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Data-driven Model Predictive Control of Nanoparticle Production in Modular Reactors

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***Abstract*— Microreactors are an essential part of modular chemical systems involved in the on-demand production of chemicals such as nanomaterials, pharmaceuticals, specialty chemicals, etc. Model-based nonlinear predictive control of microreactors is a challenging task due to the high online computational cost associated with developing and maintaining high-order first-principles nonlinear models. In this work, we propose a nonlinear data-driven model predictive control (NMPC) scheme for nanoparticle production in microreactors. In this paper, a non-linear Auto Regressive Exogenous Neural Network model (NARX-NN) is developed with the flow rates of the reactants as inputs and the peak value of the absorbance spectra (an indirect measure of the average size of nanoparticles) as output by performing a set of experiments in Corning Advanced-Flow^{TM} Reactors (AFR). Typically, producing a new desired average size nanoparticle on-demand is done by manual changes in the flow rates of reactants. In this work, a nonlinear model predictive controller using the identified NARX-NN model is formulated to track a change in the set point, the peak value of the spectra. The formulated controller with the identified NARX-NN model is demonstrated via the simulation studies. It is shown that the proposed NMPC with the NARX-NN model performs well in different scenarios of silver nanoparticle production.**

I. Introduction

In the last decade, there was a rapid shift from batch to continuous processes for large-scale synthesis in specialty chemicals and pharmaceutical manufacturing. Microreactors have gained popularity in academia and industries as they offer advantages such as higher surface-to-volume ratios and high heat and mass transfer rates [1], [2], [3]. Furthermore, these reactors allow for a high throughput design by increasing the number of reactors in parallel [2] thereby facilitating industrial applications as higher throughputs are essential for commercialization with a small footprint. The main objective of this work is to develop a data-driven model-based control approach producing different sizes of nanoparticles in microreactors.

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Microreactors are often modeled as tubular flow reactors, and the time evolution of concentrations of different species along the length of reactors is derived in terms of a set of partial or ordinary differential equations (PDE or ODEs) under certain assumptions [4], [5]. These coupled ODEs and PDEs with nonlinear kinetics are computationally expensive for online applications. Furthermore, reaction kinetics have to be identified using experimental data[6], [7]. The identification of reaction kinetics from experimental data is also a time-consuming and costly exercise. Hence, building first-principle models for real-time optimization and model-based control is not a favorable option for microreactors from a computational viewpoint [8]. On the other hand, data-driven models such as multivariate linear input-output models, nonlinear autoregressive-with-exogenous-inputs (NARX), polynomial nonlinear-autoregressive-with-exogenous-inputs (PNARX), etc. are widely used in process control[9], [10]. These data-driven models allow us to predict the quantities of interest in an online manner with less computational cost. However, the development of data-driven models requires a large amount of data.

The control of continuous microreactors leads to the multivariate and constrained control problem and hence, traditional PID control techniques will not be effective in the production of chemicals in microreactors. Recently, model-based predictive control strategies using simple first-principles models as well as data-driven models have been implemented in the literature [11], [10]. In [11], the authors developed a linear data-driven model predictive scheme for ether production. Here, the authors developed a measurement model and a control model using the partial least-squares (PLS) regression and the output-error modeling approach, respectively. An artificial neural network (ANN) has been used in developing a dynamic model for a control purpose [12]. Similarly, recurrent neural networks (RNN) have been used for the development of high-fidelity dynamic models and for designing a control strategy [13]. A successful implementation of an ANN-MPC on a continuously stirred tank reactor has been shown in [14].

The main challenge in the on-demand manufacturing of nanoparticles in microreactors is the production of different sizes of nanoparticles for various applications in a short period. Hence, the control of the microreactors is crucial for obtaining the right size of nanoparticles largely by manipulating the input flow rates and temperature [15]. Typically, the size of nanoparticles is obtained by performing Transmission electron microscopy (TEM) in an offline manner by analyzing the outlet stream of microreactors. However, TEM analysis is a tedious and time-consuming task, and it requires a dedicated facility and trained human resources. Alternatively, UV spectroscopy can be used to measure the average size of nanoparticles by analyzing the outlet stream in an online manner. Particularly, the peak of the absorbance spectra provides an indirect measurement of the average size of nanoparticles produced. In addition, it is difficult to build a first-principle model for the production of nanoparticles. Hence, an effective control strategy with a data-driven model connecting the input flow rates and/or temperature to the peak of absorbance spectra is the most suitable option for obtaining the desired sizes of nanoparticles.

