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TRAVELLING WAVES OF FILTRATION COMBUSTION WITH HOMOGENEOUS-HETEROGENEOUS REACTIONS

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ABSTRACT

An experimental study was carried out of steady-state filtration combustion waves simultaneously occurring in catalytic heterogeneous and homogeneous gas-phase reaction (travelling hybrid waves). The hybrid wave velocities and velocities of heat wave in inert porous media (LVR wave), and the maximum and equilibrium temperatures in the combustion zone were measured. It was found peculiarities of the hybrid waves. These waves travel more rapidly than the LVR waves though the maximum and equilibrium temperatures of the hybrid waves are lower than that of the LVR one. The difference in the velocities of these two types of waves is especially substantial at low filtration velocities. It has been investigated some transformations of the hybrid waves. It had been shown that it is possible for a hybrid wave to be transformed into a wave where either the heterogeneous or homogeneous reactions dominate, depending on whether the gas filtration velocity decreasing or increasing. It has been further shown that, under conditions of vanishing catalytic activity of the porous medium surface, the hybrid wave is transformed into a wave with dominant homogeneous reaction. Under transition from catalytic media to inert one there exist two options. One option is change of velocity value and another one is stabilization of the wave at the boundary of the two media. On the basis of a proposed theoretical model, a parametric analysis of the hybrid wave was carried out. An interpretation of the experimental results is presented.

Nomenclature

c - specific heat capacity,
D - diffusion coefficient,
E - activation energy,
m- porosity of medium,
k - pre-exponential factor,
Q - heat release of reaction,
R - universal gas constant,
S - catalytic surface area per unit volume,
T, θ - temperature of gas and solid,
u - wave velocity,
v - velocity of gas relative to solid,
W - reaction rate,
x - spatial coordinate.

Greek Symbols
α_T - heat transfer coefficient,
α_η - mass transfer coefficient,
λ - effective thermal conductivity,
η - mole fraction of deficient component,
κ - temperature conductivity,
ρ - density.
Subscripts
T- gas,
θ- solid,
0 - initial value,

m - maximum value,
in - wave with homogeneous reaction,
h - wave with homogeneous and heterogeneous reactions.

INTRODUCTION

Filtration combustion is usually defined as a wave process of exothermic transformation in porous medium upon gas filtration (Fig. 1). In a steady-state regime a combustion wave propagates at a constant velocity and has a characteristic structure and an equilibrium state of the system behind the combustion front. A particular feature of this type of combustion is the relative movement of phases. This usually results in the filtering gas acting both as reactant and heat carrier. Therefore the vector of the mean velocity of filtering flow is a principally important process parameter that determines not only the velocity and structure of a combustion wave, but also equilibrium states, e.g. combustion temperature.

Processes of filtration combustion can be divided in two categories according to chemical reaction type. The first one is the processes with heterogeneous reactions occurring on the surface of solid phase. The second includes processes with reactions proceeding in the gas phase of porous space. Heterogeneous reactions take place in such filtration processes as, e.g., blast furnace smelting, ore agglomeration, self-propagating high temperature synthesis [1], and propagation of heat waves in a bed of catalyst [2]. Homogeneous gas reactions occur in the filtration combustion of gases in inert porous media [3]. At present time it is known several steady-state regimes of thermal wave propagation with homogeneous gas reactions in inert porous media. They are Low Velocity Regime (LVR) and High Velocity Regime (HVR), Sonic Velocity Regime (SVR), regime of Low Velocity Detonation (LVD) and Normal Detonation with heat and pulse losses (ND) [4, 5]. From here on only one type of the regimes of gas filtration combustion will be considered - the one with low velocities (LVR waves).

Both types of filtration combustion with homogeneous and heterogeneous reactions have a number of common properties and dependencies. The most remarkable one is the possibility of realizing super- and sub-adiabatic combustion temperatures. On the other hand, there are also very distinctive and specific features. For instance, the LVR wave is characterized by the peak of abnormally high temperature in the reaction zone, possibility of transition to HVR of combustion, etc.

The rigid division of the processes into these two categories, with corresponding separate and independent development of their theories, is not always correct. In practice many filtration processes seem to include gas - phase and heterogeneous reactions proceeding simultaneously. A similar situation is discussed in certain cases of "catalytic combustion" reviewed in [6-9]. The term "catalytic combustion" is defined usually as combustion stabilized in a catalytically active porous medium acting as a part of burning device. In this case the conditions of the stabilization of a chemical reaction zone include boundary parameters at the inlet and outlet of a catalyst bed and its length. This circumstance is the principal limitation in the study of travelling waves whose properties are set under conditions of unlimited space. Practical possibility of processes with heterogeneous and homogeneous reactions proceeding simultaneously under wave combustion regime (hybrid travelling waves) was demonstrated in [10].

In the present work the structure and velocities of hybrid travelling waves were studied experimentally and theoretically. Mathematical simulation of this process includes, as the basic elements a two-temperature description and the insertion of a term relating to heat evolution in the equations of heat balance for both gas and solid phase. This allows one to determine the conditions for either of the chemical reaction types to predominate. Under such an approach, processes with only homogeneous or heterogeneous reactions, as studied previously, can be considered as particular extreme cases of hybrid waves.

