Assessment of Plastic Recycling Technologies Based on Carbon Resource Circularity Considering Feedstock and Energy Use

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Abstract

In this study, we are tackling the impact assessment of introducing plastic recycling technologies based on resource circularity for the design of a circulating plastic recycling system. For a sustainable circulation of carbon resources in the chemical industry, CO2 emitted from refineries needs to be converted back into resources and the remaining carbon needs to be substituted with bio-derived carbon and recycle-derived carbon. This study assessed the "circulating" plastic recycling technologies that convert waste plastics into raw materials that can be fed into existing facilities. The impact of plastic recycling has been evaluated in terms of greenhouse gas emissions, with a few cases evaluated from the perspective of the degree of resource circularity; however, an appropriate environmental indicator for recycling has not been constructed to date. To evaluate the impact of recycling technologies, we developed an indicator for the degree of carbon resource circulation based on the model consisting of process flows of refineries and multiple recycles. The results showed that catalytic cracking and material recycling contribute to carbon resource circulation more than other recycles.

**Keywords**: circularity indicator, carbon resource circulation, life cycle assessment, process modeling

* 1. Introduction

Regarding the circulation of carbon resources in the chemical industry, the consumption of fossil-derived resources needs to be reduced for carbon neutrality. The CO2 emitted from refineries needs to be converted back into resources and the remainder of the carbon needs to be substituted with bio-derived and recycle-derived carbon (Meng et al., 2023). In the context of carbon resource circulation, “circulating” recycles, including material recycling (MR), catalytic cracking, and monomerization, are important for converting used plastic products into chemical feedstocks and plastic resins that can be fed into existing processes such as refineries. Particularly, it was reported that the performance of catalytic cracking could be linked with improvements in environmental performance (Kikuchi et al., 2023). These technologies allow carbon resources to be used circularly and reduce the consumption of crude oil. By contrast, waste to energy mainly contributes to energetic circulation. Its main objective is to reduce greenhouse gas (GHG) emissions. Therefore, the impact of introducing recycling technologies must be evaluated in terms of both GHG emissions and contribution to carbon resource circulation. Evaluation by these two indicators will lead to an appropriate impact assessment framework for plastic recycling technologies.

In this study, we are tackling the impact assessment of introducing plastic recycling technologies based on resource circularity for the design of the circulating plastic recycling system. To date, the impact of introducing recycling has been evaluated mainly by GHG emissions, with a few cases evaluated from the perspective of the degree of resource circularity in previous studies (Lase et al., 2023). The study also reported results that an increase in material circularity generally decreases the environmental load regarding MR of plastic packaging (Vadoudi et al., 2022). However, evaluation based on the circularity of waste plastics is not sufficient. In other words, to date, an appropriate evaluation environment for plastic recycling has not been constructed. In this study, we made progress by setting the following: system boundaries, process modeling, developing an indicator for carbon resource circulation, inventory analysis, and environmental impact assessment. The system boundary includes oil refining, petrochemical processes, and several recycling processes. The unit configuration, capacity, energy consumption, and component yields of each unit refineries were set by the literature. In addition, the yields of products and energy consumption for each recycling were also set by the literature. From the above, GHG emissions were calculated by multiplying the energy consumption of each process by the CO2 emissions intensity taken from life cycle assessment (LCA) database. As for the calculation of resource circulation, the value was calculated by combining the mass of crude oil and waste plastics processed by incineration, landfill, and waste to energy.

