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Temperature control via robust compensation of heat generation: Isoparaffin/olefin alkylation

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Abstract

The temperature control of the heterogeneous alkylation, where H_2SO_4 is used as catalyst, is discussed in this paper. The problem is to design a control function to stabilize temperature in face of uncertain kinetic model. The proposed controller, whose structure resembles a PI-controller, is based on energy balance. The resulting feedback controller is robust, leads to an acceptable performance, and computes the temperature of the coolant from the refrigeration section to the reactor. The effectiveness and robustness of the designed controller for computing the coolant temperature is tested by means of simulations in such a manner that we study the effects of: (i) load disturbances, (ii) model uncertainties and (iii) initial conditions for estimation values.

Keywords: Alkylation reactors; Temperature control

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1. Introduction

The catalytic alkylation involves the addition of an isoparaffin containing a tertiary hydrogen to an olefin. Basically, alkylation process combines olefines (propylene, butylenes and/or amylenes) with paraffins, as isobutane, in presence of strong acid to produce high octane branched chain hydrocarbons. Thus, mixtures of isoparaffin, C_5–C_{16}, are alkylate products and can be divided in five families: (i) trimethylpentanes (TMP's), (ii) light ends (less than C_5–C_7 isoparaffin), (iii) dymethylhexanes (DMH's), (iv) heavy ends (higher than C_{10}) and (v) acid-soluble hydrocarbons, as conjunct polymers, ester, red oil, etc. The process is used by petroleum industry to prepare highly branched paraffins. In last 15 years, new alkylation plants were built. About 70% of this alkylate product is or will be produced using H_2SO_4 as catalyst (these new plants announcement agree well with information reported along 1990s in *Hydrocarbon Processing* journal). There are some remainder plants where HF is used as catalyst, however HF is a very hazard catalyst. Thus, although HF unit operation is well understood and there has been a large effort to ensure safe handing of HF catalyst [1]; however, H_2SO_4 units are preferable due to secure operation. Moreover, alkylation with sulfuric acid as catalyst has been studied early 1970s (see, for instance, [2]). On the other hand, large efforts are being performed to develop acid-solid catalyst (see, for instance, Simpson et al. [3], Mantilla-Ramírez et al. [4] or Clark and Subramaniam [5]); however, results in this direction are not complete yet. Although much progress has been made during past 50 years for understanding the complicated chemistry of alkylation, control of alkylation process is not an easy task if we think that alkylation chemistry is significantly different when distinct catalyst is used or when operating conditions change [6,7].

Alkylation is a quite complex chemical process in which control can result in economic and environmental benefits. There are different control problems for an alkylation plant; for instance, regulation of isoparaffin/olefin ratio, reactor temperature stabilization, etc. Alkylation involves complex kinetics and mass transfer and one of the difficulties of this class of complex chemical systems is that the kinetic terms and/or mass transfer

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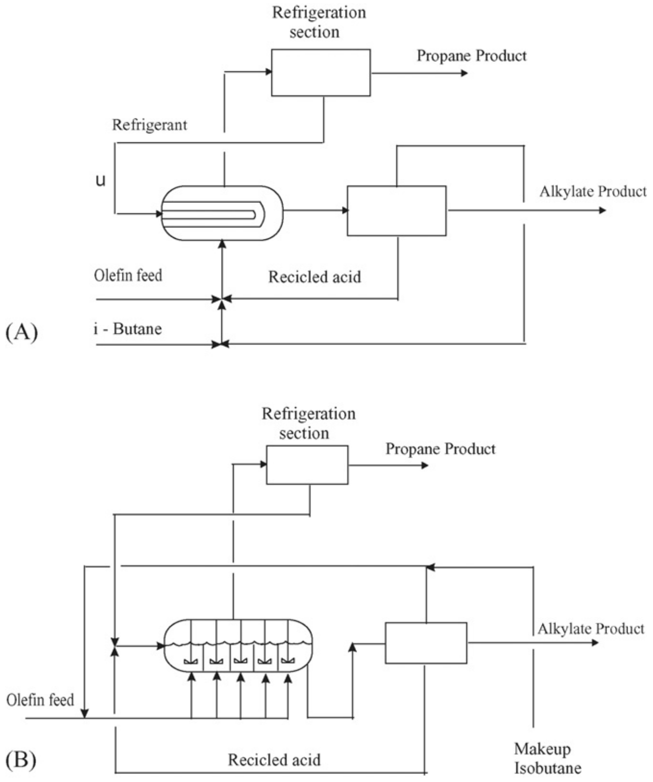

Fig. 1. Configuration or alkylation reactors. The refrigeration system basically consists in compressor and depropanizer. The main contribution is the design of an approach to the robust control temperature via heat reaction compensation. Thus, dynamical behavior of the refrigeration system is not considered.

phenomena can be required by the feedback function to compute control action (e.g., heat generation terms can be required for stabilizing temperature [8]). A refrigeration section is required to regulate the reactor temperature because alkylation reaction is highly exothermic. Indeed, reactor should be maintained at uniform temperature; i.e., temperature fluctuation less than 1 °C. Several operations can be comprised into the refrigeration system [9]. The reaction section can be constructed as contactor (STRATCO) or as a reactor chain (EXXON). However refrigeration system basically consists of compressor and depropanizer (see Fig. 1). That is, in the *refrigeration system*, the hydrocarbons that are vaporized due to the heat reaction are routed into the refrigeration compressor and, once compressed and depropanized, are returned to the reaction section. Due to very complex chemistry of alkylation, high computational effort can be required to compute the heat generation terms in such a manner that the refrigeration system reach the computed temperature for stabilizing the plant. Although high computational technology is on hand, high computational effort is undesirable. Then, a function with simple computational procedures but robustness is required to control the alkylation reactor. On the other hand, alkylation process involves mass and heat recycle streams (see Fig. 1). Recycle streams are not feedback in the control sense. Recycle streams increases the order of the characteristic polynomial with subsequent complexity. In fact, process with recycle becomes unclear at present. Few efforts has been recently devoted to understand this dynamics and control of such kind of process (see [10] and references therein; also [11–15]). Luyben [10] showed that a recycle system requires a large time constant (related to residence time) because as recycling-rate increases as instability can be induced. That is, the time response of the recycle systems is needed large in order to open-loop dynamics remains stable. Hence, plants for recycle are designed with large tanks for secure operation. Large tanks isolates sequences of units and permits the use of cascade control configuration; i.e., each downstream unit (inner loop) simply *sees* disturbances coming from its upstream loop (external loop). If such disturbances are attenuated, the reactor can be stabilized. Results in regard to dynamics and control of recycle systems are promissory; however, up today, cascade scheme is, by secure operation, best option for controlling "wide plant".

