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Online parameter identification of facet growth kinetics in crystal morphology population balance models

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Abstract

Particle shape plays an important role in many industrial applications since it can have significant impact on both, processability of particles as well as the properties of the final product. For this reason modeling of the corresponding production process is crucial for developing efficient process optimization and control strategies. The shape evolution of crystals on the process scale can be described conveniently within the framework of morphological population balance modeling. In order of being a reliable tool for the prediction of the crystal shape distribution during the production process as well as for the design of suitable control and optimal production strategies, the models require the estimation of several parameters characterizing the growth rates of the different crystal facets. This is particularly challenging due to the infinite dimensional state space of the models. In this contribution online parameter estimation for the growth rates of L-glutamic acid cooling crystallization is presented. Using a Lyapunov-based approach the parameter adaption laws are computed directly from the infinite dimensional problem formulation. It will be shown that a reasonably fast convergence of the parameter estimates can be achieved even in the presence of measurement noise using appropriate filters.

Keywords: Multivariate population balance modeling; Crystal morphology; Crystallization; Online parameter identification

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1. Introduction

Crystallization is an important class of production processes in chemical and pharmaceutical industries. It is used to produce a desired material in crystalline form from a liquid solution. Details on crystallization principles and techniques can be found e.g. in [1]. The production processes are characterized frequently by heterogeneity of the crystal ensemble with respect to crystal properties like size and shape. Those have a significant influence on the end-use property and the processability of the final product. Modeling of the corresponding dynamics is thus crucial for the design of efficient schemes for process control and optimization. It is well known that the temporal evolution of the previously described heterogeneous system can be modeled using population balances [2]. Here, morphological population balances being a special form of multivariate PBMs can be used to describe the dynamic shape evolution (e.g. [3], [4]). For the development of process control and optimization schemes the individual facets growth kinetics in the population balance have to be determined. This can be done for example by experimentally measuring the growth rates of a single crystal or a small number of crystals, which has several drawbacks that may yield biased estimates. Alternatively, the rates may be determined directly from process scale seeded crystallization experiments. Here, the temperature, solute concentration and the crystal shape distribution have to be measured. The parameters of the growth kinetics can then be estimated minimizing the error between the simulation of the morphological PB and the measurement data from experiment. In offline optimization-based parameter estimation schemes variations of the optimal parameter estimates due to changes in operation conditions are typically neglected resulting in performance deterioration. In order to overcome this problem in this manuscript the design of an online parameter estimation approach for morphological PBs will be investigated for a L-glutamic acid crystallization process [5,6].

   This contribution is structured as follows. At first the general process model will be presented. Subsequently, the online parameter adaption laws will be derived directly from the infinite dimensional process model using a Lyapunov-based approach [7]. Next the performance of the proposed parameter estimation scheme will be shown assuming ideal measurements without noise. Further, it will be made clear that in case of realistic measurement errors the online adaption has to be combined with a filtering technique. At the end, the results are summarized and possible extensions for future research are mentioned.

Nomenclature

c	solute concentration $[g/l]$
$\hat{c}$	estimated solute concentration $[g/l]$
c^*	solubility $[g/l]$
e_n, e_c	error in the number density distribution/concentration between model and process
g_L, g_W	kinetic coefficients (exponents) for facet growth
G_L, G_W	facet growth rate in length/width dimension $[m/s]$
$\hat{G}_L, \hat{G}_W$	estimated facet growth rate in length/width dimension $[m/s]$
k_L, k_W	facet growth parameters $[m/s]$
$\hat{k}_L, \hat{k}_W$	estimated facet growth parameters $[m/s]$
$\tilde{k}_L, \tilde{k}_W$	error in facet growth parameters
L	length of β-form L-GA $[m]$
m_{10}, m_{01}	mean length/width of number density distribution $[m]$
$\hat{m}_{10}, \hat{m}_{01}$	estimated mean length/width of number density distribution $[m]$
n	number density distribution $[m^{-3}]$
$\hat{n}$	estimated number density distribution $[m^{-3}]$
$P_n^{obs}, P_c^{obs}, P_m^{obs}$	error feedback gains for model $[s^{-1}]$
q_L, q_W	kinetic coefficients (exponents) for facet growth
t	time $[s]$
T	temperature $[°C]$
W	width of β-form L-GA $[m]$

