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NEW ACOUSTICAL PROPERTIES OF THE NON-EQUILIBRIUM GAS WITH THE REVERSIBLE CHEMICAL REACTIONS AND THE EXTERNAL INJECTION OF REAGENTS

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ABSTRACT

The perturbation propagation in the chemically active non-equilibrium gas mixture with the external volume pumping of the pure reagent is investigated. The external pumping of the pure reagent leads to the displacement of the chemical balance: the stationary values of the reagent concentrations are unequal to its equilibrium values. The stationary non-equilibrium mixture becomes acoustically unstable under certain conditions. The mixture behaviour and its perturbations structure depend on the external pumping of the reagent intensity, the chemical heat effect and rates of the direct and reverse chemical reactions. In the present work, we consider acoustics of such mixtures, mechanisms of the stabilization of the small perturbation growth and construct the shock wave adiabatic curves. A model of Arrhenius chemical reaction rate is used in the numerical results.

INTRODUCTION

For the last time, a large number of the theoretical and experimental works was devoted to the investigations of the gas dynamics of the non-equilibrium fluids [1]. It is connected with growing interest to this field from the applied sciences and intensive development of the plasma aerodynamics [1-11].

The gas dynamics of the non-equilibrium fluids such as vibrationally excited gas, weak ionized plasmas, three-phase fluids with non-equilibrium condensation appreciably differs from gas dynamics of the similar equilibrium fluids [1].

Under certain conditions the non-equilibrium fluid becomes acoustically active: small gas dynamic perturbations grow with the time. Acoustics of the non-equilibrium fluids was previously investigated with general point of view. It was achieved that the acoustical instability of the non-equilibrium fluids is connected with negative dispersion and negative second (bulk) viscosity [12].

As well, the anomalous behavior of the shock waves and its structure change is observed in the experiments with non-equilibrium fluids [1-11,13-17]. For the shock wave propagating through the field of the non-equilibrium gas, the shock wave amplification, the shock wave disintegration, its front acceleration and splitting, precursors, and others interesting effects are observed. All that effects do not have unified theoretical explanation and are interesting for the modern physics and applied sciences.

In the present work, the influence of the non-equilibrium on the perturbation propagation and its structure in the stationary non-equilibrium chemically active mixture with a reversible chemical reaction and an external mass reagent source is investigated. The fluid consists of two components A and B, the reversible chemical reaction has the form $\nu_A A \Leftrightarrow \nu_B B$. There are the external volume reagent source in the fluid which provides chemical non-equilibrium state and the volume mixture outflow which compensates inflow of the reagent A and supports the homogeneous stationary state. The fluid has the heat removing which compensates the chemical heating of the reaction.

NOMENCLATURE

ρ	[kg/m^3]	Density of the mixture
t	[s]	Time coordinate
z	[m]	Spatial coordinate
u	[m/s]	Velocity of the medium
m	[kg/m^{3}s]	Intensity of the reagent mass source
P	[Pa]	Pressure

U	[Ws]	Internal energy per mass unit
Q	[W/m^3]	Energy source per volume unit
M	[kg/mol]	Molar mass
x	[-]	Mol e fraction of the reagent A
Ω	[mol/m^3 s]	Chemical reaction rate
T	[K]	Temperature of the mixture
ν	[-]	Reaction order of the reagents
γ	[-]	Adiabatic exponent
R	[Ws/K/mol]	Universal gas constant
E	[Ws/mol]	Chemical energy of the reagents per mole unit

Special characters

div	[m^{-1}]	Divergence
∇	[m^{-1}]	Gradient

Subscripts

A	Reagent A
B	Reagent B
mix	Mixture
ext	External
st	Stationary state
0	Non-perturbed state

