Development of Innovative Gasification Process for Used Plastics by Using Fluidized Bed

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Abstract:

Used waste plastics flowing into ocean has become a worldwide problem. In recent years international trade in used waste plastics has been regulated. Therefore, a large amount of used plastics should be disposed domestically. On the other hand, used plastics with high calorific value could be used as an energy source. We have newly developed a gasification process of used plastics using fluidized bed. In this process, used plastics were decomposed in a fluidized bed reactor at around 600°C which was lower temperature than that used in current commercial processes. Higher calorific value gas could be attained by a gasification reaction control at lower temperature than that used in current commercial processes. Hydrogen enriched gas and catalysts were used as gasifying agent and fluid medium in the reactor, respectively, to improve gasification efficiency. High calorific value gas (LHV:5 000 kcallNm³) could be successfully produced from pyrolysis of used plastics by appropriate gasification temperature and catalyst.

1. Introduction

In recent years, marine pollution caused by used plastics has become a serious issue, with an estimated 8 million tons of plastic waste entering the world's oceans annually¹⁾. The potentially significant impact of ingestion of these plastics by marine organisms on ecosystems is a global concern²⁾. In 2018, the "Ocean Plastics Charter" was adopted at the G7 Summit held in Canada, calling for all signatory countries to make all plastics reusable, recyclable, and recoverable by 2030. Furthermore, in 2019, the Basel Convention was amended to prohibit the transboundary movement of contaminated used plastics. As a result, there is an urgent need for domestic processing of the large quantities of used plastics that were previously exported overseas for recycling purposes.

On the other hand, used plastics have a high calorific value equal to or greater than that of coal, making them a valuable potential energy source for Japan, which is a resource-limited country. One method of utilizing used plastics as a solid fuel is Refuse Derived Paper and Plastics Densified Fuel (RPF). RPF is produced by mixing and heating used plastics with wastepaper and is primarily used as a fuel for boilers in paper mills³⁾. However, there are also challenges to using RPF, such as its slow combustion rate and limited versatility as an energy source, which restrict its applications.

To address these challenges, this study aims to develop a gasification process for used plastics through thermal decomposition. Gasification is a technology that converts solid or liquid materials such as coal, waste, biomass, heavy oils, etc. into gaseous components, namely, carbon monoxide, hydrogen, carbon dioxide, methane, hydrocarbons. In this study, gasification involves the production of gaseous components, primarily light hydrocarbons ranging from C_1 to C_4 , from waste materials, in this case, used plastics. A higher calorific value would make the generated gas a more suitable fuel for various processes in steel works and could also allow effective utilization as a reducing agent in blast furnaces. Therefore, the target for the lower heating value (LHV) of the generated gas was set to be equal to or greater than the calorific value of the coke oven gas commonly used in steel works (5 000 kcal/Nm³), with the aim of producing a highcalorific gas.

One representative example of a gasification process

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using used plastics as a feedstock is the Ebara-Ube Process (EUP) developed by Ebara Corporation and Ube Corporation in Japan. This process is characterized by gasification in a fluidized bed reactor and a gas reformer using oxygen and air at temperatures ranging from 1 300 to 1 500°C to produce H_2 and CO^{4} . However, the gas obtained through the EUP process has a low calorific value due to its predominant contents of H₂ and CO. In 2005, JFE Engineering Corporation began operation of a Thermoselect gasification reforming process facility, which generates fuel gas by combusting waste materials such as combustible waste, incineration ash, and sewage sludge at a processing rate of 2.0 to 9.0 t/h at a temperature of 1 200°C using oxygen⁵⁾. However, the representative composition of the generated gas was reported to be $N_2/CO_2/H_2/CO =$ 3.3/31.9/33.2/31.4 vol%, and the estimated LHV was approximately 1 800 kcal/Nm³, indicating a low calorific value. Therefore, in this study, we focused on the decomposition temperature needed to increase the calorific value of the generated gas. Previous research reports on the gasification of used plastics have often set the decomposition temperature at 700°C or higher, which resulted in a generated gas LHV ranging from 1 000 to 3 200 kcal/Nm^{3 6-9)}. Thus, to produce a highcalorific gas with an LHV of 5 000 kcal/Nm³ or higher, the aim in this study was to suppress the decomposition of light hydrocarbons with higher calorific values compared to H₂ and CO by conducting gasification at a lower temperature.

