MONITORING OF CRYSTALLIZATION PROCESSES: A NOVEL APPROACH FOR THE SEPARATION OF TOUCHING EDGES IN CRYSTAL PARTICLE IMAGES

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Abstract

A method for separating the touching objects in the images of particulate systems is developed based on clustering in the feature space of the image. The approach adopted in this paper combines clustering in the feature space of the image to extract the characteristics of touching particle regions and the extraction of geometrical features. This combined method gives a superior performance compared to traditional watershed segmentation techniques.

Introduction

Characterization of particulate systems such as crystallization is done invariably in terms of particle size and shape distribution. However many techniques of direct measurement are not suitable for in-situ/online characterization purposes. Online measurement of particle size distribution (PSD) and shape parameters can be of utmost importance in the monitoring and control of crystallization and post-crystallization processes in the pharmaceutical and fine chemicals industries amongst others. Direct real-time visual characterization is a promising alternative to complex and expensive particle measurement systems. This characterization, which is typically executed via imaging and image analysis methods, still needs research and development particularly in overcoming the problem of adjacent touching particles.

There always exists particles touching (and overlapping) in images taken from particulate systems where no particle dispersion is applied prior to image capture. Even where particle dispersion is used to avoid the situation of particles touching, one still expects to observe images exhibiting this behavior simply due to the fact that dispersed particles will still flow together or ecliptically, and so in a dispersed sample, the probability of touching or overlapping events is reduced but not eliminated. Further, there are processes that are not amenable to dispersion, those of fragile nature like flocs and certain crystals. Moreover, it is noted that dispersion in many cases implies the use of solid particle samples. This represents a disadvantage to the particle characterization in processes like crystallization where the monitoring in real-time of crystal particle characteristics like size and shape is essential. There is thus a significant role imaging and image analysis can play in monitoring particulate systems, yet there is some way to go towards having robust real-time image-based particle characterization. A key component of a robust characterization technique is image analysis algorithms that can tackle issues related to touching particles. Such a development is the concern of the present paper.

A touching issue

The region where the particles touch will have a lesser intensity than the particle body in the captured image. However, it is not low enough to be taken as background. This means that when we binarize the image (reduce it to black-white binary format), these regions are taken in as object regions rather than background. Consequently and as far as any imaging algorithm is concerned, the two touching particles will appear as a single object in the image. In the case where a single particle touches many others, the result is one single large object in the binary image rather than many individual particles. This is detrimental to the particle size determination since the conglomerate objects will be treated as one single particle eventually generating erroneous size distributions.

Therefore to extract accurate size and shape features of particles from directly acquired images, separation of touching particles must first take place. There have been attempts, reported in the literature, to separate touching particles [1-3] but these methods employ assumptions about the shape of the imaged particles, meaning that particle shape properties are needed a priori. For instance Song and Yamamoto use the assumption that the particle is spherical simplifying their analysis and subsequently their size determination [2]. Such an assumption does not hold for most crystallization systems where particles deviate significantly from symmetrical features like sphericity or where there is need to learn about the specific particle shape features such as when analyzing crystal growth phenomena.

In this work we apply a novel image analysis approach based on clustering in multidimensional feature space to extract the characteristics of touching particle regions. A set of features that differentiate touching regions are chosen and clustered. In this multidimensional feature space, clustering is found to distinctly distinguish touching regions.

Experimental

An experimental imaging setup is used to test this methodology. We first use a polymer particle standard (PVC) of certified size distribution (Mettler Toledo USA). The PVC particles are circulated using a pump (Watson Marlow UK) through a flow cell. Images are acquired using a digital video camera (Basler A620) coupled with an Optical Zoom system (Thales Optem zoom125C). The camera is connected to the computer where data acquisition and processing is executed. The image analysis algorithm is implemented in MATLAB (Mathworks, USA) with the aid from Matlab's Image Processing Toolbox.

Methods and analyses

The approach adopted in this paper is based on clustering in the feature space of the image to extract the characteristics of touching particle regions. A set of features that differentiate touching regions are chosen and clustered. In a region where particles' edges remain unidentified within the multidimensional feature space, we ensue, after [5], the extraction of geometrical features in the binary image to determine whether these edges represent touching or rather single particles. This step ensures that no more touching particles exist in the image. Finally morphological operations are done to remove spurs and holes. This combined method gives a superior performance compared to watershed segmentation techniques previously employed [4]. A flow chart showing illustrating the sequence of steps of our approach is given in Figure 1.

