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<https://doi.org/10.5109/2349302>

出版情報 : Evergreen. 6 (3), pp.254-261, 2019-09. 九州大学グリーンテクノロジー研究教育センター
バージョン :
権利関係 :

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(Received April 22, 2019; accepted September 9, 2019).

Abstract: An instance growing of greenhouse gas emissions particularly CO₂ has been a global environmental issue that demands distinctive and practical actions to overcome. Tracking from the grass root of the great source of CO₂ emission, burning of fossil fuel can be indicted as the most influential contributor of anthropogenic activity. Thus, in addition to the reason of energy scarcity, it is necessary to massively shift the fossil fuel to renewable energy that introduces cleaner and environmentally benign energy resources. In this study, integration of biomass gasification (BG) and solid oxide fuel cell (SOFC) is applied to measure how much CO₂ emission can be reduced. However, the approach is not merely relying on renewability. It is correspondingly considered to abandon burning/combustion either partially or completely by utilization of the waste heat recovered from exhaust gas as well as excess heat from exothermic reaction to generate steam or external heat supply. In-situ utilization of CO₂ is also investigated in the systems. CO₂ emission is calculated by thermodynamic properties of each chemical compound that involves the material balance, energy balance, and chemical equilibrium. Three scenarios of BG-SOFC integration system are presented in this study. Among those 3 scenarios, lowest CO₂ emission with high electric power output and char-free of gasification products is obtained in the scenario 3.

Keywords: CO₂ emission, solid oxide fuel cell, biomass gasification, heat recovery.

1. Introduction

The environmental issue is a never-ending global subject to discuss, following the massive energy consumption that keeps showing an increase year by year. The more fuel is burned every year, the steep rebound of greenhouse gas emission is progressively accumulated over the atmosphere. If the actions in order to reduce the greenhouse gases could not at least keep the balance between emission and sequestration, our earth is most likely threatened by the consecutive effect of extreme climate change and global warming.

There are numbers of activities that release greenhouse gases (GHGs) emission either directly or indirectly into the atmospheres such as industrial processes, fossil fuel burning for transportation, deforestation, livestock, garbage treatment, fossil fuel production until its distribution, lime production, and other fossil fuel burning to generate electricity as well as refrigerant selection for air conditioning^{1,2)}. In addition to the hazardous potential of each GHGs, CO₂ seems to be the largest contributor to the global emission takes account for 65% were primarily released from fossil fuel and industrial process (Fig. 1)^{3,4)}.

Fig. 1: Global Greenhouse Gas Emission. Reprinted with permission from refs. 3 and 4. Copyright 2014 Intergovernmental Panel on Climate Change.

Naturally, the carbon cycle shows nearly net-rate between the uptake and the release as depicted in Fig. 2⁵⁾. Vegetation, ocean and the ground can store a considerable amount of CO₂ within a certain period of time, on average 10 years. This CO₂ storage will gradually discharge back to the atmosphere via decaying and respiration of the

plants. However, this natural carbon balance is disrupted by the anthropogenic activities that raises up the carbon dioxide emission with the rate of 2.1 Gt/year according to the data as of 2007-2016⁵). This fact promotes a deep concern on the concept of carbon-neutral that might be no longer sufficiently mitigate the GHGs accumulation on the earth. Basically, the concept of carbon-neutral is balancing the amount of carbon release and uptake as the consequence of human/industrial activities. The transition of fossil fuel to renewable energy has been introducing the concept of carbon-negative where the net of carbon rate is not only zero, but the carbon offset can be much greater than the release amount⁶). It is noteworthy to emphasize that CO₂ reduction is not merely focusing on the activity that avoids CO₂ release during the process nor capturing as much as CO₂ in the atmosphere and accordingly, store the CO₂ without any utilization.

Fig. 2: Global carbon dioxide budget (Gt-CO₂/year) 2007-2016⁵). Reprinted with permission from ref. 5. Copyright 2016 Le Quéré. This figure is an update of one prepared by the International Geosphere-Biosphere Programme for the Global Carbon Project (GCP), first presented in Le Quéré (2009).

