Energy System Simulation of Regenerable Molten Alkali Carbonates for Gasification of Carbonaceous Resources

by Slamet Raharjo

ENERGY SYSTEM SIMULATION OF REGENERABLE MOLTEN ALKALI CARBONATES FOR GASIFICATION OF CARBONACEOUS RESOURCES

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ABSTRACT

This paper discusses the energy system simulation of Molten Alkali Carbonates (MACs) gasifier based on our previous experimental results. They showed that the MACs can be used as catalyst and hot gas desulfurizer in a gasification of carbonaceous resources. Meanwhile, the used MACs sorbent could be regenerated either. HI-System was used for simulating the energy system of MACs in the current study. First of all, two unique components of MACs gasifier (MG) and used MACs regenerator (MR) were created and incorporated into the HI-System software. Two types of energy system including MACs system without MR and the other one with MR were constructed, simulated and compared. The result suggested that MACs system with MR achieves gross power generation efficiency of around 55 %, which is 6 % higher than that of the other one without MR due to a waste heat recirculation.

Keyword: molten alkali carbonates, hot gas desulfurization, gasification system, used MACs regeneration, power generation efficiency.

1. INTRODUCTION

Currently, increasing the power generation efficiency of a gasification system is an important issue. A combined cycle power generation featuring gas turbine, steam turbine and fuel cell as one network system integrated with the gasification facility has been considered the best method so far for increasing the power generation efficiency. Based on the feasibility study in 1995, it was concluded that an oxygen-blown gasifier would be suitable for integrated gasification fuel cell combined cycle (IGFC) because its net thermal efficiency with an oxygen-blown gasifier would be 53 % or more [1].

Generally, power generation efficiency is defined as a value which reveals how much of the energy contained in a fuel (coal or natural gas) is output as a useful product, often electricity [2]. There are two terminologies often used to express the power generation efficiency of a system, gross efficiency and net efficiency, which are based their value on higher heating value (HHV) and lower heating value (LHV). Power generation system is a network flow of mass and energy balance. It is built by connecting some unit operations (e.g. air separation unit, gasifier, gas turbine) in which each unit operation has unique physic and chemical processes, and operating conditions. The way in selecting and arranging the unit operations, as well as setting up the operating conditions determines the power generation efficiency of the system.

Fundamental study on hot gas desulfurization, regeneration and gasification characteristics with molten alkali carbonates (MACs) have been carried out in our previous works [2, 3, 4, 5]. Eutectic salt with the compositions of 43 mol% Na_2CO_3 - 57 mol% K_2CO_3 was installed in the gasifier. It can gasify carbonaceous materials and remove gaseous sulfur even at high temperatures simultaneously. Meanwhile, the introducing a mixture of steam and CO_2 at 773 K could regenerate the used alkali carbonates to around 95% [4]. The studies suggested that it is possible to develop the alkali carbonates as a main component of a gasification process. Therefore, it is necessary to continue with the study on the performance of alkali carbonates gasification as a system. In order to get the optimum condition for creating the highest possible power generation efficiency of the system, some trials of system simulation is required, which is always a time consuming work. The use of system simulator software is very helpful in dealing with this work. The use of specific simulation software, such as Aspen Plus, ChemCAD, etc., offers a powerful tool for the analysis of existing processes, synthesis of new processes, implementation of a control strategy and fast screening of process alternatives to select the best solution [6]. Generally, the system simulation models are capable of performing mass and energy balance which may be used to trace system inefficiencies to their source component thereby providing insights into component interactions within the cycles and act as pointers to system optimization trade-offs [7].

Simulation of biomass gasification in fluidized bed reactor using Aspen Plus was carried out by Nikoo et al. They could simulate the effect of temperature on the production of hydrogen and carbon conversion efficiency [7]. Doherty et al. developed a model of a biomass-SOFC system capable of predicting performance under diverse operating conditions by using Aspen plus [8]. The model performs heat and mass balances and considers ohmic, activation and concentration losses for voltage calculation. Ongi`ro *et al.* developed a computer simulation model in Aspen Plus to simulate the performance of IGCC and IGHAT cycle power plants [9]. Another study on Aspen Plus simulation was also carried out by Doherty *et al.* They studied the effect of air preheating in a biomass CFB gasifier. The model could be used to predict syn-gas composition, conversion efficiency and heating values in good agreement with experimental data [10]. However, the molten bath gasifier requires

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certain simulation condition due to its properties and behavior at high temperature. In case of MACs gasifier, new unit process (component) must be created and incorporated to the original model based on the findings from the previous experimental results. HI-System simulator was used in this study due to its source code with full access. In order to create a system simulator with full access by the users, Hijikata developed the HI-System simulator. It is not a commercial software, but with full access to its source code by using Microsoft visual C++ version 6, run under windows platform in order to get it more versatile [11].

