

Application of Waste Rubberwood Ash in Carbon Dioxide Absorption

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ABSTRACT

In upgrading raw natural gaseous fuel, CO₂ has to be removed. Waste rubberwood ash solution was applied in CO₂ absorption. The pH of the rubberwood ash suspension was max. 12.4, with the main dissolved minerals being potassium, magnesium and sodium, and no significant amounts of heavy metals were found. Rubberwood ash solution was able in a packed column reactor to remove 41% v/v of CO₂ in an N₂ stream. The highest 95.2 % CO₂ removal efficiency was achieved at the gas flow and absorbent flow rates of 600 L/h and 500 L/h, respectively, and required 35.4 kg ash/kg CO₂ removed. A concept for the application of rubberwood ash in biofuel production for biomass power plant is proposed. The discussion reveals the possibility of a good practice waste management utilizing waste ash in upgrading gaseous fuel and furthermore in environmentally benign biofuel production.

KEYWORDS

carbon dioxide; rubberwood ash; chemical absorption; packed column

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Introduction

Carbon dioxide (CO₂) is the primary greenhouse gas emitted by human activities. The main human activity that emits CO₂ is the combustion of fossil fuels, such as natural gas, crude oil, and coal, for energy to transportation or to industrial processes. According to the known geographic distribution of natural

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gas reserves, almost 40% of the global reserves are sour and about 30% have CO₂ contents in the range 15-80% (Baccanelli *et al.*, 2016). Biogas as raw material is relatively evenly distributed and can provide alternative energy. It can be produced by anaerobic fermentation and decomposition of organic matter. The proportions of components in biogas depend on substrate, temperature, and microorganisms. Typically, biogas is comprised of 35-65% CH₄, 15-50% CO₂, and low quantities of other impurities, such as nitrogen (0-5%), oxygen (0-5%), and traces of hydrogen sulfide (Sarperi *et al.*, 2014 and Eze and Agbo, 2010).

For use by vehicles as compressed natural gas or biogas, CO₂ and other impurities need to be removed in order to enhance fuel quality and maximize engine performance. CO₂ may be removed from a gaseous fuel by physical absorption, chemical absorption, biological fixation, cryogenic separation, or membrane separation. The suitability of these technologies depends on characteristics of the gas mixture (Ryckeebosch *et al.*, 2011). Chemical absorption with alkaline solution can treat gas mixture that contain acidic gases like CO₂, hydrogen sulfide, or oxides of nitrogen, among others (Bandyopadhyay and Biswas, 2012; Eze and Agbo, 2010; Lu *et al.*, 2006). This process can achieve very high removal efficiencies exceeding 95% (Ryckeebosch *et al.*, 2011), and the absorption performance depends on various conditions. Absorbents such as sodium hydroxide, potassium hydroxide, calcium hydroxide, mono-ethanolamine, methyl di-ethanolamine, or ammonia, have been used commercially. Amine derivative absorbents have high efficiency in CO₂ removal and can be recycled by heating and stripping the CO₂ out of the solution. However, these kinds of absorbents have some drawbacks, mainly degradability, so some chemicals need to be added to reduce the degradation rate, and make-up chemicals are required (Tan *et al.*, 2012).

To reduce the costs of chemical consumption in a CO₂ absorption process, inexpensive or waste substances for absorbent solution preparation are attractive. Alamdari *et al.* (2014) studied CO₂ absorption from flue gas by using distiller waste of a soda ash plant, which mainly contained CaCl₂ which formed CaCO₃ precipitate in the absorbent product. Sarperi *et al.* (2014) upgraded raw biogas to vehicle fuel by using basic oxygen furnace (BOF) slag, a by-product from the steel industry containing calcium and iron oxides, to remove simultaneously CO₂ with CaO (carbonation) and H₂S with Fe₂O₃ (desulfurization). Baciocchi *et al.* (2014) removed CO₂ from biogas by means of chemical absorption using KOH or NaOH in the first stage. In the second stage, the spent absorption solution was regenerated by waste incineration air pollution control residues, and CO₂ was captured by carbonation. Rubber is economic crop in southern Thailand, and rubberwood residues are byproducts from rubberwood industries. Rubberwood residues are used as biomass fuel in boilers at many factories located nearby the wood residue production, such as seafood industries, in rubber sheet production, and in biomass power plants. An example is seen in southern Thailand where approximately 16 tons of rubberwood ash per day is generated by a 22 MW biomass electricity power plant (Gulf Yala Green Co., Ltd., Thailand).

