

THE INVESTIGATION OF THE INFLUENCE OF OXYGEN CONTENT IN THE ATMOSPHERE ON COMBUSTION CHARACTERISTICS OF MIXTURES HYDROGEN – OXIDIZER – DILUENT

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ABSTRACT

An experimental investigation of the characteristics of combustion of rich near-limit gaseous mixtures hydrogen – oxidizer (mixture of oxygen and nitrogen) – diluent (trifluoromethane CF_3H , pentafluoroethane $\text{C}_2\text{F}_5\text{H}$, perfluorobutane C_4F_{10} , AKM agent) in a closed vessel of volume 4.2 dm^3 at oxygen contents in the oxidizer 15, 20.6 and 25% (vol.) was executed. Dependences of a maximum explosion pressure Δp_{max} and a maximum explosion pressure rise rate $(dp/dt)_{\text{max}}$ on a diluent concentration C_{rel} have been determined. It was found that for the most part of the investigated gaseous mixtures the maxima in the dependences of Δp_{max} on C_{rel} take place, but in the case of the dependences of $(dp/dt)_{\text{max}}$ on C_{rel} the maxima take place for the pentafluoroethane $\text{C}_2\text{F}_5\text{H}$ only. A qualitative interpretation of the observed results was given, which is based on two effects: a) an active participation of the diluents in a chemical conversion in a flame front (in many cases with an additional heat release); and b) an existence of two different kinetical regimes of chain-branching combustion processes. It was found that C_4F_{10} and AKM are the most effective inhibitors in relation to a flame propagation in rich near-limit mixtures hydrogen – oxidizer – diluent.

KEYWORDS: Hydrogen, Inhibition, Characteristics of combustion

INTRODUCTION

An investigation of an inertization of gaseous mixtures with an oxygen content different from that in air is interesting from a viewpoint of fire and explosion safety of technological equipment. The studies¹⁻²³ were dedicated to a decision of this problem, and their brief review was given^{1,2}. Hydrogen combustion and inertization of hydrogen contained mixtures that are very interesting because of the necessity of ensuring hydrogen safety^{1,2,5,6,9,12,13,20-23}. These studies were aimed mainly on a determination of flammability limits in mixtures hydrogen – oxidizer – diluent. But only some part of these papers describes combustion characteristics of gaseous mixtures in a closed vessel such as a maximum explosion pressure, a maximum explosion pressure rise rate, and a laminar burning velocity^{12,13,20-23}. It was found that dependences of the maximum explosion pressure Δp_{max} and the maximum explosion pressure rise rate $(dp/dt)_{\text{max}}$ on a diluent concentration C_d at a constant hydrogen content in a mixture are non – monotone. An availability of maxima in dependences of Δp_{max} and $(dp/dt)_{\text{max}}$ on C_d was explained by an active participation of fluorinated agents in combustion processes with an additional heat release in a flame front. Qualitatively analogous results were obtained^{24,25}. Combustion of mixtures $\text{H}_2 + \text{O}_2 + \text{C}_2\text{F}_6$ was studied²⁴. It was found that neither $\text{C}_2\text{F}_6 + \text{O}_2$ mixtures nor $\text{C}_2\text{F}_6 + \text{H}_2$ mixtures can support a flame propagation, but mixtures $\text{H}_2 + \text{O}_2 + \text{C}_2\text{F}_6$ can burn at some concentrations of the initial components with a formation of CF_4 , COF_2 and other fluorinated products (that is combustion of $\text{H}_2 + \text{O}_2$ mixture induced a chemical conversion of C_2F_6). It was found²⁵ that a flame propagation in mixtures $\text{C}_2\text{H}_4 + \text{air}$ in a tube also induces chemical reactions of fluorinated agents with an additional heat release. An additional heat release caused by a chemical conversion of fluorinated inhibitors was observed also in diffusion flames²⁶. In some cases fluorinated hydrocarbons can form flammable gaseous mixtures with air or oxygen^{27,28}, and this fact confirms a possibility of the additional heat release during chemical conversion of these substances in a flame front.

