# Drowning-out 결정화를 위한 쿠에트-테일러 결정화기의 테일러와류 효과 연구

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# Development of Continuous Couette-Taylor crystallizer for drowning-out crystallization of GMP Effect of Tayor vortex on GMP crystallization

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## I. Introduction

Crystallization of GMP by downing-out was investigated using methanol as an anti-solvent in Couette-Taylor crystallizer for production of GMP crystals at room temperature. Generally, the crystallinity, mean crystal size and the recovery are strongly influenced by fluid dynamic conditions via perfect mixing in the crystallizer. This paper examined the effects of mixing properties of Taylor vortex on the crystallinity, mean crystal size and the recovery of GMP solution. In order to do this, the crystallization process was observed under different working conditions by varying the concentration of reactant, the agitation speed of the inner cylinder and the geometry of reactor. We find that fluid dynamic conditions resulting from the rotating speed of the inner cylinder and the geometry of crystallizer, play the key role in the dissolution of amorphous transformation to GMP crystals and the nucleation-growth of GMP crystals. Our experimental results indicate that the crystallization of GMP can obtain one hundred percent of crystallinity and recovery at 300rpm. Particularly, the crystallization of GMP in liquid-liquid Couette-Taylor crystallizer by drowning-out can be predicted from the operating conditions.

## II. Experiments.

Generally, Taylor vortices exhibit a unique mixing behavior of excellent radial mixing combined with minimal axial mixing. In the range of the rotating speed, turbulent Taylor vortex flow appears when the rotating speed of the inner cylinder is larger than the critical value in Fig.1 [1-3].

The experiment setting is illustrated in Fig.2. First, GMP crystallization was initiated by feeding the aqueous reactants of GMP solution and methanol solution. GMP material is supplied by CJ. Cor (Korea), while methanol solution is purchased from Duksan Pure Chemical Co., LTD, (Korea) without further purification. Initially, the crystallizer is filled with deionized water and then each reactant

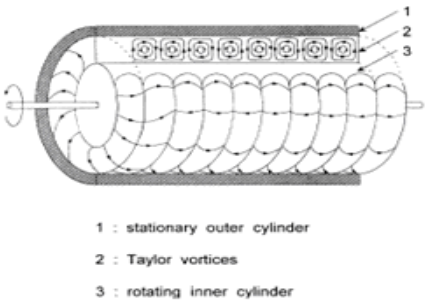


Figure. 1 Schematic picture of Taylor vortices

solution is fed into the crystallizer by pump (StepDos 08, KNF, Germany). The steady-state of Couette-Taylor crystallizer is continuously inspected by UV-Vis (JASCO, V-570, USA) at the end of the crystallizer to define the concentration of GMP in solution after filtering out by 0.45μm membrane filter (Durapore® Membrane Filter, Type HV, Ireland). The filter crystals are dried in a keep drier at room temperature. The mean crystal size and size distribution in the suspension are then analyzed using the particle size analyzer (Mastersizer/E, Malvern, USA), while the shape of each crystal is analyzed by using the Microscope (Video Microscope IT System, Sometech, USA). In addition, the crystallinity of GMP solid is measured by FT-IR (Perkin, System 2000, USA). The operation of GMP Couette-Taylor crystallizer is summarized in Table. 1.

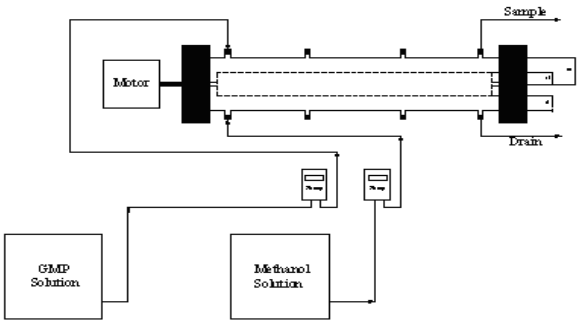


Figure 2. Schematic picture of experimental system

Table 1.

| Couette-Taylor crystallizer operation |
|---|
| Agitation speed N : 300, 500, 700 (rpm) or Energy dissipation ε: 11.3, 40.1, 92 (W/kg) |
| GMP concentration : 60, 90, 120, 150 g/l. |
| Methanol solution/GMP solution : 5:5 |
| Mean residence time : 7 (min) |
| Temperature : room temperature ($25^0$C) |
| Crystallizer's geometry $d/r_i$ : 0.11;0.25;0.34 |

## III. Results and Discussions.

The steady-state achieved after waiting for five times of the mean residence time

1. *The crystallinity* is one of the most important factors determining the amorphous transformation to GMP crystals. The transformation consists of two processes: the dissolution of amorphous and the nucleation-crystal growth of GMP crystals [12-16]. Many experiments have shown that the dissolution of amorphous, the nucleation-crystal growth of GMP crystal increase with an increased fluid dynamic condition, resulting from rotating speed N or energy dissipation ε input to the solution in crystallizer [11-16].