In this work, the goal is to design a control function in such a manner that neither the reaction heat nor kinetic nor mass transfer terms are required for stabilizing temperature. The scheme provides an estimated value of the heat generation from energy balance. Alkylation isobutane/propylene using sulfuric acid as catalyst [2] was chosen to illustrate the design of a control function for complex systems. An estimated value of the heat generation is obtained from on line measurements (output feedback) and the proposed controller does not require full information. Indeed, the proposed feedback comprises an estimation algorithm based on closed-loop heat balance. The proposed controller exploits similarities among the alkylation processes for obtaining the estimated value of heat generation by reaction. The key similarities are: (i) alkylation reaction is exothermic. Therefore, the control function represents the heat removal required for isothermic operation, i.e., the control action is *non-negative*; (ii) mass transfer from hydrocarbon to acid solution restricts the concentration in acid solution and reaction occurs in acid phase, i.e., the inlet concentration in reaction phase is a bounded time function (see below); and (iii) since wide-plant process comprises very large tanks, the process is characterized by very slow dynamics, i.e., the time constant is large. Thus, the control function in this paper computes the coolant temperature such that the alkylation reactor is stable and can be used in cascade control. The idea behind the proposed controller is that the computed coolant temperature enters to the refrigeration section as reference. The main contribution of the paper is to perform the reactor temperature regulation with least prior knowledge about the heat generation by reaction (i.e., neither kinetic nor thermodynamics terms are required) for a very complex chemical system and recycle process. The controller is a bounded PI-like feedback with dynamic estimation of uncertainties. In addition for stabilizing temperature under uncertain kinetics, the disturbance attenuation is carried out by the proposed controller.

The text is organized as follows. Next section contains a brief review on robust temperature control of complex chemical systems. Alkylation system is presented in Section 3. Problem statement is discussed in Section 4. Temperature control is analyzed in Section 5. Controller performance and tuning rules are shown in Section 6. The paper is closed with some concluding remarks.

2. Brief review on reactor temperature regulation

There are several control problems in chemical reactors. One of the most commonly studied is the temperature stabilization in exothermic monomolecular irreversible reaction A → B in a cooled continuous-stirred tank reactor (CSTR). Main theoretical questions in control of chemical reactors address the design of control functions such that, for instance: (i) feedback compensates the non-linear nature of the chemical process to induce linear stable behavior; (ii) stabilization is attained in spite of constrains in input control (as, for example, bounded control or anti-reset windup); (iii) temperature is regulated in spite of uncertain kinetic model (parametric or type of kinetics); or (iv) stabilization is achieved in presence of recycle streams. In addition, reactor stabilization should be achieved for set of physically realizable initial conditions (i.e., global stabilization is required). Some questions have been solved and among solutions the following proposals are remarkable:

(i) *Compensation of non-linear terms via state feedback*. Henson and Seborg [8] discussed the input–output linearization for temperature stabilization via non-linear control theory. Their results were a timely contribution in the sense that geometrical control framework has potential application in chemical process for global stabilization. Such a procedure is based on non-linear coordinate transformation from Lie derivatives, which, when applied to a given chemical process, can provide an input–output closed-loop linear dynamical behavior.

(ii) *Stabilization under constrained control input*. Alvarez et al. [16] and Alvarez-Ramírez and Femat [17] have reported that non-linear bounded control is capable to stabilize exothermic CSTR. Authors departed from a global stabilization under control without saturation, namely $u = \mu(x)$, toward a bounded (constrained) control. The first one (unbounded control) is based on directional derivatives along vector fields (as in [2]). Alvarez [16] proved that under certain conditions (e.g., uniqueness of the open-loop CSTR behavior) the bounded control input yields asymptotic stabilization for a given set of the initial conditions, which is the physically realizable. On the other hand, their results implies that the stabilization of exothermic CSTR cannot necessarily reach the prescribed point. From practical point of view, these results make sense since bounded input implies that required heat removal could not be reach. Hence, under bounded feedback control, a given exothermic CSTR can be stabilized at an undesired critical point.

(iii) *Stabilization against uncertain kinetic model*. (a) An extension for global stabilization of chemical reactors was proposed in [18]. That is, they proposed a feedback control such that stabilization can be achieved “whatever the initial conditions are”, which means that initial conditions are located inside the physical domain of the system CSTR. Viel et al. [18] have shown that if the sign of the reaction heat is constant the stabilization of chemical reactors can be attained in spite of unknown kinetic terms. First, the authors assumed that all states are available for feedback. Such an assumption was partially relaxed to develop an observed-based controller via temperature and concentration measurements. (b) Femat et al. [19] reported that temperature regulation in CSTR’s can be achieved against unknown kinetics only from the temperature measurements and for initial conditions belonging to the physical domain of the reactor even under bounded control actions. Triangle reaction was used to show the controller performance. Promissory results were found. However, results in Femat et al. [19] were developed for a fluidized bed reactor. This kind of reactors involves a small time constant (on the order of seconds) which, according to above discussion, does not necessarily imply cascade feedback control. In addition, one complex problem in process with small time constant is the noisy measurements.

Above results have been timely contributions for temperature regulation and reactor stabilization. However, the mechanism of the uncertainties compensation from measurements is not clear for systems with mass transfer. That is, from the control theory point of view, the design algorithms are proved and clearly established (for example, the geometrical control theory). However, from the process point of view, there is unclear meaning of the resulting controllers. Thus, there remain some questions. For example: (i) Can temperature regulation via robust feedback be interpreted from heat balance? (ii) Which is the physical meaning of the temperature stabilization against unknown kinetics via observed-based control? Questions (i) and (ii) have been solved for classical control [10]; however, robustness via geometrical control holds obscure in this sense. Here, we discuss the temperature regulation against uncertainties from measurements in terms of the heat balance for a class of rector with complex reaction and mass transfer. We show that a robust observed-based controller can be interpreted from the heat balance point of view. To this end, an alkylation reactor allows us to present the robust temperature regulation via measurements and its physical interpretation from heat balance.

3. Alkylation reactor model: kinetics, mass transfer and dynamics

The goal of this section is to show the main features in reactor that affect its dynamics.

