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# Modelling of unsteady combustion regimes for polydisperse fuels—I. Instability and auto-oscillations

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**Abstract**—A general theory of instability and self-oscillations accompanying combustion of polydisperse assemblages of particles is elaborated on the basis of a model including heat and mass balance equations and the kinetic equation governing the particle size distribution. At arbitrary burning kinetics of particles and the particle influx rate they are reduced, under the condition of ideal mixing, to only two functional integrodifferential equations for the dimensionless temperature and oxidant concentration. Steady combustion processes are shown to be unstable in many important situations. Depending on physical and regime parameters, the break of stability can be realized in either 'soft' or 'hard' manner giving rise to establishing a regular auto-oscillating or a chaotic pulsating regime, respectively. The properties of possible slightly non-linear auto-oscillations are studied in greater detail.

## 1. INTRODUCTION

COMBUSTION of dispersed fuels is usually complicated by a lot of instabilities and oscillatory phenomena being excited by various external agencies (mechanical vibrations, acoustic phenomena, electromagnetic fields, laser radiation, etc.) [1, 2]. Though the mechanisms of energy transfer to disturbances occurring in heterogeneous and homogeneous mixtures may differ essentially (e.g. strengthening of sound waves in a chemically reacting media) [3], the cardinal physical reasons for the development of instabilities and onset of oscillating regimes are the same in both cases.

However, the combustion of dispersed fuels suggests the possibility of some specific type of instability. It owes its origin to a mutual interdependence between the evolution of a burning particulate system and its heat and mass exchange with the environment. The instability of such a type takes place not only in a chemically active media but also on the collapse of metastability in superheated liquids, supersaturated solutions and supercooled melts. Under the conditions of the dominant role of the fluctuating mechanism of nucleation, the above phenomena have been analysed in refs. [4, 5]. In order to clear up the questions in which way and why the mode of reactor operation affects the behaviour of the combustion process under study as well as to find out what should be done to change the regime in a desired direction, an adequate physical model of continuous combustion is badly needed. This study is concerned with the development of instability and self-oscillations in the combustion of a polydisperse mixture.

The principal mechanism of the instability under question and the onset of self-oscillations can be represented as follows. The injection of particles into a combustor first evokes the growth of the furnace temperature which leads to the increase in the chemical reaction rate. This results in a progressive consumption of both the particles and the oxidant which may not be compensated for by their continuing influx. Then, in the course of time a further production rate is inevitable which promotes the appearance of excessive amounts of fuel and oxidant in the furnace. This again accelerates the chemical reaction and the cycle repeats itself. Thus, the system displays both a positive reverse connection (the heat production in chemical reaction) and negative one (the decrease in the chemical reaction rate due to the consumption of reacting particles). It is evident that in the kinetic regime of the combustion of a single particle, with its rate depending greatly on the temperature by the Arrhenius law and slightly on the oxidant concentration, the main reason for the onset of the instability is the positive reverse connection. In the diffusive burning regime the reaction rate is limited primarily by the injection of oxidant into the furnace and depends upon the temperature of the reacting mixture only slightly, according to an approximately linear law. Thus, in this case the break of stability and the onset of self-oscillations are caused mainly by the interaction between the burning of particles and their continuous injection. The amplitude of self-oscillations and the area over which they exist might be expected to be smaller than those in the kinetic combustion regime. In the ensuing sections the specific features of both regimes will be discussed and illustrated with reference to the stability break and self-oscillatory characteristics.

The tendency toward the instability of the type described above has been noted in refs. [6–9]. It is important to point out that publications dealing with relevant topics generally employ the method of

## NOMENCLATURE

| | |
|---|---|
| $c$, $c_1$, $c_2$ | thermal capacity of mixture, gas and disperse phase, respectively |
| $C$ | oxidant concentration |
| $f$ | particle size distribution |
| $g$ | particle influx rate |
| $G$ | oxidant mass flux |
| $k_1$ | coefficient defined by the chemical reaction |
| $k_2$ | stoichiometric coefficient |
| $q$ | squared amplitude of the basic harmonic of temperature disturbance |
| $r$ | particle radius |
| $s$ | dimensionless particle radius |
| $St$ | heat transfer Stanton number |
| $St_m$ | mass transfer Stanton number |
| $t$ | time |
| $t_*$ | dimensionless time |
| $T$ | temperature |
| $u$ | dimensionless temperature |
| $v$ | dimensionless oxidant concentration |
| $W$ | reaction heat. |

