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Effect of coolant flow rate on the dynamics of delayed recycle continuous stirred tank reactor

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ABSTRACT

Numerical bifurcation analysis of delayed recycle stream in a continuous stirred tank reactor was studied using DDE-BIFTOOL. A first-order irreversible exothermic reaction $s_1 \xrightarrow{k_{2.1}} s_2$ was considered for analyzing effect of delay on the stability of the delayed recycle system. The non-isothermal CSTR was operated at both infinite and finite coolant inlet flowrates. A constant delay was considered in the recycle stream of CSTR for concentration of reactant, and temperature. DDE-BIFTOOL solver was used for finding dependency of delays on the bifurcation parameters, and its stability characteristics. The bifurcation parameters considered were: (i) fresh feed flowrate, (ii) coolant temperature, and (iii) coolant inlet flowrate. In the absence of delay, the system exhibits the region of dynamic instability for both infinite and finite coolant inlet flowrates. For infinite coolant flowrate, the region of dynamic instability on the reactor temperature was modified as a result of delay by varying either fresh feed flowrate or coolant temperature. The steady-state multiplicity was observed on the coolant temperature by varying fresh feed flowrate at finite coolant inlet flowrate. In the absence of delay, the fold and Hopf bifurcations were observed on the multiple steady-states of coolant temperature. The concentration and temperature delays do not alter substantially the dynamic characteristics of coolant temperature at $T_{c,in} = 298$ K. However, delays can alter the region of dynamic instability for coolant temperature when coolant inlet temperature is higher than 298 K. These are the main result of present work.

1. Introduction

For the past six decades, the steady state and dynamic characteristics of continuous stirred tank reactor [1–26] (CSTR) as well as reactor-separator-recycle system [27–43] were examined, and innumerable research articles were published on the bifurcation analysis of first-order irreversible exothermic reaction. In 1953, Van Heerden [1] observed the steady-state multiplicity for the first-order irreversible exothermic reaction in a CSTR. Luss and Lapidus [3] presented a method for finding stability characteristics of the state variables in a CSTR. A detailed study on the nonlinear behavior of first-order irreversible exothermic reaction in a recycle CSTR was carried-out by Uppal et al. [5,6]. They presented the steady-state multiplicity in a parameter space by neglecting delay in the recycle stream. Kubieck et al. [9] studied the existence of isola, and mushroom regions for two inter-connected non-isothermal CSTRs. The chaotic behavior was observed by Mankin and Hudson [10] for two CSTRs connected in series with negligible delays in the recycle stream. Subsequently, many researchers investigated the bifurcation analysis of CSTR by considering various reaction schemes [11–23]. Singularity and bifurcation theory [14] was applied for finding the performance of first-order exothermic reaction in a CSTR. The chaotic behavior was observed for consecutive first-order irreversible reactions [16,17] in a CSTR through bifurcation analysis.

The nonlinear bifurcation analysis of CSTR-separator-recycle system was investigated by various researchers [27–43]. For example, CSTR-distillation column [30,31], CSTR-flash-recycle system [34], and so on. Luyben [28] presented the snowball effect in a reactor-separator-recycle system and proposed a control scheme for avoiding snowball effect. Morud and Skogestad [29] observed the instability in a reactor as a result of recycle of mass and energy with large time constant. Singularity and bifurcation theory was applied for determining nonlinear behavior of CSTR-flash recycle system [34]. Bildea et al. [41] observed the large reactor volume prevents the accumulation and infinite recycle of reactant in a reactor-separator-recycle system. Steady-state multiplicity in polymerization reactions was presented by Kiss et al. [42,43].

Few researchers [44–50] investigated the stability analysis of delayed recycle CSTR. Lehman and coworkers [46–50] studied effect of delay on the stability of first-order exothermic reaction in a

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Nomenclature

$A_{2,1}$	pre-exponential factor, s^{-1}
c_p	specific heat capacity of reaction mixture, $J\,kg^{-1}\,K^{-1}$
$c_{p,c}$	specific heat capacity of coolant, $J\,kg^{-1}\,K^{-1}$
$c_{s_1,f}$	initial concentration of reactant s_1 in the feed, $mol\,m^{-3}$
c_{s_1}	concentration of reactant s_1, $mol\,m^{-3}$
c_{s_2}	concentration of product s_2, $mol\,m^{-3}$
$E_{2,1}$	activation energy of reaction, $J\,mol^{-1}$
I	identity matrix
J	Jacobian matrix with respect to instantaneous state variables
J_τ	Jacobian matrix with respect to delayed state variables
m	number of delays considered
n	number of state variables
Q	volumetric flow rate, $m^3 s^{-1}$
Q_j	volumetric flow rate of coolant, $m^3\,s^{-1}$
r	recycle ratio ($0 < r < 1$)
R	ideal gas constant, 8.314 $J\,mol^{-1}\,K^{-1}$
s_i	symbol for reacting species i
t	time, s
T	temperature (K)
T_c	coolant temperature (K)
$T_{c,\mathrm{in}}$	coolant inlet temperature (K)
T_f	temperature (K)
UA	overall heat transfer coefficient, $Js^{-1}\,K^{-1}$
V	volume of reactor, m^3
V_j	volume of cooling jacket, m^3
z_i	state variable of index i
Greek symbols	
ρ	density of mixture, $kg\,m^{-3}$
ρ_c	density of coolant, $kg\,m^{-3}$
ψ	parameters
τ	delay, s
$(-\Delta H)$	heat of reaction, $J\,mol^{-1}$
λ	eigenvalue

