

# Controlling of mass concentration of reagents by sound in a gas with non-equilibrium chemical reactions

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## Summary

Weakly nonlinear dynamics of chemically reacting gas is under study. Nonlinear interaction of acoustic and non-acoustic types of motion are considered. Decomposing of equations bases on links of gasdynamic perturbations specific for every type of motion. The governing equation for mass fraction of reagent influenced by dominative sound, is derived and discussed. Conclusions concern equilibrium and non-equilibrium regimes of chemical reactions.

## 1 Introduction. Basic equations and starting points

The hydrodynamics of the non-equilibrium fluids is one of the new fields of modern hydrodynamics. The studies in this field began in sixties of the 20th century. They initially started in connection with the advances in laser engineering and plasma aerodynamics. Being a new field of hydrodynamics, involving advances in thermodynamics and chemistry, the theory of non-equilibrium fluids is passing through the stage of formulating the fundamental equations and for revealing new physical effects.

Attention to the non-equilibrium phenomena in hydrodynamics was firstly originated by experimental studies of anomalous dispersion and absorption of ultrasonics waves in a gas having non-equilibrium internal degrees of freedom. A reason for these anomalies is the mechanism of retarded energy exchange between the internal and translational degrees of freedom of the molecules [1,2]. The dispersion of sound in the chemically reacting gases is well-understood [2,3]. Like it takes place in gases with excited internal degrees of freedom, the equilibrium velocity of sound in a medium with reversible chemical reaction is always smaller than the "frozen" one. The situation becomes opposite during non-equilibrium chemical reactions. Besides anomalous dispersion, there observed anomalous amplification of sound, and such fluid becomes acoustically active. The similar inverse behavior was observed in media with several relaxation processes and in nonisothermic plasma.

Only recently, acoustics of gases where non-equilibrium processes take place, in particular a chemical reactions occur, became the subject of serious theoretical studies [2,4-6]. The adequate dispersion relation was firstly derived, and the basic peculiarities of sound propagation were drawn out as regard to variations in heat produced during chemical reaction [6].

The important problem to be solved is interaction of acoustic and non-acoustic motions in the chemically reacting fluids, and, in particular, controlling of mass fractions of the reagents by sound. The theoretical study is complex in view of nonlinearity of governing equations describing mass, energy and momentum balance. This study is devoted to the deriving and brief discussion of dynamic equation for the reagent mass fraction affected by dominative sound. Weakly nonlinear dynamics is considered. The nonlinear governing equation for mass fraction is obtained by means of decomposing from the general system using the links of gasdynamic perturbations specific for every type of motion, or mode. This method was worked out and applied by the author in studies of some problems of nonlinear hydrodynamics concerning acoustic heating and streaming [7,8]. We start from the linear determination of modes as specific types of gas motion in a gas where a chemical reaction takes place (Sec.2), going to the correct decomposition of equations governing acoustic and non-acoustic motions accounting for the interactions of modes (Sec.3).

The present study considers the simplest model of a gas where equilibrium or non-equilibrium chemical reaction takes place of the  $A \rightarrow B$  type. This model applies also to reacting media with complex, branching reactions [9]. There are three conservation equations, continuity, momentum and energy ones:

$$\begin{aligned} \rho \frac{d\vec{v}}{dt} &= -\vec{\nabla} P, \\ \frac{C_{V,\infty}}{R} \frac{dT}{dt} - \frac{T}{\rho} \frac{d\rho}{dt} &= Q, \\ \frac{d\rho}{dt} + \rho \vec{\nabla} \vec{v} &= 0. \end{aligned} \tag{1}$$

In the equations above,  $\vec{v}$  denotes particles velocity of a gas,  $\rho, P$  denote its density and pressure,  $T$  is temperature of a gas measured in Joules per molecule (actually the ordinary temperature multiplied by the Boltzmann constant  $\mathbf{k}_B$ ),  $C_{V,\infty}, C_{P,\infty}$  are the "frozen" molar heat capacities at constant volume and constant pressure, respectively (correspondent processes takes place at infinitely high frequencies),  $R = C_{P,\infty} - C_{V,\infty}$  is the universal gas constant,  $Q = HmW/\rho$  is the heat produced in the medium per one molecule due to a chemical reaction ( $W$  is the volume rate of formation of the reaction product  $B$ ),  $H$  denotes the reaction enthalpy per unit mass of reagent  $A$ , and  $m$  denotes the averaged molecular mass. Dynamic equation of the mass fraction  $Y$  of reagent  $A$ , and the equation of state complete the system (1):