In this work, we develop a nonlinear auto-regressive Exogeneous Neural Network model for the production of silver nanoparticles using experimental data involving two inputs and a single output. We further develop an MPC around it to achieve our desired setpoint tracking capabilities required for on-demand production of different sizes of nanoparticles. This paper further demonstrates the data-driven nonlinear MPC for tracking the peaks of absorbance spectra (a proxy for the average size of nanoparticles produced) for the production of silver nanoparticles.

The structure of the paper is as follows. Section II describes the development of NARX-Neural Network (NARX-NN) models for predicting the peak of absorbance spectra using two manipulated flow rates. The developed NARX-NN model is then used for formulating a model predictive control problem for the set-point tracking problem in Section II. Section III develops an NARX-NN model to describe the production of silver nanoparticles from the experimental data obtained in the laboratory. Section IV investigates the effectiveness of the formulated NARX-NN model predictive control for tracking a set-point change in nanoparticle production and presents the results. Finally, Section V concludes the paper and discusses the future directions.

II. Model Development and Control Formulation

This section presents the development of a data-driven model and the formulation of model predictive control.

A. Non-Linear Auto Regressive Exogenous Neural Network Model (NARX-NN)

In this section, we develop a non-linear Auto-regressive exogenous model (NARX) with one output (y) and n_u inputs ($u \in R^{n_u}$) in the system [1]. This NARX model is integrated with neural network layers to model the desired non-linearity in the plant model. This leads to nonlinear auto-regressive exogenous Neural Network (NARX-NN) models. In the production of nanoparticles using microreactors, the main goal is to develop a NARX-NN model between the manipulated flow rates and the peak of absorbance spectra (a proxy for the average size of the nanoparticles). Hence, in the formulated model, the inputs and outputs are modeled as the deviations of outputs and inputs from the reactor's desired steady-state, $\hat{y} = y - y_{ss}$ and $\hat{u} = u - u_{ss}$. Then, the NARX-NN can be expressed in Eqn. 1 as follows:

$$\hat{z}[k] = \textit{NARX-NN}\left(\begin{bmatrix} \hat{u}[k-H_w] \\ \hat{u}[k-H_w-1] \\ \dots \\ \hat{u}[k-H_w-n_u] \\ \hat{y}[k-1] \\ \hat{y}[k-2] \\ \dots \\ \hat{y}[k-n_y] \end{bmatrix}\right) \tag{1}$$

where $\hat{z}$ is the controlled output that is predicted based the past n_u manipulated inputs $\hat{u}$ applied to the system, and the previous n_y measured outputs $\hat{y}$, and H_w is the input-output delay associated with the system. The architecture neural network model is modified to exhibit the properties of chemical processes in microreactors such as zero-output response for a zero-input vector. To ensure this, we remove the biases from fully-connected linear layers of the neural network. Furthermore, steady-state data are used along with time-series perturbed data in the training set. For training the models, the custom loss functions that impose a high penalty on non-zero output for a zero-input vector can also be used. Section III describes building an NARX-NN Model for the production of silver nanoparticles using the experimental data obtained in microreactors.

B. Model Predictive Control Structure

A simplified architecture of the proposed model predictive control (MPC) structure is shown in Fig. 1. The optimizer and the prediction blocks collectively make an MPC. To identify the optimal inputs to track the programmed set-point, the MPC utilizes the NARX-NN model to predict the outputs over the prediction horizon. In the case of nanoparticle production, the MPC manipulates the flow rates of reactants to track the peak of absorbance spectra obtained at the outlet of a microreactor.

