

# A Nonlinear Observer for Semidetactable Chemical Reactions with Application to Kinetic-Rate-Constant Estimation<sup>1</sup>

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**Abstract**—In this paper we develop a nonlinear observer to estimate the concentration of reactants in a kinetic network when kinetic rate constants are unknown. When kinetic rate constants are unknown a steady-state error is present in the observer states. We thus develop an augmented observer that accounts for this steady-state error. We demonstrate this technique on numerical examples of increasing complexity, starting with a nominal case and then off-nominal cases, where input and output noise corrupt the available measurements, and finally when a kinetic rate constant evolves periodically over time with unknown dynamics.

## I. INTRODUCTION

The development of observers for nonlinear systems, within either a deterministic or stochastic framework, remains one of the most challenging and practically important problems in systems theory [1]. For the class of polynomial systems that model the concentrations of species of a chemical reaction network subject to mass-action kinetics, an observer is developed in [2]. Under a detectability assumption, the observer given in [2] provides global asymptotic observation of the steady-state concentrations, that is, the states of the observer converge, for all initial observer states and species concentrations, to the steady-state concentrations. These steady-state concentrations are generally nonzero due to the fact that the reactions of chemical networks are semistable, that is, the concentrations converge to nonzero values that depend on the initial values. Semistability for linear and nonlinear systems having a continuum of equilibria is studied in [3]–[5]. Applications to chemical kinetics and multi-agent consensus are developed in [6] and [7], respectively.

The dynamics of mass-action kinetics are governed by rate constants, and the observer developed in [2] is based on the assumption that these rate constants are known. In practice, however, rate constants may be unknown or approximately known, and it is of interest to develop an observer that can estimate these rate constants along with the species concentrations. As a step in this direction, it is possible to exploit the structure of mass-action kinetics by assuming that each rate constant is unity, where the role of the rate constant is played by an additional “fictitious” species that is constant. As expected, the observer developed in [2] fails for this

augmented reaction network due to the lack of detectability. In fact, an observer implemented for the augmented reaction network fails to estimate the unknown rate constant and, in addition, provides erroneous estimates of the steady-state concentrations.

In the present paper, we present a modification of the observer given in [2] that exploits the fact that the undetectable concentrations of the augmented network are constant. In fact, the augmented network possesses a property that we call *semidetactability*. By modifying the observer of [2], we demonstrate, for a chemical network with one unknown rate constant, the ability to estimate both the asymptotic species concentrations and the unknown rate constant. For a reaction network with two unknown rate constants, we show that an affine relationship involving the rate constants can be construed from the observer states.

The contents of the paper are as follows. In Section 2 we review the results of [2] on criteria for detectability of chemical reactions, and summarize the observer given in [2]. In Section 3 we present the McKeithan network and demonstrate the ability to observe the reactant concentrations through the output map. Next assuming that one of the kinetic rate constants is unknown, the McKeithan network is reformulated in an equivalent form where the unknown constant is assumed to be a fictitious reactant. Section 4 presents a criterion for semidetactability as well as an augmented observer for estimating the concentration of the reactants and the unknown rate constant. The augmented observer is demonstrated on several numerical examples. Section 5 explores the issue of observing reactant concentrations when more than one rate constant is unknown.

## II. PROBLEM FORMULATION

Consider the reaction network

$$AX \xrightarrow{r} BX, \quad (1)$$

where  $A, B \in \mathbb{R}^{q \times n}$ ,  $X = [X_1 \dots X_n]^T$  is a vector of species, and  $r = [r_1 \dots r_q]^T$  is a vector of kinetic rate constants. The vector of concentrations  $x = [x_1 \dots x_n]^T$ , where  $x_i$  is the concentration of the species  $X_i$ , satisfies

$$\dot{x} = f(x), \quad x(0) = x_0, \quad (2)$$

where

$$f(x) = (B - A)^T [r \circ x^A(t)], \quad (3)$$

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where  $\circ$  is the Schur product. The notation  $x^A$  denotes the vector in  $\mathbb{R}^q$  whose  $i$ th component is the product  $x_1^{A_{i1}} \dots x_q^{A_{iq}}$ . Alternatively, defining  $R = \text{diag}(r_1, \dots, r_n)$ , (3) can be written as

$$\dot{x} = (B - A)^T R x^A(t), \quad x(0) = x_0. \quad (4)$$

The output function  $y = h(x)$  is a mapping  $h : \mathbb{R}^n \rightarrow \mathbb{R}^p$  of the form

