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# Modeling and Simulation for Feasibility Study of Taylor-Couette Crystallizer as Crystal Seed Manufacturing System

**Kiho Park and Dae Ryook Yang***

*Department of Chemical and Biological Engineering, Korea University,*
*Seoul, Korea, (e-mail: dryang@korea.ac.kr)*

Abstract: Taylor-Couette reactor (TCR) consists of two concentric cylinders with small gap and typically the inner cylinder is rotating. In the concentric gap of the reactor, special patterns of flow are generated. Taylor-Couette flow has a Taylor vortex that is characterized by axisymmetric toroidal vortices. This phenomenon was first discovered by Taylor in 1923. Recently, the Taylor-Couette reactor has been used as a crystallizer and phase-transformation device. For example, the phase transformation of Guanosine 5-monophosphate (GMP) investigated by using the drowning-out crystallization method. Also, the Taylor-Couette reactor is used as a crystallization device experimentally and numerically, in which the fine particles are classified. In our previous work, the dynamic characteristics of Taylor-Couette flow and the possibility of particle classification were investigated numerically and experimentally for the case of vertical TCR in a continuous system. However, none of these studies involves the crystallization modeling due to the difficulty of simultaneous modeling of fluid dynamics and PBE.

In the present study, a CFD-PBE model is developed for verifying the performance of continuous TCR as crystallizer and classifier. A saturated ammonium sulfate solution is used as the sample substance. This CFD-PBE model is based on the Eulerian multi-phase model to describe the liquid-solid two phase flow and the discrete method PBE model including nucleation and growth kinetics. Since the PSD of TCR is much narrower than CSTR, TCR could be adjusted as crystallizer for seed manufacturing.

***Keywords**:* Taylor-Couette Crystallizer, Modeling, CFD, Seed Crystallization

## 1. INTRODUCTION

In crystallization process, seed that has narrow size distribution is needed to produce good crystals. The CSTR is popularly used in crystallization. The reactor is nearly perfect mixing so that residence time of materials inside of reactor is almost same. Regardless of particle size, crystals are mixed state at the outlet. This property has negative effect on seed size distribution.

A Taylor-Couette reactor, an alternative to common rotating devices, is often used for mixing, such as in a bioreactor or a catalytic chemical reactor. Such a reactor has also been recently applied in the field of crystallization due to its peculiar flow characteristics (Campero and Vigil, 1997, Jung et al., 2000, Lee and Lueptow, 2001, Rudman, 1998). The flow is generated in the gap between two concentric cylinders. Generally, the characteristics of this flow depend upon a dimensionless variable, Taylor number (Ta), which in itself is a function of Reynolds number (Re). When the angular velocity of the inner cylinder is increased, Ta is increased, and above a certain threshold, the Couette flow changes to another steady-state flow called Taylor-Couette flow. Taylor-Couette flow has a Taylor vortex that is characterized by axisymmetric toroidal vortices. This phenomenon was first discovered by Taylor in 1923 (Taylor, 1923). By subsequently increasing Ta, Taylor-Couette flow develops into wavy flow as the Taylor vortices become unstable. As a result, the size of the Taylor vortices changes with a certain time period named the azimuthal wave number (Abshagen and Pfister, 2008, Rudman, 1998, Weinstein, 1977, Xiao et al., 2002). If the angular velocity of the inner cylinder is further increased, turbulent flow occurs. In the turbulent Taylor-Couette flow region, the shapes of the Taylor vortices are ill-defined and the reactor provides a pronounced mixing effect (Atkhen et al., 2000, Brandstater and Swinney, 1987, Dong, 2007, Pirro and Quadrio, 2008). These regions are classified on the basis of the ratio of Re and $Re_c$ ($R' = Re/Re_c$). When $R'$ is smaller than unity, the flow is defined as Couette flow. When $R'$ is between unity and 5.5, the flow changes from Couette to Taylor-Couette flow, and when $R'$ is between 5.5 and 18, the flow is wavy flow. Finally, turbulent Taylor-Couette flow is observed when $R'$ is more than 18 (Wang et al., 2005). The value of $R'$ for these different regions depends up on the ratio of the inner cylinder radius ($r_i$) to the outer cylinder radius ($r_o$), $\eta = r_i/r_o$.

