Data-driven advances in manufacturing for batch polymer processing using multivariate nondestructive monitoring

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Abstract

Incorporation of advanced manufacturing depends on efficient strategies that can use new available sensor technologies to improve quality monitoring and process understanding. One of these new technologies is nonlinear ultrasonics, which is a multivariate nondestructive method for the characterization of produced plastic parts. Two approaches are proposed to integrate captured data for in-line quality classification, and on-line monitoring and prediction. Cluster identification is evaluated with a combination of principal component analysis (PCA) and a soft class analogy method for products with differing quality based on information contained in the multivariate ultrasonic signal, providing a cost-effective alternative to destructive testing. In the second

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approach, a state-space dynamic model using subspace identification is applied to historical process data and correlated with the ultrasonic-based quality data for quality prediction. An on-line monitoring tool was proposed, in combination with a non-parametric evaluation. Results were validated with experimental data from a polyethylene rotational molding process.

Keywords: Advanced manufacturing, Quality monitoring, Nonlinear ultrasonics, Batch dynamic modeling.

1 Introduction

Manufacturing is progressively changing to a more intelligent environment. Based on recent trends, the next decades will focus on wide spread utilization of advanced manufacturing concepts, such as big data, cyber-physical systems and cloud computing.[1] Data mining and machine learning strategies are examples of new tools that have been demonstrated essential for the efficient use of big data in process systems[2] Although these cyber resources are gradually becoming known and adopted for use, broad application of these techniques is still years away from being realized.

In polymer manufacturing, which is the focus of the present application, much emphasis has been on additive manufacturing so far,[3] with too little discussion yet on intelligent, data-driven smart manufacturing strategies for traditional processes. The concept of more intelligent data-driven manufacturing focuses on the collection and use of larger amounts of process and quality data to make decisions based on intimate understanding of the process.[4] Various data-based procedures have been investigated for both continuous and batch processes.[5,6] The concept of advanced manufacturing is becoming increasingly relevant due to the increase in the amount of digital data being recorded and stored, which in turn arises from the adoption of new sensors capable of more than traditional univariate descriptors, such as temperature, pressure and flow. Spectral sensors, for example, generate multivariate datasets that can provide a multitude of quality parameters from the manufacturing environment. On the molecular-level, structural modification has been demonstrated with the use of spectroscopic techniques, such as Raman spectroscopy to monitor changes in morphological amorphous and crystalline structures;[7] fast-Fourier transform infrared (FTIR) spectroscopy can be used for observing chemical modification;[8] and, nuclear magnetic resonance (NMR) can differentiate molecular level chain dynamics.[9]

When considering bulk nondestructive characterization methods, a study recently demonstrated the use of multivariate nonlinear ultrasonics to identify differences in the structural morphology of polyethylene (PE).[10] Linear ultrasonic characterization based on sound velocity and attenuation through the media has been traditionally applied;[11–13] however, this approach is limited by the viscoelastic nature of industrial polymers, like PE, which creates a high degree of signal attenuation that is dependent on frequency.[14] Although the adoption of newer ultrasonic methods focused on multivariate analysis is suggested, technological and economic barriers have prevented their implementation for in-line quality monitoring; in-line monitoring refers to the quality assessment after the product has been produced.

Better decision-making tools are sought that can handle larget amounts of dada, showing improvement in their assessment and prediction performance.[15] Development of these techniques has been especially focused in areas such as chemometrics[16] and image processing.[17] Orthogonal projections, such as principal component analysis (PCA), have been at the foundation of several multivariate data analysis tools. It can be used to simplify and quickly understand complex databases with an algorithm that is simple to implement and easy to compute.[18] PCA has been widely used to analyze in-line spectroscopic data of processes experiencing chemical changes[19] and to aid in the detection of tracer elements[20] in continuous compounding. PCA base models have also been demonstrated as adaptable tool for data mining in the big data era[21] In the area of polymer processing, infra-red and Raman spectroscopy sensors and univariate ultrasonic in-line measurements have been demonstrated for monitoring an extrusion process.[22] A hyperspectral imaging sensor was tested by others for continuous extrusion, and its information correlated with the final product quality by multivariate statistical tools.[23] There is even an example of multivariate sensor technology to monitor in-mold production.[24] These other applications of multivariate analysis with spectral datasets[25,26] are more familiar to the manufacturing environment, reliant on known calibration and analysis protocols. Nonlinear ultrasonics presents new challenges for the field of research since the signal contains complex morphological information on the systems being monitored. Intelligent analysis tools have yet to be demonstrated with this sensor technology for manufacturing, proving the data can be related to quality aspects of the product.

Motivated by the above considerations, this study proposes an analytical and statistical framework that can efficiently use multivariate spectroscopic data from nonlinear ultrasonics combined with process modeling to provide an in-line monitoring tool for nondestructive assessment of produced parts considering chemical and morphological changes that were caused during processing, and an on-line process modeling tool to improve understanding of the process variables and allow final quality prediction. The proposed methods were applied to a batch rotational molding manufacturing system.

The manuscript is organized as follows: Section 2 shows the experimental methods used for practical application and validation of the technique. Section 3 describes the statistical approaches used for multivariate data processing and process modeling. Section 4 demonstrates the application of the monitoring tools, where historical batch data is first used to build the models, and validation of the monitoring ability by correctly classifying desired and undesired products, and demonstration the prediction capability. Section 5 presents the concluding remarks.

2 Process Description and Quality Measurements

In this section, we describe the specific polymer manufacturing process and the destructive and nondestructive techniques utilized for characterization of the manufactured part quality.

2.1 Batch Manufacturing Process

A laboratory-scaled uniaxial rotational molding system was operated to prepare cubic samples by melting a high density polyethylene powder (Exxon Mobil HD 8660.29, supplied by Imperial Oil Ltd.). Rotation speed was kept constant at 4 RPM. Two heated panels and a compressed air supply were manipulated variables by PI controller. Temperature data corresponding to the heated panels and internal mold air was measured using K-type thermocouples and collected using a custom-written data acquisition system in Labview (National Instruments). After the powder was charged into the 90x90x90 mm cubic mold, each sample was subjected first to a heating cycle to a selected maximum temperature. In the subsequent cooling cycle, fan-circulated air was applied to the mold to solidify the part, while all manipulated variables were turned off.

It is well understood that the final product quality is largely influenced by the temperature trajectory of the molding process[27] The mold undergoes the following four phases over the course of processing (see Figure 1 for a representative profile of the internal mold temperature): (i) the adhesion phase, where the powder heats up and adheres to the surface, (ii) the melting phase, and (iii) the sintering phase, and finally (iv) the solidification phase. Of the complete sequence of precessing events, the sintering phase is the most critical that dictates product quality.[28] Depending on the duration and temperature trajectory during the sintering phase, one can produce a weak part by incomplete sintering (residual internal air bubbles present), or extensive thermo-oxidative degradation; or if the process is well controlled meet target quality showing optimal mechanical properties and no significant degradation. Accurately measuring the temperature trajectory, and understanding of how it influences the final product quality, remains as incredibly challenging problems. Traditionally, a univariate selection of batch process time or maximum internal air temperature is used to decide the end of the heating cycle. The changes in process operation (i.e. raw material variability) can be very frequent, requiring several adjustments in process variables to achieve desired quality. Monitoring approaches that are inexpensive and rapidly quantify of the product quality in-line (after the batch is finished) to eliminate defective parts, or on-line (during the batch process) are therefore highly valuable.