γ_L, γ_W	parameter adaption rate tuning parameters
ρ	crystal density $[kg\ m^{-3}]$
σ	relative supersaturation

2. Process modeling

In this manuscript seeded crystallization of β-form L-glutamic acid (L-GA) in a stirred tank reactor as presented in [5] is used as a benchmark problem. The shape of a single crystal can be described reasonably by the length L and width W of a parallelepiped (Fig. 1). Thus, depending on the ratio of both the crystal shape may vary between disks and needles. Assuming that the crystal growth is dominant and neglecting other effects like agglomeration, breakage and nucleation, the dynamics of the number density distribution $n(t, L, W)$ with respect to the two characteristic properties can be modeled using the following morphological PBM

$$\frac{\partial n(t, L, W)}{\partial t} + \frac{\partial}{\partial L}\{G_L(t, L, W)\ n(t, L, W)\} + \frac{\partial}{\partial W}\{G_W(t, L, W)\ n(t, L, W)\} = 0. \tag{1}$$

Here, the growth rates are given by

$$G_L(t, L) = k_{g_L}\ \sigma(t)^{g_L}\ L^{q_L},$$

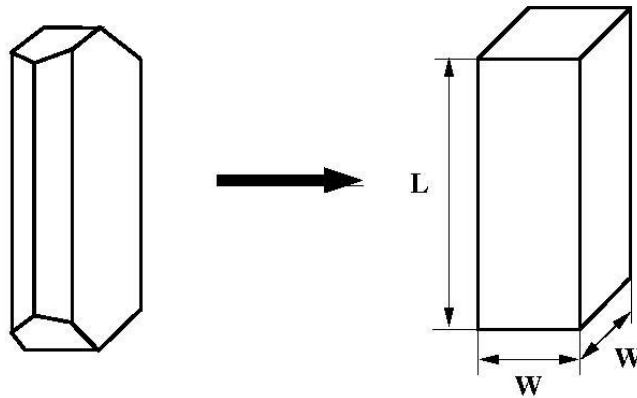$$G_W(t, W) = k_{g_W}\ \sigma(t)^{g_W}\ W^{q_W}. \tag{2}$$

Figure 1: Scheme of typical β form L-GA crystal and corresponding representation as parallelepiped [5]

It is assumed that each growth rate depends only on the crystal size in the corresponding dimension and on the relative supersaturation.

$$\sigma(t) = \frac{c(t) - c^*}{c^*} \tag{3}$$

The solubility of L-GA depends on the solute temperature and is given by the following empirical formula

$$c^* = 4.408 - 1.4644 \cdot 10^{-1}\,T + 1.786 \cdot 10^{-2}\,T^2 - 2.96366 \cdot 10^{-4}\,T^3 + 2.68138 \cdot 10^{-6}\,T^4. \tag{4}$$

Dynamics of the solute concentration can be derived from the mass balance of the liquid in the crystallizer and are given by

$$\frac{dc(t)}{dt} = -\rho \int_0^\infty \int_0^\infty (2\,W\,L\,G_W + L^2\,G_L)\,n(t,L,W)\,dL\,dW \tag{5}$$

where ρ is the crystal density. Obviously, the overall dynamical system consists of an ordinary differential equation (ODE) and partial differential equations (PDE), which are coupled. The numerical values of the parameters are given in Table 1.

Table 1: Process parameters and corresponding values

Parameter	Value	Parameter	Value
k_{g_L}	$0.6314 \cdot 10^{-6}$	k_{g_W}	$0.1943 \cdot 10^{-6}$
q_L	0.2106	q_W	0.2210
g_L	1.6602	g_W	1.5740
ρ	1540		

3. Design of the online parameter estimator

The majority of the parameter values listed in Table 1 have been identified from lab scale experiments. Thus, they can only be viewed as a rough orientation for a crystallization process on an industrial scale. Additionally, in a large scale industrial setting the process model may be not exact or the process parameters may vary during plant operation. Controller performance particularly suffers from those uncertainties as design procedure typically depend on a fully parameterized plant model. For this reason an online parameter identification procedure [7] will be designed.

