



# Enzymatic starch hydrolysis performance of Taylor-Couette flow reactor with ribbed inner cylinder

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1     **Title:**

2     Enzymatic starch hydrolysis performance of Taylor–Couette flow reactor with ribbed  
3     inner cylinder

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**Highlights**

- A Taylor–Couette flow reactor is applied to a continuous starch hydrolysis process.
- A sufficient reducing sugar yield is obtained in the Taylor vortex flow regime.
- A ribbed inner cylinder suppresses axial dispersion caused by the wavy motion of Taylor cells.
- A ribbed inner cylinder enhances the starch hydrolysis process.

**Keywords:** Taylor–Couette flow reactor; Process intensification; Starch hydrolysis process; Ribbed inner cylinder; Viscosity change; Mixing enhancement

## Abstract

In this study, a Taylor–Couette flow reactor (TCFR) was applied to starch hydrolysis accompanied with an intricate viscosity change during reaction for the purpose of process intensification. In industries, several reactors are used in starch hydrolysis, namely gelatinization, liquefaction, and saccharification. It was possible to conduct a continuous starch hydrolysis with one TCFR. In addition, a sufficient reducing sugar yield was obtained in the Taylor vortex flow regime. However, the yield decreased at a higher effective Reynolds number ( $Re_{\text{eff}}$ ) due to axial dispersion through a bypass flow generated by the wavy motion of Taylor cells. In order to immobilize Taylor vortex flow at this condition, a ribbed inner cylinder was employed which suppressed axial dispersion at the higher  $Re_{\text{eff}}$ . As a result, a higher reducing sugar yield was successfully obtained than that by using a standard cylinder, demonstrating that the optimization of TCFR geometry has the potential for process intensification.

## 1. Introduction

Recent challenges concerning the environment, energy and increasing population have heightened demand for new technologies directed at saving energy and spaces and promoting a healthy environment, among other objectives. Chemical industries have particularly focused on process intensification (PI) to address these challenges. According to Stankiewicz (2000), PI involves the development of novel apparatuses and techniques that, compared with those typically used today, are expected to advance dramatic improvements in manufacturing and processing by substantially decreasing the equipment size/production capacity ratio, energy consumption, or waste production, and ultimately resulting in less expensive, sustainable technologies.

Reay et al. (2013) offers two reactor design strategies for achieving PI: enhancement of transport rates (momentum, heat and mass), and applying the same processing experience to every molecule. Another effective PI approach is the “conversion from batch to continuous operation” because continuous reactors are typically smaller than equivalent batch reactors (Boodhoo and Harvey, 2013). Such approaches indicate that a continuous reactor with sufficient transport rates and a narrow residence time distribution (RTD) is required to achieve PI. One such reactor is the Taylor–Couette flow reactor (TCFR). Taylor–Couette flow is the flow between coaxial cylinders with

the inner cylinder rotating. Taylor (1923) initially discovered that counter-rotating toroidal vortices spaced regularly along the axis are generated when the Reynolds number ( $Re$ ) in the circumference exceeds the critical  $Re$  ( $Re_{cr}$ ). The toroidal motion enhances mixing and heat/mass transfer. In addition, all the fluid elements leaving the reactor have the same residence time when a relatively small axial flow is added (Kataoka et al., 1975). Furthermore, compared with the conventional stirred tank reactor, there is no region where the locally strong shear force is imposed. This is beneficial to processes employing shear-sensitive materials. To take advantage of these features, TCFR has been applied to various processes, e.g., emulsion polymerization (Kataoka et al., 1995), particle synthesis (Ogihara et al., 1995), photocatalytic reaction (Sczechowski et al., 1995; Dutta and Ray, 2004), enzymatic reaction (Giordano et al., 2000), bioprocess (Qiao et al., 2018) and reverse osmosis process (Lee and Lueptow, 2001), and recently, to food processes such as heat sterilization of liquid food (Forney et al., 2004; Orłowska et al., 2014; Masuda et al., 2017c; Masuda et al., 2019a, 2019b) and production of textured soy-bean meat replacers (Krintiras et al., 2016).

In order to establish PI using continuous reactors like TCFR, it is necessary to face how to consider a spatial distribution of physical quantities (e.g. density, temperature and viscosity) caused by chemical reactions. For example, in polymerization or

1 fermentation process, the viscosity increases with the reaction (Kaminoyama et al.,  
2 1997; Potumarthi et al., 2007). The change in viscosity significantly affects the fluid  
3 flow, heat/mass transfer and, consequently, the reaction performance. To investigate the  
4 possibility of PI using TCFR in the process with complicated viscosity change during  
5 the reaction, Masuda et al. (2013, 2017a) applied TCFR to enzymatic hydrolysis process  
6 of starch (biopolymer). Starch hydrolysis consists of gelatinization, liquefaction, and  
7 saccharification. As shown in Fig. 1, the viscosity intricately changes during these  
8 processes. The viscosity of starch suspension significantly increases during  
9 gelatinization. Besides, gelatinized starch shows the strong shear-thinning property.  
10 When an enzyme is added to the gelatinized starch, the viscosity decreases rapidly and  
11 breaks down into reducing sugars. From a practical viewpoint, one of the most severe  
12 points is this viscosity change. In addition, the mixing function required is different in  
13 each process. Gelatinization requires heat transfer and dispersion of starch particles.  
14 Liquefaction and saccharification require highly efficient mixing, as a small amount of  
15 enzyme is added to highly viscous gelatinized starch, and also must avoid the excess  
16 shear force to ensure the enzyme activity. Thus, the current starch hydrolysis process  
17 employs several apparatuses, resulting in a massive total size (Baruque Filho et al.,  
18 2000). Conversely, it is possible to conduct continuous starch hydrolysis using one

TCFR due to the enhanced characteristics of mixing and heat/mass transfer. Masuda et al. (2013, 2017a) successfully showed that a higher reducing sugar yield was obtained in the Taylor vortex flow regime. However, the yield decreased at a higher  $Re$ , presumably induced by axial dispersion caused by the wavy motion of Taylor vortex flow. The enhanced mixing condition at a higher  $Re$  would be desirable for decomposition of starch into sugar having a smaller molecular such as glucose or maltose. In addition, Masuda et al. (2017a) changed the diameter of the inner cylinder between gelatinization and liquefaction/saccharification, which demonstrated that shape modification of the inner cylinder has potential for the mixing enhancement of Taylor vortex flow. Other research also explored the optimization of the inner cylinder shape (Soos et al., 2007; Sorg et al., 2011). In particular, Richter et al. (2008, 2009) found that a ribbed inner cylinder stabilized the structure of Taylor vortex flow at a higher  $Re$ . This research confirmed that a ribbed inner cylinder suppresses axial dispersion even in the wavy Taylor vortex flow regime, while enhancing mixing in Taylor cells. Applying a ribbed cylinder to the starch hydrolysis process can achieve further intensification due to the stabilization of the structure of Taylor vortices.

This study aims to intensify the starch hydrolysis process using TCFR with a ribbed inner cylinder. Based on the RTD and the reducing sugar yield, performance using the

1 ribbed inner cylinder was compared to that using a standard inner cylinder. As discussed,  
2 the intricate change in the viscosity during starch hydrolysis complicates the flow  
3 condition in the apparatus. Therefore, to preliminarily investigate the effect of the  
4 ribbed inner cylinder on the performance, dextrin hydrolysis, in which there is no  
5 viscosity change during reaction, was selected as a model reaction. It is noted that  
6 dextrin solution is generally a low viscous Newtonian fluid. Next, based on the results  
7 of the dextrin hydrolysis experiments, starch hydrolysis experiments were performed.

## 9 **2. Materials and Methods**

### 10 **2.1. Experimental apparatus**

11 The Taylor–Couette flow reactor (TCFR) used in this study consisted of a rotating  
12 inner cylinder (the outer radius ( $R_i$ ): 12.5 mm) and a stationary outer cylinder (the inner  
13 radius ( $R_o$ ): 17.5 mm), as shown in Fig. 2. The length of the cylinders ( $L$ ) and the gap  
14 width ( $d$ ) were 300 mm and 5 mm, respectively. The angular velocity of the inner  
15 cylinder,  $\omega$ , was varied from 2.5 to 50 rad/s. The starch or dextrin suspension at room  
16 temperature was introduced from the inlet. The axial velocity,  $u$ , was varied from 0.145  
17 to 0.418 mm/s. The initial concentration of starch suspension or dextrin solution,  $C_0$ ,  
18 was 250 g/L. For the starch hydrolysis experiments,  $\alpha$ -amylase derived from *Bacillus*

*licheniformis* (Termamyl 120L, Novozymes) was injected at the middle point of TCFR. For the dextrin hydrolysis experiments, the injection point was 50 mm from the inlet. Thus, it should be noted that the enzyme injection point is different between each experiment. The volumetric flow rate of the enzyme,  $v_e$ , was varied from 0.1 to 1.5 mL/h. Two water jackets were included to control temperature during processing. For the starch hydrolysis experiments, two jackets were set to 85°C. For the dextrin hydrolysis experiments, two jackets were set to 65°C. Two types of inner cylinders, standard and ribbed, were used. Figure 3 depicts the ribbed cylinder schematically. The distance between each cell,  $d_{\text{cell}} (= 2d)$ , was 10 mm, indicating that the pair of vortices was segmented by ribs. The width of the ribs was 2.5 mm. The height of the ribs,  $h_{\text{rib}}$ , was 2.5 and 4.5 mm as shown in Fig. 4. To optimize rib configuration in the inner cylinder, the length of the ribbed section from the outlet,  $L_{\text{rib}}$  in Fig. 2, were varied. In the dextrin hydrolysis experiments, ribs were equipped downstream along the axis ( $L_{\text{rib}} = 250$  mm). In contrast to the dextrin solution, the starch hydrolysis experiments involved highly viscous fluids. In such systems, channeling or sedimentation would typically occur around ribs. To avoid this undesirable condition, ribs should be located in the section where the viscosity relatively decreases due to the liquefaction reaction. Thus, in the starch hydrolysis experiments,  $L_{\text{rib}}$  was set to 50 to 100 mm.