Waste ash from biomass burning gives alkaline solutions or suspensions. The ashes can be used as soil amendment and in bricks, in small amounts, but a large amount of the ash still has to be landfilled requiring neutralization with acid, because its leachate is alkaline at pH in the range 8-9 (Sakthivel *et al.*, 2012). The main components in rubberwood ash are calcium oxide (34-41%) and

potassium oxide (13-16%), with small amounts of silicon, phosphorus, manganese, iron and others (Hawa *et al.*, 2014; Dasaesamoh *et al.*, 2011). An alkaline solution prepared from the ash would contain several ions that can be used as plant mineral nutrients. CO₂ would be curtailed in the ash solution as bicarbonate species formed by reactions. Therefore, CO₂ absorption by alkaline ash solution should produce waste water that can be applied to growth of aquatic plants and algae, further fixing CO₂ into biomass instead of releasing it to the atmosphere. The minerals in the wastewater can be removed by the aquatic plants or algae. Aronsson and Ekelund studied the effects of wood ash solution on growth, photosynthesis and pigments of the freshwater moss *Fontinalis antipyretica* Hedw. The results showed that pH of the wood ash solution was about 6.2-11.3, depending on the concentration of wood ash, and the freshwater moss grew well around pH 10.8.

Thus it is possible to use alkaline rubberwood ash solution as the absorbent to remove CO₂ from a gaseous fuel mixture, and the CO₂ and minerals in the solution can then further be utilized as fertilizer for e.g. aquatic plant biomass production. The produced biomass can be utilized in the production of supplements, animal feed, or sustainable bio-fuels. The waste insoluble ash residue after separating the soluble alkaline is safe to discard environmentally friendly, as it will no longer generate toxic leachates. Based on the authors' literature search, there is no prior report on using rubberwood ash solution to remove CO₂. This research therefore aimed at exploring the feasibility and efficiency of applying rubberwood ash in the removal of CO₂ from a gas mixture.

Materials and Methods

2.1 Alkaline rubberwood ash solution preparation

Rubberwood ash (fly and bottom ash mixture) was collected from Gulf Yala Green Co., Ltd., Yala, Thailand. The ash was screened using a pilot scale screener to remove particles larger than 1.7 mm (mesh 10). To investigate the solubility of sieved ash, various amounts of ash (5-50 g) were dissolved in 100 mL tap water, stirred with a magnetic stirrer for 30 min, and the insoluble solids were separated by vacuum filtration. The pH, alkalinity and composition of the filtrate were determined. The insoluble rubberwood ash filter cake was dried at 103 °C and weighed to determine the solubility of fresh ash,

$$\text{Dissolved ash (\%)} = 100 (W - W_i) / W \quad (1)$$

where W is the initial weight and W_i is weight of the insoluble ash, respectively.

2.2 CO₂ removal from gas stream by the alkaline solution

A schematic diagram of the gas absorption system used in this study is shown in Fig. 1. An absorption column with 2 m height and 0.1 m diameter was randomly packed to contain a 1.4 m layer of Raschig rings. Nominal surface area of this packing material was 365 m²/m³. The compressed gas mixture (41% v/v CO₂ and rest N₂) was fed through the packed column from the bottom towards the top, while the alkaline solution was pumped to an injector at top of the column at a liquid flow rate measured by rotameter. The experiments had a fixed gas flow rate of 600 L/h, while the solution flow rates were in the range 200-500 L/h. Both inlet and outlet gas samples were collected at steady state for CO₂ content

determinations, and the CO₂ removal efficiency was then calculated. Used absorbent solution samples were taken from the outlet solution tank, and their CO₂ loadings were analyzed. Sodium hydroxide solution at pH similar to the rubberwood ash solution was also used as absorbent to compare the CO₂ removal efficiencies to this baseline case.