EXPERIMENTAL APPARATUS AND RESULTS

The experiments were carried out in a vertical quartz tube with a diameter of 40 *mm* (Fig. 2). The upper half of the tube with a height of 250 *mm* was filled by granulated oxide Al-Cu-Cr catalyst and the lower part by granules of alumina catalyst support. The catalyst was copper chromite $CuCr_2O_4$ supported on spherical granules of α-Al_2O_3. It was prepared by impregnation of support granules with an aqueous solution of copper dichromate followed by drying and calcination at 1300 *K* for 4 *h*. The granule diameter was 1.0 - 1.6 *mm*, and bulk density 1.46 g/cm^3. The activity of the fresh catalyst was measured in a flow recycle setup and characterized by the rate of butane oxidation in air at an initial butane concentration of 0.5 vol. % and the conversion of C_4H_{10} equal to 60%. At 593 *K* and 693 *K* the rate was respectively $0.02 \cdot 10^{-2}$ and $0.24 \cdot 10^{-2}$ cm^3/gs. As the catalyst characteristics changed under the influence of high temperatures, after each experiment the catalyst was replaced by a fresh sample.

For temperature measurement of the solid media, W-Re thermocouples with a diameter of 0.1 *mm* were used. The temperature of gas-phase was measured by Pt-Rh thermocouples of 20 microns diameter. For the elimination of catalytic effects and protection from oxidation, the thermocouples were covered by a passivating layer prepared from silica and Al-Cr-P binder. For gas temperature measurement special method was taken to exclude the contact of the thermocouple junction with the granules. For this goal the thermocouple junction was placed in the center of a ball of paraffin or ice. The balls have size of interstitial space of the porous medium and placed among granules of catalyst or inert α-Al_2O_3. Then the balls were evaporated by hot air flow.

The travelling waves were initiated at the open upper end of the tube by a ring-shaped electric heater which heated the catalyst layer of 40 *mm* height to the ignition temperature (570-600 *K)*. Then the heater was switched off, and premixed propane/air or hydrogen/air mixtures with preset composition and velocity were fed into the lower end of the tube. After certain time a heat wave was formed in the catalyst bed, which traveled downwards at constant velocity u_h. When the wave reached the lower boundary of the catalyst bed it rearranged and propagated farther (at certain parameters) also at constant velocity $u_{in} < u_h$. The wave velocity was determined by thermocouple temperature measurement data.

Wave velocity measurement. The results stationary wave velocities in catalytically active (u_h) and inert (u_{in}) porous media are given in Table 1 for C_3H_8/air at fixed flow (v_0) and varying % fuel and Fig. 3 for H_2/air at fixed (60%) fuel but varying flow rate (v_0). It is seen that the ratio of velocities in these media is rather high. For propane/air mixtures: the lower the calorific value of the mixture, the higher the difference. At sufficiently low % propane the counterflow wave propagation is possible only in the catalytic bed and impossible in the bed of inert solid. For instance, for the mixture 3% C_3H_8 at a filtration velocity of $v_0 = 0.5$ *m/s* in the catalytic bed, $u_h = 3.5 \cdot 10^{-2}$ mm/s, but the wave does not penetrate into the inert bed and is stabilized at the boundary of the two media. Maximum velocities for the both media are found in the case of stoichiometric mixtures.

As far as the dependence $u(v_0)$ is concerned, the hybrid wave has a U-form typical for inert media (Fig. 3) [4]. However the maximum of the velocity in the case of hybrid waves is substantially shifted to the region of low filtering velocities. The value $|u_h|_{max}$ is situated near a critical filtering velocity ($v^*_{0,in}$) when blow off of combustion occurs in inert porous medium. On the whole, the difference in the velocities for these two types of waves is most significant at low filtering velocities. The range of v_0 values in which the propagation of the hybrid wave in counterflow direction is possible is much wider than that for the LVR, wave travelling in inert medium. At $v_0 < v^*_{0,in}$ only hybrid waves can be realized.

Measurement of temperature of phases in hybrid wave. If the channels in porous medium are not too small, LVR waves, as was mentioned above, produce a peak of abnormally high temperatures in the chemical reaction zone, resulting from heat recuperation in the flame region. The presence of this high temperature peak in the chemical reaction zone was considered as the main evidence of gas-phase reaction proceeding in the hybrid wave [10]. Figures 4 and 5 give the results of the measurement of maximum temperatures of gas and solid for propane/air and hydrogen/air mixtures. Figure 5 also presents the corresponding maximum temperatures for LVR waves measured for hydrogen/air mixture.

For the both fuels, some common features and dependencies are observed. Firstly, both in inert and catalytically active porous media there can be a sharp difference between maximum temperatures of the two phases (temperature peak), which is the evidence of a gas - phase chemical reaction. This difference increases with increasing filtration velocity. On the contrary, at low v_0 maximum phase temperatures coincide. Secondly, in catalytically active media waves with very low combustion temperatures (as compared with LVR waves) can propagate. In the present experiments the following values of minimum combustion temperatures were observed: for 3% propane/air mixture - 850 *K;* for 65% hydrogen/air mixture - 590 *K*. For the comparison: the lowest temperature in the LVR wave formed by 65% hydrogen/air mixture is about 1170 *K*.