INITIAL SYSTEM OF THE EQUATIONS

The initial system of gas dynamic equations which describes the behavior of the investigated mixture has the form:

$$\begin{cases} \dfrac{\partial \rho}{\partial t} + div(\rho \vec{u}) = m_A + m_{mix} \\ \rho \dfrac{\partial \vec{u}}{\partial t} + \rho(\vec{u}\nabla)\vec{u} = -\nabla p + m_A(\vec{u}^A - \vec{u}) \\ \dfrac{dU}{dt} - \dfrac{p}{\rho^2}\dfrac{d\rho}{dt} = \dfrac{Q_{ext}}{\rho} \\ \dfrac{dx}{dt} = \dfrac{(M_A x + M_B(1-x))^2}{M_A \rho}\left(\dfrac{1-x}{M_A x + M_B(1-x)} m_A - \right. \\ \left. - \nu_B \Omega(x,\rho,T)\right) \\ p = \dfrac{\rho R T}{M_A x + M_B(1-x)} \\ U = \dfrac{x U_A}{M_A x + M_B(1-x)} + \dfrac{(1-x)U_B}{M_A x + M_B(1-x)} \end{cases} \quad (1)$$

The first equation is the mass balance equation. The right hand member of the first equation includes the volume mass of the pure reagent A pumping m_A and volume outflow of the reacting mixture m_{mix}.

The second equation of the system (1) is the Euler equation. Its right hand member includes the ordinary force which related with gradient of pressure and the additional force which related with inflow and outflow of the substance. The inflow and outflow substances have the different mass velocity. It leads to the additional force appearance. The additional force has the dissipation character.

Third equation of the system (1) is the energy balance equation. Let consider its right hand part. The source Q_{ext} can be represented in form:

$$Q_{ext} = Q_m + Q_k + Q_A + Q_V \quad (2)$$

Heat sources Q_m, Q_K, Q_A in (2) are related to volume inflow of the reagent and have the form:

$$Q_m = m_A(U_A{}^0 - U) \quad (3)$$

$$Q_k = m_A \frac{(\vec{u}^A - \vec{u})^2}{2} \quad (4)$$

$$Q_A = -(m_A + m_{mix})\frac{p}{\rho} \quad (5)$$

Heating (3) and (4) exist because the inflow and outflow substances have the different thermodynamic and mechanical states. Therefore, they have the different internal energy and kinetic energy per mass unit. In (3), $U_A{}^0$ [Ws/kg] is the internal energy of the mass unit of the inflow substance A. The third heat source (5) appears due to the mass exchange leading to the additional thermodynamic activity. The heat source Q_V provides the stationary state of the mixture. This source is cooling for exothermal reaction.

The fourth equation of the system (1) is the chemical reaction equation. In this equation the dynamics of the mole fraction of the reagent A is defined by the chemical rate and the intensity of the inflow m_A. This equation does not include the intensity of the outflow of the mixture m_{mix} because the outflow of the mixture leads to the outflow of the both reagents. In the present work for computing, we use the Arrhenius law of the chemical reaction rate:

$$\begin{aligned} \Omega(x,\rho,T) &= k_1\left(\frac{x}{M_A x + M_B(1-x)}\rho\right)^{\nu_A} e^{-\frac{E_f}{RT}} - \\ &- k_2\left(\frac{1-x}{M_A x + M_B(1-x)}\rho\right)^{\nu_B} e^{-\frac{E_b}{RT}} \end{aligned} \quad (6)$$

In (6) k_1, k_2 is the rate constants of the direction and reverse reactions respectively, E_f, E_b are energies of activation of the direction and reverse reactions, respectively.

However, all the analytical results have been obtained for the any chemical reaction rate model.

The pressure and the internal energy of the mixture dependences on the chemical composition of the mixture are presented in the last two equations of the system (1). Quantities U_A and U_B is the internal energy of the molecules A and B per one mole respectively. They have the form:

$$U_A = \frac{RT}{\gamma_A - 1} + E_A$$

$$U_B = \frac{RT}{\gamma_B - 1} + E_B$$

STABILITY OF THE INVESTIGATED MIXTURE

With relation to thermodynamics, the considered mixture has two degrees of freedom. We get temperature T and mass density of the mixture ρ as independent quantities.

We consider the one dimensional perturbation propagation in homogeneous stationary state of the above discussed mixture and linear stability.

