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Review

Chemical process intensification from the viewpoint of vortex dynamics

Hayato Masuda[1,*], Takafumi Horie[2], Hiroyuki Iyota[1] and Naoto Ohmura[3]

[1]Department of Mechanical and Physical Engineering, Osaka City University, 3-3-138 Sugimoto, Sumiyoshi-ku, Osaka 558-85858, Japan; [2]Department of Chemical Engineering, Osaka Prefecture University, 1-1 Gakuen-cho, Naka-ku, Sakai, Osaka 599-8531, Japan; [3]Department of Chemical Science and Engineering, Kobe University, 1-1 Rokkodai, Nada-ku, Kobe, Hyogo 657-8501, Japan.

ABSTRACT

From the viewpoint of chemical engineering, a vortex has many useful functions, such as mixing, transport, accumulation, and separation. To maximize these functions, continuous chemical reactors using vortex dynamics, such as Taylor–Couette flow reactor, oscillatory baffled reactor, and slug flow reactor, have been proposed for chemical process intensification. These reactors have been developed with a focus on efficient heat and mass transfer because their residence time distribution can be precisely controlled. This review introduces the Taylor–Couette flow reactor with modified geometry to maximize the Taylor vortex flow function for process intensification with multiphase or reactive liquid flow. A ribbed inner cylinder is utilized to intensify gas-liquid mass transfer for mixing in biopolymer processing. In addition, enhancement of the mixing of shear-thinning fluid by a conical Taylor–Couette flow is introduced. This review will inspire chemical engineers to develop new types of reactors that exploit vortices in the future.

KEYWORDS: vortex dynamics, process intensification, Taylor–Couette flow, enhancement of transport rates, continuous reactor.

*Corresponding author
hayato-masuda@osaka-cu.ac.jp

INTRODUCTION

People stir casually while adding milk to coffee or during stew cooking, to achieve uniform temperature or food ingredient distribution. In these examples, the objective of stirring is to improve the mixing and heat and mass transfer, that is, transport processes. Stirring generates vortices or eddy flows. Generally, vortices have organized structures; on the other hand, eddies consist of various large and small vortices, such as turbulent flow. The vortex or eddy flow is a powerful tool for enhancing transport rates in various manufacturing processes, as well as in cooking.

In chemical industries, the vortex flow has been traditionally utilized for various unit operations such as mixing, reaction, heat and mass transfer, separation, and distillation. In a traditional stirred vessel, used in many mixing or reaction processes, eddies are observed under turbulent conditions. The turbulent eddy flows in stirred vessels have been investigated by many researchers from experimental and numerical viewpoints [1-3]. The characteristics of eddy flow depend on the geometry of the impeller, vessel, and baffles. The turbulent eddies in stirred vessels promote mixing [4], homogenization [5], and heat and mass transfer [6, 7]. The vortex flow plays important role in mass transfer enhancement in bubble columns for gas-liquid reactions [8, 9]. A heat exchanger with a vortex generator has been utilized for heat transfer enhancement [10]. The vortex or eddy flows have long been utilized to improve chemical processes.

In recent years, the effectiveness of vortices has attracted attention in chemical process intensification as a new research focus in chemical industries. A detailed introduction of process intensification is referred to in several books [11, 12]. In short, it is a process design strategy aimed at significantly reducing the size of the process and dramatically improving its performance while reducing energy consumption. Reay *et al.* [11] and Boodhoo and Harvey [12] summarized the (i) enhancement of transport processes, (ii) uniform processing, and (iii) continuous operation as key topics for achieving process intensification. As mentioned previously, the enhancement of transport processes can be achieved by eddy or vortex flows. However, turbulent eddies are not always suitable for process intensification because of the limitation of energy consumption. Thus, the utilization of vortex flows instead of eddies is preferable for realizing process intensification.

The Taylor–Couette flow consists of structural vortices between coaxial rotating cylinders, which is one of the most effective solutions for process intensification. As is well known in the fluid mechanics [13, 14], the flow between coaxial cylinders, with the inner one rotating, exhibits a cascade type of flow transition with an increase in Reynolds number, *Re,* in the circumferential direction. Above the critical *Re* value, Re_{cr}, the counter-rotating toroidal vortices spaced regularly along the axis, known as Taylor vortices, are generated, as shown in Fig. 1 [15]. The toroidal motion of Taylor vortices within the Taylor cell enhances mixing and heat and mass transfer. Because mass transfer between pairs of Taylor cells is prevented by inflow boundaries, a pair of Taylor cells is regarded as a well-mixed stirred vessel. If a small axial flow is added, each cell operates in a single file without breakdown. Therefore, a narrow residence distribution is obtained. Because these features follow the three strategies mentioned previously, the Taylor–Couette flow system is suitable for process intensification.