(A) *Kinetic model*: First complexity in alkylation consists in the kinetic model. We have chosen the alkylation of isobutane with propylene with sulfuric acid as catalyst. A 17 reaction mechanism model was postulated in [2] to describe such an alkylation. Such a mechanism comprises 20 chemical species and including saturated hydrocarbon species. The kinetic model is based on Schmerling carbonium ion mechanism with modifications for accounting the formation of iC_9 and iC_{10} (see Table A.1). The Langley–Pike’s model [2] was validated with a scale pilot experimental CSTR under isothermal operation and several experiments were carried out over a temperature range from 290 to 330 K. There are differences in opinion concerning complexity of alkylation mechanism, then the Langley–Pike's model is not the only one possible; however, their model is good enough for illustrating the objectives of the paper. The results discussed here concern only for model including Langley–Pike's kinetics for alkylation. Experimental implementation is expected to corroborate simulation results and possibly extend to other alkylation mechanisms. Thus, the model for the kinetics taken in this contribution is given by

$$\begin{aligned}
R_1(x_1, x_2^*) &= x_{1,1} + (k_2, x_{1,13} + k_3 x_{1,15} + k_4 x_{1,16} \\
&\quad + k_5 x_{1,17} + k_6 x_{1,18} + k_7 x_{1,19} + k_8 x_{1,20}) \\
R_2(x_1, x_2^*) &= x_{1,2}(k_1[\mathrm{HX}] + k_{11} x_{1,14} + k_{15} x_{1,17}) \\
R_3(x_1, x_2^*) &= k_2 x_{1,1} x_{1,13} \\
R_4(x_1, x_2^*) &= k_3 x_{1,1} x_{1,15} \\
R_5(x_1, x_2^*) &= k_4 x_{1,1} x_{1,16} \\
R_6(x_1, x_2^*) &= k_5 x_{1,1} x_{1,17} \\
R_7(x_1, x_2^*) &= k_6 x_{1,1} x_{1,18} \\
R_8(x_1, x_2^*) &= k_7 x_{1,1} x_{1,19} \\
R_9(x_1, x_2^*) &= k_8 x_{1,1} x_{1,20} \\
R_{10}(x_1, x_2^*) &= k_9 x_{1,14} - k_{10} x_{1,10} x_{1,14} \\
R_{11}(x_1, x_2^*) &= k_{13} x_{1,12} x_{1,14} + k_{17} x_{1,20} \\
&\quad - k_{14} x_{1,11}[\mathrm{HX}] - k_{16} x_{1,11} x_{1,14} \\
R_{12}(x_1, x_2^*) &= k_{12} x_{1,17} - k_{13} x_{1,12} x_{1,14} \\
R_{13}(x_1, x_2^*) &= k_1 x_{1,2}[\mathrm{HX}] - k_2 x_{1,13} x_{1,1} \\
R_{14}(x_1, x_2^*) &= -x_{1,14}(k_9 + k_{10} x_{1,10} + k_{11} x_{1,2} \\
&\quad + k_{13} x_{1,12} + k_{16} x_{1,11}) \\
R_{15}(x_1, x_2^*) &= k_{14} x_{1,11}[\mathrm{HX}] + k_{17} x_{1,20} - k_3 x_{1,15} x_{1,1} \\
R_{16}(x_1, x_2^*) &= k_{13} x_{1,12} x_{1,14} - k_4 x_{1,16} x_{1,1} \\
R_{17}(x_1, x_2^*) &= k_{11} x_{1,14} x_{1,2} - k_5 x_{1,17} x_{1,1} \\
&\quad - k_{15} x_{1,17} x_{1,2} - k_{12} x_{1,17} \\
R_{18}(x_1, x_2^*) &= k_{10} x_{1,14} x_{1,10} - k_6 x_{1,18} x_{1,1} \\
R_{19}(x_1, x_2^*) &= k_{16} x_{1,11} x_{1,14} - k_7 x_{1,19} x_{1,1} \\
R_{20}(x_1, x_2^*) &= k_{15} x_{1,17} x_{1,2} - k_{17} x_{1,20} - k_8 x_{1,20} x_{1,1}
\end{aligned} \tag{1}$$

where $x_1 \in \mathbb{R}^{20}$ denotes the concentration vector of the chemical species (see below for notation), [HX] the sulfuric acid concentration, $k_i = k_i(x_2^*)$, $i = 1, 2, \ldots, 20$, the constant rate for any fixed value of the temperature x_2^* in interval [290, 330] and $R_i(x_1, x_2^*)$ denotes the ith reaction rate corresponding to each chemical specie. In model (1), the parameters ks are temperature functions; in this sense the kinetics can be denoted as $R(x_{1,i}; k_j(x_2^*))$, indexes $i = 1, 2, \ldots, 20$ and $j = 1, 2, \ldots, 17$ stand for chemical specie and reaction in mechanisms, respectively.

(B) *Mass transfer*: One more difficulty arises from the fact that there are two phases in the reactor: (i) hydrocarbon and (ii) acid. The reaction occurs in the acid phase while reactants are fed in hydrocarbon phase. This implies that, in order to reaction occurs, there is mass transfer from hydrocarbon to acid phase. The mass transfer is a very complex phenomenon which can involve the reaction–diffusion equation. However, such a phenomenon is beyond of the goal of this paper. Both isobutane and propylene are fed in hydrocarbon phase. Solubility of propylene in acid phase is very fast whereas mass transfer of isobutane is dictated by the smaller mass transfer rate. That is the reactor is mass-transfer-limited by isobutane. Based on the following assumptions, a simplistic model can be found for mass transfer [20]: (MT.1) The propylene concentration at the hydrocarbon phase is constant. (MT.2) The steady state of the mass transfer of $i\mathrm{C}_4$ is equal to its consumption rate in the acid phase. (MT.3) The consumption rate of the isobutane in acid phase involves the intermediate and carbonium ion rate equations. In this manner, under the above assumptions, the mass transfer results in the isobutane concentration, $C_{i\mathrm{C},a}$, which can be expressed by

$$C_{i\mathrm{C}_4,a} = \frac{K_\mathrm{d} x_{1,1}^\mathrm{h}}{1 + (z H_\mathrm{a} / k_\mathrm{L} a_\mathrm{V})} \tag{2}$$

where $C_{i\mathrm{C}_4,a}$ is the isobutane concentration at acid phase, $x_{1,1}^\mathrm{h}$ the concentration of the isobutane at hydrocarbon phase, $z = k_2 x_{1,13} + k_3 x_{1,15} + k_4 x_{1,16} + k_5 x_{1,17} + k_6 x_{1,18} + k_7 x_{1,19} + k_8 x_{1,20}$ involves the rate of the isobutane consumption and subscript ‘a’ means acid phase, and the constant K_d (distribution coefficient for $i\mathrm{C}_4$), $k_\mathrm{L} a_\mathrm{V}$ (term of mass transfer) and H_a (the acid/hydrocarbon fraction) are positive.

