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Experimental Validation of Robust Process Design and Control Based on Gaussian Mixture Densities ⋆

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Abstract: In this contribution, the effects of different degrees of uncertainty description are investigated experimentally using an exothermic chemical reaction with safety constraint on the temperature. For that purpose, two robust trajectories are designed that respect the artificially created uncertainties of the experiments either coarsely using a single multivariate normal distribution (1GMD) or in a more detailed fashion using a Gaussian mixture density (GMD) consisting of 32 multivariate normal densities (32GMD). For the optimization, the uncertainties are propagated using the unscented transformation. Both trajectories were run 71 times in an open-loop manner. The more detailed trajectory (32GMD) leads to a 9 % higher yield without increasing the risk of constraint violation. Furthermore, many experimental realizations of two robust closed-loop process control schemes are being compared. They differ again only in the degree of the underlying uncertainty description. Although the frequent corrections of the controller marginalize the advantage of a more detailed stochastic process prediction, the 4GMD-controller still allows for 3 % more educt conversion compared to the 1GMD-controller.

Keywords: Uncertain Dynamic Systems, Nonlinear Systems, Gaussian Distributions, Probabilistic Simulation, Reactor Control, Safety-Critical, Robust control, Validation

1. INTRODUCTION

Technical processes are influenced by a variety of uncertain input and model parameters. The variation of these values often has a significant impact on the development a process. The specific process behavior is especially important when safety constraints come into play. While the exact values of the uncertain parameters are not known, their probabilistic description is often available. In most cases, normal densities are used to reflect the uncertain parameters. In order to obtain a prediction of the uncertainties, the normal densities have to be propagated through the nonlinear model equations. A numerically efficient stochastic simulation is the *Unscented Transformation* (UT) presented by *Julier* and *Uhlmann*. It represents each uncertain parameter by its mean value and two so-called sigma-points. The statistical moments of the output are calculated by solving the model equations for the mean vector as well as for all sigma-points and applying an explicit formula on these solutions. It thus corresponds to a gradient-free approximation of 2^{nd} order [Julier and Uhlmann (1996); Julier et al. (2000); Nørgaard et al. (2000); van der Merwe (2004)].

The method is limited, however, in that the process variables are only represented as normally distributed, and, thus, being symmetrically uncertain. In nonlinear systems, however, normally distributed inputs will inevitably lead to distorted, asymmetrical probability densities. In order to describe arbitrary process input densities, and to better account for effects in nonlinear density propagation a Gaussian mixture density (GMD) can be used [Rossner et al. (2010)]. As here each individual density has a lower variance, nonlinear deforming effects will be less pronounced during propagation. A linear combination of normal densities can be simulated by superposition of the *Unscented Transformations* of the individual densities. This method has been proposed before [Rossner et al. (2010)] to design robust processes. Results, as with other methods, have been presented so far only with simulation studies. Hence, the primary goal of this work is to give an experimental validation.

In this contribution, the impact of different degrees of uncertainty description, single multivariate normal density and Gaussian mixture density, on open- and closed-loop process control is investigated experimentally. For that purpose a fully-automated semibatch reactor for the catalytic decomposition of hydrogen peroxide H_2O_2 is set up. Defined disturbances on the initial amount of catalyst V_0 and the cooling temperature T_M are introduced to the individual process runs. Over all process runs these disturbance samples are normally distributed. Moreover, an upper safety-constraint on the reactor temperature is introduced. This limits the production rate of the exothermic process and the process result, thus, highly depends on the prediction of the probability density along that constraint. If an overestimation of its variance can be avoided, the production capabilities can be better exploited and more yield can be expected.

In the first part of this contribution, the fully-automated chemical reactor is presented and the underlying mathematical model is introduced. Subsequently, the experimental results of both robust open-loop process designs (TP), 1GMD-TP and 32GMD-

⋆ A version in German language of this contribution has been presented in "at Automatisierungstechnik 68, Issue 1, 2014, Page 14-22".

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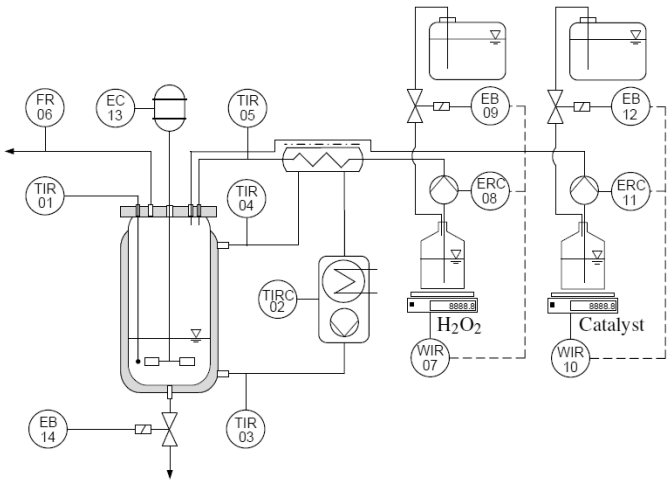

Fig. 1. P&I-Diagram of the automated reaction system. **01:** Reactor temperature. **02:** Thermostat temperature. **03:** Cooling temperature, supply. **04:** Cooling temperature, return. **05:** Feed Temperature. **06:** O_2 volume flow. **07:** Weight of buffer tank, H_2O_2. **08:** Voltage for dosing pump, Feed. **09:** Magnetic valve to storage tank of educt. **10:** Weight of buffer tank, catalyst. **11:** Voltage for dosing pump, catalyst. **12:** Magnetic valve to storage tank of catalyst. **13:** Motor voltage for stirrer. **14:** Magnetic valve to dump vessel.

