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Development of a State Estimation Environment for the Optimal Control of a Mini-plant for the Hydroformylation in Microemulsions

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A state estimation framework for a surfactant containing multiphase process for the hydroformylation of long-chained alkenes is presented. Firstly, available state estimation methods, such as the extended Kalman filter, the unscented Kalman filter and the particle filter are compared regarding their usability in processes with high model and measurement uncertainty. Subsequently, an MHE-based state estimation algorithm is introduced. This includes an approach, which handles the occurring multi-rate measurements by dividing the state estimation into two separate steps. Finally, the implementation is discussed regarding necessary requirements and the state estimation framework is applied within long-term real process operation in a mini-plant.

1. Introduction

Within the chemical industry the usage of renewable raw materials is gaining increased attention. To enable new synthesis paths using a biological feedstock, novel chemical processes using tuneable liquid multiphase systems are being developed within the collaborative research center Transregio 63. One of the process concepts is the homogeneously catalysed hydroformylation of long-chain olefines in microemulsion systems. Aiming for a fast process development and the evaluation of the concept, a mini-plant has been built at Technische Universität Berlin alongside first lab-scale investigations. In Figure 1a, the simplified process concept for the hydroformylation in microemulsions is shown. The hydroformylation reaction is run there by applying a cost-intensive rhodium-based catalyst, which shows an excellent reaction performance regarding activity and selectivity. To guarantee economic viability of the considered process, the mentioned catalyst has to be recycled efficiently. This is achieved by immobilizing it in an aqueous phase via ligand modification and by taking advantage of the distinct thermomorphic phase separation behaviour of microemulsion systems. The separation takes place in a gravity settler, developing a catalyst free product phase, a catalyst rich emulsion, and an aqueous phase. For a continuous process concept, the upper product phase is drawn off, whereas the two lower phases are recycled back to the reactor (Illner et al., 2016).
However, the operation region for this highly complex separation step is rather small and nonlinearly shifts with present disturbances. Thus, the controllability of the process is impeded. To ensure a stable operation the usage of an advanced control strategy based on dynamic real-time optimization (D-RTO) is strived for (Müller et al., 2017). This implies the availability of valid initial values for the state variables for the existing mini-plant model to achieve successful convergence of each optimization step and robustness of this approach. For this purpose, the usage of a state estimation step (SE) in interaction with the D-RTO is proposed. The integration of a SE in a D-RTO framework is show in Figure 2b. The SE (inside the box) is divided into a filtering and a simulation step. To guarantee a stable SE for the mentioned mini-plant operation, a filter, which is capable of handling a large process model, gross error, and multi-rate measurements, is required. The choice of an appropriate SE method to handle the mentioned requirements is carried out in the following.

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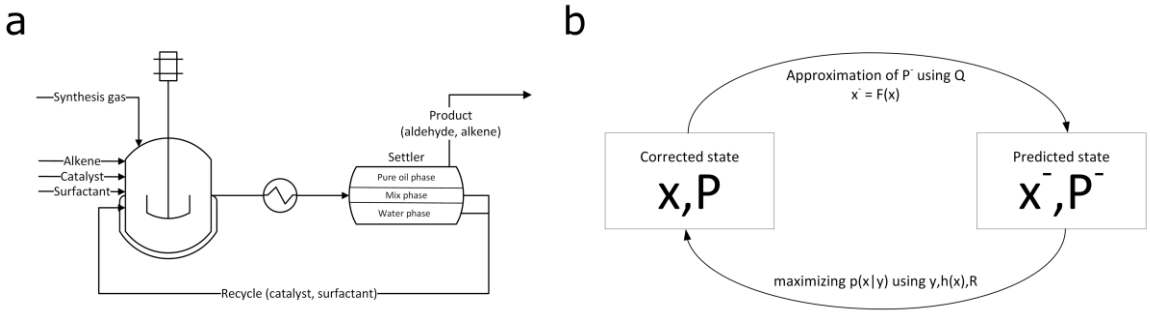

Figure 1: Simplified process concept of the hydroformylation miniplant (left, (a)) and schematic illustration of a predictor-corrector state estimation (right, (b)).