Finally, as seen from Figs. 3 and 5, under the same conditions of mixture composition and filtration velocity, the maximum temperatures are lower and wave velocities are greater (in absolute terms) in hybrid waves than those in LVR waves.

MATHEMATICAL MODEL

The model proposed for the theoretical study of hybrid waves is the system of one-dimensional equations of: heat transfer in gas phase, heat transfer in porous medium (in solid carcass), mass transfer of a deficient component of gas mixture, mass transfer through an interface, conservation of the total amount of the substance in a flow, and the perfect gas equation [11]. It is supposed that (1) the gradients of pressure caused by forced filtration or combustion process are negligibly small, (2) radiation heat transfer is taken into account by the effective coefficient of heat conductivity of the carcass, (3) porous medium is stationary, it is not deformed and does not change its mass, (4) the process is adiabatic, heat losses into outer space are absent, (5) gravitational forces do not influence the process of combustion. With these assumptions the dimensionless equations are the following:

$$\begin{gathered}
\frac{d}{d\xi}\kappa_T\frac{dy}{d\xi}-\frac{dy}{d\xi}+\alpha_T(z-y)+y_b\tau_T W_T=0,\\
\frac{d}{d\xi}\kappa_\theta\frac{dz}{d\xi}+(1-\omega)\frac{dz}{d\xi}+\alpha_T(y-z)+y_b\tau_\theta W_\theta=0,\\
\frac{d}{d\xi}\kappa_\eta\frac{d\eta_\mathrm{T}}{d\xi}-\frac{d\eta_\mathrm{T}}{d\xi}+\alpha_\eta(\eta_\theta-\eta_\mathrm{T})-\tau_\mathrm{T}W_\mathrm{T}=0,\\
\alpha_\eta(\eta_\mathrm{T}-\eta_\theta)-\tau_\theta W_\theta=0.
\end{gathered}\tag{1}$$

Here the symbols and subscripts T and η relate to gas and θ to the solid phase. For dimensionless variables, subscripts 0 and m denote parameters at initial and maximum gas temperatures respectively, α_T and α_η are coefficients of heat and mass transfer, S is specific surface area, κ_T and κ_θ are pre-exponential factors of homogeneous and catalytic reactions, m is porosity. Dimensionless variables and parameters are defined as follows:

$$\begin{gathered}
y=\frac{T-T_0}{T_m-T_0},\ z=\frac{\theta-T_0}{T_m-T_0},\ \kappa_\eta=\frac{\rho_{T,0}D}{GL},\ \xi=\frac{x}{L},\\
\kappa_T=\frac{\lambda_T}{c_{PT}GL},\ \kappa_\theta=\frac{(1-m)\lambda_\theta}{mc_{PT}GL},\ G=\rho_\mathrm{T}(v-u),\ y_b=\frac{\Delta T_b}{T_m-T_0},\\
\alpha_T=\frac{\alpha_{T,0}SL}{mc_{PT}\rho_T},\ \alpha_\eta=\frac{\alpha_{\eta,0}SL}{mG},\ \omega=\frac{v-(1+\sigma)u}{v-u},\\
\sigma=\frac{(1-m)c_{P\theta}\rho_\theta}{mc_{PT}\rho_\theta},\ L=\frac{m\lambda_{T,m}+(1-m)\lambda_{\theta,m}}{mc_{PT}G},\ \beta_T=\frac{RT_m}{E_T},\\
\tau_T=\frac{L\rho_T}{G}k_T\exp\left(-\frac{1}{\beta_T}\right),\ \beta_\theta=\frac{R\theta_m}{E_\theta},\ \tau_\theta=\frac{(1-m)L\rho_T}{mG}k_\theta\exp\left(-\frac{1}{\beta_\theta}\right).
\end{gathered}$$

Here W_T and W_θ are the rates of first order reactions after Frank-Kamenetsky transformation, other designations are standard.

For the equations (1), the following boundary conditions having obvious physical sense are used:

$$\begin{gathered}
\xi=-\infty,\quad y=z=0,\quad \eta_T=1\\
\xi=\infty,\quad y=z=y_e,\quad \eta_T=0,
\end{gathered}$$

where y_e is equilibrium dimensionless temperature that is to be determined. Then, the total balance of heat in the system gives the value of y_e [11]:

$$y_e=y_b/\omega.$$

The use of the method of counter extrapolation [12] allows one to obtain the equation for the determination of ω:

$$\frac{2}{\kappa_{\eta,m}}\left[\tau_{T,m}\left(\frac{\gamma_T}{y'}\right)^2+\frac{\alpha_\eta \tau_{\theta,m}}{\alpha_\eta+\tau_{0,m}}\left(\frac{\gamma_\theta}{z'}\right)^2\right]=1, \qquad (2)$$

where y' and z' are the fluxes extrapolated from the zone of pre-heating to the reaction zone. Here $\gamma_T=\frac{\beta_T T_m}{T_m-T_0}$, $\gamma_\theta=\frac{\beta_\theta \theta_m}{T_m-T_0}$. Equating the mass transfer coefficient α_η to zero it is not difficult to obtain from (2) the equation for the determination of ω in inert porous medium [11]:

$$2\tau_{T,m}\left(\frac{\gamma_T}{y'}\right)^2=\kappa_{\eta,m}.$$

DISCUSSION OF THE EXPERIMENTAL AND THEORETICAL RESULTS

The analytical solution of the system (1) yields the wave velocity, the distribution of temperatures and concentrations in combustion wave, contributions of homogeneous and surface reactions in total heat release and other main characteristics of the process.

