Cigarette Smoke Formation Studies

IV. The Dynamic Measurement of the Axial Density of a Burning Cigarette*

by

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INTRODUCTION

Tobacco researchers have expended considerable effort in the study of the temperature profiles within a burning cigarette (1, 2, 3). Baker (1) has produced a series of profiles illustrating the spatial temperature contours at specific time intervals in the puffing cycle. These temperature gradients are directly comparable with the basic processes taking place in the cigarette, i.e. distillation, pyrolysis and combustion. These events all have the net effect of removing atoms and molecules from the tobacco, and should be observable as a change in the absolute density as the tobacco is converted to smoke and ash. The measurement of these density changes in a burning cigarette has been accomplished using a beta ray attenuation technique.

MATERIALS AND METHODS

Apparatus

The experimental array is shown in Figure 1. A 20 mCi ^{32}P -source (New England Nuclear Corp.) was purchased as a .02 N solution of phosphoric acid. This material was transferred into an aluminum cylinder (6.35 mm by 4.76 mm) containing a thin aluminum foil window at one end. After the removal of the water by heating, the capsule was sealed. The ^{32}P -cylinder was mounted onto the source collimator so that the window was flush with the plate and concentric with the collimator hole.

Beta detection was accomplished using an organic quenched Geiger-Müller detector (Nuclear Associates, 1300 V) connected to an amplifier (Harshaw) and a timer (Harshaw), then to the multichannel scaling mode of a multichannel analyzer (Northern, NS 601). All data were held in memory until each individual run was complete, whereupon the data were transferred to punched paper tape.

The collimators are 6.35 mm thick plates, each containing a 2.38 mm hole drilled perpendicular to the plane of the plate and mounted so that both holes are parallel to the beta beam (Figure 1). The source collimator is made of aluminum, a low atomic number material to reduce bremsstrahlung production. The detector collimator is made of lead to prevent any bremsstrahlung that was produced in the aluminum or in the cigarette from reaching the detector by passage through the collimator itself. The lead detector collimator also serves to screen out any scattered beta particles. The size of the holes was chosen as a compromise between the relative counting background and spatial resolution.

An attenuation curve was established using aluminum absorbers of varying thicknesses. A mathematical model of the tobacco distribution within the cigarette has been established by Williamson et al. (4), and was used to calculate the correction that must be made to the density values obtained from the calibrated attenuation

Figure 1. Apparatus.

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curve. This correction is necessary due to the inhomogeneity of the mass distribution for the strands of tobacco in any cross-sectional view of a cigarette. This correction says that a simple homogeneous model will underestimate the density of a cigarette by 6.5% and, therefore, all empirical values were corrected using this factor to arrive at the absolute cigarette density.

A syringe-type single port smoking machine fitted with Cambridge pad and holder was employed to generate the puffing sequence. The cigarettes were smoked under standard *Coresta** conditions (one puff per minute of 35 cm³ volume over a 2-second duration).

RESULTS AND DISCUSSION

Density Measurements into the Cigarette Coal

Density data were collected on burning cigarettes under static and standardized puffing conditions. Cigarettes for density measurements into the burning coal were a typical English type Virginia blend tobacco and were 70 mm long by 8 mm diameter, similar to those used by *Baker* (1) in his studies. These were conditioned at 60% relative humidity and 21 °C before smoking. In the static burn mode, the beta beam was passed through the cigarette at a point 3.5 cm from the lighted end. Counts were taken every five seconds after a single lighting puff. This continued until after the coal and ash had passed through the beta beam. The density values for each count were calculated from the density-count rate attenuation plot. Since counts were taken every five seconds, the data plot became a density-time profile. For the static burn, the time axis is also a distance axis as the static burn rate is 5 mm per minute. Figure 2 shows the density plot arrived at in this manner. For reference, the char line position is shown as 0 mm. It is important to note that much of the decline in density (presumably due to loss of distillables) occurs before the char line region is reached. This loss of material starts at a point about 2 mm in front of the visual char line. This reduction in mass indicates that the distillables, as well as the pyrolysis products which are formed, begin to move away from a region 2–3 mm inside the paper tube with reference to the external char line. Figure 2 also includes an overlay of the temperature profile during static burn as reported by *Baker* (1). Here are seen some striking similarities, as the onset of density change correlates well with the 100–300 °C temperature range. *Baker* (1) does not show values below 300 °C so it is necessary to extrapolate to the lower values. The resolution of these density measurements does not permit the separation of the individual processes that are taking place in this region. From a point of about –1 mm to +1 mm we know that distillation and pyrolysis are occurring. *Baker's* (1) report of a high temperature region some 5 to 6 mm in front of the char line is also verified in our density data. The

Figure 2. Static burn.