2. Theoretical Background of State Estimation

The aim of state estimation is to determine a consistent set of state variables for a system, usually for the usage as initial values in a D-RTO. In many cases, these variables are not directly measured in the process. For this reason, SE uses a mathematical model of the system, which connects the measured variables with all state variables. In general, SE is using a time-discrete process model of the form

$$x(k+1) = F(x(k), u(k)) + w(k) \tag{1}$$

$$y(k) = h(x(k)) + v(k) \tag{2}$$

in which k represents the discretization point at time point $t(k)$, x are the states of the system, u the system input, and y the measured variables. F describes the discretized differential equation system related to the state variables of the system and h stands for the algebraic part of the system model, which includes relations for the measured variables y. The independent random variables $w \sim N(0, Q)$ and $v \sim N(0, R)$ are the process and measurement noise of the system with the covariance matrices Q and R.

The solution of the SE is given by the conditional probability distribution $p(x|y)$. It describes the probability of a chosen state x under the condition of already occurred measurements y. The objective of every SE method is to maximize this probability by estimating the right values for x using Bayes law. In general, one can divide the Bayesian-based SE methods into two classes (Rawlings and Bakshi, 2016).

The first class are recursively working predictor-corrector methods. The most common SE methods in this class are the Kalman filter algorithms. The schematic working procedure of SE methods of this class is illustrated in Figure 1b. A prediction step is used to forecast the state x and the covariance of the state P at a future time point. The state variables are directly predicted by using the model of the system. For the prediction of the covariance P an approximation method incorporating Q is used. The procedure of this approximation is the main characteristic of every SE method of this class. For the correction step Bayes law including the measurement data is used to maximize the probability distribution of interest (Rawlingsand Bakshi, 2016).

The second class are methods based on nonlinear programming problems (NLP). These methods use a set of measurement data from more than one time-step to estimate x by solving an optimization problem of the form:

$$\{\hat{x}(k-N), \dots, \hat{x}(k)\} = \arg \min_{x(k-N),\dots,x(k)} \phi(k-N) + \frac{1}{2} \sum_{i=k-N}^{i=k} f_{est}\big(v(i)\big) + \frac{1}{2} \sum_{i=k-N}^{i=k-1} w^T(i) Q^{-1}(i) w(i) \tag{3}$$

$$s.t. \qquad Eq.(1), \qquad Eq.(2), \qquad x^{LB} \leq x(i) \leq x^{UB}$$

Here, $\hat{x}$ are the estimated state variables, N is the number of time steps, from which measurement data is used, and ϕ are the arrival cost. The latter considers missing data from outside of the used interval. For intervals with many data points (high values of N) and a high number of measurements, the arrival cost can be neglected. The functional relation f_{est} is used to weight the deviation between the measurements y and the corresponding model variables $h(x)$. A common approach is the use of a least squares formulation. For measurement data with gross error, the least square method will most likely lead to bad estimation results. In this case different

approaches for f_{est} like M-estimators or maximum likelihood estimators have to be chosen (Hoffmann et al., 2016).

