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GASIFICATION OF COAL IN THE THERMAL ARGON PLASMA

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Key Words: High Temperature, Coal, Gasification, Plasma, Acetylene

The thermal decomposition of "Taiheiyo" coal was investigated experimentally, using a thermal argon plasma. The thermal plasma was generated by an arc discharge, and the temperature was approximately 10,000 K. The plasma and fine particles of coal were introduced into an 8 mm-i.d. tube reactor. The conversion to gasified products increased with increased plasma flow rate, and showed a maximum with the variation of input power.

The conversion of a coal particle was estimated by the energy balance. The main assumptions were that the decomposition rate was limited by heat transfer, and that the coal particle decomposed at a constant temperature. The results of this method are in good agreement with those of the experiments. The temperature profile within the particle was evaluated by numerical calculation.

Introduction

Thermal decomposition of coal using a plasma have been studied for the production of acetylene, and many papers^{2-4,8,9,15,17)} have appeared in this field. However, these results are not quantitative because they depend on many different experimental conditions, e.g., the experimental apparatus, kind of coal, input power, plasma flow rate, coal particle size, etc. Only qualitative information has been available.

In this paper, the conversion (defined in section 1.3) was quantitatively investigated. Particles of coal were injected into a thermal argon plasma in a tube, were heated by the plasma, decomposed thermally, and evolved gaseous products. Radial profiles of the temperature of a coal particle were estimated by numerical calculation, and the conversion to gaseous products was calculated by the energy balance in place of the usual numerical analysis. The assumptions used in this study were examined by numerical calculation.

1. Experiments

1.1 Experimental apparatus

The experimental apparatus consists of a plasma generator, a reacting tube, a cooling tube, a coal feeder and a gas chromatograph as shown in **Fig. 1**. **Figure 2** is a sectional diagram of the plasma generator and the reacting tube. The thermal plasma is generated by an arc discharge between a nozzle anode (8 mm i.d. and 40 mm in length) and a tungsten rod cathode (6 mm o.d. and 35 mm in height). Both electrodes are cooled by water to protect them from thermal damage. The temperature of the arc column depended upon the electric input power and the argon flow rate, and ranged from 7000 to 14,000 K. The reacting and cooling tubes are made of copper and cooled by water. Both tubes are of 8 mm i.d. The length of the reacting tube is 120 mm, and that of the cooling tube is 300 mm. Coal particles were fed to the reacting tube in an argon carrier gas through a hole located 50 mm downstream from the tip of the cathode. The coal feeder supplies fine coal particles

Received February 2, 1984. Correspondence concerning this article should be addressed to T. Honda.

Fig. 1. Experimental apparatus.

Fig. 2. Sectional diagram of plasma generator and reacting tube.

to the carrier gas flow. The feeder consists of a hopper, a gear, a speed converter, and a motor. The motor drives the gear through the converter, and controls the feed rate of coal by changing the number of revolutions of the gear in the range from 10 to 100 mg/s. The compositions of the product gases were measured by gas chromatograph, using dimethyl formamide and molecular sieve 13X columns. Helium gas was supplied into the reacting tube in argon carrier gas to measure the conversion.

1.2 Experimental procedure and conditions

First, the arc discharge was ignited and the generated thermal plasma was introduced into the reacting tube. After the argon plasma flowed steadily, fine particles of "Taiheiyo" coal were fed into the tube in argon carrier gas at a constant flow rate, 55.4 mg/s. The particles thermally decomposed in the reacting

Table 1. Experimental conditions

Argon flow rate	166–332	mg/s
Electric input power	1.9–5.2	kW
Coal diameter	60–100	mesh
	or 100–200	mesh
Coal feed rate	47.2	mg/s
Pressure in the reaction tube	101.3	Pa

Table 2. Analytical results* for "Taiheiyo" coal

Proximate analysis:	
Water	5.6 vol%
Ash	9.6 vol%
Volatile matter	44.8 vol%
Fixed carbon	40.0 vol%
Ultimate analysis:	
Ash	10.19 wt%
Carbon	69.01 wt%
Hydrogen	5.68 wt%
Nitrogen	1.05 wt%
Oxygen	13.53 wt%
Sulfur total	0.24 wt%
Heat of combustion:	6490 cal/g

* Analyzed by the Coal Mining Research Center.

tube, releasing gaseous product which were sampled through a filter and analyzed by gas chromatograph. The conversion and the mole fractions of product were determined. The reaction was carried out at atmospheric pressure. Experimental conditions and the proximate and ultimate analyses of the coal used are shown in Tables 1 and 2, respectively.

