

C-3.1.4 Co-operative Study on the Local Production and Spread of Bio-briquetting Technique

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Abstract:

In this study, the possibility of manufacturing technique of bio-briquette added slaked lime which can fix sulfur dioxide in high efficiency was investigated. The experimental results indicated: (1) there were little emissions of air pollutants from biomass combustion and emission amounts were 35-912 and 52-1764 mg for HCl and SO₂ per 1kg of biomass, respectively; (2) the breaking strength of the bio-briquette increased with increase of addition amount and lignin contents of biomasses, which were in range of 12.1-35.1%. The bio-briquette with satisfactory strength could be produced by adding 25% biomasses without any special binder; (3) it is estimated based on our experimental results that the reduction efficiency of HCl, SO₂ and dust by the bio-briquette ranged 26-61%, 82-88% and 55-83%, respectively. It is considered that the bio-briquette will be a useful countermeasure to control emission of air pollutants for civilian and small factory consumes of raw coal; (4) the solution leached from residual ash by simulated acid rain was alkaline. The residual ash was strong alkalinity due to extra amount of slaked lime and its buffering ability to acidification was very high. It is possible that residual ash of bio-briquette will be used as the neutralizers of the acidic soil and the fertilizers of nourishment supply because it contains much nourishment elements such as Ca, Mg, and K derived from added sulfur-fixation agent and biomass.

Key Words Biomass, Bio-briquette, Breaking strength, Combustion aerosol, Zero-emission

1. Introduction

It becomes very urgent for the studies on integrated control techniques for precursors of acidic precipitation in Chongqing where has suffered from the serious air pollution caused by low graded raw coal combustion with high sulfur¹⁻⁶⁾. Coal-biomass briquette (bio-briquette) which is little emissions of air pollutants from combustion and little residual matters after burning, and higher sulfur-fixation efficiency as compared with raw coal, is expected to be a better emission

control technique⁷⁾. If biomasses which are non-use wastes would be used as the vice-material of bio-briquette, the plant fiber originally contained in biomass also works as the binders of bio-briquette which increase strength except the validity as the fuel. Considering high content of the plant fiber in the food manufacturing wastes and effective use of a wastes due to abatement of load on an environment, it is expected that the food manufacturing wastes are used as the vice-material of bio-briquette. Moreover, it is possible that residual ash of bio-briquette will be used as the neutralizers of the acidic soil and the fertilizers of nourishment supply because it contains much nourishment elements such as Ca, Mg and K derived from sulfur-fixation agent and biomass, and it indicated strong alkalinity from extra amount of slaked lime. For this reason, the zero-emission cycles could become if the residual ash of bio-briquette will be sprinkled to acidic soil as the improvement agent.

In this study, in order to investigate the possibility of manufacturing technique of bio-briquette added slaked lime which can fix sulfur dioxide in high efficiency, 4 types agriculture wastes, wilds grass, sawdust and 4 types food manufacturing wastes collected in Chongqing were investigated for combustion characteristics, emissions, action as a binder to increase breaking strength of their bio-briquette. And, the reduction efficiencies of air pollutants emitted by combustion of the bio-briquette prepared from low graded raw coal were measured by the model combustion experiment. Meanwhile, the buffering ability to acidification was determined. Moreover, the possibility of integrated environmental protection strategy using bio-briquette was investigated by leaching experiment of residual ash by simulated acid rain.

2. Experimental methods

2.1 Collection and preparation of the sample

Crop wastes, woody wastes, and food manufacturing wastes which are rice straw, sorghum stalk, wheat straw, maize stalk, wild grass, sawdust, baggase dregs, beer sake lees, sorghum wine sake lees, and tofu dregs were collected in Jiangbei district of Chongqing, China. The biomasses and the coals produced in Chongqing and Chengdu of China were cut finely to 5 mm and dried.