1

2 **2.2. Sample handling and analysis**

3 Product samples were taken from the outlet of the apparatus at  $\theta = 2.5$ .  $\theta$  means the  
 4 normalized time as follows:

$$\theta = \frac{u(R_o^2 - R_i^2)\pi \cdot t}{V_r} \#(1)$$

5 According to Kataoka et al. (1995), the conversion of a chemical reaction reaches a  
 6 steady-state value after  $\theta = 2$  in a TCFR. Thus, the sampling at  $\theta = 2.5$  is adequate to  
 7 investigate the performance of TCFR.

8 To halt the liquefaction reaction, samples were immediately cooled to approximately  
 9 0°C and their pH was reduced to 2 by the addition of HCl. The sample was centrifuged  
 10 at 15,000 G for 2,400 s to separate the solid and liquid phases. To measure the  
 11 liquefaction rate ( $LR$ ) (i.e. the ratio of the liquid phase weight ( $W_{\text{liquid}}$ ) to the total weight  
 12 of the sample ( $W_{\text{total}}$ )), the solid phase ( $W_{\text{solid}}$ ) was weighed. The value of  $LR$  was  
 13 calculated as follows:

$$LR = W_{\text{liquid}}/W_{\text{total}} = (W_{\text{total}} - W_{\text{solid}})/W_{\text{total}} \#(2)$$

14 The carbohydrate composition and the reducing sugar concentration in the liquid phase  
 15 of sample were also investigated. The carbohydrate composition was analyzed using a  
 16 high performance liquid chromatography (HPLC). The HPLC column (Asahipak,

NH2P-50, Shodex<sup>®</sup>) having the inner diameter of 4.6 mm and the length of 250 mm was used with acetic acid in water (30%, v/v) as the mobile phase at 0.4 mL/min. The concentration of glucose, maltose and maltotriose in the liquid phase was quantified. Besides, the total concentration of reducing sugar in the liquid phase of sample,  $C_{rs}$ , was measured by the dinitro salicylic acid (DNS) method. Adding the DNS reagent, the sample reagent colors the sample and allows analyzing the reducing sugar concentration using a UV-VIS spectrophotometer (V-630, JASCO Corp.) at a wavelength of 540 nm.

Each experiment was repeated more than three times. Only one sample was measured and analyzed. The average values are shown in each figure.

### **2.3. Residence time distribution (RTD) measurements**

To understand the characteristics of axial dispersion, the residence time distribution (RTD) was measured using a salt-solution technique. The working fluid was an aqueous solution of 25wt% concentration of glycerin. The tracer was 200 g/L concentration of a NaCl solution. The 1 mL tracer was impulsively injected from the injection port which is located at 50 mm from the inlet. This means the actual length of TCFR in the tracer response experiments was 250 mm. The electric conductivity of the effluent was measured using an electric conductivity meter (CM-42X, DDK-TOA Corp.). The

measurement was converted to the tracer concentration from a calibration curve. The RTD was obtained by normalizing the tracer concentration,  $E_\theta$ , with the normalized time ( $\theta$ ) as follows (Richter et al., 2008):

$$E_\theta = \frac{V_r \cdot C(t)}{u(R_0^2 - R_1^2)\pi \cdot \int_0^\infty C(t) dt} \#(3)$$

#### 2.4. Definition of the effective Reynolds number ( $Re_{\text{eff}}$ )

The flow pattern of Taylor vortex flow is characterized by  $Re$ , which is defined as follows:

$$Re = \frac{\rho \omega R_i d}{\eta} \#(4)$$

where  $\rho$  [kg/m<sup>3</sup>] is the fluid density,  $\omega$  [rad/s] is the angular velocity of the inner cylinder, and  $\eta$  [Pa·s] is the fluid viscosity. Calculating  $Re$  for the starch hydrolysis reaction is more difficult. The viscosity decreases with the hydrolysis reaction. According to Masuda et al. (2017a), the drastic decrease in viscosity is observed only in the initial stage of the reaction, i.e., the rheological properties of the effluent can be regarded as the representative properties for the calculation of  $Re$ . Another challenge is the spatial change in viscosity due to the shear-thinning property of polysaccharide solutions. As Ohta et al. (2003, 2005) suggested, in non-Newtonian fluid systems, the construction of the effective Reynolds number,  $Re_{\text{eff}}$ , based on the effective viscosity

( $\eta_{\text{eff}}$ ), is necessary from a practical perspective. To properly determine  $\eta_{\text{eff}}$ , the method of estimating the effective shear-rate,  $\gamma_{\text{eff}}$ , should be considered. For a Taylor–Couette flow system, Masuda et al. (2017b) proposed an empirical correlation for the estimation of  $\gamma_{\text{eff}}$  from the angular velocity of the inner cylinder,  $\omega$ , as follows:

$$\gamma_{\text{eff}} = \left\{ 77.05n^{0.32} \left( \frac{R_i}{R_o} \right)^2 - 88.73n^{0.31} \left( \frac{R_i}{R_o} \right) + 26.85n^{0.21} \right\} \cdot \omega \quad (5)$$

The value of  $n$  is a model parameter in the Carreau model (1972), which is the rheological model used in this study:

$$\eta = \eta_0 [1 + (\beta \cdot \gamma)^2]^{\frac{(n-1)}{2}} \quad (6)$$

where  $\eta_0$  is the zero shear-rate viscosity,  $\gamma$  is the shear-rate and  $\beta$  is the characteristic time. It can be obtained by applying the rheological property of the effluent to the Carreau model (Eq. (6)). Equation (5) estimates the value of  $\gamma_{\text{eff}}$  is estimated from  $\omega$  and  $n$ . Consequently,  $Re_{\text{eff}}$  can be estimated from  $\eta_{\text{eff}}$  which is calculated by substituting  $\gamma_{\text{eff}}$  for  $\gamma$  in the Carreau model (Eq. (6)).

### 3. Results and Discussion

#### 3.1. Dextrin hydrolysis process

Figure 5 illustrates the dependence of the reducing sugar yield  $C_{\text{rs}}/C_0$  on  $Re$  and  $u$  in the dextrin hydrolysis process. According to Taylor (1923), the theoretical  $Re_{\text{cr}}$  without

axial flow is 82.2 for the radius ratio  $R_i/R_o = 0.714$  of the TCFR used. Strictly speaking,  $Re_{cr}$  depends on the axial Reynolds number,  $Re_{ax}$ , which is defined as follows (Lueptow et al., 1992):

$$Re_{ax} = \frac{\rho u(2d)}{\eta} \quad \#(7)$$

The maximum value of  $Re_{ax}$  within the experimental conditions was about 2.0. According to Lueptow et al. (1992), such low value of  $Re_{ax}$  does not affect  $Re_{cr}$ .

In Fig. 5, in all cases of  $u$ , higher  $C_{rs}/C_0$  was obtained above  $Re_{cr} (= 82.2)$ , indicating the enhancement of mixing and heat transfer due to Taylor vortex flow improved the hydrolysis reaction of dextrin. However, in all cases of  $u$ ,  $C_{rs}/C_0$  slightly decreased above approximately  $Re = 900$ . Although higher shear force at a higher  $Re$  would induce inactivation of  $\alpha$ -amylase (van der Veen et al., 2004), such extremely high shear force is not imposed in TCFR. In starch hydrolysis system, Masuda et al. (2013, 2017a) also reported the decrease in the reducing sugar yield at a higher  $Re$ , and explained this was due to the wavy motion of Taylor vortices which affects TCFR performance as a reactor because mass diffusion over cell boundaries (intermixing) is enhanced and the injection fluid (enzyme) would diffuse through a bypass flow caused by this wavy motion (Ohmura et al., 1997; Wereley and Lueptow, 1999).

