EXPERIMENTAL OBSERVATIONS OF PMMA SPHERES BURNING AT REDUCED GRAVITY[1]

JIANN C. YANG, ANTHONY HAMINS, MICHAEL GLOVER, and MICHELLE D. KING
Building and Fire Research Laboratory
National Institute of Standards and Technology
Gaithersburg, Maryland 20899 U.S.A.

Introduction

Polymer combustion is a very complicated process which involves the coupling of gas-phase combustion, melting, pyrolysis, and possible charring of the condensed phase. Although only a few studies on the combustion of a spherically shaped polymer have been conducted (refs. 1-3), there is renewed interest in the subject (refs. 4-7). At the Third International Microgravity Combustion Workshop (ref. 8), we presented a preliminary experimental plan and proposed an apparatus to study the combustion of a polymethylmethacrylate (PMMA) sphere at reduced gravity. In this paper, we describe the experimental hardware in detail, summarize our observations since the last workshop, and describe future studies. The main objective is to determine the burning rate of PMMA spheres at reduced gravity under different ambient oxygen concentrations and total pressures. The dependence of the burning rate on the initial sphere diameter will be examined. It is anticipated that the simple spherical geometry in conjunction with reduced gravity will facilitate an assessment of the effect of condensed phase behavior on polymer burning processes and can be used as a means to test polymer combustion models.

Research Approach

The reduced gravity environment was achieved by performing experiments aboard the NASA DC-9 Reduced Gravity Aircraft. The time available to carry out the experiments in both the 2.2 s and 5.18 s NASA drop towers is not long enough to observe the burning histories for spheres with initial diameters used in this study, although the quality of the low G environment in a drop tower (less than 10^{-4} G) is considerably superior to an aircraft flying a parabolic trajectory (~ 10^{-2} G). For all the experiments, the Grashof number was estimated to be less than 0.1.

Figure 1 is a photograph of the experimental package (shown inside the aircraft cabin). The experimental hardware is housed inside a rack (60 cm x 60 cm x 106 cm) which is mounted on the aircraft cabin floor. The hardware consists of a lap-top computer, a data acquisition and control system (for collecting thermocouple and G-level measurements during the experiments and for controlling the micro-step controllers and the ignition systems), a combustion chamber (an interior view is shown in Figure 2), DC power supplies, a pair of micro-step controllers and linear motors, two Hi8 mm CCD cameras, two ignition systems, a 120 V AC power strip, and a vacuum pump. Oxygen/nitrogen mixture cylinders are mounted separately in a gas bottle rack. The vacuum pump is used to evacuate the combustion products and facilitate the flushing and filling of the combustion chamber with the desired oxygen/nitrogen mixtures in the 1G tests. For the reduced gravity experiments, the overboard vent of the aircraft is used for operational convenience.

Although several different ignition methods have been explored (ref. 8) in the 1G experiments, two methods for igniting the PMMA spheres have been actively pursued, developed and used in the reduced gravity experiments. These two ignition techniques involve the application of (1) micro-torches and (2) heating coils.

Two opposing micro-torches were first used in the flight experiments in an attempt to symmetrically ignite the PMMA sphere. The micro-torch ignition system consists of a fuel reservoir with a refill port, a stainless steel sub-miniature solenoid valve for initiating the fuel flow, a metering valve, a spark ignition system, and a miniature nozzle. The fuel used is liquefied butane. This fuel was chosen over methane because of its low vapor pressure and ease of refilling. The fuel reservoir is a stainless steel cylinder with an outlet section packed with wicks in order to prevent the fuel from coming out as a flashing liquid. A two-phase fuel flow hinders the smooth operation of the micro-torch. The spark ignition circuit, which is used to ignite the gaseous fuel vapor at the nozzle, is battery operated. The initiation and the duration of the spark discharge can be controlled by the computer *via* a solid state relay. The micro-torch nozzle has a configuration similar to a Bunsen burner. Two identical ignition systems

[1]Work funded by NASA Microgravity Science and Applications Division under NASA Interagency Agreement C-32017-C.

Microgravity Combustion Workshop, Fourth (4th) International. Proceedings. National Aeronautics and Space Administration, Lewis Research Center. NASA Conference Publication 10194. May 19-21, 1997, Cleveland, OH, 1997.

are mounted oppositely on two linear motors which are used for moving the torches toward the sample for ignition and for retracting the torches away from the sample upon ignition.