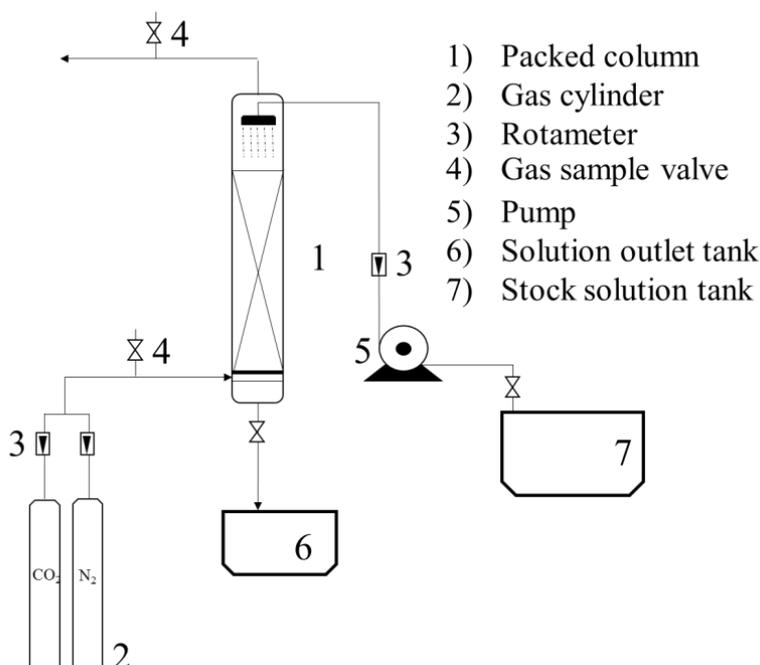


Figure 1. Schematic diagram of the experimental setup for CO₂ removal by absorption

2.3 Analysis Methods

The chemical composition of rubberwood ash was measured by x-ray fluorescence (XRF), and elements in the alkaline rubberwood ash solution were determined by inductively coupled plasma optical emission spectroscopy (ICP-OES). Alkalinity of the used absorbent solution samples was measured titrimetrically using a standard sulphuric acid solution, with end-point indicators and/or pH meter (AOAC Official Method 973.43). The concentrations of hydroxide carbonate and bicarbonate corresponding to total alkalinity were estimated using the equations proposed by Snoeyink and Jenkins (1980). The pH was measured using a standard pH meter. The gas compositions at inlet and outlet were measured by GC-TCD (Shimadzu GC 8A; Porapak Q column).

All the measurements were done in triplicates and the results were plotted and analyzed using ANOVA (analysis of variance) at 5% significance level.

Results and Discussion

3.1 Alkaline rubberwood ash solution

The rubberwood ash used in the experiments was the end product of boiler combustion of rubberwood residue, obtained from sawmilling and plantation residues such as slabs, roots, and branches. Wood ash from the boiler contained mostly minerals, but it also contains some combustible organics or other oxidizable residues, and was a mixture of fly ash and bottom ash. Fly ash is black, composed of fine particles driven out of the boiler with the flue gases: it is captured by particle filtration of the exhaust fumes. Bottom ash is a complex heterogeneous mixture of all the non-flammable, non-volatile minerals, which remain after the wood and charcoal have burned away in the furnace of a boiler. Both rubberwood bottom and fly ash must be neutralized before discharging to the environment. Traditionally this type of ash has been used by gardeners as good source of potassium and calcium carbonate, the latter acting as a liming agent that neutralizes acidic soil. However, abundant unused ash still remains with the biomass power plant.

