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Stability and Flame Spread Over Poly(Methyl Methacrylate) (PMMA) Within the Quenching Distance

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14. ABSTRACT

This report focuses on the effects of moving boundary on counter-current flame spread over poly(methyl methacrylate) (PMMA) near the flame extinction limits. Experiments were conducted with a wind tunnel and less than 19.4% oxygen (by volume) air with free stream velocity of 87 cm/s. At these conditions, the flame was observed to retreat from the leading edge after ignition and stabilize downstream, establishing a quenching distance from the leading edge. While the flame was retreating, the entire sample surface was relatively flat. As the flame stabilized, a small valley was formed near the flame leading edge and then the flame started to spread counter-currently, decreasing the quenching distance with time. The results show that the flame could not stabilize within the quenching distance when the surface was flat, molten and pyrolyzing but will spread upstream after the surface has solidified. The size of the valley increased as the flame spread upstream. It appeared that the presence of the valley stabilized the flame, perhaps by creating a stagnation/recirculating zone that increased the Damkholer number and enabled the flame to spread upstream. Flame spread in these experiments was chemically controlled and the spread rates were observed to be comparable to the regression rates. Both rates were observed to decrease with time, as the valley enlarged. On the other hand, a few tests conducted far downstream with ambient air (thermally controlled regime) showed constant spread rates, which were much larger than the regression rates.

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Stability and Flame Spread over Poly(Methyl Methacrylate) (PMMA) Within the Quenching Distance

1.0 INTRODUCTION

The suppression and extinction of boundary layer-type fires present a challenge because of the proximity of the condensed fuel to the flame. The rate of burning depends on the heat feedback to the condensed fuel and in boundary layer flames it is highest in the leading section where the flame is closest to the surface and decreases with the stream-wise distance from the leading edge. This leads to the formation of a small valley in the leading section. This report will focus on the effects of this valley on flame stability and upstream flame spread.

The two processes that determine flame spread over a condensed fuel are the heating and gasification of the virgin material and the ignition of a combustible mixture of the fuel vapor and oxidizer. Recent reviews [1-5] of flame-spread phenomenon include elaborate description of these processes and detailed discussions of the controlling mechanisms. In counter-current flame spread, two spread regimes have been defined, namely, the thermally controlled regime and the chemically controlled regime [5]. In the thermally controlled regime the spread process is controlled by the heat feedback to the fuel surface and there the spread velocity is proportional to the free stream velocity U_∞. This regime is characterized by low and intermediate velocities and/or high oxygen concentration. deRis [6] did the pioneering work in this regime. On the other hand, the chemically controlled regime is characterized by high U_∞ and/or low oxygen concentration. In this regime, the Damkholer number Da (flow time/ reaction time) in the leading section has decreased such that $Da < 1$ [7].

Chemical kinetics effects dominate over transport effects near the extinction limits. Depending on the initial value of Da, the boundary layer flame can be anchored away from the fuel leading edge, creating a quenching distance (sometimes referred to as "extinction distance"), where combustion cannot be sustained. For a flat surface, it has been established [7-9] that the flame cannot spread upstream and the quenching distance remains constant with time.

In boundary layer combustion of non-charring solids like PMMA, the surface regresses non-uniformly as the sample burns since the heat feedback is non-uniform along the stream-wise direction. This tends to create a step or valley on the surface as it burns. This report will present results of experiments that demonstrate the effects of non-uniform surface regression on flame spread over PMMA. It will show that the flame could not stabilize within the quenching distance when the surface was flat, molten and pyrolyzing but will spread upstream (decreasing the quenching distance) after the surface has solidified. The implication of this moving boundary effect is the enhancement of flame stability, making it more difficult to suppress the flame.

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Surface regression (pyrolysis propagation perpendicular to the surface) and flame spread (pyrolysis propagation parallel to the surface) are coupled. In earlier flame-spread studies [e.g., 10] with PMMA, conditions were such that the flame-spread rate is much faster than the regression rate This report will discuss flame spread in experiments where the spread rate is comparable to the regression rate.

2.0 EXPERIMENTAL

The key components of the experimental setup consist of a 15-cm^2 cross-sectional wind tunnel, a PMMA sample holder mounted on a MiniTec ® sliding mechanism and a thermocouple mounted on computer controlled Velmex® X-Y unislides (Fig.1). The wind tunnel has a 36 X 45 X 61 cm plenum at one end into which an Ametek RJ054 ® variable speed blower pumps air. Air from the blower is mixed with a known flow of nitrogen before the mixture flows into the plenum. Pressure build-up in the plenum drives the flow of the oxidizer through the wind tunnel; hence, the effects of the blower on the flow are minimized. The flow velocity in the wind tunnel is selected by adjusting the speed of the blower. The oxygen concentration in the air flow is continuously measured by sampling gas from the wind tunnel and measuring the concentration of the sample in a Beckman ® Industrial oxygen analyzer Model 755. The burning sample is positioned outside the tunnel at the center of the tunnel exit, making it easier for a thermocouple to be moved freely in and out to measure temperatures.

The sample holder is made of a 1.5-mm-thick aluminum plate (18.5 cm x 19 cm) brazed onto a 10 cm x 8 cm x 2.1cm deep cup, which holds the PMMA sample. This results to a 4-cm lip in the leading section and a 5-cm lip in the other three sides. At the measurement location, the holder is positioned with its leading edge against the tunnel exit at the center of the channel (Fig.1). A thin strip of quartz glass is placed between the PMMA sample and the walls of the holder on all four sides to prevent molten PMMA from sticking on the walls of the sample holder. The sample and holder sit on a platform mounted on a slide mechanism such that the sample can be ignited under the radiant panel located about 50 cm downstream from the tunnel exit and quickly moved to the tunnel exit after ignition. The test samples are 7.7 cm X 9.5 cm and are made from Cyro Acrylite GP ® sheet nominally 2.54 cm thick. The incoming oxidizer velocity is measured at the exit of the tunnel (measurement location) using a hot wire anemometer. More details of the setup are described in [11]. A picture of the experimental setup is shown in Fig. 2.

Air with additional nitrogen to obtain between 19.4 ± 0.1 % and 18.8 ± 0.1 % oxygen by volume flows through the wind tunnel with an exit velocity of 87 ± 1 cm/s at the center. The air velocity at the exit of the tunnel (measurement location) is measured using a hot wire anemometer and its profile across the tunnel is relatively uniform near the center. At these conditions of velocity and oxygen concentration, the flame is near the extinction limits. With U_∞ = 100 cm/s, Kodama et al. [9] predicted a mass fraction of 18% at the lean extinction limit, which is consistent with the measurements of [12].

