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TRANSLATION

THE QUESTION ABOUT MECHANISM OF INFLUENCE OF LIQUID LAYER DURING BURNING OF MIXED SYSTEMS

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THE QUESTION ABOUT MECHANISM OF INFLUENCE OF LIQUID LAYER DURING BURNING OF MIXED SYSTEMS

A. P. Glazkova

During study of burning of mixture of ammonium perchlorate (dimension of particles $< 250\mu$) with paraffin it was established [1] that dependence of burning rate on pressure has two parts: in first part to 300 atm, burning rate grows with pressure according to law $U_{\Pi} = Bp^{\nu}$, where $\nu = 1$; and second part, starting from 300 atm, where rate grows by linear law. This contradicts usual presentations about burning of mixtures, and also hypothesis expressed by O. I. Leypunskiy [2], according to which dependence of burning rate of mixture of saltpeter with carbon should decrease with growth of pressure, when diffusion mechanism becomes leading. Still more paradoxical is the fact that addition to ammonium perchlorate of certain fuels (coke, polymethylmethacrylate [1]), leading to increase of maximum (calculated) temperature of burning*, not only does not increase rate of its burning in significant range of pressures, but, on the contrary, leads to its decrease. This circumstance forces the assumption that in shown cases rate of process is

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*By calculation carried out by I. A. Tereshkin, temperature of burning of ammonium perchlorate is 1580° K; temperature of burning of its stoichiometrical mixture with coke is 3390° K, with paraffin—3280° K.

determined by transformations of the ammonium perchlorate itself and subsequent reactions with fuel have no influence on it.

Burning rate of mixtures of ammonium perchlorate with paraffin, significantly larger than that with coke, gives a basis to assume that essential factor determining burning rate of mixtures, is, besides reactionary abilities, physical properties of fuel and oxidiser, and in particular, their ability to melt while burning. If the oxidiser itself during burning changes to the liquid state (for example, perchlorate of potassium, due to forming of melted KCl), nature of fuel, supposedly, has no effect on the character and rate of burning of mixture.

To check this assumption we investigated mixtures of oxidisers with fuel which possessed various physical properties and namely, with fusible and infusible fuels. In order to exclude influence of chemical properties, experiments were also done with mixtures in which as fuel there was used dimethacrylate-triethyleneglycol in the form of a monomer (liquid) and polymer (hard, infusible, vitrious compound). Stoichiometric mixtures were pressed in portions in plexiglas pipes with an internal diameter of Ø7 mm, and wall thickness of 1 mm to densities near to theoretical; height of column of substance was more than three diameters. Dimension of particles of components in the case of mixtures on the base of ammonium perchlorate was $<250\,\mu$**. Rates and nature of burning were determined photographically, as was described earlier [1]. Experiments were conducted in a cylinder of constant pressure in an atmosphere of nitrogen.

As can be seen from Figs. 1 and 2, all studied mixtures of ammonium perchlorate with liquid or fusible fuel (paraffin, dimethacrylate-triethyleneglycol, dyne) burn faster than ammonium perchlorate, but mixture with infusible fuel (coke,

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**Combustible and oxidiser were sifted through sieve 025. Mixing of components was done manually using rubber cork. Paraffin mixtures were prepared by mixing oxidiser with solution of paraffin in gasoline "overshoe", which then was completely evacuated.

polymethylmethacrylate, polymer dimethacrylate-triethyleneglycol, naphthalene*, urotropine, cellulose) burn in significant range of pressures with smaller speeds than ammonium perchlorate itself. [We do not consider here region of pressures from 160 to 500 atm, in which burning of perchlorate (at a diameter of samples without shell which is larger than the critical diameter) is unstable and proceeds with speed ~ 0.7 — 1.0 g/cm^2 sec. At pressures of 750, 875, and 1000 atm average rate of stable burning increases to 3.3, 4.2, and 7.7 g/cm^2 sec, respectively.]

