

Filtrational combustion of gases. Present state of affairs and prospects

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Abstract - The review considers the problem of gas combustion in inert porous media. It includes the classification of steady-state combustion regimes, the structure and characteristics of thermal waves, the interaction between physico-chemical elementary processes in combustion waves, the mechanisms of reaction zone propagation. The ways of the development of the theory of this combustion type and its role in the understanding of similar combustion processes are discussed.

INTRODUCTION

During the past decade, a theory of filtrational combustion (FC), involving a new class of wave processes with exothermic chemical transformation during fluid motion in porous media, has been developed. The existence of two or three phases, a strong interphase interaction in the combustion zone and a relative motion of phases define many unique properties of FC that differ from those of conventional flames in homogeneous, gaseous and condensed systems.

These developments stimulate applications in such diverse processes as blast furnace production of steel, agglomeration of ores, in situ combustion, smoldering combustion, self-propagating high-temperature synthesis, and catalytic combustion. Until now the theory of these processes has been developed independently. Only recently, their community of ideas has been realized and the first attempts have been made to construct generalized models (ref.1).

Many FC forms and regimes are known. They depend upon the nature of porous medium and filtrating agent (fuel, oxidizer or inert material), the type of reagent filtration, whether there are phase transitions in the combustion zone and where reaction is localized, in gas or at the surface of condensed phase, and other characteristics. So far there is no nomenclature for many of the FC forms and their classification has been according to expediency.

One natural classification is according to the type of chemical reaction in the combustion wave. The present report is limited to the relatively simply, yet informative, homogeneous gas-phase reaction in a inert porous medium (filtrational gas combustion, FGC).

Although studies of FGC have been limited, a few steady-state combustion regimes have been identified over a wide range of propagation velocities of the thermal waves: low-velocity regime (LVR, $u \simeq 10^{-4} m/s$), high velocity regime (HVR), $S \simeq 10 m/s$, sound velocity regime (SVR), $S \simeq 10^2 m/s$, low-velocity detonation (LVD), $D = 800 - 1500 m/s$, and normal detonation with losses (ND), $D = 1500 - 2000 m/s$ (ref.2).

On the one hand, the functional analysis of elementary physico-chemical processes in a steady-state combustion wave allows one to understand the mechanism of chemical reaction transfer in wave motion. On the other hand, the velocity and structural characteristics of a steady-state wave front are necessary to describe the numerous accompanying non-steady-state regimes. In this report, the consideration of FGC problems concerns only the steady-state processes and, moreover, the velocity and structural aspects. The purpose of the report is to demonstrate the general and particular properties of combustion waves and to integrate the bonds between various filtrational and related combustion processes.

COMBUSTION IN LOW VELOCITY REGIME

A thermal wave in LVR can develop in a tube filled with an inert porous medium, through which a homogeneous mixture is flowing (refs.3-5). The steady-state wave velocity depends on the properties of both the gases and the porous medium (Fig.1). Depending on the flow, or filtrational, velocity, the wave can move either against, or with the flow or can become standing wave. The reverse motion of combustion wave without any changes in the gas flow direction is one of the most vivid and complex effects of FGC.

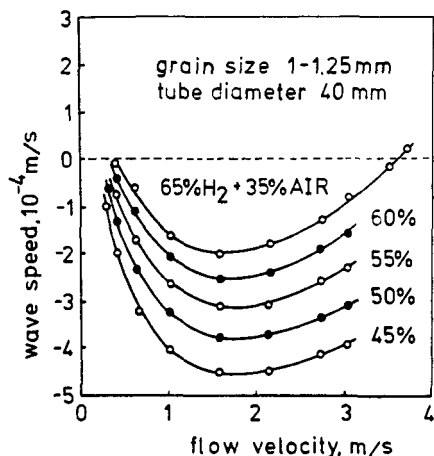


Fig.1 Dependence of wave velocity u on flow velocity v_0 for air-hydrogen mixtures.