$$\frac{dY}{dt} = -\frac{W}{\rho}, \quad P = \frac{\rho T}{m}. \tag{2}$$

## 2 Motions of infinitely small amplitude in one dimension and their decomposing

Studies of planar motion of infinitely small amplitude along axis  $OX$  use to begin with representing of every quantity  $\varepsilon$  as a sum of unperturbed value  $\varepsilon_0$  and its variation  $\varepsilon'$ , ( $|\varepsilon'| \ll |\varepsilon_0|$ ).

Following [6], we assume that the ambient stationary quantities  $Y_0$ ,  $T_0$ ,  $P_0$ ,  $\rho_0$  are maintained by a transverse pumping, so that in the longitudinal direction pointed by axis  $OX$ , the ambient medium is homogeneous. The equations of momentum, energy, mass fraction balance and continuity read:

$$\begin{aligned} \frac{\partial v'}{\partial t} + \frac{1}{\rho_0} \frac{\partial P'}{\partial x} &\equiv \frac{\partial v'}{\partial t} + \frac{T_0}{m\rho_0} \frac{\partial \rho'}{\partial x} + \frac{1}{m} \frac{\partial T'}{\partial x} = 0, \\ \frac{\partial T'}{\partial t} + (\gamma_\infty - 1) \left( T_0 \frac{\partial v'}{\partial x} - Q_T \frac{Q_0}{T_0} T' - Q_\rho \frac{Q_0}{\rho_0} \rho' - Q_Y \frac{Q_0}{Y_0} Y' \right) &= 0, \\ \frac{\partial Y'}{\partial t} + \frac{1}{Hm} \left( Q_T \frac{Q_0}{T_0} T' + Q_\rho \frac{Q_0}{\rho_0} \rho' + Q_Y \frac{Q_0}{Y_0} Y' \right) &= 0, \\ \frac{\partial \rho'}{\partial t} + \rho_0 \frac{\partial v'}{\partial x} &= 0, \end{aligned} \quad (3)$$

where  $\gamma_\infty = C_{P,\infty}/C_{V,\infty}$  denotes the frozen adiabatic exponent. The dimensionless quantities  $Q_T$ ,  $Q_\rho$ ,  $Q_Y$  are as follows:

$$Q_T = \left( \frac{\partial \ln Q}{\partial \ln T} \right)_{\rho_0, T_0, Y_0} = \frac{T_0}{Q_0} \left( \frac{\partial Q}{\partial T} \right)_{\rho_0, T_0, Y_0}, \quad Q_\rho = \left( \frac{\partial \ln Q}{\partial \ln \rho} \right)_{\rho_0, T_0, Y_0}, \quad Q_Y = \left( \frac{\partial \ln Q}{\partial \ln Y} \right)_{\rho_0, T_0, Y_0}. \quad (4)$$

An excess pressure in the first equation from (3) is expressed in terms of excess density and temperature in accordance with equation of state (the second one from (2)).

Studies of dispersion relations determining possible types of motion, start with representing of all perturbations as planar waves:

$$\varepsilon'(x, t) = \tilde{\varepsilon} \exp i(\omega t - kx). \quad (5)$$

Some intermediate steps are necessary to establish roots of dispersion relation,  $\omega(k)$ . From the third equation from the system (3) the equalities below follow:

$$\left( \frac{\partial Y}{\partial T} \right)_V = -\frac{Q_T}{Q_Y(1 + i\omega\tau_c)} \frac{Y_0}{T_0}, \quad \left( \frac{\partial Y}{\partial \rho} \right)_T = -\frac{1}{\rho_0^2} \left( \frac{\partial Y}{\partial V} \right)_T = -\frac{Q_\rho}{Q_Y(1 + i\omega\tau_c)} \frac{Y_0}{\rho_0}, \quad (6)$$

where  $V = 1/\rho$  is the specific gas volume,

$$\tau_c = \frac{HmY_0}{Q_0Q_Y} \quad (7)$$

is the characteristic time of chemical reaction. The equation of state (the second from Eq.(2)) along with the thermodynamic equality

$$\left( \frac{\partial Y}{\partial T} \right)_P = \left( \frac{\partial Y}{\partial T} \right)_V + \left( \frac{\partial Y}{\partial V} \right)_T \left( \frac{\partial V}{\partial T} \right)_P, \quad (8)$$

and Eq.(6), result in expression

$$\left( \frac{\partial Y}{\partial T} \right)_P = \frac{(Q_\rho - Q_T) Y_0}{Q_Y(1 + i\omega\tau_c) T_0}. \quad (9)$$