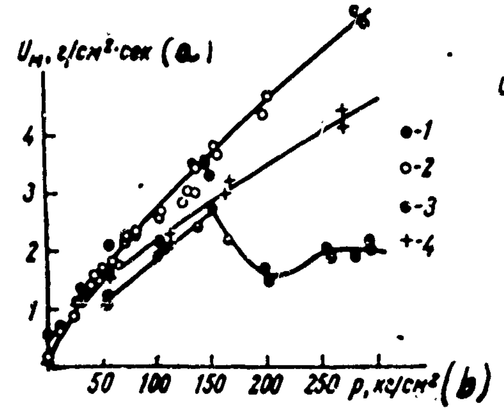

Fig. 1

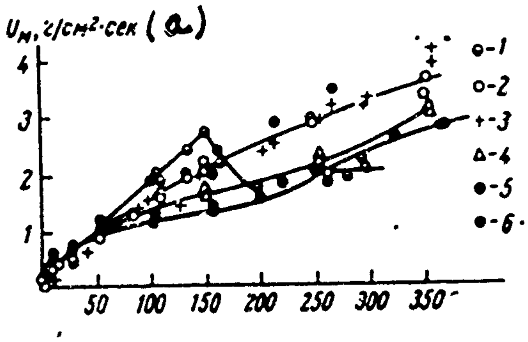

Fig. 2

Fig. 1. Dependence of burning rate on pressure for mixtures of ammonium perchlorate with fusible fuels: (1) ammonium perchlorate in plexiglas pipes, (2) with paraffin, (3) with dimethacrylate-triethyleneglycol, (4) with dyne (dimension of particles of oxidizer $>10\mu<50\mu$)
KEY: (a) glcm^2 · sec; (b) kg/cm^2.

Fig. 2. Dependence of burning rate on pressure for mixtures of perchlorate of ammonium with infusible fuels: (1) perchlorate of ammonium, (2) with coke, (3) with cellulose, (4) with naphthalene (easily sublimated), (5) with urotropine, (6) with polymer dimethacrylate-triethyleneglycol. Dimension on abscissas: p, kg/cm^2
KEY: (a) g/cm^2 · sec.; (b) kg/cm^2.

Regarding nature of burning of perchlorate of ammonium and its mixtures, in case of mixtures with infusible fuels the granular structure of front of burning, inherent to the actual perchlorate of ammonium is kept local; front of burning consists of separate micro-flashes (see Fig. 3), and in case of burning of mixtures with

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*Is melted, but is easily sublimated.

paraffin, for example, for surface there is evident a brighter luminescence than for the remaining flame, a zone of noticeable thickness, decreasing with increase of pressure and disappearing at a pressure of ~ 700 atm.

Stoichiometric mixtures on a base of perchlorate of potassium (dimension of particles $> 50\mu$ and $< 100\mu$) both with liquid and fusible fuels (paraffin, dimethacrylate-triethyleneglycol), and with infusible (naphthaline, polymer dimethacrylate-triethyleneglycol) burn with close velocities, whose law of burning can be expressed in first order of approximation by an equation of the type $U_M = 0.03275\, p$ ($[U_M]$ g/cm^2 · sec, $[p]$ kg/cm^2).

Burning of stoichiometric mixtures of ammonium nitrate with the monomer and polymer of dimethacrylate-triethyleneglycol was also studied. These mixtures do not burn at pressures below 350 atm, and in range of pressures from 350 to 1000 atm burning rates of these mixtures are practically identical**. Mixtures of heated saltpeter with the indicated monomer and polymer also do not burn at 350 atm.

When studying the combustion of mixtures one should take into account, first, the ability of one or both components*** to burn independently, and second, their ability to change to the liquid state. What role can fluidity or fusion play during burning of mixture? Main distinction of mechanism of burning of mixtures considered by Anderson [3] and Chaiken [4] from mechanism of burning of individual explosives (VV) considered by A. F. Belyayev and Ya. B. Zel'dovich [5] consists in the following. During burning of individual VV, heat emitted in the gas phase is transmitted directly to the surface of the condensed phase.

**This can be connected both with ability of the actual saltpeter to melt, and also with the fact that dimension of particles of saltpeter in our experiment ($< 100\mu$) was of the same order as the thickness of zone of heating (100μ) at 200 atm, for example, for amatole 80/20.

