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Modelling of resonance effects during burning in a continuous-flow reactor

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Abstract—The effect of pulsating laser radiation, selectively absorbed by a buffer additive, on gaseous mixture burning in an ideal-mixing continuous-flow reactor is studied in linear approximation. The relation is found between a resultant amplitude of system oscillations, frequency and outside effect amplitude. Resonance phenomena on a natural frequency and on frequency multiples of it are determined. It is shown that in the resonance vicinity on the natural frequency the amplitude-frequency response may be of both a crisis-free and hysteresis character with the amplitude of induced oscillations being proportional to the cube root of the outside effect amplitude. The results of analytical investigations are confirmed by a numerical solution of a non-linear problem.

1. INTRODUCTION

THE PROBLEM of the control of heat and mass transfer with exothermal chemical conversions, substantiation of the experiments on the simulation of various reactions necessitate finding adequate theoretical models and methods for their analysis to study laser heating dynamics of chemically active media. A theoretical investigation of the dynamics of ideal-mixing continuous-flow reactors [1–3], including that in a laser radiation field [4–6] with various types of the dependence of medium absorptivity on concentration and temperature, shows that, depending on the parameters, one, three or five stationary states or a self-oscillating mode are possible in the system. Physical parameters of the energy source greatly affect the character of its interaction with an oscillatory non-linear system [7].

Possible responses of the system with burning and without laser radiation on the disturbances of temperature, concentration and mixture feed rate are studied numerically [8–10]. It is shown that, depending on the system Q-factor, the phenomena of resonance, detector effects, synchronization or parametric excitation are observed.

The paper presents the results of approximate-analytical and numerical analysis of the effect of laser radiation periodic pulsations on oscillating burning of a gaseous mixture in an ideal-mixing continuous flow-reactor.

2. PROBLEM FORMULATION

Heat and mass transfer with exothermal chemical conversions in the ideal-mixing continuous-flow reactor is considered within the framework of the zero-dimensional thermodiffusion model. It is assumed that a gaseous mixture flow rate is constant, the reactor walls are impermeable and heat exchange with the environment occurs by the Newton law. The temperature dependence of the monomolecular reaction rate $A \to B$ obeys the Arrhenius law. Laser radiation propagates along the reactor axis through an optically thin layer of the gaseous mixture. It is assumed that laser radiation is absorbed by the buffer additive which does not participate in a chemical reaction.

In the non-dimensional variables the process is described by the following heat and substance balance equations

$$\tau > 0: \quad \frac{\mathrm{d}\theta}{\mathrm{d}\tau} = -(Pe_{\mathrm{ef}} + Bi_{\mathrm{ef}})\theta + Pe_{\mathrm{df}}\theta_{\mathrm{en}} + Bi_{\mathrm{ef}}\theta_{\infty} + QDac\exp\left(-\frac{1}{\theta}\right) + F(1 + A\sin\omega\tau) \equiv N_1(\theta, c),$$

$$\frac{\mathrm{d}c}{\mathrm{d}\tau} = -(Pe_{\mathrm{ef}} + Le_{\mathrm{ef}})(c - c_{\mathrm{en}}) - Dac\exp\left(-\frac{1}{\theta}\right) \equiv N_2(\theta, c),$$

$$\tau = 0: \quad \theta = \theta_{\mathrm{in}}, \quad c = c_{\mathrm{in}}, \tag{1}$$

where

$$Pe_{\mathrm{ef}} = 2(Pe + 2), \quad Le_{\mathrm{ef}} = 4(Le_{\mathrm{ax}} - 1),$$

$$Bi_{\mathrm{ef}} = \frac{6Bi_T}{2 + Bi_T}\lambda_{\mathrm{rax}}x_1^2$$

are the effective coefficients characterizing the reagent feed rate, mixing and heat exchange with the environment; k_0, E and $(-\Delta H)$ are the pre-exponential factor, activation energy, reaction thermal effect; R is the universal gas constant; L and R_0 the length and radius of the reactor; v the mixture feed rate; λ, c_p, $a = \lambda_{\mathrm{ax}}/\rho c_p$, D^c, α_T and ρ the coefficients of thermal conductivity, heat capacity, thermal diffusivity,

