CHEMICAL ENGINEERING TRANSACTIONS
VOL. 32, 2013
A publication of AIDIC
The Italian Association of Chemical Engineering
Online at: www.aidic.it/cet
Chief Editors: Sauro Pierucci, Jiří J. Klemeš

ISBN 978-88-95608-23-5; **ISSN** 1974-9791
DOI: 10.3303/CET1332368

Image-Based Multi-Resolution-ANN Approach for On-line Particle Size Characterization

Bing Zhang[a], Robert. Willis[a], Josè A. Romagnoli*[a], Chiara Fois[b], Stefania Tronci[b], Roberto Baratti[b]

[a] Department of Chemical Engineering, Louisiana State University, LA, USA
[b] Dipartimento di Ingegneria Chimica e Materiali Università di Cagliari, CA, Italy
jose@lsu.edu

An image-based multi-resolution sensor for online prediction of crystal size distribution (CSD) is proposed. The mean and standard deviation (std) of lognormal probability density function as the CSD can be predicted through the on-line sensor. Texture analysis, through wavelet-texture algorithm, as characteristic parameters to follow the crystal growth is utilized. Following nonlinear mappings consisting of artificial neural networks (ANNs) is incorporated using as inputs the texture information in conjunction with the available on-line process conditions. The output data for training the ANN models are measured manually at different sampling times as well as in a range of operating conditions. Validations against experimental data are presented for the NaCl-water-ethanol anti-solvent crystallization system.

1. Introduction

Crystallization is a critical part of processing in fine chemicals and pharmaceutical industries. During manufacturing, transition substances and/or end-products can exist in crystal particulate form. Crystal size, as an important external crystal structure, can largely influence the textural and physical properties of the final commercial products. A narrow CSD with desired mean size is preferred by industries. Monitoring the CSD is vital during this process for the purpose of achieving a target end-product size. Image analysis has been a very promising method for direct measurements of size, size distribution and shape aided by recent progress in high speed imaging devices and equally powerful computers at reasonable costs and the adaptability to real-time application.

The key issue of applying digital images for process monitoring and controlling is extracting relevant information correctly from disturbances in real life situation. Real-life crystal images can have agglomerating or touching and overlapping problems. Physical dispersion can be used to avoid these phenomena prior to capturing the images. This operation, however, can reduce but cannot eliminate agglomeration and overlapping events because dispersed crystals will flow together, especially in high solid concentration conditions. Zhang (2011) proposed a multi-resolution fuzzy clustering approach to separate touching and overlapping regions. This method can successfully segment when this problem is limited. This paper deals with severe touching and overlapping problems by performing texture analysis on crystal clusters aiming at accurate crystal image processing and subsequently correct control action.

In this paper, an image-based multi-resolution sensor for online prediction of CSD is proposed. The mean and standard deviation (std) of lognormal probability density function as the CSD can be predicted through the on-line sensor. A novel approach is proposed for crystal image texture analysis based on wavelet-fractal-energy algorithms. Fractal dimension (FD) as the texture analysis parameter provides a non-integer value to describe crystal growth from crystal images from the point of view of surface roughness. Furthermore, energy signatures, obtained from the wavelet coefficients, FD as well as process conditions are utilized in the approach to build ANN models towards the estimation and prediction of the mean crystal size and std from images. A series of experiments using NaCl-water-ethanol anti-solvent crystallization system were carried out to illustrate the approach.

Please cite this article as: Zhang B., Willis R., Romagnoli J., Fois C.A.M., Tronci S., Baratti R., 2013, Image-based multi-resolution-ann approach for on-line particle size characterization, Chemical Engineering Transactions, 32, 2203-2208 DOI: 10.3303/CET1332368