Until now, the Taylor–Couette flow has been applied to various processes with the aim of improving process efficiency. For example, Kataoka *et al.* [16] used a Taylor–Couette flow reactor for the emulsion polymerization process of styrene and showed that the molecular weight and size distribution of the latex particles can be precisely controlled by the flow conditions. Sczechowski *et al.* [17] and Dutta and Ray [18] utilized the Taylor–Couette flow to overcome the limited light penetration depth into a slurry in the photocatalytic reaction process. In addition, the Taylor–Courtte flow has been applied to food processing, for example enzymatic reactions [19, 20], UV inactivation of *Escherichia coli* [21], and thermal sterilization [22]. These results show that the Taylor–Couette flow improves performance compared with existing processes. However, the purpose of process enhancement is not just process improvement, but dramatic performance improvement. To achieve "intensification" beyond "improvement," it is important to maximally strengthen the function of the Taylor vortex. For example, it is known that modifying the shape or surface roughness of the outer or inner cylinders is effective for mixing enhancement [23] or drag reduction [24, 25]. This paper reviews examples of the enhancement of transport rates toward process intensification by modifying the inner cylinder shape.

Modification of inner cylinder shape

The Taylor–Couette flow is stable but not robust. The Taylor cell structure often becomes unstable in a multiphase or reactive flow with complicated changes in physical properties. Richter *et al.* [26, 27] first proposed a ribbed inner cylinder to immobilize and stabilize Taylor cells. A pair of Taylor cells is separated by ribs, as shown in Fig. 2 [28]. Generally, the mixing condition within Taylor cells under laminar Taylor vortex flow is not sufficient for a high-Schmidt-number system. Thus, an operation with a slightly higher *Re* is required to improve the mixing within Taylor cells. However, the increase in *Re* induces wavy motion of the inflow boundary; consequently, mass transfer between pairs of Taylor cells is promoted. Richter *et al.* [26, 27] showed that Taylor cells are stabilized and immobilized by ribs, even under a relatively high *Re* regime, while maintaining excellent mixing performance within the Taylor cells. This immobilization of Taylor cells by ribs is also effective for the above-mentioned multiphase or reactive flow systems.

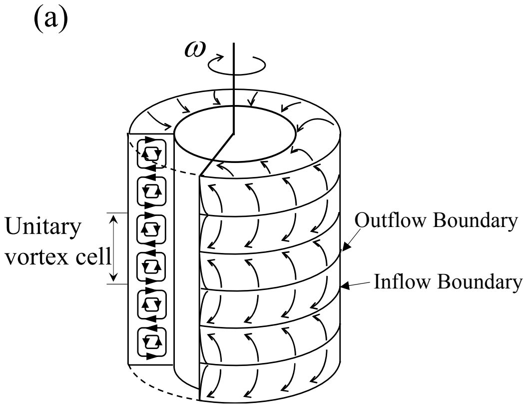

Fig. 1. Taylor–Couette flow: (a) Schematic diagram and (b) flow visualization with Kalliroscope AQ-1000 flakes [15]. With respect to (b), the left and right figures show the laminar Taylor vortex flow (*Re* = 212) and wavy vortex flow (*Re* = 994), respectively. The black bands in (b) indicate inflow boundaries.

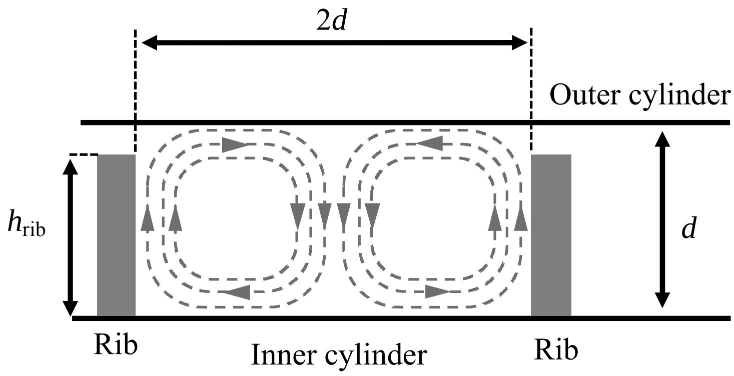

Fig. 2. Cross-sectional view of a pair of Taylor vortices between ribs [Reprinted from Matsumoto, M., Masuda, H., Hubacz, R., Horie, T., Iyota, H., Shimoyamada, M. and Ohmura, N. 2021, Chem. Eng. Sci., 231, 116270 with permission from Elsevier].

