

## Energy analysis of a CO<sub>2</sub> recycling system

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### SUMMARY

In this report, a CO<sub>2</sub> recycling system is proposed and designed for the purpose of CO<sub>2</sub> mitigation through utilization of natural energy (photovoltaic power generation). The materials for construction and unit requirements of this system are quantified and also an energy analysis is performed as one of life-cycle analysis (LCA) of this system. Concrete required for the foundation of the facilities is the largest material requirement and accounts for 62 per cent of the total amount of materials for building this system. The energy consumption needed to build the photovoltaic (PV) power generation facilities represents the largest fraction of energy consumption and accounts for 82 per cent of the energy consumption needed for building of plants. The energy balance ratio of the system is approximately 1.85. It clearly reveals that this system would be an effective way to utilize natural energy. Copyright © 2000 John Wiley & Sons, Ltd.

KEY WORDS: CO<sub>2</sub> recycling system; photovoltaic power generation; energy analysis; energy balance ratio

### 1. INTRODUCTION

The recent substantial rise in global atmospheric CO<sub>2</sub> concentration and the ensuing greenhouse effect is considered to be caused mainly by consumption of large amounts of fossil fuels due to increased human activities. In particular, power plants and ironworks consume large amounts of fossil fuels and emit large amounts of CO<sub>2</sub>. Recovery of CO<sub>2</sub> from these stationary CO<sub>2</sub> emission sources is one strategy for reducing CO<sub>2</sub> emissions.

On the other hand, CO<sub>2</sub> can be regarded as a carbon resource. Methanol (MeOH) synthesis via CO<sub>2</sub> hydrogenation has recently received much attention as one of the most promising processes for the fixation and utilization of CO<sub>2</sub>. Furthermore, MeOH utilization has promising future prospects as an alternative fuel and a chemical raw material.

We have been developing a CO<sub>2</sub> recycling system utilizing natural energy (e.g. solar energy and hydraulic energy) as a CO<sub>2</sub> mitigation system, as illustrated in Figure 1. This system is composed of five parts, (1) CO<sub>2</sub> separation/recovery via membrane separation in large amounts of CO<sub>2</sub>

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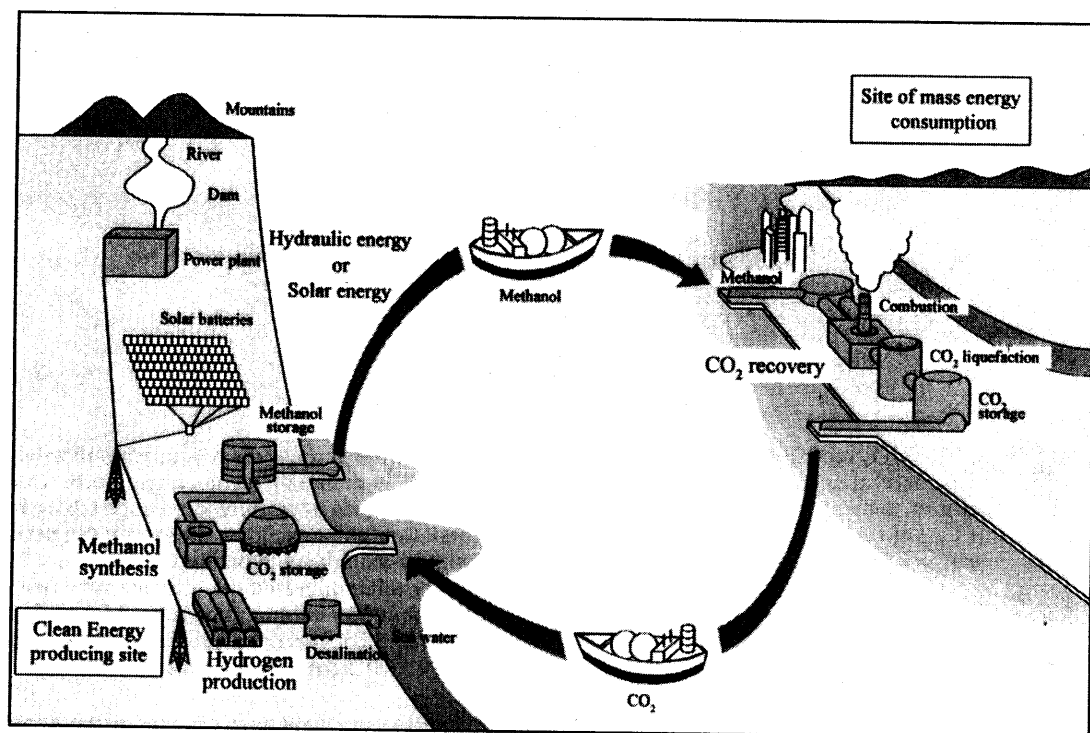