Although the micro-torch ignition system performed satisfactory, the fuel jet perturbed the flame of the burning PMMA. Consequently, the micro-torches have been replaced by two opposing heating coils. The coils are made of metal alloys with a diameter of 250 μm. To ignite a PMMA sphere, the coils are resistively heated until they glow by using a filament transformer and are moved until they just touch the sample surface. The duration that the coil is energized is controlled by the computer *via* a solid state relay; however, the duration was fixed at 2 s for all experimental conditions.

Supported PMMA spheres were used in all the flight experiments. The technique for fabricating a supported PMMA sphere has been described in ref. 8, and a photograph of the apparatus is shown in Figure 3. The supported sample is mounted at the center of a removable holder which is placed along two parallel tracks (see Figure 2). Both K-type thermocouple wire with its junction embedded at the center of the sphere and a special Al/Cr/Fe alloy wire were used to suspend the polymer sample. The diameters of the thermocouple and the special alloy wires were both 75 μm.

The experiments are controlled by a lap-top computer, except for manual operations of valves and video cameras, chamber evacuation and filling with oxygen/nitrogen mixtures, and insertion of a sample. So far, three flight campaigns have been carried out. The first involved the use of PMMA spheres with initial diameters (D_o) of 3.18 mm, 4.76 mm, and 6.35 mm, together with the micro-torch ignition system. The ambient oxygen concentration was set at 21 % in nitrogen (by volume) at cabin temperature and 0.101 MPa. In the second and third campaigns, spheres with initial diameters of 2 mm, 2.5 mm, and 3 mm and heating coils for ignition were used. Ambient oxygen concentrations were varied from 19 %, to 21 %, 25 %, and 30 %. Experiments were conducted at cabin temperature and 0.101 MPa or 0.05 MPa.

Results to Date

Experimental observation of a suspended PMMA sphere burning at reduced gravity reveals the following characteristics. Upon ignition, the sample swells, the outermost surface layer of the burning sample bubbles, and a spherical blue flame is observed, the duration of which decreases with total ambient pressure and oxygen concentration. The spherical flame subsequently becomes more luminous (yellowish color), more bubbles nucleate, the internal bubbling intensifies, and a soot shell is formed between the flame and the sphere. Violent spluttering and ejection of molten polymer from the burning sphere are observed, followed by break-up of the soot shell. The internal bubbling and ejection of material impart impulsive motion to the suspended sphere which causes it to oscillate and slide along the supported wire. The flame loses its spherical shape largely due to the relative motion of the suspended sphere to the ambient gas and partly due to variations in G level. The movement of the sample appears to be more severe for spheres with smaller initial diameters because of smaller inertia and because of the rapid propagation of the molten front through the sphere. The ejection of molten material is more severe in reduced gravity than in 1G because gravity-induced ablation of the melt (dripping of the melt) at 1G appears to counteract the ejection process. In terms of fire safety, the ejected burning material poses a potential fire hazard to adjacent objects. In cases ($D_o \leq 3$ mm) where combustion proceeds to completion during the reduced gravity period, the flame resumes its sphericity again due to a small Grashof number and smaller motion of the suspended sphere (due to surface tension).

The period of reduced gravity provided by the NASA DC-9 Reduced Gravity Aircraft is not long enough to obtain complete burning histories of spheres with initial diameters greater than 3 mm for the range of conditions tested. For spheres with $D_o > 3$ mm, the diameters of the bubbling spheres during combustion do not appear to change significantly (except some degree of swelling) during the entire reduced gravity period. For smaller spheres, complete combustion was observed, and soot strings were found attached to the suspending wire. Extinction was never observed in any of the experiments performed thus far.

Figure 4 is a video sequence showing a polymer sphere before ignition, at ignition, with a spherical blue flame, with a yellow flame, and with variation of the flame shape as combustion progresses. Note that the bubbling sphere moves randomly with respect to the suspending wire during combustion. The three components (Gx, Gy, and Gz) of the G vector are also shown in the bottom of each frame in the figure, and the date, time, and the parabolic trajectory number are depicted at the top of each frame. Figures 5 and 6 are snapshots of the break-up of the soot shell and the ejection of burning molten material, respectively. The formation of a soot shell is not surprising because the main polymer degradation product, methyl methacrylate, has a smoke point very similar to heptane, and a soot shell was observed during the combustion of heptane droplets at reduced gravity (ref. 9). The break-up of the soot shell in the figure is reminiscent of the phenomena associated with a toluene droplet burning at reduced gravity with a small drift velocity (ref. 10).

