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Abstract

Biocoalbriquette, a new artificial solid fuel, is manufactured by a mixture of coal and biomass under a high compression pressure. The combustion characteristics of biocoalbriquette were investigated in this study both experimentally and numerically. The combustion process of biocoalbriquette appears in two stages, the volatile combustion stage followed by the char combustion stage. It was found that the volatile combustion happens over the whole pellet of biocoalbriquette, whereas the char combustion proceeds in a shrinking-core mode. A volume model and a shrinking-core reaction model were introduced and modified here to simulate the two stages of combustion process. The simulation results are found to be consistent with the experimental results.

Introduction

Among the fossil fuels, coal has the highest potential as one of future energy sources in the world. Even though coal is mainly used for power generation in large scale utility boilers, the amount of coal used in domestic stoves and small scale industrial boilers is still large, especially in developing countries. The direct burning of coals, especially low grade coals with high ash, sulfur and nitrogen contents and low heat values, in small boilers and stoves causes serious pollution of the environment, such as dust, SO_x and NO_x emissions. In order to enhance effective utilization of low-grade coals and to control the emission of environmental pollutants, coal briquette had been developed. As a technique for coal combustion in small boilers and stoves, coal briquette can limit the dust emission to a great extent (Ford *et al.*, 1991, and Kim *et al.*, 1997). In recent years, biocoalbriquette has been developed as an improved technique on coal briquette, which is produced from a mixture of coal and biomass (forestry/agriculture wastes, such as bark, rice straw and cornstalk) under high compression pressure (Lu *et al.*, 1997). Experiments have shown that the biocoalbriquette has high ignition and combustion ability, low emissions of pollutants such as smoke and dust particulate (Kim *et al.*, 1997, Lu *et al.*, 1997, and Maruyama and Kamide, 1995). It was also found that the SOx and NOx emissions can be greatly decreased by mixing small amounts of desulfurizer and denitrifizater into biocoalbriquette (Naruse *et al.*, 1998). In this paper, the characteristics of biocoalbriquette combustion are investigated experimentally and numerically. The biocoalbriquette combustion process is found to appear in two stages, the volatile combustion stage and the char combustion stage. On the basis of the experimental results, a volume reaction model and a shrinking-core reaction model are adopted and modified to simulate these two combustion stages. The simulation results are consistent with the experimental results.

Experimental

The experiments were performed in an electrically heated batch furnace, schematically shown in Fig.1, which consists of temperature controllers, a digital balance, and a flue gas analyzing system. The sample was suspended by a wire linked to the digital balance and was positioned in

the center of the furnace axis. The furnace was preheated up to a predetermined temperature, and then moved upward to heat the biocoalbriquette. The digital balance continuously measured the mass loss in combustion. Five types of coal and a kind of biomass were tested in the experiments, and their properties are shown in Table 1. The tested coals have a wide range in fuel ratio (ratio of fixed carbon content to volatile content) from 0.8 to 4.3. The biomass content in biocoalbriquette and the compression pressure were varied from 0 to 100 mass % and from 49.0 to 392.3 MPa, respectively.

Results and Discussion

Experimental results

Both combustion and pyrolysis experiments were carried out, and the measured time histories of unburned and unpyrolyzed fractions of biocoalbriquette are shown in Fig.2. The combustion time history is clearly divided into two parts: the first part having a rapid mass loss and the second part a slower mass loss. Comparing to the time history of pyrolysis, the first part of combustion history can be considered corresponding to the volatile combustion, whereas the second part to the char combustion. The volatile matters in biocoalbriquette evolve when the biocoalbriquette is heated up to a temperature, and burnt in gas phase around the biocoalbriquette. The char combustion does not occur on the surface of the biocoalbriquette until the volatile combustion is almost finished. As oxygen diffuses from the surface toward the inside, the char burns to form a flame sheet which keeps moving toward the inside till the center when char is completely burnt out. The char combustion process appears in a shrinking-core reaction mode, which was conformed by the observation of cross-sectional structure of biocoalbriquette during combustion, as shown in Fig.3.

The combustion characteristics of biocoalbriquette depend on the amount of biomass addition and the coal properties. As seen in Fig. 2, the burnt out time of biocoalbriquette decreases as the amount of biomass addition increases because biomass contains more volatile matters than coal. For different kinds of coals, the coal with a higher volatile content, corresponding to a lower fuel ratio, burns in a shorter time. This can be seen from Fig. 4, which shows the burnt out times for

coal briquettes with four tested coals. The burnt out time increases as the fuel ratio of coal increases.

The char combustion takes most time of the whole biocoalbriquette combustion process. The combustion rate may depend on the diffusion rate of oxygen through both ash layer and gas boundary layer around the biocoalbriquette and/or the chemical reaction rate. To determine the char combustion rate-controlling step, the combustion experiments were conducted at the varied bulk gas temperatures from 873 to 1073 K. The profiles of unburned fraction against normalized combustion time are shown in Fig.5. The combustion profiles do not show big differences at various bulk gas temperatures, which suggests that the temperature hardly exert influence on the char combustion rate. In other words, the char combustion is not controlled by chemical reactions. It is seen in Fig. 5 that the combustion profiles locate between the gas film diffusion control and the ash layer diffusion control, and we can conclude that the char combustion process is controlled by oxygen diffusion through both the gas boundary layer and the ash layer.