Moving behavior of the crystals or particles in the Taylor-Couette reactor was also investigated by many groups. Wereley et al. simulated the motion of dilute, rigid, spherical particles in the Taylor-Couette flow using computational particle tracking in an analytic velocity filed (Wereley and Lueptow, 1999). They found that the motion of particles are significantly affected by gravity force and density ratio between particle and fluid. If the density ratio is nearly 1, neutrally buoyant particles follow fluid streamlines closely. Because of gravity force, the more density ratio increased, the motion of particles didn't follow the Taylor-Couette flow filed. Ohmura et al. studied the classification of fine particles by using Taylor-Couette reactor experimentally and numerically (Ohmura et al., 2005). They found that the large particles were

located near the outer edge of the vortices and quickly moved because of the effect of bypass flow. Therefore, they explained that the classification of particles through the use of the Taylor-Couette reactor is possible for solutions and solutes in similarly dense systems. However, there is a discrepancy between solution density and solute density in real crystallization system. The density ratio of potash alum and ammonium sulfate with water solvent is 1.65 and 1.43 at 25℃ saturated state. There is a need to investigate the effect of gravity on the crystallization in Taylor-Couette reactor.

Recently, computational fluid dynamics (CFD) program has been used to simulate crystallization process in various reactors (Cheng et al., 2012, Moguel et al., 2010, Yan et al., 2011). For example, Cheng et al. simulated the continuous precipitation of BaSO4 with a premixed feed in a stirred tank (Cheng et al., 2012). The population balance equation is closed with the multi-class method. Although there are some numerical diffusion, the predicted crystal size distribution (CSD) is in good agreement with the experimental measurement.

In this study, the characteristics of crystallization in Taylor-Couette crystallizer are investigated and compared with the CSTR for the seed crystallizer. In order to simulate crystallization process in Taylor-Couette crystallizer, multi-phase model and population balance model are solved simultaneously using commercial CFD software (Fluent 6.3.26). Effects of gravity on the crystallization is investigated by changing slope of Taylor-Couette crystallizer. Also, other factors like temperature, flow rate, and rpm of inner cylinder influencing the crystallization are investigated and analyzed.

## 2. THEORETICAL BACKGROUND

### 2.2.1 Crystallization process

Crystallization is the process of formation of solid crystals precipitating from a solution, melt or more rarely deposited directly from a gas. The crystallization of solid phase from liquid and gaseous solutions can only occur if there are some degree of supersaturation or supercooling in the system. The supersaturated state means that the solution has to contain more solute entities dissolved than it would contain under the equilibrium. This can be achieved by various methods, with solution cooling, addition of a second solvent to reduce the solubility of the solute, chemical reaction and change in pH being the most common methods used in industrial practice. The degree of supersaturation or deviation from the equilibrium saturated condition is the prime factor controlling the crystallization process and is the driving force for both the initial nucleation step and the following crystal growth.

### 2.2.2 Nucleation

Nucleation is the initiation of a phase change in a small region, such as formation of a solid crystal from a liquid solution. It is a consequence of rapid local fluctuations on a molecular scale in a homogeneous phase that is in a state of metastable equilibrium. Total nucleation is the sum effect of primary nucleation and secondary nucleation.

Primary nucleation is the initial formation of a crystal for all cases of nucleation, homogeneous or heterogeneous, in systems that do not contain crystalline matter. Homogeneous nucleation is not influenced in any way by solids and heterogeneous nucleation induced by foreign particles. Homogeneous nucleation rarely occurs in practice due to the high energy necessary to begin nucleation without a solid surface to catalyse the nucleation.

Primary nucleation has been modelled with the following (Vedantam et al., 2006).

$$B = \frac{dN}{dt} = k_n (c - c^*)^n$$

where $B$ is the number of nuclei formed per unit volume per unit time, $N$ is the number of nuclei per unit volume, $k_n$ is a rate constant, $c$ is the instantaneous solute concentration, $c^*$ is the solute concentration at saturation and $n$ is an empirical exponent.

Secondary nucleation is the formation of nuclei in the vicinity of crystals present in a supersaturated system.

Secondary nucleation has been simply modelled with the following (Vedantam et al., 2006).

$$B = \frac{dN}{dt} = k_1 M_t^{\,j} (c - c^*)^n$$

where $k_1$ is a rate constant, $M_t$ is the suspension density, $j$ is an empirical exponent.

### 2.2.3 Crystal growth

Once the first small crystal, the nucleus, forms it acts as a convergence point for molecules of solute touching the crystal so that it increases its own dimension in successive layers. There is no simple or generally accepted method of expressing the rate of crystal growth, since it has a complex dependence on temperature, supersaturation, size, system turbulence and so on. However, for carefully defined conditions crystal growth rates may be expressed as a mass deposition rate $R_G$ ($kg/m^2s$), a mean linear velocity $\bar{v}$ (m/s) or an overall growth rate G (m/s).

Growth rate has been simply modelled with the following (Gu and Fahidy, 1985).

$$G = \frac{dm}{dt} = K_g (c - c^*)^n$$

where $K_g$ is a rate constant.

## 3. MODELING

Ammonium sulfate ($(NH_4)SO_4$) is an inorganic salt with a number of commercial uses. The primary use of ammonium sulfate is as a fertilizer for alkaline soils. It is also used as an

agricultural spray adjuvant for water soluble insecticides, a laboratory use for purifying proteins, a food additive and the preparation of other ammonium salts. 30℃ supersaturated solution was used for the simulation.