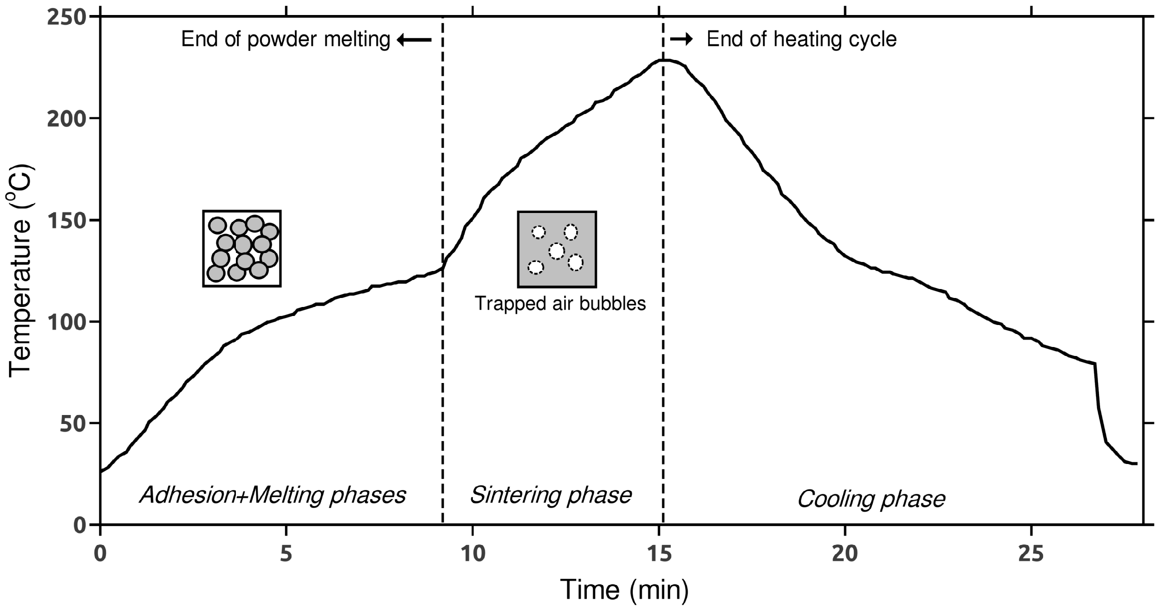


Figure 1: Rotational molding batch internal air mold temperature profile

2.2 Destructive characterization

In order to validate the quality of each sample, two traditional destructive tests were performed to evaluate the physical properties of produced parts. The extent of melt consolidation from sintering (i.e. removal of trapped air bubbles during the process) was evaluated using a falling weight dart impact test (ASTM D5420) as is typically done by rotomolding processors. The impact data was used for classification purposes in this study where a sample was considered fully sintered if its impact value was above 0.41 Joules (J); this threshold was determined as 80% of the maximum impact energy measured for optimal parts (0.51 J). To evaluate the thermo-oxidative degradation, an oscillatory rheology test was performed on a square cut, 30x30 mm, from a part in a DHR 2 parallel plate rheometer (TA Instruments). Complex viscosity estimation was obtained with a frequency sweep from 0.1-100 rad/s at a temperature of 190 ^oC. Data was converted using the Cox-Merz transformation and used the Cross model to estimate the value of zero-shear viscosity. Samples were classified as degraded if the zero-shear viscosity was above 8160 Pa.s, or 20% higher than the supplied material before processing (approximately 6800 Pa.s).

The practical execution of the described characterization methods was limited to a small set of samples from a series of produced parts. Information obtained from a sampled group might not portray the real quality of all batch runs. Total cost of the quality assessment procedure is increased by the use of specific equipments and specilized procedures for different quality tests that need to be executed separetely (sintering and degradation). This motivates the use of alternative, less expensive, quick and non-destructive test methods.

2.3 Ultrasonic characterization

Nonlinear ultrasonics have been shown in recent work[28] to be an effective tool that can be correlated with traditional quality tests to evaluate both sintering and degradation effects on a rotational molded part. The ultrasonic measurements were carried out after the sample was removed from the mold and cooled to room temperature. Figure 2 presents the schematic of the two ultrasonic transducers (F30a - broadband and R15 - resonant, Physical Acoustics Corp.) positioned on one of the external surfaces, at a distance of 35 mm apart. A high vacuum grease (Dow Corning) was used to affix the sensors to the surface. A series of pulses (10 cycles-burst) were introduced to an uncut molded sample, with controlled frequencies from 135 to 165 kHz, in 1 kHz ascending frequency steps. The signal was sampled at an acquisition rate of 4 MHz (using a National Instruments data acquisition board). The spectrum used in the analyses was from all 31 captured signals for the same sample after Fourier transformation.

In this manuscript, the validity of nonlinear ultrasonics as a viable alternative to destructive characterization for quality classification is illustrated. The second relevant contribution is a framework with the ability to predict the product quality on-line, enabling process corrections and control as appropriate. In the next section, we describe the modeling tools that

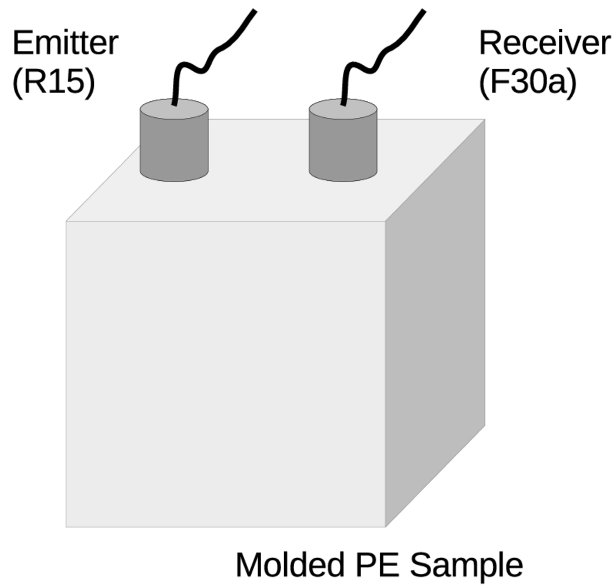


Figure 2: Nondestructive ultrasonic measurement schematic

we utilized to achieve these objecives.

3 Data-driven classification and modeling approaches

We address the monitoring problem for a general scenario where the nondestructive multivariate sensor for product characterization is available at the end of a batch process, with specific application to the rotomolding process. To this end, two approaches are proposed, one for in-line quality classification and the second for on-line quality prediction and process visualization. The first approach focuses on the ultrasonic data evaluated from a produced part at the end of every batch run for an in-line classification. Strategies described in Section 3.1 and 3.2 demonstrate a data processing methodology to reduce the complexity of the signal and show how to improve classification between qualitative classes. This nondestructive classification tool can be applied in the situation where traditional characterization tests are to be minimized. The second approach combines process modeling and non-parametric evaluation, explained in Section 3.3 and 3.4, for on-line prediction of the final part quality. Correlation between a dynamic model with the ultrasonic reduced space, allows at the current state of the process, to predict the final product quality, that can be validated using the ultrasonic sensor once the process is concluded and the part is formed. This decision support tool can help understand causes of process variability and be a foundation for quality control strategies.

