

Thermal Analysis of Tobacco on a View Point of Kinetic Approach

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Thermogravimetry (TG), derivative thermogravimetry (DTG) and differential scanning calorimetry (DSC) of tobacco and its some major components were carried out in helium atmosphere at a wide range of heating rates. One of the principal objects of the present study is to obtain apparent kinetic parameters useful in predicting the weight loss during the thermal decomposition process of tobacco.

The DTG curves showed that the complex thermal decomposition process of tobacco could be divided into four reaction zones. Assuming that the weight loss in each zone arises from the decomposition of a single solid component following n -th order Arrhenius kinetics, four sets of apparent kinetic parameters were determined from the TG-DTG curves by Kissinger's and Coats-Redfern's kinetic plots. The observed kinetic equations have been found to be very good empirical descriptions of the weight loss during the thermal decomposition process of tobacco.

In addition to kinetic analyses, the general characteristics of the thermal analytical curves of tobacco, as well as the contribution of tobacco components to them, have been discussed.

1. INTRODUCTION

Kinetic parameters, *i.e.* reaction order, pre-exponential factor and activation energy, for the thermal decomposition of tobacco should be essential factors influencing the weight loss or the density gradient in the evaporation-pyrolysis zone of a smoldering cigarette¹⁾ and consequently, the formation of smoke components.²⁾⁻⁴⁾

Kinetic approaches to the thermal decomposition process of tobacco have just started recently²⁾⁻⁵⁾ in the course of studies on the mechanism of smoldering and smoke formation of a cigarette. On the basis of a postulated mechanism of tobacco pyrolysis, Baker²⁾⁻⁴⁾ has derived kinetic parameters affecting the formation rates of carbon monoxide, carbon dioxide and some other low molecular weight gases produced when a tobacco

decomposes on heating. In another work, Tiller and Gentry⁵⁾ have determined the kinetic parameters for the bulk decomposition process of a reconstituted tobacco using a differential thermal analysis (DTA) technique.

However, the kinetic parameters reported in these papers²⁾⁻⁵⁾ do not necessarily serve to predict the overall weight loss of tobacco solid occurring during the thermal decomposition or the smoldering process of a cigarette. This is the first reason that the present study has been undertaken. In this study, thermogravimetry (TG), derivative thermogravimetry (DTG) and differential scanning calorimetry (DSC) of Bright, Burley and Matsukawa tobaccos and several major tobacco components were carried out in an inert atmosphere at a wide range of heating rates. Applying n -th order isothermal homogeneous kinetic equations, apparent kinetic parameters for the thermal decomposition of each tobacco were determined from the TG-DTG curves. The kinetic equations proposed have been found to be very good empirical descriptions of the weight loss during the

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thermal decomposition process of tobacco. In addition to the kinetic analyses, thermal decomposition patterns of tobacco and its related components have been characterized qualitatively based on the thermal analytical curves.

2. EXPERIMENTAL

2.1 Materials Used

Three types of tobacco leaves, *i.e.* Bright, Burley and Matsukawa tobaccos, as well as cellulose, hemicellulose, lignin and pectin isolated from Burley tobacco by conventional methods^{6),7)} were used. All samples were dried at 80°C for 2 hours and then ground to powder passing through a 80 mesh sieve. The powders were kept dry in a desiccator.

2.2 Apparatus and Procedures

Thermal analyses were performed using a simultaneous differential scanning calorimeter-thermobalance (Rigakudenki Co., 8085-D1) and a rapid heating thermobalance equipped with an infrared image furnace (Shinkū-Rikō Co., TG-3000). Both apparatus were provided with a DTG unit. The former was used for the thermal analysis at a slow heating rate below or equal to 20°C/min, and the latter was used at a relatively rapid heating rate above 20°C/min. The latter was employed by considering the heating rates (100–500°C/min) of the evaporation-pyrolysis zone in a smoldering cigarette.

Both TG-DTG curves obtained from the two different apparatus but at the same heating rate of 20°C/min agreed very well with each other. Consequently, a consideration of the dependence of TG-DTG curves on the apparatus is inferred to be unnecessary.

