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THE EFFECT OF A CATALYTIC AGENT ON THE CHARACTERISTICS OF THE COMBUSTION ZONE OF A CONDENSED SUBSTANCE

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Wright-Patterson Air Force Base, Ohio

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THE EFFECT OF A CATALYTIC AGENT ON THE CHARACTERISTICS OF THE COMBUSTION ZONE OF A CONDENSED SUBSTANCE

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Moscow

One of the methods of regulating the combustion rate of condensed substances is the introduction of small additives into the fuel composition. A large number of works have been devoted to the investigation of the effect of these additives on the combustion mechanism [1-6]. The question of the site of the effect of a catalytic agent in a combustion wave is of great interest to investigators. The following zones in a combustion wave are known [7]: the zone of heating in the condensed phase (k-phase), the decomposition zone near the surface of the burning powder in which gasification and the primary reactions take place, and the gaseous zone in which the combustion reactions are completed. A number of authors [8-10] consider the gaseous zone to be the site of catalytic agent action. There are works, in which the authors have examined the k-phase and the decomposition zone as the probable site of the catalytic agent action [11, 12]. Finally, the authors of work [13] consider, that the catalytic agent acts both in the gaseous as well as in the condensed phases. The conclusions of these works are

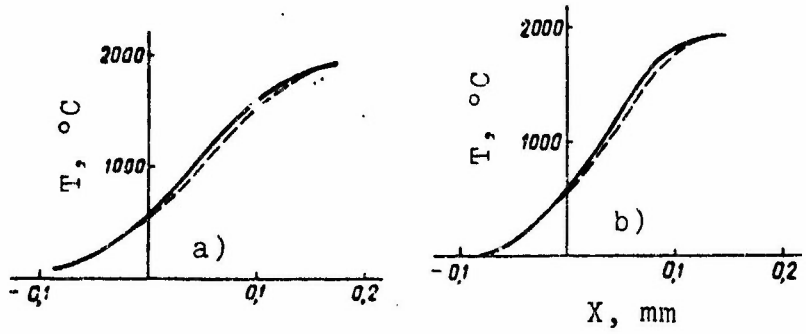
based on indirect considerations. Measurements of the effect of a catalytic agent on the temperature distribution in the combustion zone and on the surface temperature (T_n) have not yielded results [14, 15].

The purpose of the present work is to obtain data on the effect of a catalytic agent on the process of the thermal interaction of the combustion zones and to indicate the site of the effect of the combustion catalytic agent. For this, the temperature distribution in the combustion wave and T_n were determined on the burning surface of model mixtures with catalytic agents and without them.

The main experiments were carried out on a mixture of ammonium perchlorate (APC) with polymethyl methacrylate (PMMA) with an excess of oxidizer (α) 0.6, without a catalytic agent and with 1% Fe_2O_3 at pressures of 5-60 at. The mean arithmetic dimensions of the APC particles were equal to 4 μm, of the PMMA - 8 μm, and of the Fe_2O_3 - 5 μm. Control experiments were also carried out at a pressure of 40 at for the APC mixture (40% - less than 50 μm, 60% - with dimensions of 130-160 μm) with a rubber-like binding (RLB) with α = 0.38, without a catalytic agent, with 1% Fe_2O_3 and with 3.8% ferrocene. The temperature distributions were determined with tungsten-rhenium thermocouples with thicknesses of 7; 3.5, and 2.5 μm [16], the surface temperature - with thermocouples with small weights [17], the combustion rate - by motion picture photography. The mixture with PMMA has considerable combustion rates and temperatures gradients near the surface, thus, to eliminate errors connected with the thermal lag of the thermocouples, the experimentally obtained temperature distributions in the gaseous phase were reconstructed with the following formula

$$T_i = T'_i + \frac{u\tau_0}{2\Delta x}(T'_{i+1} - T'_{i-1}),$$

where u is the combustion rate; τ_0 is the thermocouple time constant [16]; Δx is the calculation interval with respect to the X coordinate; T_i' is the temperature being measured.



The temperature distribution for the mixture combustion zone (p = 30 at); the broken line is the measured temperature, the solid line is the temperature recalculated with formula (1): a) PMMA + APC + Fe_2O_3; b) PMMA + APC.

The figure presents typical temperature distributions, on the basis of which the reconstruction is demonstrated, taking the thermocouple lag into account (thickness 7 μm).

Table.

p, at	Composition	u, mm/s	T_n, °C	$\dot{T}_n$, deg/μm	Δ	q, cal/g	Q, cal/g	Φ, kcal/cm³·s
5	Б	4,0	400	14,0	1,0	27	101	33
	С	5,0	400	14,6	1,7	22	106	43
13	Б	5,5	500	25,0	1,3	40	118	81
	С	7,0	500	20,0	4,0	25	133	83
30	Б	8,0	650	19,0	1,8	27	176	91
	С	14,0	700	12,0	5,5	10	206	100
60	Б	7,0	700	55,0	2,1	93	123	230
	С	14,4	700	25,0	3,5	21	193	210
Error, %		±4	±13	±12	±23	±13	±16	±12