3.1 Principal component analysis (PCA)

Nonlinear ultrasonic analysis implies the interpretation of harmonics, thus requiring that the captured signal must be converted from the time domain into the frequency domain. Instead of traditional ultrasonic analysis that focuses on amplitude and sound velocity calculations, in nonlinear ultrasonic analysis, changes in peak amplitude from different frequencies are correlated with structural characteristics.[28] To achieve this, principal component analysis (PCA) was applied to first reduce the dimensionality of the ultrasonic multivariate data without losing important information, using Equation 1 below:

$$\mathbf{U} = \mathbf{T}\mathbf{P}^t + \mathbf{e} \tag{1}$$

where $\mathbf{U}$ is the matrix with ultrasonic spectra organized in rows from different batches; $\mathbf{T}$ is the concatenated scores vectors, $\mathbf{P}$ is a matrix with loading vectors, and $\mathbf{e}$ is the matrix of residuals. The reduced score space is able to capture the essence of the information available in the ultrasonic measurement, and can be interpreted using the loadings vector to understand the importance of frequencies. The PCA model forms the basis of the classification strategy described next.

3.2 Soft independent modeling of class analogy (SIMCA)

Ultrasonic spectroscopic data from rotational molded parts has been correlated with traditional destructive tests to evaluate both sintering and degradation problems.[28] For most spectroscopic techniques applied to predict product quality, a calibration model derived from controlled conditions based on a design of experiments and results from destructive tests are required. In practical industrial applications, a calibration model is often either unavailable or infeasible. Thus, a flexible and efficient method is proposed for quality classification based only on nondestructive data of historical samples, that can later be validated with secondary (destructive) tests. The soft independent modeling of class analogy (SIMCA) tool was utilized. The method utilizes the square prediction error (SPE) values calculated from PCA models to achieve classifcation.[29]

Algorithm 1 describes in details the steps required to create and update a classification model for different quality classes. Selection of initial clusters can be done manually through the visual inspection of the scores plot of the base PCA model or through a cluster algorithm to improve reproducibility Specifically, for the rotational molding process, three classes were used as a starting point for the SIMCA algorithm, in order to utilize the ultrasonic measurements alone to identify the presence of these different quality products. Any new sample may be determined to belong to a particular class based on the SPE values, and the PCA models updated at the end of one set of new measurements to enable subequent classification.

Algorithm 1 SIMCA algorithm

1. Create base PCA model with all available data.
2. Identify initial cluster points from the scores space, separate data into defined classes
   (a) Calculate separate PCA models for each class.
3. Classification of a new element
   (a) Calculate standard prediction error (SPE) for each PCA model. Resultant matrix $\mathbf{SPE} \in \mathbb{R}^{l \times o}$, where l is the number of variables and o is the number of groups.
   (b) Compare calculated values and locate in similar class using SPE limit values or find the combination of vector $\mathbf{a}$, where $\mathbf{a}_i \in \mathbb{R}^o$, to satisfy the objective function: $min \sum_{i=1}^{l} \mathbf{SPE}[a, i]$.
4. Model improvement: If the classification of a specific group meets the criteria, incorporate the new sample into the classified group; Repeat step 2 with the new dataset built. If a newly introduced sample does not fit in any previous groups, consider the creation of a new class.

This approach only requires data from the ultrasonic test and can be applied to any classification that allows differentiation by the physical characteristics that influence the ultrasonic spectrum, allowing a simple interpretation of a complex data set. As indicated earlier, supplementary destructive tests can be conducted to give meaningful physical labels after the fact. In the present examples, these could include, for instance, target quality (meeting desired specifications), incomplete sintering and degraded.

3.3 Subspace identification for dynamic batch process modeling

The development of the on-line monitoring tool was based on a dynamic model that was able to predict the trajectories of process variables (the internal mold temperature for the present application), for a candidate future manipulated input trajectory. Furthermore, the prediction from the dynamic model could then be utilized as the basis to predict the utlrasonic spectrum (described in the Section 3.4).

Batch manufacturing processes have traditionally been treated with a rigid recipe approach, requiring a design that would be specially selected for each material and equipment. A flexible tool to model and understand variability in these processes was proposed with the application of subspace identification.[30] The model identification procedure outlined in[30,31] yielded a linear time-invariant dynamic model that enabled prediction of process outputs (based on candidate future inputs trajectories), and generalized the subspace identfication procedure[5] for batch process operation.[32] In contrast to previous PLS based modeling approaches that uses the concept of maturity to represent a time invariant space for batches with different lengths,[33] no batch time alignment was required for the proposed approach. The identified model took the form of Eqs.2-3 below:

$$\mathbf{x}^d_{k+1} = \mathbf{A}\mathbf{x}^d_k + \mathbf{B}\mathbf{u}_k \tag{2}$$

$$\mathbf{y}_k = \mathbf{C}\mathbf{x}^d_k + \mathbf{D}\mathbf{u}_k \tag{3}$$

where $\mathbf{x}_k \in \mathbb{R}^n$ represents the process state at different sampling instants, k; $\mathbf{u}_k \in \mathbb{R}^m$ and $\mathbf{y}_k \in \mathbb{R}^l$ denote the input and output values at sampling instant k, respectively; and matrices $\mathbf{A} \in \mathbb{R}^{n\times n}$, $\mathbf{B} \in \mathbb{R}^{n\times m}$, $\mathbf{C} \in \mathbb{R}^{l\times n}$, $\mathbf{D} \in \mathbb{R}^{l\times m}$, m input variables, and l measured variables. The model relied on subpsace states to appropriately capture the process dynamics over the training data. For the purpose of prediction for a new batch, the subspace states needed to be first estimated, and in the present work, this was done using a Luenberger observer, as shown in Eq. 4 (see,[34] for more details).

$$\hat{\mathbf{x}}[k+1] = \mathbf{A}\hat{\mathbf{x}}[k] + \mathbf{B}\hat{\mathbf{u}}[k] + \mathbf{L}(\mathbf{y}[k] - \hat{\mathbf{y}}[k]) \tag{4}$$

where the hat mark denotes the observer prediction and $\mathbf{L}$ is the observer gain determined by ensuring that the matrix $(\mathbf{A} - \mathbf{LC})$ is stable. From any point in time for a new batch (after the state estimator has converged), this model could be used to predict the process outputs for any candidate input trajectory, and thus could be utilized as a on-line monitoring tool.

One of the contributions of the present work was to utilize the underlying dynamic model to build an associated quality model that was able to predict, and visualize the final quality, enabling its use as a quality monitoring tool.

3.4 On-line final quality projection

A combination of the ultrasonic data processing, described in Section 3.1, and the dynamic modeling, presented in Section 3.3, was used to develop an on-line monitoring tool to correlate process state and the final product quality.

The proposed new approach connected subspace state variables with the reduced variables related to the final quality (ultrasonic frequency data after PCA) from historical batch data collected for training. More specifically, multiple linear regression was utilized to build a model (see Eq. 5).

$$\mathbf{T} = \mathbf{R_a}\hat{\mathbf{x}}_\mathbf{f} + \mathbf{R_b} \tag{5}$$

where $\mathbf{R_a} \in \mathbb{R}^{n \times o}$ and $\mathbf{R_b} \in \mathbb{R}^{n \times o}$ are the multiple linear regression coefficient matrices that correlate the final state variable, $\hat{\mathbf{x}}_\mathbf{f}$, a vector obtained from Equation 4 with the scores, $\mathbf{T}$, from the projection (Equation 1) of ultrasonic spectra to a reduced space, where n is the order of linear time invariant system and o is the order of projected scores space.

This relationship was not built on mechanistic understanding, but relied on the assumption that all the information about the process was captured in the state vector. Thus, the final quality depend on the final state vector as well. This was also the basis of how the states were determined in the subspace identification approach.

