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The effect of periodic disturbances of inlet parameters on the performance of a chemical reaction

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Abstract—The aim of the present work is to study possible responses of a system with combustion to different disturbances under the conditions when the initial regime is characterized by damped oscillations. Mathematically the problem is formulated by a system of ordinary differential heat and mass balance equations. Damped oscillations, appearing in a model on instantaneous perturbation, are characterized by the natural frequency of the system, while the ability to remember perturbations as oscillations is represented by the quality factor. Linear analysis allows one to relate the parameters of a chemically reacting system. Resonance properties of the system with combustion are studied, the resonance phenomena are established at frequency multiples and subharmonics, the beating and parametric excitation of oscillations are found, and the system sensitivity to the shape of the perturbative signal and different perturbative parameters is studied. The results obtained can be used for controlling combustion intensity in a continuously stirred tank reactor.

AN EFFICIENT means for studying combustion of a fuel mixture in a continuous stirred tank reactor has proved to be the zero-dimensional model [1, 2]. It allows one to separate the parametric regions of non-unique, auto-oscillation and damped oscillation regimes analytically in a linear approximation [1]. The aim of the present work is to study possible responses of a system with combustion to different disturbances under conditions when the initial regime is characterized by damped oscillations.

Mathematically, the problem is formulated by a system of ordinary differential equations of heat and mass balance written in terms of non-dimensional variables

$$\frac{\mathrm{d}\theta}{\mathrm{d}Fo} = -4\theta+\delta\eta\exp\left(\frac{\theta}{1+u\theta}\right) \equiv f_1(\theta,\eta)$$

$$\frac{\mathrm{d}\eta}{\mathrm{d}Fo} = -Le(\eta-\eta_0)-\delta\gamma\eta\exp\left(\frac{\theta}{1+u\theta}\right) \equiv f_2(\theta,\eta)$$

$$Fo = 0;\quad \theta = \theta_{\mathrm{H}},\quad \eta = \eta_{\mathrm{H}} \tag{1}$$

where

$$\theta = \frac{E}{RT_0^2}(T-T_0),\quad \eta = \frac{C}{C_{\mathrm{H}}}$$

$$Fo = \frac{at}{l^2},\quad u = \frac{Rt_0}{E}$$

$$\delta = \frac{E}{RT_0^2}\frac{q}{\lambda}l^2k_0C_{\mathrm{H}}\exp\left(-\frac{E}{RT_0}\right)$$

$$\gamma = \frac{RT^2}{E}\frac{C_v\rho}{qC_{\mathrm{H}}}$$

$$Le = \frac{D}{a}.$$

Here T, C and t are the temperature, mixture concentration and time; E, q and k_0 are the activation energy, reaction thermal effect and the pre-exponential factor in the expression for the reaction rate constant; R is the universal gas constant; λ, D, a, C_v and ρ are the thermal conductivity, mass diffusivity, thermal diffusivity, heat capacity and the density; Le, δ and γ are the Lewis number, the Frank–Kamenetsky parameter and the burning parameter, respectively.

Damped oscillations, appearing in model (1) on instantaneous perturbation, are characterized by the natural frequency of the system, while the ability to remember perturbations as oscillations is represented by the quality factor.

Linear analysis allows one to relate the natural oscillation frequency and the quality factor to the parameters of a chemically reacting system.

Let us perform linear transformations of system (1) with the matrix

$$\mathrm{A} = \begin{bmatrix} \left(\frac{\partial f_1}{\partial\theta}\right)_s & \left(\frac{\partial f_1}{\partial\eta}\right)_s \\ \left(\frac{\partial f_2}{\partial\theta}\right)_s & \left(\frac{\partial f_2}{\partial\eta}\right)_s \end{bmatrix}.$$

Then the characteristic matrix equation can be written as

$$v^2-\sigma v+\Delta = 0$$

where

NOMENCLATURE

a thermal diffusivity
A amplitude
c_v heat capacity
C concentration
D molecular diffusion coefficient
E activation energy
f_1, f_2 functions
Fo dimensionless time
k_0 pre-exponential factor
l length of reactor
Le Lewis number
q heat of reaction
Q quality factor
R universal gas constant
t time
T absolute temperature
u dimensionless parameter.

Greek symbols
A matrix of linear transformations
γ parameter of burning
δ Frank–Kamenetsky parameter
θ dimensionless temperature
η dimensionless concentration
λ thermal conductivity
ν eigenvalues
ρ density
σ trace of the matrix
ω frequency
ω_0 natural frequency.

