C -4. 1. 3 Studies on Integrated Control Techniques for Precursors of Acid Precipitation in South-eastern Region in China

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[Abstract] Many regions in China are suffered from serious acidic precipitation caused by the combustion of coal. In this study, we are introducing the effective techniques in Chongqing district as typical case, to reduce the emission of sulfur dioxide and other components by coal combustion. That is "coal-biomass briquette", which is produced from pulverized-coal, biomass such as straw, and slaked lime as desulfurizing agent, under high pressure (3-5 ton/cm ^{2}).

From the field surveys in Chongqing, we gathered the data on physico-chemical properties, available amout and cost for the raw materials of coal-biomass briquette. The expecting reduction rate of the emitted SO2 was 80-90% in the preliminary test. We introduced the coal-biomass briquette technology including manufacturing and uses into the district and confirmed the high sulfer fixation, the high energy efficiency and the the versatility of coal-biomass briquett. At the same time, the evaluation systems of flue components from the cooking stoves for briquette or charcoal were developed and the emission factors for SO2, NOx, CO and CH4 for the combustion of briquette were calculated in some cases.

Key Words Desulfurization, Improvement of fuel, Coal, Briquette, Flue, Chongqing.

1. Introduction

Chongqing district in south-western China is one of the most polluted area with SO2 and dust which emitted from the local combustion sources of low graded raw coal (Sulfur content=ca.2~6%) for the use of domestic and industrial activities. These pollutants currently causes serious acidic deposition, conspicuous human damage, and various kinds of environmental deterioration.

In this study, we introduced an effective technique called "coal-biomass briquetting process" in Chongqing as a typical case, to reduce the SO2 emittion caused by the small many combustion sources of low graded raw coal. The "coal-biomass briquette" are produced from pulverized-coal, biomass such as various woody wastes and agricultural wastes (e.g. barks, sawdust, bagasse, beet pulp, rice husk etc.), and slaked lime as a sulfur fixation agent, under high roll line pressure (3 ~ 5 ton cm-1). The briquetting process, combustion characteristics and its sulfur fixation efficiency of the briquette and its applicability as one of the integrated control techniques against acidic deposition are chiefly discussed, on the basis of the testing results.

After the introduction of coal- biomass briquetting technology including manufacturing and uses into the district, the designs for coal burning test stoves and a blueprint of a manufacturing plant's specifications were set about by the local scientists and engineers with Japanese side assistances. The concurrent technology transfer of the techniques to Chongqing or China is also the important targets of this study. The sulfur fixation and energy efficiencies, briquetting and combustion characteristics of coal-biomass briquette are chiefly discussed.

2. Experimental

2.1 Emission of gaseous pollutants from the combustion of briquette.

Experiment of combustion follows the method of already reporte. As fuel, we used one kind of Japanese briquette and four kinds of Chinese one. Flue gas is gathered by the hood above the cooking stove, and sucked into the duct and analyzed (see Fig. 1). The components of flue gases (CO, CH_4, NOx, SO_2, etc.) were measured by Micro-sensor Gas Analyzer (Model 200, made by Nippon Tylan) and Combustion Gas Analyzer (Type GA-60, made by Madur Elect.) and Multi-Gas Monitor (Type 1302, made by Bruel & Kjaer) and NOx meter (Model 42S, made by Thermo Electron). The flow velocity of flue gas in duct was measured by pitot tube and precise differential manometer. The temperature of flue gas and glowing briquette were measured by thermocouple.