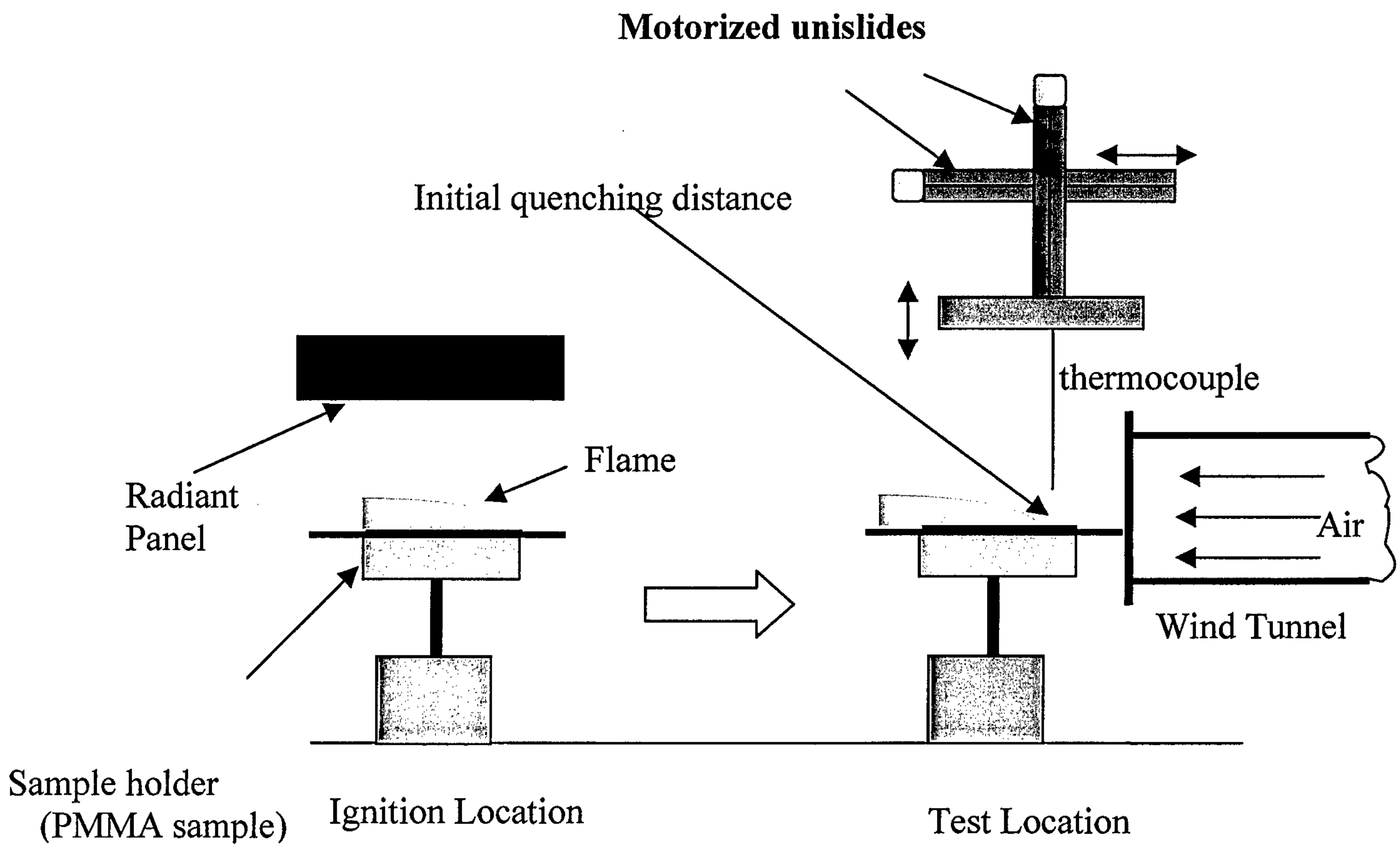


Figure 1: A Schematic of the experimental setup illustrating the process of establishing the quenching distance on the PMMA sample surface.