Since the atmosphere of pyrolysis zone inside a smoldering cigarette has been reported to be oxygen deficient,⁸⁾ all of measurements were carried out in an atmosphere of helium at a flow rate of 60 ml/min. Other conditions are as follows:

Reference material : calcined alumina

(α -Al₂O₃),

Weight of specimens: 10 ± 0.1 mg,

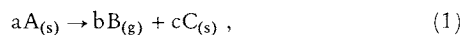
Heating rate : 2.5–20°C/min for the conventional TG-DTG-DSC, and 20–240°C/min for the rapid heating TG-DTG,

Temperature range : 70–580°C

The upper limit of heating rate was chosen taking into account the reliable responsiveness of the thermobalance used.

3. DESCRIPTION OF REACTION KINETICS OF PYROLYSIS

In general, pyrolytic process of a solid material can be expressed by the following simplified equation:



and the rate of disappearance of solid reactant A_(s) can be expressed by the following Arrhenius equation:

$$\frac{d\alpha}{dt} = Z \exp\left(-\frac{E}{RT}\right) f(\alpha), \quad (2)$$

where the symbols are defined in the Nomenclature section. The term $f(\alpha)$ is a function of weight loss, and its form depends on the mechanism of the reaction. However, it is often found^{9)–13)} that $f(\alpha)$ can be represented by:

$$f(\alpha) = (1 - \alpha)^n. \quad (3)$$

The following equation may be used to relate weight loss of A_(s) to fraction decomposed,

$$\alpha = \frac{W}{W_c}. \quad (4)$$

Various methods^{11)–17)} for kinetic analysis of thermal analytical curves have been developed by applying the isothermal homogeneous kinetic equation expressed by Eq. 2. Among them two typical methods simple to use are applied to the kinetic analysis of the TG-DTG curves of tobacco decomposition. They are the methods of Kissinger¹¹⁾ and Coats-Redfern.¹⁴⁾

Kissinger's method is based on the dependence of DTA or DSC peak temperature (T_p) on the linear heating rate. In this method, the following relation is utilized for first-order reaction kinetics:

$$\log\left(\frac{T_p^2}{\phi}\right) = \log\left(\frac{E}{RZ}\right) - \frac{E}{2.303RT_p}. \quad (5)$$

A plot of $\log(T_p^2/\phi)$ against $1/T_p$ should yield a straight line whose slope and intercept are $E/2.303R$ and $\log(E/RZ)$, respectively. Therefore,

E and Z can be calculated from the linear plot. Since T_p is the temperature at which reaction rate reaches the maximum, this method may also be applied to the kinetic analysis of TG-DTG curves.

The method of Coats-Redfern utilizes the following equation for n -th order kinetics; for $n=1$:

$$\log \left[\frac{1-(1-\alpha)^{1-n}}{T^2(1-n)} \right] = \log \left[\frac{RZ}{\phi E} \left(1 - \frac{2RT}{E} \right) \right] - \frac{E}{2.303RT}, \quad (6)$$

and for $n=1$

$$\log \left[-\frac{\ln(1-\alpha)}{T^2} \right] = \log \left[\frac{RZ}{\phi E} \left(1 - \frac{2RT}{E} \right) \right] - \frac{E}{2.303RT}. \quad (7)$$

A plot of either $\log \left[\frac{1-(1-\alpha)^{1-n}}{T^2(1-n)} \right]$ against $1/T$, where $n=1$, or $\log \left[-\frac{\ln(1-\alpha)}{T^2} \right]$ against $1/T$, where $n=1$, should result in a straight line with a slope $-E/2.303R$ for the correct value of n . The value of $\log [RZ(1-2RT/E)/\phi E]$ is practically constant for most values of E over the temperature range where reaction generally occurs. Therefore, the approximate value of Z can be calculated from the extrapolated intercept of the linear plot using the value of E obtained from the slope and the average temperature of the reaction zone.

4. RESULTS AND DISCUSSIONS

4.1 Thermal Analytical Curves of Tobacco

Figure 1 shows typical TG-DTG-DSC curves of Bright and Burley tobaccos heated at 20°C/min. General features of the thermal analytical curves of Matsukawa tobacco are almost similar to those of Burley tobacco and they are omitted in Fig. 1. Since tobacco consists of many components and its thermal decomposition involves a great number of pyrolytic reactions, the change in tobacco weight and endothermic and/or exothermic peaks observed on the thermal analytical curves should be considered as the summation of various pyrolytic reactions of respective components. Therefore, the resulting thermal analytical curves are rather complicated. However, it may be sure that the