Subscripts
a thermal conductivity
D molecular diffusion
H initial
s steady state
x chemical reaction
0 inlet.

$$\sigma = \left(\frac{\partial f_1}{\partial \theta}\right)_s + \left(\frac{\partial f_2}{\partial \eta}\right)_s$$

and

$$\Delta = \left(\frac{\partial f_1}{\partial \theta}\right)_s\left(\frac{\partial f_2}{\partial \eta}\right)_s - \left(\frac{\partial f_2}{\partial \theta}\right)_s\left(\frac{\partial f_1}{\partial \eta}\right)_s$$

are the trace and the determinant of the matrix, respectively. The subscripts refers to the steady states of system (1) which satisfy the conditions

$$f_1(\theta_s, \eta_s) = f_2(\theta_s, \eta_s) = 0.$$

With $\sigma < 0$ and $\sigma^2 - 4\Delta < 0$ the solution of the linear equations

$$\dot{\mathbf{X}} = \mathbf{A}\mathbf{X}$$

for small deviations of system (1) from equilibrium positions has the form of damped oscillations with the frequency

$$\omega_0 = \sqrt{\left(\Delta - \frac{\sigma^2}{4}\right)}. \tag{2}$$

The quality factor of an oscillatory system is calculated by the relation

$$Q = \frac{\omega_0}{-\sigma}. \tag{3}$$

When $\sigma \to 0$, damping is weak, i.e. the oscillatory system has a high quality factor. When $\sigma^2 - 4\Delta \to 0$ (see equation (2)), the system losses its oscillatory properties.

In the case of a high quality factor for $u \to 0$, equation (2) is reduced to

$$\omega_0 \approx \sqrt{\left(\frac{4(4Le + \delta\gamma\exp\theta_s)^2 - (\delta\gamma\exp\theta_s)\dfrac{(4Le)^2\eta_0}{\gamma}}{4Le + \delta\gamma\exp\theta_s}\right)}. \tag{4}$$

Since the Lewis number is proportional to the ratio of the characteristic thermal and diffusive relaxation times Fo/Fo_D, and the quantity $(\delta\gamma \exp \theta_s)^{-1}$ is proportional to the ratio between the chemical and thermal times Fo_x/Fo_a, equation (4) relates the natural oscillation frequency to the characteristic times of the process. Equation (4) can easily yield the conditions for the existence of oscillations at a high quality factor of the system

$$4\left[1 + \frac{\delta\gamma\exp\theta_s}{4Le}\right]^2 \frac{\gamma}{\eta_0}\frac{1}{\delta\gamma\exp\theta_s} > 1 \tag{5}$$

or

$$4\left[1 + \frac{1}{4}\frac{Fo_D}{Fo_x}\right]^2 \frac{Fo_x}{Fo_a}\frac{\gamma}{\eta_0} > 1.$$

According to equation (3), the quality factor of the oscillatory system for this case can be found from the relation

$$Q = \sqrt{\left(\frac{4(4Le + \delta\gamma\exp\theta_s) - (4Le)^2(\eta_0/\gamma)[(4Le/\delta\gamma\exp\theta_s) + 1]}{\{4(Le+1) + (\delta\gamma\exp\theta_s)[1 - 4Le(\eta_0/\gamma)/4Le + \delta\gamma\exp\theta_s]\}^2} - \frac{1}{4}\right)}. \tag{6}$$

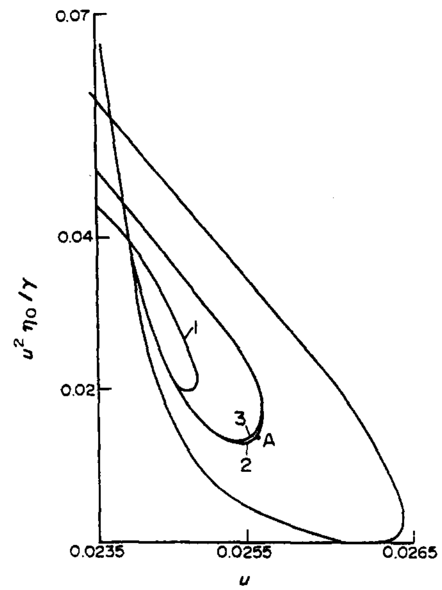

FIG. 1. The spatial pattern of lines of the same quality factor projected onto the plane $u^2\eta_0/\gamma$, u. The lines $\sigma^2-4\Delta = O(1)$ correspond to $Q = 0$, line 2 corresponds to $Q = 25$, and the line $\sigma = 0$ to $Q = \infty$.