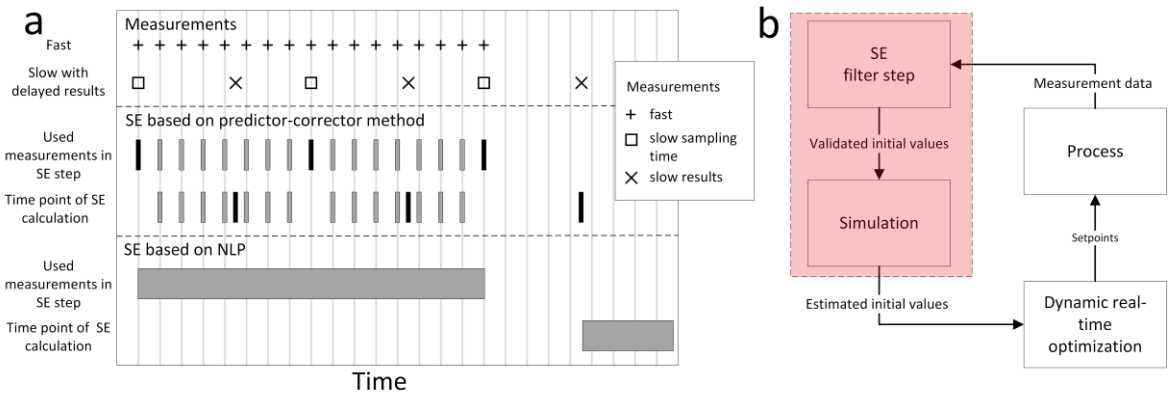

Figure 2: Measurement usage and calculation behaviour of different state estimation algorithms. The black bars refer to predictor-corrector SE steps where slow measurements are available (left (a)). Framework of the integration of a state estimation step in a dynamic real-time optimization environment (right (b))

In Figure 2a, the general measurement usage of the mentioned SE classes is shown. The exemplary data consists of multi-rate measurements with delayed incoming results for slow measurements like concentration data. The SE based on predictor-corrector methods only uses one data set per estimation step (bars in the middle segment), whereas the SE based on an NLP uses measurement data of multiple past time points (bars in the lower segment). The black bars in the middle segment are linked to time points with slow measurements available. It becomes clear, that the slow measurement data causes a delayed SE calculation at the respective time points. This fact needs to be considered for the implementation of SE in a D-RTO environment (see Figure 2b), given that the D-RTO step needs initial values for a future time step.

3. Case study on different predictor-corrector state estimation methods

As mentioned above, SE based on predictor-corrector methods uses an explicit formulation with no optimization steps needed. Under ideal conditions, this fact should cause a more stable SE because solving optimization problems always contains the possibility of failing, especially for highly nonlinear models and non-convex problems. Additionally, predictor-corrector methods use a model of the system with less equations because only one time-step is simulated. The model size of SE based on NLP is related to the number of time steps incorporated in the optimization problem. This leads to lower computing effort of predictor-corrector SE in comparison to SE based on NLP. However, the estimation result of an explicit formulation is only determined by the covariance of the process and measurement noise Q and R. Hence, these statistical parameters have to be accurate.

To investigate the influence of these covariances on the estimation result, a case study is performed. A continuous stirred-tank reactor, holding two compounds i is modeled together with first order reactions and a constant rate parameter k_j and the reactor level L is used as measurement variable:

$$\frac{\partial n_i}{\partial t} = F_{i,in} - F_{i,out} + \nu_i \cdot k_j \cdot \frac{n_i}{n_1 + n_2}, \qquad L = \left(\frac{n_1 \cdot M_1}{\rho_1} + \frac{n_2 \cdot M_2}{\rho_2}\right) \cdot \frac{4}{\pi \cdot d^2}, \qquad i \in \{1,2\},\ j \in \{1,2\} \tag{4}$$

For the integration of measurement and process noise into the system, a true and a faulty model is formulated. Both models use different values for the rate parameter k_j. The true model ($j = 1$) is used for the generation of level measurements by adding random noise based on the covariance R to simulation results. To implement process noise to the system the faulty model ($j = 2$) is used in the SE calculations.

As one can see in Figure 3a, the chosen approach generates changing deviations between the true and the faulty model over time. This behaviour is also expected for the process noise in real applications. The determination of Q for the case study can be carried out by using the deviations between the true and the faulty model. A good guess of Q was achieved by setting the standard deviations of the process noise to the largest deviation of the two models. Based on the standard deviations, the covariance Q can be calculated.