In this study, the influence of biocoalbriquette shape and compression pressure on the combustion rate was also investigated. A cylindrically shaped biocoalbriquette, with the dimension 2R in both diameter and height, was seen to have nearly the same combustion process as a spherically shaped biocoalbriquette with the diameter 2R. The influence of the compression pressure on the combustion rate appears in the way that the burnt out time slightly increases as the compression pressure increases. This is because the compression pressure has an influence on the porosity in ash layer of the burned part of biocoalbriquette and then on the oxygen diffusion in the ash layer, which will be discussed next.

Models and simulation

As discussed above, the biocoalbriquette combustion process occurs in two different stages, i.e. the volatile combustion stage and the char combustion stage. To simulate the combustion process, two models are necessarily developed for these two stages.

Volume reaction model. Since the biocoalbriquette has a rapid mass loss in the volatile combustion process, the volatile evolution can be considered occurring volumetrically all over

the biocoalbriquette. Therefore, a volume reaction model (Arai *et al.*, 1981) can be adopted to describe the volatile combustion process, and the reaction rate is expressed as follows;

$$dx/dt = k(1-x)^n \tag{1}$$

Here, t is the reaction time [s], k the reaction rate coefficient [s^{-1}], x the reacted mass fraction [-], and n the reaction order [-]. Both k and n depend on the coal type and the biomass addition, and the correlation are expressed as follows on the basis of the experiments,

$$n = n_0 - 4.2X_{bm} \tag{2}$$

$$n_0 = 1.69 + 1.061FR \tag{3}$$

$$k = k_0 + 0.007X_{bm} \tag{4}$$

$$k_0 = 0.0039 - 0.00061FR \tag{5}$$

Here X_{bm} is the biomass content (by mass base) in biocoalbriquette [-], and FR the fuel ratio [-].

Shrinking-core reaction model. As discussed above, the char combustion happens in the shrinking-core mode, and the combustion rate is controlled by the oxygen diffusion through both the gas boundary and the ash layer. Introducing a shrinking-core reaction model (Levenspiel, 1972), we can simulate the char combustion process. When the flame sheet moves from the surface toward the unburned core with a radius of r_c, the elapsed char combustion time t_c can be calculated as follows;

$$t_c = \frac{R}{AC_0}\left[\frac{1}{3}\left(\frac{1}{K_c} - \frac{R}{D_c}\right)\left(1 - \frac{r_c^3}{R^3}\right) + \frac{R}{2D_c}\left(1 - \frac{r_c^2}{R^2}\right)\right] \tag{6}$$

Here, R is the radius of the biocoalbriquette [m], K_c the oxygen diffusion coefficient in the gas boundary layer [$m \cdot s^{-1}$], D_c the effective oxygen diffusion coefficient in the ash layer [$m \cdot s^{-1}$], A the volume of reactable char per mole-oxygen [$m^3 \cdot mol^{-1}$], and C_0 the oxygen concentration in atmosphere [$mol \cdot m^{-3}$]. If assuming that the reactable char consists of carbon, hydrogen, nitrogen, and sulfur, A can be calculated as

$$1/A = \rho_0(1 - Ash_0 - Moisture)(1 - VM_{(bb.T)})(C/12 + N/14 + S/32 + H/4) \times 10^3 \tag{7}$$

where ρ_0 is the density of biocoalbriquette [$kg \cdot m^{-3}$], Ash_0 the ash fraction in biocoalbriquette [-],

Moisture the moisture fraction in biocoalbriquette [-], $VM_{(bb.T)}$ the volatile fraction in biocoalbriquette [-], and C, N, S, and H are contents of carbon, nitrogen, sulfur, and hydrogen in char [-], respectively.

The effective oxygen diffusion coefficient in the ash layer, D_c, is closely related to the porosity in ash layer, whereas the porosity in ash layer in turn depends on the ash properties of both coal and biomass, the compression pressure, and the amount of biomass addition. As seen in Fig. 6, when the compression pressure increases, the porosity in ash layer decreases at first and then keep nearly unchanged. On the other hand, the porosity in ash layer increases almost linearly as the amount of biomass addition increases. The correlation between the porosity and the fuel ratio is shown in Fig. 7. With and without biomass addition, the porosity in ash layer always decreases as the fuel ratio increases. This is because the more volatile is contained in coal briquette and biocoalbriquette, the more porosity will be produced after devolatilization. It was also observed that the diameter of biocoalbriquette did not change during the combustion. On the basis of experimental results and observations, the porosity in ash layer, ε_{ash}, can then be calculated as

$$\varepsilon_{ash} = 1 - Ash_0 \frac{\rho_0}{\rho_{ash}} \tag{8}$$

$$Ash_0 = Ash_{bm} X_{bm} + Ash_{coal}(1 - X_{bm}) \tag{9}$$

$$\rho_{ash} = \frac{\rho_{ash(coal)} \rho_{ash(bm)} [Ash_{coal}(1 - X_{bm}) + Ash_{bm} X_{bm}]}{\rho_{ash(bm)} Ash_{coal}(1 - X_{bm}) + \rho_{ash(coal)} Ash_{bm} X_{bm}} \tag{10}$$

Here ρ_{ash}, $\rho_{ash(coal)}$, and $\rho_{ash(bm)}$ are the real densities of biocoalbriquette ash, coal ash, and biomass ash, respectively. Ash_{coal} and Ash_{bm} are the ash fractions in coal and biomass, respectively.