2.2 Combustion experiment

In **Fig. 1**, 1.000 g (or 0.500 g) of the sample in the quartz boat was put in the center of quartz combustion tube maintained at 500°C. Then, 1.0 l/min of burning gas was flowed from a flow controller to electric furnace, electric furnace was heated from 500°C to 800°C and the experimental sample was burned for 20 minutes at 800°C. Temperature in combustion tube was monitored by a thermo-couple. The exhaust gas from combustion was diluted by 4.0 l/min of purified air. The particles emitted from the sample combustion were collected with a quartz filter (2500QAT-UP, Pallflex Co.). A part of diluted gaseous exhaust (1.0 l/min) was introduced to impinger, which was filled with 150 ml of absorption solution (2.7 mM Na₂CO₃ /0.3 mM NaHCO₃ (eluent solution for anion analyses in ion chromatograph)). Then, Cl⁻, NO₂⁻, NO₃⁻, SO₃²⁻ and SO₄²⁻ concentrations in the absorption solution were measured with an ion chromatograph

(DX-100, Dionex Corp. of U. S. A.).

A part of the filter collected aerosols is extracted by an ultrasonic cleaner. Then, Cl^- , NO_2^- , NO_3^- , SO_3^{2-} and SO_4^{2-} (Ion Pac AS12A), Na^+ , NH_4^+ , K^+ , Ca^{2+} and Mg^{2+} (Ion Pac CS12A) concentrations in the extracted solution were measured with an ion chromatograph. The acid neutralizing capacity (ΔC_b) to acidification was investigated by an auto-titration apparatus⁹⁾. Ten ml of extracted solution is titrated until pH 5.6 with 0.01M NaOH or 0.01M HCl. ΔC_b is defined as negative value when OH is added.

2.3 Measurement of lignin content in the biomass

According to JIS P8008 method, the biomass is extracted with the ethanol-benzene mixture (1:2,v/v) by Soxhlet's extractor for 6 hours. The filtrated sample is left in 20°C-thermostatted water bath for 4 hours after 15 ml of 72 % sulfuric acid was added, then boiled at flask with a condenser for cooling for 4 hours after 560 ml of distilled water was added, filtrated with the glass

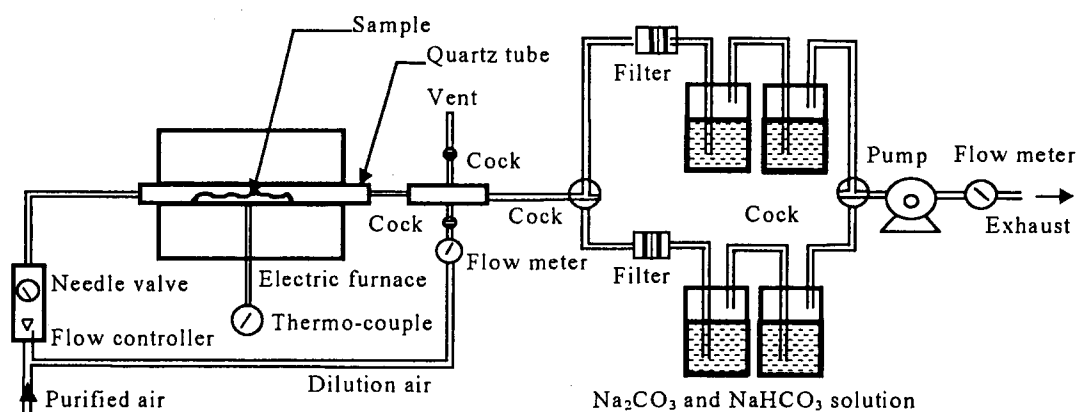


Fig. 1. Experimental equipment for combustion test.

filter, washed with the distilled water, dried at $105\pm 3^\circ\text{C}$ for 4 hours. Then, the mass of the residual substance is weighted. The lignin content (%) is calculated.

