

Acoustic chemometrics - monitoring of chemical production processes

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Abstract

Acoustic chemometrics is a new Process Analytical Technology (PAT) approach for on-line monitoring of industrial processes. Acoustic chemometrics concerns capturing system vibration characteristics, e.g. generated by a manufacturing process or by transportation flow. The resulting vibrations can be measured by non-intrusive, "clamp-on" sensors. Acoustic signatures carry embedded information about physical and chemical parameters, such as composition (oil, fat, ammonia, buttermilk, glycol, ethanol), mixing progress, fiber length, flow, density, temperature as well as system state. For extraction and quantification of these types of specific analytes and parameters of interest, domain transforms (FFT, WT) and PLS-regression is essential for multivariate calibration (process chemometrics).

Acoustic chemometrics is here applied for monitoring of industrial production processes, a feasibility study of fluidized bed granulation of a fertilizer product (urea) (Semi-Industrial Pilot Plant: SIPP), illustrating the main acoustic chemometrics features and benefits. We also present examples of monitoring of ammonia concentrations also caused by turbulent flow. We finally discuss monitoring for visualization of critical situations - early operator warnings. With acoustic chemometrics it is possible to monitor both process state and product quality for industrial process control. Relevant early warnings trigger the process operator to change relevant process parameters to control product quality or to prevent critical shut-down situations.

Successful validation of these types of PLS-prediction models signify that acoustic chemometrics has matured into a proven on-line technology in the Process Analytical Technologies (PAT) domain.

Keywords: Acoustic chemometrics, process monitoring, PAT (Process Analytical Technologies), ammonia, urea, multiphase flow, crystallization

1. Introduction

In this study we show the development of a new method to monitor industrial granulation processes [1], as well as an industrial application example where acoustic chemometrics is used to predict concentrations of ammonia. Earlier papers [2, 3, 4] describes the first forays of small-scale pilot experiments, and gave an introduction to the acoustic chemometric approach both for liquid flow in pipelines and industrial granulation processes.

An introduction to acoustic chemometrics has been published earlier [3, 4]; Acoustic chemometrics concerns capturing passive system vibration characteristics, e.g. from two-phase systems (gas-solids/liquid-solids) generated by a manufacturing process or by transportation (flow in pipelines). The resulting vibrations generated by the process itself can be easily measured by non-intrusive, "clamp-on" sensors (accelerometers). Vibrations/acoustic emission (audible noise) from industrial processes is often considered as audible noise only, but in this paper we show that the vibrations or "noise" contains relevant information for processes monitoring purposes.

Intensive signal processing is necessary to extract relevant information. The raw signals are preprocess by Fourier transformation and the resulting spectra we call acoustic signatures. Acoustic signatures carry embedded information about a whole range of system-relevant physical and chemical parameters e.g. composition (oil, fat, ammonia, glycol, ethanol), mixing progress, fiber length, flow, density, temperature - as well as system state. For extraction and quantification of these types of specific analytes and parameters of interest PLS-regression [6] is essential to extract relevant information regarding the parameter of interest by calibration of a regression model based on empirical acoustic data and reference values for the parameter(s) of interest.

Acoustic chemometrics for fluid flow quantification:

Acoustic emission from fluid flow trough an orifice plate inserted in a pipeline contains information which can be used to predict parameters of interest e.g. composition [4]. Acoustic signatures from fluid flow are affected by several factors e.g. flow rate, differential pressure over the orifice plate, static pressure downstream etc. Several application examples showing application examples of e.g. trace concentrations of oil in water has been reported [4]. Figure 1 shows liquid flow trough an orifice plate. The relatively high differential pressure loss leads to cavitation downstream of the orifice plate. Cavitation occurs when the local hydrostatic pressure is lower that the vapor pressure and micro bubbles are generated as can be seen in figure 1. The vibrations/sound produced by the micro bubbles which are affected by surface tension, viscosity, both static and differential pressure.

Oil in water reduces the surface tension of the water and generates a dramatic change in the vibrations/sound emitted from the orifice.

Vibrations are recorded using acoustic sensors (accelerometers) which can be mounted on the surface of the pipeline. The sensors are easy to install and are so-called non intrusive which means there are no need for drilling holes through the pipeline.



Figure 1. Liquid flow through an orifice plate, note cavitations downstream generated by the relatively high pressure loss causing the local pressure to drop below the vapor pressure. (photo by Lienhard & Stephenson)

Acoustic chemometrics for monitoring of particles in granulation processes

This approach is here used in order to gain acoustic measurements of vibrations produced by process equipment or product (particle) movement in a semi-industrial granulator, used in the experiments to produce a suite of specialized fertilizers; the present study focus on Urea.

Granulation of Urea is a complex process, which is controlled by experienced process operators. The parameters used to monitor the granulation process are so-called standard process measurements such as temperature, pressure and flow. The standard measurements have no information (or are only very indirectly related) to e.g. particle size, clogging of the reactor or the accumulating depository layering on the bottom plate – and often with a quite unacceptable delay time. A sample of layering cake on the bottom perforated plate, taken out of the reactor after several days in production is shown in figure 2, already a serious process impediment. When the layering cake develops further, the perforated bottom plate of the reactor necessarily becomes increasingly clogged with a resultant fluidization airflow decrease. Decreased fluidization in turn leads to a situation with less agitation of the particles; the result is often deformation of big lumps, which can quickly lead to a shutdown of the reactor, and a significant economic loss (reactor downtime and general production flow stoppage during clean-up).

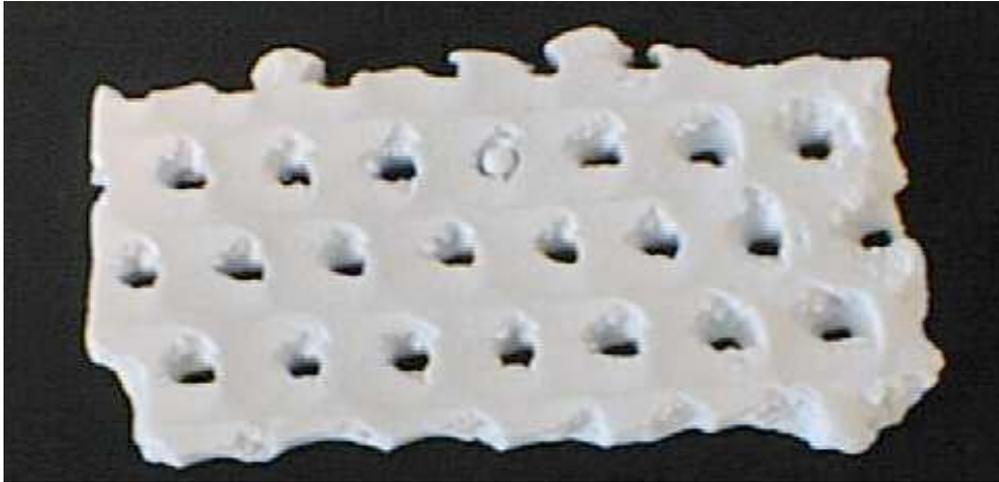


Figure 2. Layering on the perforated bottom plate in the semi industrial granulator after several days of production.

