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Diffusion of Gaseous Components through the Wrapper of a Cigarette*

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INTRODUCTION

The diffusion of carbon monoxide from a burning cigarette has been a subject for study by several groups of workers. During a puff, CO is generated both at the coal and in the pyrolysis region immediately behind the coal (1). In the nearly isothermal region of the rod, commencing about one centimeter behind the char line, mixing and dilution of the gases occur as well as diffusional exchange of the gases through the wrapper with the atmosphere. The early work of *Owen* and *Reynolds* (2) and *Newsome* and *Keith* (3) indicated that an appreciable amount of CO generated at the coal was lost to the atmosphere by diffusion through the paper. Other work has shown (4, 5, 6) that as much as 30% of the CO generated in the coal region could diffuse out. *Baker* and *Crellin* (7) have recently measured the diffusion of CO from unlit cigarettes as a function of the degree and type of ventilation. They also developed simplified models which predict the diffusion of CO from cigarettes.

In each of these studies, however, the losses of CO were determined indirectly by attributing to diffusion the difference in measured CO delivery from that expected if the paper served merely to allow the smoke to be diluted. Moreover, in those studies in which unlit cigarettes have been used without a device to simulate the pressure drop of the coal, the effect of this pressure drop upon the flow of dilution air is ignored.

Partly as a result of the introduction and increased popularity of low delivery cigarettes, the mechanisms for both the generation and diffusion of CO have become topics of renewed interest. *Mattina* and *Selke* (8) discussed the effect of modifications to the cigarette wrapper on the generation of CO. During the course of their work, it became apparent that diffusion might in fact be responsible for some of the effects which the authors ascribed to differences in the generation of CO resulting from changes in the character of the burning coal. Therefore, it was decided to measure directly the amount of CO diffusing from a cigarette. This work was performed on unlit cigarettes using a simulated puffing regime and equipped with a device to mimic the coal pressure drop encountered in experiments with lit cigarettes.

The diffusion of gases from a burning cigarette is dependent upon the vacuum distribution within the cigarette. In general, this vacuum distribution will be different for lit and unlit cigarettes. The use of unlit cigarettes under steady state flow conditions is attractive from the standpoint of precision and ease of measurement. For this reason, an artificial coal consisting of a length of capillary tubing was employed on unlit cigarettes used in these experiments in order to achieve pressure drops similar to those existing in the coal region of a lit cigarette. All measurements simulated the first puff, when diffusion is most important.

MATERIALS AND METHODS

Cigarette paper permeabilities were measured by the *Coresta* method (9). This technique expresses the volume flow of air through a one square centimeter area of paper when a pressure drop of 10 cm of water is applied across the paper. Thus, *Coresta* permeability, reported in units of cm/min, represents a face velocity.

The CO and CO_2 were research quality grades supplied by Matheson. The CO was 4.5 v/v % in nitrogen and the CO_2 13.1 v/v % in air. These concentrations were chosen because they approximate the concentration of these species in the mainstream flow one centimeter downstream from the pyrolysis region in a lit cigarette.

CO and CO_2 concentrations were measured using a Perkin-Elmer model 881 gas chromatograph equipped with a hot wire detector and a 5.0 ml gas sampling