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The basic results of the experiments with the PMMA mixture and their processings are presented in the figure. The letter Б designates the composition without the catalytic agent, the letter C - with the catalytic agent. The average probable relative errors of the indicated magnitudes are indicated in the last line of the table. The combustion rate at 30 and 60 at upon the introduction of the catalytic agent increases approximately by two times. The magnitude of $T_п$ increases with an increase in pressure, and for both mixtures within the limits of experimental error it is identical. The experimentally obtained depth of the heating zone and of the chemical reactions in the k-phase is 30-80 μm. The ratio of this magnitude to the theoretical $X_м = a/u$ ($a = 1.2 \cdot 10^{-3}$ cm^2/s [7]) is designated in the table by Δ. The relative expansion of the zone in the k-phase (Δ) for the Б mixture attains 2.1 at 60 at, which is usual for mixed systems. For the C mixture Δ can attain 5.5. Such a severe expansion of the zone in the k-phase for the mixture with the catalytic agent is the first characteristic feature of the action of the catalytic agent in this system. The temperature gradient in the gaseous phase for the φ_n surface (in degrees (°C) per micron), found with respect to the slope of the temperature distributions near the surface, is for the Б mixture greater than for the C mixture (at pressures, where the catalytic agent increases the combustion rate, i.e., approximately above 10 at). The difference in the heat supply q in the case of thermal conduction from the gaseous phase into the condensed phase is even more sharply expressed for the Б and C mixtures ($q = \lambda \cdot \varphi_n / m$, λ was calculated with respect to the composition of the combustion products and was equal to $1.3 \cdot 10^{-4}$; $1.7 \cdot 10^{-4}$; $2.6 \cdot 10^{-4}$ cal/cm·s·deg at temperatures of 400, 600, and 1000°C respectively). An estimate shows, that the heat supply to the k-phase surface by radiation does not exceed 2-4 cal/g and thus we did not take it into account. So, the second characteristic feature of the catalytic agent effect in this system is the decrease in the heat supply from the gaseous phase into the condensed phase upon the introduction of the catalytic agent.

The heat release in the near-surface layer[1] (n.s.l.) was calculated in accordance with the following equation

$$Q = c(T_n - T_0) - q + q',$$

where Q is the heat release for the original mixture, cal/g; c is the average specific heat of the k-phase (0.3 cal/g·deg); T_0 is the initial temperature of the mixture; q' is the heat of the phase transition in the APC (19.4·0.723 = 14 cal/g [18]).

It is apparent from the table, that for both systems there is considerable heat release in the near-surface layer (100-200 cal/g). If at pressures of up to 30 at the difference in Q for the Б and C mixtures is within the limits of experimental error, then at a pressure of 60 at the magnitude of Q for the C mixture exceeds the Q of the Б mixture by more than 1.5 times. Thus, the third characteristic feature of the catalytic agent effect in this system is the increase in the heat release of the n.s.l. upon the introduction of the catalytic agent.

Finally, calculation of the average rate of heat release in the gaseous phase (Φ) with respect to the experimental temperature distributions was carried out (an approximate estimate is possible with formula $\Phi = c_p m \Phi_n$, $c_p = 0.3$ cal/g·deg; a more accurate estimate - by averaging the curve $\Phi = \Phi(x)$ with respect to [19]). It is evident from the table, that within the limits of the errors Φ is identical for both systems. Thus, the fourth characteristic feature is: the catalytic agent does not change the average rate of heat release in the gaseous phase.

[1]The near-surface layer is the reaction layer of the k-phase and the layer of the gaseous phase (5-10 um), unresolvable by the thermocouple, adjacent to it.

These results point to the near-surface layer as the site of catalytic agent action. It is of interest to check, whether the catalytic agent action is the purely mechanical loosening of the k-phase reaction layer during the rapid heating, to which the k-phase is subjected in the combustion wave. To check this assumption, instead of Fe_2O_3, Al_3O_3 (sic) was employed in the very same quantity and at approximately the same dispersity. The experimental data for the mixture with Al_2O_3 corresponds more to the data of the Б mixture. The magnitude of Δ is less than 2, which points to the chemical action of the Fe_2O_3 catalytic agent. The conclusion obtained concerning the site of the Fe_2O_3 catalytic agent action in the system with PMMA can be understood, if we estimate the time that the catalytic agent particles remain in the different zones with respect to the experimental temperature distributions. The time that the catalytic agent particles remain in the gaseous phase (taking into account their lag and the variability of the gas temperature) is 10^{-4} s, whereas the sojourn time in the k-phase reaction layer is equal to 10^{-2}-10^{-3} s. If we consider, that the time of the diffusion of the reagents to a catalytic agent particle ($\tau_д$) [20] for Fe_2O_3 of 5 μm dimensions is ~10^{-2} s, then it becomes clear, that one of the reasons for the absence of catalytic agent action in the gaseous phase is that its sojourn in the volume of the gaseous phase is extremely short. Moreover, since $\tau_д$ exceeds the sojourn time even in the k-phase reaction layer, it is necessary to assume, that only part of the substance reacts on the catalytic agent, and these reactions are essential for increasing the combustion rate.

With the introduction of Fe_2O_3 and ferrocene into the APC (mixed) + RLB composition, the effects are within the limits of the probable errors, thus it is possible to discuss only the trends of the average values of the magnitudes being measured. The average values of Φ increase upon the introduction of catalytic agents by approximately two times, however, q decreases, in spite of the twofold increase in the rate. The magnitudes of Q and Δ

increase. The thermal fluxes Qm and qm also increase, however, both in absolute magnitude as well as with respect to its increase Qm is by an order of magnitude greater than qm.

Thus, the iron-containing catalytic agents, Fe_2O_3 and ferrocene, in the investigated mixtures can act both in the near-surface layer, as well as in the main volume of the gaseous phase, but only the catalytic effect in the near-surface layer has an effect on increasing the combustion rate.

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