Chemistry of Cigarette Burning Processes*

by

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SUMMARY

Cigarette-burning and the smoke-formation processes and smoke composition are important topics for understanding cigarette performance. This paper proposes the molecular formulas representing the active components of bright, burley, and Oriental tobaccos and a basic chemistry model of the cigarette burning processes. Previous knowledge of the cigarette burning processes and smoke formation helped to establish parameters in deriving the basic chemistry equations. The proposed chemistry provides a brief view of the mechanisms of the cigarette burning during puffing and interpuff smoldering, and can be used to interpret and predict the smoke composition for cigarettes made from bright, burley, and Oriental tobaccos. Based on the proposed chemistry, the effect of ventilation on smoke component deliveries is discussed and the reaction heat of the puffing process is estimated. [Beitr. Tabakforsch. Int. 21 (2004) 105–110]

ZUSAMMENFASSUNG

Informationen über die Prozesse beim Abbrennen von Zigaretten und der Rauchentstehung sowie die Zusammensetzung des Rauches sind wichtig für das Verständnis der Eigenschaften von Zigaretten. Diese Arbeit beschreibt die aktiven Komponenten des Bright, Burley und Orienttabaks anhand ihrer molekularen Formeln und schlägt ein chemisches Modell für das Abbrennen von Zigaretten vor. Auf der Grundlage bereits bekannter Informationen über die beim Abbrennen von Zigaretten und bei der Rauchentstehung stattfindenden Prozesse konnten Parameter für die grundlegenden chemischen Gleichungen abgeleitet werden. Die vorgeschlagenen chemischen Prozesse geben Einblick in die während eines Zuges und der Glühphase zwischen den Zügen stattfindenden Abbrennmechanismen der Zigarette

und können dazu herangezogen werden, die Zusammensetzung des Rauches von Zigaretten aus Bright, Burley und Orienttabaken zu interpretieren und vorherzusagen. Die Auswirkung der Filterventilation auf die Zusammensetzung des Rauches wird auf der Basis der vorgeschlagenen chemischen Vorgänge diskutiert und die beim Abrauchen entstehende Reaktionswärme wird geschätzt. [Beitr. Tabakforsch. Int. 21 (2004) 105–110]

RESUME

Les processus de la combustion des cigarettes et de la formation de la fumée ainsi que la composition de la fumée sont des paramètres importants pour comprendre la performance des cigarettes. Cette étude propose les formules moléculaires représentant les composants actifs du tabac Virginie, Burley et Oriental ainsi qu'un modèle chimique des processus de la combustion des cigarettes. La connaissance des processus de combustion des cigarettes et la formation de la fumée ont aidé à établir des paramètres à partir des réactions chimiques de base. La chimie proposée fournit un bref aperçu des mécanismes de combustion pendant les bouffées et entre les bouffées et peut servir à interpréter et prédire la composition de la fumée de cigarettes fabriquées avec du tabac Virginie, Burley et Oriental. Au moyen des processus proposés, l'effet de la ventilation sur la composition de la fumée est discuté et le dégagement de la chaleur au cours des bouffées est estimé. [Beitr. Tabakforsch. Int. 21 (2004) 105–110]

INTRODUCTION

Cigarette-smoke formation and smoke composition are major characteristics of cigarette performance. Fundamental research on the mechanisms of cigarette burning and smoke formation has been performed by a number of

Table 1. Percentage of carbon transferred to total smoke of the 1R1 cigarette (11) based on tobacco consumed (MS = mainstream; SS = sidestream; TPM = total particulate matter)

Component	MS gas	MS TPM	SS gas	SS TPM	Butt
Bright lamina	13.7	11.5	60.2	11.2	3.3
Burley	13.0	7.7	62.6	11.4	5.3
Oriental	12.1	12.3	58.5	12.9	4.4

Table 2. Carbon, nitrogen, and calculated ash content of typical cigarettes

Component	Carbon % (11)	N % (19)	Calculated ash %
Bright cigarette	40.8	1.6	10.2
Burley cigarette	37	3.2	17.0
Oriental cigarette	38	2.2	15.6

investigators over several decades (1–15). The research has been targeted to the following basic questions: What affects cigarette smoke formation and composition? How much air participates in the reaction? What is the contribution of each cigarette component in the combustion process? What is the mass balance when a cigarette is smoldering or being puffed?