* 1. Materials and methods
		1. System boundary

The production flows of petroleum products and basic chemicals derived from oil refining and petrochemical processes to produce plastics are shown in Figure 1. At the end-of-life, plastic wastes are treated for resource recycling (i.e., MR, mechanical recycling, monomerization, or catalytic cracking) or other purposes (energy recovery (ER), gasification, blast furnace reduction, or coke oven reduction). The manufacturing of plastic products and the use of the products were excluded from the system boundary of this study. The oil refining converts crude oil into naphtha and other products through separation and refining. The petrochemical process uses the naphtha to produce basic chemicals such as olefins and benzene, toluene, and xylene (BTX). The catalytic cracking converts waste plastics into hydrocarbons, which are similar in composition to conventional oil refineries. The steps include mixing with solvents, cracking reaction, and distillation. The cracking products were separated into four groups based on their carbon chains. Product A (C1 to C4) was similar to gas products used as raw materials at cryogenic separation. Product B (C5 to C7) and product C (C8 to C9) were fed as naphtha substitutes into naphtha cracking and catalytic reforming, respectively. Product D (>C10) contained the heavy content that must be fed to fluid catalytic cracking (FCC) for further cracking. Product distribution depends on the composition and decomposition conditions of the waste plastics. MR produces recycled pellets by washing, crushing, sorting, and pelletization. The mechanical recycling produces recycled pellets with improved quality by sorting and removal of impurities and solid-phase polymerization. The monomerization produces recycled pellets by depolymerization and repolymerization. The ER generates electricity from incinerating waste plastics. Other processes, including ER, produce energy sources.



Figure 1 System boundary including the plastic recycling system and refineries

* + 1. Process modeling and parameter setting

The model was created for the system boundary diagram shown in Figure 1 to analyze the impact of each unit process on the overall process, the relationship between unit processes, the contribution of technology performance at refineries and recycles, and the flow of resources when the system boundary is considered as a series of resource loops. For this model, the unit process configuration of the oil refining and petrochemical process, in addition to the capacity, the component yields, and the energy consumption per throughput of the unit process, were determined based on the reports on an existing refinery (Yoshitome et al., 2022). The production of basic chemicals and petroleum products was calculated by tracking the production of each unit process. We were also able to confirm the flow of fossil-derived feedstocks and waste plastics-derived feedstocks and calculate the throughput for each unit process capacity. Regarding 

Figure 2 Process modeling

catalytic cracking, energy consumption per amount of waste plastics to be processed and the yields of cracking products were estimated by process simulation using Aspen HYSYSTM. The product yields, which depend on the decomposition temperature and the composition of waste plastics, were collected from literature values (Keller et al., 2022). Energy consumption per amount of waste plastics to be processed and yield of products at MR and ER were established based on reports (JaIME, 2022). The composition of waste plastics can be arbitrarily changed. Basically, it was assumed that polypropylene (PP), polyethylene (PE), and polystyrene (PS) account for at least 60% of the total, and polyethylene terephthalate (PET), polyvinyl chloride (PVC), and other types account for the rest based on the current composition of Japan. Additionally, catalytic cracking was set to process mixed PP, PE, and PS, and MR was set to process pure PP or PE, and monomerization was set to process pure PET in this model. On the other hand, other recycling including ER, were assumed to process all types of waste plastics. The substitution rate of recycled resins for petroleum-derived resins was set to 0.5 based on the literature (Schwarz et al., 2021) for those by MR, and 1.0 for those by monomerization considering that polymerization process is included with reference to the study (Ghosh et al., 2023).

* + 1. The indicator of carbon resource circulation

In this study, the indicator for evaluating the impact of a recycling process introduction based on the degree of carbon resource circulation is named the recycling system circularity indicator for plastics (RSCIPlastics). The RSCIPlastics was developed by modifying the product circularity indicator (PCI) (Bracquené et al., 2020). RSCIPlastics is presented in Eq. (1), and the linear flow index (LFI) is constructed as Eq. (2).

|  |  |
| --- | --- |
| $$RSCI\_{Plastics}=1-LFI$$ | (1) |
| $$LFI=\frac{M\_{α}+ M\_{β}+ M\_{γ}}{M\_{α,liner}+ M\_{β,liner}+ M\_{γ,liner}}$$ | (2) |