A measurement system that can predict the thickness of layering cake, particle size, or give an early warning of lump formation is thus highly wanted. Similarly, an acoustic chemometrics prediction facility for general process state monitoring is of equally critical importance.

One of the major goals of the feasibility study is to relate process state trends, presented as chemometric score plots, to specific conditions/qualities of the product inside the reactor. The process operators can then use this “new” information to better operate the process, with an ultimate objective to significantly reduce costly shutdown situations. On-line measurements of particle characteristics such as particle size distribution together with properties of the liquid feed to the sprayer nozzles makes it manifestly easier to control the process.

The results concentrates on the results from an experimental trial period of several months, involving a suite of induced deviations of the general production process in order to learn as much as possible about the feasibility of acoustic chemometrics.

Experimental

The objectives of the experiments reported in this paper are divided into 3 major themes:

1. Investigate different sensor positions on (in) a semi-industrial granulator
2. Assessment of the feasibility of acoustic chemometrics to:
 - Concentration of Urea melt
 - Crystallisation point temperature of liquid Urea feed to the granulator
 - Moisture content in the granules
 - Monitoring of ammonia concentration flowing in pipelines
3. Monitor the overall granulator process state, to detect critical situations and to visualize these situations as early warnings in an operator-friendly fashion (lump formation and clogging of the bottom plate are the most important mishaps in the industrial production setting).

The experimental equipment consists of a semi-industrial pilot fluidized bed reactor, illustrated in figure 3, which highlights five different sensor positions [A, B, C, D]. All the four sensors are mounted with screw-fittings onto a metal surface (in order to secure a stable sensor pick-up efficiency). Sensor position A is mounted onto an *orifice plate* on the main supply line of liquid urea to the reactor nozzles, following Esbensen et al. 1999 [4]. Sensor positions B, C and D are mounted directly onto the wall of reactor chambers 1, 2 and 4 respectively.

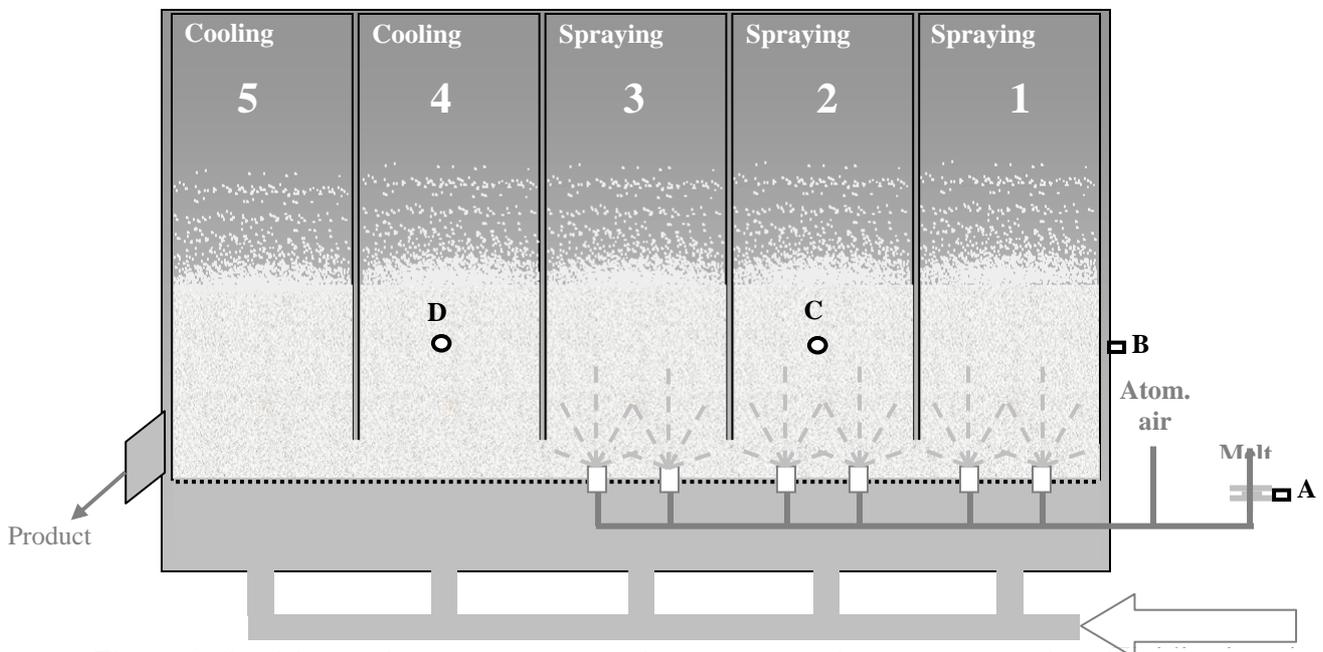


Figure 3. Semi-industrial granulator used in all the experiments reported in this paper. Sensor positions A, B, C and D are indicated.

The semi-industrial granulator displayed in figure 3 is identical to an industrial full-sized granulator except for size, which is 1:10 roughly. The granulator is divided into five chambers, three injection (spraying) chambers and two cooling chambers. The injection chambers each have several nozzles where liquid urea is sprayed into the granulator at a certain process temperature. The bottom of the reactor is a perforated plate, which allows fluidization air to jet into the reactor, to interact with the growing particles and keep all particles in the bed in vigorous agitation. The cooling chambers are used to cool down the granules before they exit as the final product: urea granules with a specified size and size range (important parameters for agro-industrial product use).

Sensor A is mounted onto an orifice plate inserted in the main supply pipeline for liquid urea. The orifice has a smaller hole-diameter than the pipeline, which induces *turbulence* in the flowing urea downstream the orifice. The vibrations produced by this turbulence will be detected by sensor A. Sensors B, C and D are mounted on the vertical wall on the granulator, about 30 cm above the perforated bottom plate; they are supposed to detect *vibrations* produced by the granules when they interact with the reactor wall. Thus sensors B, C and D are used to monitor the process conditions inside the granulator, while sensor A is used to monitor the liquid supply of urea. The sensors used in this trial are four high-temperature accelerometers.

The present measurements were recorded in a "piggy-back" mode, as other process experiments - in themselves not related to acoustic chemometrics - were carried out. This resulted in many days with stable conditions in the reactor, and no particular variations in the acoustic signals. Therefore there were only a limited number of days (hours), which display the necessary *variation in process parameters*, which are necessary for successful multivariate calibration. These still turned out to constitute a satisfactory basis for the present full feasibility study however.