BOYD *et al.* (3) studied the dependence of the gas-phase composition of smoke on the combustion temperature of tobacco products. JOHNSON *et al.* (6) studied the distributions of some components in tobacco smoke including ammonia, carbon monoxide, carbon dioxide, carbonyls, and hydrocarbons. They used the hydrocarbon information to predict combustion temperatures. JENKINS *et al.* (11) used ^{14}C to study the contribution of carbon to each smoke phase from ingredients in the 1R1 Kentucky reference cigarette. The smoke distribution of radioactivity from each of the labeled cigarettes was measured and multiplied by the actual amount of carbon consumed during burning to obtain the carbon distribution in smoke phases. YAMAMOTO *et al.* (15) studied the effect of chemical constituents on the formation rate of carbon monoxide in bright tobacco.

BAKER *et al.* (16), BROWN *et al.* (17), and NORMAN (18) studied ventilation effects on the smoke composition. These efforts significantly improved the understanding of cigarette burning processes and smoke formation. However, a more comprehensive understanding of the chemistry of cigarette burning processes and smoke formation is necessary to extend these research achievements for predicting the chemical composition of cigarette smoke.

This paper proposes the molecular formulation of the active component of tobacco materials and uses the mass balance involved in cigarette burning processes to propose a model chemistry of cigarette burning processes. Previous knowledge of cigarette burning and smoke formation helped to establish the parameters used in deriving the chemistry equations. The chemical reactions proposed can be used to interpret and predict the mainstream and sidestream smoke composition of a defined cigarette and offers a simple method to explore the effects of cigarette parameters such as tobacco components, ventilation, coal temperature, and other factors on cigarette-smoke composition.

CHEMISTRY OF CIGARETTE BURNING PROCESSES

When a cigarette is smoked, it undergoes puffing and inter-puff smoldering processes. As the cigarette is burning, a receding burning front is observed, which has a constant temperature (approx. 450 °C) and divides the burning cigarette into two zones, the burning zone and the pyrolysis zone (20). During smoldering, the main reaction – among distillation, pyrolysis and pyrosynthesis – in the burning zone is high-temperature (600–850 °C) char combustion. The oxygen participating in the reaction in the burning zone is coming through diffusion and natural convection. In the pyrolysis zone behind the paper char line, due to the heat transferred from the burning zone, the moisture in the tobacco evaporates and tobacco undergoes a series of pyrolysis reactions and is continuously converted into volatile and semivolatile smoke components and char. Hot gas flows out of the burning zone at the top of the coal near the paper char line with a temperature of about 350 °C. Due to the sudden drop of the temperature of the hot gas, semi-volatiles and some volatiles will condense to the sidestream smoke aerosol. As a result of relatively long contact times between incoming air and the burning coal, the chemical reactions occurring during smoldering may reach a steady state. The chemical composition of the sidestream smoke is affected by tobacco components, coal length, and coal temperature.

During puffing, the hot gases generated in the burning zone flow through the tobacco column to heat the tobacco and carry the pyrolysis and distillation products to form the mainstream smoke. Since the contact time of air in the burning coal is relatively short and hot gas flows through the tobacco column and filter, the chemical composition of the mainstream smoke will be affected not only by tobacco components, coal length, and coal temperature, but also the puff volume, ventilation level, and the filter design.

JENKINS *et al.* (11) used ^{14}C to study the contribution of carbon in each smoke phase from each gradient in the 1R1 Kentucky reference cigarette in 1980. The smoke distribution of radioactivity from each of these labeled cigarettes was measured in all of the smoke components, resulting in the carbon distribution of each cigarette in all the smoke phases as shown in Table 1.

Based on the analytical results of carbon and nitrogen contents in tobacco, it is proposed here that the molecular formula of the active component of tobacco material can be represented as $\text{C}_{30}\text{H}_{50}\text{O}_{23}\text{N}_n$. For burley, n equals 2, for bright and Oriental, n equals 1. The rest or the inactive component of tobacco material is ash. The tobacco ash contents as shown in Table 2 were calculated from the measured C and N contents and the formula above. For example, if the carbon content of a bright cigarette is 40.8%, the content of $\text{C}_{30}\text{H}_{50}\text{O}_{23}$ will be 88.2%, the nitrogen content is 1.6%, therefore, the ash content is 10.2%.