1.3 Experimental results

Acetylene, methane, hydrogen, and carbon monoxide were detected by gas chromatograph as product gases. No tar was detected under the present conditions. Therefore, the conversion was defined by the ratio of gasified weight of the supplied coal to initial weight.

Changes in conversion were first examined, choosing a coal feed rate which ranged from 11 to 102 mg/s. The conversion decreased at feed rates over 50 mg/s because the plasma temperature decreased due to the heat capacity of the coal particles. Therefore, a coal feed rate of 47.2 mg/s was used in this study.

Figure 3 shows the effects of the argon flow rate on the conversion for two coal particle sizes and an electric input power of 3.2 kW. The conversion increases with argon flow rate and is inversely proportional to the square of the coal particle diameter. The solid curves in this and the next figure show the calculated results, which will be described later. Figure 4 shows the effects of the electric input power on the conversion at an argon flow rate of 277 mg/s. The conversion increases with input power under 4 kW because of the increase in plasma temperature, but decreases over 4 kW.

Fig. 3. Conversion vs. argon flow rate

Fig. 4. Conversion vs. electric input power.

Figures 5 and 6 show the mole fraction changes of the product gases for a coal size of 60–100 mesh. They show that the mole fractions do not significantly depend on either the argon flow rate or the input power.

2. Calculations

The conversion can be evaluated by solving, the unsteady heat conduction problem within a coal particle. This requires information concerning the trajectory of the particle.

2.1 Trajectory of a particle in a field of non-uniform temperature and velocity in the reacting tube

As the temperature and velocity distributions of the plasma flow in the reacting tube were known to be a function of the plasma flow rate and the input power,¹⁰⁾ the trajectory of the particle was numerically calculated by solving the following momentum equations.

$$\frac{dV_r}{dt} = -\frac{3}{4} c_D \frac{\rho_p}{\rho_c D_c} \{V_r^2 + (u - V_z)^2\}^{1/2} V_r \quad (1)$$

$$\frac{dV_z}{dt} = \frac{3}{4} c_D \frac{\rho_p}{\rho_c D_c} \{V_r^2 + (u - V_z)^2\}^{1/2} (u - V_z) - g \quad (2)$$

The collision with the wall is assumed to be perfectly

Fig. 5. Mole fractions vs. argon flow rate.

Fig. 6. Mole fractions vs. electric input power.

Table 3. Properties of "Taiheiyo" coal

				Units
T_d	723 ¹⁾	T_{sub}	3672 ¹³⁾	K
ΔH_d	7.41 ⁵⁾	ΔH_{sub}	59.8 ¹⁴⁾	MJ/kg
ρ_c	750 ⁷⁾	ρ_d	450*	kg/m ³
c_{pc}	1050 ⁷⁾	c_{pd}	1050**	J/kg·K
k_c	0.256 ⁷⁾	k_d	0.154*	W/m·K

* $750 \times 0.6 = 450$; $0.256 \times 0.6 = 0.154$.

** Assumed to be equal to c_{pc} .

elastic. The drag coefficient C_D is given in the literature,¹¹⁾ and the properties of the coal are shown in Table 3. The residence time of the particles in the reacting tube was numerically estimated at about 5 ms.

2.2 Thermal decomposition of a fine particle of coal in a field of uniform temperature and velocity

First, the amount of thermal decomposition of a

Fig. 7. Radial temperature profiles in a coal particle.

Fig. 8. Changes in conversion with time.

spherical particle of coal in a field of uniform temperature and velocity was calculated along with time by solving a one-dimensional unsteady heat conduction problem. The phase change and the evolution of gaseous products were included as the result of thermal decomposition.^{5,6,8,12,16} The assumptions used in this calculation are:

- a) the decomposition rate is limited by the heat transfer from the plasma to a particle,
- b) radiation is negligible,
- c) only gaseous products are released from the coal particle (no tar was observed),
- d) the coal particle decomposes at a constant temperature, and
- e) the diameter of the coal particle does not vary during the decomposition.

The diameters of the spherical particles were 0.1 and 0.2 mm corresponding to the size of 100–200 and 60–100 mesh, respectively, used in the experiment.

2.2.1 Temperature profiles in a coal particle using numerical analysis The radial profiles of temperature in a 0.2 mm-diameter particles were calculated by numerical analysis and are shown in Fig. 7, where the parameter is the reaction time. The procedures of the numerical method are given in Appendix. The initial temperature of the particle is 300 K, and the plasma temperature is 8000 K. First, the particle is heated, and then decomposes at 0.2 ms. At time 30 ms, the surface of the particle begins to sublimate and the

Fig. 9. Definition of τ and c' .

radius is decreasing to 0.18 mm from the initial value of 0.20 mm.

