C-3.1.3 A Study on Control Techniques for the Emission of Acid Precipitation Precursors from the Stoves for Domestic Uses

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Abstract. The evaluation systems of flue gas components from the combustion of coal briquette or charcoal in cooking stoves were developed. The emission factors (EF) of SO2, CO, NOx and CH_4 were calculated from the combustion tests of coal briquette using at some cities in China and in Japan. EFs of CO from the briquette combustion were 40-150 mg/g-fuel, which is extremely high compared to 2-3 ug/g for coal combustion in power plants. The EF of SO_2 for the briquette in Chongqing district, of which the S content is 2.6%, is 59 mg/g-fuel. The SO_2 emission factors for others were similar and comparatively low (0.9-15 mg/g-fuel). EFs of NOx and CH4 are 0.4-2.6 and 0.4-6.2 mg/g-fuel, respectively.

The emission of CO could be reduced to below 20% by putting of a ceramic disk above the briquette with 2-3 cm space. For reduction of SO_2 emission, we tried three techniques, that is, (a); putting lime layer on mesh [or (b); a lime disk] above briquette with ca. 1 cm space, (c); adding lime powder to coal powder and mixing to form a flat-ball of briquette. The results shows that (c) is most effective and the reduction rate of SO2 emission was more than 85% with 5-10%(w/w) lime addition, (a) is second and the rate was 45-60%, and (b) did not show obvious reduction effect.

Key Words; control technology, stove, domestic uses, coal, briquette, flue, China

1. Introduction

The main row material of briquette is coal which is most abundant fossil fuel and distributed all over the world. Advantages of the briquette use are easy to control the combustion and to keep high temperature for long time (up to 50 hours for one briquette unit). Therefore, it is expected to use more in developing countries where is suffering from the luck of domestic fuel and had to leave the remained forest in countryside.

However, the briquette combustion produces lot of pollutants. The air in rooms using briquettes is especially polluted by CO and dust, in addition any ventilation systems are not used in many cases in developing countries. Also, the combustion of briquette cause air pollution in town.

In this study, we construct 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, and developed some reduction techniques for the emission of CO and SO_2.

2. Experimental

2.1 Material and apparatus

The material for combustion experiments were collected from Japan and China. They are shown in Table 1 with the results of element analysis. Combustible carbon (C), hydrogen (H), nitrogen (N) were analyzed by a CHN Analyzer, and total sulfur by JIS M 8818, and ash by JIS M 8812.

Experimental methods for combustion test were already reported[1]. Flue gas is gathered by the hood above the cooking stove, and sucked into the duct and analyzed. The components of flue gases were measured as follows, [N_2, O_2, CO_2, CO, CH_4]; Microsensor Gas Analyzer (Model 200, made by Nippon Tylan), SO_2; SO_2 Analyzer (Model 43C, Thermo-Electron Co.), and NOx; NOx gas analyzer (Model NOA-7000, Shimadzu Co.). 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 monitored by thermocouple. All of them were recorded by computers and a recorder.

2.3 Estimation of emission factors from the combustion experiments.

The emission factors (EF,w/w) for each compounds emitted from the fuel combustion were estimated from the following equations (1)-(4).

$EF1=[\Sigma (T\cdot Cn\cdot F)\cdot Mnw]/[V\cdot Wt]$ -----(1)
$EF2=[\Sigma (T\cdot Cn\cdot F)\cdot Mnw]/[V\cdot Wb]$ -----(2)
$EF4=[\Sigma (T\cdot Cn\cdot F)\cdot Mnw]/[V\cdot Wc]$ -----(3)
$EF3n=[Cn\cdot Mnw]/[C_{CO2}\cdot 12]$ -----(4)

Where;

T ; Period fot the process. Cn ; Concentration of a n-component.
C_{CO2} ;CO2 concentration in flue gas. F ; Flow rate of flue gas.
Mnw ; Molecular weight of n-component V ; Volume of 1 mol.
Wt ; Weight (dry base) of fuels. Wb ; Weight of ash or residue after combustion.
Wc ; Burned carbon amount estimated from the concentration of CO2 in flue gas.

These (1)-(3) equations are for emission factors based on (1); total fuel, (2); combustion material, (3); burned carbon amount. Equation (4) is a EF for the every time of a measurement.

If we could maintain the period for the process (T) and the flow rate of flue gas, eq.(1)- (4) is simplified as eq.(5).

$EF1=Ta\cdot F\cdot Mnw\cdot Anv/[V\cdot Wt]$ -----(5)

Where;

Ta ; Period of the start to end of the combustion of fuel.
Anv ; Mean concentration of n-components.