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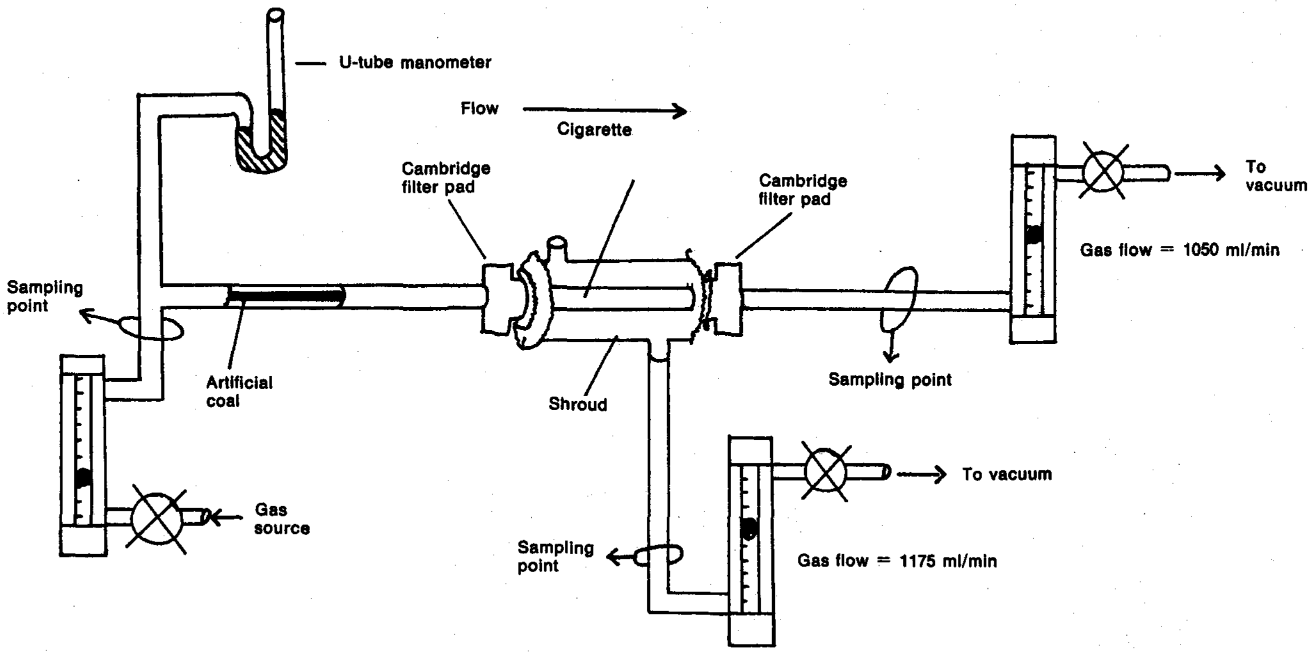
Figure 1. Schematic diagram of experimental apparatus.



valve (GSV). Columns were stainless steel, nine feet long and packed with 30/60 mesh, 5Å molecular sieve.

The apparatus used to measure diffusion of CO or CO_2 is shown in Fig. 1. Each end of the test cigarette was inserted to a depth of 5 mm into a Cambridge filter pad (CFP) holder. The remainder of the cigarette was surrounded by a glass shroud measuring 60 mm × 15 mm. The shroud was fitted at each end with a rubber dental dam seal, and was swept continuously with air via two 6 mm orifices. The shroud was exhausted through the orifice at the upstream end at a rate of 1175 ml/min. This flow rate was found to be optimal for gas chromatograph sensitivity over the range of CO concentrations encountered in these experiments, and was high enough to prevent the attainment of an appreciable concentration of CO in the shroud. Drawing air through the shroud at this velocity produced no measurable alteration in the dilution air flow into the cigarette.

Gas flow into the cigarette was controlled by a two stage tank regulator with fine control being provided by a needle valve. The flow was measured with a rotameter and adjusted to maintain atmospheric pressure at the entry to the artificial coal. Pressure was measured with a water filled U-tube manometer. This system insured that CO was not being forced through the cigarette. As a further check on the validity of this procedure, replacement of the gas feed system with a plastic bag filled with the standard gas mixture produced identical experimental results. Carbon monoxide was bubbled through a humidifying solution, whereas carbon dioxide was not humidified.

The artificial coal consisted of a segment of capillary tubing and the upstream CFP holder. The length of tubing was selected so that the pressure drop across both it and the CFP holder would be 5 cm of water at a flow rate of 1050 ml/min. This pressure drop corresponds to the pressure drop across the coal region during a puff of a popular commercial non-filter cigarette wrapped in standard citrate paper. It was determined in a separate set of experiments that the pressure drop across the coal region for a lit cigarette varies nearly linearly with mainstream flow. Therefore, the artificial coal should approximate the pressure drop response of the coal region of a lit cigarette.