The optimizer's objective is to minimize the deviation from the setpoint, denoted by $\hat{r}[k]$, while ensuring constraints on the system. The objective function or loss function is formulated in Eqn. 2 as follows:

$$V[k] = \sum_{j=H_w}^{H_p-1} \left\|\hat{r}[k+j]-\hat{z}[k+j]\right\|^2_{Q(j)} + \sum_{j=0}^{H_u-1} \left\|\Delta\hat{u}\right\|^2_{R(j)} \tag{2}$$

where $\|\mathbf{a}\|^2_W = \mathbf{a}^T\mathbf{W}\mathbf{a}$, H_p is the prediction horizon, H_u is the control horizon and Q and R are the weight matrices associated with the objectives of deviation from the set-point and movement suppression, respectively. The loss function in the vector form can be rewritten by defining the controlled output $\hat{Z}$, reference trajectory vector $\hat{\tau}$, and input vector $\hat{U}$ as follows:

$$V[k] = \left(\hat{\tau} - \hat{Z}\right)^T Q \left(\hat{\tau} - \hat{Z}\right) + \Delta U^T R \Delta U \tag{3}$$

with

$$\hat{Z} = \begin{bmatrix}\hat{z}[k+H_w] & \hat{z}[k+H_w+1] & \dots & \hat{z}[k+H_p-1]\end{bmatrix}^T \tag{4}$$

[1]Note that the methodology can be extended to multiple inputs and outputs straightforwardly.

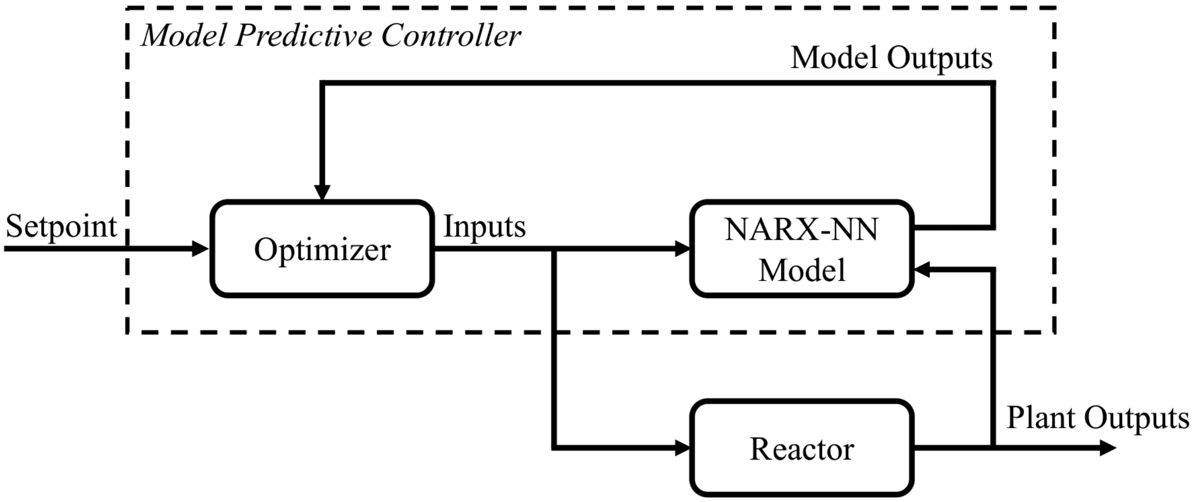

Fig. 1. Control structure depicting the information flow inside the MPC and in between the MPC and reactor

$$\hat{r} = \begin{bmatrix} \hat{r}[k + H_w] & \hat{r}[k + H_w + 1] & \ldots & \hat{r}[k + H_p - 1] \end{bmatrix}^T \tag{5}$$

$$\hat{U} = \begin{bmatrix} \hat{u}[k] & \hat{u}[k+1] & \ldots & \hat{u}[k + H_u - 1] \end{bmatrix}^T \tag{6}$$

Here, ΔU is the change between consecutive inputs. Furthermore, the following lower and upper bounds on the inputs and change in the inputs are formulated as constraints:

$$\hat{U}_{min} \leq \hat{U}_{opt} \leq \hat{U}_{max} \tag{7}$$

$$\Delta U_{min} \leq \Delta U_{opt} \leq \Delta U_{max} \tag{8}$$

As the NARX neural network model is built to predict only one step in the future, the computation of a larger horizon requires the backtracking of the previous optimal inputs. The optimizer's objective function repeatedly calls the prediction model to determine the intermediate variables. We have used `scipy.optimize.minimize` function in `Python` to minimize this loss function. Given the function's properties, we use the 'Powell' method to solve the constraint optimization problem formulated in Eqn. (3) with constraints in Eqns. (7), and (8).