2. Methodology for the Image-based multi-resolution Sensor

The overall architecture of online crystal size distribution prediction by Image-Based Multi Resolution Sensor is given in Figure 1. The crystallization can be carried out in a reactor with suitable volume and designs that allow temperature detection, anti-solvent addition and crystal suspension circulation. To maintain desired crystallization manufacturing parameters, temperature and anti-solvent addition speed in this case, control instruments such as feed pump or equipment with control function are welcomed. Crystal suspension circulates through a cell at which the crystal images are taken by a camera connecting to a microscope and a computer. The images will be stored in a database in the computer and gone through image analysis. The essential and sequence of steps for implementing the proposed methodology for CSD prediction from crystal images are shown in rectangular boxes in Figure 1. The sensor needs a prerequisite ANN which can be built at several crystallization conditions with corresponding images. An input image is treated as a 2D array of pixel intensities. A thresholding algorithm is applied for extracting features of interest which are the crystal clusters in this work. This is accomplished through a series of three sub-steps: a) detection of crystal edges by a threshold value differentiating them from the background, b) detection of the locations of the crystal clusters with the help of x-y coordinates on the binary image of crystal clusters, and c) determining and extracting the intensity values belonging to the crystal clusters. The information data of the clusters is then restored into a vector, being processed to generate the texture features by means of wavelet-fractal-energy algorithm. In this regard, it is first decomposed by wavelet transformation at several levels into details and an approximation. The detail from lower decomposition level and the approximation are considered as the high and low frequency noise to be removed. The remained details are then used for finding their variance and 2-based logarithm of the variance. The 2-based logarithm of the variance at each scale and decomposition scales are then used to calculate the Hurst exponent and consequently the FD. The texture features as well as crystallization conditions (flow and temperature) are then used as inputs in an ANN for the prediction of crystal mean size or standard deviation. In the following all the components of the methodology are briefly explained.

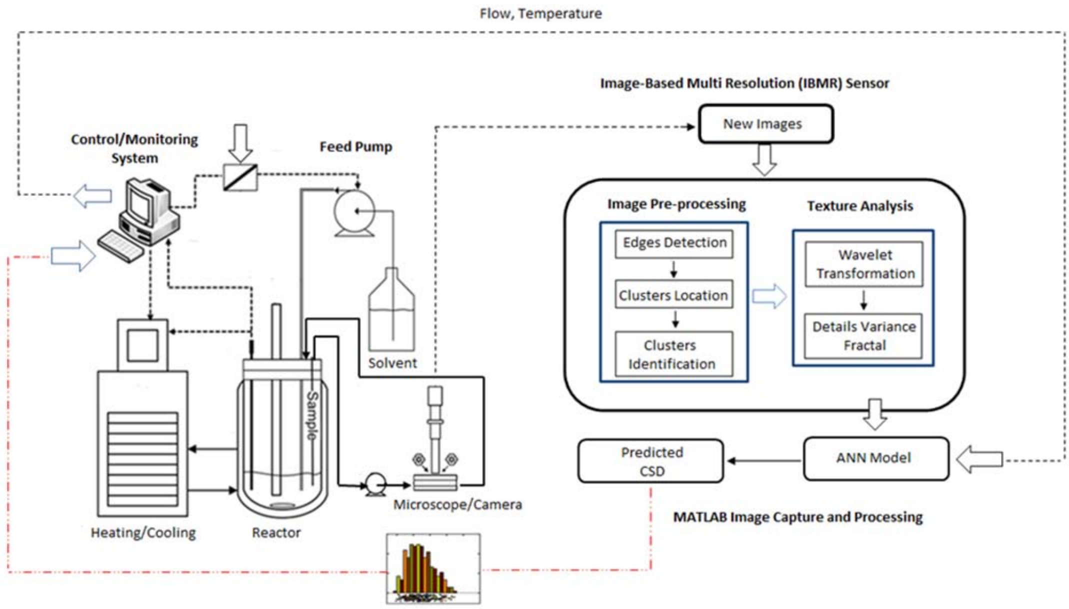

Figure 1: Overall architecture of CSD prediction

3. Texture analysis by wavelet transformation

Wavelet transform, a multi-resolution technique, can extract textural characteristics from crystal images. It can decompose a signal into several details and an approximation. Details carry the characteristic information such as edges of distinct objects distinguishing its corresponding signal from others. The approximation, on the other hand, usually reflects the intensity variance generated by lighting or illumination. Thus extracting features from details is more appropriate than from the approximation. As the number of crystal edges is the distinction between small and large crystals in images, texture features can be obtained from the point of energy distribution.