HI-System simulator is based on steady-state model, element of energy system is transformed into component. Each component is arranged manually on the screen so that energy system can be constructed by connecting one component to another. This simulator especially focuses on a coal-fired power plant evaluation, primarily pulverized coal-fired power (PFC), coal gasification combined cycle (IGCC) and pressurized fluidized bed combustion (PFBC) power generation system. HI-System component receives the input variable conditions from the inlet side, and determines the output conditions at the outlet side. In this program, it is assumed that there is no heat and work loss on lines/pipes connecting between the components. However, it is possible to consider the pressure loss by setting it into the component so that the mass, energy and momentum can be satisfied conserved. Inside the source code of HI-System, enthalpy and entropy of each element are defined as a function of temperature [11].

2. METHODOLOGY

Previous experimental data on the fundamental characteristics of MACs gasification process such as optimum temperature, sodium - potassium sulfide ratio, regeneration efficiency, etc. were required for setting up the new components and simulation condition. Original HI-System software does not have MACs gasifier (MG) and used MACs regenerator (MR) components. Therefore, the new components must be created and incorporated into the original HI-System software prior to simulations.

Simulation conditions refer to some certain restrictions that must be fulfilled when doing system simulation. In this MACs gasification system, some simulation conditions are applied including:

- Operating temperature of MG must be maintained at higher than 700 °C, which is the melting point of alkali carbonates ($43Na_2CO_3.57K_2CO_3$) [2].
- Operating temperature of MR must be maintained at around 500 °C, which is the optimum regeneration temperature under a mixture of CO_2 and steam as the regeneration agent [4].
- Gas turbine used in this simulation treated as a 1700 °C-class GT which is a super high temperature gas turbine with a maximum operating temperature of 1700 °C.
- Steam turbine has a maximum operating temperature of 700 °C.

Input conditions for HI-System simulation includes an arrangement of a network which connects the component to each other creating a system flow and input parameters of each feeder and component. A description of each component utilized in HI-System simulation is given in **Table-1**, while coal characteristics and input parameters are given in **Tables-2** and **3**, respectively. Two types of MACs gasification system flow were constructed, simulated and compared in this paper as follows:

Table-1. Description of HI-System components.

Symbol	Component name	Description
F	Feeder	Feeding the fluid (including solid fuel)
AD	Adder	Connecting the fluid from two pathways.
ASU	Air separation unit	Separating atmospheric oxygen and nitrogen
CP	Compressor	Increasing temperature and pressure of the gas
MG	MACs gasifier	Simulating gasification and desulfurization reaction
MR	Used MACs regenerator	Simulating regeneration reaction of used MACs
AS	Ash separator	Separating ash from fluid
SE	Separater	Separating gas from liquid or solid
GBNR-TB	Gas turbine	Extracting the working gas by burning the syngas in order to get the external power (electricity)
HEX	Heat exchanger	Performing the heat exchange between the two pathways independent of each other
HEX-TB	Steam turbine	Extracting the working gas by using heat for steam in order to get the external power (electricity)
PM	Pump	Compressing the liquid (water)

Table-2. Properties of coal used in HI-System simulations.

Proximate analysis	dry. wt%	ultimate analysis	d.a.f. wt%
Moisture	3.7	Carbon	69.8
VM	31.8	Hydrogen	4.9
FC	50.5	Nitrogen	1.8
Ash	14.0	Sulfur	0.5
		Oxygen	5.4

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Table-3. Input parameters for HI-System simulations.

