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A method of suppressing chaos in a chemical reactor and the use of chaos for identifying the reactor model

Marek Berezowski*

Cracow University of Technology, Faculty of Chemical Engineering and Technology, 30-155 Kraków, ul. Warszawska 24, Poland

A method of suppressing chaotic oscillations in a tubular reactor with mass recycle is discussed. The method involves intervention in the temperature of the input flow by the recirculation flow and the temperature set from the exterior. The most advantageous solution was proved to be heat coupling elimination and maintenance of the reactor input temperature on the set level. Moreover, the reactor model was identified on the basis of a chaotic solution, as it provides the biggest entropy of information.

Keywords: chemical reactor, recycle, chaos, control, fractals, identification

1. INTRODUCTION

In chemical engineering there are many systems and apparatuses that can operate in a chaotic way. One group consists of various types of chemical reactors and systems constructed from them, including both CSTRs (Berezowski et al., 2009; Berezowski, 2017; Chien-Chong Chen, 1996; Zukowski et al., 2000), and tubular reactors: with dispersion, without dispersion, adiabatic, non-adiabatic, heterogeneous, homogeneous, etc. (Antoniades et al., 2001; Berezowski et al., 2015; Berezowski, 2000; Berezowski, 2001; Berezowski, 2009; Berezowski, 2013; Elnashaie et al., 1995; Femat et al., 2004; Jacobsen and Berezowski, 1998; Luss et al., 1966; Rehacek et al., 1998; Russo et al., 2006). One of such reactors is a homogeneous, non-adiabatic tubular chemical reactor without dispersion and with mass recycle. Its schematic diagram is presented in Fig. 1.

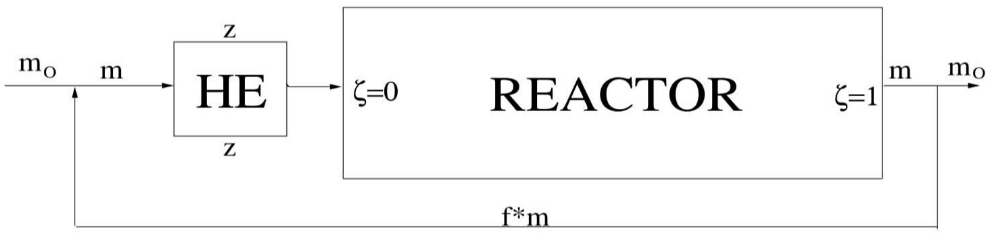

Fig. 1. Schematic diagram of the reactor with recycle

The reactor can generate various types of dynamic behaviour, in the simplest case, stationary states. There may also be periodic, quasiperiodic and chaotic oscillations (Berezowski, 2000; Jacobsen et al., 1998). The Feigenbaum diagram of steady states is presented in Fig. 2, where the influence of cooling medium θ_H on

* Corresponding author, e-mail: marek.berezowski@pk.edu.pl

the degree of raw material conversion at the reactor outlet $\alpha(1)$ is shown. Steady states, but also periodic and chaotic oscillation are visible in the diagram. From the process point of view, chaotic oscillations are the most unfavourable, or even dangerous events, because in such a case the process may become unpredictable. Likewise, it may be inferred from Fig. 2. that there are cases where the amplitudes of chaotic oscillations have a very big range, i.e. $0.15 < \alpha(1) < 1$, which may be especially unfavourable.