Figure 1. Concept of CO<sub>2</sub> recycling system.

emitted from stationary source, (2) tanker transportation of the CO<sub>2</sub> to a coast close to a desert, (3) hydrogen (H<sub>2</sub>) production by water electrolysis generated by such as a natural energy, (4) MeOH synthesis from CO<sub>2</sub> and H<sub>2</sub>, and (5) the transportation of the MeOH produced to energy consumption site and/or as a chemical product.

In this study, we propose utilizing PV power generation as a natural energy source for water electrolysis, and also a conceptual design for this system, and we quantify materials for construction and unit requirements of this system. Furthermore, in order to measure the energy-effectiveness of this system, an energy analysis (energy consumption and energy balance ratio) was performed as one of LCA.

## 2. SYSTEM DESCRIPTION

### 2.1. Outline of the CO<sub>2</sub> recycling system

Figure 2 shows the major materials and energy balances for this CO<sub>2</sub> recycling system. As an example of the application of this system of CO<sub>2</sub> recycling, we applied it to a 1000 MW coal-fired power plant as the stationary CO<sub>2</sub> emission source. The aim is that 467 t/h of CO<sub>2</sub> out of 778 t/h of the total CO<sub>2</sub> emissions be recovered and 323 t/h (2.6 Mt/year) of MeOH be produced.