In this manner the dynamical model can be obtained from mass and heat balance for the open-loop system [10]. STRATCO reactor (see Fig. 1a) was chosen for modeling under the following assumptions. (D.1) Reactor is perfect mixed and volume in the reactor is constant; (D.2) reaction is carried out in the acid phase and kinetics is given by Eq. (1); (D.3) mass transfer is given by: (i) expression (2) for isobutane while propylene remains constant. Parameters of Eq. (2) are constant. It should be noted that the kinetic model (1) was obtained for isothermic process. Then, according to the results by Langley and Pike [2], the temperature dependence of the isobutane/propylene alkylation is given by $k_i(x_2) = k_{0,i} \exp(-\alpha_i / x_2)$, where x_2 denotes the temperature, $k_{0,i}$ the pre-exponential factor and $\alpha = E_\mathrm{a}/R_\mathrm{g}$ (E_a and R_g are, respectively, the activation energy and the constant of the idea gases, $R_\mathrm{g} = 8.314\,\mathrm{J\,mol^{-1}\,K^{-1}}$). The values of these parameters are shown in Table A.1 Note that neither the products nor olefinic intermediate nor carbonium ions are feed into the reactor. In this way, the dynamical model of the alkylation reactor is given by

$$\left.\begin{aligned}
\dot{x}_{1,1} &= \theta(x_{1,1}^\mathrm{IN} - x_{1,1}) - R_1(x_1, x_2) \\
\dot{x}_{1,2} &= \theta(x_{1,2}^\mathrm{IN} - x_{1,2}) - R_2(x_1, x_2) \\
\dot{x}_{1,i} &= -\theta x_{1,i} + R_i(x_1, x_2); \quad i = 3, 4, \ldots, 20
\end{aligned}\right\} \tag{3.a}$$

$$\dot{x}_2 = \theta(x_2^\mathrm{IN} - x_2) + \beta\left[\sum_{i=1}^{17} R_i(x_1, x_2)\right] - \gamma(x_2 - u) \tag{3.b}$$

where $x := (x_1, x_2) \in \mathbb{R}^{21}$, $x_1 \in \mathbb{R}^{20}$ is the concentration of the chemical species, $x_2 \in \mathbb{R}$ the temperature in the reactor and

$(x_{1,1}^{\mathrm{IN}}, x_{1,2}^{\mathrm{IN}}) \in \mathbb{R}^2$ is the concentration feeding of isobutane and propylene, respectively. $R_i'(x_1, x_2)$, $i = 1, 2, \ldots, 17$ is the ith rate laws in Table A.1 with Arrenhuis-like temperature dependence. γ stands for the heat transfer coefficient of coolant system and β is the reaction enthalpy, whose value has been experimentally obtained [21]. θ is the inverse of the residence time. One should note that the concentration fed $x_{1,1}^{\mathrm{IN}}$ and $x_{1,2}^{\mathrm{IN}}$ are restricted by the mass transfer. That is, $x_{1,1}^{\mathrm{IN}}$ corresponds to the isobutane concentration at acid phase $C_{i\mathrm{C}_4,a}$, which is computed from Eq. (2) whereas $x_{1,2}^{\mathrm{IN}}$ is constant due to mass transfer of propylene is fast.

4. Temperature regulation problem

In order to state the control problem, the following property of the alkylation reactor is presented:

(P.1) Consider an exothermic continuous-stirred tank reactor with temperature dependence Arrenhuis-type, there is a stable equilibrium point such that, under the isothermic operation (i.e., as reactor temperature x_2^* is constant). The above property was theoretically studied for CSTR's by [22] and guarantees the existence of, at least, one stable equilibrium point (see Appendix A). Of course, multiple equilibrium points can be found in chemical reactors. Such an assumption also implies that reactor stabilization can be achieved via temperature regulation (see [8,15,17,18]). Thus, according to property (P.1), it is enough to design a feedback controller from subsystem (3.b) in order to attain the reactor temperature stabilization at any equilibrium point. This makes sense in terms of the heat balance. That is, in order to control the reactor temperature, the most important term to be compensated in Eq. (3.b) is the heat generation due to reaction, namely $\beta \sum_i R_i'(x_1)$, which, according to discussion in previous sections, can be often uncertain. Also the inlet heat term, θx_2^{IN}, contributes to the reactor temperature; however, small contribution is expected for highly exothermic reactions. In addition, inlet heat term is not hard to compute or estimate because it does not comprise neither kinetic nor mass transfer terms.

From heat balance, we can compute the heat removal by jacket along time such that reactor operates in constant temperature x_2^* (isothermic operation). Thus, from heat balance, the heat removal to induce steady state in reactor temperature is as follows:

$$\begin{gathered}(\text{Heat removal by jacket}) = (\text{heat generation by reaction}) \\ - (\text{heat entering by inlet flow}) + (\text{heat leaving by outlet flow}) \\ \text{or} \qquad (4) \\ -\gamma(x_2 - u) = -\beta \sum_{i=1}^{m} R(x_1 - x_2) - \theta x_2^{\mathrm{IN}} + \theta x_2\end{gathered}$$

where γ, θ and β are defined in Eqs. (3.a) and (3.b). Here, the reaction rate $R(x_1, x_2)$ corresponds to kinetics in Table A.1 including temperature dependence (for example, Arrenhuis-type).