Experimental cigarettes were fabricated 8.4 cm long, 2.5 cm in circumference, and made of bright, burley, or Oriental tobaccos. The cut-widths of the tobacco fillers were 30 cuts per inch. The filters used in the cigarettes were 2.1-cm long cellulose acetate filters (8.0 denier per filament, 40,000 total denier) with filtration efficiencies of 27% for “tar” and 22% for nicotine. All cigarettes were smoked under Federal Trade Commission (FTC) conditions

Table 3. Selected mainstream data from the experimental cigarettes ^a

Component	Puffs	W_{bt} (mg)	CO (mg)	CO ₂ (mg)	"Tar" (mg)	Nicotine (mg)	Volatiles (mg)
Bright cigarette	9.5	711	14.7	53	35	3.4	6.1
Burley cigarette	6.1	565	12.7	48	24	1.9	5.5
Oriental cigarette	12.8	878	13.9	67	38	1.2	5.4

^a The total length burnt during smoking, L_{bt} , is 5.6 cm.

Table 4. Selected sidestream data from the experimental cigarettes

Component	LBR ^a	CO (mg)	CO ₂ (mg)	"Tar" (mg)	Nicotine (mg)	NH ₃ (mg)	Volatiles (mg)
Bright cigarette	0.460	57	534	37	8.3	7.0	13.6
Burley cigarette	0.656	41	370	24	5.1	9.8	11.5
Oriental cigarette	0.364	68	665	44	2.3	72	14.0

^a LBR = linear burn rate of cigarette during smolder (cm/min).

(with 35 cc puff volume, 2 s puff duration, and 1 puff per minute), the total length burnt during smoking, L_{bt} , is 5.6 cm. The smoking performances of these cigarettes are shown in Table 3 and the sidestream data of these cigarettes are shown in Table 4. The volatiles in the tables include hydrocarbons, NO, HCN, and the volatile organic compounds measured (the data shown in Tables 3 and 4 are the average of five replicates using the Fourier Transform Infrared (FTIR) method for vapor-phase analysis and the Gas Chromatography (GC) method for nicotine and "tar" analysis [22,23]). The total mass consumed W_{bt} , includes both burnt tobacco and paper.

If a cigarette is smoked under FTC conditions, the total mass consumed during the interpuff smoldering process, W_s (mg), may be estimated by

$$W_s = (pfn - 0.5) \cdot \frac{58}{60} \cdot \frac{W_{bt}}{L_{bt}} \cdot LBR \quad [1]$$

where pfn is puff count, L_{bt} and W_{bt} , are total length (cm) and mass burned (mg) during smoking, respectively. The number of puff intervals for a single cigarette is the integer part of the puff number. However, the puff count of a designed cigarette pfn is an average value of several tests, so the simplest estimate of the number of puff intervals is $(pfn - 0.5)$. Thus, the total mass consumed during puffs, W_p (mg), will be

$$W_p = W_{bt} - W_s = W_{bt} - (pfn - 0.5) \cdot \frac{58}{60} \cdot \frac{W_{bt}}{L_{bt}} \cdot LBR \quad [2]$$

By using the above information and the experimental data from burley, bright, or Oriental cigarettes, the chemistry of the cigarette burning processes is proposed. During a puff, the chemical reactions of the burning processes can be written as follows:

For burley cigarettes (Bu. cig.),

M.W.	806	176	84	264	306	196	132
Bu. cig.	1104						
W_p (mg)	213	33.9	16.2	50.9	58.9	37.8	25.3
Data (mg)			12.7	48		31.4	

where M.W. denotes the molar weight of reactants and products of the reaction. The pseudo-molar weights of the experimental cigarettes should include its ash content (17.4%) and moisture content (12%). The burned mass of the cigarettes calculated from Eqn. [2] can be used to calculate the oxygen amount consumed and the weight of the products in Eqn. [3]. The fourth term on the right hand side of the equations is the lump-sum of organic and inorganic products which includes "tar", nicotine, and volatiles in the vapor phase given in Table 3. It should be noted that the major components of the lump-sum products are "tar" and nicotine. "Tar" and nicotine count for 83–88% of the lump-sum products in mainstream smoke, and 73–77% in sidestream smoke. The char derived during puffing is not all consumed due to the short contact time between air and the coal.

For bright cigarettes (Bt. cig.),

M.W.	792	192	84	264	306	222	108
Bt. cig.	1003						
W_p (mg)	204	39	17.1	53.7	62.2	45.2	22.0
Data (mg)			14.7	53		44.5	

For Oriental cigarettes (Or. cig.),

M.W.	792	256	84	352	306	222	84
Or. cig.	1066						
W_p (mg)	200	48	15.7	66	57.4	41.7	15.7
Data (mg)			13.9	67		44.6	

In interpuff smoldering, the proposed chemical reactions of the burning processes can be expressed as follows. The unburned char derived during puffing will be consumed in interpuff smoldering, resulting in higher oxygen consumption than that in free smoldering.

