

ISOTHERMAL CARBONIZATION OF *EUCALYPTUS CAMALDULENSIS* DENHN.

ปฏิกิริยาการทำถ่านไม้ยูคาลิปตัสที่อุณหภูมิคงที่

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ABSTRACT

The carbonization of Eucalyptus camaldulensis Denhn. was carried out by isothermally heating at selected temperature between 200°-800° C in a flow nitrogen atmosphere within 10 h. Four specimens (3 × 3 × 4 cm each, from 53 months old planted eucalypts) were carbonized in an electrical furnace at a given time and temperature. It was found that the average yield and some properties of charcoal depended on temperature. The charcoal yield was 21.5-91%. The heat of combustion was 4,600-7,480 cal/g. The fixed carbon, volatile matter and ash contents were 19.5-91.5%, 7-80% and 0.2-2%, respectively. Statistical analysis revealed linear relationships between carbonization temperature from 300° to 600° C versus heat of combustion, fixed carbon and volatile matter contents. The results of this study will be useful for the selection of a carbonization condition of the eucalypt for optimal charcoal yield, heat content, fixed carbon as well as volatile matter contents.

บทคัดย่อ

การศึกษาปฏิกิริยาการทำถ่านไม้ยูคาลิปตัสพันธุ์คามาลดูลเลนซิสโดยการอบด้วยความร้อนภายใต้บรรยากาศของไนโตรเจนที่อุณหภูมิคงที่ระหว่าง 200°-800° ซ. ภายในระยะเวลา 10 ชม. ไม้ตัวอย่างที่ใช้ศึกษาได้มาจากไม้ที่ปลูกขึ้น อายุ 53 เดือน มาแปรรูปให้มีขนาด 3 × 3 × 4 ซม. ในการทำปฏิกิริยาแต่ละครั้งใช้ไม้ตัวอย่างจำนวน 4 ชิ้นอบในเตาเผาไฟฟ้าตามอุณหภูมิและเวลาที่กำหนด ผลการทดลองพบว่าผลผลิตและคุณสมบัติของถ่านแปรผันตามอุณหภูมิที่ใช้ทำปฏิกิริยา ผลผลิตถ่านมีปริมาณ 21.5-91%

ความร้อนของการสันดาป 4,600-7,480 แคลอรี/ก. ปริมาณคาร์บอนเสถียร 19.5-91.5% ปริมาณสารระเหย 7-80% และปริมาณขี้เถ้า 0.2-2% การวิเคราะห์ความสัมพันธ์ทางสถิติของคุณสมบัติของถ่านเหล่านี้ ที่อุณหภูมิ 300°-600° ซ. ทราบว่ามีความสัมพันธ์เป็นแบบเส้นตรง ผลการศึกษาในครั้งนี้น่าจะใช้เป็นแนวทางในการเลือกอุณหภูมิในการผลิตถ่าน เพื่อให้ได้ผลผลิตถ่าน ค่าความร้อน ปริมาณคาร์บอนเสถียร และปริมาณสารระเหยที่เหมาะสม

INTRODUCTION

Energy from wood is still essential for rural Thai people. The consumption of cooking charcoal in Thailand was over three million tons in 1983 which the major demand was found in the rural area⁵ while the natural forest is unable to supply wood to produce large quantity of charcoal. The objective of this research is to find out an optimum condition to produce cooking charcoal from *Eucalyptus camaldulensis*.

There are several publications reported about residual mass and charcoal properties from pyrolysis of gymnosperm and angiosperm specimens under a nitrogen atmosphere. They confirmed that both residual mass and charcoal properties depended on rate of increasing temperature and final pyrolyzed temperature.

Beall² pyrolyzed redwood specimens at heating rates of 1° to 50° C/min to end temperature of 250°-600° C. The mass loss of a slow heating rate was greater at the end temperature lower than 400° C. However, it was found almost no effect of heating rate to mass loss at the end temperature of 600° C. Slocum et al.⁴ pyrolyzed oak and hickory specimens at heating rates of 1.5° to 130° C/h to the maximum temperature between 250° - 800° C. The heating rate varied with maximum temperature. The mass loss depended on maximum temperature. They also reported that the residence time at temperature between 400° - 800° C slightly affected to mass loss of both species. Charcoal yield would be reduced less than 10% when a residence time at 800° C was extended to 114 h.

The effect of carbonization temperature between 200° - 1,000° C to charcoal yield and elementary composition of charcoal was summarized by Wenzl.⁶ The charcoal yield and carbon content were approximate 33% and 90%, respectively. Acesita³ carbonized *Eucalyptus grandis* specimens at 300°, 500° and 700° C. The fixed carbon contents were 68, 86 and 92%, respectively. Baileys and Blankenhorn¹ pyrolyzed wood specimens of red oak, southern yellow pine, black cherry and hybrid poplar under a nitrogen atmosphere at average heating rate of 3° C/min to selected final temperature up to 700° C. They confirmed that the residual mass and heat of combustion depended on final temperature. The linear equations between pyrolyzed temperature versus heat of combustion were reported with respect to wood species. The coefficient of determination of each equation was high.

MATERIALS AND METHODS

Preparation of specimens and a furnace

About 250 specimens, $3 \times 3 \times 4$ cm each, were prepared from 8 trees of 53 months old *Eucalyptus camaldulensis*. The eucalypts were planted at Rangsit, Changwat Pathum Thani with a spacing of 2.4×1.8 m. All specimens were oven-dried in an electrical oven at $105 \pm 3^\circ$ C to obtain oven-dry weight of each specimen before carbonization.

An electrical furnace for carbonization of the eucalypt specimens has a chamber space of $8 \times 11 \times 26$ cm and it can be heated up to $1,000^\circ$ C. An additional temperature controller by a thermocouple with precision of $100 \pm 3\%$ was connected to the furnace. Temperature profile of the furnace was recorded prior to carbonize the specimens. The temperature profile was shown in Figure 1.

Carbonization

At a given time and temperature of carbonization, 4 specimens were heated isothermally under a nitrogen atmosphere after the temperature profile had been recorded. All specimens were placed among 15 cm from the rear of the furnace. The given time and temperature of carbonization were up to 10 h and 800° C respectively as follows.