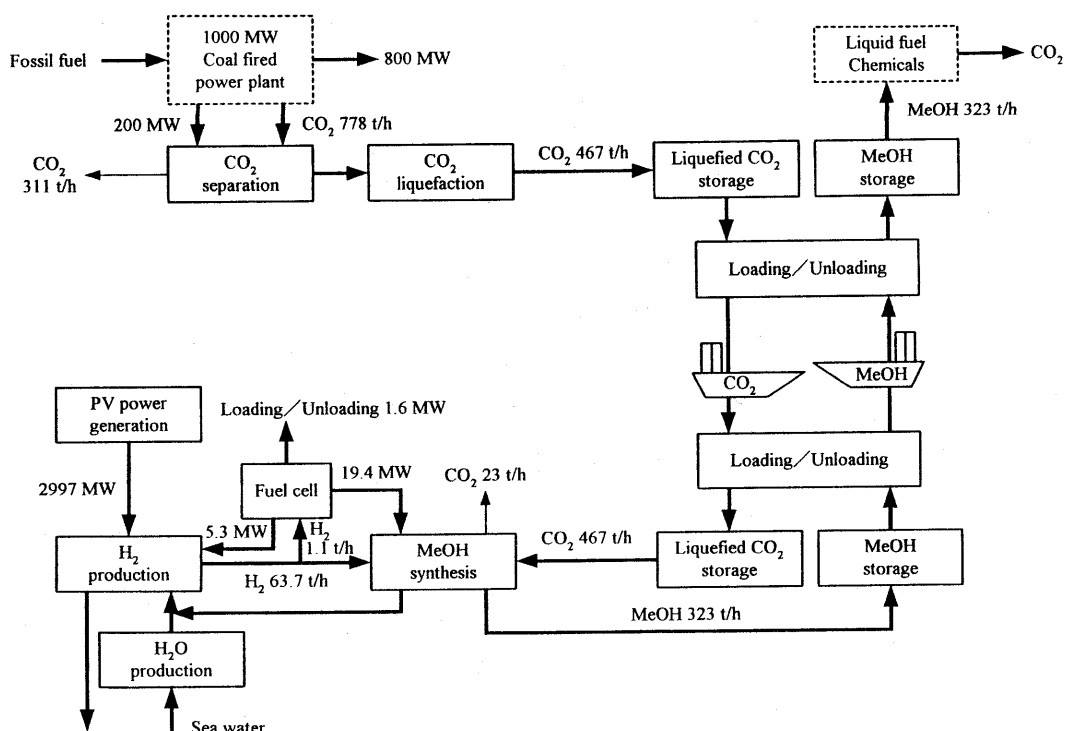


Figure 2. Major materials and energy balances for CO<sub>2</sub> recycling system.

The CO<sub>2</sub> is separated and recovered from the emitted CO<sub>2</sub> from a coastal zone power plant by polymer membranes, and then liquefied and stored in Japan (Tokyo bay area). It is then transported to the MeOH synthesis site overseas (Port Hedland in Australia) by tanker. CO<sub>2</sub> is hydrogenated to MeOH with H<sub>2</sub> produced by water electrolysis using a catalytic hydrogenation reaction. The electricity for water electrolysis (H<sub>2</sub> production) is generated by PV power generation in a desert (Great Sandy desert in Australia). The MeOH produced at the clean energy producing site is then transported to a mass energy consuming site in Japan, where MeOH is used as a fuel and/or a chemical raw material.

## 2.2. Process (Plant) design

This system is classified into five main processes (plants).

1. CO<sub>2</sub> separation (including liquefaction).
2. H<sub>2</sub> production.
3. MeOH synthesis.
4. CO<sub>2</sub>/MeOH transportation and storage (including loading and unloading).
5. PV power generation.

These processes are now being designed using some new technologies, which can potentially be performed in near future (about 2015). The specifications of processes and the design factors of the

major equipment (based on our experimental data) used for each process are summarized as follows.

**2.2.1. CO<sub>2</sub> separation (including liquefaction).** 467 t/h of CO<sub>2</sub> is separated and recovered from 778 t/h of CO<sub>2</sub> emitted from a coal-fired power plant at coastal zone by high performance polymer separation membranes (Tokuda *et al.*, 1996), distilled, liquefied and stored. The recovery ratio of 60 per cent in this case was determined to minimize energy required for CO<sub>2</sub> separation.

The dust is removed from the combustion exhausted gas before CO<sub>2</sub> gas separation, and 60 per cent of CO<sub>2</sub> gas is separated recovered from this gas by separation membranes. The CO<sub>2</sub> gas concentration of separated gas is approximately 60 per cent (dry basis). The separated gas is desulphurized, dehumidified and liquefied before the distillation. In the distillation tower, the bottoms product is 99.5 mol% liquefied CO<sub>2</sub>, and also is distilled as 99.92 wt% CO<sub>2</sub> at the flash drum of the tower bottom and is then cooled and stored (0.98 MPa, 233 K).

The major design factors of the CO<sub>2</sub> separation membrane equipment are listed below.

Membrane type: Cardo-type polyimide asymmetric hollow fibre.

CO<sub>2</sub>/N<sub>2</sub> separation factor: 35 (—).

CO<sub>2</sub> permeability (flux):  $5 \times 10^{-4}$  cm<sup>3</sup>/cm<sup>2</sup> sec cmHg.

Separation temperature: 323 K.

Separation pressure: 0.01 MPa on the permeated side.

0.1 MPa on the gas feeding (non-permeated side)

Liquefied CO<sub>2</sub> purity: 99.92 (wt%).