The existence of the homogeneous stationary state requires the following conditions:

$$m_A + m_{mix} = 0 \tag{7}$$

$$Q_{ext} = 0 \tag{8}$$

$$\frac{1 - x_0}{M_A x_0 + M_B(1 - x_0)} m_A - \nu_B \Omega(x_0, \rho_0, T_0) = 0 \tag{9}$$

In the further investigation, we do not take into account the volume additional force in the right hand member of the second equation of the system (1). The condition (7) represents the mass balance. Condition (8) represents the energy balance and can be satisfied by respective cooling Q_V. We can find the stationary value of the mole fraction of the reagent A $x_{st}(\rho, T)$ from equation (9). The stationary value of pressure can be found by means of the state equation $p_{st}(\rho, T) = p(\rho, T, s_{st})$. It must be taken into account that the values x_{st} and p_{st} differ from its respective equilibrium values $x_e(\rho, T)$, $p_e(\rho, T)$ and equal them only under condition $m_A = 0$.

The intensity of the reagent A inflow m_A. takes part of the non-equilibrium degree of mixture.

Let consider the state of the mixture with density ρ_0, temperature T_0, pressure $p_0 = p_{st}(\rho_0, T_0)$, mole fraction of the reagent A $x_0 = x_{st}(\rho_0, T_0)$ and flow speed $\vec{u}_0 = 0$. We denote that values as unperturbed state.

To linearization of (1) we use $a = a_0 + a^{(1)}(z,t)$, where a is any thermodynamic quantity. Linearization of the first and the second equations of the system (1) under current conditions does not differ from its linearization for ordinary equilibrium fluid. Linearization of the equation of the chemical reaction gives the following equation:

$$x^{(1)} + \tau_{eff} \frac{\partial x^{(1)}}{\partial t} = \frac{x_0}{\rho_0} x_{st\rho} \rho^{(1)} + \frac{x_0}{T_0} x_{stT} T^{(1)} \tag{10}$$

In (10), the dimensionless derivatives $x_{st\rho}$ x_{stT} are defined by following expressions:

$$x_{st\rho} = \frac{\rho_0}{x_0} \left(\frac{\partial x_{st}}{\partial \rho}\right)_T$$

$$x_{stT} = \frac{T_0}{x_0} \left(\frac{\partial x_{st}}{\partial T}\right)_\rho$$

These expressions are calculated by means of (9) under stationary state of the fluid:

$$\left(\frac{-M_A}{(M_A x_{st} + M_B(1 - x_{st}))^2} m_A - \nu_B \left(\frac{\partial \Omega}{\partial x}\right)_{\rho,T}\right)\left(\frac{\partial x_{st}}{\partial \rho}\right)_T = \nu_B \left(\frac{\partial \Omega}{\partial \rho}\right)_{x,T}$$

$$\left(\frac{-M_A}{(M_A x_{st} + M_B(1 - x_{st}))^2} m_A - \nu_B \left(\frac{\partial \Omega}{\partial x}\right)_{\rho,T}\right)\left(\frac{\partial x_{st}}{\partial T}\right)_\rho = \nu_B \left(\frac{\partial \Omega}{\partial T}\right)_{x,\rho}$$

The effective time relaxation τ_{eff} is defined by equation:

$$\frac{1}{\tau_{eff}} = \frac{1}{\tau_r} + \frac{1}{\tau_m}$$

The typical time of the pure reagent pumping has the following form:

$$\tau_m = \frac{\rho_0}{m_A}$$

The relaxation time of the chemical reaction is defined by its rate:

$$\tau_r = \frac{M_A \rho_0 x_0}{\nu_B (M_A x_0 + M_B(1 - x_0))^2 \Omega_0 \Omega_x} \tag{11}$$

In equation (11), the following symbols were introduced:

$$\Omega_0 = \Omega(x_0, \rho_0, T_0)$$

$$\Omega_x = \frac{x_0}{\Omega_0} \left(\frac{\partial \Omega}{\partial x}\right)_{\rho,T}$$

Let consider linearization of the energy balance equation:

$$-\frac{T_0}{x_0 x_{stT}}\left((C_{V0} - C_{V\infty}) - \frac{\tau_{eff}}{\tau_Q} C_{VQ}\right)x^{(1)} + C_{V\infty}\tau_{eff}\frac{\partial T^{(1)}}{\partial t} + (C_{V0} - C_{V\infty})T^{(1)} =$$

$$= \frac{p_0}{\rho_0^2}\tau_{eff}\frac{\partial \rho^{(1)}}{\partial t} + \frac{T_0}{\rho_0 p_T}\left(c_{0T}^2 (C_{p0} - C_{p\infty}) - p_\rho (C_{V0} - C_{V\infty}) - \right.$$

$$\left. - \frac{p_\rho x_{stT} - p_T x_{st\rho}}{p_\rho} \frac{p_0}{\rho_0 T_0} \frac{c_{0T}^2 - p_\rho}{x_{st\rho}}\right)\rho^{(1)}$$