A mixture of rubberwood fly ash and bottom ash was used in this study, and the XRF analysis results are shown in Table 1. Si and Ca were the main inorganic components with small amounts of Mg, K, P, Al and Fe, and trace amounts of Mn, S and Sr. Hawa *et al.* (2014) and Dasaesamoh *et al.* (2011) reported similar results, with the main oxide component in rubberwood ash analyzed by XRF being calcium oxide. The crystalline phase of rubberwood ash characterized by x-ray diffraction (XRD) contained mostly calcite and quartz with arcanite as a minor component. These minerals are common in biofuel ashes (Pöykiö *et al.*, 2014). Moreover, the amounts of heavy metals observed in our ash samples were very similar to those reported by Masae *et al.* (2013). The chemical composition of rubberwood ash depends on several factors, such as tree species, soil type, nutrients, growth environment, age, and part of the tree. Maneerung *et al.* (2015) reported that the furnace temperature is an important factor affecting the composition of wood ash, and that ash from wood biomass contains large amounts of calcite, which can be directly transformed into lime by simple calcination at temperatures exceeding 800 °C. At temperatures of 650 °C and below, calcite and quartz were the most abundant crystalline phases, but also fairchildite was present in relatively small amounts.

Table 1 Elements in the rubberwood ash and in its aqueous extract

Elements	Rubberwood ash (%)	Alkaline extract solution (mg/L)
Si	26.8	12.3
Al	1.7	<0.003
Fe	1.1	0.1
Ca	25.6	9.1
Mg	7.4	32.2
Na	<0.001	62.0
K	6.2	1,777
P	2.7	6.5
S	1.5	0.8
Mn	0.4	<0.001
Cl	<0.001	97.0
Cu	<0.001	0.01
Sr	0.1	0.01

¹ Loss of Ignition (LOI)

The pH of alkaline ash solution and solubility of ash at different ash to water ratios are shown in Fig. 2. The pH of filtrate solution increased with ash to water ratio, and gradually became stable with ash to water ratios above 0.03. The pH increased because of basic buffering, whose mechanism with pH above 7 is the dissolution of carbonate or bicarbonate minerals, and leaching of cations and hydrogen carbonate ions. The exchange of hydrogen ions with base cations (potassium ion, sodium ion, magnesium ion, and calcium ion) accompanied by the leaching of the base cations with sulfate ions dominate the acid buffering process (Jiang *et al.*, 2016). Typically, the leachate of an ash is alkaline, and leachate neutralization requires the use of an acid. If alkaline rubberwood ash is applied as an absorbent that replaces commercial alkali, then acid is not required, and also the release of contaminants by leaching and their environmental impacts from residue disposal are reduced. The insoluble ash after extraction with water can be discarded or utilized as filler in concrete or in lightweight bricks.

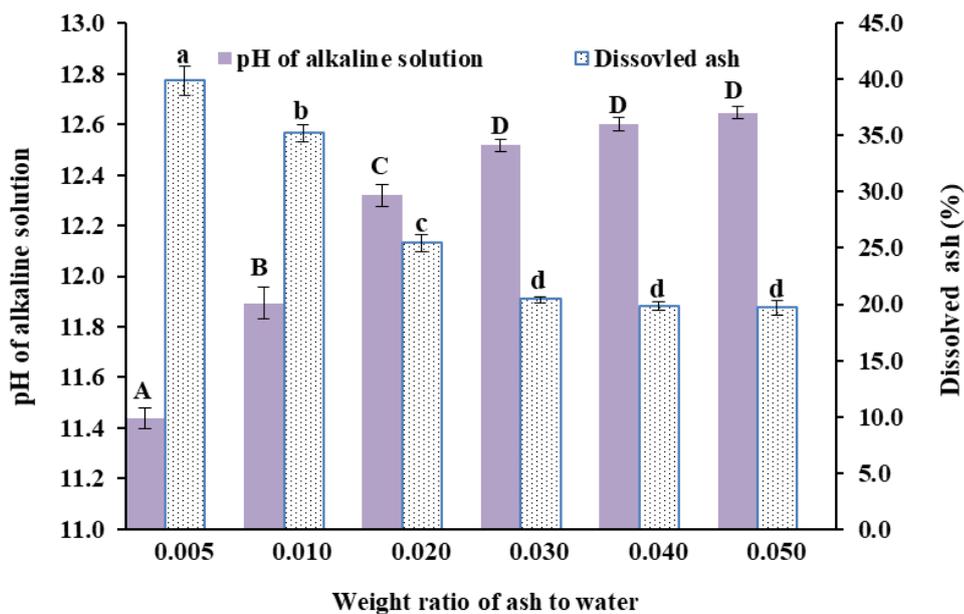


Figure 2. pH of aqueous rubberwood ash extract and apparent solubility of ash at various rubberwood ash to water weight ratios