Thus far, carbon-negative goal can be technically achieved by the following methods:

1. CO₂ capture by direct air capture (DAC)

Basically, the idea of DAC is imitating the principle of photosynthesis by using a chemical that has high affinity to CO₂, such as sodium hydroxide or calcium. When the air passes through the DAC system, CO₂ will be selectively scrubbed by the chemical, setting free the nitrogen, oxygen and another trace gases contained in the air. Subsequently, CO₂ can be easily stripped off from the chemical by applying high pressure. This CO₂ accordingly, can be utilized in the cement or plastic manufacturing process or sequestered underground⁷). The feasibility aspect shows the inefficiency of the system due to the huge

amount of energy required to capture the entire CO₂ in the atmosphere⁸). American Physical Society reports that in order to capture billion tons' scale of CO₂ by DAC, at least 10 GW of power is required which is equal to 3 times the capacity of the largest US nuclear plant⁹).

2. Biochar production

Production of biochar can favorably mitigate the CO₂ release from organic matter. Biomass is the starting material to produce biochar through pyrolysis, an environmentally friendly thermochemical process that avoids combustion during the process. By converting biomass into biochar, CO₂ emission can be prevented and another benefit can be obtained from the biochar utilization as a soil amendment. The carbon stored in biochar accounts for 41-64% of overall carbon avoided¹⁰⁻¹²).

3. BECCS (Bio-energy Carbon Storage and Sequestration)

BECCS technology integrates the biomass as a source of bio-energy and geological carbon storage by the loading of CO₂ underground. Unfortunately, in fact, this technology is less economical rather than a windmill and solar farm for CO₂ cutting¹³). It is also still unknown for how long and how large the capacity of our ground can keep CO₂ without any leakage to the air.

4. Ocean liming

Ocean liming is carried out by adding CaO to the ocean leading to the chemical reaction between CaO and CO₂ from the atmosphere to form CaCO₃¹⁴). Simultaneously, another environmental impact emerges due to the increase of pH resulting in acidification of the ocean.

5. Ocean fertilization

The main aim of this method is to increase the uptake of CO₂ from the atmosphere by the biological marine process. CO₂ is forcibly pumped into the deep ocean to supply the requirement of marine phytoplankton photosynthesis. In addition to the CO₂, essential micronutrient such as iron is also supplied to the ocean to give fertilizing effect for the marine plant growth¹⁵⁻¹⁷).

6. Synergistic biomass-nuclear process

This method is proposed by M. Hori¹⁸), which integrates the utilization of nuclear and biomass-based energy. Unlike the conventional CO₂ reduction by biomass utilization, the author suggests to include nuclear power based to supply the energy needs during biomass conversion to biochar and biofuel. He claimed that by using that system, the carbon removal efficiency increases at about 60% compared to the conventional one. Minimization of CO₂ emission can be actualized by replacing natural gas usage to generate heat or electricity¹⁸).

The shift of power generation or fuel and chemical production from fossil fuel to renewable sources would be advantageous for the CO₂ mitigation. Therefore, biomass is selected as the most potential feedstock since, in the natural cycle, biomass can absorb the CO₂ back from the atmosphere during the planting. In this study, combustion/burning would be abandoned or highly minimized to suppress and avoid the large emission of CO₂. There are very few studies highlight the significance of burning/oxidation in the unit operations of the systems they presented, associated to the contribution on CO₂ emission¹⁹⁻²¹). CO₂ is greatly emitted as the result of oxidation of fuel to generate heat and electricity. The highly endothermic gasification reaction obviously requires much amount of heat to enable the syngas production. Most of biomass gasification prefer partial oxidation to obtain the affordable and inexpensive large heat supply by feeding air with certain air to fuel ratio. On the other hands, direct oxygen/air feeding as the gasifying agent is less beneficial due to effect of nitrogen dilution over the producer gas. The other source of CO₂ pollutant in gasification is char burning. By means of char burning in separate unit, i.e., combustor, effect of dilution is easily prevented and the exothermic heat of combustion can be transferred to facilitate gasification. However, it may enhance the CO₂ production. In order to maintain the CO₂ level in the atmosphere, CO₂-emitting industrial-activity is commonly followed by a costly CO₂ sequestration. Rather than adding the unit of CO₂ isolation such in the case of sequestration, recycling and in-situ utilization of CO₂ in the system is much more economically efficient.