Here,

$$\tau_Q = \frac{T_0}{Q_{ext0}}$$

is the characteristic time of heating, $c_{0T}^2 = p_\rho + p_x x_{st\rho}$ is the low-frequency isothermal dimensionless velocity . The specific heat capacities are defined by following expressions:

$$C_r = \frac{1}{T_0}\left(\frac{\partial U}{\partial x}\right)_T$$

$$C_{VQ} = \frac{x_{stT} Q_{extx}}{\rho_0} + \frac{Q_{extT}}{\rho_0}$$

$$C_{V0} = C_{V\infty} + x_0 x_{stT} C_r - \frac{\tau_{eff}}{\tau_Q}\frac{1}{\rho_0} Q_{extT}$$

$$C_{pQ} = -\frac{Q_{extT}}{\rho_0} - \frac{Q_{extx}}{\rho_0}\frac{p_\rho x_{stT} - p_T x_{st\rho}}{p_\rho + p_x x_{st\rho}} + \frac{Q_{ext\rho}}{\rho_0}\frac{p_T + p_x x_{stT}}{p_\rho + p_x x_{st\rho}}$$

$$C_{p0} = C_{p\infty} + \frac{p_\rho x_{stT} - p_T x_{st\rho}}{p_\rho + p_x x_{st\rho}}\left(C_r x_0 + p_x \frac{p_0}{\rho_0 T_0}\frac{1}{p_\rho}\right) -$$

$$- \frac{\tau_{eff}}{\tau_Q}\frac{1}{\rho_0}\frac{p_\rho Q_{extT} - p_T Q_{ext\rho}}{p_\rho + p_x x_{st\rho}}$$

where

$$p_\rho = \frac{\rho_0}{p_0}\left(\frac{\partial p}{\partial \rho}\right)_{x,T}$$

$$p_T = \frac{T_0}{p_0}\left(\frac{\partial p}{\partial T}\right)_{x,\rho}$$

$$p_x = \frac{x_0}{p_0}\left(\frac{\partial p}{\partial x}\right)_{\rho,T}$$

$$Q_{ext\rho} = \frac{\rho_0}{Q_{ext0}}\left(\frac{\partial Q_{ext}}{\partial \rho}\right)_{x,T}$$

$$Q_{extT} = \frac{T_0}{Q_{ext0}}\left(\frac{\partial Q_{ext}}{\partial T}\right)_{x,\rho}$$

$$Q_{extx} = \frac{x_0}{Q_{ext0}}\left(\frac{\partial Q_{ext}}{\partial x}\right)_{\rho,T}$$

From the linearized system of the equation (1), we obtaine the following dispersion relation

$$\frac{\omega^2}{k^2} = \frac{p_0}{\rho_0}\frac{(p_\rho + p_x x_{0\rho})C_{p0} - (p_\rho + p_x x_{0\rho})C_{pQ} / i\omega\tau_Q - i\omega\tau_{eff}\, p_\rho C_{p\infty}}{C_{V0} - C_{VQ} / i\omega\tau_Q - i\omega\tau_{eff0} C_{V\infty}}$$

The previous denotations were introduced in order to lead the dispersion relation for the investigated mixture to the form of the dispersion relation of typical non-equilibrium fluids [18]. By means of the obtained dispersion relation and chemical rate model (6), we can find conditions of instability, dispersion and second (bulk) viscosity for the investigated mixture.