3.1 Wavelet transformation

Discrete wavelet transformation (DWT) is an efficient and accurate way used in this context since an image can be considered as discrete data. DWT calculates at selected scale a and location b parameter (Addison 2002). They are discretized in such a manner that a and b are linked. The scale a is generally discretized in a logarithmic way a_0^m where m is an integer. Each location b can be reached in discrete steps n (an integer) from an origin. It is also proportional to the scale a_0^m. Thus it can be represented as $nb_0a_0^m$. In wavelet transformation, a wavelike function called wavelet $\psi_{m,n}$ is used to transform a signal or function $f(x)$ in space or time into another form by convolution. Discrete wavelet transformation $T_{m,n}$ is known as the detail coefficients or high-frequency components. The approximation coefficients $S_{m,n}$, like the detail coefficients generated by wavelet functions, are given by convoluting with the function $f(x)$ and another set of functions called scaling functions $\phi_{m,n}(x)$. Taking multi-resolution into consideration, we can write the function $f(x)$ as a sum of approximation of the function at arbitrary scale index m_0 and detail function from scale m_0 to $-\infty$, using the approximation coefficients and detail coefficients:

$$f(x) = \sum_{n=-\infty}^{+\infty} S_{m_0,n}\phi_{m_0,n}(x) + \sum_{m=-\infty}^{m_0}\sum_{n=-\infty}^{+\infty} T_{m,n}\psi_{m,n}(x) \tag{1}$$

3.2 Wavelet energy signature

After wavelet decomposition, the coefficient variance at scale m is calculated using the following equation:

$$\langle T_{m,n}^2\rangle_m = \frac{\sum_{n=0}^{2^{M-m}-1}\left(T_{m,n}\right)^2}{2^{M-m}} \tag{2}$$

According to this expression, wavelet coefficient variance can be explained as the average energy wrapped up per coefficient at each scale. In this context, the detail variance is used as the wavelet energy signature.

3.3 Fractal-Wavelet Feature

Fractal dimension (FD) is a statistical real number that measures how complicated a fractal is. Fractals are objects that possess self-similar property, where fractals appear identical at different scales or numerical or statistical measures of fractals are consistent across scales. The reason why FD is selected as the texture feature is that the dynamics of crystal growth follows a fractal process and crystals in images exhibit statistical self-similarity in reality. To calculate FD, we here use fractal-wavelet method which requires not only wavelet decomposition but also fractional Brownian motion (fBm) function in our work. The FD of an fBm function can be calculated through the relationship equation:

$$FD = D_E + 1 - H \tag{3}$$

Here D_E is the Euclidean dimension. The Hurst exponent, H, has a value between 0 and 1 characterizing fBm. It is an indicator to reflect the smoothness of the fBm function: the higher the Hurst exponent is, the smoother the fBm function is. For an fBm, H also scales to wavelet power spectra $P_w(f_m)$ and frequency f_m. Meanwhile, the wavelet power spectra is related to the variance of discrete wavelet coefficients, and the frequency is inversely proportional to the wavelet scale a_m. The logarithmic plot of variance of discrete wavelet coefficients provides a method of deciding Hurst exponent from the slope of the plot.

$$log_2\left(\langle T_{m,n}^2\rangle_m\right) = (2H+1)m + constant \tag{4}$$

Where, the constant depends both on the wavelet function and the Hurst exponent.

4. Artificial neural network

Artificial Neural Networks (ANNs) have been explored as mathematical models to process information. ANNs have a strong ability in capturing complex input/output relationships. The network is composed by a number of nodes or units connected to the inputs and to the outputs. The signals pass throw the connections and are scaled using appropriate weights, obtained through a learning procedure aimed to obtain good generalization capability of the network.

In our work, three-layer feed-forward neural networks (FNNs) are utilized as a nonlinear mapping to predict the particle characteristics from the texture analysis and operating conditions. An input layer $\boldsymbol{x}$ consisting of sensory neurons, a hidden layer of computational neurons and an output layer $\boldsymbol{y}$ of target neurons. The hidden layer $\boldsymbol{a}$ is connected with the input layer through a Tan-Sigmoid transfer function with weights $\boldsymbol{W}1$ and biases $\boldsymbol{b}1$, as:

$$\boldsymbol{a} = f(\boldsymbol{W}1\boldsymbol{x} + \boldsymbol{b}1) \tag{5}$$

The output layer has the similar connection to the hidden layer with a linear transfer function:

$$\boldsymbol{y} = \varphi(\boldsymbol{W}2\boldsymbol{a} + \boldsymbol{b}2) \tag{6}$$

FNN is a supervised neural model, which has been developed using Matlab Neural Network Toolbox. The network parameter estimation, generally referred to as neural network training, has been developed by means of the Levenberg-Marquardt optimization algorithm, using the mean square error (MSE) between the network outputs and the targets as objective function. Training the net can be started by first randomly initializing the weights.

