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Sparsity-based image monitoring of crystal size distribution during crystallization

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To facilitate monitoring crystal size distribution (CSD) during a crystallization process by using an in-situ imaging system, a sparsity-based image analysis method is proposed for real-time implementation. To cope with image degradation arising from in-situ measurement subject to particle motion, solution turbulence, and uneven illumination background in the crystallizer, sparse representation of a real-time captured crystal image is developed based on using an in-situ image dictionary established in advance, such that the noise components in the captured image can be efficiently removed. Subsequently, the edges of a crystal shape in a captured image are determined in terms of the salience information defined from the denoised crystal images. These edges are used to derive a blur kernel for reconstruction of a denoised image. A non-blind deconvolution algorithm is given for the real-time reconstruction. Consequently, image segmentation can be easily performed for evaluation of CSD. The crystal image dictionary and blur kernels are timely updated in terms of the imaging conditions to improve the restoration efficiency. An experimental study on the cooling crystallization of α-type L-glutamic acid (LGA) is shown to demonstrate the effectiveness and merit of the proposed method.

1. Introduction

Monitoring the crystal size distribution (CSD) during a crystallization process is very important for control optimization to obtain the desired product quality and production efficiency [1]. With the rapid development of process analytical technology (PAT) in the past decade [2], a few real-time measurement methods have been explored for measuring CSD based on using the laser diffraction (LD), ultrasound attenuation (UA), and focused beam reflectance measurement (FBRM) technologies [3,4]. In particular, FBRM has been increasingly applied for on-line monitoring of crystal size distribution termed as cord length distribution [5,6], which is mainly effective for spherical particles. However, these technologies cannot offer two-dimensional details of crystal size or shape. By comparison, high-speed optical imaging devices have also been gradually adopted for crystal size measurement and shape identification in the recent years [2,7]. Real-time image analysis has therefore become intensively appealed for monitoring crystallization processes.

Based on using an invasive or non-invasive imaging system for monitoring a crystallization process, a small number of real-time image analysis methods were explored for measuring crystal size or CSD [2,8]. By using a non-invasive imaging system, Larsen et al. [9] presented a model-based object recognition algorithm to extract crystal size information for the α-glycine crystallization process from the captured images; A synthetic image analysis method was developed for in-situ crystal type identification and size measurement in the recent paper [10]. By comparison, an invasive imaging system named particle vision and measurement (PVM) was adopted to develop a comprehensive image analysis [11] on the crystal size of monosodium glutamate during crystallization. In addition, a flow-through cell imaging device was used to estimate CSD [12], based on crystal image segmentation using the wavelet transform and fuzzy C-means clustering strategy. The device was further extended to take in-situ crystal images from two perpendicular directions [13], such that a faster image analysis algorithm was proposed to classify particles and count the particle sizes. A multivariate image analysis method was combined with a classical image technique for in-situ estimation of CSD [14]. To address the problem of out-of-focus particles provoking degraded imaging for in-situ monitoring, Presles et al. [15] developed an optimization strategy into the image analysis to acquire better particle characterization. To effectively extract the moving particle information for on-line measurement of particle size, Chen et al.

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[16] proposed a fast image processing algorithm for correction of imaging illumination and binarization in a two-phase flow. To tackle the recognized challenge of estimating the crystal growth rate from real-time captured images, a few advanced image processing techniques were presented to estimate the crystal length distribution specifically for needle-shaped crystals [17]. Agimelen et al. [18] adopted the mean aspect ratio of all the particles in the captured image by PVM to reduce the computation effort for particle size estimation. However, little work had been devoted to improve the captured image quality against the influence from the solution turbulence and time-varying illumination background with respect to the crystallization progress, which should be envisaged for effective particle extraction and CSD estimation during the crystallization process.

To eliminate the influence from particle motion, solution turbulence, uneven illumination background, and imaging noise, it is necessary to restore the true crystal images from the captured images in a fast manner. A synthetic sparsity-based image analysis strategy is therefore proposed in this paper for real-time monitoring of CSD with high efficiency and accuracy. Firstly, the noise in a captured crystal image is filtered out by using an image dictionary established in advance. Then the edges of a crystal shape in the captured image are determined in terms of the salience information defined from the denoised crystal images. These edges are used to determine the blur kernel for reconstruction of a denoised image. To this end, image segmentation is proceeded for evaluation of CSD subject to uneven illumination background. Experimental results are shown to demonstrate the proposed image analysis method for in-situ measurement of CSD for an L-glutamic acid (LGA) crystallization process.