Figure 7 shows the temperature measurements obtained from suspending a thermocouple wire with the junction initially located at the center of the sphere for conditions of 25 % oxygen and 0.101 MPa. The plateau in the temperature trace for D_o = 3.18 mm is the result of the thermocouple junction being displaced to the exterior of the sphere by impulsive motion. The sharp rise in the temperature is due to the sphere being dislodged from the suspending thermocouple during the 1.8 G maneuver of the aircraft. The non-existence of the plateau in the temperature trace for larger spheres, together with the video records, reaffirms the observation of less impulsive motion for larger spheres because the location of the thermocouple junction remains relatively stationary with respect to the burning sphere and the melting front may not have propagated to the junction (*i.e.*, the junction is still embedded in solid phase polymer).

Because of the impulsive motion of the suspended bubbling PMMA spheres and the non-spherical flame shapes, it is very difficult to obtain accurate information on the transient sphere and flame diameters. However, for spheres that burn to completion over the reduced gravity period, the average burning rates can be obtained from the burning times (t_b). Given the initial mass of a sphere and assuming the mass loss due to the embedding of a supported wire to the sample is negligible, the average mass burning rate can be calculated by dividing the initial mass of a sphere (m_i) by t_b. The burning times were obtained from the video record, using frame-by-frame analysis. The uncertainty of the measurements was less than ± 30 ms.

Figures 8 and 9 summarize the burning rate results for some of the successful runs performed using the coil ignition system. The effects of total ambient pressure and oxygen concentration on the burning rate are not apparent due to the large data scatter. However, a general trend emerges, with the average burning rate increasing with the initial sphere diameter. One plausible explanation of the data scatter is that the random ejection of molten material represents a mass loss which is not accounted for in the burning rate calculations. Another reason for the scatter may be partly due to the fixed (2 s) contact ignition period. Ignition delay is expected to vary for different sphere diameters and ambient oxygen concentrations. Although the coil ignition time can be tailored to different experimental conditions, a prohibitively large number of reduced gravity experiments may be required in order to determine the optimum ignition times. Since the ignition events at reduced gravity may be different from those at 1 G, the ignition times cannot be *a priori* determined from 1 G experiments. In order to ensure ignition under various experimental conditions, a 2 s contact ignition time was used and found to be quite satisfactory. If the ignition delay time is shorter than the contact time, unnecessarily prolonged contact of the heating coils with the sample surface would undoubtedly increase the burning rate. Depending on the experimental conditions, the 2 s ignition time is approximately 10 % to 20 % of the burning time. Ignition remains one of the most challenging issues that will be continuously addressed during the course of this study.

Research Plans

We plan to continue to focus on combustion of suspended spheres. Based on the observations from the suspended burning spheres, experiments using unsupported spheres need to be re-evaluated. These experiments may prove to be not feasible with PMMA because the impulsive motion of the sphere imparted by the spluttering and ejection of molten material can cause the unsupported sphere to drift out of the field of view of the cameras, making observations impossible. We are planning to examine other polymeric materials which may exhibit distinct burning behavior. In our next flight experiments, several polypropylene spheres (3 mm) will be tested. In 1 G experiments, polypropylene does not exhibit bubbling. We are also working closely with Dr. K. Butler (of NIST), who is developing a polymer combustion model (also funded by NASA, MSAD) that incorporates the formation and growth of bubbles. Burning rate predictions from this model will be compared to our observations.