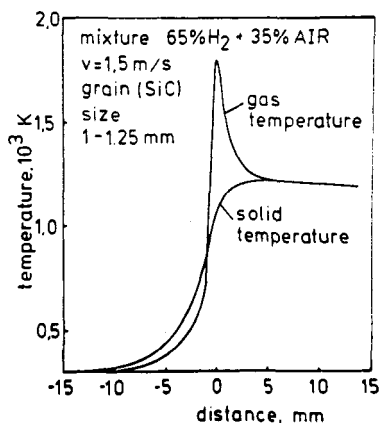


Fig.2 Temperature profiles in combustion wave in gas and porous medium.

Another characteristic effect of FGC is temperature heterogeneity in the burning wave, i.e. a considerable difference in phase temperature (Fig.2). In the case of a moderate internal heat exchange, in the temperature evolution of a moving gas element there is a moment when the rate of heat release in the gas phase becomes higher than the rate of heat supply from the solid phase. The gas-phase reaction proceeds as in a thermal explosion. In the reaction zone an anomalous high temperature peak occurs.

In the case of an intense interphase heat exchange the rate of the chemical reaction is limited by the heat exchange and the temperature of a gas element cannot strongly differ from the solid phase temperature. Due to comparatively low combustion temperatures upon the counter motion of the wave, wide zones of chemical reactions are possible (refs.6,7). In the limiting case of a strong interphase heat exchange, a two-temperature wave structure degenerates into a one-temperature structure. On the other hand, when the heat exchange is rather weak, LVR converts into HVR (see below).

The other important characteristic of LVR is heat recuperation in the combustion zone, as the source of the enthalpy excess of the reacting gas. Due to a high thermal conductivity, the heat is effectively transferred along the solid porous medium to the preheating zone in which, via the interphase heat exchange, it passes from the solid to the gas (Fig.2, $-4 < x < -1$ mm). Further, the heat is transferred via convection to the zone of chemical reaction and thermal relaxation. Here it returns to the solid (Fig.2, $0 < x < 4$ mm). Recuperation leads to enthalpy excess and higher temperatures and rates of chemical reaction that occur in the corresponding homogeneous flame.

The rearrangement of internal processes relative to the standing wave is related to the wave motion and, especially, to the change in its direction. The recuperation cycle gives rise to a new convective heat flow due to a relative motion of a solid phase. As a result, upon change from the up to the down stream, the efficiency of a recuperation cycle increases. This is the difference between moving and stabilized waves. The latter have been considered in detail by T.Takeno et al. (refs.8-10).

Compared to homogeneous combustion, in LVR the combustion temperature is determined not only by thermo dynamic gas properties but also by the properties of the porous medium and the filtration velocity. This reflects the fact that the LVR wave is not only the heat generator that transforms the chemical energy into the thermal one, but also the distributor of the last between the phases. Owing to such an unusual function of the combustion wave, its equilibrium temperature can be lower, higher, or equal to the temperature of homogeneous gas flame.

The main rules of steady-state waves have been established: the parameters upon which wave velocity depends (ref.3); the thermal structure has been studied (ref.5); the mechanisms of flame propagation and quenching have been determined (ref.11). Moving (refs.6, 12) and stabilized (refs.13-17) waves have been described mathematically. Such questions as flame front stability (ref.18), the chemical structure of waves (refs.19,20), the nature of waves with phase transitions (ref.21) and homogeneous and heterogeneous reactions (ref.22) have been less well studied. Many aspects of LVR, i.g., non-one-dimensional and non-steady-state waves, LVR with accompanying pressure wave, etc. have not been studied.