This work is aimed on an experimental investigation of an influence of diluents of various chemical natures (mainly fluorinated hydrocarbons) on combustion characteristics of mixtures hydrogen-oxidizer (mixture of nitrogen and oxygen) at various oxygen concentrations in the oxidizer. Rich near-limit mixtures were considered (that is the mixtures, in which hydrogen concentration is lower than the upper flammability limit at a given diluent content not more than 1% (vol.)).

This choice of the object for investigation was made due to two reasons. Firstly, according to existing theoretical and experimental studies²⁹⁻³¹ an adiabatic flame temperature is approximately the same for various near-limit flames (that is the maximum explosion pressure for these flames propagating in closed vessels should be also approximately the same). It is interesting to check this empirical regularity for the case of inhibited flames. Secondly, according to theoretical and experimental studies³²⁻³⁴ an abrupt decrease of the maximum explosion pressure occurs at some diluent concentration, which can be rather far from a minimum inertization concentration (the concentration at a peak point of a flammability curve). In this study we check also this regularity for rich near limit flames of hydrogen – oxidizer – diluent. We consider only rich mixtures, because for lean hydrogen contained mixtures with H₂ concentration less than 8% (vol.), the so called bubble regime of flame propagation is typical³⁵⁻³⁷. In this regime a pressure increase at combustion in a closed vessel can be negligible, and an investigation of an influence of diluents on the maximum explosion pressure and the maximum explosion pressure rise rate is not interesting.

EXPERIMENTAL

Experiments were carried out on a set-up “Variant” described in^{38,39}. This set-up has a spherical reaction vessel of volume 4.2 dm³ (internal diameter 0.2 m). Hydrogen was used as a fuel, and pentafluoroethane C₂F₅H, trifluoromethane CF₃H, perfluorobutane C₄F₁₀, and AKM agent were studied as diluents. The AKM agent is the mixture of hydrocarbons with a C=C bond proposed in the Institute of Structural Macrokinetics of Russian Academy of Sciences. The main part of the AKM agent (not less than 50 % (vol.)) is propylene C₃H₆, the rest are propane and butane. The gaseous mixtures were prepared immediately in the reaction vessel in the following way. Required quantities of hydrogen and diluent were introduced into the preliminary evacuated up to a residual pressure 0.5 kPa reaction vessel by partial pressures. If air is an oxidizer, then it was introduced up to atmospheric pressure. If the oxidizer is an oxygen-nitrogen mixture with an oxygen content different from that in air, then a required quantity of additional nitrogen (nitrogen enriched atmosphere) or oxygen (oxygen enriched atmosphere) was added and finally air up to atmospheric pressure. After a time interval 5 min (required for a qualitative mixing) an ignition source was initiated.

Air and oxygen-nitrogen mixtures with O₂ content 15 and 25 % (vol.) were used as oxidizers. An ignition was made in the centre of the reaction vessel by means of an electrical spark with energy near 1 J. Pressure in the reaction vessel at a flame propagation was measured by the pressure detector “Karat – DI” with a time constant 10⁻³ s. Signal from the pressure detector was supplied to a computer. Experiments were carried out at room temperature and atmospheric pressure. The maximum explosion pressure Δp_{\max} and the maximum pressure rise rate $(dp/dt)_{\max}$ were determined from measured pressure-time curves. Rich near-limit mixtures of hydrogen – (O₂ + N₂) – diluent were investigated, in which the hydrogen concentration differs from that at an upper flammability limit at a given diluent content non larger than 1 % (vol.). The flammability curves used for the experiments are shown in Figs. 1 to 4^{1,2}.