$$h(x) = x^C = \begin{bmatrix} x_1^{c_{11}} x_2^{c_{12}} \dots x_n^{c_{1n}} \\ \vdots \\ x_1^{c_{p1}} x_2^{c_{p2}} \dots x_n^{c_{pn}} \end{bmatrix} \in \mathbb{R}^p, \quad (5)$$

where

$$C = \begin{bmatrix} c_{11} & c_{12} & \dots & c_{1n} \\ \vdots & \vdots & \vdots & \vdots \\ c_{p1} & c_{p2} & \dots & c_{pn} \end{bmatrix} \in \mathbb{R}^{p \times n}. \quad (6)$$

#### A. Detectability

Let  $\xi$  satisfy

$$\dot{\xi} = f(\xi), \quad \xi(0) = \xi_0. \quad (7)$$

*Definition 1:* The system (2) is detectable if, for every pair of trajectories  $x(t)$  and  $\xi(t)$  of (2) and (7), respectively, such that  $x_0$  and  $\xi_0$  are nonnegative and such that  $h(x(t)) = h(\xi(t))$  for all  $t \geq 0$ , it follows that  $x(t) - \xi(t) \rightarrow 0$  as  $t \rightarrow \infty$ .

Loosely speaking, detectability means the unobservable subspace of (2) is asymptotically stable. The following result is proven in [2]. Let  $\mathcal{R}$  and  $\mathcal{N}$  denote range and null space, respectively.

*Theorem 1:* The system (2) is detectable if and only if

$$\mathcal{R}(B^T - A^T) + \mathcal{R}(C^T) = \mathbb{R}^n. \quad (8)$$

Note that (8) is equivalent to

$$\mathcal{N}(B - A) \cap \mathcal{N}(C) = \{0\}, \quad (9)$$

and, if (2) is detectable, then

$$\text{rank } C \geq n - \text{rank}(B - A), \quad (10)$$

However, (10) does not imply (8). The following result is proved in [2].

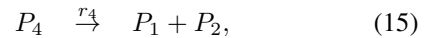
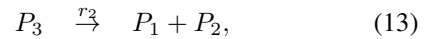
*Theorem 2:* Consider the system (2) with solution  $x(t)$ , assume that (8) is satisfied, and consider the observer

$$\dot{z} = f(z) + C^T[h(x) - h(z)], \quad (11)$$

where  $z(0)$  is nonnegative. Then  $x(t) - z(t) \rightarrow 0$  as  $t \rightarrow \infty$ .

### III. OBSERVER CONSTRUCTION FOR THE MCKEITHAN NETWORK

Consider the McKeithan network [2]



where  $P_i$  are reactants. The corresponding matrices  $A$ ,  $B$ , and  $R$  are

$$A = \begin{bmatrix} 1 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \end{bmatrix}, \quad B = \begin{bmatrix} 0 & 0 & 1 & 0 \\ 1 & 1 & 0 & 0 \\ 1 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix}, \quad (16)$$

$$R = \text{diag} [ r_1 \quad r_2 \quad r_3 \quad r_4 ]. \quad (17)$$

Using  $A$ ,  $B$ ,  $R$  with (3) we obtain

$$\dot{x}_1 = -r_1 x_1 x_2 + r_2 x_3 + r_4 x_4, \quad (18)$$

$$\dot{x}_2 = -r_1 x_1 x_2 + r_2 x_3 + r_4 x_4, \quad (19)$$

$$\dot{x}_3 = r_1 x_1 x_2 - (r_2 + r_3) x_3, \quad (20)$$

$$\dot{x}_4 = r_3 x_3 - k_4 x_4. \quad (21)$$

Letting

$$h(x) = [x_1 x_2^2 \quad x_1 x_4]^T, \quad (22)$$

it follows that

$$C = \begin{bmatrix} 1 & 2 & 0 & 0 \\ 1 & 0 & 0 & 1 \end{bmatrix}. \quad (23)$$

Since  $\text{rank}(B - A) = 2$  and  $\text{rank } C = 2$  it follows that (10) is satisfied. Furthermore, (8) holds, and thus (18)–(22) is detectable. We use the observer (11) with  $r = [6 \quad 0.5 \quad 7 \quad 1]^T$ ,  $x_0 = [1 \quad 3 \quad 3 \quad 2]^T$ , and  $z_0 = [2 \quad 25 \quad 20 \quad 1]^T$ . The concentrations of the species and the states of the observer are shown in Figure 1.