COMBUSTION IN HIGH VELOCITY REGIME

With increasing pore size there is a transition from LVR into HVR (ref.2). Transition proceeds by a jump, with wave velocity increases of 3 to 4 orders of magnitude. The basic change in the mechanism of combustion wave propagation is related to the reduced thermal coupling of gas with solid in the reaction zone. In this case, there is no need in the continuous supply of chemical energy to the reaction zone via the filtrational flow. For combustion, the energy of the stationary gas in a single pore is sufficient. Therefore, HVR can be excited in a semiclosed tube containing a porous medium and a combustible mixture (ref.23,24). When the mixture is ignited at the open end, the flame then moves uniformly under constant pressure after some time. The typical dependencies of flame propagation velocity on the initial pressure and mixture composition in various porous media - polyurethane foam (PF), steel balls (SB) and foil (FPM) are depicted in Figs.3 and 4.

Thermal wave structure can be presented as a pulse of some average temperature in the gas and a smooth small rise of temperature in the porous medium. The pulse front is caused by heat release, and the pulse back is due to the completion of chemical reaction and gas cooling as a result of thermal interaction with the porous medium.

Experimental and theoretical estimations suggest the chemical reaction is localized in separate hot centers, which alternate with the local volumes of the cool products (ref.26). In a long combustion zone (a few cm in normal conditions), the temperature and concentration fields are rather inhomogeneous. The total combustion time (the residence time of centres of chemical reaction in the combustion zone) and the duration of a thermal pulse are of the same order of magnitude. This

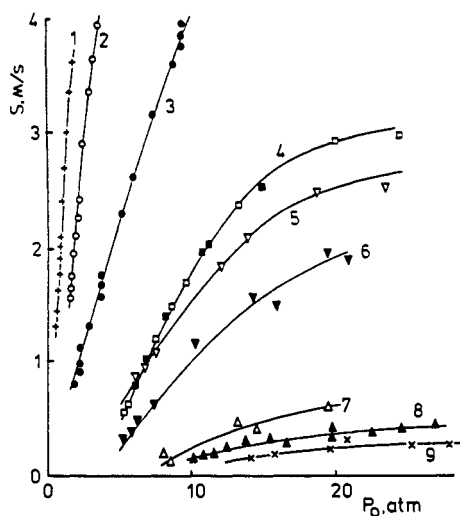


Fig.3 Dependence of wave velocity

S on initial pressure in a porous media and air - methane mixtures:

- 1 - 9.5%, FMP $d = 4.5$ mm; 2 - 9.5%, PF $d = 2.8$ mm; 3 - 9.5% SB $d = 2.2$ mm;
- 4 - \square , 8.5%; \blacksquare , 9% SB $d = 1.15$ mm;
- 5 - 9.5%; 6 - 8%; 7 - 7.25%; 8 - 7%;
- 9 - 6.8% SB $d = 1.15$ mm (ref.25).

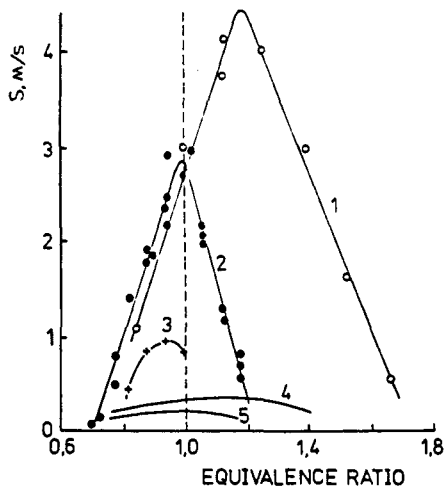


Fig.4 Dependence of wave velocity S in porous media (1 - 3) and burning velocity (4, 5) for air mixture of propane and methane on equivalence ratio: 1 - propane, $p_0 = 0.1$ MPa, FPM $d = 4.0$ mm; 2 - methane, $p_0 = 0.6$ MPa, SB $d = 2.2$ mm; 3 - methane, $p_0 = 0.6$ MPa, SB $d = 1.15$ mm; 4 - propane, $p_0 = 0.1$ MPa; 5 - methane, $p_0 = 0.6$ MPa (ref.25).

structure depends on the processes of combustion and cooling under conditions of strong turbulence in the porous medium. However, the characteristics of turbulence, the sources of its appearance and the conditions of chemical reaction, differ here, probably, from those in usual turbulent flames. In the porous medium, the turbulence can arise from the directed filtrational gas flows, due to a pulsatory character of combustion itself, as a result of the local processes of gas burning and expansion in the combustion zone. The narrowing and broadening of pore channels determine the effects of high-speed pulsed jets and the quenching and reinitiation of the chemical reaction.