Semi-industrial reactor experiments and results

Concentration of urea melt:

A model for urea concentration in the melt sprayed into the granulator was developed based on acoustic spectra recorded from sensor position A, during a trial period of 5 hours. Sensor A is mounted onto an orifice plate inserted in the main supply pipeline of liquid urea (full information about the principles of acoustic chemometrics in fluid flow systems using orifice plates can be found in [4]). The reference values used to calibrate the model are based on pressure measurement on the pipeline, which is used to *calculate* the concentration in a standard fashion accepted by the industry involved. The reference in figure 4 is an indirect measure of the concentration; it is not even converted to concentration [%], as we can equally well show the prediction performance using this industrial concentration measures for both predicted and for measured (reference) values.

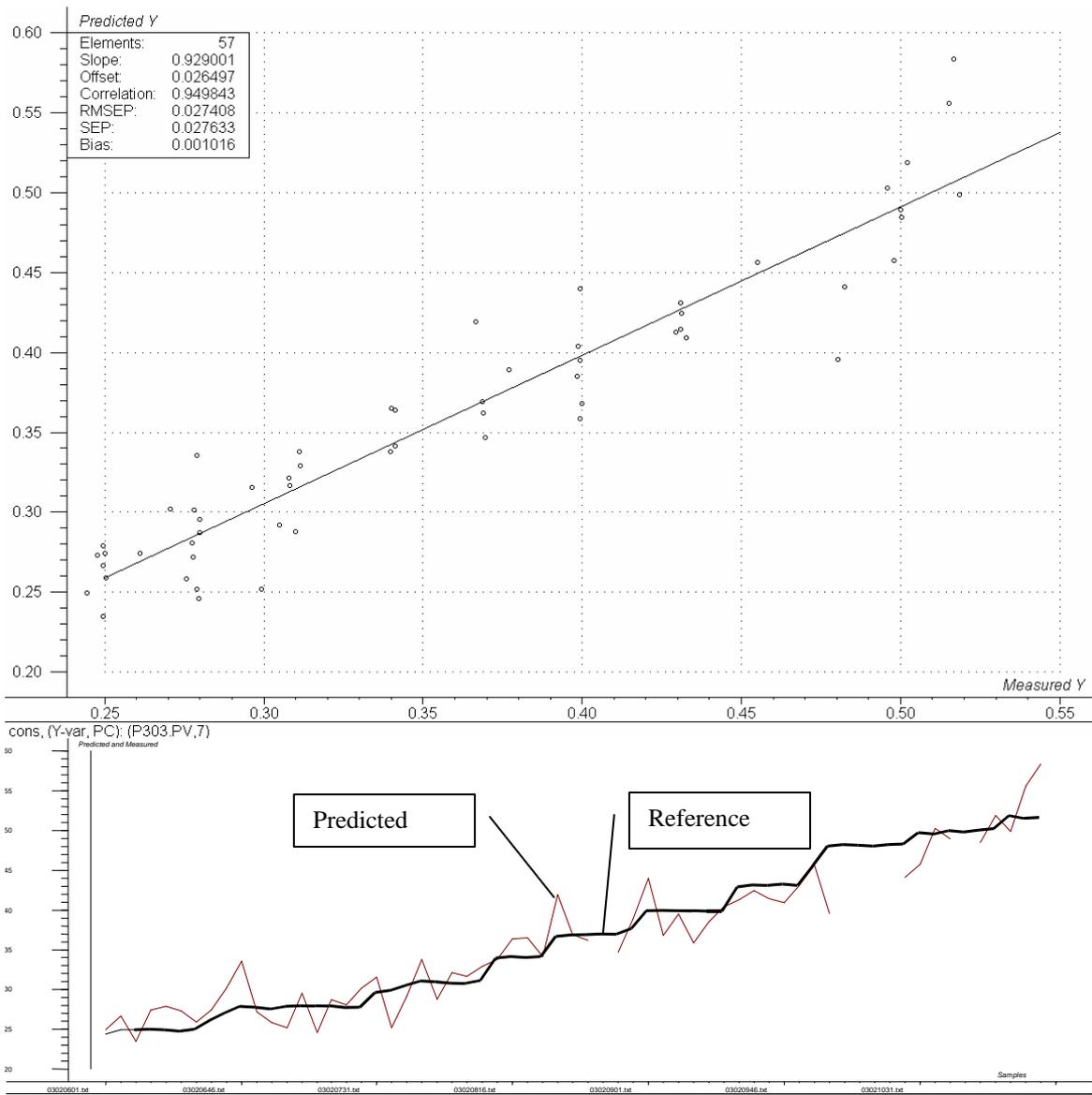


Figure 4. PLS-1 model for urea melt concentration sprayed into the granulator in chamber 1, 2 and 3. Sensor A was used in this model, which is based on 7 components. The model was validated with 10-segmented cross validation. Predicted vs. measured (top) and predicted vs. time (bottom).

The model presented in figure 4 shows that it is possible to get a satisfactory prediction of the concentration of the liquid urea. Slope = 0.93 - Relative RMSEP = 13%.

Acoustic data were also calibrated against the average particle size as well as the spread (variance) of particle sizes, which were calculated from laboratory sieving samples. The results for average particle size were on an equal satisfactory footing as those shown above (not shown here). While the laboratory analysis of particle size and – variances were of quite satisfactory accuracy, the actual physical sampling of the products were suboptimal w.r.t. *representative sampling* in the sense of the

Theory of Sampling, Gy [7, 8] due to very tight resources in the present pilot study, which had one overriding constraint: no adverse interference with the ongoing industrial production process. The particle spread constitute the prime parameter for further development of the present approach as soon as it has been included in the industrial on-line monitoring scheme.

Multivariate Statistical Process Control (MSPC) monitoring as an early warning of critical situations (shut down situations):

One of the main objectives in this project was to assess the acoustic chemometric potential to monitor the general process state with an aim to give a so-called “early warning” if a critical situation occur in the bed. A critical situation in the bed is often a result of lump formation and/or layering on the bottom plate of the reactor. 15. February 2001 such a critical situation occurred in the bed, which was (definitely) not according to the experimental plan, and thus very “welcome”. Lump formation which probably been building over several hours, suddenly resulted in an uncontrolled shutdown around 16:30 the same day. Analysis of the acoustic data recorded in the period immediately leading up to this critical period shows an interesting *trend line*, figure 5.