As property (P.1) holds, the heat removal (4) implies that the jacket temperature $u = (\gamma x_2 - \beta \sum_i R_i'(x_1, x_2) - \theta x_2^{\mathrm{IN}} + \theta x_2)/\gamma$ induces the steady state in subsystem (3.b) from measurements of reactor temperature and assuming that model of reaction rate has no errors. This implies that the heat accumulation and generation are required in order to compute the heat removal. Thus, regarding (4), there are two drawbacks: (i) heat generation involves the reaction laws which, according to discussion in previous sections, are uncertain and (ii) although the jacket temperature computed from heat balance (4) induces temperature steady state, the reactor dynamics is not regulated in a desired value (that is the isothermic operation does not assure uniqueness of the stable equilibrium points in chemical systems, e.g., autocatalysis, see Chapter 5 in [23]). On contrary, controller from heat balance (4) has the following advantages: (i) a linear closed-loop behavior can be induced and the convergence rate can be directly related to the control gain, (ii) if a gradient term $K_{\mathrm{c}}(x_2 - x_2^{\mathrm{r}})$ is added to the feedback, then the coordinate of the (unique) equilibrium point x_2^* are located at the constant x_2^{r} (which is stable for suitable control parameter value). In fact, the jacket temperature $u = (\gamma x_2 - \beta \sum_i R_i'(x_1, x_2) - \theta x_2^{\mathrm{IN}} + \theta x_2 + K(x_2 - x_2^{\mathrm{r}}))/\gamma$ can also be derived from Lie derivative of the measured temperature along vector field of (3.a) and (3.b); i.e., as $L_{\mathrm{g}} L_{\mathrm{f}2}^{0} = \gamma$ and $L_{\mathrm{f}} x_2 = \gamma x_2 - \beta \sum_i R_i'(x_1, x_2) - \theta x_2^{\mathrm{IN}} + \theta x_2$ and the gradient terms corresponds to the desired dynamics [8,24]. Thus, one has that the combined jacket temperature to stabilize the reactor becomes: $u = (\gamma x_2 - \beta \sum_i k_i R_i'(x_1) - \theta x_2^{\mathrm{IN}} + \theta x_2 + K_{\mathrm{c}}(x_2 - x_2^{\mathrm{r}}))/\gamma$. Such control law can be interpreted in terms of the classical control theory (indeed the term $K_{\mathrm{c}}(x_2 - x_2^{\mathrm{r}})$ is a proportional control action [25,10]) and exactly corresponds to the non-linear state feedback designed by geometric control theory (compare above equation with results by Henson and Seborg [8]). Nevertheless, the term $K_{\mathrm{c}}(x_2 - x_2^{\mathrm{r}})$ is known as high-gain control and undesirable behavior can be induced; as, for example, scattering.

All above advantages and drawbacks should be considered to design the control function for the alkylation reactor. In this way, the temperature control problem in alkylation can be worded *for finding the jacket temperature such that the heat balance (4) is held to reach the desired temperature value despite kinetic uncertainties*. This implies that reaction laws should be estimated in order to obtain a feedback control against errors in kinetic model. In what follows a feedback controller is proposed for accounting the heat generation with least prior information about the kinetic and chemical mechanism but by exploiting structure from heat balance. The main idea behind our proposal is to obtain an estimated value of temperature dynamics due to an estimated value of the heat generation by reaction.

5. Estimation of the heat generation

Let us define the time variable $\eta(t) = \beta \sum_i R_i'(x_1(t), x_2(t))$ toward obtainment of estimated value of heat generation term. Such a variable describes the generation heat due to reaction along the solution of the dynamical model ((3.a) and (3.b)). That is, at time $t \in \mathbb{R}$, the heat generation is evolving according with rule $\beta \sum_i R_i'(x_1(t), x_2(t))$ along time and such an time-evolution can be seem as the variable $\eta(t)$. In other words, the heat reaction is a continuous-time assignation $\eta : \mathbb{R}^{21} \to \mathbb{R}$ whose time derivative $\dot{\eta}$ is defined for all $x \in \mathbb{R}^{21}$. In this sense, subsystem (3.b) can be rewritten as follows:

$$\dot{x}_2 = \theta(x_2^{\mathrm{IN}} - x_2) + \eta + \gamma(x_2 - u) \qquad (5.a)$$

$$\dot{\eta} = \beta \sum_{j=1}^{21} \frac{\partial \left(\sum_{i=1}^{21} R_i'(x_1(t)) \right)}{\partial x_j} f_j(x) \qquad (5.b)$$

where $f_j(x)$, for $i = 1, 2, \ldots, 21$, stands the right side terms in dynamical model (3.a) and (3.b). Note that time-derivative of the augmented state η can be derived by Lie derivative as follows $\dot{\eta} = L_f R_i'(x_1(t), x_2(t))$. Thus, the heat balance (4) can be interpreted in terms of the extended temperature system ((5.a) and (5.b)). One should note that, if the reaction law terms are uncertain, then the time derivative of the augmented state η is also uncertain. Thus, although $\dot{\eta}$ describes the dynamics of the heat generation term, the augmented state cannot be available for feedback. That is, the augmented state η cannot be measured from system ((5.a) and (5.b)). As matter of fact, Eqs. (5.a) and (5.b) stand an intermediate dynamical system toward final controller such that final controller remains the main features of the heat balance (4), i.e., $u = (\gamma x_2 - \eta - \theta x_2^{\mathrm{IN}} + \theta x_2 + K_c(x_2 - x_2^{\mathrm{r}}))/\gamma$. However, system ((5.a) and (5.b)) has the property that it provides information about the directional derivative of the heat generation terms along the reactor trajectory. That is, the augmented state indicates when an uncertain term increases or decreases at any time $t \in [t, t + \Delta t]$. Now, since η is not available for feedback an estimation procedure is needed. Thus, following the ideas reported by Alvarez-Ramírez and Femat [17], a state observer can be designed for system ((5.a) and (5.b)) from the output $y = x_2$ to obtain

$$\dot{\hat{x}}_2 = \theta(x_2^{\mathrm{IN}} - y) + \hat{\eta} - \gamma(y - u) + g_1(y - \hat{x}_2) \qquad (6.a)$$

$$\dot{\hat{\eta}} = g_2(y - \hat{x}_2) \qquad (6.b)$$

where $(\hat{x}_2, \hat{\eta})$ are estimated values of the reactor temperature x_2 and the augmented state η, which, by definition is equal to heat generation term, $\beta \sum_i R_i'(x_1(t))$. The parameters g_1 and g_2 are such that the polynomial $P_2(\lambda) = \lambda^2 + g_1\lambda + g_2$ is Hurwitz. In this manner, the jacket temperature can be computed from the output y and the estimated value of generation heat term as follows:

$$u = \frac{\gamma y - \hat{\eta} - \theta x_2^{\mathrm{IN}} + \theta y + K_c(y - x_2^{\mathrm{r}})}{\gamma} \qquad (7)$$

where parameters are defined as above.