Temperature, $^\circ$ C	Time, h
200	1, 2, 4, 7, 10
250	$\frac{1}{2}$, 1, 2, 4, 7
300	$\frac{1}{4}$, $\frac{1}{2}$, $\frac{3}{4}$, 1, 2, 4, 6
350	$\frac{1}{6}$, $\frac{1}{4}$, $\frac{1}{3}$, $\frac{1}{2}$, 1, 6
400	$\frac{1}{12}$, $\frac{1}{6}$, $\frac{1}{4}$, $\frac{1}{2}$, 1, 6
500	$\frac{1}{12}$, $\frac{1}{6}$, $\frac{1}{4}$, $\frac{1}{2}$, 1, 6
600	$\frac{1}{6}$, $\frac{1}{4}$, $\frac{1}{2}$, 1, 6
800	$\frac{1}{2}$, 1, 3, 6

At the end of each carbonization all remaining specimens were taken from the furnace and cooled in aluminium desiccators under nitrogen atmosphere for 1 h. The specimens were then weighed. The charcoal yield was calculated as follow.

$$\text{Charcoal yield} = \frac{\text{residual weight}}{\text{ovendry weight}} \times 100$$

Analyses of charcoal

About $\frac{1}{3}$ of each remaining specimen from a given time and temperature was ground into powder and then mixed together for the following analyses.

Heat of combustion

The heat of combustion of charcoal was determined by using an adiabatic oxygen bomb calorimeter, Parr no. 1341 and bomb no. 1108, according to the method of ASTM D 2015-72. The heat of combustion was reported based on ovendry weight of charcoal.

Proximate analyses

The proximate analyses of charcoal were determined by using an electrical furnace, Hoskins type FD 203° C, according to the ASTM D 1726-64 procedure. The fixed carbon, volatile matter as well as ash contents were reported based on ovendry weight of charcoal. The fixed carbon content was the amount of carbon that is not volatilized at a temperature of 950° C for 6 min, minus the amount of ash remaining after the charcoal was completely combusted in the furnace at a temperature of 750° C for 6 h. The fixed carbon content was calculated from the following relation:

$$\% \text{ of fixed carbon} = 100 - \% \text{ of volatile matter} - \% \text{ of ash.}$$

RESULTS AND DISCUSSION

Charcoal yield

Residual mass of carbonized specimens was revealed in Figure 2. The remaining specimens, at temperature less than 250° C, were not completely carbonized even though the reaction time was as long as 7 h. The residual mass was still decreasing. Between 300° - 350° C, observed specimens were completely carbonized after 30 min. The residual mass was almost constant during 30 min to 6 h of carbonization. The observed specimens could be completely carbonized within 15 min when carbonization was taken at temperature between 400° - 600° C.

The residual mass from the carbonized specimen was almost constant after 30 min of carbonization. At 800° C, the carbonization was very fast. The remaining specimens could not be removed from the furnace within 30 min of carbonization because they simultaneously combusted due to high temperature and volatile matter content. Unlike lower temperature, the residual mass was decreased with respect to time of carbonization.

The average charcoal yield and charcoal properties of each carbonization temperature were reported in Table 1. The average yield was determined by extrapolation of respective constant value of a given temperature as shown in Figure 3, the determination of charcoal yield at 500° C for example. The relationship between average charcoal yield versus carbonization temperature was probably a curve as shown in Figure 4. The average charcoal yield at temperature between 300-350° C was 39-47%. The carbonization of remaining specimens at these temperatures had begun to be completed. The average charcoal yield was reduced to 24.5-34.5% when carbonization had been taken at temperature between 400° - 600° C. At 800° C, the temperature was too high for carbonization. The charcoal yield was as less as 21.5%.

Fixed carbon contents

Fixed carbon contents from all specimens with respect to time and temperature of carbonization are shown in Figure 5. At temperature less than 250° C, the contents were much less than usual cooking charcoal. The contents increased slightly with respect to time of carbonization. At temperature higher than 300° C, the fixed carbon contents were almost constant after carbonization longer than 30 min. The effect of time for carbonization to fixed carbon contents, after specimens has been completely carbonized, was only slightly.

The average fixed carbon contents of remaining carbonized specimens were reported in Table 1. The relationship between carbonization temperature (300° -600° C) versus respective fixed carbon contents was linear as shown in Figure 6. The linear equation for this relation as well as other properties and its quality are revealed in Table 2.

The fixed carbon content for usual cooking charcoal should be greater than 65%. The result from this experiment indicated that the carbonization temperature for *Eucalyptus camaldulensis* must be higher than 400° C in order to obtain cooking charcoal with qualified fixed carbon content.

Volatile matter content

Volatile matter contents from all specimens at time and temperature of carbonization are revealed in Figure 7. The contents decreased with respect to time and temperature of carbonization except temperature higher than 400° C. The volatile matter content at a given temperature decreased slightly after carbonization had been completed.

The average volatile matter contents were reported in Table i. Figure 8 and Table 2 revealed the linear relationship between 300°-600° C of carbonization temperature versus volatile matter contents.

The normal cooking charcoal should compose of volatile matter content less than 30% in order to provide smokeless charcoal. The result from this experiment indicated that the carbonization temperature of *Eucalyptus camaldulensis* must be greater than 400° C so that the charcoal contained qualified volatile matter content for cooking.

Ash content

All remaining specimens from this carbonization composed of ash content less than 2.5%. It was difficult to conclude that the content depended on time and temperature. However, the ash content of most remaining specimens was 1-2%. The usual cooking charcoal allowed to compose of ash less than 5%.

Heat of combustion

Heat of combustion from all carbonized specimens with respect to time and temperature was shown in Figure 9. The heat content depended on time and temperature of carbonization similar to charcoal yield, fixed carbon as well as volatile matter contents. It appeared that the heat content of remaining specimens increased slightly after 1 h of reaction time at temperature higher than 300° C.

The average heat of combustion with respect to carbonization temperature was shown in Table 1. Results from regression analysis according to Figure 10 and Table 2 revealed that the relationship between carbonization temperature (300° - 600° C) versus heat content was linear. The heat of combustion of usual cooking charcoal should not be less than 6,600 cal/g. Therefore, carbonization temperature of the eucalypt should be higher than 400° C.