Direct utilization of syngas is proposed in this study in order to cut the process that is likely becomes the source of CO₂ emission such as storing and transportation of intermediate product to the next manufacturing unit. Fuel cell is capable of complete conversion of syngas into power with additional methane reforming ability. It is well-known as a clean energy production that only emits water as the side product. SOFC is typical of less expensive and more efficient fuel cell^{22,23}). SOFC can be fueled by hydrogen as well as carbon monoxide and methane to generate electricity. Another advantageous feature of SOFC is its high resistance to the impurities and high operating temperature that allows recuperation of the heat^{22,23}). The integration of BG and SOFC is expected to favorably mitigate the CO₂ emission. The approach to reducing CO₂ emission is not merely focusing on the renewability and integration, but also relying on the steam and heat generation by exhaust heat recovery extracted from exothermic reaction in the system. Moreover, the in-situ utilization of CO₂ for gasifying agent may greatly shrink the CO₂ pollutant.

The syngas composition may influence the composition of exhaust gas releases from SOFC since oxidation also occur inside the cell. In fact, SOFC is not as clean as expected, some CO₂ is generated either. From this study, we may discover how much contribution of

gasifier, combustor, and SOFC, respectively to the CO₂ emission.

2. Case Study

Some scenarios of integration between BG and direct utilization of syngas to generate electricity via SOFC is presented in this study. By excel-based thermodynamic calculation, the carbon footprint is tracked throughout the core process in biomass gasification. SOFC model is not calculated in detail.

In the entire process to convert biomass to generate energy, CO₂ is emitted directly and indirectly. Direct CO₂ emission in this study is reckoned as the chemical process that produces CO₂ as the side product other than the producer gas (CO, H₂, CH₄, and H₂O). In the syngas production, CO₂ is released during the water-gas shift reaction and combustion of the char to supply heat of endothermic reaction as presented in Table 2. SOFC also releases considerable amount of CO₂ during the oxidation of CO. However, this reaction rate is much slower than water gas shift reaction²³). Once the water is generated as a result of hydrogen-powered fuel cell, it will instantly initiate the reaction with CO and subsequently be converted to hydrogen and carbon dioxide.

While indirect CO₂ emission is triggered by the combustion of fuel to generate electricity, heat, steam and other utilities in order to support the running of overall processes. The abundance of CO₂ emission is created mainly in the gasifier to supply the endothermic heat of gasification process. Meanwhile, supplying air directly to the gasifier to allow partial combustion may lead to the dilution effect of nitrogen. In order to reduce the energy consumption in gasification, the heat is normally provided by burning fuel which largely causes CO₂ emission. Minimization of CO₂ emission can be achieved by eluding combustion as much as possible during the process. Therefore, the demand of endothermic heat of gasification can be fulfilled from various alternative ways such as, utilization of solid heat carrier that potentially transfers the heat from char combustor or energy recuperation from SOFC heat release to provide high temperature of gasifying agent. In this case, the gasifying agent has a double role as the heat carrier and gasifying agent. CO₂ mitigation is also realized by replacing steam with CO₂ as the gasifying agent.

Assumptions and given input data:

Within the boundary of the system, the following assumptions are made:

- The system is in steady state.
- The air composition is 21% O₂ and 79% N₂.
- The pressure of the system is 1 atm.
- No heat loss in the system.
- For the basis of calculation, the flow rate of the biomass input is given in 1 mole/s of carbon.
- Methane in the output stream of gasifier is fixed to 1 wt% and it is not in equilibrium.

- H_2 , CO and CH_4 are fully converted to product in SOFC.
- SOFC efficiency to produce electricity is 55%.
- Gasifier is operated at the temperature range of 500-800°C.
- Inlet temperature for fresh air, water and CO_2 is 25°C.
- Outlet temperature of SOFC exhaust gas is 1000°C.
- Temperature difference between the outlet cold fluid and hot fluid from heat exchanger (HX) is assumed to be 100°C.

The other input data are as follows:

Table 1. Input data for calculation.

