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Calibration model maintenance in melamine resin production: Integrating drift detection, smart sample selection and model adaptation

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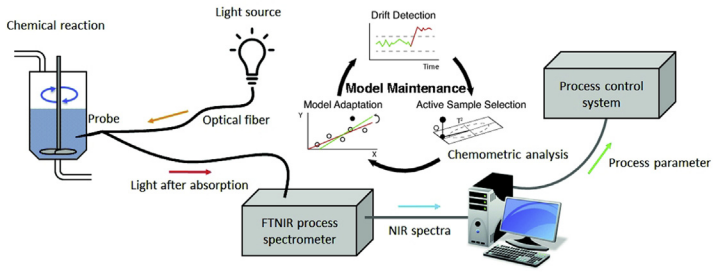
HIGHLIGHTS

- High-performance prediction of turbidity point in melamine resin production based on FTNIR spectra; cost savings of manual measurements.
- Drift detection based on committee disagreement over ensemble PLS models (significant outperformance of SoA methods).
- Weighted supervised model adaptation based on actively selected samples.
- Unsupervised model adaptation based on antecedent/consequent decomposition of calibration models.
- Significant reduction of model errors (produced by static calibration models) with economic model adaptation schemes.

GRAPHICAL ABSTRACT



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ABSTRACT

The physico-chemical properties of Melamine Formaldehyde (MF) based thermosets are largely influenced by the degree of polymerization (DP) in the underlying resin. On-line supervision of the turbidity point by means of vibrational spectroscopy has recently emerged as a promising technique to monitor the DP of MF resins. However, spectroscopic determination of the DP relies on chemometric models, which are usually sensitive to *drifts* caused by instrumental and/or sample-associated changes occurring over time. In order to detect the time point when *drifts* start causing prediction bias, we here explore a universal drift detector based on a faded version of the Page-Hinkley (PH) statistic, which we test in three data streams from an industrial MF resin production process. We employ *committee disagreement* (CD),

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computed as the variance of model predictions from an ensemble of partial least squares (PLS) models, as a measure for sample-wise prediction uncertainty and use the PH statistic to detect changes in this quantity. We further explore supervised and unsupervised strategies for (semi-)automatic model adaptation upon detection of a drift. For the former, manual reference measurements are requested whenever statistical thresholds on Hotelling's T^2 and/or Q-Residuals are violated. Models are subsequently re-calibrated using weighted partial least squares in order to increase the influence of newer samples, which increases the flexibility when adapting to new (drifted) states. Unsupervised model adaptation is carried out exploiting the dual antecedent-consequent structure of a recently developed fuzzy systems variant of PLS termed FLEXFIS-PLS. In particular, antecedent parts are updated while maintaining the internal structure of the local linear predictors (i.e. the consequents). We found improved drift detection capability of the CD compared to Hotelling's T^2 and Q-Residuals when used in combination with the proposed PH test. Furthermore, we found that active selection of samples by active learning (AL) used for subsequent model adaptation is advantageous compared to passive (random) selection in case that a drift leads to persistent prediction bias allowing more rapid adaptation at lower reference measurement rates. Fully unsupervised adaptation using FLEXFIS-PLS could improve predictive accuracy significantly for light drifts but was not able to fully compensate for prediction bias in case of significant lack of fit w.r.t. the latent variable space.

1. Introduction

1.1. Motivation and state-of-the-art

Melamine formaldehyde (MF) is an important class of thermosetting polymers, which have found wide industrial application as coating and adhesive materials owing to their high thermal- and chemical stability and physical resistance [1]. The properties of MF are largely determined by the curing behavior and the degree of polymerization and cross-linking of the final product [2]. It is therefore of pivotal interest to supervise the degree of polymerization (DP) during MF resin production and to determine the optimal time point when the polymerization reaction should be stopped. The water tolerance method and determination of free melamine concentration by liquid chromatography (LC) are well established and widely used to assess the DP of MF resins but produce significant cost in terms of material and human resources [3,4]. In addition, the delay between sample collection and retrieval of the critical process information harbors the risk of missing the optimal time point when the reaction has reached the desired DP [5]. In practice, MF condensation processes are therefore often run below the theoretically possible speed in order to minimize this risk at the expense of suboptimal allocation of resources.

Non-destructive estimation of free melamine content in MF resins by vibrational spectroscopy has been demonstrated previously and holds promise for high-frequency in-line monitoring of MF condensation [3]. Pawlicek et al. have recently established an online monitoring and control system for MF resins based on Fourier-transform near infrared (FT-NIR) spectroscopy and demonstrated the feasibility of accurate spectroscopic estimation of the turbidity point during the condensation process [4]. However, it is widely appreciated that chemometric models, usually required for spectroscopic estimation of critical process parameters, often have a limited time-span during which they deliver reliable predictions on new samples (e.g. spectra) and thus need regular supervision and maintenance [6]. The breakdown of model reliability might occur either due to instrumental changes, changes in environmental conditions during spectra acquisition or changes in sample composition (e.g. due to changing raw material composition) [7]. In any case, the relation between FT-NIR spectra and the target values eventually gets altered by such *drifts*, which might ultimately lead to prediction bias.

Maintenance of chemometric models can be roughly divided in two tasks: i) Detection of the time point when any drift starts causing prediction bias (i.e. drift detection) and ii) model adaptation once a drift has been detected. Timely detection of drifts is crucial to avoid cost-intensive manufacturing errors, defective production or even complete shutdown of the process [8]. Re-calibration of chemometric models according to a static schedule using regularly collected reference measurements could eventually compensate for the occurrence of drifts to some extent, but requires time-intensive modelling/validation cycles and disproportional efforts in terms of reference analytics. We have therefore recently proposed a self-adaptive calibration modelling strategy employing non-linear fuzzy PLS models in order to automate off-line re-calibration and validation [5]. However, despite implicit drift handling [9], full supervised model adaptation employed in Refs. [5,10,11] require a high number of reference measurements and is thus of little practical relevance. In an attempt to reduce the amount of reference measurements Cernuda et al. [12] proposed an active learning (AL) based approach to request (manual) reference measurements for new samples when the corresponding spectra violate upper control limits for Hotelling's T^2 statistic or Q-residuals. Therein, the authors employed a sliding window approach featuring user-defined thresholding of the rate at which reference measurement should be conducted. However, despite dramatic reduction of reference measurements, the method performs permanent sample selection and model re-calibration which may lead to superfluous reference measurements. In addition, employing a fixed window size might be problematic as large windows typically lack the ability to keep pace with the dynamics induced by the drift while important sources of process variability might be disregarded when using small ones. Other works on self-adaptive (evolving) chemometric models have been proposed in Refs. [10,11], but also require the full knowledge of target measurements and are thus not applicable in our context. Active learning for off-line chemometric calibration has been proposed in Refs. [13,14]. However, these approaches are not directly applicable for online model adaptation. For this reason, we here combine active learning along with explicit drift detection: Initially trained chemometric models are updated according to a complete on-line, single-pass active learning concept only once a drift has been detected. In addition, we also explore unsupervised model adaptation exploiting the dual antecedent-consequent structure of the recently proposed fuzzy systems variant of PLS (FLEXFIS-PLS) in order to reduce prediction bias.

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