

## **Nonviable carbon neutrality with plastic waste-to-energy**

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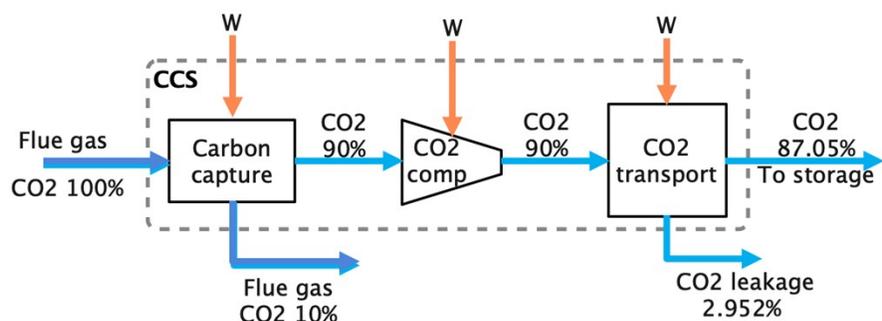
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## Carbon mitigation methods

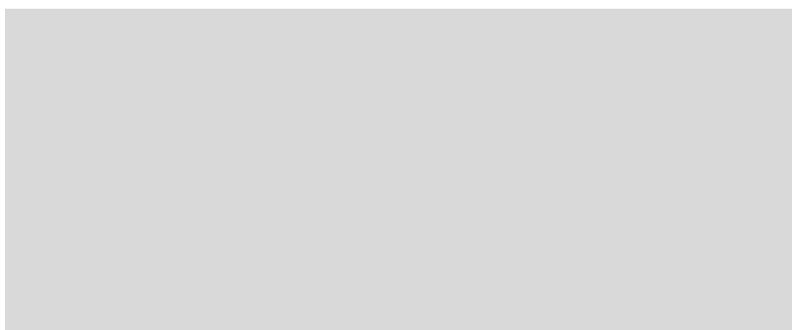
Other than CCS, there are methods for carbon mitigation, such as carbon capture and utilization (CCU), bioenergy with carbon capture and storage (BECCS), and carbon recycling. Carbon-based chemicals and fuels can be produced from the captured CO<sub>2</sub> by CCU. BECCS captures CO<sub>2</sub> from the atmosphere by photosynthesis, and the biomass can be converted into other chemicals. Carbon recycling literally recycles carbon within the chemical production processes from the captured CO<sub>2</sub>. These methods are also modeled, and the results showed that these methods do not have significant advantages over CCS in some respects per method. In addition, adding more chemical processes for the captured carbon will accompany economic issues. The details of modeling methods and results are discussed as follows.

In this study, considering power consumptions and leakages, the CO<sub>2</sub> removal efficiency and net power consumption of CCS were 87.05% and 892.8 MJ/ton-CO<sub>2</sub>, respectively. As shown in Fig. S1, CCS was modeled as carbon capture, transportation, and storage. This modeled CCS is compared with three other additional routes for carbon mitigation, which are carbon capture and utilization (CCU), bioenergy with carbon capture and storage (BECCS), and carbon recycling by CCU.



**Fig. S1.** Schematic of carbon capture and storage (CCS) unit.

For carbon capture and utilization (CCU), various carbon-based chemicals can be synthesized using captured CO<sub>2</sub> from the flue gas. The lifetime of CO<sub>2</sub> storage of the produced chemicals varies for materials. For example, CO<sub>2</sub>-based polyurethanes can be used as a construction material with a CO<sub>2</sub> storage lifetime from several decades to centuries while CO<sub>2</sub>-based organic carbonates, methanol, and urea have a CO<sub>2</sub> storage lifetime of several months.<sup>1</sup> The CCU stage to the synthesis of fuel gas or chemical was investigated, as shown in Fig. S2. Between many carbon-based chemicals, CO, methane, and methanol were investigated in this study because of relatively higher technology readiness over other chemicals.<sup>2</sup>



**Fig. S2.** Schematic of carbon capture and utilization (CCU).

The energy efficiency of 60% and selectivity of 90% were assumed based on the literature,<sup>2</sup> and the energy requirements for CO, methane, and methanol synthesis were 15.3 GJ/ton-CO, 28.41 GJ/ton-methane, and 21.93 GJ/ton-methanol, respectively. The power consumption for carbon capture of 0.0950 kWh/kg-CO<sub>2</sub> was included in the net power consumption. Thus, the total power consumption of CCU was 8.71–13.26 GJ/ton CO<sub>2</sub>, as shown in Table S1, which is 10–14 times higher than the power consumption of CCS. In addition, the CO<sub>2</sub> removal efficiency of CCU is 81%, which is 6.05% lower than that of CCS, and CCU cannot be a permanent CO<sub>2</sub> storage method. However, CCU could be an alternative for carbon mitigation for CO<sub>2</sub> emitting sources far from CO<sub>2</sub> storage sites and without CO<sub>2</sub> transport infrastructures.