A close correlation was found between the effective oxygen diffusion coefficient in the ash layer, D_c, and the porosity in ash layer by the experimental results, as shown in Fig. 8, and can be described as

$$\log D_c = 1.757 \varepsilon_{ash} - 5.5546 \tag{11}$$

The oxygen diffusion coefficient in the gas boundary layer, K_c, was determined by experiments, and was found to be almost the same in the tested range of biocoalbriquette

diameter as

$$K_c = 3.0 \times 10^{-2} \tag{12}$$

Comparison between prediction and experiments. The above-mentioned models were applied to simulate the combustion processes of three kinds of biocoalbriquettes, with two types of coal and different amounts of biomass addition. The simulated combustion processes are shown in Fig. 9, together with the corresponding experimental results for comparison. The good agreements can be seen between the simulations and the experiments for these three biocoalbriquettes in both stages of volatile combustion and char combustion. It reveals that the developed two models should be suitable for the simulation of the combustion process of biocoalbriquette.

Conclusions

The combustion process of biocoalbriquette can be divided into two stages, the volatile combustion stage followed by the char combustion stage. At the volatile combustion stage, the volatile evolution occurs over the whole pellet of biocoalbriquette. A volume model has therefore been adopted to simulate the volatile combustion, and the reaction rate, correlating to the biomass addition and the coal property, was determined by experiments. The char combustion appears in the shrinking-core mode, with a flame sheet moving from the surface toward the center of the biocoalbriquette. The char combustion rate was found to be controlled by oxygen diffusion through both the gas boundary layer and the ash layer, and the formulas were derived to calculate the oxygen diffusion coefficients on the basis of experiments. A shrinking-core reaction model can then describe the char combustion process. The combustion processes of different kinds of biocoalbriquettes have been simulated by the developed two models, and the simulation results are consistent with the experiments. The developed models for biocoalbriquette combustion in this paper will provide useful information for the production and application of biocoalbriquette, such as the design of boiler for biocoalbriquette combustion.

Acknowledgments

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Figure and Table Captions

Table 1 Properties of tested coal and biomass

Fig.1 Experimental apparatus

Fig.2 Influence of biomass content on burning profile

Fig.3 Change of cross-sectional structure during biocoalbriquette combustion

Fig.4 Influence of coal type on burning out time

Fig.5 Normalized combustion profiles under different furnace temperatures

Fig.6 Porosity in ash layer as functions of biomass content and compression pressure

Fig.7 Porosity in ash layer vs. different coal type

Fig.8 Correlation between diffusion coefficient and porosity at various experimental conditions

Fig.9 Comparison of calculations and experiments

Table 1 Properties of Tested Coals and Biomass

Sample		Proximate analysis [mass %, d. a. f.]			Fuel Ratio	Ultimate analysis [mass %, d. a. f.]				Real density [kg/m^3]	Calorific value [kJ/kg]
		Ash	VM	FC	[-]	C	H	N	S	ρ_{ash}	Q_H
Coal	SB	2.7	53.6	43.7	0.8	71.8	5.2	1.6	1.3	2650	25400
	BR	17.6	32.7	49.2	1.5	81.9	5.1	1.8	0.4	2790	25700
	BJ	20.5	19.6	59.9	3.1	83.9	4.2	1.0	1.6	—	26700
	SV	16.7	19.4	63.9	3.3	90.2	4.9	1.0	0.3	2770	28300
	DS	15.3	16.1	68.6	4.3	84.3	4.2	1.1	2.1	2650	27500
Biomass	Bark	2.6	90.6	6.8	0.02	42.5	5.5	0.5	0.0	2660	18200