Relationships between heat of combustion versus fixed carbon and volatile matter contents

Relationship between some charcoal qualities of the same specimens was not commonly reported. From this experimental result, it was indicated that the linear relationship between heat of combustion versus fixed carbon content as well as volatile matter content were confirmed from carbonized specimens at temperature between 300° - 600° C as shown in Figure 11. The correlation coefficient of each line was 1.00.

SUMMARY

The carbonization of *Eucalyptus camaldulensis* was made at temperature between 200° - 800° C within 10 h of reaction time. Two hundred and fifty specimens, 3 × 3 × 4 cm each, were prepared from 8 trees of planted eucalypt at 53 months old. A carbonization was taken from isothermally heating of 4 specimens in an electrical furnace under a nitrogen atmosphere at a given time and temperature. Residual mass as well as some charcoal properties, i.e. heat of combustion, fixed carbon and volatile matter contents, were determined from the remaining specimens. These charcoal properties depended on carbonization temperature. The charcoal yield was 21.5-91%. The heat of combustion was 4,600-7,480 cal/g. The fixed carbon, volatile matter as well as ash contents were 19.5-91.5%, 7-80% and 0.2-2%, respectively. The details of charcoal yield and properties from each carbonization temperature were summarized as follows.

At temperature lower than 250° C, all specimens were not completely carbonized although the carbonization had been taken as long as 7 h. The charcoal yield was more than 67%. The heat of combustion and fixed carbon content of the products were less than 5,500 cal/g and 35%, respectively. The products composed of more content of volatile matter.

At temperature between 300° - 350° C all specimens were completely carbonized within 30 min. The average charcoal yield, heat of combustion, fixed carbon and volatile matter contents were almost constant after 30 min of reaction time. The charcoal yield was 39-47%. The heat of combustion, fixed carbon and volatile matter contents were 6,200-6,420 cal/g, 57-62% and 37.5-42%, respectively. These charcoal properties were little poorer than usual cooking charcoal.

Between 400° - 600° C, all specimens were completely carbonized within 15 min. The average charcoal yield, heat of combustion, fixed carbon and volatile matter contents were 6,620-7,480 cal/g, 68-87% and 11-31%, respectively. These physical properties were similar to usual cooking charcoal.

At 800° C, the temperature was too high to produce charcoal. The charcoal yield decreased with respect to reaction time. The heat of combustion, fixed carbon as well as volatile matter contents were not much better than those charcoal properties at 600° C.

Statistical analysis revealed linear relationships between carbonization temperature between 300° - 600° C versus heat of combustion, fixed carbon as well as volatile matter contents. The linear equations for these relationships are as follow:

heat of combustion vs temperature,

$$Y = 4,885 + 4.402 X :$$

fixed carbon content vs temperature,

$$Y = 26.63 + 0.1025 X :$$

volatile matter content vs temperature,

$$Y = 73.99 - 0.1067 X :$$

heat of combustion vs fixed carbon content,

$$Y = 3,743 + 42.93 X :$$

heat of combustion vs volatile matter content,

$$Y = 3,937 - 41.23 X :$$

The correlation coefficient of each equation is 1.00.

The results from this experiment suggested that the optimal carbonization temperature for *Eucalyptus camaldulensis* at 53 months old should be 400° - 500° C for producing of cooking charcoal with qualified yield and properties.

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Table 1. Average charcoal yield and its properties from carbonization of *Eucalyptus camaldulensis*

Carbonization temperature (° C)	Charcoal yield (%)	Volatile matter content (%)	Fixed carbon content (%)	Heat of combustion (cal/g)
200	91	80	19.5	4,600
250	67	64	35	5,500
300	47	42	57	6,200
350	39	37.5	62	6,420
400	34.5	31	68	6,620
500	27	19	79.5	7,170
600	24.5	11	87	7,480
800	21.5	7	91.5	7,430

Table 2. Regression analysis of carbonization temperature (300° - 600° C) and respective charcoal properties.

Items	a_0	a_1	$s_{y.x}$	s_0	s_1
Fixed carbon content vs temperature	26.63	.1025	1.22	2.24	5.05×10^{-3}
Volatile matter content vs temperature	73.99	-.1067	1.24	2.27	5.13×10^{-3}
Heat of combustion vs temperature	4,885	4.402	57.50	105.8	0.24
Heat of combustion vs fixed carbon content	3,743	42.93	27.82	80.35	1.123
Heat of combustion vs volatile matter content	7,937	-41.23	29.81	35.11	1.156

Note The correlation coefficient of each relationship was 1.00.

$$Y = a_0 + a_1 X$$

$s_{y.x}$ = Standard error of estimate (of Y on X)

