

SMOLDERING OF CIGARETTES

Fernando de Souza Costa

Laboratório Associado de Combustão e Propulsão
Instituto Nacional de Pesquisas Espaciais
Caixa Postal 01, Cachoeira Paulista, 12630-000, São Paulo, Brasil
fernando@cptec.inpe.br

Abstract. A simplified theoretical model describing the natural smoldering of cigarettes without drawing is developed. The steady propagation of infinitesimally thin fronts of drying, pyrolysis and char oxidation is calculated, considering circumferential heat losses and the convective-diffusive processes in the boundary layer. A smoldering transfer number is defined and the distribution profiles of density and temperature inside the cigarette are determined and compared to experimental data. Theoretical burning rates (~ 3 mm/min) and pyrolysis lengths (~ 10 mm) of cigarettes with no draw presented a good agreement with experiments.

Keywords. smoldering, cigarettes, pyrolysis, char oxidation, burning.

1. Introduction

Burning of cigarettes is a major cause of household and forest fires. According to Ohlemiller et al. (1995) most deaths in fires in the US are consequence of the ignition of soft furnishings by cigarettes. Therefore it is important to analyse the natural smoldering of cigarettes in order to predict the ignition propensity of materials and reduce fire hazards.

Several researchers studied the mechanism of smoldering in cigarettes. Smoldering is a flameless burning process occurring in charring materials.

Gugan et al. (1963) developed a theoretical one-dimensional model of smoldering cigarettes with puffing. Their model predicted an unstable shape of the smoldering front in time which is not verified experimentally. They measured temperature profiles using x-ray pyrometry, the distribution of hot gases by schlieren photography and consumption patterns for a variety of smoking conditions.

Gugan (1966) predicted the combustion zone shape in a simplified diffusion controlled model and determined burning rates during puffs 10 times larger than between puffs (no draw), and found a calorific value for tobacco/air stoichiometry of 15740 kJ/kg.

Summerfield et al. (1978) developed a steady-draw one-dimensional model assuming a one-step char oxidation reaction and one-step pyrolysis. They compared model predictions of the effects of flow rate and oxygen concentration on burning rate and pressure drop with experiments and found a good agreement.

Muramatsu et. (1979) developed a one-dimensional mathematical model of the evaporation-pyrolysis processes inside a naturally smoldering cigarette, considering Arrhenius kinetics for evaporation of water and pyrolysis of tobacco, volumetric char oxidation, internal heat transfer, using an effective thermal conductivity, and considering heat losses due to free convection and radiation. They identified a pyrolysis-evaporation zone range of 373-773 K and measured temperature profiles using thermocouples and density profiles using β radiation, obtaining a good agreement with the theoretical values.

Muramatsu (1981) determined several physical characteristics of tobacco during smoldering, such as specific heats, thermal diffusivities and heat released. The amounts of oxygen consumed and carbon oxide formed during smoldering were measured. A two-dimensional theoretical model was developed, yielding a good agreement to experimental results. The natural smolder rates measured were about 26 times slower than smolder rates with puffing, with 65 % of the carbon in the cigarettes converted to CO and CO₂, generating 6 to 10 moles of CO₂ for each mol of CO produced.

Gann et al. (1988) made a research project comprising several concurrent, interactive studies related to cigarette smoldering, ranging from computational, fundamental to empirical. They have made basic research into the physics of the ignition process to highly empirical studies of the manifest cigarette features affecting ignition. Several experiments were performed to analyse cigarette burning at bench and full scale. A two dimensional computer model (CIGARET 25) was developed to predict the temperature and density profiles inside a cigarette. An initial effort was made to develop a new test method to evaluate the ignition propensity of cigarettes.

Mitler and Walton (1993) developed a computer model (CIGARET) to calculate the time-dependent behavior of a cigarette smoldering quietly in the air, away from surfaces. The model incorporates diffusion and convection of gases, as well as the kinetics of char oxidation. It calculates the internal heat fluxes, as well as the internal distributions of temperatures, gas velocity and oxygen concentration. Another code (SUBTRACT) was also developed to determine if a two-layer solid, with an air gap in between, exposed to a moving heat source would ignite. The model successfully simulated the thermal runaway.

This work presents a new simplified analytical model of a smoldering cigarette without drawing assuming a steady one-dimensional propagation of infinitesimally thin fronts of drying, pyrolysis and char oxidation with circumferential heat losses, coupled to diffusive-convective processes in the boundary layer.