NOMENCLATURE

a	thermal diffusivity
A	amplitude
c_p	heat capacity
c	weight fraction of initial substance
D^c	mixing
E	activation energy
F	laser radiation intensity
I_0	laser beam intensity
k_0	pre-exponential factor
L	reactor length
r_0	laser radius
R	universal gas constant
R_0	reactor radius
t	time
T	mixture temperature
v	mixture feed rate.

Greek symbols

α_T	heat transfer coefficient
$(-\Delta H)$	reactor thermal effect
δ	radiation absorption factor in the medium
λ	thermal conductivity
ρ	density
ω	frequency
ω_0	natural frequency.

Subscripts

ax	axial direction
en	reactor entrance
r	radial direction
in	initial
s	stationary
∞	environment.

mixing, heat transfer, and the density; I_0 and r_0 the constants of the laser beam intensity and radius; δ the coefficient of radiation absorption in a medium; t the time. Subscripts in, en and ∞ refer to the initial state, reactor entrance and the environment; r and ax denote radial and axial direction.

The case is considered when weakly damping oscillations having the frequency of ω_0 are realized in the independent system ($A = 0$).

3. APPROXIMATE ANALYTICAL STUDY BY THE PERTURBATION METHOD

Expand the right-hand sides N_i (θ, c) into the Taylor series in terms of the powers of small deviations $\xi_1 = \theta - \theta_s$ and $\xi_2 = c - c_s$ ($|\xi_1| \ll \theta_s$, $|\xi_2| \ll c_s$) from the equilibrium state determined by the conditions

$$N_1(\theta_s, c_s) = 0, \quad N_2(\theta_s, c_s) = 0 \quad \text{at } A = 0$$

and restrict ourselves to the non-linear terms of the expansion to the third order inclusive. Then, equations (1) will be rewritten in the form

$$\tau > 0: \quad \frac{d\xi_1}{d\tau} = a_{11}\xi_1 + a_{12}\xi_2 + Q\Phi(\xi_1, \xi_2) + AF \sin \omega\tau,$$

$$\frac{d\xi_2}{d\tau} = a_{21}\xi_1 + a_{22}\xi_2 - \Phi(\xi_1, \xi_2),$$

$$\tau = 0: \quad \xi_1 = \theta_{in} - \theta_s, \quad \xi_2 = c_{in} - c_s, \tag{2}$$

where a_{ij} are the coefficients of the linear transformation matrix

$$\Phi(\xi_1, \xi_2) = \Phi_1\xi_1^2 + \Phi_2\xi_1\xi_2 + \Phi_3\xi_1^3 + \Phi_4\xi_1^2\xi_2. \tag{3}$$

3.1. *Solution by the method of slowly varying amplitudes* [11]

Restricting ourselves to the first non-linear term $\Phi(\xi_1, \xi_2) \approx \Phi_1\xi_1^2$, equations (2) will be reduced to the form

$$\frac{d^2\xi_1}{d\tau^2} + \omega_0^2\xi_1 = W\left(\xi_1, \frac{d\xi_1}{d\tau}\right) + K \sin(\omega\tau + \psi) \tag{4}$$

and describe induced damped oscillations.

When $\omega \neq \omega_0$ the solution of equation (4) is determined by the amplitude h_1 and shift of phases ϕ_1 of the resultant oscillations

$$\xi_1 = h_1 \sin(\omega_0\tau + \phi_1) + k \sin(\omega\tau + \psi),$$

$$k = \frac{K}{\omega_0^2 - \omega^2}. \tag{5}$$

Introduction of two unknown functions h_1 and ϕ_1 instead of one ξ_1 in equation (4) requires that additional limitations of the form

$$\frac{dh_1}{d\tau} \sin(\omega_0\tau + \phi_1) + h_1 \frac{d\phi_1}{d\tau} \cos(\omega_0\tau + \phi_1) = 0 \tag{6}$$

should be imposed on them. Equations (4) and (6), after substitution of (5) into them, are solved relative to $dh_1/d\tau$ and $h_1(d\phi_1/d\tau)$. As a result of averaging, when h_1 and ϕ_1 are considered constant, a system of the differential equations is obtained for 'slow' amplitude and phase variations

$$\frac{dh_1}{d\tau} = G_1(h_1, \phi_1), \quad h_1\frac{d\phi_1}{d\tau} = G_2(h_1, \phi_1).$$

