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# Flow Field of Stirred Tank Used in the Crystallization Process of Ammonium Sulphate

**Zhenxing Zhu and Hongyuan Wei***

School of Chemical Engineering, Tianjin University, Tianjin, 300072, China.

* Corresponding author, E-mail: david.wei@tju.edu.cn

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**ABSTRACT:** This paper gives a brief introduction to the crystallization process and basic theory of computational flow dynamic (CFD), before discussing CFD simulation of batch crystallization of ammonium sulphate. Three types of impellers and two dimensions of tanks were applied in observing the changes of flow field. A popular CFD code, Fluent, was used to simulate stirred tanks used in the process, and the sliding mesh (SM) method was adopted to describe the rotating phenomenon. After comparison of the simulation results, an optimal crystallizer can be obtained.

**KEYWORDS:** CFD, ammonium sulphate, crystallization, flow field, sliding mesh.

## INTRODUCTION

Crystallization is an important unit operation in chemical industry since it often brings the final product to the market. Particle size distribution (PSD) is an important parameter for the solid product. Prediction of particle size distribution in a crystallization process is a key problem in the study of industrial crystallization field. However, it is difficult to predict the local particle size distribution in a tank because the concentration of solid suspension strongly depends on the tank geometry and the operating conditions. For decades, the Mixed Suspension Mixed Product Removal (MSMPR) model has been widely used for that purpose[1]. The assumption of ideality in the MSMPR model makes it difficult to attain MSMPR conditions in an industrial scale crystallizer since it is difficult to get uniform suspension of the different sizes of particles[2]. In practice, there are other types of crystallizers to be applied, for example a baffled tank with different kinds of stirred impellers. Draft tube baffled (DTB) crystallizer can provide an upward flow in the center of crystallizer and a circular flow in the whole tank, by which a relatively well mixed flow field can be obtained to achieve a preferable PSD[3].

Crystallization processes are frequently carried out batchwise in baffled vessels operating under turbulent flow conditions. Such processes are strongly influenced by the hydrodynamic and mixing characteristics, which, in turn, depend on the impeller configuration[4]. Many experimental investigations on hydrodynamics and mixing in vessels of different geometrical configurations agitated by various types of impeller were carried out in the past[5,6]. Over the last two decades, many CFD modelling studies of turbulent flow in agitated vessels with and without baffles have been reported in the literature[5,7,8]. However, most of these studies have focused on the flow generated by a Rushton turbine in baffled vessels, which could only present radial flow. A common radial impeller and two axial impellers have been investigated in this paper. In studies carried out until the late 1990s, the rotating impeller was not explicitly modeled and available experimental flow data for a given impeller type was imposed as boundary conditions at the surfaces of the impeller swept region. This approach is referred to as the impeller boundary condition (IBC) method. In the early 1990s, fully predictive modeling methodologies, known as the sliding-mesh (SM) method[9], the inner–outer (IO) method[10] and the multiple-frames of reference (MFR) method[11], were developed which allowed explicit modeling of the impeller geometry without recourse to any experimental data. The SM method is based on a transient flow calculation procedure, while the other two involve steady-state flow calculations. In this paper, the SM method was adopted to simulate the interaction between various impellers and baffles.

## MODELING APPROACH

In the simulation of the flow inside the crystallizer for ammonium sulphate, equations for the conservation of momentum and turbulence quantities have been solved. The variables solved include Cartesian velocity components, pressure and turbulence quantities using the continuity and Navier-Stokes equations:

$$\frac{\partial \rho}{\partial t} + \nabla \bullet (\rho \vec{U}) = 0 \qquad (1)$$

$$\frac{\partial(\rho\vec{U})}{\partial t}+\nabla\bullet(\rho\vec{U}\otimes\vec{U})=-\nabla P+\nabla\bullet(\vec{\tau})+\rho\vec{g} \qquad (2)$$

Since the flow is in the turbulent regime, the quantities in the above equations are Reynolds-averaged, and $\vec{\tau}$ is the effective turbulent stress tensor. By applying the eddy viscosity hypothesis, the Reynolds stresses can be linearly related to the mean velocity gradients in a manner analogous to the relationship between the stress and strain tensors in laminar Newtonian flow:

(3)

where the effective viscosity is sum of the laminar and turbulent viscosities:

$$\mu_{\text{eff}}=\mu_{\text{L}}+\mu_{\text{T}} \qquad (4)$$

The turbulent viscosity in the liquid phase is calculated using the standard $k$-$\varepsilon$ turbulence model[12]:

$$\mu_{\text{T}}=C_{\bullet}\rho\frac{k^2}{\varepsilon}\sqrt{a^2+b^2} \qquad (5)$$

Turbulence kinetic energy $k$ and turbulence dissipation rate $\varepsilon$ are solved based on the liquid phase velocity field.