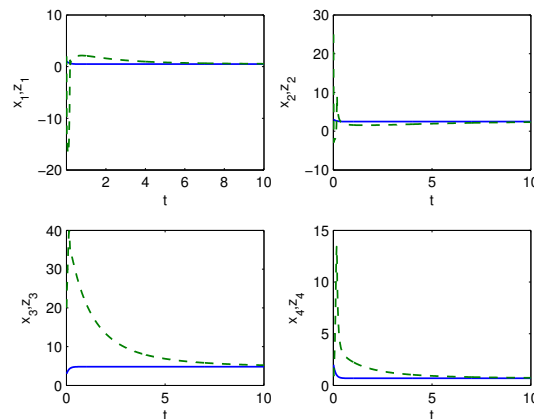
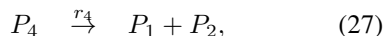
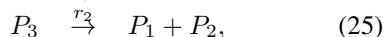
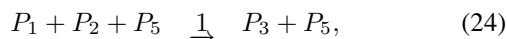


Fig. 1. Comparison of the concentrations  $x$  (solid line) with observer states  $z$  (dashed line)

Next we consider (12)–(15), where  $r_1$  is unknown. To

estimate  $r_1$  we reformulate the McKeithan network in the equivalent form



where the rate constant of (24) is 1, and  $r_1$  is given by the fictitious concentration  $P_5$ . The matrices  $A$  and  $B$  from (18)–(21) become

$$A = \begin{bmatrix} 1 & 1 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 \end{bmatrix}, \quad B = \begin{bmatrix} 0 & 0 & 1 & 0 & 1 \\ 1 & 1 & 0 & 0 & 0 \\ 1 & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 1 \end{bmatrix}, \quad (29)$$

$$R = \text{diag} [ 1 \quad r_2 \quad r_3 \quad r_4 \quad 1 ], \quad (30)$$

and thus the concentrations satisfy

$$\dot{x}_1 = -x_5 x_1 x_2 + r_2 x_3 + r_4 x_4, \quad (31)$$

$$\dot{x}_2 = -x_5 x_1 x_2 + r_2 x_3 + r_4 x_4, \quad (32)$$

$$\dot{x}_3 = x_5 x_1 x_2 - (r_2 + r_3) x_3, \quad (33)$$

$$\dot{x}_4 = r_3 x_3 - k_4 x_4, \quad (34)$$

$$\dot{x}_5 = 0. \quad (35)$$

Note that (31)–(34) are identical to (18)–(21) with the exception that  $r_1$  is replaced by  $x_5$ , which by (35) is constant. Since  $\text{rank}(B - A) = 2$ , (10) is satisfied only if  $\text{rank } C \geq 3$ . More seriously, note that if (8) is satisfied, then  $h(x)$  must be a function of  $x_5$ ; otherwise (8) always fails. Since  $r_1$  is unknown and cannot be an argument of  $h(x)$  it follows that (31) – (35) cannot be detectable.

Although (31)–(35) and (22) do not satisfy the detectability requirements, we simulate the observer system (11) to examine the steady-state error. Figure 2 shows that all of the observer's states have a steady-state error. Furthermore, Figure 3 shows that the observer state  $z_5$ , which is the rate constant estimate, remains at its initial value.

#### IV. SEMIDETECTABILITY

The failure in detectability for (31) – (22) is due to the inclusion of the dynamics of the rate constant, specifically,  $\dot{x}_5 = \dot{r}_1 = 0$ . We now introduce the concept of semidetectability, which refers to a system whose unobservable subspace is semistable.

*Definition 2:* The system (2) is semidetectable if, for every pair of trajectories  $x(t)$  and  $\xi(t)$  of (2) and (7), respectively, such that  $x_0$  and  $\xi_0$  are nonnegative and such that  $h(x(t)) - h(\xi(t))$  is constant for all  $t \geq 0$ , it follows that  $\lim_{t \rightarrow \infty} (x(t) - \xi(t))$  exists.