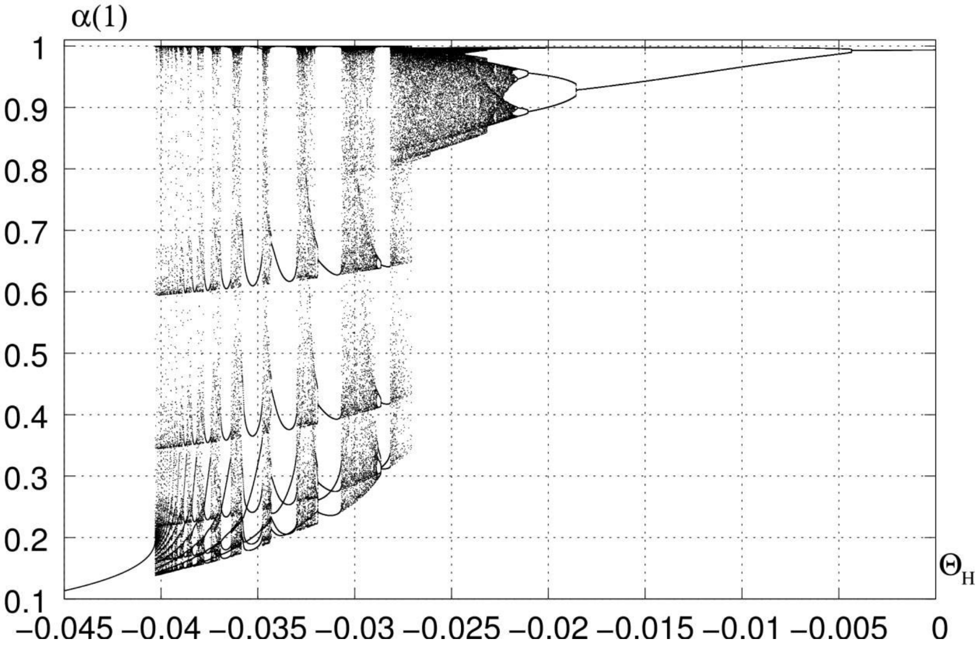

Fig. 2. Feigenbaum diagram of the reactor without control ($h = 1$)

Therefore, it would be expedient to design an apparatus in the manner enabling the suppression of chaos. One of the solutions is to assume such working parameters of the reactor for which it does not operate chaotically. In consideration of the process, this is not always possible. Thus, the remaining option is to design appropriate control to suppress oscillations and reduce them to the stationary state, for example, by intervening into the reactor inlet temperature $\theta(0)$.

2. REACTOR MODEL

The mathematical model of the discussed reactor is as follows (Berezowski, 2000; Jacobsen et al., 1998):

- mass balance:

$$\frac{\partial \alpha}{\partial \tau} + \frac{\partial \alpha}{\partial \varsigma} = \phi(\alpha, \theta) \tag{1}$$

- heat balance:

$$\frac{\partial \theta}{\partial \tau} + \frac{\partial \theta}{\partial \varsigma} = \phi(\alpha, \theta) + \delta(\theta_H - \theta) \tag{2}$$

Assuming that a single reaction of the A → B n-th order occurs in the reactor, the kinetic function of the reaction has the form:

$$\phi(\alpha, \theta) = \mathrm{Da}(1-\alpha)^n \exp\left(\gamma \frac{\beta\theta}{1+\beta\theta}\right) \tag{3}$$

where: $0 \leq \varsigma \leq 1$.

Due to the presence of the recirculation loop, the mathematical model of the reactor must be supplemented with proper boundary conditions concerning the coupling of mass and heat:

$$\alpha(\tau, 0) = f\alpha(\tau, 1) \tag{4}$$

$$\theta(\tau, 0) = f\theta(\tau, 1) \tag{5}$$

From the mathematical point of view, this model is discrete, rendering in the solution a horizontal wave of variables α and θ (Jacobsen et al., 1998). According to scientific publications, in some cases the wave may have a chaotic character, which was also shown in Fig. 2 (Berezowski, 2000).

The above-mentioned control changes boundary condition (5) and transforms it to:

$$\theta(\tau, 0) = (1 - h)z + hf\theta(\tau, 1) \tag{6}$$

where: $0 \leq h \leq 1$, $0 \leq f \leq 1$.

In the above equations, z is the set control temperature, whereas h is the coefficient determining the contribution of temperature $\theta(\tau, 1)$ and temperature z in the control process (see SYMBOLS). Parameter f is the recycle coefficient designating which part of the product flow is reversed. Thus, for $h = 0$ the heat recycle is completely shut off (complete absence of heat coupling), and only mass coupling remains, expressed by condition (4). Then, the influence of temperature $\theta(\tau, 1)$ is zero. However, for $h = 1$ the system has full recycle, both as far as mass and heat are concerned. The influence of temperature z is zero and the control of the reactor is completely shut off. As can be seen from formula (6), the above control is realised through a heat exchanger, which is affected by a stream with a temperature $f\theta(\tau, 1)$ and control stream with constant temperature z. The flow with temperature $f\theta(\tau, 1)$ is a mixture of the recycle stream with the raw material stream. A stream with a temperature defined by the formula (6) flows out of the exchanger. It should be added that this control takes place at the design stage of the installation, not in real time.