The aim of system ((6.a) and (6.b)) is to estimate the motion of the heat generation, which is the uncertain term. Fortunately, estimator ((6.a) and (6.b)) and feedback function (7) leads to feedback control with PI-like structure. To prove this, we can take the Laplace transformation to obtain the transfer function of the system ((6.a), (6.b) and (7)). Thus, we have that the transfer function of the (heat compensation) control law becomes: $C(s) = u(s)/y(s) = K_c[(1 + (1/\tau_{\mathrm{IN}}s)) + (K_{\mathrm{E}}/(s^2 + k_1 s))]$. Moreover, note that as $\hat{\eta}(t) \to \eta(t)$ as the controller (7) behaves like the heat compensation (4). That is, $\hat{\eta} \to \eta$ for all $t > 0$ then controller (7) is the linearizing feedback control. In addition, note that estimator ((6.a) and (6.b)) is a linear dynamical system with constant parameters. Hence, the transfer function of the estimator can be computed to get $\hat{\eta}(s) = G_{\mathrm{E}}(s)y(s) + G_{\mathrm{d}}(s)x_2^{\mathrm{IN}}(s)$ where

$$G_{\mathrm{E}}(s) = \frac{K_{\mathrm{E}}}{s(\tau_{\mathrm{E}}s + 1)}; \qquad G_{\mathrm{d}}(s) = \frac{K_{\mathrm{IN}}}{\tau_{\mathrm{IN}}s + 1} \qquad (8)$$

and the parameters becomes $K_{\mathrm{E}} = g_2(\theta + \gamma - g_1 + K_c)/(\gamma + \theta - g_1)$, $\tau_{\mathrm{E}} = 1/(\gamma + \theta - g_1)$, $K_{\mathrm{IN}} = g_2 K_c$ and $\tau_{\mathrm{IN}} = 1/(\gamma + \theta - g_1)$. Transfer function $G_{\mathrm{E}}(s)$ depends on the control and estimator gain. That is, the estimator and controller should be designed together (i.e., there is no separation principle).

Fig. 2 shows the block diagram of the closed-loop process under above feedback. In regard the transfer functions $G_{\mathrm{d}}(s)$ and $G_{\mathrm{E}}(s)$, one should note that: (a) $G_{\mathrm{d}}(s)$ has low-pass structure. Indeed, the cut frequency is $\omega_c = 1/\tau_{\mathrm{IN}}$; and increases as the estimation constant g_1. Then, high frequency disturbances from inlet temperature $x_2^{\mathrm{IN}}(t)$ can be filtered by means of parameters tuning of the estimator ((6.a) and (6.b)). Nevertheless, as the control constant K_c increases the gain K_{IN} increases. These facts can involve an estimation/regulation tradeoff. However, under appropriate tuning, for suitable values of the control parameters the noisy measurements cannot affect the uncertainties estimation. (b) Fig. 3 shows the Bode diagram for several values of the parameters K_c, g_1 and g_2. It is easy to see that there is a frequency ω^*, which depends on the control and estimation parameters, such that for any $\underline{\omega} < \omega^*$ the estimated value $\hat{\eta}(s)$ in more sensitive to output $y(s)$ than disturbance $x_2^{\mathrm{IN}}(s)$ but there is any $\bar{\omega} < \omega^*$ $\hat{\eta}(s)$ is more sensitive to disturbances $x_2^{\mathrm{IN}}(s)$ than output $y(s)$. That is, $|G_{\mathrm{E}}(\omega \mathrm{j})| > |G_{\mathrm{d}}(\omega \mathrm{j})|$ for any $\omega < \omega^*$. Therefore, since an accurate estimated value $\hat{\eta}(t)$ is needed in order to proposed controller behaves like linearizing feedback, a tuning procedure is required in order to estimated value of uncertain term $\hat{\eta}(s)$ be not sensitive to external disturbance $x_2^{\mathrm{IN}}(s)$.

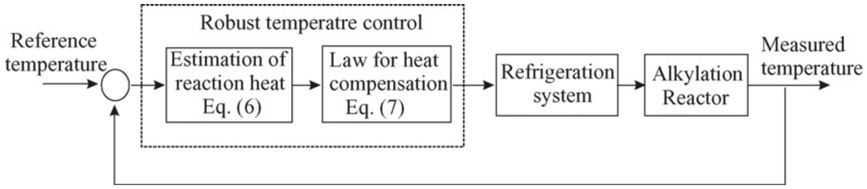

Fig. 2. Block diagram for the complete closed-loop behavior. Here, the block corresponding to the refrigeration system is neglected.

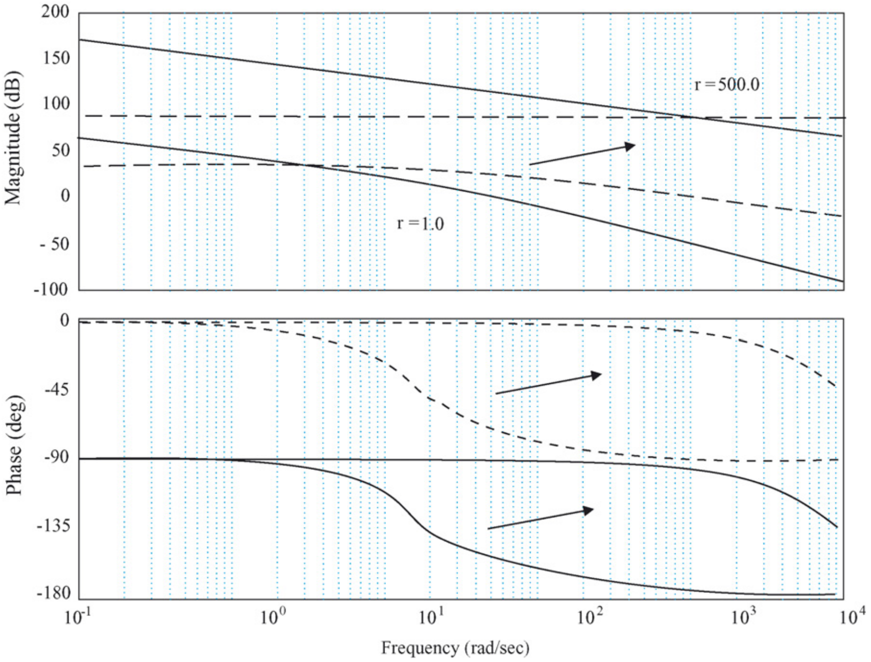

Fig. 3. Bode diagram of the closed-loop under the estimation/compensation control approach. Closed-loop system is expected to be non-sensitive to high frequency signals.

