabe[17] or Colombet et al. [20] to account for the effect of a finite Reynolds number. For a fully contaminated bubble, the mass transfer rate can be described using the mass transfer relation established for a solid spherical particle [21–23]. For partially contaminated bubble surface, bubble dynamic [24] and mass transfer [25,26] can be estimated from normalised drag coefficient proposed by Sadhal and Johnson [27] based on the stagnant cap model.

Since in oxidation processes the chemical reaction in liquid phase induced non-isothermal flow conditions, it is presumed that some interactions may occure between the mass transfer of a solute and the phase change of the hydrocarbon liquid or vapor. In the literature, the study of gas/liquid transfer interaction with phase change has been mainly conducted for two different bubbly flow configurations: for bubble growth under boiling in the presence of non condensible gases and for bubble growth in supersaturated liquids. For boiling, at least since the 50's, it is admitted that dissolved gas interact with phase change mechanism affecting mainly the global heat transfer. It is shown from experimental studies that dissolved gas tend to increase the heat transfer coefficient compared to degassed liquids cases [28–30]. This enhancement of global heat transfer can be attributed to a larger number of active nucleation sites especially for nucleate boiling conditions [30]. As a consequence, to get reproducible results when studying phase change in bubbly flows, it is necessary to control the non-condensible gas concentration in the liquid phase and to degas the liquid if only phase change is studied. The dynamic of mass transfer in supersaturated liquid due to a rapid decompression has been studied by Payvar [31] or Bisperink and Prins [32] for desorption of carbon dioxide in liquids. By neglecting vapor pressure compared to noncondensible gas pressure in the bubble, Payvar found a good agreement with a model based on radi evolution from Rayleigh equation for the case of rising bubbles, while for the case of bubble growth from a cavity, Bisperink and Prins [32] found that applying the penetration theory to predict interfacial mass flux from Fick's law is sufficient to predict bubble growth rate. In both works where interaction of solute transfer and phase change should be present, the effect of the vapor pressure on the mass transfer is neglected. For the same application, Divinis et al. [33] have studied bubble growth in supersaturated solutions from