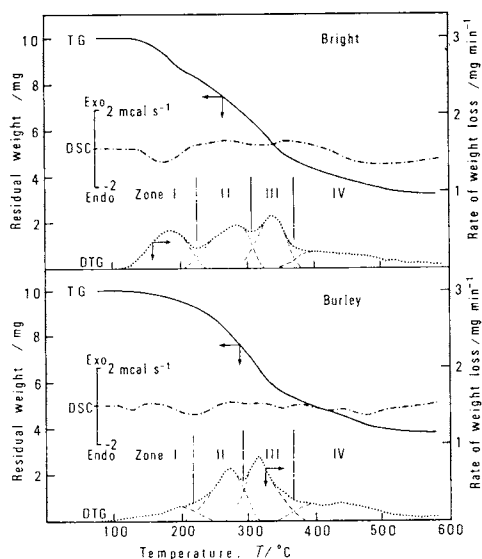


Fig. 1 TG-DTG-DSC curves of Bright and Burley tobaccos heated at 20°C/min in the atmosphere of helium.

shapes of the thermal analytical curves of tobacco will be governed to a considerable extent by the decomposition of certain major components in it.

Based on the characteristic peaks observed on the DTG curves, the thermal decomposition process of tobacco could be divided into four major reaction zones (I–IV) similarly to the results reported by others^{18),19)} as shown in Fig. 1. Prior to kinetic analyses, it will be relevant to review briefly the general characteristics of the thermal analytical curves divided into four zones.

Zone I: A discernible difference in the shapes of the TG-DTG curves is observed among tobacco types. Weight loss is significant for Bright tobacco, but insignificant for Burley and Matsukawa tobaccos. The weight loss in this zone has been mainly attributed to the vaporization^{18)–21)} of hydrated water and volatile materials and the decomposition^{5),18)} of sugars in tobacco. Some workers^{20),21)} have reported that most of such difference in the TG-DTG curves is accounted for by the content of volatile materials in tobacco. However, it seems reasonable to suppose that this will mainly result from the decomposition of sugars, since Bright tobacco normally contains much more sugars (15–30, wt%) than Burley and Matsukawa tobaccos.

Zone II and Zone III: In both zones a consider-

able weight loss is observed for all of tobaccos tested, however, differences in DTG peak temperature and its height are found among tobacco types. As will be mentioned later, most of the weight loss in Zone II appears to be attributable to the decomposition of pectin, hemicellulose, lignin, etc. and to the vaporization of high boiling materials, while that in Zone III seems to depend largely on the decomposition of relatively thermostable materials such as cellulose and lignin.

It is interesting to note that the thermal decomposition in Zone III is likely to be related to the combustibility of tobacco. Figure 2 shows the relationship between the rate of natural smoldering of cigarette and the DTG peak temperature observed in Zone III. Each plot in Fig. 2 represents different material in terms of tobacco type and stalk position. As can be seen from Fig. 2, the smoldering rate increases as the peak temperature becomes lower.

Zone IV: Weight loss proceeds at a slower rate compared with those in another zones, and any clear peak cannot be observed on the DTG curves. The weight loss is probably due to the decomposition of lignin and some other relatively thermostable materials and to the carbonization of the char remaining after thermal decomposition in the preceding zones.

As seen from Fig. 1, neither distinguishable endothermic nor exothermic peaks appear on DSC curves, except a small and broad endothermic peak observed in Zone I for Bright tobacco.

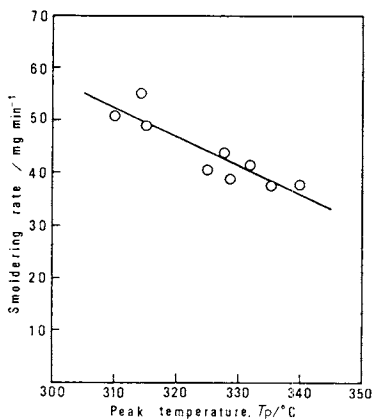


Fig. 2 Relation between DTG peak temperature in Zone III and naturally smoldering rate of cigarette.

General features of the thermal analytical curves mentioned above were unchanged over the range of heating rates examined. However, these curves were shifted to higher temperatures and the resolution of adjacent DTG peaks decreased with increase in heating rate, although it was not so remarkable as the peaks coalesced.

4.2 Thermal Analytical Curves of Some Tobacco Components

Hemicellulose, cellulose, pectin and lignin occupy 5–8%, 10–20%, 3–10% and 2–3% of tobacco leaf, respectively, by dry weight basis, and the sum of these cell-wall materials makes up a considerable portion of tobacco. Therefore, the thermal decompositions of these components should affect the shapes of the thermal analytical curves of tobacco. In this section, the influence of these components on the thermal analytical curves of tobacco has been studied.

