Heat and Mass Transfer from Arrays of Flame Jets to a Rotating Cylinder and its Application to the Surface Modification of Polymer Films

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INTRODUCTION

The coupling that occurs between the fluid mechanics and the chemical kinetics at the interface of a gas and a surface is central to many processes, including semiconductor fabrication, turbine blade degradation, and adhesive tape manufacturing. During a critical process step in the fabrication of adhesive tapes, a combustion reaction is employed to alter the surface of the tape's polymer film substrate. A schematic of the process is shown in Figure 1a; the film is routed between a ribbon burner and a rotating, water-cooled drum. Species present in the post-flame gases oxidize the surface, increasing the wettability and surface energy of the film.^[1] The flame treatment process is used to modify several billion square meters of polymer film each year. In this study, the modification of polypropylene film exposed to a laminar, premixed methane - air flame is investigated using experimental and numerical techniques. The goal of the study is the understanding of the gas phase phenomena occurring between the flame jets and the moving impingement surface, the effects of the fluid mechanics on treatment efficacy, and the identification of gas phase species responsible for treatment.

EXPERIMENTAL APPARATUS

A schematic of the laboratory simulation of the flame treatment apparatus is shown in Figure 1. In the process, one side of 0.04 mm thick polypropylene film is exposed to a laminar premixed methane - air flame. The backside contacts a rotating steel roll that is 25 cm in diameter and is cooled by recirculating water. This experimental apparatus is designed to observed the fluid mechanics occurring between the hot combustion gases and the cool rotating roll. For safety and practicality reasons, the polymer film is not part of the investigational facility; the rows of flame jets impinge directly on the cylinder surface. The industrial burner is shown in Figure 1b, and is a series of 35 $cm \ge 1$ cm corrugated stainless steel ribbons packed into a cast-iron housing. Conical flames 2 mm in diameter are produced; four staggered rows comprised of 150 conical flames per row span the 35 cm burner width.^[1]

Experiments were conducted under typical film processing conditions with the burner producing 11.7 kW (60,000 Btu/hr), and the burner-to-roller separation distance set at 10 mm. Temperature measurements were made between the burner and roller surfaces using an R-type thermocouple mounted to a three-axis traverse. For observation of the flow developing between the flame and the rotating roller, a color Schlieren imaging system was assembled. By employing a bulls-eye Schlieren stop, radial temperature gradients around the cylinder are colored from high (red) to low (blue).^[2]







RESULTS AND DISCUSSION

A fundamental issue when applying impinging jets in any heat or mass transfer process is the need to maximize not only the heat or mass transfer to the surface, but to also maintain uniformity across the surface.^[3] In the case of multiple impinging jets, the choice of separation distance between the burner and impingement surfaces is a critical parameter in obtaining optimal (uniform and high) transfer. Multiple jets interact with one another prior to impingement as well as after striking the wall. To prevent non-homogeneous surface treatment, uniform heat and mass transfer requires that the gases from these jets are well mixed prior to impingement. The combustion reaction increases the complexity of the problem. To observe jet mixing, a thermocouple was used to take measurements of the temperature field existing between the burner and roller; radiation corrected results are shown in Fig. 2.^[4]

High temperature gradients are observed at low distances above the burner (Fig. 2a), as little interjet mixing has occurred, and a significant amount of unreacted gas exists at this low position. At greater heights above the burner, more of the combustion reactants are consumed, and hot-gas mixing prior to impingement becomes more pronounced (Fig. 2b). An environment with moderate gradients is present at a height of 8 *mm* above the burner surface where more uniform gas phase conditions exist.

Figure 2c depicts how the observed temperature field effects the resulting polypropylene treatment. One diagnostic technique for measuring wettability is observation of the contact angle that a drop of water makes with a surface; the lower the contact angle, the higher the wettability and surface energy. For comparison, untreated polypropylene has a receding contact angle of 103° . As seen if Fig. 2c, a wettability profile that is nearly uniform across the film is obtained when treatment occurs at a separation distance above 8 *mm*. Treatment at lower separation distances results in non-homogeneous surface energy, with variations that match the jet-to-jet spacing of the flames produced by the burner (~ 2.5 mm). It is interesting to note that a higher treatment level is observed at a lower separation distance, but poor treatment uniformity prohibits processing at this level.^[1]

The problem of uniform surface treatment presents a compromise. It is commonly believed that the polymer is modified by surface oxidation from reactive radicals in the post-flame gases.^[1] The concentration of these radicals is low and decreases with distance from the flame zone. The large separation distance required for uniform treatment decreases the availability of reactive radicals that promote treatment. By making measurements like those shown in Fig. 2, designers can better address burner optimization for the flame treatment process.