## Case Studies

The sample of ammonium sulphate for crystallization was provided by the outgrowth from producing Caprolactam, which is an important raw material of petrochemicals, for example Nylon. The mass fraction, supersaturation, density and relative viscosity of ammonium sulphate in solution are 45% , 2.9004 g/(100g solvent), 1241.6kg/m$^3$ and 3.32×10$^{-3}$Pa.s respectively.

In the last few years, batch vacuum-evaporation and cooling crystallization has been the most popular method for the preparation of high purity ammonium sulphate. The normal mode of operation is by evaporation of a saturated solution of ammonium sulphate in water at a constant temperature of 70 °C and a fixed vacuum at 0.04 MPa, because the temperature of crystal point is about 70 °C. Then the initial crystal population in the cell is grown out by cooling the suspension down. However, the effect of vacuum on the flow field in the tank is so slight that it can be negligible in simulation. There is a slightly boiling phenomenon in the crystallizer to evaporate solvent, when the solution was heated to 80 °C under a fixed vacuum at 0.04 MPa, in order to produce the supersaturation. Then the system was slowly cooled under the vacuum of 0.04 MPa till the crystals were generated at the temperature of 70 °C. During this process, boiling was gradually stopped and never occurred in the following stages. The simulation was performed from this point to describe the flow field of crystallization process without boiling.

It was confirmed, that the velocity distribution did not change significantly when the particle phase was involved, through a confidential multi-phase simulation where Euler-Euler model was applied to describe the interaction between crystal particles and solution. Therefore the assumption that the flow field does not change with the involvement of crystals is reasonable.

To simplify the problem, the simulation was performed in a single phase and the operating pressure and temperature were given constant values of 0.04 MPa and 70 °C, respectively, in the process of CFD simulations, because the flow field at the crystal point was a definitive aspect for the crystallization results.

The crystallizers, inculuding a pilot scale crystallizer with a capacity of 200 ml, a baffled tank with a capacity of 300 l and a same scale DTB, were defined as simulating models. Both a common impeller which was constructed with a shaft and two symmetrical vertical half-crescent blades and often used in laboratory, and a pitch blade turbine (PBT) impeller, which was constructed with a shaft and 4 blades angled up from horizontal at 45 ° were used as impeller models. A clear scale-up procedure of the crystallizer for ammonium sulphate was indicated from this arrangement and the advantages of PBT impeller were also described in this process. A high performance industrial crystallizer can be investigated by the computing results of 300 l DTB with PBT impeller for their structural comparability subsequently.

## Flow Field of Crystallizers by CFD Simulations and Discussion

To know how well the simulation works for the flow field of crystallizers in Fluent6.2, unsteady solver using the $k$-$\varepsilon$ turbulence model was used to simulate the turbulence flow distribution in a stirred tank. The $k$-$\varepsilon$ turbulence model was used for the whole tank. The problem was studied three-dimensionally. SM technology was used to simulate the transient flow in the mixing tank. An approximate steady-state result was obtained by taking a sufficiently long simulation time.

Before the main simulations are commenced it is necessary to consider the mesh required to obtain mesh independent results. The flow field obtained by CFD method for a certain volume is definitely affected by amount of meshes. More meshes will produce more accurate results and consume more time. Four deferent meshing levels were attempted for each case (see Table

1) to validate if the grid was independent in simulation. The results from levels 1 and 2 were quite different from those from levels 3 and 4, and the maximum deviation of the velocity magnitude of all the measuring points in the crystallizers in all cases between levels 3 and 4 was less than 3%. However, the computing speed for each step in level 4 was much slower than that in level 3. The meshing level 3 was chosen in all the cases below. Furthermore, confidential Laser Doppler Anemometry (LDA) results produced for the crystallizer indicate that these simulation results are reasonable.