The burning rates, pyrolysis lengths and temperature profiles are calculated and compared to experimental values.

2. Model Description

Figure 1 shows a schematic of a smoldering cigarette. The main hypotheses assumed in the present analysis are: 1) 1D problem; 2) no drawing; 3) steady state; 4) infinitesimally thin fronts of drying, pyrolysis and char oxidation; 5) volatiles and tar exit through the smoldering end without reacting with the char; 6) ash layer thickness does not affect the flow; 7) char is pure carbon; and 8) paper burns at the same rate as the tobacco.

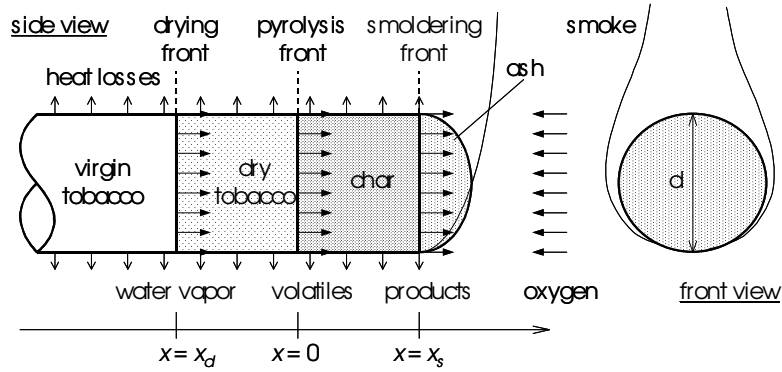


Figure 1 – One-dimensional smoldering of a cigarette.

2.1. Chemical Reactions

Pyrolysis of tobacco and other cellulosic materials produces a reactive char through a complex interplay of chemistry, heat and mass transfer. The char oxidation process involves chemisorption of oxygen in the reactive sites of the char, which is highly exothermic and takes place at relatively low temperatures, to form surface oxides. Smoldering combustion in general can be strongly enhanced or suppressed by inorganic materials which affect the number of active sites and, consequently, the rates of surface oxidation (Shafizadeh and Sekiguchi, 1984).

It is assumed in the analysis that pyrolysis occurs at a fixed temperature, $T = T_p$, following the single step reaction $\text{tobacco} \rightarrow \text{char} + \text{volatiles} + Q_p$, where Q_p is the heat of pyrolysis. This idealization corresponds to an infinite activation energy of pyrolysis kinetics. This reaction is considered slightly endothermic, with $Q_p \sim 300 \text{ kJ/kg}$ and $T_p \sim 600 \text{ K}$ (Kanury and Holve, 1982).

In the present analysis the char is assumed to be made of pure carbon. The primary surface reactions (Zhou, 1993) for char oxidation are:



A general chemical equation for the primary surface equations can be written as



where $Q_s = 218614(2 - 2a) + 393965(2a - 1) \text{ kJ / kmol}$ is the heat of smoldering, and $1/2 < a < 1$. The oxidizer/fuel mass ratio for reaction R3 is $\beta = 8a/3$ which varies from $4/3$ to 2 and indicates the proportion of CO and CO_2 being produced. The char oxidation rates for reactions R1 and R2 are given, respectively, by

$$\dot{m}_{c,1}'' = (12/32)k_1\rho_s Y_{\text{O}_{2,s}} \quad (\text{kg/s/m}^2) \quad (1)$$

$$\dot{m}_{c,2}'' = (12/16)k_2\rho_s Y_{O_{2,s}} \quad (\text{kg/s)/m}^2 \quad (2)$$

where $k_1 = A_1 \exp(-E_1/RT_s)$ and $k_2 = A_2 \exp(-E_2/RT_s)$ are reaction constants. A_1 , E_1 , A_2 and E_2 are kinetic constants, T_s is the surface or smoldering temperature (K), ρ_s is the surface gas density (kg/m^3) and R is the universal gas constant ($= 8314 \text{ J/Kmol/K}$). Xie and Liang (1997) adopted $A_1 = 7.6 \times 10^3 \text{ m/s}$, $A_2 = 1.9 \times 10^7 \text{ m/s}$, $E_1 = 146 \text{ MJ/kmol}$ and $E_2 = 198 \text{ MJ/kmol}$, following Adomeit et al. (1985). Zhou (1993) reports that Alexeyev and Pomerantsev found that the activation energy for char and coke oxidation E_1 can vary from 88 to 100 MJ/kmol and $E_2 = 1.1E_1$, with pre-exponential factors $A_1 = k^* \exp(E_1/R/T^*)$ and $A_2 = k^* \exp(E_2/R/T^*)$, where $k^* = 100 \text{ m/s}$ and $T^* = 2600 \text{ K}$.