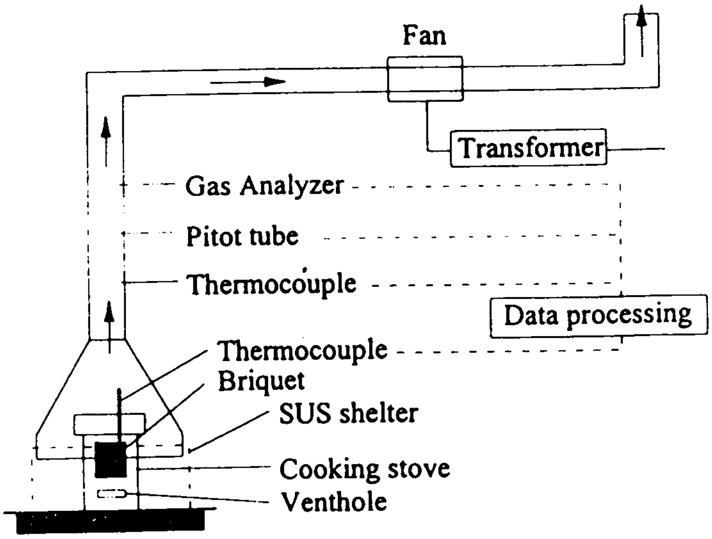

Fig 1. Combustion test system

2.2 Trial production of coal-biomass briquette

With a view to developing coal-biomass briquette production techniques, a testing machine was installed in Hokkaido Industrial Research institute. This machine is a small-size roll press for high pressure briquetting (3 ~ 5 ton cm-1). In this test machine, the raw material mixtures same as tableting tests is fed forcedly into between rolls by a vertical screw. Low graded raw coal samples (S% = ca. 3) used for test production are selected from the largest Chongqing Nanton collierys. Biomass such as sawdust, kaoliang (Chinese sorghum), corn stalk or husk, bagasse and rice husk were collected at Chongqing rural sites. Up to now, this machine has been used for investigating the briquette-ability of various Chongqing run of mine coals. They are Nanton coal, Tianfu coal, Sonzao coal and so on.

In the briquetting test for Chongqing coals, the coals were used without any coal cleaning techniques, but sulfur fixation agents were added in these coals, together with biomasses. In FY1996, during the adjustment of briquetting machine installed in demonstrating plant under construction in Jiangbei District of Chongqing, preliminary testing production of coal-biomass briquettes were carried out for both of coal-biomass briquettes (Japanese combination) and the improved coal briquettes (Chongqing's combination).

2.3 Breaking tests for coal-biomass tablet and briquette

Tableting tests were carried out on coal-biomass particle mixtures for basic study of their briquette-ability from the relation of briquetting conditions, breaking strength and density of tablet obtained. Low graded raw coal samples from the Nanton collierys and biomasses such as sawdust and wood dust were used for the test. Tableting test was carried out with a small cylindrical mold (d = 25 mm), where 3 g of each mixed coal-biomass sample was placed (See Fig.2).

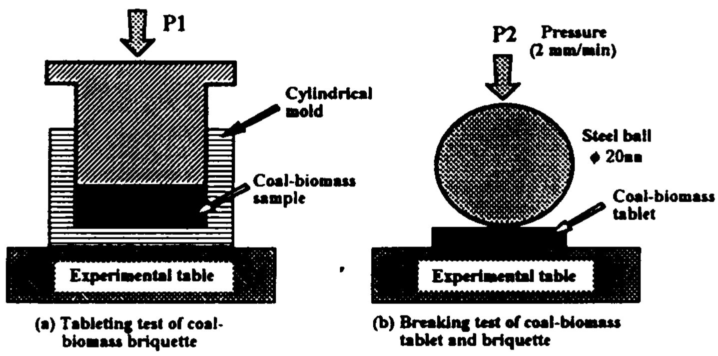

Figure 2. Tableting and breaking tests.

2.4 Burning tests and questionnaire survey for coal-biomass briquette

In order to investigate components and characteristics of coal-biomass briquette, burning testing with a domestic stove and chemical analysis were performed, and microscopic observations was conducted on an internal texture of coal-biomass briquette. Sulfur emissions were calculated from content in combustion residues and total sulfur in test fuel when sulfur fixation agent (e.g. $Ca(OH)2$, CaO or $CaCO3$) were added. In addition, the preliminary questionnaire survey on the use of the coal-biomass briquette was carrid out to the inhabitants of Jiangbei District, Chongqing. At the same time, several tests for the estimate of the reduction rate of $SO2$ in the combustion of coal-biomass briquette were performed by Sichuan prefectural professional institution. Combustion behaviors of coal-biomass briquette and raw coal were also determined.

3. Results and discussion

3.1 Emission of gaseous pollutants from the combustion of briquette.

The main row material of briquet is coal which is most abundant fossil fuel and distributed all over the world. Most advantage of the briquette use is easy to control the combustion and to keep high temperature for long time(up to 50 hours for one briquette unit). However, the combustion of briquette causes sometime air pollution in houses or in town. In this study, we constract the experimented systems for the estimate of emission of gaseous pollutants from the cooking stoves, and calculate the emission factors of CO, CH_4, SO_2, NOx for the combustion of briquette in some cases.

