Insights into reaction-diffusion behaviors of n-butane partial oxidation on catalyst pellet

Yao Shi, Xuezhi Duan*

State Key Laboratory of Chemical Engineering, East China University of Science and Technology,

130 Meilong Road, Shanghai 200237, China

**Corresponding author: xzduan@ecust.edu.cn*

Highlights

- Reactor-pellet coupled model on n-butane partial oxidation is successfully established.
- CLT-1.65 pellet enhances $C_4H_2O_3$ yield by 2.8% with a 34.3% reduction in catalyst loading.
- Employing CLT-0.55/CLT-2.2 pellet grading structure can lower hot spot temperature by 21 K.

1. Introduction

Maleic Anhydride (MA, $C_4H_2O_3$) is a crucial unsaturated organic acid anhydride intermediate widely employed in industries such as pesticides, pharmaceuticals, papermaking, and so on. Industrial production of MA typically involves the oxidation of n-butane at high temperature and atmospheric pressure, where n-butane and oxygen react over the vanadium-phosphorous oxide catalysts in fixed-bed reactors. Considering n-butane exhibits low reactivity, the reaction typically occurs at temperature exceeding 400°C. On one hand, the desired product MA is prone to over-oxidation at high temperature, leading to the formation of by-products CO_2 and CO within catalyst pellets. On the other hand, as an exothermic reaction, significant reaction heat would release at the front end of reactor, resulting in the formation of local hot spots, which is detrimental to MA selectivity. Therefore, regulation of diffusion path and hot spot temperature within the reactor are crucial for improved MA selectivity and yield.

2. Methods

A steady-state two-dimensional reactor-pellet coupled model is employed for investigation, with the schematic diagram illustrated in Figure 1. A realistic packing structure of cylindrical catalyst pellets within a single tube is generated by employing the Bullet Physics Library in Blender 3.6 as depicted in Figure 1(a). Subsequently, the radial bed voidage is extracted and integrated into the reactor-pellet coupled model through interpolation functions in Figure 1(b). The coupled model predominantly comprises two scales: the reactor and the pellet scale, with coupling occurring through mass and energy fluxes. The catalyst pellet is segmented into two regions: the catalyst layer and the inert area, and reactions take place solely in the catalyst layer region. Additionally, a pellet grading reactor model is further established as illustrated in Figure 1(c). The reactor models established above are solved in the commercial software COMSOL Multiphysics.

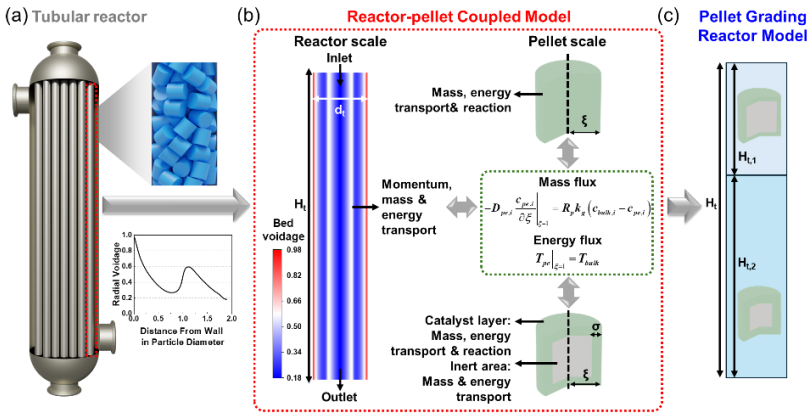

Figure 1. Schematic diagram of (a) tubular reactor, (b) reactor-pellet coupled model and (c) pellet grading reactor model.

3. Results and discussion

The reaction-diffusion behaviors of 5.5 mm catalyst pellets within the reactor were first investigated by employing pellet-reactor coupled model. At the hot spot (z = 0.34 m) as illustrated in Figure 2(a), $C_4H_2O_3$ concentration exhibits an initial increase followed by a subsequent decrease along the radial direction of the pellet. The enlarged pattern of $C_4H_2O_3$ concentration in Figure 2(b) also indicates that n-butane was over-oxidized to undesired CO_2 and CO in the interior zone of the pellets, and it is imperative to reduce the catalyst layer thickness (CLT) to inhibit over-oxidation for improved $C_4H_2O_3$ yield.