The case study was carried out for three different filter methods. An Extended Kalman (EKF) filter approach from Rawlings et al. (2006) was used. This filter uses a linearization of the model to predict the covariance of the state P at the next time step. Moreover, the Unscented Kalman filter (UKF) from Wan et al. (2000) was used. Here, P is approximated by sampling the response of the system at five different sigma points. Furthermore, a particle filter (PF) based on an approach from Liu et al. (1998) was formulated. This method is similar to the UKF, but instead of sigma points it uses randomly generated particles based on P and Q to evaluate the system at the next time step. For the case study 100 particles were used in the PF.

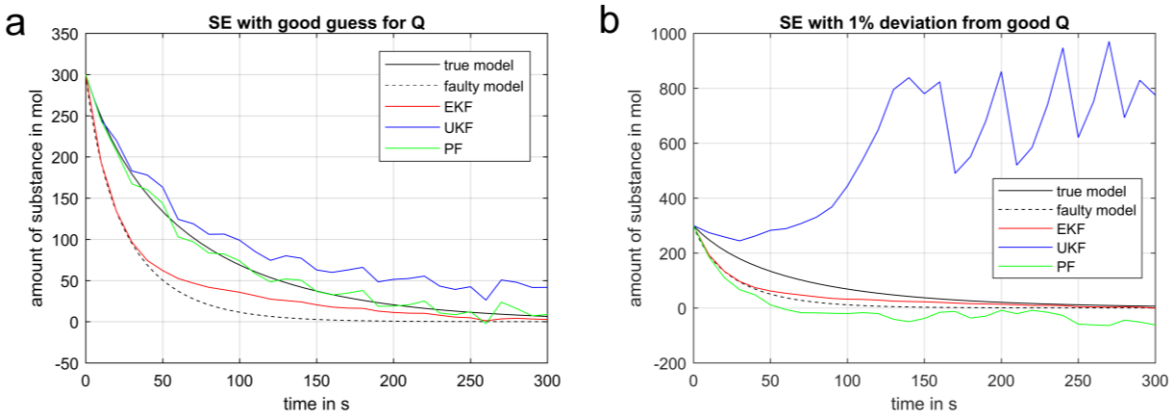

Figure 3: Results of different predictor-corrector SE algorithms with a good guess of the covariance matrix of the process noise (left (a)) and results of different predictor-corrector SE algorithms with a deviation in Q of 1% from the values used in Figure 3a (right (b))

The results of the case study are shown in Figure 3. For the SE presented in Figure 3a, the covariance Q was determined with the method mentioned above. As one can see, the EKF and the UKF fail in estimating the true state of the system. A reason for that could be the large nonlinearities especially in the faulty model. The particle filter is able to estimate the true state of the system. Because of the high number of used sample points the PF manages to handle the nonlinearities of the system. This high number of sample points also leads to a much higher calculation time in comparison with EKF and UKF (factor 106 and 42 in comparison with EKF and UKF). However, in areas with small deviations between the true and the faulty model the results of the PF are inaccurate. This issue arises from the used determination approach for Q, because the used value of this covariance is based on system states with large process noise.
For small perturbations on the covariance Q even the PF fails in estimating the true model. In Figure 3b the results for a deviation of 1 % on the value of the used Q from Figure 3a are shown.
It can be concluded that the EKF and the UKF are not able to produce sufficient estimation results for the observed system. Furthermore, the used values of Q have to be very accurate to maintain appropriate state estimation results with the PF.

