Chemical Engineering Science 60 (2005) 1787 – 1792

Chemical Engineering Science

www.elsevier.com/locate/ces

Shorter Communication

Analytical relationship determining the oscillation period of heat-integrated homogeneous tubular chemical reactor without dispersion

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Received 10 August 2004; received in revised form 7 November 2004; accepted 8 November 2004

Abstract

The derived mathematical formulae enable the exact calculation of the period of temperature and concentrations oscillations of a non-adiabatic homogeneous tubular reactor with the external shell-and-tube heat exchanger. The derivations refer to both cocurrent and countercurrent exchanger.

Keywords: Chemical reactors; Nonlinear dynamics; Oscillations

1. Introduction

The problem of temperature–concentration oscillations in chemical reactors is essential for the design and control of reactors. Accordingly, the value of the oscillation period may have a significant influence on process dynamics and production quality. In some cases, in absence of the possibility of rapid heat removal, sudden temperature changes may cause temporary preheating of feed, product or reactor. On the other hand, the buffer elements of an industrial system may be insufficient to suppress unfavourable long-period oscillations.

Tubular chemical reactors with thermal feedback in an external shell-and-tube heat exchanger are commonly used in industrial processes. Because of the presence of feedback, complex static and dynamic phenomena (Subramanian and Balakotaiah, 1996; Bildea and Diman, 1998), including chaos (Berezowski et al., 2000; Jacobsen and Berezowski, 1998a), may occur in such reactors. The mathematical formulae derived in the paper enable the exact calculation of the period of temperature–concentration oscillations in the above-mentioned system. The reactor assumed in the analysis is a homogeneous reactor without dispersion, cooled internally by constant temperature fluid. The derived relationships refer both to the cocurrent and countercurrent heat exchanger. In the case of a reactor with the cocurrent exchanger, the obtained algebraic expression enables explicit determination of the oscillations period. In the case of a countercurrent exchanger, this period is determined as an iterative solution of the corresponding algebraic relation.

2. Model

Let us consider the model of a tubular homogeneous chemical reactor with the external shell-and-tube heat exchanger. The mathematical form of the model results from the following corresponding balances:

mass balance of *i*th component in the reactor:

$$\frac{\partial \alpha_i}{\partial \tau} + \frac{\partial \alpha_i}{\partial \xi_R} = \phi_i, \quad i = 1, N, \tag{1}$$

energy balance in the reactor:

$$\frac{\partial \Theta_R}{\partial \tau} + \frac{\partial \Theta_R}{\partial \xi_R} = \phi_\Theta, \tag{2}$$

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doi:10.1016/j.ces.2004.11.006

energy balance in the external exchanger-tube of feed:

$$\sigma_F \frac{\partial \Theta_F}{\partial \tau} + \frac{\partial \Theta_F}{\partial \xi_E} = \delta_E(\Theta_P - \Theta_F), \tag{3}$$

tube of products:

$$\sigma_P \frac{\partial \Theta_P}{\partial \tau} + \varepsilon \frac{\partial \Theta_P}{\partial \xi_E} = \delta_E(\Theta_F - \Theta_P). \tag{4}$$

The coefficient “ε” is equal to 1 if the exchanger is cocurrent, whereas for the countercurrent exchanger it is -1.

The required boundary relationships, resulting from the external thermal feedback, are as follows:

for the system with the external cocurrent exchanger ($\varepsilon = 1$):

$$\begin{aligned} &\Theta_R(0,\tau) = \Theta_F((1,\tau),\\ &\Theta_F(0,\tau) = 0,\\ &\Theta_P(0,\tau) = \Theta_R(1,\tau), \end{aligned} \tag{4a}$$

for the system with the external countercurrent exchanger ($\varepsilon = -1$):

$$\begin{aligned} &\Theta_R(0,\tau) = \Theta_F((1,\tau),\\ &\Theta_F(0,\tau) = 0,\\ &\Theta_P(1,\tau) = \Theta_R(1,\tau). \end{aligned} \tag{4b}$$

In the next part of the paper, mathematical relationships are derived to facilitate strict determination of the values of temperature and concentrations oscillations period in the discussed system. In accordance with the balance form of the model of the reactor in (Eqs. (1) and (2)) assumed above, the delay contributed by the reactor equals one, irrespective of the reactor kinetics (Jacobsen and Berezowski, 1998b; Berezowski and Jacobsen, 1998). Accordingly, the next step is to determine the delay caused by the exchanger.

3. Analytical relationship determining the oscillation period for the cocurrent exchanger

By the introduction of deviation variables

$$x_F = \Theta_F - \Theta_{Fs}, \quad x_P = \Theta_p - \Theta_{Ps} \tag{5}$$

(subscript “s” refers to the stationary state), the deviation balance equations of the cocurrent heat exchanger take the form

$$\sigma_F \frac{\partial x_F}{\partial \tau} + \frac{\partial x_F}{\partial \xi_E} = \delta_E(x_P - x_F), \tag{6}$$

$$\sigma_P \frac{\partial x_P}{\partial \tau} + \frac{\partial x_P}{\partial \xi_E} = \delta_E(x_F - x_P). \tag{7}$$

After Laplace transformation with respect to time, the following system is obtained:

$$\frac{\mathrm{d}X_F}{\mathrm{d}\xi_E} = -(\delta_E + s\sigma_F)X_F + \delta_E X_P, \tag{8}$$

$$\frac{\mathrm{d}X_P}{\mathrm{d}\xi_E} = \delta_E X_F - (\delta_E + s\sigma_P)X_P \tag{9}$$

the solution of which is

$$X_F(1,s) = \delta_E \frac{\exp(\lambda_2) - \exp(\lambda_1)}{\sqrt{s^2(\sigma_F - \sigma_F)^2 + 4\delta_E^2}} X_P(0,s), \tag{10}$$

where λ_1 and λ_2 are eigenvalues of the form

$$\lambda_1 = \frac{-s(\sigma_F + \sigma_P) - 2\delta_E - \sqrt{s^2(\sigma_F - \sigma_P)^2 + 4\delta_E^2}}{2}, \tag{11}$$

$$\lambda_2 = \frac{-s(\sigma_F + \sigma_P) - 2\delta_E + \sqrt{s^2(\sigma_F - \sigma_P)^2 + 4\delta_E^2}}{2}. \tag{12}$$

Defining the transfer function of the cocurrent exchanger as

$$K(s) = \frac{X_F(1,s)}{X_P(0,s)}, \tag{13}$$

the following expression is obtained:

$$\begin{aligned} K(s) = \delta_E &\frac{\exp\left(\frac{1}{2}\sqrt{s^2(\sigma_F - \sigma_P)^2 + 4\delta_E^2}\right) - \exp\left(-\frac{1}{2}\sqrt{s^2(\sigma_F - \sigma_P)^2 + 4\delta_E^2}\right)}{\sqrt{s^2(\sigma_F - \sigma_P)^2 + 4\delta_E^2}}\\ &\times \exp\left(-s\frac{\sigma_F + \sigma_P}{2}\right)\exp(-\delta_E). \end{aligned} \tag{14}$$

It directly follows from the above equation that the delay due to the exchanger equals

$$d = \frac{\sigma_F + \sigma_P}{2}. \tag{15}$$

Hence, the basic period of temperature–concentration oscillations of the whole system is expressed as

$$O = 2(1 + d) = 2 + \sigma_F + \sigma_P. \tag{16}$$

Accordingly, the oscillation period of the system with cocurrent exchanger does not depend on the parameters of the reactor. This period has been labelled “the basic period” because in the case of N-period oscillations, the period is $N * O$.