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PROJECT SQUID

TECHNICAL REPORT ARC-8-P

TECHNIQUES FOR THE STUDY OF COMBUSTION OF BERYLLIUM AND ALUMINUM PARTICLES

By

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February 1964

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Technical Report ARC-8-P

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A COOPERATIVE PROGRAM OF FUNDAMENTAL RESEARCH
AS RELATED TO JET PROPULSION
OFFICE OF NAVAL RESEARCH, DEPARTMENT OF THE NAVY

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DEPARTMENT OF AEROSPACE ENGINEERING
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ABSTRACT

Minimum ignition temperatures for single beryllium particles, under total pressures of up to 50 atmospheres, range from 2380° to 2650°K depending on the partial pressure of oxygen in the ambient gas. Atmospheric pressure photographic studies of fine details of aluminum combustion have also been made and are discussed qualitatively.

The work described herein deals with a fundamental investigation of ignition and combustion of single metal particles of diameters ranging from 30 to 45 microns. Although different experimental procedures have been used, the essence of this study has always been the same, and it is quite simple. Metal particles of known sizes and shapes are suddenly plunged into hot gases of known physical and chemical characteristics, and their behavior is observed.

Techniques needed for this work were to a large extent developed and used previously in a study of combustion of aluminum particles, and much of this development and application has been described in earlier publications.[1,2]

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The purpose of this paper is (1) to present the application of a previously developed technique to the study of beryllium particle ignition, and (2) to describe a recently developed technique for observation of fine details of metal particle burning which has so far been applied to aluminum only. Efforts are now being made to extend this technique to combustion of beryllium.

1. Ignition of Beryllium

a. Background

The main experimental problems in the study of beryllium ignition and combustion are the high ignition temperature of the metal and the high toxicity of both the metal and its combustion products. The combustion characteristics of the metal are largely determined by the thermodynamic properties of the metal and its oxide (BeO), listed in Table I. Since BeO forms a protective layer, one might expect, in analogy to the results previously obtained with aluminum,[1] that the ignition process will be influenced by the melting of beryllia. It is interesting to note that the normal boiling point of the metal is quite near the melting point of the oxide, and this may also be a determining factor in ignition. Indeed, as will be seen below, techniques necessary for a quantitative study of beryllium combustion must allow generation of controllable ambient temperatures approaching these high values. The boiling point of beryllia, about 4100°K, presumably determines the metal flame temperature.

b. Experimental Procedure

The procedure was to admix small amounts of the metal powder to a combustible mixture consisting of a fuel and an oxidizer, which was then burned

in loosely tamped powder form (about 1 gm/cc) in a pressurized bomb. The oxidizer was ammonium perchlorate. The fuel was trioxymethylene, a volstile compound, which makes even quite fuel-lean, relatively cool, mixtures flammable at atmospheric pressure. The smount of beryllium added, 0.05 per cent by weight, was so small that it did not perturb the thermodynsmic properties of the oxidizer-fuel mixture to any appreciable extent. A dilute stream of metal particles thus ignited and burned in the combustion product gases conaiating of known proportions of H_2O, CO_2, N_2, HCl, O_2, and free atoms and radicals. Temperature of the gases for several constant pressures wss computed by an IBM-7090 program with the usual assumption of equilibrium adiabatic conditions, and it was also measured, at atmospheric preasure, by the spectral line reversal method. The measured temperatures were 60 to 70°K below the computed ones.

Safe handling of beryllium and its combuation products presents a major problem. An appaiatus conforming to stringent industrial hygiene requirements, shown in Fig. 1, has therefore been constructed which allows the beryllium-containing powder to be ignited in sealed capsules. Dilution required for diaposal of combustion producta ia 1 part of beryllium in s million parts of water or 25 micrograms of beryllium in one cubic meter of air. The combuation products contained in the sir-tight burner are therefore flushed first with a stream of nitrogen through dry filter, and then with generous amounts of water, all without opening the burner. Frequent analytical safety checks are necessary.

The procedure of determining the ignition limit aa a function of ambient gas temperature and oxygen content can be illustrated by reference to Fig. 2

which describes these ambien conditions corresponding to mixtures of ammonium perchlorate and trioxymethylene at three pressures; the percentage of free oxygen in the ambient gas is virtually independent of the total pressure so that a single oxygen-content curve suffices for all pressures. As long as the mixture is fuel-lean, progressive addition of fuel to oxidizer increases the temperature and decreases the oxygen content of the product gas. The procedure is continued up to the temperature at which a significant fraction of the metal particles ignite. This temperature and the corresponding partial pressure of oxygen are the ambient conditions which define the ignition limit of the metal.

c. Results

The results of ignition of beryllium particles, size range 30-45μ, are shown in Fig. 3. The total pressure in the above described experimental procedure is not an independently variable parameter, and it varies from point to point in the figure from 2.4 to 50 atmospheres. It can be seen that the metal ignites at temperatures as low as 2380°K when the partial pressure of oxygen is several atmospheres, and only above 2650°K when it is down to about 0.1 atm; this means that in fuel-rich mixtures the temperature limit for ignition of beryllium approaches the melting point of BeO (2820°K). Thus it appears possible that, in a close analogy to the case of aluminum, the temperature of the beryllium particle rises by self-heating above the ambient temperature to the melting point of the oxide, at which point ignition occurs. Since the normal boiling point of beryllium is slightly below the melting point of the oxide, boiling of the metal introduces an additional process to reckon at low pressures. Further work, which is in progress now, may give

basis for firmer judgment.

The mode of combustion of beryllium particles was also found to be affected by the ambient conditions. Large excess of oxygen appears to accelerate the burning. At low oxygen content, low pressures, and high temperatures, metal particle tracks are sharp and straight on photographic exposures, a characteristic of surface combustion of metals. At high oxygen content, high pressures, and low temperatures, the light from the burning particle is often intermittent and fairly diffuse, characteristic of gaseous combustion. These trends are illustrated by photographs taken through the window of the closed bomb (Fig. 4).