3. CALCULATIONS AND ANALYSIS OF THE RESULTS

The following parameter values were assumed for the calculations: Da = 0.075, $n = 1.5$, $\beta = 2$, $\gamma = 15$, $\delta = 1.5$, $f = 0.5$, $\theta_H = -0.03$. At first, the influence of coefficient h and temperature z on chaos suppression was examined. To obtain this, a fractal diagram of the steady states with coordinates z and h was constructed, see Fig. 3.

The third coordinate is conversion degree $\alpha(1)$, which is not a numerical axis but a colour. Accordingly, the principle is that the darker the green, the higher value of conversion degree $\alpha(1)$. Hence, it may be inferred from the Figure that for small values of variable z and any value of parameter h, the process has a rapid, i.e. chaotic nature. Chaos disappears only for bigger values of z. Concurrently, it should be noticed that the process is not chaotic for $h = 0$ and $z > 0.09$. Such conditions are favourable for the design of control, as the boundary condition takes the following form:

$$\theta(\tau, 0) = z \tag{7}$$

causing the heat coupling to be completely shut off and only the mass coupling remaining. This, in turn, makes the mathematical model one-dimensional with one state variable α. Then, variable θ completely depends on α.

In the next step (assuming that $h = 0$), the quantitative influence of variable z on $\alpha(1)$ was investigated, in accordance with Fig. 4.

