

## IGNITION AND COMBUSTION OF ALUMINUM AND ZINC IN AIR

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The ignition and combustion processes of aluminum have been investigated relatively widely. However, the existing experimental data do not permit these processes to be described completely. This is explained by the fact that, because of methodological difficulties in the investigation of relatively high-temperature ignition and combustion processes of aluminum, few quantitative characteristics have been measured and these frequently indirectly. In particular, when using the procedure based on gas burners, tracking facilities, furnaces, simulator engines, etc., instead of the temperature of the particles at the instant of ignition, the temperature of the ambient gas medium has been observed. The mechanisms of self-heating of a metal particle can be determined with sufficient reliability only by measuring the temperature change of a particle.

The basis of the procedure used in this paper lies in the measurement of the temperature of a metal sample during its heating up, ignition, and combustion, using thermocouples. Ignition of samples of aluminum was carried out using an induction heating method, making it possible to heat up the samples to high temperatures (~2400°C). The ignition and combustion processes of zinc were also investigated. As the main features of the high-temperature oxidation of aluminum and zinc coincide, a similarity in the nature of the course of their ignition and combustion might be expected. However, these processes take place in the case of zinc at significantly lower temperatures. This has made it possible to carry out the investigations in the case of zinc, both under conditions of induction heating and by heating in a hot oxidant gas stream, which allowed the results obtained by the two methods to be compared.

Spherical samples of zinc (2 to 3 mm) and aluminum (3 to 4 mm), with a 99.8 and 99.5% purity, respectively, were used for the experiments. In the initial state, the samples were covered with an oxide film formed at room temperature. The investigations were conducted by heating up the samples in an inductor (with cold gas blowing over them) and in a stream of hot gas blown through a tubular electric furnace.

The scheme of the experiments under induction heating conditions is described in [1]. In order to carry out the experiments in the hot oxidizing gas stream, a type T-40/600 tubular electric furnace, with a working space diameter and length of 4 and 75 cm, respectively, was used; this was installed vertically. The maximum working temperature was 1200°C, which allowed this same gas flow temperature to be obtained. The oxidizing gas was fed in from a cylinder and was directed from below upward. The upper part of the furnace was sealed with an insert of quartz glass, with openings for the gas outlet and for introducing the sample. The samples investigated, suspended on a thermocouple, were inserted in the furnace at a distance of 10 cm from the end.

Air was used as the oxidizing gas, with a flow velocity of 15-20 cm/sec. All experiments were conducted at atmospheric pressure. In the course of the experiments, the temperatures of the sample and gas were measured in the stream at a distance of 2 to 4 mm from the sample.

The temperatures were measured with tungsten-rhenium thermocouples with diameters of 0.35 and 0.05 mm. The ends of the thermocouples were not welded, but were fixed into the sample in previously drilled holes; i.e., contact between them was not direct but through the metal being studied. The signal from the thermocouples was applied to an amplifier with a high input resistance (70 kΩ), and then recorded on an N-105 loop oscillograph.

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TABLE 1

Region	$t_g, ^\circ\text{C}$	$t_s, ^\circ\text{C}$	$\tau_g, \text{sec}$	Remarks
I	896	—	—	Ignition absent
	915	—	—	
	928	—	—	
	930	—	—	
II	925	874	—	Ignition and extinction
	930	883	—	
	940	885	—	
	962	890	—	
	970	875	—	
III	980	886	9,1	Ignition and combustion
	980	885	8,0	
	1020	870	5,4	
	1056	880	4,6	
	1290	880	1,25	

The absolute measurement error of the temperature by the thermocouple in the experiments on aluminum amounted to  $\pm 15^\circ$  (over the range of temperature variation 20-2500°C), and in the case of zinc  $\pm 7^\circ$  (over the range 20-1200°C).

A photocell with a set of light filters was used to record the instant of the appearance of a flame (flash). Colored light filters covered the infrared region of the spectrum and neutral filters controlled the sensitivity of the system. The temperatures of the sample and of the gas and the instant of flash were recorded simultaneously on the loop oscillograph.