Calculated velocities of combustion wave versus filtration velocity for catalytically active and inert media are given in Fig. 6. In the calculations for inert media, it was assumed that $\alpha_{\eta,0}=m_\theta=0$. In both cases, the following values of parameters were taken: $\rho_{T,0}=0.5$, $\rho_\theta=1500\ kg/m^3$; $c_{PT}=2500$, $c_{P\theta}=1000\ J/(kg\cdot K)$; $\lambda_T=0.1$, $\lambda_\theta=1\ W/(m\cdot K)$; $E_T/R=2\cdot 10^4$; $E_0/R=7500K$; $k_T=10^{12}$, $k_0=5\cdot 10^6 1/s$; $m=0.5$; $\alpha_{T,0}=10^4 W/(m^2K)$; $\alpha_{\eta,0}=4kg/(m^2s)$; $S=3000\,1/m$; $T_0=300\ K$; $Q/c_{PT}=1100\ K$.

Fig. 6 shows that the calculated values $u(v_0)$ for catalytic and inert media qualitatively correlate with the experimental ones: the characteristic U - shape dependencies are reproduced, $|u_{h,m}|>|u_{in,m}|$ and the position of $|u_{in,m}|$ is shifted with respect to $|u_{h,m}|$ to the region of lower values of v_0.

Figure 7 presents the calculated dependencies of the maximum temperatures of the phases on the filtration velocity for catalytic and inert porous media. It can be seen that at low the v_0 maximum temperatures of gas and solid phase coincide. As the filtration velocity increases, the difference in the maximum temperatures of the two phases in the wave front appears. This means that, apart from the catalytic heat source, a homogeneous one appears that is due to the concurrent change in the ratio of the rates of chemical reactions.

For the quantitative estimation of this change, values of energy release due to homogeneous and catalytic reactions were introduced:

$$I_T=\tau_T\int_{-\infty}^{+\infty}W_T d\xi,\ I_0=\tau_0\int_{-\infty}^{+\infty}W_0 d\xi,$$

that represent the fractions of heat release caused by homogeneous and catalytic heterogeneous reactions in the total heat balance.

The dependencies $I_T(v_0)$ and $I_\theta(v_0)$ are illustrated by Fig. 8. It is seen that two regimes of combustion are realized: with predominant gas-phase and heterogeneous heat release, in the ranges of high and low filtration velocities respectively. The interpretation of this fact is connected with different temperature dependencies of gas-phase and heterogeneous reactions. Figures 5 and 7 show that the maximum phase temperatures in the vicinity of which the main homogeneous and surface reactions occur increase with the increasing filtration velocity. Therefore at low v_0 catalytic reactions with low activation energies should prevail, whereas at high v_0 the homogeneous ones with high activation energies should be predominant. At some extent the temperature heterogeneous depends not only on the temperature sensitivity of gas-phase and heterogeneous reactions but on the porous medium dispersity too.

The comparative analysis of catalytic, LVR and hybrid waves shows that the latter have dual properties. On the one hand the properties of catalytic waves are exhibited (possibility of low-temperature process), on the other hand the LVR waves properties (temperature heterogeneity in the zone of chemical reaction). This duality reflects the fact of the combined form of heat release when the two reactions, gas-phase and heterogeneous ones, proceed simultaneously and compete. This concurrent interaction has both a material and heat basis, because not only temperatures (in the range of parameters when ($T_m = \theta_m$), but also the initial reagents are common for the two reactions.

In the mathematical analysis the combined heat release is accounted for by including heat sources into the two energy balance equations which have wave properties. This circumstance predetermines a number of interesting possibilities of hybrid waves.

In the first place, there exists the possibility of the transfer from the regime of the filtration combustion with heterogeneous reaction to the one with a prevailing gas - phase one, and vice versa upon the change of the physical parameters. Such transitions from catalytic wave to LVR one upon the increase of filtration velocity are demonstrated by Figs. 4, 5, 7 and 8. It is of interest that in this case the change of the physical parameter (v_o) results in the change of the type of dominating chemical reaction. In the second place, there is possibility of analogous transitions upon the change of chemical parameters. This possibility is realized in the experiments presented here on the transition of the wave from catalytic porous medium into inert one. In this case only the change of the surface reactivity, with all other parameters being unchanged, can lead to the change of combustion regime and physical characteristics of the process (e.g. T_m, θ_m equilibrium temperature T_e). As this takes place a paradoxical fact is observed, i.e. at the transition of the boundary of the two media wave velocity $| u |$ decreases, while maximum and equilibrium temperatures increase (Figs. 3, 5). From practical viewpoint this change of regimes is very important for the understanding of the dynamics and development of uncontrolled explosions in catalyst production industry.

