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EDITED MACHINE TRANSLATION

HEAT AND MASS TRANSFER (SELECTED ARTICLES)

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PREPARED BY:

TRANSLATION DIVISION FOREIGN TECHNOLOGY DIVISION W-APB, ONIO.

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detonation transition was studied in a constant volume bomb by pressure recording and high speed photography. The hexogen samples were compacted into plexiglass cases and ignited by an electric wire or a powder charge. The results showed that the deflagration-to-detonation transition under increasing pressure takes place according to the following order: normal combustion; perturbed combustion; ejection of particles into the flame zone, which is accompanied by interruption of luminosity; accelerated combustion of the ejected particles, which generates a pressure increase above the burning surface; and gas penetration into the pores, which leads, in case of a pure explosive, to a detonation and, in case of an explosive phlegmatized with paraffin wax, to accelerated combustion of the charge. Phlegmatization of the charge decelerates the developemtn of this process and eliminates the deflagration-to-detonation transition. A new type of deflagration-to-detonation transition was found which is caused by the self-ignition of semiproducts and gases in the pores of the charge. Original article has: 5 figures. English translation: 7 pages.



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INVESTIGATION OF THE STRUCTURE OF SURFACE OF BURNING OF MODEL MIXTURES OF SOLID FUELS

P. F. Pokhil and L. D. Romodanova

For a deeper understanding of the mechanism of burning a solid fuel mixture we investigated the structure of the surface forming with the burning of a model of stoichiometrical mixtures with oxidizer as a base: ammonium perchlorate, unmelted, spontaneously inflammable at a temperature of 633-653°K; potassium perchlorate melted with decomposition at 883°K, and sodium perchlorate melted at 755°K, decomposed at 778°K.

Different fuels were selected differing in their physical chemical properties: graphite unmelted and undecomposable; tungsten melted at a temperature of $\sim 3473^{\circ}$ K; napthalene, melted and sublimated at 353°K; melted with decomposition at 533°K, amber and malonic dibasic, saturated; fatty acids (amber acid - $T_{\Pi\Pi}$ = 458°K, $T_{\Pi\Pi}$ = 508°K; malonic acid - $T_{\Pi\Pi}$ = 408.6°K).

Study of the structure of the surface forming with burning of these mixtures was conducted with the aid of a microscope (amplification of 10-80 times). There was studied the surface forming with burning of these mixtures in a vacuum, in air and under pressure.

The mixtures were pressed to a maximum density with a diameter of the charge at 0.005 m.

All the compositions studied by us except composition $KClO_4 + W$ (tungsten) in a vacuum (pressure $p \sim 10^{-2}$ mm Hg) at room temperature do not burn stably and with removal of the incandescent spiral die out.

At atmospheric pressure the compositions were extinguished by compressed air. Under pressure the inflaming composition was extinguished by a rapid drop in pressure in a bomb. The methods of carrying out the experiments in a vacuum and under pressure was described in the preceding works [1, 2].

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$\frac{Results of Investigation of Mixtures with the Oxidizer}{NH_4Cl0_4} \frac{as a Base}{}$

Study of the structure forming with burning of a model of stoichiometrical mixtures with the oxidizer $NH_{\rm H}ClO_{\rm H}$ (with the dimension



Fig. 1. Oxidizer $NH_{4}ClO_{4}$: a) - 1 - fuel-malonic acid; 3 - fuel - starch; 2, 4 oxidizer; b) fuel-tungsten (1), graphite (2); 3 napthalene; c) crystals $NH_{4}ClO_{4}$: large (1), small (2). of particles from 40 to 300 µm) and combustible organic dibasic saturated fatty acids, malonic and succinic, and starch as a base showed that in a vacuum (p $\sim 10^{-2}$ mm Hg) under the action of a heated spiral on the surface of the charge the fuel is melted and decomposed, and the crystals of the oxidizer are preserved and therefore come forward above the surface of the mixture. Between the emerging crystals of the oxidizer there are depressions in which fuel was found, as was shown schematically on Fig. la. Besides separate crystals, on the surface there are observed aggregates of crystals with a dimension of ~0.0004 m.