### 3.1. Geometric Condition

All simulations were carried out in grid with 2D axial symmetry. The geometry of structure is shown in Fig 1. Although there may be a difference between real 3D and 2D axial symmetry, when considering the entire crystallizer its impact is negligible.

Basically axial flow direction is same with gravity direction and slope of crystallizer is changed to analyze the effect of gravity.

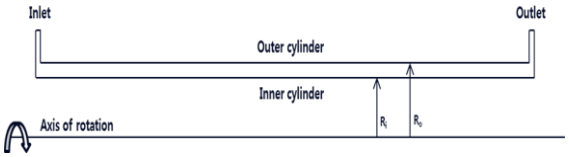


**Figure 1. Geometry structure of TCC model and slope**

### 3.2. Boundary Conditions

Inlet condition was chosen as a velocity inlet so that volumetric flow rate varied from 10ml/min to 40ml/min and inlet concentration and temperature are maintained 0.8 and 30℃. Mean residence time (τ) of fluid is defined by the volumetric flow rate. Outlet condition was selected as a pressure outlet and the pressure was set 1 atm at the same atmospheric pressure. Wall temperature was maintained 20℃ for cooling crystallization. The wall roughness constant was ignored and shear condition was chosen as a no slip condition. The inner cylinder was rotated at some radian/sec by using swirl condition. In this conditions, $Re_c$ is 167.8 and the inner cylinder rpm was calculated at 37.84 to simulate under the same condition in Ohmura et al (Ohmura et al., 2005).

## 4. RESULTS AND DISCUSSION

The TCC is compared to the CSTR for seed crystallizer. The CSTR was simulated using MATLAB code in a perfect mixing state and same operating conditions.

Figure 2 shows the PSD of the TCC and the CSTR. From the Fig. 2a), we can know that the TCC has more narrow size distribution than the CSTR. Fig. 2b) represents normalized number density. By normalizing the number density, we can qualitatively compare the PSD. The normalized number density is defined as

$$\text{normalized number density} = \frac{number\ density}{\sum number\ density} \quad (1)$$

In comparison with the PSD of the CSTR, the TCC has PSD which small size and over size crystals are removed. The discrepancy results from the difference between the features of two crystallizers. The CSTR has same residence time for all crystals, but residence time of crystals in the TCC depends on particle size as mentioned above.

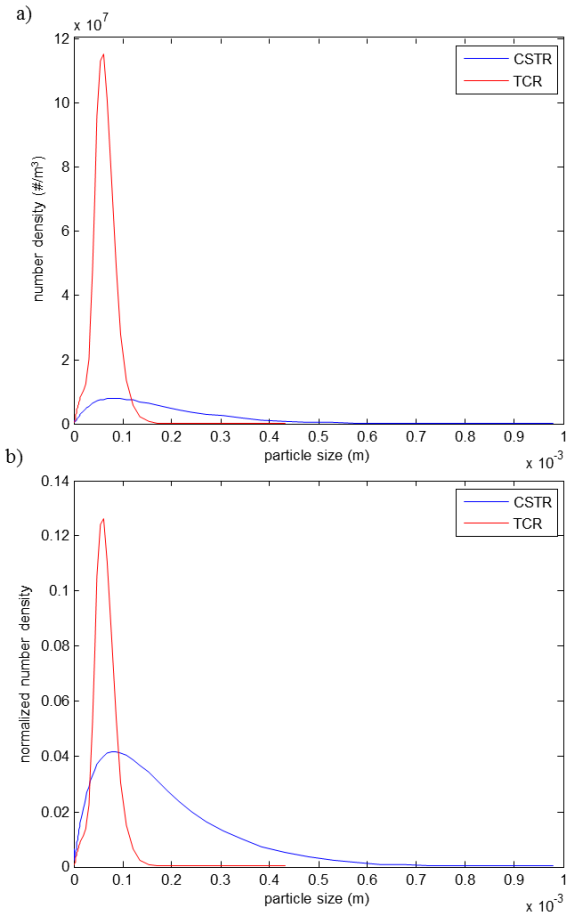


**Figure 2. The PSD of the TCC and the CSTR: (a) number density (b) normalized number density**

In order to get seed crystals which have narrow size distribution, the CSTR needs additional step like as control of metastable limit. The TCC can easily produce seed crystals by its features.

## 5. CONCLUSION

The Taylor-Couette crystallizer was modelled and simulated with CFD. For describing crystallization, population balance equations was also modelled. For satisfying the target of seed manufacturing, Taylor-Couette crystallizer was compared with CSTR crystallizer. Particle size distribution of TCR is much narrower than CSTR seriously, so the feasibility of TCR for seed manufacturing process could be verified. This results could be used as reference data if someone would want to make seed manufacturing process.

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