III. NARX-NN Model Development for Nanoparticle Production in Microreactors

In this work, the production of silver nanoparticles in Corning Advanced-Flow^{TM} Reactors (AFR) is considered. The aim is to produce silver nanoparticles (AgNPs) with predetermined specifications in the Corning AFR. The size of silver nanoparticles is provided in terms of the peak of the predetermined absorbance spectra of the nanoparticles. The materials used are Silver Nitrate ($AgNO_3$), Sodium Borohydride ($NaBH_4$), and Tri-sodium citrate (TSC) are mixed in a Corning AFR to produce silver nanoparticles. The experimental setup incorporates a UV spectrophotometer to measure the absorption spectra of the nanoparticles produced at the exit of the microreactors, and the peak value of absorbance spectra is used as the proxy for an average size of nanoparticles.

In this experiment, the manipulated variables are the flow rates of silver nitrate and sodium borohydride, while the controlled variable is the peak values of the absorption spectra of silver nanoparticles. Initially, we fix the inputs and let the reactor reach its steady state, after which we apply perturbations in the inputs (pseudorandom binary sequence (PRBS)) and measure the response of the system (peak values of absorbance spectra at the exit of microreactors) for obtaining experimental data. Fig. 2 shows the experimental measurements obtained after applying perturbations in the flow rates of both reactants for an experiment. The sampling time for measuring the inputs and outputs is 60 seconds.

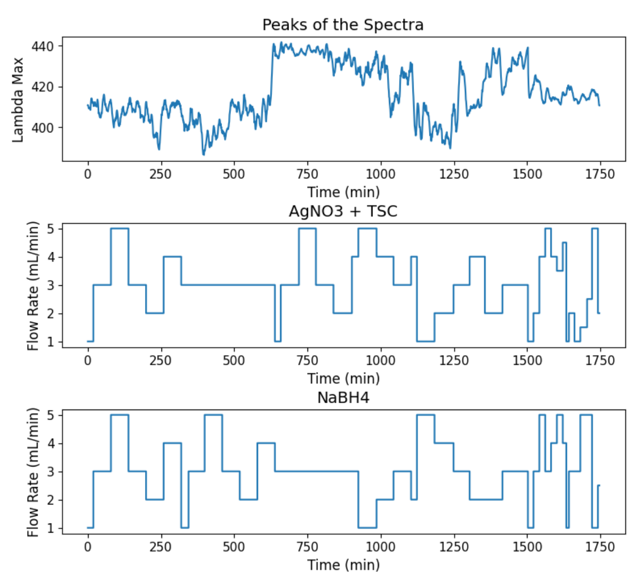

Fig. 2. Perturbation data generated through the silver nanoparticle synthesis experiment. The first subplot 1 shows the absorption spectra peak values across the time horizon. Following subplots 2 and 3 show the corresponding input flow rates over the time horizon

For training the NARX-NN model, the perturbation data is split into training and validation datasets, with the ratio being 85%-15%. Additionally, the steady-state flow rates and the steady-sate peak values of absorbance spectra are subtracted from the inputs and output, respectively, to obtain the data in the deviation form.

The implemented NARX-NN model assumes that the output is dependent upon the current input and the past two outputs. This configuration was chosen as it demonstrated a high R^2-value on 10-fold cross-validation in comparison to other configurations considering multiple past inputs and more outputs. It was observed that an enhanced model fit allowed superior MPC performance. This relation is exhibited in Fig. 3. The delay associated is less than the sampling time and hence, it is ignored.

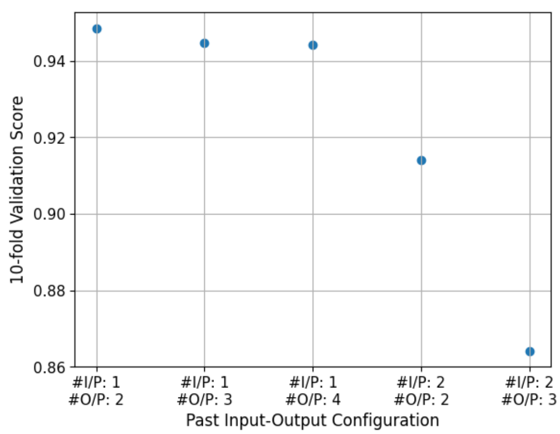

Fig. 3. Performance of various input-output configuration with respect to R^2-value in 10-fold cross-validation test. The model was trained for 20 epochs

$$\hat{z}[k] = \textit{NARX-NN}\left(\begin{bmatrix} \hat{u}_{AgNO_3}[k] \\ \hat{u}_{NaBH_4}[k] \\ \hat{y}[k-1] \\ \hat{y}[k-2] \end{bmatrix}\right) \tag{9}$$