The turbulent character of combustion and the possibility of thermal quenching of flame in the most rapid pulsations due to interphase heat exchange have been assumed in elementary model of flame propagation in HVR (ref.25). According to this, the flame velocity obeys the formula: $Re = k Z^{-3/2} Pe^3$, where $Re = (S - S_u) d / \nu$, $Pe = S_u d / \kappa$, $Z = E(T_b - T_0) / (RT_b^2)$ are the Reynolds, Pelet, and Zeldovich numbers, respectively. It is seen that the velocity of flame S depends not only on the gas characteristics (burning velocity S_u , the coefficient of molecule exchange ν , κ , but also on the characteristics of porous medium, i.e. the characteristic pore size d , and is independent of its thermophysical properties. This model, however, neglects Lewis number effects. They manifest as a shift of cape points on the curves of velocity in the region of rich propane and lean hydrogen flames (Fig.4). These effects probably depend upon of selective diffusion (ref.28).

Thus, compared to LVR, the more important factor in HVR is not thermal but aerodynamic interphase interaction and its consequences. Thermal interaction has actually no direct effect on the chemical reaction except for the most rapid pulsations. Therefore, in the zone of chemical reaction, there is no recuperation of heat that is typical of LVR.

In HVR the role of filtrational flow is different and it can be absent. In other cases, it can be caused by either external forces or combustion itself as e.g. upon non-steady-state combustion in a closed vessel (ref.23). The flow not only transfers the combustion zone in the porous medium but also increases the turbulent burning velocity by increasing turbulent intensity. However, the filtrational flow is neither the only nor the main reason for turbulence generation. On the other hand, combustion in HVR differs from turbulent combustion in free space. This, particularly, is verified by the above strong dependence of wave velocity of the burning one ($S \sim S_u^3$).

The above mentioned brief data on HVR demonstrate a rather interesting phenomenon of combustion, many aspects of which remain to be studied. Studies of non-steady-state flames, effects of Lewis number, combustion in closed vessels (ref.2), thermal explosion (ref.27) and the configuration and nature of the boundaries of the regime (ref.28) have been only started. The mathematical simulation of phenomena, the conditions of regime initiation, the behavior and regularities of flames at the boundaries of structurally different media have not been studied.

COMBUSTION OF GAS IN THE SOUND VELOCITY REGIME

The theory of filtration (ref.29,30) suggest a pressure wave (filtration zone), formed under the action of the forces of interphase friction, must exist in front of the combustion front in the porous medium. In HVR, due to the relatively low velocities, the zone of filtration must be large compared with that of combustion and hence the process of combustion can be considered to be isobaric.

Another situation holds for velocities of the order of that of sound ($10^2 m/s$). In this case, the filtration zone can be compared to the combustion one, and chemical reaction proceeds at high pressures and temperatures. This regime of steady-state combustion propagating in the range of sound velocities (SVR) was, probably, first described in papers (refs.31,32). The SVR regime is commonly entered from that of HVR when the wave velocity in HVR increases, e.g. with increases in initial pressure or burning velocity or pore size (Figs.5,6).

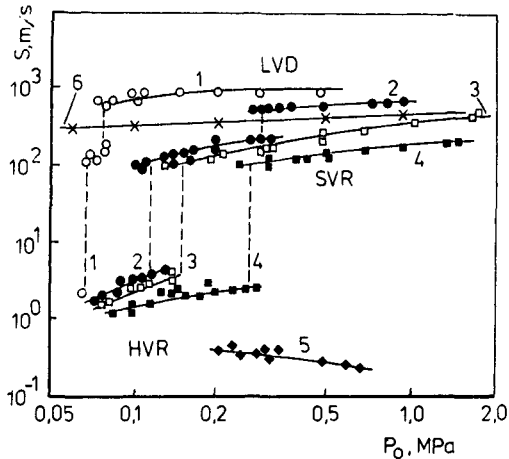


Fig.5 Dependence of wave velocity S on initial pressure for air - hydrogen mixtures. Porous medium - fraction of steel shot 2.6-3.6 mm. Content of H_2 in mixture (%): 1 - 20, 2 - 18, 3 - 17, 4 - 15, 5 - 10. Dotted line - sound velocity in porous medium (ref.32).

