CATALYTIC WET OXIDATION OF ETHANOL IN AN INTEGRAL TRICKLE-BED REACTOR OPERATED WITH LIQUID FLOW MODULATION

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Abstract– **The effect of liquid flow modulation on the performance of an integral trickle-bed reactor used for carrying out the catalytic wet oxidation of ethanol is analyzed. Particular emphasis is devoted to examine whether the induced periodic variations in conversion are affected by prolonged operation. A Pt/γ-Al_20_3 catalyst (1% w/w, mean particle diameter dp = 0.003 m) is employed for the experiments. Liquid flow is open/closed periodically by means of a programmable logic controller with given splits and at different total cycle period. The reactor operates isothermally at 70°C during the whole cycles. The time dependence of the outlet ethanol concentration in the integral TBR is examined. In addition, the average conversion obtained with periodic operation is compared to that of the steady state at different cycle periods. It is found that the history of the operation mode has influence on the attained conversion, pointing to the existence of hysteresis affecting the particle wetting.**

Keywords – **trickle-bed reactors, liquid flow modulation, periodic operation, ethanol oxidation**

I. INTRODUCTION

Trickle-bed reactors (TBRs) are extensively used for many industrial applications, mainly in the refinery industry but also for new unconventional chemical and biochemical processes. Actual applications of these reactors have extended largely in various fields, including wastewater treatment, fine chemical, biochemical and electrochemical processes (Dudukovic *et al.*, 1999). Among these new applications, the suitability of trickle-bed reactors for the catalytic wet oxidation (CWO) of organic compounds has been recently noticed (Pintar *et al.*, 1997; Horowitz *et al.*, 1999; Béziat *et al.*, 1999; Fortuny *et al.*, 1999). CWOs in three-phase reactors are particularly adequate for wastewater treatment when the concentration of organic compounds is too high or toxic to microorganisms (Matatov-Meytal and Sheintuch, 1998).

Traditionally, TBRs have been designed to operate under steady-state conditions. However, several recent studies have demonstrated that an unsteady-state (periodic) operation can yield better performance of these reactors, particularly for gas-limited reactions (Lange *et al.*, 1994; Castellari and Haure, 1995; Khadilkar *et al.*, 1999; Turco *et al.*, 2001; Boelhouwer, 2001).

For CWO processes, the low solubility of oxygen in water may lead to a control of the gaseous reactant, specially for moderate to high temperature conditions (Tukac *et al.*, 1999). Hence, high pressures are generally required. An alternative way to increase the accessibility of oxygen to the active sites arises from the possibility of having direct contact of the gaseous phase with the catalyst surface due to partial wetting of the catalysts. This can be achieved in trickle-bed reactors by low liquid flow rates or by using hydrophobic catalysts (Horowitz *et al.*, 1999). Flow modulation of the liquid phase also leads to a lower wetting efficiency for the dry period. Then, the CWO in a TBR may be improved by periodic operation.

Therefore, in this work, the effect of liquid flow modulation on the performance of a TBR used for carrying out the CWO of ethanol is analyzed. A mini-pilot scale integral TBR packed with a Pt/γ-Al_20_3 catalyst is employed for the experiments. Variations in the mean conversions obtained with periodic operation, in comparison with the attainable steady-state conversion, are determined for different modulation parameters. In addition, the time dependence of the outlet ethanol concentrations in the integral TBR is examined. Also, taking into account that only a partial oxidation of ethanol can be expected, intermediate products, like acetaldehyde and acetic acid, have been determined in the reactor effluent. This information will help to understand the influence of liquid flow modulation on the selectivity in a system of consecutive reactions.

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