To construct an amplitude–frequency response, the stationary states are considered

$$G_1(h_{1s}, \phi_{1s}) = 0, \quad G_2(h_{1s}, \phi_{1s}) = 0.$$

The obtained stationary solutions (h_{1s}, ϕ_{1s}) are

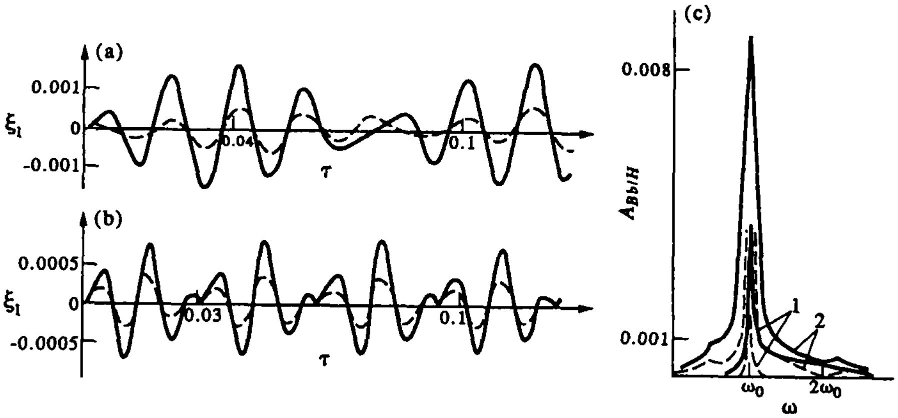

FIG. 1. Time-variation of ξ_1 for $\omega = 0.8\omega_0$ (a) $1.5\omega_0$ (b) and amplitude-frequency response (c) for $A = 0.001$ (1), 0.01 (2).

checked for stability by the Routh–Hurwitz criterion [11] and are used to calculate ξ_1 by equation (5).

The dashed line in Figs. 1(a) and (b) shows the time-variable deviations of ξ for $A = 0.01$. As is seen, the character of resultant oscillations depends on the perturbation frequency. If the frequency of the effect is close to the frequency ω_0 then beats occur (Fig. 1(a)). At $\omega = 1.5\omega_0$ periodic oscillations of a complex shape are realized in the system (Fig. 1(b)). In Fig. 1 the dashed line presents the dependence of the amplitude $A_{\text{ind}} = (\xi_{1\max} - \xi_{1\min})/2$ of the system resultant oscillations on the frequency ω for two amplitudes of the outside effect. In the case of stationary oscillations the maximum amplitude values for the period were plotted. The analytical solution is seen to predict resonance bursts at multiple frequencies. When $\omega \to \omega_0$, the amplitude A_{ind} infinitely increases (this is, however, imposed due to the limited amount of the fed reagent). To specify the value of the amplitude of the induced oscillations at $\omega = \omega_0$ and to reveal possible linear effects with the resonance in the vicinity of the natural frequency, system (2) with allowance for all non-linear terms of functions (3) expansion is studied by the multiscale method.