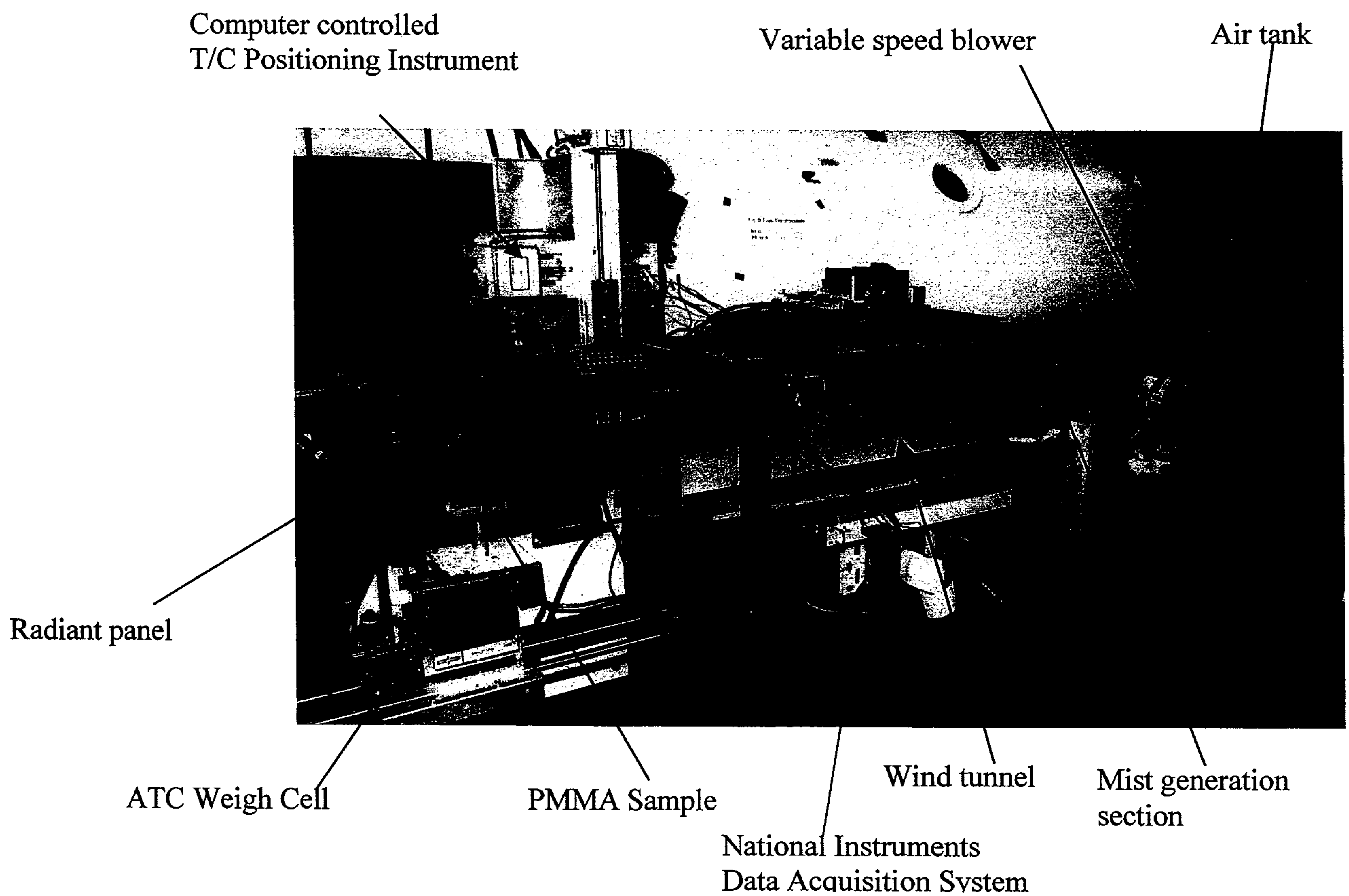


Figure 2: Picture of the Experimental Setup

The sample surface is ignited by uniform radiation from a radiant panel at the ignition location and the flame retreats downstream soon after it is moved to the exit of the tunnel (Fig.1). An R-type thermocouple, 75 μm in diameter, is quickly lowered into the molten layer as the flame retreats. The thermocouple continuously measures the temperature of a point on the molten/solidifying layer within the quenching distance as the flame stabilizes downstream and as the stable flame spreads upstream within the quenching distance. To reveal the sample surface profile at the relevant times in the experiment, separate tests were conducted, where the flame was extinguished as it is retreating (stage 1), as it stabilizes (stage 2) and after it had spread upstream for some time (stage 3).

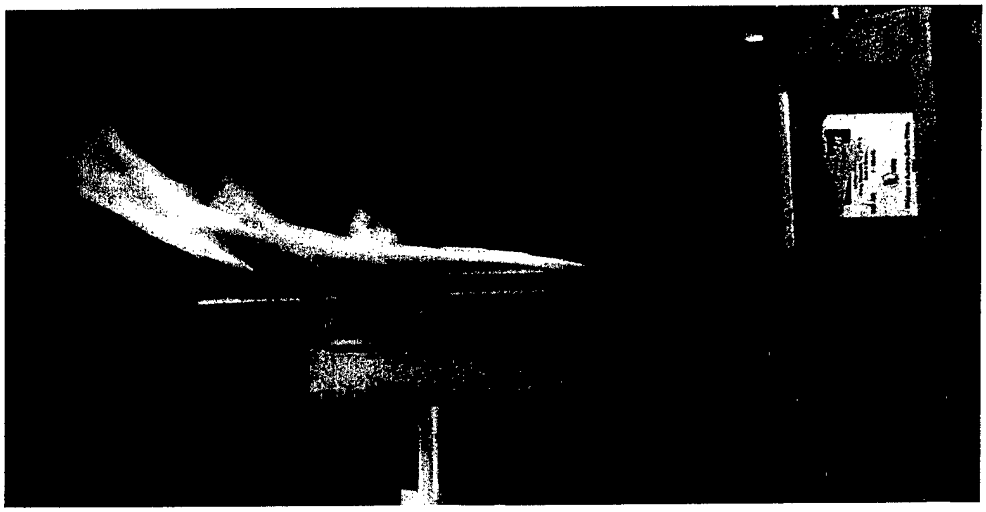
Since PMMA surface regresses as the sample burns, a set of tests was conducted to obtain the depth of surface regression and in these tests, samples of known initial thickness were burned for a fixed time before the flame was extinguished. After the sample cools down, the change in thickness along the centerline was measured at various stream-wise (X) locations with a digital micrometer (accuracy ± 0.003 mm).

3.0 RESULTS AND DISCUSSIONS

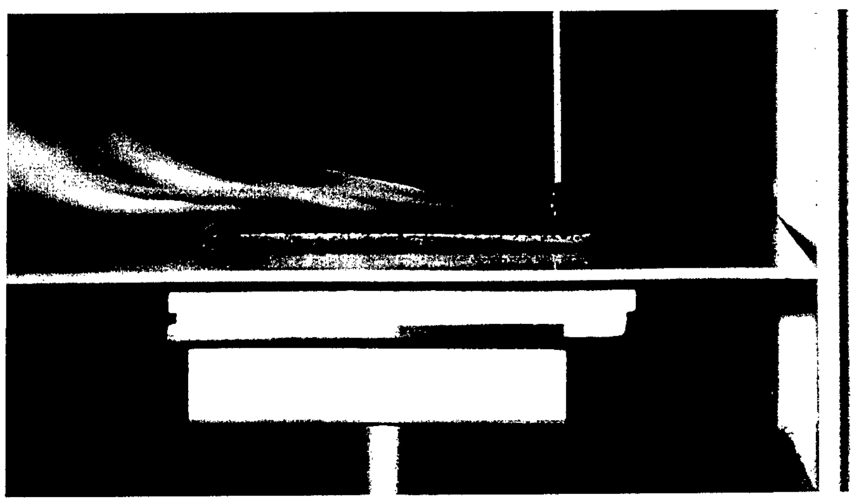
To establish a boundary layer flame over the entire PMMA sample, the surface is heated under a radiant panel far downstream, where the air velocity is significantly smaller than at the exit of the tunnel, Fig.1. It is important to ensure that the entire surface is pyrolyzing as indicated by intense bubbling before the flame is ignited. In addition it is necessary to ensure that a stable flame is established quickly (few seconds) so that the shape of the solid surface remains flat. After ignition, the sample is quickly moved to the exit of the tunnel.

Figure 3a shows the flame as the sample is brought to the exit of the wind tunnel. It shows a relatively smooth flame near the leading edge region, which gets wrinkled and unsteady downstream. The wrinkling could be as a result of the movement or as a result of buoyancy effects, since the plate surrounding the sample was still very hot having been heated by the radiant panel. The buoyancy effects die down as the metal lip cools to ambient within a short time. Figure 3a shows that the flame covers the entire sample, including the leading edge region, implying that this region is molten and pyrolyzing at this stage. One may also see a flat fuel surface under the flame. The fuel surface sticks out of the sample holder by about 1 to 2 mm so that the surface does not regress below the metal lip during the test, preventing the formation of an artificial trough.