The Lyapunov-based online estimation uses the following modified plant model, which runs in parallel to the actual process (see Fig. 2)

$$\frac{\partial \hat{n}}{\partial t} = -\frac{\partial}{\partial L}\{\hat{G}_L\,n\} - \frac{\partial}{\partial W}\{\hat{G}_W\,n\} + P_n^{obs}\,(\hat{n} - n),$$

$$\frac{d\hat{c}}{dt} = -\rho \int_0^\infty \int_0^\infty \left(2\,W\,L\,\hat{G}_W + L^2\,\hat{G}_L\right)n(t,L,W)\,dL\,dW + P_c^{obs}\,(\hat{c} - c). \tag{6}$$

where $\hat{n}$ and $\hat{c}$ are the particle shape distribution and the solute concentration estimated from the modified plant model. From this point on it is assumed that only the parameters k_{g_L} and k_{g_W} are unknown such that the unknown growth rates are given by

$$\hat{G}_L(t,L) = \hat{k}_{g_L}\,\sigma(t)^{g_L}\,L^{q_L}, \tag{7}$$

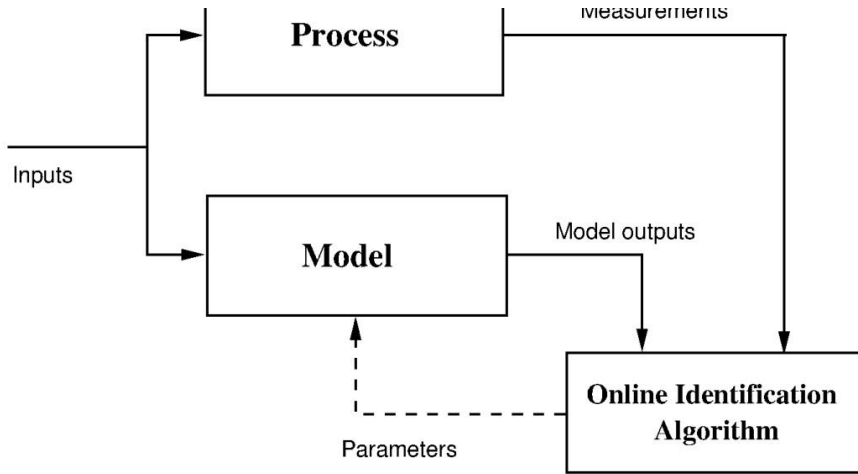$$\hat{G}_W(t, W) = \hat{k}_{gw}\, \sigma(t)^{gw}\, W^{qw}.$$

Figure 2: Online estimation scheme

The parameters P_n^{obs} and P_c^{obs} are additional tuning factors and can be interpreted as model error feedback gains. In a first step, the estimation errors of the crystal shape number distribution, the solute concentration and the parameters are introduced.

$$
\begin{aligned}
e_n &= \hat{n} - n \\
e_c &= \hat{c} - c \\
\tilde{k}_{g_L} &= \hat{k}_{g_L} - k_{g_L} \\
\tilde{k}_{g_W} &= \hat{k}_{g_W} - k_{g_W}
\end{aligned}
\tag{8}
$$

Combining (1) and (5-6) the error dynamics can be derived.

$$
\frac{\partial e_n}{\partial t} = -\frac{\partial}{\partial L}\{\tilde{k}_{g_L}\sigma^{g_L}L^{q_L}n\} - \frac{\partial}{\partial W}\{\tilde{k}_{g_w}\sigma^{gw}W^{qw}n\} + P_n^{obs}e_n
$$

$$
\frac{\partial e_c}{\partial t} = -\rho \int\limits_0^\infty \int\limits_0^\infty \left(2\,\tilde{k}_{g_w}\sigma^{gw}\,W^{1+qw}\,L\; +\tilde{k}_{g_L}\,\sigma^{g_L}\,L^{2+q_L}\,\right)n(t,L,W)\,dL\,dW + P_c^{obs}\,e_c
\tag{9}
$$

In order to design suitable adaption laws for the parameter estimates the following Lyapunov functional is chosen

$$
V = \tfrac{1}{2}\int_0^\infty \int_0^\infty e_n^2\, dL\, dW + \tfrac{1}{2}\, e_c^2 + \frac{1}{2\,\gamma_L}\,\tilde{k}_{g_L}{}^2 + \frac{1}{2\,\gamma_W}\,\tilde{k}_{g_w}{}^2.
\tag{10}
$$