The objective of the tuning procedure is to find the value of the parameters K_c, g_1 and g_2 such that the magnitude of $G_d(s)$ be smaller than magnitude of $G_E(s)$ for any ω less than the given frequency ω^*. Classical tuning methods include criteria based on trial and error and procedures which involve to determine process models and use heuristic basis [26,27]. Here, since process model is available, we use heuristic basis to derive a tuning procedure via frequency response. Let $r > 1$ be an integer number such that $g_1 = -2rK_c$ and $g_2 = (rK_c)^2$. The number r denotes the estimation/regulation ratio; that is, for a given control gain K_c (which stands the regulation rate), as r increases as the roots of the polynomial $P_2(\lambda) = \lambda^2 + g_1\lambda + g_2$ are shifted to left within open left-hand complex plane. Fig. 3 shows Bode plots for several estimation/regulation ratio r and control gains K_c. Then, since $|G_E(\omega j)| > |G_d(\omega j)|$ for any $\omega < \omega^*$.

6. Numerical simulations

Here, the performance of the heat compensation controller is tested. To this end, the model parameters were taken as follows [2]: $x^{\mathrm{IN}}_{1,1} = 862.5\,\mathrm{mol\,m^{-3}}$, $x^{\mathrm{IN}}_{1,2} = 145.4\,\mathrm{mol\,m^{-3}}$, $[\mathrm{HX}] = 1776\,\mathrm{mol\,m^{-3}}$, $x^{\mathrm{IN}}_{2} = 283\,\mathrm{K}$, $\theta = 8.33\mathrm{e}{-4}$, $\beta = 0.3266\,\mathrm{K\,m^3\,mol^{-1}}$. The controller ((6.a), (6.b) and (7)) was interconnected with non-linear model ((3.a) and (3.b)) to numerically simulate its performance.

The idea behind the proposed feedback is the estimation of the generated heat by reaction, then simulations are aimed to show how the reactor temperature stabilization is affected by the initial value of the estimated heat, $\hat{\eta}(0)$. Fig. 4 shows the reactor temperature and the computed coolant temperature for several initial values $\hat{\eta}(0)$. Here the estimation parameter was arbitrarily chosen $L = 0.5$. Note that as the estimated value decreases the convergence to the reference temperature, 283 K, is reached. Fig. 5 shows the same effect for the value of the estimation parameter $L = 5$. By comparing both Figs. 4 and 5,

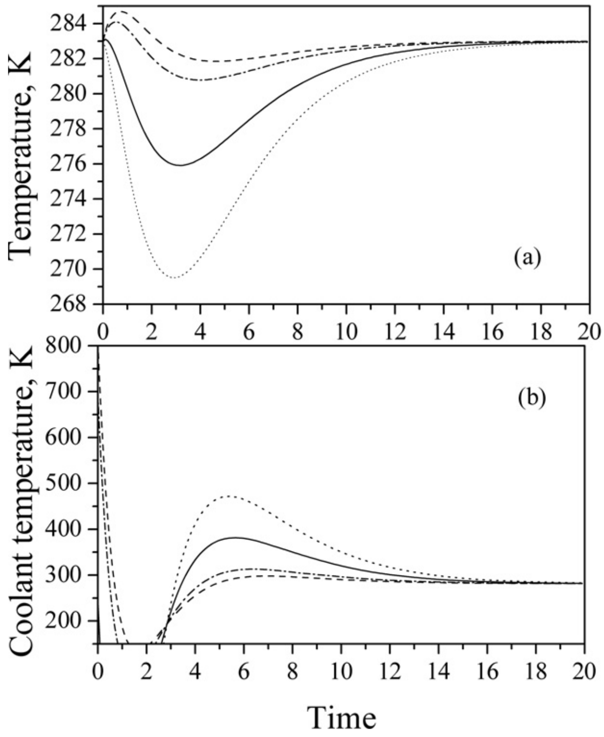

Fig. 4. Effects of the initial value of the reaction heat for $L = 0.5$. Dotted line, $\hat{\eta}(0) = 10.0$; solid line, $\hat{\eta}(0) = 5.0$; dash–dotted line, $\hat{\eta}(0) = 1.0$; dashed line, $\hat{\eta}(0) = 0.0$. Small initial values of the heat reaction lead to better performance than large values.

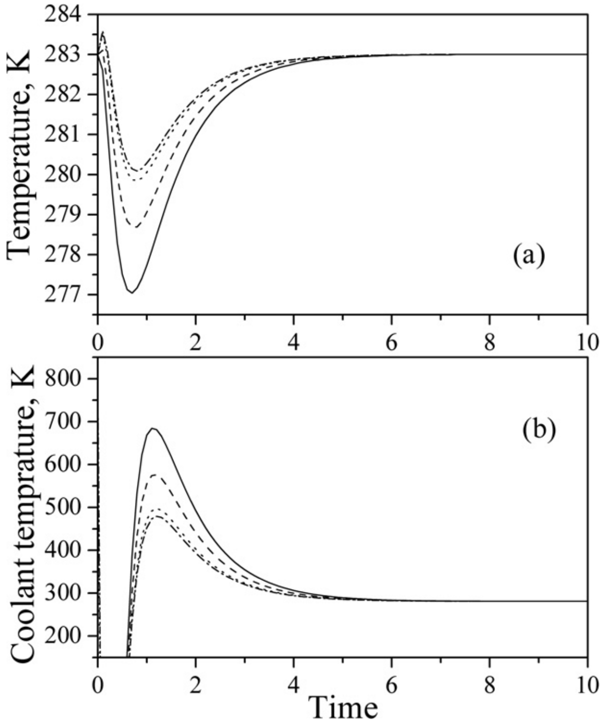

Fig. 5. Effects of the initial value of the reaction heat for $L = 5.0$. Note that as L increases as faster reactor temperature convergence (compare with Fig. 4). Solid line, $\hat{\eta}(0) = 10.0$; dashed line, $\hat{\eta}(0) = 5.0$; dotted line, $\hat{\eta}(0) = 1.0$; dash–dotted line, $\hat{\eta}(0) = 0.0$. In regard initial value of estimated value $\hat{\eta}(0)$, the effect is similar than for $L = 0.5$.

we can observe that as the value of L in creases as the faster convergence; however, the control cost for faster convergence is the saturation of the coolant temperature. Thus, a trade off between fast convergence and control performance is found.