Gas mixtures were drawn through the cigarette by a vacuum pump and needle valve and monitored with a rotameter. The flow rate was maintained at 1050 ml/min to simulate a two-second 35 ml puff. The effect of using a ventilated filter on the cigarette was mimicked by decreasing the mainstream air flow through the mouth end of the cigarette such that the difference between 1050 ml/min and the mainstream flow would represent the dilution flow into the filter vents.

Each gas flow stream was sampled by inserting the gas sampling valve on the gas chromatograph in series with the stream. Since the gas flow rate in each stream was known, the CO flow rate could be calculated from the measured concentration of the aliquot. Sampling points are indicated in Fig. 1. For each experiment mass balance of CO was achieved, thus insuring the absence of leaks in the system. In the following tables, the flow of CO through each segment of the apparatus is presented for various situations. The relative amount of CO diffusing from the cigarette, defined as

$$\% \text{ CO diffusing} = \frac{\text{CO diffusion flow through wrapper (ml/min)}}{\text{CO flow through artificial coal (ml/min)}} \times 100,$$

is also presented. This method of reporting the data, while not showing actual concentrations, does display the effect of diffusion on the delivery of CO.

RESULTS AND DISCUSSIONS

Diffusion of CO through Porous Papers

Experimental results of the diffusion of CO from cigarettes wrapped in naturally porous papers covering a wide range of permeabilities are presented in Table 1. Clearly, mass balance is achieved in each case. It can be seen that for a wrapper of high permeability, relatively more CO diffuses out of the cigarette (as defined) than for a wrapper of lower permeability. However, while the concentration of CO in the gas stream entering the cigarette remains constant, the absolute amount drawn from the coal is much less for very porous papers because of the greater flow of dilution air through the wrapper. Consequently, while the percentage of the CO diffusing increases slightly, the absolute amount of CO diffusing through the wrapper for very porous papers is greatly reduced.

Table 1. Diffusion of carbon monoxide from a cigarette as a function of wrapper permeability.

Wrapper permea-bility (cm/min)	Flow* through artificial coal (ml/min)	CO flows			% CO diffusing
		through artificial coal (ml/min)	de-livered (ml/min)	through wrapper (ml/min)	
20	810	37.4	27.5	9.9	26
52	570	25.7	19.1	6.9	27
120	340	14.9	10.1	4.7	32

* Delivery flow maintained at 1050 ml/min.

Diffusion of CO through Perforated Papers

Experimental results of the diffusion of CO through the wrappers of cigarettes wrapped in electrically perforated paper are presented in Table 2. Also listed are gas flows for cigarettes wrapped in papers of equivalent natural permeability. It is apparent from the data that relatively more CO diffuses from a cigarette wrapped in paper of natural permeability than for cigarettes wrapped in a paper of equivalent permeability achieved principally by perforation. This difference in relative diffusion is a consequence of the dissimilar pressure drop - flow characteristics of perforated and porous papers, and does not result from any change in the diffusional resistivity of paper.

Table 2. Diffusion of carbon monoxide from naturally porous and perforated wrappers.

Wrapper permea-bility (cm/min)	Flow through artificial coal (ml/min)	CO flows			% CO diffusing
		through artificial coal (ml/min)	de-livered (ml/min)	through wrapper (ml/min)	
20	810	37.4	27.5	9.9	26
120	330	14.9	10.1	4.7	32
120 (EP)*	240	11.3	8.1	2.7	24
160 (EP)*	208	9.5	7.4	2.1	22

* Base paper permeability is 30 cm/min.
EP = electrically perforated.