It is seen that at small input concentrations η_0 the quality factor is determined only by the characteristic times of the process

$$Q \to \sqrt{\left(\frac{4(Fo_a Fo_D + 4Fo_a Fo_x)Fo_x Fo_D}{[4(Fo_a Fo_x + Fo_D Fo_x) + Fo_a Fo_D]^2} - \frac{1}{4}\right)}.$$

Along with the system response to an instantaneous perturbation, it is interesting to study the resonance properties of a chemically active medium under the action of periodic external perturbations. Since resonance properties are exhibited by systems with high quality factors, let us consider such systems. The resonance properties of non-linear system (1) will be studied numerically. For $\delta = 1.394$, $\gamma = 0.199$, $u = 0.0252$, $Le = 0.1$ and $\eta_0 = 4$ the natural frequency and the quality factor (according to equations (4) and (6)) are $\omega_0 = 2$ and $Q = 25$. In this case, condition (5) for the existence of an oscillatory process with a high quality factor holds. In the parametric space (δ, γ, u, Le, η_0) the point with the specified coordinates (see Fig. 1) located near the boundary $\delta = 0$. Owing to the perturbation acting upon system (2), it acquires a number of specific properties. Figures 1–4 illustrate the influence of periodic perturbations of concentrations at the reactor inlet on combustion according to the law

$$\eta_0 = \eta_{01}[1 + A\sin(\omega Fo)]. \tag{7}$$

From Fig. 2 it is seen that if the action frequency is close to the natural one, a heating regime is realized (line 1). When the action has the frequency $\omega \approx 2\omega_0$, an oscillatory regime is realized with the frequency ω_0, i.e. the parametric excitation of oscillations takes place (Fig. 2(a), line 2). This phenomenon can be used for amplifying natural frequency oscillations. The amplitude of resulting oscillations is about twice as high as the initial amplitude of damped oscillations in the unperturbed system. It was found that at a frequency close to 3/2 times of the natural frequency, one can observe steady state subharmonic oscillations different in shape and amplitude (see Figs. 2(b) and (c)) which are realized when passing to this frequency from the lower (b) and the higher (c) frequencies ω. It should be noted that this phenomenon was observed within the frequency range $\omega = 2.933$–2.950 at the natural frequency $\omega = 2$.

Along with the sine perturbation (7), the action of periodic signals of rectangular

$$\eta_0 = \eta_{01}\left\{1 + \frac{4A}{\pi}\left[\sin(\omega Fo) + \frac{1}{3}\sin(3\omega Fo) + \cdots\right]\right\} \tag{8}$$

and triangular

$$\eta_0 = \eta_{01}\left\{1 + \frac{2A}{\pi}\left[\sin(\omega Fo) - 0.5\sin(2\omega Fo) + \frac{1}{3}\sin(3\omega Fo) - \cdots\right]\right\} \tag{9}$$

shapes on system (1) was also considered.

Figure 3(a) shows the phase plots at different perturbative signal shapes [1, equation (8); 2, equation (7); 3, equation (9) for $A = 0.2$; $\omega = 2$].

It should be noted that in the case of actions (8) and (9), as well as for (7), resonance appears at a frequency close to the natural one. However, the forced oscillation amplitude rises with an increase in the profile fullness (see curves 3–1 in Fig. 3(a)).

The exothermal chemical reaction is more sensitive to temperature perturbations than concentration perturbations at the reactor inlet. Even in the case of a weak effect of temperature pulsations

$$T = T_{01}[1 + A\sin(\omega Fo)] \tag{10}$$

on combustion at a frequency differing from the resonance frequency ($\omega = 2.95$; $A = 0.01$), the amplitude of forced oscillations is four times higher than that for concentration perturbations according to law (7) (see lines 1 and 2 respectively in Fig. 3(b)).

The amplitude–frequency characteristics of the system with combustion allow one to illustrate other non-linear properties of system (1). Figure 4 shows such curves for perturbations of form (7) at different values of $A = 0.02$ (1), 0.05 (2), 0.1 (3) and 0.2 (4). It should be noted that in the case of complex steady state oscillations (see, for instance, line 1 in Fig. 2(a)), the maximum (for the period) amplitudes were plotted. As is seen from Fig. 4, at all the action amplitudes there is a resonance of frequency ω_0. As the action amplitude increases (see curves 2 and 4), the resonances are more pronounced at frequency