The emission factor of CO was 100-200mg/g and the highest amount in pollutants emitted from combusted briquette even if air supply was rich. CH_4, SO_2, NOx emission factors were nearly equal, and under 15 mg/g. Inside the combusted briquette, CO was richer than CO_2 and the CO concentration was at several %, however the CO level was immediately reduced to ppm order after exposure to the air. This phenomenon shows that CO is formed coal particle in burning briquette at first and it is oxidized to CO_2 with oxygen outside of fuel, that is, the two steps combustion process is occured.

Table 1 Emission factor for each compound [mg/g·fuel]

Fuel	Air supply	Compound			
		CO	NOx	SO_2	CH_4
Japanese briquet	lean	61-141	0.17-8.0	1.1-5.0	0.80-2.5
	rich	46-120	0.41-2.0	2.4-7.2	0.46-6.2
Japanese charcoal	lean	92	0.76	1.2	0.55
	rich	103-322	0.64-1.4	0.67-0.89	0.42
Chinese briquet	rich	74-93	2.4-2.6	5.3-9.3	0.13-7.8
Reference					
Coal (Power plant)		*0.00023**	*4.37***	——	——
Coal (Domestic)		*0.023**	——	——	——

**Air quality criteria for carbon monoxiide. (1972)*
***J.E.A. Tissosankabutusouryoukiseimanuaru.(1992)*

3.2 Tablet-ability of coal-biomass mixture

The coal-biomass briquette has a strength enough to withstand ordinary handling without using any special binder. This is because its biomass may act as a binder (Maruyama et al.; 1980 and 1985) . This effect may also be expected for many biomasses such as bagasse, beet pulp, rice husk, and wood waste.

Table 2 shows the results of tablet test conducted by using various kinds of biomasses. All of biomasses are capable of acting as a binder, but the bagasse shows especially excellent chara- cteristics as a binder. The breaking strength of coal- biomass tablets increased with increasing ratio of biomass, while decreased with increasing grain size of coal. Each of the Chong- qing coals is different in briquette-ability, but any coal-biomass briquette shows the breaking strength of 64 kg at least or more. Thus, coal- biomass briquette has a satisfactory strength without addition of any special binder.

Table 2. Properties of Chongqing coal-biomass tablets.

Coal/Biomass§	Biomass (wt.%)	Breaking strength (kg)
Nanton coal/sawdust	10	64
Nanton coal/sawdust	20	114
Nanton coal/sawdust	30	239
Nanton coal/wood dust	10	90
Nanton coal/wood dust	20	160
Nanton coal/wood dust	30	300

§ Particle size of wood dust < 2mm, particle sizes of raw coal and sawdust : < 3 mm.

3.3 Analytical results of raw coals and biomasses

Table 3 shows the results of chemical analysis of biomasses (corn stalk, sawdust and kaoliang), three kinds of raw coals and their coal-biomass briquettes from three main collierys in Chongqing. As is clear from this table, all of the Chongqing coals contain a high percentage of combustible sulfur that may cause air pollution. The constituents of the combustible sulfur is mainly inorganic sulfur such as pyrite (FeS_2), and partly organic sulfur bound coal substance. The mineral sulfur of the former type is, unlike other inorganic substances such as clay mineral and quartz, hardly removed by the ordinary coal cleaning because it exists as small particles and disperses in Tianfu coal substance.

Table 3 Analytical results of raw coals, biomass and coal-biomass briquette (Biocoal)※ of Chongqing.

Item \ Coal/biomas	Tianfu coal		Nanton coal		Sonzao coal		Biomasses※※※		
	Coal	Biocoal	Coal	Biocoal	Coal	Biocoal	Corn stalk	Sawdust	Kaoliang
Moisture (%)	1.2	2.0	3.1	3.3	1.2	2.0	7.3	6.5	7.7
Ash (%)	22.1	23.4	30.2	30.9	14.4	17.6	6.3	0.9	14.9
Volatile matter (%)	17.5	28.2	17.0	27.2	14.6	26.0	82.4	91.0	73.8
Fixed carbon (%)	59.2	46.4	49.7	38.6	69.8	54.4	4.1	8.1	3.6
Calorific value (kcal/kg)	6,510	5,680	5,600	4,890	7,180	6,190	4,200	4,440	4,200
Total sulfur (%)	2.40	1.89	3.30	2.69	2.50	2.05	0.70	0.07	0.26
Incombustible S (%)	0.73	1.61	0.40	2.39	0.21	1.68	0.08		0.05
Combustible S (%)	1.67	0.28	2.90	0.30	2.29	0.37	0.62		0.21
HGI※※	95		122		114				

※ Coal-biomass briquette (Biocoal) : Raw bituminous coal (Coal) 80 wt.%, bagasse 20 wt.%, while sulfur fixation agent (Ca/S = 1.5) was added.