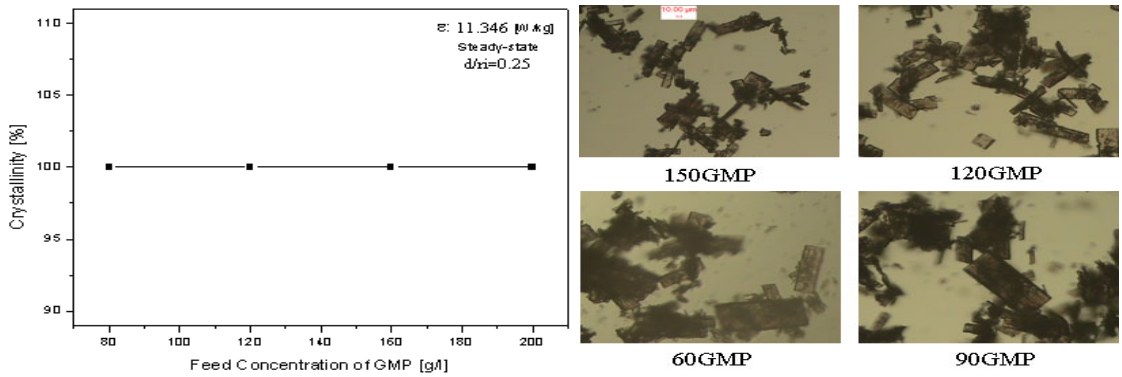


Fig 3. Correlation of crystallinity with feed concentration and GMP crystals at steady-state

With perfect suspension fluid in the Couette-Taylor crystallizer [1-9], the results indicated that the crystallinity obtained one hundred percent for various feed concentration at 300rpm (ε: 4.1677 W/kg) in Fig.3.

2. *The recovery* of GMP solution in Couette-Taylor crystallizer measured with respect to the concentration of GMP solution, is displayed in Fig.4-5. According to Fig.4, the increased concentration of feed GMP solution, the recovery decreases at the same energy dissipation. However, the results in Fig.5-Fig.6 indicate that the higher energy dissipation via the raised rotating speed of inner cylinder and the reduced geometry of crystallizer via $d/r_i$, the recovery of GMP solution increase. Because the higher energy dissipation exhibites the higher suspension fluid, leading to the higher nucleation and crystal growth of GMP crystal. Thus, the concentration of the GMP solution decreases [11].

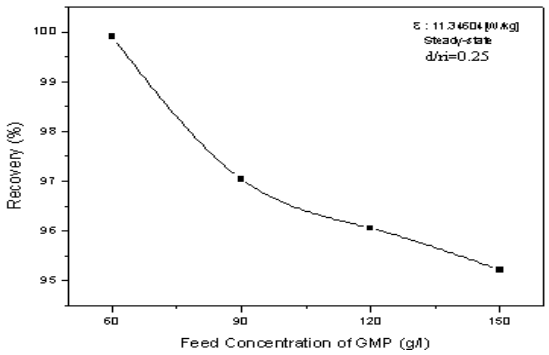


Fig 4. Variation of recovery with feed concentration at steady-state

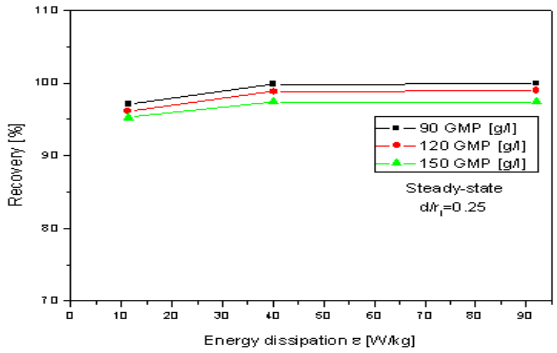


Fig 5. Variation of recovery with energy dissipation at steady-state

3. *Mean crystal size*. The fluid dynamic conditions influence not only on the crystallinity, recovery but also on the mean crystal size of GMP crystallization due to the effects on the nucleation. An increased nucleation creates more nucleis, thereby GMP crystals will have small size. Therefore, the results in Fig.7 indicate that the mean crystal size decreases as energy dissipation increases under the same condition operation.

