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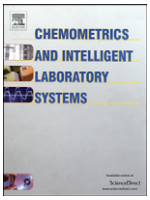

The effect of flow rate in acoustic chemometrics on liquid flow: Transfer of calibration models

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ABSTRACT

Previous studies on acoustic chemometrics on liquid flow have demonstrated that flow rate, accelerometer location and temperature affect the passive acoustic spectra and prediction results. Changes in the flow rate result in spectral variations, causing the resultant local calibration model to perform poorly predicting new samples measured at other flow rates. Developing good and robust calibration models can be done using several approaches. Global calibration methods were discussed in a previous study on acoustic chemometrics on liquid flow. In the present paper preprocessing techniques such as multiplicative scatter correction (MSC) and piecewise multiplicative scatter correction (PMSC), and variable selection methods such as interval partial least squares (iPLS) regression and powered partial least squares (PPLS) regression are studied. The preprocessing and variable selection methods were validated on two experimental data sets from passive acoustic measurements of liquid flow through an orifice plate. Acoustic spectra were registered at four different accelerometer locations. The liquids were two-component mixtures of sucrose and water, and three-component mixtures of ethanol, sucrose and water. MSC resulted in the improvement of model performance predicting new (preprocessed) samples measured at other flow rates. Sucrose prediction in two-component mixtures and ethanol prediction in three-component mixtures were improved in terms of bias and correlation coefficients respectively. Absolute bias values for sucrose prediction were in the range of 0.84–2.57 wt.% for spectra preprocessed by MSC compared to 1.17–22.38 wt.% for the uncorrected spectra using an accelerometer located at the orifice plate and the highest of studied flow rates as calibration flow. Correlation coefficients for prediction of ethanol were in the range of 0.80–0.97 for MSC spectra compared to 0.76–0.97 for the uncorrected spectra using an accelerometer located at the orifice plate and the highest of studied flow rates as calibration flow. Limited systematic improvement was observed for the sucrose and water prediction in three-component mixtures. PMSC slightly improved sucrose and ethanol prediction in the three-component mixture compared to MSC. iPLS regression indicated some intervals in acoustic spectra which were less affected by flow rate fluctuations. Regression using these intervals instead of full acoustic spectra resulted in lower prediction errors for sucrose, ethanol and water prediction in three-component mixtures compared to full spectra models. PPLS regression on frequency (peak position) matrix derived from full acoustic spectra did not determine any peaks robust to flow rate fluctuations. Effect of the flow rate on positions of the peaks important for chemical composition was difficult to establish. However, a shift to lower frequency with increasing flow rate could be observed.

1. Introduction

Acoustic chemometrics on liquid flow have been reported as a promising measurement method having several potential industrial applications [1–4]. The method is based on registering vibrations from the liquid flow in the pipe using “clamp-on” accelerometers [2,5]. Acoustic emission spectra are then analyzed using multivariate regression.

Previous studies on acoustic chemometrics on liquid flow [2,6] have demonstrated that flow rate, accelerometer location and temperature affect the passive acoustic spectra and prediction results. Changes in the flow rate and, to a lesser extent, temperature result in amplitude change and frequency shift in spectra, causing the local calibration model to perform poorly predicting new samples measured at other flow rate and temperature [6].

Developing good and robust calibration models can be done using different approaches. The ideal approach is to minimize fluctuations in the process, but in some cases this possibility is limited. Other well known methods are model updating by use of new measurements (global calibration), spectral preprocessing, and spectral transfer

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methods. A global calibration approach, including samples measured at various flow rates into the model, has been already discussed in a previous paper [6]. Global calibration models for each accelerometer location were made by inclusion of all samples from the three levels of temperature and flow in the single model. Sucrose in the two-component mixture, and ethanol, sucrose and water in the three-component mixture could be well predicted at all flow levels and all accelerometer location by the global models. However, the models required 8 to 10 PLS factors. It seemed that changes in the flow rate caused multiplicative changes in the overall amplitude of the acoustic spectra. Also some peak shifts and peak transformations as result of flow variations were observed. Preprocessing of the spectra can probably deal with the observed flow induced changes in amplitude. The aim of the present paper is to develop robust regression models by preproccesing of the acoustic spectra and variable selection.

1.1. Spectral preprocessing and spectral transfer methods

Typically spectral preprocessing techniques are used to handle linear spectral effects like light scattering and similar. In acoustic chemometrics on liquid flow the nature of physical effects on spectra is different from light spectroscopy. However, changes in acoustic spectra can be similar to the changes in electromagnetic spectra in terms of amplitude and frequency. It might be possible to implement some preprocessing and spectral transfer methods used in light spectroscopy on acoustic spectra.

