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Recognizing overlapped particles during a crystallization process from *in situ* video images for measuring their size distributions

Ola Suleiman Ahmad^a, Johan Debayle^a, Nesrine Gherras^a, Benoît Presles^a, Gilles Févotte^{a,b}
and Jean-Charles Pinoli^a

^aÉcole Nationale Supérieure des Mines de Saint-Etienne, LPMG, UMR CNRS 5148,
158 cours Fauriel, 42023 Saint-Etienne cedex 2, France.

^bUniversité Lyon 1, Campus de la Doua,
43 Boulevard du 11 Nov. 1918, 69622 Villeurbanne Cedex, France.

ABSTRACT

This paper presents a method to recognize polygonal-shaped particles (i.e. rectangles, regular/irregular prisms) among agglomerated crystals from *in situ* images during a crystallization process. The aim is to measure the particle size distributions (PSD), which are key measurements needed for the purification operations and the quality control of chemical products or drugs. The method is first based on detecting the geometric features of the particles identified by their salient corners. A clustering technique is then applied by grouping three correspondent salient corners belonging to the same polygon. The efficiency of the proposed method is tested on particles of Ammonium Oxalate during batch crystallization in pure water. Particle size distributions are calculated, and a quantitative comparison between automatic and manual sizing is performed.

Keywords: Crystallization, Salient Corner Detection, Grouping Method, Shape Recognition, Particle Size Distributions.

1. INTRODUCTION

Crystallization is a chemical engineering process which is used in time-continuous mode in order to produce solid products such as sodium, ammonium sulfate and potassium chlorides, and in batch mode to fabricate fine chemicals and drugs. It consists of two major events. The first event is the nucleation, where the solute molecules distributed in the solvent aggregate and increase the concentration of the solute, then stabilize under certain conditions to form the nuclei. The second event is the crystal growth, where the nuclei grow and reaches a critical size to finally form one or different crystal structures called polymorphisms. Controlling the particle shape and size distributions (PSD) is very important to precisely evaluate the quality of the final drugs and chemical products, and to the downstream processes.¹

The main challenge to achieve a successful and robust characterization of the PSD by image analysis is the segmentation process, which aims to separate the particles from the background and ones from each others. It becomes a critical problem in crystallization because of several reasons, such as: the perspective projection of the 3D crystal shape onto the image plane, the blurred appearance of unfocused particles, the heterogeneity and the anisotropy of the solute, the degree of agglomeration or overlap, and the random variation in size of the observed particles. Many methods have been proposed to detect the crystals for monitoring the particle size distribution. An algorithm² based on line segment detection³ was proposed to detect rectangular crystals by assuming that the rectangular object can be constructed from two or more line segments. In their preceding work,⁴ the algorithm is modified to be used to detect prismatic crystals basing on 3D-model fitting. In both applications, the results was satisfied in detecting accurately the overlapped particles from images but in low solids concentrations, where the particles are small enough with no high degree of overlap. However, the performance of the algorithm declines