1. Baker, R. R.: High Temp. Sci. 7 (1975) 236–247.

plateau that occurs from +2 to about +6 mm into the coal agrees with *Baker's* (1) high temperature region of 775 °C. This region in the coal is predominantly composed of carbon undergoing oxidation. The continuous process of oxidation finally gives rise to the high temperature region at +6 mm, where the last vestiges of carbon are removed. The conversion to inorganic ash is concurrent with a final drop in density before the ash begins to sag and drops out of the beta beam as indicated on the figure just beyond +12 mm.

Figure 3. Puffing burn.

1. Baker, R. R.: High Temp. Sci. 7 (1975) 236–247.

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In a study of the puffing effects, cigarettes were smoked in the standardized manner until they had passed through the beta beam. Figure 3 shows these data overlayed with Baker's (1) temperature distribution in the coal, 4.0 seconds after the end of the puff cycle. This temperature profile was chosen for descriptive purposes since, in actuality, our density measurements are an average of static and puffing effects over the entire range. Here again the temperature zones appear to correlate with the observed density changes. A comparison of the puffing density curve with that obtained during static burn shows that the density change during puffing occurs over a longer region of the cigarette, from -2 to $+4$ mm, whereas in static burn, this region was from -2 to $+2$ mm.

This faster drop in density as a function of length indicates the temperature gradient during static burn to be steeper (occurring in a shorter rod length) than in the puffing mode. A logical finding since the flow of hot gases from puffing would be expected to elongate the temperature gradient.

Measurements into the Tobacco Rod

In an attempt to examine in more detail the density changes that may occur in the tobacco rod behind the coal, a series of density determinations were made using the Kentucky Reference 1R1 cigarette. The method employed was to take density measurements at several points along the rod, making note of the position of the puffs. Cigarettes were placed in such a manner that the beam passed through the diameter at 55 mm as shown in Figure 4. Puffs were taken every 5 mm, starting at the first 5 mm from the lighted end.

Counting data were taken every 4 seconds until a series of five cigarettes had been assayed in the same manner. All data were then averaged. Figure 5 is a plot of this averaged density profile as a function of rod distance from the char line. One can easily see that after the second puff there is a definite increase in density per puff as the coal nears the measurement point. Puff 1 and puff 2 show no change, indicating that there is very little condensation from the vapor state taking

Figure 5. Density in unburnt tobacco rod.

place at distances of 20 mm beyond the char line. However, after the second puff, one can see that the rate of density change in the rod during the static burn (lines through the data points are linear least square fits), changes as a function of puff count. There is a definite slope change from puff 2 through puff 5. These changes in the tobacco rod density can be attributed mostly to the buildup of condensed water (plus some organic volatiles) from the vapor phase of the smoke and its subsequent re-evaporation. In order for the rate of re-evaporation of this water to change as the rod position nears the coal, one can assume that there is either an increased tobacco temperature or a change in the composition of the atmosphere over the tobacco strands. Egerton (2) has shown that at a distance of about -10 mm, the temperature of the tobacco rod is still about 50°C . Since the rate of evaporative loss of water starts to change in this range (12 – 15 mm), the increase in tobacco temperature may be a major factor accounting for the variation in rate of water loss. Also of unknown magnitude is the effect of the change in the relative humidity of the atmosphere over the tobacco strands. This atmosphere is constantly changing due to a loss of moisture by diffusion. Both of these factors probably account for this fluctuation in rate of water loss from the tobacco column.

Figure 6. Static burn moisture loss.

In another experiment the cigarette was puffed four times to encourage the buildup of condensables on the tobacco column and then permitted to burn in a static mode for 1 to 2 min through the beta beam. These data, shown in Figure 6, indicate that a total decline of about 2.5 mg/cm^3 in density takes place in the first forty seconds. During the next approximately forty seconds the density remained constant, and over the final forty seconds, a density increase of about 2.5 mg/cm^3 was observed. Finally, with the passage of the coal, the sudden loss in density occurs. The cause of the late increase in density is only speculative at this stage, but may be due to the condensation of tobacco distillables that have a relatively high vapor pressure (compared to water). It has been demonstrated (5) that material buildup in the butt occurs mostly in the first 10 mm behind the char line. These condensed constituents may account for this observed increase in density.

Figure 4. Reference points on the cigarettes.

Figure 7. Single puff resolution.

A more detailed density profile was desired on the changes taking place within the tobacco rod during and after a single puff. Figure 7 shows data collected from a Kentucky Reference 1R1 cigarette using a 1 second count rate. This single puff resolution plot is the average data from 5 replicate runs taken for the 3rd puff at a point 15 mm behind the char line. One can see a steady decline in density until the onset of the puff marked by the arrow. At this point an immediate increase in density occurs due to condensing water, followed by a slow decline in the density relatable to re-evaporation of moisture.

The appearance of a smooth increase in density during the puff could also be interpreted as evidence that water is uniformly distributed within the 35 cm³ puff volume. If water were more concentrated in the beginning or later portion of the 35 cm³ puff volume, an irregular shape would have been observed in the density increase.