We next discuss some details of these steps including preprocessing, binarizng, processing to separate the touching regions and then show the improvement this approach by comparing results for size distributions determined from images with and without separation and by a comparison against traditional watershed segmentation technique.

Preprocessing

The acquired images bring with them much external undesired noise. Median filtering is done on the image with a kernel size of [3x3] to remove random noise. This filter is an order statistic filter which replaces the value of a pixel by the median of the pixel values in a small neighborhood [6]:

$$f(x,y) = \text{median}\{g(u,v)\} \qquad (1)$$

where $(u, v) \in S(x,y)$ the neighborhood around (x,y). The median filter has the advantage that it does not blur the image compared to smoothening filters. Figure 2a shows the original image with median filtering.

Thresholding

The subsequent step is making the image binary (black and white). The quantitative estimation of various image features are obtained from the binary image which will have the object areas as white and background as black (1 and 0 respectively). The conversion from the gray scale image to binary image is essentially a classification problem. When this is done based only on the pixel intensity, it is normally known as thresholding. The grayscale image (I_g) and binarized image (I_{bw}) can be represented by equations 2 and 3 respectively.

$$I_g = \{x \mid 0 \leq x \leq U\} \qquad (2)$$

$$I_{bw} = \{x \mid x \in (0,1)\} \qquad (3)$$

Where U is the upper limit of the intensity which has a value of 255 for a grayscale image. Thresholding of the enhanced image is done using Otsu's method [7]. In this method the pixels are divided into two classes such that the mean of each class is as separated as possible and the variance within each class is as least as possible. The binarized image is shown in Figure 2b. It can be seen that most of the touching regions are accounted for as belonging to the object regions (white) resulting in a single large particle in the center of the image. The separation of these particles is necessary representative particle size estimation.

Separation of touching particles

The original image (Figure 2a) in its grayscale form contains useful information. We exploit this information specifically the fact that touching regions in the grey-scale image exhibit more rapid variation in intensity than the object body. This indicates a probable high value for the gradients in these regions. The gradient is due to less light being captured from the valley regions between touching particles. A typical feature which is used to trap regions where difference in intensity is prominent is the range operator. The intensity range is the difference between maximum and minimum values of intensity in a small neighborhood around each pixel. However these features are not only strong enough to completely capture the touching regions but also have random high values in the background and object areas.

Another novel feature was employed here to capture the touching regions more decisively. Instead of directly calculating intensity ranges in the image, two fuzzy clusters are defined based on intensity alone (Figures 2c and 2d). The difference between total membership values for each cluster in a small neighborhood around each pixel is then calculated. This can be considered as a special kind of aggregate range (R'):

$$R' = \sum(X_i) - \sum(Y_i) \quad i = 1,\ldots,n \qquad (4)$$

where X and Y are the two clusters and n is the number of pixels in the neighborhood.

The value of R' approaches n in object and background areas while it approaches zero in the boundary and touching regions. Since some of the absolute values of R' are near zero, we take the log transformation of R' values and represent this data as an image shown in Figure 2e. The next step is to cluster the range values to obtain two distinct clusters as shown in Figures 2f and 2g. This result shows two images with distinctly high and low values for touching boundary regions, respectively. We use the lower value cluster (Figure 2g in its binary format) for the next step of superimposition on the previously binary image (Figure 2b) obtained via thresholding. Finally, morphological operations like filling holes are executed to enhance the result before size counting is done.