s_0 = Standard error of regression coefficient a_0

s_1 = Standard error of regression coefficient a_1

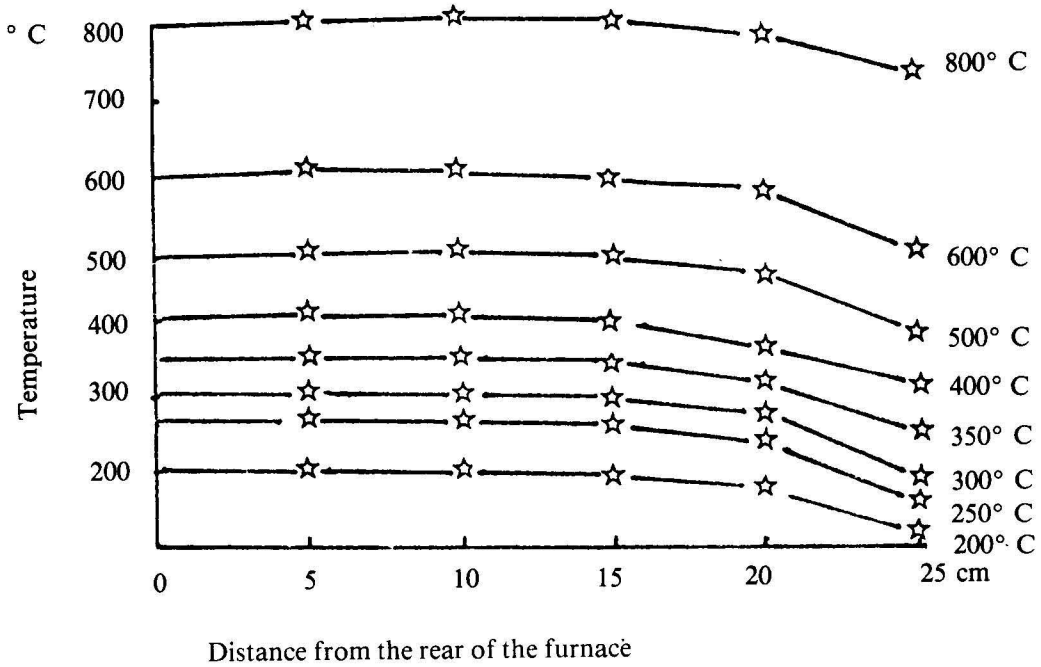


Fig. 1 Temperature profile of the furnace.

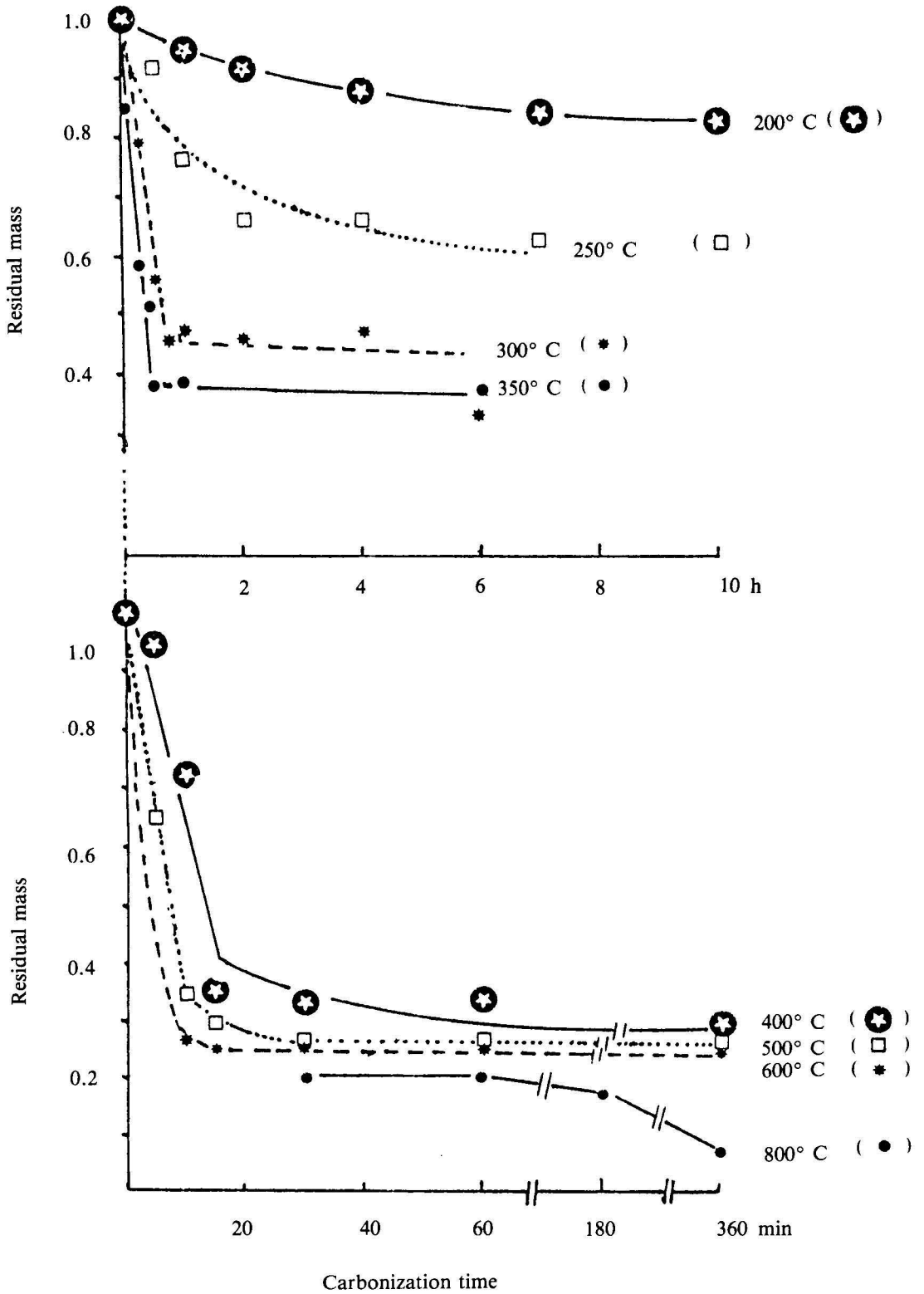


Fig. 2 Average residual mass from carbonization.