Finally, the proposed qualitative mathematical model describes rightly the main regularities of hybrid waves observed in the experiments. For quantitative describing it is necessary to detail the mathematical model, to take into account many additional factors such as detailed kinetics of chemical reactions, temperature dependence of the parameters of heat and mass exchange, multi-dimensionality etc. But this is the subject of future studies.

CONCLUSION

The phenomenon of travelling hybrid waves, by the number of parameters and variety of properties, is more complex than related phenomena of catalytic waves and waves of filtration gas combustion. The latter ones can be considered as particular cases of hybrid waves with heterogeneous and homogeneous reactions dominating correspondingly. Hybrid waves, while having the properties of catalytic waves and the waves of gas filtration combustion, possess specific features. Among these the most interesting one is the possibility of their transformation into catalytic waves or waves of gas-phase combustion in inert porous medium. The study of hybrid waves allows one to understand the role of homogeneous reaction in catalytic processes and the role of surface reaction in processes of filtration gas combustion. From practical point of view the study of hybrid waves allows the diagnostics of emergency cases caused by uncontrolled explosions in catalytic processes and helps to develop new methods of control over filtration gas combustion. The vivid example of the latter is the method to increase the stability of gas combustion using a catalytic igniter installed in a burner [13].

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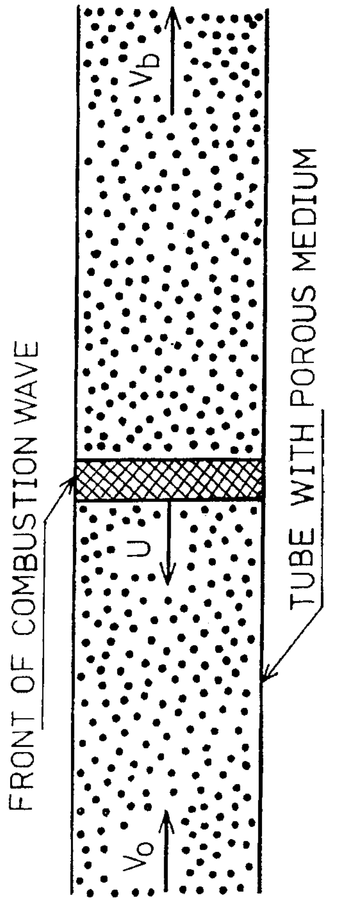

Fig. 1. The scheme of filtration combustion. v_0- filtration velocity, u - combustion wave velocity, v_b- combustion products velocity