***In the case when one of components of mixture is able to independently burn, regularities of its burning cannot help but affect regularities of burning of mixture.

During burning of mixtures in condensed phase there is first of all heat from gas reactions proceeding near surface of particles (as result of decomposition of oxidizer). From zone with maximum temperature, due to remoteness of this zone from surface, and also presence near particles of primary oxidizer, or, as R. F. Chaykin calls it, "buffer" flame, heat is practically not transmitted to the condensed phase and does not introduce therefore its own contribution to the burning of system. This phenomenon is analogous to that which was observed during burning of liquid nitroglycol [6], when appearance of "secondary", "hotter" flame did not influence either burning rate of its dependence of it on pressure.

Fig. 3. Photographs of burning of perchlorate of ammonium and its mixtures at 200 atm: a) with coke, b) pure, c) with paraffin.

If during burning of mixture, fuel, for example, is able to change to liquid state, then the liquid formed will envelop surface of particles of oxidizer. Every particle of oxidizer in zone of reaction in this case will be in contact not with separate particles of fuel, but with layer of melting of definite thickness.

Transition into liquid state involves, as it were, "crushing" in the course of burning of one of the components to very small dimensions—practically to separate molecules, as a result of which their reaction ability is sharply increased. Gaseous products of primary reaction of disintegration of oxidizer have to pass through layer of melt formed, giving to it part of heat and forcing it partially to decompose or to evaporate. Zone with maximum temperature in this case will be located nearer to surface of condensed phase*. Heat from this zone can be now transmitted back into condensed phase, since surface of oxidizer is not altogether bare, and buffer flame enveloping it either will not be formed at all (if thickness of zone of melting is larger than dimension of particles) or its role decreases. Schematically this is shown in Fig. 4.

Thus presence of liquid phase during burning of mixture partially or completely liquidates buffer flame and moves zone with maximum temperature to surface of condensed phase, which involves increase of temperature in zone of reaction for surface of condensed phase. In brief, depending upon physical properties of components, the system (fuel-oxidizer) can burn either as mixture or analogously as individual VV; and since for majority of individual VV studied by us dependence of burning rate on pressure is near to linear, this allows explanation of linear section of curve $u = f(p)$ during burning of paraffin mixture.

Alternate path of transformation of mixture into "individual" explosive is realized for sufficiently small dimensions of particles of components of mixture; it was considered by N. N. Bakhman [7]. Indeed, our experiments showed that a stoichiometric mixture of perchlorate of ammonium (dimension of particles $1-10\,\mu$) with gas black burns according to linear law $U_M = 0.13 + 0.0115\ p$ in range of pressures to 350 atm. In accordance with the above, dimension of particles of

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*Preliminary experiments on defining burning temperature of mixtures of ammonium perchlorate with low-melting monomer (m.p. = 52°) and its infusible polymer were conducted by our request in laboratories of P. F. Pokhila; they showed that in case of mixture with monomer, distance from zone with maximum temperature to surface of condensed phase at all studied pressures (20, 40, 60 atm) is less than for mixtures with polymer in these conditions.

Author takes this opportunity to express gratitude to V. M. Mal'tsev for carrying out these experiments.

oxidizer must be reflected in burning rates of mixture in smaller degree if fuel is easily melted, than in the case when fuel is infusible.

In Fig. 5 are data of experiments on influence of dimension of particles on burning rate of mixtures of ammonium perchlorate with fusible monomer (m.p. = 52°) and its infusible polymer. Solid curve without points shows $U_M = f(p)$ for mixture with polymer with dimension of oxidizer particles <250μ; curve 3 refers to the same mixture, but with dimension of particles of oxidizer 1—10μ. Burning rates of mixtures differ by approximately two times. Dotted curve depicts