Typical TG-DTG-DSC curves of hemicellulose, cellulose, pectin and lignin from Burley tobacco are illustrated in Figure 3. A prominent weight loss of hemicellulose (Fig. 3a) takes place in the temperature range of about 200–320°C with a maximum rate at 275°C, followed by a gradual weight loss leaving about 40 wt% of carbonized char at 580°C. The DSC curve exhibits a broad and small exotherm, but its peak temperature shifts approximately 20°C to higher side than that of DTG.

Thermal analytical curves of pectin (Fig. 3c) exhibit similar pattern to those of hemicellulose, however, the exothermic peak on DSC curve is appreciably sharp and its peak temperature coincides with that of DTG curve. Although the profiles of TG-DTG curves of hemicellulose and pectin determined in this study agree well with those reported by Ohnishi *et al.*,^{22),23)} the profiles of DSC curves are in conflict with their results. The DSC curve reported by Ohnishi *et al.* for purified tobacco-stalk xylan²²⁾ in a helium atmosphere exhibits neither endotherm nor exotherm, and that for purified tobacco-leaf pectin²³⁾ exhibits a small endotherm. This discrepancy may be ascribed in part to the effects of inorganic salts remaining in test substances as impurities (even though the impurities were not examined in this study). It has been well known^{9),24)–26)} that

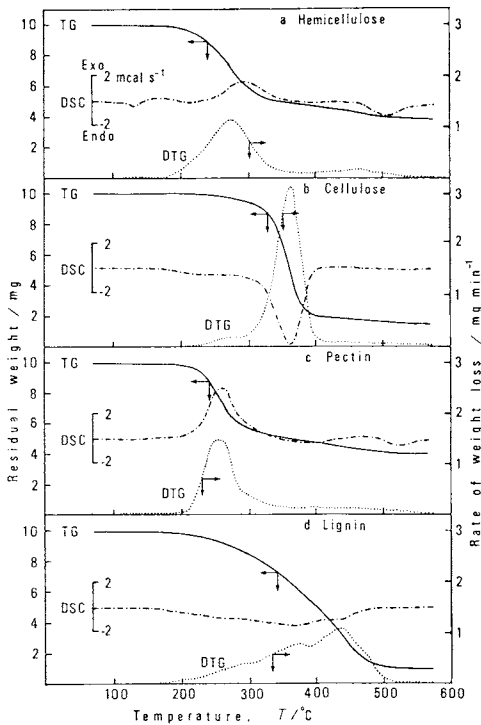


Fig. 3 TG-DTG-DSC curves of tobacco components heated at $20^\circ\text{C}/\text{min}$ in the atmosphere of helium.

the presence of inorganic impurities or additives influence pyrolytic processes of cell-wall materials.

The TG-DTG curves of cellulose (Fig. 3b) show that a weight loss starts at about 220°C . A rapid decrease in weight occurs in the temperature range of about $300\text{--}400^\circ\text{C}$, showing a peak at 360°C . Then this rapid decline is followed by a very slow decrease, finally leaving 15 wt% of carbonized char. The DSC curve of cellulose exhibits a deep endotherm corresponding to a weight loss. These results for cellulose agree with those reported by others.^{9),27)}

The TG-DTG curves of lignin (Fig. 3d) show that a weight loss occurs over a wide range of temperatures, $170\text{--}500^\circ\text{C}$, leaving 10 wt% of carbonized char at 580°C . The DTG and DSC curves are rather complicated, the former exhibits two broad peaks at 320°C and at 437°C , and the latter shows a broad and small endotherm.

Thus, it may be expected that the decompositions of hemicellulose and pectin, and cellulose in tobacco contribute to a weight loss of tobacco occurring in Zone II and Zone III, respectively,

and the decomposition of lignin contributes to those in Zones II, III and IV. However, any peak corresponding to endotherm or exotherm observed in each tobacco component cannot be clearly observed on the DSC curve of tobacco (Fig. 1). In the thermal decomposition process of tobacco, pyrolytic reaction of each component will be overlapping with concurrent pyrolytic reactions and/or vaporization of other components. Consequently, endotherm and exotherm must overlap and compensate each other.