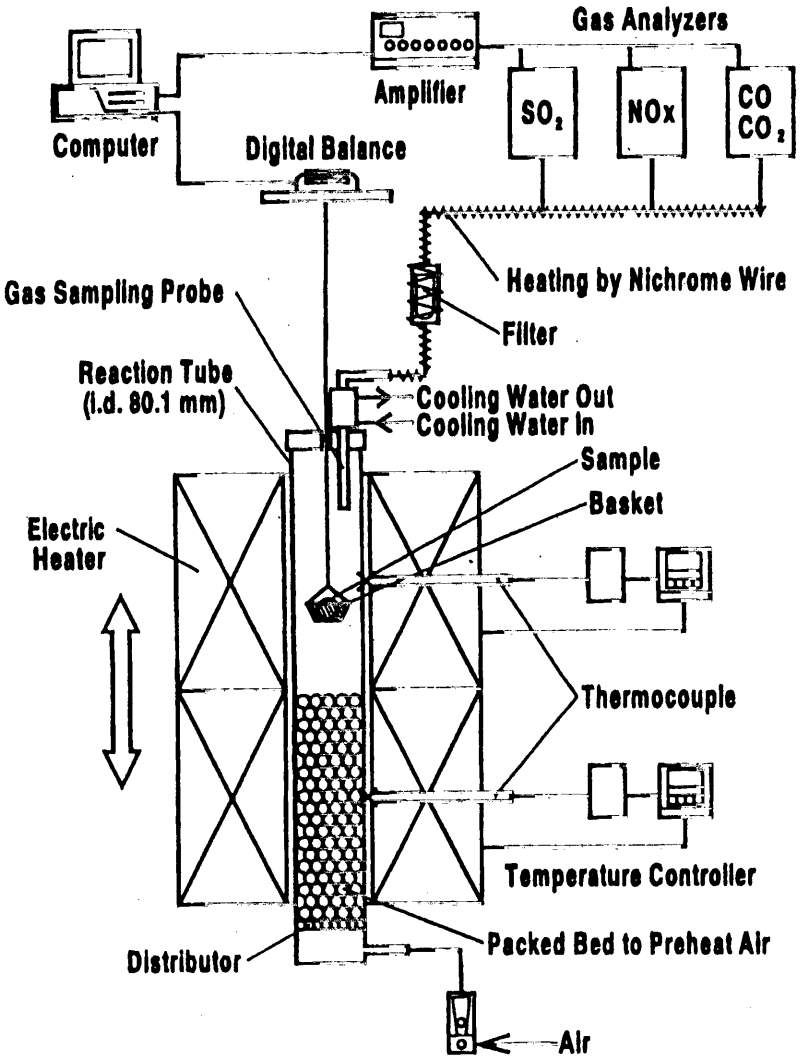

Figure 1 Experimental apparatus

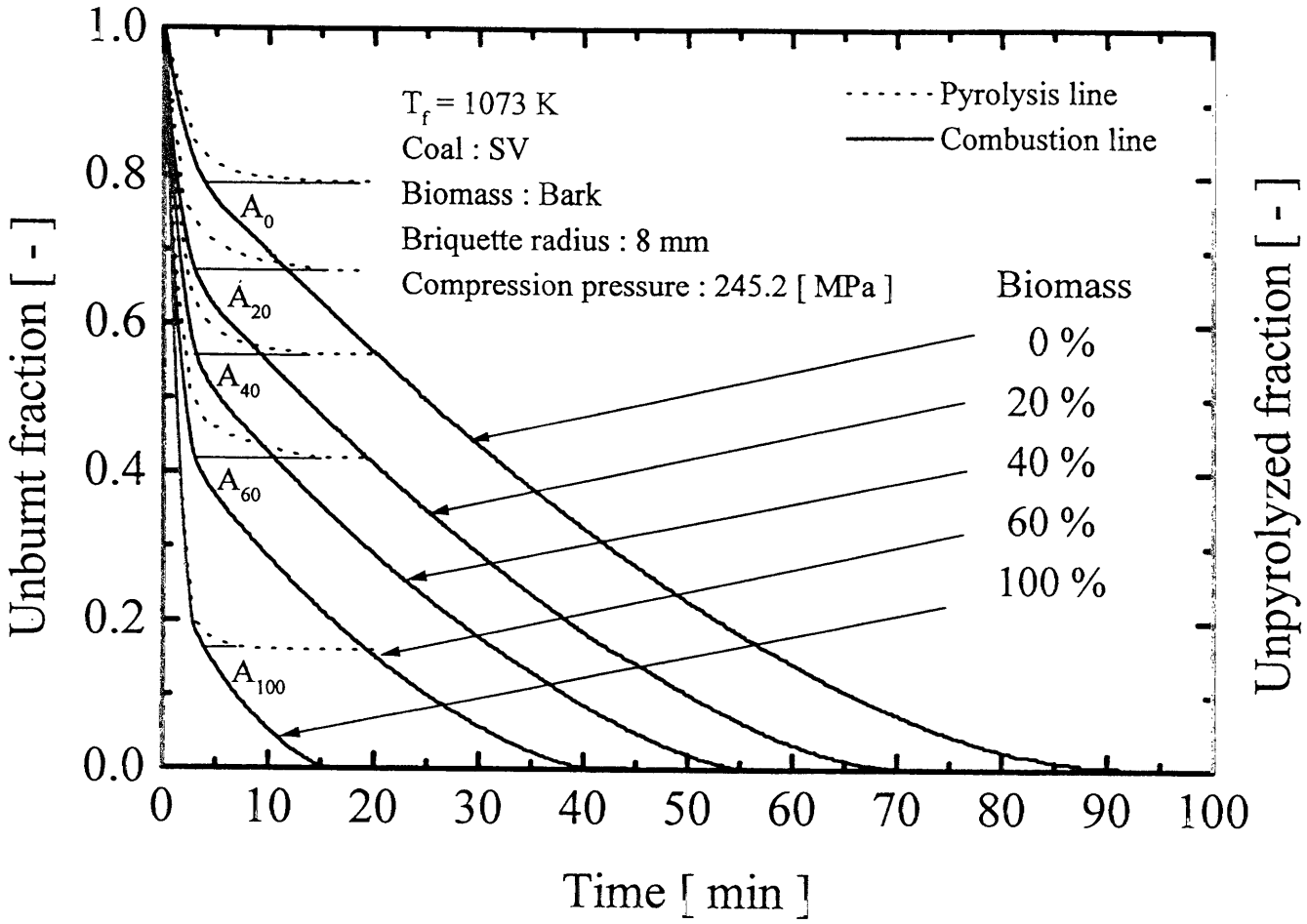

Fig.2 Influence of biomass content on burning profile

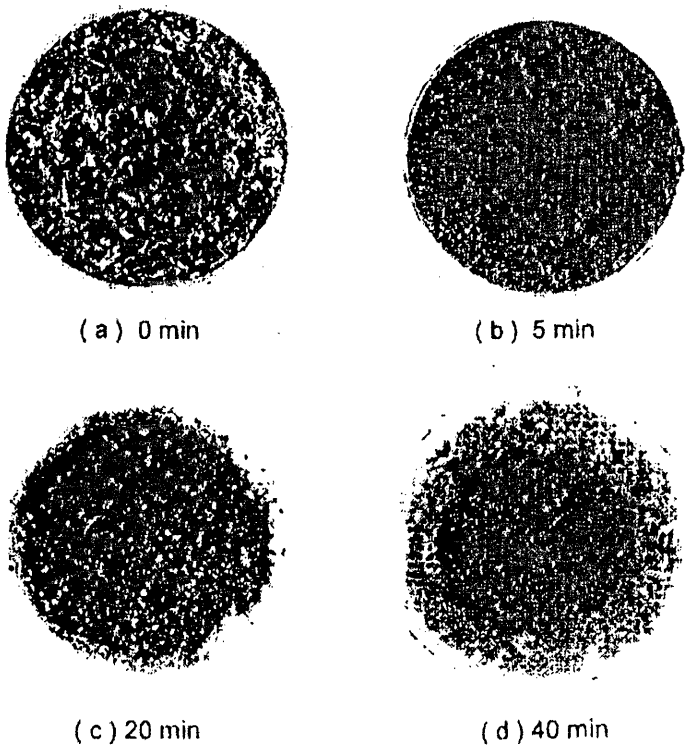