The model architecture comprises one hidden layer constructed using three neurons, with a rectified linear unit activation function to incorporate the desired non-linearity. The model is trained using the 'ADAM' optimization algorithm with the mean-squared prediction error as the loss function for training the NARX-NN model. For the termination criteria, we train the model until the R^2-a value of at least 0.95 is achieved. The trained model is independently validated through 10-fold cross-validation. The R^2-value of the validated model is 0.9561. This value indicates that the model between the peak value of absorbance and the flow rates of reactants is a good fit. The developed NARX-NN model performance on independent experimental data is shown in Fig. 4.

Additionally, we perform the residual analysis to understand how the model describes the system. The residuals are differences between the one-step-ahead predicted output from the model and the measured output from the validation data set. The residual analysis plots are shown in Fig. 5 and Fig. 6 and it indicates that the performance of the model is satisfactory.

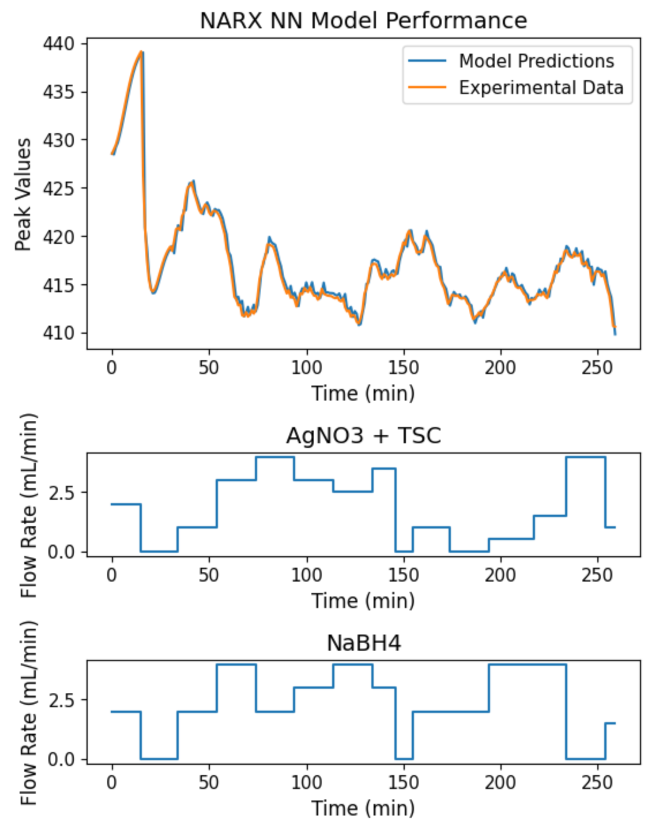

Fig. 4. Performance of NARX neural network against the validation data. The model displayed has an R^2-value of 0.9672

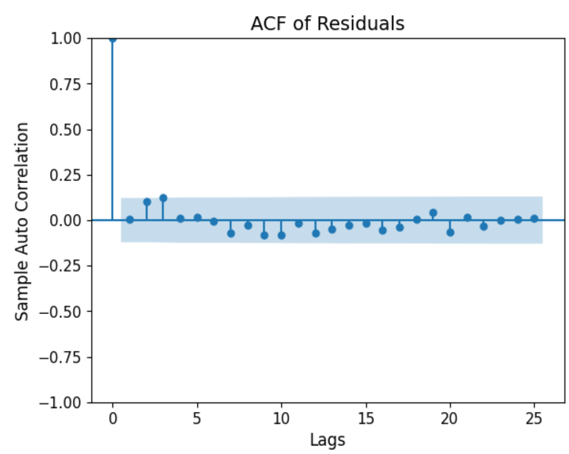

Fig. 5. Autocorrelation of the NARX-NN model residuals

The model also retains the defined steady state by predicting no change in the output (peak value) for the zero-input vector in the deviation form. In other words, the constant values of the flow rates of reactants do not change the peak value of the measured spectra. Hence, the developed NARX-NN Model also provides physically meaningful predictions.