**Table 1.** grid dividing of each simulating case (elements of mesh)

| Mesh level | 200ml crystallizer | 300l crystallizer without draft tube | 300l crystallizer with draft tube |
|---|---|---|---|
| 1 | 10,000 | 20,000 | 30,000 |
| 2 | 30,000 | 60,000 | 80,000 |
| 3 | 60,000 | 80,000 | 100,000 |
| 4 | 100,000 | 100,000 | 120,000 |

## Flow Field of Crystallizer with Common Impeller

In a pilot scale experiment, the power input into crystallizer is always enough to form a comparatively uniform flow in the whole tank. Therefore, a 350 ml unbaffled tank with 200 ml effective volume was taken into practice and agitated with a two blades common impeller whose diameter is a half of the diameter of tank at a speed of 200 rpm (see Fig 1).

The boundary conditions are: the wall and bottom of tank is no-slip wall, the top of tank is symmetry, the shaft and impeller is no-slip wall, the sliding surface of both inner and outer parts are interface. There are a total of 60,000 elements generated for both inner and outer mesh files of the crystallizer.

The time step of iteration was 0.005 s to satisfy the rule that the courant number should be no more than 1 for an unsteady case. The Courant number is a dimensionless number that compares the time step in a calculation with the characteristic time of transit of a fluid element across a control volume:

$$C = \frac{\Delta t}{\Delta x_{cell} / v_{fluid}} \qquad (6)$$

where $C$ is courant number, $\Delta t$ is time step, $\Delta x_{cell}$ is size of the smallest mesh and $v_{fluid}$ is the speed of the fluid. Regenerating the formula above by changing the Cartesian co-ordinates into cylindrical co-ordinates, the time step $\Delta t$ can be calculated by:

$$\Delta t = (\Delta n \times N)^{-1} \qquad (7)$$

where $\Delta n$ is the number of meshes in $q$ direction when the volume meshes were divided into $r \times \theta \times z$, and $N$ is a rotation speed of impeller. In this case, D$n$ is 50 and $N$ is 200 rpm, or 3.333 rps, so the theoretical time step is 0.006 s. Therefore, the chosen time step 0.005 s was less than 0.006 s, and complied with the demand of the computing precision.

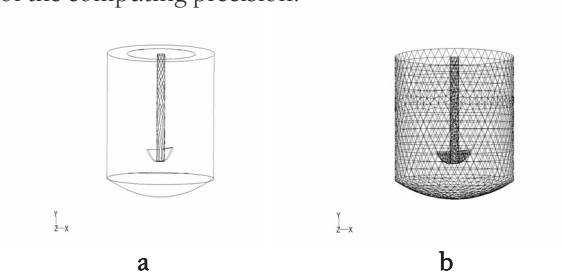


**Fig 1.** Geometry and meshes of a 350 ml crystallizer with a common impeller. **a** Geometry of crystallizer. **b** Meshed volume of crystallizer.

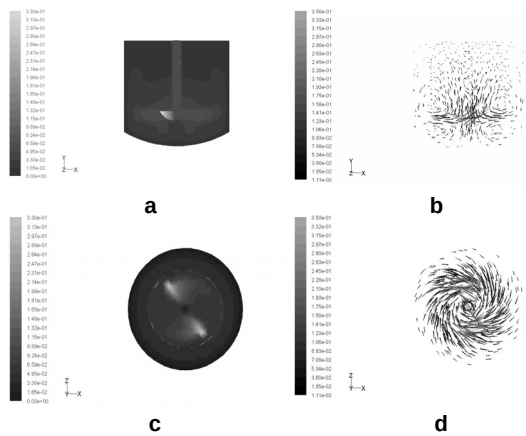


**Fig 2.** Flow field of a 350 ml crystallizer with common impeller at 100 s (velocity/(m/s)). **a** Velocity magnitude distribution of the middle vertical plane in crystallizer. **b** Velocity vector distribution of the middle vertical plane in crystallizer. **c** Velocity magnitude distribution of the middle horizontal plane in crystallizer. **d** Velocity vector distribution of the middle horizontal plane in crystallizer.

An approximate steady-state flow would come into being at about 100 s. The simulation results were shown in Fig 2.