Heat capacities under constant volume  $C_V$  and pressure  $C_P$  depend on frequency  $\omega$ :

$$C_P = C_{P,\infty} + mHR \left( \frac{\partial Y}{\partial T} \right)_P = C_{P,\infty} + \frac{mHR(Q_\rho - Q_T) Y_0}{Q_Y(1 + i\omega\tau_c) T_0}, \quad (10)$$

$$C_V = C_{V,\infty} + mHR \left( \frac{\partial Y}{\partial T} \right)_V = C_{V,\infty} - \frac{mHRQ_T Y_0}{Q_Y(1 + i\omega\tau_c) T_0}.$$

Finally, the dispersion relations for three non-entropy modes and the entropy (or thermal) one read [6]:

$$\omega^2 = k^2 \frac{T_0 C_P}{m C_V} = k^2 \frac{T_0}{m} \left( \frac{C_{P,\infty} + \frac{(Q_\rho - Q_T)\tau_c Q_0 R}{1 + i\omega\tau_c}}{C_{V,\infty} - \frac{Q_T\tau_c Q_0 R}{1 + i\omega\tau_c}} \right), \quad \omega = 0. \quad (11)$$

Actually, the first equation from (11) determines, besides acoustic branches, one non-wave root responsible for rate of increase in mass fraction of reagent. The quantities below indexed by 1 relate to the sound progressive in the positive direction of axis  $OX$ , those indexed by 2 and 3 - to the leftwards progressive sound, and the remaining non-zero root, correspondingly. The fourth root corresponding to the entropy, or thermal, mode equals zero.

The explicit formulae linking  $\omega$  and  $k$  for non-entropy modes (Eq. (11)) are fairly complex. As for both branches of sound, the dispersion relations depend on a ratio of sound period and characteristic time of chemical reaction  $\tau_c$ . It is reasonable to consider acoustic frequency large compared to the reverse duration of a chemical reaction:  $|\omega_{1,2}\tau_c| \approx |ku_\infty\tau_c| \gg 1$ . Under this condition, approximate acoustic roots of dispersion relation are following:

$$\omega_1 = u_\infty k - i \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{2\gamma_\infty T_0}, \quad (12)$$

$$\omega_2 = -u_\infty k - i \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{2\gamma_\infty T_0},$$

where  $\gamma_\infty = \frac{C_{P,\infty}}{C_{V,\infty}}$  denotes the frozen adiabatic exponent,  $u_\infty = \sqrt{\gamma_\infty \frac{T_0}{m}}$  is the frozen sound velocity. The approximate roots of dispersion relations for both acoustic branches are derived in [6]. Under assumption of slow variations of heat release with temperature and density,  $|Q_T| \ll 1$ ,  $|Q_\rho| \ll 1$ , the third non-wave root gets a form:

$$\omega_3 = i \left( \frac{Q_0(Q_\rho - Q_T)(\gamma_\infty - 1)}{\gamma_\infty T_0} + \frac{1}{\tau_c} \right). \quad (13)$$

Basing on approximate roots of dispersion relation, it is easy to establish links connecting perturbations of density, temperature, mass fraction and velocity, specific for every type of motion. For the progressive in the positive direction of axis  $OX$  sound, the links are following:

$$v'_1 = \left( \frac{u_\infty}{\rho_0} - \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{2\gamma_\infty \rho_0 T_0} \int dx \right) \rho'_1,$$

$$T'_1 = \left( \frac{T_0(\gamma_\infty - 1)}{\rho_0} - \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{u_\infty \rho_0} \int dx \right) \rho'_1, \quad (14)$$

$$Y'_1 = \left( \frac{Q_0(Q_\rho + (\gamma_\infty - 1)Q_T)}{Hmu_\infty \rho_0} \int dx \right) \rho'_1,$$

