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Ultrasonic sensor for predicting sugar concentration using multivariate calibration

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ABSTRACT

This paper presents a multivariate regression method for the prediction of maltose concentration in aqueous solutions. For this purpose, time and frequency domain of ultrasonic signals are analyzed. It is shown, that the prediction of concentration at different temperatures is possible by using several multivariate regression models for individual temperature points. Combining these models by a linear approximation of each coefficient over temperature results in a unified solution, which takes temperature effects into account. The benefit of the proposed method is the low processing time required for analyzing online signals as well as the non-invasive sensor setup which can be used in pipelines. Also the ultrasonic signal sections used in the presented investigation were extracted out of buffer reflections which remain primarily unaffected by bubble and particle interferences.

Model calibration was performed in order to investigate the feasibility of online monitoring in fermentation processes. The temperature range investigated was from 10 °C to 21 °C. This range fits to fermentation processes used in the brewing industry. This paper describes the processing of ultrasonic signals for regression, the model evaluation as well as the input variable selection. The statistical approach used for creating the final prediction solution was partial least squares (PLS) regression validated by cross validation. The overall minimum root mean squared error achieved was 0.64 g/100 g.

1. Introduction

In many biotechnological processes it is of great interest to detect the concentration of ingredients in the working medium. Further, monitoring and control of such industrial processes need reliable online measuring devices. Those sensor systems should have the ability to be robust, easy to use and non-invasive, even when it comes to food related applications. Typically, the sensor equipment used for analyzing fluid properties is invasive to the medium of interest [1–4]. Over the last few decades the importance of ultrasonic sensors for such applications have become more and more standard [5,6]. Several groups have studied the possibility of ultrasonic devices for measuring sugar concentration in various different ways [7,8]. Further, frequency and time domain representation of ultrasonic features is reported to contain information about the changes in density of respective fluids. In literature there are several possibilities presented which use indirect prediction of acoustic impedance via reflection coefficient combined with ultrasonic velocity estimated from time of flight measurement to finally calculate the density of the fluid [9–14]. However, relevant information in relation to changes in sugar concentration and therefore density changes should be visible in acoustic impedance. Extracting this feature based on the known physical relations with the presented setup is quite difficult. Nevertheless, one possibility to extract the feature impedance is based on the decay of temporal echo amplitudes in the buffer material [10]. Other possibilities presented in literature are based on the frequency domain. Further, influences like superposition as well as signal resolution do have a high impact on those approaches. Despite all that, literature reports difficulties in predicting impedance accurately under the given circumstances [15,16]. In this work, the information was gathered using several acoustic features calculated on time and frequency domain representation. These were extracted on buffer reflections of ultrasonic signals. Therefore, influences caused by i.e. gas bubbles can be avoided by only analyzing the signal reflection. Although those signal parts do not penetrate the medium, they still carry medium information in the reflection coefficient. Generally, the chosen features individually capture the information included in each signal. This results in a new feature based multivariate representation covering signal attenuation as well.

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Nomenclature

a	regression parameters for temperature dependence (–)
$\boldsymbol{B}$, $\boldsymbol{b}$	matrix/vector of regression parameters (–)
b_0	first regression parameter (g/100g)
BW	bandwidth (–)
cen	centroid (–)
cf	crest factor (–)
EDFT	extended discrete Fourier transform
eng	energy (–)
ent	entropy (–)
f_s	sampling rate (Hz)
h	leverage of sample (–)
k	number of iterations extracting the latent vectors
kur	kurtosis (–)
m	number of variables (–)
mag	magnitude (–)
MLR	multiple linear regression
n	number of samples (–)
N	number of data points in ultrasonic signal sequence (–)
NIPALS	nonlinear iterative partial least squares
$\boldsymbol{p}$, $\boldsymbol{P}$	loading vector/matrix of X
PMMA	poly(methyl methacrylate)
PVDF	polyvinylidene flouride
PC	principal component
PCA	principal component analysis
PLS(R)	partial least squares (regression)
$\boldsymbol{q}$, $\boldsymbol{Q}$	loading vector/matrix of Y
RMSECV	root mean squared error of cross-validation (g/100g)
$\boldsymbol{S}$	diagonal matrix with standard deviation
s^2	sample variance
ske	skewness (–)
spr	spread (–)
T	temperature (°C K)
$t(A_i)$	reflection of ultrasonic pulse
$\boldsymbol{t}$, $\boldsymbol{T}$	score vector/matrix of $\boldsymbol{X}$
$\boldsymbol{u}$, $\boldsymbol{U}$	score vector/matrix of $\boldsymbol{Y}$
VIP	variable importance in the projection
$\boldsymbol{w}$, $\boldsymbol{W}$	weighted loading vector/matrix
$\boldsymbol{X}$	predictor matrix
$\boldsymbol{Y}$	target matrix