Intensification of gas–liquid flow processes

The Taylor-Couette flow has been utilized as a fermenter for bioprocess intensification [29]. Such processes incorporate an aeration operation. The mass transfer characteristics and flow dynamics of gas–liquid two-phase Taylor–Couette flow have been reported by many researchers [30-37]. However, the fundamental problem of rising bubbles by buoyancy significantly affecting the structure and dynamics of the Taylor–Couette flow has not yet been addressed. Masuda *et al.* [38] utilized an inner cylinder equipped with ribs to suppress the buoyancy effect in a gas-liquid two-phase flow. Fig. 3 presents photographs of the flow pattern when the rotational speed of the inner cylinder, *n*, is 190 rpm, and the volumetric air flow rate, V_G, is 5.0(10−6 m3/s. As shown in Fig. 3, with increasing rib width (hrib), the cell boundaries were found more clearly. This means that the Taylor–Couette flow is stabilized by the ribbed inner cylinder owing to the enhancement of the centrifugal force. A stabilized Taylor cell can capture more bubbles than a normal cylinder. Fig. 4 shows the mean residence time of the bubbles under each condition. According to Djéridi *et al.* [30] and Atkhen *et al.* [31], the phenomenon of capturing bubbles within the Taylor cells is observed in the high Re regime. The advantage of the ribbed cylinder is that the Taylor cells capture more bubbles even under relatively low Re conditions. Furthermore, as shown in Fig. 5, the specific interfacial area, which is one of the most

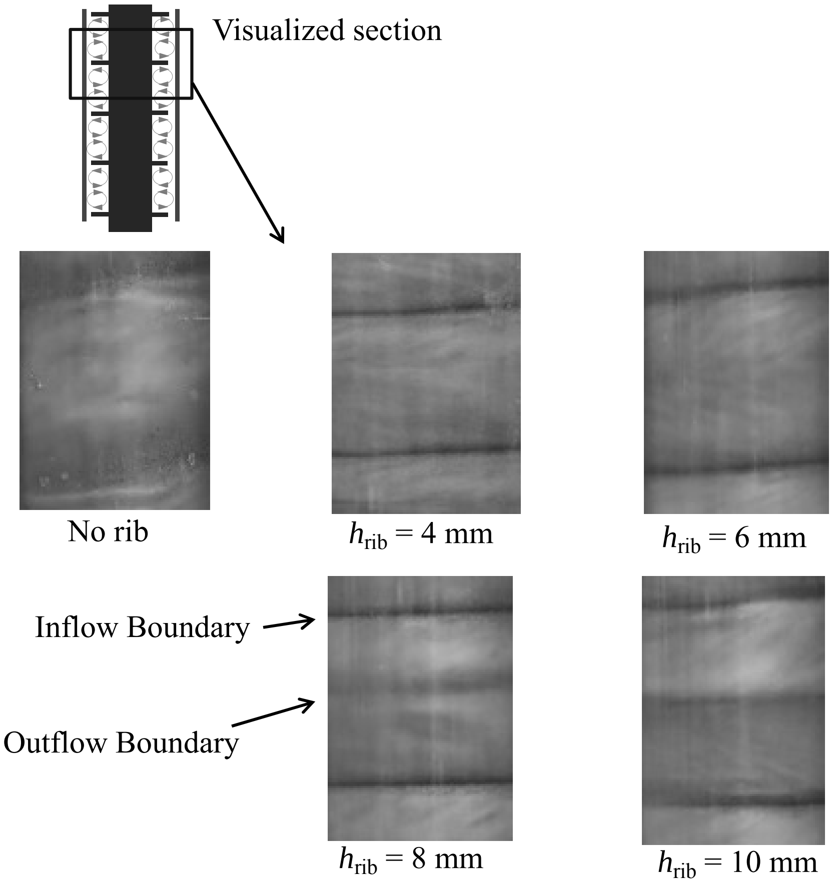

Fig. 3. Photographs of flow patterns at the same rotational speed of the inner cylinder at n = 190 rpm and $V_G = 5.0\times10^{-6}$ m^3/s [Reprinted with permission from Masuda, H., Zheng, T., Horie, T. and Ohmura, N. 2013, J. Chem. Eng. Japan, 46, 27].