2.4 Bio-briquette production and measurement of their breaking strength

The mixtures of coal and biomass added the slaked lime ($\text{Ca/S}=2$) is manufactured experimentally to tablet type bio-briquette (12×7 mm ϕ , 1 g) under 4 tons/ cm^2 of pressure using high-pressure jack machine (Iuchi Seieidou Corp.) showed in Fig. 2 at the laboratory. Beside, the mixtures of the coal (70~85%) and the biomass (15~30%) added the sulfur fixation agent $\text{Ca}(\text{OH})_2$ ($\text{Ca/S}=2$) were produced to the bio-briquettes of an almond (22×35 mm ϕ , ca. 8 g) or pillow type (40×45 mm ϕ , ca. 30 g) by the manufacturing procedure shown in Fig. 3 with the rolling compressor under $3\sim 5$ tons/ cm^2 pressure in Chongqing and Chengdu of China, and Hokkaido of Japan. And, for each kind of tablets and bio-briquettes, the breaking strength was measured by axis pressure testing machine (Maruto Testing Machine Corp.) with a steel ball (10

mm ϕ) in diameter.

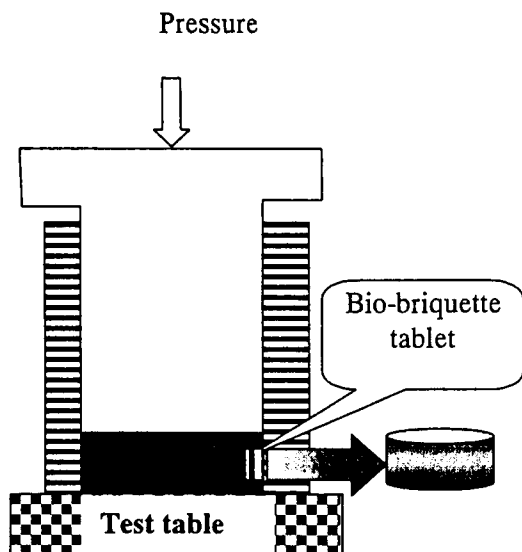


Fig. 2. Tableting test of coal-biomass.

matter after burning bio-briquette added slaked lime in mixture of baggase and raw coal from Chongqing of China based on the equivalent ratio $Ca/S = 2$. SAR used in this experiment was prepared by H_2SO_4 and ion exchange water, their pH were 3.0, 4.0, 5.0 and 5.6. One gram of combustion ash of the bio-briquette was put into the glass column (26 mm ϕ) with the filter. Then, the solution leached from the column was collected after adding SAR in 50 ml/h rate (about 95 mm/h rainfall). The total addition of SAR was 5,300 mm, nearly equal with the total rainfall during 7 years (the annual rainfall in Chongqing is about 800 mm). Then, Na^+ , NH_4^+ , K^+ , Ca^{2+} and Mg^{2+} concentrations in the leaching solution were measured with an ion chromatograph. The acid neutralizing capacity (ΔC_b) to acidification was determined by an auto-titration method. ΔC_b is defined as positive value when H^+ is added.

3. Results and discussion

3.1 Composition of raw coals

As is clear from **Table 1**, all of the coals except Chengdu raw coal 1 contained a high percentage of the sulfur and ash. The most of sulfurs contained in coal are combustible sulfur, which are emitted to atmosphere by their combustion. Thus, the serious air pollution is occurred in Chongqing due to the complicated terrain and unfavorable meteorological condition for diffusion of air pollutants emitted from combustion of coal with high sulfur and ash without any treating.

2.5 Leaching experiment of combustion ash of bio-briquette

The compositions of leaching solution of bio-briquette combustion ash by SAR were measured by leaching experiment¹⁰. The ash sample was the residual