Figure 3 Change of cross-sectional structure during biocoalbriquette combustion

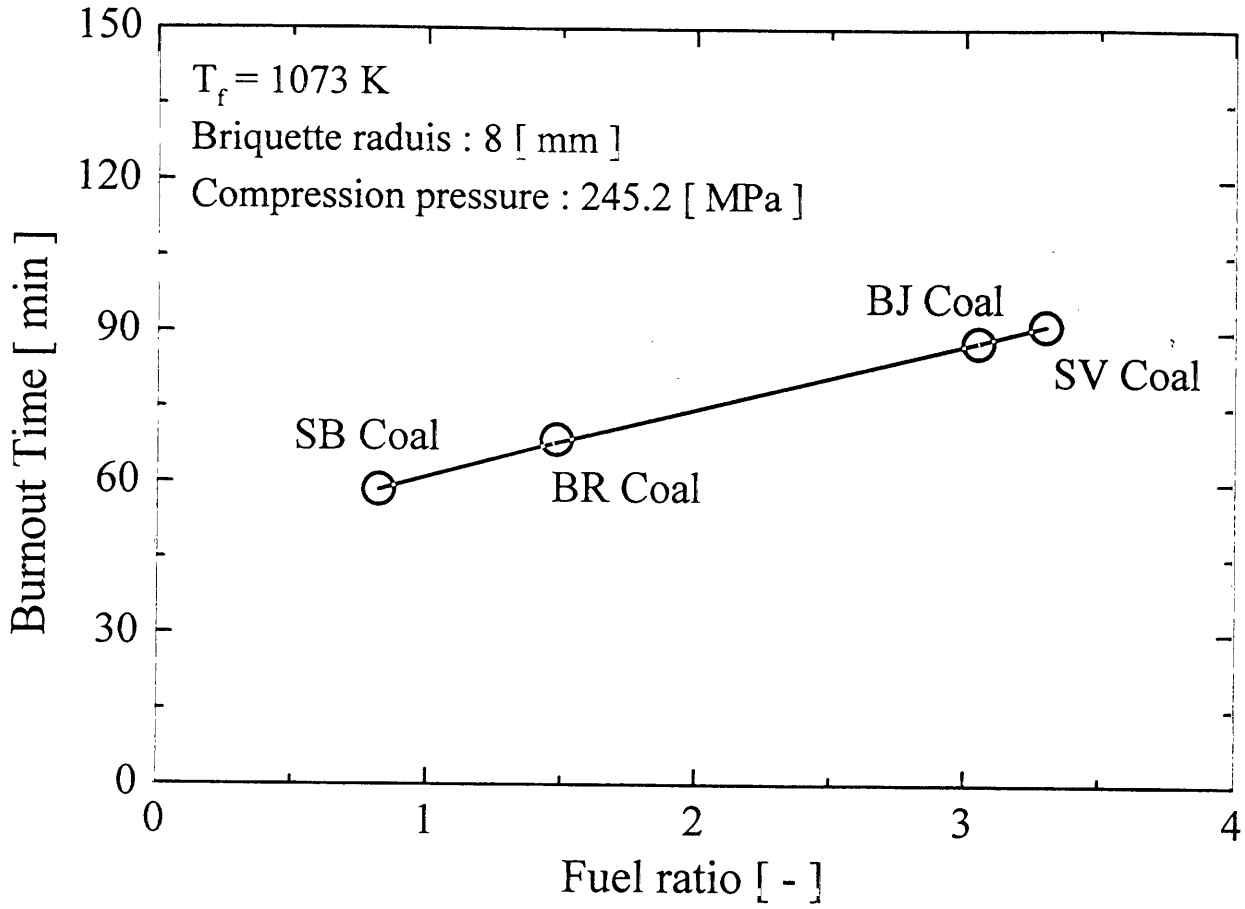

Fig.4 Influence of coal type on burning out time

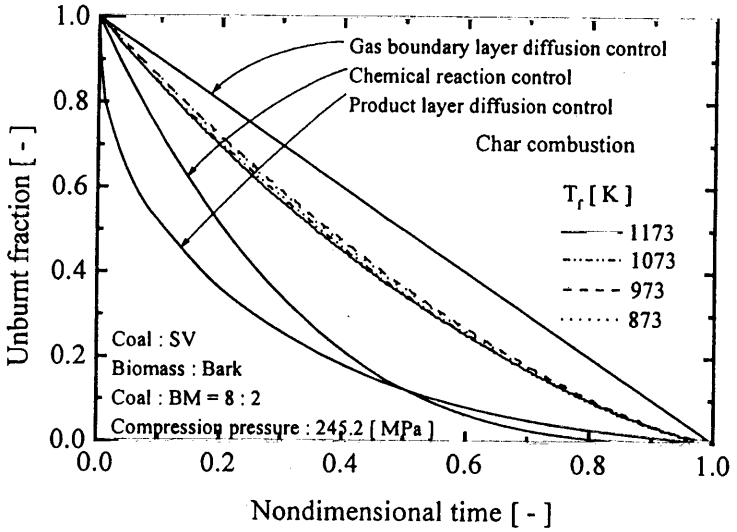