Further author information:
Ola Suleiman Ahmad: E-mail: ahmad@emse.fr

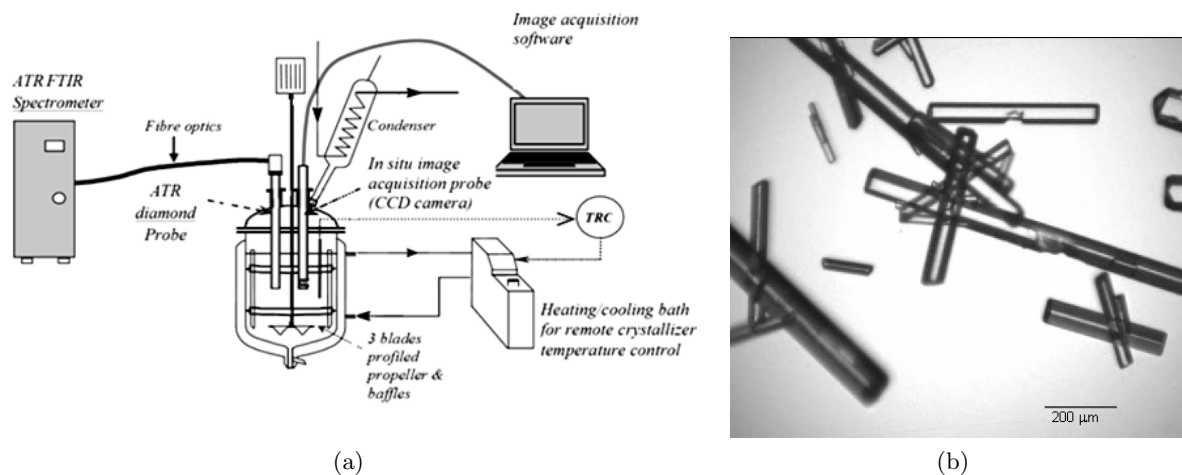


Figure 1. (a) Experimental setup of batch crystallization process. (b) A gray level image of Ammonium Oxalate crystals in pure water.

in high solids concentrations and with the existence of unfocused objects. A multi-scale segmentation method⁵ was proposed and found to be effective for extracting crystals from unclear background of varied illumination, but the overlapped particles are detected as one object which is unwanted for the present paper setting. In the recent work⁶ related to the last one,⁵ the concave points are extracted from the detected particles⁵ which are defined as markers for a marker-controlled watershed algorithm⁷ in order to separate the overlapped crystals. The method showed efficiency in separating the attached particles even at high solids concentrations, but it was not sufficient to detect the overlapped crystals of polygonal shapes. A restoration method⁸ was proposed for detecting unfocused crystals from a segmented image⁷ for achieving an accurate measurement of the particle size distribution. However, the method detects the overlapped crystals as individual ones.

The goal of this paper is to outline an applied efficient method, based on image analysis,^{9,10} that will be able to separate the overlapped particles of rectangular and regular/irregular shapes during the batch crystallization process at different solids concentrations. This paper is organized as follows. The experimental setup of the crystallization process is presented in section 2. The image segmentation method of overlapped particles is introduced in section 3. The application of this method for analyzing the particle shape and size distributions is described in section 4. Finally, a conclusion and perspectives are drawn in section 5.

2. EXPERIMENTAL SETUP

The experimental setup of the batch crystallization process of Ammonium Oxalate crystals is conducted in pure water.¹¹ Figure 1(a) illustrates the materials and the imaging system of the experiment. The reactor is a 2L, double-jacketed, glass vessel provided by a condenser. The mixer is a 3 blades, profiled, propeller, and baffles (Mixell TT) driven by a motor with a speed of 300 rpm which is sufficient to obtain a homogeneous suspension. The temperature of the crystallizer is controlled by a heating/cooling bath provided with an external probe of temperature $Pt100\Omega$ in a feedback control way.

The *In situ* imaging system consists of a probe (EZProbe sensor)^{8,12} equipped with a CCD monochrome camera with high resolution up to $4\mu m^2$ per square pixel, and 640×480 pixels per image. The video images are acquired by a frame grabber with a rate of 25 fps. Images of particles are formed using a transmitted light conducted within the probe by optical fibers from the light source. Figure 1(b) illustrates an *in situ* image example of Ammonium Oxalate crystals obtained during this experiment.

3. IMAGE SEGMENTATION METHOD

A method based on the geometric properties of the particle shapes has been developed to detect the overlapped rectangular and regular/irregular prismatic crystals by the assumption that each particle can be constructed from

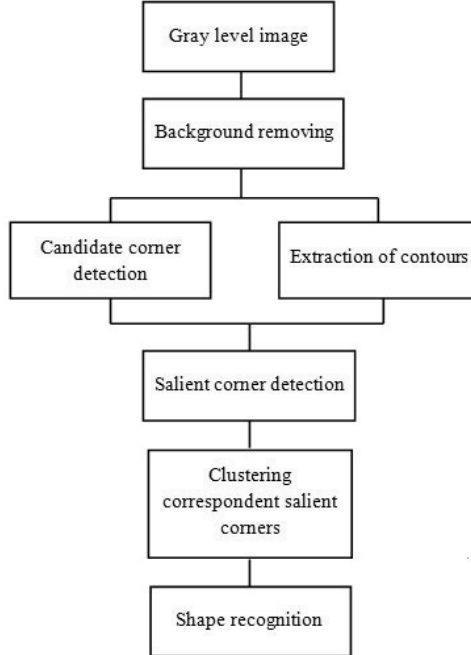


Figure 2. The diagram of the proposed method

grouping three of their correspondent salient corners that belong to the same particle. A computer simulation process has been presented in order to validate the method, and then experiments have been implemented on the real images.