When the ratio of rubberwood ash to water increased, the percentage of ash that was dissolved decreased, with corresponding increase in the portion of insoluble ash. In practice, high ash to water ratios make it difficult to stir the ash-water mixture. After ratio 0.03 of sifted rubberwood ash to water, apparent solubility of ash was stable, so this ash to water ratio was used in further absorption experiments. Composition of the filtrate solution, analyzed by ICP-OES, is shown in the third column of Table 1. The main metal detected in the

alkaline solution was K (1,777 mg/L), followed by Cl, Na, and Mg, with smaller amounts of Si, Ca, P, and trace amounts of S, Fe, Cu and Sr. It can be seen that the calcium content in rubberwood ash is highest among the soluble components, but its 9 mg/L presence in the filtrate is relatively low. Moreover, the filtrate had 62 mg/L of sodium and 97 mg/L chloride, while these chemical species are not dominant in the ash. Possible some of the ions observed in the alkaline rubberwood ash filtrate came with the tap water. Sodium salt and chlorides are used in tap water treatments. These results are similar to Pöykiö *et al.* (2014), who characterized biomass-based bottom and fly ash fractions from a large-sized power plant of an integrated pulp and paper mill complex, and the extractable concentrations of elements in bottom ash and fly ash using water extraction, showing that potassium and calcium were present in the water.

3.2 Carbon dioxide removal from gas stream by alkaline solution

Efficiency of the alkaline solution (prepared from rubberwood ash) as absorbent that removes CO₂ from a mixed gas stream was investigated in a laboratory-scale packed column scrubber. The CO₂ removal efficiency with 600 L/h fixed gas flow rate, using alkaline solution prepared with rubberwood ash to water weight ratio 0.03, under alkaline solution flow rates 200-500 L/h, is shown in Fig. 3. As expected, increasing the solution flow rate improved the efficiency. Providing fresh solution more rapidly kept the system further away from equilibrium, and caused higher reaction rates. Moreover, high solution flow rates spread the solution on the packing surfaces better, increasing the effective interfacial area of solution and gas, and improving interfacial mass transfer.

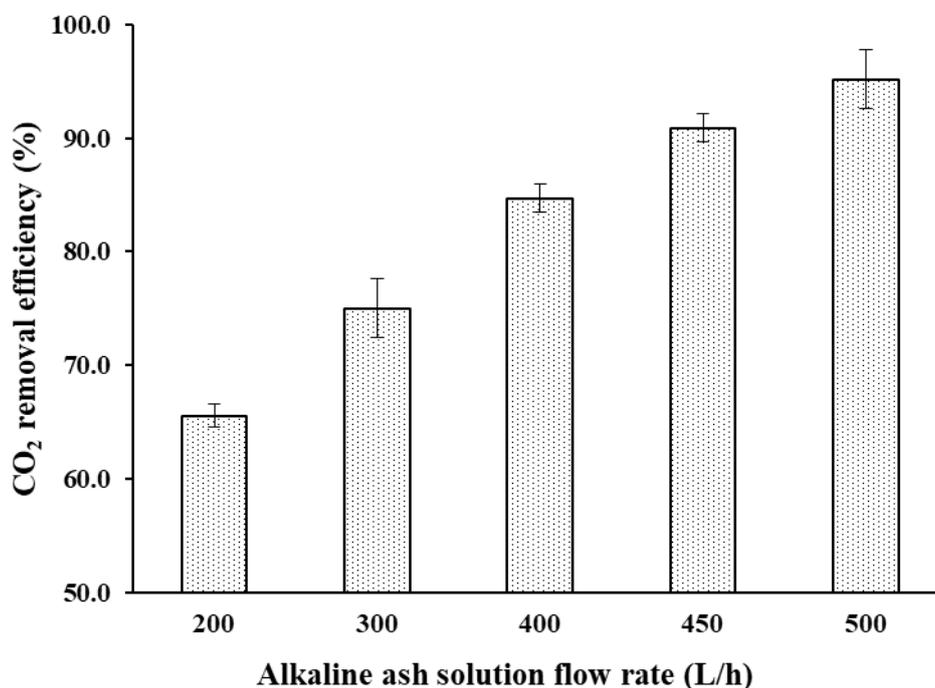


Figure 3. Carbon dioxide removal efficiency at various flow rates of the alkaline solution, with the gas flow rate fixed at 10 L/min