TP, are shown. The more detailed prediction of process uncertainties using 32 normal densities leads to a 9 % higher productivity compared to the 1GMD design. Hereafter, online-optimization is introduced, and, thus, the results of the closed-loop process designs are presented. Due to the frequent corrections of the controller uncertainties have less impact on the process result. However, the control design based on the more detailed uncertainty description still leads to a 3 % higher yield compared to the one based on a single normal distribution. The contribution finally ends with a brief discussion of the obtained results.

2. EXPERIMENTAL SETUP

In order to run a large number of specifically disturbed processes with a high repeatability a fully-automated reactor system has been set up. This allows for running the semibatch reaction all around the clock, and, thus, enables the realization of all process runs in a timely manner. For each disturbance sample, the different control designs are run in an alternating fashion to ensure that remaining non-modeled disturbances affect both designs similarly.

Fig. 1 shows the experimental setup with all sensors and actuators using a piping and instrumentation (P&I) diagram. The jacket-cooled reactor has a diameter of $d = 0.1\,\mathrm{m}$ and holds a maximum of 2 L. When a process is initiated the disturbance samples for the initial volume of catalyst V_0 and the cooling temperature T_M are read from the database. In order to ensure an isoperibolic [1] process campaign, the cooling temperature T_M is send to the thermostat (02) and the initial volume V_0 is used as set-point for a pump-scale-controller of the catalyst. The dosage of the catalyst is realized with a tolerance of 0.1 g. In order to reach the starting reactor temperature $T(t_0) = T_\mathrm{M}$ more quickly, the stirrer (13) is activated with the dosage of the catalyst. Moreover, before the catalyst enters the reactor it is pre-cooled or -heated by a heat exchanger embedded in the reactor cooling circuit. This heat exchanger is also used to ensure a defined feed temperature $T_\mathrm{feed} = T_\mathrm{M}$. Once the temperature of the catalyst has reached the cooling temperature with a tolerance of $\Delta T_0 = 0.5\,\mathrm{K}$ the process is initiated. Now, the sensor signals are being written to the database using the sample interval Δt_meas. At the same time, the feeding profile q_f of the specific process run is read from the database and transferred to the feed-controller (07, 08) using a zero-order-hold scheme. The stirrer is used to homogenize the reaction mixture, to improve the heat transfer to the cooling jacket, and to avoid an over-saturation of dissolved oxygen for a more direct measurement of its evolution rate (06) during the model identification experiments (not shown). Once the end of the process t_end is reached, the reaction will be cooled down to the exit temperature T_ab and then released via the magnetic relief valve (14). The weight controlled buffer tanks for educt H_2O_2 and the catalyst are automatically refilled by opening the valves (9,12) to the tanks. Because the tanks are positioned higher than the buffer vessels this procedure is driven by hydrostatic pressure. A new process cycle is initiated as soon as the control system is connected to the next prepared database.

[1] The disturbance is constant during each process run. Over all runs these values are normally distributed.

3. PROCESS MODEL

The chemical decomposition of hydrogen peroxide (H_2O_2, Index 'hp') involves at least four substances. Due to the presence of potassium iodine (PI) the educt H_2O_2 will be catalytically decomposed into the products $H_2O^{(l)}$ (Index 'w') and $O_2^{(g)}$ (Index 'o'). Oxygen leaves the reactor as exhaust gas. The exothermic reaction has reaction rate r and reaction enthalpy ($-\Delta H_\mathrm{R}$). It can thus be written as:

$$\mathrm{H_2O_2} \xrightarrow[r]{\mathrm{PI}} \mathrm{H_2O} + \frac{1}{2}\mathrm{O_2} + (-\Delta H_\mathrm{R}). \tag{1}$$

In this reaction scheme, PI is modeled as a perfect catalyst that is not being consumed. This is a slight simplification compared to the more complex reaction scheme of the Bray-Liebhafsky reaction [Bray and Liebhafsky (1931); Liebhafsky and Mohammad (1933); Schmitz (2011)] which also involves other substances, e.g., the formulation of Iodine I_2. The reaction enthalpy in (1) is approximately $(-\Delta H_\mathrm{R}) = 100.4$ kJ/mol (Steudel et al. (2008)). Since both educt and catalyst concentration influence the reaction rate r, the rate has been modeled as a reaction of second order:

$$r = k_0\, e^{\frac{-E_\mathrm{A}}{RT}}\, c_\mathrm{hp}\, c_\mathrm{PI} \qquad \left[\frac{\mathrm{mol}}{\mathrm{L\,s}}\right]. \tag{2}$$

Here, c_hp and c_PI are the molar concentrations of hydrogen peroxide and potassium iodine, respectively. The constants $k_0 \approx 10^9\,\mathrm{L/mol/s}$ and $E_\mathrm{A} \approx 54\,\mathrm{kJ/mol}$ (Liebhafsky and Mohammad (1933)) represent the rate factor and the activation energy, respectively, and allow for the temperature depending description of the reaction rate according to *Arrhenius*. The factor k_0, however, will still be treated as a free parameter in order to create a degree of freedom for the model identification based on the obtained data.

Mole Balance

Based on the description of the reaction rate r the amounts of the reactants can be balanced. In a semibatch process the rate of the amount n_i, $\dot{n}_i$, of substance i depends on the reacting amounts $\dot{n}_{\mathrm{R},i}$ as well as the feeding $\dot{n}_{\mathrm{f},i}$: