LITERATURE CITED

1. M. A. Korchagin and V. A. Podergin, "Study of chemical transformations during combustion in condensed systems," Fiz. Goreniya Vzryva, **15**, No. 3, 48-53 (1979).
2. X-ray Powder File, ASTM, Philadelphia (1972).
3. L. S. Berezhnoi, in: Multicomponent Oxide Systems [in Russian], Naukova Dumka, Kiev (1970), pp. 64-65.
4. M. N. Shaskol'skaya, in: Crystallography [in Russian], Moscow High School, Moscow (1975).
5. Yu. L. Pliner and G. F. Ignatenko, in: Reduction of Metal Oxides by Aluminum [in Russian], Metallurgiya, Moscow (1987).
6. V. V. Aleksandrov and M. A. Korchagin, "Mechanism and macrokinetics of reactions in ultrarapid synthesis."

ENHANCING THE PROPAGATION VELOCITY OF A FLAME FRONT IN AN ALUMINUM AEROSUSPENSION

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Results are presented from experimental and theoretical studies of the effect of particle microcapsuling on the major operational characteristics and phenomena associated with the ignition and combustion of aluminum aerosuspensions. It is found that microcapsuling can be used to enhance the reactivity of an aerosuspension and to lower the amount of the condensed phase in the combustion phases while conserving the initial dispersivity of the particles and the energy characteristics of the aluminum—air mixture.

Powdered aluminum (PA) is widely used in various engineering processes. Depending on the features of these processes, the specifications for the initial PA may differ greatly. At present, particle microcapsuling is one of the most promising methods for obtaining powdered materials with the required parameters. The purpose of this article is to analyze the effect of microcapsuling on the major operational characteristics and phenomena associated with the ignition and combustion of aerosols of PA particles.

Previous studies have shown that coatings obtained by chemosorption and polymerization of organic compounds on the surface of particles have the best characteristics [1]. In choosing the composition and type of coatings it was noted that they should be thermally stable, water repellant, and contain chemically active fluorine compounds among their pyrolysis products. These requirements are satisfied best by fluorine-containing organic compounds. Powdered ASD-4 aluminum was used as an initial products. The powder had the dispersion characteristics illustrated in Fig. 1 by plots of the probability distributions γ of the number and volume of the particles.

In order to establish the best type of coating we studied PA microcapsulated with different chemical compounds. The mass fraction of the coating does not exceed 4% of the mass of the initial PA. For convenience in analyzing the results given here, we introduce the following arbitrary designations: A1—ASD-4 for the initial PA; A2 — $Si[OCH_2(CF_2{-}CF_2)_3H]_4$, A3 — $Cl_2Si[OCH_2(CF_2{-}CF_2)_2H_2]_2$, A4 — $(CH_2{=}CH{-}CH_2{-}O)_2Si[OCH_2(CF_2{-}CF_2)_2H_2]$; A5 — for polymethyl fluoracrylate (PMFA); and A6 for polymethyl methacrylate (PMMA). The polymer structure and the thermal properties of these compounds and their breakdown products have been discussed in detail elsewhere [2].

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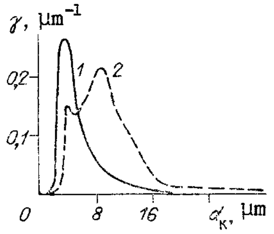

Fig. 1. Distributions of the amount (1) and volume (2) of the particles.

TABLE 1

Type of coating	d_{ij}, μm			
	d_{10}	d_{30}	d_{32}	d_{43}
A1	4.3	5,6	7,3	8,5
A3	4,2	5,2	6,4	7,6
A4	5.2	6,1	7,2	8,2
A5	4,5	5,4	6,4	7.3
A6	4.4	5,7	7,4	8,8

One of the major characteristics of powdered materials is their dispersivity. Since here we are studying a microcapsulated PA with a polydisperse distribution, we shall use the following statistical average diameters as basic geometric characteristics: mean number d_{10}, mean volume d_{30}, the Sauter diameter d_{32}, and mean mass d_{43} [3]. In order to determine the effect of microcapsuling on the fractional composition of the different batches of modified powdered Al, the powder was subjected to a dispersion analysis by means of sampling using an optical microscope. It is evident from the data in Table 1 that the average particle sizes of all the types of modified PA were essentially the same. Since in practice the particle size is the controlling parameter for various engineering processes, the retention of the initial dispersivity of the PA is fortunate.