	Biomass	Char
C, %daf	46.8	83
H, %daf	6.4	3
O, %daf	46.8	13
LHV, kJ/kg	18000	33000
Cp, kJ/kg/K	0.3	
T, °C	25	

Modelling:

The thermodynamic calculation involves material balance, energy balance and chemical equilibrium. The material balance is calculated based on each element account in every unit operation. For heat/energy balance calculation, the following information and formula are required:

$$Cp^\circ = A + Bt + Ct^2 + Dt^3 + E/t^2 \quad (1)$$

$$H^\circ - H_{298}^\circ =$$

$$At + Bt^2/2 + Ct^3/3 + Dt^4/4 - E/t + F - H \quad (2)$$

$$S^\circ = A \ln(t) + Bt + Ct^2/2 + Dt^3/3 - E/(2t^2) + G \quad (3)$$

Shomate equation²⁴⁾ (Eqs. 1-3) is the most widely used and accurate polynomial regression to estimate the thermo-chemical data. The coefficients are available in the NIST Chemistry Web Book²⁵⁾. Cp, Cp° , H° , H_{298}° , and S° represent heat capacity (J/mol/K), standard enthalpy (kJ/mol), standard enthalpy at 298 K (kJ/mol), and standard entropy (J/mol/K), respectively. Where t denotes temperature (K)/1000.

The gasifier modelling assumes that water-gas shift reaction is in chemical equilibrium. The chemical equilibrium can be obtained by the following equations:

$$\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ \quad (4)$$

$$\Delta G^\circ = -RT \ln K \quad (5)$$

ΔG° , ΔH° , and ΔS° is the change of standard Gibbs free energy, the change of standard enthalpy and the change of standard entropy, respectively. K and R denote chemical

equilibrium constant and ideal gas constant, respectively.

2.1 Scenario 1

Fig. 3: Integration of Allothermal BG-SOFC with steam generated from exhaust heat recovery (scenario 1).

The scheme of integration process in the scenario 1 can be seen from Fig. 3. The main gasification process occurs in the gasifier and combustor. The char remained in the gasifier is combusted separately in the combustor to avoid the tar formation that leads the problem in the gasifier. This gasification system is familiarly termed as allothermal gasification^{26,27)}. Indirect feeding of air to the combustor instead of gasifier beneficially prevents the dilution effect of nitrogen that may lower the yield of syngas. Combustor mainly plays a role as external heat source using circulating alumina sand to transport the heat from combustor to gasifier. The high temperature of the exhaust gas from combustor is recuperated to provide steam that is further fed to the gasifier to directly contact with the feedstock. SOFC also releases high temperature of exhaust gas as high as 1000°C to generate additional steam that is required for gasification. SOFC electrical efficiency in this system is assumed at the value of 55%. CO_2 is obtained as the exhaust gas as a result of oxidation of char in combustor and oxidation of CO as well as water gas shift reaction in SOFC.

2.2 Scenario 2

Fig. 4 presents the schematic diagram of scenario 2. Slightly different from the scenario 1, in the scenario 2 CO_2 is utilized to replace steam as the gasifying agent. CO_2 consumption is beneficial for the purpose of greenhouse gas reduction. The heat from combustor exhaust gas is used to preheat the fresh CO_2 before

Table 2. Reactions occur during BG-SOFC process

Process	Chemical reaction
Gasification with steam	$biomass + H_2O \rightarrow char + H_2 + CO + CO_2 + CH_4 + H_2O$
Gasification with CO ₂	$biomass + CO_2 \rightarrow char + H_2 + CO + CO_2 + CH_4 + H_2O$
Water-gas shift reaction	$CO + H_2O \rightarrow H_2 + CO_2$
Combustion of char	$char + O_2 \rightarrow CO_2 + H_2O$
SOFC	$2H_2 + O_2 \rightarrow 2H_2O$ $2CO + O_2 \rightarrow 2CO_2$ $CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O$ $CO + H_2O \rightarrow H_2 + CO_2$ $CH_4 + H_2O \rightarrow CO + 3H_2$

entering gasifier. The presence of HX-2 is removed from the system since the enthalpy to preheat gasifying agent is sufficiently supplied by HX-1.

Fig. 4: Integration of Allothermal BG-SOFC with CO₂ preheated by exhaust heat recovery (scenario 2).

2.3 Scenario 3

Fig. 5: Integration of Allothermal BG-SOFC with CO₂ preheated by exhaust heat recovery without combustor (scenario 3)

In the scenario 3, the presence of combustor as the external heat source is removed from the system (Fig. 5) to reduce CO₂ emission due to the combustion of char.