For the following result, assume that for a kinetic reaction network with  $n$  concentrations, the last  $n_r$  equations describe

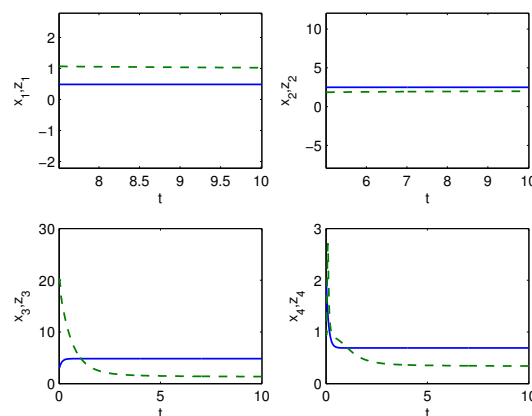


Fig. 2. Comparison of the concentrations  $x$  (solid line) and the observed system states  $z$  (Dashed line)

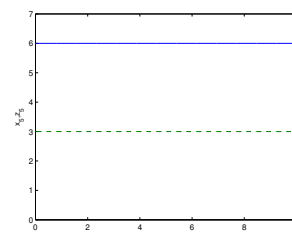


Fig. 3. Comparison of the concentration  $x_5$  (solid line) and the observed system history  $z_5$  (Dashed line)

the time rate of change of the unknown rate constants, which are necessarily equal to zero. Let  $e_i$  denote the  $i$ th row of the identity matrix.

*Theorem 3:* Assume that

$$\mathcal{N}(B - A) + \mathcal{N}(C) = \mathcal{R}([e_{n-n_r+1} \cdots e_n]), \quad (36)$$

where  $n_r$  is the number of unknown rate constants, and

$$\text{rank } C \geq n - \text{rank}(B - A). \quad (37)$$

Then (2) is semidetectable

We now modify (11) to account for the steady-state error by introducing a function  $\delta$  of the measurement residual. Let  $\|\cdot\|$  denote the Euclidean norm.

*Theorem 4:* Assume that (2) is semidetectable, and let  $n_r = 1$ . Consider the observer

$$\dot{z} = f(z, \hat{r}) + C^T (h(x) - h(z)), \quad (38)$$

$$\dot{\hat{r}}_1 = \delta(h(z) - h(x)), \quad (39)$$

where

$$\delta(h(x) - h(z)) = \text{sign} \left( \sum_{i=1}^p [h_i(z) - h_i(x)] \right) \|h(x) - h(z)\|, \quad (40)$$

and  $z(0)$  and  $\hat{r}_1(0)$  are nonnegative. Then  $x(t) - z(t) \rightarrow 0$  and  $\hat{r}_1(t) \rightarrow r_1$  as  $t \rightarrow \infty$ .

### A. Numerical Examples

In the following simulations the true initial state is  $x_0 = [1 \ 3 \ 3 \ 2 \ 6]^T$  and the initial observer state is  $z_0 = [2 \ 25 \ 20 \ 1 \ 3]^T$ . Furthermore, the rate constants are  $r = [1 \ 0.5 \ 7 \ 1 \ 1]^T$  and the measurements are  $h(x) = [x_1 x_2^2 \ x_1 x_4 \ x_2 x_4]^T$ . We evaluate the performance of the observer for several scenarios including the nominal case with zero noise as well as in the presence of white process and measurement noise. Finally, we test the observer in the presence of a disturbance that creates an unknown time-varying kinetic rate constant.

Equations (2) and (5) are the nominal chemical reaction system. Reconsider (2) and (5) as

$$\dot{x} = f(x) + w, \quad (41)$$

$$y = h(x) + v, \quad (42)$$

where  $f$  is given by (3),  $y \in \mathbb{R}^p$ , and  $w \in \mathbb{R}^n$ ,  $v \in \mathbb{R}^p$  are process and measurement noise, respectively. For the nominal case  $v = w = 0$ .

*Example 4.1:* For the nominal case we choose  $w = v = 0$ . The concentration histories of the reactants are

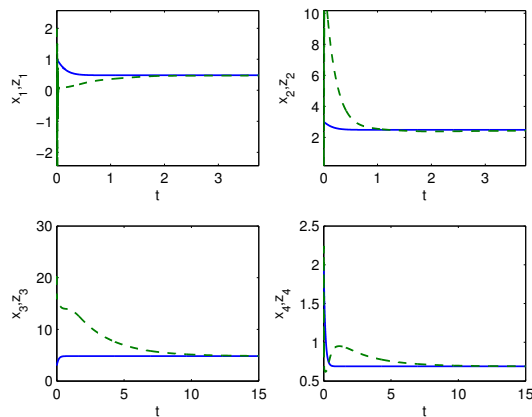


Fig. 4. Comparison of the concentrations  $x$  (solid line) and the observed system history  $z$  (dashed line)

shown in Figure 4, and the kinetic rate constant estimate is shown in Figure 5. For this example, the observer drives