MACs gasifier (MG)	
Gasification ratio	0.9
Temperature [°C]	>700
Pressure [atm]	30
Coal [kg/s]	2
Na_2CO_3 [kg/s]	2
K_2CO_3 [kg/s]	2
Used MACs regenerator (MR)	
Regeneration ratio	0.95
Temperature [°C]	500
Gas turbine	
Inlet temp. [°C]	1700
Adiabatic efficiency	0.9
Steam turbine	
Inlet temp. [°C]	700
Adiabatic efficiency	0.9

a) MACs basic system.

This is a standard oxygen-blown gasification system. MG was installed instead of common gasifier, while MR was not installed. **Figure-1** shows the system flow.

b) MACs system with regenerator.

This system features Oxygen-blown gasification with MG and MR components installed. The flue gases (Steam and CO_2) inside the system were introduced into the MR component to convert alkali sulfides to alkali carbonates. The regenerated alkali carbonates were recycled into the MG component. This system is an improvement of the basic system. The system flow is illustrated in **Figure-2**.

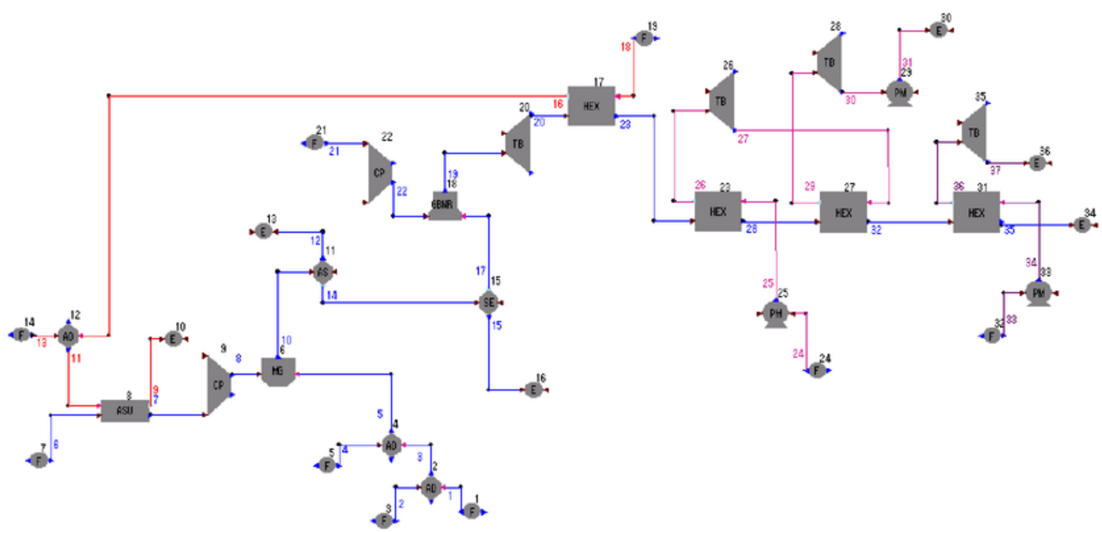

Figure-1. System flow of MACs basic gasification system.

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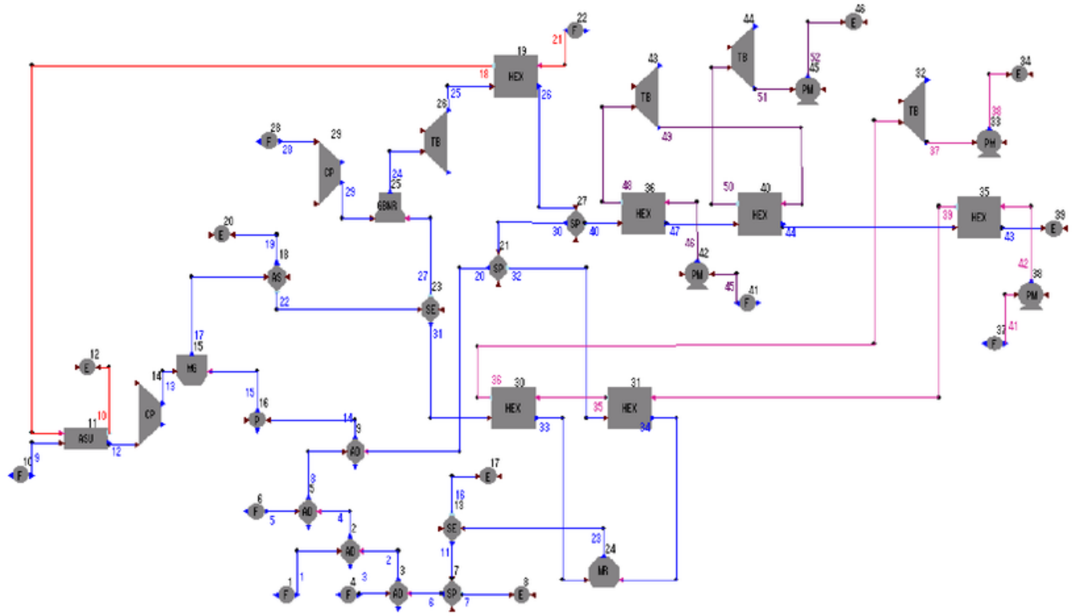