In summary, we have demonstrated a technique using beta attenuation to measure dynamically the density profile of a burning cigarette. The density changes which occurred have been correlated with known temperature profiles of cigarettes. In addition, the buildup and re-evaporation of condensable water on the tobacco rod is observed. Detailed density analysis of the rod immediately before the coal has passed shows evidence for the buildup of the condensable portion of smoke in the tobacco butt.

SUMMARY

Much research has gone into establishing the temperature gradients that exist inside a burning cigarette. These temperature gradients should correspond to changes in the density of the tobacco column due to the condensation and subsequent re-evaporation of volatiles resulting from pyrolysis, combustion, and final ash formation. These dynamic measurements on the burning cigarette were made using a modified beta thickness gauge. A beam of collimated beta particles from a ³²P-source is projected through the side of a cigarette at a fixed position. The cigarette is allowed to burn past the focused beta beam. The number of transmitted particles is dependent upon the density of the mass penetrated by the incident particles, and is reflected as a change in the number of counts detected per unit time.

A density profile of the burning cigarette has been established for both the puffing and static modes. These density changes are correlated with published tempera-

ture profiles. The buildup of the volatile materials (mostly water) on the tobacco rod behind the pyrolytic region is presented. The rate of evaporative losses of these condensables is shown to be dependent on puff count (cigarette length). A detailed density profile of the condensing materials from a single puff has been established. The relationships of these density changes to current smoke formation theories are discussed.

ZUSAMMENFASSUNG

Auf die Untersuchung der Temperaturgradienten in einer brennenden Zigarette ist viel Forschungsarbeit verwendet worden. Es wird vermutet, daß Beziehungen zwischen den Temperaturgradienten und Veränderungen in der Dichte des Tabakstranges bestehen. Diese Dichteänderungen sind auf die Kondensation und die erneute Verdampfung flüchtiger Verbindungen zurückzuführen, die durch Pyrolyse, Verbrennung und schließlich durch Aschebildung entstehen. Die hier beschriebenen dynamischen Messungen an der brennenden Zigarette wurden unter Benutzung eines modifizierten Beta-Dickenmeßgerätes durchgeführt. Ein Bündel ausgeblendeter Beta-partikel aus einer ³²P-Quelle wird an einer bestimmten Stelle seitlich durch die Zigarette projiziert. Die Zigarette brennt dabei bis hinter den fokussierten Betastrahl. Die Anzahl der wieder ausgetretenen Partikel ist abhängig von der Dichte der von den einfallenden Partikeln durchstrahlten Masse und zeigt sich in einer Veränderung der Zählrate, die je Zeiteinheit beobachtet wird.

Sowohl während des Abrauchvorganges als auch während des statischen Abbrennens sind Dichteprofile erstellt worden. Die beobachteten Dichteänderungen werden zu in der Literatur veröffentlichten Temperaturprofilen in Beziehung gesetzt. Die Ansammlung von flüchtigem Material (meistens Wasser) im Tabakstrang hinter der Pyrolysezone wird dargestellt. Es zeigt sich, daß die Größe der Verdampfungsverluste der kondensierbaren Substanzen abhängig ist von der Zugzahl (Zigarettenlänge). Ein detailliertes Dichteprofil der kondensierenden Stoffe eines einzelnen Zuges wird dargestellt. Die Zusammenhänge dieser Dichteänderungen mit den aktuellen Theorien über die Rauchbildung werden diskutiert.

RÉSUMÉ

Beaucoup de recherches ont été faites pour déterminer les gradients de température existant à l'intérieur d'une cigarette en combustion. Ces gradients de température devraient correspondre à des changements de densité du boudin de tabac, dus à la condensation suivie par la réévaporation de matières volatiles provenant de la pyrolyse, la combustion et finalement la formation de cendres. On a capté ces données dynamiques d'une cigarette en combustion à l'aide d'une jauge d'épaisseur aux rayons bêta (modifiée). On projette un faisceau de particules bêta provenant d'une source de ³²P à travers le côté d'une cigarette et ce à un endroit fixe. La cigarette

peut se consumer jusqu'à derrière le faisceau bêta. Le nombre de particules transmises dépend de la densité de la masse que pénètrent les particules incidentes, et est observé par la modification du nombre d'enregistrements par unité de temps.

On a établi un profil de densité de cigarette dans les deux états, soit combustion active (pendant la bouffée) ou passive (intervalle statique). Les changements de densité observés ont été mis en rapports avec des profils de température publiés. On présente la formation des produits volatiles (en majorité de l'eau) sur le boudin de tabac en deçà de la région pyrolytique. Le taux de pertes par évaporation de ces produits condensables semble dépendre du nombre de bouffées (longueur de la cigarette). On a établi un profil de densité détaillé des produits de condensation d'une bouffée. On discute les relations entre ces changements de densité et les théories courantes de formation de fumée.

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