In this study, converter gas, a byproduct gas of steelmaking, was used to incorporate H_2 into the gasifying agent. Converter gas is emitted from the converter during the refining process. While converter gas contains very little H_2 , it has a high CO concentration of 65 vol%. Therefore, to increase the H_2 concentration, a shift reaction, as shown in Eq. (1), was performed by adding water vapor before introducing the gas into the reactor.

In this paper, we investigated the effects of catalysts and the decomposition temperature on the gasification of used plastics using a gasifying agent obtained by subjecting converter gas to a shift reaction with the aim of producing a high-calorific gas with a lower heating value (LHV) of 5 000 kcal/Nm³ or higher.

2. Experimental Method

2.1 Fluidized Bed Gasification Equipment

To elucidate the fundamental behavior of plastic



Fig. 1 Experimental equipment

gasification, we created a test apparatus for gasification of used plastics by using a fluidized bed reactor and conducted experiments. The fluidized bed reactor is characterized by a uniform temperature distribution compared to other reactors¹⁰). **Fig. 1** shows the configuration of the experimental apparatus.

The apparatus consisted of a fluidized bed gasifier, a gasifying agent supply system, a plastic feed system, and a purification device for the gas product. The gasifier can be heated to 650°C by an external heating system. A gas dispersion plate made of sintered metal with 5-mesh openings was installed at the bottom of the gasifier, and the gasifying agent was supplied from below the dispersion plate to fluidize the bed of solid particles loaded on it. Water, N₂, CO₂, and H₂ were sent to a preheater by way of flow control devices and preheated to 150°C. The water was vaporized in the preheater before being supplied. The plastic feed system was designed to supply plastic quantitatively from a feeding hopper installed on top of the gasifier by means of a vibrating feeder. The height (L) to diameter (D) ratio of the fluidized bed, defined as L/D, was used as an indicator for determining the flow stability of the fluidized bed. The amount of catalyst was adjusted to maintain a constant value of L/D in all experiments. A gasifying agent preheating section was provided to prevent a decrease in the temperature of the fluidized bed due to the supply of the gasifying agent.

2.2 Experimental Conditions

2.2.1 Fluid medium

In this experiment, silica sand, which is widely used as a fluid medium in fluidized beds, was employed together with off-gas (OG) dust and γ -alumina, which are expected to promote gasification due to their catalytic effects. Silica sand of grade 7 with particle sizes ranging from 80 to 300 μ m was used. OG dust is an iron-rich dust which is discharged from a converter, but in this study, it was pre-classified to remove particles with sizes ranging from 45 μ m to 200 μ m to prevent ejection from the gasifier. γ -alumina, which is commonly used as an adsorbent and industrial catalyst, was used in the form of spherical particles of γ -alumina with sizes ranging from 75 μ m to 150 μ m. The true density, which affects the flow state, showed variations depending on the type of fluid medium. OG dust, which contains iron, had the highest true density, followed by γ -alumina and silica sand. The bulk density of γ -alumina was smaller than that of silica sand.

2.2.2 Pyrolysis temperature

The pyrolysis temperature of plastics in the fluidized bed was determined considering the thermal decomposition mechanism of plastics. The thermal decomposition mechanism of plastics, estimated based on reports by Moon et al. and Narobe et al., is shown in **Fig. 2**^{11–12)}.

Plastics are composed of polymer chains of hydrocarbons, and when heat is applied, these chains are broken and decomposed. Initially, solid plastics, as shown in Fig. 2 (a), are heated to around 400°C, resulting in decomposition into liquid oils and tars with carbon numbers ranging from 5 to 50 (Fig. 2 (b)). Further heating to 500 to 600°C leads to decomposition into light hydrocarbon gases with carbon numbers ranging from 1 to 4 (Fig. 2 (c)), while heating above 700°C results in CO₂, H₂, and CO (Fig. 2 (d)). Thus, when attempting to produce gas with a calorific value of more than 5 000 kcal/Nm³, thermal decomposition should not progress beyond the state in Fig. 2 (c), considering the low calorific values of the CO₂, CO, and H₂ which are generated when the decomposition temperature exceeds 700°C. Therefore, assuming that one characteristic feature of the process developed in this study is suppression of the decomposition temperature to below 700°C to limit the progress of thermal decomposition to the state in Fig. 2 (c), the gasification behavior of plastics at decomposition temperatures of 550°C, 600°C, and 650°C was investigated.

