European Research Studies Journal
Volume XXIV, Special Issue 2, 2021

pp. 587-596

Industrial Tomography Platform for Diagnostics and Control of the Crystallization Process

Submitted 24/03/21, 1st revision 11/04/21, 2nd revision 20/05/21, accepted 30/06/21

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Abstract:

Purpose: *The aim of the article is an industrial system platform for diagnostics and control of the crystallization process with the use of tomographic technologies.*
Design/Methodology/Approach: *Various methods are used to study crystallization processes. Here, the tomographic method has been applied.*
Findings: *Tomography of industrial processes is a harmless, non-invasive imaging technique used in various industrial in-process technologies. It plays an important role in continuous data measurement for better understanding and monitoring of industrial processes, providing a fast and dynamic response that facilitates real-time process control, fault detection and system malfunctions.*
Practical Implications: *Sensor networks with their feedback loops are fundamental elements of production control. A critical difference in the mass production of chemicals, metals, building materials, food and other commodities is that common process sensors provide only local measurements, e.g. temperature, pressure, fill level, flow rate or species concentration. However, in most production systems such local measurements are not representative of the entire process, so spatial solutions are required. Here the future belongs to distributed and image sensors.*
Originality/Value: *The concept of a system based on industrial tomography represents a solution currently unavailable on the world market, in its assumptions and effects it has a legitimate character of innovation on a global scale. At the same time, it means the creation of a new, fundamentally different from those available on the market, universal product in the technological sphere. It is an innovative, efficient tool for diagnostics and process control.*

Keywords: *Electrical capacitance tomography, ultrasound tomography, diagnostic and control, sensors, process tomography, crystallization process.*

JEL codes: *C61, C88, L23.*

Paper type: *Research article.*

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1. Introduction

Advanced automation and control of production processes play a key role in maintaining competitiveness. While expensive process equipment and production lines can be considered the heart of industrial production, control systems and information technology are its brain. They provide the flexibility to quickly adapt production processes to changing customer requirements and ensure safety and efficiency at the lowest possible resource and energy costs. Hence, the development and application of advanced process control is one of the most effective levers for immediate and long-term gross energy savings, improved product quality, increased process safety and greater production flexibility, and will provide security and support economic growth in conventional and emerging fields.

The idea to use tomography to monitor industrial processes emerged in the 1990s (Beck *et al.*, 1997*)*. Since then, various tomographic methods have emerged, including: electrical, magnetic, optical, ultrasonic, microwave and radioactive tomography. The common feature of all these techniques is the high measurement speed. However, until now it has not been possible to perform real-time reconstruction of phase, flow velocity, temperature, pressure and chemical distribution properties. The recent development of intelligent massively parallel computing architectures at affordable costs currently enables real-time data processing in a production environment. This allows process tomography to be transformed into a powerful sensor solution in the new system concept presented - tomography-based process control. This concept requires new data processing strategies and a correct extension of classical control theory, as the latter is not sufficiently developed for large amounts of sensor data and has to be created on a non-parametric criterion. For this purpose, knowledge-based control and diagnostic system strategies based on fuzzy logic, neural networks and deep learning algorithms must be developed.

2. Basic of the Crystallisation Process

Crystallisation is a widely used process for purification and particulate production. Most industrial processes use nucleation-cooled crystallisation, in which nucleated crystals of a specific compound are added to a reactor and supersaturation of the liquid is produced by cooling. Cooling crystallisation is used if the solubility changes significantly with temperature and if the compound is thermally stable. Alternatively, supersaturation can be produced by the addition of an antisolvent, that is, a solvent with a significantly lower solubility, or by a chemical reaction, for example as a result of a change in pH. One of the objectives in manufacturing is to control product properties, for example particle size and particle size distribution, purity, residual solvent content, crystallinity and polymorphic form, hydrate or solvate. To some extent, product properties can be manipulated by changing process parameters. A crystal size distribution that meets the assigned specifications, for example, maximum average particle size, minimum coefficient of variation, etc., can

be obtained by growing the given seed crystals and, for example, by minimising nucleation, that is, by keeping the system in an unstable zone, provided that agglomeration and cracking can also be avoided. The operating conditions to obtain the desired product properties can be selected by trial and error or by model-based process optimisation.