Greek symbols

| | |
|---|---|
| $\alpha$ | effective heat transfer coefficient |
| $\rho$, $\rho_1$, $\rho_2$ | density of mixture, gas and dispersed phase, respectively |
| $\omega$ | oscillation frequency. |

Subscript

| | |
|---|---|
| $s$ | steady state regime. |

moments of the particle size distribution function or the method of fractions. In these methods a real polydisperse assemblage of particles is subdivided into a great number of separate fractions corresponding to narrow size ranges. The first method is far from being satisfactory except for the case when the burning rate of a particle does not depend on its size. This requirement radically restricts the class of problems which may be tackled by the method. The method of fractions invariably involves cumbersome and tedious calculations and frequently does not provide the necessary accuracy.

Evolution equations obtained in the present paper for a system of burning particles provide an opportunity to carry out comprehensive analytical and numerical analyses of unsteady combustion regimes without invoking any supplementary hypothesis about both the particle burning kinetics and the particle influx rate. It should be noted that an earlier related work is available [10] which forms some background for the problems under study. The ideas given in that work have been essentially generalized and extended in the present paper.

## 2. PHYSICAL MODEL

Consider combustion of a solid or liquid fuel in the form of a polydisperse assemblage of solid particles or droplets. Physical and chemical parameters are assumed to be homogeneous throughout a combustion chamber under study. In practice the homogeneity is attained by means of intensive mixing. Agglomeration and breakage of burning particles are supposed to be negligible. Heat exchange between the combustion chamber and the surroundings is described phenomenologically with the help of an effective heat transfer coefficient. Let heat and mass balance, which governs the oxidant concentration, in the furnace per unit volume of the mixture be described by the following system of equations:

$$\rho c \frac{\mathrm{d}T}{\mathrm{d}t} = \alpha(T_* - T) + 4\pi k_1 W \int_0^\infty f(t,r) \left|\frac{\mathrm{d}r}{\mathrm{d}t}\right| r^2 \,\mathrm{d}r \tag{1}$$

$$\frac{\mathrm{d}C}{\mathrm{d}t} = G(C) - 4\pi k_1 k_2 \int_0^\infty f(t,r) \left|\frac{\mathrm{d}r}{\mathrm{d}t}\right| r^2 \,\mathrm{d}r \tag{2}$$

which is supplemented with a kinetic equation governing the evolution of the size distribution of the polydisperse assemblage of particles

$$\frac{\partial f}{\partial t} + \frac{\partial}{\partial r}\left(\frac{\mathrm{d}r}{\mathrm{d}t} f\right) = g(r). \tag{3}$$

The dependence of the influx rate of particles on the particle size may be arbitrary in principle. The coefficient $k_1$, being determined by the reaction mechanism, depends on the stoichiometry of the reaction and on the specific surface of the particles. At the cost of an appropriate definition of the parameters $\alpha$ and $T_*$, the term $\alpha(T_* - T)$ can be used to describe not only the heat flux to the external media, but also the fluxes initiated by the withdrawal of the reaction products and by the influx of non-heated particles. Fluctuations of the particle burning rate are assumed to be negligible. Otherwise, the kinetic equation (3) should involve an appropriate diffusive term. The thermal capacity of the mixture can be written as

$$\rho c V = \rho_1 c_1 V_1 + \rho_2 c_2 V_2 \tag{4}$$

where $V$, $V_1$ and $V_2$ are the volume of the combustion chamber, the free gas volume and the volume of the dispersed phase, respectively. If the particles are spherical

$$V_2 = \frac{4}{3}\pi \int_0^\infty f(t,r) r^3 \,\mathrm{d}r. \tag{5}$$

Thus equation (4) is arranged to yield

$$\rho c=\frac{1}{V}\left[\rho_1c_1V+\tfrac{4}{3}\pi(\rho_2c_2-\rho_1c_1)\int_0^\infty f(t,r)r^3\,\mathrm{d}r\right]. \tag{6}$$

The moments of the particle size distribution function are regarded to be finite due to the condition of sufficiently rapid decrease of the $f(t, r)$ in the limit $r\to\infty$. Since just the developed stage of the process, when the influence of initial conditions becomes less significant, is of principal interest, the initial condition can be written as $f(t, r) = 0$ without loss of generality.