CSTR with an assumption of infinite coolant flowrates. They found that a delay in the recycle stream does not alters the dynamic characteristics of the system. A detailed study on the dynamic behavior of first-order irreversible reaction in the delayed CSTR-flash-recycle system was carried-out by Balasubramanian et al. [51–53]. A delay independent stability was determined analytically by controlling the fresh feed flowrate entering into an isothermal CSTR. Furthermore, a delay dependent stability was observed by controlling the effluent flowrate. For non-isothermal CSTR, the switching of stability characteristics from unstable to stable and vice-versa was recognized. Dynamic behavior of CSTR-mechanical separator-recycle system sustaining first-order irreversible exothermic reaction [54] was studied using DDE-BIFTOOL [55,56] with an assumption of infinite coolant flowrates. DDE-BIFTOOL [55,56] is a MATLAB based solver for numerical bifurcation analysis of delay differential equations. New isola regions were found at large delays. Furthermore, the switching of stability characteristics from unstable to stable state, and vice versa was observed for infinite coolant flowrate. Here, dimensionless parameters were considered in the bifurcation analysis.

Uppal et al. [5,6] presented the bifurcation analysis of delayed recycle CSTR with negligible delays in the recycle stream for first-order irreversible exothermic reaction. Infinite coolant inlet flowrate was assumed for bifurcation analysis, and observed that the system admits fold as well as Hopf bifurcations on the steady-state multiplicity. Likewise, Balasubramanian et al. [51–53] presented the effect of delay on the stability of first-order irreversible exothermic reaction in CSTR-flash-recycle system with an assumption of an infinite coolant inlet flowrate. A delay dependent stability was analytically proved by controlling the effluent flowrate using a flow controller. Previously, researchers studied the bifurcation analysis of first-order irreversible exothermic reaction in a CSTR with an assumption of an infinite coolant flowrate. That is, the exit and inlet coolant temperatures are alike for finding the heat of cooling in a reactor. In the earlier studies, the dynamics of coolant temperature was neglected. The coolant flow rate entering into a reactor must be finite in the realistic situation and reactor temperature of an exothermic reaction must be controlled at a desired value by manipulating the coolant inlet flowrate. Thus, the dynamics of coolant temperature in a reactor should be included in the bifurcation analysis. Various researchers used the dimensionless parameters for bifurcation analysis of delayed recycle systems and the realistic delay values were not included. Therefore, we decided to use realistic delays up to 180 seconds in the bifurcation analysis for better understanding of its effect on the process variables.

Besides, this article describes a comprehensive numerical bifurcation analysis of first-order exothermic reaction in the CSTR-mechanical separator-recycle system with delays up to 180 s using DDE-BIFTOOL for both infinite and finite coolant inlet flowrates.

2. Model description

In the following, the mass and energy balance equations which describe the steady-state and dynamic performance of the CSTR-mechanical separator-recycle system are presented.

A first-order irreversible exothermic elementary reaction was considered for analyzing the steady- state and dynamic performance of the delayed recycle stream in a reactor. The stoichiometry of elementary reaction [57,58] can be represented as

$$s_1 \xrightarrow{k_{2,1}} s_2 \tag{1}$$

In Eq. (1), the kinetic constant $k_{2,1}$ indicates the formation of product s_2 from reactant s_1 by virtue of chemical reaction. According to law of mass action kinetics [57,58], the reaction rates for disappearance of reactant s_1 is

$$r_{s_1} = -k_{2,1} c_{s_1} \tag{2}$$

The constitute relationships considered for the nonlinearity of delayed recycle system are: (i) Arrhenius law of kinetics, and (ii) heat of cooling in a jacketed CSTR. According to Arrhenius law, the temperature dependency of kinetic constants can be represented as

$$k_{2,1} = A_{2,1} \exp\left(-\frac{E_{2,1}}{RT}\right) \tag{3}$$

Fig. 1 illustrates the schematic diagram of CSTR-mechanical separator recycle system. Here, the CSTR was connected with a mechanical separator for the separation of product s_2 from unconverted reactant s_1. The latter was recycled back to the reactor. A constant delay was assumed in the recycle stream for both concentration of reactant, and temperature. The non-isothermal operation of the reactor was considered in the bifurcation analysis.

The exothermic heat released by the reaction mixture in a jacketed CSTR during the reaction must be removed by the coolant flow in a jacket. The convective and conductive heat transfer between the cooling jacket and reactor should be included for finding the heat of cooling requirements. Moreover, the logarithmic mean temperature difference between the cooling medium and reaction mixture must be accounted in the heat of cooling calculations. Thus, the formula [59] for finding the heat of cooling in a