Shortly after the flame gets to the exit of the channel, where the velocity is significantly higher than at the ignition location, the boundary layer flame is dislodged from the sample leading edge and starts retreating downstream (Fig. 3b). The leading edge of the flame was observed to oscillate back and forth over the molten surface of PMMA as it retreats from the sample leading edge, similar to what Kodama et al. [9] observed. For the purposes of discussion we define this state where the flame was dislodged from the sample leading edge and retreating downstream as stage 1. The thermocouple, which is about 15 mm downstream from the sample leading edge along the centerline, is quickly



(a)



(b)

Figure 3: (a) Picture of the flame anchored at the sample leading edge and being moved to the measurement location. (b): Picture of the flame dislodged and retreating downstream soon after it gets to the measurement location; thermocouple measuring temperatures as it dips into the molten layer.

lowered into the molten surface at this stage. Figure 3b shows the thermocouple passing through the retreating bluish flame and one can see that the surface is still relatively flat at this stage. This will be shown more clearly later. Soon the leading edge of the flame stops oscillating and stabilizes, then anchors at some distance (quenching distance) from the sample leading edge. Thereafter, a small step is formed at the location of the flame leading edge and the flame starts to spread upstream (stage 2). In this test, where the air has 19.4% oxygen, the quenching distance is about 20 mm.

Figure 4a shows the flame in stage 2, anchored about 5 mm downstream from thermocouple. While the leading edge of the flame was retreating, the molten polymer was exposed to ambient air and was cooling down slowly. The melt has a thermal diffusivity 1600 times smaller than that of the hot gas adjacent to the surface, therefore, it takes a long time for it to cool down. Consequently, the surface could still release a significant amount of fuel vapor during the transient stabilization process. The rate of vaporization, however, decreases with time as the flame stabilizes. In the present experiments, the reaction rates ($\propto$ 1/time) are lowered by the presence of nitrogen and the flow rate is increased as the sample is brought to the exit of the channel. Therefore, the Damkholer number Da, which is flow time/reaction time, decreases as the flame is brought to the tunnel exit, causing the flame to retreat and stabilize at the quench distance.

In boundary layer flames, the heat feedback to the surface is largest near the leading edge of the flame where the boundary layer thickness is smallest and decreases with stream-wise distance. Since the surface regression rate is roughly proportional to the heat feedback, the regression rate is non-uniform along the surface, being highest near the flame leading edge. Therefore, as time progresses, the shape of the surface changes as the depth and shape of the valley or step formed near the flame leading edge increases and changes. The flame appears to anchor behind the step. With time, it was observed that the leading edge of the flame and valley moved upstream, decreasing the quenching distance. Figure 4b shows a picture of the flame several minutes later (stage 3) as it approaches and engulfs the thermocouple, which was located 5mm upstream from where the flame stabilized. It also shows that the surface is no longer flat and the edge of the valley has moved past the thermocouple toward the leading edge. The flame could now stabilize behind the step at the location of the thermocouple but it could not stabilize there while the surface was flat (stage 1). The surface profile at the various stages will be shown more clearly later. The above observations suggest that the formation of a step stabilizes the flame and enables it to spread counter currently in Fig. 4b. It is well established in the literature that flame is stabilized behind a step and this has been used extensively in burner design [13]. These experiments were repeated numerous times with various initial quenching distances established with various combinations of flow velocity and oxygen concentration and each time the flame was observed to spread upstream decreasing the quenching distance. Therefore, one can infer that the moving boundary effects lead to step formation, which was critical for the stabilization and spread of the lean limit flame. The flame also could not spread on the hot melt surface due to too low Da but was able to spread on a cooler solid surface due to the moving boundary effect, which helped to increase the Da.

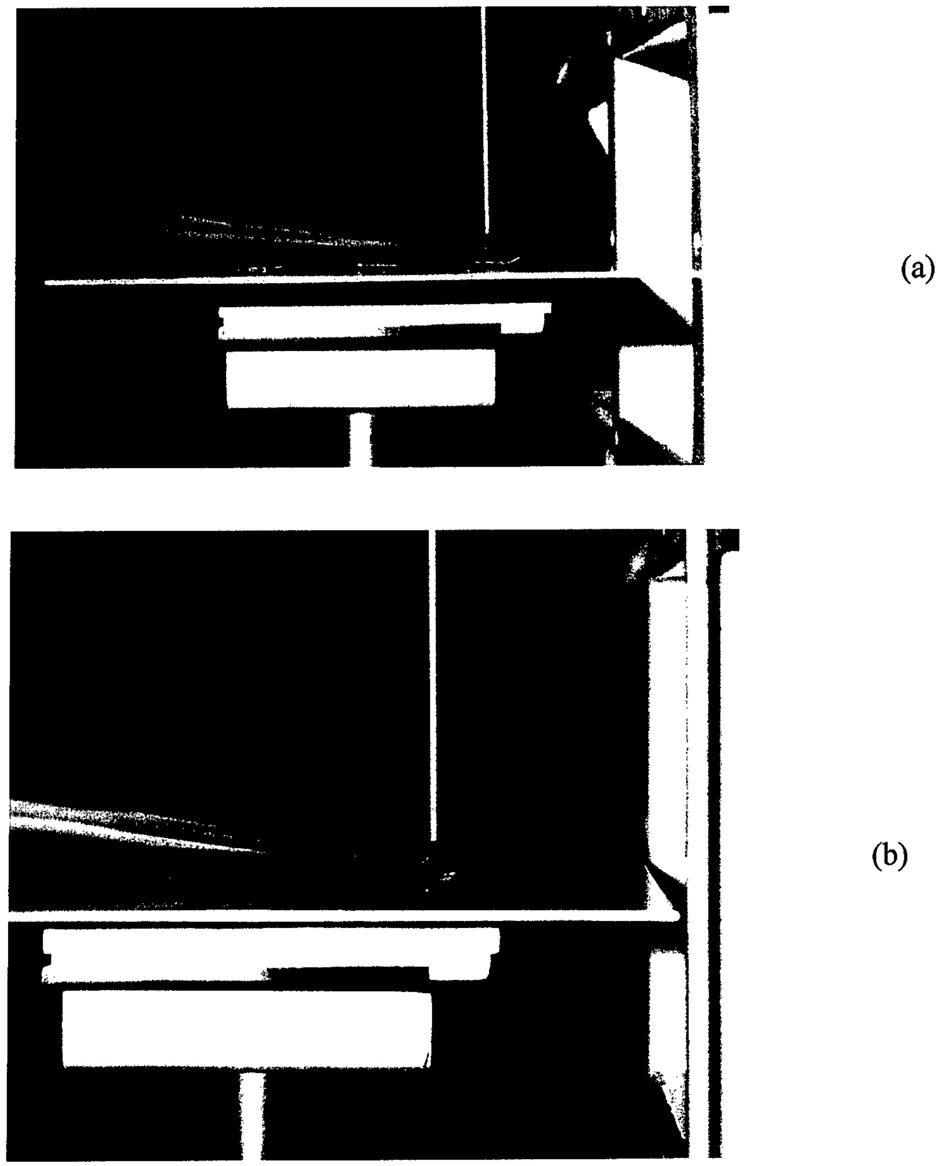


Figure 4: (a) Picture of the Flame stabilized about 5mm downstream of the thermocouple location. (b) Picture of the Flame spreading upstream up to the location of the thermocouple.