**2.2.2. H<sub>2</sub> production.** The H<sub>2</sub> produced by solid polymer electrolyte (SPE) water electrolysis (Kondoh *et al.*, 1996; Kato *et al.*, 1998) is compressed in a pipeline and transported to a MeOH synthesis facility. H<sub>2</sub> for use at night is stored in a pipeline during the daytime. 64.8 t/h of H<sub>2</sub> are produced by water electrolysis using the electricity, which is supplied from PV power generation.

In order to produce the pure water for water electrolysis, sea water is pretreated (filtration and pH control), and desalted through a reverse osmosis (RO) membrane in a coastal zone. The desalted water is transported through a pipeline to a H<sub>2</sub> production facility, built 300 km from a coastal zone, and organic substances and other contaminants are removed by activated carbon. The pure water is produced by ion exchange and supplied to the electrolyser.

The major design factors of the water electrolysis are listed below.

Method: SPE water electrolysis.

Electrocatalyst: a Ru – Ir-based composite catalyst.

Electric power consumption: 3.95 kWh/N m<sup>3</sup>-H<sub>2</sub>.

Electrolysis temperature: 353 K.

**2.2.3. MeOH synthesis.** This plant would be built in a coastal zone in the vicinity of a port. This plant consists of two major facilities, the MeOH synthesis unit and the fuel cell (FC) unit. 323 t/h (2.58 Mt/year) of MeOH are synthesized from H<sub>2</sub> and CO<sub>2</sub> by catalytic hydrogenation. The required electricity for MeOH synthesis is supplied from the FC (phosphoric acid type, conversion efficiency: 60 per cent based on HHV).

CO<sub>2</sub> is hydrogenated to MeOH with H<sub>2</sub> produced by water electrolysis and the highly activated catalyst (Kanai *et al.*, 1996; Saito *et al.*, 1996; Ushikoshi *et al.*, 1997).

Liquefied CO<sub>2</sub> is vaporized and mixed with H<sub>2</sub> and heated and the mixed gas is fed to the reactor. 1 mol MeOH is synthesized from 3 mol H<sub>2</sub> and 1 mol CO<sub>2</sub> at 5 MPa and 523 K. As by-products, water, methyl formate and higher alcohols are produced. The reacted gases are cooled and separated into liquid products and unreacted gases by a gas-liquid separator. The liquid products are distilled as 99.5 wt% MeOH and stored. The unreacted gases are recycled to the reactor.

The major design factors of the MeOH synthesis are listed below.

Method: catalytic hydrogenation.

Reactor: fixed-bed gas-phase reactor.

Catalyst: Cu/ZnO-based catalyst (e.g. Cu/ZnO/ZrO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub>/Ga<sub>2</sub>O<sub>3</sub>).

MeOH selectivity: 99.8%.

Reaction conditions: 5 MPa, 523 K.

MeOH purity: 99.5%.

**2.2.4. CO<sub>2</sub>/MeOH transportation and storage (including loading/unloading).** The CO<sub>2</sub> is liquefied and stored in Japan, loaded and transported to a clean energy-producing site overseas by a dual-purpose CO<sub>2</sub>/MeOH tanker (Sakamoto, 1998) and unloaded. The produced MeOH is loaded and transported to a site of mass energy consumption in Japan.

The major design factors of the transportation and storage facilities are listed below.

(a) *Tanker (two ships):*

Transport capacity: 149 000 m<sup>3</sup>.

Cargo tank: 38.7 m  $\varnothing$   $\times$  5 (heat insulating).

(b) *CO<sub>2</sub> storage tank:*

Capacity: 34 000 m<sup>3</sup>  $\times$  6 (Japan).

30 000 m<sup>3</sup>  $\times$  6 (overseas).

(c) *MeOH storage tank:*

Capacity: 51 800 m<sup>3</sup>  $\times$  4 (Japan).

45 600 m<sup>3</sup>  $\times$  4 (overseas).

**2.2.5. PV power generation.** The PV plant (Sakamoto, 1998) is built at a distance of 300 km from a port and adjoins the facility of H<sub>2</sub> production, and supplies the generated electricity (2997 MW) to the facility of H<sub>2</sub> production.