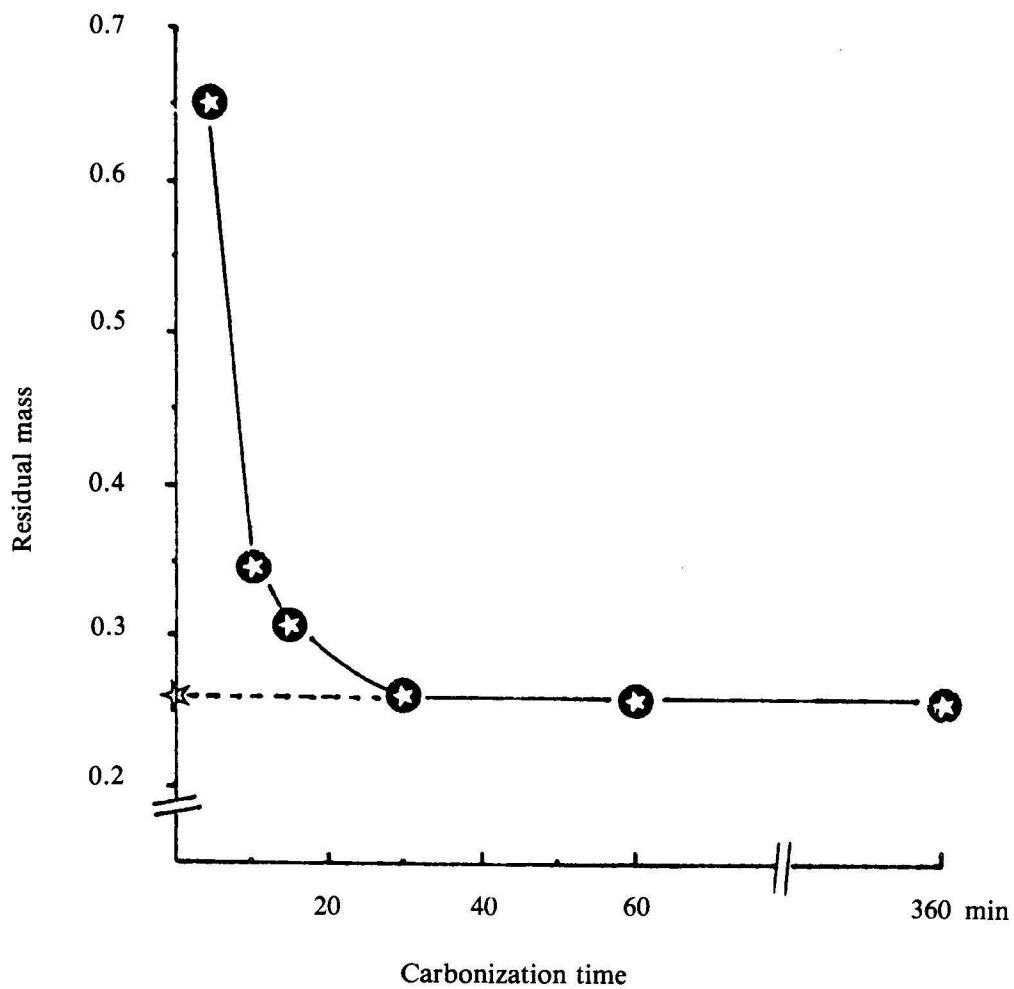


Fig. 3 Determination of average charcoal yield at 500° C.

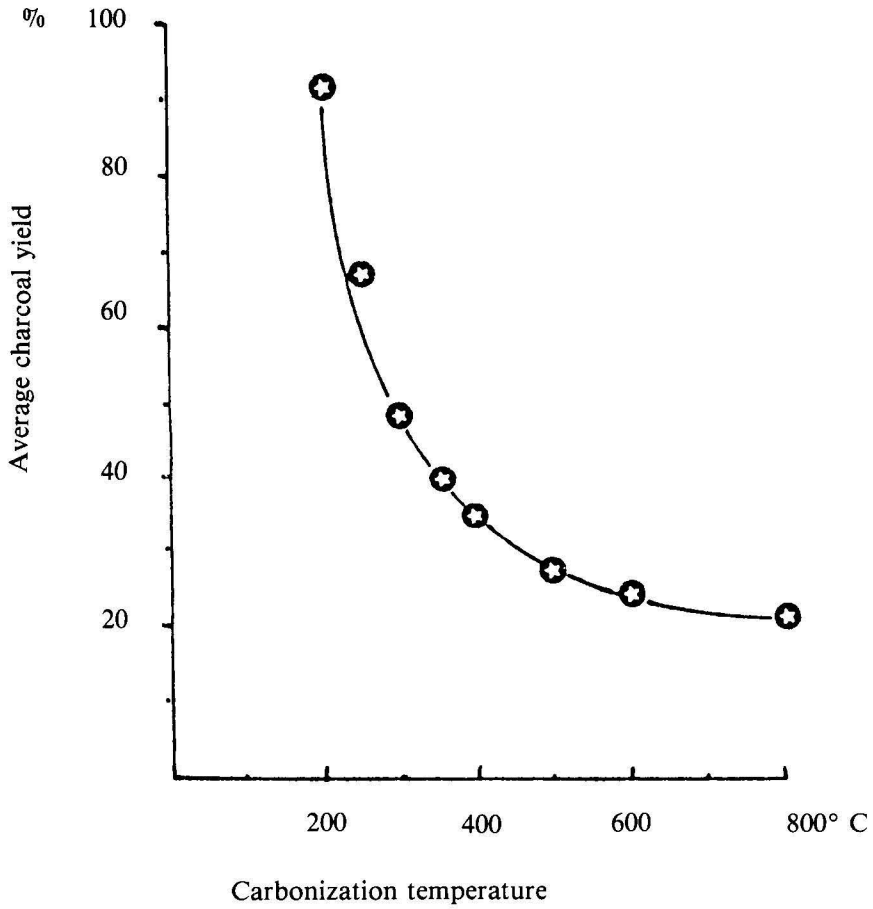


Fig. 4 Relationship between average charcoal yield versus carbonization temperature.