Table 1-1. Elemental contents of fuels mode in Japan, Unit;%

Fuel*	Combustible			Total	Ash
	C	H	N	S	
Charcoal; N= 4					
Mean =	82.00	1.82	0.34	<0.01	5.2
S.D.=	1.77	0.32	0.15	--	--
Bio-briquette	67.40	4.15	0.57	0.14	12.8
Normal-briquette	47.10	1.55	0.95	0.45	27.5
Chakka-briquette*-1					
Upper part	62.50	2.60	0.73	0.22	14.0
Lower part	52.70	1.59	0.58	0.45	26.5
Chakka-briquette-2					
Core part	67.40	1.45	0.80	0.59	25.1
Peripheral part	70.80	1.93	0.81	0.33	17.8
Ippatsu-briquette**					
Ignition part	54.70	1.99	4.08	<0.01	28.7
Body part	48.60	1.58	0.48	0.42	25.0

* Chakka-briquette ; In order to get easily fire, the compositionof the upper part of a briquette are changed.

** Ippatsu-briquette ; It has a ignition part on the top.

Table 1-2 Elemental contents of fuels mode in China, Unit;%

Fuel*	Combustible			Total	Ash
	C	H	N	S	
Sian bri. -1	43.20	0.42	0.48	0.38	--
Sian bri. -2	63.90	1.84	0.80	0.53	32.3
Peking bri.-1	59.30	<0.01	0.25	0.24	--
Peking bri.-2	61.40	<0.01	0.22	0.19	--
Ch'angsha bri.	44.00	0.88	0.63	0.60	--
Chongqing bri.	46.80	2.07	0.68	2.60	43.5
M.coal*;Tianfu	59.20	--	--	2.40	22.1
Nanton	49.70	--	--	3.30	30.2
Sonzao	69.80	--	--	2.50	14.4
Chengtu n= 5					
Mean =	49.48	2.4	0.59	0.32	40.2
S.D. =	3.07	0.27	0.16	0.05	1.9

Note, * ; Raw coal for briquette.

3. Results and Discussion

3.1 Fuel property

The fuels used in this study are listed in Table 1-1 and 1-2 with the elemental contents. The total amounts of them are below 100%. The rest of the sum could be water which seemed to be connected with mineral components and was not removed by the dry process with a desiccater.

The contents of combustible hydrogen and nitrogen were low and in similar level in each briquette. However, the sulfur contents were changed much, especially the briquette from Chongqing showed high level.

The ash amount of briquettes are abundant, especially Chinese briquettes are exceeded 30% and some ones reach 43%, although Japanese ones are around 25%. The amount of ash in charcoal is low. Table 3 show the original fuel weight and the residue after combustion. The original weight of Japanese briquette are heavy compared to the Chinese ones, of which two or three pieces of them are used together in a stoves to maintain combustion for longer time with sequential addition.

The charcoal was burned out mostly, only 4% was remained after combustion as shown in Table 3. However, in the case of the briquette from Peking, only 40% was burned. Especially the "flat ball" briquette, which was made from muddle grain of briquette with water, showed the low efficiency in combustion. Generally Chinese briquettes are weak in the physical strength and they are easily broken during shipping to Japan. Then, they were used in the experiments as the flat balls of briquettes.

Table 2-1 Elemental contents of fuel ash after combustion experiments, Unit; %

Fuel*		Combustible			Total	Ash
		C	H	N	S	
Japanese Charcoal	Ash-1	7.96	0.20	<0.01	0.63	59.4
	Ash-2	12.40	<0.01	<0.01	0.61	--
	Ash-3	22.00	<0.01	0.01	0.24	--
Japanese bio-briquette		1.24	0.13	<0.01	0.72	95.4
Ippatsu-bri. n= 7						
	Mean =	0.92	--	--	0.95	97.7
	S.D.=	0.43	--	--	0.22	0.5
Sian bri. n= 8						
	Mean =	5.91	--	--	0.31	87.9
	S.D.=	4.81	--	--	0.17	3.9
Peking bri. n=4						
	Mean =	8.36	--	--	0.27	84.4
	S.D.=	4.15	--	--	0.08	6.3
Ch'angsha bri.Ash		2.60	<0.01	<0.01	0.23	--
Chongqing bri.Ash		15.80	0.02	0.10	0.58	85.5

3.2 Ash

The elemental amounts of ash of various fuels show in Table 2. The carbon content in charcoal ash is high, however the actual amount of carbon in the ash is little because the little amount of ash. The ash of Chinese briquette contains much unburned carbon, then the amount of waste results in increase. The much amount of ash in briquette cause the loss of energy and efficiency in each process such as carrying, storage, combustion and waste.