5. Case study

5.1 Experiment

A set of anti-solvent crystallization experiments at both constant and changed conditions was carried out with flow rate and temperature as the operating variables. For constant conditions, three different values were chosen: 0.7 ml/min, 1.5 ml/min, 3 ml/min as flowrate and 10°C, 20°C, 30°C as temperature. For validation purposes and additional run for a typical temperature and flowrate profile was performed. At the start up conditions, the crystallizer was filled with the saline solution composed of 100 g of water and 34 g of NaCl. The solvent solution was made up of 95% of ethanol and was added by the time to the initial solution using a peristaltic pump. As for the crystals growth examination, this had been performed choosing to work in an online fashion by using a peristaltic pump to direct the solution into a glass cell (Figure 1).

A laboratory scale software/hardware framework for capturing crystal images for this case study was setup. The experimental setting utilizes a USB microscope camera (model MD900) with a resolution of 1280 x 960 pixels, which fits into the side tube on the side of the microscope with one of the supplied adapters and connects to a computer. The AMSCOPE software is utilized (off-line) for manual measurement (individual particle analysis) utilized for training and validation. The magnification used is 25x which corresponds to 0.775 Microns/pixel. This conversion factor was used to manually measure individual crystal sizes on each image. Images were taken at different crystallization stages (30, 60, 90, 120, 180, 240, 300, 360, 420 and 480 min for flowrate of 0.7 and 1.5 ml/min, 10, 15, 20, 30, 60, 90, 120, 180, 240 and 300min for flowrate of 3.0 ml/min. At each crystal growth stage, a set of at least 10 images capturing different amount of crystals were utilized.

5.2 Texture analysis on crystal images

A sample image from crystallization at 10 °C and 0.7 ml/min at 30 min is used to illustrate the method of FD estimation from crystal images. The original grey crystal image (Figure 2a) in TIF format was imported in Matlab for image analysis. To make calculations simpler, the image intensities have been normalized with intensity value into a range of 0 to 1. Otsu's method multiplied by a parameter 1.1 generated a threshold value of 0.7182 for the normalized image; implying pixels with intensity value below 0.7182 belong to edges. Threshold method gave birth to a binary image shown in Figure 2b. The holes presented in the edge detection binary image correspond to crystal bodies in the original image. A morphological operation was used to fill the holes and generated the image shown in Figure 2c which provides the location information of crystal clusters. The locations of each crystal cluster in crystal cluster binary image are the same as those in the original image since the size of the former image is the same as the latter. Identifying crystal clusters was achieved through determining the locations of crystal clusters and storing them in an empty image. The detected crystal clusters are depicted in Figure 2d. The intensities of the crystal clusters were then rearranged in a sequence of the same column order connecting with the next row as a vector with a length of 224203. The intensity vector was the input data for the wavelet-fractal analysis. In this study, wavelet function 'db3' was chosen and decompositions at six levels were performed. We select details from decomposition level 3 to 6 for further analysis. The variance of the detail coefficients are 0.011, 0.083, 0.26, and 0.49 respectively. Performing a least-square linear fit for 2-based logarithm of the detail variance and its corresponding decomposition level, we found the slope was 1.7976. Using equations (4) and the previously calculated slope, we calculated the Hurst exponent to be 0.3988 and FD, according to equation (3), to be 1.6012 when 1 was adopted as the Euclidean dimension for 1-D situation.

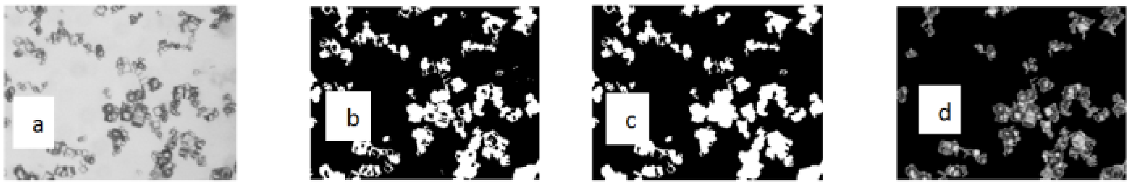

Figure 2: (a) Original, (b) Edge detection binary image, (c) Crystal clusters binary image, (d) Crystal clusters image.