References

1. Essenhigh, R. H. and Dreier, W. L., "Combustion Behavior of Thermoplastic Polymer Spheres Burning in Quiescent Atmosphere of Air," *Fuel*, **48** (1969), pp. 330-342.
2. Waibel, R. T. and Essenhigh, R. H., "Combustion of Thermoplastic Polymer Particles in Various Oxygen Atmospheres: Comparison of Theory and Experiment," *Fourteenth Symposium (International) on Combustion*, The Combustion Institute, Pittsburgh, 1973, pp. 1413-1420.
3. Raghunandan, B. N. and Mukunda, H. S., "Combustion of Polystyrene Spheres in Air," *Fuel*, **56** (1977), pp. 271-276.
4. Chung, S.L. and Tsang, S.M., "Soot Control During the Combustion of Polystyrene," *J. Air Waste Manage. Assoc.*, **41** (1991), pp. 821-826.
5. Chung, S. L. and Lai, N. L., "Suppression of Soot by Metal Additives During the Combustion of Polystyrene," *J. Air Waste Manage. Assoc.*, **42** (1992), pp. 1082-1088.
6. Panagiotou, T. and Levendis, Y., "A Study of Combustion Characteristics of PVC, Poly(styrene), Poly(ethylene), and Poly(propylene) Particles under High Heating Rates," *Comb. & Flame*, **99** (1994), pp. 53-74.
7. Okajima, S., Kawakami, T., and Raghunandan, B. N., "Extinction of Fuel Particles Burning Under Microgravity," Work-in-Progress Poster Session, *Twenty-sixth Symposium (International) on Combustion*, The Combustion Institute, Pittsburgh,

1996.
8. Yang, J. C., and Hamins, A., "Combustion of a Polymer (PMMA) Sphere in Microgravity," *Third International Microgravity Combustion Workshop*, pp. 115-120, April 11-13, 1995.
9. Hara, H., and Kumagai, S., "Experimental Investigation of Free Droplet Combustion Under Microgravity," *Twenty-third Symposium (International) on Combustion*, The Combustion Institute, Pittsburgh, 1990, pp. 1605-1610.
10. Avedisian C. T., Yang, J. C., and Wang, C. H., "On Low Gravity Droplet Combustion," *Proc. Roy. Soc. London*, **A 420** (1988), pp. 183-200.

Figure 1. Experimental package.

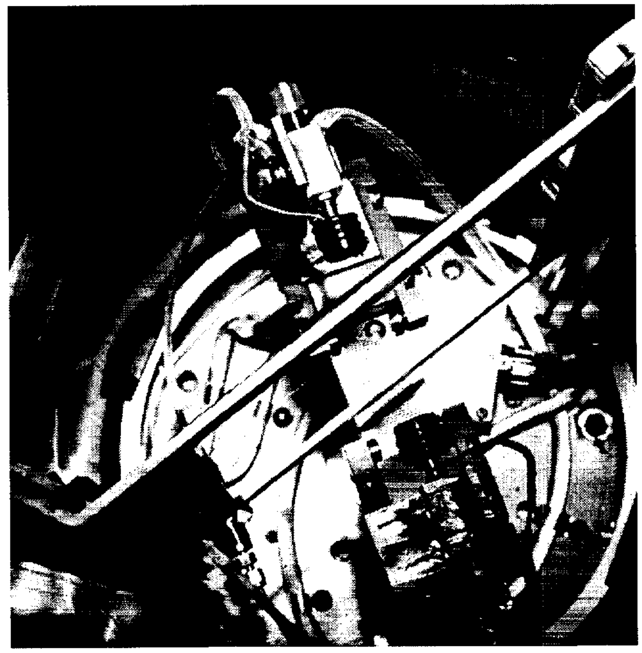

Figure 2. View of interior of combustion chamber.

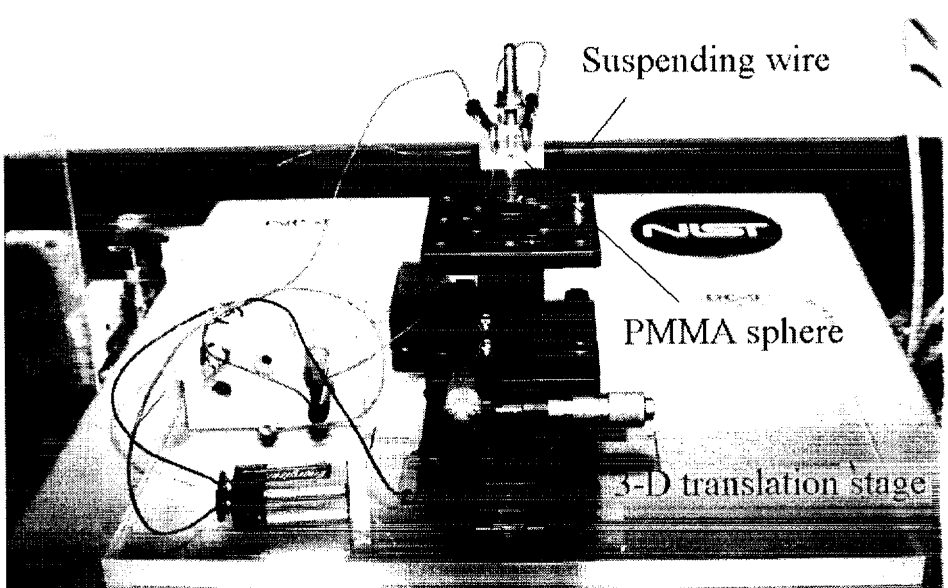

Figure 3. Apparatus used to embed a suspending wire inside a PMMA sphere.