The particle burning rate may be represented in a general case in the following form

$$\frac{\mathrm{d}r}{\mathrm{d}t}=-b(T,C)F(r) \tag{7}$$

where $b$ and $F$ are the arbitrary functions of $T$, $C$ and $r$, respectively. The particular expressions of $b$ and $F$ can be found elsewhere [11, 12].

## 3. PARTICLE SIZE DISTRIBUTION. BASIC EVOLUTION EQUATIONS

Introduce the following variables and parameters

$$u=(T-T_0)/T_0,\quad u_*=(T_*-T_0)/T_0,$$

$$Z=fF,\quad v=(C-C_0)/C_0,$$

$$t_*=\frac{1}{Bb_s}\int_0^t b\,\mathrm{d}t,\quad s=\frac{1}{Bb_s}\int_0^r\frac{\mathrm{d}r}{F},$$

$$B=(b_s^4g_0F_0^4)^{-1/5} \tag{8}$$

where $T_0$, $C_0$ and $F_0$ are the characteristic values of the temperature, oxidant concentration and functions $g$ and $F$, respectively; $b_s = b(T_s, C_s)$ and $B$ is the time scale of the process.

The notation used is the same in spite of the change of the arguments, i.e.

$$b(u,v)=b[T(u),C(v)],\quad g(s)=g[r(s)],$$

$$F(s)=F[r(s)],\quad f(t_*,s)=f[t(t_*),r(s)]. \tag{9}$$

In view of equations (8), equations (1)–(3) assume the following form:

$$\rho c\frac{b}{b_s}\frac{\mathrm{d}u}{\mathrm{d}t_s}=\alpha B(u_*-u)$$

$$+4\pi k_1WB^2b_sbT_0^{-1}\int_0^\infty Z(t_*,s)F(s)r^2(s)\,\mathrm{d}s, \tag{10}$$

$$\frac{b}{b_s}\frac{\mathrm{d}v}{\mathrm{d}t_*}=\frac{BG(v)}{C_0}$$

$$-4\pi k_1k_2B^2b_sb\int_0^\infty Z(t_*,c)F(s)r^2(s)\,\mathrm{d}s \tag{11}$$

$$\frac{\partial Z}{\partial t_*}-\frac{\partial Z}{\partial s}=\frac{b_sBgF}{b}. \tag{12}$$

By using the method of refs. [4, 5] and applying the Laplace transform to equation (12), one obtains

$$pZ_p-\frac{\mathrm{d}Z_p}{\mathrm{d}s}=BgFb_s\left(\frac{1}{b}\right)_p,\quad Z_p\div Z,$$

$$\left(\frac{1}{b}\right)_p\div\frac{1}{b}. \tag{13}$$

Applying the inverse Laplace transform to the solution

$$Z_p=Bb_s\left(\frac{1}{b}\right)_p\int_0^\infty g(x)F(x)\,\mathrm{e}^{p(s-x)}\,\mathrm{d}x \tag{14}$$

of equation (13) and changing the order of integration in equation (14) yield the Laplace–Mellin formula

$$Z(t_*,s)=Bb_s\int_0^\infty\frac{g(x)F(x)\,\mathrm{d}x}{b[u(t_*+s-x),v(t_*+s-x)]}. \tag{15}$$

In accordance with equations (8), the solution (15) determines the particle size distribution in the form of the functional depending on the dimensionless temperature and oxidant concentration.

Introducing the new variables and the Stanton parameters

$$y=(u-u_s)/u_s,\quad z=(v-v_s)/v_s,$$

$$St=\alpha(u_s-u_*)/\rho_sc_su_s,$$

$$St_m=v_sBG_s/C_0,\quad St_*=\alpha u_*B/\rho_sc_su_s,$$

$$H=4\pi(\rho_2c_2-\rho_1c_1)B^2b_sX/3V\rho_sc_s,$$

$$X=\int_0^\infty\left(\int_s^\infty g(x)F(x)\,\mathrm{d}x\right)r^3(s)\,\mathrm{d}s \tag{16}$$

and taking into account equations (10) and (11), one obtains a system of functional integrodifferential equations governing the dynamics of the temperature and oxidant concentration in the combustion chamber