Important advantages were found with the combination of the two techniques for on-line monitoring. The use of a subspace based time invariant model allowed the quality model to be applied for all batches regardless of the duration of the batch trial, without the need for an alignment variable. Any nonlinearities in the process dynamics were captured (as best as they were expressed in the available data) through the choice of the number of states and the resultant subspace model. The use of the reduced space rather than the full spectrum for the ultrasonic multivariate signal, captured the essence of the information contained in the spectrum, and thus a linear relationship between these two sets of variables ended up being sufficient to predict the final quality.

One important contribution of the present work was the development of the visualization tool. While the subspace states have been demonstrated to be important in capturing the process dynamics,[30] evolution of the subspace states by themselves did not lend itself readily to physical interpretation beyond the outputs they predicted. In the present work, therefore, a visualization tool was developed, aimed at predicting the utlrasonic spectrum at the end of a batch trial. Note that the ulstrasonic spectrum, with its correlation to the final part quality, was much easier to interpret and be read by practitioners (see[35] for another instance of off-line process visualization tool).

Another tool that utilized the proposed correlation between subspace states and the reduced space of the utlrasonics was demonstrated as an alternate to the classification steps in Section 3.2. A univariate non-parametric classification approach was utilized next to classify final quality based on the on-line reduced space projection of the ultrasonic signal. The task was then, for a new batch, to identify in the known samples which ones were most similar to the newly produced part. This approach did not require an extensive calibration based on previous data points. In order to execute this search and recognition, the use of the k nearest neighbors (k-NN) algorithm was proposed. The objective was to find the closest, or most similar, cases for a certain number of observations, k, thus minimizing the value of the Euclidean distance, d defined as follows:

$$d = \sqrt{\sum_{a=1}^{k} \sum_{b=1}^{p} (\hat{t}_b - t_b^a)^2} \tag{6}$$

where p is the number of components of the reduced space, $\hat{t}$ is the projected score from the current state variable from Eq. 5, and t^a is the score from the a-th closest sample from the database of historical batches.

With this non-parametric evaluation, the process data extracted from the subspace identification could be used to explore all the previous batch data available for prediction of product quality. Therefore, with new data being available, the prediction could be improved. However, larger available datasets also require more computing power for the search, thus justifying the work being done in a reduced spaced (i.e. data processing using PCA).

4 Results

4.1 In-line quality monitoring

Recall that univariate analysis of an ultrasonic signal, focused on time domain estimation of amplitude and sound velocity, is traditionally used for characterization or process monitoring. In order to demonstrate the effectiviness of the proposed approach, the traditional methodology was used as comparison. Figure 3 shows the ultrasonic amplitude for several batch runs with known final qualities separated into three different categories: incomplete sintering, with residual internal air bubbles present; degraded, with extensive thermo-oxidative degradation due to long exposure to heat; and, meeting target quality, for samples showing optimal mechanical properties and no significant degradation. It was possible to observe a clear distinction between samples with incomplete sintering and the other groups, since the air bubbles present caused an increase in signal attenuation that reduced the final amplitude. However, no clear distinction was notable between the target and degraded groups. Also, even if a threshold value for amplitude was selected for differentiating each group, some samples would fall outside the defined groups due to natural variation in the amplitude that was related to the experimental procedure (for example, measurement sensitivity to the application of coupling the vacuum grease between transducer and surface of the sample). Thus, in the case of this study, relying only on the univariate analysis (i.e.

signal amplitude) would not allow for a clear in-line classificaion between quality groups.

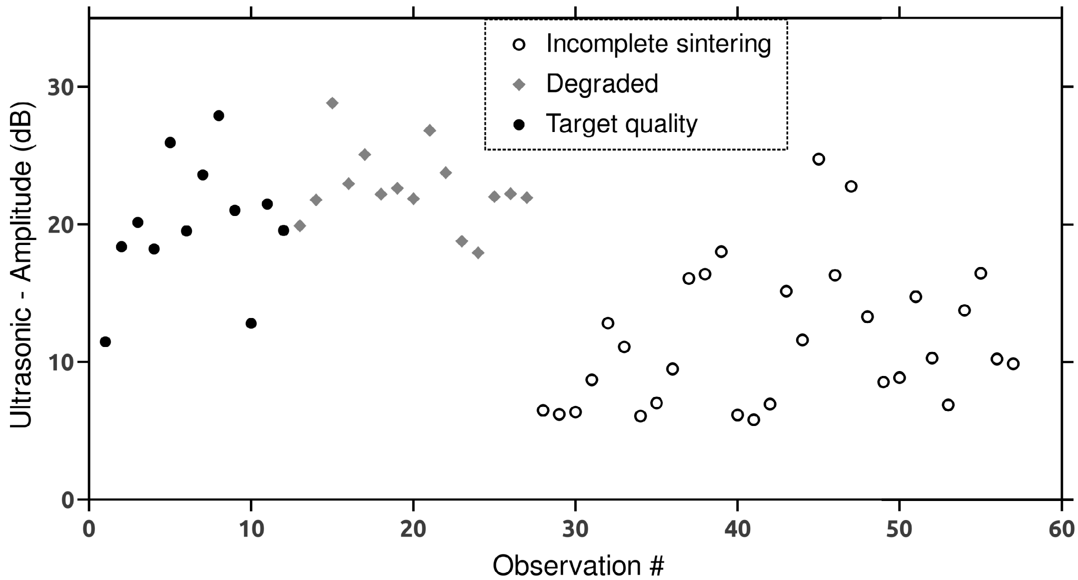


Figure 3: Maximum time-domain ultrasonic amplitude of rotational molded polyethylene samples (symbols indicate different quality groups defined based on destructive tests)

Using the same samples that were classified into the three categories previously mentioned, Figure 4 shows a score map of the first two components of a PCA model constructed from the ultrasonic spectra from groups of molded samples. Of the available samples, 31 batch runs were used for training purposes. The data available to build the model included measurements from each batch horizontally aligned as 31 vectors with each line representing a frequency from the ultrasonic spectra with a total of 2500 from 0 to 1000 kHz. The number of components in the PCA model was determined such as a minimum of 90% of variance explained the ultrasonic spectra matrix data located after the inflection of a scree plot. A group of 7 samples was reserved for validation, not to be included in the calibration of the model. As can be seen from the projection, some clusters can be identified to help create the base groups, but did not elucidate all the differences between classes. Note that since the final class for each marked group was unknown, groups were numbered and not labeled. This first clustering using a general PCA model with the whole group of samples thus serve as the starting point for the classification. The SIMCA algorithm described in Section 3.2

was performed to create PCA projections from each individual group of samples (details on the performed calculation can be seen in Appendix A).