2. Photographic Study of Particle Combustion

a. Experimental Technique

In this technique metal particles are injected into laminarly flowing hot gases, generated by a flat-flame burner. The burner and the properties of the gases it generates were described in detail before.[1,2] Briefly, the hot gases consist of known proportions of H_2O, CO_2, N_2, and O_2, which are independently variable to a significant extent; the temperature can be varied from 1900 to 2800°K; the total pressure is restricted to 1 atmosphere.

The diagnostic technique for studying fine details of particle combustion consists of still photography through a magnifying lens system. The optical system has a standard 48 mm focal-length objective lens and a 10X magnifying eyepiece. Photographs are taken by a Reflex Leitz Microscope camera using 35 mm TRI X film. The depth of focus is 0.5 mm and the field of view 4.5 mm. The optical system is mounted on a cathetometer at a slight angle off the horizontal so that the vertically rising particles cross the

focal plane at a shallow angle. This system gives a magnification of film of 8 X. Satisfactory prints have been obtained with magnification factors up to 50. Positioning of the lens at various heights above the burner plate thus allows observation of the entire history of the particle from ignition to burnout.

b. Results

The photographic technique has been applied to combustion of spherical aluminum particles, size range of 30-30μ, and has revealed many complex features of the process. It had been found previously[2] that there are distinct effects on combustion of aluminum particles of both oxygen and water vapor in the hot ambient gases. Oxygen promotes vigorous combustion, and if its concentration is sufficiently high, there is fragmentation of burning particles. It had also been found that in virtual absence of water vapor there was much diffuse luminosity surrounding the burning particle, suggesting extensive vapor phase reaction, while in the presence of water in amounts larger than about 0.1 atm the luminous reaction is confined to a region closely adjacent to the particle. These effects have now been observed in close detail, as illustrated by Figures 5, 6, 7, 8, and 9.

Figures 5 and 6 show successive stages of particle combustion in low-moisture and high-moisture gases respectively. The oxygen content in both cases is relatively low so that burning is smooth (no fragmentation). It can be seen that the stages in the two figures are qualitatively similar. In the early stages following ignition, frames a and b in both figures, the burning appears even. The central intensely luminous core -- 30-50μ wide, corresponding approximately to the particle diameter -- is surrounded by a less luminous

region. The striking difference between Figures 5 and 6 is the appearance of the outer region, which is much wider and more diffuse in the absence of water. This difference in appearance persists into the later stages (Fig. 5c and 6c) in which the particles rotate. Fig. 6d records a particle at or near burnout; rotation has ceased, there is no outer region, and luminosity of the central core is dwindling. These progressive stages are typical of burning aluminum particles in hot gases whose oxygen content is too low to cause fragmentation.

Figures 7, 8 and 9 show comparable phenomena in high-moisture and low-moisture gases, side by side. The temperature in these figures is about 2400°K, and there is sufficient oxygen to cause some particles to undergo fragmentation (Fig. 9). The difference in the amount of diffuse luminosity between the adjacent photographs differing in amounts of water vapor is quite pronounced in all three figures.

There does not seem to be a ready explanation for these complex processes. However, we found in our earlier work[2] that, while in the presence of appreciable amounts of water vapor combustion of aluminum particles is accompanied by formation of hollow bubbles (reported earlier by Fassell *et al*[3]), there is no evidence of such a process in low-moisture gases. This fact and the fact that in the presence of water vapor the outer, less luminous, region is sharply delineated on the outside (e.g. Figures 6b and 7b), suggest that the outer layer luminosity may represent different things. In the presence of water, where combustion is confined to a region close to the surface and takes place asymmetrically with formation of bubbles, the dim outer layer is perhaps a relatively cool condensed phase portion of the burning system. In

the absence of water the burning is probably true vapor phase reaction, which indeed one should expect for aluminum on the basis of the relative volatilities of the metal and the oxide.[4]

Work is now in progress to apply the gas-burner photographic techniques to combustion of beryllium particles.

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1. R. Friedman and A. Maček, Combustion and Flame, 6, 9 (1962).

2. R. Friedman and A. Maček, Ninth Symposium (International) on Combustion, pp 703-712. Academic Press, 1963.

3. W. M. Fassell, C. A. Papp, D. A. Hildenbrand, and R. P. Sernka, Solid Propellant Rocket Research, pp 271-278. Academic Press, 1960.

4. I. Glassman, 14th Annual Meeting of American Rocket Society, Washington, D. C., November 16-20, 1959.

TABLE I

Thermodynamic Properties of Be and BeO

(JANAF Thermochemical Data)

	Be	BeO
Melting point (^{o}K)	1556	2823
Boiling point (^{o}K)	2757	(4123)
Heat of fusion (kcal/mole)	2.8	(14)
Heat of Vaporization (kcal/mole)	71.1	(143.5)
Density (gm/cc)	1.82	3.02