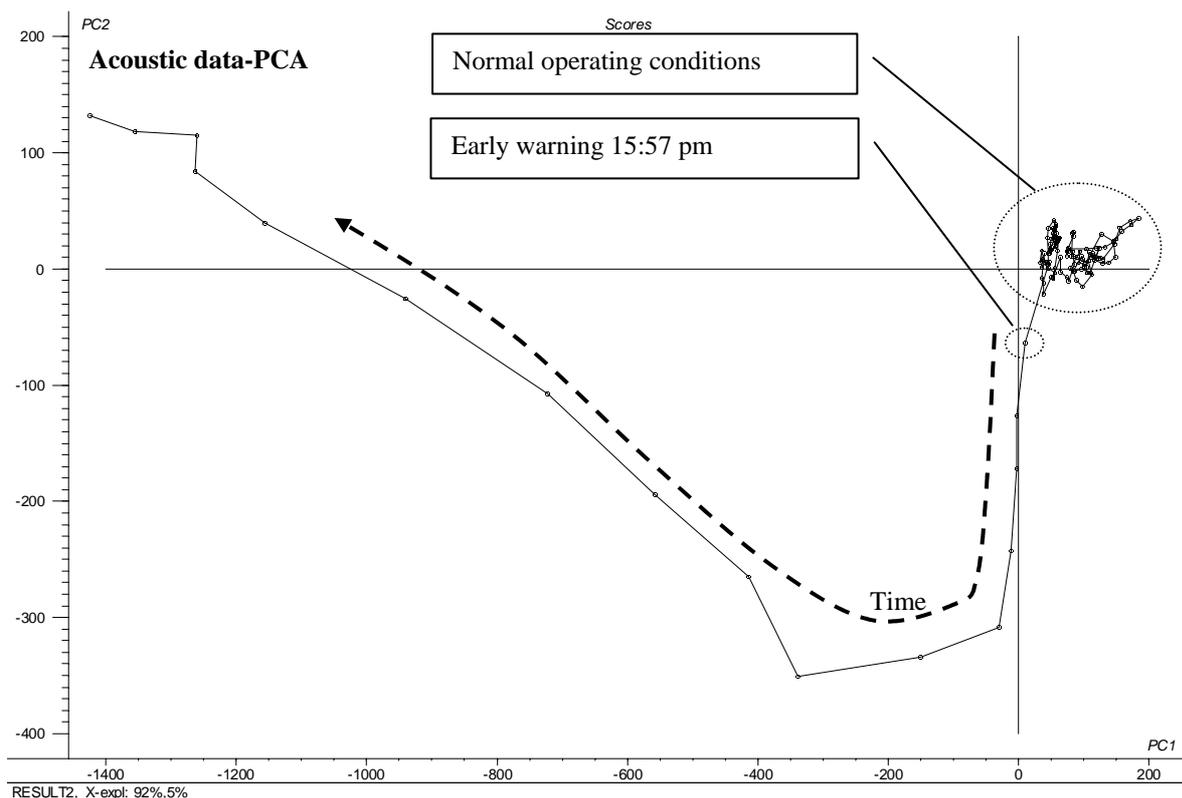


Figure 5. Score plot of PCA-components 1 and 2. Sensor B was used because lump formation first started in chamber 1. Acoustic warning appears as early as 15:57, approximately 30 minutes before reactor shutdown. Compare with Fig. 7, which shows the same situation as delineated by traditional process parameters only

From the trending score plot in figure 5 it can be seen that an operator (in an on-line situation) would have had early warning at least 30 minutes before the shutdown. According to the extensive, process-specific operator experience, this would very likely have been enough time to take the necessary preventive action, needed to bring the granulator back into normal conditions without a shutdown occurring.

Figure 6 shows the loadings from the PCA analysis, which clearly shows that the progress in the direction of principal component 1 in the figure above is caused by lump formation in chamber 1 measured by sensor B.

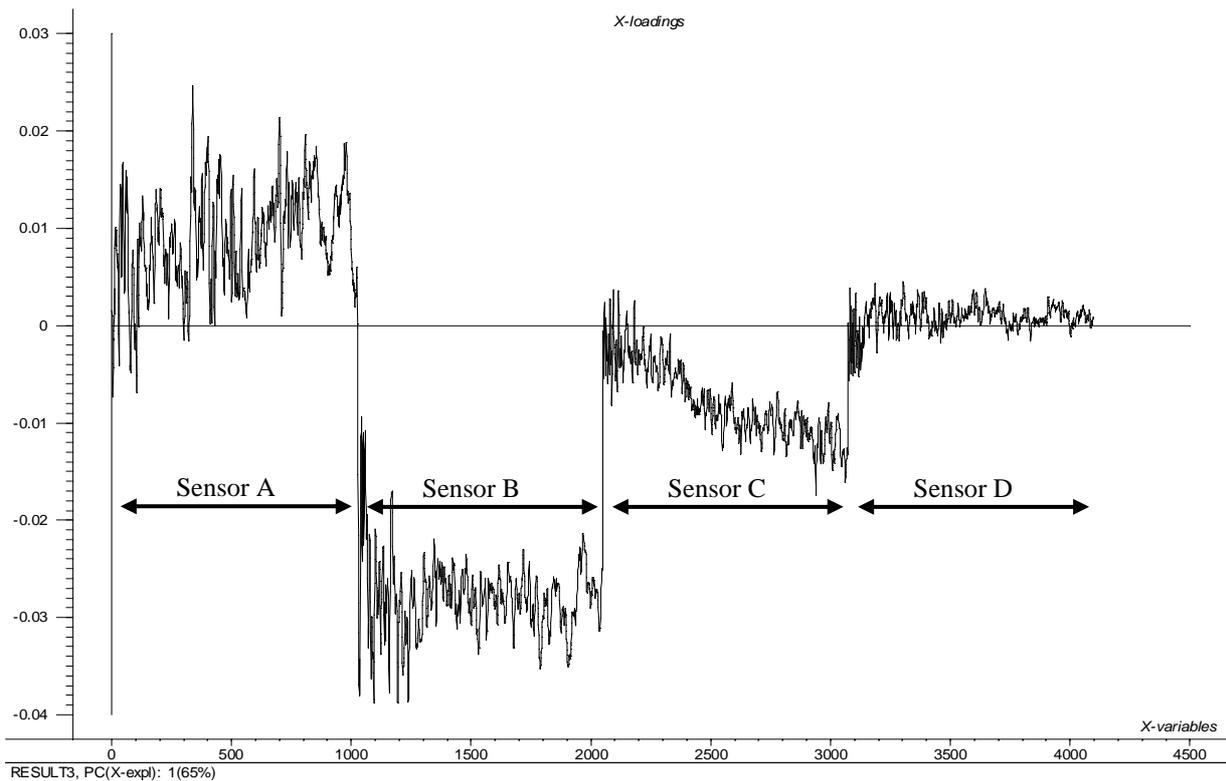


Figure 6. Loadings for principal component 1 shows that the lump formation started in chamber 1 (sensor B).

A similar PCA-analysis was performed on the standard process data only for the same time period. Note that here the warning first shows up at 16:20, which cannot be called “early”, but rather late, probably too late, according to the same process operator consensus. The results from the analysis of the process data can be seen in figure 7 below for comparison.