Table 2-2 Element contents of flat ball briquette after the adding of slaked lime into briquette grains, Unit; %

Fuel*	Combustible			Total	Ash
	C	H	N	S	
Mixing ratio of slaked lime into briquette, %					
0	8.80	0.30	0.05	0.21	88.9
2	7.20	0.20	0.05	0.67	89.8
5	5.10	0.10	0.05	0.84	91.8
10	6.20	0.10	0.05	0.97	89.6

Note; Flat ball briquette.: Pieces of briquette were broken into grains and certain amount of slaked lime was added to them and mixed with water, and formed flat ball briquette (Dia meter ca.5cm)

Table 3. Weight of each briquette and ash after combustion experiments.

		Fuel W*,g	Ash W*,g	Comb.W*,g	Ratio*,%	Num.S*
Japanese	Mean=	463.4	16.6	446.8	96.2	5
Charcoal	S.D.=	256.4	8.8	248.9	0.9	
Ippatsu-	Mean=	1286.0	322.8	963.2	74.9	9
bri.	S.D.=	15.7	15.7	15.1	1.1	
Sian	Mean=	615.0	217.3	397.7	64.4	6
bri.	S.D.=	31.3	21.7	50.3	5.5	
Peking	Mean=	976.5	455.0	521.5	52.7	2
bri.	S.D.=	97.5	19.0	116.5	6.7	
Peking	Mean=	81.6	61.1	20.5	25.5	8
F.B bri.**	S.D.=	16.7	13.8	3.2	2.3	
Ch'angsha	Mean=	103.2	66.1	37.1	36.1	17
F.B bri.**	S.D.=	13.1	10.9	6.3	5.2	

Note, *; Fuel W: fuel wight (one brick of a briquette) before combustion.
Ash W: ash weight after combustion. Comb.W: The weight of burnt parts.
Ratio: The ratio of Comb.W to Fuel W.
Num.S: Number of sample to get data.
**; F.B. Bri.= Flat Ball Briquette, which was made a ball of muddled brain of briquette with water and was dried.

3.3 State of the compounds emitted through each stage of combustion process.

The trends of concentration of pollutants in flue gas were classified three kinds, that is, first one is a peak in a beginning stage such as CH_4, second is by-modal as CO and NOx and third is a peak in a middle as SO_2. Some example can be observed in Fig. 1 and Fig.2. However, the trend of emission factors were different, whose peaks were appeared in the latter stage when the combustion became gentle.

3.4 Estimate of emission factors

The emission factors (EFs) of CO, CH_4, SO_2, NOx based on the measured data are shown in Table 4. EFs of CO from the briquette combustion were 40-150 mg/g-fuel, which is extremely high compared to 2-3 ug/g for coal combustion in power plants. The EF of SO2 for the briquette in Chongqing district, of which the S content is 2.6%, is 59 mg/g-fuel. The SO_2 emission factors for others were similar and comparatively low (0.9-15 mg/g-fuel). EFs of NOx and CH_4 are 0.4-2.6 and 0.4-6.2 mg/g-fuel, respectively. CO mission was the highest amount of pollutant even air supply was rich. SO_2, NOx, CH_4 emission ones were nearly equal, and were under 20 mg/g-fuel.

Table 4. Emission factors of each compounds for same fuels [mg/(g・fuel)].

Fuel	Air supply	Compounds			
		CO	NOx	SO_2	CH_4
Japanese charcoal	Lean	92	0.76	1.2	0.55
	Rich	103-322	0.64-1.4	0.67-0.89	0.42
Japanese bri.	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 bio-bri.	Rich-1	40	0.4	2.4	1.2
	Rich-2	70	0.2	3.5	2.2
Chinese bri.*	Rich	74-93	2.4-2.6	5.3-9.3	0.13-7.8
Chongqing bri.	Rich	87	2.2	59	1.0

Note, *Chinese bri.: Chinese briquette from Peking, Sian, Chengtu.

3.5 Reduction techniques of the emission of pollutants

CO is the most major pollutant for briquette combustion. In Japan, the ceramic disk have been put above the briquette in a stove with 2-3 cm space (the small space is called "second combustion chamber (SCC)", here), in order to reduce the CO emission. SCC has eight holes (diameter; ca.1cm) in the side wall of a stove.

We confirmed the effect of the ceramic disk or SCC. The results of experiments show in Fig. 1, in which the disk reduced CO emission to half, in addition SCC added 5% reduction. The Chinese stoves, which we collected in Peking, Sian, Chengtu, has not such structure or system.