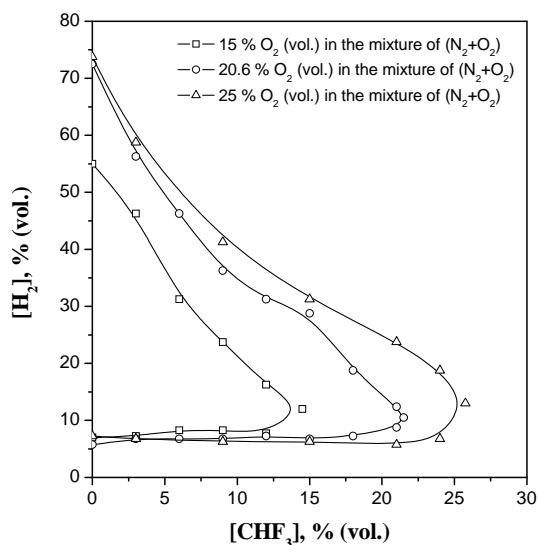


FIGURE 1. Flammability limits in mixtures $H_2 - (O_2 - N_2) - CHF_3$ at the oxygen content in the oxidizer 15 (\square), 20.6 (\circ), and 25 (Δ) % (vol.) (data [1,2]).

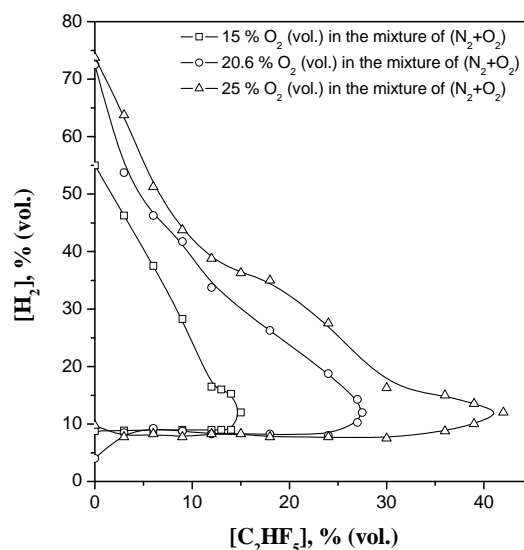


FIGURE 2. Flammability limits in mixtures $H_2 - (O_2 - N_2) - C_2F_5H$ at the oxygen content in the oxidizer 15 (\square), 20.6 (\circ), and 25 (Δ) % (vol.) (data [1,2]).

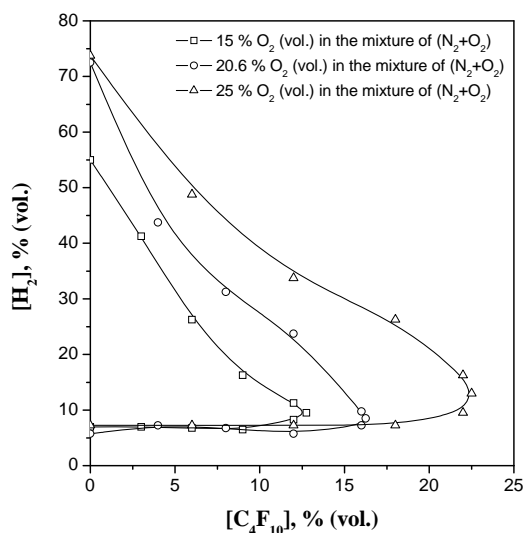


FIGURE 3. Flammability limits in mixtures $H_2 - (O_2 - N_2) - C_4F_{10}$ at the oxygen content in the oxidizer 15 (\square), 20.6 (\circ), and 25 (Δ) % (vol.) (data [1,2]).