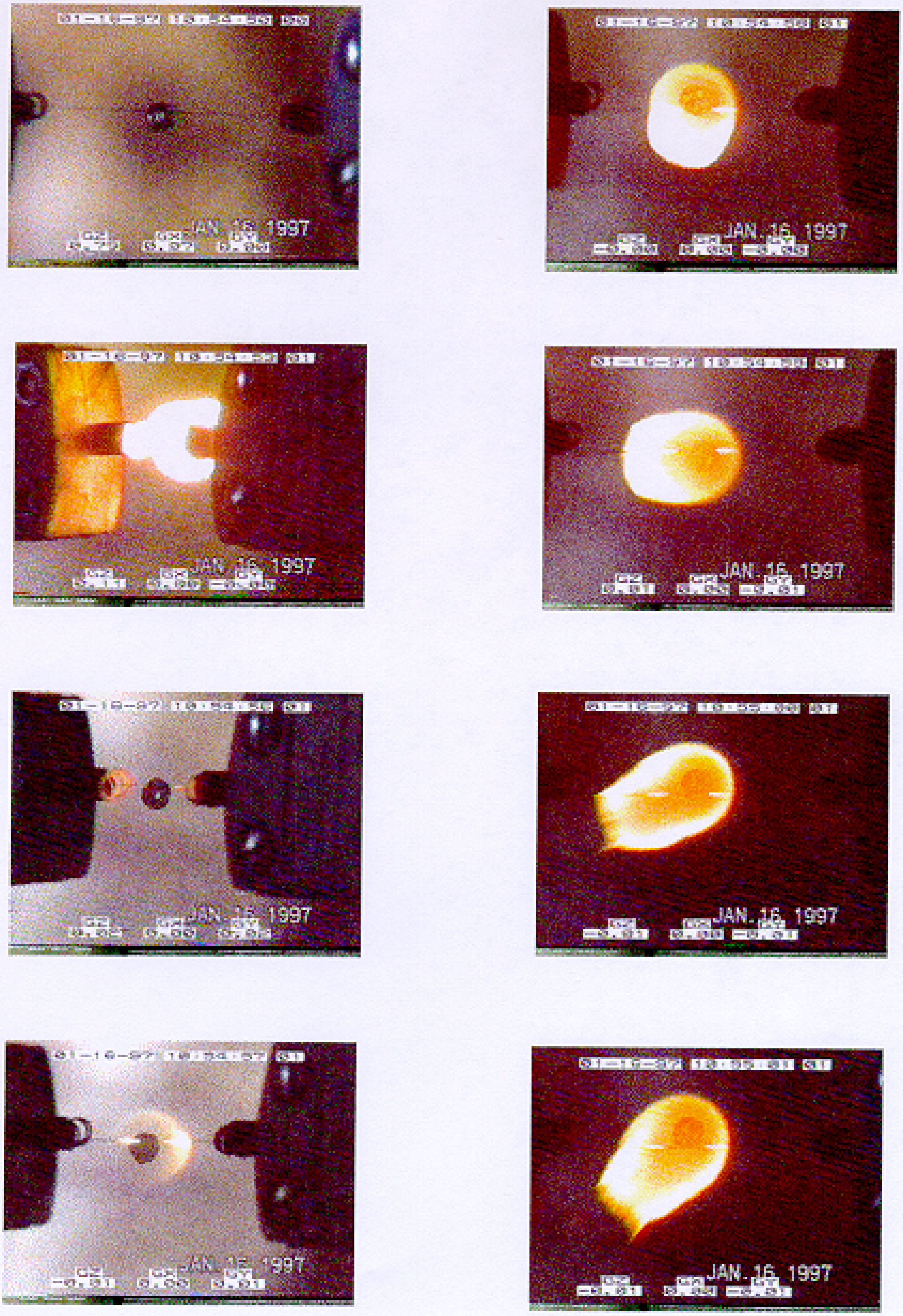

Figure 4. A video sequence showing a burning PMMA sphere (D_o = 3 mm at 19.9 % O_2 and 0.101 MPa).