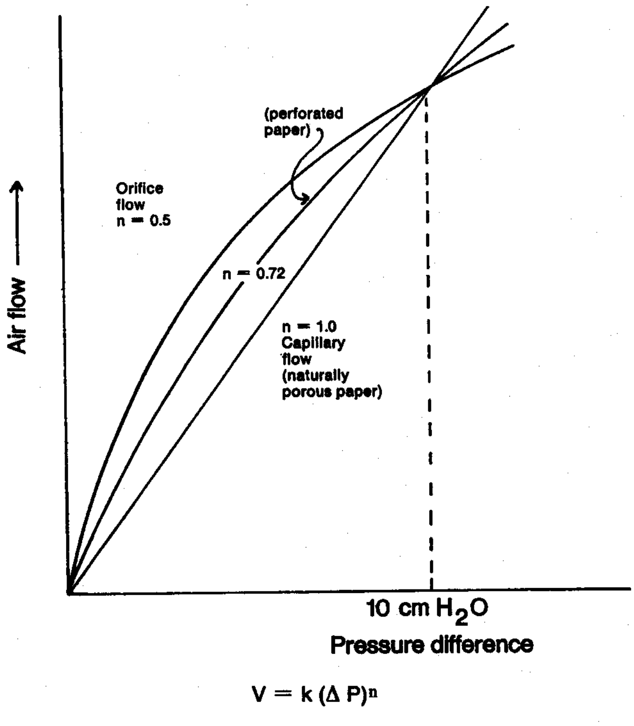
Fig. 2 shows the air flow characteristics of a perforated and a porous paper of equal *Coresta* permeabilities. Air flow through cigarette paper is described by the general equation:

$$V = k(\Delta P)^n,$$

where
- n = pressure drop exponent,
- k = permeability constant for specific paper,
- V = air velocity,
- ΔP = pressure difference across the paper.

Porous paper has a pressure drop exponent of unity, indicating that air flow through naturally porous paper is mainly through the capillaries in the paper. For perforated papers, n is equal to about 0.72 indicating a mixed flow regime of air flow through both capillaries and larger channels (perforations). Because the pressure drop across the wrapper of a cigarette is generally less than the 10 cm of water used in the *Coresta* test method, more dilution air will flow through the wrapper of the cigarette with the perforated paper than through the wrapper of a cigarette having naturally porous paper of equivalent *Coresta* permeability. In other words, there is a lower flow rate through the artificial coal for the cigarette wrapped in perforated paper.

Figure 2. Flow - pressure drop relationship for porous and perforated papers.



As shown by *Baker* and *Crellin* (7), perforating a given base paper to different levels of permeability does not significantly alter the diffusional resistance of the paper since the relatively large holes increase only slightly the diffusional pathways provided by the fine-scale capillarity of the paper, which remains unchanged. These perforations, being more than an order of magnitude larger than the natural capillaries, contribute radically to permeability but add only insignificantly to the total open area available. Furthermore, the velocity of the air entering these holes is high, thus reducing the contribution of that area to outward diffusion.

As a consequence of the increased dilution of the cigarette having the perforated wrapper, the pressure drop across the wrapper at any point along the rod will be less than at the corresponding point in the cigarette wrapped in naturally porous paper. Because of the decrease in the velocity of dilution air through the fine-scale pores of the paper (against which CO diffuses), the resistance to diffusion is reduced. However, despite this reduction in the resistance to diffusion, less CO diffuses out because the concentration of CO (i. e. the driving force for diffusion) at any point in the cigarette wrapped in perforated paper will be lower than in one wrapped in porous paper.

Diffusion through the Wrapper of Ventilated Filter Cigarettes

Experimental results for the effect of ventilation through the filter on the diffusion of CO through the rod are presented in Table 3. The simulation of filter tip ventilation was achieved as discussed in the section on Materials and Methods. Separate experiments, in which the shroud was placed around the perforated filters of several commercial cigarettes, showed that there was no measurable diffusion of CO through the filter vents.

In Table 3, filter tip dilution is expressed relative to a non-vented filter cigarette as

$$\% \text{ tip dilution} = \frac{(1050 \text{ ml/min}) - (\text{delivery flow})}{1050 \text{ ml/min}} \times 100 .$$

The data in Table 3 are interesting in two regards. First, the percentage of CO diffusing increases markedly upon increasing dilution air flow through the filter tip. Second, for equivalent degrees of dilution, relatively more CO diffuses through the wrapper of the cigarette having a vented filter tip than for a non-filter cigarette wrapped in permeable paper. This can be seen by comparing the relative diffusion values in Table 3 and by interpolation of the data in Tables 1 and 2 for cigarettes having the same flow through the artificial coal. For example, at 45 % tip dilution, the CO loss from diffusion for a vented filter cigarette is 42 % as contrasted with approximately 26 % for equivalent dilution achieved using a permeable wrapper.