Results and Discussion

Most of the touching particles are separated by this process as shown in Figure 2h. There are still touching edges left in the image after this procedure. A geometric feature of the initial binary image and the separated image is used to locate the touching regions present. This is based on the fact that wherever particles are touching, the boundary curves of the particles form a wedge shape [5]. Moreover, these wedges occur in pairs in opposite orientation. This "wedgness" of the boundary can be quantified by parameters called center of gravity (COG) and eccentricity of a boundary pixel. Center of gravity is the average relative coordinates of boundary pixels in a small neighborhood around the particular boundary pixel. Eccentricity is the Euclidean distance from the boundary pixel to its COG. These quantities are illustrated in Figure 3. If this eccentricity is above a certain threshold for a boundary pixel, we can consider it as lying in a high wedge region. For each wedge region, one representative point is used along with its orientations. Then matching points on opposite wedges are paired. Each pair counts as a possible touching region in the image. For the image shown in Figure 2b with no separation, we had observed eight such pairs while after separation as shown in Figure 2h, these were reduced to two (circled).

The particle size distributions were next quantified from the images using standard image counting techniques. The comparison of PSD of the objects in the image before and after separation is given in Figures 4 and 5. The effect of touching on the PSD is clearly evident in this comparison where the PSD of the image with no separation is biased towards higher sizes as expected due to the agglomeration of image object.

Further our results are compared against traditional watershed segmentation technique results. Figure 6 shows the separation under watershed segmentation while Figure 7 shows the corresponding calculated PSD. It can be seen that watershed method suffers from over segmentation and hence a large number of small particles. Our method shows superior and far less error in separation compared to traditional watershed segmentation technique.

Conclusions and future work

A method for separating the touching objects in the images of particulate systems was presented. The method successfully used clustering in the feature space along with geometrical features of the image to arrive at a separation of particles resulting in enhanced PSD quantification. This combined method gave better results than traditional watershed segmentation techniques.

It should be pointed out our approach presented here for particle separation carries a limitation and a side effect. The limitation, as mentioned above, is associated with the incomplete separation (two particle edges were left without separation). Improvement here is undergoing. The side effect of our approach is associated with the imperfect superimposition which is observed to erode particle areas along the periphery and thus results in under-sizing. Improvement in separation method to reduce this over erosion is under investigation.

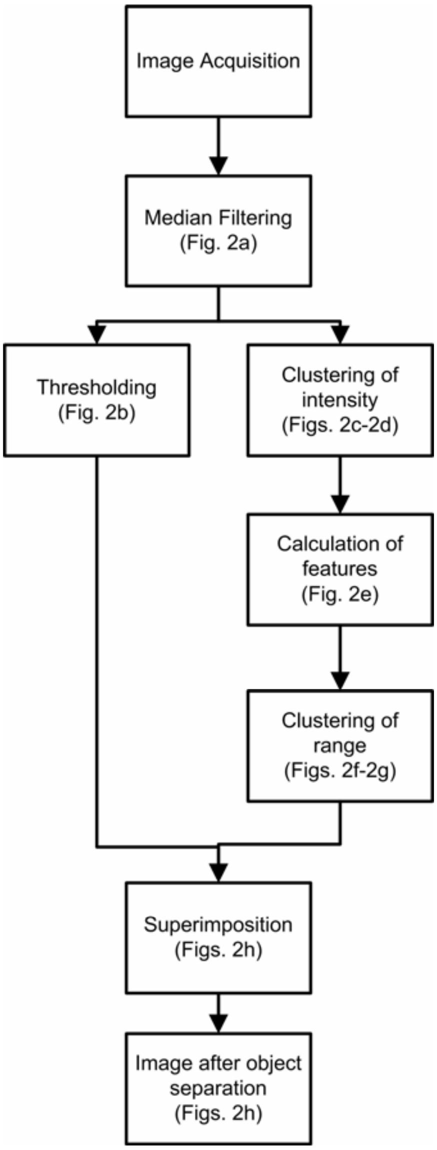

Fig. 1. Sequence of steps in the image object separation analysis.