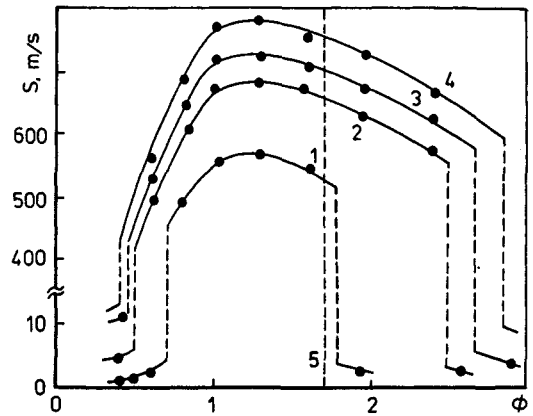


Fig.6 Dependence of wave velocity S on equivalence ratio Φ for air-hydrogen mixtures. Porous medium - polyethylene granules, mean size of pore channels - 1 mm. Initial pressure (MPa): 1 - 0.1; 2 - 0.2; 3 - 0.3; 4 - 0.5 (ref.28).

The regime changes, as a rule, by jumps. A pressure peak in the combustion zone with a smooth rise and drop of pressure is typical of SVR. The maximum pressure does not exceed the final pressure attained in a gas explosion in a closed vessel. With air-hydrogen mixtures, combustion is substantially incomplete.

Experiments on flame propagation in tubes with periodic obstacles (rough tubes, channels with repeated narrowings and obstacles) are related to the phenomena under discussion (refs.33-36). Like porous media, these systems have an increased hydraulic resistance characterized by a blockage relationship $BR = 1 - (d_2/d_1)$ where d_1 and d_2 are the diameters of tubes and orifices, respectively.

Fig.7 demonstrates the steady-state velocities of flame propagation of air-propane mixtures in tubes with periodical obstacles. It is seen that, as in porous media, regimes are possible with velocities of about $10^2 m/s$ and transitions occur by jumps from one velocity regime to another, including supersonic regimes with velocities of $10^3 m/s$. Fig.8 depicts pressure distribution during flame propagation in a system of 19 connected vessels. In the region of steady-state propagation ($t = 25 - 27 ms$) the pressure wave is observed to have the form of the "thermal wave" with a smooth rise of pressure. Pressure drop behind the front is small due to the relatively weak cooling of combustion products.