for the progressive in the negative direction of axis  $OX$  sound they are

$$\begin{aligned} v'_2 &= \left( -\frac{u_\infty}{\rho_0} - \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{2\gamma_\infty\rho_0T_0} \int dx \right) \rho'_2, \\ T'_2 &= \left( \frac{T_0(\gamma_\infty - 1)}{\rho_0} + \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{u_\infty\rho_0} \int dx \right) \rho'_2, \\ Y'_2 &= - \left( \frac{Q_0(Q_\rho + (\gamma_\infty - 1)Q_T)}{Hmu_\infty\rho_0} \int dx \right) \rho'_2. \end{aligned} \quad (15)$$

The third type of motion, originating from chemical reaction in a gas, possesses links:

$$\begin{aligned} v'_3 &= \left( \frac{1}{\rho_0\tau_c} \int dx + \frac{Q_0(\gamma_\infty - 1)(Q_\rho - Q_T)}{\gamma_\infty T_0\rho_0} \int dx \right) \rho'_3, \\ T'_3 &= - \left( \frac{T_0}{\rho_0} \right) \rho'_3, \quad Y'_3 = \left( \frac{u_\infty^2}{H\rho_0(\gamma_\infty - 1)} \right) \rho'_3. \end{aligned} \quad (16)$$

The last series represent the entropy mode:

$$v'_4 = 0, \quad T'_4 = - \left( \frac{T_0}{\rho_0} \right) \rho'_4, \quad Y'_4 = \left( \frac{Q_0\tau_c(Q_\rho - Q_T)}{Hm\rho_0} \right) \rho'_4. \quad (17)$$

These links do not depend on time and keep constant at every moment of gas evolution. They are in general integral, reflecting the dispersive properties of chemically reacting fluid. A perturbation of every dynamic variable may be expressed in terms of one from specific excess densities,  $\rho'_1$ ,  $\rho'_2$ ,  $\rho'_3$  or  $\rho'_4$ . The overall velocity, pressure, density and mass fraction of reagent, equal a sum of specific parts:  $v'(x, t) = v'_1(x, t) + v'_2(x, t) + v'_3(x, t) + v'_4(x, t)$ , and so on.

In the flow of infinitely small amplitudes governed by Eqs (3), the dynamic equations for the excess densities of every mode are independent and decompose completely from the system. Governing equations for all four excess densities take forms:

$$\begin{aligned} \frac{\partial \rho'_1}{\partial t} + u_\infty \frac{\partial \rho'_1}{\partial x} - \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{2u_\infty^2 m} \rho'_1 &= 0, \\ \frac{\partial \rho'_2}{\partial t} - u_\infty \frac{\partial \rho'_2}{\partial x} - \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{2u_\infty^2 m} \rho'_2 &= 0, \\ \frac{\partial \rho'_3}{\partial t} + \left( \frac{Q_0(Q_\rho - Q_T)(\gamma_\infty - 1)}{\gamma_\infty T_0} + \frac{1}{\tau_c} \right) \rho'_3 &= 0, \\ \frac{\partial \rho'_4}{\partial t} &= 0. \end{aligned} \quad (18)$$

That obviously coincides to the roots of dispersion relations  $\omega_1, \dots, \omega_4$  established by Eqs (11)-(13). Amplitudes of excess acoustic quantities increase when

$$Q_0(Q_\rho + (\gamma_\infty - 1)Q_T) > 0, \quad (19)$$

and decrease otherwise. Inequality (19) determines an area of irreversibility of a chemical reaction. It is easy to verify, that it establishes also an inequality as follows:

$$u_\infty^2 - u_0^2 = \frac{T_0}{m} \left( \frac{C_{P,\infty}}{C_{V,\infty}} - \frac{C_{P,0}}{C_{V,0}} \right) = \frac{(\gamma_\infty - 1)Q_0(Q_\rho + (\gamma_\infty - 1)Q_T)T_0\tau_c}{m(Q_0Q_T\tau_c(\gamma_\infty - 1) - T_0)} < 0, \quad (20)$$

if  $C_{V,0} = C_{V,\infty}(1 - Q_0 Q_T (\gamma_\infty - 1) \tau_c / T_0) > 0$  ( $C_{P,0}$  and  $C_{V,0}$  denote quantities correspondent to infinitely slow processes following from Eqs (10) when  $\omega = 0$ ). The condition of a chemically reacting gas be acoustically active in the non-equilibrium regime, inequality (19), was firstly obtained by Molevich [6]. An adequate analysis of dispersion characteristics of sound in the equilibrium and non-equilibrium regimes was given in the paper [6] as well.