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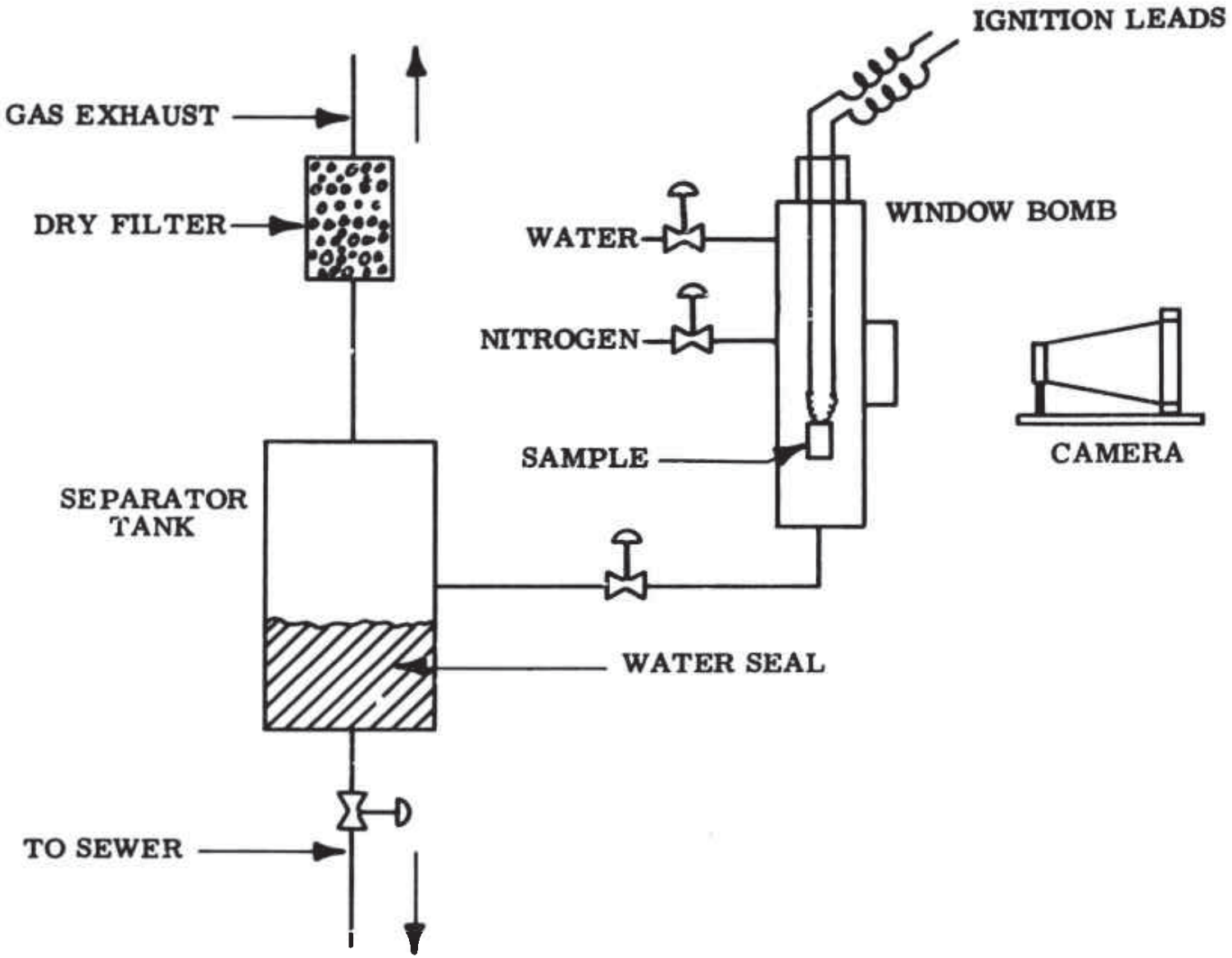