6. ANN models and prediction

The topology structure of ANN was designed to be a three-layer network, which including seven input neurons/variables (temperature, flowrate, wavelet energy signature at decomposition level 3 to 6, FD) and one output/target neuron which is the manually measured mean size or std. The gradient and the number of validation checks are the training termination criteria. Images from the constant conditions comprise of the training set for the ANN model while those from the changed conditions are the testing set. For images coming at the same stage from a specific condition, the corresponding outputs, shared the same value. The predicted size for each stage from a specific condition was the average of generated values by the ANN model for corresponding images. The prediction validity, defined as how well the predicted outcomes obtained from the ANN model fit the experimental values, can be confirmed by the root mean square error ($RMSE$), R^2statistics and sum of squared residuals (SSR) between exp, short for experimental, and $pred$, short for predicted values, as:

$$RMSE = \sqrt{\frac{\sum(exp-pred)^2}{n}}, \qquad R^2 = 1 - \frac{\sum(exp-avg)^2}{\sum(exp-pred)^2}, \qquad SSR = \sum(exp-pred)^2 \tag{7}$$

Two ANN models were built to predict the mean size and std respectively. The manually measured mean size was used as the outputs for the former. Standard deviation from the best global model using nonlinear Fokker-Planck Equation (Grosso et al. 2001) were adopt as the targets for the latter. The statistical characterizations of the size prediction for training and testing sets are listed in Table 1.

Table 1: Statistical parameters of ANN for prediction for training, and testing set.

	Mean size prediction			Standard deviation prediction		
Predicted size by ANN	RMSE	R^2	SSR	RMSE	R^2	SSR
10℃, 0.7ml/min	4.18	91%	174	2.68	62%	72
10℃, 1.5ml/min	9.28	51%	861	2.67	91%	72
10℃, 3.0ml/min	6.66	76%	354	3.97	45%	110
20℃, 0.7ml/min	4.62	88%	213	1.35	94%	18
20℃, 1.5ml/min	8.76	68%	767	2.62	83%	69
20℃, 3.0ml/min	6.23	89%	310	2.00	92%	28
30℃, 0.7ml/min	7.5	89%	564	1.18	87%	14
30℃, 1.5ml/min	11.3	74%	1276	0.97	64%	8
30℃, 3.0ml/min	10.7	75%	912	4.39	60%	135
Changed conditions (test)	5.62	96%	319	2.12	91%	45

Figure 3 shows how the CSD model predictions compare to the raw histogram experimental data and the estimated probability density function (pdf) of the raw data. They show the samples taken at 20, 30, 60, 90, 120, 180, 240, 300, 360, 420 and 480 minutes for the crystallization experiment with changed conditions. As illustrated in the figures, the image-based multi-resolution sensor shows an excellent performance in matching both the smoothed data and the raw data histograms.

7. Conclusions

An image-based approach of texture analysis combining thresholding and wavelet-fractal with ANN for the prediction of CSD was proposed and applied on a case study. This method could successfully and automatically identify crystal clusters and estimate the texture by means of FD and wavelet energy signature. The relationship of FD and crystal mean size had been extracted and built as an ANN model for predicting crystal mean size as well as for standard deviation. The experimental results attest the potential application of the proposed method for crystal production process monitoring and control given good quality of images by high speed cameras. The number of input variables of ANN can be further investigated to improve the predictions.