$$\left[1+\frac{H}{X}\int_0^\infty\left(\int_s^\infty g(x)F(x)\left(\frac{b_s}{b}-1\right)\mathrm{d}x\right)r^3(s)\,\mathrm{d}s\right]$$

$$\times\frac{b}{b_s}\frac{\mathrm{d}y}{\mathrm{d}t_*}+St+y(St+St_*)-\frac{4\pi k_1WB^2b_s^2b}{\rho_sc_sT_0u_s}$$

$$\times\int_0^\infty\left(\int_s^\infty\frac{g(x)F(x)\,\mathrm{d}x}{b(t_*+s-x)}\right)F(s)r^2(s)\,\mathrm{d}s=0, \tag{17}$$

$$\frac{b}{b_s}\frac{\mathrm{d}z}{\mathrm{d}t_*}-St_mG(v)+4\pi k_1k_2B^2b_s^2b$$

$$\times\int_0^\infty\left(\int_s^\infty\frac{g(x)F(x)\,\mathrm{d}s}{b(t_*+s-x)}\right)F(s)r^2(s)\,\mathrm{d}s=0. \tag{18}$$

For a steady-state combustion regime equations (15), (17) and (18) yield simple correlations

$$f_s = BF^{-1}(s)\int_s^\infty g(x)F(x)\,\mathrm{d}x,$$

$$St = 4\pi k_1 WB^2 b_s^2 Y/T_0\rho_s c_s u_s$$

$$St_m = 4\pi k_1 k_2 b_s^2 B^2 Y,$$

$$Y = \int_0^\infty \left(\int_s^\infty g(x)F(x)\,\mathrm{d}x\right)F(s)r^2(s)\,\mathrm{d}s. \quad (19)$$

It is worth noting that equations (19) make it possible to calculate parameters $St$ and $St_m$ (and, hence, $\alpha$) if experimental data are available on pertinent characteristics of steady combustion regimes.

Since equations (17) and (18) are highly non-linear, it is necessary to analyse their solutions numerically by reversing the functions $t_*(t)$ and $s(r)$ at each step of integration. However, an opportunity will be demonstrated for avoiding rather cumbersome numerical calculations while studying unsteady combustion regimes in the immediate vicinity of the neutral stability surface of steady-state processes by using perturbation methods.

## 4. STEADY-STATE REGIME STABILITY

Next, attention is turned to the stability of the steady-state combustion regimes (19) relative to small fluctuations of temperature and oxidant concentration. Assuming $|y| \ll 1$, and $|z| \ll 1$, one obtains the following system of equations in the linear approximation:

$$\frac{\mathrm{d}y}{\mathrm{d}t_*} + y[St_* + St(1-R_u)] - R_v\,St\,v + \frac{R_u\,St}{Y} \times \int_0^\infty \left(\int_s^\infty g(x)F(x)y(t_*+s-x)\,\mathrm{d}x\right)F(s)r^2(s)\,\mathrm{d}s + \frac{R_v\,St}{Y}\int_0^\infty \left(\int_s^\infty g(x)F(x)z(t_*+s-x)\,\mathrm{d}x\right) \times F(s)r^2(s)\,\mathrm{d}s = 0 \quad (20)$$

$$\frac{\mathrm{d}z}{\mathrm{d}t_*} + z(R_v\,St_m - G_v) + St_m\,R_u y - \frac{R_u\,St_m}{Y}\int_0^\infty \left(\int_s^\infty g(x)F(x)y(t_*+s-x)\,\mathrm{d}x\right) \times F(s)r^2(s)\,\mathrm{d}s - \frac{R_v\,St_m}{Y} \times \int_0^\infty \left(\int_s^\infty g(x)F(x)z(t_*+s-x)\,\mathrm{d}x\right) \times F(s)r^2(s)\,\mathrm{d}s = 0 \quad (21)$$

$$R_u = u_s\frac{\partial \ln b}{\mathrm{d}u}\bigg|_{u=u_s},\quad R_v = v_s\frac{\partial \ln b}{\mathrm{d}v}\bigg|_{v=v_s},\quad G_v = v_s\frac{\partial \ln G}{\mathrm{d}v}\bigg|_{v=v_s} \quad (22)$$