Figure 5 Normalized combustion profiles under different furnace temperatures

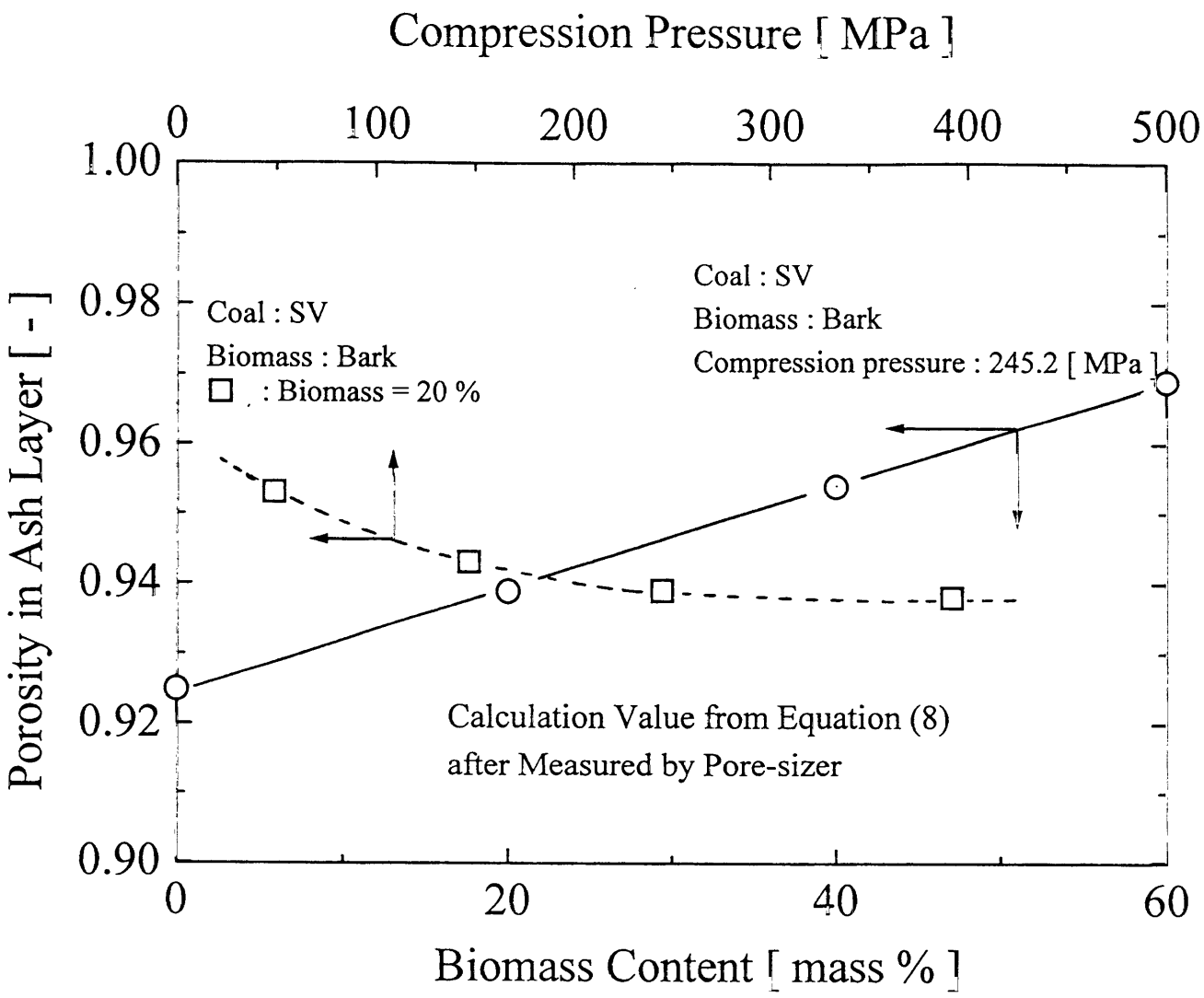

Fig.6 Porosity in ash layer as functions of biomass content and compression pressure