Thus, the total char consumption rate is calculated from

$$\dot{m}_c'' = \dot{m}_{c,1}'' + \dot{m}_{c,2}'' = k\rho_s Y_{O_{2,s}} \quad (3)$$

where $k = 0.75(0.5k_1 + k_2)$.

2.2. Mass Transfer Equation for Oxygen

Using a reference frame on the smoldering surface, at $x = x_s$, the equation for mass transfer of oxygen at any point in the gas phase is

$$\dot{m}_{O_2}'' = \dot{m}'' Y_{O_2} - \rho_g D \frac{dY_{O_2}}{dx} \quad (4)$$

where $\dot{m}_{O_2}''$ is the mass flow rate of oxygen per unit area, $\dot{m}''$ is the mass flow rate of the bulk flow per unit area, Y_{O_2} is the oxygen mass fraction, ρ_g is gas mixture density and D is the oxygen diffusivity in the mixture. Since $\dot{m}'' = \dot{m}_f'' = \dot{m}_c'' + \dot{m}_v'' + \dot{m}_w''$, where $\dot{m}_f''$ is the rate of cigarette consumption per unit area, $\dot{m}_v''$ is the mass flow rate of volatiles from pyrolysis and $\dot{m}_w''$ is the mass flow rate of water vapor, and considering that $\dot{m}_{O_2}'' = -\beta \dot{m}_c''$, it can be shown that solution of Eq. (4) is given by

$$\dot{m}_{TB}'' = \frac{\rho_\infty D_\infty}{\delta} \ln(1 + B_s) \quad (5)$$

where ρ_∞ is the ambient density, D_∞ is the oxygen diffusivity in the ambient, δ is the diffusion layer thickness and B_s is the mass transfer number for smoldering combustion:

$$B_s = \frac{Y_{O_{2,\infty}} - Y_{O_{2,s}}}{\beta \rho_c / \rho_f + Y_{O_{2,s}}} \quad (6)$$

where $Y_{O_{2,\infty}}$ is the oxygen mass fraction in the ambient, $Y_{O_{2,s}}$ is the oxygen mass fraction at the smoldering surface, ρ_f is the cigarette density and ρ_c is the char density. The transfer number represents the ratio of a driving force for smoldering divided by a resistance to smoldering. It is useful for calculating the blowing factor in the thermal boundary layer.

The film thickness δ can not be determined without a detailed solution of the gas-phase fluid-mechanics problem, and therefore the mass consumption rate can not yet be calculated precisely using Eq. (5).

The heat transfer coefficient in the boundary layer, with correction for blowing, is calculated from

$$h = h_0 (1/B_s) \ln(1 + B_s), \quad (7)$$

where h , h_0 are the heat transfer coefficients ($\text{W/m}^2/\text{K}$) with and without blowing, respectively.

The viscous and thermal boundary layer thickness, δ_L , for natural convection along a vertical wall at constant temperature, in laminar flow, can be obtained from

$$\delta_L = 3.93zGr^{-1/4}Pr^{-1/2}(Pr + (20/21))^{1/4}, \quad (8)$$

valid for Grashof number $Gr < 5 \times 10^9$ – Pr is the Prandtl number. Assuming heat and mass diffusion boundary layers of equal thicknesses, and neglecting blowing effects, Eq. (8) can be used as a estimate for the diffusion layer thickness along the smoldering end, assuming laminar flow.

2.3. Energy Equations for the Cigarette

The initial temperature within the cigarette is T_0 . There is no reaction inside the char zone, but only on its boundaries. The energy equation is first solved for the char zone, $0 < x < x_s$, and then coupled to the energy equation for the preheating zone, $x < 0$, assuming no circumferential heat losses.