The method (see Fig. 2) is divided into two main parts involving the detection of salient corners from the gray scale image, then recognizing the shape by clustering the correspondent salient corners that satisfy certain geometric conditions.

3.1. Salient Corner Detection

Many approaches have been developed for the corner detection because of its importance in computer vision, and pattern recognition applications. Some methods^{13–15} extract the corners directly from the gray level image. However, they are not sufficient for the present work since the orientation of salient corners is also required (see Eq. 1). Other methods^{16–18} extract the boundaries of the objects and then detect the corners from the planar curves or arc segments. Although, the performance of some of these methods depends on the robustness of extracting the arc segments from the gray level image.¹⁷

A salient corner C_i is defined, in this work, by the intersection of its two associated edges in the image spatial support D as follows:

$$C_i = \left\{ (x, y, \vec{u}_1, \vec{u}_2)_i : (x, y) \in D, \|\vec{u}_1\|^2 = 1, \|\vec{u}_2\|^2 = 1 \right\} \quad (1)$$

The salient corner detection method is based on extracting the candidate corners from the gray level image, and then on analyzing these corners in the edge image (see Fig. 2) to finally identify the salient corners. Firstly, a pre-processing on the gray level image is needed to increase the contrast and to remove the illumination heterogeneity of the background (see Fig. 3(c)), then the candidate corners are detected from the restored image by Rosten method¹⁵ as illustrated in Figure 3(d). Secondly, the binary edge image is obtained using a multi-scale edge detection,⁵ because it shows efficiency in extracting the boundaries of the semi-transparent overlapped particles whereas the strength of the edge is affected by the overlap (see Fig. 3(e)). Canny detector¹⁹ is used to get the edges from different scales, then a set of morphological operations²⁰ and edge cleaning are done to extract the final contour of the overlapped particles (see Fig. 3(f)). Finally, the direction of the salient corners is obtained