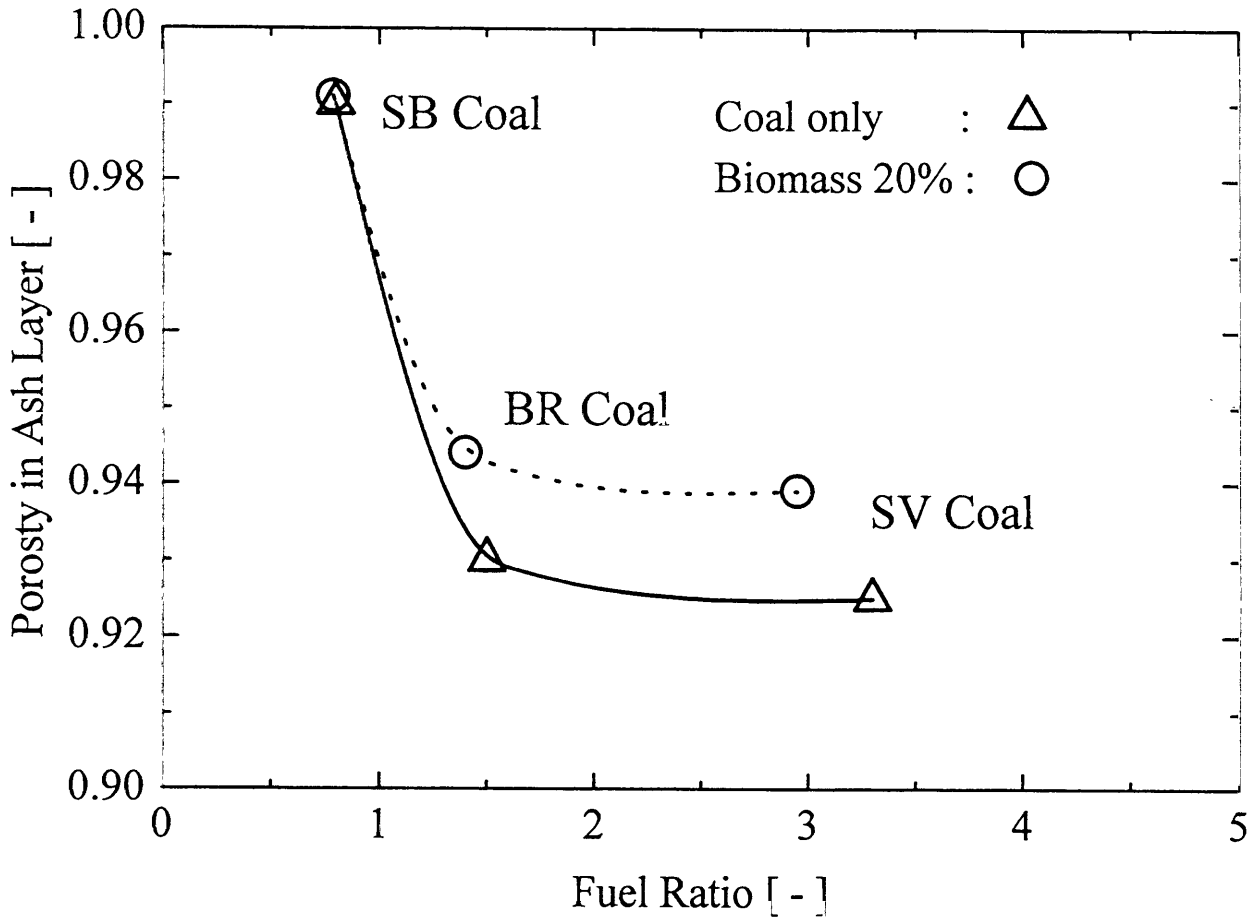

Fig.7 Porosity in ash layer vs. different coal type

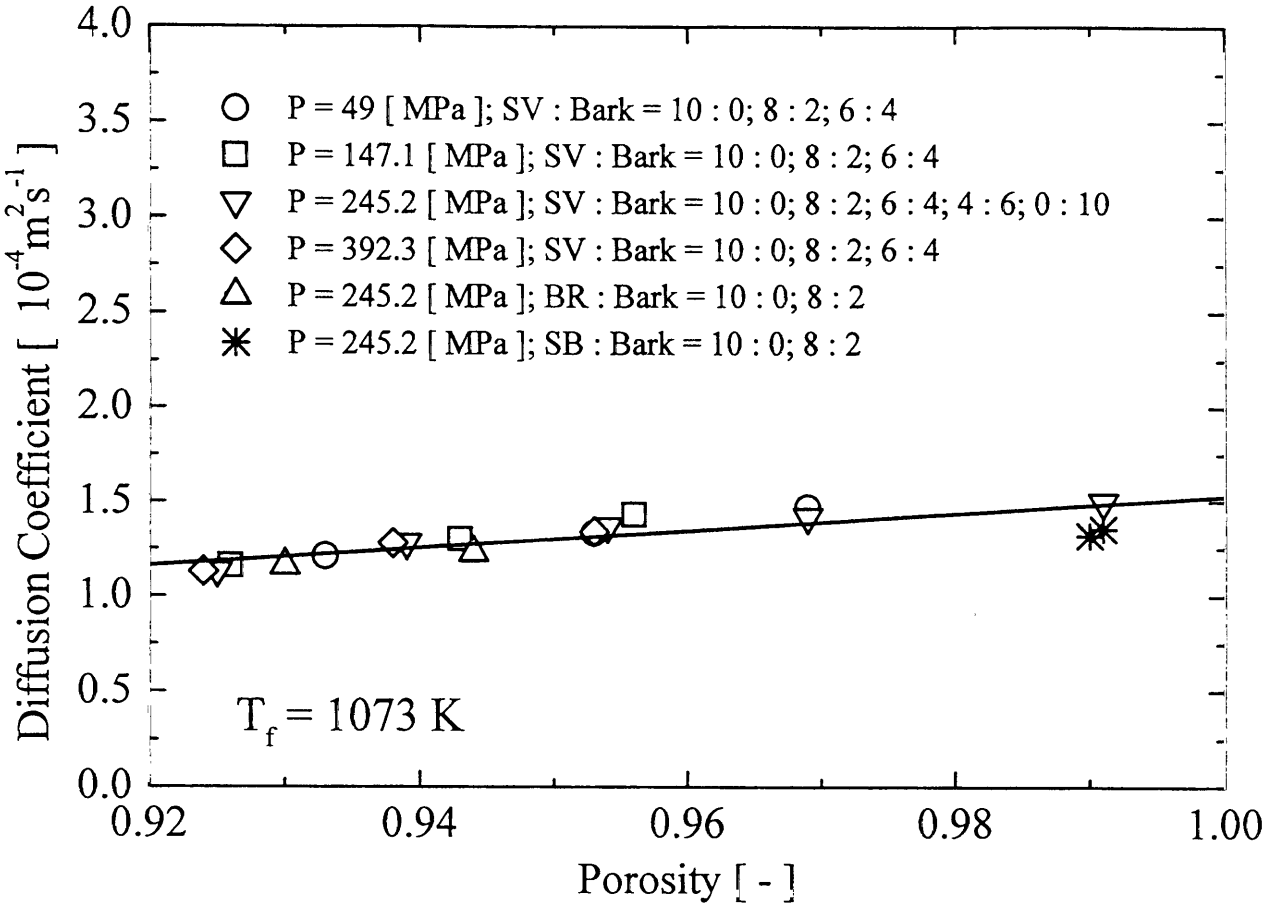

Fig.8 Correlation between