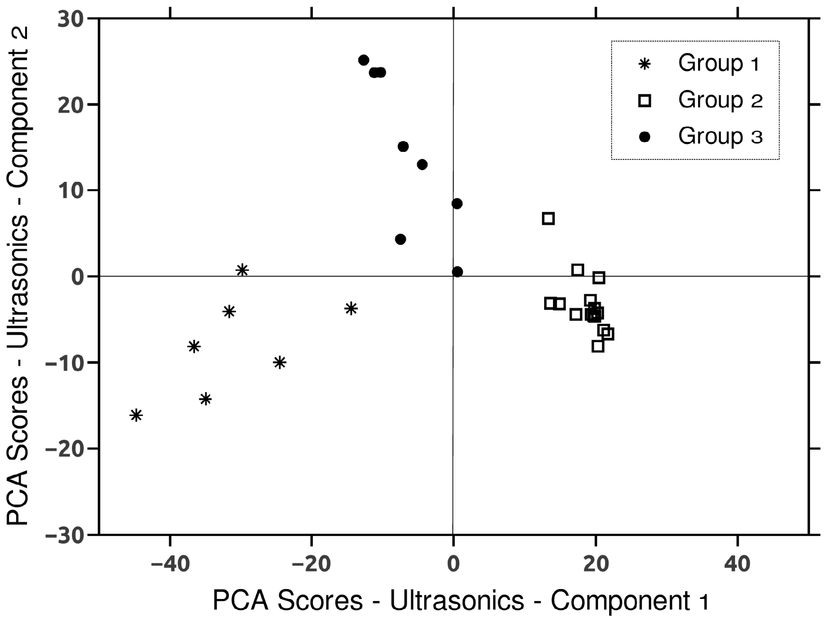


Figure 4: Projection of PCA scores from experimental batch samples using ultrasonic spectra data (different classes indicated by marker color and format)

Although the selection of the samples contained in each group was based on the scores of the ultrasonic PCA, the experimental validation (see Table 1) demonstrated that each class was mainly populated by samples of different quality groups (incomplete sintering, target and degraded). As a practical scenario, only a small number of samples from each group is necessary to be tested in order to determine a quality label. For the clusters observed in Figure 4, Group 1 correctly predicted most of the degraded samples with 5 samples being characterized as target, representing a type 2 error. Group 2 was populated with all parts characterized as incomplete sintering. And Group 3 had a combination of target (valid prediction) and degraded samples. These clear distinctions based only on ultrasonic spectra differences representd a significant evidence supporting the proposed in-line classification method. The approach did not require extensive calibration with destructive methods, but only one or two samples from the respective groups needed to be tested for the purpose of labeling.

Table 1: SIMCA groups label

	Experimental validation		
Group	Label	Correct	Type 2 error
1	Degraded	7	5
2	Incomplete sintering	14	0
3	Target	3	1
	Total	24	6

Another instructive analysis of each constructed PCA model was an evaluation of their loadings. Figure 5 shows the ultrasonic projection of the loadings considering all scores to be zero (center or average sample representation). Considering the region of the harmonic peaks, the amplitude at the primary frequencies (same as the generated pulses) was lowest in the case of incomplete sintering, and increased for the target and degraded cases. This amplitude variation was expected from the increase in density caused by a reduction in air bubbles. In the case of the third harmonic frequency range, there is also an increase in amplitude that follows the previous pattern between classes, however considering the amplitude ratio value, there is another distinction, with an increase from the target to degraded group. The variation in this nonlinear parameter, harmonics amplitude ratio, has been demonstrated as a sensitive indicator of morphological changes such as those related with thermo-oxidative degradation.[28] All of this analysis considered only the clusters of groups based on given class attributes and data from ultrasonic spectra. The loadings could also be used to identify significant frequencies related with quality features. For example, the observation of the amplitude signal around 450 kHz can be used for detection of degraded samples

Validation of the classification approach using the SIMCA methodology is demonstrated in Table 2. Classification used the value of SPE as a reference for classification into the labeled groups defined previously. All samples except one (Sample 6) were sucessefuly classified with compared experimental validation test. It is possible to argue that the classification covering Group 3 accomodated samples with target quality and with some level of degradation, but not representing the same structural change as observed in degraded samples classified

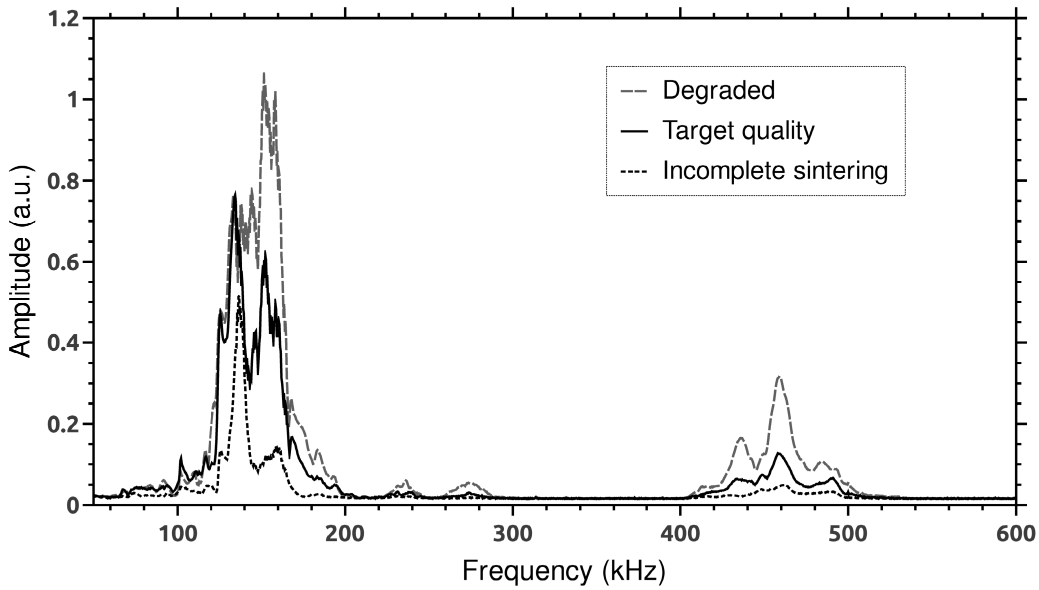


Figure 5: Ultrasonic spectra projected from loadings of PCA models of different quality groups

in Group 1. A reclassification of the experimental limits or the separation of Group 3 into two subgroups could be tried to improve classification. Recall that the traditional univariate descriptors simply did not allow any classification, and thus not attempt at validation was made. In contrast, the proposed approach was excellent in its ability to classify samples.

Table 2: SIMCA groups classification

	SPE Values				
Sample	Group 1	Group 2	Group 3	Classification	Experimental validation
1	2590	4663	**1779**	Target	Target
2	**2568**	19665	13618	Degraded	Degraded
3	2877	**1932**	2486	Inc. Sintering	Inc. Sintering
4	2911	**2044**	2614	Inc. Sintering	Inc. Sintering
5	2670	**1567**	2386	Inc. Sintering	Inc. Sintering
6	3638	38187	**2062**	Target	Degraded
7	6351	40058	**3736**	Target	Target

4.2 On-line quality monitoring and prediction

4.2.1 Process modeling and projection validation

First step for on-line approach is the process modeling using subspace identification. Following the approach described in Section 3.3, a third order state-space model was created using subspace identification for the process data available from historical rotational molding batches. Data included measurements of the internal air mold and heaters duties at 10 seconds interval. The input variables (x-variables of the model) were the heaters duties values and the output variable modeled (y-variable) was the internal air mold temperature. In Figure 6, the fit using the dynamic model is shown for two representative batches.