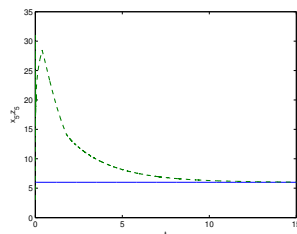


Fig. 5. Comparison of the concentration  $x_5$  (solid line) and the observed system history  $z_5$  (dashed line)

the steady-state error to zero. Furthermore, since the rate constant estimate approaches the true value, it follows that  $h(x(t)) - h(z(t)) \rightarrow 0$  as  $t \rightarrow \infty$ .

*Example 4.2:* For the off-nominal case we include process and measurement noise. The noise is zero mean, with a signal to noise ratio of 20, where the amplitude of the process noise is determined by evaluating the standard deviation of the chemical concentrations over the time period and the amplitude of the process noise is the concentration standard deviation divided by the SNR. The concentrations of the

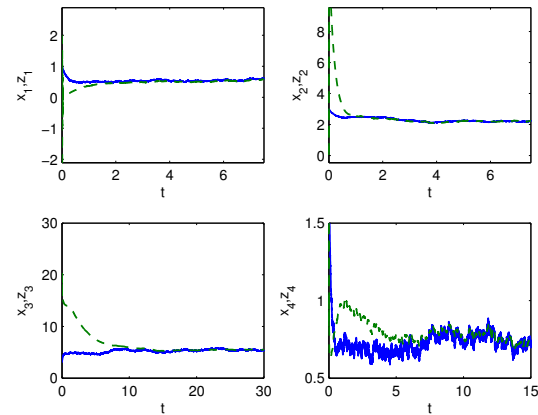


Fig. 6. Comparison of the concentrations  $x$  (solid line) and the observed system history  $z$  (dashed line)

reactants are shown in Figure 6, and the kinetic rate constant estimate is shown in Figure 7. In the presence of noise, the

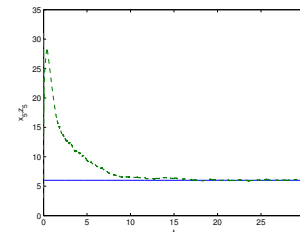


Fig. 7. Comparison of the concentration  $x_5$  (solid line) and the observed system history  $z_5$  (dashed line)

observer estimates the kinetic rate constant asymptotically and without offset.

*Example 4.3:* Reconsider (41) and (42) with the addition of a periodic disturbance. The system becomes

$$\dot{x} = f(x) + w + d, \quad (43)$$

$$y = h(x) + v, \quad (44)$$

where  $d$  is an exogenous disturbance. This case is motivated by chemical reactions of space weather systems where kinetic rate constants can fluctuate throughout a given period depending on a number of factors including exposure to the sun, and fluctuations in temperature [8]. In this example the rate constant is perturbed by an unknown sinusoidal process  $d(t) = 0.0005\sin(2\pi t 0.0001)$ . The concentration history of the reactants is shown in Figure 10, and the kinetic rate constant estimate is shown in Figure 11.

*Example 4.4:* This example demonstrates observer performance in the presence of a disturbance to the rate

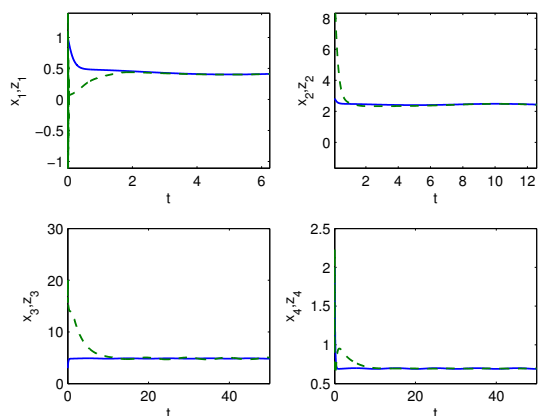


Fig. 8. Comparison of the concentrations  $x$  (solid line) and the observed system history  $z$  (dashed line)