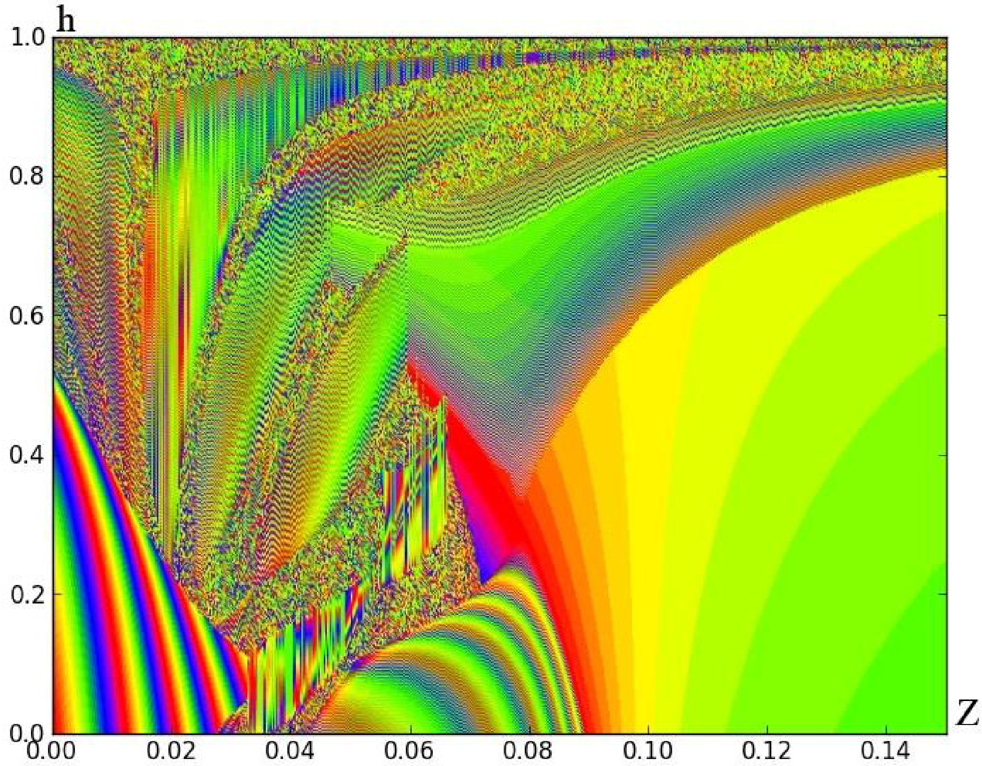

Fig. 3. Fractal graph of the steady states of the reactor with control

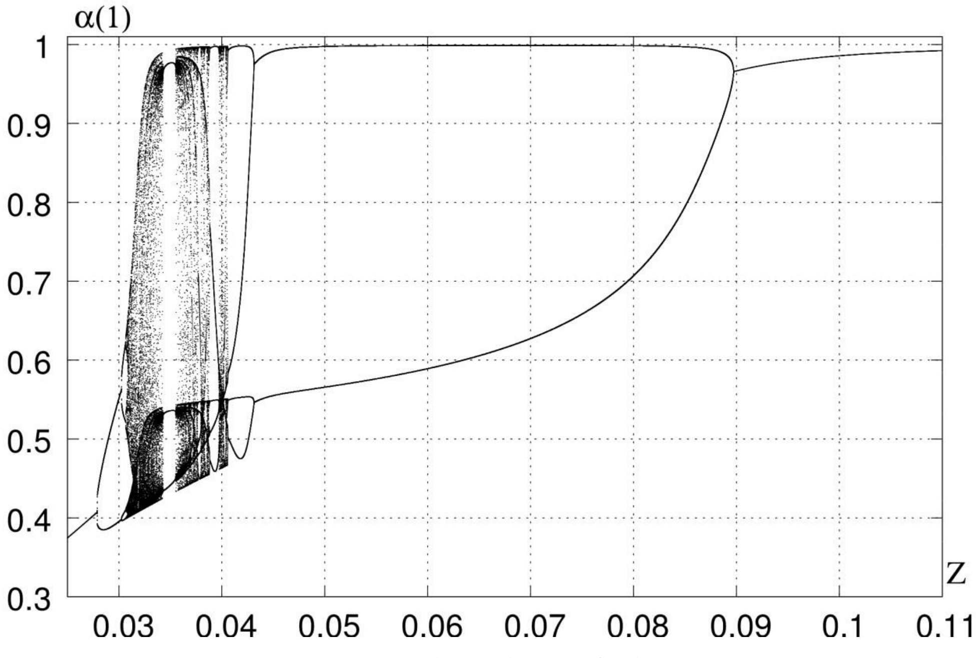

Fig. 4. Feigenbaum diagram for $h = 0$

It may be observed in Figure 4 that for $z > 0.09$ the reactor works in the stationary state, which is characterized by a very high conversion degree $\alpha(1)$. To verify the results, the graph of Lyapunov exponent λ was plotted (Fig. 5).

Positive values of the exponent confirm chaos, whereas the negative ones exclude it. Thus, accepting the following values of control parameters: $h = 0$ and $z = 0.1$, the assumptions are fully guaranteed, i.e. chaos suppression, reduction of the reactor to stationary operation, provision of the highest degree of conversion.