2. Image analysis

Analysis of real-time crystal image aims at obtaining the details of CSD. The whole process of image analysis strategy for real-time CSD measurement is shown in Fig. 1, including image preprocessing, image restoration, and image segmentation, which are presented in the following subsections, respectively.

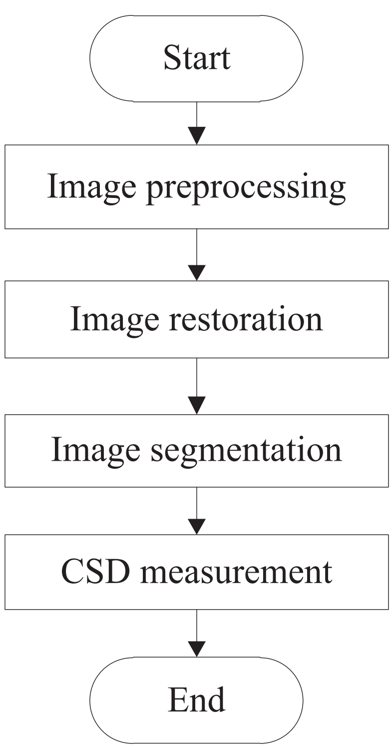

Fig. 1. The flow chart of image analysis.

2.1. Image preprocessing

In general, the size of a captured image depends on the resolution of an imaging system. Typically, when the size of a captured image becomes larger, a longer processing time is needed for on-line image analysis. To alleviate the time delay, an efficient method based on the wavelet transform [19] is adopted for downsizing the captured images while maintaining necessary information for real-time analysis. Given an original image $f(x, y)$ with the size $M \times N$, a two-dimensional discrete method is used for the wavelet transform based on the biorthogonal wavelet function [19]. Denote by m the row, by n the column, and by j the scale. The discrete wavelet transform for $f(x, y)$ is defined by

$$\begin{cases} A_\varphi(j, m, n) = \dfrac{1}{\sqrt{MN}} \sum_{x=1}^{M} \sum_{y=1}^{N} f(x, y)\varphi_{j,m,n}(x, y) \\ A_\psi^i(j, m, n) \\ \quad = \dfrac{1}{\sqrt{MN}} \sum_{x=1}^{M} \sum_{y=1}^{N} f(x, y)\psi_{j,m,n}^i(x, y), \qquad i = \{H, V, D\} \end{cases} \tag{1}$$

where

$$\begin{cases} \varphi_{j,m,n}(x, y) = 2^{j/2}\varphi(2^j x - m, 2^j y - m) \\ \psi_{j,m,n}^i(x, y) = 2^{j/2}\psi^i(2^j x - m, 2^j y - m), \\ \qquad i = \{H, V, D\} \end{cases} \tag{2}$$

where $\psi_{j,m,n}^i(x, y)$, $i = \{H, V, D\}$ are used to identify three directional edges including horizontal, vertical, and diagonal directions, respectively. Then the original image $f(x, y)$ is decomposed into four parts at the scale j: a low frequency component $A_\varphi(j, m, n)$ which is used for approximating $f(x, y)$, and three high frequency components $A_\psi^i(j, m, n)$ to be removed.

2.2. Image restoration

Generally, a crystallization process is involved with solution agitation. Although a high-speed camera can be used to reduce the capture time for imaging, solution turbulence interferes with real-time imaging, causing difficulty to discern the outlines of crystals. To deal with the problem, a restoration algorithm is proposed to remove noise and blurs from the captured images, in order to facilitate the subsequent image segmentation and CSD measurement.

The key idea of image restoration is to establish a degradation model to recover the crystal images from the captured images, and then use a mathematical method of solving the inverse problem to obtain the optimal approximation of the original image. Due to that the related parameters (additive noise and blur kernel) could not be known in advance, a blind restoration method is explored here, including degradation model construction, image filtering, dictionary learning, salient edge estimation, blur kernel estimation, image deconvolution, as detailed in the following subsections.

2.2.1. Degradation model construction

It is assumed that in a captured crystal image with a short time exposure, the blur kernel is space-invariant. That is, the degradation model is simplified as

$$Y = k*X + v \tag{3}$$