DIFFERENCES BETWEEN THE COMBUSTION OF ALUMINUM DROPLETS IN AIR AND IN AN OXYGEN-ARGON MIXTURE*

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When aluminum droplets were burned in air and in its nitrogen-free analog, 20% oxygen in argon, keeping other conditions identical, marked differences between the combustions appeared and will be described in this communication.

Freely falling aluminum droplets, 377μ in diameter, were formed and ignited with single millisecond pulses from a Nd-glass laser by the technique described elsewhere[1]. The droplets were formed from squares of aluminum foil 1.02 mm on a side and 20μ thick supported on crossed aluminum fibers placed at the laser focus. The gas mixtures in both cases were at a total pressure of 625 ± 5 torr, and were flushing slowly upward through a windowed combustion chamber 11 cm on each side and 36 cm high. The heating was performed in a manner that permitted observation of the light emitted by the burning particle from ignition to burn-out. The emission of radiation was recorded as a time exposure with a 102 x 127 mm view camera, and with an RCA 7102 (S-1 surface) photomultiplier-oscilloscope

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combination. A narrow band interference filter that peaked at 8300A was placed before the photomultiplier, which viewed the falling particle from a distance of 3.7 meters.

Time-exposed photographs of the trajectories of droplets burning in the two gaseous atmospheres are shown in Figures 1a and 1b. The trajectory of the aluminum droplet burning in air (Figure 1a) indicates vigorous spinning and shows jetting and fragmentation in the lower portion of the trace. In the nitrogen-free combustion, the trace is broader and somewhat shorter, with completely smooth edges. The trace shows no signs of a spinning particle as in the combustion in air.

The photomultiplier traces of the light emitted by the particles burning in the two atmospheres are also different as shown in Figures 2a and b. In air there is usually an initial sharp rise of about 15 msec duration after which the trace oscillates rapidly, often at frequencies as high as 10^4 cycles per second. After a gradual second increase there is a very irregular emission of light which decays altogether at about 350 msec for the droplet diameter used here.

In the 20% oxygen-argon atmosphere, however, the trace rises sharply after the laser pulse, and then decays smoothly over a somewhat longer time of 400 msec. The peak intensity is about 60% greater than for the droplet that burned in air. Two irregularities are seen in this trace - the change in slope at about 200 msec and the tiny peaks just as the particle extinguishes. Both features are highly reproducible.

When the photomultiplier traces were recorded, 2.5 mm-wide tapes were placed horizontally across the front window of the combustion chamber; they were 38 mm apart vertically. Dips formed in the oscilloscope trace each time the particle passed behind a tape, and allowed us to correlate distance of fall with both time and intensity of light emission. These dips are visible in Figures 2a and 2b.

Distances of fall were determined from traces like those in Figures 2a and 2b, and have been plotted in Figure 3 against the square of time. The straight line has been drawn for a body accelerating gravitationally from rest, where the distance would be $1/2gt^2$. The droplets fell slightly faster than gravitational at the beginning of the oxidation in both atmospheres, but then gradually slowed as time increased. The droplet that burned in 20% oxygen-argon fell considerably more slowly than the droplet that burned in air, and virtually stopped before extinguishment.

When the droplets were allowed to impact glass plates at similar stages of the combustions in the two atmospheres, marked differences again appeared, as shown in the photomicrographs in Figure 4. Here the droplet that had burned in the oxygen-argon mixture was a bare aluminum sphere surrounded by a circular smoke pattern. (The sphere had an indentation, presumably due to contraction on freezing.) The droplet that had been ignited in air, however, showed a glass-like hemispherical cap attached to one side of the aluminum sphere, forming a pronounced bilobate structure. Also in air, the smoke pattern on the glass plate was definitely non-symmetric and confined to the area nearest the aluminum end of the particle.

We now attempt to explain the following characteristics of the burning aluminum droplets: (1) the droplets spin in air but not in O_2/Ar; (2) the droplets jet in air but not in O_2/Ar; (3) the droplets radiate less intensely in air than in O_2/Ar; (4) the droplets fall more rapidly in air than in O_2/Ar, and (5) the total burn time is shorter in air than in O_2/Ar. We suggest that the accumulation of the cap-like lobe of condensed phase product in air but not in O_2/Ar is responsible for the variations in behavior in each case.