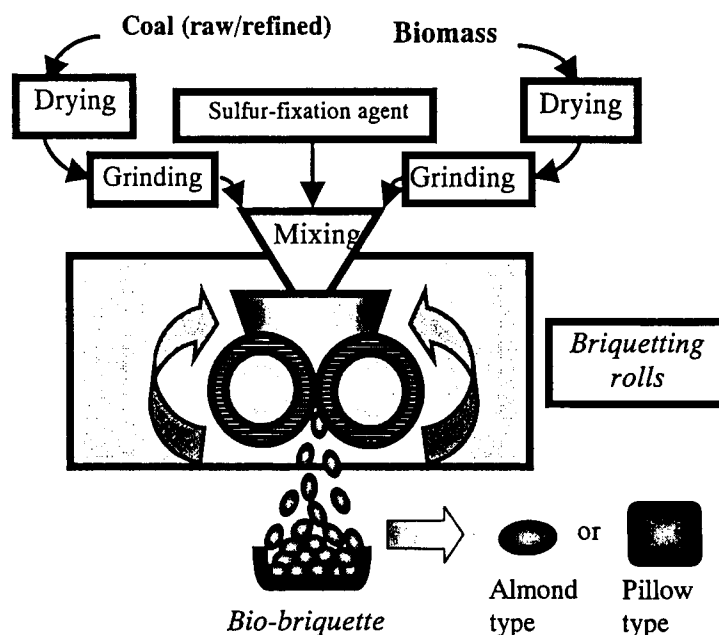


Fig. 3. Flow sheet of bio-briquette production.

Table 1. Compositions of coal used in production of bio-briquette (%)(dry basis).

Coal sample	Ash	Volatile matter	Fixed carbon	Sulfur		
				Combustible	Incombustible	Total
Chengdu raw coal 1 ^{a)}	23.0	8.7	66.5	0.40	0.11	0.51
Chengdu raw coal 2 ^{a)}	29.2	15.9	54.9	2.03	0.67	2.70
Chengdu raw coal 3 ^{a)}	28.6	11.9	59.5	2.64	0.31	2.95
Chongqing raw coal 1 ^{a)}	29.8	12.7	57.5	2.47	0.26	2.73
Chongqing refined coal 2 ^{b)}	16.0	19.7	64.3	1.11	0.11	1.22
Chongqing raw coal 3 ^{b)}	38.6	20.9	40.5	2.16	0.37	2.53

^{a)} Anthracite coal; ^{b)} Bituminite coal.

3.2 Air pollutants from biomasses combustion and basic analysis of biomass

HCl and SO₂ emission amounts from combustion of 1 kg biomass in **Table 2** based on the data estimated from Cl⁻, SO₃²⁻ and SO₄²⁻ concentration in collecting solution for exhaust gas were in the range of 35~912 and 52~1764 mg, respectively. Combustible sulfur content of the biomass and the food manufacturing wastes were low, sawdust gave the lowest content, and beer sake lees gave the highest one in comparison with a wastes of crops. In a manufacturing process of beer, it is thought to add enzyme containing sulfur as a cause of SO₂ exhaust from beer sake lees combustion which was a little high. Considering the residual ash content, content of the volatile

Table 2. Emission of the air pollutants from biomass combustion and basic analysis of biomass (dry basis).

Biomass	Emission (mg/kg-biomass)		Combustible sulfur ^{a)}	Ash	Volatile matter	Fixed carbon
	HCl	SO ₂				
Sorghum wine sake lees	136	91	0.005	4.3	82.5	13.2
Baggass dregs	150	228	0.011	2.8	84.6	12.6
Tofu dregs	153	1208	0.060	4.4	83.1	12.5
Beer sake lees	354	1764	0.088	5.5	81.6	12.9
Sawdust	80	114	0.006	1.7	83.9	14.4
Rice straw	912	220	0.011	17.0	69.0	14.0
Wheat straw	406	386	0.019	8.2	85.9	5.9
Maize stalk	203	599	0.030	12.5	79.9	7.6
Sorghum stalk	799	789	0.039	4.2	82.6	13.2
Wilds grass	35	52	0.003	21.7	67.7	10.6

^{a)} Combustible S was defined as sulfur emitted from biomass combustion.

matters and the concentration of pollutants emitted from the biomass combustion, sawdust was the best suitable vice-material for bio-briquette of biomasses investigated in this work. Since there are little air pollutants from food manufacturing wastes combustion, it is assumed that the food manufacturing wastes have the availability as a binder of bio-briquette considering effective use of the wastes and abatement of load on an environment.