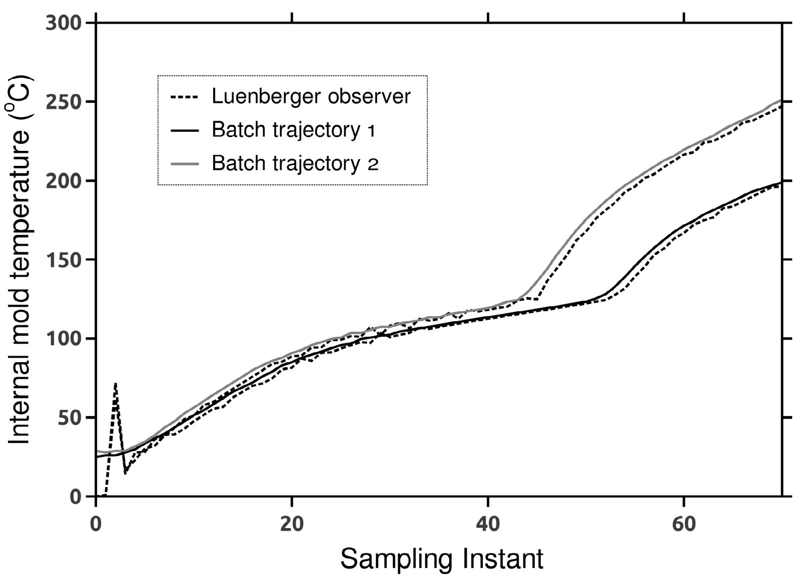
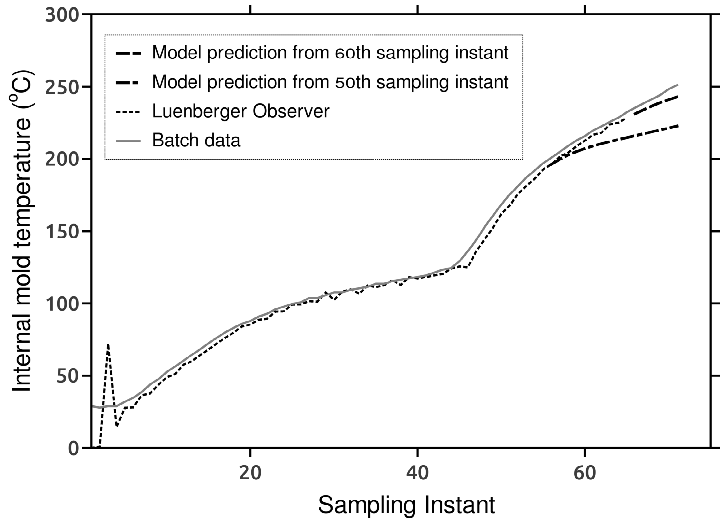
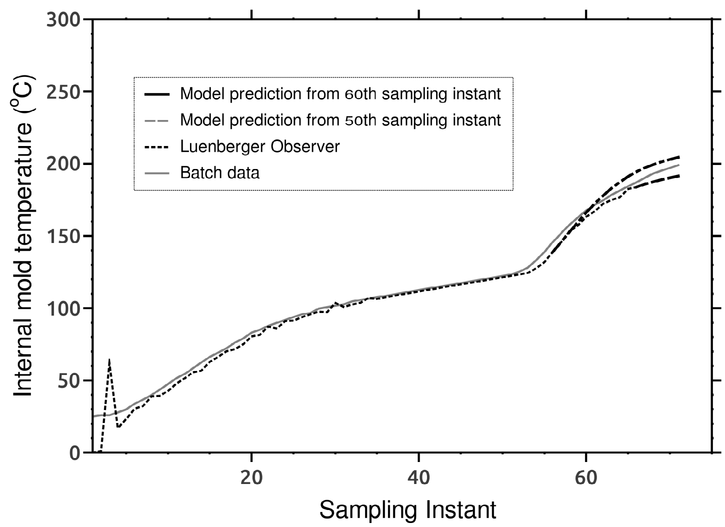


Figure 6: Batch internal air temperature profile for two validation batches until the instant of heating stage termination

Figure 7 shows the validation results using the identified model. In these results, the initial duration of a new batch is determined by a Lungberger observer to estimate the state of the subspace model (thus imbuing the modeling approach with learning characteristics). After the states have converged (corroborated via the convergence of estimated outputs to the measured outputs), the measured ouput trajectory was predicted for the remainder of the batch. The predictions were done starting from 3 different time points in the batch to demonstrate the improved ability of the model to predict process evolution behavior farther into the batch run.



(a) Batch 1



(b) Batch 2

Figure 7: Validation for dynamic model

4.2.2 On-line process visualization

Having illustrated the ability of the subspace model to capture the process dynamics, the objective of this section is to demonstrate the ability to predict the spectrum of the molded part. To this end, first, a model is built between the terminal subspace states, and the reduced ulstrasonic spectrum for the training batches (as described in Section 3.4). Thus, a multiple linear regression model was determined between the final state variables, containing 3 vectors with the size of the number of batches, and the ultrasonic spectra data in a reduced dimension after PCA, considering the scores of 8 components aligned with the size of the number of batches. A total of 21 batch runs (with measured process variables and ultrasonic spectra from the molded part) were used to calibration of the model. A group of 7 samples, that were not included on the original model calculation, were separated for validation.

The results for this proposed correlation showed excellent prediction capability. In particular, for validation purposes, two batches were chosen which corresponded to a incomplete sintering and degraded parts, respectively, exhibiting significantly different spectra. The model however, was able to predict very well the spectrum in each case utilizing the final subspace states alone, as can be seen in Figures 8 and 9. The ability to predict dynamic behavior (shown in section 4.2.1), along with the ability to predict terminal spectrum based on the terminal states built confidence in the possibility of using the dynamic model together with the multiple regression model for the purpose of on-line monitoring.

Figure 10 shows the trajectory of a batch run for the first two components of the ultrasonic spectrum based on the process trajectory with batch time. In this figure, the states at any given point in time are used to compute the terminal scores. As the process progressed, the states got closer to the terminal states, and thus the ability to predict the terminal PCA scores kept continually improving. In other words, with the progression of the sintering phase and increase in temperature during the heating phase, the process got closer to the finished product, and thus closer to the region of model validity. The apparent chaotic path presented by the process trajectory can be explained as being either a phenomena

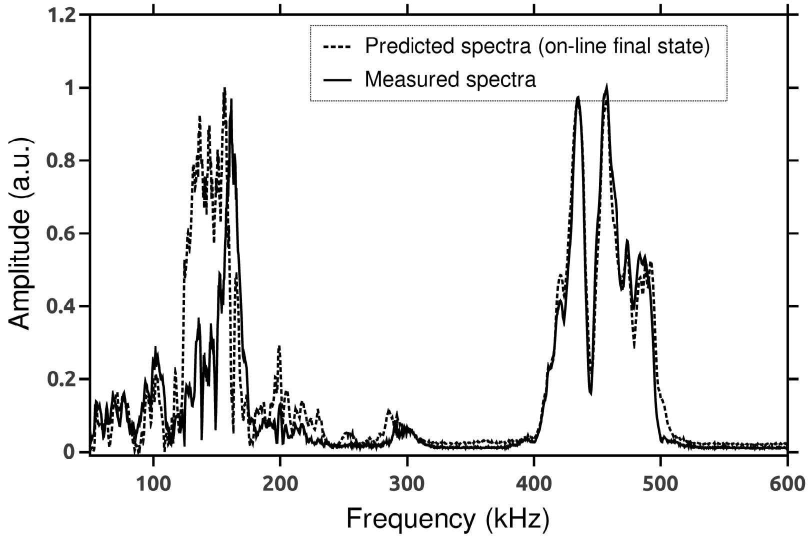


Figure 8: Experimental validation of the ultrasonic spectra projection from incomplete sintering sample