Heat supply for the endothermic reaction in gasification is substituted by SOFC. Circulation of solid

heat carrier occurs between gasifier and SOFC. Preheating of CO₂ before entering gasifier occur in the heat exchanger with the SOFC exhaust gas as the hot fluid that provides the sensible heat for CO₂. The scenario 3 demonstrates the absence of burning in the system. However, unavoidable oxidation still exists in the SOFC.

3. Results and Discussion

In the previous studies, it was found that the major share of CO₂ emission in the biomass/coal to syngas/electricity is the direct emission during gasification in the range of 79-85% of the entire CO₂ emission in every stage²⁸⁾. Therefore, in this study particularly focuses on the gasification process. In the most presented cases²⁸⁻³⁰⁾, chemical/fuel production is always equipped with the unit of CO₂ sequestration, and hence it will significantly eliminate CO₂ emission up to 72-100%²⁹⁻³¹⁾ depending upon the technology of CO₂ isolation. However, the technology requires a costly high pressure separation and further transportation of the liquid CO₂ to the next treatment site or storage site³¹⁾. Hence, in this study in-situ utilization of CO₂ is studied to evaluate how much the reduction of CO₂ emission can be performed.

The calculation is obtained by collecting thermal properties of each chemical substance obtained from NIST Chemistry Web book²⁵⁾ and constructing equations that include material balance, energy balance, and chemical equilibrium. Basically, the result refers to the theoretical yield and performance parameter that is able to achieve. Cold gas efficiency (CGE) is the common parameter to investigate the performance of gasifier on how effective it is to convert the fuel energy content into the chemical. According to the result in Table 3, carbon in the biomass is stored in three forms of final product, those are CO₂, char and electric power. These three products show a resultant as an effect of CO₂ utilization/circulation and energy recuperation. Integration of BG and SOFC involves the conversion of chemical energy of biomass to syngas and further is transformed into electric power and

Table 3. The comparison of CO₂ emission and performance of the scenario 1, scenario 2 and scenario 3.

		Gasifying agent	T gasifier, °C	kg-CO ₂ /kg-syngas	CGE	Char (wt%)	Power generation kW/mol-C biomass
scenario	1	H ₂ O	500	2.81	0.89	11.72%	227.02
			600	2.94	0.87	12.97%	220.38
			700	3.08	0.84	14.22%	213.69
			800	3.23	0.82	15.49%	206.96
	2	CO ₂	500	1.50	0.84	16.81%	214.22
			600	1.56	0.82	18.02%	207.30
			700	1.63	0.79	19.20%	200.55
			800	1.70	0.76	20.35%	194.01
	3	CO ₂	500	0.96	1.22	0.00%	310.32
			600	0.96	1.22	0.00%	310.32
			700	0.96	1.22	0.00%	310.31
			800	0.96	1.22	0.00%	310.31

heat. The heat dissipation of SOFC, and then, is recovered to preheat the gasifying agent. The scenario 1 tends to release more CO₂ among the other two scenarios.

It results in the slightly higher CGE and power than that of the scenario 2, however, the char is lower than the scenario 2. CO₂ emission is an accumulation of exhaust gas emitted from combustor and SOFC. Although the syngas composition in the scenario 1 is greatly dominated by hydrogen, SOFC is responsible for the highest share of CO₂ emitter (Fig. 6).

Despite the relatively lower char yield in the scenario 1, CO₂ emission contribution primarily originated from SOFC (see Fig. 6) at the range of 43-50% of CO₂ emission. In the integration of BG-SOFC, char is required to supply endothermic heat of gasifier by combustion. Therefore, at the 500°C of gasifier temperature, only a few of char amount is required to provide the heat for gasification. The low char yield indicates the high biomass conversion to syngas (see Fig. 7). Thus the electric power generated from SOFC increases as the operating temperature decreases. In the scenario 1, the optimum operational condition that gives the lowest CO₂ emission, highest CGE and power generation is achieved when the gasifier temperature is 500°C.