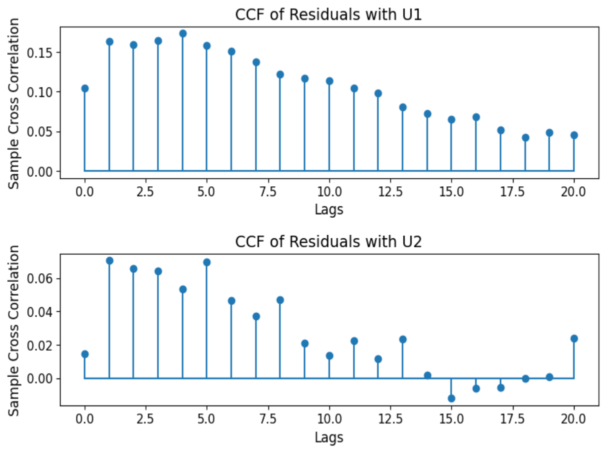

Fig. 6. Cross correlation of the NARX-NN model residuals with the inputs

IV. NARX-NN Model Predictive Control (MPC) for the Production of Silver Nanoparticles of Desired Sizes

After obtaining satisfactory performance of the developed NARX-NN model, we use this model for a one-step predictor in the model predictive controller formulated in Section IIB. In this section, we perform simulation studies to analyze the set-point tracking capabilities of the proposed MPC using the NARX-NN model in the production of silver nanoparticles of different sizes. The applications of nanoparticles depend on the average size of nanoparticles. For example, nanoparticles with a size of less than 5 nm are used for catalysis-related applications. Hence, obtaining nanoparticles of a particular size is an important objective of the control of the production of silver nanoparticles. Since the peak values of absorbance spectra are used to measure the average size of particles indirectly, the set point for the lambda max (maximum absorbance) must be tracked by manipulating the flow rates of $AgNO_3$ + TSC stream and $NABH_4$ stream independently. Hence, in this simulation study, we first investigate the set point tracking problem.

It is important to track the peak values of the absorbance spectra of the nanoparticles obtained at the exit of the microreactors. For the first simulation scenario, we have set the values of prediction horizon $H_p = 6$ and control horizon $H_u = 3$. The weights $Q = 100$ and $R = 500$ are taken. The performance of the MPC for the following configuration is shown in Fig. 7.

As observed, the NARX-NN MPC successfully tracks the reference trajectory provided and changes in the setpoints. However, the observed settling time is longer than desired in practice. The situation can be enhanced by decreasing the weight R to allow greater movement between consecutive input vectors. Then, maintaining all the other parameters, the weight R is reduced to 0.5.

The performance of the MPC for the following configuration is shown in Fig. 8. As observed, the model has become more aggressive, and convergence to the setpoint is approximately 15 minutes. However, the changes in consecutive inputs are significant.

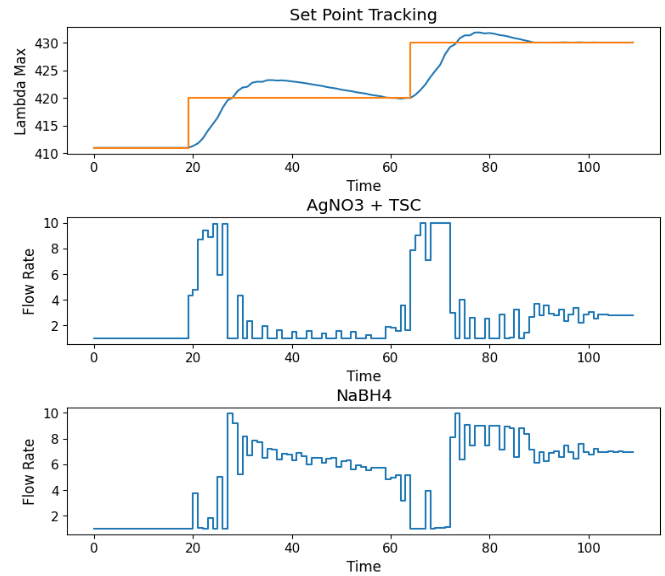

Fig. 7. Case 1: MPC set-point tracking simulation. Lambda Max is the peak value of absorbance spectra at the exit of the microreactor. The flowrates are in milliliters per minute, while the iterations are in line with the sampling time of 1 minute