This PV system consists of two major facilities (a PV array unit and a power conditioning system). The PV arrays are assumed to be a fixed flat-plate type and the rated power of a module in PV arrays is 120 W at 20.0 V (nominal voltage), and 50 of these modules are connected in series (output: 1000 V) and also 2 are connected in parallel as one minimum output unit (output: 12 kW).

The required total cell (module) area is determined by the mean daily load (H<sub>2</sub> production) and the mean solar radiation value of Great Sandy desert in Australia (6.76 kWh/m<sup>2</sup>-day, Lat. 20°23' S, Long. 118°37' E).

The power conditioning systems consist of electrical devices (e.g. d.c.-d.c. converters and d.c.-a.c. inverters). The d.c.-d.c. converter transforms the PV generator voltage to the d.c. voltage (rated voltage: 500 V) for the electrolysis and supplies it the electrolyser. The d.c.-a.c. inverter (efficiency: 98 per cent) converts the generated d.c. from the PV generator to a.c. (rated voltage: 6600 V) for a.c. load equipment (e.g. compressor, pure water production and pumps in the facility of H<sub>2</sub> production), and transforms it to the levels of the load equipment.

The major design factors of the PV module are listed below.

*PV module:*

Cell: Polycrystalline – silicon.

Conversion efficiency: 20%.

Nominal voltage: 20 V.

Dimensions: 1200 (*L*) × 500 (*W*) × 50 (*H*) (mm).

Weight: 7 kg.

Tilt angle: 20° (facing due south).

### 3. METHODOLOGY OF ENERGY ANALYSIS

#### *Energy consumption*

Each process was divided into six stages as follows:

1. materials.
2. manufacture of equipment.
3. building of plant.
4. transportation of equipment and materials.
5. maintenance of plant.
6. running of plant.

Regarding each process, the energy consumption required in each stage was evaluated from the preceding conceptual design data (equipment, unit requirements utilities, etc.), and is expressed in terms of thermal energy (J: joule), which was considered as primary energy (e.g. 9.419 MJ/kWh, conversion efficiency, 38.2 per cent). The energy consumption required for constructing plants was the sum of the energy consumed for materials used for plant building, equipment manufacturing, equipment transportation and on-site maintenance of buildings and equipment.

It is assumed that the energy consumption for equipment manufacture was 20 per cent of the energy consumption of materials (i.e. 1. materials). The plant building energy consumption was assumed to be 10 per cent of that of materials. The equipment transportation energy consumption was calculated by the fuel amount consumed by each transporting unit. The energy consumption for equipment maintenance was incorporated at 5 per cent of the total of materials, equipment manufacture, equipment transportation and building maintenance energy consumption. The PV plant was assumed at 1 per cent of the aforementioned total energy consumption.

The energy consumption of materials was calculated by multiplying the weight of each material and fuel by each specific energy consumption (SEC, e.g. MJ/t-material) and summing these. The SECs of materials, fuels and utilities were quoted from the author's study (Sakamoto, 1998).

The amounts of materials, equipment and unit requirements for building and for running of each plant were quantified from the preceding plant design data.

#### *Energy balance ratio (EBR)*

In order to measure energy-effectiveness of this system, EBR which is a very important index (Uchiyama and Yamamoto, 1991) was estimated. For this system, EBR can be defined

as follows:

$$EBR = E_{out}/E_{in}$$

where  $E_{in}$  is the total annual energy supplied to this system and calculated by the total energy consumption (i.e. the total energy input to this system for construction and running) and the plant lifetime (Sakamoto, 1998), and  $E_{out}$  is the annual energy generated by MeOH and calculated by the annual production amount of MeOH (2.584 Mt/year) and the calorific value (22.69 MJ/kg-MeOH).