Since the only difference between the two reactive systems reported here consists in the simple substitution of argon for

nitrogen the conclusion seems inescapable that nitrogen is participating in the combustion as a reactive species. Recently, Michel, Perez y Yorba and Collongues[2] have obtained aluminum oxynitrides by melting a mixture of alumina and aluminum in air with an induction heater. The similarity of their system to the one reported here suggests that the product which accumulates on aluminum droplets burning in the presence of nitrogen is some aluminum-oxygen-nitrogen species of as yet undetermined composition. (An analogous reaction occurs when zirconium droplets burn in air[4].) The assumption[5] that the product lobe of the bilobate configuration shown in Figure 4a is aluminum oxide does not seem justified in view of the experimental findings reported here.

Comparison of Figures 1a and b and 4a and b indicates that the spinning seen on the particle track from aluminum burning in air results from the accumulation of the condensed-phase product lobe on the droplet's surface. This restricts the available surface from which aluminum may vaporize. The combination of droplet dissymmetry and directional vaporization of aluminum generates a tangential force on the droplet that results in the particle spin seen in Figure 1a. Since no product accumulates on the droplet's surface when aluminum burns in 20% oxygen in argon, the particle trajectory is spin-free (Figure 1b).

The jetting seen toward the bottom of Figure 1a when the specimen burns in air also seems to arise from the accumulated lobe of product. When quenched specimens similar to the one shown in Figure 4a are examined with an optical microscope, considerable frothiness is seen in the product globule suggesting the formation of a gas. Later in the burning, the frothiness leads to the formation of larger bubbles which burst at the surface. These gas-related processes appear on our photographic traces as either jetting or fragmentation of the droplet in the terminal stages of combustion.

The greater radiant emission from the droplets that burn in air (Figure 2a and b) may be attributed to the incomplete flame front around the droplets that burn in air, due to the reduced and directional vaporization of aluminum. It is thought that the primary source of radiation in burning aluminum droplets emanates from the cloud of incandescent smoke particles formed in the combustion[6]. Since the cloud of particles around the droplet is apparently reduced significantly by the presence of the product lobe, as seen when Figures 4a and 4b are compared, it is not surprising that the light intensity measured photoelectrically is lower for the droplet that burned in air than for the droplet that burned in O_2/Ar.

This perturbation of the symmetrical cloud of particles by the product cap probably explains the greater rate of fall of droplets that burn in air than in O_2/Ar (Figure 3). The cloud of hot gases that surround the burning droplets seems to add a drag on buoyancy force opposed to gravity. Since this cloud is reduced significantly in the combustion in air, the fall rate should then increase as the buoyancy force is reduced.

An apparent paradox arises, however, when burning times in the two atmospheres are compared. Thus, in spite of the reduced rate of vaporization of aluminum when the combustion occurs in air, due to the product lobe on the droplet's surface, the droplet actually burns in a shorter time overall. A possible explanation for this is that the droplet can react with both oxygen and nitrogen in air, but only with oxygen in O_2/Ar. Thus the droplet can reach a state of complete oxidation more rapidly in air than in O_2/Ar. Perhaps in air, part of the reaction with the atmosphere occurs in the vapor phase flame front, while the rest occurs at the surface of the product lobe, with oxygen and nitrogen passing into the lobe from the gas phase and aluminum diffusing across the metal-product interface.

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CAPTIONS FOR FIGURES

Figure 1. Time-exposed track photographs of 377μ-diameter aluminum droplets as they burned in free fall (a) in air and (b) in a mixture of 20% oxygen in argon, with total gas pressures in both cases at 625 torr. Short traces emanating from holder arise from tiny droplets formed from crossed aluminum fiber support. Note the typical erratic trace that appears when an aluminum droplet burns in air.

Figure 2. Oscilloscope records of RCA 7102 photomultiplier output when it viewed 377μ-diameter aluminum droplets burning (a) in air and (b) in a mixture of 20% oxygen in argon at a total pressure of 625 torr. Central traces were recorded with second beam of dual beam oscilloscope at an amplification 0.1 that used for other trace. Sharp dips in trace were formed as droplet passed behind strips of tape placed horizontally across front of combustion chamber. Intensity scales are identical in both traces.