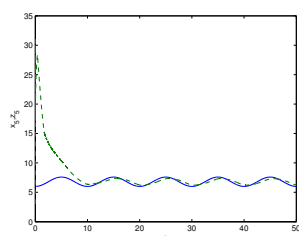


Fig. 9. Comparison of the concentration  $x_5$  (solid line) and the observed system history  $z_5$  (dashed line)

constant and in the presence of noisy measurements. The rate constant is perturbed by an unknown sinusoidal process given by  $d(t) = 0.0005\sin(2\pi t 0.0001)$ . Furthermore the states and measurements are disrupted by white noise with a signal to noise ratio of 20. The concentration history of

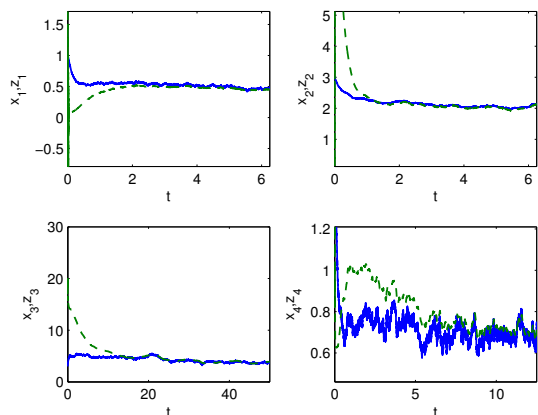


Fig. 10. Comparison of the concentrations  $x$  (solid line) and the observed system history  $z$  (dashed line)

the reactants are shown in Figure 10, and the kinetic rate constant estimate is shown in Figure 11.

## V. MULTIPLE UNKNOWN RATE CONSTANTS

We now use Theorems 3 and 4 to observe the concentrations of the reactants when two rate constants are unknown.

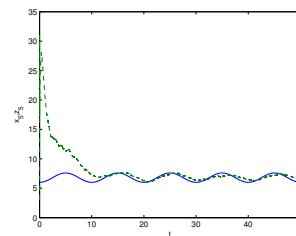
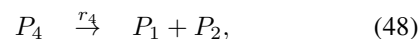
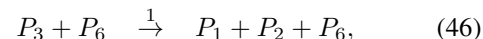
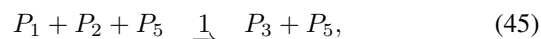


Fig. 11. Comparison of the concentration  $x_5$  (solid line) and the observed system history  $z_5$  (dashed line)

We thus reformulate the McKeithan network as



whose concentrations satisfy

$$\dot{x}_1 = -x_5x_1x_2 + x_6x_3 + r_4x_4, \quad (50)$$

$$\dot{x}_2 = -x_5x_1x_2 + x_6x_3 + r_4x_4, \quad (51)$$

$$\dot{x}_3 = x_5x_1x_2 - (x_6 + r_3)x_3, \quad (52)$$

$$\dot{x}_4 = r_3x_3 - k_4x_4, \quad (53)$$

$$\dot{x}_5 = 0, \quad (54)$$

$$\dot{x}_6 = 0. \quad (55)$$

To satisfy Theorem 3 we choose

$$h(x) = [x_1x_2^2 \quad x_1x_4 \quad x_2x_4 \quad x_3^2]^T, \quad (56)$$

and to implement Theorem 4 with  $n_r = 2$  we choose

$$\dot{\hat{r}} = \begin{bmatrix} \delta(h(z) - h(x)) \\ 0 \end{bmatrix}, \quad (57)$$

where  $\hat{r} = [\hat{r}_1 \quad \hat{r}_2]^T$ . We simulate the system with  $x_0 = [1 \ 3 \ 3 \ 2 \ 7 \ 2]^T$ ,  $z_0 = (2 \ 25 \ 20 \ 1 \ 3 \ 4)$ , and  $r = [1 \ 1 \ 7 \ 1 \ 1]^T$ . Figure 12 shows that the observer (38) and (57) asymptotically observes the concentrations of the reactants, despite two unknown rate constants. Figure 13 shows that the asymptotic value of the estimate  $x_5$  is not equal to  $r_1$ . Furthermore,  $x_6$ , which is  $\hat{r}_2$ , does not change since there are no dynamics associated with it. These results suggest that, since the combination of  $\hat{r}_2$  and the asymptotic value of  $\hat{r}_1$  satisfy  $h(z(t)) - h(x(t)) \rightarrow 0$  as  $t \rightarrow \infty$  it follows that there exist multiple solutions for the unknown rate constant pair. We test this conjecture by analyzing the observer equations, which are analogous to (50) – (55) at steady state, that is,  $\dot{z} = 0$ . We reformulate the steady-state concentrations and kinetic rate constants as