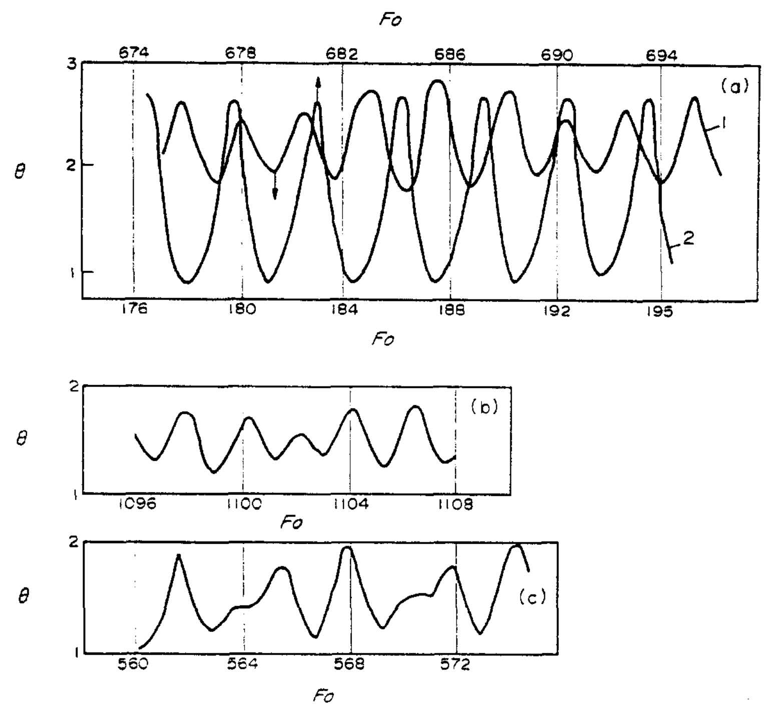

Fig. 2. Steady state oscillations under the action of form (7). $A = 0.1$. (a) $\omega = 2.53$ (1), $\omega = 4$ (2); (b) $\omega = 2.94$; (c) $\omega = 2.94$.

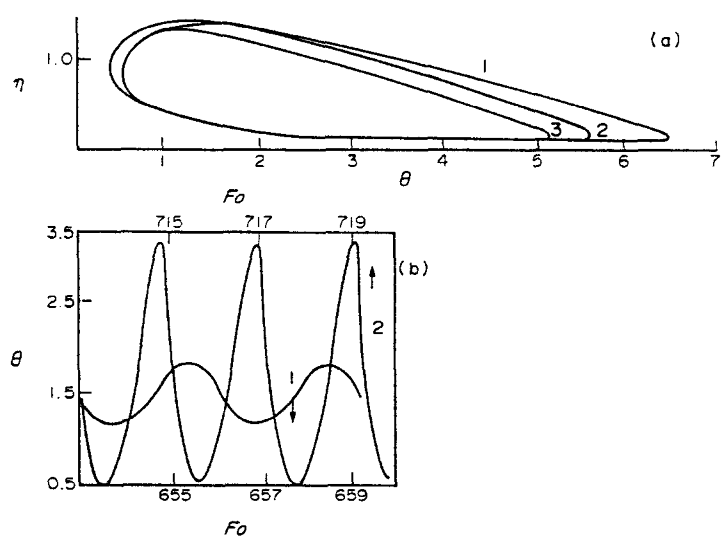

Fig. 3. The combustion sensitivity to the shape of the perturbative signal (a) and to different perturbative parameters (b).

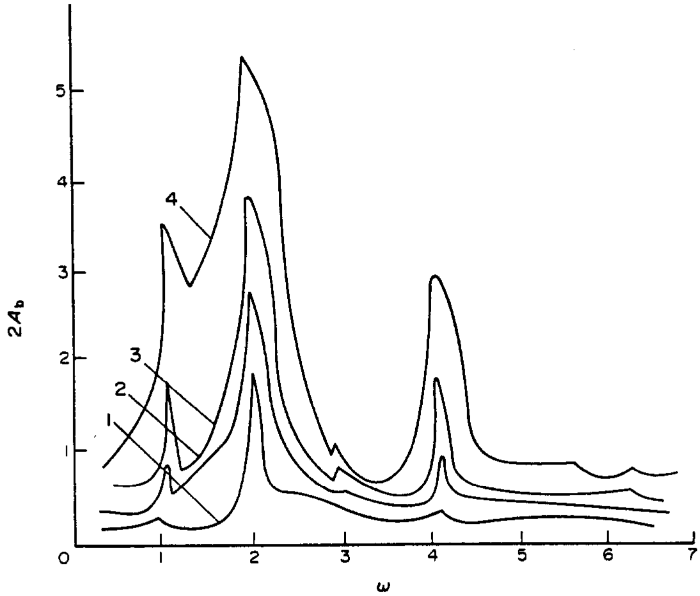

FIG. 4. The amplitude–frequency characteristics of the system with combustion with perturbations of form (7).

multiples and subharmonics: at $\omega = 2\omega_0$, $\omega = \omega_0/2$, and then at $\omega = 3\omega_0$. As the perturbation amplitude increases, the resonance frequencies decrease and the resonance peaks become asymmetrical.