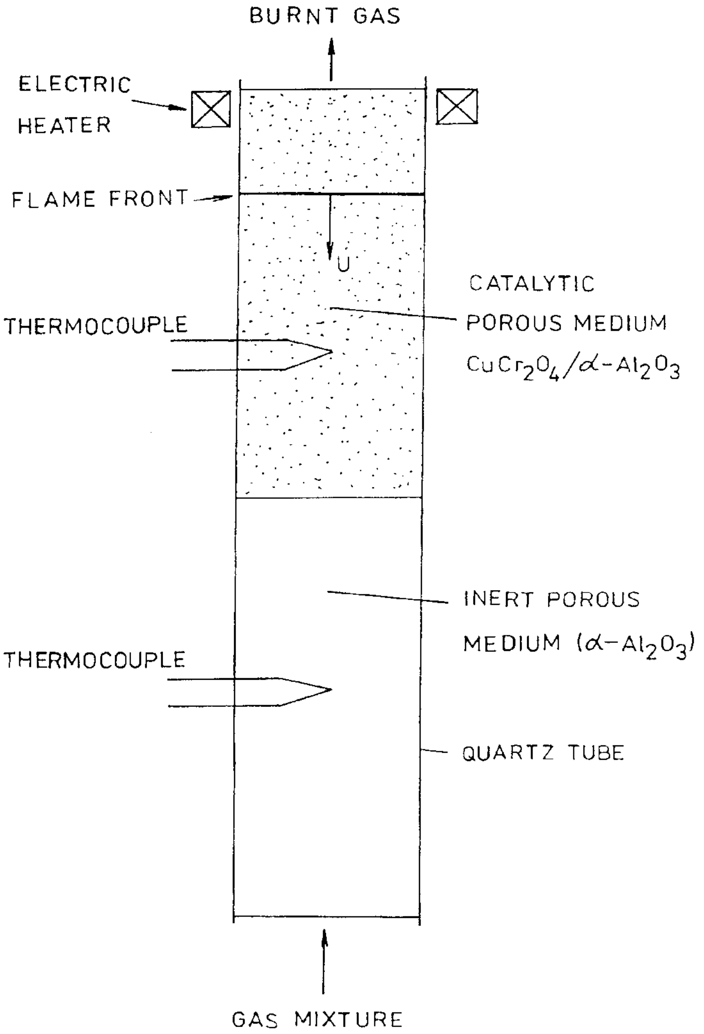

Fig. 2. The tube for hybrid travelling waves investigation

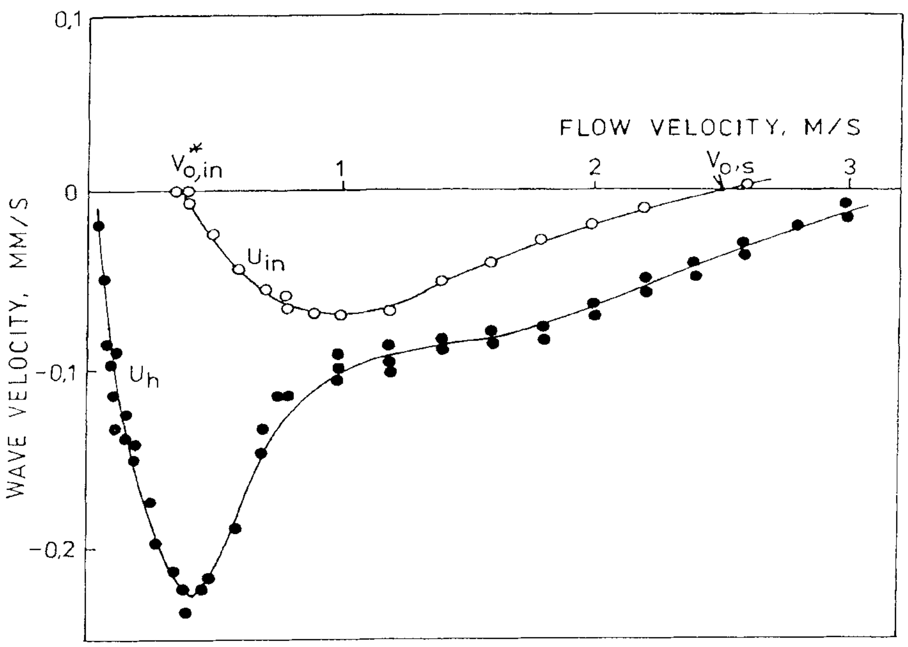

Fig. 3. The dependence of wave velocity in inert (u_{in}) and catalytic active (u_h) porous media on filtration velocity of reacting gas. Reacting gas: 65% H_2 + air. $v^*_{0,in}$ – critical velocity of reacting gas, $v_{0,s}$ – velocity of reacting gas corresponding to stationary state of the combustion wave.

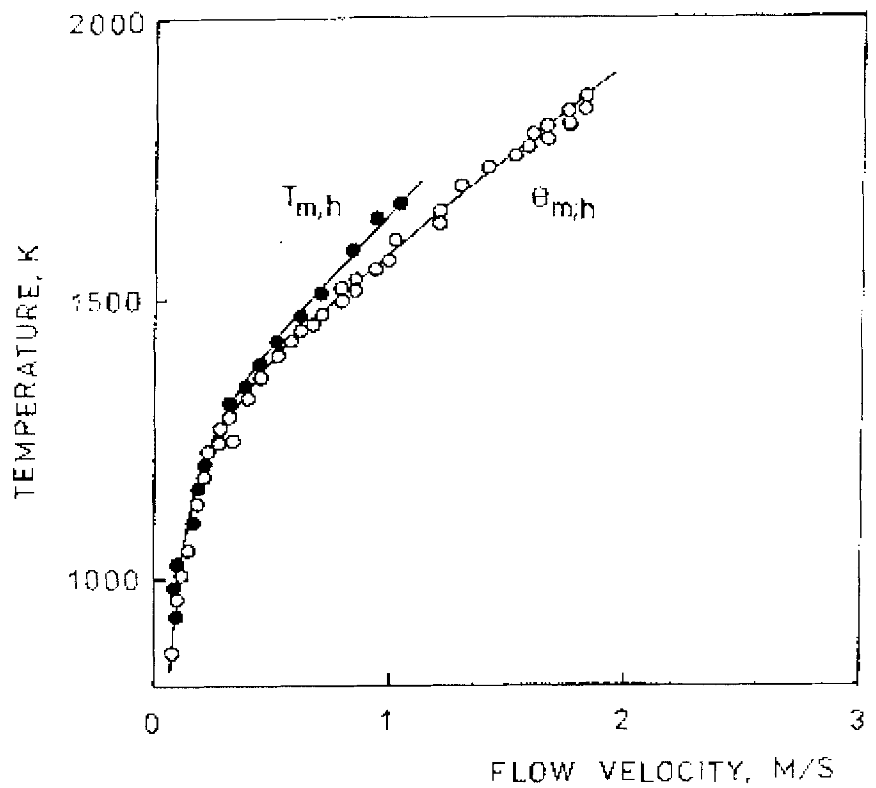

Fig. 4. The dependence of maximum gas ($T_{m,h}$) and porous medium ($\theta_{m,h}$) temperatures in the wave with homogeneous-heterogeneous reaction on filtration velocity of gas mixture. Reacting gas: 3% C_3H_8 + air.