### 3 Controlling of chemical reaction by sound in a weakly nonlinear flow

#### 3.1 Weakly nonlinear dynamics of reacting gas and decomposing of governing equations

Since the quadratic nonlinear terms are of importance while studying of weakly nonlinear flows, only these terms will be hold in the system of governing equations. The governing system (1) with account for (2) within accuracy of quadratic nonlinear terms rearranges in one dimension to the following system:

$$\begin{aligned} \frac{\partial v'}{\partial t} + \frac{T_0}{m\rho_0} \frac{\partial \rho'}{\partial x} + \frac{1}{m} \frac{\partial T'}{\partial x} &= -v' \frac{\partial v'}{\partial x} + \frac{T_0 \rho'}{m\rho_0^2} \frac{\partial \rho'}{\partial x} - \frac{T'}{m\rho_0} \frac{\partial \rho'}{\partial x}, \\ \frac{\partial T'}{\partial t} + (\gamma_\infty - 1) \left( T_0 \frac{\partial v'}{\partial x} - Q_T \frac{Q_0}{T_0} T' - Q_\rho \frac{Q_0}{\rho_0} \rho' - Q_Y \frac{Q_0}{Y_0} Y' \right) &= -v' \frac{\partial T'}{\partial x} - (\gamma_\infty - 1) T' \frac{\partial v'}{\partial x}, \\ \frac{\partial Y'}{\partial t} + \frac{1}{Hm} \left( Q_T \frac{Q_0}{T_0} T' + Q_\rho \frac{Q_0}{\rho_0} \rho' + Q_Y \frac{Q_0}{Y_0} Y' \right) &= -v' \frac{\partial Y'}{\partial x}, \\ \frac{\partial \rho'}{\partial t} + \rho_0 \frac{\partial v'}{\partial x} &= -v' \frac{\partial \rho'}{\partial x} - \rho' \frac{\partial v'}{\partial x}. \end{aligned} \quad (21)$$

Actually, the nonlinear right-hand parts of Eqs (21) include also terms referring to the second-order derivatives of  $Q$ :  $\partial^2 Q / \partial T^2$  and so on, which are not taken into account by the present study, confining the changes in  $Q(\rho, T, Y)$  by differential and hence restricting the accuracy of conclusions.

The mathematical content of the method which has been worked out by the author, was applied previously to some problems of nonlinear hydrodynamics [7,8]. It points out a way to decompose weakly nonlinear equations basing on properties of linear modes. The main idea is to keep linear links of modes unchanged (Eqs (14)-(17)) in investigations of weakly nonlinear dynamics. It is easy to verify, that acting at the first equation from (21) by operator  $-\frac{Q_0(Q_\rho + (\gamma_\infty - 1)Q_T)}{Hmu_\infty^2} \int dx$ , multiplying the second, third and fourth ones by  $-\frac{Q_0(Q_\rho - Q_T)\tau_c}{Hm^2u_\infty^2}$ ,  $1 - \frac{Q_0(\gamma_\infty - 1)(Q_\rho - Q_T)\tau_c}{mu_\infty^2}$ ,  $\frac{Q_0(\gamma_\infty - 1)(Q_\rho - Q_T)\tau_c}{Hm\rho_0\gamma_\infty}$ , correspondingly, and taking a sum of final equations, reduces all terms including perturbations of first, second and fourth modes in the linear part yielding in the governing equation for  $Y'_3$ . Nonlinear right-hand part of the final equation includes the mixed quadratic terms of all types of motion responsible for the modes interaction. Let the progressive in the positive direction sound be dominative. That means,

that characteristic amplitude of its excess density in the considered domain is much greater than that of other modes:

$$\rho_{A,1} \gg \rho_{A,n}, \quad n = 2, 3, 4 \quad (22)$$

So that, only quadratic acoustic terms in the nonlinear part may be considered. They may be expressed in terms of  $\rho'_1$  using links (14).