As a parameter for characterizing the intensity of ignition and combustion of the powders that were tested, we have used the propagation velocity of a flame front, W_f, through an aerosuspension of PA particles. The experiments were conducted at atmospheric pressure on a apparatus which has been described in detail in [4]. The experimental technique is as follows: Al powder is continuously sprayed upward in a vertically positioned cylindrical working volume of length 1 m and diameter 0.088 m. The spraying device produces a uniform concentration of PA in the working volume, as confirmed by special sampling. The Al particles settle under the action of gravity. Pairs of oppositely positioned lamps and photocells were used to determine the speed of settling. When the working volume is completely filled with an aerosuspension, an ignition trigger in the form of an electric arc between carbon electrodes was applied to the open end of the working volume. A flame propagated uniformly upward from the open end to the closed end and the combustion products were drawn out into a receiver which served to maintain a constant pressure in the working section.

In this case the flame front propagates laminarly at a velocity that was determined from the time intervals between the signals from the photocells which were mounted in special windows spaced 100-mm apart along the length of the working section. Then the propagation velocity of the flame front in motionless PA was calculated by taking the sum of the settling velocity of the aerosuspension (0.8-0.9 m/sec) and the velocity of the flame front relative to the working section. The systematic error in determining W_f was 10.2%. The ratio of the components, which is characterized by the excess oxide coefficient α, was kept at 0.25. This value of a was chosen because in practice overrich aerosuspensions of PA particles were used quite often. In addition, this value of α is of interest for the prediction of explosion or fire hazards.

Our studies showed that microcapsuling of the PA particles makes it possible to increase W_f by a factor of 1.5-2 (Fig. 2). The rise in the reactivity is greater for a higher molar fraction g_F of fluorine in the coating (Fig. 3). The greatest effect is observed when the aluminum particles are microcapsulated with PMFA.

The improved combustibility of PA coated by fluorine-containing organic compounds is explained, in our opinion, by the following factors. During the initial stage of heating of the capsulated particles (up to temperatures of T = 700-900 K), the heat-resistant coating protects the Al from oxidation and the thickness of the oxide film on the particle surface does not change. As

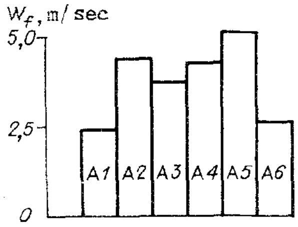

Fig. 2

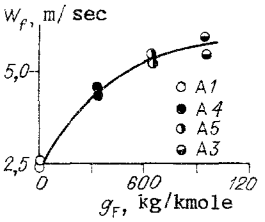

Fig. 3

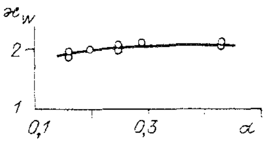

Fig. 4

Fig. 2. Propagation velocity of a flame front through aerosuspensions of modified PA; $\alpha = 0.25$.

Fig. 3. Dependence of W_f on the molar content of fluorine in the coating; $\alpha = 0.25$.

Fig. 4. $\varkappa_W$ as a function of the excess oxidant coefficient.

a result, after the coating disintegrates the diffusion resistance of the Al_2O_3 surface film is lower than for particles of pure PA [5]. Furthermore, when $T > 700$ K the disintegration of the organic coating may be accompanied by the release of atomic fluorine. Then an exothermic reaction of aluminum with fluorine, $Al + 3F \rightarrow AlF_3 + 56$ MJ/kg, takes place and intensifies the overall heat release process, leading to an increase in the burning velocity of the particles.* The resulting aluminum trifluoride facilitates the destruction of the continuity of the Al_2O_3 oxide layer. Given all these factors, we may conclude that the deposition of fluorine-containing layers leads to a reduction in the induction period for ignition of PA particles and to a growth in the reactivity, which in turn causes an acceleration in the propagation of the flame front.

We note that W_f was essentially unchanged for an aerosuspension of aluminum particles coated with PMMA. Since PMMA does not contain fluorine, this observation confirms the dominant role of the interaction between fluorine and aluminum in increasing the reactivity of PA microcapsulated with fluorine-containing compounds.

We have studied the effect of the excess oxidant on the efficiency of the modifications, as evaluated from the relative increase in the propagation velocity of the flame front, $\varkappa_W$. Here $\varkappa_W$ is the ratio W_f/W_f^0, where W_f^0 is the propagation velocity of the front through an aerosuspension of the original PA particles. As can be seen from Fig. 4, the effectiveness of the modification is almost independent of α. This is because when α is changed the total amount of fluorine in the aerodispersed system changes proportionately, but its mass concentration relative to the PA remains constant.