Fig. 1 APPARATUS FOR COMBUSTION OF BERYLLIUM UNDER PRESSURE

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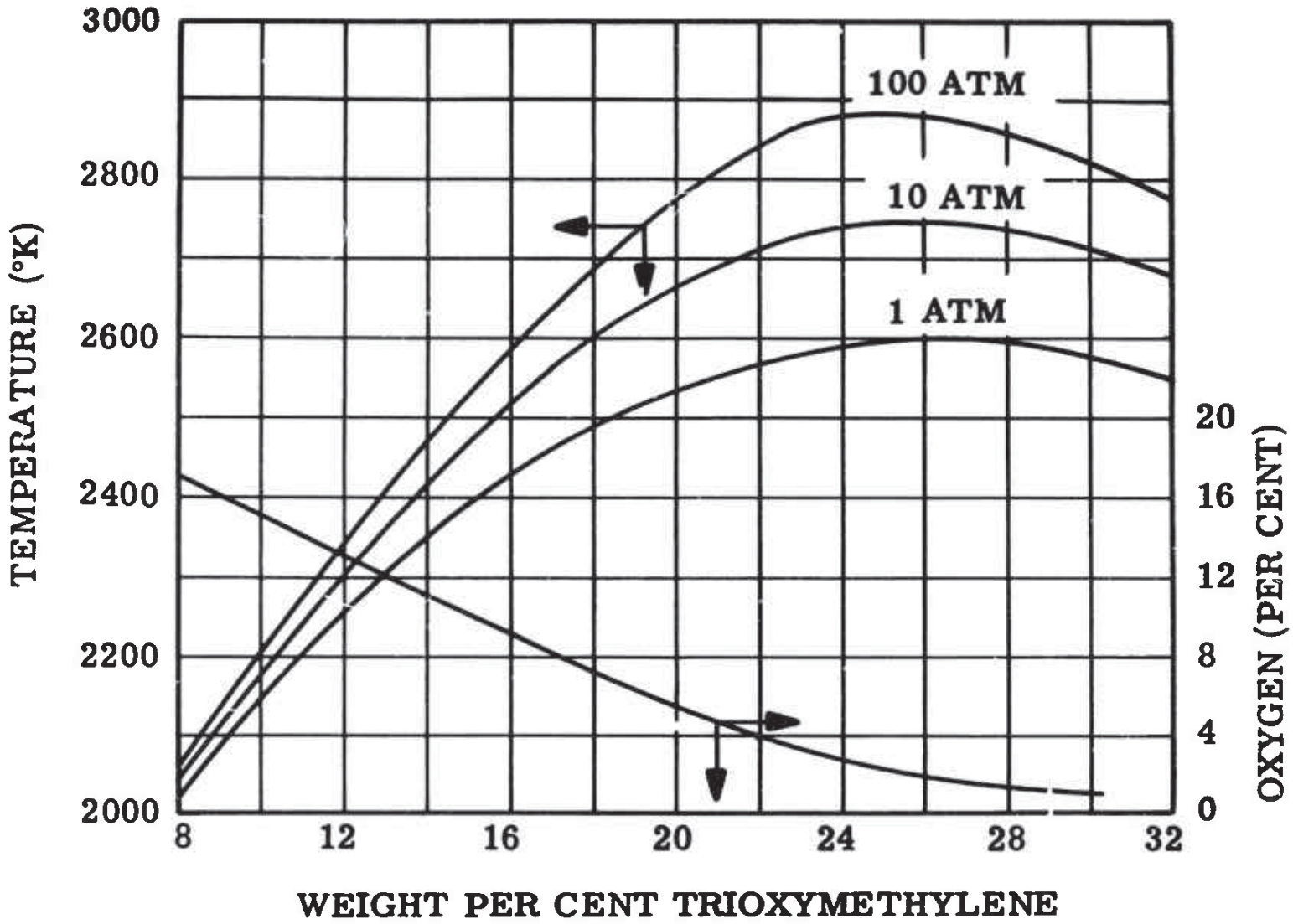
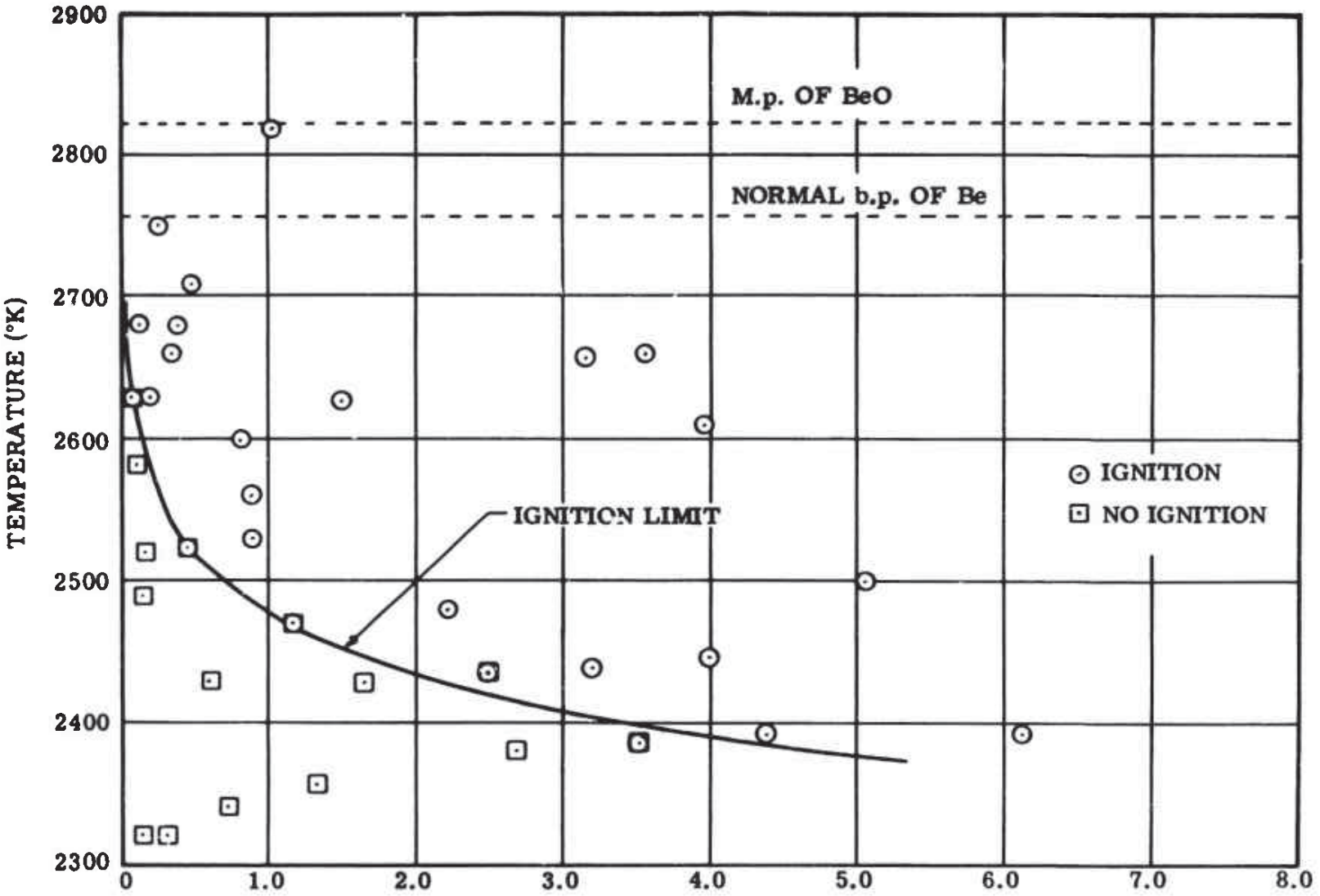


Fig. 2 **CALCULATED ADIABATIC FLAME TEMPERATURE OF AMMONIUM PERCHLORATE-TRIOXYMETHYLENE MIXTURES AT SEVERAL PRESSURES**



IGNITION OF BERYLLIUM PARTICLES (DIAMETER RANGE 30-45 MICRONS)

Fig. 3.