According to Richter et al. (2008), using a ribbed inner cylinder is effective to

1 suppress the wavy motion even at a higher  $Re$ . Figure 6 shows the flow visualization at  
2  $Re = 905.1$  (a) without rib and (b) with ribs ( $h_{rib} = 4.5$  mm). In flow visualization  
3 experiments, it should be noted that an aqueous solution of 25wt% glycerin was used  
4 and no axial flow was imposed. The flow was visualized by adding small amount of  
5 Iridin<sup>®</sup> (Iridin<sup>®</sup> silver-white pigment, MERCK) to the glycerin solutions. This  
6 substance consists of small, light reflecting slabs, which arrange themselves along  
7 streamlines due to the viscous forces (Richter et al., 2008). As shown in Fig. 6 (a), dark  
8 streaks correspond to the inflow boundary wave in the case of no rib. This wavy motion  
9 causes intermixing and bypass diffusion. As a result, axial dispersion is induced.  
10 Conversely, in the case of ribbed inner cylinder, a pair of vortices is segmented and  
11 immobilized by the rib (Fig. 6 (b)). Thus, axial dispersion is expected to be suppressed  
12 even the wavy Taylor vortex flow regime. In order to investigate the cross-sectional  
13 structure of Taylor vortices, numerical simulation was conducted based on the method  
14 reported (Masuda et al., 2017b). The density and viscosity of aqueous solution of  
15 25wt% glycerin was used for the simulation. Figure 7 shows the cross-sectional view of  
16 velocity field at  $Re = 905.1$  (a) without rib and (b) with ribs ( $h_{rib} = 4.5$  mm). It should be  
17 noted the aspect ratio in the simulation differed from the experimental condition. As  
18 shown in Fig. 7, the suppression of wavy motion at the inflow boundary was also

1 confirmed.

2 In order to clarify the characteristics of TCFR with or without ribs, RTD ( $E_\theta$  curve)  
3 at  $Re = 905.1$  and  $Re_{ax} = 2.0$  was investigated as shown in Fig. 8. The fitting lines based  
4 on the tanks-in-series model (Eq. (8)) were also drawn (Levenspiel, 1999):

$$E_\theta = \frac{N}{(N-1)!} (N\theta)^{N-1} \exp(-N\theta) \quad (8)$$

5 where  $N [-]$  is the number of tanks in series. In the case of no rib ( $h_{rib} = 0$  mm), the RTD  
6 similar to a continuous stirred tank reactor (CSTR) model was obtained. Although the  
7 RTD at  $h_{rib} = 2.5$  mm becomes marginally sharp compared with the standard inner  
8 cylinder, there was no clear effect of ribs on the suppression of axial dispersion.  
9 Remarkably, the sharp RTD, i.e., plug flow type RTD, was obtained at  $h_{rib} = 4.5$  mm due  
10 to the robust immobilization and stabilization of Taylor vortices. The values of  $N$ , which  
11 is obtained by fitting Eq. (8) to the experimental results (RTD), were 2 (no rib), 4 ( $h_{rib} =$   
12 2.5 mm), and 17 ( $h_{rib} = 4.5$  mm), respectively. In the tracer response experiments, 25  
13 pairs of vortices are ideally formed. Because one pair of vortices is regarded as one  
14 stirred vessel (Ohmura et al., 1997), the ideal value of  $N$  with no axial dispersion is 25.  
15 Thus, it was found the RTD at  $h_{rib} = 4.5$  mm close approaches the ideal condition.

16 The degree of axial dispersion was evaluated based on the dispersion coefficient (**D**).

17 The differential equation representing the dispersion model is as follows:

$$\frac{\partial C}{\partial \theta} = \left( \frac{\mathbf{D}}{uL} \right) \frac{\partial^2 C}{\partial z^2} - \frac{\partial C}{\partial z} \#(9)$$

where the dimensionless group ( $\mathbf{D} / uL$ ), called the vessel dispersion number, is the parameter that measures the extent of axial dispersion (Levenspiel, 1999). By numerically solving Eq. (9) using the results of tracer response experiments, the dispersion number was estimated. The dispersion number in TCFR with or without ribs is shown in Fig. 9. In the case of  $h_{\text{rib}} / d = 0.9$  (i.e.  $h_{\text{rib}} = 4.5$  mm), the significantly low dispersion number was successfully obtained and it is closely to the value in the ideal cascade of 25 stirred vessels (the dotted line in Fig. 9). Thus, the rib with  $h_{\text{rib}} = 4.5$  mm was used for the dextrin/starch hydrolysis experiments.

Figure 10 illustrates the effect of the ribbed inner cylinder on  $C_{\text{rs}}/C_0$  at  $u = 0.418$  mm/s in the dextrin hydrolysis experiment. As expected, the ribbed inner cylinder suppressed the decrease in  $C_{\text{rs}}/C_0$  at a higher  $Re$ . In addition, using the ribbed inner cylinder, the sufficient  $C_{\text{rs}}/C_0$  was obtained at a relatively low  $Re$ . In the case of ribbed inner cylinder, as shown in Fig. 4, a pair of vortices (Ekman cells) between ribs is formed even below  $Re_{\text{cr}}$  due to Ekman boundary layers (Czamy et al., 2003). Therefore, the ribbed inner cylinder is expected to improve mixing at a lower  $Re$  and suppress axial dispersion at a higher  $Re$ .

### 3.2. Starch hydrolysis process

Figure 11 depicts the dependence of the liquefaction rate ( $LR$ ) on  $Re_{\text{eff}}$  at  $u = 0.418$  mm/s in the standard and ribbed inner cylinders ( $h_{\text{rib}} = 4.5$  mm). In the case of ribbed inner cylinder, two types of the length of ribbed section from the outlet ( $L_{\text{rib}}$ ) was investigated. In the Taylor vortex flow region, starch liquefaction was conducted sufficiently and continuously in both cylinders. In the standard inner cylinder, the  $LR$  increased with the increase in  $Re_{\text{eff}}$ . In contrast, the ribbed cylinder largely exhibited no dependence of  $LR$  on  $Re_{\text{eff}}$ . Remarkably, higher  $LR$  was obtained even at a relatively low  $Re_{\text{eff}}$  when employing the ribbed inner cylinder. As described in the previous section, it is considered that pairs of vortices between ribs enhanced mixing of gelatinized starch and enzyme.

Figure 12 shows the dependence of  $C_{\text{rs}}/C_0$  on  $Re_{\text{eff}}$  at  $u = 0.418$  mm/s in the standard and ribbed inner cylinders in the starch hydrolysis process. For the standard inner cylinder, the decrease in  $C_{\text{rs}}/C_0$  at a higher  $Re_{\text{eff}}$  was observed, although a high value of  $C_{\text{rs}}/C_0$  was obtained in the Taylor vortex flow regime. For the ribbed inner cylinder, there was no decrease in  $C_{\text{rs}}/C_0$  at a higher  $Re_{\text{eff}}$ . On the contrary, at  $L_{\text{rib}} = 50$  mm,  $C_{\text{rs}}/C_0$  slightly increased with the increase in  $Re_{\text{eff}}$  even above  $Re_{\text{eff}} = 1,000$ . Therefore, it is considered that the ribbed inner cylinder suppresses axial dispersion at a higher  $Re_{\text{eff}}$

even in the starch hydrolysis process. In another case where  $L_{\text{rib}} = 100$  mm,  $C_{\text{rs}}/C_0$  remained constant with an increase in  $Re_{\text{eff}}$ , indicating that in this case a sufficient mixing condition for starch hydrolysis was achieved even at low  $Re_{\text{eff}}$ .

Figure 13 illustrates the dependence of the yield of small saccharides,  $C_{\text{ss}}/C_0$ , on  $Re_{\text{eff}}$  at  $u = 0.418$  mm/s with the standard and ribbed inner cylinders in starch hydrolysis process.  $C_{\text{ss}}$  is the concentration of small saccharides and is calculated as follow:

$$C_{\text{ss}} = C_{\text{g}} + C_{\text{m}} + C_{\text{mt}} \quad (10)$$

where  $C_{\text{g}}$ ,  $C_{\text{m}}$  and  $C_{\text{mt}}$  are the concentration of glucose, maltose, and maltotriose, respectively. As evident in Fig. 13, above  $Re_{\text{eff}} = 500$ , a significantly higher value of  $C_{\text{ss}}/C_0$  was obtained using the ribbed inner cylinder compared with that using the standard inner cylinder. Thus, it was determined that a mixing enhancement suppressing axial dispersion using the ribbed inner cylinder further intensifies the starch hydrolysis process.

As with the result of  $C_{\text{rs}}/C_0$  shown in Fig. 12, the dependence of  $C_{\text{ss}}/C_0$  on  $Re_{\text{eff}}$  differed between  $L_{\text{rib}} = 50$  and 100 mm. From a practical perspective,  $Re_{\text{eff}}$  and  $L_{\text{rib}}$  should be optimized. For example, Fig. 13 indicates that the comparable value of  $C_{\text{ss}}/C_0$  was obtained in  $Re_{\text{eff}} = 397$ ,  $L_{\text{rib}} = 50$  mm and  $Re_{\text{eff}} = 91$ ,  $L_{\text{rib}} = 100$  mm. In general, increasing the number of ribs equipped would increase power consumption. Therefore,

$Re_{\text{eff}} = 397$ ,  $L_{\text{rib}} = 50$  mm appears to be a more efficient condition than  $Re_{\text{eff}} = 91$ ,  $L_{\text{rib}} = 100$  mm. However, a higher  $Re_{\text{eff}}$  condition naturally requires higher power consumption. Therefore, in order to optimize starch hydrolysis using the ribbed inner cylinder, it is necessary to investigate power consumption at each condition. Besides, the optimization of  $h_{\text{rib}}$  is also beneficial for process design in the future. Nevertheless, it can be concluded that the ribbed inner cylinder is effective for the production of small saccharides even in relatively high starch concentration system.