The volatile matter and the ash of biomass in **Table 2** were in the range of 65.5~85.9% and 1.7~25.5%. The biomasses with 80 % or higher volatile matter and 6.0 % or less ash content were sawdust, sorghum stalk, and food manufacturing wastes. In consideration of the low ash content

and the high volatile matter content, these food manufacturing wastes could be used as a vice-material of bio-briquette. Because the decomposition combustion precedes at a early combustion stage of solid fuel such as the coal and the biomass, the thermal decomposition gas (volatile matters) of the possible inflammability in an oxidation atmosphere is bring about, and this generated substance is ignited by an ignition source. The ignition temperature of solid fuel containing much volatile matters is generally low. When the biomass with lower ignition temperature is mixed with coal, the thermal decomposition products are generated at much lower temperature than the ignition temperature of coal ¹¹⁾. Beside, it is consider that the agriculture wastes will be better vice material when the bio-briquette is produced since the food manufacturing wastes were being used to feed the animals. Moreover, there were large amounts of biomass from agriculture wastes because the weather was suitable for production of agriculture in Chongqing. From the investigating results in **Table 3**, it was as known that total yearly output of woody wastes and agriculture wastes amounted to ca. 18.0 million tons. This data is equal to consumption of coal in Chongqing. Therefore, it is considered that biomass is enough to produce bio-briquette.

Table 3. Results of the research for biomass output in Chongqing.

Biomass	Output (10 ⁶ t)
Rice straw	2.0 ~ 2.2
Wheat straw	1.0 ~ 1.2
Corn	2.3 ~ 3.0
Weeds	~ 3.2
Maize stalk and straw	2.5 ~ 2.7
Wastes of other plants	~ 5.4
Woody dust	~ 0.3
Total outputs	16.4 ~ 18.0

3.3 Lignin content

The biomasses work as a binder when the bio-briquette is produced. It is considered that the binder work of the biomass is induced by the sticking of pulverized coal due to softening of lignin and hemicellulose contained in it. Lignin contents of biomasses in **Table 4** were in range of 12.1~33.3%, and the food manufacturing wastes were lower in comparison with the plants wastes.

Table 4. Lignin content of the biomasses (%) (dry basis).

Biomass	Wheat straw	Sorghum stalk	Maize stalk	Sorghum wine sake lees	Rice straw
Lignin content	33.3	30.4	28.8	25.7	24.4

Biomass	Sawdust	Baggass dregs	Wild grass	Beer sake lees	Tofu dregs
Lignin content	22.4	21.7	21.0	15.7	12.1

3.4 Breaking strength of bio-briquette

The briquetting characteristics of biomass-coal were influenced by the carbonization degree, the charcoal nature of coal, the kind and the addition amount of biomass as the vice-material. These results were summarized as following.

(1) When the bio-briquette is manufactured from the coal added sawdust (25%), the breaking strength was concerned to the kind of coals. Refined coal was higher than that of raw coal. And, for the breaking strength, both of tablet and bio-briquette was 40 kg or more although the bio-briquette produced in the

local area was not strong compared with the thing which produced in Hokkaido Industry Research Institute. Thus, the bio-briquette with satisfactory strength could be produced by adding 25% biomasses without any special binder.

(2) The breaking strength of the tablet type bio-briquette was concerned to the lignin contents of biomasses added (Fig. 4), increased with increase of lignin contents of biomasses except baggase, which is sugar-rich food. Therefore, it is considered that baggase is the best vice-material for the production of bio-briquette because it showed especially excellent binder work in 10 kinds biomasses and there are not much air pollutants emitted from its combustion.

(3) The food manufacturing wastes with lower lignin content could be used also as the binder by mixed to the biomass with high lignin content (Fig. 5).

(4) When the biomass works as the binder, the strength of the bio-briquette increases with increase of addition amount of biomass (Fig. 6). The addition of 15~30% biomass gave the strength enough for normal handling, the bio-briquette containing 20% wheat straw or sawdust showed the breaking strength of 50 kg or more.