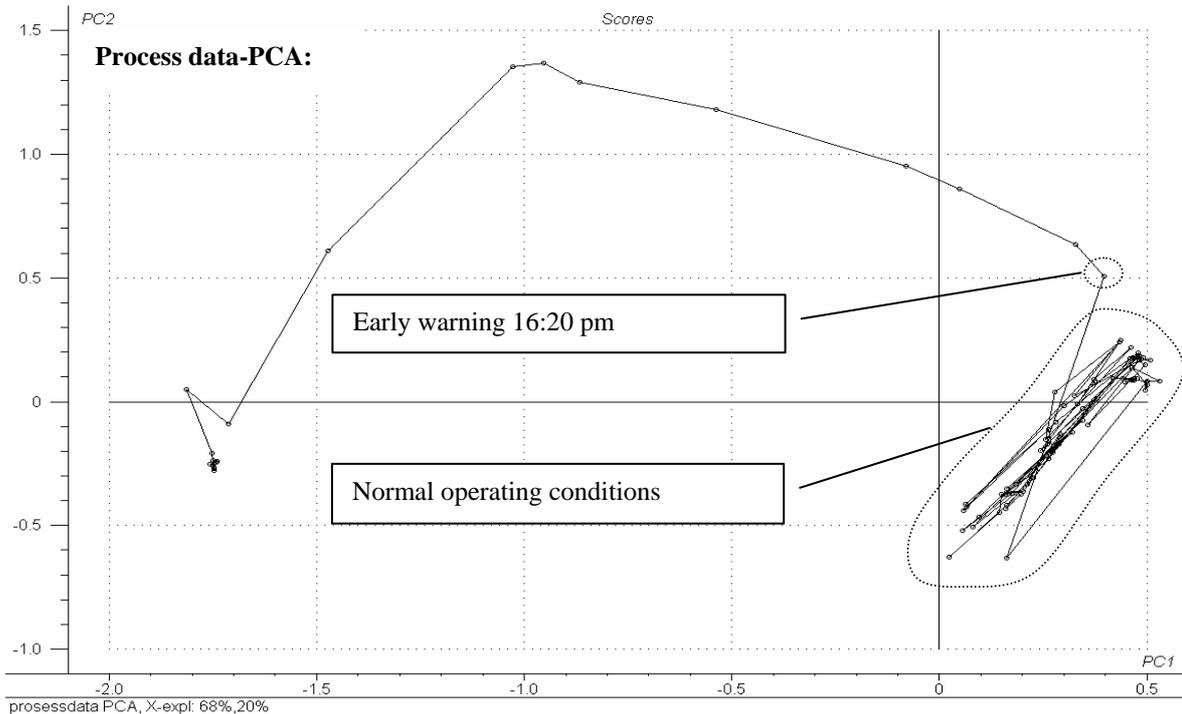


Figure 7. Score plot of PCA-component 1 and 2 of the process parameters alone. A warning that something is wrong first appears 16:20, only some 10 minutes before shutdown of the reactor, which is usually deemed insufficient for process recovery.

This means that the acoustic chemometric approach is much more sensitive to changes in the process state(s) of the bed reactor than the traditional process data alone. Of course an implemented acoustic chemometrics process monitoring facility would use both these sets of parameters together with appropriate chemometric data analysis (PCA, PLS).

Model calibrated on data from the full trial period of 5 months:

A model for granule moisture content was calibrated with data from the full 5-months trial period.

Granule moisture content

To test the acoustic chemometric potential to predict granule moisture content, the same 1032 object 5-month data set was used, where the first 900 objects were used for calibration and the last 132 as a test set [6]. The data matrix was also further re-samples slightly because the acoustic data had to be calibrated against laboratory tests of moisture content which were only available with a relatively low sampling rate; still plenty of results were at hand to allow a full assessment of the prediction performance re.granule moisture. The results can be seen in fig. 8.

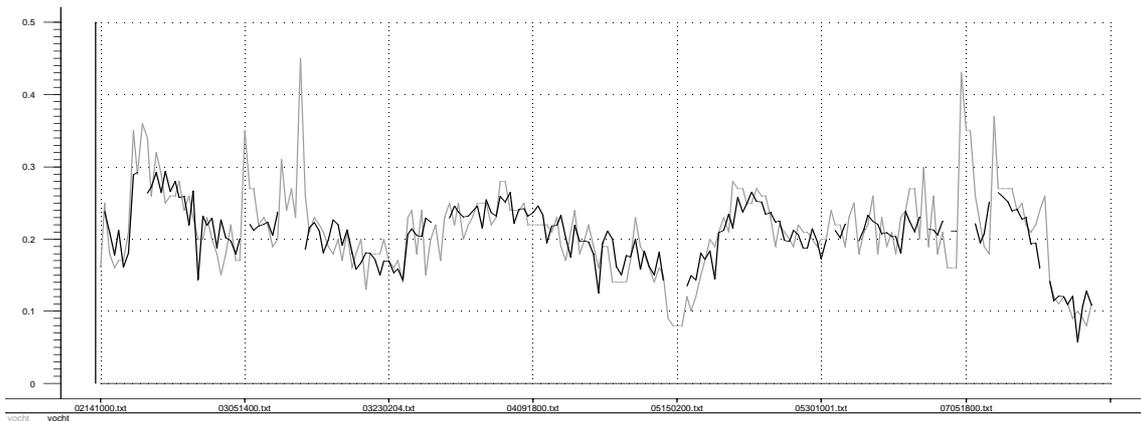


Figure 8. Predicted vs. measured plot for granule moisture contents, calibrated on data from five months of production. Sensor B was used in this model (8 PLS-components). The model was validated with 10-segment segmented cross validation. Predicted vs. production time. Grey curve: measured, black curve: predicted

In spite of the fact that this model has a relative RMSEP of 14%, the general ability to pick up the important production trend changes is already at an acceptable level.

Ammonia concentration – Industrial application example

An experimental industrial setup for prediction of ammonia concentration directly from acoustic spectra has been tested in a full scale industrial plant. Figure 9 shows the bypass loop with the orifice plate. The acoustic sensor was mounted onto the orifice plate [4]. To ensure constant differential pressure and temperature of the ammonia two pressure transmitters and one temperature transmitter was used in conjunction with the orifice plate. Reference samples was taken at the sample valve shown in figure 9.

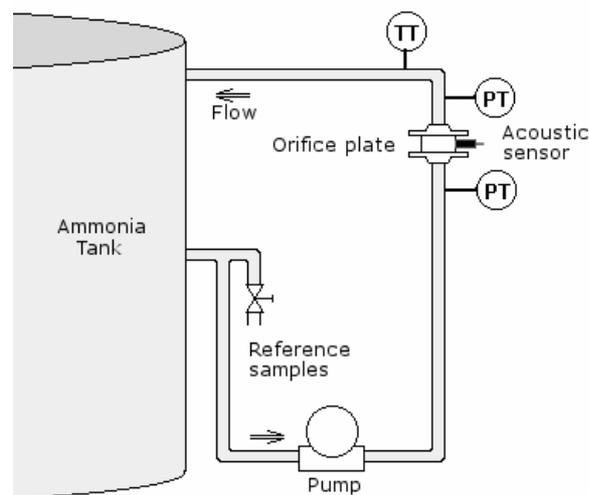


Figure 9. Experimental setup for assessment of acoustic chemometrics ability to predict concentration of ammonia directly from acoustic emission generated by cavitation/turbulent flow. Full-scale industrial pilot study.