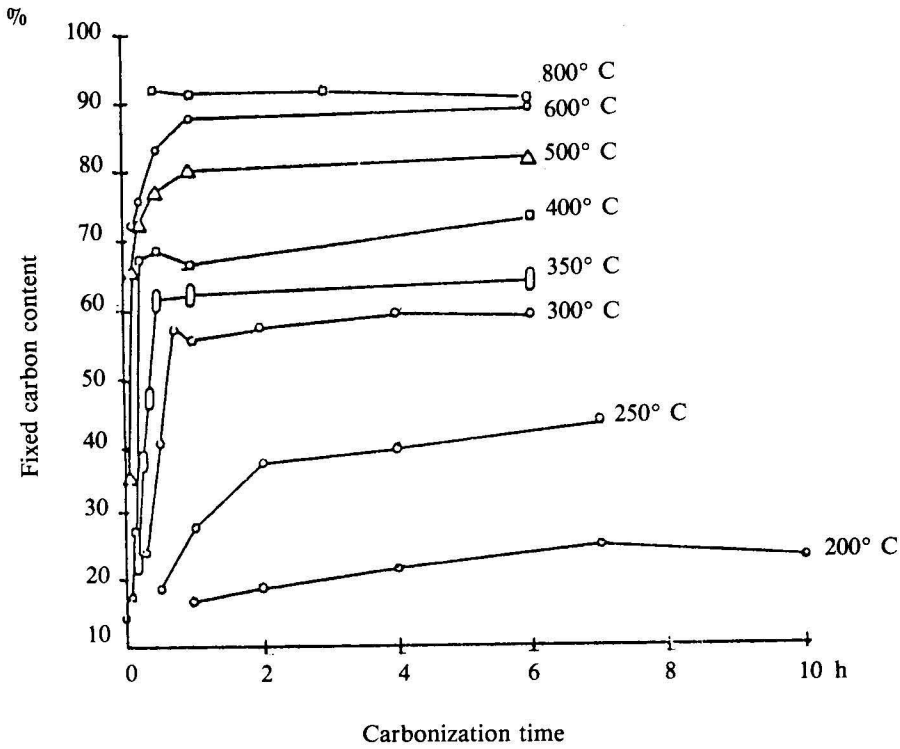


Fig. 5 Fixed carbon content in carbonized specimens.

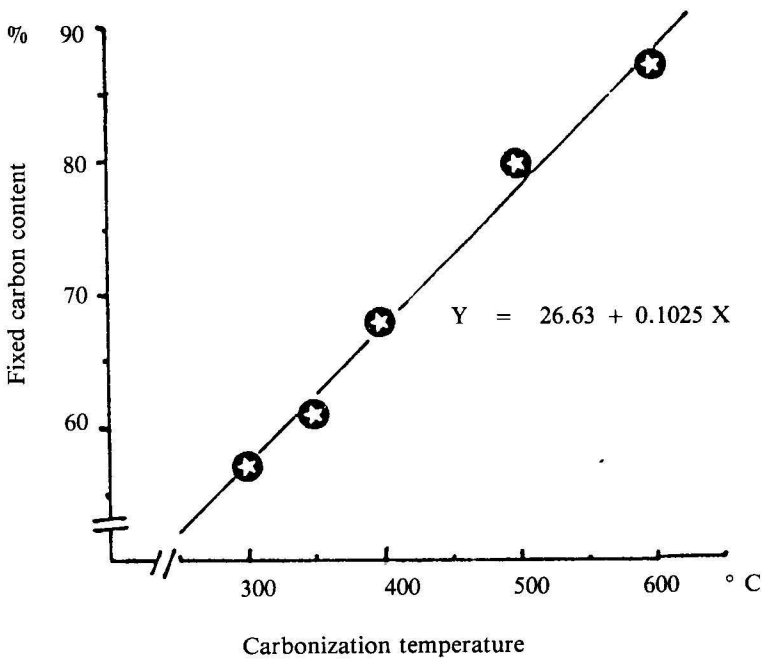


Fig. 6 Relationship between fixed carbon content versus carbonization temperature.

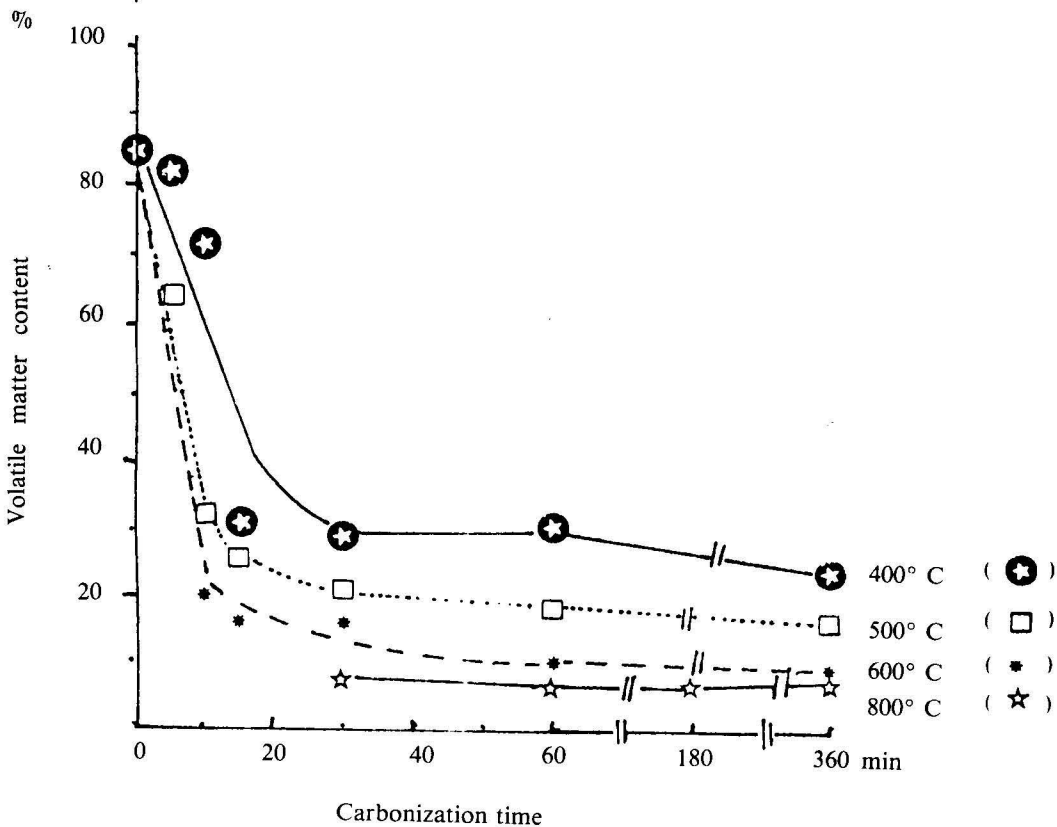
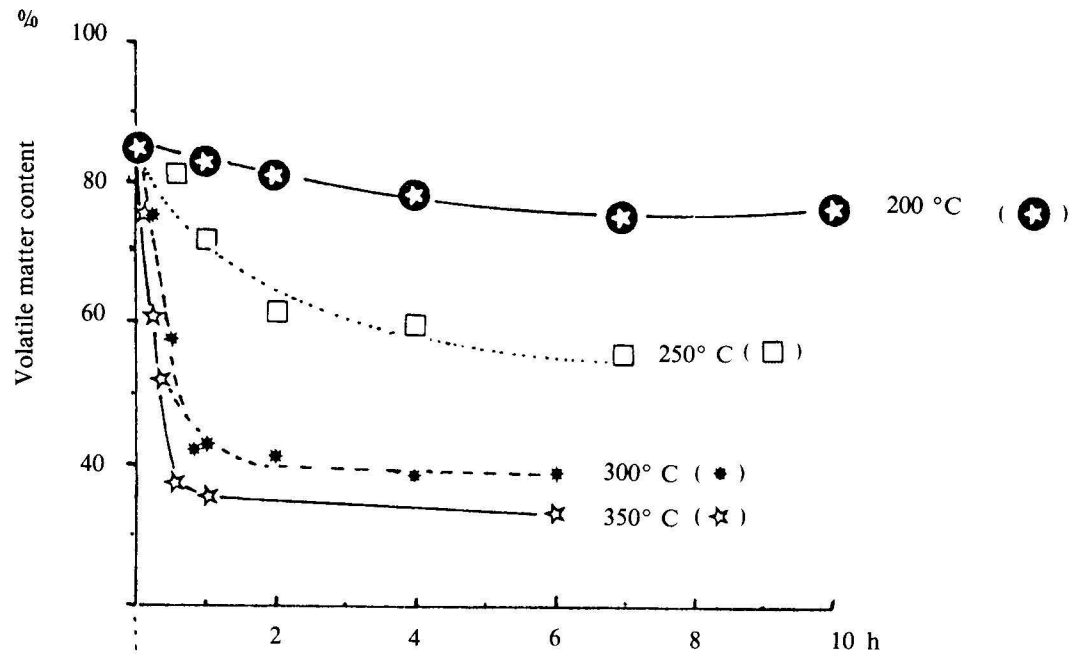