For reduction of SO_2 emission, we tried three techniques, that is, (a); putting lime

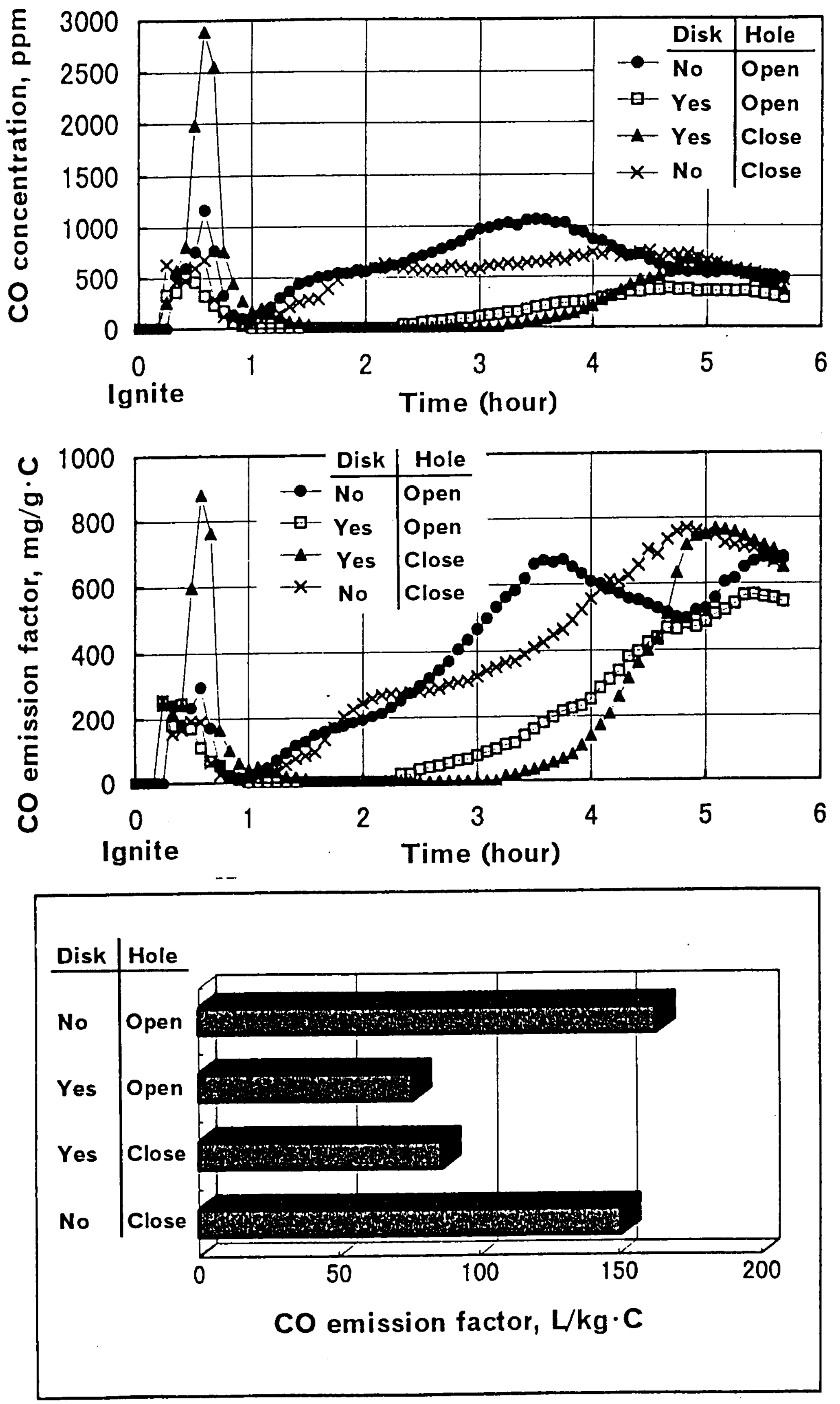

Fig. 1 The effect of a ceramic disk and holes in the "second combustion chamber" in the Japanese stove on CO emission.