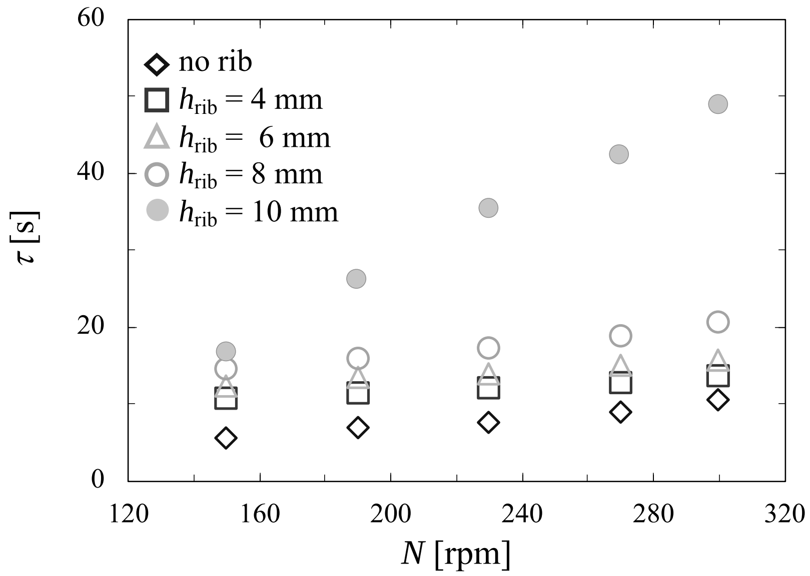

Fig. 4. Mean residence time of gas rotational speed of the inner cylinder at $V_G = 5.0\times10^{-6}$ m^3/s [15].

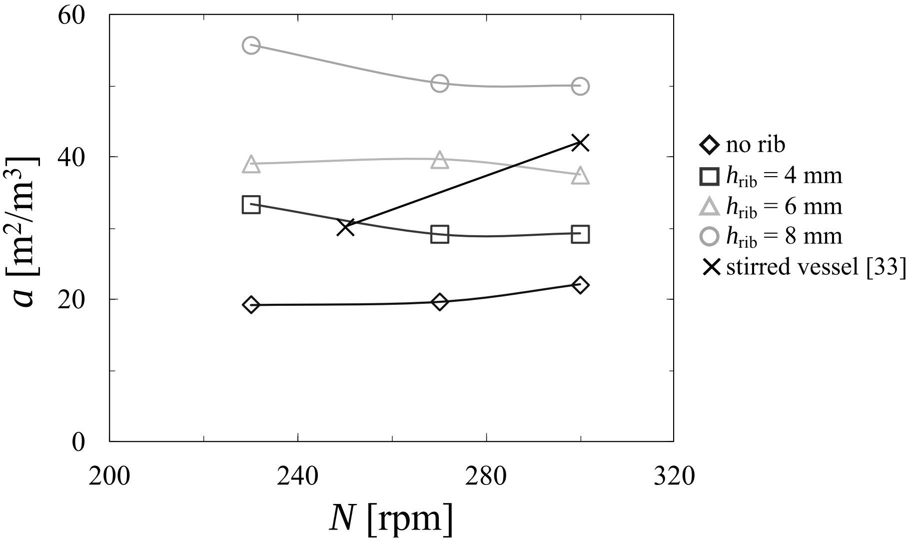

Fig. 5. Specific interfacial area between gas and liquid against the rotational speed of the inner cylinder at $V_G = 5.0\times10^{-6}$ m^3/s [15]. A comparison with a stirred vessel is also shown [33].

important factors for mass transfer, is enhanced up to three times. Fig. 5 also shows a comparison of the interfacial area in the Taylor–Couette flow with that in a typical stirred tank. Although a strict comparison is difficult because of the difference in various operational conditions, the Taylor–Couette flow with a ribbed inner cylinder is found to be an effective solution to intensify gas–liquid mass transfer.