The impingement of hot flame gases upon the cool, rotating cylinder gives rise to complex fluid dynamics that have been shown to play a central role in surface modification. Through color Schlieren imaging, visualization of this flowfield has been performed; results are shown in Fig. 3. The circular dark area is the rotating chill roll with the light colored burner positioned directly below it. While Figs. 3a and 3c appear to show flames spanning the entire chill roll, the actual flame zone exists in a small area located directly between the burner and roller, as observed in Figs. 3b and 3d. The high gradient red areas indicate mixing between post-flame gases and ambient.

At low rotational speed (10 *rpm*, Figs. 3a and 3c), the boundary layer is balanced upstream (right) and downstream (left) of the burner. At higher rotational speed (450 *rpm*, Figs. 3b and 3d), the flow becomes more turbulent, the upstream boundary layer thickens, and an upstream recirculation zone is observed in Fig. 3d, while the downstream flow remains relatively well behaved. The upstream interaction between the ambient fluid entrained by



Figure 3: Schlieren images at 10 RPM (a and b), and 450 RPM (c and d)

the rotating chill roll and the hot gases emanating from the burner implies that relatively little mass transfer occurs in the upstream direction. In contrast, a favorable pressure gradient exists downstream, and downstream boundary layer separation is delayed; this may marginally promote mass transfer in this direction.

The observed flow structure plays a significant roll in the problem of non-homogeneous treatment, as burner design does not appear to take the observed fluid mechanics into account. Due to their close proximity, the two central rows of flames (Fig. 1b) compliment each other well in providing homogeneous gas conditions across the burner surface; they are barely distinguishable in Figs. 3b and 3d. In contrast, the upstream and downstream ports fail to balance one another as the effectiveness of the upstream ports is reduced from mixing with ambient. The temperature measurements and Schlieren imaging show that non-uniform treatment is caused by two complementary factors: insufficient inter-jet mixing prior to impingement at low burner-to-surface separation distance, and suboptimal burner design to account for the non-uniform flow around the rotating cylinder.

The fluid mechanics observed in the previous figures impart significant strain on the methane - air flame. The strain applied when a flame jet strikes a stagnation surface has been observed to cause unexpected secondary reactions in the boundary layer, reactions may be relevant to flame treatment.^[5] A key element in understanding the modification of polymer surfaces by flames is identification of the gas phase species responsible for treatment. To distinguish such species, the SPIN application of the CHEMKIN software library was used to model this process.^[6]

The SPIN software solves the two-dimensional, steady-state equations governing laminar, chemically reacting flow impinging against an infinite disk. It was initially designed for the study of chemical vapor deposition processes used in microelectronics fabrication. In the numerical experiments conducted in this work, a methane - air flame impinges on a stationary, inert surface. The gas is initially at 300 *K* and flowing at 100 *cm/sec*; the surface is held at 400 *K*. The code employs the GRI 2.11 chemical kinetics mechanism^[7] (49 species, 277 reactions) to calculate the temperature, velocity, and species concentration across the 10 *mm* computational domain. Results are shown in Fig. 4.

In Fig. 4a, the general structure of the flame is evident. Gases initially at 100 *cm/sec* slow down before igniting at the flame speed of 40 *cm/sec* near x = 5 mm. The heat released in the combustion reaction boosts the velocity to 225 *cm/sec*; the gases again slow down before stagnating at the wall (x = 0 mm). The temperature exceeds 2000 *K* after the flame zone, but heat transfer to the surface reduces the gas temperature to 400 *K*. Fig. 4b shows the behavior of flame radicals in the high gradient zone near the wall (x < 1 mm). The radicals most commonly associated with polymer surface modification, atomic hydrogen, atomic oxygen, and hydroxyl radical, are reduced in concentration in this zone, reaching values of 215, 3, and 112 *ppm*, respectively. At this low concentration, it appears unlikely that atomic oxygen is a significant contributor in polymer surface modification. However, the same reactions responsible for the decreases in O, H, and OH radicals cause sharp increases in the concentration of hydroperoxy radical (HO₂) and hydrogen peroxide (H₂O₂). These highly reactive species reach concentrations of 14 and 33 *ppm* at the surface, implying that they may play a limited role.



Figure 4: a) Temperature and velocity profiles for the impinging flame jet; b) species concentration at the stagnation surface; c) species concentration and treatment level versus flame equivalence ratio

Figure 4c shows how these species behave under varying flame equivalence ratios. Previous work has shown that film wettability is maximized at $\phi = 0.93$. The SPIN simulations show a correlation between the concentration of the radicals OH, HO₂, and H₂O₂ and this wettability peak. Atomic hydrogen is believed to impede oxygenation of the polymer surface, and its steep increase at $\phi > 0.95$ may explain the drop in wettability under rich conditions. Further information on the chemical mechanism involved in polymer film treatment is found in Ref. [1].

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