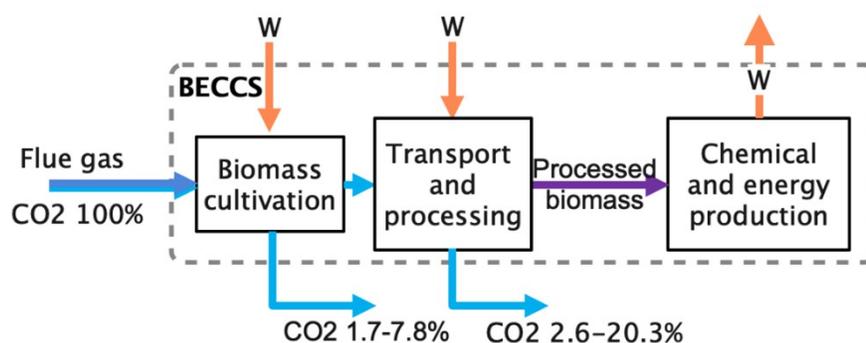
**Table S1.** The amount of synthesized material, power consumption, and power generation of carbon capture and utilization (CCU).

Synthesized material	Material recovery (kg/kg-CO <sub>2</sub> )*	Power consumption for synthesis (GJ/ton-CO <sub>2</sub> )*	Total power consumption (GJ/ton-CO <sub>2</sub> )
CO	0.57	12.20	11.32
Methane	0.33	9.30	8.71
Methanol	0.66	14.35	13.26

\* Denominator of the unit is the amount consumed for synthesis.

BECCS was modeled, which captures CO<sub>2</sub> from the atmosphere via biomass photosynthesis (Fig. S3). The grown biomass can then be utilized for chemical and energy production, where carbon is emitted.<sup>3</sup> The carbon can be captured on-site and stored via CCS or emitted into the atmosphere. Ideally, this method may achieve carbon balance. However, the production and disposal of fossil-based plastic will increase, and plastic waste-to-energy will produce more carbon influx into the atmosphere. Therefore, the amount of biomass has to be increased to achieve carbon neutrality. For example, the total amount of plastic waste generation was 353.29 Mt in 2019, of which 29% was incinerated.<sup>4</sup> Assuming 63.36% of C of the plastic<sup>5</sup> and 98% of oxidation, 233.26 Mt-CO<sub>2</sub> would be produced in 2019. In the same manner, 1277.76–1858.56 Mt-CO<sub>2</sub> production could be estimated from incinerating 50% of 1100–1600 Mt plastic waste in 2050.<sup>6,7</sup> The land required for BECCS is 0.1–0.6 ha per capturing 1 ton CO<sub>2</sub>-eq.<sup>8</sup> Therefore, the total land area to remove CO<sub>2</sub> from the incineration of plastic waste would have been 23.33–

139.96 Mha in 2019 and would be 127.78–766.66 Mha in 2050, which are 0.5–2.9% of the total land for agriculture in 2019 and 2.6–15.6% in 2050. Thus, BECCS requires significant land use. In addition, BECCS is subject to biomass feedstock, land type, supply chain of biomass, and type of biomass utilization, even necessitating eventual CCS after utilization of the biomass.



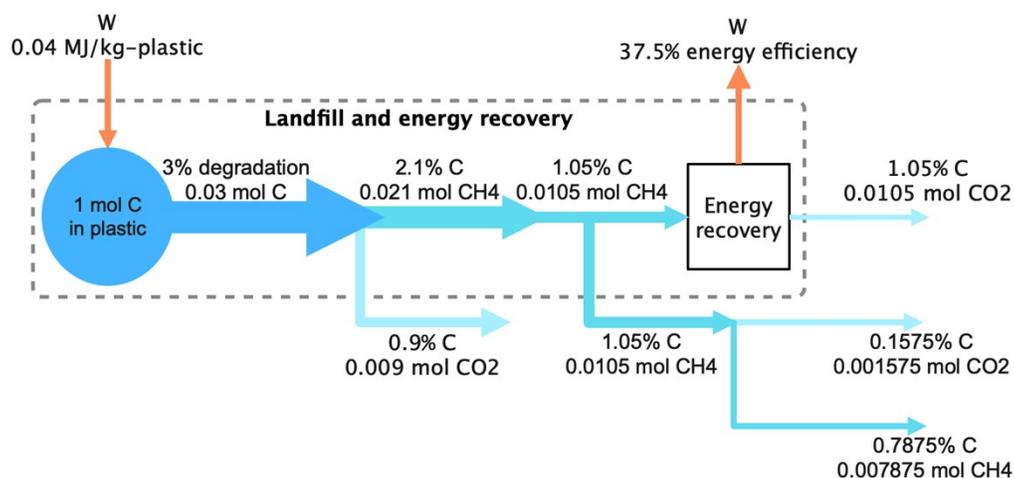
**Fig. S3.** Schematic of bioenergy with carbon capture and storage (BECCS).