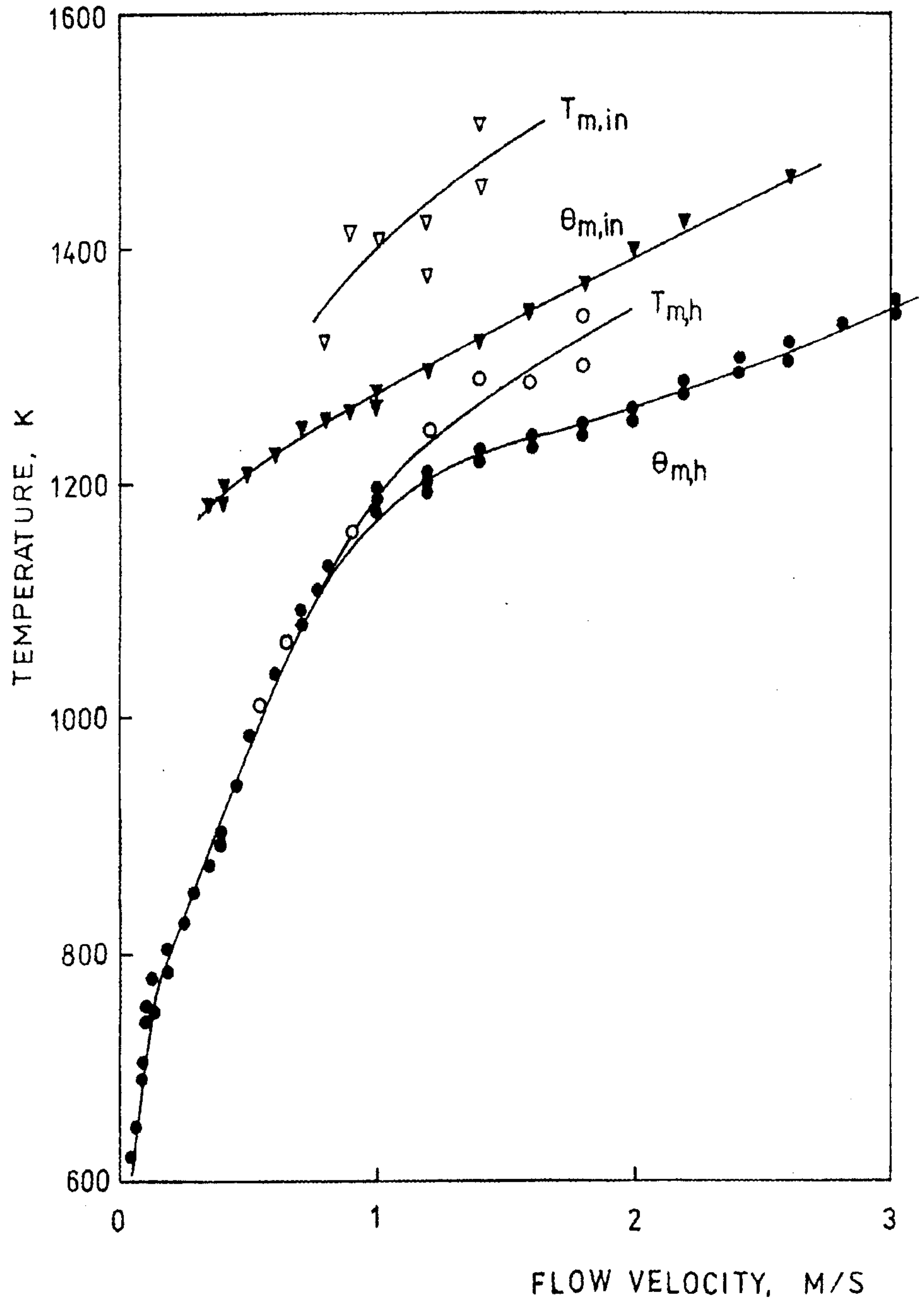

Fig. 5. The dependencies of maximum gas (T_m) and porous medium (θ_m) temperatures in the waves with homogeneous-heterogeneous (circle symbols) and homogeneous (triangular symbols) reactions on filtration velocity of gas mixture. Reacting gas: 65% H_2 + air.

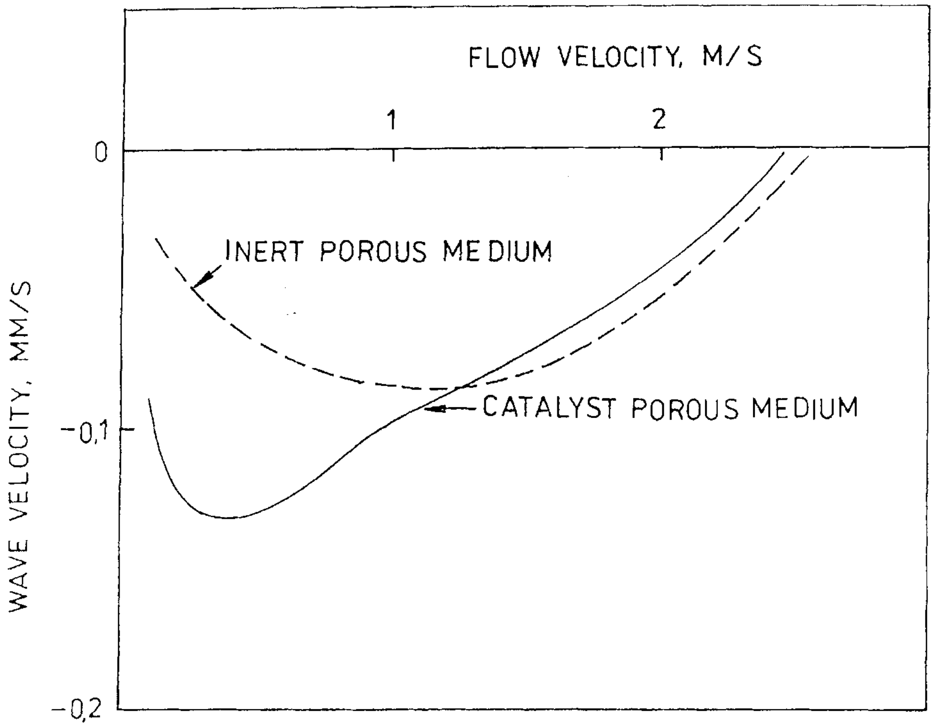

Fig. 6. Theoretical dependencies of wave velocity in inert and catalytic active porous media on filtration velocity of reacting gas.