As will be shown in the next section, this large increase in CO diffusion in a vented filter cigarette is a consequence of both the decreased average pressure drop across the rod wrapper and of a higher average concentration of CO in the rod.

Table 3. Effect of perforated filter on CO diffusion through a cigarette wrapper*.

Delivery flow (ml/min)	Flow through artificial coal (ml/min)	Filter tip dilution (%)	CO flows: through artificial coal (ml/min)	CO flows: delivery (ml/min)	CO flows: through wrapper (ml/min)	% CO diffusing
1050	810	0	37.4	27.5	9.9	26
600	450	43	19.1	10.9	8.2	42
390	240	63	10.8	6.0	5.2	56

* Wrapper permeability is 20 cm/min.

Factors Affecting the Diffusion of CO through the Wrapper of a Cigarette

Owing to the difficulty of rigorous quantitative application of diffusion equations to a system with a moving frame of reference (i.e. the dilution air entering the cigarette), there will be no attempt in this paper to utilize equations derived from first principles. Rather, diffusion of CO from a cigarette will be described in terms of parameters most closely associated with different modes of cigarette ventilation: pressure drop across the cigarette wrapper, pressure drop at the coal, and gas concentrations within the cigarette. However, because the cigarette behaves as a system, it is difficult to uncouple any one parameter and determine its effect on gaseous diffusion (e.g. changing the pressure drop across the coal will change the flow of dilution air into the cigarette, against which diffusion of CO takes place).

The type and amount of burn chemical applied to the cigarette paper will change the pressure drop across the coal region of a lit cigarette during a puff as well as the amount of CO generated at the coal region (8). However, in the diffusion experiments described in this paper, these two effects are easily uncoupled by using a fixed concentration of CO. The effect on coal pressure drop of some common burn additives is given in Table 4. It should be noted that the levels of burn additives used to generate the data in Table 4 represent extremes that would not be encountered in commercial cigarettes.

Table 4. Increase in Δ P across the coal region during a puff for various burn chemical additives.

Chemical treatment of cigarette wrapper	Average Δ P* across coal plus one cm of rod behind char line (cm H_2O)
none	3.0
1.5 % citrate	3.5
5.0 % citrate	5.0
2.5 % urea	3.0
3.0 % MAP**	6.0

* For 700 ml/min flow entering at coal; paper permeability is 20 cm/min.

** Mono-ammonium phosphate.

Table 6. The effect of vacuum level across paper and CO concentration in rod on the diffusion of carbon monoxide.

Wrapper permeability (cm/min)	ΔP_{AM}* across wrapper (cm H_2O)	Mole fraction CO			Molar diffusion flow (mmole/min)	(Molar diffusion flow)/ ($\bar{X}_{CO}$) (min^{-1})
		$X_{through\ coal}$	$X_{delivered}$	$\bar{X}_{CO}$		
120	4.8	0.045	0.010	0.023	0.21	9.1
120	4.8	0.036	0.0083	0.019	0.17	8.9
20	6.7	0.045	0.027	0.035	0.37	10.5
20	6.7	0.039	0.022	0.030	0.29	9.7
20	4.0	0.045	0.028	0.036	0.46	12.8
20	6.7	0.045	0.026	0.035	0.39	11.1
20	14.2	0.045	0.022	0.032	0.26	8.1

* arithmetic mean.

Levels of less than 1 % on the cigarette paper are more common.

The functional dependence of diffusion of CO on changes in coal pressure drop is presented in Table 5. Changes in coal pressure drops were simulated either by removing the artificial coal ($\Delta P_{coal} = 0$) or by changing the length of the capillary. A comparison of Tables 4 and 5 shows that, for both filter and non-filter cigarettes, the difference in vacuum level caused by change in the burn additives will cause only a minor change in the relative amount of CO diffusing from the cigarette. This change is less than the increase in CO generated in the coal region with increases in burn additive.