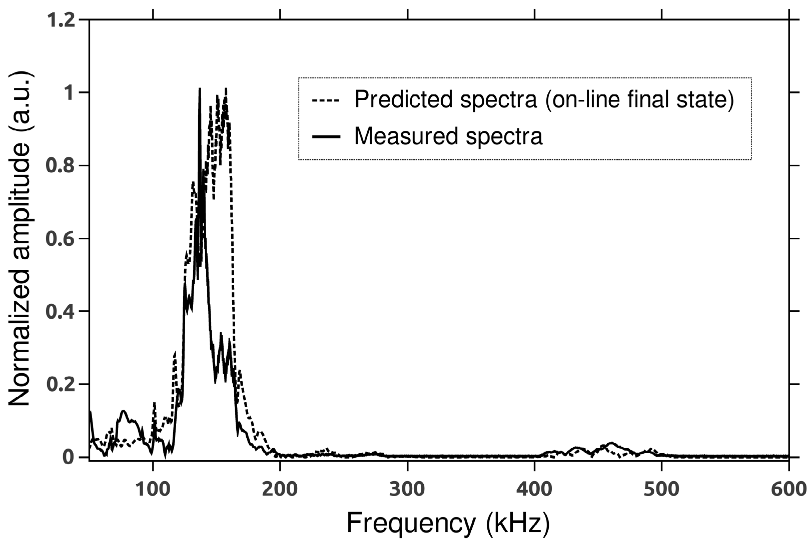


Figure 9: Experimental validation of the ultrasonic spectra projection from degraded sample

or a visualization limitation. As a phenomena, rotational molding has different processing phases, as described in Section 2.1, that can be registered as shifts in the process trajectory. And the projection represented in Figure 10 is a limited view of a multivariate system on a two-dimensional space. An ideal visualization would take in account more components but would be unpractical to preview. This on-line process monitoring combined measured process data with a projection of product quality, wich gave a useful visualization of the process state that goes beyond the collection of univariate measurements.

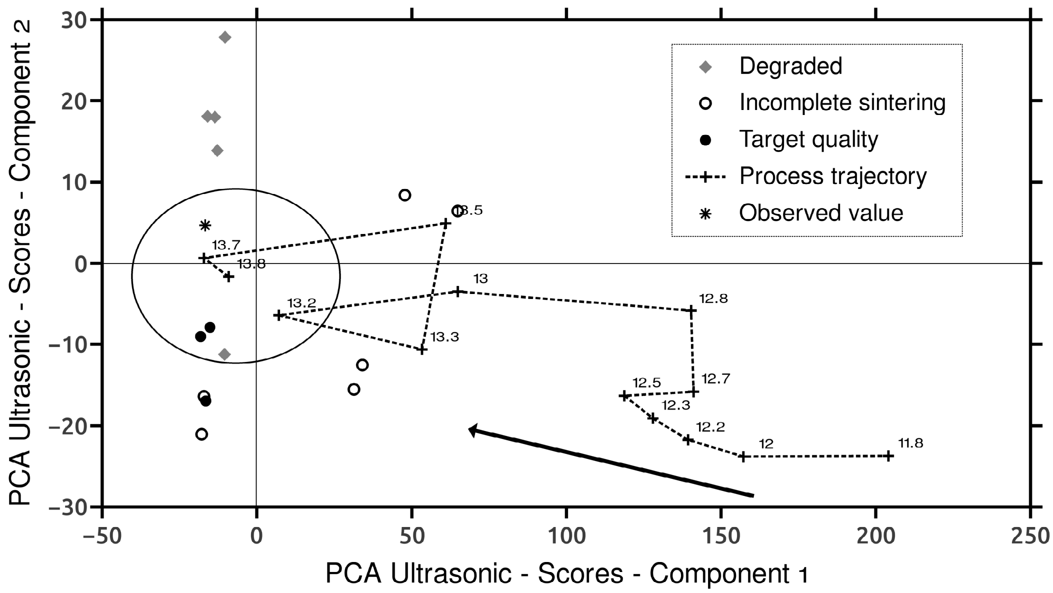
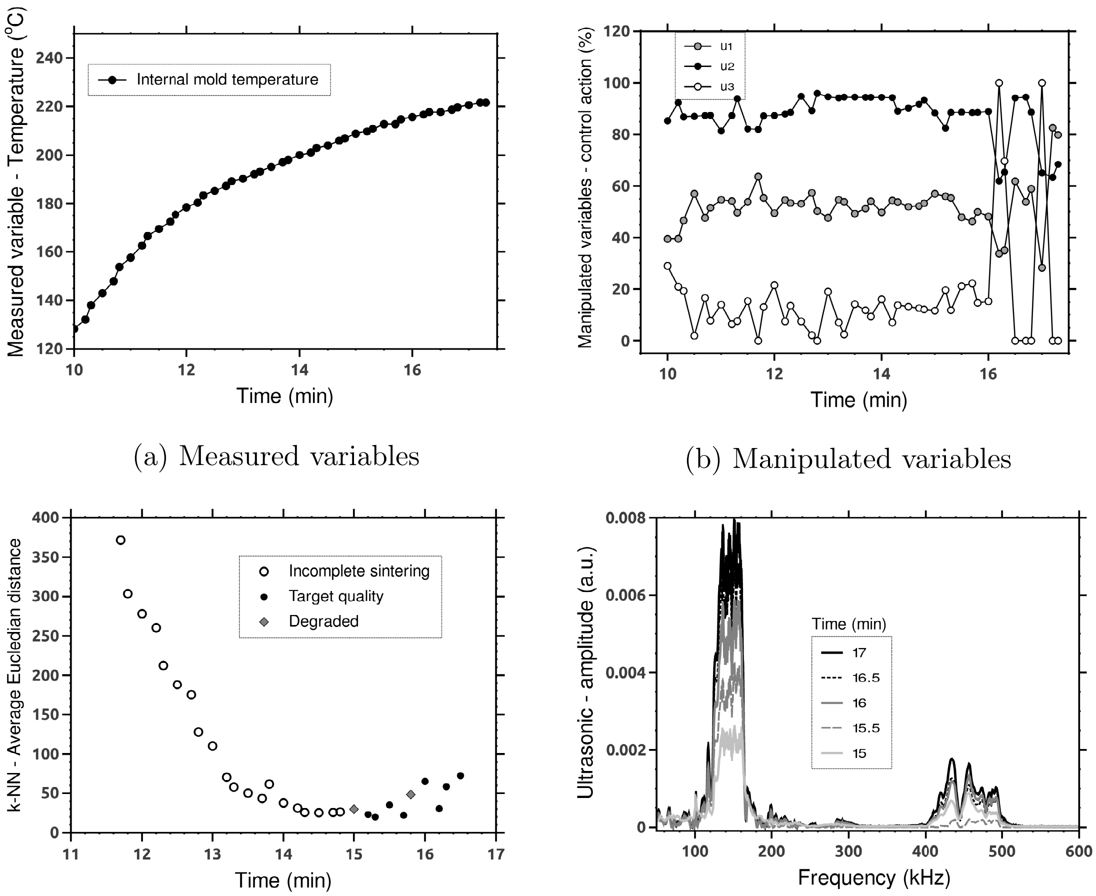


Figure 10: Process trajectory considering state-space variables at each sampling instant to project first two components of the reduced PCA ultrasonic spectra for a final degraded sample

The next set of figures show another visualization tool. In particular, a tool capturing the evolution of molding through the various phases as the batch run proceeded. To build the tool, first the classification method using the k-NN algorithm described in Section 3.4, with a k=3 was built. The value of the 'k' parameter can be decided based on the characteristics of the historical database. For the current proposed model, an odd number was selected to allow only a single prediction. The value also had to be smaller than the number of samples from the smallest quality group to predict, otherwise, no sample would be classified in that group. As the number of available batches can be increased this value could be optimized based on the lowest occurrence of prediction errors. The predicted ultrasonic spectra were converted from the reduced space to the frequency domain using the PCA loadings, and the absolute values of the amplitudes were used (to avoid meaningless negative values). Figure 11 showed the evolution of a particular batch. On the k-NN plot (top right corner), it is possible to see a gradual decrease in the calculated distance from the beginning of the sintering phase with a prediction of incomplete sintering quality, when it shifts temporarily to degraded quality, but then reaches target as a class prediction. The projected ultrasonic spectra from each sampling process instant shifts from incomplete sintering, with an increase in amplitude as the primary frequency increased, and a reduction of the amplitude ratio based on the third harmonic peak amplitude.