with the parameter $R_u$ representing the dimensionless energy of activation. Expressing $y$ and $z$ in the form $y = y_0\exp(nt_*)$ and $z = z_0\exp(nt_*)$ gives the spectral equation

$$(n+St+St_* - R_u\,St\,I)(n - G_v + R_v\,St_m\,I) + St\,St_m\,R_u\,R_v\,I^2 = 0 \quad (23)$$

where

$$I = 1 - \frac{1}{Y}\int_0^\infty \left(\int_s^\infty g(x)F(x)\,\mathrm{e}^{n(s-x)}\,\mathrm{d}x\right)F(s)r^2(s)\,\mathrm{d}s. \quad (24)$$

Equation (23) becomes applicable to the neutral stability surface when $n = \mathrm{i}\omega^0$, where $\omega^0$ is a rational quantity. The correctness of introducing the above representation of $y$ and $z$ for equations (20) and (21), which do not belong to the objects of the classical stability theory, when studying the stability of the steady combustion processes is corroborated by the possibility of reducing these integrodifferential equations to an autonomous system of ordinary differential equations governing $y, z$ and moments of the particle size distribution of different orders [13]. Provided that $F = 1$ or that a finite number of moments is taken into account at an arbitrary $F$, this system becomes finite. Thus, equation (23) actually determines the neutral stability surface of steady-state combustion regimes in the space of parameters $R_u$, $R_v$, $St$, $St_m$, $St_*$ and $G_v$, depending also on the functional $Y$.

### 4.1. *Kinetic regime of burning*

Now, consider kinetic regimes of burning which approximately satisfy the Arrhenius law $b \sim \exp(-E/RT)$, $F = 1$. Though equation (23) has the rational root $n = 0$, the corresponding instability condition $St + St_* < 0$ cannot be implemented in real processes. However, the oscillating instability with respect to disturbances having some non-zero frequency $\omega^0$ (here and hereafter the zero superscript marks the quantities that refer to the neutral stability surface) is also possible. Traces of the neutral stability surface and the corresponding oscillation period in the plane of parameters $R_u$ and $St$ are shown in Figs. 1 and 2. Calculations have been carried out for functions $g(r)$ of the following types

$$g(r) = g_0 P(r_0 - r) \quad \text{(a)}$$

$$g(r) = g_0(1 - r/r_0)P(r_0 - r) \quad \text{(b)} \qquad (25)$$

where $P$ is the Heaviside function.

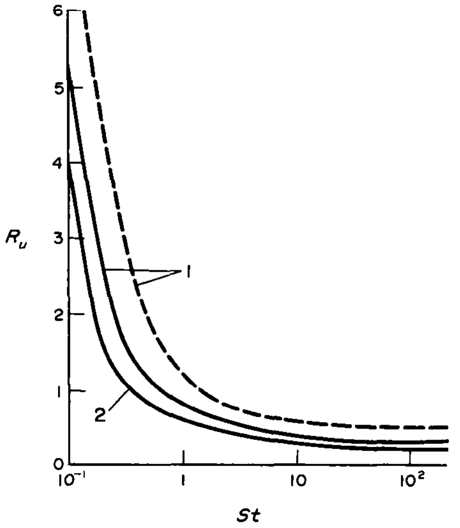

FIG. 1. The curves of neutral stability for the kinetic regime of burning and particle influx rate, equation (25), (a) and (b) correspond to solid and dashed curves; $s_0 = s(r_0) = 10$; 1, $St/St_* = -0.7$; 2, $St/St_* = -0.8$.

The instability region is determined by the inequality $R_u > R_u^0$. When a hyperplane $St = \text{const.}$ (along which the heat production does not change) intersects the neutral stability surface, the instability grows due to the increase of the derivative of $b$ with respect to $u$, that is, due to an increasing temperature dependence of the chemical reaction rate. The corresponding critical value $R_u^0$ of the parameter $R_u$ is defined by the conditions of heat and mass exchange of the polydisperse assemblage of particles with the external media, thermal capacity, fuel influx rate and by its dispersivity. From Figs. 1 and 2 one can easily see that the increase in the parameter $St_m$, that is, the growth of the influence of the oxidant concentration on the process, is a stabilizing factor. This effect has the following explanation. The injection of particles into the furnace first evokes an increase in its temperature and, hence, leads to the growth of the chemical reaction rate and to progressive burning of both the particles and the oxidant. Thus, the decrease in heat production may occur not only as a result of the particle burning being not compensated by their injection, but also due to the decrease in the oxidant concentration exerting an additional stabilizing effect.

