Gas-liquid mass transfer and hold-up in a bench-scale Robinson-Mahoney reactor at industrially relevant operating conditions

Jeroen Lauwaert$, Chetan S. Raghuveer, Joris W. Thybaut

Laboratory for Chemical Technology, Ghent University, Technologiepark 914, B-9052 Ghent, Belgium

$ Current address: Industrial Catalysis and Adsorption Technology, Ghent University, Valentin Vaerwyckweg 1,
B-9000 Ghent, Belgium

Introduction

At three-phase conditions Continuous Stirred Tank Reactors (CSTRs) typically provide better mixing and catalyst wetting in comparison to plug flow reactors. Hence, CSTRs constitute a potent tool for catalyst performance evaluation and intrinsic kinetics determination at such conditions [1, 2]. A specific type of fixed-basket, three-phase "Robinson-Mahoney" (RM) CSTR was proposed by Mahoney *et al.* [3]. The volumetric gas-liquid mass transfer coefficient, k_La, along with the gas and liquid hold-up, ε_G and ε_L are the most relevant parameters for assessing the reactor performance. Among others, they allow drawing an adequate picture of the actual phase distribution within the reactor. The main objectives of this work are to determine k_La, ε_G and ε_L in the RM reactor at industrially relevant conditions, *i.e.*, between 523 to 583 K and 2.5 to 5.5 MPa and inlet H_2/Halpasol™ ratio in the range of 5 to 250 $m^3\ m^{-3}$, and compare them with typically, more easily, measured values at ambient conditions, i.e., at 298 K and 0.1 MPa with a H_2-Water mixture. A correlation that allows capturing the effect of the agitator rotation speed, $N_{agitator}$, on the k_La is proposed and compared to a literature reported correlation [4]. Liquid phase hold-up data are used to propose an adequate image of the phase distribution in the reactor and compare it to the one calculated based on the feed composition.

Procedures

Intrinsic kinetics determination making use of a RM reactor at three phase conditions as well as a detailed lay-out of the set-up have been discussed in our previous work [1]. Here k_La has been determined via the so called 'dynamic gas absorption technique' where the pressure decrease as a result of agitating the mixture is monitored as a function of the time. The liquid hold-up is investigated by imposing a step change or injecting a tracer impulse in the liquid feed to the reactor. k_La and ε_L are estimated by minimizing an objective function employing a non-linear least squares algorithm [5].

Results and Discussion

k_La was found to increase from $1.22\ 10^{-2}\ s^{-1}$ to $2.14\ 10^{-2}\ s^{-1}$ with $N_{agitator}$, from 8.33 to 25 rps, see Figure 1 (left). While initially the increase is only moderate, it increases more rapidly once a threshold agitator speed has been reached. This evolution can be regarded as having to overcome an initial, minimum resistance induced by the reactor internals, such as the catalyst basket consisting of concentric metal gauzes with a very fine mesh filled with inert alumina material. Once this initial resistance has been overcome, a high turbulence regime is entered wherein k_La increases more rapidly with the agitator speed. The observed experimental behaviour was fitted by using a three parameter equation, $k_La = 1.77\ 10^{-7} * N_{agitator}{}^{3.4} + 1.21\ 10^{-2}$. This correlation is in line with earlier reported correlations based on less relevant gas liquid mixtures and 'ambient' operating conditions.

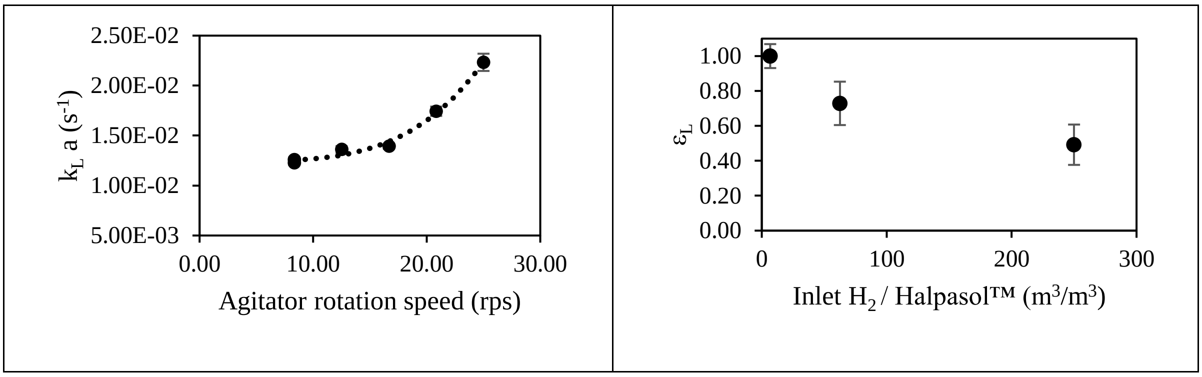

Figure 1. (Left)Evolution of the volumetric gas-liquid mass transfer coefficient, k_La, as a function of the agitator rotation speed in a three-phase bench-scale RM reactor for a H_2-Halpasol™ mixture. (Right). Liquid hold-up, ε_L, dependency in a H_2-Halpasol™ mixture for a RM reactor.

At industrially relevant conditions, a liquid hold-up of about 70% was observed irrespective of the temperature and total pressure. The volumetric inlet H_2-Halpasol™ ratio was the determining factor for the liquid hold-up: in the range from 5 to 250 m^3 m^{-3} it resulted in a decrease in liquid hold-up from 100 to 50%. At ambient conditions and with a H_2-water mixture, a more pronounced change in the volumetric inlet gas-liquid ratio, *i.e.*, from 0 to 580 m^3 m^{-3}, was necessary to achieve a similar reduction in liquid hold-up. This could be attributed to differences in fluid properties such as surface tension and viscosity rather than the operating conditions. The experimentally measured liquid hold-up always exceeded the one obtained from vapour-liquid equilibrium calculations based on the volumetric inlet gas liquid ratio. The results indicate a reactor in which the liquid constitutes the continuous phase, while the gas is present as a disperse phase, although at extremely high volumetric inlet gas-liquid ratios the gas phase may start to adopt a continuous character.

Conclusions

Experimental studies for calculating the volumetric gas-liquid mass transfer coefficient, k_La, and liquid hold-up, ε_L, in a three-phase RM reactor were performed at industrially relevant as well as at ambient operating conditions. k_La increased with increasing agitator speed, reaching a maximum value of 2.23 10^{-2} at 25 rps for H_2-Halpasol mixture at industrially relevant operating conditions. The corresponding correlation between k_La and the agitator rotation speed was in line with earlier literature proposed correlations for H_2 - gasoil/water mixture at atmospheric pressure and room temperature.

The most influential factor on the liquid hold-up was identified to be the inlet volumetric gas-liquid ratio rather than the operating temperature and pressure. The difference in the inlet gas-liquid ratio required to achieve a similar reduction in the liquid hold-up at ambient compared to industrially relevant conditions could be attributed to the differences in the properties of the fluids used in these experiments rather than to the actual operating conditions. In all experiments, high liquid hold-ups were measured, indicating the liquid as the continuous phase in the reactor rather than the gas phase as could have been expected from thermodynamic calculations.

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