For a control volume inside the char zone the differential equation for energy conservation is

$$\left[\dot{m}_c'' c_c + \dot{m}_v'' c_{p,v} + \dot{m}_w'' c_{p,w} \right] \frac{dT}{dx} = \lambda_{c,eff} \frac{d^2 T}{dx^2} - \frac{4h'_c}{d} (T - T_\infty) \quad (9)$$

where $\lambda_{c,eff}$ is the char effective heat conductivity, h'_c is the average heat transfer coefficient around the char, d is the cigarette diameter and c_c , $c_{p,v}$ and $c_{p,w}$ are the specific heats of char, volatiles and water vapor, respectively. It is assumed that volatiles, water vapor and char are in thermal equilibrium and that the temperature is constant across the cigarette circular cross section. The effective heat conductivity includes a conduction term and a radiation term:

$$\lambda_{c,eff} = \lambda_{c,cond} + \lambda_{c,rad} \quad (10)$$

All participants, char, volatiles and water vapor, contribute to conduction:

$$\lambda_{c,cond} = (1 - \phi) \lambda_c + \phi \lambda_{v,w} \quad (11)$$

with ϕ denoting porosity, λ_c the char thermal conductivity and $\lambda_{v,w}$ an average thermal conductivity of volatiles and water vapor. For an optically dense medium, the linearized radiant transport can be approximated by the coefficient $\lambda_{c,rad} = 16\sigma d_{p,c} T_s^3 / 3$ where $d_{p,c}$ is the average pore diameter of char, $\sigma = 5.67 \times 10^{-8} \text{ W/m}^2/\text{K}^4$ is the Stefan-Boltzmann constant and T_s is the smoldering temperature which is not known yet. The boundary conditions for the char zone are

$$T(0) = T_p \quad (12)$$

$$\lambda_{c,eff} \left. \frac{dT}{dx} \right|_{x_s^-} = \dot{q}_s'' = \dot{m}_c'' Q_s - h(T_s - T_\infty) - \epsilon_c \sigma (T_s^4 - T_\infty^4) \quad (13)$$

where $\dot{q}_s''$ is the heat flux at the smoldering surface. The first term on the right side of Eq. (9) corresponds to heat generation by char oxidation; the second term corresponds to convective heat losses; and the third term corresponds to radiative heat losses, with ϵ_c being the char emissivity. The radiation term does not need to be linearized here.

Assuming that the drying front coincides with the pyrolysis front, i.e., $x_d = 0$, and assuming no flux of volatiles inside the pre-heating/vaporization zone, the energy conservation equation in the pre-heating/vaporization zone takes the form

$$\dot{m}_f'' c_f \frac{dT}{dx} = \lambda_{f,eff} \frac{d^2 T}{dx^2} - \frac{4h'_f}{d} (T - T_\infty) \quad (14)$$

where c_f is the specific heat, $\lambda_{f,eff}$ is the effective heat conductivity and h'_f is the average heat transfer coefficient around the preheating zone of the unburned cigarette. The boundary conditions are

$$T(0) = T_p \quad (15)$$

$$T(x \rightarrow -\infty) = T_0 \quad (16)$$

with the coupling condition

$$\lambda_{f,eff} \left. \frac{dT}{dx} \right|_{0^-} = \lambda_{c,eff} \left. \frac{dT}{dx} \right|_{0^+} + \dot{m}_v'' Q_p + \dot{m}_w'' Q_w, \quad (17)$$

where we define the variable $Q_w = Q_v - c_{p,w}(T_p - T_b) - c_{p,l}(T_b - T_0)$ which is the heat required to vaporize the moisture in the tobacco and increase its temperature to the pyrolysis temperature. Q_v is the heat of vaporization of water, taken as negative, T_b is the vaporization temperature of water and $c_{p,l}$ is the specific heat at constant pressure of liquid water.

