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# Residence time distribution and reaction rate in the horizontal rotating foam stirrer reactor

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## HIGHLIGHTS

- A multistage horizontal rotating foam stirrer reactor is used for hydrogenation purposes.
- The reactor model consists of stirred tanks in series with backflow and dead volume.
- The reactor exhibits a dead volume of 23% and a backflow ratio between 1 and 2.
- Plug flow behavior is achieved when six stages are used.
- The selectivity of the hydrogenation is enhanced due to plug flow behavior.

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## ABSTRACT

The performance of a multistage horizontal rotating foam stirrer reactor in semi-continuous operation was studied for the selective hydrogenation of functionalized alkynes to alkenes, an important process in the fine chemicals industry. This new type of multiphase reactor consists of a horizontal vessel compartmentalized by vertical baffles and equipped with an impeller in each compartment. The impeller is a donut-shaped foam block, which is also used as catalyst support. The advantage of this reactor configuration compared to batch slurry reactors is the better catalyst handling, since the catalyst is fixed on the stirrer. In addition, a higher selectivity towards the desired product is achieved as a result of a narrower residence time distribution. A reactor model consisting of stirred tanks in series with backflow and dead volume was used to describe the liquid flow behaviour. The effects of liquid flow rate, backmixing and number of stages for the hydrogenation reaction are discussed, and optimal operation conditions are suggested.



## 1. Introduction

Catalytic gas–liquid–solid reactions occur in a broad range of industrial application areas and constitute the basis for the manufacturing of a large variety of intermediate and consumer-end products. These reactions are often carried out in stirred tank slurry reactors where small catalyst particles (typically $<50\ \mu m$) are suspended in the liquid medium through which gas is dispersed. The small dimensions of the catalyst particles provide catalyst utilization factors that approach unity. However, additional cost for catalyst separation and loss of active phase from the catalyst due to attrition are the main drawbacks (Cybulski and Moulijn, 2006). Reactor designs using structured catalysts have been considered as potential replacement for the traditional slurry reactors. Examples are the monolithic stirrer reactor (Hoek et al., 2004; de Lathouder et al., 2006; Boldrini et al., 2012; Joshi and Parikh, 2008; Albers et al., 1998; Ramos-Fernandez et al., 2011) and the rotating foam stirrer reactor (Tschentscher et al., 2010a, 2010b, 2011b, 2012, 2011a; Leon et al., 2011, 2012a, 2012b) in which monolithic and solid foam structures are mounted on the stirrer shaft as impeller blades, respectively. These structured catalysts provide a large liquid–solid interfacial area and short diffusion paths within the catalyst, while at the same time avoiding the necessity of filtration of small catalyst particles. As a result of the application of the regular structure, the scale-up to industrial relevant size is considered to be easier (Roy et al., 2004). Compared to monoliths, the mixing in solid foams is strongly increased, especially in directions perpendicular to the flow, due to the continuous disruption of the flow pattern by the tortuous three-dimensional structure of the foam (Scheffler and Colombo, 2005).

The rotation of a donut-shaped foam block has shown enhanced mass transfer rates compared to the foam blade stirrer, resulting in a more efficient use of the catalyst. Moreover, the

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surface area available for catalyst deposition is significantly increased (Tschentscher et al., 2010a, 2010b). Examples of applications where the foam block stirrer have been proposed for use in heterogeneous catalysis include hydrogenation (Leon et al., 2012b) and oxidation (Tschentscher et al., 2011a) reactions as well as fermentation reactions (Amin and Doelle, 1989). Although for this vertically oriented foam stirrer reactor higher productivities have been achieved when compared to the conventional slurry reactor, the horizontal rotating foam stirrer reactor has been proposed to further increase the supply of the gas reactant to the foam catalyst surface, which is often the rate limiting step in the above mentioned reactions. In this configuration, a donut-shaped foam block is mounted on a horizontal shaft and the reactor is partially filled with liquid (Fig. 1a). Due to the centrifugal force at high rotational speeds, the gas is separated from the catalyst surface only by a very thin liquid film. The alternating contact of the solid foam with the gas and liquid phases enhances the rate of gas–solid mass transfer (Leon et al., 2013). This concept has been previously applied in the biotechnology area (Karapantsios et al., 1993). A horizontal axially rotated packed bed, the so-called rotating biological contactor, has been used as a gas–liquid contact device for the biological treatment of wastewater either from food production plants (Borghei, 1981) or from oil and chemical facilities (Hirata and Hosaka, 1991). The rotating bed is partially immersed in liquid, while the rest is exposed to the atmosphere. Air is trapped in the packing, thus facilitating liquid aeration and bio-oxidation. The results showed that the organic removal efficiency of the horizontal axially rotated packed bed is well placed in comparison to most efficient aerobic processes.