Note that there was no experimental validation of the spectrum predicted during this particular batch- i.e., the processing was not terminated at say 12 minutes into the batch. However, the training and, more importantly, validation samples did include batches that were terminated at different times into the batch, and that demonstrated the reasonableness of the tool to predict the final spectrum at different times during the batch. Furthermore, the model-based classification also matched with the experimental classification.

We next demonstrate the application of the proposed tool for detection of process conditions for a sample prepared outside of specifications. For this specific case, Figure 12 shows a different heating cycle with a higher heating rate that allowed the internal mold temperature to reach high values in a shorter time. The result was a shift in the classification by the k-NN search algorithm, from incomplete sintering to a degraded sample being projected after 11 minutes of batch run, at which time the internal temperature was already above 220 ^oC. The proposed monitoring tool also allowed for further process data interpretation beyond the final product quality classification. For the profile shown in Figure 12, it is notable that the heating conditions shifted the projections from incomplete sintering directly to degraded sample. In contrast to the example shown in Figure 11, the sample did not go through the target quality condition. This questions the traditional view for the rotational molding process that the quality depends only on either selection of the time to stop the heating cycle based on a fixed heating rate or based on the peak internal air temperature. In other words, a process run using the traditional understanding would not yield the target quality for either of these samples. It is understood that sintering and thermo-oxidative degradation



(a) Measured variables

(b) Manipulated variables

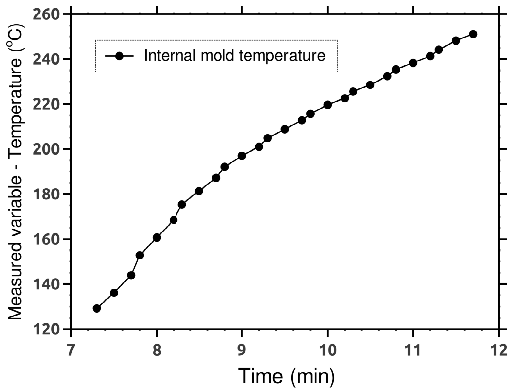
(c) k-NN classification

(d) Ultrasonic spectrum projection

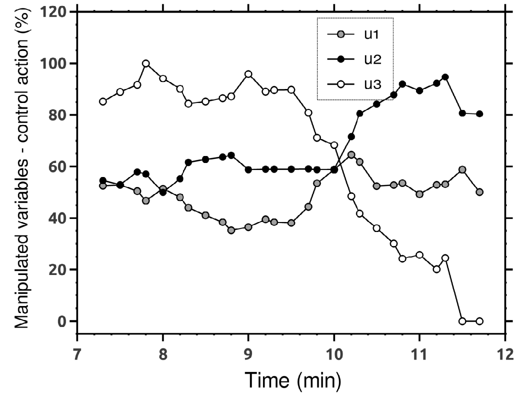
Figure 11: Process monitoring for a target quality sample

in rotational molding are parallel processes influenced by the combination of time and temperature.[36–38] Thus, it is understandable that some heating profiles can accelerate or reduce either of these two processes and might not be fully explained by monitoring of individual variables. In summary, the proposed multivariate monitoring approach combines sufficient process knowledge without the need of mechanistic determination to provide an estimation of the progression of the final quality during the batch run.

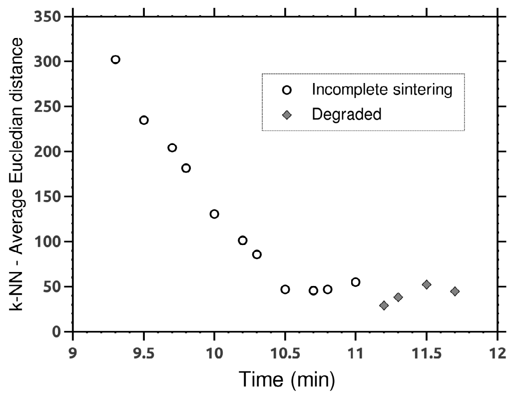
In order to demonstrate the adaptability of the proposed statistical tools for quality monitoring, Table 3 groups the predictions and experimental verification for samples of the validation group, with all product classes being accurately predicted. To the knowledge of the authors, there is no other data-driven on-line method capable of accurately predicting



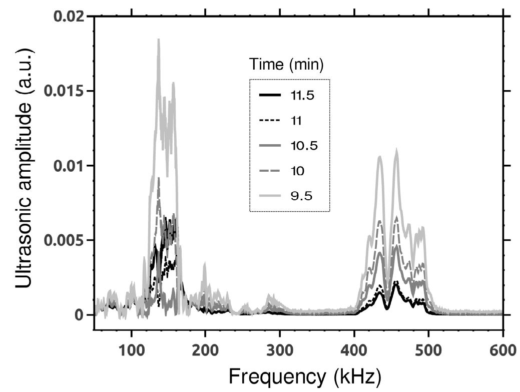
(a) Measured variables



(b) Manipulated variables



(c) k-NN classification



(d) Ultrasonic spectrum projection

Figure 12: Process monitoring for a degraded quality sample

both degradation and sintering quality for rotationally molded samples.

Variation between batches of different manufacturing units are expected due to the nature of heaters and oven designs that can be used. Considering the long term use of the same unit, the developed model can be used to detect significant deviations that might be cause to failures on heaters or sensors. A possible source of batch to batch variation and noise was the ultrasonic measurement with use of coupling agent. Recommendation for multiple points of measurement and crosschecking with different sensors can be done to reduce the introduction of noisy data.

Table 3: Results for classification prediction and experimental measurements for validation group

	On-Line Prediction		Experimental validation				
Sample	k-NN	Prediction	Impact	Incomplete sintering?	Viscosity	Degraded?	Observed
1	95	Target	0.50	No	8088	No	Target
2	234	Incomplete	0.31	Yes	6645	No	Incomplete
3	62	Target	0.43	No	7522	No	Target
4	264	Degraded	0.65	No	13418	Yes	Degraded
5	52	Target	0.51	No	6966	No	Target
6	45	Degraded	0.51	No	11365	Yes	Degraded
7	82	Incomplete	0.37	Yes	6447	No	Incomplete

5 Conclusions

Data-driven approaches based on a more intelligent manufacturing framework that uses multivariate data from nonlinear ultrasonics for in-line quality classification and an on-line monitoring tool were demonstrated and validated. Classification based on multivariate statistical analysis have shown efficiency even with minimal qualitative input, to confirm the validity of important structural properties contained on the multivariate ultrasonic spectrum tested from rotomolded polyethyelene parts. The proposed correlation between the termination state-space from the end of the heating phase and the reduced space ultrasonic spectrum was applied for the on-line visualization and quality prediction. The data-driven tools based on operational data and a nondestructive quality measurement described in this document have strong potential to be used not only for quality evaluation but also to improve process understanding. The integration of batch modeling and multivariate data projection for on-line quality prediction was the basis for development of new batch control strategies. Some practical challenges still need to be addressed for future improvement of the methods, such as the reduction of prediction error, control of data noise and a protocol for optimization with larger datasets. However, the presented work strongly support the use of nonlinear ultrasonics as a viable sensor technology for incorporation to advanced manufacturing practices for batch processing in polymer industry.

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