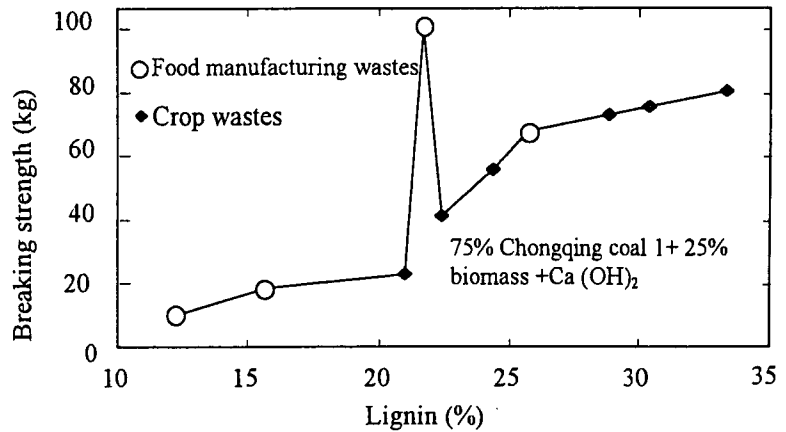


Fig. 4. Relationship between the breaking strength of bio-briquette and the lignin content of biomass.

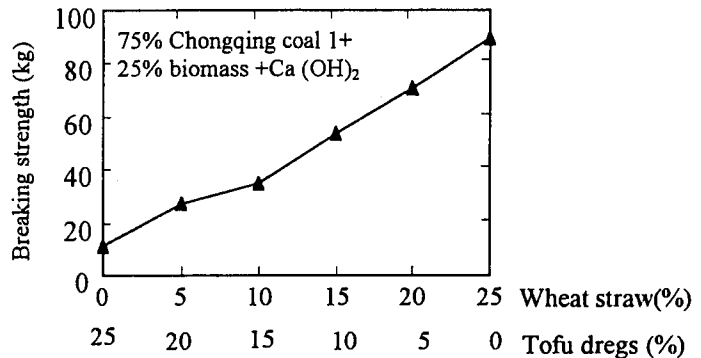


Fig. 5. Relationship between breaking strength of bio-briquette and mixed biomasses.

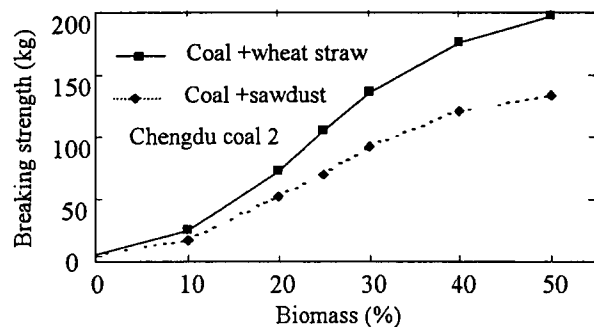


Fig. 6. Relationship between the breaking strength of bio-briquette and addition of biomass.

3.5 Reduction efficiency of air pollutants emission by the bio-briquetting of raw coal

In Table 5, compared with raw coal, air pollutant emissions from the bio-briquette combustion were reduced largely, their reduction efficiency for HCl, SO₂ and dust ranged 26~61%, 82~88% and 55~83%, respectively. From experimental results, it is known that the reduction efficiency of dust for bituminite coal was more excellent than anthracite coal. While, the reduction efficiency for HCl was lower because HCl emission from biomass combustion was higher, and chloride in coal and biomass was decomposed easily under 800°C. It is expected that air pollutants emission such as SO₂ is reduced drastically with bio-briquette when it is popularly used in some districts such as Chongqing where have suffered from serious air pollution and acid rain.

Table 5. Emission of the air pollutants from coal and their bio-briquette combustion, and reduction efficiency of pollutants by bio-briquetting (dry basis).