The time-dependent thermocouple data (uncorrected for radiation) measured as the flame retreats and then spreads upstream (Figures 3a to 4b) are shown in Fig 5. The first several data points on the left hand side of the curve were taken during stage 1 as the flame was retreating and the thermocouple was passing through it (Fig. 3b). A fairly rigid thermocouple (75 μm diameter) was used to ensure that the bead goes into the melt/froth layer. The thermocouple was lowered into the melt until the bead goes through the melt/froth layer and the thermocouple starts to bend. The thermocouple was moved slowly through the retreating flame to show that the surface was gasifying at this location, the gases were hot and the flame could not sustain there. PMMA is known to pyrolyze at temperatures on the order of 650 to 700 K [14,15]. Furthermore, the pyrolysis rate drops by a factor of 100 as the temperature drops by about 60 K, since PMMA pyrolysis has high activation energy. Therefore, for the purpose of this discussion we defined the pyrolysis zone as 600 to 700 K temperature range in Fig. 5. Hence, the thermocouple is assumed to be in the PMMA condensed phase when it reads temperatures below 600 K. During this time the thermocouple readings represent real time changes in the temperature inside the condensed phase at the thermocouple location.

About 100 seconds into the test the thermocouple reads the lowest temperature (close to 400 K). This corresponds to stage 2, when the flame stabilized about 5 mm downstream from the thermocouple and it is farthest from the thermocouple. Meanwhile, a step is formed where the flame stabilizes and starts spreading upstream. As the flame spreads, heat is transferred from the flame to the solid ahead of the flame front. This raises the temperature at the thermocouple location from about 400 K to the pyrolysis temperature in about 200 seconds as shown in Fig. 5. As the flame creeps up to the thermocouple location, the polymer pyrolyzes exposing the thermocouple bead to the hot gases. Thereafter, the thermocouple measures the hot gas temperatures of the flame. The valley has now extended beyond the thermocouple location as seen in Fig. 4b (stage 3).

To examine the surface profiles more closely during each of the three stages described above, separate experiments were conducted, where the flame was extinguished during each stage. The sample was allowed to cool down and was cut stream-wise along the centerline to show the surface profile. These experiments were performed with 18.8% ± 0.1% oxygen concentration so as to obtain a significant quenching distance. Figure 6a shows the picture of the surface profile as the flame front was retreating from the sample leading edge (stage 1). It shows that the entire surface is flat and that the degree of pyrolysis during the ignition process and the short burn time had a negligible effect on the shape of the surface. Figure 6b shows the surface profile when the flame had just stabilized about 45 mm downstream from the sample leading edge. The surface was still nearly flat except for a tiny step at the location where the flame has just anchored. At this location, the reaction time is expected to be comparable to the flow time. The flame did not stay long enough at this location to create a deeper step before it was extinguished. Figure 6b also shows a tiny step formed at the leading edge of the sample before the flame was dislodged in this experiment. This tiny step was not adequate to stabilize the flame at the leading edge of the sample where the effective velocity was higher. At a fixed oxygen concentration and free stream velocity, the flame stability may depend both on the distance from the leading edge and on the size of the step for small size step.

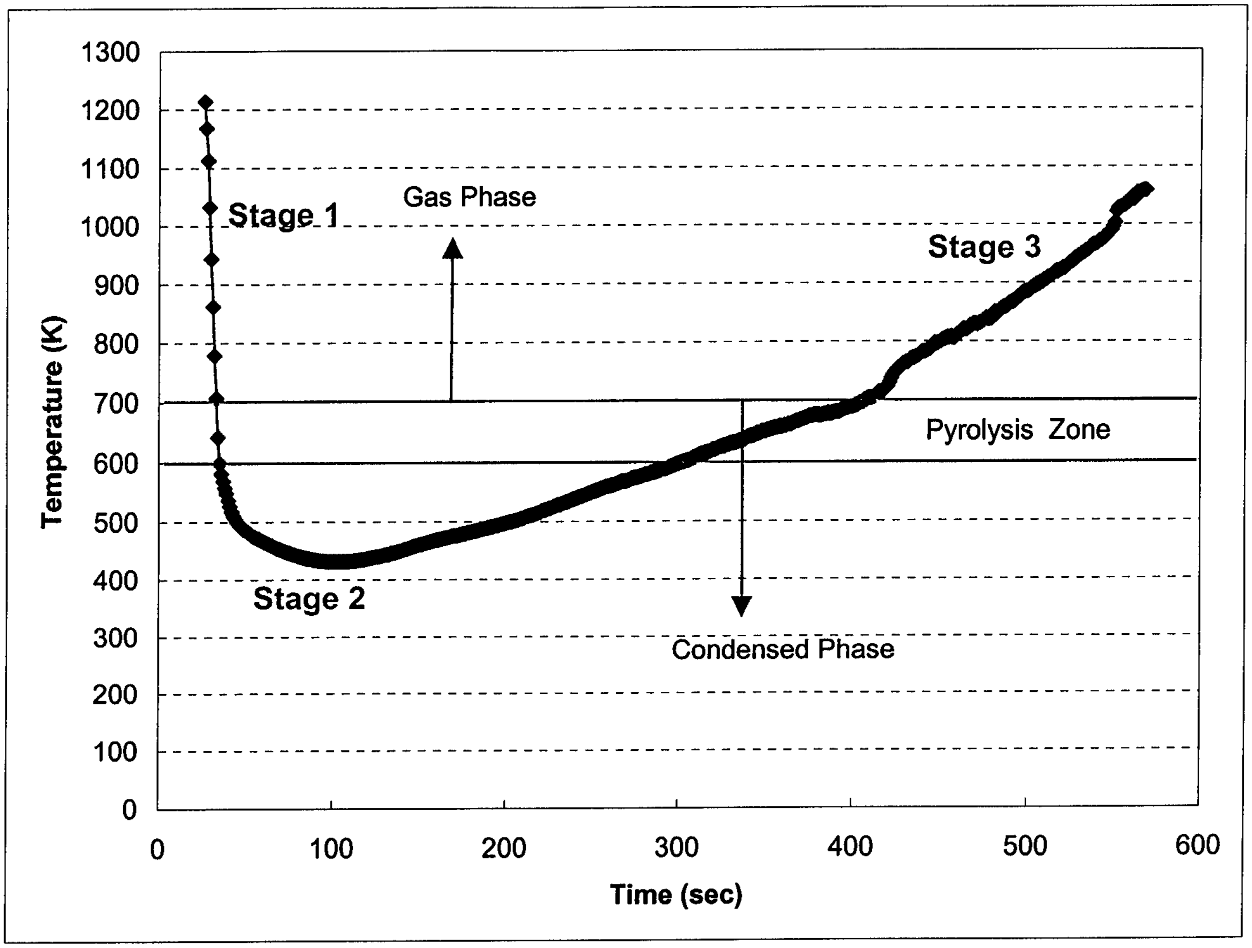


Figure 5: Time dependent temperature of a point within the quenching distance as the flame retreats (stage 1), stabilizes (stage 2) and spreads within the quenching distance (stage 3).