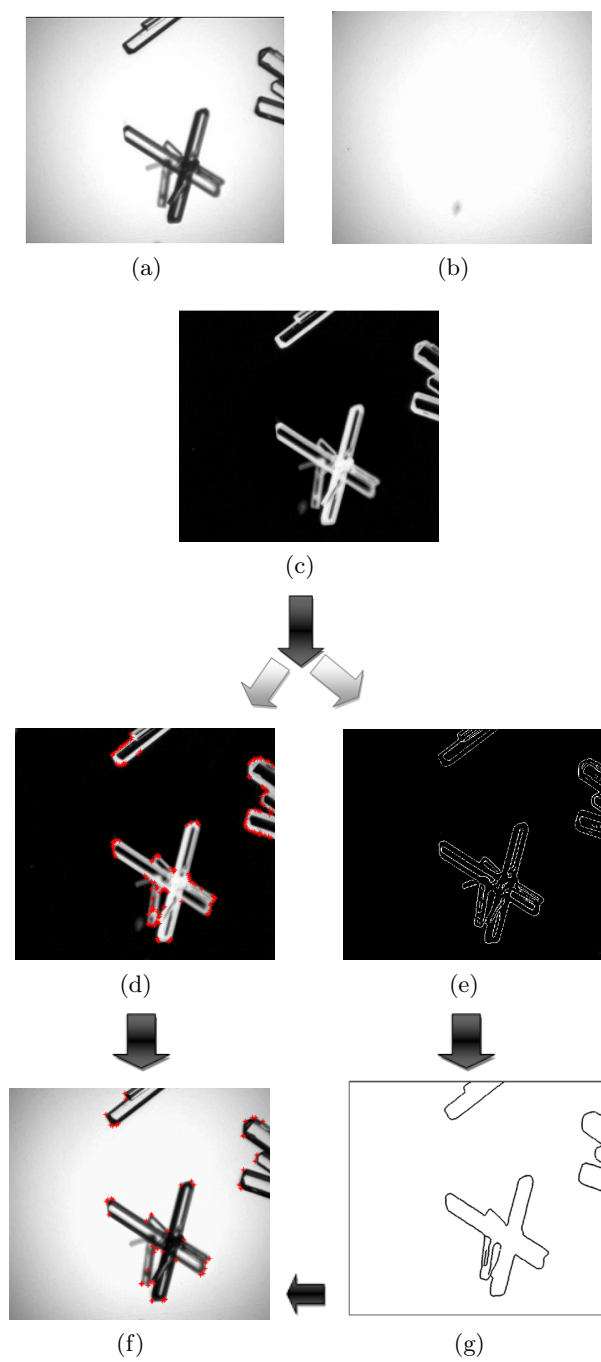


Figure 3. Example of the salient corner detection applied on a real image of Ammonium Oxalate crystals. (a) Original gray level image. (b) Background image. (c) Removing the background from image (a). (d) Candidate corners image. (e) Multi-scale edge detection. (f) Edge image after contour completion and edge cleaning steps. (g) Salient corners image.

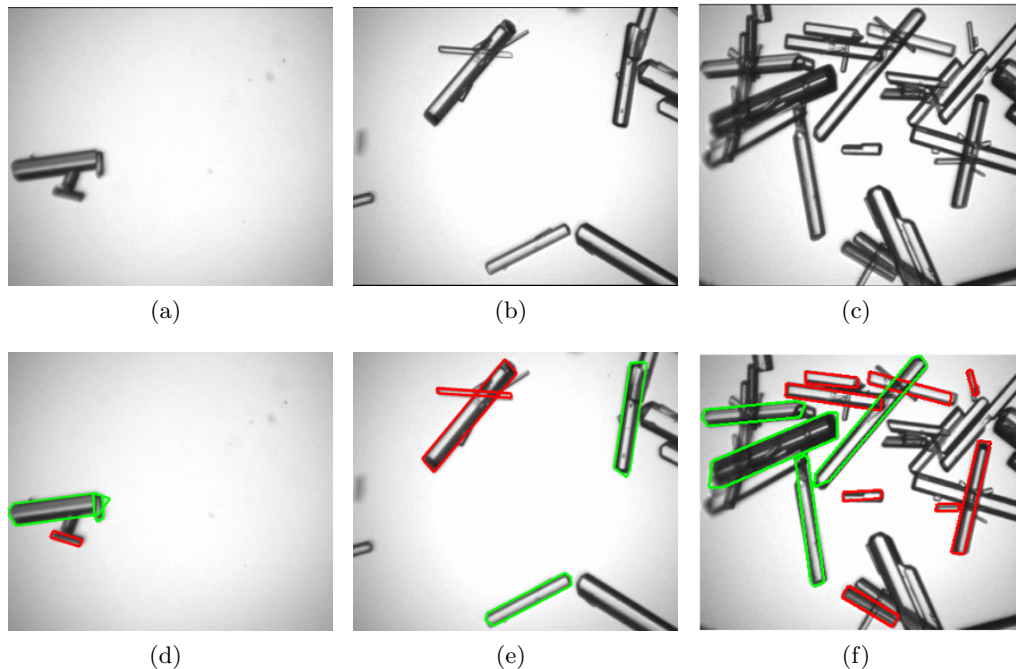


Figure 4. Examples of real images of overlapped Ammonium Oxalate crystals in low, intermediate and high solid concentrations respectively. (a)-(c). Original images. (d)-(f). Recognized particles

from the directional correlation function, denoted E , which is defined in Equation 2:

$$\forall \theta \in [0, 5^\circ, 10^\circ, \dots, 360^\circ[: E(\theta) = \sum_{x=1}^M \sum_{y=1}^N W_{C_i}(x, y) \times T_\theta(x, y) \quad (2)$$

where W_{C_i} is the binary edge image restricted to a window of size $M \times N$ pixels centered at corner C_i , and T_θ is a line template oriented at θ° .