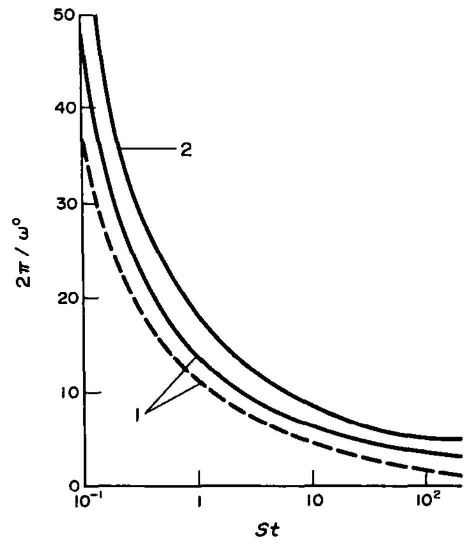

FIG. 2. The oscillation period on the neutral stability curve; nomenclature is the same as in Fig. 1.

### 4.2. *Diffusive regime of burning*

Assume, for the sake of simplicity, that the particle influx rate is a delta-function corresponding to an assumption that monodisperse particles are injected (it should be noted that this does not mean that the fuel in the combustion chamber is also monodisperse). Traces of the neutral stability surface in the plane of the parameters $R_u$ and $St$ for various values of parameters $St_m$ and $R_v$ are presented in Fig. 3. For this case $b = b_0 T^{3/2} C$ and $F = 1/r$. It is clearly seen that the decrease in the parameter $St_m$ (i.e. the increase

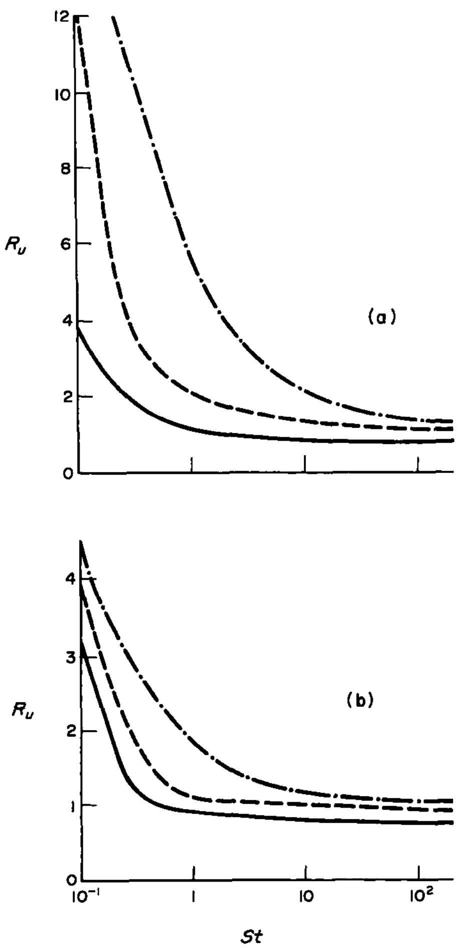

FIG. 3. The curves of neutral stability for the diffusive regime of burning; (a) $R_v = -10$, (b) $R_v = -1$. Solid curve corresponds to $St_m = -0.01$, dashed curve to $St_m = -0.1$, dashed–dotted curve to $St_m = -1$.