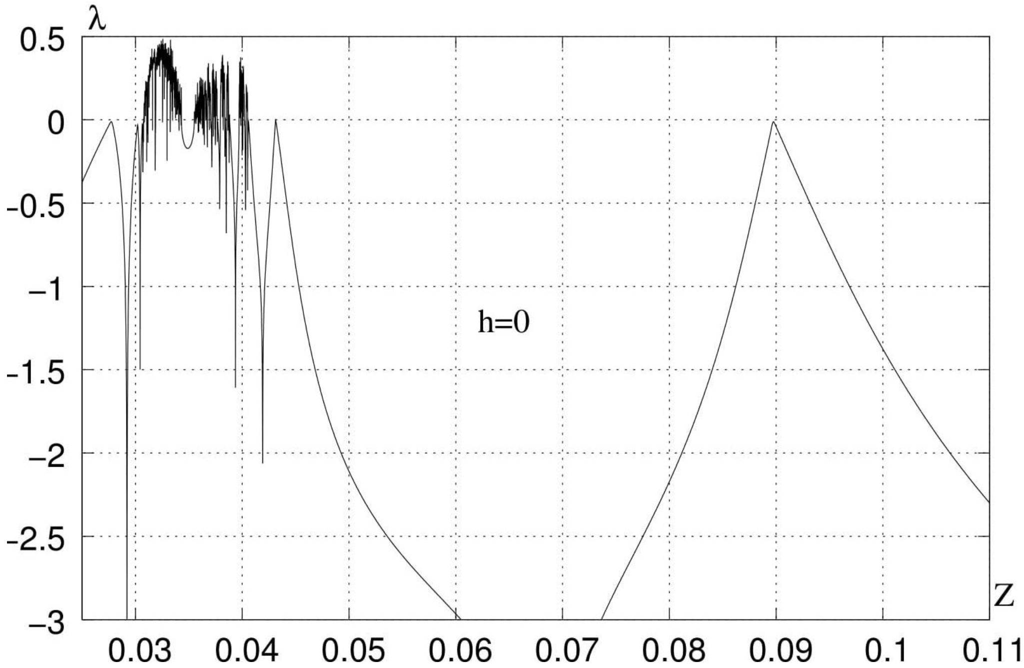

Fig. 5. Diagram of Lyapunov exponent for $h = 0$

4. IDENTIFICATION OF THE REACTOR MODEL

As proved above, if $h = 0$ is assumed, the reactor model becomes one-dimensional and is expressed by means of differential equations (1)–(2) and boundary condition (7). The inlet temperature of the reactor $\theta(\tau, 0)$ does not depend then on the outlet temperature of the reactor $\theta(\tau, 1)$, i.e. it does not depend on itself and it is equal to the temperature of the external factor z (Eg. (7)). It would be useful to solve this model in an explicit analytical form. Chaos gives such a chance. In the examined model, chaos occurs for small values of z (Figs. 3 and 4). When the system is introduced into a chaotic state, it has a very high value of the entropy of information (theoretically, infinitely high) (Bizon et al., 2012 – Fig. 3). This means that the system generates the maximum quantity of information about its model. To illustrate this phenomenon, popular logistic equations $x_{k+1} = rx_k(1 - x_k)$ may be evoked, the right side of which is a reversed parable. By constructing a graph $(x_k x_{k+1})$ numerically for parameter r that secures chaos, this parable is derived (Wikipedia, Logistic Map; Berezowski, 2001 – Fig. 6; Foryś et al., 2003 – Fig. 1).

In the paper, the above method was used for the discussed reactor. Assuming that $z = 0.037$, graph $[\alpha_k(1),\ \alpha_{k+1}(1)]$ was derived, see Fig. 6.

It is an attractor comprising inconsistent points that form one layer, which proves that we are dealing with a one-dimensional model. If the model were multi-dimensional, a fuzzy cloud comprising many layers would be observed in the figure (theoretically, comprising an infinite number of layers), typical for Henon attractor (Berezowski, 2009 – Fig. 1; Berezowski, 2006 – Fig. 2; Berezowski, 2001 – Fig. 10). The presented graph has a significant cognitive importance. The graph line (comprising very many inconsistent points) constitutes an explicit geometric form of the solution of the reactor model. If we wrote equation of a line from Fig. 6 in the form $\alpha = F(\alpha)$, we would have an explicit form of the reactor model, without differential equations. In the dynamic process it is $\alpha_{k+1} = F(\alpha_k)$. Similar to the logistic equation, where $x = rx(1 - x)$ is an inverted parabola (Wikipedia, Logistic Map). A similar approach was used in (Berezowski et al., 2014; Lawnik et al., 2014).