#### 4. Conclusions

A Taylor–Couette flow reactor (TCFR) was applied to dextrin and starch hydrolysis processes to achieve process intensification. In both processes, the reducing sugar yield increased in the Taylor vortex flow region, because the toroidal motion of Taylor vortices enhanced the mixing of substrates with an enzyme. However, the yield decreased at a higher Reynolds number ( $Re$ ) and effective Reynolds number ( $Re_{\text{eff}}$ ). The residence time distribution (RTD) was broad at a higher  $Re$  in the tracer response experiment. Thus, it was determined that the axial dispersion induced by the wavy motion of Taylor cells at higher  $Re$  and  $Re_{\text{eff}}$  caused the decrease in the reducing sugar yield.

1 In order to immobilize Taylor cells at a higher  $Re$ , a ribbed inner cylinder was used.  
2 A relatively sharp RTD was obtained using this cylinder. Because the ribbed inner  
3 cylinder suppressed axial dispersion, there was no decrease in the reducing sugar yield  
4 at higher  $Re$  and  $Re_{\text{eff}}$ . Furthermore, in the starch hydrolysis process, a significantly  
5 higher yield of small saccharides was obtained using the ribbed inner cylinder compared  
6 than that using the standard inner cylinder.

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6    **Nomenclature**

7     $E_\theta$     normalized tracer concentration, –

8     $C$     concentration, g/L

9     $C_g$     glucose concentration, g/L

10     $C_m$     maltose concentration, g/L

11     $C_{mt}$     maltotriose concentration, g/L

12     $C_{rs}$     reducing sugar concentration, g/L

13     $C_{ss}$     small saccharide concentration, g/L

14     $C_0$     initial concentration of substrate, g/L

15     $D_z$     dispersion coefficient, m<sup>2</sup>/s

16     $d$     gap width, mm

17     $d_{cell}$     distance between each cell, mm

18     $E_0$     normalized tracer concentration, –

1	$h_{\text{rib}}$	height of ribs, mm
2	$L$	length of cylinders, mm
3	$LR$	liquefaction rate, –
4	$L_{\text{rib}}$	length of ribbed section from outlet, mm
5	$N$	number of tanks, –
6	$n$	model parameter in Carreau model, -
7	$u$	axial velocity, mm/s
8	$v_e$	volumetric flow rate of enzyme, mL/h
9	$Re$	Reynolds number, –
10	$Re_{\text{ax}}$	axial Reynolds number, –
11	$Re_{\text{cr}}$	critical Reynolds number, –
12	$Re_{\text{eff}}$	effective Reynolds number, –
13	$R_i$	inner cylinder radius, mm
14	$R_o$	outer cylinder radius, mm
15	$t$	time, s
16	$V_r$	reactor volume, mL
17	$W_{\text{liquid}}$	liquid phase weight, g
18	$W_{\text{solid}}$	solid phase weight, g

1	$W_{\text{total}}$	total weight, g
2	$z$	axial position, mm
3		
4	<i>Greek letters</i>	
5	$\beta$	characteristic time, s
6	$\dot{\gamma}$	shear-rate, 1/s
7	$\dot{\gamma}_{\text{eff}}$	effective shear-rate, 1/s
8	$\eta$	viscosity, Pa·s
9	$\eta_{\text{eff}}$	effective viscosity, Pa·s
10	$\theta$	normalized time, –
11	$\rho$	density, kg/m <sup>3</sup>
12	$\omega$	angular velocity of inner cylinder, rad/s
13		
14		
15		
16		
17		
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**Fig. 1.** Intricate change of rheological properties during starch hydrolysis.

**Fig. 2.** Taylor–Couette flow reactor.

**Fig. 3.** Picture of ribbed inner cylinder.

**Fig. 4.** Cross-sectional view of a pair of Taylor vortices between ribs.

**Fig. 5.** Dependence of  $C_{rs} / C_0$  on  $Re$  at various  $u$  and  $C_0 = 250$  g/L in dextrin hydrolysis experiments. The results in this figure were obtained using the standard inner cylinder.

The solid line shows  $Re_{cr} = 82.2$ .

**Fig. 6.** Flow visualization of Taylor–Couette flow with (a) standard and (b) ribbed inner cylinder ( $h_{rib} = 4.5$  mm) at  $Re = 905.1$ . An aqueous solution of 25wt% glycerin with small amount of Iriodin<sup>®</sup> was used for flow visualization experiments.

**Fig. 7.** Cross-sectional view of velocity field obtained using numerical simulation with (a) standard and (b) ribbed inner cylinder ( $h_{rib} = 4.5$  mm) at  $Re = 905.1$ . The density and

viscosity of aqueous solution of 25wt% glycerin was used.

**Fig. 8.** RTD in TCFR with each cylinder at  $Re = 905.1$  and  $u = 0.418$  mm/s. Solid lines (fitting lines) correspond to tank-in-series model lines. An aqueous solution of 25wt% glycerin was used as the working fluid.

**Fig. 9.** Effect of normalized rib height ( $h_{rib} / d$ ) on dispersion number at  $Re = 905.1$  and  $u = 0.418$  mm/s.

**Fig. 10.** Dependence of  $C_{rs} / C_0$  on  $Re$  with standard and ribbed inner cylinder at  $u = 0.145$  mm/s in dextrin hydrolysis experiments.

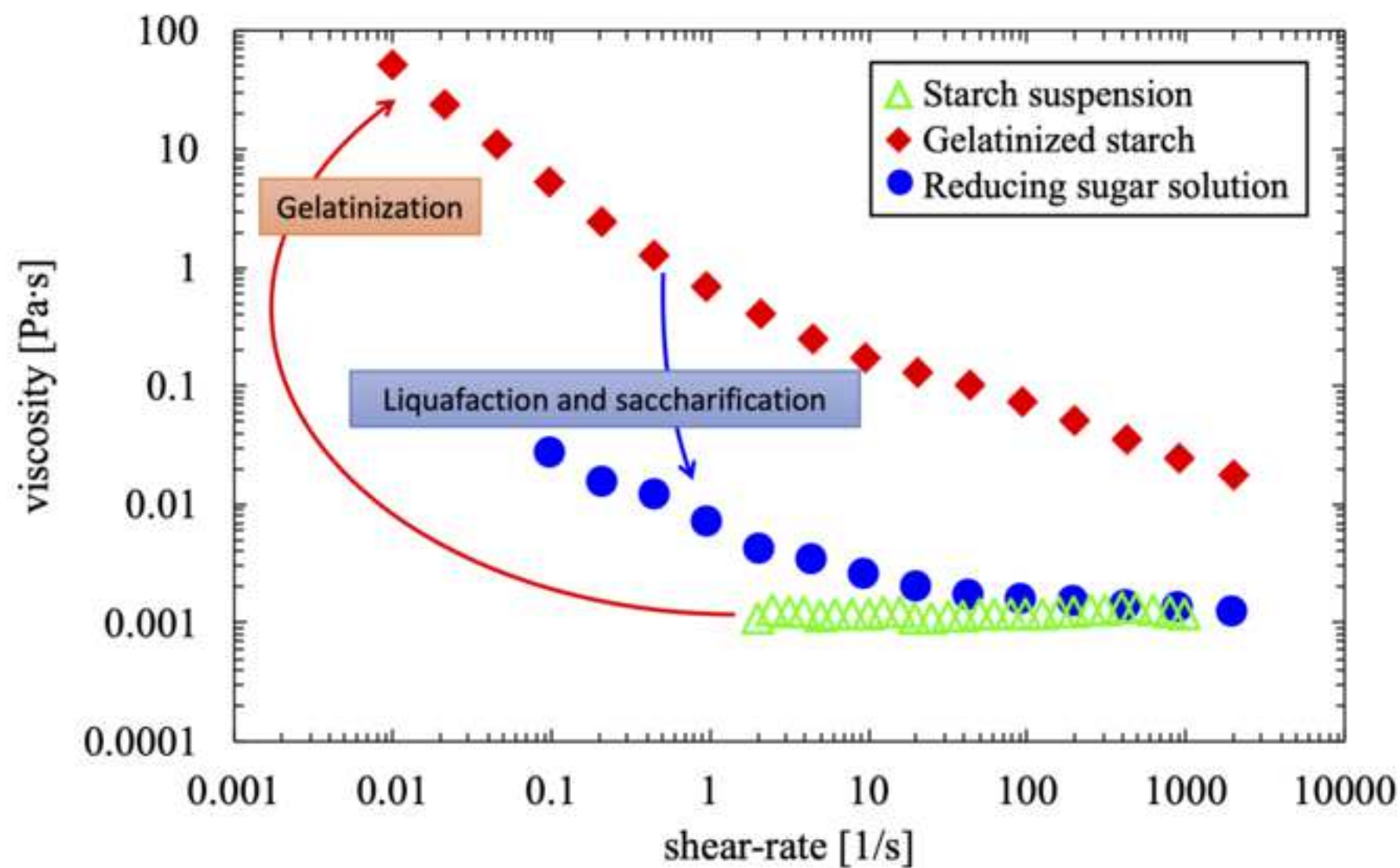
**Fig. 11.** Effect of  $Re_{eff}$  on  $LR$  with three types of cylinders ( $L_{rib} = 0, 50, 100$  mm) at  $u = 0.240$  mm/s in starch hydrolysis experiments.