※※ The hardgrove grindability index (HGI) may be taken as one of the indicators for the breaking strength of coal briquettes. If HGI > 60, it is considered that this kind of coals will be easily briquetted.

※※※ Particle sizes of raw coal and biomasses : < 3 mm.

3.4 Sulfur fixation efficiency and analytical results of coal-biomass briquette

As shown in Table 3, the deduc- tions of combustible sulfur of Nanton, Tianfu and Sonzao coals were deter- mined, which were 0.30/2.90=10.3%, 0.28/1.67=16.8% and 0.37/2.29=16.2%, respectively, when $Ca(OH)_2$ (Ca/S=1.5) were added. It is expected from this results that reduction rate of the emitted SO2 will be 80~90% by the coal-biomass briquette technique.

From the test by Sichuan prefectural professional institution, it is known that the breaking strength of coal-biomass briquette (Japanese combination) is higher than that of the improved coal briquettes (Chongqing's combination) (Table 4). Both of these briquettes show good sulfur fixation efficiencies and the SO2 reduction rates of which are over 85%. It should be emphasized the excellence in ignition-ability for Japanese combination and in waterproof property for Chongqing's combination.

Table 4 Initial results of testing production.

(1) Coal-biomass briquette (Japanese combination)	
Sawdust	~25%
Zhongliangshan coal (S%=3.8)	~75%
Sulfur fixation agent : $Ca(OH)_2$	Ca/S = ~1.5
Weight	~30 g
Breaking strength	50~90 kg
Reduction rate of SO_2	~85%
Excellent in ignitability	
(2) Coal briquette (Chongqing's combination)	
Binder	10~20%
Zhongliangshan coal (S%=3.8)	80~90%
Sulfur fixation agents	Ca/S = ~2.0
Weight	~28 g
Breaking strength	40~50 kg
Reduction rate of SO_2	~90%
Waterproof	

Particle sizes of raw coal and sawdust : < 3 mm.

3.5 Mechanism of coal-biomass briquetting process

From microscopic observation, we found a specimen of the internal texture of coal-biomass (woody) briquette obtained by polishment. It is found that both coal and biomass grains are oriented in a direction and coal grains in a surface part of coal-biomass briquette are finer in its sizes. The voids among coal grains are more compact with biomass grains, where seem to be plastic-deformed in briquetting process. It can be deduced that the consolidation of coal-biomass mixture in the roll press takes place under compressive force with shearing strain. This implies that coal grains, while being partially broken, are subjected to plastic deformation together with biomass grains and that, in the end, the biomass grains having received a large plastic deformation act as the binders of coal-biomass briquette at the compression stage.

Thus, the biomass constituent serves as binder in coal-biomass briquette. This effect has been recognized in biomass fines such as baggase, sawdust, Kaoliang, corn stalk or husk, rice husk, beet pulp and so on, having a rheological property similar to wood fines. Therefore, these biomasses can be expected to use as raw materials for coal-biomass briquette production. The microscopic observation of the internal structure of coal-biomass briquette shows the important role of biomass as binder on briquetting process.

3.6 Grind-ability and briquette-ability of Chongqing coals

In our study, fine coal particles are observed on the surface coal-biomass briquette. This fact tells that unlike the biomass showing large plastic deformation, the coal particles have partially been broken in the briquetting process. In case of the highly pressed briquetting, it is therefore assumed that the briquette-ability of coal is greatly influenced by the coal strength. The hardgrove grind-ability index (HGI) can be taken as an indicator for the breaking strength of coal because it shows a deep correlation with the breaking strength of coal particles. The higher value of HGI mean more briquettable. Nanton coal and Tianfu coal of Chongqing which have higher HGI were mainly used in production of a coal-biomass briquette in this study. The results show that the HGI can be used for the actual indicator in briquette production (Table 3).

3.7 Burning test of raw coals and coal-biomass briquette

The combustion test of coal-biomass briquette with a commercially available domestic coal stove in Chongqing shows that the use of coal-biomass briquette results a very low smoke density and little smell over a wide range of combustion load during combustion. Figure 3 shows the variation of water temperature within the testing pan which were observed upon combustion test for on the raw coal and coal-biomass briquette.