To investigate the effects of catalyst layer thickness, the $C_4H_2O_3$ yield and hot spot temperature of pellets with varying catalyst layer thicknesses were compared in Figure 2(c). Compared to CLT-5.5 pellet, CLT-1.65 pellet demonstrates the 34.3% reduction in catalyst loading, with only 5.5% decrease in C_4H_{10} conversion, while it exhibits a significant improvement of 6.9% in $C_4H_2O_3$ selectivity, thereby achieving the highest $C_4H_2O_3$ yield of 37.6%. However, the hot spot temperature of CLT-1.65 pellet is still high. To strike a balance between the lower hot spot temperature and higher $C_4H_2O_3$ yield, the pellet grading approach loaded with CLT-0.55/CLT-2.2 pellets were proposed in Figure 2(d). As illustrated in Figure 2(e), compared to CLT-5.5 pellet, employing CLT-0.55/CLT-2.2 pellets can enhance the $C_4H_2O_3$ yield by 2.6% while simultaneously reducing the hot spot temperature by 21K.

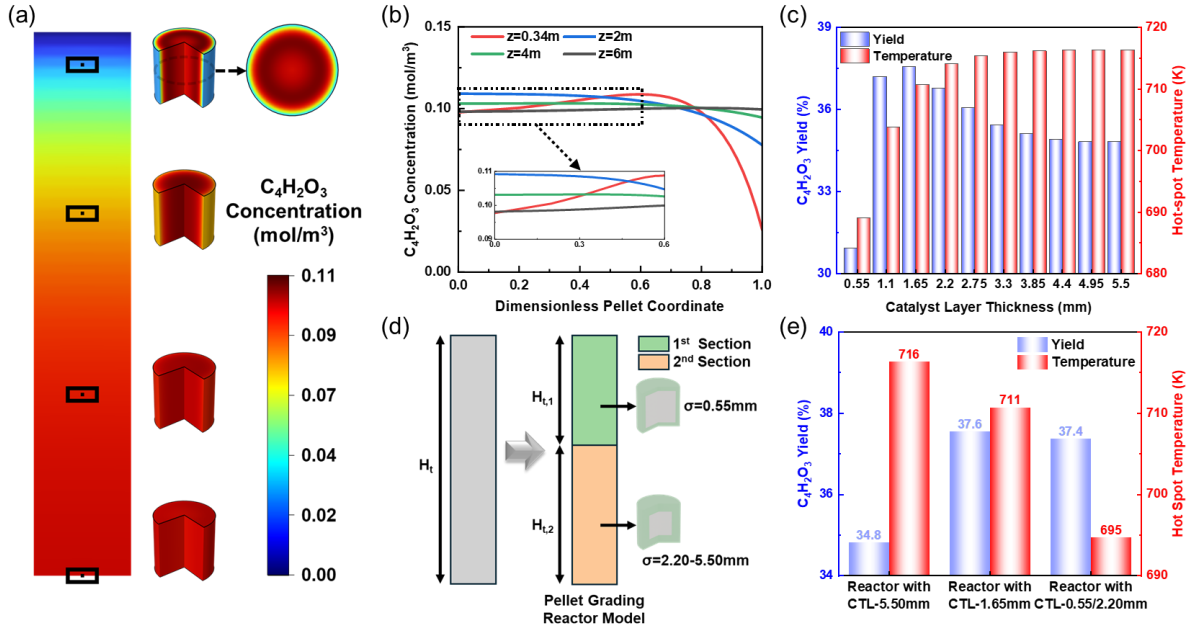

Figure 2. (a) $C_4H_2O_3$ concentration profile at reactor and pellet scales, (b) $C_4H_2O_3$ concentration along the pellet radial direction, (c) $C_4H_2O_3$ yield and hot spot temperature with respect to catalyst layer thickness, (d) schematic diagram of pellet grading reactor model, and (e) comparison of $C_4H_2O_3$ yield and hot spot temperature.

4. Conclusions

In this work, the reactor-pellet coupled model and pellet grading reactor model have been successfully established to investigate the reaction-diffusion behaviors of n-butane partial oxidation process. Reducing the catalyst layer thickness of pellet can effectively inhibit the over-oxidation of n-butane, and CLT-1.65 pellet stands out with the highest $C_4H_2O_3$ yield. Employing CLT-0.55/CLT-2.2 pellet grading packing structure can significantly reduce the hot spot temperature while keeping the $C_4H_2O_3$ yield almost unchanged.

References

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Keywords

N-butane partial oxidation; hot spot temperature; mass transfer.