#### 4. CONCLUDING REMARKS

##### *Materials for construction*

Table I shows the materials required for constructing this system (excluding materials with less than 0.001 wt% of the total weight). The total amount of the materials accounts for 12 888 kt. The amount of concrete is the largest and accounts for 62.1 per cent (8001 kt) of the total amount of

Table I. Materials required for plant construction.

Materials	Weight	
	(t)	(%)
Carbon steel	3 545 120	27.506
Stainless steel	12 001	0.093
Copper	214 825	1.667
Aluminium	2939	0.023
Titanium	3000	0.023
Polyethylene	8657	0.067
Polyvinylchloride	16 586	0.129
Epoxy resin	327	0.003
Phenol resin	5342	0.041
Separation membrane	288	0.002
FRP	215	0.002
Butyl rubber	2922	0.023
Catalyst electrode	250	0.002
Glass	141	0.001
Concrete	8 001 039	62.080
Calcium silicate	3183	0.025
Polyurethane foam	586	0.005
Carbon	98	0.001
Paint	2866	0.022
Paper	600	0.005
Wood	23 422	0.182
Asphalt	349	0.003
PV module	1 043 560	8.097
Total	12 888 316	100

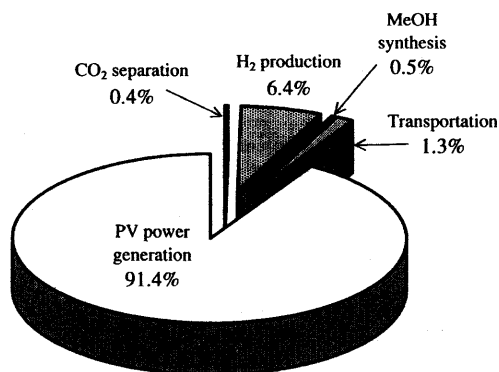


Figure 3. Weight fraction of materials for each plant required for construction.

materials, those of carbon steel and PV modules follow and account for 27.5 and 8.1 per cent (3545 and 1044 kt), respectively. The concrete is used as the foundation for plants, and the carbon steel is mainly used for the structure of plants and the frames of PV arrays. Copper is used as the cable, which supplies electricity to the facilities.

Figure 3 shows the weight fraction of material for each plant required for constructing this system. The amount of materials required for constructing the PV power generation is the largest and accounts for 91.4 per cent (11 777 kt) of the total amount of materials. That of H<sub>2</sub> production follows and accounts for 6.4% (829 kt). Regarding the PV power generation, the amounts of concrete, carbon steel, PV modules and copper are 7645, 2845, 1044 and 208 kt, and account for 64.9, 24.2, 8.9 and 1.8 per cent of the amount of materials for PV power generation plants, respectively. Regarding the H<sub>2</sub> production, the amounts of concrete and carbon steel are 195 and 592 kt, and account for 23.5 and 71.4 per cent of the amount of materials for the H<sub>2</sub> production plants, respectively, and 68 per cent (566 kt) of carbon steel is used as the equipment and pipes.

#### *Annual consumption for running*

Table II shows the major annual consumption for running this system, excluding the amount of electricity (regarding electricity balance, see Figure 2). Almost of materials are consumed for the water treatment for pure water production of the H<sub>2</sub> production. The catalyst is consumed for MeOH synthesis. All steam is consumed for the distillation involved in the CO<sub>2</sub> separation process. The heavy oil is consumed as fuel of the tanker.

#### *Energy analysis*

Table III shows the energy consumption of each process for this system. Regarding the energy consumption for building plants, PV power generation comprises the largest energy-consumption process (12 003 TJ/year). The amount of energy consumption of this process accounts for 81.7% of the total energy consumption for building plants (14 687 TJ/year), and those of H<sub>2</sub> production and CO<sub>2</sub> separation follow and account for 15.2 and 0.7 per cent (2229 and 106 TJ/year), respectively.



Table II. Annual consumption for running.