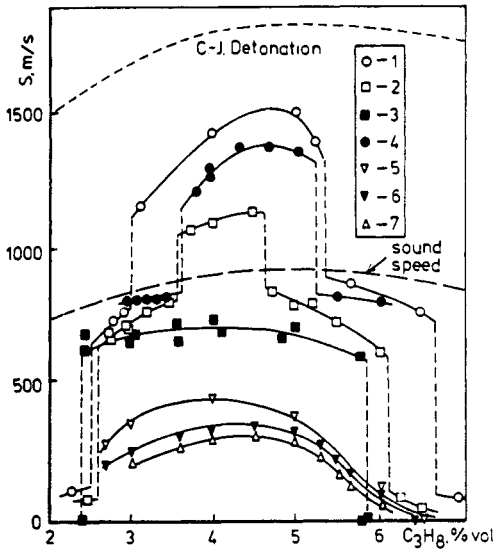


Fig.7 Dependence of wave velocity S on equivalence ratio for air - propane mixtures in tubes with periodic obstacles: 1 - $d_1 = 30$ cm, $BR = 0.43$; 2 - $d_1 = 15$ cm, $BR = 0.5$ (ref.33); 3 - $d_1 = 5$ cm, $BR = 0.43$; 4 - $d_1 = 15$ cm, $BR = 0.39$ (ref.34); 5 - $d_1 = 11.5$ cm, $BR = 0.91$, $p_0 = 0.1$ MPa; 6 - $d_1 = 11.5$ cm, $BR = 0.96$, $p_0 = 0.2$ MPa; 7 - $d_1 = 11.5$ cm, $BR = 0.96$, $p_0 = 0.1$ MPa (ref.35).

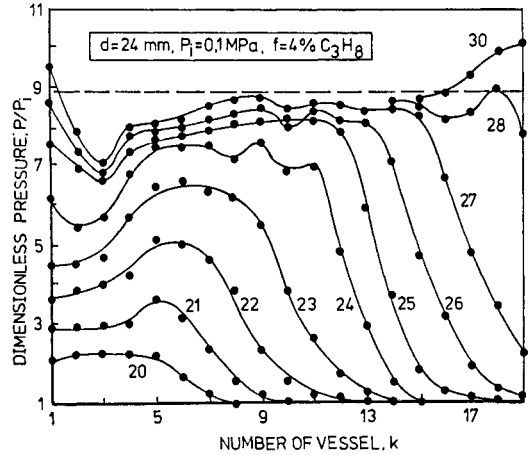


Fig.8 Pressure distribution for flame propagation in a 19 chamber system of connected vessels. Number on curves denotes time in ms from moment of mixture ignition. Dotted line maximum pressure upon combustion in closed vessel (ref.35).

The limits of the regime of steady-state SVR hardly have been studied. In (ref.29) it is shown that in SVR not only the boundaries with complete flame quenching can exist, but also those determined by transition to HVR. If the parameters leading to increases S in SVR are changed, transitions to supersonic combustion regimes are possible (Fig.5).

Turbulent motion of gas and its compressibility suggest that the conductive-convective mechanism of reaction transfer in SVR can be accompanied by an alternative inductive mechanism that involves the quenching of chemical reaction followed by its reinitiation (self-ignition) upon mixing of unburnt and burnt gas. In this case two facts should be mentioned. First, unlike in HVR, in SVR an aerodynamic quenching with the characteristic time d/u where u , d are the pulsation velocity and the pore size, respectively, is more probable than convective quenching. Second, heating and self-ignition of unburnt gas with a short delay upon its compression are impossible and this excludes direct initiation of the chemical reaction.

Compared to LVR and HVR, SVR has been less well studied. A few papers on the pressure wave structure (ref.32), velocity characteristics (ref.29,32), propagation limits (ref.29), mathematical simulation (ref.36) only outline the phenomena. Systematic studies of all aspects of this combustion regime are necessary, including the establishment of the parameters that determine the velocity, structural characteristics and regularities, limit nature, etc.

On the other hand, SVR is of special interest in combustion theory as an example of strong turbulent combustion, involving convective and inductive mechanisms of reaction transfer in the cases of both filtration with the highest gas velocities and of the intermediate combustion regime between deflagration and detonation.

LOW VELOCITY DETONATION

When the velocity of a shock wave, propagating in a porous medium is lower than that usually associated with detonation and the temperature of unburnt gas behind the shock wave is insufficient for direct initiation of the chemical reaction with a short delay, then steady-state combustion regimes are possible with local repeated initiation of the reaction upon interaction of the shock wave with the porous medium. LVD first was observed by K.I.Shchelkin in rough tubes (ref.37). Further, LVD with the velocities of 0.5 to 0.7 D_0 , where D_0 is the detonation velocity in a smooth tube, was recorded in the porous medium (ref.38). Using the method of initiation of detonation by the shock wave of variable intensity (ref.39) it was shown that gas ignition follows the scheme of Ya.B.Zeldovich, upon reflection of the shock wave from elements of the porous medium.