2.2.3 Gasifying agent

As a condition for stable operation with all the fluid media used in this experiment, the gasifying agent quantity was fixed at 4.0 L/min. Next, as the gasifying agent composition, hydrogen was used because it was considered effective in promoting the decomposition of plastics, as mentioned earlier. For this purpose, converter gas (LDG) was reformed by shift conversion to amplify the hydrogen concentration, and the resulting



Fig. 2 Mechanism of gasification process of waste plastics by thermal decomposition

Table 1 Gas composition

Gas	Flow rate (NL/min)	N ₂ (vol%)	CO ₂ (vol%)	CO (vol%)	H ₂ (vol%)	H ₂ O (vol%)
LDG	1.63	18	15	65	1	1
Steam	2.37	0	0	0	0	100
Gasifying agent	4	7	30	2	25	36

gas was used.

LDG at room temperature $(25^{\circ}C)$ and steam heated to 250°C were supplied to the shift converter at a mixing ratio determined so as to achieve a temperature of 430°C after the shift reaction, which is the maximum usable temperature of the shift converter. The gas compositions before and after the shift reaction are shown in **Table 1**.

Based on the calculated gas composition after the shift reaction, the gasifying agent composition for this experiment was determined. Since CO is not used as a gasifying agent and is not expected to have a significant impact on the experimental results, the gasifying agent composition was set as $N_2/CO_2/H_2/H_2O = 9/30/26/35$ vol%.

2.2.4 Plastic materials

 Table 2 shows the plastics used in this study and their properties.

2.3 Experimental Method

After heating the fluidized bed to the specified temperature with an external heater, a gasifying agent consisting of preheated N_2 , CO_2 , H_2 , and H_2O at 150°C was introduced from the bottom of the fluidized bed. Plastic was then supplied to the fluidized bed at a rate of 300 g/h using a vibrating feeder. The gas generated

Plastic	Size (mm)	LHV (calculated) (kcal/kg)	C (wt%)	H (wt%)	N (wt%)
Crashed plastics	0.6~2.0	9 754	77.5	12.7	0.1
O (wt%)	Cl (wt%)	Fe (wt%)	Ash (wt%)	Moisture (wt%)	PE ^{**1} (wt%)
4.5	0.1	0.1	5.1	0.3	77
PP ^{*2} (wt%)	PS ^{**3} (wt%)	PVC ^{**4} (wt%)	PET ^{*5} (wt%)	Insoluble (wt%)	
16	<1	1	<1	5	

Table 2 Property of plastics

*1 : Polyethylene *2 : Polypropylene *3 : Polystyrene*4 : Polyvinyl chloride *5 : Polyethylene terephthalate

by the decomposition of the plastic was collected periodically through a gas backflow system, and the contents of N₂, CO₂, H₂, CO, and light hydrocarbons (with carbon numbers ranging from 1 to 4) were measured using a gas chromatograph with a thermal conductivity detector (GC-TCD). The evaluation parameters for the generated gas were the LHV in kcal/Nm³ and the carbon-to-gas conversion ratio in wt%. LHV is an indicator of the heat generation capacity of the generated gas, and was defined by the following equation based on the calculation method specified in JIS K2301 8.2.

$$H_0 = \frac{\sum H_i R_i}{100}$$

 H_0 represents the lower heating value (LHV) of the generated gas (kcal/Nm³), H_i represents the LHV of each gas component in the generated gas (kcal/Nm³), and R_i represents the volumetric content of each gas component in the generated gas (vol%). The carbon-to-gas conversion ratio indicates the proportion of carbon in the plastic that is transferred to the generated gas and serves as an indicator of gas generation efficiency. The carbon-to-gas conversion ratio based on the calculations reported by Koyama et al.¹³.

$$\eta = \frac{\sum \left(V_i \times \frac{12}{22.4} \times C_i \right) - \sum \left(V_j \times \frac{12}{22.4} \times C_j \right)}{W_p \times \frac{C_p}{100}} \times 100$$

 η represents the gasification rate of the feedstock (wt%), V_i represents the production rate of each gas component in the generated gas (NL/h), C_i represents the number of carbon atoms in each gas component in the generated gas (-), V_j represents the quantity of each gas component in the gasifying agent (NL/h), C_j represents the number of carbon atoms in each gas component in the gasifying agent (-), W_p represents the

amount of plastic used in the experiment (g/h), and C_p represents the carbon content of the plastic (wt%). The number of carbon atoms in each gas component refers to the number of carbon atoms contained per gas molecule, which is 1 for CH₄ and 3 for C₃H₈.

