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Modeling of a three-phase industrial batch reactor using a hybrid first-principles neural-network model

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Abstract

We present an industrial case study of a three-phase reaction system in a batch reactor. For the successful modeling and prediction of the plant-scale performance a hybrid model is used. Data from different scales were available for developing the model. In order to model the large-scale production process the first principles model was extended with neural network models to identify the missing parameters.

Keywords hybrid modeling, neural networks, three-phase reaction, laboratory and industrial scale, scale-up

1. Introduction

In order to develop a reliable three-phase model all information available from the lab-scale experiments and from the industrial-scale process is used. To take advantage of process knowledge and process measurements a hybrid first-principles neural network model has been proposed. The key factors in modeling this batch process involve the reaction mechanism and kinetics, dissolution rates, solubility equilibrium, global mass-balance equations for the liquid and solid phases, and the calculation of missing removal rates. The different types of model components and measurements data are combined into

a hybrid first-principles neural network model. It will be demonstrated how the developed models are used to analyze and improve the investigated process.

2. Problem statement

In the beginning of the process operation, until the complete dissolution of component A, the reactor system consists of three phases: solid, liquid and gas. Four equilibrium reactions in series take place in the liquid phase and a catalyst is used in solubilized form. The reaction scheme is as follows:

$$A_s \rightleftharpoons A_l \quad (1)$$

$$A_l + B \rightleftharpoons C + D \quad (2)$$

$$B + C \rightleftharpoons E + D \quad (3)$$

$$B + E \rightleftharpoons F + D \quad (4)$$

$$B + F \rightleftharpoons P + D \quad (5)$$

where A_s and A_l represent component A in solid and liquid phase, respectively. Raw materials are component A and B; components C, E, F are intermediates and P is the desired product. Besides the reaction kinetics there are two major phenomena to be modeled: the dissolution of component A into the liquid phase and the removal of coupled product D (scale dependent).

3. Methodology

3.1. Small-scale process model

On the small scale experimental data from 1 kg laboratory experiments are available in the form of time-variant temperature and pressure profiles and measured concentrations for components A, B, E, F, and P. Due to the operating conditions and the small reaction mass we assume that the component D holdup in the liquid phase is zero on this scale. As a consequence reverse reactions do not take place and the kinetic model will comprise only the four forward kinetic equations. The complete model description is found in L. L. Simon et al. [1]. For this model there are five parameters to be fitted: the solid-liquid mass transfer coefficient and the four rate constants at reference temperature.

3.2. Large-scale process model

The industrial process is operated in a 6 m^3 vessel with the same initial mass fractions of component *A* and *B* as the experiments on the 1 kg scale. However, as will be demonstrated below the assumption of component *D* concentration being zero does not hold true on this scale and a large-scale process model is needed. To complement the first-principle part the mass balances in the reaction model are extended with the liquid phase mass balance of component *D*:

$$\frac{dn_D}{dt} = -\frac{dn_B}{dt} - r_{removal} \tag{6}$$

where n_D is the component *D* mole number in liquid phase [mol], and $r_{removal}$ is the time-variant removal rate of component *D* during the process [mol / s]. The latter parameter is the unknown in the first-principles model and is calculated with two neural networks, each describing one of the two stages of process operation. The connection between the first principles part and the neural network is made in the serial way [2].

3.2.1. Modeling of the first stage of process operation

During the first stage component *D* is condensed after removal from the reactor and is collected in a storage tank. The goal of the neural network model *NN1* for this stage is to create a dynamic mapping between process advancement, measured in form of accumulation of component *D* in the storage tank, a process operation specific variable (i.e. temperature) and the removal rate of component *D* from the reactor. In order to achieve this goal a one-step ahead feed-forward neural network is proposed. The inputs to the model are the accumulated (condensed) mass of component *D* in the storage tank and the reactor temperature, which is needed due to the fact that the production rate of component *D* is directly influenced by it. The neural network output is the accumulation rate in the storage tank, which was calculated by differentiating the accumulated mass. The implemented black-box model is a Bayesian type of neural network.

3.2.2. Modeling of the second stage of process operation

In the second stage the accumulation rate of component *D* is not available anymore because it is not condensed; instead concentration measurements are taken and similarly to stage one the evacuation rate is the unknown parameter in the first-principles part. Due to the fact that the removal rate of component *D* is not measured, usual training methods such as back propagation are not possible.

The identification of the component *D* removal during this stage is posed as an inference or parameter estimation problem from the concentration measurements. As a solution to the parameterization of the removal rate and inference of component *D* in liquid phase a feed-forward neural network structure (*NN2*) is proposed. This structure has as input the component *P* yield and as output the component *D* evacuation rate. By setting the product *P* yield as the input the normalization of the mass of product *P* was achieved, by this the model can be used for extrapolation as will be shown later on. The assumption behind this normalization is that the system will have the same behavior at the same yield values and its behavior does not depend on the mass of reactants present in the reactor. This assumption is only valid in a certain range around the operating points used for the *NN2* model development.
The calculation of the removal rate of component D is posed as an optimization problem [3] with the goal of minimizing the overall hybrid-model predicted concentration deviations by manipulating the weights and biases of this second neural network (*NN2*) subject to the constraints formulated in the given first-principles model and evacuation rates calculated from *NN1*. The hybrid model structure is presented in Figure 1.