Here, γ_L and γ_W are positive real tuning parameters. One can easily obtain that the Lyapunov function is positive definite and vanishes only for exact parameter estimates and if the shape number distribution and the solute

concentration of the model converge to the ones of the real plant. Applying standard Lyapunov stability theory yields that stability of the proposed estimator scheme can be achieved by guaranteeing that the first time derivative of V is negative semidefinite for all time points and vanishes for $V = 0$. Calculating the first time derivative of the Lyapunov functional along the system trajectory gives

$$
\frac{dV}{dt} = \iint P_n \, e_n^2 \, dL \, dW + P_c \, e_c^2
$$
$$
+ \tilde{k}_{g_L} \left(\frac{\dot{\hat{k}}_{g_L}}{\gamma_L} - \sigma^{g_L} \iint \frac{\partial L^{q_L} \, n}{\partial L} \{ \quad \} e_n \, dL \, dW - \rho \, \sigma^{g_L} e_c \iint L^{2+q_L} \, n \, dL \, dW \right)
$$
$$
+ \tilde{k}_{g_W} \left(\frac{\dot{\hat{k}}_{g_W}}{\gamma_W} - \sigma^{g_W} \iint \frac{\partial W^{q_W} \, n}{\partial W} \, e_n \, dL \, dW - 2 \, \rho \, \sigma^{g_W} e_c \iint L \, W^{1+q_W} \, n \, dL \, dW \right).
\tag{11}
$$

In order to guarantee the negative definiteness of the first time derivative of the Lyapunov functional V the adaption laws are chosen as

$$
\dot{\hat{k}}_{g_W} = \gamma_W \left(\sigma^{g_W} \iint \frac{\partial}{\partial W} \{ W^{q_W} \, n \} e_n \, dL \, dW + 2 \, \rho \, \sigma^{g_W} e_c \iint L \, W^{1+q_W} \, n \, dL \, dW \right),
$$
$$
\dot{\hat{k}}_{g_L} = \gamma_L \left(\sigma^{g_L} \iint \frac{\partial}{\partial L} \{ L^{q_L} \, n \} e_n \, dL \, dW + \rho \, \sigma^{g_L} e_c \iint L^{2+q_L} \, n \, dL \, dW \right),
\tag{12}
$$

Resulting in

$$
\frac{dV}{dt} = \int_0^\infty \int_0^\infty P_n \, e_n^2 \, dL \, dW + P_c \, e_c^2,
\tag{13}
$$

which is negative semidefinite for $P_n^{obs} < 0$ and $P_c^{obs} < 0$.

4. Evaluation of online identification procedure

The presented online parameter estimation approach has been implemented numerically using MATLAB/Simulink. For this reason the partial differential equations for the process (1) and the model (6) were transformed to a large-scale system of ordinary differential equations using a two-dimensional finite volume scheme. Details on the application of a finite volume scheme for the discretization of PDEs can be found for example in [8]. The double integrals were approximated using a two-dimensional trapezoidal rule. For the process the parameters given in Table 1 are used. The initial solute concentration is

$$
c(0) = 27.26 \, g/l
$$

and for the initial crystal shape number density distribution of the process is assumed to correspond to a two-dimensional normal distribution

$$
n(0, L, W) \sim N(\mu, \Sigma)
$$

with mean and covariances given as

$$
\mu = (10 \cdot 10^{-5}, 5 \cdot 10^{-5}), \qquad \Sigma = 10^{-10} \begin{pmatrix} 4 & 0 \\ 0 & 1 \end{pmatrix}.
$$

The cooling rate is chosen as $1.5\ K/min.$ It is assumed that the models initial values for shape number density distribution and solute concentration differ from the corresponding process values by a factor of 0.9. Furthermore, the initial values for the unknown parameters, i.e. the initial guesses, are only known very roughly.
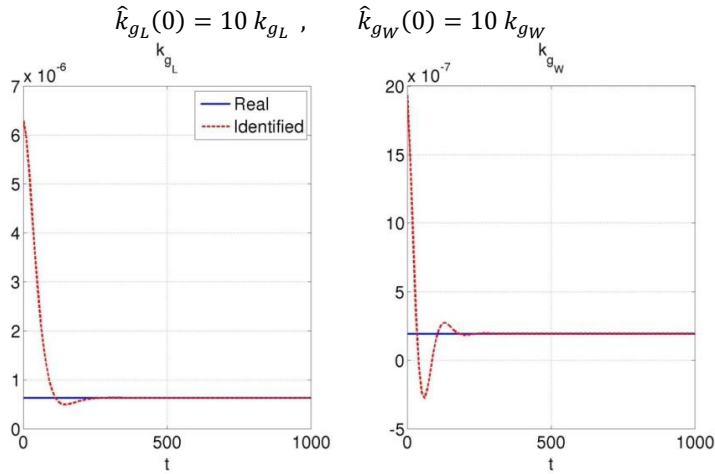
$$\hat{k}_{g_L}(0) = 10\ k_{g_L}\ , \qquad \hat{k}_{g_W}(0) = 10\ k_{g_W}$$

