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Design of a two-stage fluidized bed reactor for preparation of diethyl oxalate from carbon monoxide

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A B S T R A C T

In this study, a two-stage fluidized bed reactor (TS-FBR) was designed as a new technology to produce diethyl oxalate (DEO) from carbon monoxide (CO) based on the catalytic coupling reaction. Computational fluid dynamics (CFD) approach was adopted to obtain the detailed flow field, the bubble behaviors in the TS-FBR, and further demonstrate the feasibility of the TS-FBR used in producing DEO from CO. Furthermore, the whole preparation process of DEO from CO using the optimum TS-FBR as the central unit is simulated using an advanced software tool, realizing the proper matching of coupling reaction and regeneration reaction. As a result, the realization of the whole preparation process is presented.

Keywords: CO coupling reaction; DEO; TS-FBR; CFD; Process simulation

1. Introduction

Ethylene glycol (EG), one of the most important alcoholic compounds, is widely used, particularly in the field of polyester, synthetic fibre, paint industry, etc. (Eugene and Andre, 2001; Shoaeifar et al., 2007). Recently, a green process for the preparation of EG independent of petroleum was developed based on coal (Li et al., 2005; Meng et al., 2003; Xu et al., 2008a). It was first industrialized in China in 2009 (Qian, 2009). This green process is a promising method to convert coal to high-value chemicals and its key part is the CO coupling-regeneration circulation system, shown in Eqs. (1) and (2):

$$2CO + 2RONO \xrightarrow{\text{Coupling reaction}} (COOR)_2 + 2NO \quad (1)$$

$$2ROH + 2NO + \tfrac{1}{2}O_2 \xrightarrow{\text{Regeneration reaction}} 2RONO + H_2O \quad (2)$$

where R can be the methyl-group, the ethyl-group or the butyl-group. In this study, R is the ethyl-group. Thus RONO is ethyl nitrite (EtONO, EN), $(COOR)_2$ is diethyl oxalate ($(COOEt)_2$, DEO), and ROH is ethanol. In addition, Eq. (1), i.e. the coupling reaction of the synthesis of DEO from CO and EN, occurs on supported metal catalyst, while Eq. (2), i.e. the regeneration reaction of the preparation of EN from ROH, NO and O_2, does not need any catalyst. EN produced from regeneration reaction is then recycled as feed to the coupling reaction. This CO coupling-regeneration circulation process/system can be shown as the total reaction equation in Eq. (3).

$$2CP + 2ROH + \tfrac{1}{2}O_2 \xrightarrow{\text{Overall reaction}} 2(COOR)_2 + H_2O \quad (3)$$

The coupling-regeneration circulation process, especially the coupling reaction, attracted great interests (Chatterjee et al., 2002; Gao et al., 2002, 2005; Ji et al., 2009; Le Gall et al., 2001; Li et al., 2005; Meng et al., 2003, 2004; Qian, 2009; Wu et al., 2003; Xiao et al., 2000; Xu et al., 1995, 2008a). Most of them focused on the coupling reaction catalysts (Chatterjee et al., 2002; Gao et al., 2002, 2005; Ji et al., 2009; Le Gall et al., 2001; Wu et al., 2003; Xu et al., 1995) and mechanisms (Ji et al., 2009; Xiao et al., 2000). There are few studies on the coupling reactors, particularly on the fluid dynamics in the reactors. Recently, Wang et al. (2000a,b) developed a fixed-bed reactor model of coupling reactor of CO to DEO by using advanced software tools of Aspen and Pro-II. Xu et al. (2008a) established a

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