In Refs.40,41 it was shown that the LVD velocity increases with increasing initial pressure, pore size, and mixture reactivity. It is unclear yet if there is an explicit boundary between LVD and normal detonation, or whether the transition to ND is always smooth. As for the lower boundary, three situations are possible, namely the transition to SVR, HVR, or cessation of the detonation. In this case, the transition to SVR or HVR has, as a rule, a threshold hysteresis character (Fig.5).

A characteristic feature of LVD is the inductive mechanism of reaction transfer in the detonation wave. However, due to the low velocity, the mechanism of normal cellular detonation with cross ignition waves is not realized in LVD. The reaction is transferred forward by the leaders of "hot-spots" followed by a turbulent front. Gas self-ignition in "hot-spots" can be caused by the reflection of shock waves from the elements of the porous medium, and the interaction of shock waves with each other, and the vortex mixing of unburnt and burnt gas. This conclusion is based on model experiments with periodic obstacles (ref.43,44). However, the explanations of these phenomena in such systems and in porous media are not fully adequate. Novel approaches are needed to establish the dominating mechanisms of gas ignition in LVD.

Although the general reason for the lowering of velocity in LVD, the momentum and energy losses, is long realized, so far there is no understanding in the character of heat, gas-dynamic, and chemical structure of the wave. If the steady-state velocity results from the equilibrium of elementary physico-chemical processes, what are these processes and what is their interconnection? In what cases is this balance violated and in what way is this related to the boundaries of the LVD regime? The answers to these questions are important not only for better understanding of LVD in inert porous media, but also for that of the regimes of non-ideal detonation in more complex heterogeneous systems, liquid aerosols, combustible dusts, foams, etc.(ref.45).

FURTHER DEVELOPMENT OF FILTRATIONAL COMBUSTION OF GASES

One useful direction of FGC development would be a deeper study of the above-mentioned regimes in a porous media, as discussed above. Of interest are studies that connect FGC with other related processes, namely, catalytic combustion waves, combustion of porous and dispersed media, filtrational combustion of liquids, etc.

1. One of the basic questions is the existence and behavior of hybrid waves, i.e. FGC involving parallel homogeneous and heterogeneous chemical reactions. The regularities and specific features of such waves can be revealed in comparative experiments with one and the same porous medium, but with different surface states. The surface can be modified by applying a catalyst (ref.22).

Fig.9 gives the dependencies of wave velocity through catalytically active and inert porous media on the velocity of filtration for a 5% air-hydrogen mixture. The strongest difference in the velocities is observed for low flow velocities. There is a velocity range within which only catalytic waves can exist. It is shown (ref.46) that, as in LVR, the phase temperatures are quite different and the difference decreases with decreasing flow velocity due to the decrease of the contribution of volumetric reaction to the total balance of heat release (Fig.10). It is interesting, that at the same flow velocity, the equilibrium temperature in the inert medium is higher than in the catalytic one. This is caused by

the difference in wave velocities and the dependence of the equilibrium temperature on the wave velocity. These and other regularities of hybrid waves are satisfactorily described by a mathematical model that takes into account homogeneous and heterogeneous reactions (ref.46).

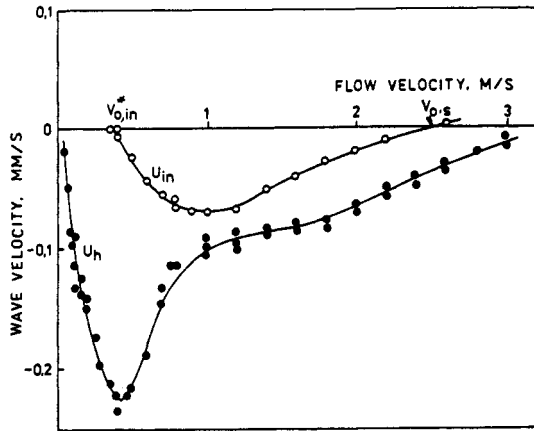


Fig.9 Dependencies of wave velocities in active U_h and inert U_{in} porous media on flow velocity. Catalytic medium - $CuCr_2O_4/\alpha - Al_2O_3$. Inert medium - $\alpha - Al_2O_3$. Granule size $\alpha - Al_2O_3$ 1 - 1.6 mm.