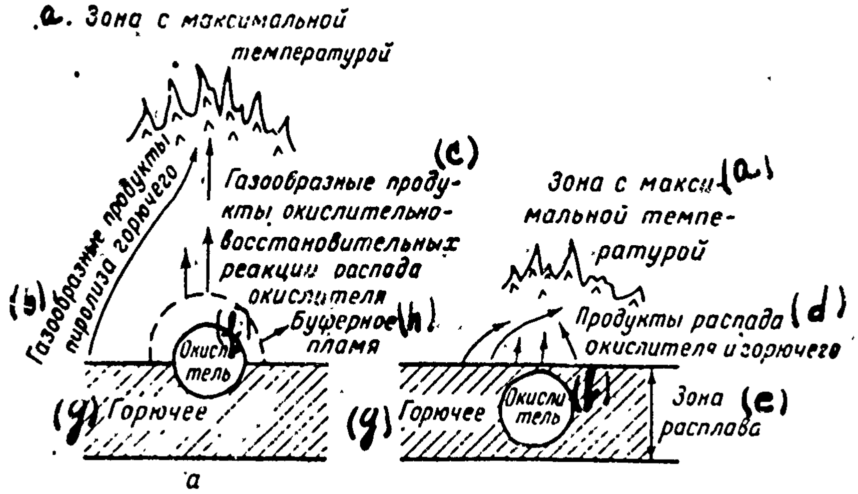

Fig. 4. Schematic image of burning of mixtures: (a) with formation of a buffer flame per Andersen [3]; (b) without formation of buffer flame, due to presence of liquid component
KEY: (a) Zone with maximum temperature; (b) Gaseous products of pyrolysis of fuel; (c) Gaseous products of oxidation-reduction reactions of decomposition; (d) Products of decomposition of oxidizer and fuel; (e) Zone melting; (f) Oxidizer; (g) Fuel; (h) Buffer flame.

$U_M = f(p)$ for analogous mixture with dimension of particles NH_4ClO_4 <250μ, but with monomer, and curve 2—$U_M = f(p)$ for mixture with monomer for dimension of particles of NH_4ClO_4, equal to 1—10μ. From the figure it is clear that in case of fusible monomer burning rates of mixtures practically coincide, in spite of the fact that dimensions of particles of oxidizer strongly differed from the viewpoint expressed by our hypothesis that it is possible to consider influence of dimension of particles during burning of mixtures with liquid or fusible components as a result of increase or decrease of role of buffer flame during burning of mixture. Incidentally, it is necessary to note that increase of pressure leading to decrease of thickness of heated layer, will lead (at constant dimension of particles) to relative increase of dimension of particles in comparison with zone of heating. Disappearance of bright luminescent stripe during burning of paraffin mixture at 700 atm can be connected with this influence.

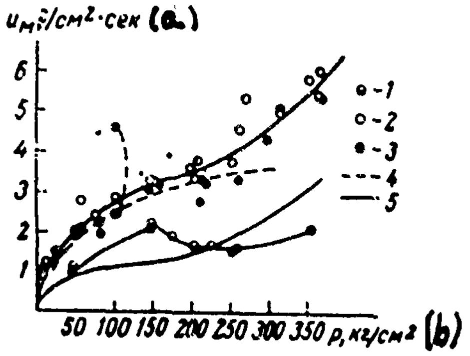

Fig. 5. Influence of dimension of particles on dependence of burning rate on pressure: (1) NH_4ClO_4, dimension of particles 1 — 10μ; (2) NH_4ClO_4 (dimension of particles 1—10 μ) + monomer; (3) NH_4ClO_4 (dimension of particles 1—10μ) + polymer; (4) NH_4ClO_4 (< 250μ) with monomer; (5) NH_4ClO_4 (< 250μ) with polymer.
KEY: (a) 2/cm^2 · sec; (b) kg/cm^2.

Author considers it his pleasant duty to express gratitude to K. K. Bobolev, in whose laboratories experiments were carried out, and also to his own comrades in the work, L. G. Bolkhovitinov and A. D. Margolin for discussion.

Conclusions

→ a study was made of ~~1. There was studied~~ the burning of mixtures on a base of perchlorates of ammonium and potassium with fusible and infusible fuel in range of pressures of 1 — 350 atm. If while burning, one of the components of the mixture changes to the liquid state, then the mixture burns according to a mechanism similar to the mechanism of burning of individual explosive substances.

~~2.~~ A Hypothesis is expressed about mechanism of influence of liquid layer during burning of mixture systems. ( )

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