Similar measurements were conducted during heating up of the samples in the inductor. The conditions under which the experiments were conducted in this case differed significantly from the conditions of heating of the samples in the hot gas stream. When heating up the samples in the inductor, the gas stream was blown in at room temperature (at a velocity of 15-20 cm/sec), and the samples were heated up by the release of heat from induction heating in the surface layers of the metal sample (skin-effect mechanism). This method ensured that the samples were heated right up to the boiling point of aluminum (2410°C), i.e., up to considerably higher temperatures than in the case of heating in the tubular furnace.

In the experiments, depending on the gas temperature (during heating in the hot stream) or on the intensity of the heating in the inductor, the ignition (or lack thereof) the metal samples was observed. In both cases, the temperature-time curve of the samples was recorded by means of the thermocouples.

When investigating the samples of zinc in the hot air flow, the heating curves shown in Fig. 1 were obtained. The section *ab* corresponds to melting; *bc* corresponds to further heating-up of the liquid sample, covered by a solid-oxide film. At the points *c* and *c'*, the appearance of a flame around the sample is observed, which is accompanied by an intense loss of finer condensed products in the stream and by an increase in the rate of heating of the sample. At this point, the photocell records a sharp change of brightness. The temperature of the sample corresponding to this point is the flash temperature ( $t_s$ ) and is simultaneously the critical ignition temperature [2]. The sections *cd* and *de* are the onset of combustion and the combustion of the sample, respectively. In the case of total combustion of the sample, on arrival at the combustion cycle at the point "d," its temperature reached 910°C, and in the case of combustion with extinction the temperature at this point reached 900-904°C.

In the case of combustion with extinction, hollow compact oxide spheres, covered inside with a layer of metal and on the outside with a relatively thin friable layer of oxide were observed on the thermocouples; this, obviously, is the result of deposition of finely dispersed particles. In the case of total combustion, the thickness of the oxide spheres was considerably less and the presence of metal was not observed.

The experimental data, corresponding to these different cases of behavior of the zinc samples with a diameter of 2 mm, are given in Table 1. The temperature of the gas flow  $t_g$  is divided into three regions. In the first of them, ignition is not observed and the temperature of the sample reaches a maximum value, after which some decrease is observed (see Fig.

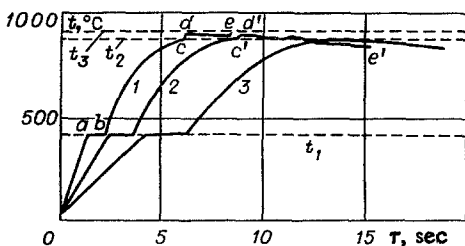


Fig. 1

Fig. 1. Curves of temperature of samples of zinc versus time, for their ignition and combustion in a hot air flow ( $t_1$  and  $t_3$  are the melting and boiling points of Zn;  $t_2$  is the flash temperature): 1) ignition and total combustion of sample; 2) ignition and extinction; 3) no ignition.

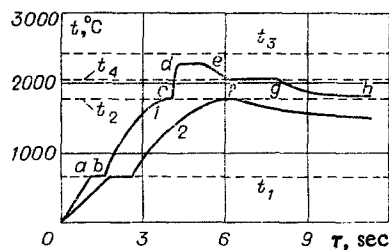


Fig. 2

Fig. 2. Curves of temperature of samples of aluminum versus time, in the case of their induction heating in a stream of cold air ( $t_4$  is the melting point of  $Al_2O_3$ ): 1) Ignition with subsequent extinction of combustion; 2) no ignition.

TABLE 2

Re-gion	$dt/d\tau$ , °C/sec	$t_{max}$ , °C	$t_z$ , °C	$t_{cm}$ , °C	Remarks
I	337	1420	—	—	Ignition absent
	351	1470	—	—	
	367	1560	—	—	
	373	1670	—	—	
	379	1765	—	—	
II	377	—	1780	2160	Ignition and combustion with extinction
	395	—	1775	2190	
	455	—	1785	2250	
	575	—	1770	2260	
	695	—	1770	2290	

1, curve 1). This decrease may be due to an increase of heat loss by radiation because of an increase of the coefficient of emittance during growth of the oxide film on the sample.