The larger the crystals, the greater they come forward above the surface of burning. The protruding crystals are partially covered by products of pyrolysis of fuel (for example, carbonized particles of starch).

An analogous picture is observed on the surface of these mixtures with burning in air. With an increase in pressure crystals of the oxidizer protruding above the surface, decrease, and at pressures greater than $p \sim 30$ atm (abs.) at the place where there were crystals of ammonium perchlorate depressions, craters, will be formed (Fig. la).

This phenomenon is explained in the following way. It is known [3] that the maximum pressure, lower than which ammonium perchlorate does not burn stably, at room temperature is equal to 20-30 atm (abs.). Therefore, with burning of mixtures in the region of low pressures, when ammonium perchlorate does not burn independently and the temperature of the decomposition of it is higher than the temperature of decomposition of the fuel on the surface crystals of $NH_{\rm H}ClO_{\rm H}$.

At a pressure greater than the limiting, when ammonium perchlorate is capable of stably burning independently (without a cluster), it vanishes faster than the cluster and on surface there will be formed depressions - craters (on the place where crystals of $\rm NH_4ClO_4$ were

located) *)}^{\$**}*

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On the liquidated surface of the mixture $NH_{4}ClO_{4}$ + napthalene (Fig. 1, 3) the oxidizer is stored. The accumulation of the oxidizer on the surface is caused by the great difference between melting points and sublimation of napthalene and the temperature of the thermal decomposition of $NH_{4}ClO_{4}$. Therefore, in the process of

burning of the mixture, when the temperature on the surface is equal to $\sim723^{\circ}$ K, the napthalene vanishes in the surface layer earlier than that of the oxidizer.

It is characteristic that on the surface and in the surface layer between crystals of $NH_{4}ClO_{4}$ there are observed products of various degree of decomposition of napthalene, which will be formed, apparently, as a result of the reaction of gaseous products of decomposition of the oxidizer with products of decomposition and the liquid phase of the fuel - napthalene.

During the study of the burning of a mixture with an oxidizer base of ammonium perchlorate and combustible tungsten it was established that this mixture inflames and burns stably with ignition from an incandescent spiral at room temperature in the pressure region of $p \ge 5$ atm (abs.). On the surface of the extinguished sample (Fig. 1b) there is stored tungsten and products of its oxidation. The mixture with a base of NH₄ClO₄ and graphite does not burn stably in the region of pressures investigated by us up to 100 atm (abs.). With visual observation of the surface of the mixture after influence of incandescent spiral, it is clear that an accumulation of graphite occurs on it (Fig. 1b).

For mixtures with an oxidizer of ammonium perchlorate and fuels studied by us - malonic and succinic acids, napthalene, starch, graphite, tungsten - the ultimate pressure (lower than which the mixture does not stably burn and dies out) was determined depending upon the dimension of particles of $40-50 \ \mu m$ and $200-300 \ \mu m$ (see table).

From the table it is clear that the higher ultimate pressure for mixtures with a base of melted fuel of the type of malonic and succinic acids the less the particles of the oxidizer.

This phenomenon, apparently, is explained by the fact that the small particles are easily covered by a film of melted fuel, which protects and hampers heating of the oxidizer up to a temperature of violent decomposition (Fig. 1c).

Large particles of $NH_{4}ClO_{4}$ (Fig. 1c) are partially covered by a film of fuel, and their upper part is free. With burning of the mixture of $NH_{4}ClO_{4}$ + napthalene on the surface there is stored $NH_{4}ClO_{4}$, and the ultimate pressure of stable burning does not depend on the dimension of particles of the oxidizer.

Starch, as is known, spreads weakly with heating of it up to a temperature of decomposition, and the ultimate pressure of stable burning of the mixture $NH_{ll}ClO_{ll}$ + starch does not depend therefore on

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the dimension of the particle for the oxidizer.

Ultimate pressure of stable burning does not depend on the dimension of particles of the oxidizer and for mixtures on a base of ammonium perchlorate of refractory fuels of tungsten and graphite.