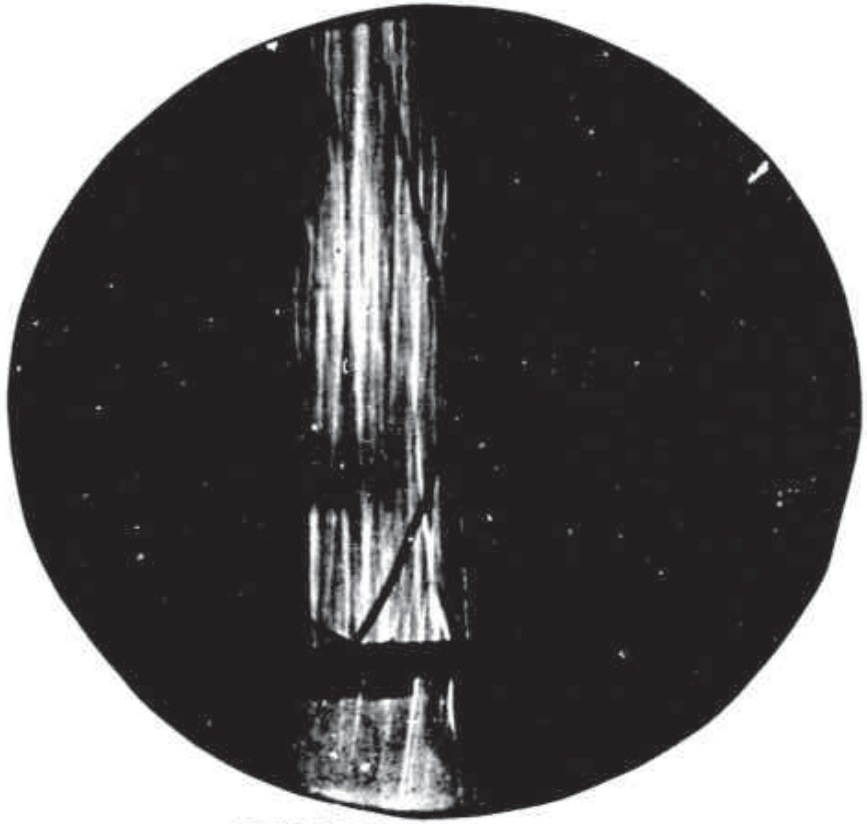
13% $C_3H_6O_3$

87% NH_4ClO_4

T = 2380°K

O_2 Partial Pressure = 6.12 Atm.

700 # psig



21.5% $C_3H_6O_3$

78.5% NH_4ClO_4

T = 2660°K

O_2 Partial Pressure = 0.335 Atm.

100 # psig

Fig. 4. COMBUSTION OF BERYLLIUM PARTICLES IN A CLOSED BOMB

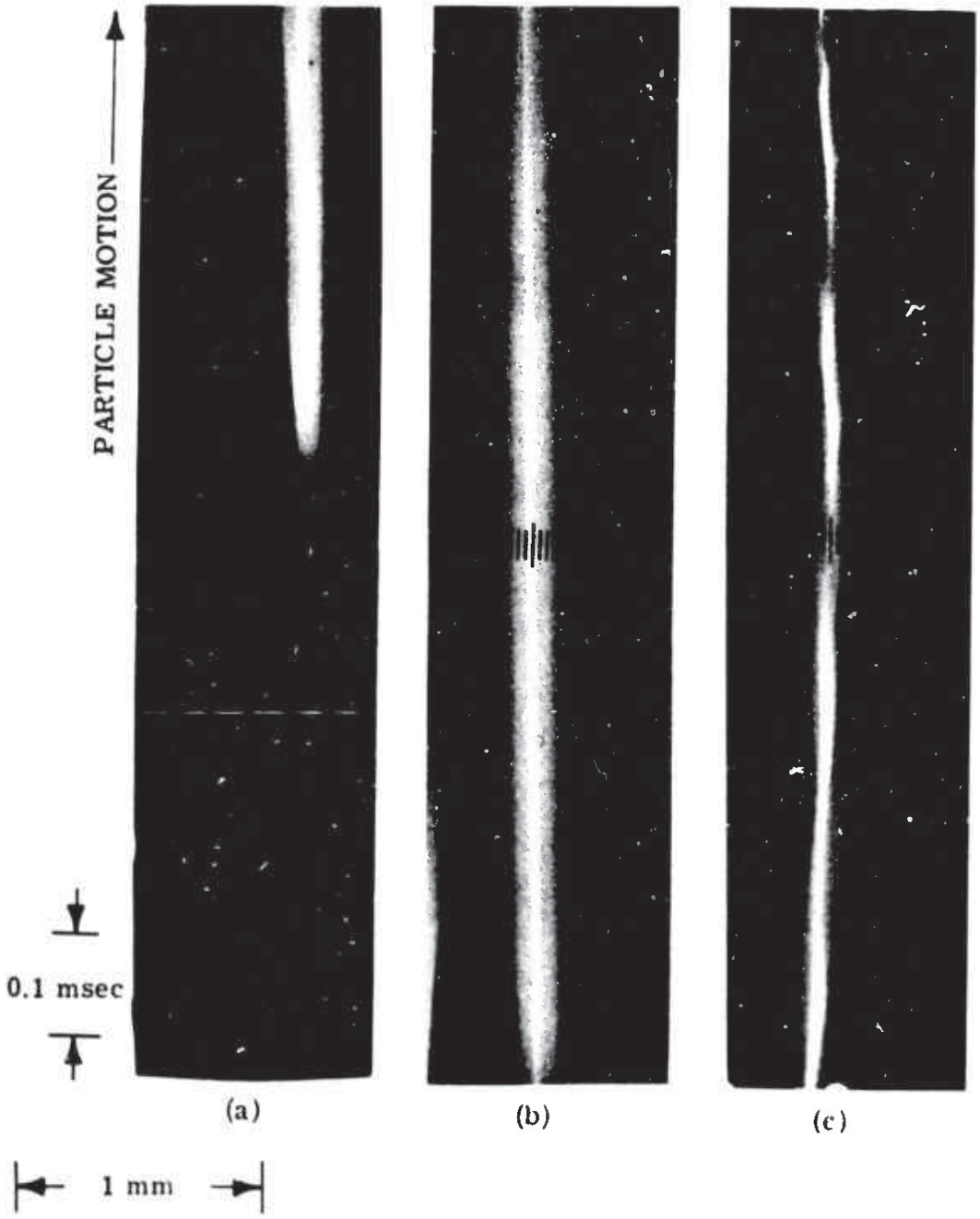


Figure 5. Time exposure of vertically moving, burning single Al particles, diameter range 30-35μ. Ambient gas properties: T = 2530°K, O_2 = 7.9%, H_2O = 0.5%. Residence time of particle in gas is:

(a) 10.4 msec to 11.4 msec (ignition at 11.0 msec)
(b) 11.8 msec to 12.8 msec
(c) 13.2 msec to 14.2 msec (near burnout)

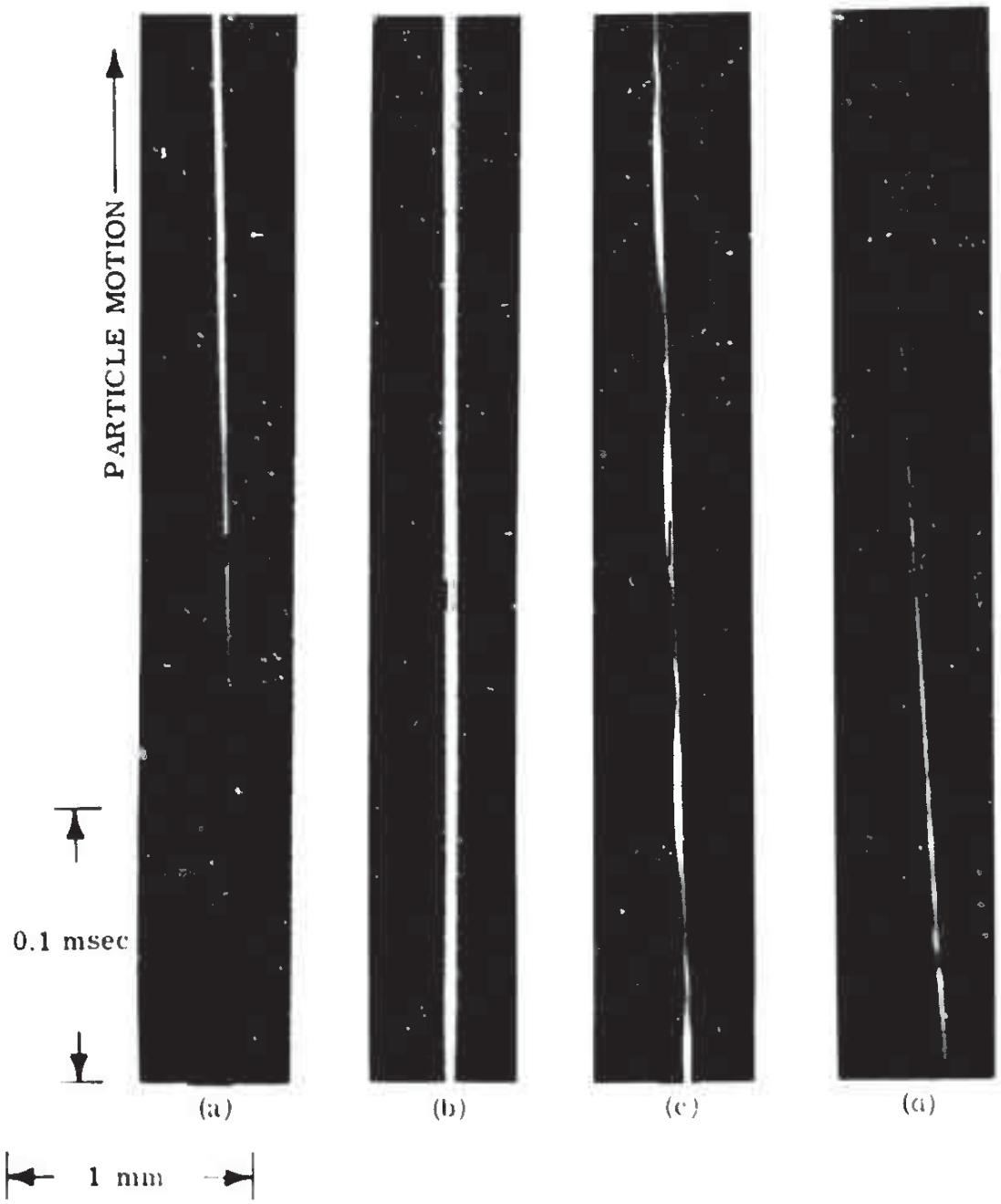


Figure 6. Time exposure of vertically moving, burning single Al particles, diameter range 30-35-. Ambient gas properties: T 2500 K, O_2 15.8%, H_2O = 17.4%. Residence time of particles in gas is:

(a) 7.5 msec to 7.9 msec (ignition at 7.6 msec)
(b) 8.6 msec to 9.0 msec
(c) 9.7 msec to 10.1 msec (rotation)
(d) 10.8 msec to 11.2 msec (near burnout)