Calibration of the model for prediction of ammonia concentrations

Acoustic spectra were calibrated using Partial Least Squares regression PLS-R with six ammonia concentration levels, each characterized by 5 replicate acoustic measurements. Figure 10 shows the concentration levels spanning 0 to 8 % of ammonia concentrations.



Figure 10 Ammonia concentration reference levels 0, 0.5, 1, 2, 5 and 8 % ammonia, 5 replicate measurements on each level.

Results

Figure 11 shows the PLS-R prediction results validated with cross validation.

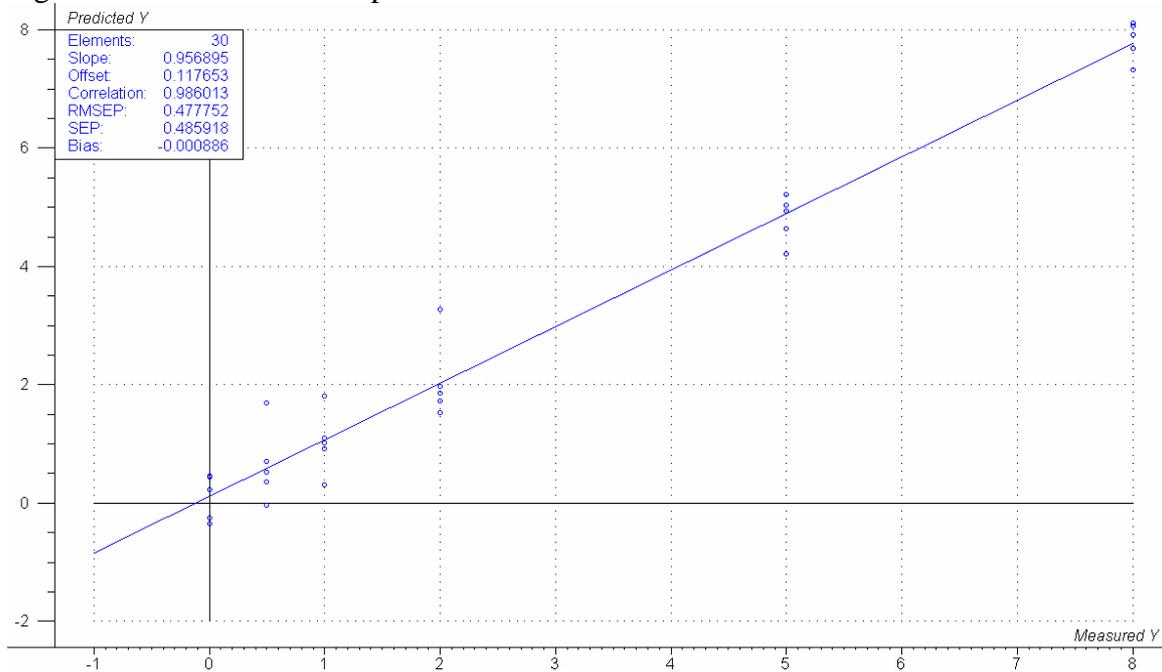


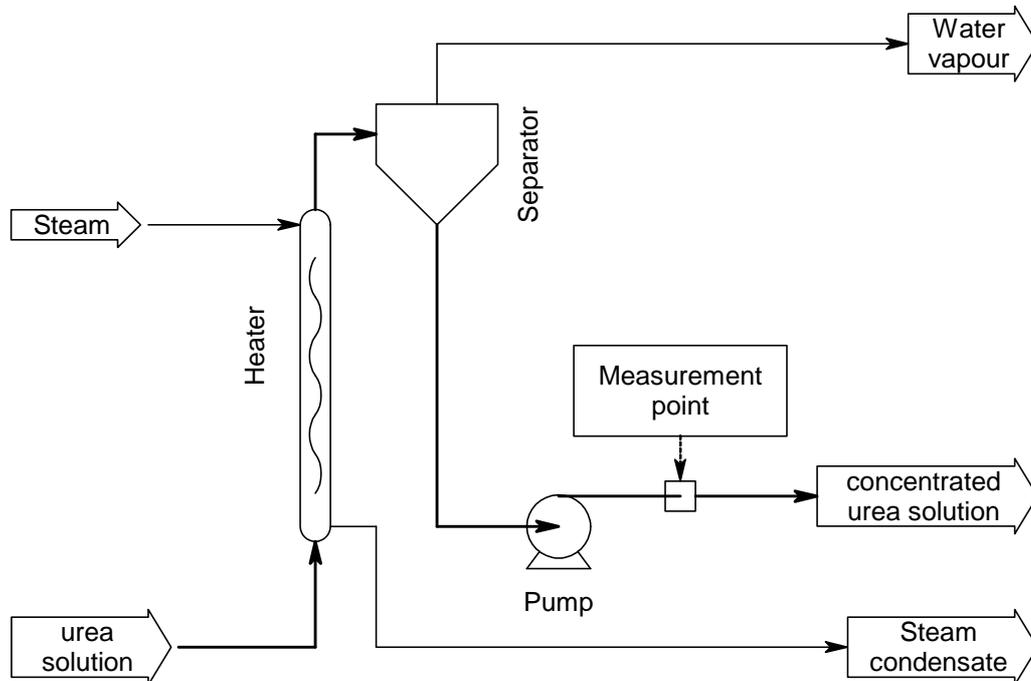
Figure 11 Prediction results for ammonia validated with 2-segment cross validation, RMSEP = 0.48 % ammonia.

This pilot study included only 6 concentration levels. With a 5-component PLS-model there is a potential danger for modeling over-fit. Even a 2-segment cross-validation

is no absolute guarantee [6]. However, there would certainly appear promising possibilities for further, significantly extended calibration work.

Prediction of Urea crystallization point temperature – Industrial experiments

X-matrix: Acoustic spectra from sensor located on orifice plate in the UREA feed to the granulator, Fig. 12.



Flowsheet of the liquid feed to the Urea granulation process



Figure 12. Acoustic sensor mounted on orifice plate in liquid feed line to granulator

A PLS-regression model based on X (acoustic spectra) and Y (crystallisation-temperature) was established. The X-matrix contains 12 objects, each with 1024 variables (frequencies 0 - 25 kHz). An overview of the X-data is shown in figure 13, in which can be appreciated systematic changes in the acoustic signatures following the object (samples) succession.