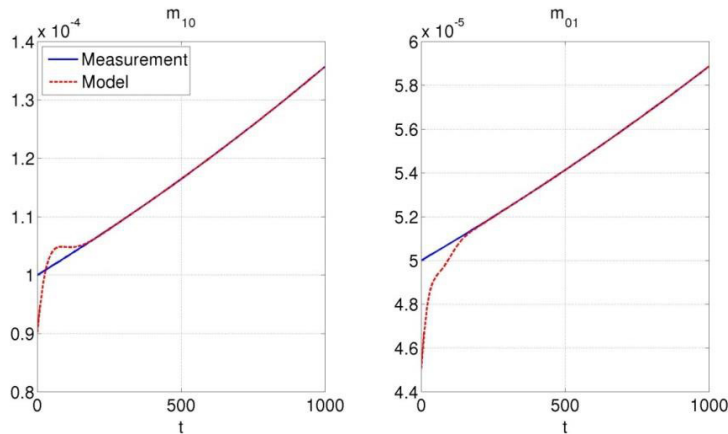
Figure 3. Parameter estimates for ideal measurements

Figure 4. Moment estimates for ideal measurements

4.1. Ideal measurements without noise

In a first step the performance of the online parameter estimation algorithm is shown for the case of ideal measurements without measurement noise. Though this scenario is not realistic as experimental data is always corrupted by measurement uncertainties it is well suited to show the general performance of the algorithm and to study the effects of different choices of the tuning parameters on the dynamics of the parameter estimates. Simulation studies indicate that the parameter estimates convergence rate mainly depends on the adaption rate factors γ_i while the ratio of error feedback factors and adaption rate factors affect the damping or the smoothness of

the parameter estimate dynamics. Thus, by an appropriate choice of the tuning parameters a reasonable convergence rate can be achieved. In Fig. 3 the corresponding parameter estimate dynamics are shown for

$$\gamma_L = \gamma_W = 1 \cdot 10^{-19}, P_n^{obs} = -0.05, P_c^{obs} = -0.025.$$

It can be seen that the parameter estimates converge to the real values within 250 s and are only slightly overshooting. Additionally the model crystal shape number density distribution converges to the one of the process reasonably fast, as shown in Fig. 4.

4.2. Noise corrupted measurements

In order to come up with a more realistic setup the measurements are now considered to be corrupted with additive white noise. Due to the noise the performance of the proposed online parameter estimation algorithm is expected to deteriorate. In order to achieve comparable performance as in the noise-free setting the model has been extended including the first order moments of the distribution

$$m_{10} = \int_0^\infty \int_0^\infty L \, n \, dL \, dW,$$

$$m_{01} = \int_0^\infty \int_0^\infty W \, n \, dL \, dW,$$

(14)

resulting in two additional model equations

$$\frac{d\hat{m}_{10}}{dt} = \hat{k}_{g_L} \sigma^{g_L} \int_0^\infty \int_0^\infty L^{q_L} \, n \, dL \, dW + P_m^{obs} \, (\hat{m}_{10} - m_{10}),$$

$$\frac{d\hat{m}_{01}}{dt} = \hat{k}_{g_W} \sigma^{g_W} \int_0^\infty \int_0^\infty W^{q_W} \, n \, dL \, dW + P_m^{obs} \, (\hat{m}_{01} - m_{01}).$$

(15)

Using the extended estimator model the Lyapunov function V has to be extended by errors in the two first moments

$$V = \frac{1}{2} \int_0^\infty \int_0^\infty e_n^2 \, dL \, dW + \frac{1}{2} \, e_c^2 + \frac{1}{2} (\hat{m}_{10} - m_{10})^2 + \frac{1}{2} (\hat{m}_{01} - m_{01})^2 + \frac{1}{2 \gamma_L} \tilde{k}_{g_L}^2 + \frac{1}{2 \gamma_W} \tilde{k}_{g_W}^2,$$