The final dynamic equation takes a form:

$$\begin{aligned} \frac{\partial Y'_3}{\partial t} + \left( \frac{Q_0(Q_\rho - Q_T)(\gamma_\infty - 1)}{\gamma_\infty T_0} + \frac{1}{\tau_c} \right) Y'_3 = \\ - \frac{Q_0(Q_\rho + (\gamma_\infty - 1)Q_T)}{Hm\gamma_\infty \rho_0^2} \rho_1'^2 + \frac{(\gamma_\infty - 1)(\gamma_\infty - 2)}{\gamma_\infty} \frac{Q_0(Q_\rho - Q_T)\tau_c u_\infty}{Hm\rho_0^2} \rho_1' \frac{\partial \rho_1'}{\partial x}. \end{aligned} \quad (23)$$

The governing equation for the sound itself also becomes modified in a weakly nonlinear flow. To decompose it, reducing all terms of the second, third and fourth modes, one has to act at the first equation from (21) by  $\frac{\rho_0}{2u_\infty} + \frac{Q_0\rho_0(\gamma_\infty - 1)(Q_\rho + Q_T(\gamma_\infty - 1))}{2mu_\infty^4} \int dx$ , at the second one by  $\frac{\rho_0}{2mu_\infty^2} + \frac{Q_0\rho_0(\gamma_\infty - 1)(3Q_\rho + Q_T(\gamma_\infty - 3))}{4m^2u_\infty^5} \int dx$ , and at the third and fourth ones by  $-\frac{H\rho_0(\gamma_\infty - 1)}{2u_\infty^3\tau_c} \int dx$  and  $\frac{1}{2\gamma_\infty} - \frac{Q_0(\gamma_\infty - 1)(-3Q_\rho + 3Q_T + (2Q_\rho - 3Q_T)\gamma_\infty)}{4\gamma_\infty mu_\infty^3} \int dx$ , correspondingly. Calculating a sum of resulting expressions, leads to the final governing equation:

$$\frac{\partial \rho_1'}{\partial t} + u_\infty \frac{\partial \rho_1'}{\partial x} - \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{2u_\infty^2 m} \rho_1' = - \frac{(\gamma_\infty + 1)u_\infty}{2\rho_0} \rho_1' \frac{\partial \rho_1'}{\partial x}. \quad (24)$$

The sound is dominative, so that in the nonlinear part of Eq. (24) only acoustic terms are hold.

### 3.2 Variations in mass concentration of reagent caused by sound

Equations (23) and (24) govern dynamics of dominative sound excess density and mass concentration of reagent, its part belonging to the third non-wave mode. In accordance to procedure of deriving, they are valid when sound progressive in the positive direction of axis  $OX$  keeps dominative in comparison with other modes, hence, Eqs (22) are valid. That determines the temporal and spacial domains of formulae validity, because nonlinear distortions accumulate with time and may make other modes to enhance due to nonlinear transfer of mass, energy or momentum.

The further analysis needs foundation of an acoustic excess density in time and space accordingly to Eq.(24). An excess acoustic density and other acoustic quantities (Eqs (14)) may be periodic or aperiodic. The method or linear combining of equations bases only on links of specific perturbations valid for any type of sound and does not need any temporal averaging.

For the preliminary estimations, periodic in time solution of the linear equation (the first one from Eqs (18)) will be considered:

$$\rho_1'(x, t) = \rho_{A,1} \sin(\omega_1(t - x/u_\infty)) \exp(\alpha x), \quad \rho_1'(0, t) = \rho_{A,1} \sin(\omega_1 t), \quad (25)$$

where  $\alpha = \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{2u_\infty^3 m}$ .





Substituting Eq.(25) in the right-hand part of Eq.(23), averaging the result over sound period along with replacing of  $\rho_{A,1}/\rho_0$  by acoustic Mach number  $M$ , yield in the following equation:

$$\frac{\partial Y_3'}{\partial t} + \left( \frac{Q_0(Q_\rho - Q_T)(\gamma_\infty - 1)}{\gamma_\infty T_0} + \frac{1}{\tau_c} \right) Y_3' \approx \left\langle \frac{\partial Y_3'}{\partial t} + \left( \frac{Q_0(Q_\rho - Q_T)(\gamma_\infty - 1)}{\gamma_\infty T_0} + \frac{1}{\tau_c} \right) Y_3' \right\rangle = -\frac{\alpha u_\infty^3}{H\gamma_\infty(\gamma_\infty - 1)} M^2 \exp(2\alpha x). \quad (26)$$

Eq. (26) is written on within the leading accuracy. Square brackets denote averaging over the sound period. Taking in mind, that characteristic period of sound is much less than characteristic time of chemical reaction  $\tau_c$ , the averaging over sound period would give only small correction to  $Y_3'$ , and may be omitted in the left-right side.