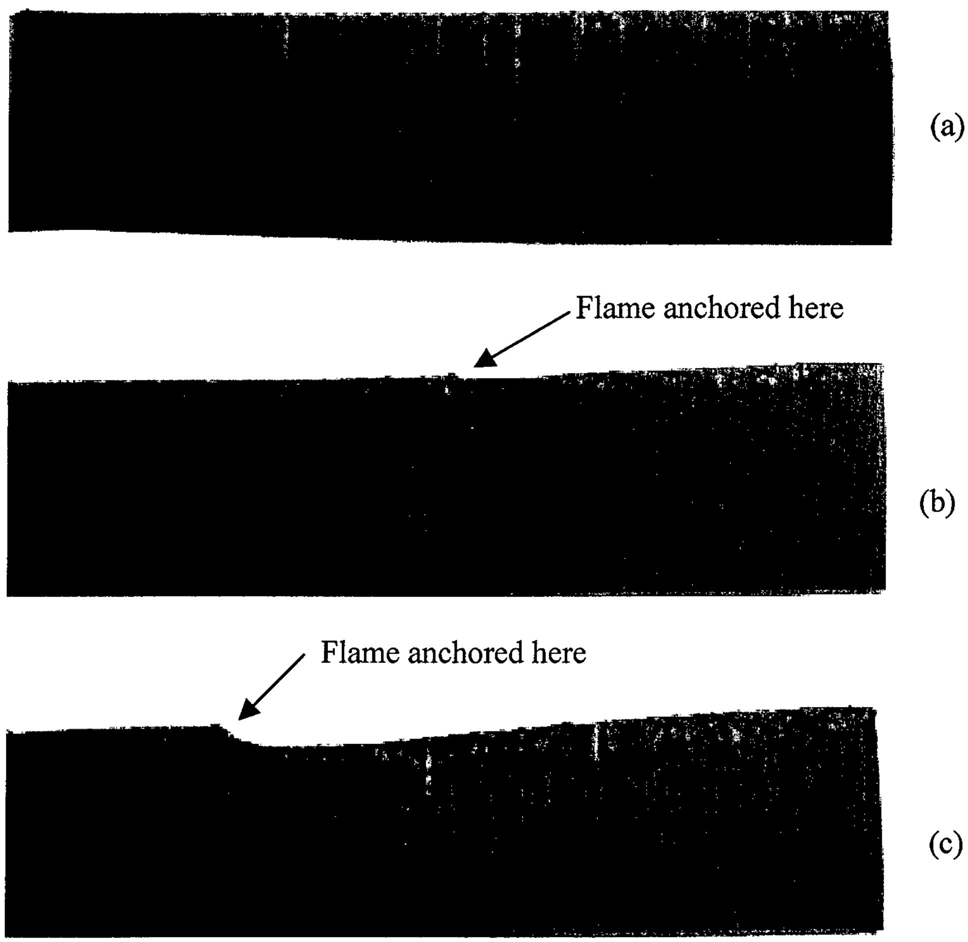


Figure 6: Sample surface profiles for tests with 18.8% Oxygen and flame extinguished (a) while flame was retreating from the leading edge (stage 1); (b) just as flame stabilized (stage 2) and (c) after flame has spread upstream for 5 minutes (stage 3).

Therefore, a bigger step than seen in Fig. 6b would be needed to stabilize the flame within the quenching distance. In Fig. 6c we show a picture of the surface profile in an experiment where the flame stabilized and was allowed to spread upstream for 5 minutes before it was extinguished (stage 3). In this experiment, the flame initially stabilized nearer the sample leading edge than in Fig. 6b, creating an initial quenching distance less than 45 mm. This indicates that the length of the quenching distance can vary between experiments even when the oxygen concentrations and inlet velocities are the same. Figure 6c shows a bigger valley than in Fig. 6b, indicating that a bigger valley is needed to sustain the flame as it spreads upstream closer to the leading edge. This is further illustrated in Figs. 7a-c.

Figures 7a - c show sample surface profiles in tests with 19.4% oxygen, where the flame was allowed to spread upstream for 5, 10 and 20 minutes, respectively. Like the flame described in Fig. 4a and b, the flames in Figs. 7a - c stabilized about 20 mm downstream from the sample leading edge and as the flames spread upstream the valleys get deeper and broader. The initial point of stabilization (X=20 mm) was exposed to the highest flux from the flame for the longest time than any other part of the surface during the flame spread process. Therefore, the deepest point (highest regression distance) in the valley is expected to occur approximately at the initial point of flame stabilization. As the flame gets closer to the leading edge, it encounters increasing air velocity. Therefore, a bigger valley is needed for the flame to be stable. The surface profiles in Figs. 7a - c clearly show increased depth of valley with time that enables the flame to sustain and spread.

Figure 8 shows the profile of the moving boundary with time. The surface regression shown in Figure 7a - c are plotted in Fig 8 against the distance X from the sample leading edge such that zero regression indicates flat part of the surface. Therefore, the points on the abscissa near the leading edge correspond to the leading edge of the valleys in Fig. 7 a - c. The depth of the valley increases sharply with X and reaches a maximum at the point where the flame supposedly initially stabilized. Following the maximum, the depth of the valley tapers down slowly as we approach the trailing edge. The curvature increases with time, as the valley gets bigger. The high curvature of the valley may lower the air velocity, especially at large time, and may create a local circulation zone. The reduced local velocity U would increase Da above the critical value and stabilize the flame. Da is proportional to U^{-2}; hence, a small change in U will cause a larger change in Da. Flame spread in the current experiments is in the chemically controlled regime where spread velocity increases sharply with Da [5]. Therefore, one needs only a small decrease in U (which the step can provide) to make the flame spread upstream within the quenching distance.

