Application of Computational Fluid Dynamics in Industrial Crystallization

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Abstract. Computational Fluid Dynamics (CFD) simulates a multi-phase flow field by means of numerical calculations and becomes a knot. A powerful tool for crystal process simulation. This paper summarizes the application of CFD technology in the coupling of solving methods, crystallization phenomenon, reaction crystallization process, different operation modes and proportional amplification in the simulation of industrial crystallization, and points out that the development of a suitable model will have a high application value.

1. Introduction
Computational Fluid Dynamics (CFD) is a numerical simulation technique that uses fluids as a research object. Computer simulations provide information about certain fluids under certain conditions. CFD contains a variety of knowledge and related technologies such as mathematics, computer science, engineering, and physics. It integrates these knowledge and technologies on a specific problem and forms an analog model of fluid flow [1].

The purpose of the CFD simulation is to make predictions and gain knowledge of the issues to be studied. The theoretical prediction comes from the solution of the mathematical model rather than the result of an actual physical model. The mathematical model is mainly composed of a set of differential equations. The solution of these equations is the result of the CFD simulation. CFD is an advantageous tool for studying various fluid phenomena and for designing, operating, and studying various flow systems and flow processes. With the rapid development of computer science, modern numerical technology, and fluid mechanics theory, CFD is used to model chemical processes, the design and amplification of chemical reactors and crystallizers are examples of the combination of modern high-tech and traditional science.

2. Introduction to CFD Technology
The basic idea of numerical solution using CFD technology [2] is to use a series of finite discrete points (e.g., nodes, nodes) that are physically continuous fields (such as concentration field, velocity field, etc.) in space and time coordinates. Instead of a set of values on top of it, set up an algebraic equation (called a discrete equation) on the relationship between the values of the variables at these discrete points, and solve the established algebraic equation to obtain the approximate value of the solved variable.
2.1. Basic Control Equations of Fluid Flow
Any flowing fluid follows the law of conservation of mass, the law of conservation of momentum, and
the law of conservation of energy [3]. If the flow contains a mixture or interaction of different
components, the system also observes the conservation of components. If the flow is in a turbulent
state, the system must also comply with the additional turbulent transport equations. The control
equation is a mathematical description of these conservation laws. The motion of the fluid can be
described by the partial differential equation (equation) or the integral equation (equation) of the
conservation law. These equations are called the fluid motion equations. Using certain methods and
means, according to the mathematical model of the special properties of fluid flow (such as turbulence
model, combustion model, multiphase flow model, etc.), boundary conditions, and initial conditions, a
closed system of equations is constructed to mathematically describe specific flow fields and fluids.
Flow law [4]. Solving can get the fluid motion information in the system and the physical process
information related to the fluid flow.

2.2. Numerical Solutions of Control Equations
Because of the nonlinearity and complexity of the motion control equations, it is almost impossible to
directly solve the above equations. However, the information described by the equations is also very
important. Therefore, numerical calculation methods must be used to solve these control equations.
The commonly used numerical solution methods include finite difference method, finite volume
method, finite element method, boundary element method, finite analysis method, spectrum analysis
method, numerical integral transformation method, lattice-Boltzmann method, etc. [5]. The finite
volume method uses the integral conservation equation as a starting point, divides the calculation area
into a series of control volumes, and integrates the differential equation to be solved for each control
volume to obtain a discrete equation. The finite volume method is easy to apply to simulate fluid
motion with complex boundary regions. It is the most widely used method in computational fluid
dynamics. The CFD, PHOENICS, STAR-CD, and FLUENT in the popular large-scale commercial
CFD software are all based on the finite volume method.

3. Application of CFD in Industrial Crystallization
The crystallization process is a complex process of heat transfer and mass transfer. In different
physical (hydrodynamics, etc.), chemical (component composition, etc.) environments, the control
steps of the crystallization process may change and may reflect different crystallization behaviors, all
complicating the mathematical model of the crystallization process [6]. The establishment of a suitable
mold model is of great significance for optimizing the crystallization operation, designing a new type
of crystallizer, and developing a new process for amplifying the crystallization process. The rapid
development of CFD technology not only makes solving complex model equations faster, but also
promotes the progress of model simulation.