4.3 Kinetic Analyses of TG-DTG Curves of Tobacco

As mentioned above, the thermal decomposition process of tobacco has apparently been divided into four zones (I–IV). Thereupon, following a Baker's report,⁴⁾ DTG curves were resolved by hand into four regions as illustrated in Fig. 1 by dashed lines, in order to estimate the weight loss within each zone or its ratio to the total weight loss up to 580°C . Although the total weight loss and the contribution of respective zone to that varied slightly with heating rate, the average over the heating rates examined was used in subsequent kinetic analyses. These values are listed in Table 1.

The following assumptions have been made to obtain apparent kinetic parameters, even though the observed weight loss in each zone arises from various pyrolytic reactions and vaporization of many components.

(1) Weight loss in each zone arises from the decomposition of a single solid component. Therefore, tobacco consists of four solid components different from one another.

(2) With a rise in temperature, each solid component in tobacco is continuously converted into volatile smoke and carbonized char following the scheme given by Eq. 1.

Strictly speaking, there remain some theoretical problems in the assumptions, however, such simplification is one of the practical approaches to determine the apparent kinetic parameters useful in predicting the rate of overall weight loss of tobacco during the complicated thermal changes.

In any event, it is almost impossible to determine the kinetic parameters which may admit no argument or can account for the reaction mechanism of tobacco decomposition clearly. Thereupon,

Table 1. Apparent kinetic parameters for the thermal decomposition of tobacco (see Nomenclature)

	Bright	Burley	Matsukawa
Zone I	*	**	***
n	1	1	
E (kcal mol ⁻¹)	20.2	20.8	
A (min ⁻¹)	3.76 × 10 ⁹	6.04 × 10 ⁹	
Weight loss (mg)	1.6	0.5	0.3
Zone II	**	**	**
n	1	1	1
E (kcal mol ⁻¹)	24.5	25.6	20.1
A (min ⁻¹)	1.03 × 10 ¹⁰	4.38 × 10 ¹⁰	2.45 × 10 ⁸
Weight loss (mg)	1.8	1.8	2.2
Zone III	*	*	*
n	1	1	1
E (kcal mol ⁻¹)	45.7	39.8	40.2
A (min ⁻¹)	3.59 × 10 ¹⁶	7.91 × 10 ¹⁴	5.19 × 10 ¹⁴
Weight loss (mg)	1.1	1.7	1.8
Zone IV	**	**	**
n	3	3	3
E (kcal mol ⁻¹)	25.2	27.9	29.1
A (min ⁻¹)	2.81 × 10 ⁸	1.58 × 10 ⁹	3.67 × 10 ⁹
Weight loss (mg)	1.9	1.7	1.8
Residual weight (mg)	3.6	4.3	3.9

* Measured by Kissinger's method. ** Measured by Coats-Redfern's method.
 *** Not determined because the weight loss is a very small amount.

assuming a first-order reaction relationship, we tried to determine the apparent kinetic parameters for the decomposition in each zone from the dependence of the DTG peak temperature on the linear heating rate (*i.e.*, Kissinger's method). However, since any clear DTG peak was not found in Zones I (except Bright tobacco) and IV over the heating rates tested, kinetic analyses for these zones were carried out separately by Coats-Redfern's method. In both analyses, actually measured average heating rates over the temperature range of 100° to 580°C were used instead of programmed ones.

Figure 4 shows Kissinger's kinetic plots for Zone I, II and III of Bright tobacco. The plots for Burley and Matsukawa tobaccos, though they are not shown in Fig. 4 because of overcrowding, are similar to those for Bright tobacco. The Kissinger's plot for each zone gives a straight line except Zone II. The weight loss in Zone II will be later analyzed again by Coats-Redfern's method. Kinetic parameters

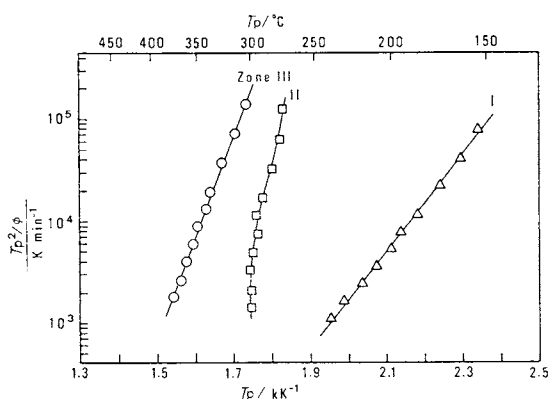


Fig. 4 Kissinger's kinetic plots for the first-order thermal decomposition in Zones I, II and III of Bright tobacco.

determined from the straight line by the least squares method are summarized in Table 1.