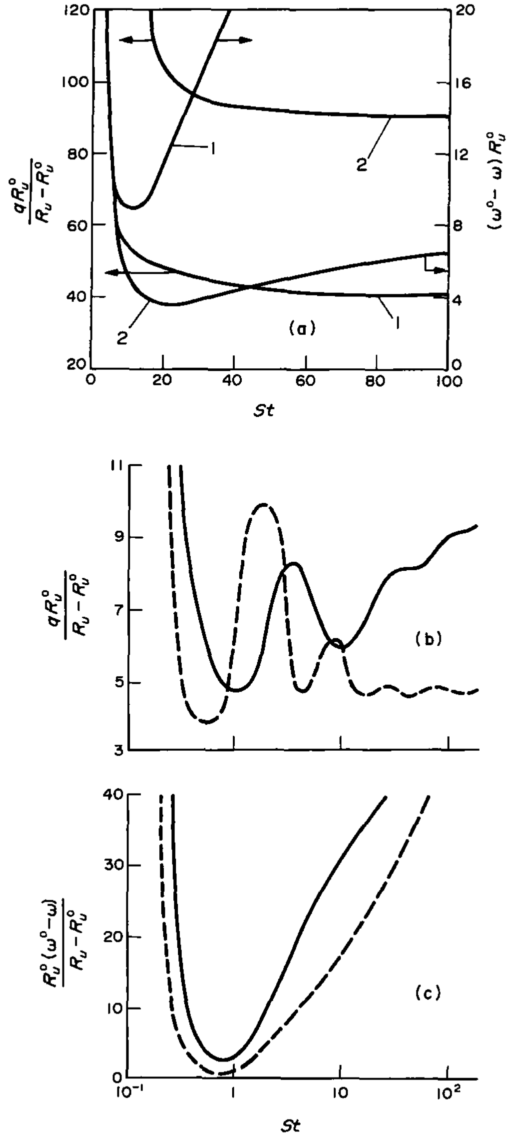

FIG. 6. The squared oscillation amplitude and the frequency shift depending on the Stanton number and the supercriticality. (a) The kinetic regime of burning, $s(r_0) = 10$, $g(r) = g_0P(r_0-r)$ : 1, $St/St_* = -0.7$; 2, $St/St_* = -0.8$. (b), (c) The diffusive regime of burning, $R_v = -0.01$; solid curve, $St_m = -0.01$; dashed curve, $St_m = -0.1$.

responding to the unsteadiness of the thermal capacity of the mixture and to the influence of the oxidant concentration oscillations, leads to a fall in the amplitude and to a rise in the frequency of self-oscillations.

The self-oscillations of the temperature and of the oxidant concentration give rise to oscillations of important processing parameters, such as the total number and mass of particles in the furnace, their mean size and surface, which are governed by the moments of the particle size distribution function of the appropriate order are to be calculated directly by using equation (15). The non-linearity of the system under consideration results in the differences between the time-averaged temperature, oxidant concentration and moments of the particle size distribution function in auto-oscillating regimes and the corresponding quantities peculiar to steady-state ones. This is to be explained by the influence of the non-zero terms $A_0$ and $B_0$ in the expansions for $y$ and $z$, equations (32). As an example, the condition for the decrease in the mean mass of the fuel dispersed throughout the combustion chamber in self-oscillating regimes as compared with that in the corresponding steady-state one of the same heat production assumes the following form:

$$A_0R_u^0 - 4A_1^2(R_u^0)^2 + 2Q_2A_1^2 < 0. \qquad (36)$$

Direct verification of inequality (36) shows that it is valid over a broad range of physical and regime parameters. The mean mass of the unburnt fuel in the furnace decreases proportionally to the supercriticality and reaches several per cent even at the supercriticality of the order of $10^{-2}$–$10^{-1}$, with the heat production rate being regarded as constant. It should be kept in mind that condition (36) has been derived for a slight supercriticality. Thus, the latter conclusion may appear to be wrong if applied outside the immediate vicinity of the stability boundary.

## 6. CONCLUDING REMARKS

The above reasoning and calculation evidence that the origination of auto-oscillations due to the inherent inner instability of steady combustion regimes is able to change important operational characteristics of furnaces to a considerable extent without any variations in constant external conditions. In these circumstances, one may expect that the same will be even truer when these conditions are being varied in accordance with some prescribed periodic law. This can bring about, first, the change in the form of the stability region and also in the properties of the resulting auto-oscillating regimes and, second, the establishment of forced periodic or quasi-periodic regimes specific for non-linear oscillating systems. Both expectations happen to be true. External periodic influences can stabilize combustion systems and eliminate random pulsations being undesirable from technological considerations. They are also able to excite forced oscillations. Within new regions of instability of artificially excited oscillations the parametric modulation causes phenomena of frequency locking and the occurrence of quasi-periodic oscillating processes. This gives an opportunity for causing a rather drastic influence on the performance of combustion devices in practice in order to shift their operational characteristics in a desired direction. This is a main topic of the second part of this paper.

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