Intensification of reactive liquid flow processes

The change in physical properties, such as density or viscosity, during processing significantly affects flow dynamics. With respect to the density change, it is well known that the Taylor–Couette flow dynamics and structure are destabilized by buoyancy resulting from the density difference in thermal convection or air flow systems [39, 40]. From the viewpoint of chemical engineering, reactive liquid flow accompanied by viscosity change should be discussed because the viscosity change with reaction is often encountered in many polymerization or fermentation processes. This viscosity change complicates the flow field, and consequently, destabilization of the Taylor cells is induced.

Masuda *et al*. [20, 28, 41] investigated the potential of the Taylor–Couette flow for process intensification of starch processing as an example of process with an intricate viscosity change during a chemical reaction. The starch suspension shows a significant increase in viscosity by heating (gelatinization). Consequently, enzymes are added to gelatinized starch to decompose into small sugars, which are used in various industries such as food, pulp, and biotechnology. The viscosity rapidly decreases after gelatinized starch comes into contact with the enzyme. The intricate changes in the shear viscosity are shown in Fig. 6. Because of the viscosity change, a series of starch processes using one apparatus is difficult. In industry, several apparatuses are often utilized [42]. Masuda *et al.* [20, 41] applied a Taylor–Couette flow with a normal inner cylinder for starch processing and showed that continuous and efficient hydrolysis is possible. However, the sugar yield decreased at a higher *Re* regime because of the destabilization of the Taylor cell. To overcome this destabilization, Matsumoto *et al.* [28] introduced a ribbed inner cylinder for starch processing. They experimentally confirmed that an ideal plug-flow type of residence time distribution was obtained with a ribbed inner cylinder even at a relatively high *Re*. As a result, as shown in Fig. 7, a decrease in the reducing sugar yield at higher effective *Re* (Re_{eff}) was prevented. Here, L_{rib} is the length of the ribbed section from the outlet. It is noted that Re_{eff} based on the effective viscosity is used because the viscosity spatially varies due to the non-Newtonian property of the fluid. Furthermore, Fig. 8 indicates that the sum of the yields of small sugars (glucose, maltose, and maltotriose), C_{ss},

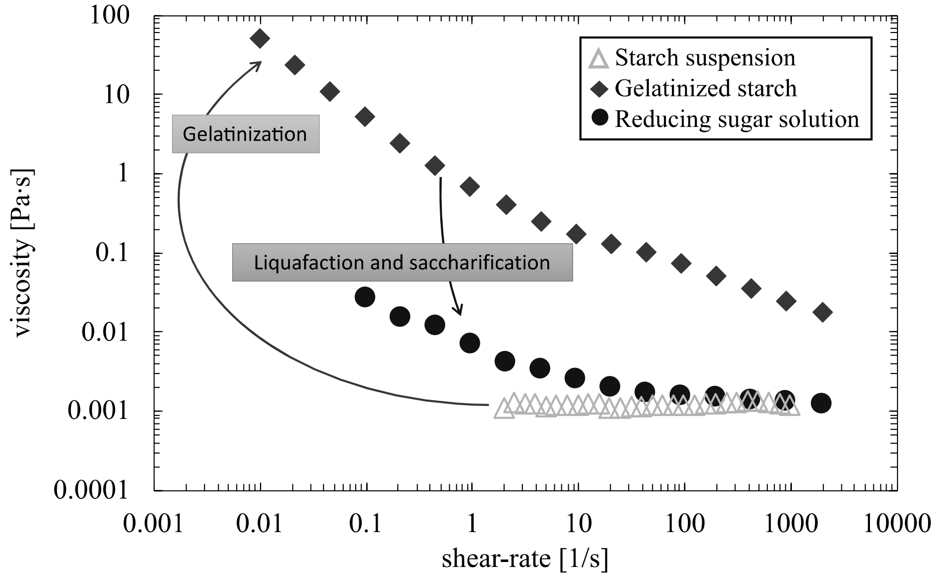

Fig. 6. Intricate change of rheological properties during starch hydrolysis [Reprinted from Matsumoto, M., Masuda, H., Hubacz, R., Horie, T., Iyota, H., Shimoyamada, M. and Ohmura, N. 2021, Chem. Eng. Sci., 231, 116270 with permission from Elsevier].