A solution satisfying initial condition  $Y_3'(t = 0) = 0$ , with  $\beta$  replacing  $\frac{Q_0(Q_\rho - Q_T)(\gamma_\infty - 1)}{\gamma_\infty T_0} + \frac{1}{\tau_c}$ , looks:

$$Y_3'(x, t) = -\frac{\alpha u_\infty^3}{H\beta\gamma_\infty(\gamma_\infty - 1)} M^2 \exp(2\alpha x) (1 - \exp(-\beta t)). \quad (27)$$

An equilibrium chemical reaction presupposes  $\alpha < 0$ . At any concrete time  $t$ , an excess mass fraction of reagent decreases towards zero with distance from the transducer keeping positive during equilibrium regime. The temporal behavior of  $|Y_3'|$  depends on sign of  $\beta$  : if

$$\frac{1}{\tau_c} + \frac{Q_0(Q_\rho - Q_T)(\gamma_\infty - 1)}{\gamma_\infty T_0} > 0, \quad (28)$$

it does not grow with time, otherwise, it does. Note, that heat capacity at zero frequency following from Eqs (10), is:

$$C_{P,0} = C_{P,\infty} + \frac{Q_0(Q_\rho - Q_T)(C_{P,\infty} - C_{V,\infty})\tau_c}{T_0}. \quad (29)$$

Both inequality (28) and the temporal decrease of excess mass fraction of reagent absolute value, follow from positiveness of  $C_{P,0}$ . The negative values of  $\beta$ , though possible in strongly non-equilibrium reactions, contradict to the frames of the present investigation: they mean considering enough large values  $|Q_T|$ ,  $|Q_\rho|$  which do not belong to the limitations accepted. Moreover, it is necessary to take into account inhomogeneity of background temperature and other quantities in strongly non-equilibrium regime, making the mathematic content of the theory fairly complex.

In the non-equilibrium acoustically regime,  $\alpha > 0$ , and excess mass fraction of reagent decreases with distance from the transducer keeping negative in accordance to Eq. (27). This equation presupposes infinitely large absolute values of  $Y_3'$ , but its validity is confined by the regard on dominative sound:  $|\rho_3'| = \left| \frac{H\rho_0(\gamma_\infty - 1)}{u_\infty^2} Y_3' \right| \ll |\rho_1'|$ , which rearranges in the following inequality:

$$\left| \frac{\alpha u_\infty}{\beta\gamma_\infty} M \exp(2\alpha x) (1 - \exp(-\beta t)) \right| \ll 1. \quad (30)$$

The next condition concerns weak nonlinearity of a flow:  $|Y_3'| \ll Y_0$ :

$$\left| \frac{\alpha u_\infty^3}{H\beta\gamma_\infty(\gamma_\infty - 1)} M^2 \exp(2\alpha x) (1 - \exp(-\beta t)) \right| \ll Y_0. \quad (31)$$



Eqs (30), (31) determine the domain of distances from transducer at any concrete time  $t$ , where the formulae above are valid.

## 4 Concluding remarks

The main result of study is Eq.(23) which describes dynamics of this part of mass fraction of reagent  $A$  which is not a wave quantity,  $Y_3'$ . An excess mass fraction of reagent, specific for sound,  $Y_1'$ , varies in time and space accordingly to Eq.(24) and links from Eqs (14) during sound propagation (analogously, a quantity coming from other sound branch,  $Y_2'$  does). As for the part of mass fraction, correspondent to the entropy motion, it is a small quantity of order  $Max(Q_T^2, Q_\rho^2)M^2$ , if caused by dominative sound, and therefore exceeds permissible accuracy.