In recent years, the use of in situ monitoring tools in crystallisation processes has become increasingly popular, also as a result of the FDA's process analytical technology initiative. Attenuated total reflectance Fourier transform infrared (ATR-FTIR) spectroscopy has been used to measure liquid phase concentrations in precipitation crystallization, cooling and anti-solvent processes. The focused reflection beam method (FBRM) has been used in crystallization processes to monitor the solid phase, either to follow the evolution of the chord length distribution or to monitor the particle size distribution using a suitable reconstruction method. Furthermore, it has been used to detect the onset of particle formation or the point of complete dissolution.

Sarkar *et al.* (2006) and Trifkovic *et al.* (2008) recently estimated the nucleation and growth kinetics of paracetamol by combining population equilibrium modelling and non-linear regression with in situ supersaturation monitoring using ATR-FTIR and measuring the chord length distribution using FBRM. In the same work, optimal antisolvent flow rate profiles were obtained using nonlinear constrained single- and multi-objective optimisation techniques. The same group also applied the multi-objective optimisation method to optimise batch cooling and reactive crystallisation processes, determining the optimal cooling and flow rate profiles, respectively. The application of FBRM to model-based optimisation of particle size distribution in a batch cooling process for paracetamol was also previously presented.

Although many publications focus on offline optimal control, i.e., calculating the optimal trajectory for antisolvent or temperature, (Nagy *et al.,* 2008) designed the crystallisation process of a pharmaceutical antisolvent by feedback concentration control based on ATR-FTIR. A comparison of open-loop temperature control and closed-loop saturation control showed that the latter solution, although more difficult and complex to implement, is less sensitive to disturbances such as changing seed quality or contaminant profile than the former and is therefore more favourable for industrial applications.

3. Optimising Energy Efficiency

Energy efficiency measures and improvements are prioritised by companies today, due to rising energy prices and energy policies being implemented. Rising energy prices, on a national scale, have a devastating impact on the competitiveness of manufacturing companies. Industries located in countries with colder climates, where production is temperature-dependent, are even more affected by rising energy prices. These negative effects can lead to companies moving abroad or being forced

to reduce production. On the other hand, if a company faces increased energy costs, this can increase the incentive to take action on energy efficiency measures. Energy efficiency measures can have a positive impact on the total cost to the company and can lead to increased productivity, thus giving higher profits.

3.1 Optimising the Time and Volume Efficiency of the End Product

Optimisation can also be done from a time perspective (Worlitschek and Mazzotti, 2004). By optimising the time it takes to produce a product, it is possible to find the optimum balance between time and quantity in the crystallisation process. One way to achieve time efficiency is to optimise the quantity of product created at a given moment in the production process. In the case of crystallisation, the mixing speed and flow rate of the crystallising liquids can be optimised in the time-space. In other words, we are looking for process parameter settings for which a minimum is found according to the following:

$$min(c_1\mu_1(t) * R_1 + c_2\mu_2(t) * R_2) - \eta K, \quad (1)$$

for each t. Here $\mu_1(t)$ and $\mu_2(t)$ are time-dependent parameters that are changed as a result of changing the device parameter settings, η is the process yield, and K is the theoretical maximum value of the product produced using the mass of components R_1 and R_2. Finding the minimum in such a space will allow us to find the maximum production for a given moment of time. Monitoring and adjusting these parameters over time will allow the maximum number of crystals to be produced for a given period of time.

The total efficiency of the process can be determined by summing the efficiencies with the appropriate weights:

$$\text{Efficiency} = \sum_{n}^{N} w_n * E_n \quad (2)$$

where w_n are the weights determined by the company for a given optimized parameter e.g. energy efficiency, E_n is the efficiency of a given parameter, N the number of all parameters. In order to find the optimal parameters, statistical analysis can be applied.

3.2 Statistical Models for Optimisation

In order to assess energy consumption in factories, various statistical methods can be used. To get specific information about whether the energy consumption of a factory at a certain time is energy efficient or not, the energy consumption can be compared to historical consumption where conditions were similar. Statistical models based on past energy data can show when energy consumption was higher or when there was

more variability in consumption. Before data can be analysed for correlations and conclusions can be drawn, they must be evaluated. Only data that are relevant to the model are considered.

3.3 Determination of Process Efficiency for Crystallisation

A number of parametric studies of the crystallisation process were carried out, taking into account the variation of parameters such as the reactant infusion rate μ_1 and the stirring rate μ_2. The analysis of the laboratory tests allowed us to find a set of parameters for which the yield of the size of crystals produced by the crystallisation process is the highest.