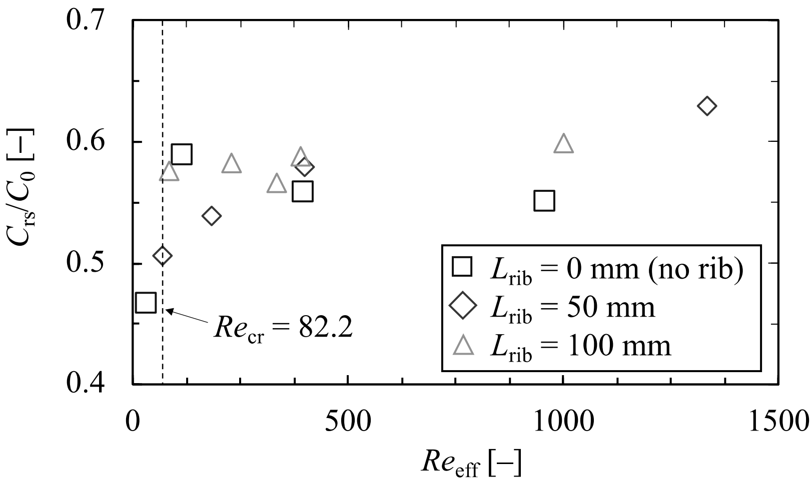

Fig. 7. Effect of Re_{eff} on C_{rs} / C_0 with three types of cylinders (L_{rib} = 0, 50, 100 mm) at u = 0.240 mm/s in starch hydrolysis experiments [Reprinted from Matsumoto, M., Masuda, H., Hubacz, R., Horie, T., Iyota, H., Shimoyamada, M. and Ohmura, N. 2021, Chem. Eng. Sci., 231, 116270 with permission from Elsevier].

significantly increased using the ribbed inner cylinder. This result indicates that the ribbed inner cylinder intensifies micromixing within Taylor cells while maintaining the stability of the Taylor cell structure.

These approaches are effective for maintaining the structure of the Taylor cell during processing. However, they do not address the problem of fluidity decrease due to the complex rheological properties of polymeric fluids such as gelatinized starch. The non-Newtonian property of the polymeric fluid causes the viscosity distribution in the Taylor–Couette flow apparatus. This viscosity distribution induces segregation of the region between the well-mixing and poor-mixing regions [43, 44]. To prevent this segregation, the Taylor cell should not be localized.

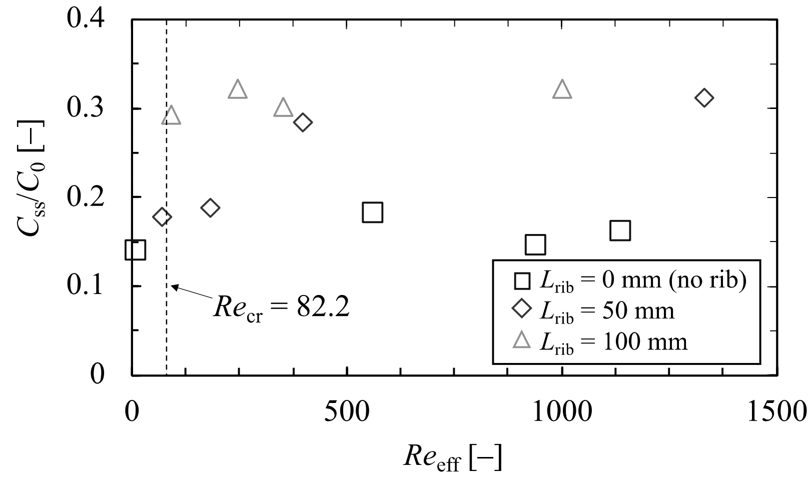

Fig. 8. Effect of C_{ss}/C_0 on *LR* with three types of cylinders (L_{rib} = 0, 50, 100 mm) at u = 0.240 mm/s in starch hydrolysis experiments [Reprinted from Matsumoto, M., Masuda, H., Hubacz, R., Horie, T., Iyota, H., Shimoyamada, M. and Ohmura, N. 2021, Chem. Eng. Sci., 231, 116270 with permission from Elsevier].