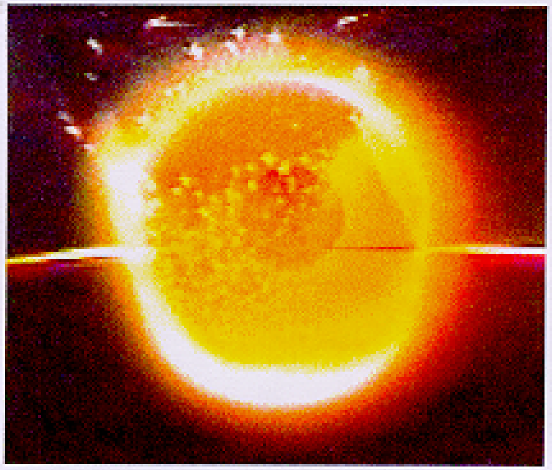

Figure 5. Break-up of a soot shell.

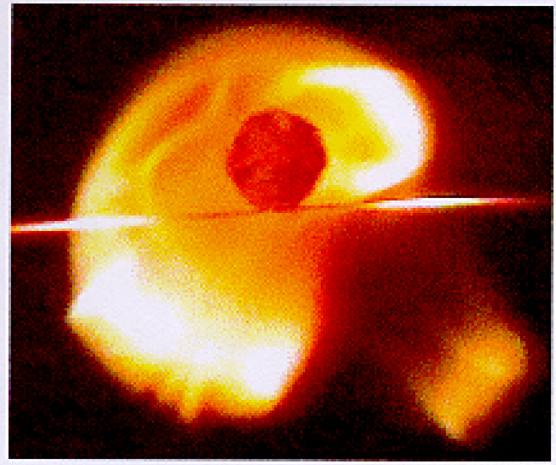

Figure 6. Ejection of molten material.

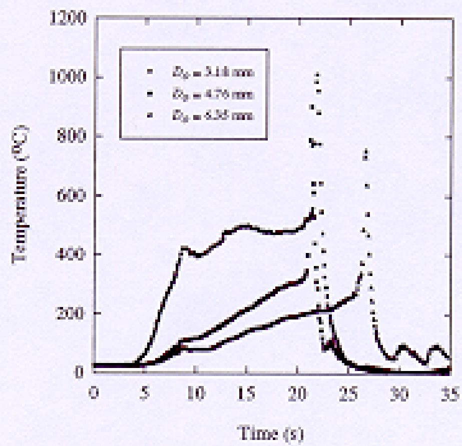

Figure 7. Temperature trace with thermocouple junction initially at the center of a burning sphere with different D_o.

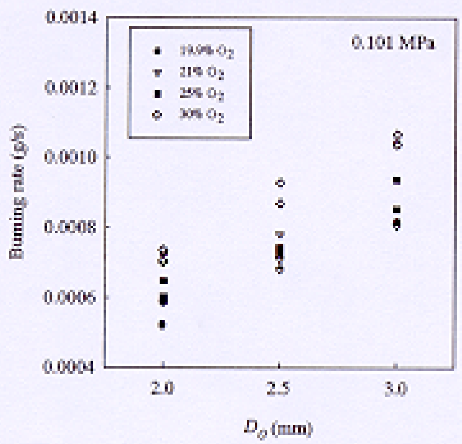

Figure 8. Burning rate as a function of D_o at different O_2 levels at 0.101 MPa.

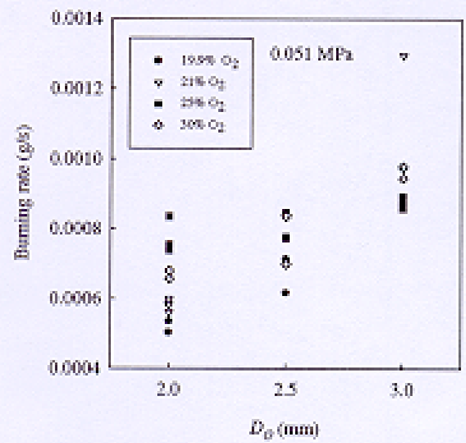

Figure 9. Burning rate as a function of D_o at different O_2 levels and 0.051 MPa.