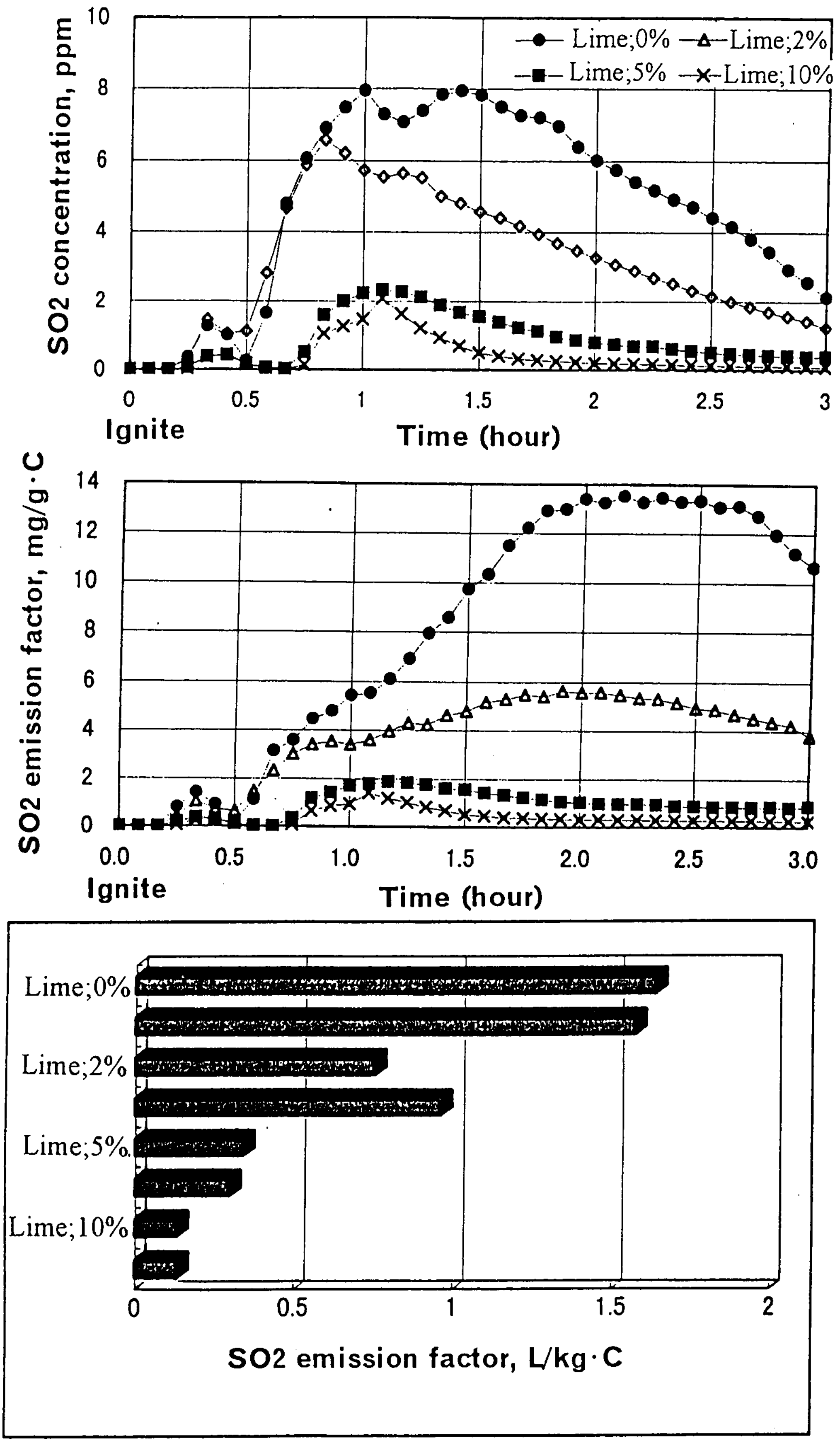

Fig.2 The relationship between the mixing ratio of lime to briquette grain and the SO2 emission.

layer on mesh [or (b); a lime disk] above briquette with ca. 1 cm space, (c); adding lime powder to coal powder and mixing to form a flat-ball of briquette. The results shows that (c) is most effective and the reduction rate of SO_2 emission was more than 85% with 5-10%(w/w) lime addition (See Fig.2), (a) is second and the rate was 45-60%, and (b) did not show obvious reduction effect. In Table 2-2, S in the briquette seemed to be caught with lime and to stay in the ash.

3.6 Importance of the briquette combustion in the total emission of each pollutants.

The total amount of pollutants emitted from the briquette combustion in China was estimated based on the emission factors in this study. They are; CO: 3-6 Tg/year, NOx: 0.06-0.18, CH4: 0.01-0.3. The CO emission from briquette combustion would be ca. 1% of total CO emission (500 Tg/year) in the world. SO_2 emission was estimated to be ca 0.9 Tg/year, based on the ratio of used amount of coal in China [2-4].

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