**Fig. 12.** 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.

**Fig. 13.** 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.

Figure1

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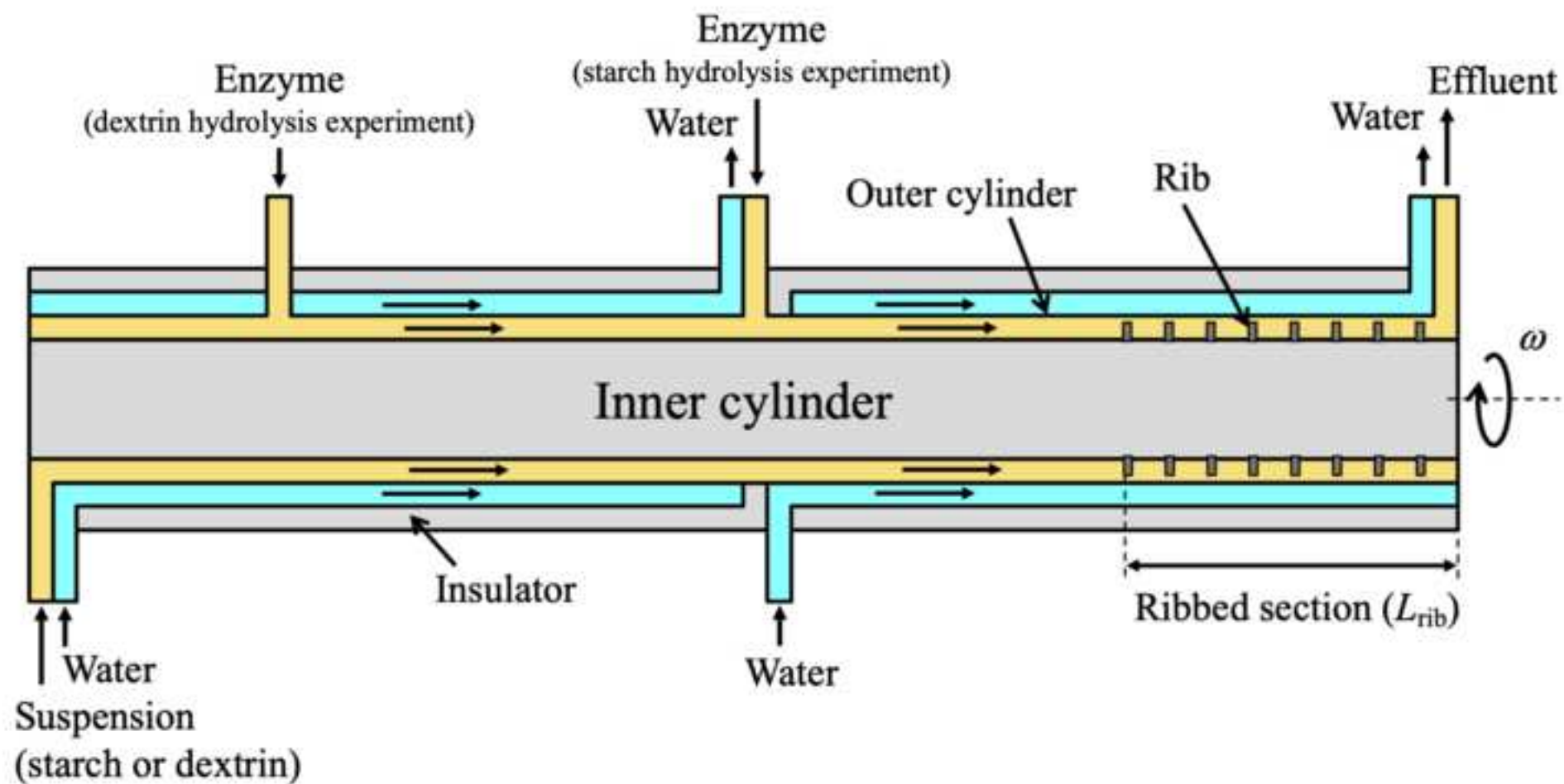


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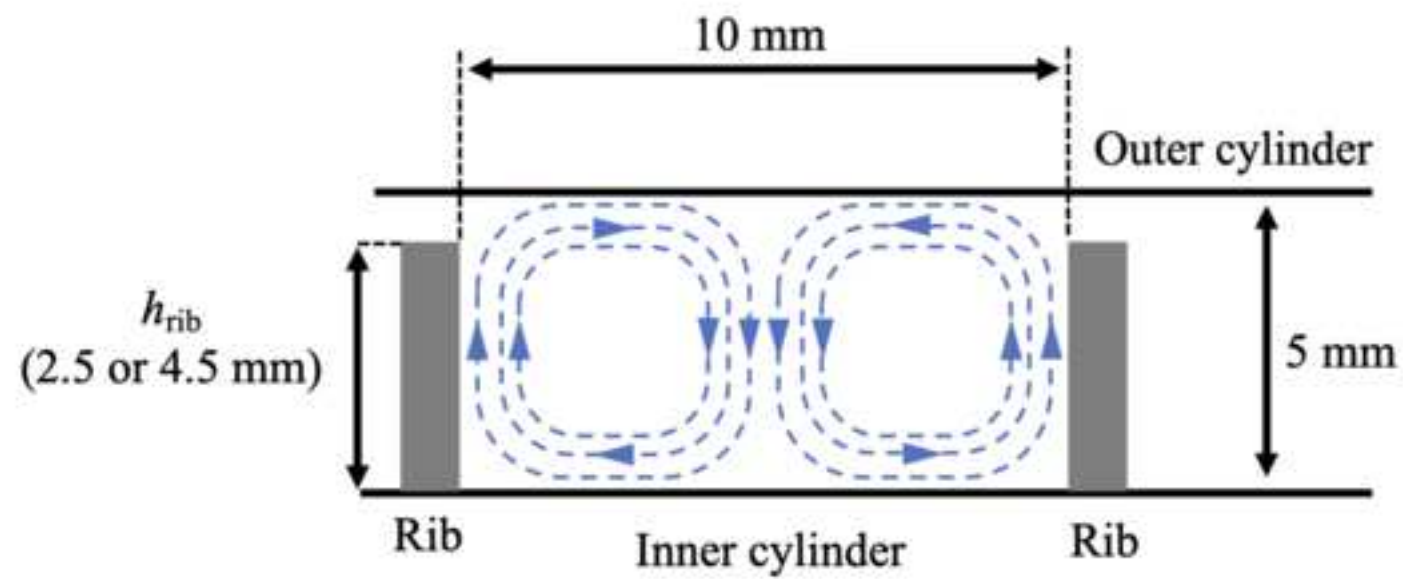