The diffusion of CO from a cigarette is directly dependent on the driving force for diffusion, i.e. concentration of CO within the rod during the puff. To describe the correct average concentration of CO over the length of the rod requires integration of the effects of dilution and loss by diffusion. As an approximation it is proposed that the logarithmic mean mole fraction be used. That is,

$$\bar{X} = \frac{X_{CO}\ \text{entering} - X_{CO}\ \text{leaving}}{\ln (X_{CO}\ \text{entering}/X_{CO}\ \text{leaving})},$$

where X represents the mole fraction of CO. A comparison of the relative diffusion of CO versus molar diffusion flow and log mean mole fraction is presented in Table 6. As expected, the molar diffusion of CO decreases as the permeability of the wrapper increases. The first four entries show that, for a given paper, the quantity (molar diffusion flow/$\bar{X}_{CO}$) is nearly independent of changes in concentration of CO fed into the artificial coal. Therefore, the molar diffusion flow of CO should be linearly related to the concentration of CO generated in the coal region.

The effect on CO diffusion of vacuum level, presented for simplicity as the arithmetic mean pressure across the wrapper, is also given in Table 6. An increase in pressure drop across the wrapper will increase the velocity of dilution air through the wrapper and, as a consequence, retard the molar CO diffusion flow through the wrapper. The dependence of diffusional resistance upon the velocity of dilution flow is shown by relating the quantity (molar diffusion flow/$\bar{X}_{CO}$) to the pressure differential across the paper. As expected, the data in Table 6 indicate that this quantity decreases as the pressure drop across the wrapper increases.

The above observations explain the dramatic increase in the diffusion of CO observed when the cigarette is ventilated through the filter. In the vented filter cigarette, the mainstream velocity through the cigarette is reduced. This decrease in pressure drop across the wrapper results in a decreased flow velocity of dilution air in through the wrapper. Similarly, less diluting air means that the average concentration of CO in the rod is greater. As discussed above, both these factors serve to increase the molar CO diffusion flow through the wrapper and, consequently, the relative amount of CO diffusing through the wrapper is increased.

Table 5. The effect of coal pressure drop on diffusion of carbon monoxide*.

Coal ΔP drop (cm H_2O)	Flow through artificial coal (ml/min)	Delivery flow (ml/min)	CO flows			% CO diffusing
			through artificial coal (ml/min)	delivered (ml/min)	through wrapper (ml/min)	
0	890	1050	40.1	29.0	10.4	26
3.2	800	1050	35.7	26.9	8.8	25
11.1	600	1050	27.0	22.2	5.8	21
0	460	700	20.7	13.2	7.9	38
6.4	105	700	4.7	3.1	1.7	36
0	240	390	10.8	5.2	6.0	56
3.6	190	390	8.6	3.8	4.8	56

* Wrapper permeability is 20 cm/min.

The Diffusion of CO_2 from Cigarettes Wrapped in Naturally Porous Papers

Presented in Table 7 are data describing the diffusion of CO_2 from cigarettes wrapped in naturally porous and perforated cigarette papers. It can be seen from a comparison of Tables 2 and 7 that the dependence of the relative diffusion of CO_2 on permeability of the paper closely follows that of CO. However, the magni-

Table 7. Diffusion of carbon dioxide from naturally porous and perforated papers.

Wrapper permeability (cm/min)	Flow through artificial coal (ml/min)	CO_2 flows			% CO_2 diffusing
		through artificial coal (ml/min)	delivery flow (ml/min)	through wrapper (ml/min)	
20	740	97.5	75.7	21.0	22
52	570	74.1	57.3	16.8	23
120	330	42.9	32.2	11.8	28
120 (EP)	220	28.6	21.9	7.0	24
160 (EP)	200	26.0	19.3	5.2	20

EP = electrically perforated.

tude of the relative diffusion is, as expected, less. A comparison of CO and CO_2 diffusivities with that expected from Graham's Law is in Table 8. As can be seen from these data, the ratios of CO/CO_2 diffusivities are close to that expected theoretically.