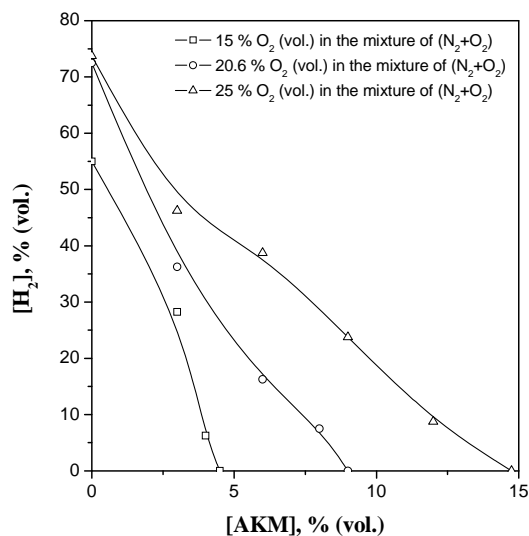


FIGURE 4. Upper flammability limits in mixtures $H_2 - (O_2 - N_2) - AKM$ at the oxygen content in the oxidizer 15 (\square), 20.6 (\circ), and 25 (Δ) % (vol.) (data [1,2]).

RESULTS AND DISCUSSION

Results of a determination of the maximum explosion pressure Δp_{\max} are presented in Figs. 5 to 7, and of the maximum explosion pressure rise rate $(dp/dt)_{\max}$ in Figs. 8 to 10.

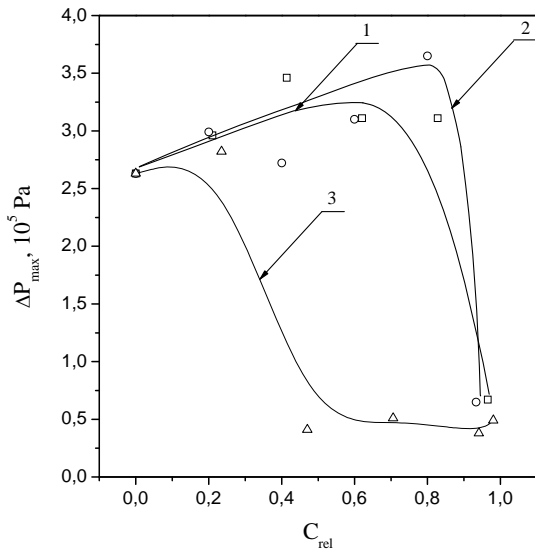


FIGURE 5. Dependence of the maximum explosion pressure on the relative diluent concentration at the oxygen content in the oxidizer 15 % (vol.). Diluents: 1 (\square) – CF_3H ; 2 (\circ) – $\text{C}_2\text{F}_5\text{H}$; 3 (Δ) – C_4F_{10} .

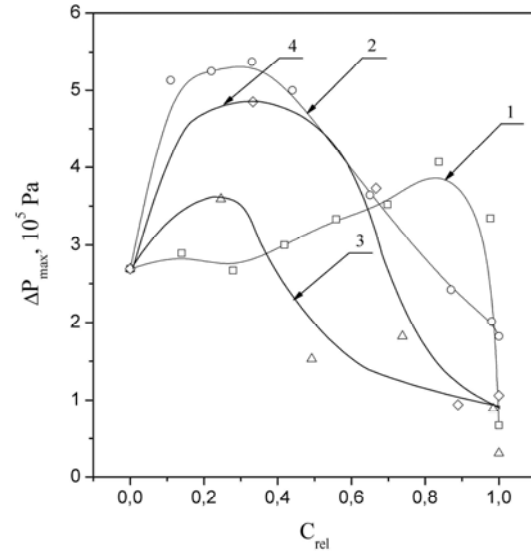


FIGURE 6. Dependence of the maximum explosion pressure on the relative diluent concentration at the oxygen content in the oxidizer 20.6 % (vol.). Diluents: 1 (\square) – CF_3H ; 2 (\circ) – $\text{C}_2\text{F}_5\text{H}$; 3 (Δ) – C_4F_{10} ; 4 (\diamond) – AKM.