4. Developed Moving-Horizon state estimation environment

The case study results show that the covariance Q has to be maintained very accurately in every process state for SE based on predictor-corrector methods. In real processes none true state of the system is known and because of that the approach to guess Q as mentioned above is not applicable in this case. Therefore, the state estimation environment used in the D-RTO of the multiphase hydroformylation process of long-chained alkenes has been developed based on an NLP. The optimization problem formulation for this case is shown in Eq(3). Because of the uncertainties in the determination of Q, the respective term is neglected for the SE. This leads to the case that the occurring process noise is treated as measurement noise in the NLP.
The resulting SE problem contains 75 states (component hold ups of all units in the system), which have to be estimated. Six uncertain mass flows have to be estimated. The usable measurement data consists of four level measurements (measured every second) and concentration measurements of five components at three positions (measured every one, two, and four hours with a delay of 45 minutes of the results) in the process.
Preliminary investigations on the observability of all states and uncertain flows were carried out by determining the sensitivity $\partial\hat{y}/\partial x_{est}$ of all measurement variables $\hat{y}$ with respect to all estimated variables x_{est}. This sensitivity shows whether changes of the estimated variables have an influence on the objective function. If this is not the case the respective variable cannot be estimated in SE based on NLP. The carried-out investigations

show that 10 states are not observable. These states are not included in the variables of the optimization problem and can only be used without the SE corrections in D-RTO.

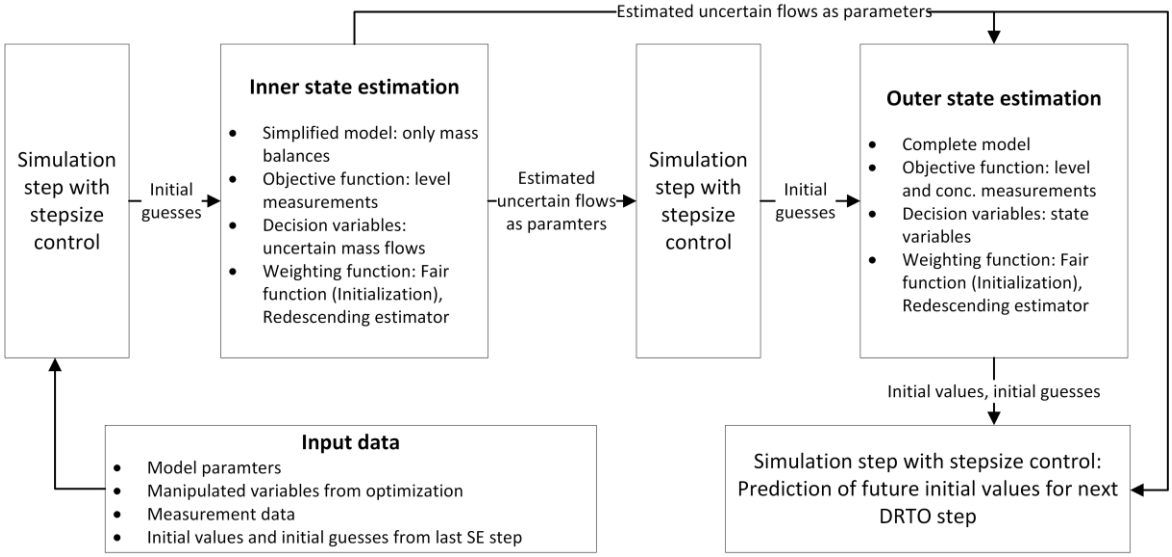

Figure 4: Simplified framework of developed Moving Horizon state estimation environment