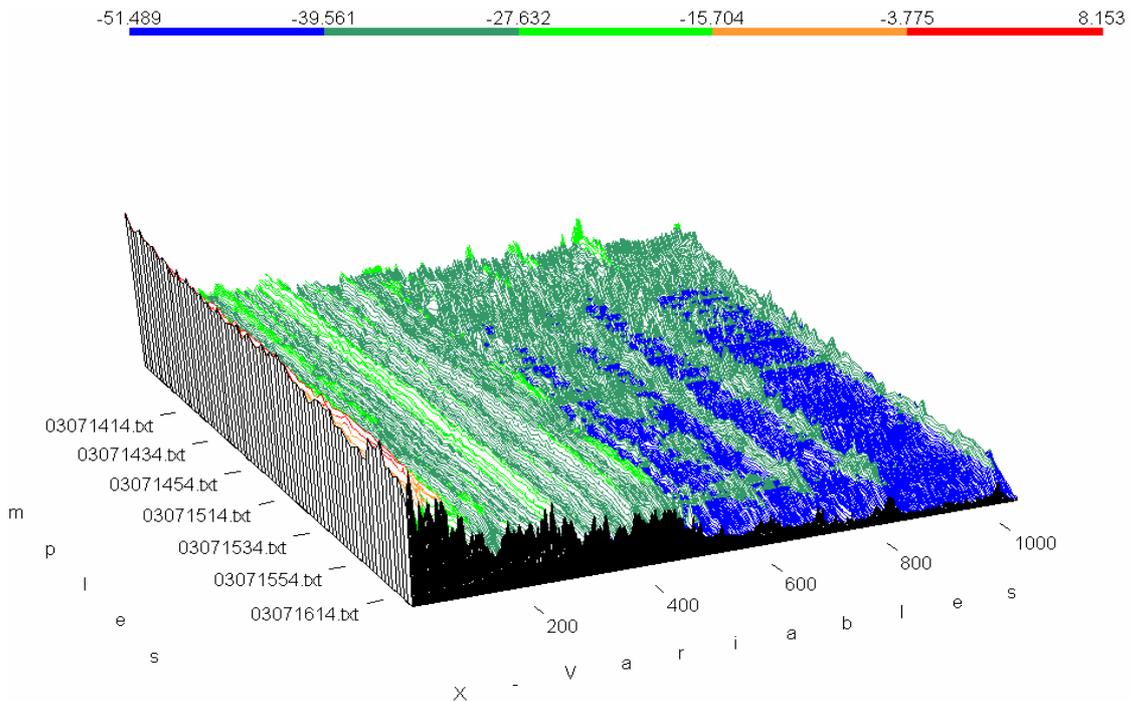


Figure 13. X data matrix of acoustic spectra used in calibration

The objects span a nominal urea concentration range: 85.5 to 91.4 %. The PLS model will be built on experimental Y-reference values (crystallisation-temperature), spanning 92 - 107 °C. A model for urea concentrations can also be established following appropriate laboratory data (not shown here).

Results from multivariate calibration ($Y = \text{xx-temperature}$):

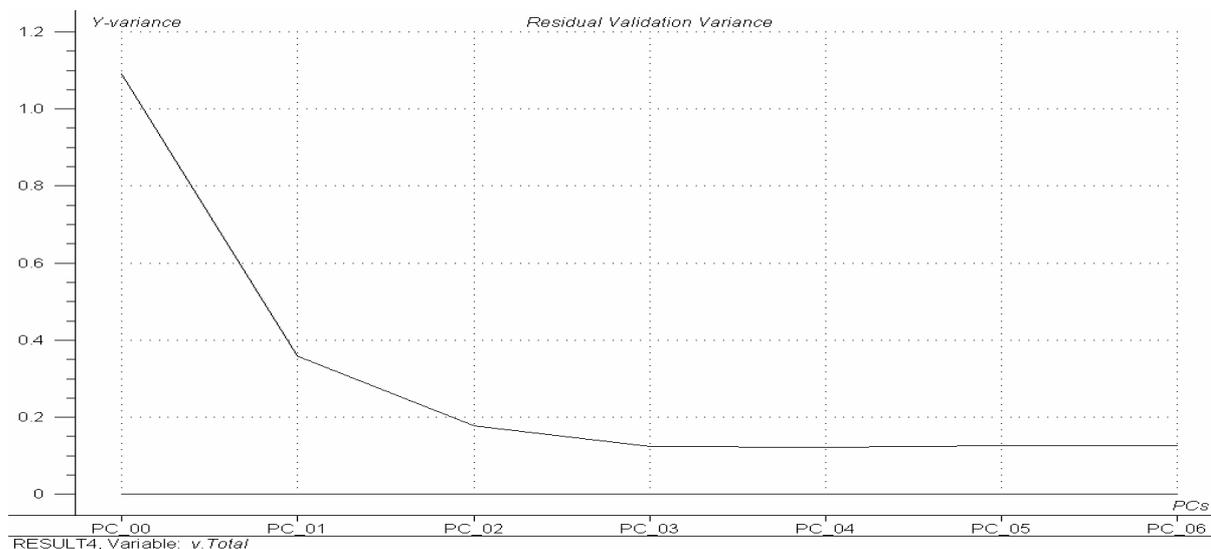


Figure 14. Y-validation variance modelled vs. number of components in the model

The crystallisation-temperature model (no outliers) is able to describe 87% of the Y-variance with 3 PLS-components, Fig. 15; this is a satisfactory modelling with this relatively small calibration range for crystallisation temperatures.