We define the efficiency of the process as the ratio of the mass of crystals after optimisation to the mass of crystals before optimisation:

$$Efficienty = \frac{m_0}{m_{n0}} \tag{3}$$

where m_0 is the mass of crystals after optimisation and m_{n0} is the mass of crystals before optimisation.

After the experiment the results were obtained the weight of the crystals was 158.3 for the optimised process and 130.6 for the non-optimised process. From the experiment, it was found that the average crystal size was increased by 21.2% due to the optimisation.

4. Models for Crystallisation

The crystallization process is described by the equation:

$$c_1\mu_1(t) * R_1 + c_2\mu_2(t) * R_2 = \eta K, \tag{4}$$

where c_i are the material constants, μ_1 is the injection velocity of liquid 1 into the tank and μ_2 is the rotational velocity of liquid 2 in the tank, and R_i are the mass coefficients of the liquid. On the right side of the equation, we have two quantities K - the mass of the crystallized substance and the crystallization yield. The aim is to achieve the highest possible crystallisation efficiency by optimising the dynamic parameters of the mixing of the liquid (1).

4.1 Regression

From the measurement data we have, we can draw up the dependence of the injection speed of liquid 1 on the rotation speed of liquid 2. A mathematical function can be fitted to such data to extrapolate the data to estimate the optimum values of

the mixing speed and the functionally related injection speed. We propose a polynomial model:

$$\mu_1 = \sum_i^N a_i \mu_2^i + b \tag{5}$$

Alternatively, a polynomial with arguments that are logarithms (due to the data run):

$$\mu_1 = \sum_i^N a_i (log\mu_2)^i + b \tag{6}$$

Having obtained a well-fitted function, we can reduce the dimensionality of our problem. We insert the obtained approximate function into the equation of crystallisation, so the left-hand side of our equation depends only on the variable μ_2. It can now be represented as:

$$F(\mu_2) = \eta K \tag{7}$$

We therefore look for parameters for which the left-hand side of the equation reaches a maximum, which is the same as the maximum crystallization yield on the right-hand side of the equation. The maximum satisfies the conditions:

$$\frac{dF}{d\mu_2} = 0 \tag{8}$$

$$\frac{d^2F}{d\mu_2^2} < 0 \tag{9}$$

The derivatives needed can be calculated numerically or analytically. We solve the resulting equation by any numerical method. The point found determines the maximum crystallization yield.

5. Model Using Tomography

In this approach, we use a tomographic apparatus, which is equipped with a tank used to carry out the crystallisation process. The EIT, ECT (Rymarczyk *et al.*, 2021) and UST (Sikora and Polakowski, 2016) techniques allow us to image a cross-section of the vessel and determine the approximate amount of crystallised substance.

The crystallisation is determined by integrating the images from three tomographic techniques and averaging them, which is described by the formula below.

$$\eta K = \frac{1}{3}\sum_{i}^{3}\int_{S}\tilde{f}_i(x,y)dS \tag{10}$$

where f_i is the value of the reconstruction image derived from the i-th technique. It should be mentioned that in order to perform this type of operation, we must first normalise the reconstruction. Otherwise, one of the images may have a leading role and falsify the result.

We normalise the images to a standard distribution by subtracting the mean and dividing by the standard deviation

$$\tilde{f}_i = \frac{f_i - \bar{f}_i}{\sigma_i} \tag{11}$$

In this way, we obtain the right-hand side of the crystallisation equation. Moreover, we can follow the evolution of this process over time. However, in this method we only focus on the final data, where the fluid velocities remain constant throughout the process. Going further, using a number of measured data we can draw up a functional dependence of effective crystallization on velocity.

As a result, we get the surface area K as a function of two velocity parameters. It is therefore necessary to find the maximum of the obtained surface area.

If the measured data do not show any clear maximum, interpolation of the obtained surface area, e.g. by means of polynomials, should be used. It is then possible to extrapolate a short distance from the approximated area to find an increase in efficiency.

5.1 Dynamic Model

This approach is a generalisation of the previous point using tomography. In this model, we are interested in tracking changes over time as the crystallization process continues. The crystallisation equation is therefore given an additional degree of freedom:

$$c_1\mu_1(t) * R_1 + c_2\mu_2(t) * R_2 = \eta(t)K. \tag{12}$$

In this model, by taking a single measurement we divide it into a series of time steps Δt apart. Again as in the previous model we measure (at each time step) the crystallisation using tomographic techniques. One measurement series draws a curve in parameter space. With a series of measurements, the curves merge into a surface that we can analyse similarly to the previous method.

