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Narrow residence time distribution in tubular reactor concept for Reynolds number range of 10–100

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ABSTRACT

For chemical reactions, which require residence times of several hours, enhanced heat transfer, or narrow residence time distribution (RTD), good radial mixing combined with poor axial mixing in laminar flow regime has long been desired by industry and R&D. The main goal of this work is to obtain the narrowest RTD curve in a continuously operated reactor at Reynolds numbers smaller than 100. By using a stepwise method the most promising reactor type was chosen to meet the requirements. Design parameters of this reactor, the coiled flow inverter (CFI), were characterized and their effects on RTD were experimentally investigated. Design of CFI includes several straight helix modules, where the tubular reactor is coiled around a coil tube. After each straight helix module, the coil direction is changed by a 90°-bend. As a starting point for designing a CFI reactor for specific applications, the “best performance” design space diagram was investigated. Regarding narrowing RTD, the diagram gives the user the design space for the CFI reactor, which leads to the best performance. The most significant design parameter regarding a narrow RTD was experimentally determined as number of bends. By using a CFI design consisting of 27 bends at volume flow rate of 3 mL/min, which corresponds to Reynolds number of 24 and mean residence time of 2.6 h, a Bodenstein number over 500 was achieved. Beside its narrow RTD behavior, CFI is a compact and cost-efficient reactor concept, which is flexible to scale-up and implement for different processes, even for single-use applications.

1. Introduction

In chemical industry many processes are driven in continuous operation mode because a continuously operated process may serve for constant product quality, better process control and higher production rates compared to batch operation mode. In the case of continuous operation mode, most reaction systems require efficient heat transfer and a narrow residence time distribution (RTD) to achieve the desired reaction rates, yield, selectivity, and product quality. Especially for kinetically controlled reaction systems, where side reactions or consecutive reactions may take place, a narrow RTD is essential. Consequently, good radial mixing combined with poor axial mixing is desired within the continuously operated reaction system because improper mixing can result in low product quality (Vashisth and Nigam, 2008). Within this work, the combination of good radial mixing and poor axial mixing is defined as effective mixing for an easier speech.

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Nomenclature

A_t	absorbance value [mAU]
A_0	maximum absorbance value [mAU]
Bo	Bodenstein number [–]
D_{ax}	axial dispersion coefficient [$m^2\,s^{-1}$]
d_c	coil diameter [m]
d_{ct}	coil tube diameter [m]
d_i	inner tube diameter [m]
d_o	outer tube diameter [m]
Dn	Dean number [–]
$E(\theta)$	dimensionless exit-age distribution curve [–]
$F(\theta)$	dimensionless concentration [–]
f_c	friction factor of a CFI reactor [–]
f_s	friction factor for laminar straight tube [–]
L	reactor length [m]
p	pitch distance [m]
r_c	coil radius [m]
Re	Reynolds number [–]
R_w	relative width [–]
T	Torsion parameter [–]
T^*	modified Torsion parameter [–]
t	time [min]
$\bar{\upsilon}$	average flow velocity [$m\,s^{-1}$]
Greek	
η	dynamic viscosity [$kg\,m^{-1}\,s^{-1}$]
Θ	dimensionless time [–]
ρ	density [$kg\,m^{-3}$]
τ	mean residence time [min]

When using a continuously operated reactor or mixer, it would be advantageous to have a scalable system with a wide range for applications, from laboratory scale with its tiny flow rates (few mL/min) up to container pilot plants or large-scale manufacturing processes (Bieringer et al., 2013). That system could be used from the early phases of process development without changing process conditions. In most cases microreactors are used for this field of application as shown by Kockmann et al. (2008) and Roberge et al. (2009). Due to the fact that most microreactors are quite costly, those systems may not be the first option, if an application with a long reaction or mixing time of 2 h or more is required. Examples of applications for reactions with long residence times are discussed by Roberge et al. (2009) (type C reactions) or Johnson et al. (2012) (continuous reaction with 12 h of residence time).

For applications that need long residence times at continuous operation mode, a cost efficient reactor implies low Reynolds numbers, which will result in smaller reactor length. Therefore, another reactor concept is investigated, which should be suitable for continuous reaction and mixing applications, especially for long residence times from minutes to hours with a narrow RTD at low Reynolds number ($Re < 100$). Furthermore, an easy scale-up, a wide operation window regarding flow rates and component system, low investment costs, and a compact design should be characteristic for the reactor concept. If possible, the reactor should be built with decisive single-use components for applications in pharma and fine chemicals to reduce cleaning effort and quality control.

This paper presents a method how a suitable reactor for the above defined requirements is found based on a short description of the state-of-the-art-technologies. Subsequently, the most promising reactor concept is defined in detail and a diagram is presented that visualizes the "best performance" design space of the reactor for a specific flow rate in terms of narrow RTD. An introduction to the design of experiments and a detailed experimental characterization of the reactor including discussion of the results follow before the article ends up with a conclusion.

2. Investigation of a suitable reactor concept

As described in the previous part, the aim of this paper is to design a continuously operated reactor, which provides a narrow RTD at laminar flow regimes ($Re < 100$) with long residence times of up to several hours. Furthermore, the reactor should combine the attributes of compact design, low investment costs, easy scale-up, wide operating window and single-use-technology.

Regarding a continuous flow reactor, one might consider to use a straight tube reactor as a first idea. In the case of laminar flow ($Re < 2300$) the characteristic parabolic velocity profile is formed in the straight tube reactor. The Reynolds number Re is calculated according to Eq. (1) where ρ is the density, $\bar{\upsilon}$ is the average flow velocity, d_i is the inner tube diameter and η is the dynamic viscosity.

$$Re = \frac{\rho \cdot \upsilon \cdot d_i}{\eta} \tag{1}$$

The parabolic velocity profile leads to a very broad RTD because the fluid elements nearby the wall have low velocities and thus very long residence times compared to the fast flowing fluid elements in the center of the tube. Due to the broad RTD, the straight tube is not a suitable reactor to provide the above defined requirements. However, by decreasing the inner tube diameter and/or increasing the volume flow rate, it is possible to reach turbulent flow regime ($Re > 2300$). The advantage of turbulent flow is that the strong radial mixing can be maintained within the tube. Due to the strong radial mixing effects, RTD behavior in turbulent flow regime is close to an ideal plug flow reactor. The RTD behavior of an ideal plug flow reactor is the narrowest RTD that can be maintained by using a continuous mode operation (Fogler, 2006). Strong disadvantages are the long tube and high pressure losses in the case of long residence times due to the very high flow velocities. Consequently, a reactor with turbulent flow regime fails the requirement of a compact reactor design in case of long residence time.

An alternative to achieve a narrow RTD at laminar flow condition with its low flow velocities and smaller reactor size is to enhance the mixing within the reactor. Different reactors can result in identical RTD behavior, which is caused by similar types of mixing. Therefore a method was formed, which takes a look at different flow reactor concepts. By elimination of inconvenient concepts, the method leads to the most suitable laminar flow reactor that provides RTD behavior close to the ideal plug flow reactor as well as compact design and easy scale-up. Fig. 1 reflects the overall representation of this approach, which was built up during the literature survey.

In Fig. 1 each layer of the pyramid model consists of different names of methods. Starting from the bottom layer of the pyramid, which includes the two broad categories of mixing, the pyramid model ends up at the top layer with the most