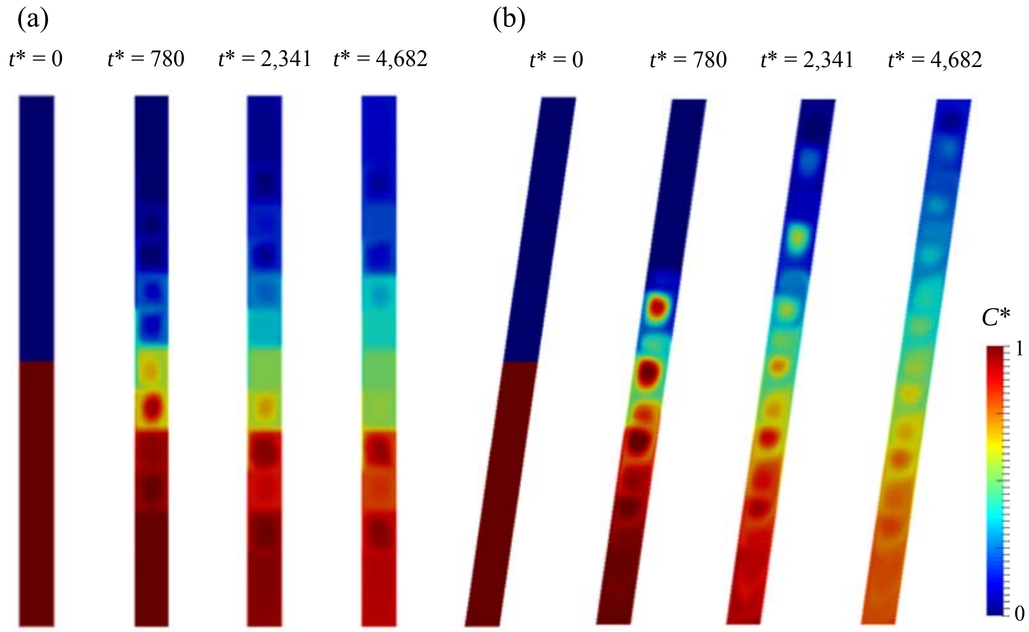

Fig. 9. Passive scalar transport process with time at Re_{eff} = 200 and n = 0.3: (a) cylindrical and (b) conical systems [Reprinted from Masuda, H., Iyota, H. and Ohmura, N. 2021, Chem. Eng. Technol., 44, 2049 with permission from John Wiley and Sons]. In each figure, left and right sides correspond to the inner and outer cone (cylinder) surfaces, respectively.

One solution is the global circulation of the Taylor cell in a rotating cone system. Wimmer [45] first reported that a meridional flow caused by the axial variation of centrifugal force causes the Taylor cells to circulate in the entire region. This large circulation while maintaining the structures of the Taylor cells has the potential to enhance mixing and heat and mass transfer in non-Newtonian fluid systems. Masuda *et al.* [46] preliminarily investigated the mixing performance of a conical Taylor–Couette flow with shear-thinning fluids. Fig. 9 shows the passive scalar transport conducted by the numerical simulations using cylinder and cone systems. The value of *n* indicates the strength of the shear-

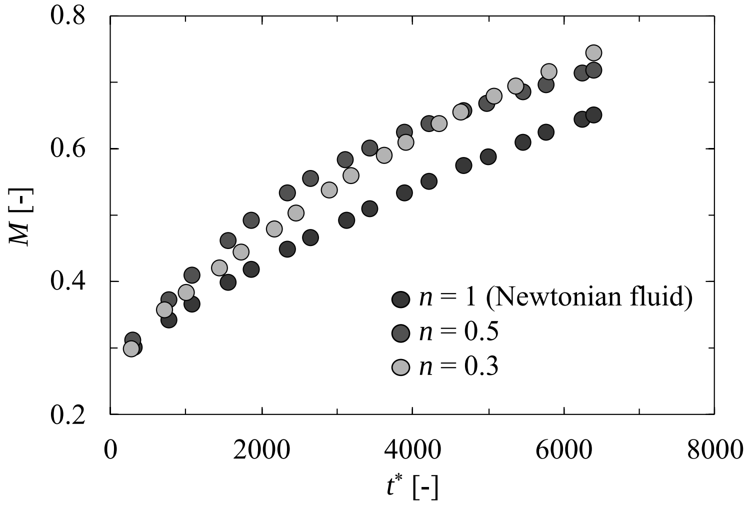

Fig. 10. Distributive mixing efficiency as a function of the non-dimensional time (t^*) in each fluid system at $Re_{\mathrm{eff}} = 200$ [Reprinted from Masuda, H., Iyota, H. and Ohmura, N. 2021, Chem. Eng. Technol., 44, 2049 with permission from John Wiley and Sons].