The residence time of the particle in the reacting tube is about 5 ms, as mentioned in section 2.1, and the plasma temperature of 8000 K is considered to be the highest under these experimental conditions. Therefore, the particle does not sublimate under the present conditions. This supports assumption e. At 5 ms, the temperature of the particle is about 1100 K, and the amount of radiation from the surface is one-hundredth at most of that of the heat transfer from the plasma. This supports assumption b.

The thick curves in Fig. 8 show some typical calculated results of the conversion of a 0.2 mm-diameter particle as a function of time elapsed. The parameters in this figure are the plasma temperature and velocity. The conversion increases rapidly in regions of high temperature and velocity. However, the calculated conversions do not exceed 0.4 because only the volatile matter is assumed to decompose. The thin lines, which also show calculated results, are mentioned in the next section.

2.2.2 Evaluations of heating time and conversion rate using energy balance From the above results shown by thick curves, the following two periods may be considered:

- 1) the heating period τ_j in which the particle is heated up to the decomposition temperature and does not decompose, and
- 2) the decomposition period in which the particle decomposes and the conversion increases linearly up to 0.4 (the conversion rate is given from the slope as a constant c'_j).

The heating time τ_j and the conversion rate c'_j are defined as shown in Fig. 9, and the two are obtained from the energy balance between a coal particle and the plasma as follows.

The energy balance during the heating period is

$$\frac{1}{6} D_c^3 \rho_c c_{pc} (T_d - T_i) = D_c^2 h^0 (T_p - T_s) \tau_j \quad (3)$$

where 1) the temperature in the coal particle is

assumed uniform, and 2) the surface temperature T_s is also assumed to be given as the arithmetic mean of T_i and T_d . And, h^0 is given by Eq. (A-8). τ_j is obtained from this equation as a function of T_p and velocity.

The energy balance during the decomposition period is

$$\begin{aligned} & \frac{1}{6} D_c^3 \rho_c c'_j \{ \Delta H_d + c_{pd}(T_s - T_d) \} \\ & = D_c^2 h^0 \frac{B}{\exp(B) - 1} (T_p - T_s) \end{aligned} \quad (4)$$

The heat flux at the surface is given by

$$k_d \frac{T_s - T_d}{D_c/2} = h^0 \frac{B}{\exp(B) - 1} (T_p - T_s) \quad (5)$$

c'_j and T_s are calculated from Eqs. (4) and (5). These evaluations require iteration of the calculation because the blowing parameter B is a function of c'_j .

The values of τ_j and c'_j are shown by the thin solid lines in Fig. 8 for $D=0.2$ mm. The required time for decomposition is one fourth that for $D=0.1$ mm. No changes in particle temperature can be observed experimentally, so that the results obtained by the energy balance method are comparable with those by numerical analysis. As the thin lines agree with the results of numerical analysis as shown by the thick curves in Fig. 8, the energy balance method is of similar quality for estimation of the conversion as is numerical analysis. The energy balance method was used in evaluation of the conversion afterwards.

2.3 Calculation of conversion using heating time and conversion rate

τ and c' are estimated from the results of τ_j and c'_j obtained by the energy balance method under the corresponding temperature and velocity conditions obtained from the calculation of the trajectory of a particle.

The total amount of heat from the plasma flow to a coal particle in the heating period Q is given by

$$\frac{Q}{M_c c_{pc}} = \int \frac{dT}{dt} dt = \sum_{j=1}^1 \frac{T_d - T_j}{\tau_j} \Delta t \quad (6)$$

where Δt is a time increment and τ_j is the heating time corresponding to the temperature and velocity conditions at the j -th time segment. As the amount of heat required to heat up the particle to the decomposition temperature is given by $M_c c_{pc}(T_d - T_i)$, the following equation is obtained.

$$\sum_{j=1}^m \frac{\Delta t}{\tau_j} = 1 \quad (7)$$

where m is determined so as to satisfy Eq. (7) and denotes the time of the end of the heating period. The heating time τ is obtained as $m\Delta t$.

In the decomposition period, the conversion at time

$n\Delta t$ is obtained by the following equation.

$$\sum_{j=m+1}^n c'_j \Delta t = c \quad (8)$$

where c'_j is the conversion rate at time $j\Delta t$ obtained by the energy balance method for a uniform field that has the same temperature and velocity as the actual field in the reaction tube. The effects of the injection velocity of the particle on the conversion were numerically examined, and are not significant except for the very low-velocity region. The error caused by the injection velocity is considered to be small in this experiment.