The highest observed CO₂ removal efficiency was 95% at alkaline solution flow rate 500 L/h, which gives L/G ratio 83.3 L/m³. Such high L/G ratio results in large pressure drop across the column, so a high-powered blower is required. However, the measured pressure drop was less than 8 cm H₂O. Removal of CO₂ by absorption into alkaline solution needs high L/G ratio because the solubilities of alkaline species (hydroxide and carbonate) in rubberwood ash are low. There are some other ways to enhance the CO₂ removal, such reducing the gas flow rate or increasing the packed bed height.

The pH and alkalinity of the alkaline solution before and after CO₂ absorption are shown in Table 2. It can be seen that the pH after CO₂ absorption was 7.2, which is almost neutral. In contrast, the total alkalinity before and after scrubber were not significantly different, but the form of alkalinity was changed from hydroxide and carbonate to bicarbonate species, by the following overall reactions (Eq. (2)-(3)),

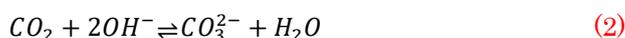


Table 2 pH and alkalinity of the alkaline solution of 30 g/L sifted ash (ash to water ratio of 0.03) before and after CO₂ absorption

Ash solution	pH	Total alkalinity (mg CaCO ₃ /L)	Approximate concentration (mM)		
			OH ⁻	CO ₃ ²⁻	HCO ₃ ⁻
Initial	12.4±0.04	2520±10	16.6±0.2	17.6±0.01	0
After CO ₂ absorbed	7.2±0.05	2533±5	0	0	50.67±0.1

It should be noted that metal carbonates and bicarbonates of sodium or potassium were solubilized in the absorbent solution, while some carbonates and bicarbonates of calcium, silicon, and magnesium precipitated due to their poor solubility.

Many processes can separate CO₂ from a gas mixture, and if the CO₂ is not used as an industrial gas to regenerate some material, it will contribute to atmospheric emissions. This research proposed the alternative of storing CO₂ as biomass, specifically in aquatic plants or algal biomass, because the used alkaline solution contains some macronutrients (P, K, S, Mg, Ca), micronutrients (Fe, Mn, Cu, Cl, Na, Al, Si), and bicarbonate species. With CO₂ abatement by storage into biomass, not only can minerals or heavy metals be removed from wastewater, but also new biomass is generated and it can be further used in renewable bioenergy production.

To compare the efficiency of the alkaline absorbent extracted from ash with a commercial alkaline absorbent, NaOH was chosen in this study as a baseline case for comparisons. pH is the main parameter characterizing the absorbent for reaction (2), so the NaOH solution was adjusted to the same pH 12.4 as the ash extract, and then fed into the column at the same operating conditions as used with the ash extract. The results clearly show that the absorbent solution obtained from extracting rubberwood ash had almost double the efficiency of sodium

hydroxide in CO₂ removal, at the same conditions. The CO₂ removal efficiency reached by alkaline rubberwood ash solution was 95% while sodium hydroxide reached 55%, as shown in Fig. 4., corresponding to 0.43 and 0.24 kg/h removal rates, respectively. The rubberwood ash extract was more alkaline than the NaOH solution, in the titrimetric capacity while not in pH, so it has more capacity for CO₂ acid absorption. In practice, NaOH can be prepared to much higher molar concentration due to good solubility, and this could give very efficient CO₂ removal, but regenerating the NaOH would be costly or impossible. The chemical quantity required for removal of one unit of CO₂ was also calculated. The amount of NaOH required is 2.08 kg NaOH/kg CO₂ removed (2.2 mol NaOH/mol CO₂ removed), which is much less than the 7.08 kg dissolved ash/kg CO₂ removed, or 35.4 kg sifted ash/kg CO₂ removed for rubberwood ash. In addition, an alkaline solution can remove other acidic gases such as H₂S, SO_x and NO_x. Tippayawong and Thanompongchart (2010) reported that CO₂ and H₂S were absorbed by NaOH, and remaining H₂S after biogas upgrading was below the detection limit (almost 100% removal efficiency), while CO₂ was removed with over 90% efficiency. Kuroпка (2011) studied the removal of nitrogen oxides from flue gases by sodium hydroxide. It was determined that over 90% of the nitrogen oxides were absorbed when adding an oxidant into the NaOH solution.