Methods for spectral preprocessing can be divided in two groups [7]:

1. Techniques that require no prior information related to the reference measurements e.g. first and second derivatives, multiplicative scatter correction (MSC) [8], extended multiplicative scatter correction (EMSC) [9], piecewise multiplicative scatter correction (PMSC) [10] and finite impulse response (FIR) [11].
2. Techniques that take into account at least one of the dependent variables. Orthogonal signal correction (OSC) [12] is one of such methods.

Spectral transfer methods include such techniques as direct standardization (DS) [13] and piecewise direct standardization (PDS) [13,14]. However, in industrial application the spectral transfer methods are not always suitable since measurements of standard samples under both primary and secondary conditions are required. In the real world processes no such standard samples would be available. In addition the process parameters change randomly and continuously making the application of spectral transfer methods difficult or impossible. The present study focuses on spectral preprocessing methods which are more relevant for the real world process application.

1.2. Variable selection techniques

There is no prior knowledge for the most relevant peaks in the acoustic spectra, whose shifts follow the expected trends and reflect the changes in the fluid system giving the best prediction results. Therefore a variable selection may be necessary to apply. Powered partial least squares (PPLS) regression [15] and jack-knife based method for variable selection [16] has been successfully applied in the reported studies [4,5].

Changes in flow rate and temperature cause amplitude and frequency changes in the acoustic spectra [2,6]. However, it might be possible that some parts of the spectra are robust to flow and temperature fluctuations. PPLS regression and jack-knife based method are not optimal to apply in this case, because these methods select single variables, not segments of the spectra. Interval PLS (iPLS) regression [17] can probably be applied to investigate the robustness of parts of the spectra to the changes in flow rate and temperature.

There are numerous studies reporting the application of the preprocessing techniques as MSC and PMSC, and variable selection methods in light spectroscopy. However, we could not find any reports discussing spectral preprocessing on acoustic spectra. The aim of this paper was to study if preprocessing techniques and variable selection methods could improve the performance of local models to predict new samples measured at other flow rate than calibrated for. Temperature effect is not discussed in the present paper because of its minor influence on spectra and prediction results [6].

2. Materials and methods

2.1. Experimental design

The study was performed on two experimental data sets containing passive acoustic measurements of designed two-component sucrose–water (data set 1) and three-component ethanol–sucrose–water (data set 2) mixtures. The same data were analyzed in the previous paper [6].

2.1.1. Two-component mixtures (data set 1)

Sucrose (Danisco Sugar, Copenhagen, Denmark) and tap water were mixed obtaining 17 samples with the following sucrose content: 0, 2, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 18 and 20 wt.%, and giving a standard deviation $SD_y = 5.48$ wt.% The standard deviation here describes dispersion of samples in the experiment.

2.1.2. Three-component mixtures (data set 2)

Ethanol (denatured, Arcus Kjemi AS, Vestby, Norway), sucrose (Danisco Sugar, Copenhagen, Denmark) and tap water were mixed according to a simplex lattice design. Sucrose and ethanol content were varied in 2.5 wt.% intervals giving 45 samples and standard deviation $SD_y = 5.53$ wt.% The ethanol and sucrose added up to a total of 20 wt.% for each sample.

2.2. Acoustic measurements

Acoustic vibration was measured using accelerometers (Model 7259B-10 and Model 7259B-100, Endevco Corporation, San Juan Capistrano, USA) placed at four different locations on a pipe. Locations 1 and 2, both at the orifice plate, were chosen as suggested in the original study on acoustic chemometrics on liquid flow [2]. The accelerometer at location 2 was Model 7259B-100 which was ten times more sensitive compared to accelerometers at other locations (Model 7259B-10). Accelerometer 3 was located at the assumed strongest turbulence region downstream the orifice. Accelerometer 4 was placed at the flow region 140 mm (6 pipe diameters) downstream the orifice where the liquid flow is less turbulent compared to the flow at the orifice. For more details and illustration of the instrumental set up please refer to the previous paper [6]. The registered vibration signal was filtered by high-pass (3000 Hz) and low-pass (60,000 Hz) filters. Signal conditioning, recording of time series and Fast Fourier transform (FFT) of the time signal were done by a Process Signature Analyser (PSA-100i, Telemark University College, Porsgrunn, Norway). The acoustic spectra for each sample were obtained as an average of 1000 Fourier transformed time domain signals.

The passive acoustic measurements for the two-component sucrose–water mixtures (data set 1) were performed according to a full factorial design with the following factors:

- Flow rate (3 levels): 1.4×10^{-4}, 1.5×10^{-4} and 1.6×10^{-4} $m^3 s^{-1}$, corresponding to a pressure drop of 1.5, 1.7, and 2.0 bar or a flow velocity of 21.2, 22.5 and 24.0 ms^{-1} through the orifice
- Temperature (3 levels): 25.0, 30.0 and 35.0 °C
- Accelerometer location (4 locations)

Each design point was represented by two-component mixture design made in 2 fully independent replicates.