CONCLUSIONS

The information presented in this paper enables the following conclusions to be made.

During the first puff the diffusion of gases from a cigarette can be quite significant, accounting for 30% of the CO and 24% of the CO_2 generated in the coal region.

There is little difference in the amount of relative diffusion of CO from cigarettes wrapped in papers of widely differing degrees of natural porosity.

For a given level of dilution on the first puff, relatively more CO diffuses from a cigarette where ventilation is achieved through the filter than for a cigarette where ventilation is achieved by increasing the wrapper permeability.

Burn chemicals applied to the paper have a minor effect on the diffusion of CO from the cigarette.

SUMMARY

The diffusion of CO and CO_2 through the wrapper of cigarettes was studied by direct measurement under steady-state conditions, simulating the conditions which obtain during puffing.

Cigarettes made with porous paper, with perforated paper and with ventilated filters were studied. The diffusion of CO and CO_2 from cigarettes was found to be strongly influenced by the mode of cigarette ventilation. The greatest effect of diffusion on CO delivery was found for cigarettes where ventilation was achieved by ventilating the filter.

The influence on diffusion of the type and amount of burn chemical applied to the paper is described as well as changing the average pressure drop across the cigarette. It was found that normal levels of burn chemicals

Table 8. Comparison of diffusion of carbon monoxide and carbon dioxide as a function of wrapper permeability.

Wrapper permeability (cm/min)	% CO_2 diffusing	% CO diffusing	% CO diffusing / % CO_2 diffusing
20	22	26	1.2
52	23	27	1.2
120	28	32	1.1
120 (EP)	24	24	1.0
160 (EP)	20	22	1.0

$$\frac{D_{CO}}{D_{CO_2}} \left\{\frac{M.W._{CO}}{M.W._{CO_2}}\right\}^{-0.5} = \left\{\frac{44}{28}\right\}^{0.5} = 1.25$$

D: diffusion constant
M.W.: molecular weight
EP: electrically perforated

added to the paper will only slightly decrease the fraction of CO diffusing through the wrapper.

ZUSAMMENFASSUNG

Die Diffusion von CO und CO_2 durch den Papiermantel der Cigarette wurde durch direkte Messung bei stationärer Strömung untersucht, wobei die Bedingungen während des Rauchens simuliert wurden.

Es wurden Cigaretten mit porösem Papier, mit perforiertem Papier und mit ventilierten Filtern untersucht. Die Diffusion von CO und CO_2 aus der Cigarette heraus ist stark abhängig von der Art der Ventilation der Cigarette. Die CO-Ausbeute wurde am stärksten durch die Diffusion beeinflußt, wenn die Cigarette durch den Filter ventiliert wurde.

Es wird über die Beeinflussung der Diffusion sowohl durch Art und Menge der Zuschläge zum Papier zur Brandsteuerung als auch durch Änderung des durchschnittlichen Druckabfalls über die gesamte Cigarettenlänge berichtet. Es zeigte sich, daß die Mengen der normalerweise im Cigarettenpapier befindlichen Zuschläge den Anteil an CO, der durch das Cigarettenpapier nach außen diffundiert, nur wenig vermindert.

RÉSUMÉ

La diffusion du CO et CO_2 à travers le papier de la cigarette a été étudiée par mesure directe en régime stationnaire simulant les conditions dans lesquelles les bouffées sont tirées.

Des cigarettes faites avec du papier poreux, du papier perforé et avec des filtres ventilés ont été étudiées. La diffusion du CO et du CO_2 hors de la cigarette est fortement influencée par le mode de dilution de celle-ci. L'effet le plus prononcé de la diffusion sur le rendement en CO a été constaté sur les cigarettes dont la dilution était assurée par le filtre.

On décrit l'influence exercée sur la diffusion non seulement par le type et la quantité d'additifs du papier, mais aussi par la modification de la résistance moyenne

au tirage sur toute la longueur de la cigarette. Il a été trouvé que les quantités habituelles d'additifs du papier diminuent seulement de façon minime la fraction de CO diffusée à travers le papier.

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