In figure 1, the dependence of minimum damping constant of the sound waves

$$\alpha_{\min} = \left(\frac{RT_0}{p_0}\right)^{\nu_A - 1}\sqrt{\frac{RT_0}{M_A}}\frac{1}{k_1}\min_{\omega}(\mathrm{Im}(k))$$

on the intensity of the pumping of the reagent A is represented. Here,

$$F = \frac{m_A}{k_1 M_A}\left(\frac{RT_0}{p_0}\right)^{\nu_A}.$$

In fields $F_1 < F < F_2$ or $m_{A\min} < m < m_{A\max}$, where the damping constant is negative, the mixture becomes acoustically unstable.

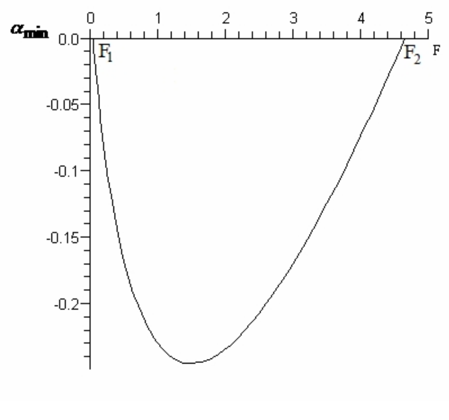

Figure 1 Minimum damping constant of the sound waves

This result differs from the similar result for other non-equilibrium fluids for which usually only one boundary value of the non-equilibrium degree exists. Let consider physical contents of the first and second boundary values.

Acoustical instability of the non-equilibrium fluids takes place due the positive feedback between the non-equilibrium degree and local perturbations in the fluids [18]. For example, in the vibrationally excited gas the local temperature excess leads to decrease of the VT relaxation time. Decrease of the relaxation time leads to additional transfer of the non-equilibrium vibrationally energy to the translational degrees of freedom. Thereby, the local temperature excess leads to the futher temperature increase. In the case of the investigated mixture, we consider the chemical reaction equation. The local increase of the temperature ΔT in the first order described by the following equation:

$$\frac{dx}{dt} \sim [\nu_B k_1 (\frac{x}{M_A x + M_B(1-x)}\rho)^{\nu_A} e^{-\frac{E_f}{RT}}(E_b - E_f) - \\ - \frac{1-x_0}{M_A x_0 + M_B(1-x_0)} m_A E_b]\Delta T \qquad (12)$$

If $m_A = 0$, the behaviour of the reaction is defined by the first term of the right part of (12). Its sign is defined by sign of the expression $(E_b - E_f)$. For the exothermal reaction, this sign is positive and the local temperature increase leads to the increase of the reverse chemical reaction and heat absorption in the reaction. For the equilibrium chemical reactions, equation (12) demonstrates the Le Chatelier's Law.

If the external source of the pure reagent A exists then after the first boundary value $m_{A\min}$ The second term dominates over the first in (12). The temperature increase leads to the increase of the direct reaction and heating. The external inflow and outflow execute the effect which similar to the existence of the additional reverse chemical reaction which leads to the appearance of the positive feedback.

The second component of the right hand side of the (12) is proportional to quantity $1 - x_0$, i.e., to the mole fraction of the product of the reaction. The chemical balance displaces to the substance A with increase of m_A and mole fraction of the product of the reaction nonlinearity decreases. That leads to positive feedback disappearance. Therefore, in the investigated mixture the second boundary value $m_{A\max}$ exists.

SHOCK ADIABATS

The instability of the investigated mixture leads to the growth of the small perturbations. Small perturbations increase with the time and evaluate to the shock waves. With growth of the temperature in the shock wave the non-equilibrium degree which defined, for example, as the relative deviation of the stationary mole fraction $x_{st}(\rho,T)$ from its equilibrium value $x_e(\rho,T)$ decreases. That leads to perturbation stabilization.

Further, we consider the stationary shock wave structure and its dependence on the shock wave velocity m_A and q.

Let use the method of the shock adiabat to define the shock wave structures. We will use the well known Zeldovich model for the shock wave structure. Accordingly that theory the change of the thermodynamic quantities on the shock wave front occurs sharply. In the further flow, there is the relaxation zone (chemical reaction zone) and zone of the homogenous stationary state.