Activation energy, 20.2 kcal mol⁻¹ (1 cal = 4.1840 J), for Zone I of Bright tobacco seems to be suggestive of decomposition of sugars, since the

value is comparable to those (18–26 kcal mol⁻¹) for the thermal decomposition of glucose and fructose reported by Örsi.²⁸⁾ Kinetic parameters, 39.8–45.7 kcal mol⁻¹ and 7.91–3.60×10¹⁶ min⁻¹, for Zone III are the same order as those (44.7 kcal mol⁻¹, 2.34×10¹⁵ min⁻¹) for the first-order weight loss of a linter cellulose under isothermal conditions in the range from 273° to 318°C, reported by Hirata *et al.*²⁹⁾ Tiller and Gentry⁵⁾ have calculated the kinetic parameters for the thermal decomposition of a reconstituted tobacco from its DTA curves, and found the kinetic parameters corresponding to the decomposition of cellulose to be 43 kcal mol⁻¹ and 3.48×10¹⁵ min⁻¹. These values also agree well with those obtained in this paper.

Coats-Redfern's kinetic plots were made on the data ranging in the fraction of reactant decomposed, α , from 0.3 to 0.7 by changing the value of reaction order, n , from 0 to 4.0 in steps of 0.5. In Zone II, as well as in Zone I for Burley tobacco, reaction orders ranging from 1.0 to 2.0 give higher correlation coefficients than others over the heating rates examined, regardless of tobacco type. While, in Zone IV, the highest correlation coefficient is observed for a reaction order of 3.0, although reaction orders of 2.5 and 3.5 also give high correlation coefficients. Thus, the best fitting empirical reaction orders for Zones I and II and Zone IV are considered to be first- and third-order, respectively. Such the best fitting Coats-Redfern's plots for Zones II and IV of Bright tobacco are illustrated in Figure 5.

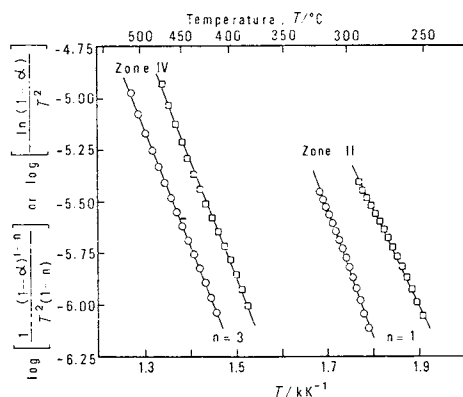


Fig. 5 Coats-Redfern's kinetic plots for the thermal decomposition in Zones II and IV of Bright tobacco.

Heating rate: —□—□— 20°C/min
—○—○— 240°C/min

Observed kinetic parameters varied with heating rate, especially those for Zone II. For example, the value of activation energy for Zone II of Bright tobacco changed from 21.7 kcal mol⁻¹ ($\phi=2.5^\circ\text{C min}^{-1}$) to 27.5 kcal mol⁻¹ ($\phi=240^\circ\text{C min}^{-1}$). This variation suggests³⁰⁾ that the nature of thermal decomposition process essentially is not so simple as to be governed by a single kinetic equation. In the present study, the average has been used as the experimental value for kinetic parameter. These are summarized in Table 1.

Theoretical TG curves derived numerically from the kinetic parameters shown in Table 1 were compared with experimental master TG curves over a wide variety of heating rates. In general, the theoretical curves approximate to experimental ones as illustrated in Figure 6. The agreement between theoretical and experimental curves demonstrated that the observed kinetic parameters are useful in predicting the overall weight loss of tobacco solid during the smoldering process of a cigarette.

For an example, Figure 7 compares theoretical and experimental profiles of density and temperature in the evaporation-pyrolysis zone inside a naturally smoldering cigarette. The experimental profiles were measured using thermocouples and a beta-ray attenuation technique. The theoretical profiles were derived from a one-dimensional mass- and heat-transfer model. The model includes decomposition of tobacco solid obeying n -th order kinetics specified in this study, as well as evaporation of water, heat transmission by conduction, radiation and free convection, etc.