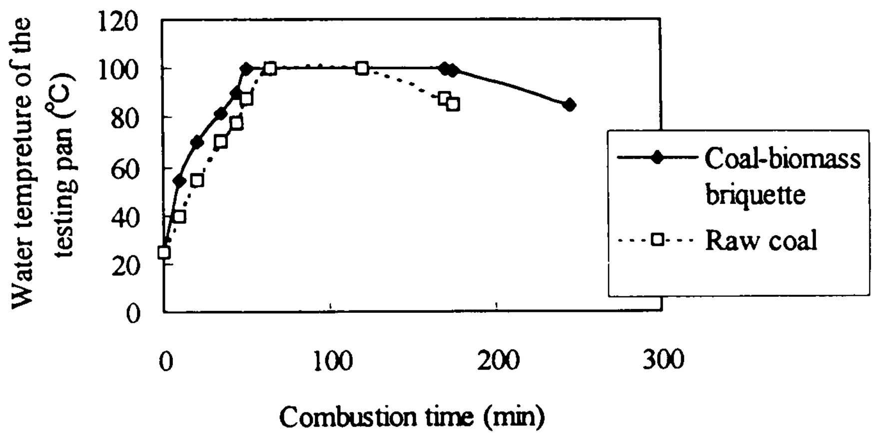

Figure 3 Combustion process of raw coal and Coalbiomass briquette.

Table 5 Combustion behaviors of coal-biomass briquette and raw coal.

Parameters and/or items	Symbol	Unit	Coal-biomass briquette	Raw coal
Time till ignition 1) (ignitability)	T_D	min	13	20
Time till burning	T_S	min	36	45
Flame holding duration	T_W	min	120	51
Cookable duration	T_K	min	212	133
Total combustion duration	T_R	min	248	178
Total steam generation 2)	D_{zz}	g	4,720	2,920
Heat efficiency 3)	η	%	25.5	20.5
Average heating power	H_P	$g \cdot min^{-1}$	22.2	21.9
Average entrance temperature of the cooking stove	t_K	℃	586	
Average wall temperature of the cooking stove	t_b	℃	95	91.2

1) 0.55 kg of ignitable materials were applied for the ignition.
2) 5.98 kg of water for boiling.
3) The heat efficiency was very low because the Chongqing's cooking stove with low heat efficiency was used in this burning test.
4) Both 3.0 kg of coal-biomass briquette and raw coal were used for the test.

As is clear from the results of Table 5, the coal-biomass bri- quette shows higher ignition-ability and longer periods in com- bustion and heating duration than the raw coal. This is an essential property for combustion control in a small-sized stove. At the same time, low smoke density from the combustion of coal-biomass briquette, as compared with the raw coal, is due to the effect of the addition of biomass and the smaller particle size. The smoke generated from combustion of coal-biomass briquette with primary air may be burnt completely with secondary air. It is clear from this study that the use of the coal-biomass briquette can reduce the emission of dust and sulfur, as compared with its raw coal.

3.8 Development of special coal-biomass briquette combustors

Coal stoves attached storage room are suited for the coal-biomass briquette. However, it is advantageous in that its combustion efficiency is about 60 % at the highest. Therefore, the development of special made combustors for coal-biomass briquette must be promoted so as to be fit for the characteristics of coal-biomass briquette. The new type combustor madem for trial, which is a kind of space heater, is in the basis on the way in which the coal-biomass briquette is burnt less with primary air and mainly with secondary air. As a result, significant improvement of heat efficiency can be achieved with little smoke generated over a wide range of combustion load.

4. Conclusions

The coal-biomass briquette is a kind of composite solid fuel having excellent combustion characteristics which can be summarized as follows:

a) The emisson of smoke and SO2 can be significantly reduced by mixing some biomass and sulfur fixation agents in it.

b) The property can be improved such as easy ignition, higher combustion effeciency and easy combustion control.

c) The clinker formation during combustion is rare, thus the ash could be disposed easily.

d) It generates less dust during handling and carrying because of the surface viscosity.

The reduction effects of the fuel improvement on the emission of materials which are caused acidic precipitation and air pollution was made clearified from the experiments and field survey in the laboratory and the actual plant constructed in Chongqing, Chaina. In the second stage, the construction of the bigger production plant for coal-biomass briquette and the social system for use it will be key. We shall continue to support the concurrent technology transfer of coal-biomass briquetting technique to Chongqing or other district in Chaina.