From the standpoint of optimizing the parameters and estimating the efficiency of different engineering processes, there is some interest in analyzing the effect of the modifying materials on the burning characteristics of the PA. In order to do this we have carried out a set of thermodynamic calculations using a special program which makes it possible to determine the equilibrium parameters of multicomponent heterogeneous systems [7]. As criteria for comparing the different modifications of Al powders we use the thermodynamic combustion temperature T* and the relative mass content of the condensed phase (c-phase) in the combustion products, z_c. The data of Table 2 show that for all the batches of modified PA, when the mass fraction of the coating z_p is raised to 6% the value of T* decreases by 0.4-0.6%. This happens because the fraction of Al in the modified aluminum—air system is reduced because of the coating material.

Since the calculations were done for an overrich aerodisperse system ($\alpha = 0.25$), the combustion process is characterized by the formation of both the oxide and the nitride of aluminum in the condensed state. For the initial PA their mass concentrations in the c-phase are 0.031 and 0.183, respectively. The nitride content is rather large because under these conditions nitrogen is not inert with respect to aluminum and the nitriding reaction has a significant effect on combustion. The possibility of producing condensed AlN during combustion of overrich aerosuspensions has been demonstrated experimentally before [8, 9].

We have found that for PA capsulated with fluoropolymers that the c-phase content is reduced by 2.3-25%. In all cases the ratio of the mass concentrations of condensed aluminum nitride and oxide is essentially the same. In some engineering devices, such as industrial burners, the presence of the c-phase in the combustion products leads, first of all, to a reduction in the energy characteristics and, second, to a deterioration in the environmental indices. In this regard the observed effect can be

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*This situation is in accord with the results of a study of the effect of fluorine on the ignition and combustion of PA [6].

TABLE 2

z_p	T^*, K for					z_c for				
	A2	A3	A4	A5	A6	A2	A3	A4	A5	A6
0	2540	—	—	—	—	0,214	—	—	—	—
0.02	2536	2536	2536	2536	2534	0,196	0.197	0,198	0.197	0.219
0,04	2532	2532	2531	2532	2528	0.178	0.181	0,182	0.181	0,224
0,06	2528	2528	2527	2528	2522	0,160	0.165	0,166	0.164	0,227

used to improve the operational characteristics of PA. When a coating based on PMMA is used, on the other hand, the content of the c-phase increases by 2-6%. This is because the composition of this coating material is poor in oxidant elements (molar fraction 32%) and is rich in fuel elements. Thus, K is lower and the chemical equilibrium of the system is shifted toward increased formation of the c-phase.

In conclusion, microcapsuling PA by fluorine-containing organic compounds (while retaining the initial dispersivity) makes it possible to increase the reactivity by a factor of 1.5-2 by reducing the induction period for ignition as a result of keeping the thickness of the oxide film on the capsulated particle constant during the initial phase of heating and as a result of increased heat release during ignition. An analysis of thermodynamic calculations shows that microcapsuling PA particles causes almost no change in the energy characteristics of aluminum—air mixtures and makes it possible to reduce the amount of the c-phase in the combustion products.

LITERATURE CITED

1. I. D. Solodovnik, Microcapsuling [in Russian], Khimiya, Moscow (1980).
2. I. L. Knunyants and V. A. Ponomarenko (eds.), Fluoropolymers [in Russian], Mir, Moscow (1975).
3. V. Ya. Bulanov, L. I. Kvater, T. V. Dolgal', et al., Diagnostics of Metal Powders [in Russian], Nauka, Moscow (1983).
4. V. M. Kudryavtsev, A. V. Sukhov, A. V. Voronetskii, et al., "A study of high-temperature two-phase flows," in: High-Temperature Gas Flows, Production and Diagnostics [in Russian], Khar'kov (1983), Vol. 3, p. 79.
5. P. F. Pokhil, A. F. Belyaev, Yu. V. Frolov, et al., The Combustion of Powdered Metals in Active Media [in Russian], Nauka, Moscow (1972).
6. T. I. Kolpakov and V. I. Glukhov, "A study of low-temperature oxidation of aluminum," in: The Physics of Aerodispersed Systems [in Russian], Kiev (1977), Vol. 16, p. 60.
7. G. V. Sinyarev (ed.), A Method, Universal Algorithm, and Program for Thermodynamic Calculations of Multicomponent Heterogeneous Systems [in Russian], Moscow, Proc. MVTU, No. 268 (1978).
8. A. G. Alekseev, I. V. Sudakova, and T. I. Tsidelko, "An experimental study of the dynamics of the interaction of oxygen with metals during explosions of aerosuspensions of metal powders," in: The Physics of Aerodispersed Systems [in Russian], Kiev (1986), Vol. 30, p. 20.
9. D. A. Yagodnikov, A. V. Sukhov, V. I. Malinin, et al., "The role of the nitriding reaction in flame propagation through overrich metal—air mixtures [in Russian], Vestn. MGTU, Ser. Mashinostr. (1990), No. 1, p. 121.