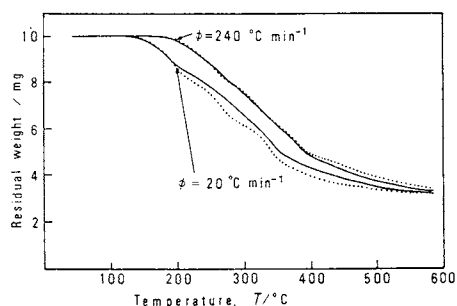


Fig. 6 Comparison of theoretical and experimental TG curves for Bright tobacco.

— Experimental,
..... Theoretical

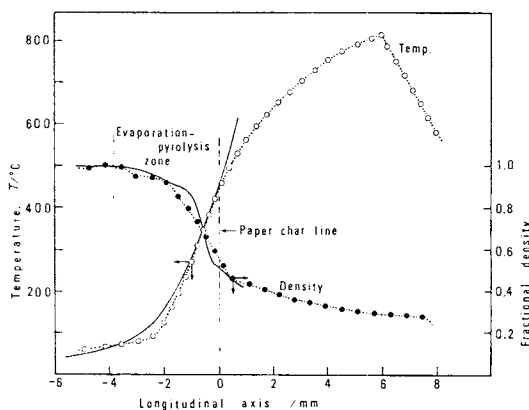


Fig. 7 Comparison of theoretical and experimental density and temperature profiles in the evaporation-pyrolysis zone of a naturally smoldering cigarette containing Burley tobacco.

○—○—○—○ Experimental,
●—●—●—● Theoretical

Further details of the model and the experimental procedures will be published in near future.¹⁾

As can be seen from Fig. 7, the theoretical profiles of density and temperature in the evaporation-pyrolysis zone agree closely with the experimental ones.

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NOMENCLATURE

$A_{(s)}$: reactant (solid phase, #)
 $B_{(g)}$: product (gas phase, #)
 $C_{(s)}$: product (solid phase, #)
 E : activation energy (cal mol⁻¹,
 1 cal=4.184 J)
 R : gas constant (1.98 cal mol⁻¹ K⁻¹)
 T : temperature (K or °C)
 T_p : peak temperature on thermal analytical
 curves (K or °C)
 W_c : weight loss at completion of reaction (mg)
 W : weight loss up to time, t , or temperature,
 T (mg)
 Z : pre-exponential factor (min⁻¹)

a, b, c : stoichiometric coefficients in Eq. 1 (#)
 t : time (min)
 α : fraction of $A_{(s)}$ decomposed at time, t ,
 defined by $\alpha = W/W_c$ (#)
 ϕ : linear heating rate (K min⁻¹ or °C min⁻¹)

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IUPAC 第30回総会報告

隔年ごとの上記総会は、今年は9月3日から10日までスイスのダボスにおいて開催された。筆者の参加した熱力学委員会での議論の中から、熱測定学会に関連の深いいくつかの事項についてとりいそぎ簡単に報告したい。

1. グリーンブック(「物理・化学量および単位」に関する記号と述語の手引)へのAppendixについて

従来から、Appendix IVとして、(1)化学熱力学における“標準状態”の定義・記号の明確化、(2)熱力学データ表における関数について、の2点が論議され、いろいろ問題があるため結論が出なかったが、今回も(1)標準状態圧力として従来の101 325 Pa(1 atm)を捨てて、あらたに1 bar (=0.1 MPa)を採用すべきだ、との強い提案があり、激論の末、影響が他分野にも及び大きいことを考慮してペンディングとなった。(2)熱力学関数として、いわゆる自由エネルギー関数の有用性には異論はなく、その符号も ϕ (phi) を用い

ることで一致したが、名称については前回の米国グループよりの提案、“Tempered Gibbs Energy”は反対が多く、あらためて“Giauque function”または“phi function”とすることでほぼ合意した。最終決定は次回に持ち越された。(これらについては、のちほど、本誌に詳しい報告をさせていただくつもりである。)

2. 次の第6回国際熱力学学会は、明1980年8月26-29日に東独メルゼブルグで開催されるが、世話人のRätzsch教授より、東独入国のVisa取得の関係から、参加希望者は全員、明年1月1日までに参加申込みと東独への入国申請を提出してほしい、との要請があった。従来の慣例よりはるかに早い時期なので注意していただきたい。

3. その次の第7回国際熱力学学会は、McGlashan教授の招請を受けて、1982年夏に、ロンドンで開催されることに内定した。

(東大工・高橋洋一)