Figure 3. Distance of fall plotted against the square of time for aluminum droplets burning in air and in a mixture of 20% oxygen in argon at a total pressure of 625 torr. Initial droplet diameter was 377 microns.

Figure 4. Photomicrographs of aluminum droplets that had impacted glass plates during combustion (a) in air and (b) in a mixture of 20% oxygen in argon. Note the bilobate structure of the droplet that had burned in air, and the bare metallic appearance of the droplet that had burned in O_2/Ar.

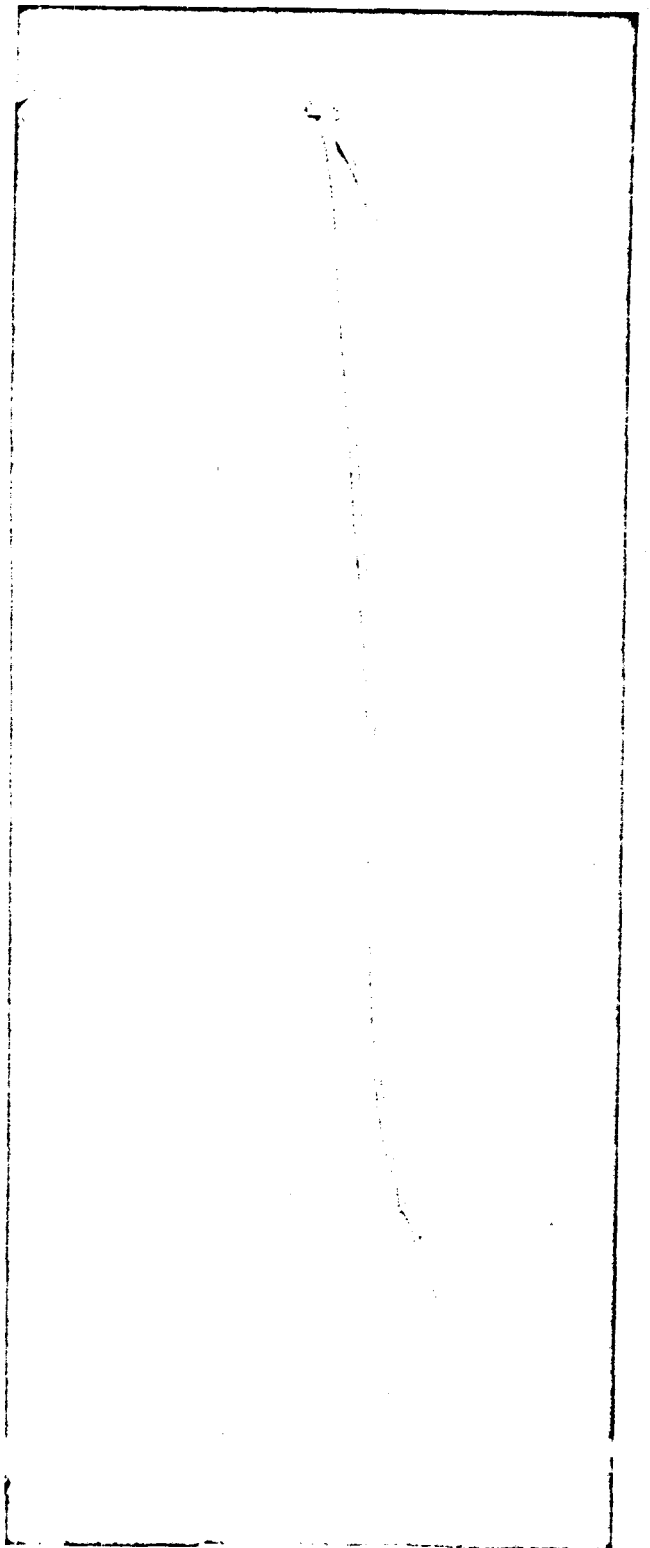

Figure 1a

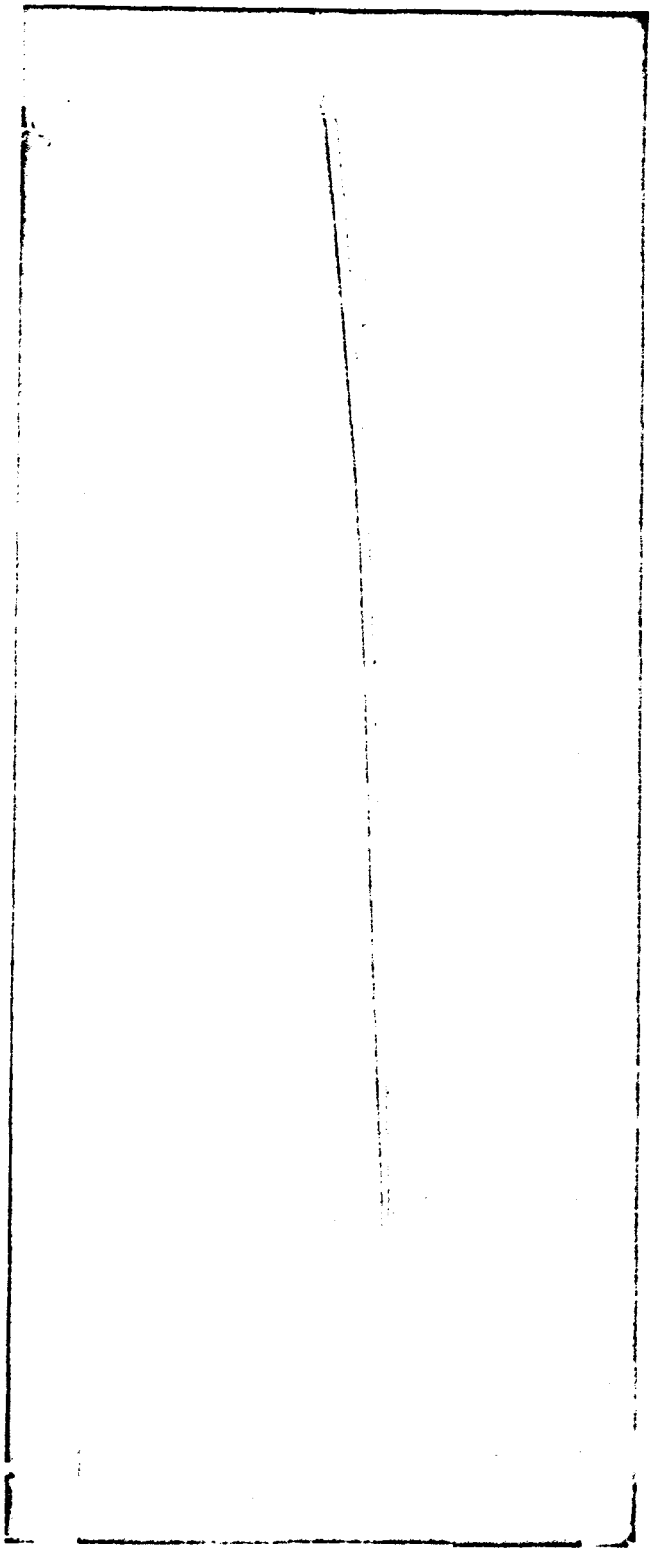

Figure 1b

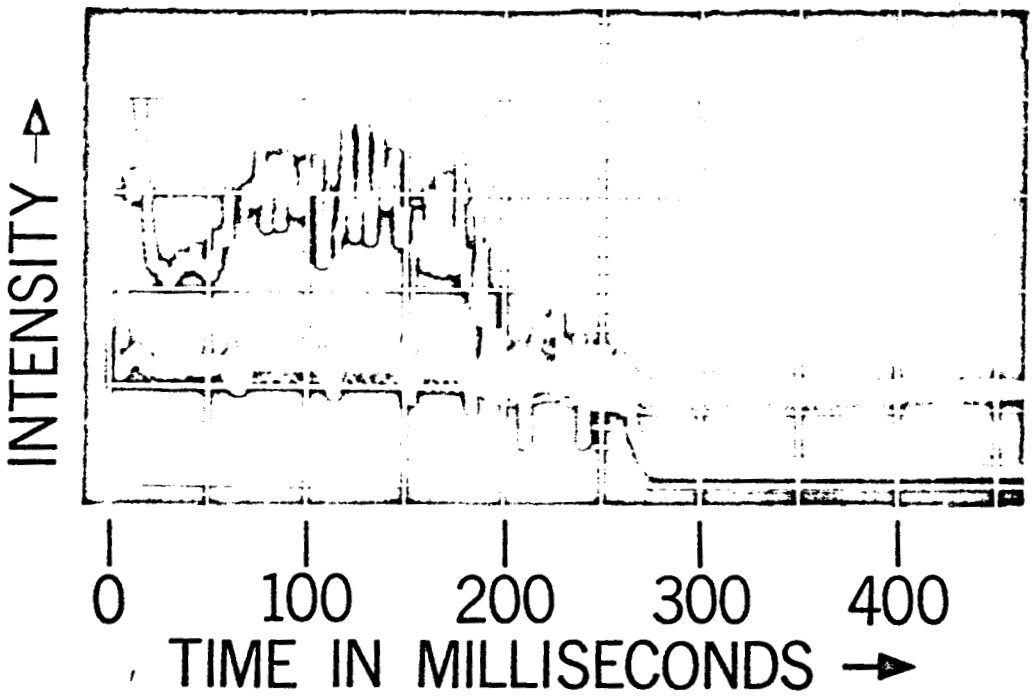

Figure 2a

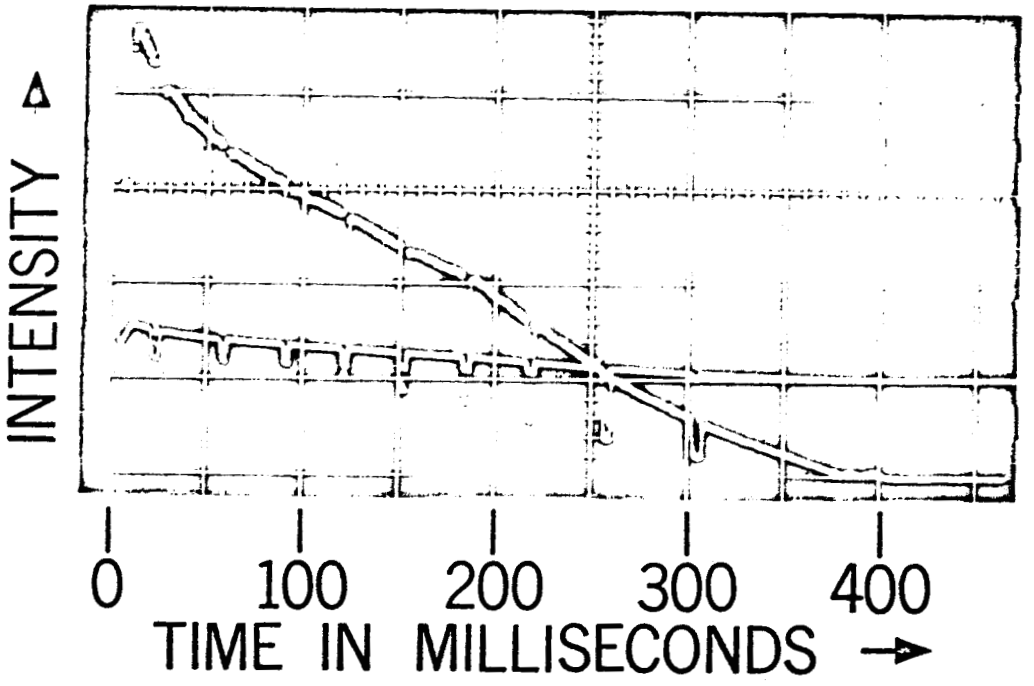

Figure 2b

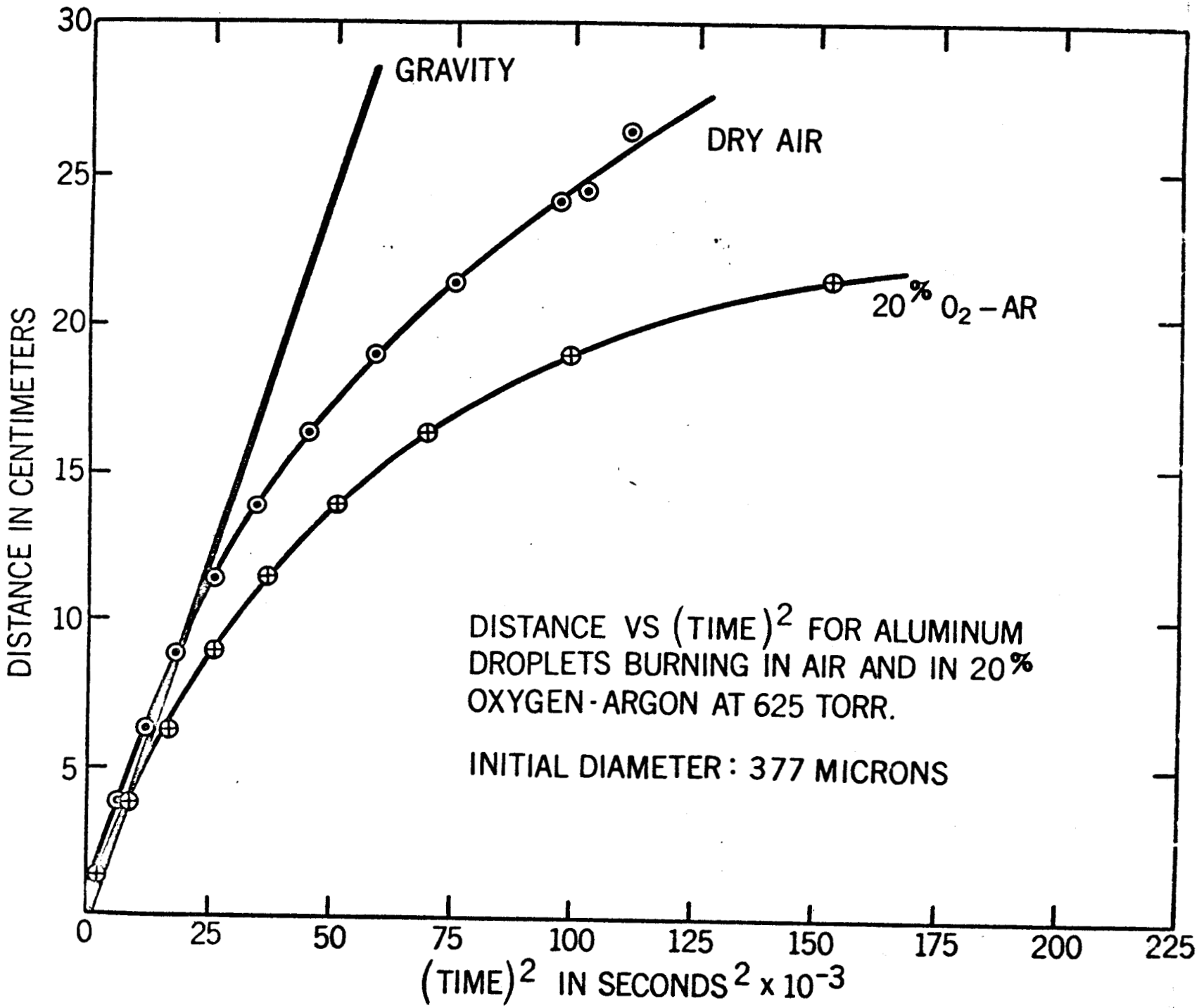
GRAVITY
DRY AIR
20% O_2 – AR
DISTANCE VS $(TIME)^2$ FOR ALUMINUM DROPLETS BURNING IN AIR AND IN 20% OXYGEN-ARGON AT 625 TORR.
INITIAL DIAMETER: 377 MICRONS
DISTANCE IN CENTIMETERS
$(TIME)^2$ IN SECONDS^2 x 10^{-3}
0
5
10
15
20
25
30
0
25
50
75
100
125
150
175
200
225