It has been discovered that such kind of a radial impeller would cause an intense flow in the radial direction and a weak flow in the axial direction, which would lead to the breakage of crystals and inadequate suspension of solids in a larger scale crystallizer.

## Flow Field of Crystallizer with PBT

To reduce the breakage of crystals cased by the interaction between impeller and baffles, a 300 l baffled

tank with 250 l effective volume was stirred by an axial impeller PBT, whose diameter had been shortened to one third of the diameter of tank (see Fig 3).

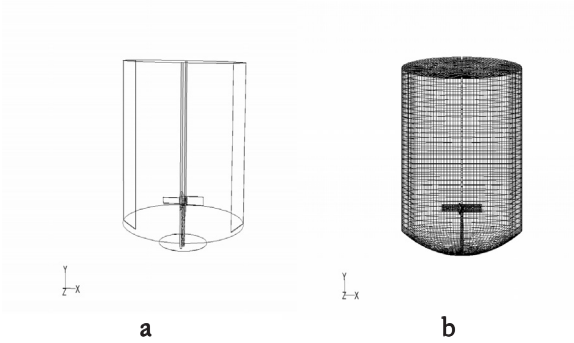


**Fig 3.** Geometry and meshes of a 300 l crystallizer with PBT. **a** Geometry of crystallizer. **b** Meshed volume of crystallizer.

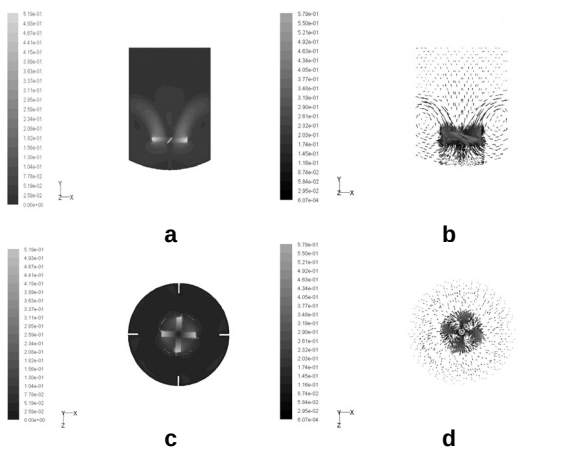


**Fig 4.** Flow field of a 300 l crystallizer with PBT at a speed of 200 rpm at 100 s (velocity/(m/s)). **a** Velocity magnitude distribution of the middle vertical plane in crystallizer. **b** Velocity vector distribution of the middle vertical plane in crystallizer. **c** Velocity magnitude distribution of the middle horizontal plane in crystallizer. **d** Velocity vector distribution of the middle horizontal plane in crystallizer.

The boundary conditions are: the wall and bottom of tank is no-slip wall, the top of tank is symmetry, the shaft and impeller is no-slip wall, the 4 baffles are also no-slip wall and the sliding surface of both inner and outer parts are interface. There are 80,000 elements for the meshed tank, which used the same meshing scheme. Then the steady state flow field at a speed of 200 rpm at 100 s can be obtained and listed in Fig 4.

When the rotation speed was 200 rpm, the shear stress caused by a radial flow declined obviously, but the flow field in the tank was not uniform especially at the top of the tank. There are alternative methods to improve the mixing effect, such as by increasing rotation speed or changing the geometry of tank. By raising the

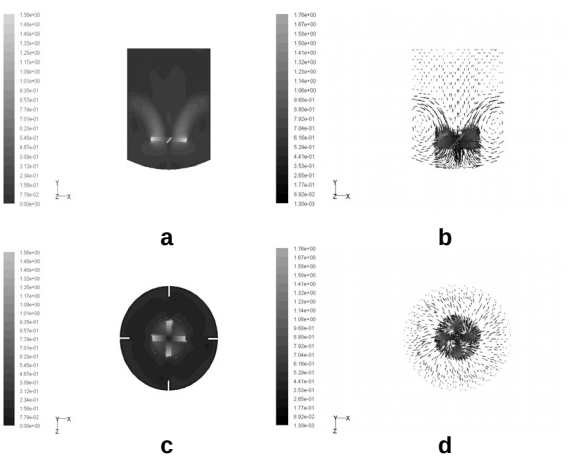


**Fig 5.** Flow field of a 300 l crystallizer with PBT at a speed of 600 rpm at 100 s (velocity/(m/s)). **a** Velocity magnitude distribution of the middle vertical plane in crystallizer. **b** Velocity vector distribution of the middle vertical plane in crystallizer. **c** Velocity magnitude distribution of the middle horizontal plane in crystallizer. **d** Velocity vector distribution of the middle horizontal plane in crystallizer.