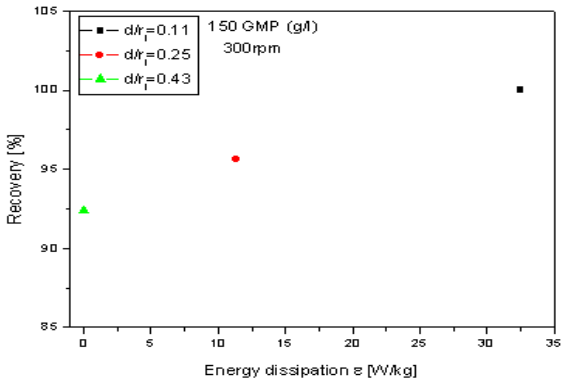


Fig. 6 Mean size of GMP crystal with energy dissipation

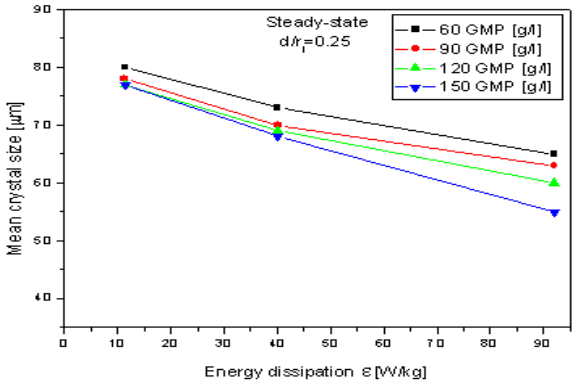


Fig. 7 Mean size of GMP crystal with energy dissipation

## IV. Conclusion

GMP crystallization in Couette-Taylor crystallizer is investigated with different operating conditions, such as feed concentration, agitation speed of the inner cylinder and geometry of crystallizer. It has been found that the crystallinity, mean size, and the recovery strongly depend on the fluid dynamic conditions via energy dissipation ε input to GMP solution. Due to the unique behavior of Taylor vortex with perfect mixing, the experiment results show that the crystallization of GMP obtained up to one hundred percent of crystallinity and recovery at 300rpm (feed concentration=60 GMP g/l, $d/r_i$=0.25) in the Couette-Taylor crystallizer within only 7 minutes of mean residence time.

## V. References

[1] Sung Hoon Kang, Woo-Sik Kim, Journal of Crystal Growth 254 (2003) 196-205.
[2] Wang Mo Jung, Woo-Sik Kim, Chemical Engineering Science 55 (2000) 733-747.
[3] Dong-Myung Shin, Woo-Sik Kim, Chemical Engineering of Japan 35 (2002)1083-1090
[4] Taesung Jung, Woo-Sik Kim, Crystal Growth & Design 4 (2004) 419-495
[5] Kataoka, Futagawa, Chemical Engineering of Japan, 8 (1975) 472-476.
[6] Kataoka, Takigawa, AIChE Journal 27 (1981) 504-508.
[7] G.. Desmet, G.V. Baron, Chemical Engineering Science 51 (1996) 1287-1298.
[8] M. Rudolph, R. M. Lueptow, Physica D 121 (1998) 163-174.
[9] R. A. Leonard, G.J. Bernstein, AIChE Journal 27 (1981) 495-503.
[10] L. J. Forney, R. A. Holl, Seperation Science and Technology, 37 (13), 2967-2986 (2002).
[11] Roger Davey, Jonh Garside, From Molecules to Crystallizers, Oxford University Press (2000).
[12] Shoji Maruyama, Hiroshi Ooshima, Jyoji Kato, Chemical Engineering 75(1999) 193-200.
[13] Shoji Maruyama, Hiroshi Ooshima, Chemical Engineering Journal 81 (2001) 1-7.
[14] R.J. Davey, J. Richards, Journal of Crystal Growth 71 (1985) 597-601.
[15] P.T. Cardew, R.J. Davey, Pro. R. Soc. Lond. A 398, 415-428 (1985).
[16] R.J. Davey, P.T. Cardew, Journal of Crystal Growth 79 (1986) 648-653.