The less CO₂ emission can be favored when CO₂ is subjected as the gasifying agent as presented in the scenario 2 which shows the number of CO₂ emission decreases to half of the steam gasifying agent. Gasifier in the scenario 2 shows negative CO₂ emission with the share 14% of CO₂ emission (Fig. 6). However, it is accompanied by less electric power. Thus, the scenario 2 is inferior than in the scenario 1. In other words, in situ utilization of CO₂ with less heat recovery is not beneficial to improve the electrical power output and efficiency of gasifier performance.

Meanwhile, the scenario 3 seems more promising than other two scenarios. In the scenario 2 combustor supplies 90% and 97% of total heat requirement for gasification at gasifier temperature of 500°C and 800°C, respectively. While the rest of the heat is compensated by SOFC. This fact attributes to the high heat content of combustor carried by the high thermal conductivity of alumina sand. Hence, the presence of combustor as the primary heat

Fig. 6: CO₂ emission distribution in each unit operation

source of gasifier can be replaced by SOFC that releases high temperature up to 1000°C. It is confirmed that omitting combustion in the system can greatly reduce CO₂ emission.

Interestingly, a complete conversion of biomass by CO₂ gasifying agent can be realized in the scenario 3 remaining no char at the end of the gasification process. Replacement of combustor by SOFC as endothermic heat supply for gasifier, results in unnecessary of char presence. In scenarios 1 and 2, char is required to provide exothermic heat after it is combusted in the combustor. This heat accordingly subjected to supply the heat of endothermic gasification. This result is confirmed by Pohorel'y et al³²⁾ who found a substantial increase in carbon conversion and energy efficiency when CO₂ is fed to the gasifier. CGE is above the unity (1) implies that the total heat contained in the producer gas is greater than the heat of feedstock³³⁾. The percentage of CO₂ reduction in the gasifier of the scenario 3 (Fig. 6) is greater than the scenario 2 since the total CO₂ emission is much lower after the elimination of combustor. Apart from the operating temperature of gasifier, it releases the lowest CO₂ emission with high electric power output.

Fig. 7 exhibits the distribution of syngas product from the scenarios 1, 2 and 3. From this bar diagram, it can be concluded that syngas composition for dry reforming (the scenarios 2 and 3) predominantly produces CO rather than H₂. This result is in agreement with the finding on the gas evolution product as the effect of CO₂ introduction in the gasification of biomass that showed hydrogen production shrinkage, while CO production enhances³⁴⁻³⁶⁾. The high CO content in SOFC feeding stream may increase the possibility of water gas shift reaction that produces H₂ and CO₂ as well as CO oxidation that furthermore contributes to the enhancement of CO₂ emission. However, the CO₂ emission level of dry reforming (the scenarios 2 and 3) is still below the level of CO₂ emission of the scenario 1 due to in-situ utilization. In fact, CO₂ is consumed during gasification in the scenarios 2 and 3.

The realization of the scenario 3 requires a modification of SOFC design that allow indirect contact of cell and alumina sand so that recuperation of excess heat of SOFC is possibly attained. According to this result, the author believes that additional unit of CO₂ utilization in BG-SOFC system such as chemical production can effectively reach carbon negative goal with or without sequestration. In the wider scope of either analysis or integration system, the benefits of CO₂ reduction will be doubled with the co-production of high-added value product such as chemicals and power which inevitably increases the economic value of the entire process.

Fig. 7: Syngas distribution at varies temperature.

4. Conclusions

The approach of CO₂ reduction has been proposed by the integration of biomass gasification and solid oxide fuel cell, in addition to the in-situ utilization of CO₂ formed in the process. The approach introduces the elimination of combustion/burning as the heat/ energy supply in the system. The energy recuperation/ heat recovery can be supplied by the heat from exothermic reaction/ exhaust gas. Biomass gasification produces syngas that is further used to generate electric power by solid oxide fuel cell, the clean renewable energy. Carbon is stored in the form of char, electric power and exhaust gas. The scenario 3 seemingly shows the most preferable option as lowest CO₂ emission can be attained, and char as the undesirable product can be fully converted to syngas, and hence, maximized electric power is possibly gained. In the future scenario, modification of the scenario 3 is expectedly resulting in completely-free CO₂ emission or even carbon negative emission. The realization can be achieved by eliminating CO reduction in SOFC and utilize it for higher added value product.

Acknowledgements

The author would like to show the gratitude to the Advanced Graduate Program in Global Strategy for Green Asia for the financial support provided by the Kyushu University.

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