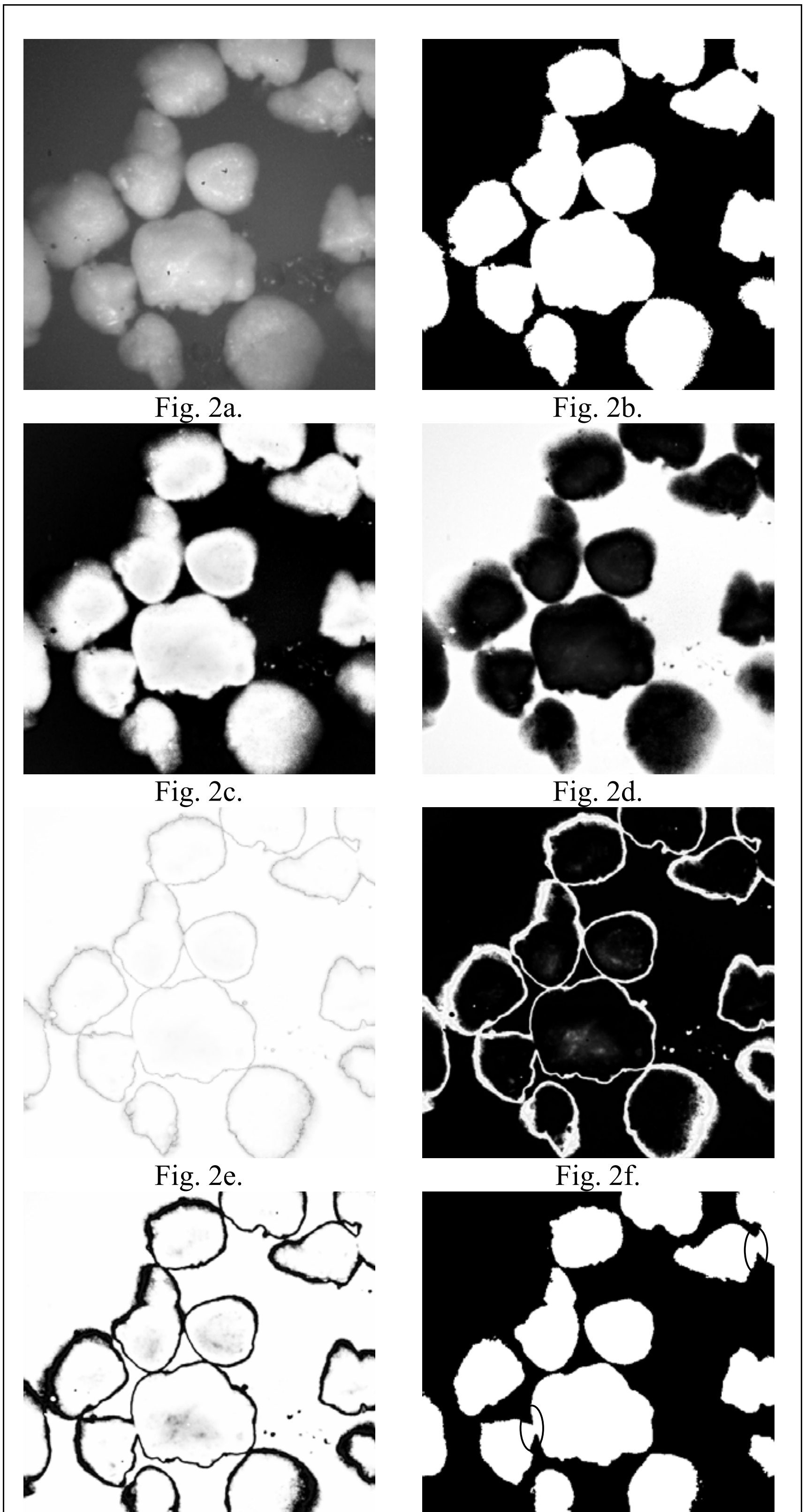

Fig. 2a. Fig. 2b.

Fig. 2c. Fig. 2d.

Fig. 2e. Fig. 2f.

Fig. 2g. Fig. 2h.

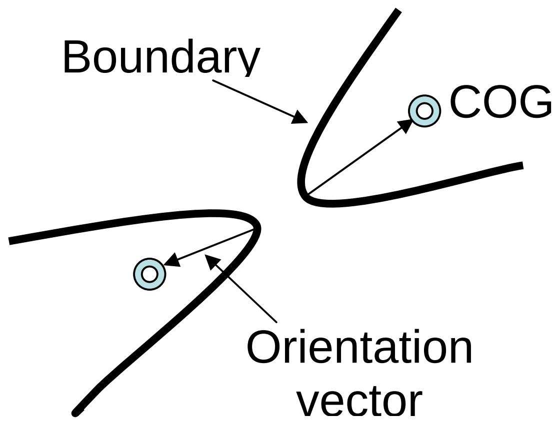

Fig. 3. Center of Gravity (COG) and Eccentricity (Magnitude of Orientation Vector).