2.4. Solution of the Problem

Using the energy equations for the char and pre-heating zones, with the boundary and coupling conditions described, and considering that $\dot{m}_v'' = U(\rho_d - \rho_c)$, $\dot{m}_w'' = U(\rho_f - \rho_d)$, $\dot{m}_c'' = U\rho_c$ and $\dot{m}_f'' = U\rho_f$, where ρ_d is the dry tobacco density, the following expressions for the temperature profiles are obtained:

$$T = T_\infty + c_1 e^{r_1 x} + c_2 e^{r_2 x}, \quad 0 < x < x_s, \quad (18)$$

$$T = T_\infty + c_1' e^{r_1' x}, \quad -\infty < x < 0, \quad (19)$$

where

$$x_s = \frac{1}{r_1} \ln \left[\frac{\dot{q}_s'' / \lambda_{c,eff} - r_2 (T_s - T_\infty)}{(c_2 - T_p + T_\infty)(r_2 - r_1)} \right], \quad (20)$$

$$c_1 = T_p - T_\infty - c_2, \quad (21)$$

$$c_2 = \frac{(T_p - T_\infty)(\lambda_{f,eff} r_1' - \lambda_{c,eff} r_1) - (\rho_d - \rho_c) U Q_p - (\rho_f - \rho_d) U Q_w}{\lambda_{c,eff} (r_2 - r_1)}, \quad (22)$$

$$r_{1,2} = \left(a \pm \sqrt{a^2 + 4b} \right) / 2, \quad (23)$$

$$a = (\rho_c c_c + (\rho_d - \rho_c) c_{p,v} + (\rho_f - \rho_d) c_{p,w}) U / \lambda_{c,eff}, \quad (24)$$

$$b = 4h_c' / d \lambda_{c,eff}, \quad (25)$$

$$c_1' = (\lambda_{c,eff} (c_1 r_1 + c_2 r_2) + U(\rho_d - \rho_c) Q_p + U(\rho_f - \rho_d) Q_w) / \lambda_{f,eff} r_1', \quad (26)$$

$$r_1' = \left(a' + \sqrt{a'^2 + 4b'} \right) / 2, \quad (27)$$

$$a' = \rho_f c_f U / \lambda_{f,eff}, \quad (28)$$

$$b' = 4h_f' / d \lambda_{f,eff}. \quad (29)$$

The smoldering rate U is given implicitly by the expression:

$$\frac{\dot{q}_s'' - r_1 \lambda_{c,eff} (T_s - T_\infty)}{[\dot{q}_s'' - r_2 \lambda_{c,eff} (T_s - T_\infty)]_{r_1}^{r_2}} = \frac{(T_p - T_\infty)(\lambda_{f,eff} r_1' - \lambda_{c,eff} r_1) - (\rho_d - \rho_c) U Q_p - (\rho_f - \rho_d) U Q_w}{[(T_p - T_\infty)(\lambda_{f,eff} r_1' - \lambda_{c,eff} r_2) - (\rho_d - \rho_c) U Q_p - (\rho_f - \rho_d) U Q_w]_{r_1}^{r_2}} \quad (30)$$

Equation (30) gives the burn rate U in terms of smoldering temperature, T_s , and oxygen mass fraction at char surface, $Y_{O_2,s}$. Two other relations are required to solve the problem, e.g., Eqs.(3) and (5).

3. Results and Discussion

Table 1 shows experimental data for several tobacco types (Muramatsu, 1979) obtained at $T_{amb} = 293$ K. The last column of Table 1 shows the cigarette densities when pyrolysis is supposed to be completed, at $T = 750$ K. According to Muramatsu data, above 750 K the volumetric char oxidation starts and reduces char densities to about 20% of the initial values at maximum temperatures of about 1100 K for all cigarettes.

Table 2 presents reference data from Muramatsu (1979), Kanury and Holve (1982) and Gann et al. (1988). Using Eqs.(3), (5) and (30) with data from tables 1 and 2, theoretical smolder rates were determined for the 5 cigarette types studied by Muramatsu. It was assumed in the simulations that $B_s = 0$, i.e., that the process was diffusion controlled.

Table 3 shows a comparison of theoretical results given by the present model and the experimental burning rates. As seen in Table 3 there is an excellent agreement for all cigarette types except for flue cured cigarette. This can be

possibly explained by the presence of inorganic substances which can increase or decrease reaction rates and, therefore, affect the heats of smoldering.

Table 1 – Experimental data for free burning cigarettes obtained by Muramatsu (1979).

Tobacco	Packing density (kg/m ³)	Water Content (%) dry basis	Density for $T = 750\text{K}$	Max. Temp. (K)
Flue cured	281	13.11	$0.52\rho_f$	1090
Burley	256	10.90	$0.55\rho_f$	1100
Matsukawa 1	210	11.34	$0.63\rho_f$	1090
Matsukawa 2	239	11.34	$0.63\rho_f$	1095
Matsukawa 3	293	11.34	$0.63\rho_f$	1100

Figure 2 depicts theoretical and experimental temperature profiles for 3 different cigarettes (FC = flue cured; BU = burley; M2 = Matsukawa 2). As can be seen in fig. 2, despite the differences among the curves, the char/pyrolysis lengths, which are defined for temperatures above 600K, have values around 10 mm giving a good agreement between theory and experiments.