Sample	Emission (mg/g-coal)			Combustible S%	Reduction efficiency (%)		
	HCl	SO ₂	Dust		Dust	HCl	SO ₂
Chengdu raw coal 1 ^{a)}	0.05	8.01	0.69	0.40			
B.B ^{c)} (coal+sawdust)	0.04	1.02	0.31	0.05	55	30	83
Chengdu raw coal 2 ^{a)}	0.20	40.62	2.37	2.03			
B.B ^{c)} (coal+sawdust)	0.10	4.16	0.90	0.21	62	35	85
Chengdu raw coal 3 ^{a)}	0.15	52.83	2.82	2.64			
B.B ^{c)} (coal+sawdust)	0.06	6.65	1.02	0.33	64	49	82
Chongqing raw coal 1 ^{a)}	0.39	49.49	2.25	2.47			
B.B ^{c)} (coal+sawdust)	0.11	5.48	0.79	0.27	65	61	84
Chongqing refined coal 2 ^{b)}	0.12	22.20	8.93	1.11			
B.B ^{c)} (coal+sawdust)	0.12	2.06	2.18	0.10	76	54	87
B.B ^{c)} (coal+rice bran)	0.28	1.93	1.60	0.10	82	26	88
B.B ^{c)} (coal+maize stalk)	0.30	2.67	1.59	0.13	82	31	83
B.B ^{c)} (coal+tofu dregs)	0.14	2.99	1.51	0.15	83	50	82

^{a)} Anthracite coal, ^{b)} Bituminite coal, ^{c)} Bio-briquettes (B. B.) were produced from 75 wt% of raw coal and 25 wt% of biomass by the addition of sulfur-fixation agent (Ca(OH)₂)(Ca/S=2.0)

The aerosol emissions from anthracite coal and bituminite coal combustion range 0.39~2.82 and 5.03~14.30 mg/g-coal with our experiments device, respectively. However, the aerosol emissions from the bio-briquette combustion are lower than the raw coal, range 0.31~1.02 and 1.5~1.6 mg/g-coal for the bio-briquette produced by anthracite coal and bituminite coal, respectively. All water-soluble solutions of coal and bio-briquette combustion aerosols were acidic. Their ΔC_b were negative value. However, compared with the particulate emission from the raw coal, acidity of the particulate matter emitted from the bio-briquette combustion was lower, SO₂ emission was little. It is considered that the impact on the environment from the bio-briquette combustion may be a little.

Major water-soluble anion of coal combustion aerosols was SO₄²⁻, which accounted for 90% of total anions, major cations were H⁺ and NH₄⁺. Water solution of the combustion aerosol was called nearly as the sulfuric solution. Compared with the raw coal, H⁺ and SO₄²⁻ concentration of

bio-briquette combustion aerosols were reduced largely. However, Cl⁻ concentration increased fractionally caused by higher emission from the biomass because HCl emission from biomass combustion was higher than raw coal.

3.6 Zero-emission by using bio-briquetting

It is considered that that residual ash of the bio-briquette would be used as the fertilizers of nourishment supply because it contains much nourishment elements such as Ca, Mg, K by added slaked lime and biomass and also as the neutralizer of acidic soil since there is strong alkalinity from extra amount of slaked lime. For these reasons, bio-briquette can be used as not only the control techniques for precursors of air pollution, but also its ash can improve the soil. Then, biomass products waste after harvesting will be used as the vice-material of the bio-briquette production further. Thus, it is possible that load on an environment from the bio-briquette could become minimum.

3.6.1 pH changes and the buffering ability of leaching solution by simulated acid rain (SAR)

pH changes on various kinds pH SAR and the buffering ability (ΔC_b) of leaching solution by pH 5.6 (SAR) is shown in Fig. 7. pH3 SAR could not acidify the ash by the rainfall of 2500 mm, only up to 4.5 on 5000 mm or more rainfall. Beside, pH of all leaching solution by pH4, 5, 5.6 SAR were near 8.0, presented strong alkalinity. Finally, it is considered that the leaching solution has the high buffering ability to acidification based on ΔC_b changes of leaching solution by pH 5.6 (SAR). Thus, it is assumed that the combustion ash of bio-briquette could neutralize acidic soil through sprinkling it to the region where is suffered from acid rain. And, the heavy metal such as Al³⁺ which is poisonous for the plant is hard to leach under alkaline condition. Furthermore, it was reported that slaked lime was sprinkled to acidic soil to rise the growth nature of wheat in the part area of China, and it is considered that it can make activity of the toxic heavy metal such as Al in soil decline¹²⁾.