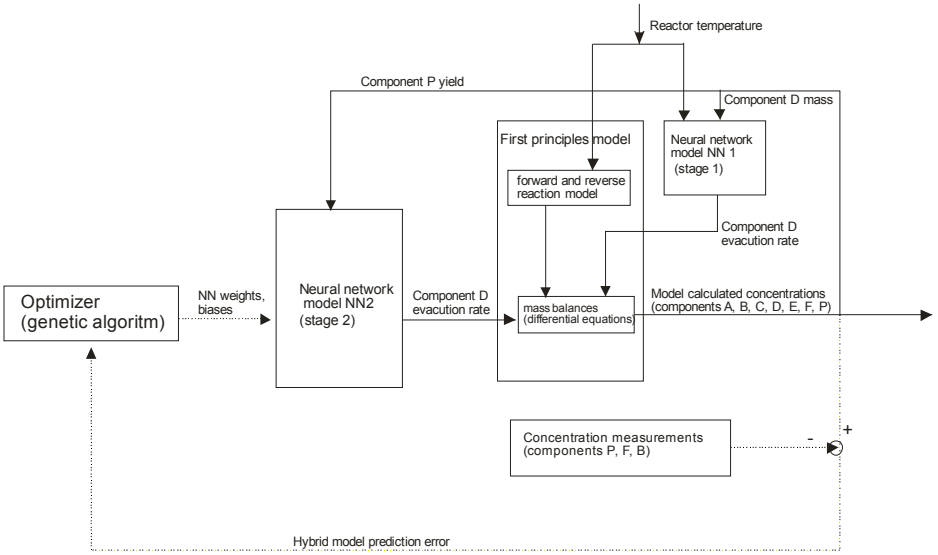

Figure 1 Overall structure of the hybrid model (continuous lines indicate the connections between the sub-models) and the optimization problem (dashed lines).

3.3. Results& discussions

3.3.1. Small- scale process model results

A comparison of the modeled concentrations and the experimental data as obtained in fitting the reaction kinetics and solid-liquid mass transfer coefficient to the small-scale laboratory experiments is shown in Figure 2. The model describes the data well and the degree of explanation R^2 is about 98%.

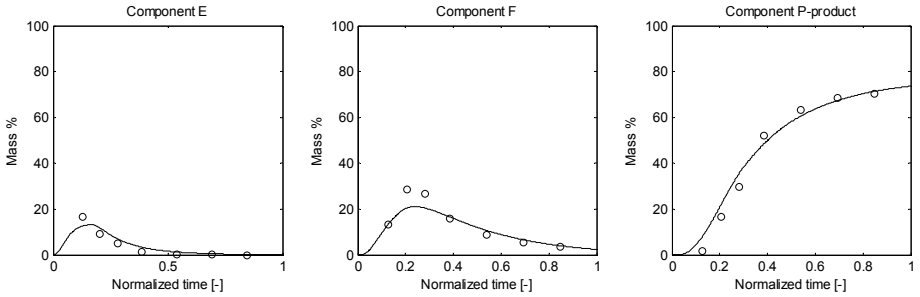

Figure 2: Comparison of measurements from the laboratory experiments (circles) and corresponding fitting results as obtained with the small-scale process model (solid line).

3.3.2. Large-scale process model results

It is important to describe the concentration of component *F* as accurately as possible because it defines the termination criteria for the batches in plant operation. The hybrid model describes the low component *F* values at the end of the batch well. However, the model does not describe the decrease in component *F* mass fraction from its peak value at the beginning of the batch well. A reason for this might be that the catalyst activity in the large-scale reactor is not exactly the same as in the laboratory experiments. Although the exact reason could not be identified it was concluded that a re-fitting of the kinetic parameters on large scale is required.

The kinetic parameters were re-fitted with the forward reactions model. The fitting resulted in a value for the 4^{th} reaction constant being 30% smaller than in the small-scale model while the other reaction constants are unchanged. The decrease in component *F* mass fraction from its peak value is described much better with this new set of parameters. Therefore, in the next modeling step the *NN2* in the hybrid model is re-optimized on the basis of the updated first-principles model.

By this a very good agreement between plant measurements and modeled component mass fractions is obtained (Figure 3). In particular component *F* is

now modeled well all over the whole range of available measurements.

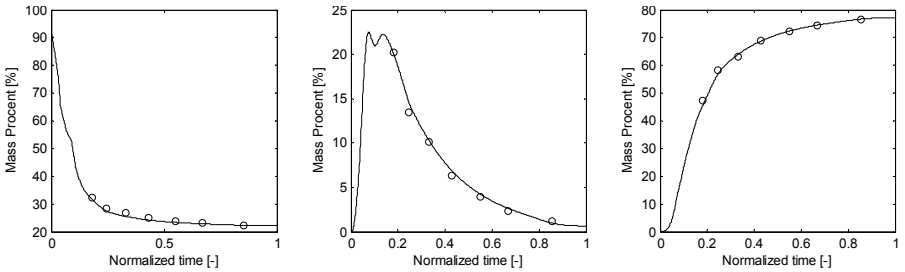

Figure 3: Comparison of component mass fractions as obtained with the large-scale hybrid model including updated kinetic constants and updated NN2 (solid line) and plant measurements (circles).

4. Conclusions

In this work the modeling and improvement of a complex industrial batch reactor has been presented. Here a first-principle approach is applied for modeling reactions and dissolution of one reactant that is introduced into the reactor in solid form. In order to model the large-scale production process this model was extended with neural network models to identify the missing parameters. In addition a re-fitting of the kinetic parameters on plant scale was required. With this hybrid model a good prediction of the concentration courses in the industrial reactor was obtained.

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