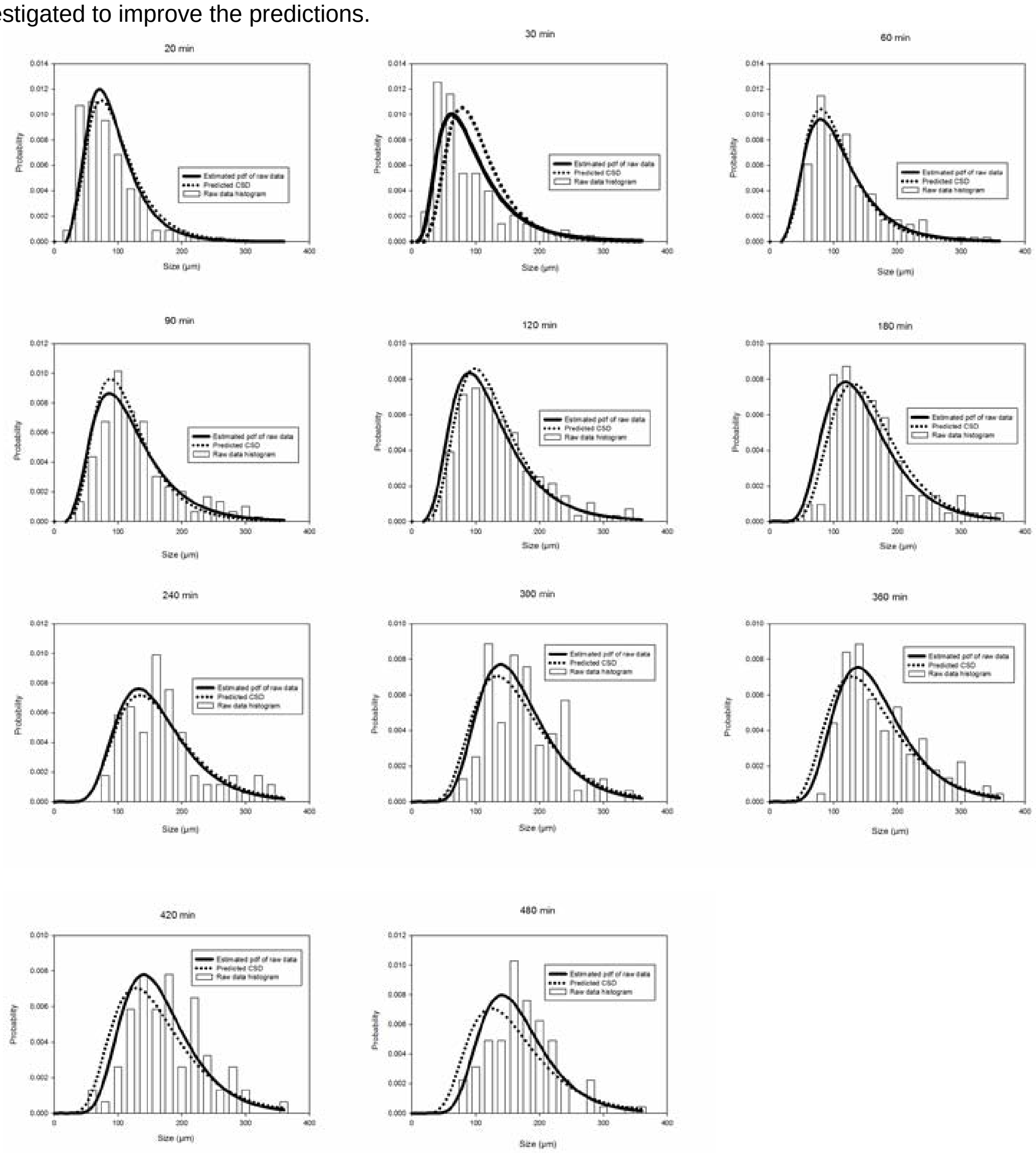

Figure 3: Comparison predicted CSD, raw histogram and the smoothed approximation with the manually measured lognormal distribution parameters at each sampling time from the crystallization at changed operation conditions.

Acknowledgements

J. Romagnoli kindly acknowledges the financial support by NSF through the Award # 1132324

References

Addison P. S., 2002, The Illustrated Wavelet Transform Handbook. Napier University, Edinbrugh, UK: Institute of Physics Publishing Bristol and Philadelphia.

Grosso M., Cogoni G., Baratti R., Romagnoli J.A., 2011, Stochastic Approach for the Prediction of PSD in Crystallization Processes: Formulation and Comparative Assessment of Different Stochastic Models, Industrial & Engineering Chemistry Research, 50, 2133-2143.

Zhang B., Abbas A., Romagnoli J. A., 2011, Multi-resolution fuzzy clustering approach for image-based particle characterization for particle systems, Chemometrics and Intelligent Laboratory Systems, 107, 155-164.