In previous work (Leon et al., 2013), we have presented the hydrodynamics and the external mass transfer characteristics of the horizontal rotating foam stirrer reactor. This paper aims to demonstrate the catalytic application of such foam stirrer configuration in the fine chemical industry. The selective hydrogenation of a functionalized alkyne (3-methyl-1-pentyn-3-ol) to alkene is chosen as a model reaction. Since plug flow conditions along with high intensity mixing are desired for achieving high conversion, selectivity and mass transfer rates, a multistage horizontal rotating foam stirrer reactor is used in semi-continuous operation. The horizontal reactor vessel is divided in several compartments by vertical donut-shaped baffles and multiple foam block structures are mounted on the same stirrer shaft (Fig. 1b). Key issues in designing multistage mixing tanks include tank volume, length/diameter ratio, interstage baffle opening size, and inlet–outlet locations. The reactor volume is based on the desired mean residence time. The aspect ratio is determined on the basis of the desired number of stages. The interstage baffle opening must be sized based on acceptable level of exchange flows between stages. A larger opening increases the exchange flow and thus, the backmixing. Increasing the backmixing brings the multiple stages close to a single CSTR. 15% lower efficiency has been reported for a backflow ratio of 10 in hydrogenation reactions performed in multistage agitated contactors (Zhang et al., 2005, 2010; Pan et al., 2009). The inlet and outlet should be located at opposite ends of the mixing tank for best plug flow conditions (Hemrajani and Tatterson, 2004). This paper presents the residence time distribution in the horizontal rotating foam stirrer reactor as a function of the rotational speed, liquid flow rate and the number of compartments. Based on the RTD data, the macromixing behavior of the reactor is described and a model to estimate the properties of the effluent during the selective hydrogenation of 3-methyl-1-pentyn-3-ol is presented.

## 2. Experimental section

### 2.1. Horizontal rotating foam stirrer reactor

Experiments in a single stage horizontal rotating foam stirrer reactor were performed in a 1.4 l glass reactor with an inner diameter of 200 mm and a length of 45 mm. As foam block stirrer, cylindrical shaped aluminum foams with an inner diameter of 13 mm, an outer diameter of 150 mm and a thickness of 10 mm were used. The aluminum solid foams were purchased from ERG Aerospace (for RTD measurements) and from Alantum (for hydrogenation reactions). The foam pore size used was 20 ppi (pores per linear inch). The reactor was equipped with four equidistant baffles placed inside the reactor wall (baffle dimensions: 2 mm of thickness, 10 mm of width, 45 mm length) as well as four tilted baffles placed on each reactor lids as shown in Fig. 1a (baffle dimensions: 1 mm of thickness, 13 mm of width, 72 mm length, tilted angle 35°). The tilted baffles reduce the dry sections observed previously by tomography studies (Leon et al., 2013) by forcing the liquid to flow into the center of the foam packing, leading to high mass transfer rates. The multistage horizontal rotating foam stirrer reactor was built by repeating two or four times the single stage above described (Fig. 1b). The stages were separated by vertical donut-shaped baffles with a central circular opening of 25 mm inner diameter and a thickness of 2 mm. The interstage baffle opening size was chosen in order to minimize the backmixing. In multistage agitated gas–liquid contactors equipped with Rushton turbines, low backmixing is typically achieved when the baffle central opening area is less than 2% of the cross section area of the reactor column (Vidaurri and Sherk, 1985; Magelli et al., 1982; Zhang et al., 2005, 2007; Takriff et al., 2000; Xu et al., 2005; Vidaurri et al., 1983). The tilted baffles described above on the reactor lids were also placed on the vertical donut-shaped baffles. Each compartment was provided with a foam block stirrer mounted on the same shaft. The power consumption was calculated using the voltage and the current measured by an ES300-series power supply (Delta Elektronika B.V.).

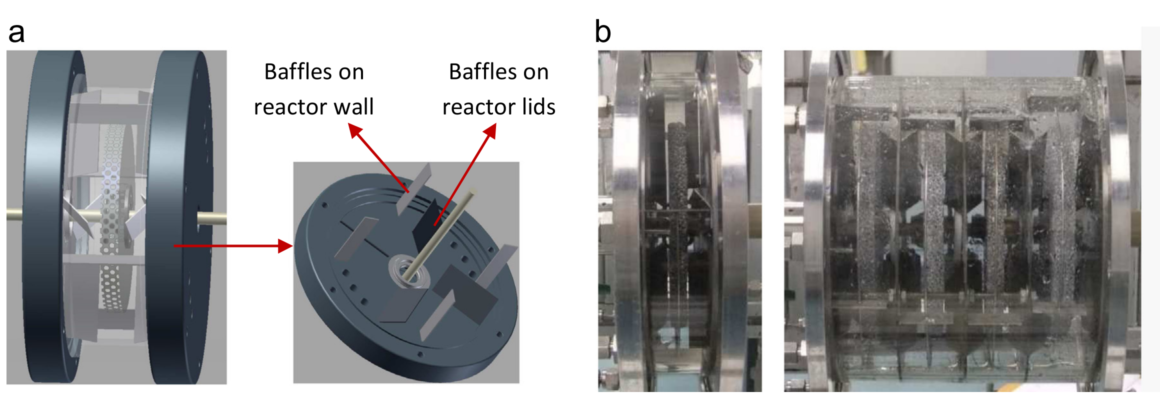


**Fig. 1.** Horizontal rotating foam stirrer reactor: (a) single stage, (b) multiple stages.