3.2. *Solution by the multiscale method* [12]

Assume that

$$t_n = \varepsilon^n \tau, \quad n = 0, 1, 2, \ldots,$$

$$\begin{pmatrix}\xi_1\\ \xi_2\end{pmatrix} = \varepsilon\begin{pmatrix}\xi_{11}\\ \xi_{21}\end{pmatrix} + \varepsilon^2\begin{pmatrix}\xi_{12}\\ \xi_{22}\end{pmatrix} + \cdots, \quad \begin{pmatrix}\xi_{1n}\\ \xi_{2n}\end{pmatrix} = \sum_{m=-n}^{n}\begin{pmatrix}\xi_{1n}^{(m)}(t_1, t_2, \ldots)\\ \xi_{2n}^{(m)}(t_1, t_2, \ldots)\end{pmatrix} \exp(im\omega_0 t_0), \quad (7)$$

$\omega = \omega_0 + \varepsilon^2\omega_2 + \cdots$, $F = \varepsilon\gamma_1 + \varepsilon^2\gamma_2 + \cdots$. The coefficients γ_n, $n = 1, 2, \ldots$ are determined from the conditions of the solvability each order in terms of ε. Having substituted expressions (7) into system (2) and equated the coefficients at the same powers of ε, obtain the following differential equations for the first order in terms of a small parameter

$$\frac{\partial \xi_{11}^{(1)}}{\partial t_0} = a_{11}\xi_{11}^{(1)} + a_{12}\xi_{21}^{(1)} - \frac{i}{2}A\gamma_1 \exp(i\omega_2 t_2),$$

$$\frac{\partial \xi_{21}^{(1)}}{\partial t_0} = a_{21}\xi_{11}^{(1)} + a_{22}\xi_{21}^{(1)}. \quad (8)$$

The condition for problem (8) solvability yields

$$\gamma_1 = 0, \quad \begin{pmatrix}\xi_{11}^{(1)}\\ \xi_{21}^{(1)}\end{pmatrix} = \begin{pmatrix}1\\ u_{21}^{(1)}\end{pmatrix} Y, \quad Y = Y(t_1, t_2, \ldots)$$

or, otherwise

$$\begin{pmatrix}\xi_{11}\\ \xi_{21}\end{pmatrix} = \begin{pmatrix}\bar{Y}\,e^{-i\omega_0 t_0} + Y\,e^{i\omega_0 t_0}\\ \bar{u}_{21}^{(1)}\bar{Y}\,e^{-i\omega_0 t_0} + u_{21}^{(1)} Y\,e^{i\omega_0 t_0}\end{pmatrix}.$$

For the second order in terms of ε the solvability condition has the form

$$\gamma_2 = 0, \quad \frac{\partial Y}{\partial t_1} = 0,$$

$$\begin{pmatrix}\xi_{12}\\ \xi_{22}\end{pmatrix} = \begin{pmatrix}\bar{u}_{12}^{(2)}\bar{Y}^2 e^{-2i\omega_0 t_0} + u_{12}^{(0)}|Y|^2 + u_{12}^{(1)} Y^2 e^{2i\omega_0 t_0}\\ \bar{u}_{22}^{(2)}\bar{Y}^2 e^{-2i\omega_0 t_0} + u_{22}^{(0)}|Y|^2 + u_{22}^{(2)} Y^2 e^{2i\omega_0 t_0}\end{pmatrix}.$$

For the third order in terms of ε the solvability condition is reduced to the equation

$$\frac{\partial Y}{\partial t_2} - \kappa_1 Y|Y|^2 + \frac{i}{\kappa_0}A\gamma_3 \exp(i\omega_2 t_2) = 0. \quad (9)$$

Assume $Y = (H/\varepsilon)\exp(i\omega_2 t_2)$, then the differential equation (9) is changed to

$$\frac{dH}{d\tau} + i(\omega - \omega_0)H - \kappa_1 H|H|^2 + i\kappa_2 = 0, \quad \kappa_2 = \frac{AF}{\kappa_0}.$$