However, we are interested in the dynamics of the process, since time-varying parameters μi can generate crystallization increments of different severity. In the first approach, we propose to fix one of the parameters, for example μ_1 and vary the mixing speed μ_2 in time.

By performing the experiment with varying velocities we track their increments in successive time steps, similarly with the crystallisation, which we calculate by tomography as in the second model. We define increments of each quantity in a standard way

$$\Delta\mu_i = \mu_i^{(t+1)} - \mu_i^{(t)} \tag{13}$$

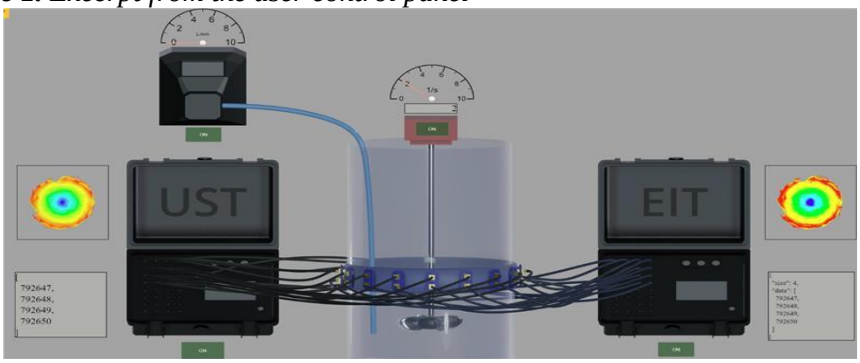
$$\Delta\eta = \eta^{(t+1)} - \eta^{(t)} \tag{14}$$

We then create a space of increments, where for every increment in velocity there is a corresponding change in crystallization. Of course, the crystallization increments are non-negative, so we can only talk about its acceleration or deceleration.

The data collected in this way may be analysed in terms of the search for the maximum increment of crystallisation. This can be done in a simple way - by searching through the data set, or in a more complex way. By approximating the data with a mathematical function and then extrapolating beyond the set of collected data we can look for further increases and use this knowledge in planning further measurements. It is possible that the most efficient crystallisation can be achieved at variable mixing speeds.

A tomograph has been prepared for the crystallisation study, with a user control panel. It contains simple information to edit the device's operating parameters, stop and start devices such as: ultrasonic tomograph, impedance tomograph, variable speed agitator, pump with adjustable pumping speed. The panel allows viewing of reconstructed images as well as process control and steering.