Thus, the resonance properties of a system with combustion have been studied, the resonance phenomena have been established at frequency multiples and subharmonics, the beating and parametric excitation of oscillations have been found, and the system sensitivity to the shape of the perturbative signal and different perturbative parameters has been studied. The results obtained can be used for controlling combustion intensity in a continuously stirred tank reactor.

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EFFET DES PERTURBATIONS PERIODIQUES DES PARAMETRES D'ENTREE SUR LA PERFORMANCE D'UNE REACTION CHIMIQUE

Résumé—On étudie les réponses possibles d'un système avec combustion à différentes perturbations quand le régime initial est caractérisé par des oscillations amorties. Mathématiquement le problème est formulé par un système d'équations aux dérivées partielles pour les bilans de chaleur et de masse. Des oscillations amorties, apparaissant dans un modèle de perturbations instantanées, sont caractérisées par la fréquence naturelle du système tandis que l'aptitude à mémoriser des perturbations comme des oscillations est représentée par le facteur de qualité. Une analyse linéaire relie les paramètres d'un système en réaction chimique. Des propriétés de résonance du système avec la combustion sont étudiées; les phénomènes de résonance sont établis à des fréquences multiples et subharmoniques; on trouve le battement et l'excitation paramétrique des oscillations, la sensibilité du système à la forme du signal perturbateur et à différents paramètres perturbateurs. Les résultats obtenus peuvent être utilisés pour contrôler l'intensité de la combustion dans un réacteur agité en continu.

DER EINFLUSS PERIODISCHER STÖRUNGEN VON EINGANGSPARAMETERN AUF DEN ABLAUF EINER CHEMISCHEN REAKTION

Zusammenfassung—Ziel der vorliegenden Arbeit ist es, die mögliche Reaktion eines Verbrennungssystems auf unterschiedliche Störungen zu beschreiben, wenn das ursprüngliche Verhalten durch gedämpfte Schwingungen charakterisiert wird. Mathematisch beschrieben wird das Problem durch ein System gewöhnlicher Differentialgleichungen für den Wärme- und Stofftransport. Gedämpfte Oszillationen, die nach einer einmaligen Störung auftreten, sind durch die Eigenfrequenz des Systems bestimmt. Der Qualitätsfaktor beschreibt die Fähigkeit, Störungen als Oszillationen wiederzuerkennen. Mittels linearer Analysis werden die Parameter des chemisch reagierenden Systems untersucht. Resonanzeigenschaften des Systems mit Verbrennung werden untersucht und bei Vielfachen und Subharmonischen von Frequenzen nachgewiesen. Außerdem ist die Empfindlichkeit des Systems bezüglich der Form des störenden Signals und verschiedener Störparameter von Interesse. Die Ergebnisse können zur Regelung der Verbrennungsintensität in einem kontinuierlich betriebenen Tankreaktor verwendet werden.

ВЛИЯНИЕ ПЕРИОДИЧЕСКИХ ВОЗМУЩЕНИЙ ВХОДНЫХ ПАРАМЕТРОВ НА РАБОТУ ХИМИЧЕСКОГО РЕАКТОРА

Аннотация—Целью данной работы является исследование возможных откликов системы с горением на различные возмущения в условиях, когда первоначально режим характеризуется затухающими колебаниями. Математическая задача формулируется системой обыкновенных уравнений баланса тепла и массы. Затухающие колебания, возникающие в модели при мгновенном возмущении, характеризуются собственной частотой системы, способность помнить возмущения в виде колебаний—добротностью. Линейный анализ позволяет связать параметры химически реагирующей системы. Исследованы резонансные свойства системы с горением, установлены явления резонанса на кратных частотах и субгармониках, обнаружены биения и параметрическое возбуждение колебаний, изучена чувствительность системы к форме возмущающего сигнала и к различным возмущающим параметрам. Полученные результаты могут использоваться для управления интенсивностью горения в непрерывном реакторе идеального смешения.