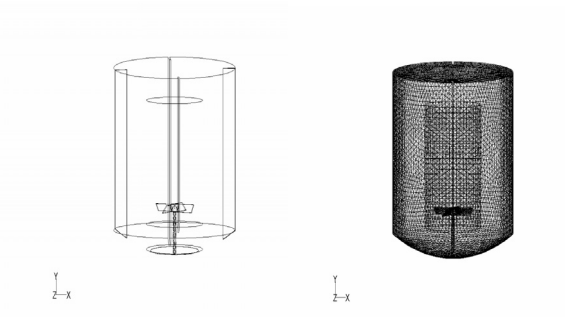


**Fig 6.** Geometry and meshes of a 300 l DTB crystallizer with PBT. **a** Geometry of DTB crystallizer. **b** Meshed volume of DTB crystallizer.

rotation speed to 600 rpm (Fig 5), the acceleration of the rotation speed could not evidently mend flow state but accompany a distinct increase of shear stress.

A popular way to change the flow in the tank was to place inside it a draft tube to construct a DTB crystallizer, the geometry and meshes of which was shown in Fig 6. The same boundary condition was used except that the draft tube is no-slip wall. The DTB crystallizer with 100,000 meshes was circulated at a speed of 200 rpm, and then a new steady flow was gained 60 s later (see Fig 7).

The flow in the crystallizer has been radically altered by addition of a draft tube. An upward flow in the center of tank and a circular flow in the whole tank have been generated, which not only kept a relatively low shear stress in the radial direction, but also

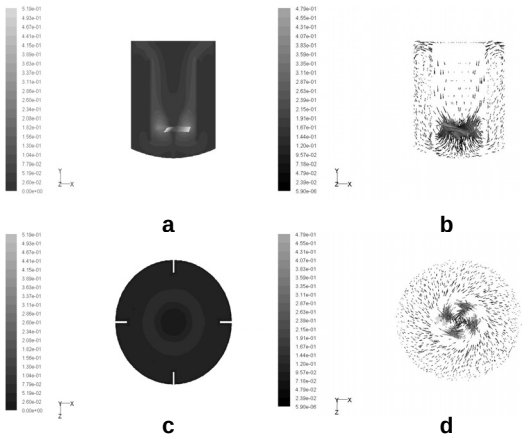


**Fig 7.** Flow field of a 300 l DTB crystallizer with PBT at a speed of 200 rpm at 60 s (velocity/(m/s)). **a** Velocity magnitude distribution of the middle vertical plane in crystallizer. **b** Velocity vector distribution of the middle vertical plane in crystallizer. **c** Velocity magnitude distribution of the horizontal plane above draft tube in crystallizer. **d** Velocity vector distribution of the horizontal plane above draft tube in crystallizer.

produced a uniform flow to a certain extent especially at the top of the vessel. Furthermore, a static region has been formed at the top of crystallizer outside the draft tube (Fig. 7, a and b) where there is a preferable platform for the separation of vapor and liquid with valid vacuum evaporation crystallization.

## Conclusion

A pilot scale crystallizer agitated by a common radial impeller for the crystallization of ammonium sulphate was simulated by using CFD as the basis of the larger scale crystallizers. To reduce the shear stress in the radial direction which causes breakage of crystals, a typical axial impeller PBT was adopted in a 350 l vessel. However, the flow inside the tank does not satisfy the requirements of crystallization, while the shear stress in the radial direction was lowered. Instead of increasing the rotation speed from 200 rpm to 600 rpm, adding a concentric draft tube inside the kettle to create a DTB crystallizer was applied to reform the flow field. It has been demonstrated that a DTB crystallizer with an axial impeller could provide a suitable flow field for a vacuum evaporation crystallization of ammonium sulphate, which would be a reference to build an industrial crystallizer.

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