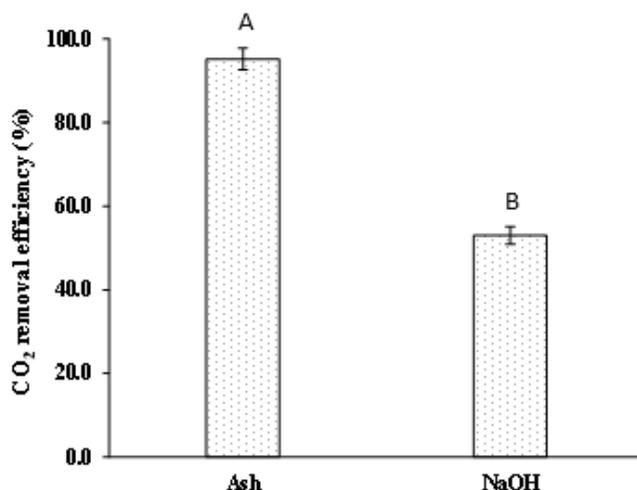


Figure 4. Comparison of carbon dioxide removal efficiency between alkaline rubberwood ash extract and aqueous sodium hydroxide as the absorbents, both with initial pH 12.4

Generally, if the packed column height is increased, the gas-liquid contact area increases and improves removal efficiency. Overall the gas film transfer unit height (H_{OG}) shows the dependence of efficiency on bed height. A low H_{OG} means the higher efficiency, as lower packing height is needed for the fixed removal rate, and this reduces investment costs and operating costs from driving the gas stream through the system. The efficiency of CO₂ removal from gas mixture by absorption found in this study is compared to prior literature in Table 3. The “inefficiency” in terms of H_{OG} , when using alkaline ash extract in this study, was about twice that of the other studies, because of the low L/G ratio and the poor solubility of ash.



However, the use of waste ash has both economic and environmental benefits. It should be noted that 60% of the ash sample was able passed through sieve mesh 10. At using 0.03 sifted ash to water ratio, the dissolution was 20%, so only 12% of original ash was used as absorbent in this study. This study used only ash extract and discarded the insoluble for the reason of preventing clogging of packed column. However, due to oxide of Ca, Si, Mg containing in the rubberwood ash, using ash suspension could be enhancing specific CO₂ removal and increase ash recycling quantity.

Table 3 Comparison of current study to prior studies on CO₂ removal from a gas mixture by absorption, using a packed column for scrubber

Reference	Absorbent	L/G ratio (L/m ³)	CO ₂ concentration (%)		Packed bed height (m)	HOG (m)
			Inlet	Outlet		
Zeng <i>et al.</i> , 2013	Aqueous NH ₃ 4.54M (77 g/l)	400	15.0	0.6	0.6	0.19
Tippayawong <i>et al.</i> , 2010	NaOH 0.1 M (4.0 g/L)	1,000	46.8	3.2	0.7	0.26
	Ca(OH) ₂ 0.1 M (7.4 g/L)	1,000	46.8	4.0	0.7	0.28
	MEA 0.1 M (6.1 g/L)	1,000	46.8	1.3	0.7	0.20
This work	Ash solution (6.12 g/L)	83.3	41.0	2.0	1.4	0.46