Results of Investigation of Mixtures with Oxidizers KClO_{li} and NaClO_{li} as a Base

For all mixtures investigated by us on oxidizers KClO_4 and NaClO_4 with burning a formation on the surface of the charge of a melted layer is observed. The reaction between the oxidizer and fuel begins in the melted layer or on its surface.

The mixture KClO_{\downarrow} + tungsten in a vacuum (p $\sim 10^{-2}$ mm Hg) at room temperature burns stably at a rate of ~ 0.0022 m/s.

It was not possible for us to extinguish this mixture neither in a vacuum nor in a region of increased pressures. In order to study the surface forming during burning the oxidizer $NaClo_4$ was taken. Mixtures on this oxidizer burn less intensively and are extinguished at a pressure of p < 1 atm (abs.) and in their properties the oxidizers are very close.

With the burning of this mixture there will be formed a melt from $NaClO_4$ and the product its decomposition NaCl. On the surface of



Fig. 2. Oxidizers $KClO_4$, $NaClO_4$. a) fuel; malonic and succinic acid, napthalene, starch; 1 - products of decomposition of fuels; 2 - melt of $KClO_4$ with KCl or

NaCl04 with NaCl;

3 - melted fuel; b) fuel; tungsten, graphite; 1 - fuel, and products of it oxidation; 2 - melt of KClO₄ with KCl or NaClO₄ with NaCl.

the melt there is disposed tungsten and products of its oxidation, i.e., on the surface of the melt there is stored thermoresistant fuel because of the great difference between temperatures of decomposition of the oxidizer and the fuel. Under the action of the incandescent spiral on surfaces of mixtures $KClO_{4}$ + graphite and $NaClO_{4}$ + graphite there is also observed an accumulation of graphite (Fig. 2a).

Mixtures of KClO_4 with graphite and NaClO₄ with graphite do not stably burn in the investigated region of pressures up to 100 atm (abs.). Mixtures of KClO_4 + napthalene and NaClO₄ + napthalene in a vacuum at $p \sim 10^{-2}$ mm Hg do not burn. They burn stably in air and at higher pressure.

On surface of the burning, consisting of melted $KC10_{4}$ and KC1 or $NaCl0_{4}$ and NaCl, there are located dark-brown products of decomposition of napthalene.

Under the melt of $KClO_{\downarrow}$ there is melted fuel - napthalene, and on their boundary - a layer of products of decomposition of napthalene of brown colors.

An analogous picture is observed on the surface of burning of mixtures KClO_4 and NaClO_4 with succinic acid and starch (Fig. 2a). For all mixtures with fusible (KClO_4) or infusible (NH_4ClO_4) oxidizers and fusible fuels in the surface layer of the melt are observed a great quantity of small bubbles, forming due to gaseous products of decomposition of the oxidizer and fuel and the liquid-phase oxidation of fuel as a result of occurring reactions in the condensed phase.

In the surface layer of the melt reactions are possible not only between gaseous products of decomposition of the oxidizer and gaseous products of the decomposition of the fuel, but also between gaseous products of the oxidizer and melted fuel in the reaction layer of the condensed phase, i.e., liquid-phase oxidation is carried out.

If the mixture consists of a fusible oxidizer and infusible fuel or of infusible oxidizer and infusible fuel, then the reaction of interaction between products of decomposition of the oxidizer and fuel flows on the surface of the condensed phase of the fuel.

In work [4] it is shown that the mechanism of burning NH_4ClO_4 is analogous to the mechanism of the burning of powders and mixture solid fuels [1, 2]. In heating of the charge of NH_4ClO_4 with burning there proceeds ~95% heat liberated in the reaction layer of the condensed phase, and about 5% heat proceeds from the zone of burning of the stack gas mixture.

Mixtures with oxidizers KClO_4 and NaClO_4 as a base burn considerably more intense than do mixtures on the basis of oxidizer NH_4ClO_4 . This distinction is explained by the fact that the temperature on the surface of the burning mixtures on the basis of oxidizers KClO_4 and NaClO_4 , equal to $\sim 600-700^\circ\text{C}$, are considerably higher than temperatures of the surface of mixtures on the basis of NH_4ClO_4 equal to 500°C , and besides this chlorides of metals K and Na forming during decomposition of KClO_4 and NaClO_4 are catalysts of the process of burning [5].