diffusion coefficient and porosity at various experimental conditions

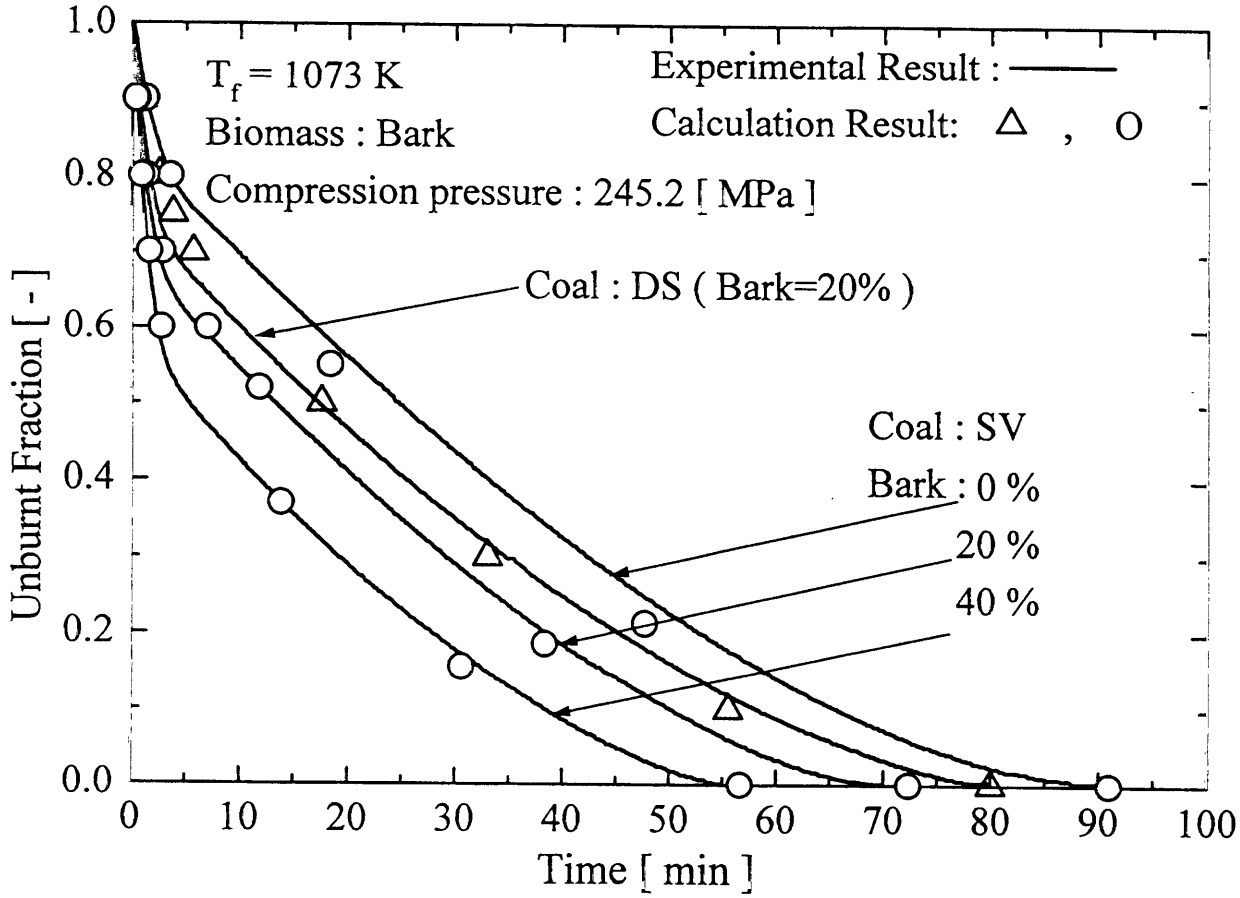

Fig.9 Comparison of calculations and experiments