The stated problem may be solved by the method of a small parameter, when $Re\ \kappa_1 < 0$, i.e. a mild form of instability is realized in the system, therefore we shall restrict ourselves to this case. With the help of scalar transformations

$$\nu = \omega - \omega_0, \quad H = Z\sqrt{-\frac{|\nu|}{Re\,\kappa_1}}, \quad \tau = \frac{\tau}{|\nu|}$$

$$\frac{dZ}{d\bar{\tau}} + iZ\,\mathrm{sign}\,\nu + (1+i\gamma_1)Z|Z|^2 + i\gamma_2 = 0, \quad (10)$$

where $\gamma_1 = (Im\,\kappa_1/Re\,\kappa_1)$ is the system parameter determined by the coordinates of a stationary state

$$(\theta_s, C_s), \quad \gamma_2 = \kappa_2\sqrt{-\frac{Re\,\kappa_1}{|\nu|^3}}$$

is the variable quantity dependent on the outside effect amplitude and frequency.

As an example, consider the case of $\nu > 0$. Introduce $Z = h_2 \exp(i\phi_2)$, where the new variables h_2 and ϕ_2, respectively, are the amplitude of induced oscillations and the shift of phases. Then from equation (10) a system of equations is obtained

$$\frac{dh_2}{d\bar{\tau}} = -h_2^3 - \gamma_2 \sin\phi_2,$$

$$h_2\frac{d\phi_2}{d\bar{\tau}} = -h_2 - \gamma_1 h_2^3 - \gamma_2\cos\phi_2.$$

Stationary solutions of the obtained system are determined from the equations

$$tg\,\phi_{2s} = \frac{h_{2s}^2}{1+\gamma_1 h_{2s}^2},$$

$$D(h_{2s}) = (1+\gamma_1^2)h_{2s}^6 + 2\gamma_1 h_{2s}^4 + h_{2s}^2 - \gamma_2^2 = 0, \quad (11)$$

which show that the amplitude of the secondary mode h_{2s} is proportional to the cube root of the outside effect amplitude γ_2. From equation (11) and from the condition of the function $D(h_{2s}) : (\partial D/\partial h_{2s}) = 0$ extremum existence the equation is derived for the boundary of the non-uniqueness of the amplitude of induced oscillations on the plane 'system parameter–action parameter' in the form

$$27(1+\gamma_1)^2\gamma_2^4 + 4(9+\gamma_1^2)\gamma_1\gamma_2^2 + 4 = 0.$$

Figure 2(a) presents a wedge-like curve, plotted by this equation, within which (region 2) the non-unique forced oscillation modes are possible, while outside this curve (region 1) the unique modes occur. For example, on the perturbation of the system with the parameter $\gamma_1 = -0.2979$ ($A = 0.001$, curve 1 on Fig. 2(b)) at each frequency the unique mode of induced oscillations is realized. In the case of the effect with the amplitude $A = 0.01$ on the system with the parameter $\gamma_1 = -5$ (curve 2 in Fig. 2(b)) the phenomena is observed that is typical of non-linear systems—the stationary amplitude jump on a slow quasi-stationary passage of the 'hanging' portion of the resonance curve. Within the range of the effect frequency (ν_1, ν_2), depending on the direction of its variation, oscillations that are different in amplitude and shape may be realized. The dashed line indicates virtually non-realizable unstable modes.

With allowance for all the computations obtained the following expressions for the deviation from the stationary values of temperature

$$\xi_1 = 2h_{2s}\sqrt{\left(\frac{|\nu|}{Re\,\kappa_1}\right)}\cos\Omega - h_{2s}^2\frac{|\nu|}{Re\,\kappa_1}$$

$$\times\,(u_{12}^{(0)} + 2Re\,u_{12}^{(2)}\cos 2\Omega + 2Im\,u_{12}^{(2)}\sin 2\Omega)$$

and concentration

$$\xi_2 = 2h_{2s}\sqrt{\left(-\frac{|\nu|}{Re\,\kappa_1}\right)}(Re\,u_{21}^{(1)}\cos\Omega + Im\,u_{21}^{(2)}\sin\Omega)$$

$$-h_{2s}^2\frac{|\nu|}{Re\,\kappa_1}(u_{22}^{(0)} + 2Re\,u_{22}^{(2)}\cos 2\Omega + Im\,u_{22}^{(2)}\sin 2\Omega).$$

where $\Omega = \phi_{2s} + (\omega_0 + \nu)\tau$ is the phase of induced oscillations.