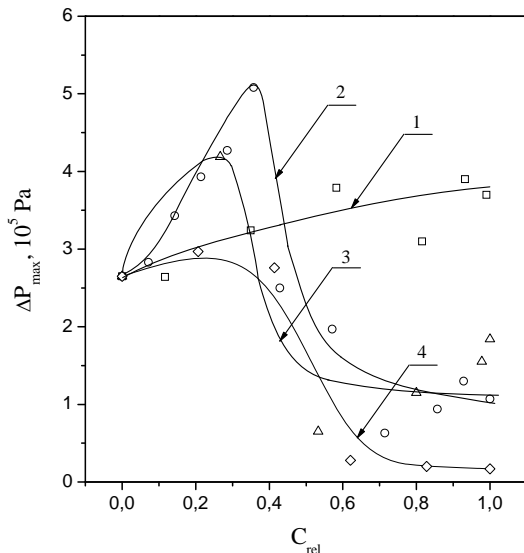


FIGURE 7. Dependence of the maximum explosion pressure on the relative diluent concentration at the oxygen content in the oxidizer 25 % (vol.). Diluents: 1 (\square) – CF_3H ; 2 (\circ) – $\text{C}_2\text{F}_5\text{H}$; 3 (Δ) – C_4F_{10} ; 4 (\diamond) – AKM.

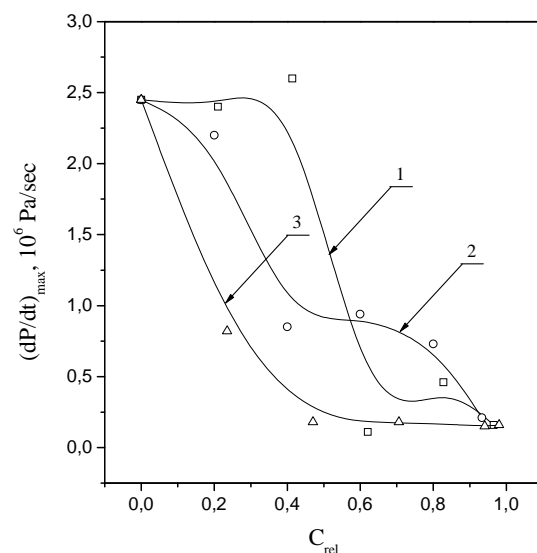


FIGURE 8. Dependence of the maximum explosion pressure rise rate on the relative diluent concentration at the oxygen content in the oxidizer 15 % (vol.). Diluents: 1 (\square) – CF_3H ; 2 (\circ) – $\text{C}_2\text{F}_5\text{H}$; 3 (Δ) – C_4F_{10} .

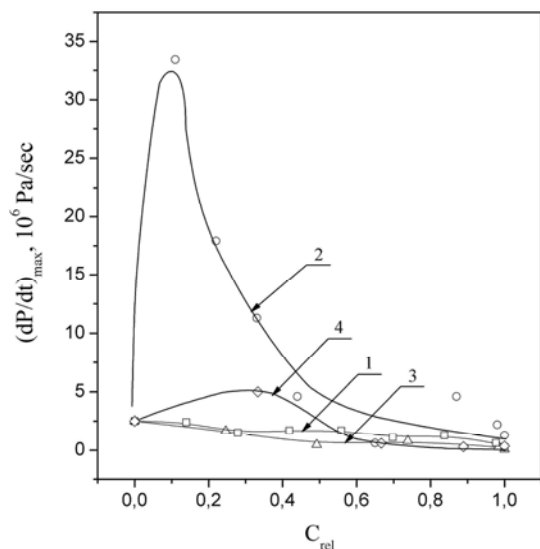


FIGURE 9. Dependence of the maximum explosion pressure rise rate on the relative diluent concentration at the oxygen content in the oxidizer 20.6 % (vol.). Diluents: 1 (□) – CF₃H; 2 (○) – C₂F₅H; 3 (Δ) – C₄F₁₀; 4 (◇) – AKM.