During the development of the SE an issue regarding the multi-rate measurements appeared. Because of the high number of level measurements in comparison to the slow concentration measurements, the influence of the concentrations on the objective function was very small. With the additional degree of freedom arising from the usage of the uncertain mass flows, the solving of the optimization problem was very unstable.
To overcome this problem a two-stage SE approach was developed. A simplified framework of this approach is formulated in Figure 4. One can see that the SE problem is separated into an inner and an outer SE problem. The inner SE uses a model only based on mass balances of the system and only uses the fast level measurements in the NLP. Here the uncertain flows are estimated and treated as parameters in further SE process. The outer SE uses the whole process model with level as well as concentration measurements in the NLP. Between the consecutive SE steps, simulations for the generation of initial guesses are carried out.
For the usage in long-term process operation the two-step SE approach was formulated as MHE based on the work of Hoffmann et al. (2016). The MHE uses a horizon of past measurement data of four hours. The process model is fully discretized with orthogonal collocation on finite elements (FE) with a length of each FE of 15 minutes. Because of the long horizon length, the arrival cost in Equation 3 is neglected. This leads to an objective function formulation only based on the measurement noise. As functional relation for the weighting of the measurement error (f_{est}), the redescending estimator is used to eliminate gross error from the measurements. For stability reasons the Fair function is used for initialization (Hoffmann et al., 2016).
The developed MHE environment was initially tested with measurements based on the process model with the same characteristics (frequency of measurements, gross error) as in real plant operation. In this validation case the SE was able to estimate the true state of the system.
Furthermore, the MHE environment was tested with real plant data from long-term mini-plant operation. Some exemplary results are shown in Figure 5. Here, for visualization purposes variables with available measurements are shown. One can see measured levels of different tanks of the mini-pant and the estimated values of the regarding measurement variables. The results regarding the fast-measured levels are good and one can see that the large gross error especially in the measurements of L_{u14} and L_{u3} has no influence on the associated variable. Whereas the results concerning the concentrations show some problems. Especially between the particular measurements the concentration courses show unphysical behavior. Because this issue does not arise when measurements based on the process model are used, the plant-model mismatch appears to be too high to receive appropriate estimation results. The assumption of neglecting the process noise term (last term in Eq. 3) from the objective function is not feasible for the used model. To improve the SE quality either the plant-model mismatch has to be reduced or a reliable formulation of Q has to be incorporated in the SE.

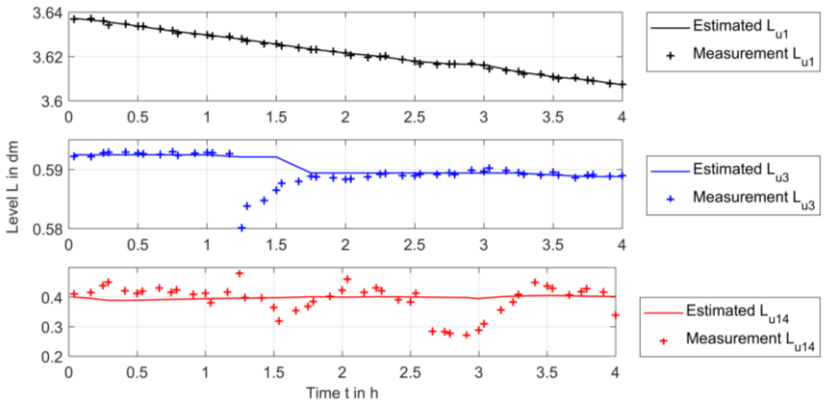

Figure 5: Exemplary results of estimated levels of developed MHE with real mini-plant data

5. Conclusions

This contribution shows the development of a state estimation environment for a hydroformylation process of long-chained alkenes. SE based on predictor-corrector methods and on NLP have been tested. For the predictor-corrector SE methods it can be stated, that the particle filter shows good estimation results. This can only be guaranteed with accurate values of the covariance matrix of the process noise Q at every state of the system.

Because accurate values for Q are not available for the investigated hydroformylation process, SE based on NLP is used. The developed MHE environment shows good results for measurement data based on the model. With real plant data the results are only partially appropriate for the usage in a D-RTO because the mismatch between the used model and the real process is too high.

To overcome this issue, one possibility would be a reduction of this plant-model mismatch by further adjusting the plant model to the real process. In many cases this is not possible, because the complexity of the resulting model would be inappropriate for the usage in real-time applications like D-RTO. Another possibility would be the incorporation of the process noise term in the SE optimization problem (as formulated in Equation 3). However, again a reliable formulation of the covariance of the process noise is needed for this approach. It is suggested to carry out investigations on techniques for the provision of Q for SE in real processes by using long-term plant data.

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