3.6.2 The basic ions of the leaching solution by SAR

The leaching solution by SAR contained a large amounts of Na⁺, NH₄⁺, K⁺, Mg²⁺ and Ca²⁺ based on analytical data, which they were about 0.2, 24, 1, 2, 1000 ppm on 9 mm rainfall. Ca²⁺ and Mg²⁺ concentration were still high although amount of Na⁺, NH₄⁺ and K⁺ decreased rapidly with the increase of rainfall, near 7 ppm and 0.4 ppm on 5000 mm rainfall.

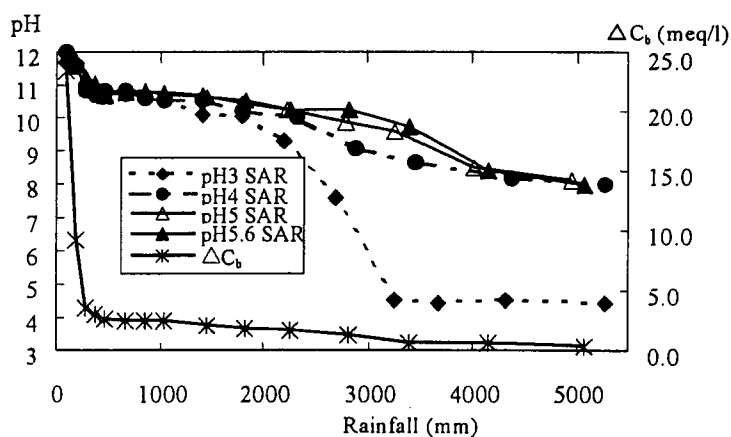


Fig. 7. Variation of pH and ΔC_b in solution of residue ash leached by simulated acid rain.

A large amounts of basic cations existed in leaching solution of bio-briquette ash shown in Fig. 8, total amounts of cations ($\text{Na}^+ + \text{NH}_4^+ + \text{K}^+ + \text{Ca}^{2+} + \text{Mg}^{2+}$) nearly equal with exchanged basic ion of the soil in Chongqing ($0.15\sim 0.25 \text{ meq/g-soil}$)¹²⁾ on 5000 mm rainfall. The concentration of Ca^{2+} accounted for 90% or more of total cation. It is reported that the forest decline have occurred in European due to the decrease of basic ions along with exhaust regulation of the atmospheric dust and the useful amount of Ca^{2+} and Mg^{2+} in the forest soil¹³⁾. Therefore, it is considered that the residual ash could improve the lack situation of basic ions such as Ca^{2+} and Mg^{2+} .

4. Conclusion

Biomass such as crop wastes and food manufacturing wastes can be used in the production of bio-briquette because there were little emissions of air pollutants and its action as a binder. It is estimated based on our experimental results that the reduction efficiency of HCl, SO_2 and dust by the bio-briquette ranged 26~61%, 82~88% and 55~83%, respectively. The solution of residual ash leached by simulated acid rain was alkaline. Its buffering ability to acidification was very high. It is possible that residue ash of bio-briquette will be used as the neutralizers of the acidic soil and the fertilizers of nourishment supply because it contains much nourishment elements such as Ca, Mg, K and Na.

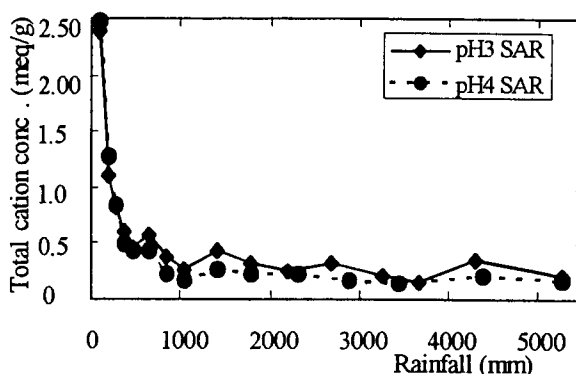


Fig. 8. Total basic ions in solution leached from residual ash by simulated acid rain.

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