Figure 3

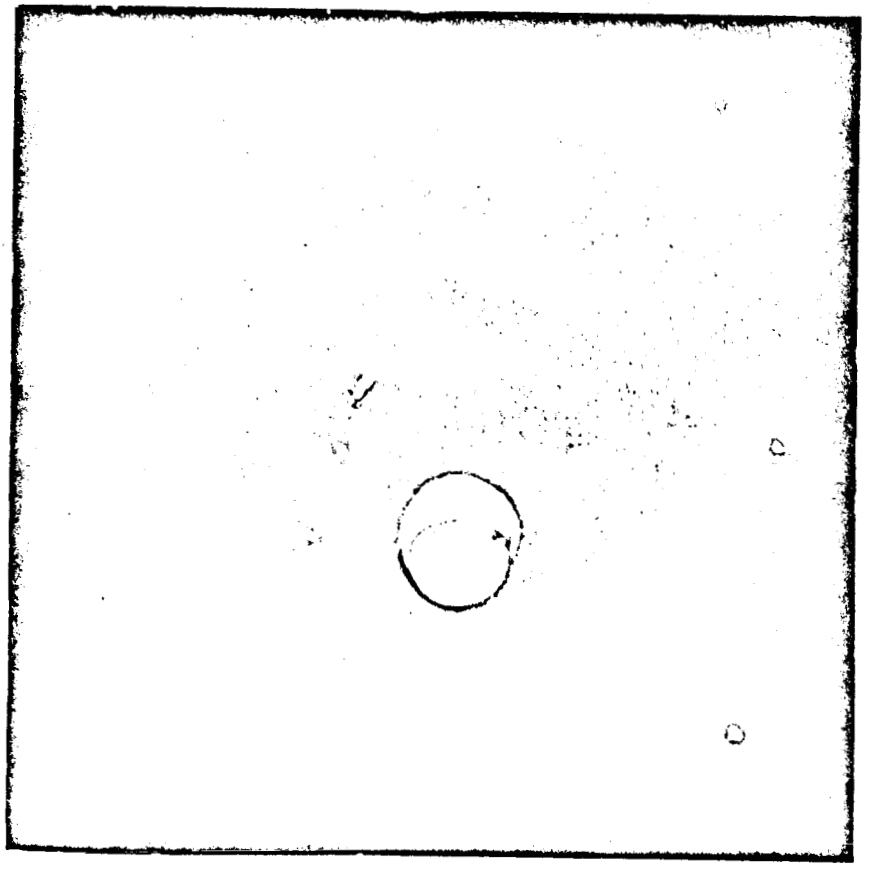

Figure 4a

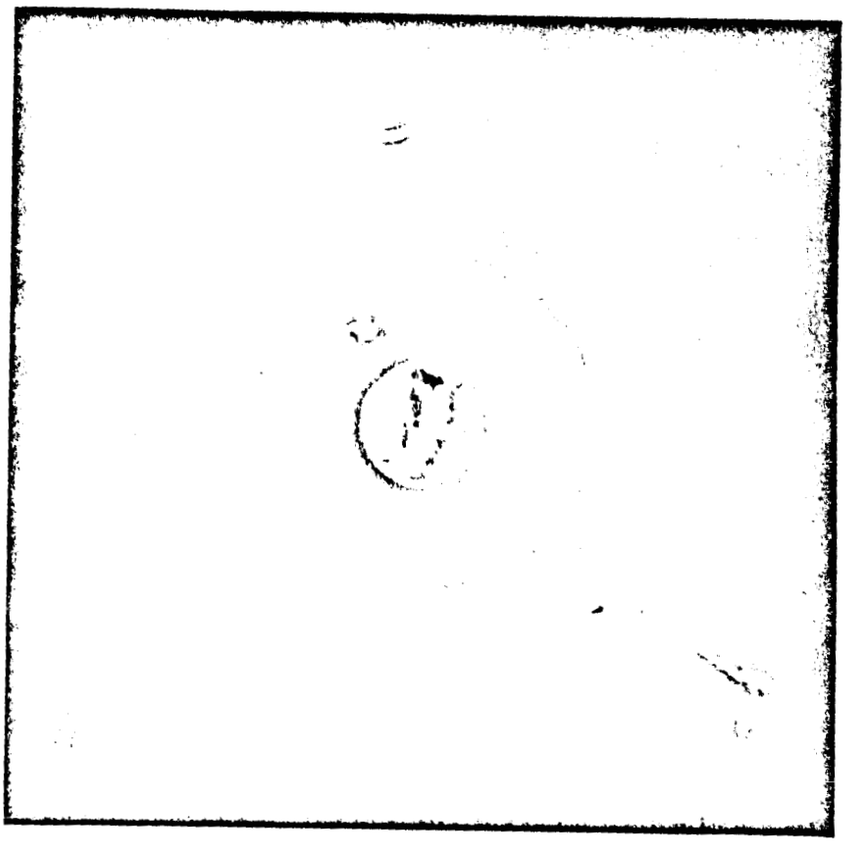

Figure 4b