For burley cigarettes, the number of carbon molecules to be burnt in the interpuff smoldering, $\alpha_1 = 11 \times W_p / W_s = 6.6$ and the number of oxygen molecules consumed $\beta_1 = 20.3 + 0.93 \cdot \alpha_1$, Eqn. 6.

M.W.	806	79	844	124	1151	306	125	23
Bu. cig.	1104							
W_s (mg)	352	25.3	267	39.3	367	97.6	40.0	7.2
Data (mg)				41	370		40.6	9.8

Equation 6. Chemistry in interpuff smoldering for burley cigarettes

M.W.	792	43	755	112	1038	306	122.6	11.3
Bt. cig.	1003							
W_s (mg)	507	22	382	56.6	525	155	62	5.7
Data (mg)				57	534		59.7	7

Equation 7. Chemistry in interpuff smoldering for bright cigarettes

M.W.	792	24.7	710	106	981	306	122.6	11.3
Or. cig.	1066							
W_s (mg)	678	15.7	452	67.4	624	195	78	7.2
Data (mg)				68	665		60.3	7.2

Equation 8. Chemistry in interpuff smoldering for Oriental cigarettes

Table 5. Puff count calculation ^a

Ventilation	0%	20%	40%	60%	80%
Cigarette A	7.4	7.7 (7.8)	8.0 (8.3)	8.5 (8.9)	9.1 (9.5)
Cigarette B	5.4	5.7 (5.8)	6.1 (6.2)	6.7 (6.8)	7.2 (7.4)
Cigarette C	7.9	8.7 (8.4)	8.9 (9.0)	9.8 (9.7)	10.4 (10.5)

^a A = L_{bt} : 5.6 cm, LBR: 0.57 cm/min; B = L_{bt} : 4.8 cm, LBR: 0.62 cm/min; C = L_{bt} : 5.6 cm, LBR: 0.51 cm/min. The numbers in brackets are calculated values.

For bright cigarettes, the number of carbon molecules to be burnt in the interpuff smoldering $\alpha_2 = 9 \times W_p/W_s = 3.6$, and the number of oxygen molecules consumed, $\beta_2 = 20.3 + 0.93 \cdot \alpha_2$, Eqn 7.

For Oriental cigarettes, the number of carbon molecules to be burnt in the interpuff smoldering $\alpha_3 = 7 \times W_p/W_s = 2.1$, and the number of oxygen molecules consumed $\beta_3 = 20.3 + 0.93 \cdot \alpha_3$, Eqn. 8.

For cigarettes with filter ventilation, the number of carbon molecules α should be determined by the ratio of W_p to W_s of the ventilated cigarettes to reflect the effect of ventilation. As W_p approaches zero, α becomes zero, Eqns. [6] to [8] become the chemistry of free smoldering processes. The proposed chemical reactions take into account the carbon distribution data in smoke phases by JENKINS *et al.* (11). For example, for burley cigarettes, the calculated carbon distributions in the gas phase and TPM of the mainstream smoke are 13% and 12%; and that of the sidestream

smoke are 63% and 12%, respectively. Including the original moisture in tobacco, the total delivery of water is about 42% of the tobacco weight. It can be seen from these equations that the composition of the tobacco types, including the nitrogen content, has an obvious impact on the cigarette burning process.

VENTILATION EFFECT

Based on the ventilation-effect studies by BAKER *et al.* (12,16), BROWN *et al.* (17), and NORMAN (18), the effect of ventilation level, v , on the puff count of a given cigarette (under FTC smoking conditions) may be empirically expressed as,

$$pfn = \frac{pfn|_{v=0}}{(1 - kv)} \quad k = 1 - \frac{pfn|_{v=0}}{\left(\frac{L_{bt}}{LBR} + 0.483\right)} \quad [9]$$

Once the puff number of the non-ventilated cigarette, $pfn = pfn|_{v=0}$, is known, the puff number of cigarettes with any ventilation level can be calculated. Table 5 illustrates the calculated puff count under FTC smoking conditions and the experimental values for Cigarettes A, B, and C. The total weight consumed during puffing and interpuff smoldering can be determined using Eqns. [1] and [2]. The corresponding smoke component delivery in mainstream smoke can then be determined by the proposed chemical reactions of the burning processes using Eqns. [3] to [5].