The correlation function of the salient corners has two major peaks that refer to its associated edges. Other corners such as junctions will have more than two dominant peaks, while the correlation values at the isolated points will be near zero with no significative peaks. Consequently, all corners which have less or more than two dominant peaks are removed. So an image of salient corners is obtained as illustrated in Fig. 3(g)

3.2. Shape Recognition from clustering method

Following the salient corner detection, a clustering technique is performed for extracting the final shape of the particles. The procedure depends on searching three correspondent corners that satisfy geometric conditions and thus belong to the same particle.

The clustering technique is developed for recognizing rectangular and regular/irregular prismatic overlapped particles, whereas the general shape of the Ammonium Oxalate crystals is shown to be like rectangular or prismatic shape with faces of different growth rate under the experimental conditions.²¹

The method is evaluated on 150 images acquired in low, intermediate and solids concentrations, successively (see Fig. 4) and a quantitative comparison with the visual recognition by a human expert is illustrated in Table 1. The results show that the method is efficient in detecting the particles in low, intermediate, and even high solids concentrations. Indeed, more than 75% of the particles could be sightly recognized, which is sufficient for monitoring the size distributions.

Table 1 Quantitative comparison between automatic and visual recognition for 150 images of overlapped Ammonium Oxalate particles:

Particles (numbers)	Low overlap	Intermediate overlap	High overlap
Manually recognized (N_V)	130	149	374
Automatically recognized (N_A)	125	137	327
Right N_R	125	122	281
Missed $N_M = N_V - N_A$	5	12	47
False $N_F = N_A - N_R$	0	15	46
Right to automatic recognition ratio in percentage (N_R/N_A)%	100%	89.1%	85.9%
Missed to visual recognition ratio in percentage (N_M/N_V)%	3.9%	8.1%	12.6%
False to automatic recognition ratio in percentage (N_F/N_A)%	0%	10.9%	14.1%
Right to visual recognition ratio in percentage (N_R/N_V)%	96.2%	81.9%	75.2%

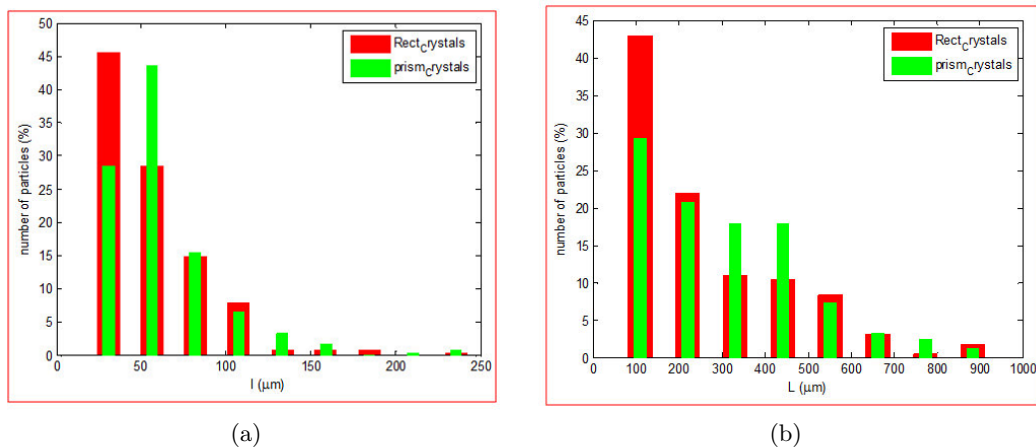


Figure 5. Particle length and width distributions. (a) Particle width distribution of prismatic and rectangular crystals. (b). Particle length distribution of prismatic and rectangular crystals.