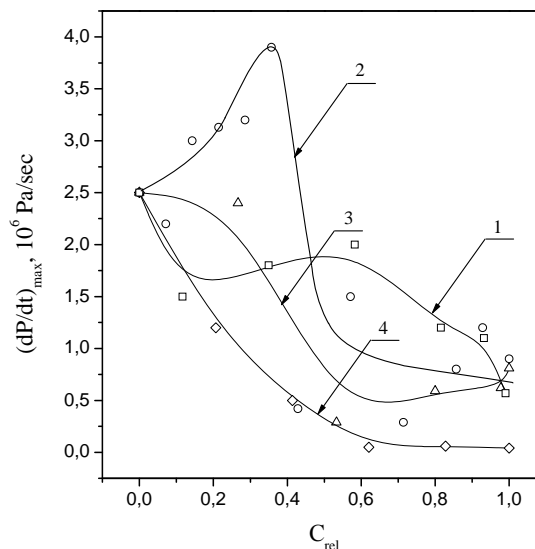


FIGURE 10. Dependence of the maximum explosion pressure rise rate on the relative diluent concentration at the oxygen content in the oxidizer 25 % (vol.). Diluents: 1 (□) – CF₃H; 2 (○) – C₂F₅H; 3 (Δ) – C₄F₁₀; 4 (◇) – AKM.

It can be seen that the curves characterizing a dependence of Δp_{\max} on a relative diluent concentration C_{rel} have as a rule maxima (C_{rel} is a ratio of a diluent concentration in a mixture to the value of this concentration at a peak point of a flammability curve – see Figs. 1 to 4). Positions of these maxima depend both on type of the diluent and on an oxygen content in the oxidizer [O₂]. The higher the oxygen content [O₂], the more clearly the position is expressed a maximum in a dependence of Δp_{\max} on C_{rel} . For example, the maximum explosion pressure Δp_{\max} is increased at an inserting of C₂F₅H into a flammable mixture from 0.27 MPa ($C_{\text{rel}} = 0$) to 0.36 MPa ($C_{\text{rel}} = 0.8$, and this quantity corresponds to the maximum of Δp_{\max} as a function of C_{rel} at the oxygen concentration in the oxidizer 15 % (vol.) (Fig. 5). But at [O₂] = 25% (vol.) the analogous increase of Δp_{\max} occurs from 0.27 to 0.51 MPa (Fig. 7). Quantitatively the same results were observed for other diluents (Figs. 5 to 7). The least expressed maxima with the following rapid decrease of Δp_{\max} at an elevation of C_{rel} (larger than the extremum value) is realized in the case of perfluorobutane C₄F₁₀.

Another character of curves can be seen in the case of a dependence of $(dp/dt)_{\max}$ on C_{rel} (Figs. 8 to 10). The remarkably expressed maxima take place only for C₂F₅H, but for the other diluents this maxima are either absent (C₄F₁₀, AKM) or are expressed very slowly (CF₃H). A drop of $(dp/dt)_{\max}$ at the diluent concentration higher than the extremum value occurs very rapidly.

According to the existing theoretical approaches²⁹⁻³¹ an adiabatic flame temperature for near limit mixtures is approximately the same for various mixture's compositions. This regularity leads to an approximate constancy of the maximum explosion pressure for these mixtures. The experimental data of our study do not confirm this semiempirical rule (see Figs. 5 to 7), that is the applicability of this rule is rather narrow.

As it was mentioned above, the most part of the dependences of Δp_{\max} on C_{rel} have maxima (Figs. 5 to 7). According to^{22,23} the availability of such maxima is caused by an active participation of the diluents in processes of a chemical conversion in a flame front with an additional (in comparison with mixtures without diluents) heat release. This regularity is valid not only for AKM (which is a

flammable gas), but also for nonflammable fluorinated agents. In the case of mixtures, which are far from near limit ones, the maxima take place as a rule not only for Δp_{\max} , but also for $(dp/dt)_{\max}$ ^{22,23}. But in our study dedicated to near limit flames, the maxima for $(dp/dt)_{\max}$ occur only for C_2F_5H . Thus an inhibition of near limit flames can be reached more easily than of mixtures far from the limit despite of the additional heat release.