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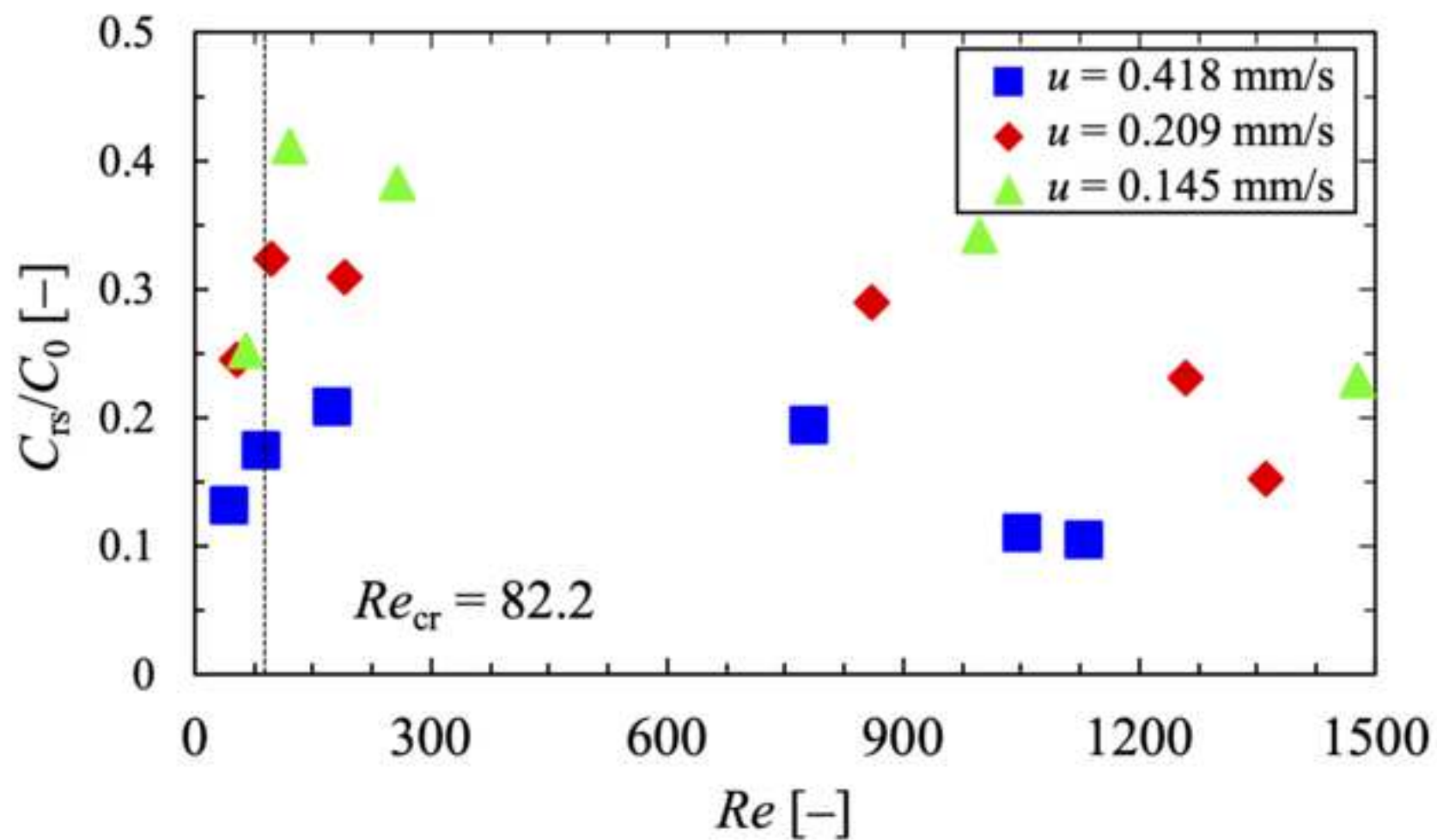
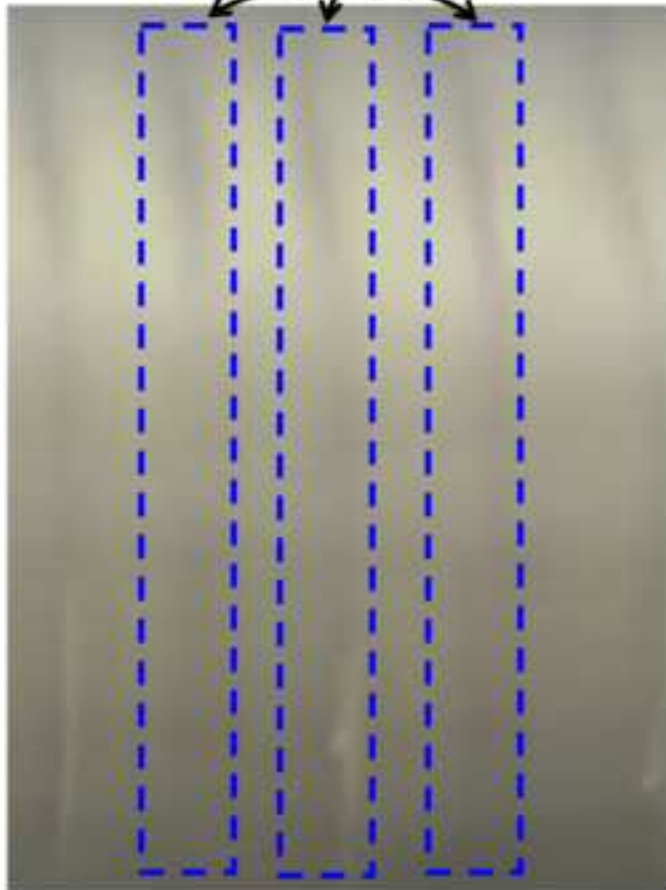


Figure6  
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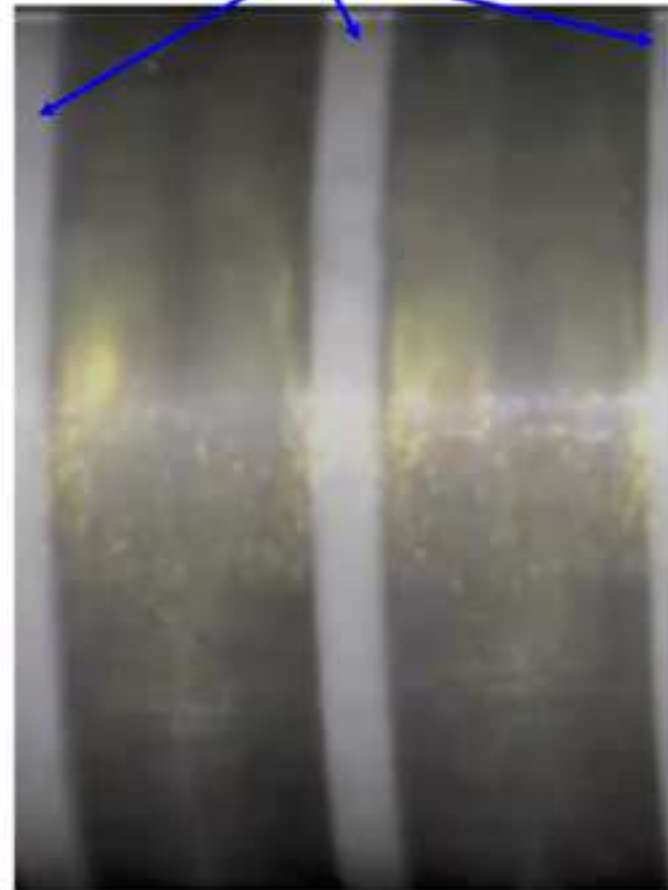
(a)

Wavy motion at the inflow boundary



(b)

Rib

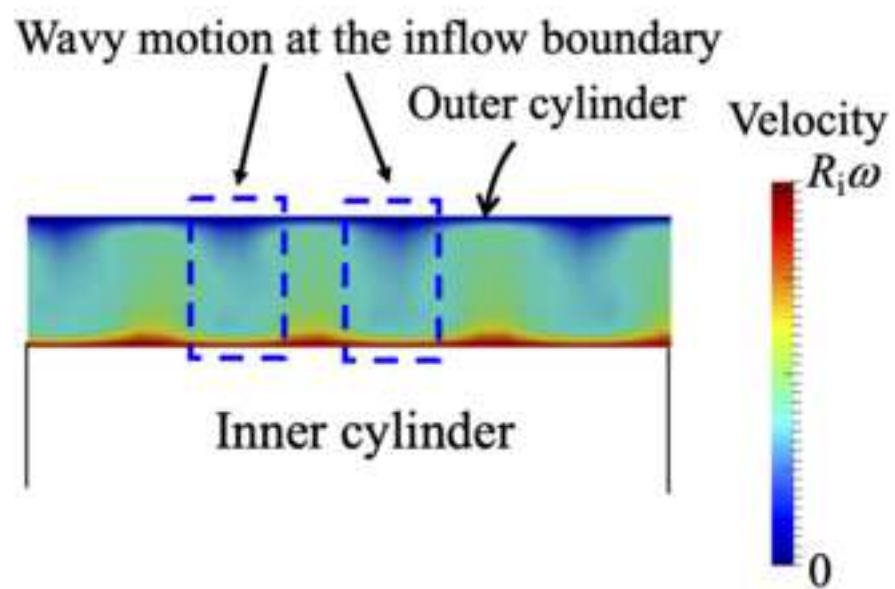


A pair of vortices

Figure7

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(a)



(b)