The counter-current flame spread rate within the quenching distance can be approximated from the surface profiles in Fig 7 or Fig. 8. In Fig. 7 the flame front was approximately at X=11, 6 and 2 mm from the sample leading edge (X=0) in 5, 10 and 20-minute tests, respectively. The time-averaged spread rate can be obtained as $\Delta X/\Delta$time. Thus, between

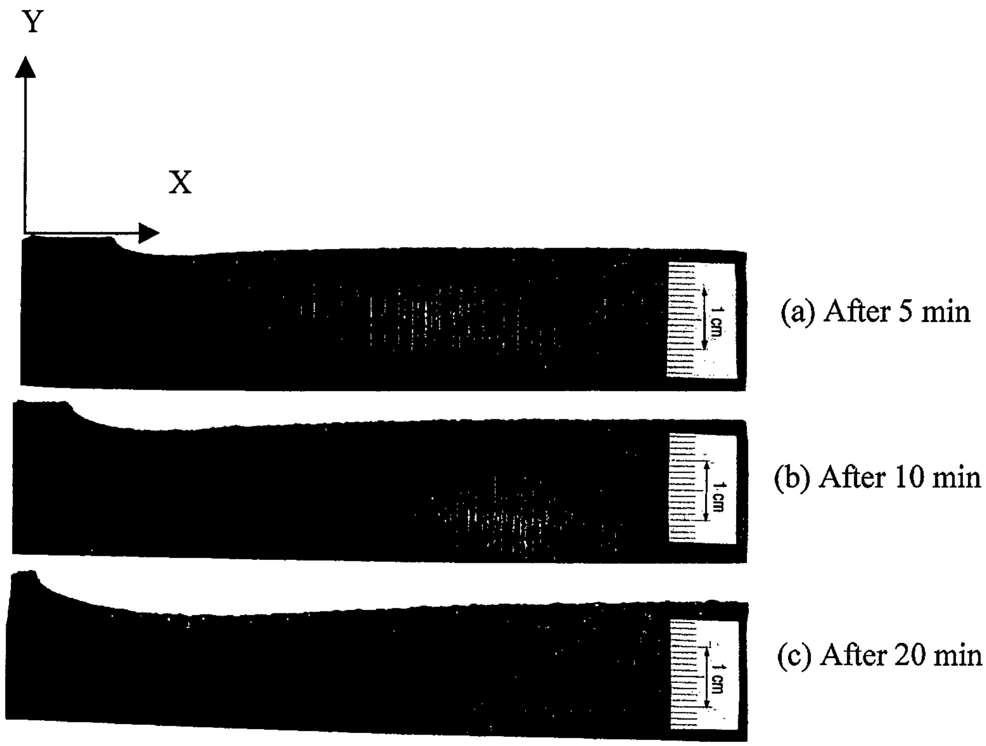


Figure 7: PMMA Surface profiles after (a) 5 min, (b) 10 min and (c) 20-minute burns; 19.4% oxygen, flame spreading within X< 20 mm.

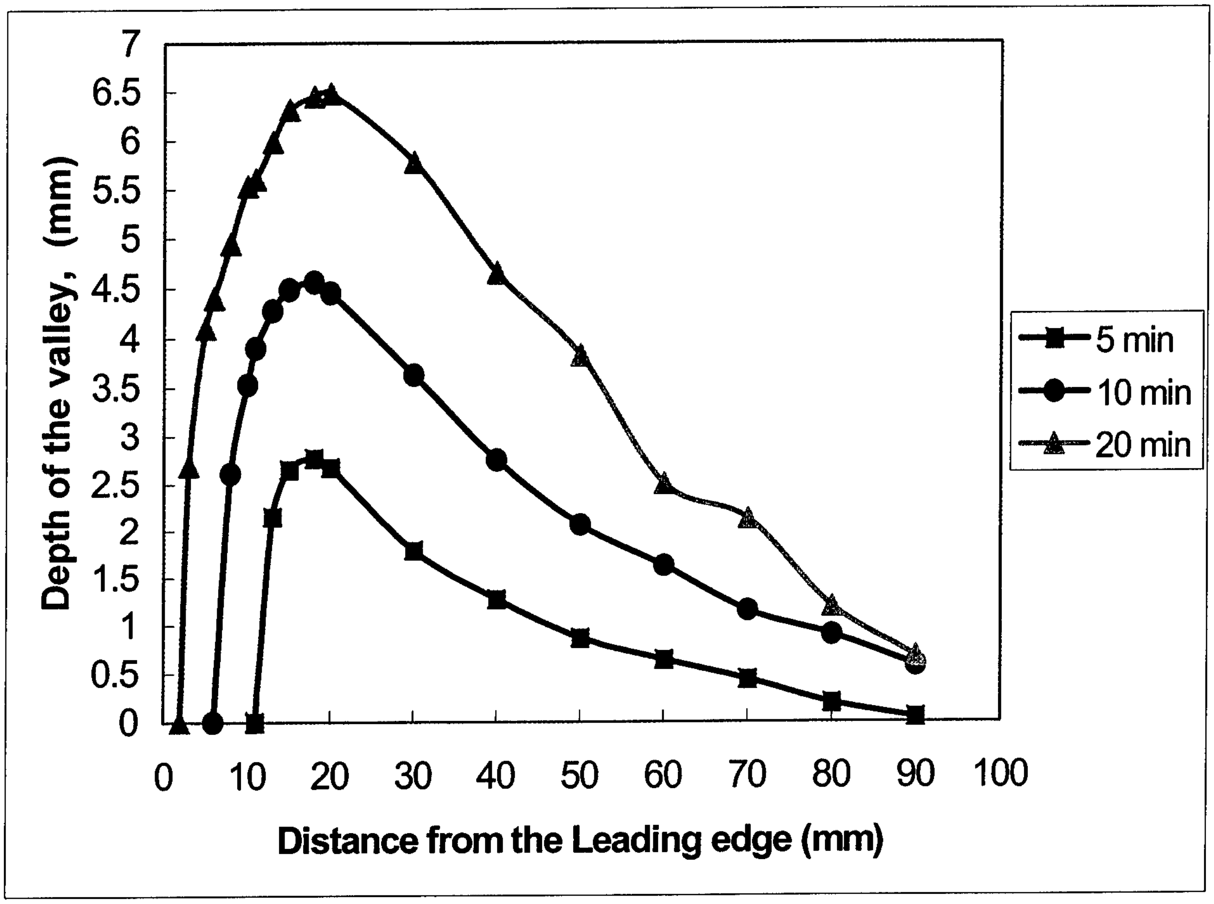


Figure 8: Regression depth at various stream-wise locations for tests lasting 5, 10 and 20 minutes, 19.4% oxygen.

5 minutes and 10 minute after ignition, the time-averaged spread rate is about 1 mm/min; while between 10 minutes and 20 minutes, it is about 0.4 mm/min. Thus, the flame spread rate decreases with time. Furthermore, from Fig. 8 one can obtain time-averaged regression rates at any X location as - depth of valley /time. Thus, the peak regression rates from Fig. 8 are about 0.6, 0.45 and 0.32 mm/min in 5, 10 and 20-minutes tests, respectively. The peak regression rates are also decreasing with time. This is another effect of moving boundary, which we reported in more detail in another paper [11]. It is pertinent to note that the spread rate is comparable to the regression rate under the current conditions and the moving boundary effects are significant. Most existing theories that predict flame spread and burning rates assume a stationary surface. A two-dimensional moving boundary analysis would require that the heat flux balance be written along a normal to the surface rather than along the vertical axis Y. Figures 7 and 8 can be used to estimate the normal velocity to the moving boundary at different times.