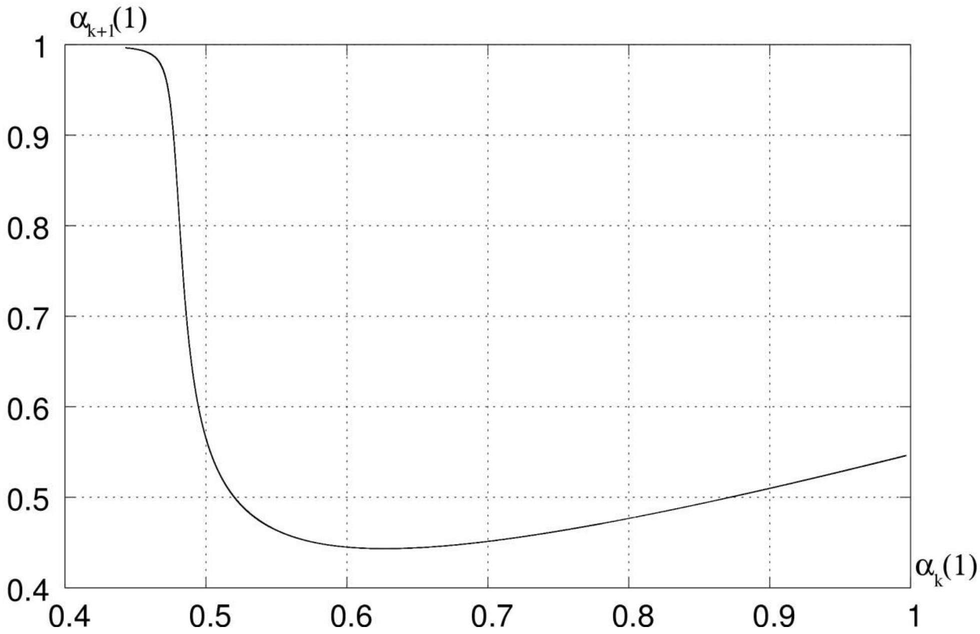

Fig. 6. Attractor for $h = 0$

5. CONCLUSIONS

Chemical reactors may operate in steady states, in periodic oscillations, quasiperiodic oscillations, as well as in chaotic states. In principle, chaos is disadvantageous, as it leads to unpredictability of the process, which may be dangerous. For this reason, it is beneficial to devise a method that enables the liquidation of chaos in favour of the stationary state with a high degree of conversion. This objective was achieved in the paper. It was proved that the most convenient manner of control is to maintain the temperature at the reactor input on a set level. However, a proper selection of the temperature values is required to avoid consecutive chaos generated by the control.

The reactor model was also identified in the paper, taking advantage of the fact that chaos renders the biggest entropy of information. As a result, the line illustrating an explicit mathematical model of the apparatus was derived.

SYMBOLS

c_p	heat capacity, kJ/(kg·K)
C_A	concentration of component A, kmol/m^3
Da	Damköhler number $\left(= \dfrac{V_R(-r_0)}{\dot{F}C_0}\right)$
E	activation energy, kJ/kmol
f	recycle coefficient
$\dot{F}$	volumetric flow rate, m^3/s
h	control coefficient $\left(= \dfrac{1}{1+\delta_z}\right)$
HE	control heat exchanger
$(-\Delta H)$	heat of reaction, kJ/kmol
k	reaction rate constant, 1/s(m^3/kmol)$^{n-1}$
L	length, m

m	mass flow, kg/s
n	order of reaction
$(-r)$	rate of reaction ($= kC^n$), kmol/(m^{3}s)
R	gas constant, kJ/(kmol·K)
t	time, s
T	temperature, K
V	volume, m^3
x	position, m
z	dimensionless control temperature $\left(= \dfrac{T_z - T_0}{\beta T_0}\right)$

Greek letters

α	degree of conversion $\left(= \dfrac{C_{A0} - C_A}{C_{A0}}\right)$
β	dimensionless number related to adiabatic temperature increase $\left(= \dfrac{(-\Delta H)C_{A0}}{T_0 \rho c_p}\right)$
γ	dimensionless number related to activation energy $\left(= \dfrac{E}{RT_0}\right)$
δ	dimensionless heat exchange coefficient $\left(= \dfrac{A_q k_q}{\rho c_p \dot{F}}\right)$
λ	Lyapunov exponent
θ	dimensionless temperature $\left(= \dfrac{T - T_0}{\beta T_0}\right)$
z	dimensionless position $\left(= \dfrac{x}{L}\right)$
ρ	density $\left(= \dfrac{\text{kg}}{\text{m}^3}\right)$
τ	dimensionless time $\left(= \dfrac{\dot{F}}{V_R} t\right)$

Subscripts

0	refers to feed
H	refers to temperature of cooling medium
R	refers to reactor
z	refers to control

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