3.3 Proposed concept: use of rubberwood ash extract in alternative biofuel production

Most rubberwood ash from the power plant using rubberwood residue as fuel feedstock is discarded as waste. Approximately 16 ton/day of ash is generated by a 22 MW power plant. For the ash disposal, suitable land area is required, and the leachate needs to be neutralized by adding acid. An alternative scheme for waste ash utilization is proposed in Fig. 5. The water soluble compounds in ash are extracted to produce an alkaline filtrate solution, and the stable insoluble ash can be safely discarded to environment. The alkaline solution is then fed to an absorber to remove the acidic impurities in biogas. The upgraded biogas is fed to a combined-cycle power plant for electricity generation. Burning rubberwood or biomass also causes SO₂ and NO_x emissions, while the limits imposed on biomass power plants include: not over 60 ppm SO₂, 200 ppm NO_x, or 120 mg/m³ dust. So an alkaline absorption system is required for treatment of the flue gases containing NO_x, SO₂, and CO₂, which reduces also greenhouse gas emissions. The minerals rich wastewater, namely used alkaline absorbent, along with biogas plant effluents can be used in aquatic plant cultivation ponds. Select aquatic plants or macroalgae have potential to produce biofuels, such as *Chara* spp. (Baltrėnas and Misevičius, 2015). This wastewater treatment generates biomass that can be used as a substrate in biogas production. This conceptual scheme would combine waste conversion to energy with zero waste discharge, and a strongly reduced CO₂ emission.

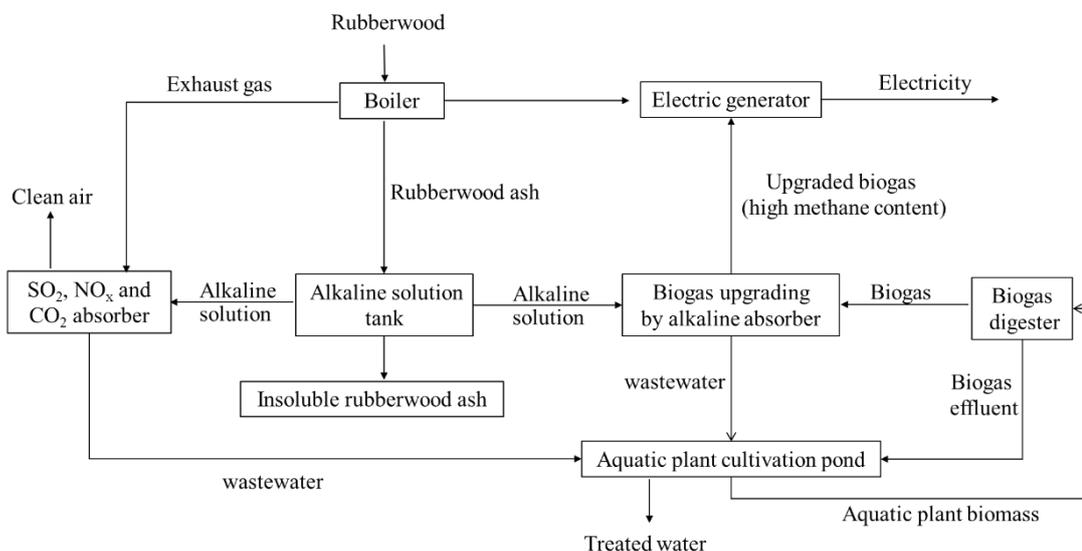


Figure 5. Conceptual scheme for application of rubberwood ash in scrubbing biogas, combined with alternative biofuel production for a biomass power plant

Conclusion

The alkaline solution prepared as rubberwood ash extract had pH up to around 12.4, with 20% dissolution of the ash, and the remaining insoluble ash could be safely discarded as it should no longer give toxic leachates to groundwater. The CO₂ removal efficiency reached with the alkaline solution obtained was better than 95%. The macronutrients and micronutrients in the alkaline solution, together with bicarbonate species from CO₂ scrubbing, make the absorbent effluent suitable for renewable generation of aquatic plant biomass. An integrated concept proposal for waste conversion to sustainable energy on industrial scale is discussed. Economic and environmental analysis of the technology demonstrated in laboratory scale will be pursued in our further work.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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