RSCIPlastics is indicated as dimensionless numbers from 0 to 1, where 1 means complete carbon resource circulation. In this study, the complete carbon resource circulation is defined as “the state in which all waste plastics are processed by circulating recycles in resource loop and are used as raw materials for basic chemicals and plastic resins, not used for fuels". This indicator was mainly calculated by LFI, which indicates the degree of resource use in a linear economy. LFI was composed of mass of crude oil input ($M\_{α})$, mass of waste plastics that are incinerated or landfilled ($M\_{β})$, and mass of waste plastics that are processed by waste to energy ($M\_{γ})$. Also, LFI was calculated as a percentage in Eq. (2), with the denominator being the value obtained when the resources are used in a completely linear economy with the numerator being the value obtained when recycling processes are introduced. To calculate $M\_{α}$, $M\_{β}$, and $M\_{γ}$, the distribution ratio of waste plastics to each recycling process, yields of products, and others are also used as parameters.

A previous study (Matos et al., 2023) showed that indicators for circularity that focus on specific materials and products have been developed. However, the indicators do not adapt to system boundaries that cover multiple recycling processes and include design variables of recycling processes. RSCIPlastics covered the system boundary shown in Figure 1, which can include existing refineries, as well as multiple recycling processes for waste plastics. This indicator makes it possible to evaluate each recycling process per processing of 1 kg of waste plastics comparatively and evaluate recycling systems per processing the total amount of waste plastics. Plastic resins are considered to be final products, and the resource loop starts at the stage after the collection of waste plastics within the system boundary. As described above, RSCIPlastics is characterized by the fact that it does not take into account the contribution of the recycling process to energetic circulation, but makes possible an evaluation focused on material-to-material.

* 1. Results and discussion

Figure 3 shows the evaluation results of recycling technologies based on two indicators, RSCIPlastics and GHG emissions reduction. The upper right corner of the graph indicates a greater GHG reduction and a higher RSCIPlastics, which represents a better plastic recycling path. The graph also identifies any potential burden shift between GHG reduction and carbon resource circulation. We show that although circulating recycles are inferior to waste to energy in terms of GHG emission reduction, they have an advantage in contributing to carbon resource circulation. This result can be attributed to the fact that catalytic cracking and MR can convert waste plastics into chemical feedstocks and plastic resins on a material-to-material basis when compared to other recycling technologies. When we consider scenarios that move the plot to the upper right, the results are calculated under the assumption that energy sources for all recycles are fossil-derived, so if renewable energy resources can replace these, the plot can move to the right. In other words, there are other ways to increase GHG emission reductions besides implementing the recycling process itself. In addition, to move the plot upward, it is important to introduce recycling technologies like catalytic cracking and MR. Therefore, in the context of carbon resource circulation, it is essential to promote the implementation and introduction of such recycling technologies. Another important aspect is that the arrangement of these plots also changes depending on the composition of the waste plastics. In this figure, the composition of waste plastics was determined based on the current composition in Japan, and the results were calculated under the condition that all technologies process waste plastics with this composition. This was done to standardize the processing conditions for comparison of multiple recycling technologies. In the future, when we consider the strict introduction of these recycling processes, it would be useful to analyze what kind of condition or composition of waste plastics can be collected and sorted for a more precise evaluation.



Figure 3 Results based on carbon resource circularity and GHG emission reductions

* 1. Conclusions

In this study, the model that linked the process flow based on existing refineries with the process flow of multiple recycling processes was constructed. In addition, RSCIPlastics, an indicator for evaluating the impact of introducing a recycling technology from the perspective of carbon resource circulation, was developed. As a result, we confirmed the impact of introducing recycling in terms of not only GHG emission reduction, but also its contribution to resource circularity. It shows the importance of increasing the amount of waste plastics treated by catalytic cracking and MR in promoting carbon resource circulation. However, because the evaluation results are highly dependent on the state and composition of waste plastics, it is also essential to set up multiple scenarios that take these factors into account in detail. In the future, we need to clarify the supply and composition of waste plastics by material flow analysis and proceed to set up multiple scenarios. Also, RSCIPlastics does not take into consideration the quality of plastic resins. Therefore, in future studies it would be preferable to identify the destination products of plastic resins and incorporate parameters that indicate changes into physical properties.

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