Thus from the obtained results it follows that the initial stage of burning for different mixtures of solid fuel is carried out in the reaction layer of the condensed phase and is completed by the formation of stack gas fuel mixture.

Conclusions

1. In the region of pressures of p < 30 atm (abs.) on the surface of the burning mixture on the basis of an infusible oxidizer $NH_{\mu}Clo_{\mu}$ with fusible fuel (or fuel decomposing at a temperature close to the temperature of decomposition of the oxidizer) crystals of $NH_{\mu}Clo_{\mu}$ come forward, and the more distinct the larger the crystals. At a pressure of $p \ge 30$ atm (abs.), the reverse is observed, namely: at the place where there protruded crystals of ammonium perchlorate depressions, craters, were formed.

2. In the process of burning, thermoresistant fuels (graphite, tungsten) are stored on the surface of mixture. If the temperature of decomposition of the oxidizer NH_4ClO_4 is higher than the temperature of sublimation or decomposition of the fuel, for example napthalene, the oxidizer is stored on the surface of the mixture.

3. In the surface layer of the melt of melted oxidizers and melted fuels there are observed numerous small bubbles which form due to gaseous products of decomposition of the oxidizer and fuel and liquid-phase oxidation of the fuel.

4. The ultimate pressure, lower than which the mixture does not stably burn (dies out), for the mixture on the basis of unmelted ammonium perchlorate depends on the dimension of particles of the oxidizer and physical properties of the fuel.

5. The mixture KClO_4 + tungsten stably burns in a vacuum of $p \sim 10^{-2}$ mm Hg at a temperature of $T \geq 5^\circ\text{C}$. The mixture NH_4ClO_4 + + tungsten stably burns at room temperature in the region of pressure $p \geq 5$ atm (abs.). Mixtures KClO_4 + graphite and NH_4ClO_4 + + graphite do not burn stably in the region of pressures up to $p \sim 100$ atm (abs.) investigated by us.

6. Experimentally it is shown that the initial stage of burning of mixtures of solid fuel is carried out in the reaction layer of the melt of the condensed phase or on the surface of thermoresistant fuels (tungsten, graphite) with total positive thermal effect and is completed by the formation of a stack gas mixture, which burns in the zone above the surface of mixtures to the end products of burning.

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THE DISTURBANCE OF NORMAL CONDITIONS OF THE BURNING OF POROUS CHARGES OF EXPLOSIVE SUBSTANCES

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It is known [1, 2] that the burning of porous charges can occur normally only under certain conditions. The lowering of the density of the charge, the increase in its dimensions and the increase in pressure under which burning occurs all promote the breakdown of normal burning and the transition of it to explosion. In this work the process of the appearance of explosion during burning of porous charges under conditions of increasing pressure was investigated.

Method

There was used a bomb of constant volume of 200 cm³. A change in the pressure in the process of burning was recorded by a piezotransducer through a cathode-ray oscillograph and photorecorder. Registration was produced also of the process of burning by its glow by a high-speed movie camera [SKS-1] (CHC-1) and photorecorders, high-speed and standard which could operate simultaneously. All recordings were synchronized. Charges of hexogene was used, which was pressed in transparent plexiglas glasses with an internal diameter of 0.008 m and a height of up to 0.06 m. There were also used charges of flegmatized hexogene (0.5, 2 and 10% by weight of paraffin). Ignition was produced or from electricspiral or from combustion of a batch of black powder.

Normal Burning

The opinion exists [3, 4] that normal burning of melting explosives to which a certain degree of hexogene belongs, can occur only until the layer of melt completely closes the pores of the charge.¹ For

¹As a result, porous charges of unmelted substances, in general, should normally burn, which contradicts the experiment [5].

checking this assumption we conducted the following experiments.

1. The difference of pressures in the volume of the bomb and in the pores of the sample was recorded [6]. Irrespective of the dependence on the pressure in the bomb the difference of pressures did not exceed $\sqrt{3}$ atm.

2. The charge through the durable incombustible grid had escape throigh the lower face to the atmosphere. With pressure in the bomb of the order of 1.5-2 atm the melt burst and products of burning were ventilated through the sample, which led to damping of the burning. The same charge with a closed lower end burned normally up to a pressure of the order of 25 atm.