Over the temperature range 930–980°C, ignition with subsequent extinction of combustion was observed. Above 980°C, the samples are ignited and burn completely. The flash temperature is independent of the temperature of the surrounding gas and amounts to  $880 \pm 10^\circ\text{C}$ , but during combustion  $\tau_g$  varies from 9.1 to 1.25 sec with increase of the air temperature. Consequently, with increase of temperature of the surrounding medium, a significant increase of the rate of combustion occurs.

During heating up of the samples of zinc in the inductor, where they were blown with cold air, the same three situations were obtained, depending on the quantity of induction heat fed into the samples. The measured flash temperature also amounted to  $880 \pm 10^\circ\text{C}$ .

It can be concluded that in the processes investigated — ignition and combustion — the method of heating does not have a marked effect, but they are determined by the temperature of the sample. In the investigations this allows induction heating to be used the same as heating by a hot gas stream.

The samples of aluminum could not be ignited in the furnace; therefore, the investigation was carried out only in the inductor. The temperature curves of the heating-up of the samples of aluminum, while blowing them with cold air, are shown in Fig. 2. Qualitatively they are similar to the curves shown in Fig. 1 for the ignition and combustion of zinc in hot air. The difference is observed in the quantitative characteristics, in that for aluminum only two types of curves are obtained. The curve related to the case of complete combustion of the sample could not be obtained (because of procedural difficulties); however, on the possibility of its existence, an extension of the section of stable combustion  $dc$  with increase of the intensity of heating has been shown, and the approach of the temperature of the sample during combustion to the boiling point of aluminum, and the reduction of the quan-

tity of unburned aluminum in the residues observed on the thermocouples. The latter had a form similar to the residues obtained in the experiments with zinc in the case of combustion with extinction. The hollow, compact (vitreous), oxide spheres were covered on the inside with a layer of metal and on the outside with a thin friable layer of oxide.

During heating-up of the samples of aluminum, starting from  $\sim 1450^{\circ}\text{C}$ , the loss of condensed reaction products into the stream was observed on the section bc. This confirms that the oxide film becomes permeable at lower temperatures than the ignition temperature of the metal. Below the permeable oxide film, its state is understood to be one in which it insulates the samples from the surrounding atmosphere not completely, but has constantly fluctuating-through-pores or fissures.

With aluminum, just as with zinc, a kink in the temperature curve is observed at the point c, which indicates a sharp intensification of the removal of condensed reaction products into the flow. The photocell at this point records a sharp change of brightness — a flame appears round the sample. The section cd is the arrival at the combustion regime, de is combustion, efgh is the extinction of the sample, and fg corresponds to solidification of the oxide.

The appearance of a similar plateau on arrival of the sample at the combustion regime also might be expected; however, it was absent on the section cd, which can be explained by the relatively small quantity of oxide on the sample. Obviously, only the accumulation of a considerable quantity of aluminum oxide on the samples in the course of combustion conditions the effect of its phase transition on the temperature of the samples.

The experiments to investigate the interaction of aluminum with air (with induction heating), just as in the case of the interaction of zinc with air, were conducted at different intensities of sample heating. The intensity of heating ( $dt/d\tau$ ) was determined by the slope of the temperature curves on section Oa (see Fig. 2).

The results of the experiments for samples with a diameter of 3.5 mm are given in Table 2. Ignition is absent right up to the maximum temperature of the samples  $t_{\text{max}} = 1765^{\circ}\text{C}$  (ignition occurs if this temperature is exceeded), so that as a result of changing the intensity of heating from 377 to  $695^{\circ}\text{C}/\text{sec}$ , the temperature of the sample at the steady-state cycle of combustion  $t_{\text{com}}$  (section de in Fig. 2) also increases from 2160 to  $2290^{\circ}\text{C}$ , thus approaching the boiling point of the metal. The flash temperature  $t_{f1}$ , remains almost constant. This is observed also in the case of ignition of zinc. For the aluminum samples with a diameter of 3 to 4 mm,  $t_{f1} = 1780 \pm 20^{\circ}\text{C}$ .

From consideration of the ignition process of samples of aluminum and zinc, which is characterized by the kink in the temperature curve, the increase of brightness (recorded by the photocell), and also the visually observed sharp intensification of the removal of condensed products into the stream, it can be concluded that ignition of these metals, obviously, is due to vapor-gas reactions.