Fig. 7 Volatile matter content in carbonized specimens.

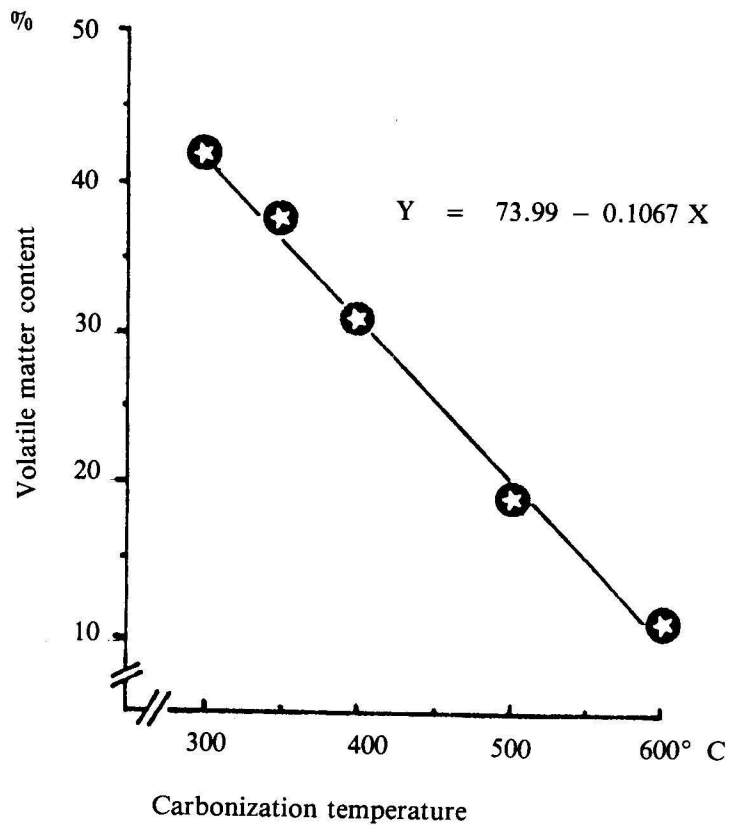


Fig. 8 Relationship between volatile matter content versus carbonization temperature.