3. Calculation of the condition of continuity of the layer of melt on the surface of charge, where it was assumed that the filling of the pores occurs due to the increase in specific volume of the melted substance, gives the following expression for p_{μ} -critical pressure of the disturbance of continuity of the layer of melt:

 $p_{\bullet}^{n} \leq \frac{12/\delta\lambda_{0}(\rho - \rho_{\bullet})}{(1 - \delta)D\rho}, \text{ if } u = Bp^{n},$

where u - burning rate of explosive; B - constant; δ - relative density; λ_0 - thickness of the layer of melt at 1 atm; D - average

dimension of particles of explosive; $\rho_{\#}$ — density melt of explosive; ρ — density of solid explosive. For hexogene with D = 200 µm and δ = 0.7 we have $p_{\#}$ % 3 atm. If we consider the influence of pressure of gases in the volume of the bomb on the layer of melt, then the true value of $p_{\#}$ will appear even less. Thus with burning at raised pressures (especially under conditions of increasing pressure) the layer of melt at low pressures ceases to play a role in the absolute barrier, protecting the pores from the penetration of gases. Filtration of gases of burning in pores of the charge should appear. However, photographic recordings of the burning show that in these conditions the process nevertheless continues to proceed normally. Consequently, the penetration of gases of burning in pores of charges in these conditions still does not signify penetration of burning.

What conditions must be fulfilled for penetration of burning in pores of charges? In the case of an ideal bomb of constant pressure (density of charging is equal to zero) the condition of disturbance of the front of burning consists in the commensurability of dimensions of unevenness (diameter of pores) and thickness of the thermal layer [7], i.e.,

$$p_{4}^{n} = \left(\frac{\lambda b}{c_{p}B} \sqrt{\frac{m}{\kappa}}\right),$$

where λ - coefficient of thermal conduction; b and n - constants; c_p - heat capacity; m - porosity; k - permeability

Strictly speaking, this formula is applicable only to infusible explosives.

In one of the wide-spread cases — with burning under increasing pressure — normal burning of a porous sample occurs until gases filtrable into pores of the charge heat the substance to a temperature of gasification in depth greater than the thickness of the thermal layer.

Development of the Burning Process Under Increasing Pressure

The conducted investigation exposed the following general picture of the transition of the normal burning of porous explosive charges



a) pure substance, $\delta = 0.7$; b) flegmatization of 2% parafin, $\delta = 0.7$. Dimensions of particles $\lambda \sim 200 \ \mu m$. ing of porous explosive charges into an explosion under conditions of increasing pressure.

Filtration of gases into pores leads on a certain stage of the process to the penetration into pores of burning. Channels of filtration, as the most prepared and heated, become the most favorable directions of the propagation of burning. As photographic recordings show, (Fig. 1), after a section of normal burning there appears disturbed burning, characterized by an eroded front and glow pulsations. The experiment showed that these conditions are caused by the process of penetration of burning in depth of the charge by separate most favorable

of burning in depth of the charge by separate most favorable pores, which leads to ejection from the surface and accelerated combustion in the flame of small portions (separate grains and pieces) of the charge. This phenomenon determines burning (Fig. 2). With an increase in pressure scales of the phenomenon increase. With a certain next ejection there occurs so strong a destruction of the front of burning that

Fig. 2. Highspeed movie film of the process of ejection from the surface of the charge of explosive particles, $\delta = 0.7$.

With an increase in pressure scales of the phenomenon increase. With a certain next ejection there occurs so strong a destruction of the front of burning that the secondary flame is rejected upwards. There appears a break in the glow, which is fixed on the After a period of induction, photographic recordings. which, as the experiment showed, linearly increases with an increase in dimensions of the grain, ignition occurs in particles of the explosives along the large surface. Rapid combustion creates above surface of the charge a jump in pressure, which forces the hot gases of burning with great speed to penetrate in depth of the charge. Simultaneously there occurs adiabatic compression of gas pockets in pores of the The results of penetration of gases of charge. burning will depend on what kind of heat reserve of gases there is, how fast in depth of the charge this heat will be given off, what the gas-permeableness of the charge is, and what is the period of delay of combustion of the given substance in the given conditions.