Figure-2. System flow of MACs gasification system with regenerator.

3. RESULT AND DISCUSSIONS

3.1 Create and incorporate new components

MACs gasifier (MG)

MG component was created for simulating a molten alkali carbonates gasifier. As suggested by the previous experimental results, this component is capable of simultaneously doing gasification and desulfurization process of carbonaceous resources. Some simulation conditions are applied to this MG component including:

a) Gasification ratio is set at > 0.9. It refers to how many proportion/ratio of carbon in coal is converted to CO and CH_4, while the rest is CO_2.

b) Sulfur content in coal is all converted to H_2S and COS with the ratio of 0.92 and 0.08. The ratio between H_2S and COS in raw syngas is based on typical data of EAGLE project.

MG component has two input lines and one output line. Each line contains variables of mass, energy and pressure. The illustration of mass and energy flow are given in **Figure-3**. Mass (M), energy (H) and pressure (P) in input and output can be balanced as shown in the following equation:

Mass Flow:

M_{in1} = *Coal + O_2 + recycled flue gas (CO_2, H_2O)*

M_{in2} = *Coal + O_2 + Fresh and regenerated $43Na_2CO_3.57K_2CO_3$ (molten alkali carbonates)*

M_{out1} = *Gasified gas (H_2, CO, CH_4, CO_2, H_2O, H_2S, COS) + drainage (M_2S + Ash)*

Enthalpy:

$H_{in1} + H_{in2} = H_{out1}$

Pressure:

$P_{out1} = M_{in1} (P_{in1}, P_{in2}) \sim 15\ atm$

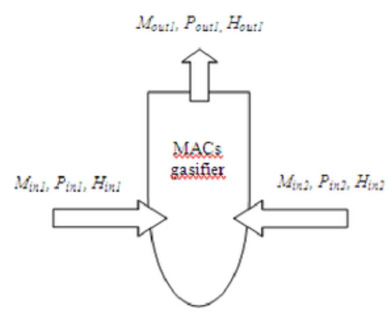

Figure-3. Mass and energy flow of MG component.

The relation between energy (enthalpy), mass and temperature can be expressed by the famous equation $H = m \times C \times T$, where *H*: energy required in Joule, *m*: mass in grams, *C*: specific heat capacity in J/g/°C and *T*: temperature in °C.

Partial oxidation, combustion and desulfurization reactions were simulated inside MG as described in **Figure-4**. The partial oxidation and combustion are simulated according to reaction $C+1/2O_2 \rightarrow CO$, $C+H_2O \rightarrow CO+H_2O$ and $1/2O_2excess+CO \rightarrow CO_2$. Hot gas desulfurization by molten alkali carbonates is also simulated in this component. Sulfur species in coal is first converted to H_2S and COS, then reacted with Na_2CO_3 and K_2CO_3 according to reaction [2]: $0.43Na_2CO_3+0.57K_2CO_3+0.92H_2S+0.08COS \rightarrow 0.43Na_2S +0.57K_2S+0.92H_2O+1.08CO_2$.

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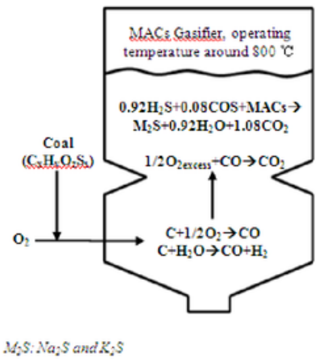

Figure-4. Chemical reaction scheme inside MG component.