The flame spread experiments described here were conducted under lean limit conditions (small Da) where the spread mechanism is chemically controlled unlike in the more traditional flame spread experiments where the Da is large and the flame is stable on the flat surface. A few experiments were conducted with 21% oxygen and air velocity of 84 cm/sec for comparison with the lean limit flame spread tests. In these experiments, the first 75 mm of the sample length was shielded from the radiant panel during pre-pyrolysis and ignition. Therefore, the boundary layer flame was formed on the last 20 mm of the sample before the sample was brought to the exit of the air channel. It was then allowed to spread counter-currently for 5, 10, and 15 min. The surface profile at the end of each test is shown in Fig. 9. From Fig. 9 the peak regression rates are found to be 0.28, 0.28, 0.38 mm/min in 5, 10, and 15 minutes, respectively. The regression rates do not seem to change significantly with time. The spread rates are obtained from the points at zero regression depth to be about 3 mm/min between 5 and 10 minutes and between 10 and 15 minute. The spread rate does not seem to change significantly with time, consistent with measurements reported in the literature [7]. Although this method of measuring spread rate is not "traditional", the results are consistent with those [7, 10, 16] measured in the "traditional" way under similar conditions. Traditionally flame spread has been measured by visually determining the time it takes the flame front to move a known distance or by determining the time it takes for two points on the surface, a known distance apart, to attain the solid gasification temperature.

The spread rates are about 10 times higher than the regression rates, unlike the result obtain in the lean limit flame spread tests. Furthermore, the peak regression rates are significantly lower in Fig. 9 than in Fig. 8 (0.28 vs. 0.6 mm/min.) despite higher oxygen concentration in Fig. 9. Obviously, the spread rate and the regression rate are coupled in the moving boundary problem such that as the spread rate increases the regression rate decreases since at high spread rates, the leading edge of the flame spends less time at any X location.

The results of these experiments have demonstrated that the formation of a valley (moving boundary) is critical for the counter-current spreading of a lean limit boundary layer flame within the quenching distance. The flame would not stabilize within the

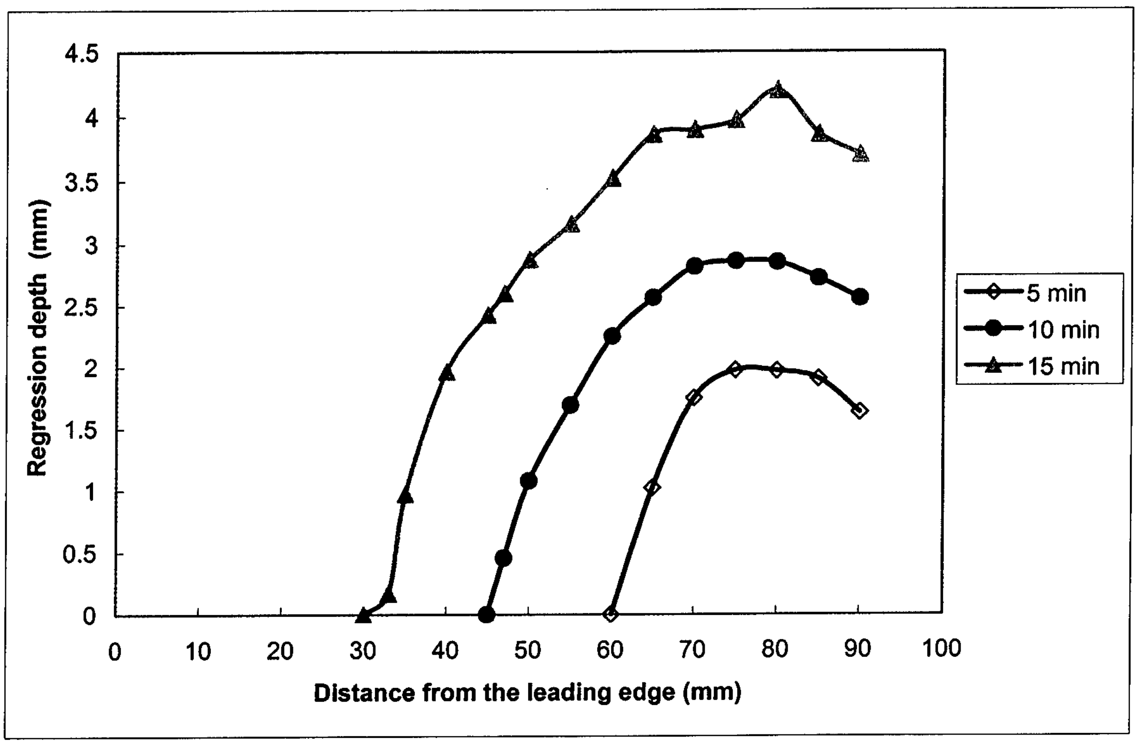


Figure 9: Regression depth with distance from the sample leading edge at various times, 21% Oxygen, and flame ignited 75mm from the Leading edge.

quenching distance while the surface was flat, as we have shown. The effectiveness of steps and baffles as flame stabilizers have been recognized in the gas turbine industry, where burner designs exploit the enhanced stability associated with combustion in a re-circulation zone [13]. The presence of a step, baffle plate or bluff body in a flow path creates a re-circulation zone where conditions are favorable for flame holding, e.g., lower velocity, enhanced mixing of fuel, air and hot combustion products and augmented heat transfer to the condensed fuel surfaces [13]. The presence of a step may be advantageous for flame holding and stabilization, but it becomes a disadvantage in flame suppression and extinction. Takahashi et al. [17] studied the stabilization and suppression of methane flames formed behind a step in a wind tunnel. A re-circulation zone was formed behind the step and they showed that the minimum mass fraction of the agent required to extinguish the flame increased with the volume of the re-circulation zone. It is therefore expected that effects of a moving boundary would reduce the effectiveness of suppressing agents in boundary layer fires over non-charring solids.

4.0 CONCLUSIONS

The results presented above reveal the effects of a moving boundary on counter-current boundary layer flame spread over a non-charring solid near the flame extinction limits. Under this condition the flame spread rate is time dependent and comparable in magnitude to the surface regression rate, unlike in the traditional flame spread experiments where Da is large. Tests were conducted at low oxygen concentration ($\leq$19.4%) and air velocity of 87 cm/s. This created an initial quenching distance ($\geq$20 mm) where the flame could not sustain while the surface was pyrolyzing and flat. As the flame stabilized, a small step is formed under the leading edge of the flame where the heat feedback is the highest. This small step was observed to grow deeper and bigger with time. The step (valley) appears to play a critical role in the flame stabilization and spread within the quenching distance, where the flame could not stabilize when the surface was flat. Therefore, the moving boundary affects flame spread and stability significantly and may make it difficult to extinguish the flame. There is a need to include moving boundary effects in current theories of flame spread and burning rate.

5.0 ACKNOWLEDGEMENT

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