Carbon can be recycled by carbon recycling, which recycles a portion of produced CO<sub>2</sub> during recovery processes back to the synthesis plant. In addition, CO<sub>2</sub> can be recycled for energy recovery processes like gasification and incineration. Carbon recycling reuses CO<sub>2</sub> before CCS. Compared to CCU, carbon recycling should employ a highly efficient process to convert recycled CO<sub>2</sub> into value-added products.<sup>9</sup> Moreover, permanent CO<sub>2</sub> storage is not plausible even with carbon recycling. Therefore, eventually, CCS unit should be included considering a cradle-to-grave point of view.

As discussed above, there exist other carbon mitigation methods. However, plastic waste-to-energy will significantly increase carbon emissions in the future. Therefore, CCS could be a practical way for carbon mitigation for plastic waste-to-energy and is considered as the carbon mitigation method in this study.

## Comparison with landfill

The carbon flow of degradation of landfilled plastic could be estimated according to the previous study.<sup>10</sup> The degradation rate of landfilled plastic was 3% in 100 years. 70% of degraded carbon was methane, and the remaining 30% was emitted. 50% of the methane underwent gas engine with energy recovery of 37.5%. The other 50% of the methane was emitted, and 15% of the methane was emitted as CO<sub>2</sub> owing to soil oxidation in the top layer. Besides, 0.04 MJ/kg-plastic of electricity was needed for landfill management. The carbon and energy flow are demonstrated in Fig. S4. Applying global warming potential (GWP) 25 on methane, the total emission was 0.34 g CO<sub>2</sub>-eq per 1 g of carbon in landfilled plastic. Approximately, non-recyclable and mixed plastic in the previous study<sup>10</sup> contained 0.6560 and 0.6935 g of C per 1 g of plastic. Thus, the total emission of landfill and energy recovery of biogas for 100 years were 0.22 and 0.24 g CO<sub>2</sub>-eq/g. Electricity generation at the gas engine was 262.5 MJ per 1 kg of carbon, which is 0.17 and 0.18 MJ/kg-plastic for non-recyclable and mixed plastic, respectively. Counting the power consumption for landfill management, the total power generation of landfill and energy recovery systems is 0.13–0.14 MJ/kg-plastic. Thus, the system emitted 5972.10–6069.69 g CO<sub>2</sub>-eq/kWh. The net energy efficiency of the system was 12.2–13.7%, regarding the degraded 3% of plastic. To sum up, 1 kg of landfilled plastic can generate 0.038–0.039 kWh of electricity and 0.22–0.24 kg of CO<sub>2</sub>-eq.



**Fig. S4.** Carbon and energy flow of landfill and energy recovery from plastic.

While, WtE methods in this study provide mean energy efficiency of 11.9–37.8% and produce mean life cycle analysis (LCA) greenhouse gas (GHG) emission of 87.16–2348.19 g CO<sub>2</sub>-eq/kWh, depending on the modeled eight scenarios. Thereupon, 1 kg of plastic, which were subject to landfill analysis, would generate 1.06–3.39 kWh of electricity and 0.25–3.78 kg CO<sub>2</sub>-eq of GHG. Examining the scenarios with CCS, 1 kg of plastic would generate 1.06–2.85 kWh of electricity and 0.25–0.33 kg CO<sub>2</sub>-eq of GHG.

WtE methods with CCS can generate 27–73 times more electricity while producing a similar amount of GHG. The large discrepancy in the electricity generation between the landfill and WtE is due to the tiny degradation rate of the landfill. Because the landfilled plastic only degrades 3% for 100 years, WtE methods, which handle the whole amount of landfilled plastic, can generate more electricity. Furthermore, the emission during landfill and WtE with CCS are similar. Therefore, the transition from landfill to WtE would enable more energy recovery while maintaining a similar emission level. Likewise, avoided emissions due to electricity would be more significant.

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