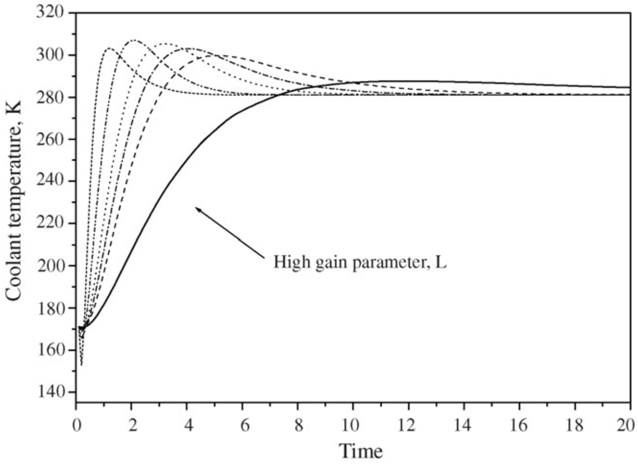

Fig. 6. Effect of increasing the high gain parameter L on the performance of the control action. The response of the coolant temperature is faster as L increases. However, a very large value of L can induce oversooh and saturation. On contrary, a very low value on L can induce off set at steady state. The arrow in picture indicates the direction of the effect; that is, L was increased as the arrow direction shows.

The effect of increasing the estimation parameter L is shown in Fig. 6. After extensive simulations, we found that the performance satisfies the Integral Square Errors and Control (ISEC) performance index (see Chapter 7 in [24]) corresponds to $L = 1.0$.

Note that, for large value of estimation parameter L, the control input (coolant temperature) can be saturated due to finite-time escape of the control signal (peaking phenomenon). Saturation is a non-linear function that induces discrepancy between the computed and actual control input. Such a discrepancy is often related to actuator constraints [16,28]. Thus, the feedback is broken and the process behaves as an open-loop plant with the possible performance degradation. Indeed, the degradation can include windup phenomenon in presence of integration actions. Since the estimation of the heat generation is computed from (6.a) and (6.b) (i.e., it includes integration parts), an antireset-windup strategy can be needed. We do not find reset windup in the extensive simulations. This can be associated to the slow dynamics of the process and low value of the estimation gain; however, the possibility of appearing the reset windup phenomenon should be considered as well.

7. Concluding remarks

Here, a control law for chemical reactors had been proposed. The controller was designed from compensation/estimation of the heat reaction in exothermic reactor. In particular, the paper is focused on the isoparaffin/olefin alkylation in STRATCO reactors. It should be noted that control design from heat compensation leads to controllers with same structure than non-linear feedback. This fact can allow to exploit formal mathematical tools from non-linear control theory. Moreover, the estimation scheme yields in a linear controller. Thus, the interpretation for heat compensation/estimation is simple in the context of process control.

On the other hand, the proposed approach has structure of low pass filter; see Eq. (8). Thus, we can expect that the closed-loop response is not sensitive to high frequency signals (as, for example, by noisy measurements or fluctuations in fluids mechanics by agitation). Although Fig. 3 depicts the frequency response, the robustness of the closed-loop should be guaranteed by weighting functions. To this end, H_∞ theory can be exploited. One more issue about robustness is related to delay by lag transport. In this direction, the question is how the delay by lag transport affects the closed-loop performance? These issues are beyond the goals of the present contribution, and are being analyzed to be reported elsewhere.

Acknowledgments

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Appendix A

Values of kinetic parameters are given in Table A.1.

Table A.1
Reaction mechanism and rate constants at 330 K with 95% of H_2SO_4 as catalyst

Reactions	Rate law, $R'(x_1)$	Preexponential factor[a]	Activation energy[b]
Initiation			
$C_3{}^{2-} + HX \xrightarrow{k_1} C_3{}^{+}X^{-}$	k_1AB	1.01×10^{7}	2.35
$C_3{}^{+}X^{-} + iC_4 \xrightarrow{k_2} C_3 + iC_4{}^{+}X^{-}$	k_2AB	2.07×10^{11}	0.0
Primary			
$iC_4{}^{+}X^{-} + C_3{}^{2-} \xrightarrow{k_{11}} iC_7{}^{+}X^{-}$	$k_{11}AB$	1.99×10^{17}	2.36
$iC_7{}^{+}X^{-} + iC_4 \xrightarrow{k_5} iC_7 + iC_4{}^{+}X^{-}$	k_5AB	4.20×10^{10}	0.0
Self-alkylation			
$iC_4{}^{+}X^{-} \xrightarrow{k_9} iC_4{}^{2-} + HX$	k_9A	3.92×10^{4}	0.40
$iC_4{}^{+}X^{-} + iC_4{}^{2-} \xrightarrow{k_{10}} iC_8{}^{+}X^{-}$	$k_{10}AB$	5.63×10^{19}	4.10
$iC_8{}^{+}X^{-} + iC_4 \xrightarrow{k_6} iC_8 + iC_4{}^{+}X^{-}$	k_6AB	5.35×10^{10}	0.0
Destructive alkylation			
$iC_7{}^{+}X^{-} \xrightarrow{k_{12}} iC_7{}^{2-} + HX$	$k_{12}A$	7.49×10^{5}	1.08
$iC_7{}^{2-} + iC_4{}^{+}X^{-} \xrightarrow{k_{13}} iC_5{}^{2-} + iC_8{}^{+}X^{-}$	$k_{13}AB$	3.64×10^{21}	5.73
$iC_5{}^{2-} + HX \xrightarrow{k_{14}} iC_5{}^{+}X^{-}$	$k_{14}AB$	1.62×10^{11}	3.69
$iC_5{}^{+}X^{-} + iC_4 \xrightarrow{k_3} iC_5 + iC_4{}^{+}X^{-}$	k_3AB	3.29×10^{10}	0.0
$iC_6{}^{+}X^{-} + iC_4 \xrightarrow{k_4} iC_6 + iC_4{}^{+}X^{-}$	k_4AB	4.04×10^{10}	0.0
$iC_7{}^{+}X^{-} + C_3{}^{2-} \xrightarrow{k_{15}} iC_{10}{}^{+}X^{-}$	$k_{15}AB$	3.72×10^{17}	2.59
$iC_{10}{}^{+}X^{-} + iC_4 \xrightarrow{k_8} iC_{10} + iC_4{}^{+}X^{-}$	k_8AB	6.68×10^{10}	0.0
$iC_5{}^{2-} + iC_4{}^{+}X^{-} \xrightarrow{k_1} iC_9{}^{+}X^{-}$	$k_{16}AB$	4.26×10^{19}	2.65
$iC_9{}^{+}X^{-} + iC_4 \xrightarrow{k_7} iC_9 + iC_4{}^{+}X^{-}$	k_7AB	6.02×10^{10}	0.0
$iC_{10}{}^{+}X^{-} \xrightarrow{k_{17}} iC_5{}^{2-} + iC_5{}^{+}X^{-}$	$k_{17}A$	4.45×10^{11}	8.40

[a] $cm^3\,g^{-1}\,mol^{-1}\,s^{-1}$.
[b] $kcal\,g^{-1}\,mol^{-1}$.

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