A dependence of $(dp/dt)_{\max}$ on C_{rel} characterizes a heat release rate at a flame propagation in a gaseous mixture in a closed vessel. This heat release rate depends both on an effective heat of combustion (taking into account the additional heat release due to a chemical conversion of a diluent in a flame front) having a maximum as a function of C_{rel} (Figs. 5 to 7) and a laminar burning velocity characterizing a rate of the release of this effective heat of combustion. An absence of maxima in the dependences of $(dp/dt)_{\max}$ on C_{rel} in the most part of our experiments indicated that the effect of a diminishing of the laminar burning velocity due to presence of a diluent prevails over the effect of an increase of the additional heat release. Therefore the maxima in the dependences of $(dp/dt)_{\max}$ on C_{rel} are absent in most part of our experiments. Because the maxima in the dependences of $(dp/dt)_{\max}$ on C_{rel} usually take place for mixtures, which composition is far from flammability limits^{22,23}, a conclusion can be made that inhibition of near-limit flames occurs more easily than of flames far from flammability limits.

For the most part of the curves characterizing the dependences of Δp_{\max} and $(dp/dt)_{\max}$ on C_{rel} a rapid decrease of the maximum explosion pressure and the maximum explosion pressure rise rate at an increase of the relative diluent concentration over the certain threshold takes place (see for example curves 3 in Figs. 5 to 7, curve 1 in Fig. 8, curves 2 in Figs. 9 and 10 as the most typical cases). These experimental data confirm an existence of two different kinetic regimes of chain branching combustion, which differ by a qualitative character of dependences of a heat release rate q_+ and its derivative dq_+/dT on temperature T . These kinetic regimes were discovered, explored and confirmed by many experimental data (see³²⁻³⁴ and works cited there).

It is interesting to reveal, what a diluent among tested in this study is the most effective for reduction of combustion characteristics of rich near-limit hydrogen – oxidizer – diluent mixtures. It is followed from the data on Figs. 5 to 10 that the most effective is perfluorobutane C_4F_{10} (curves 3) and AKM agent (curves 4). For C_4F_{10} the maxima in the dependences of Δp_{\max} and $(dp/dt)_{\max}$ on C_{rel} are absent in the most cases, and the drop of these parameters with an increase of C_{rel} occur rather rapidly. The analogous properties has AKM agent. At the same time the worst inhibitive properties were found for pentafluoroethane C_2F_5H (curves 2). This conclusion is followed from the availability of the maxima in all determined dependences of $(dp/dt)_{\max}$ on C_{rel} (for other diluents such regularity is not observed in all cases), and thus effect is probably due to a slow inhibitive action of C_2F_5H on the laminar burning velocity. It should be noted²³ that it is hardly possible to propose a universal inhibitor, which is effective for decreasing of all combustion characteristics of gaseous mixtures of any composition (independent on concentrations of fuel and inhibitor, oxygen content in an oxidizer and a chemical nature of fuel). Therefore the conclusions on relative effectiveness of various agents are referred only for the investigated gaseous mixtures, the experimental conditions and the determined combustion characteristics.

CONCLUSIONS

In this study an experimental investigation of combustion characteristics of rich near limit mixtures hydrogen – oxidizer – diluent in a closed vessel for various oxygen concentrations in the oxidizer was executed. Dependences of a maximum explosion pressure Δp_{\max} and a maximum explosion pressure rise rate $(dp/dt)_{\max}$ on a diluent concentration C_{rel} were determined. It was found that the dependences of Δp_{\max} on C_{rel} have maxima in the most cases, but at the same time such maxima in the dependences of $(dp/dt)_{\max}$ on C_{rel} take place only for pentafluoroethane C_2F_5H . A qualitative interpretation of the obtained results was proposed based on the concepts of an active participation of the investigated agents in a chemical conversion in a flame front with an additional heat release and of an existence of

two kinetic regimes of chain branching combustion. It was shown that C₄F₁₀ and AKM are the most effective among tested in this study inhibitors in relation to combustion of rich near-limit mixtures hydrogen – oxidizer – diluent.

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