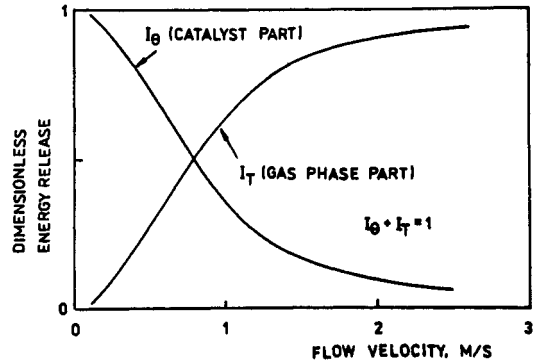


Fig.10 Fractions of volumetric and surface reactions in the total balance of heat release depending on flow velocity.

2. In the processes of filtrational combustion, the filtrational agent can be either gas and liquid. Liquids have a higher densities, heat capacities per unit volume and higher coefficients of interphase heat transfer. In addition, the liquid creates a number of new factors, such as phase transitions, surface tension, and gravity.

Recently, the possibility has been demonstrated of wave propagation in filtrational combustion of a liquid, with gas-phase chemical reaction (ref.21). Fig.11 shows the dependence of wave velocity u and the rate of hydrazine decomposition S_u on the filtrational velocity v_0 . The dependence of $u(v_0)$ displays a U-like shape, as for FGC. Increase of v_0 leads to intensification of combustion. With decrease of v_0 , the rate of N_2H_4 decomposition decreases, and for some critical value of v_0^* , combustion ceases.

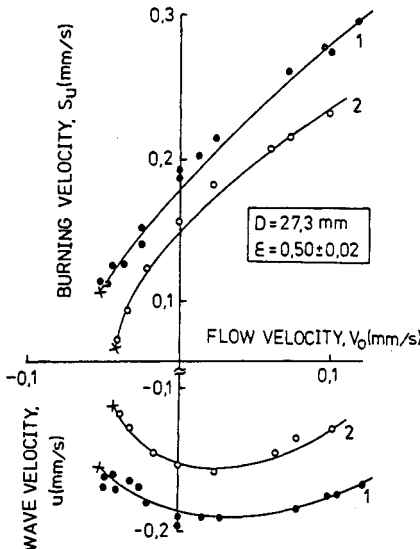


Fig.11 Dependence of wave velocity u and the rate of hydrazine S_u decomposition in inert porous medium on flow velocity v_0 . Porous medium - fraction of SiC 1 - 1.2 mm (1) and 2.0 - 2.5 mm (2).

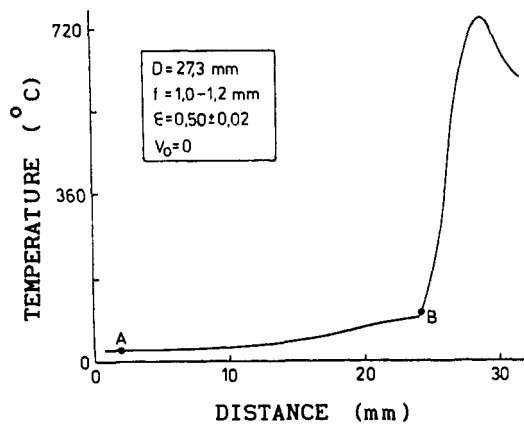


Fig.12 Temperature profile upon filtrational decomposition of hydrazine.

Of special interest is the wave structure. In a space wave structure there is a long region of two-phase flow (gas-liquid zone). It corresponds to the anomalously wide zone of preheating (AB, Fig.12), determined by capillary filtration and convective heat-and-mass exchange of burnt gas and hydrazine vapors deep into the zone (ref.47).

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