Up to now, it has been supposed that a surface reaction is responsible for ignition of metals. On the basis of this assumption, a series of theoretical models has been constructed. For aluminum, in particular, models exist with losses of the shielding properties of the oxide film (fusion or spallation) [3, 4], and a model with a variable thickness of the oxide film [5].

It can be seen in Fig. 2, curve 1 that the ignition temperature of the samples is considerably lower than the melting point of the oxide. Likewise, spallation cannot be the cause of ignition of aluminum because, as shown above, the oxide film on the samples is not protective long before ignition.

Experiments were carried out with the complete removal of the oxide film from the surface of the metal at temperatures immediately before the ignition temperature ( $1300\text{--}1500^{\circ}\text{C}$ ). For this, the samples of aluminum were previously heated in a stream of argon. In this way, aluminum devoid of oxide film is obtained. With the subsequent replacement of the argon by oxygen or air, a new oxide film grows rapidly on the sample; however, ignition of the sample and a significant rise of temperature were not observed. This phenomenon raises doubt on the essential role of a surface oxidation reaction in the ignition of aluminum.

In order to explain the effect of the thickness of the oxide film on the ignition temperature, samples of aluminum were previously oxidized at variable temperatures and for variable times so that the thickness of the film was increased up to  $20\ \mu$ . Then the samples

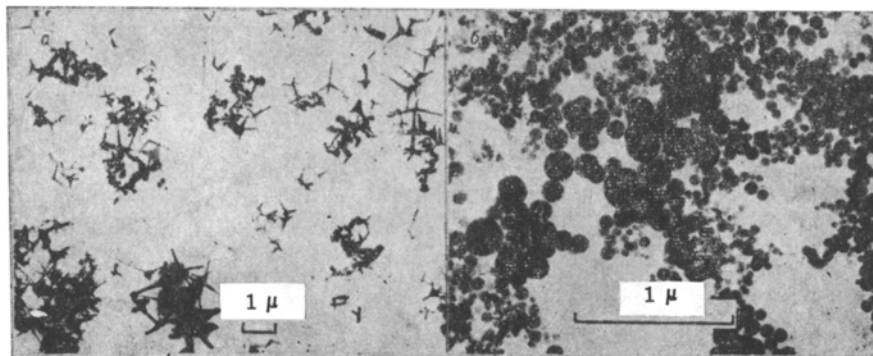


Fig. 3. View of condensed vapor-gas reaction products during the combustion in air of (a) zinc and (b) aluminum.

were ignited by the usual method. It was found that the ignition temperature was unchanged by comparison with the samples without previous oxidation. This result confirms the conclusion drawn earlier about the permeability of the oxide film in the period preceding ignition. The film on zinc in this period also is permeable [2].

During the investigation of the combustion of aluminum and zinc in this paper, two significantly different mechanisms have emerged: a stable cycle of combustion and combustion with extinction. They appear fully during the combustion of zinc.

From a comparison of the results obtained, it can be concluded that if the ambient conditions ensure that after ignition the metal is heated up to temperatures close to the boiling point, then its complete combustion takes place. If the case is otherwise, combustion dies down with time and the metal is only partially burned. Thus, in the case considered, the conditions under which combustion proceeds, and not the fact of ignition of the metal, determine its complete or incomplete combustion.

An analysis of the finely dispersed vapor-gas reaction products, carried away in the stream, has been carried out (Fig. 3).<sup>\*</sup> The combustion products of zinc consist of needle-shaped formations with sizes of not less than 3  $\mu$ . During the combustion of aluminum, hollow spherical reaction products are formed, with  $d \sim 0.5 \mu$ . The presence of these products, and also the analysis of the oxide residues in the thermocouples, indicate that during the combustion of aluminum and zinc in air, the reaction proceeds both on the surface of the metal and also in the gas space surrounding it. Part of the space reaction products is deposited on the surface.

In the scheme for describing the combustion of aluminum, the authors of [6] start from the possibility of simultaneous existence of surface and vapor-phase reactions. However, the application of this scheme is confined essentially to small particles. In the present paper, a similar course of the combustion process has been shown also for large-scale samples of aluminum.

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<sup>\*</sup>The photographs were taken by V. I. Shevtsov and V. P. Fursov.