2.4 Comparison with the experimental values

The results of the energy balance method are shown as solid curves in Figs. 3 and 4. These results are in good agreement with the experimental values. Therefore, the assumptions of this calculation are considered to be reasonable. Figure 3 shows that conversion increases with plasma flow rate. This can be explained by the fact that a high plasma flow rate causes a high temperature of the plasma flow. The calculated conversions, solid curves in Fig. 4, monotonically increase with input power. However, the experimental results have maxima. The decrease in the conversion observed experimentally over 4 kW can be considered to be caused by the fusion of the particle surface, which prevents product gas release from the coal.

Conclusions

Hydrogen, methane, acetylene, and carbon monoxide were observed as product gases of thermal decomposition of "Taiheiyo" coal, using a thermal argon plasma. The experimentally observed conversion increases with the argon plasma flow rate and has a maximum against the input power. Experimentally observed changes in mole fractions were not significantly large.

The variations in conversion with argon flow rate and input power were calculated and agreed with the experimental results within the experimental error limits, using a combination of the energy balance method and the results of the trajectory calculation of a coal particle. The heat transfer limiting step and a constant decomposition temperature are assumed in the calculation.

Appendix

The temperature distribution in a coal particle is obtained by solving a one-dimensional unsteady heat conduction equation:

$$\rho_c c_{pc} \frac{\partial T}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 k_c \frac{\partial T}{\partial r} \right) \quad (A-1)$$

where the initial condition is

$$T = T_i \quad \text{at} \quad t = 0 \quad (A-2)$$

and the boundary conditions are as follows.

$$\frac{\partial T}{\partial r} = 0 \quad \text{at } r = 0 \quad (\text{A-3})$$

$$h(T_p - T) = k_c \frac{\partial T}{\partial r} \quad \text{at } r = R \quad (\text{A-4})$$

When the temperature reaches the decomposition temperature, the following boundary condition is added.

$$T = T_d \quad \text{at } r = r' \quad (\text{A-5})$$

where r' is the radial position of the reacting spherical interface. Furthermore, when the surface temperature of the particle reaches the sublimation temperature of carbon, one more boundary condition,

$$T = T_{\text{sub}} \quad \text{at } r = R \quad (\text{A-6})$$

is added and the sublimation is taken into consideration.

The heat transfer coefficient is obtained as follows.

$$h = h^0 \frac{B}{\exp(B) - 1} \quad (\text{A-7})$$

where

$$h^0 = \frac{k_p}{D_c} Nu = \frac{k_p}{D_c} (2 + 0.6 Re^{1/2} Pr^{1/3}) \quad (\text{A-8})$$

B in Eq. (A-7) is a blowing parameter showing the gas blowing effect on h and is given by

$$B = \frac{\rho_w u_w c_{pw}}{h^0} = \frac{D_c}{6} c' \frac{c_{pw}}{h^0} \quad (\text{A-9})$$

where $\rho_w u_w$ is estimated from the experimental decomposition rate c' as shown on the right-hand side of Eq. (A-9).

Equation (A-1) is converted to a finite difference equation and solved by the explicit method, since Eq. (A-1) is parabolic.

Nomenclature

B	= blowing parameter	[—]
c	= conversion	[—]
c'	= conversion rate	[s ⁻¹]
c_D	= drag coefficient	[—]
c_p	= specific heat	[J/kg · K]
D	= diameter of cooling tube	[m]
D_c	= diameter of coal particle	[m]
g	= gravitational acceleration	[m/s ²]
ΔH	= enthalpy change	[J/kg]
h	= heat transfer coefficient	[W/m ² · K]
h^0	= h without blowing	[W/m ² · K]
k	= thermal conductivity	[W/m · K]
M	= mass	[kg]
m'	= mass flow rate of argon	[kg/s]
Nu	= Nusselt number	[—]
P	= partial pressure	[Pa]
Pr	= Prandtl number	[—]
Q	= accumulated amount of heat to a particle	[J]
Re	= Reynolds number	[—]

r	= radial coordinate	[m]
r'	= radius of reacting interface	[m]
T	= temperature	[K]
t	= time	[s]
Δt	= time increment	[s]
u	= plasma flow velocity	[m/s]
V	= particle velocity	[m/s]
z	= axial coordinate	[m]

ρ	= mass density	[kg/m ³]
τ	= heating time	[s]

<Subscripts>

c	= coal
d	= decomposition, or decomposed part of coal
i	= initial
j	= j -th time segment, or uniform field
p	= plasma
r	= radial coordinate
s	= surface
sub	= sublimation
w	= wall
z	= axial coordinate

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