Figure 1. Ventilation effect on mainstream smoke component delivery

Figure 1 shows the effect of ventilation on the relative deliveries of smoke components in mainstream smokes, such as CO_2 , NH_3 , “tar”, and nicotine. As can be seen from Figure 1, the impact of tobacco type on the relative delivery of mainstream components is very little. However, the impact of tobacco type on the relative delivery of sidestream components is significant as shown in Figure 2. The impact of tobacco type on sidestream CO or CO_2 delivery is less than that on other components, as determined by the proposed chemistry for the interpuff smoldering processes, Eqns. [6] to [8].

It should be noted that Figure 1 and Figure 2 only consider the effects of dilution and puff count, but not any other effects. For example, the CO diffusion through the cigarette paper during puffs is not accounted for in the mainstream CO delivery and most of the water vapor in the mainstream smoke condensed onto the tobacco filler will eventually go to the sidestream smoke. It was observed that CO delivery was reduced more as compared with CO_2 delivery when the ventilation level increases (4,17,18). This is because, as the incoming air flow rate is low or the ventilation level is high, the contact time between the air and the coal becomes longer, and part of the sidestream smoke might be drawn to mix with mainstream smoke. It is also possible that the chemistry of the puff burning processes might vary with the ventilation level, since the coal temperature varies. Thus, it is necessary to investigate the ventilation effect on the chemistry of the puff burning processes in future studies.

HEAT OF CIGARETTE COMBUSTION

BOYD *et al.* (3) studied the dependence of the gas-phase composition of smoke on the combustion temperature of tobacco products. JOHNSON *et al.* (6) used the smoke component information obtained to predict the coal temperature. In a previous paper, a mathematical model of the

Figure 2. Ventilation effect on sidestream smoke component delivery

cigarette smoldering process was developed (20). This model predicts the free smoldering speed, or linear burn rate, and the temperature and density profiles in the pyrolysis zone of the smoldering cigarettes. The model also estimates the coal length and the maximum coal temperatures during free smoldering. The results show that air consumed in the smoldering process is about 2.6 times the mass of tobacco burnt. WAYMACK *et al.* (21) found the heat of smoldering combustion for tobacco, $\Delta H(\text{tobacco})$, is about 1.68 cal/mg for the 78 cigarette designs that were investigated.

For a free smoldering cigarette, if the burn weight of tobacco is 1000 mg, the air consumed will be 2600 mg, or 619 mg of oxygen (19.3 mmol). Therefore, the heat generation based on oxygen consumed, $\Delta H(\text{O}_2)$, will be:

$$\Delta H(\text{O}_2) = \Delta H(\text{tobacco}) \cdot 1000/19.3 = 87.1 \text{ cal/mmol oxygen.}$$

This value is similar to the literature value of 89.0 cal/mmol of oxygen consumed (9).

The char derived during puffing is not all consumed, so it is not suitable to estimate the reaction heat during puffing based on the heat of smoldering for tobacco. Therefore, it is better to estimate the reaction heat during puffing based on oxygen consumption, by assuming the same heat generation based on oxygen consumed as in free smoldering. The estimated reaction heat during puffing for these cigarettes is shown in Table 6. The values were calculated by Eqns. [3] to [5] using the fixed heat generation of 87.1 cal/mmol oxygen consumed.

CONCLUSIONS

The chemical reactions of cigarette burning processes during puffing and smoldering are proposed based on the mass balance in cigarette burning processes and earlier studies in the literature. The predicted deliveries of some

Table 6. Calculated reaction heat of cigarette during puffing
(based on 87.1 cal/mmol oxygen consumed)

Cigarette	Calculated ash %	Reaction heat during puffing
Burley	17.0	0.43 cal/mg
Bright	10.2	0.52 cal/mg
Oriental	15.6	0.65 cal/mg

smoke components agree fairly well with the experimental data for bright, burley, and Oriental experimental cigarettes.

The effect of ventilation is obvious on relative delivery of smoke components in both mainstream and sidestream smoke. The impact of tobacco components on the relative delivery of mainstream constituents is small, but it is significant on the relative delivery of sidestream constituents. Based on the heat generation in free smoldering, the reaction heats of the burning process during puffing are estimated to be 0.43, 0.52, and 0.65 cal/mg for burley, bright, and Oriental experimental cigarettes, respectively.

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