Being slowly varying quantity,  $Y_3'$  changes the background mass concentration  $Y_{0,new} = Y_0 + Y_3'$ . The analogy of this phenomenon with acoustic heating is obvious. The acoustic heating in fluids with standard attenuation is known as a slow process of isobaric increase in temperature due to nonlinear loss in acoustic energy [10,11]. It itself influences sound by means of forming a new background of sound propagation  $(\rho_0, T_0)$ . In contrast, the new background mass fraction  $Y_{0,new}$  does not influence propagation of high-frequency sound (Eq.(24)) but influences low-frequency sound ( $\omega_{1,2}\tau_c \ll 1$ ) which is out of frames of the present study. The dispersion relations for low-frequency acoustic branches are following:

$$\begin{aligned}\omega_1 &= u_0 k - i \frac{H^2 k^2 m R Y_0^2 (Q_T C_{P,0} + (Q_\rho - Q_T) C_{V,0})}{2 Q_0 Q_Y^2 C_{V,0}^2}, \\ \omega_2 &= -u_0 k - i \frac{H^2 k^2 m R Y_0^2 (Q_T C_{P,0} + (Q_\rho - Q_T) C_{V,0})}{2 Q_0 Q_Y^2 C_{V,0}^2},\end{aligned}\quad (32)$$

where  $u_0 = \sqrt{\frac{T_0 C_{P,0}}{m C_{V,0}}} = \sqrt{\frac{T_0 (C_{P,\infty} T_0 Q_Y + R Y_0 H m (Q_\rho - Q_T))}{m (C_{V,\infty} T_0 Q_Y - R Y_0 H m Q_T)}}$  is the low-frequency sound velocity. So that, the new background mass concentration  $Y_{0,new}$  influences both low-frequency sound velocity and attenuation (or amplification) of sound. It easy to establish, that stability conditions for high-frequency and low-frequency sound are identical:  $\alpha > 0$  means unstable, non-equilibrium regime.

This study considers dynamic equations governing different types of motions in a chemically reacting gas without taking into account for its thermal and viscous (standard) attenuation. The terms, reflecting these phenomena (originating from the stress tensor and energy flux associating with thermal conductivity), should complete the momentum and energy equations from the system (1). The equation describing sound dynamics, accounting for the standard attenuation, takes a form

$$\frac{\partial \rho_1'}{\partial t} + u_\infty \frac{\partial \rho_1'}{\partial x} + \frac{(\gamma_\infty + 1) u_\infty}{2 \rho_0} \rho_1' \frac{\partial \rho_1'}{\partial x} - \alpha u_\infty \rho_1' - \frac{b}{2} \frac{\partial^2 \rho_1'}{\partial x^2} = 0, \quad (33)$$

where  $b = b_V + b_T$  is the diffusivity of sound,  $b_V = (4\eta/3 + \xi)/\rho_0$ ,  $b_T = \kappa(1/C_{V,\infty} - 1/C_{P,\infty})/\rho_0$ . Quantities  $\eta$ ,  $\xi$  and  $\kappa$  denote shear viscosity, bulk viscosity, and thermal conductivity, respectively. The standard attenuation always leads to a linear damping of sound during its propagation. Balance of two last terms in Eq.(33)) decides about amplification or damping of

a sound: if, for periodic sound,  $\alpha - b\omega_1^2/2u_\infty^3 < 0$ , the amplitudes of sound quantities decrease, otherwise, they increase during sound propagation.

The study is devoted to chemically reacting ideal gases by means of using of state equation for an ideal gas. The results may be easily expanded at other fluids by involving of correspondent equation of state instead of that for an ideal gas.

All equations in this study are derived within accuracy up to the terms of order  $M^2$  inclusively. Accuracy of conclusions are restricted by considering of only first partial derivatives of heat release  $Q(\rho, T, Y)$ . The conclusions are valid at confined temporal and spatial domains, where sound keeps still dominative. An important problem to overcome is spacial inhomogeneity of ambient quantities of a fluid which enhance simultaneously with increase of partial derivatives of the heat power produced in a gas with respect to density and temperature,  $Q_\rho$  and  $Q_T$ . Spacial inhomogeneity of ambient quantities essentially complicates mathematical analysis but may lead to new physically significant conclusions, as it was discovered relatively to gas with excited internal degrees of freedom. Preliminary estimations revealed that spacial inhomogeneity essentially expands an area of existence of non-equilibrium gas [12].

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