Figure 1. *Excerpt from the user control panel*

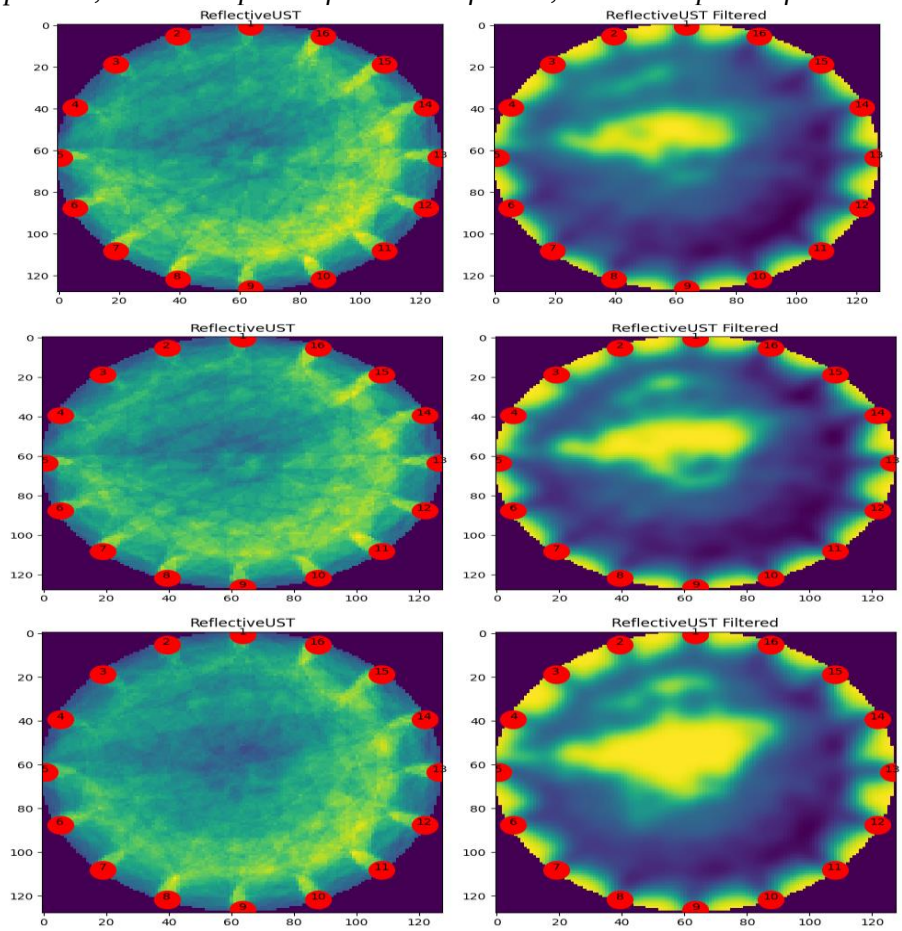


Source: *Own study.*

6. Measurement Results Obtained

A technological process was carried out where the process was monitored using an Ultrasonic Tomograph. The process consisted in the gradual addition of solution A to a tank containing solution B. The reaction resulted in the formation of product C and water. The observed crystallisation was distinguishable in the image reconstruction using the reflected wave imaging technique.

Figure 2. *Evolution of the process with time. The top panel shows the beginning of the process, the middle panel after a time of 2 min, the bottom panel after 5 min*



Source: *Own study.*

7. Conclusions

As part of the development work, it was assumed that a set of solutions would be created to test the functionality for reaction tanks with different parameters and sizes. For this purpose, plastic tanks with the following dimensions were used:

- 200mm diameter, 250mm height, 3mm thickness;
- 200mm diameter, 500mm height, 3mm thickness;
- 400mm diameter, 250mm height, 4mm thickness.

In the first phases of development work focused on the EIT/ ECT technology for this purpose a set of capacitive electrodes made of copper foil covered with insulating varnish was developed and mounted on one of the test tanks. Such a solution allowed to minimise the influence of the wall thickness on the sensitivity of the measurement system and to ensure durability during the tests. The electrodes with their fixings were mounted in two rows of 32 electrodes each, 50mm apart in a vertical plane. This creates two measuring rings. Additionally UST electrodes were mounted between EIT electrodes. The whole solution was prepared so as to enable connection to the measuring system via standard SMB connectors.

Measurements were carried out under laboratory conditions. The measurements are aimed at experimental confirmation of correct operation of all functional elements of the system. The measurements were conducted, among others, for the crystallisation process and were performed using the developed measuring systems within the demonstration platform. Technological processes were analysed by means of 3D spatial and 4D spatial-temporal measurements.

References:

Beck, M.S., Byars, M., Dyakowski, T., Waterfall, R., He, R., Wang, S.J., Yang, W.Q. 1997. Principles and Industrial Applications of Electrical Capacitance Tomography. Measurement and Control, 30(7). https://doi.org/10.1177/002029409703000702,

Nagy, Z.K., Chew, J.W., Fujiwara, M., Braatz, R.D. 2008. Comparative performance of concentration and temperature controlled batch crystallizations. Journal of Process Control, 18(3-4), 399-407. https://doi.org/10.1016/j.jprocont.2007.10.006.

Rymarczyk, T., Maj, M., Kamia, K., Gołąbek, M. 2019. The Fermat rule for Detection of Anomalies in Ultrasound Transmission Tomography. In Applications of Electromagnetics in Modern Techniques and Medicine. Polskie Towarzystwo Zastosowań Elektromagnetyzmu.

Sarkar, D., Rohani, S., Jutan, A. 2006. A. Multi-objective optimization of seededbatch crystallization processes. Chemical Engineering Science, 61(16), 5282-5295. https://doi.org/10.1016/j.ces.2006.03.055.

Sikora, J., Polakowski, K. 2016. Podstawy matematyczne obrazowania ultradźwiękowego. Lublin: Politechnika Lubelska.

Trifkovic, M., Sheikhzadeh, M., Rohani, S. 2008. Kinetics estimation and single and multi-objective optimization of a seeded, anti-solvent, isothermal batch crystallizer. Ind. Eng. Chem. Res., 47(5), 1586-1595. https://doi.org/10.1021/ie071125g.

Worlitschek, J., Mazzotti, M. 2004. Model-based optimization of particle size distribution in batch-cooling crystallization of paracetamol. Crystal Growth & Design, 4(5), 891-903. https://doi.org/10.1021/cg034179b.