Used MACs regenerator (MR)

MR is a component for simulating the regeneration reaction of used MACs. Some simulation conditions applied to this component are:

a) Regeneration efficiency is set at 0.95. It refers to the ratio of alkali sulfides (Na_2S and K_2S) converted to alkali carbonates (Na_2CO_3 and K_2CO_3)[4].

b) Based on desulfurization simulations and experiments in our previous study, sulfur content in gaseous sulfurs is converted mainly to alkali sulfides (Na_2S and K_2S) at around gasification temperature 1173 K) [2].Therefore, the composition of alkali sulfides in used MACs is similar to alkali carbonates, $0.43Na_2S$ and $0.57K_2S$.

The mass and energy flow illustration are given in **Figure-5**. Mass (M), energy (H) and pressure (P) in input and output can be balanced as shown in the following equation:

A. Mass Flow:

M_{in1} = *Used MACs (Na_2S, K_2S) from MG*

M_{in2} = *CO_2 + steam from flue gas (exhaust of gas turbine)*

M_{out1} = *Na_2CO_3, K_2CO_3, H_2S*

B. Enthalpy:

$H_{in1} + H_{in2} = H_{out1}$

C. Pressure:

$P_{out1} = M_{in1} (P_{in1}, P_{in2}) \sim 1\ atm$

The alkali sulfides (Na_2S, K_2S) formed inside MG from desulfurization process are directed into MR. CO_2 and steam from flue gas are injected into MR to regenerate the used MACs.

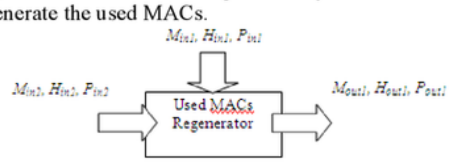

Figure-5. The mass and energy flow illustration of MR component.

3.2 Simulation process

The flowchart output of the system simulation for MACs basic gasification system and MACs gasification system with regenerator are given in **Figures-1** and **2**, respectively. The calculation results of mass flow rate of each element and thermodynamic properties on each line are given in **Tables-4** and **5**, respectively. HI-System also provides the performance calculation of each component and system. It can be referred to **Table-6**. It displays heat input of coal, electrical input for ASU and compressor and electrical output from gas turbine and steam turbine. Gross electrical output is calculated by subtracting the electrical output from gas turbine and steam turbine with the electrical input for ASU and compressor, because in HI-System their electrical input is considered as part of electrical power used in practical turbine. Furthermore, by comparing the electrical output with the heat input without considering the heat and pressure loss, theoretical gross power generation efficiency on HHV basis can be calculated.

MACs basic system simulation is aimed for evaluating the basic system of MACs gasification process. As illustrated in **Figure-1**, air separation unit (ASU) is used to produce oxygen to initiate the partial oxidation of coal inside MG component to produce synthesis gas. Then, it is used by the combined-cycle power generation to produce electricity. **Table-4** shows that line 10 in **Figure-1** had no O_2 and gaseous sulfurs (H_2S and COS). It was caused by the consumption of O_2 to produce CO and H_2 in MG component. The gasification process was conducted at relatively low temperature (around 880°C) as indicated in **Table-5** for line 10. Meanwhile, the H_2S and COS produced from the gasification reaction are captured by the alkali carbonates (Na_2CO_3 and K_2CO_3) to form alkali sulfides (Na_2S and K_2S). Therefore, the gasified gas is readily free of gaseous sulfurs at high temperature and directed into the gas turbine to produce electricity.

Table-5 shows that temperatures of exhaust gas from gas turbine (lines 20 and 25) in all MACs systems are still high enough to power steam turbine to generate the additional electricity. This table also confirms that all simulation results meet the simulation conditions including the operating temperatures for MG component (lines 10 and 17), the inlet temperatures for gas turbine (lines 19 and 24), the inlet temperatures for steam turbine (lines 26, 29, 36; lines 36, 48, 50) and the operating temperatures of used MACs regenerator (lines 23).