Table 1.

WAVE VELOCITIES IN CATALYTIC AND INERT POROUS MEDIA

V_0 = 0,5 M/S

	% C_3H_8 IN AIR MIXTURE						
	2,0	2,5	3,0	3,25	3,5	4,0	4,5
$U_h \cdot 10^2$ MM/S	—	1,5	3,5	5,0	7,0	10,0	9,0
$U_{in} \cdot 10^2$ MM/S	—	—	—	1,0	3,0	4,0	3,0

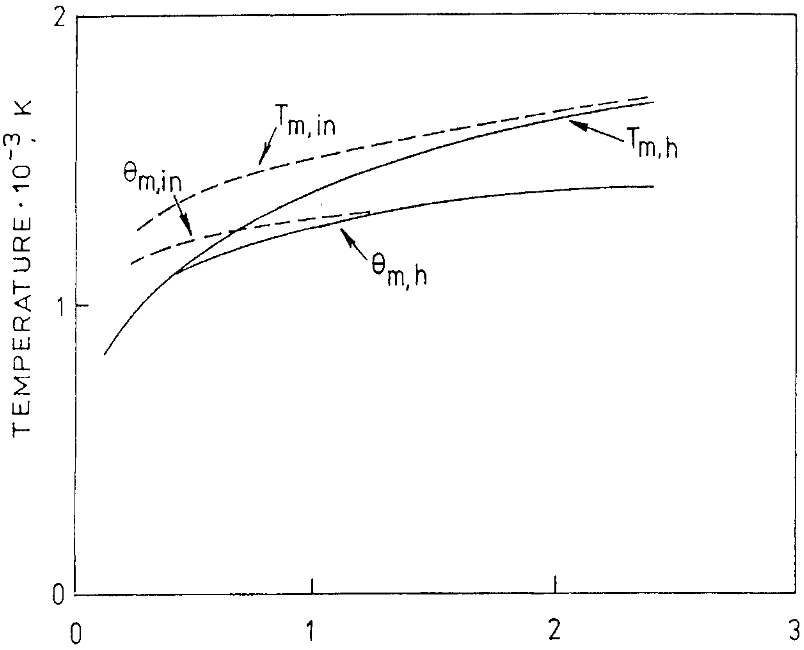

Fig. 7. Theoretical dependencies of maximum gas (T_m) and porous medium (θ_m) temperatures in the waves with homogeneous-heterogeneous (lines) and homogeneous (dashed lines) reactions on filtration velocity of gas.

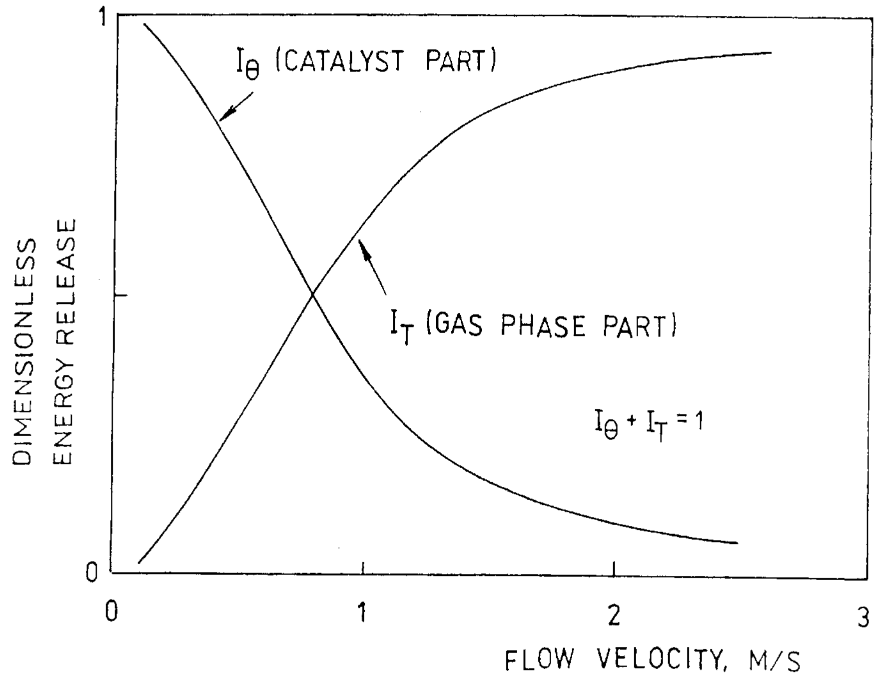

Fig. 8. The dependencies of dimensionless parts of heat release in the volume (I_T) and on catalytic surface (I_θ) on filtration velocity of reacting gas.