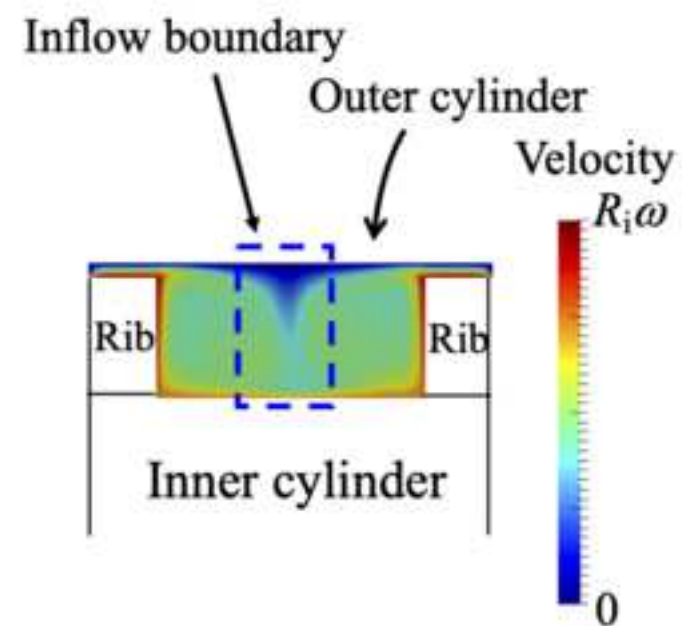


Figure8

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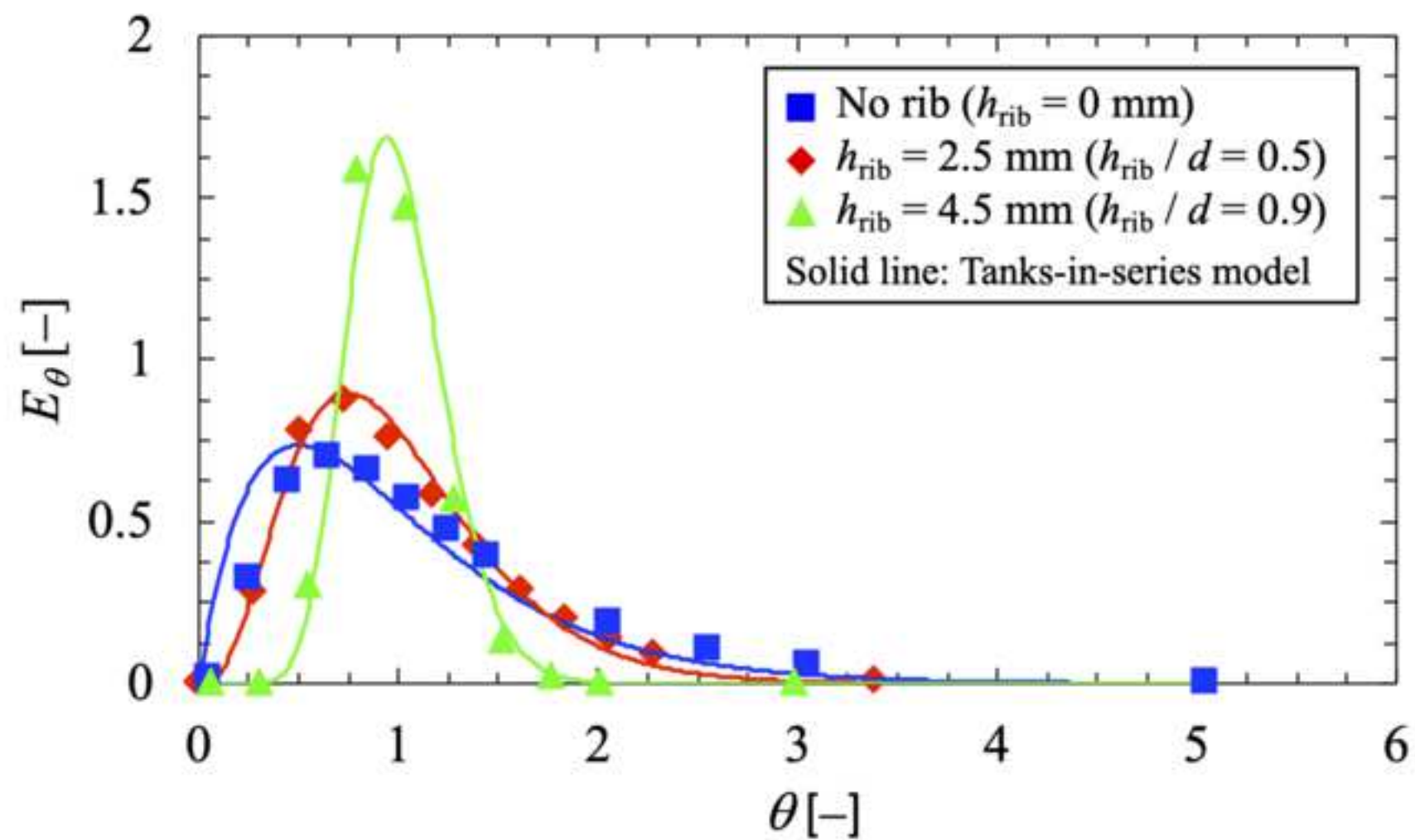


Figure9

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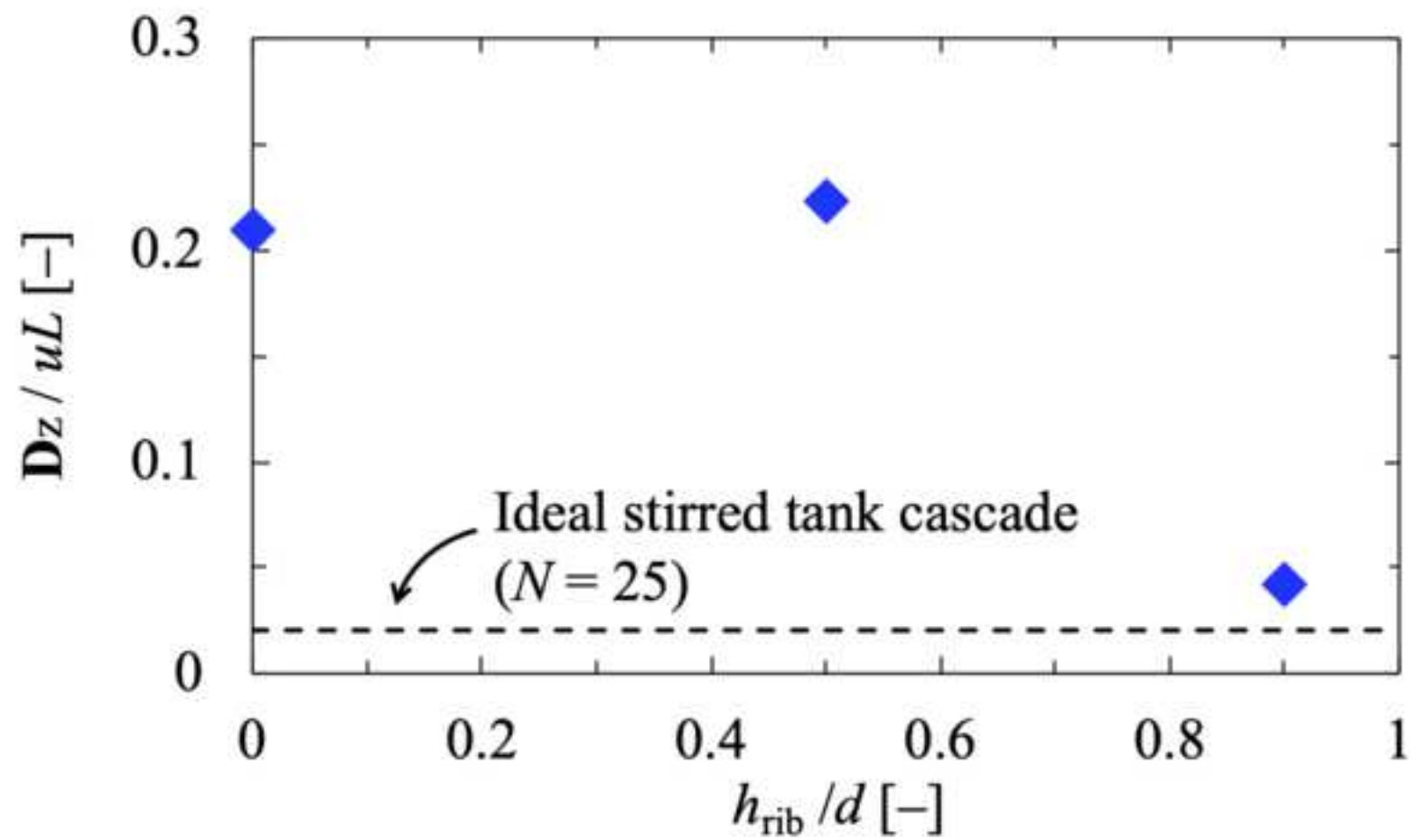


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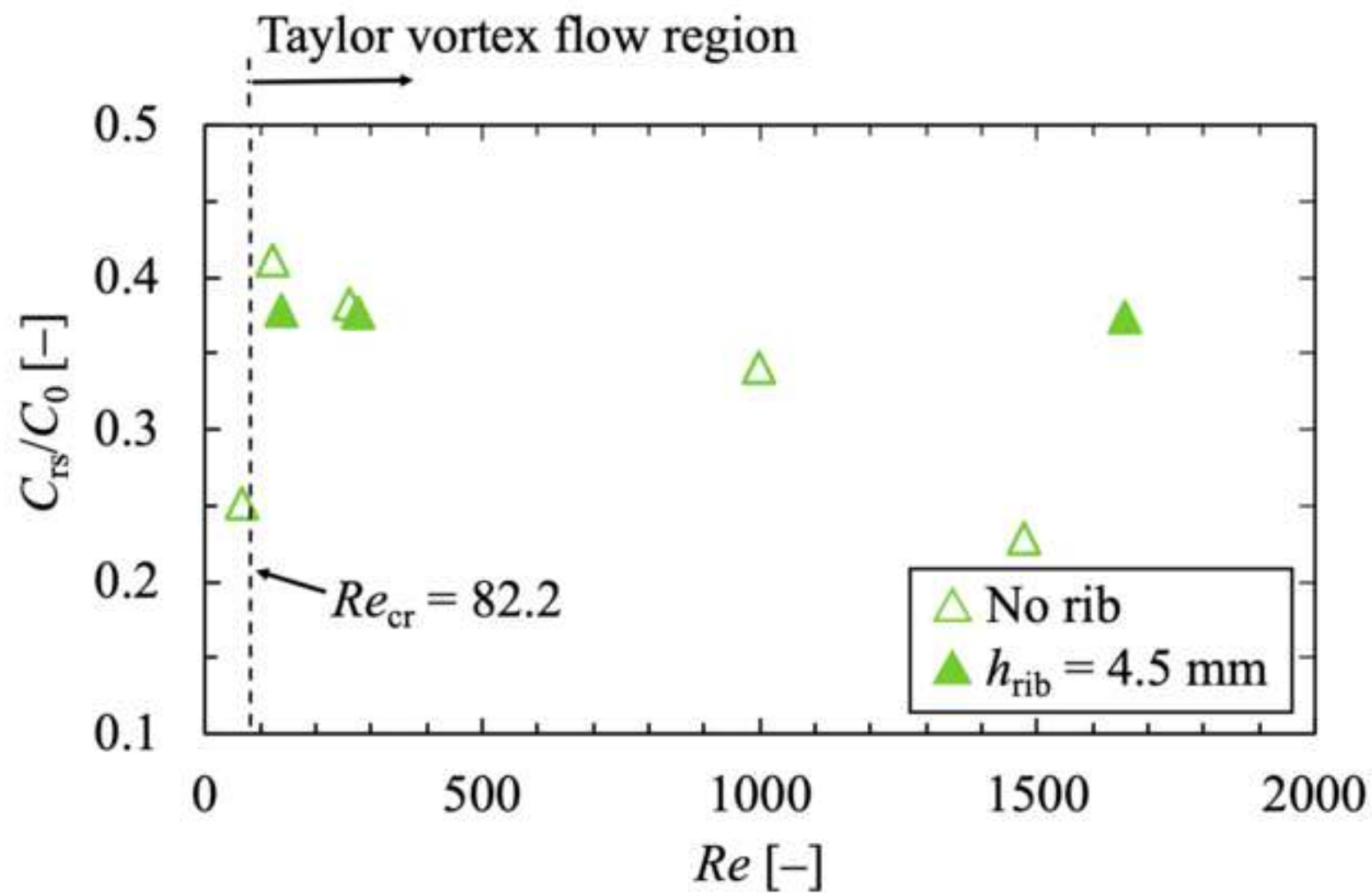