Influence of a Stabilizer

Introduction of a stabilizer in this case did not change the

P., | am 50 40 • 2 30 20 15 Q7 Q8 Q3 10 6 05 Fig. 3. Dependence p_# on relative density & for pure and flegmatized explosive: a) pure explosive; b) 0.5% parafin; c) 2% parafin; d) 10%

parafin.

pressure of the appearance of disturbed burning (Fig. 3) and the general picture of the development of the process with the exception of one, however, essential moment; even with the introduction of a total of 0.5% parafin instead of explosions there occurred only accelerated combustions of the remaining substance. As compared to the pure substance flegmatized explosive has smaller pores (with identical density), which decreases the specific surface of the charge. The period of delay of ignition of the ignited substance with 2% parafin increases approximately The filtration of gases occurs according twice. to enlarged pores. But unlike the pure substance the flegmatized pores inflame considerably slower, the rise in pressure in them is less, and the development of the explosive process is delayed (Fig. 4). Ejection of the flegmatized substance into the zone of the flame leads to its great cooling, since the parafin in its fusion removes heat. On the stage of burning of rejected particles the stabilizer promotes rapid heat removal for the filtering gases, hampers ignition of the charge along the great surface, which in the case of a pure substance produced explosions. Consequently, the basic effect of introduction of the stabilizer consists in the influence on the explosive stage of the

process that explosions are converted to accelerated combustion without mechanical effect. This conclusion in its meaning is analogous to the well-known position of the theory of sensitivity of explosives: the stabilizer basically affects the process of development of the explosion from the focus.



Fig. 4. High-speed photograph of the process of appearance of perturbed burning of flegmatized explosive: hexogene, 10% parafin. On the penultimate frame one can see damping of the glow above the surface of the charge.

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GRAPHIC NOT REPRODUCIBLE

Self-Ignition of Half-Finished Products of Burning in Pores of the Charge

On charges of pure hexogene with a dimension of the particles of about 200 μ m there are frequently observed a very unusual type of transition of normal burning into explosion. Under conditions when on the surface of the sample normal burning occurs, suddenly without the usual section of perturbed burning there is an explosion. The unfolding of this process by a high-speed photorecorder showed that at a depth of 3-5 mm from the surface there appears the focus of the glow from which upwards and downwards spreads the explosive process with speeds of 300-800 and 200-600 m/s respectively. Local pressure at point of the formation of the focus is estimated at $10^5 - 5 \cdot 10^5$ N/m²

Texplosions do not reach to end of the sample, and part of the substance 2 remains whole.

Further investigation permitted assuming that in this case self-ignition takes place of half-finished products of burning,

Fig. 5. Photographic recording of appearance of an explosion in pure hexogene in depth: $\delta = 0.75$, D = 200 m, speed of evolement 200 m/s. penetrating into pores of the charge, which are enriched due to the exothermic decomposition of walls of the pores induced by the heating by filtrating gases of burning. Because of the initial cooling action of the walls the process of self-ignition is braked as compared to its development above the surface of the charge. Figure 5 gives a typical recording of such a process. The dependence $p_{\#}$ for this case of transition of normal burning into explosion on the speed of accretion of pressure is expressed by a straight line the slope of which gives the characteristic time of the order of 0.7 s, and time of development of the process in the pores.

Conclusions

1. Transition of normal burning into explosion under conditions of increasing pressure occurs according to this scheme: normal burning — perturbed burning — ejection of part of the charge into the zone of the flame accompanied by a temporary cessation of glow, accelerated burning of rejected substance creating a jump in pressure above the surface of the charge, — breakthrough of hot gases into pores of the charge leading in the case of a pure explosive to an explosion but in the case of a flegmatized explosive, to an accelerated burning of the remaining part of the charge.

2. Flegmatization explosives leads to braking of the process of development and excludes, as a rule, transitions of burning into an explosion.

3. There is revealed a new type of transition of normal burning to explosion through self-ignition of half-finished products of burning and gasification in pores of the charge.

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