4. PARTICLE SIZE DISTRIBUTIONS

Following the segmentation process, the particle size distributions are illustrated for 3 sets of video sequence images, each set contains 50 images, which are acquired in low, intermediate and high solids concentrations. The length and width of each detected crystal is calculated for all image sets and the size distributions are evaluated²² for illustrating the frequency of each length and width parameters of the crystals during the experiment (see Fig. 5). The results show that the particles have a tendency to exhibit a prismatic shape in high agglomeration, whereas the small particles exhibit a rectangular shape. Small particles refer to the nucleation process and the beginning of the experiment. Thus, the Ammonium Oxalate crystals have a rectangular shape in low concentrations, then the faces grow to take a prismatic shape in high concentrations. This analysis is important for characterizing the flow properties, dissolution rate, agglomeration tendencies, mixing and demixing properties, filterability, etc.²³

A quantitative comparison between automatic and manual sizing is performed (see Fig. 6) using the cumulative distribution function (CDF), which is independent of the bins width or number contrary to the particle size distribution function. The normalized error is calculated for both length and width cumulative distributions:

$$Err = \sum \frac{|Cumh_M - Cumh_A|}{Cumh_M} \quad (3)$$

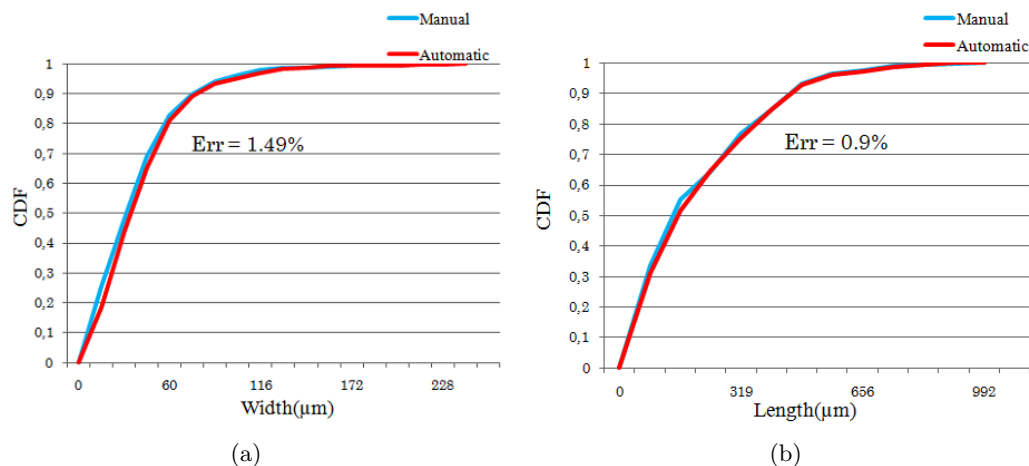


Figure 6. Comparison of cumulative distribution functions (CDF) of particles length and width for 150 in-situ images. (a) Comparison of particles width distributions. (b). Comparison of particles length distributions.

where $Cumh_M, Cumh_A$ are the cumulative distributions of manual and automatic sizing, respectively.

The results are satisfying and show that the proposed automatic recognition method could be used for analyzing the particle size distributions of the overlapped particles.

5. CONCLUSION AND PERSPECTIVES

A recognition method of overlapped particles based on salient corner detection and geometric grouping has been described and applied. The method is proposed to recognize rectangular, regular and irregular prismatic shapes. Then, the particle size distributions are measured on a set of video sequence images in different solid concentrations. A quantitative comparison between the present method and the visual recognition by a human expert is performed. The results show that the method can recognize generally more than 96%, 81% and 75% of the particles recognized by the human vision in low, intermediate and high solids concentrations, respectively. The cumulative distribution functions of particles width and length are calculated for both manual and automatic sizing. The maximum normalized difference between the cumulative distribution functions was 1.5%. The method can be generalized to detect parallelograms or other convex quadrilateral forms. It could be modified for recognizing other geometric polygons by clustering their correspondent salient corners according to the geometric properties of the desired shape. In the difficult cases of overlap, when the image contains a large number of semi-transparent overlapped particles, it becomes difficult to recognize accurately the particles, which might decrease the recognition percentage. So, our future work is to ameliorate the method by estimating the shape using the likelihood function and the clustering propability of two or three salient corners that belong to the same polygonal shape.

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