The equilibrium shock adiabat represents on the $p-V$ plane the stationary states which associated with the final state of the fluid after the shock wave propagation. To obtain the equation of the equilibrium shock adiabat let suppose that the final state of the fluid is homogenous and stationary. Thereby, we obtain the following system of the equations for the equilibrium shock adiabat:

$$x\left(\frac{\rho_0 D^2 + p_0}{\rho} - \frac{\rho_0{}^2 D^2}{\rho^2}\right)\frac{1}{\gamma_A - 1} + \frac{x}{M_A x + M_B(1-x)}E_A +$$
$$+(1-x)\left(\frac{\rho_0 D^2 + p_0}{\rho} - \frac{\rho_0{}^2 D^2}{\rho^2}\right)\frac{1}{\gamma_B - 1} + \frac{(1-x)}{M_A x + M_B(1-x)}E_B +$$
$$+\frac{\rho_0 D^2 + p_0}{\rho} - \frac{\rho_0{}^2 D^2}{2\rho^2} = x_0 \frac{p_0}{\rho_0}\frac{1}{\gamma_A - 1} + \frac{x_0}{M_A x_0 + M_B(1-x_0)}E_A +$$
$$+(1-x_0)\frac{p_0}{\rho_0}\frac{1}{\gamma_B - 1} + \frac{(1-x_0)}{M_A x_0 + M_B(1-x_0)}E_B + \frac{D^2}{2} + \frac{p_0}{\rho_0}$$

$$\nu_B k_1\left(\frac{x}{M_A x + M_B(1-x)}\rho\right)^{\nu_A} e^{-\frac{E_f}{RT}} -$$
$$-\nu_B k_2\left(\frac{1-x}{M_A x + M_B(1-x)}\rho\right)^{\nu_B} e^{-\frac{E_b}{RT}} = \frac{1-x}{M_A x + M_B(1-x)}m_A$$

Here, we use the chemical reaction rate model (6).

The frozen adiabat represents on the $p-V$ plane the states which associated with the sharp shock wave front. The reagents are conserved during gas passage through the sharp front.

The results of the numerical simulation of the equilibrium and frozen adiabats are presented in figure 2. The parameters in figure 2 correspond to the unstable initial state.

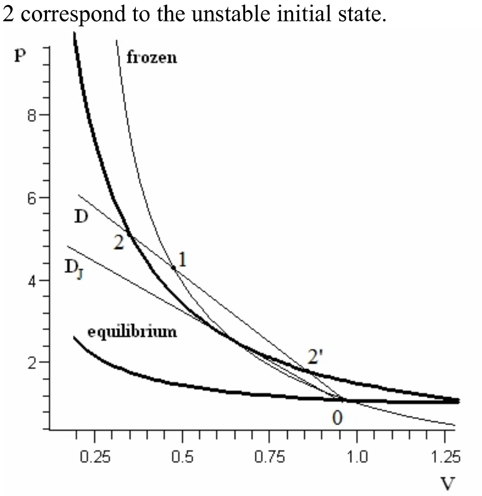

Figure 2 Equilibrium and frozen shock adiabats

The equilibrium adiabat consists of two branches. Any shock wave speed D are defined by the slope of the chord which passes through the initial point 0. The tangent of the chord is proportional to the D^2. One can obtain from the conservation laws that the point associated with current flow state locates on the chord. Thereby, gas from the initial point 0 passes to the frozen adiabat by jump (point 1). Further, gas slowly relaxes to the final point on the equilibrium adiabat (point 2 or 2').

We show that the equilibrium shock adiabat for the chemical reactive mixture with reversible chemical reaction and external source of the pure reagent is similar to one for the non-equilibrium gas with external source of the vibrational energy [19]. So, we can use the shock wave profiles obtained in [19]. Among them the profiles with smooth increase or smooth decrease after sharp front, self-sustained pulse and Chapman-Jouguet wave. For the Chapman-Jouguet wave, the chord which defines the shock wave speed D_J concerns upper branch of the equilibrium adiabat.

CONCLUSION

The behavior of the gas dynamic perturbations in the stationary non-equilibrium chemically active mixture with reversible chemical reaction and external pure reagent source is similar to one in the stationary non-equilibrium vibrationally excited gas with external vibrational pumping. These fluids have the similar equilibrium shock adiabat with two branches.

In the chemically reactive mixture, there are two boundary values of the pumping defining the interval of parameters associated with acoustical instability.

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