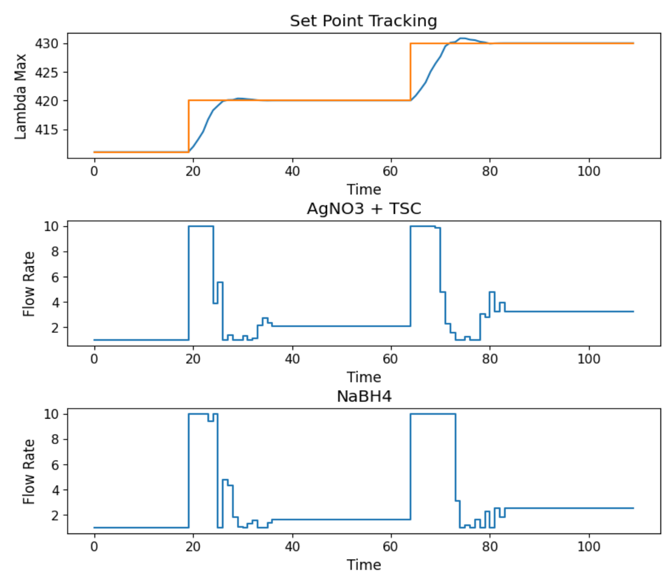

Fig. 8. Case 2: MPC set-point tracking simulation

To further improve the settling time, we now increase the prediction and control horizon from 6 and 3 to 8 and 5, respectively. We maintain the weights Q and R at 100 and 0.5, respectively, in this simulation. The performance of the MPC for the following configuration is shown in Fig. 9.

As observed, the response is further enhanced with the now approximate settling time of around 10 minutes, which is fine in practice.

V. Conclusions and Future Works

In this work, a non-linear auto-regressive exogenous model with Neural Networks (NARX-NN) has been developed for

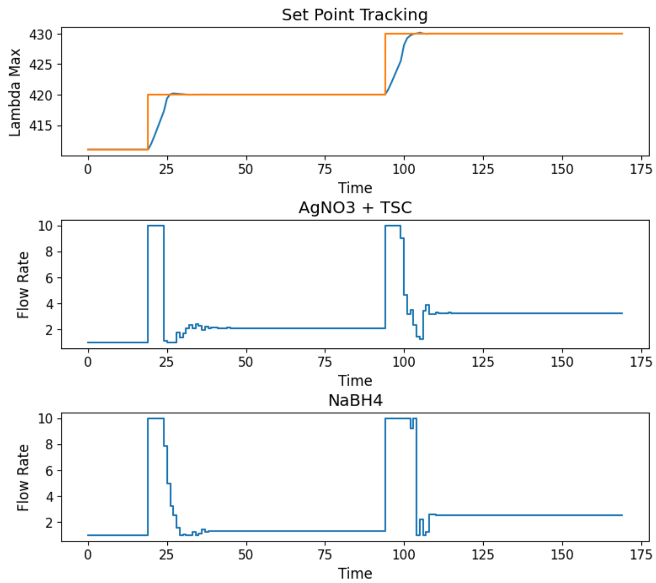

Fig. 9. Case 3: MPC set-point tracking simulation

the production of nanoparticles in microreactors. It is shown that the developed NARX-NN model allows for predicting the peak of absorbance spectra at the exit of microreactors using the flow rates of reactants. The developed NARX-NN model is then used to formulate a nonlinear model predictive control strategy for tracking set point changes in the production of nanoparticles. A NARX-NN model has been developed using experimental data obtained from the production of silver nanoparticles in the Corning Advanced-Flow^{TM} reactors. The developed NARX-NN model for the production of silver nanoparticles has been used in the NMPC formulation for tracking the change of the average size of nanoparticles via tracking in the peak values of absorbance spectra at the exit of the microreactors via a set of simulation studies. The simulation to identify the optimal tuning of the MPC has revealed that the controller is effectively settling to the desired setpoint in approximately 10 minutes.

In the future, the proposed NARX-NN model-based predictive control will be integrated into the experimental setup for the production of silver nanoparticles of different sizes using the Corning reactors, and the size of silver nanoparticles will be validated using the TEM analysis. Furthermore, the framework will be extended to reject unknown disturbances that affect the average size of nanoparticles. We will also develop control strategies for obtaining narrow particle size distributions using the absorbance spectra.

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