thinning property. The axial transport of mass is promoted in the conical case. Fig. 10 shows the distribution of mixing efficiency with time for each fluid. Note that $n = 1$ corresponds to a Newtonian fluid. The detailed mechanism for mixing enhancement with a shear-thinning fluid is described in a previous paper [46]. In this review, it is emphasized that the Taylor–Couette flow with modified geometry intensifies the mixing of rheologically complex fluids.

Other reactors using vortex dynamics

The segmented tubular reactor (SFTR) is also an effective reactor for process intensification. This reactor uses two non-miscible phases to create individual microvolumes of previously well-mixed reactants [47]. The segmented microbatch reactor circulates through a tube with identical residence times within the tubular reactor. In addition, there is no backmixing. Thus, the SFTR is also regarded as the ideal plug-flow reactor. By leveraging these characteristics, the SFTR is mainly applied in particle synthesis processes [48–50]. Another attractive reactor for process intensification is the oscillatory baffled reactor (OBR). The OBR consists of a tube with equally spaced baffles. In addition, oscillations are imposed on the net flow. The interaction of the pulsed flow with baffles creates complex hydrodynamics with recirculating vortices in the OBR, resulting in effective mixing and heat and mass transfer, as well as plug flow in a relatively low *Re* regime [51]. Several studies on the superiority of the OBR have been reported. In gas-liquid two phase flow, a higher gas hold-up and larger specific interfacial area are obtained compared with traditional stirred vessels [52, 53]. In addition, several researchers have applied the OBR to various chemical processes and demonstrated its superiority over traditional processes such as enzymatic saccharification [54], crystallization [55], and polymerization [56].

Similar to the Taylor–Couette flow reactor, both reactors enable enhanced transport rates and uniform continuous operation, and are expected to continue to play an important role in process intensification research. In this review, the main topic is the enhancement of mixing or mass transfer by vortices. However, the function of vortex flow is not only mixing but also accumulation and separation [57]. In the future, a new process that applies vortex dynamics in many directions will be developed.

CONCLUSIONS

This paper discusses process intensification, which is still developing, and the importance of vortex dynamics is discussed with regard to the Taylor–Couette flow. As described in the introduction, the Taylor–Couette flow has been applied to various processes. If the target is a small performance improvement, simply applying the Taylor–Couette flow is sufficient. However, to dramatically improve the performance (what is called a "quantum leap"), which is the original purpose of process intensification, it is necessary to maximize the function of vortex dynamics.

This review introduces a ribbed inner cylinder as a method for intensifying mass transfer and mixing by immobilizing and stabilizing vortices in a system where the vortex structure tends to be unstable, such as multiphase or reactive flow. The gas-liquid mass transfer and mixing processes were selected as successful examples. Although there is room for optimization in the shape of the ribs and the equipment method, it is possible to maximize the inherent performance of Taylor-Couette flow even in severe systems.

In addition to the Taylor–Couette flow reactor, other reactors also exploit vortices, such as the SFTRs and OBRs. With the vortex function as the starting point, a completely new type of reactor or process could be developed in the future.

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflict of interest.

ABBREVIATIONS

a	[m^2/m^3]	specific interfacial area
c^*	[–]	non-dimensional concentration
C_{rs}	[g/L]	reducing sugar concentration
C_{ss}	[g/L]	small saccharide concentration
C_0	[g/L]	initial concentration of substrate
d	[mm]	gap width
h_{rib}	[mm]	height of ribs
L_{rib}	[mm]	length of ribbed section from outlet
M	[–]	distributive mixing efficiency
N	[rpm]	rotational speed of inner cylinder
n	[–]	model parameter
Re	[–]	Reynolds number
Re_{cr}	[–]	critical Reynolds number
Re_{eff}	[–]	effective Reynolds number
t^*	[–]	non-dimensional time
u	[mm/s]	axial velocity
V_G	[m^3/s]	volumetric gas flow rate

Greeks

τ	[s]	mean residence time bubbles

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