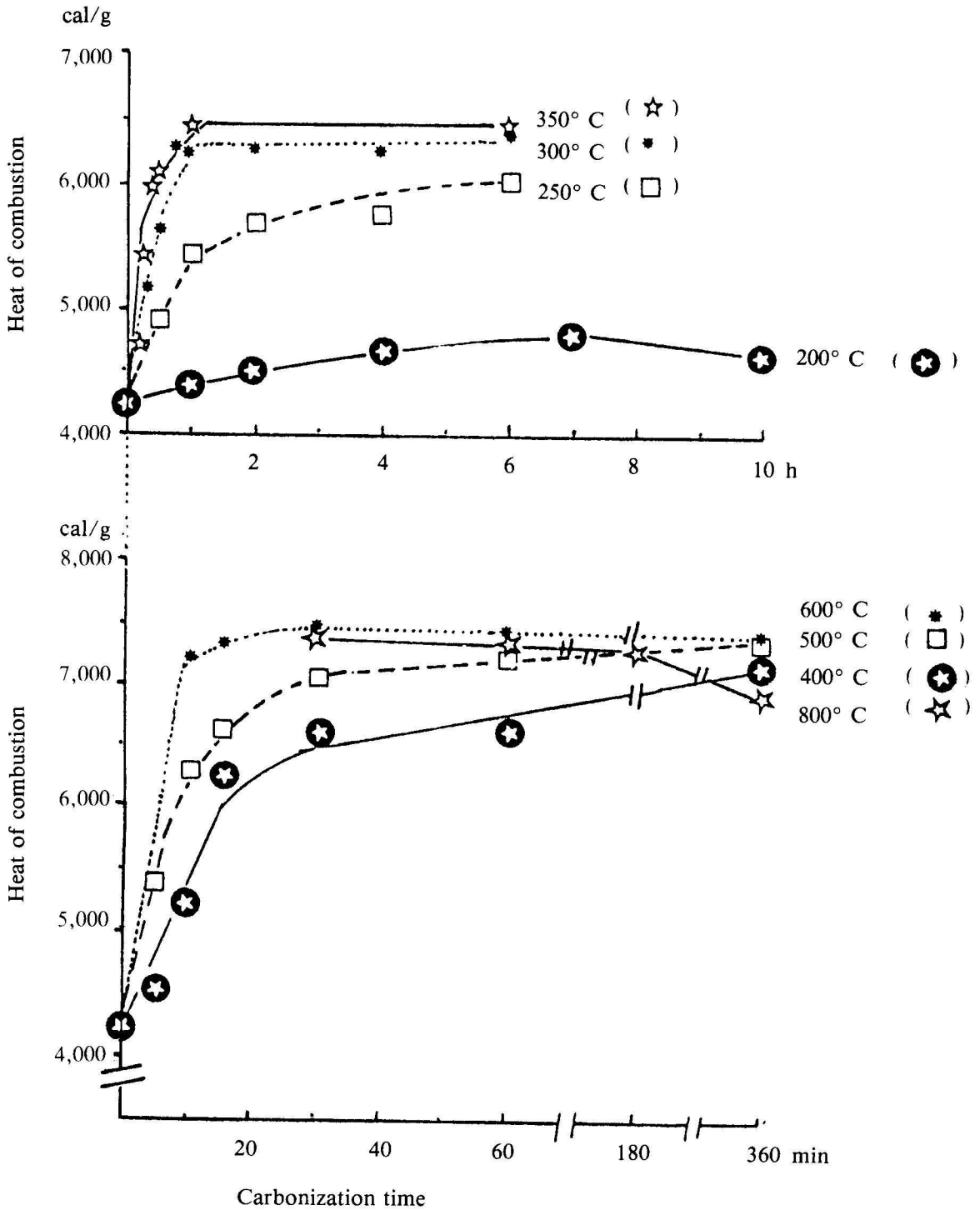


Fig. 9 Heat of combustion in carbonized specimens.

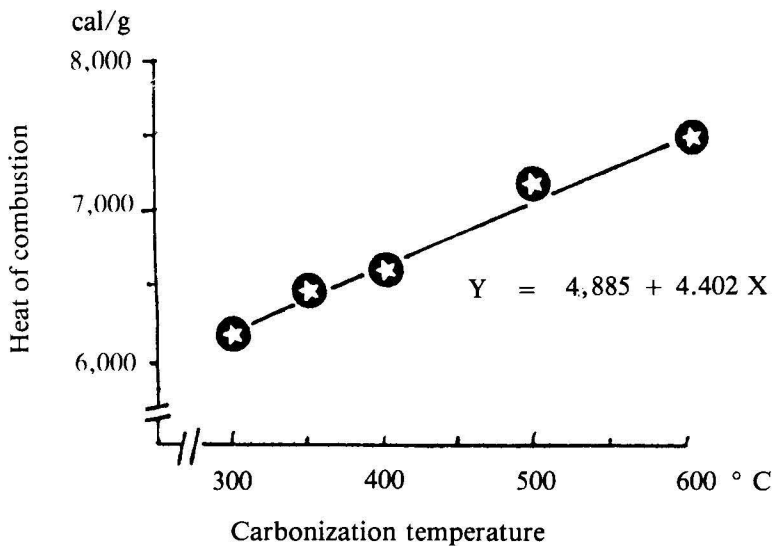


Fig. 10 Relationship between heat of combustion versus carbonization temperature.

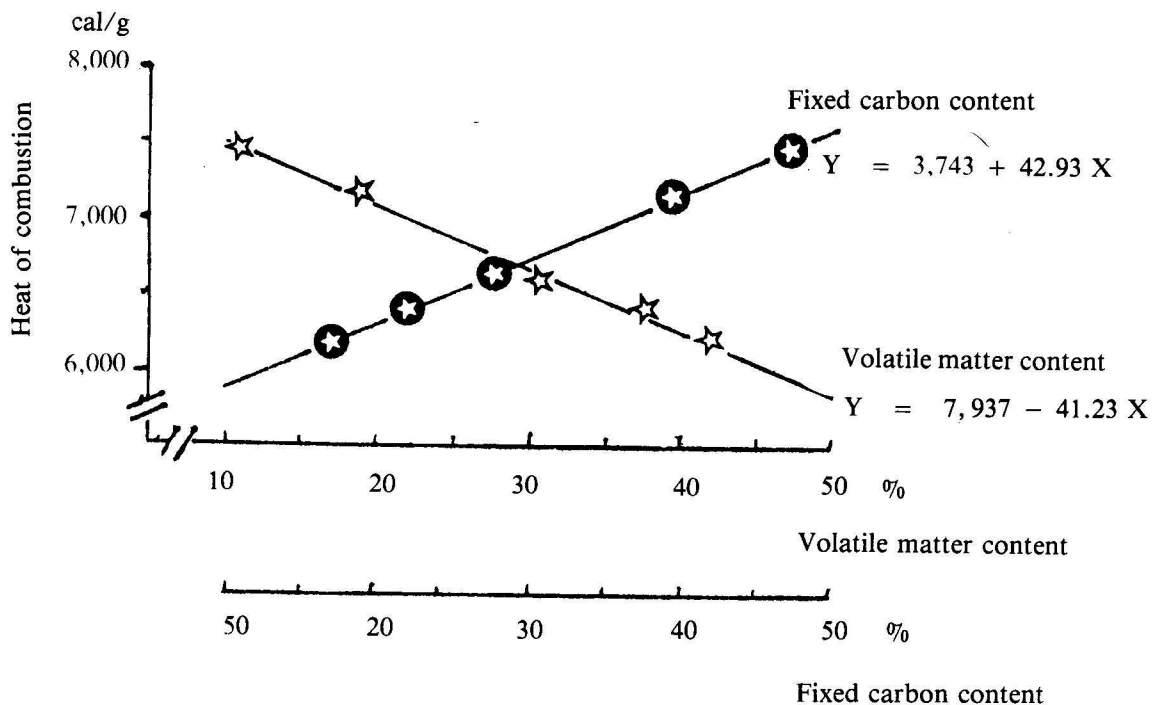


Fig. 11 Relationship between heat of combustion versus volatile matter content and fixed carbon contents.