(16)

resulting in two extended adaptation laws

$$\dot{\hat{k}}_{g_W} = \gamma_W \left(\sigma^{g_W} \iint \frac{\partial W^{q_W} \, n}{\partial W} \, e_n \, dL \, dW + 2 \, \rho \, \sigma^{g_W} \, e_c \iint L \, W^{1+q_W} \, n \, dL \, dW - (\hat{m}_{01} - m_{01}) \, m_{q_W} \right),$$

$$\dot{\hat{k}}_{g_L} = \gamma_L \left(\sigma^{g_L} \iint \frac{\partial L^{q_L} \, n}{\partial L} \, e_n \, dL \, dW + \rho \, \sigma^{g_L} \, e_c \iint L^{2+q_L} \, n \, dL \, dW - (\hat{m}_{10} - m_{10}) \, m_{q_L} \right).$$

(17)

The corresponding simulation results can be seen in Fig. 6 and Fig. 7. As in the ideal measurement scenario, the parameter estimation errors decrease at first but, due to the stochastic measurement uncertainty, the parameter estimates do not converge to their real values. Instead the estimates themselves reflect the stochastic process behavior. Applying a simple filter is however sufficient to overcome this problem (dashed black lines).
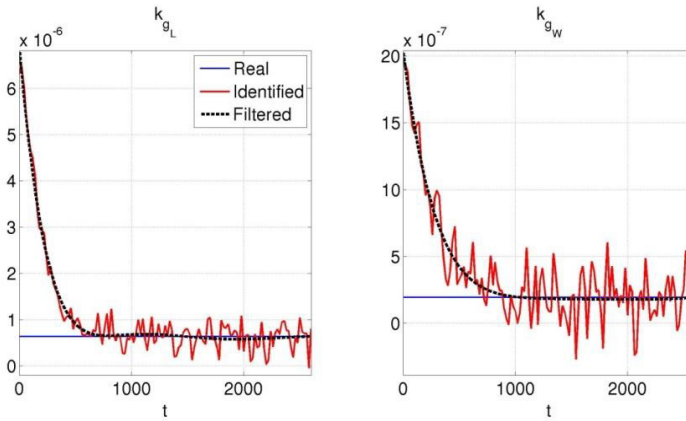

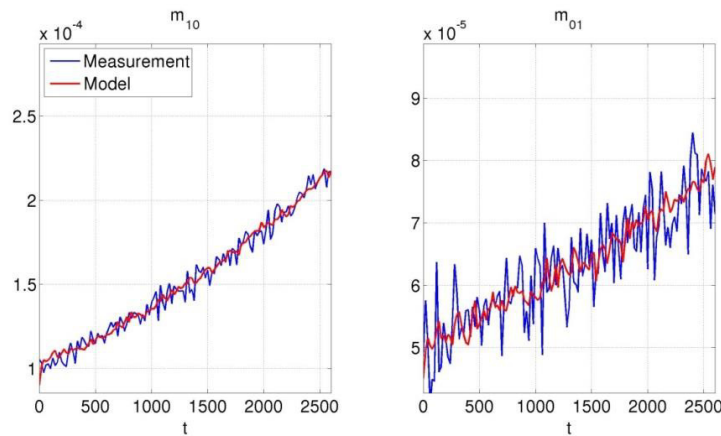Figure 5. Parameter estimates for noise corrupted measurements

Figure 6. Moment estimates for noise corrupted measurements

5. Summary/Conclusion, Future Work

In this manuscript online estimation of facet growth kinetics was studied for L-glutamic acid crystallization. It has been shown that the proposed Lyapunov-based adaption laws allow a reasonable fast estimation of the unknown model parameters in the case of ideal measurements. Additionally, an extension of the algorithm was presented to deal with noise corrupted measurements. Future work will be concerned with further analysis of tuning parameter effects on the estimation error dynamics as well as further analysis of the effects of stochastic measurements. In addition, the approach will be extended to the estimation of parameters depending directly on the shape dimensions.

Finally, the proposed online parameter estimation algorithm is planned to be used for real lab-scale experiments and as a part of an adaptive control scheme.

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