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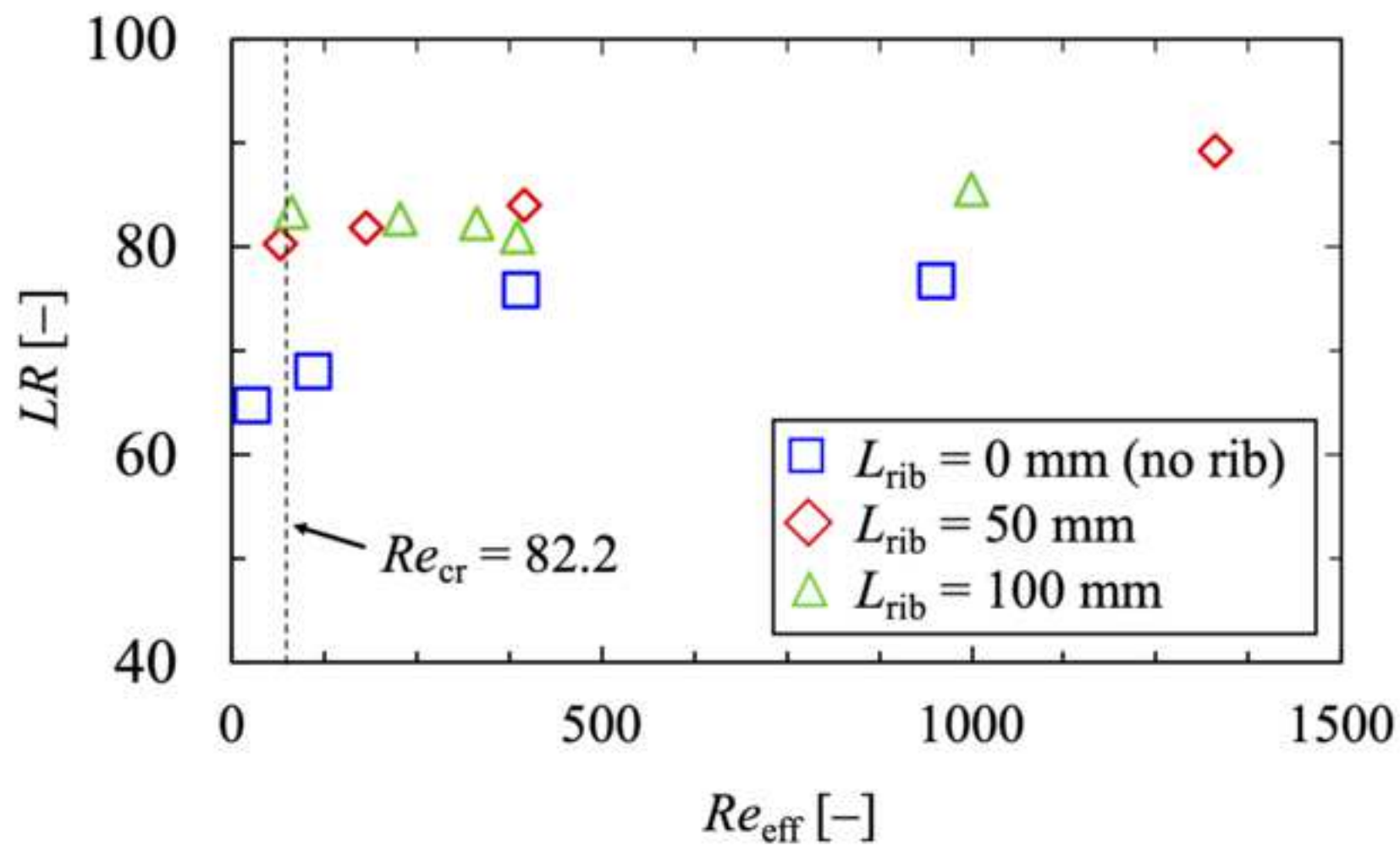


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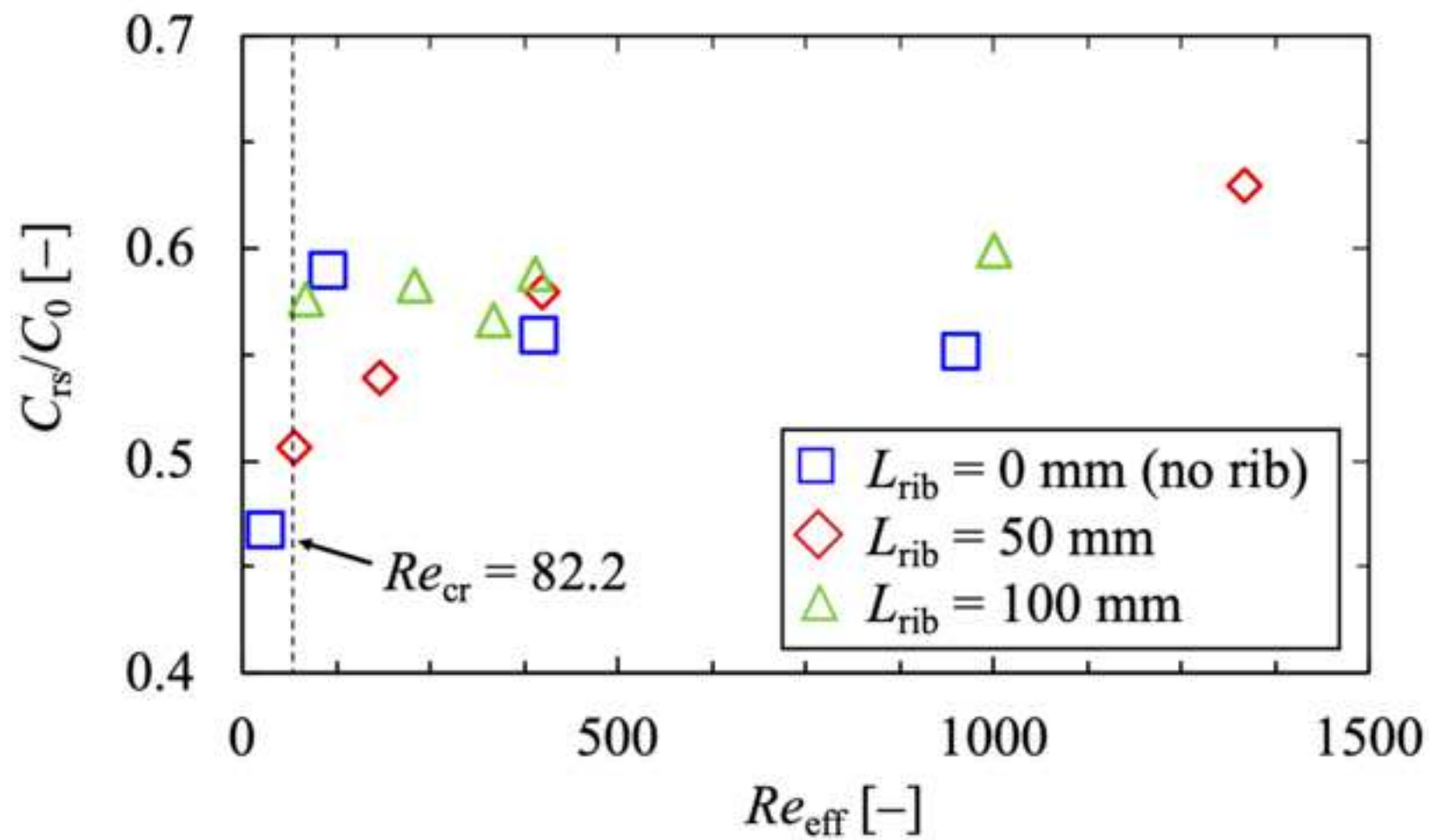


Figure13

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