The assumption of pyrolysis fronts coinciding with the vaporization fronts eliminates an inflection point in the theoretical curves. Nevertheless, the theoretical burning velocities present a good agreement with experiment since they are mainly dependent on smoldering temperatures and radiation characteristics. Densities and smoldering temperatures in the simulations were chosen according to the experimental data.

Table 2 – Properties and reference data used in the simulations.

Property	Value	Property	Value
Q_p (kJ/kg)	-300	T_b (K)	373
Q_s (kJ/kg)	13700	T_p (K)	600
Q_v (kJ/kg)	-2445	T_o (K)	293
c_f (kJ/kg/K)	2.50	T_∞ (K)	293
c_c (kJ/kg/K)	0.67	ϵ_c (-)	0.97
$c_{p,v}$ (kJ/kg/K)	1.10	ϵ_f (-)	0.97
$c_{p,w}$ (kJ/kg/K)	2.02	ϕ_c (-)	0.8
$c_{p,l}$ (kJ/kg/K)	4.17	ϕ_f (-)	0.8
λ_c (W/m/K)	0.041	$d_{p,c}$ (m)	0.005
λ_f (W/m/K)	0.147	$d_{p,f}$ (m)	0.0005
λ_v (W/m/K)	0.043	ρ_f (kg/m ³)	210-293
λ_w (W/m/K)	0.047	moisture (%)	10-14
$Y_{O_2,\infty}$ (-)	0.23	ρ_c (kg/m ³)	130-185

Table 3 – Comparison of theoretical and experimental data.

Tobacco	Experimental smolder rate (mm/min)	Theoretical smolder rate (mm/min)
Flue cured	2.65	3.56
Burley	3.80	3.73
Matsukawa 1	3.79	3.74
Matsukawa 2	3.23	3.34
Matsukawa 3	2.87	2.77

The present model can be easily extended to include a separate front of vaporization or distributed zones of vaporization, pyrolysis and char oxidation, which is under work. However the main interest here is to present a simplified method and relatively easy and fast method to estimate the smoldering rates of cigarettes with no draw.

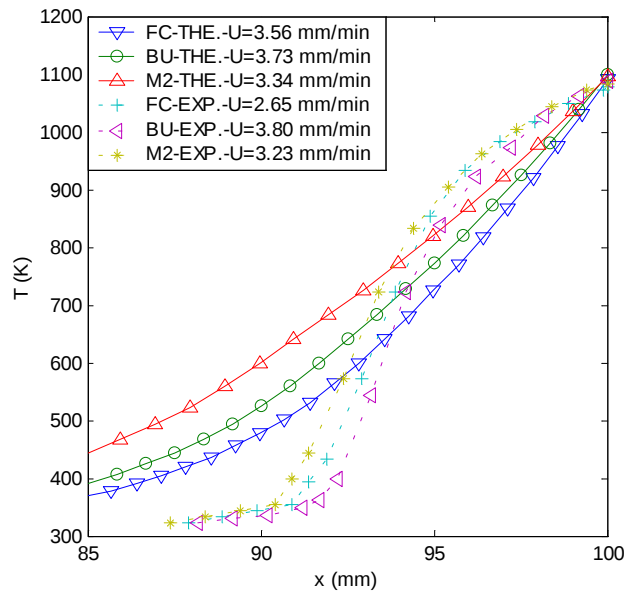


Figure 2 – Theoretical and experimental temperature profiles along cigarettes.

4. Conclusions

A simplified analytical method to estimate burning rates of cigarettes has been developed, based on the steady propagation of infinitesimally thin fronts of drying, pyrolysis and char oxidation with circumferential heat losses, coupled to the diffusion processes in the boundary layer. A smoldering transfer number was defined and the distribution profiles of density and temperature inside the cigarette were determined and compared to experimental data. Theoretical burning rates (~ 3 mm/min) and pyrolysis lengths (~ 10 mm) have a good agreement with the experimental data by Muramatsu (1979).

5. Acknowledgement

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