$$Qr = 0, \quad (58)$$

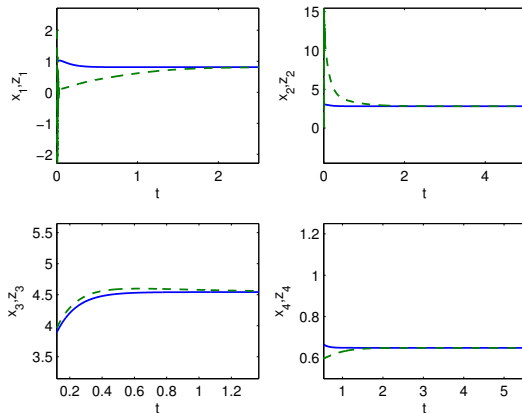


Fig. 12. Comparison of the concentrations  $x$  (solid line) and the observed system history  $z$  (dashed line)

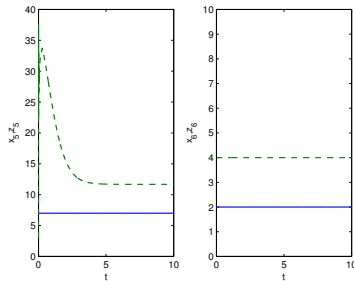


Fig. 13. Comparison of the concentration  $x_5$  (solid line) and the observed system history  $z_5$  (dashed line), the concentration of  $x_6$  and the observed system history  $z_6$  is also shown.

where  $r = [\hat{r}_1, \hat{r}_2, 1, 7, 1, 1]^T$  and

$$Q = \begin{bmatrix} -z_{1,ss}z_{2,ss} & z_{3,ss} & 0 & z_{4,ss} & 0 & 0 \\ -z_{1,ss}z_{2,ss} & z_{3,ss} & 0 & z_{4,ss} & 0 & 0 \\ z_{1,ss}z_{2,ss} & -z_{3,ss} & -z_{3,ss} & 0 & 0 & 0 \\ 0 & 0 & z_{3,ss} & -z_{4,ss} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{bmatrix}. \quad (59)$$

Solving (58) for  $\hat{r}_1$  and  $\hat{r}_2$ , yields an affine subspace of possible unknown rate constant pairs, which for this case is given by

$$\hat{r}_1 = \frac{z_{3,ss}}{z_{1,ss}z_{2,ss}}\hat{r}_2 + \frac{7z_{4,ss}}{z_{1,ss}z_{2,ss}}. \quad (60)$$

Substituting the steady state values obtained from the observer, the affine subspace for this example is

$$\hat{r}_1 = 2.33\hat{r}_2 + 2.33. \quad (61)$$

Figure 14 shows the loci of rate-constant combinations that satisfy (58). The initial rate constant guess along with the true and observed values are also plotted in Figure 14. Since  $z_6$  does not change,  $z_5$  tends to the value on the loci such that the pair are a solution to (58).

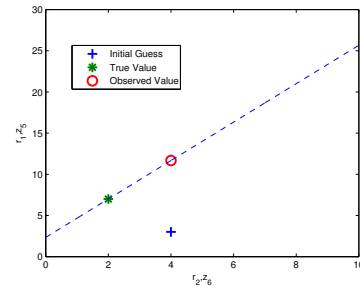


Fig. 14. Affine loci of possible unknown rate constant pairs.

## VI. CONCLUSION

We have demonstrated a method for estimating a single unknown rate constant in a chemical reaction. In order to estimate a reaction rate constant the constant is treated as a fictitious reactant that participates in the chemical reaction network. It is demonstrated that by using this method, the system of differential equations that describe the reaction is not detectable. However, these augmented systems are semidetectable, meaning the limit of the observation residual exists. Based on this knowledge a nonlinear observer is constructed to account for the steady-state error. The observer robustness was tested under a variety of conditions, including influence due to white noise and exogenous disturbances to the rate constant, which is motivated by space weather systems. For chemical concentrations with multiple unknown rate constants, the modified observer can estimate the unknown rate constants up to an affine subspace determined by the asymptotic estimate of the species concentrations.

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