3.1. Coupling solution method of particle balance equation in the process of simulated crystallization
The grain number density distribution is an important characteristic parameter of crystal products, and
taking into account the crystal growth rate, nucleation rate, coalescence and fragmentation phenomena
in the grain balance equation is an effective way to accurately describe the crystallization process and
obtain the grain number density distribution of crystal products. Means. Therefore, how to solve the
particle balance equation in the CFD simulation is an important factor in the simulation. The most
critical issue is to solve the coupled solution of the particle balance equation and the flow equation.
In general, crystals have a wide range of particle number density distributions in crystallization,
and different size crystals have their own flow fields. It is therefore unrealistic to describe crystal grain
balance equations with crystals of different sizes in an infinite number of crystal sizes. In order to
simplify the particle balance equation, many researchers have used the method of moment-moment to
simulate the crystallization process, especially the precipitation crystallization process [7 - 17], and
obtained the average properties of the crystal product. In the moment-based method, in order to
examine changes in crystal size distribution due to coalescence and fragmentation, consideration should be given to changes in internal coordinates such as particle size in the particle balance equation. Researchers have proposed some methods to examine the aggregation and crushing of cores\textsuperscript{[18-21]}. However, all methods are based on the assumption of uniform suspension.

The solution to the balance equation of the grain number used in the CFD environment needs further development. Another method that may solve the equation of crystallization balance in the simulation is the MUSIG (Multi-Size Group - Model) model. This model was first proposed by Luo in 1998\textsuperscript{[22]} for the simulation of gas-liquid systems. The MUSIG model defines a two-phase flow. The dispersed phase defines a group of bubbles of different sizes, and simulation can obtain information on the volume fraction distribution of bubbles of different sizes. However, the flow field of the dispersed phase is simulated by the average particle size of the bubble size group, that is, the flow fields obtained by different size bubbles are the same. If this model is applied to a solid-liquid system, the size distribution information of the crystal can be obtained. However, due to the same flow field in the dispersed phase, the motion characteristics of crystals of different sizes cannot be examined. In addition, the phenomenon of crystal coalescence and fragmentation needs further development. The model, in particular, lacks phenomena such as the crystal growth and the difference method.

3.2. Analog Crystallization
In order to accurately describe the crystallization process in the flow field, it is necessary to understand the physical model of the crystallization phenomenon, such as the crystal growth rate, nucleation rate, coalescence and fragmentation phenomena. Years of research have shown that the hydrodynamic state in the mold has a critical influence on the crystallization process\textsuperscript{[23]}. Therefore, many phenomena related to fluid dynamics can be studied using CFD simulations. Zller et al.\textsuperscript{[24]} studied the growth of single-sized crystals in solution in 1999. Mori et al.\textsuperscript{[25]} simulated the crystal growth rate by considering the mass transfer between the solution and different size crystals. The mass transfer coefficient was estimated by the model that defined the local crystal slip velocity and the local energy consumption rate. The nucleation rate is considered to be directly related to the energy of the moving particles, and the motion can be obtained by CFD simulation. Therefore, Ten Cate et al.\textsuperscript{[26]} proposed a model to study the single crystal in the flow field, and obtained crystal collision frequency and energy information that can be used to predict the secondary nucleation rate and particle wear according to the model. Liiri et al.\textsuperscript{[27]} simulated the nucleation rate due to the collision of crystal-propeller and crystal-crystallizer through CFD. Roelands et al.\textsuperscript{[28]} used CFD to simulate the mixing time of the reactants to determine the nucleation rate in the precipitation crystallization. The CFD study of the nucleation and growth model is still in its infancy, and researchers are still looking for better models to simulate the growth and nucleation in the crystallization process.

The phenomenon of coalescence can be described in the CFD environment because it is related to the local shear rate of the fluid. Madec et al.\textsuperscript{[29]} used the Monte Carlo method directly in the CFD environment to describe the coalescence process in a batch reactor. Some researchers have also developed simulation models for the phenomenon of coalescence in precipitation crystallization\textsuperscript{[30–31]}.

3.3. Simulation reaction crystallization process
Most of the CFD simulations are focused on reaction crystallization, or precipitation crystallization. Wei et al.\textsuperscript{[17]} reported for the first time in 1997 that the reaction crystallization process was modeled using the CFD method. The particle size balance equation and the crystallization nucleation and growth kinetics were integrated into the CFD main solver. The simulation results were visually reproduced. The supersaturation of crystallinity in the duct reactor crystallizer and the distribution of the crystal phases, and a comprehensive evaluation of the operating conditions, such as the effect of the feed flow ratio on the particle size distribution of the product. Later, Jaworski et al.\textsuperscript{[10]} simulated the continuous reaction crystallization process. Wei \textsuperscript{[7]}, Zauner \textsuperscript{[9]} and Marchisio \textsuperscript{[11]} simulated the semi-batch reaction crystallization process. In reaction crystallization, the reaction time is usually very fast. For example, the reaction time in a semi-batch stirred crystallizer simulated by Vicum et al.\textsuperscript{[16]}
is only 9 s to 10 s, which is much less than the mixing time. In order to examine the micro-mixing process during the reaction crystallization process, some researchers [11-15] adopted the PDF (Probability Density Function) method, in which the component concentration is described by a multidimensional density function.

3.4. The application of different operating modes in the crystallization process
CFD simulations of the crystallization process of batch cooling solutions are still relatively rare. Yang et al. [32] used CFD to control the batch crystallization process while considering heat transfer. The crystal growth and nucleation rates were taken into account in the simulated grain balance equation, and the 5th-order equations were defined using the method of the order-moment. Kouougoula et al. [33] developed a compartment model to simulate the intermittent cooling crystallization process based on the results of CFD simulation of heat transfer and solid suspension. This model has a good application prospect in the simulation of intermittent cooling crystallization process.

3.5. Simulation of the proportional amplification and mixing for the crystallization process
Other CFD simulation studies are directed at the structure of the crystallizer. Since the crystallizer structure seriously affects the crystallization process, it is useful to study the magnification of the crystallizer in the CFD environment. In addition to its economic advantages, CFD simulations can examine crystallization processes in molds of different sizes and provide a reference basis for the design of the mold. Zauner et al. [8, 37] introduced the model of fine-particle-mixing and intermediate-size mixed-bonded-grain balance to study the effects of mixing conditions on crystal size distribution in the reaction crystallization process. Wei et al. [38] studied the effect of energy input on the reaction crystallization process with different size ratios using the grain-equilibrium equation model with a step-size moment. Since the basic model of the simulated crystallization process is still in development, the scaled-up results have not been satisfactory. The mixing in the crystallizer is an important factor affecting the crystallization process. The crystal suspension in the crystallizer and the distribution of the suspension density are important information for controlling the crystallization process. Supersaturation and the corresponding crystal growth and nucleation rates are directly affected by the local crystal suspension density. Sha et al. [36, 39-40] studied the relationship between the suspension density distribution in the crystallizer and the suspension density distribution at the outlet of the product using a hierarchical function. Therefore, the effect of the suspension density distribution on the size of the crystal product during continuous crystallization was simulated.

4. Conclusion
CFD technology is widely used in many fields. In some fields, it has become a mature tool for research and design. However, the application of CFD to the industrial crystallization process is still in its infancy. Due to the complexity of the crystallization process, it is difficult to conduct a comprehensive study of this process with general test methods and thus no reliable results can be
obtained. Therefore, the application of computational fluid dynamics technology in industrial crystallization process, research and development of a suitable model, so that the basic phenomena and basic laws in the crystallization process can be studied in the multi-phase flow field, thereby enhancing the scientific and reliability of the research results. With high theoretical value and wide application value, its development prospect is very broad.

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