Consumption	(t/year)
<i>Materials</i>	
Activated carbon	88
Molecular sieve	10
Catalyst	135
Anthracite	11
RO element	2
Ion exchange resin	21
NaOH	720
HCl	262
FeCl <sub>3</sub>	10
NaHSO <sub>3</sub>	365
<i>Utilities</i>	
Steam	71 693
Heavy oil	32 940

Regarding the energy consumption of materials, the energy consumption of PV power generation is the largest, and accounts for 87.2 per cent (6799 TJ/year) of the total energy consumption of materials and 46 per cent of the total energy consumption for building plants. That of H<sub>2</sub> production follows and accounts for 10.6 and 5.6 per cent (825 TJ/year), respectively.

In the PV power generation process, the energy consumption of PV modules is the largest and accounts for 59 per cent (4012 TJ/year), and those of carbon steel and copper follow and account for 33 and 4 per cent (2252 and 278 TJ/year), respectively. This is because the SEC of the PV module is approximately 5 times as large as that of carbon steel, and results from the reduction process for producing high-purity silica from quartz sand which consumes a large amount of energy. The PV module production process is therefore costly in terms of energy consumption.

On the other hand, in the H<sub>2</sub> production process, the energy consumption of carbon steel for the equipment and pipes is the largest and accounts for approximately 81 per cent (672 TJ/year).

Regarding the energy consumption for plant running, the energy consumption of H<sub>2</sub> production is the largest (226 229 TJ/year) of the energy consumption for plant running. Those of CO<sub>2</sub> separation and transportation follow (15 293 and 1819 TJ/year). In H<sub>2</sub> production, the most energy (226 193 TJ/year) is consumed as electricity for the electrolysis of water. In CO<sub>2</sub> separation, the most energy (15 070 TJ/year) is consumed as electricity for the pumps. In transportation, 76 per cent of the energy (1379 TJ/year) is consumed as fuel for the tanker.

In case of offsetting the electricity generated by the PV power generation, CO<sub>2</sub> separation process comprises the largest energy-consumption process (15 399 TJ/year). The amount of energy consumption of this process accounts for 48.6 per cent of the total energy consumption (31 711 TJ/year), and those of PV power generation and H<sub>2</sub> production follow and account for 37.9 and 7.1 per cent (12 003 and 2257 TJ/year) of the total energy consumption, respectively.

Regarding the EBR of this system,  $E_{in}$  is 31 711 TJ/year from the preceding estimate (i.e. the total annual energy consumption of this system). On the other hand,  $E_{out}$  which is the annual energy generated by MeOH calculates 58 626 TJ/year. Hence, the EBR is approximately 1.85.

Consequently, this analysis indicates that this system is not only a CO<sub>2</sub> recycling system but also an effective energy supply system.

Table III. Energy consumption of each process.

	CO <sub>2</sub> separation	PV power generation	H <sub>2</sub> production	MeOH synthesis	Transportation	Total
			(TJ/year)			
Energy consumption for building plants						
Materials	40.5	6798.9	825.4	29.7	98.5	7792.9
Equipment manufacture	8.1	1359.8	165.1	5.9	19.7	1558.6
Transportation	0.3	394.4	41.6	3.2	4.6	444.2
Construction	4.1	679.9	82.5	3.0	9.8	779.3
Maintenance	52.9	2769.9	1114.6	41.8	132.7	4111.9
Subtotal	105.8	12002.8	2229.2	83.6	265.4	14686.8
	%	(0.7)	(15.2)	(0.6)	(1.8)	(100.0)
Energy consumption for running plants						
	15293.2	(- 227783.5)	226229.3	1465.9	1819.1	244807.5 <sup>†</sup>
	%	(6.2)	(92.4)	(0.6)	(0.7)	(100.0)
Total <sup>‡</sup>	15399.0	12002.8	2256.6	88.4	1964.0	31710.8
	%	(48.6)	(7.1)	(0.3)	(6.2)	(100.0)

<sup>†</sup>Excluding energy consumption of PV power generation.<sup>‡</sup>Offsetting the electricity generated by PV power generation.

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