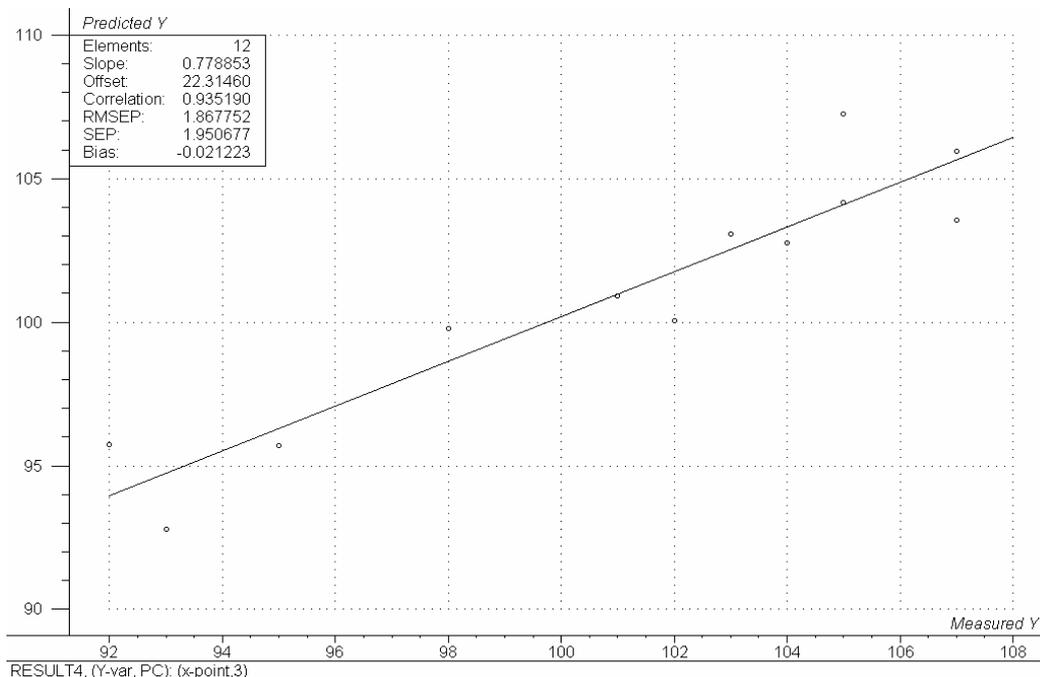


Figure 15. Predicted vs. measured plot for UREA crystallisation-temperature

The *Predicted vs. Measured* plot in Fig. 15 shows the degree to which the acoustic signatures are able to predict crystallisation-temperature: RMSEP := 1.87 °C corresponds to a relative prediction uncertainty of approx. +/- 4% (2 STD).

Figure 16 shows an alternative illustration of the accuracy of the prediction values, when compared to reference values in their process time relationships (N.B. truncated Y-axis).



Figure 16. Predicted vs. measured UREA crystallisation-temperature in relation to the granulator experiment setting up an urea concentration gradient

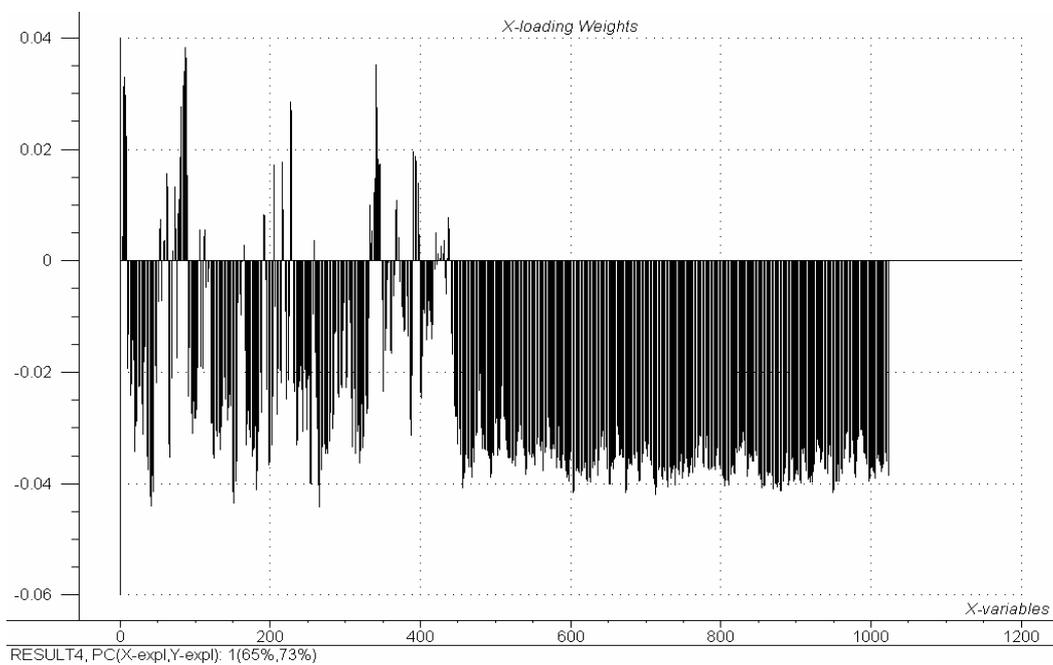


Figure 17. Loading-weights for PLS component 1

The prediction model is characterised by 1024 regression coefficients, alternatively 3 x 1024 loading-weights spectra from the full 3-component PLS-model. From Figure 17 it can be observed several well-defined frequency bands in the lower 0-10 kHz range contribute significantly to the model, while all higher frequencies shows extensive redundancy. This is a reflection of stable acoustic signatures, well correlated with the changing crystallisation-temperature.

The experimental crystallisation-temperature prediction model was evaluated using 12-segment cross-validation (full cross-validation); this can be considered acceptable for such "small sample" data sets [6], although only for indicative estimates.

This pilot study shows that there would appear to be good prospects to predict crystallisation-temperature directly from the acoustic signatures of the liquid feed into the granulator with an indicated prediction error (RMSEP = 4 % relative, 2 STD).

Discussion & Conclusion

Sensor positions, Sensor A (on the orifice plate) and sensor B (chamber 1) showed the overall best results for the industrial granulator. Acoustic chemometrics can be used to predict, a.o. fluidization airflow, reflux of fines to the reactor, granule moisture content and general process states. The first models were calibrated with data from one week only. Each week several other experimental parameters were changed so that the conditions were only comparable within this timeframe. The resulting predictions for the parameters investigated gave satisfactory results in this context.

Since the overall objective was to predict parameters on-line, the next step was to calibrate the models with data from a longer period of time to span all variations that can occur for industrial production reactor runs. We conclude that acoustic chemometrics provide the process operators with useful information which can be used to run the process with less critical shutdowns.

5-month calibration models:

These results, considering the *extensive* compounding of the underlying non-acoustics process conditions that had to be accepted, can be considered *promising* for the potential of acoustic chemometric on-line granulation process monitoring for airflow and liquid urea concentration. There remains a certain amount of focused calibration work before more precise predictions of moisture can be expected.

All the above experiments have been set up so as to be realistic scenarios w.r.t. industrial MSPC-monitoring of granulation processes (MSPC: = Multivariate Statistical Process Control). The pilot plant granulator is operated exactly as the industrial scale counterpart. But the 5-month experiments included much more severe variation than what will usually be found in an otherwise stable industrial production situation of similar duration - e.g. it is *normally* not necessary to change nozzles, or to change formulation (products) with similar short intervals; also additives are certainly not changed as often as in the present trial campaign - all of which goes to show that the strength of the present positive validation assessments are in fact strong indeed.

Another argument for the above is that all the most important parameter validations (airflow, moisture and liquid concentration) are *both* based on identical 10-segment cross-validations as well as proper *test sets*, Esbensen (2001) [6]. The latter have here been displayed in their time-dependent fashions, which are most relevant for the industrial process operators.

Process state monitoring:

We are currently experimenting further with different types of data visualization plots of the type shown in Figures 6-7. We are involving process operators directly in this work, and their preliminary conclusions are generally positive. More experience (e.g. as to the usefulness of the "trend score-plots", "residual variance plots" etc.) is needed however before these prototype acoustic chemometric MSPC development will achieve complete acceptance from this critical target group. It is relevant however that the system described herein is already implemented on-line in the SIPP reactor control room.

Together with papers [2,3,4,5] we have here taken an important first step towards transforming on-line MSPC acoustic chemometrics from an experimental concept to a proven technology.

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