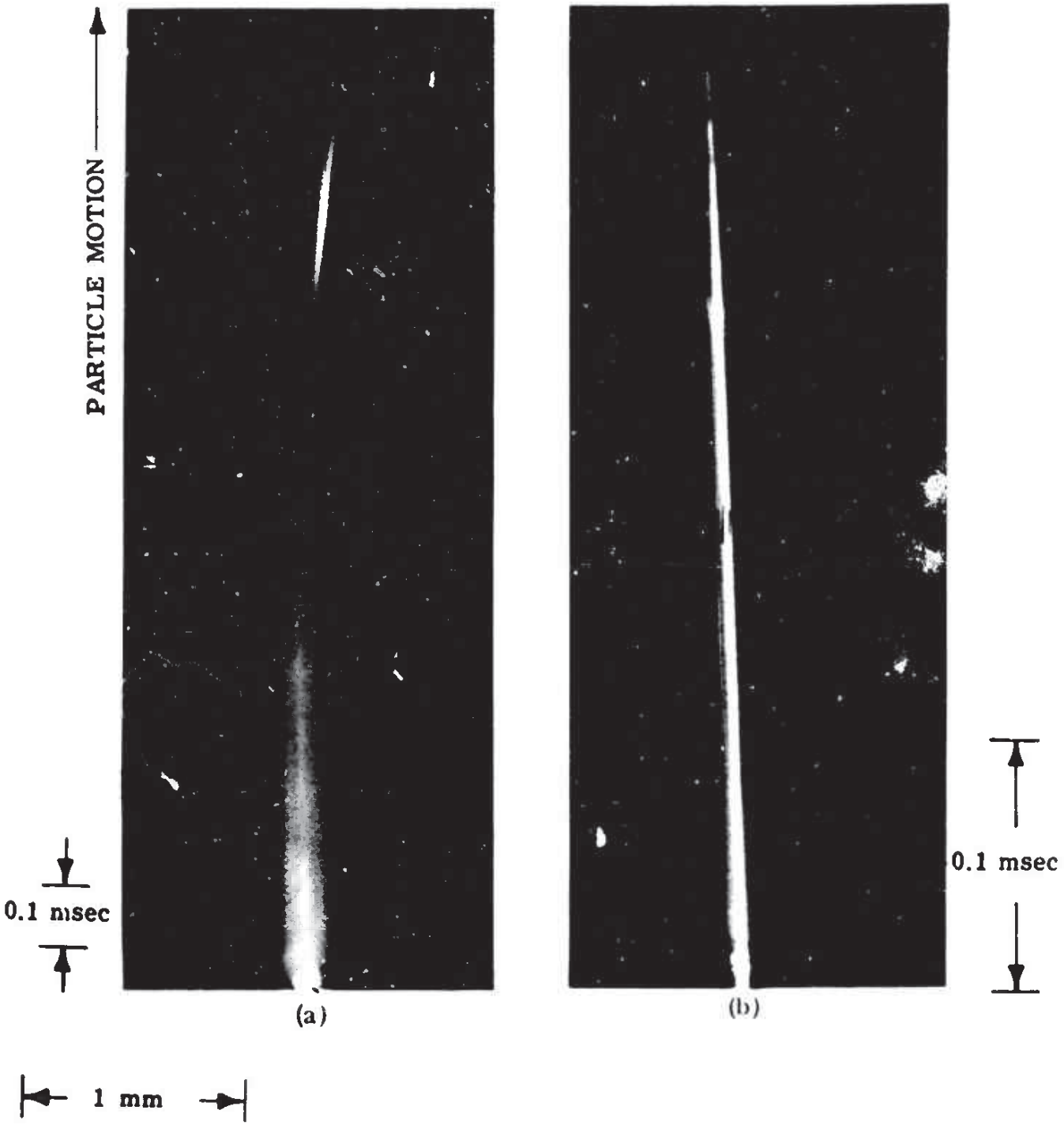


Figure 7. Time exposure near burnout of vertically moving, single Al particles, diameter range 30-35-, burning in gases with following properties:

(a) T = 2390 K, O_2 = 34%, H_2O = 0.6%
(b) T = 2410 K, O_2 = 32%, H_2O = 17%

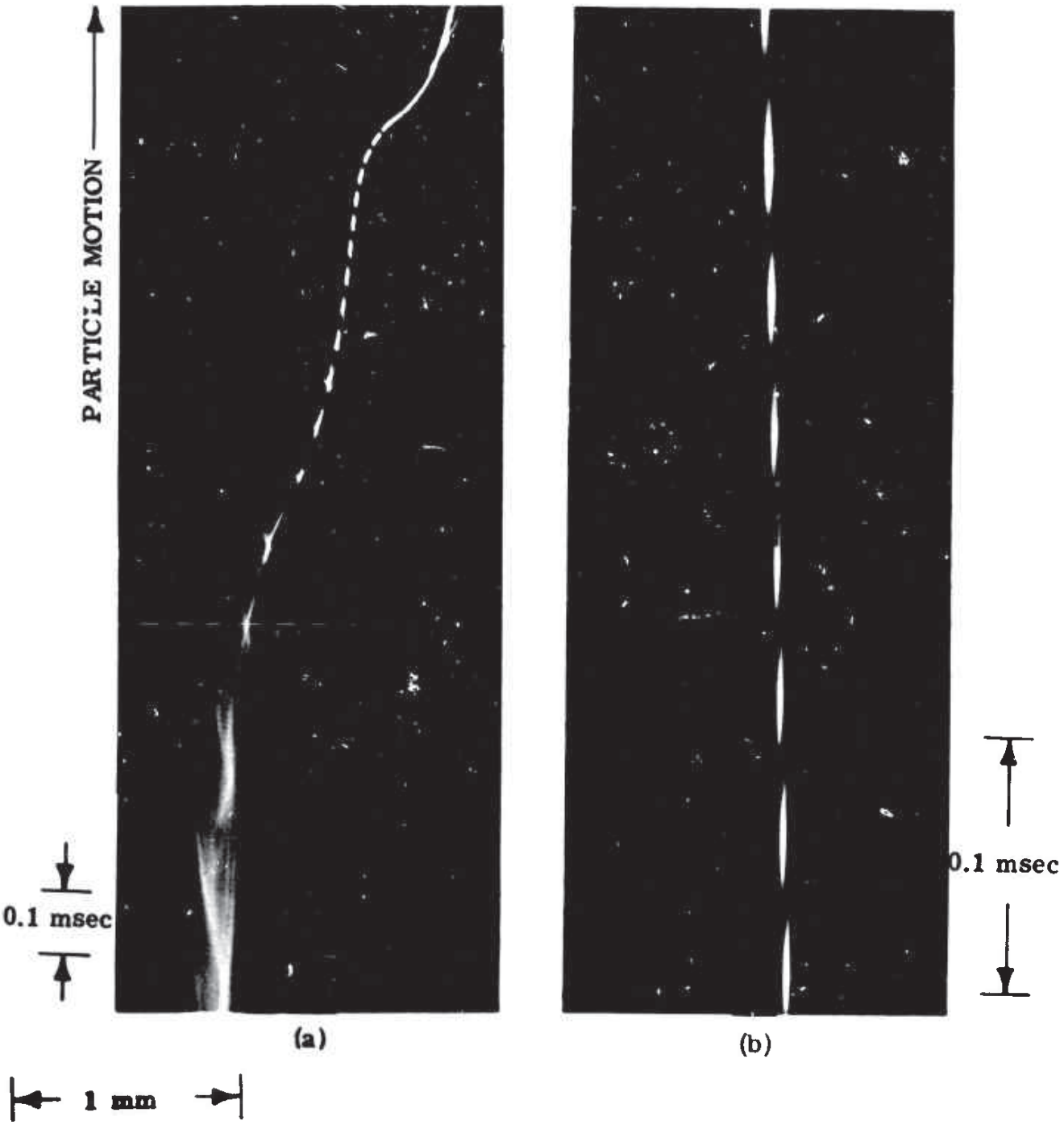


Figure 8. Time exposure of vertically moving, rotating, single Al particles, diameter range 30-35μ, burning in gases with following properties:

(a) T = 2390°K, O_2 = 34%, H_2O = 0.6%
(b) T = 2410°K, O_2 = 32%, H_2O = 17%

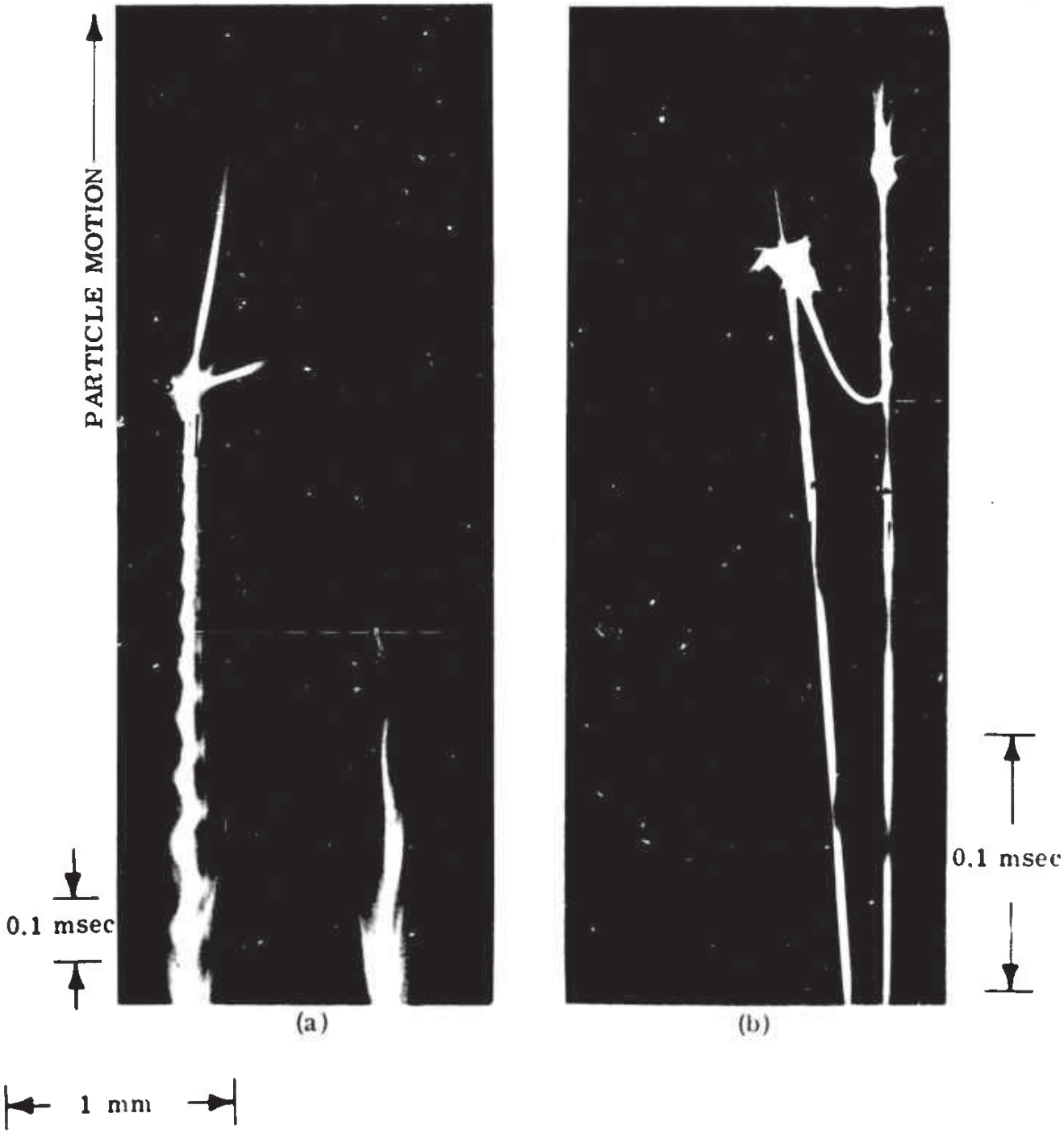


Figure 9. Time exposure at burnout (with fragmentation) of vertically moving single Al particles, diameter range 30-35-, burning in gases with following properties:

(a) T = 2390 K, O_2 = 34%, H_2O = 0.6%
(b) T = 2410 K, O_2 = 32%, H_2O = 17%