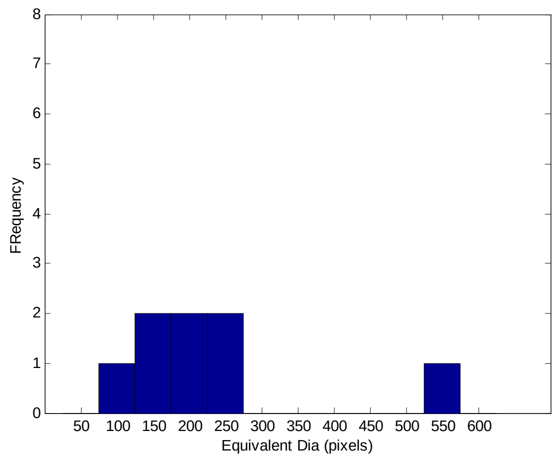

Fig. 4. PSD of image before separation.

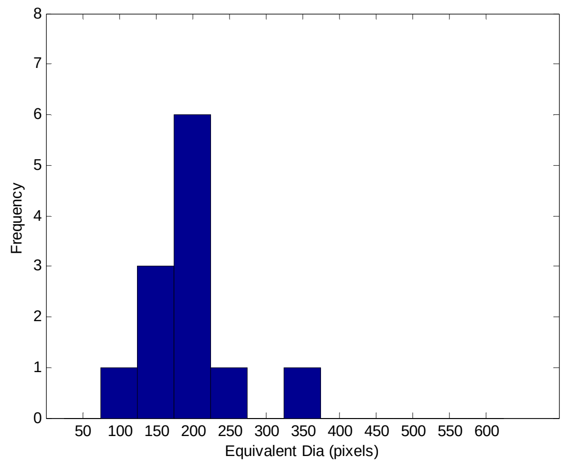

Fig. 5. PSD of separated image.

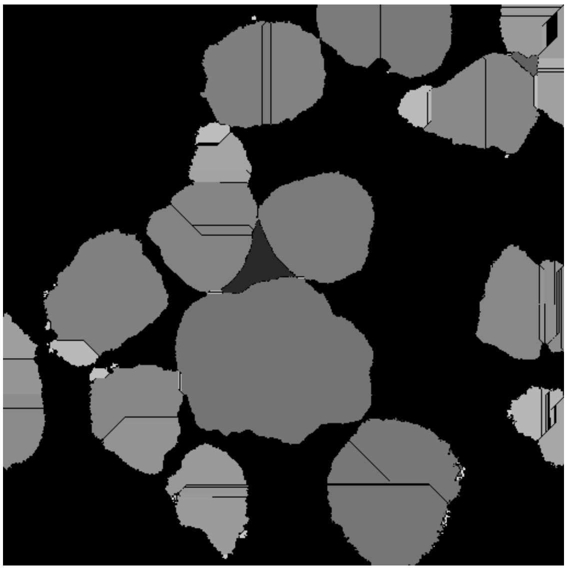

Fig. 6. Result of Watershed segmentation.

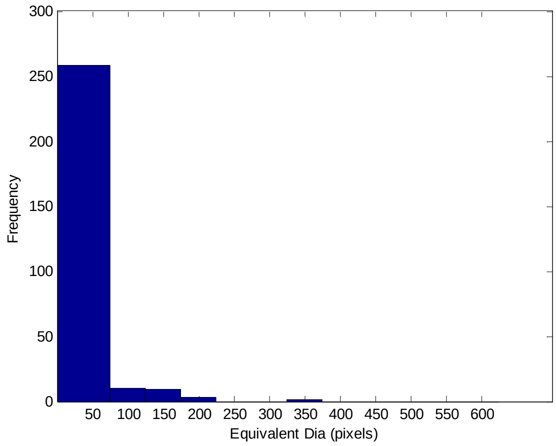

Fig. 7. PSD from Watershed segmentation.

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