4. NUMERICAL ANALYSIS OF THE OUTSIDE EFFECT ON BURNING

To check the results of the approximate analysis, problem (1) was solved on a computer by the Runge–Kutta method. As an example, in Figs. 1(a) and (b) time-distribution of the deviation ξ_1 of temperature θ from the stationary temperature θ_s for $A = 0.01$ and

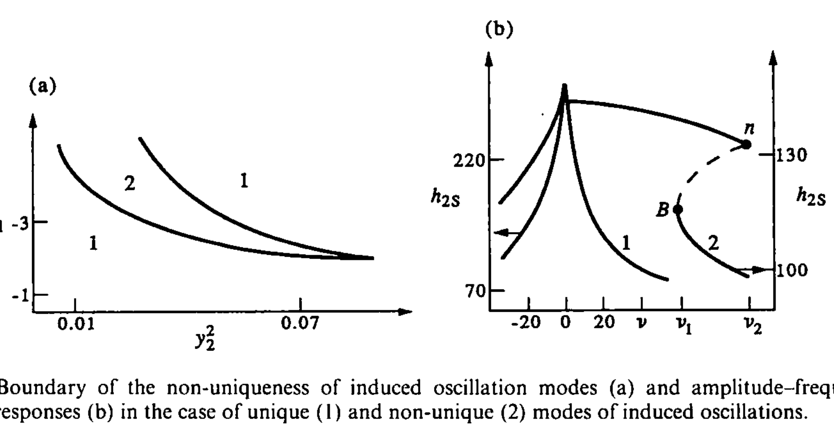

FIG. 2. Boundary of the non-uniqueness of induced oscillation modes (a) and amplitude–frequency responses (b) in the case of unique (1) and non-unique (2) modes of induced oscillations.

different ω (solid lines) is presented. As is seen, the numerical experiment confirms the character of the modes and the period of oscillations predicted analytically (5). In Fig. 1(c) the comparison is given of the amplitude–frequency responses of system (1) obtained by an approximate-analytical method using equation (5) (dashed lines) and by a numerical experiment (solid lines). As is seen, at small amplitudes the resonance on the natural frequency (line 1) takes place, the increase in the outside effect amplitude leads to the origination of the resonance on the frequencies $\omega = 0.5\omega_0$ and $\omega = 2.1\omega_0$ (line 2) that is in agreement with the results of analytical studies. In this case, non-linear effects are observed with the growth of A, namely, the reduction of resonance frequencies and asymmetry of peaks. For the chosen parameter of system (1) and the effect parameters from region 1 of Fig. 2(a) the unique mode of induced oscillations is realized numerically (see curve 1 in Fig. 2(b)). Here, the amplitude in the vicinity of the resonance on the natural frequency, as has been predicted, grows in proportion to the cube root of the outside effect amplitude $A_{\text{ind}} = 0.042\sqrt[3]{A}$.

Thus, the results of the analytical study are in agreement with the solutions of the initial non-linear system.

5. CONCLUSION

In the non-linear approximation the effect of the oscillating laser radiation, selectively absorbed by the buffer additive, on gaseous mixture burning in the ideal-mixing continuous-flow reactor is investigated. Using the method of slowly varying amplitudes, with allowance for the square-law expansion terms in the lumped-parameter model, the dependence of the amplitude of induced oscillations on the frequency and outside effect amplitude is obtained. The resonance frequencies are identified.

Application of the model with allowance for non-linear terms of the expansion up to the third order inclusive enable to conclude that, depending on the parameters of burning and outside effect (amplitude and frequency) both hysteresis and crisis-free amplitude–frequency responses are possible in the vicinity of the resonance on the natural frequency. The regions of non-uniqueness of induced oscillations are discriminated. The amplitude of induced oscillations is shown to increase proportionally to the cube root of the outside amplitude.

The results of analytical investigations are confirmed by the experiment.

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