Subscripts, exponents

^	"predicted"
a,i,j	counter
s	scaled
s	spectral
t	temporal

Analyzing material properties as control variables for industrial applications based on physical modelling is not always possible due to the lack of knowledge. This leads to the use of multivariate statistics. These methods have been used for years in several fields when dealing with large volumes of data [17]. For evaluating data based on its statistical variance, multivariate regression models, such as principal component regression (PCR) or PLS, are used to handle the large amount of mostly collinear variables. These methods are used for correlation of target values ($\boldsymbol{Y}$) with direct measurable descriptor variables ($\boldsymbol{X}$). The background of such approaches is multiple linear regression (MLR). In this contribution partial least squares regression (PLSR) is used for modelling the variations in ultrasonic signals transmitted through aqueous solutions with varying maltose concentrations. This method uses a reduced number of latent variables compared to the descriptor variables found by cross correlation of variance in $\boldsymbol{Y}$ and $\boldsymbol{X}$.

It was shown earlier, that changing concentration of a dissolved substance causes changes in the fluid properties such as density and bulk modulus. This directly influences the properties of the ultrasonic waves travelling through the fluid [18]. It was reported, that features of ultrasonic signals like reflection coefficient is frequency dependent [18]. Developing a physical relationship between pulse distortion and the fluid properties is quite complicated due to the complexity of the system. Because of this the usage of multivariate data analysis is of benefit in cases of fast modelling of the phenomena of interest [18]. With the aid of multivariate analysis, it is possible to extract the most dominant information with respect to density. At the same time, the noise caused by arbitrary influences such as temperature inaccuracies, superposition, and bubble induced distortion will be discarded. This is the goal of the presented study. It was shown earlier in a similar approach using PLS, that it is possible to predict substance concentrations using ultrasonic signal features [18].

This study presents a system, which is fully non-invasive. Further, it is less dependent on influences caused by bubble interferences or particles suspended in the medium of interest. A signal with corresponding wavelength travelling through the fluid could lose energy caused by scattering or dissipation at these bubbles or particles [19]. Up till now, the calibration covers a defined temperature range and is capable of detecting sugar concentration. The method is based on feature extraction of ultrasonic pulses and multivariate regression based on PLS. Up to the author's knowledge, the combination of both parts is new in the field of ultrasonic measurements. Finally, the method is simple and easy to implement. It is also possible, to extend the detection on ternary mixtures containing ethanol as well. Benefit of this extension would be the possibility to use just on sensor device as well as uncoupling the detection from any relation to the process behavior like in Resa et al. or Cha and Hitzmann [20–22].

2. Materials and methods

2.1. Experimental

The experimental setup as well as the measuring principle is explained schematically in Fig. 1 including photography of the setup (middle). The shown container is indirectly heated over a tempering mantle which is supplied with tempering fluid by an external thermostat. To investigate maltose-water mixtures the container is first filled with the solution of interest. Additionally, the solution is heated/cooled continuously as well as permanently mixed by a magnetic stirrer to reach a temperature distribution as homogeneous as possible. The ultrasonic signals are recorded over a temperature range from 10 up to 21 °C at constant container pressure over a time frame of around 3 h. The temperature points used in this study for calibration were extracted in steps of 0.5 K out of this continuous spectrum.

The in-house produced piezo electric transducer (built using a piezo ceramic with a center frequency of 2 MHz) is used for creating an ultrasonic pulse by excitation with a rectangular electrical pulse (width of 200 ns, amplitude of 5 V). After passing container wall (wall 1) and fluid the pulse is reflected at the backside (wall 2) and caught by the transducer which works as a receiver in the