MACs system with regenerator is an improvement from the basic system. As displayed in **Figure-2**, MR component is installed so that the used MACs can be regenerated and recycled. Additionally, flue gas, which contains steam and CO_2 is also recycled into the MG component and utilized as regeneration agent for MR component. **Table-4** shows a decrease of Na_2S and K_2S and an increase of Na_2CO_3 and K_2CO_3 in lines 17 and 23, which confirms that Na_2S and K_2S are converted into Na_2CO_3 and K_2CO_3 in this simulation.

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Table-4. Mass flow-rate of each material.

		MACs basic		MACs with regenerator		
Line		10	19&20	17	24&25	23
Exhaust from component		MG	GBNR-TB	MG	GBNR-TB	MR
Element[kg/s]	N_2	0.08	28.86	1.21	28.43	4.55
	O_2	0	5.77	0	5.29	0.85
	H_2	0.07	0	0.07	0	0
	H_2O	0.14	0.88	0.15	0.89	0.14
	CO_2	0.32	5.09	0.23	5.11	0.81
	CO	2.94	0	3.01	0	0
	CH_4	0.06	0	0.06	0	0
	H_2S	0	0	0	0	0.01
	COS	0	0	0	0	0
	Na_2S	0.01	0	0.01	0	0.0004
	K_2S	0.02	0	0.02	0	0.001
	Na_2CO_3	1.99	0	2.49	0	2.50
	K_2CO_3	1.98	0	2.48	0	2.50

Table-5. Thermodynamic properties on each line.

MACs basic system			MACs system with regenerator		
Line	**P (atm)**	**T (°C)**	**Line**	**P (atm)**	**T (°C)**
10	30	880	17	30	766
19	30	1682	23	1	480
20	1	691	24	30	1719
26	350	645	25	1	710
27	100	428	36	350	540
29	100	539	37	0.05	33
30	0.05	33	48	350	662
36	350	508	49	50	346
37	1	100	50	50	530
			51	0.05	33

3.3 Power generation efficiency

The main outcome of HI-System software is to assess possible high efficient system by simulating its energy balance. As given in **Table-6**, theoretical gross power generation efficiency on HHV basis of MACs basic system can be calculated as high as 49 %. When the basic system is improved by adding the MR component and CO_2 recycle in MACs system with regenerator, the power generation efficiency can be increased by a margin of around 6 %. The recycling process of flue gas and used MACs would increase the power generation efficiency more due to waste-heat utilization and an increase of syngas production. This utilization of used MACs for many times would increase the economic value.

Current simulation with HI-System software for MACs system with regenerator, which results in gross power generation efficiency (HHV basis) of around 55 % without the addition of fuel cell component and calculation of heat and pressure loss, is considered comparable with the gross power generation efficiency (HHV basis) of IGFC, which is around 55 %. However, new component addition of fuel cell into the HI-System software for future simulation of the MACs gasification system fuel cell combined cycle (MGFC) is necessary.

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Table-6. Performance calculation of each component and system.

MACs basic								
Components	F(coal)	ASU	CP1	GBNR-TB	CP2	HEX-TB1	HEX-TB2	HEX-TB3
Win [kW]	0	3,887	0	0	20,658	0	0	0
Wout [kW]	0	0	0	41,953	0	572	2,049	4,708
Hin [kW]	50,941	0	0	0	0	0	0	0
Gross electrical output (Wout-Win) [kW]					24,737			
Gross power generation efficiency on HHV basis [%]					49			
MACs system with regenerator								
Components	F(coal)	ASU	CP1	GBNR-TB	CP2	HEX-TB1	HEX-TB2	HEX-TB3
Win [kW]	0	3,512	0	0	19,541	0	0	0
Wout [kW]	0	0	0	41,631	0	6,887	801	1,820
Hin [kW]	50,941	0	0	0	0	0	0	0
Gross electrical output (Wout-Win) [kW]					28,086			
Gross power generation efficiency on HHV basis [%]					55			

4. CONCLUSIONS

Molten Alkali Carbonates (MACs) basic system features the standard system without any materials recycling. Theoretical gross power generation efficiency on HHV basis of MACs basic system can be calculated as high as 49 %.

MACs system with regenerator features MACs basic system with regenerated alkali carbonates and CO_2 recycle. The power generation efficiency can be increased by a margin of around 6% compared to the MACs basic system because of waste-heat utilization and an increase of syngas production. This utilization of used MACs for many times would increase the economic value.

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