3. Experimental Results

3.1 Trends in Plastic Gasification Experiments

Fig. 3 shows the composition of gas product when crushed plastic was used as the feedstock, OG dust was used as the fluidizing medium, and the temperature was set at 600°C (feedstock supply time: 1.5 h, sampling interval: 30 min, average data from 3 samplings).

In this experiment, formation of light hydrocarbons with high calorific values in the gas product was confirmed, resulting in a significant increase in heat generation. Since water vapor was removed by a trap, the generated gas only contained saturated water vapor, which was present in trace amounts and therefore was not included in this graph. When compared to the gas produced by the Thermoselect method, it can be observed that the low gasification temperature suppresses the production of CO and H_2 .

When the gasifying agent was changed to 100% N₂, LHV was 2 713 kcal/Nm³, and when it was changed to 26 vol% H₂ and 74 vol% N₂, LHV was 4 361 kcal/Nm³. The content of light hydrocarbons (C₁-C₄) was 17.3 vol% when N₂ was used as the gasifying agent, but was 25.1 vol% when the gasifying agent included H₂, indicating that H₂ has an amplifying effect on heat generation (as a result of amplification of light hydrocarbons).



Fig. 3 Composition of gas product

3.2 Influence of Decomposition Temperature and Catalyst

To investigate the influence of the decomposition temperature on the heat generation of the generated gas and the gasification rate (carbon-to-gas conversion ratio), an experiment was carried out using OG dust. The results are presented in **Fig. 4**.

At a gasification temperature of 550°C, both LHV and the gasification rate were low. The gasification rate also remained low across the entire temperature range, presumably due to the low catalytic activity of the OG dust.

Next, to investigate the influence of the catalyst on heat generation and the gasification rate, an experiment was conducted with different catalysts at a decomposition temperature of 600°C. The results are shown in **Fig. 5** (feedstock supply time: 1.5 h in all cases).

The experimental results using OG dust showed the highest LHV, generating a high-calorific gas with an LHV of 5 000 kcal/Nm³ or higher. In addition, the process using OG dust exhibited a higher gasification rate compared to silica sand, indicating the catalytic effect of OG dust. However, when compared to γ -alumina, the gasification rate was lower than when using the γ -alumina fluidizing medium. The causes and potential solutions for this lower gasification rate are discussed in the following section.



Fig. 4 Effect of temperature on gasification



Fig. 5 Effect of catalyst on gasification

4. Discussion

4.1 Effect of Catalyst

The catalytic mechanism of OG dust when used as a catalyst during the thermal decomposition and gasification of used plastics was investigated. Although the main component of OG dust is iron oxide, according to Funai et al., iron oxide acts as a catalyst for the decomposition of plastics and heavy oils in the presence of high-temperature steam¹⁴. A conceptual diagram of the catalytic behavior of iron oxide is shown in **Fig. 6**.

It is estimated that the decomposition of plastics occurs through an alternating process of consumption of the lattice oxygen in the iron oxide and replenishment of oxygen-deficient parts by the oxygen generated by the decomposition of water molecules. The hydrogen generated in this process is believed to be utilized in the hydrogenation decomposition reaction of plastics, thereby promoting the production of light hydrocarbon gases. Upon examining the composition of the generated gas in the experiments using OG dust, it was found that the amount of light hydrocarbon gas increased compared to when silica sand was used. Therefore, use of OG dust was considered to be effective for generating a high-calorific gas. Since the main component of OG dust is iron oxide, it was hypothesized that if the iron oxide in the OG dust could be effectively utilized as a catalyst, further improvement in the gasification rate could be achieved. The following experiment was conducted to verify this assumption.

A small-scale experimental apparatus with a fluidized bed diameter of 19 mm was used. Polypropylene (PP) was supplied at a rate of 3 g/h for 1 h, and the decomposition temperature was set at 600°C. The gasifying agent was a mixture of $Ar/CO_2/H_2 = 40/3/$ 30 vol%, which was supplied at a flow rate 60 ml/min. Silica sand, FeO, Fe₃O₄, and Fe₂O₃ iron oxide reagents were used as catalysts, with 1 g of each mixed with 10 g of fluidized silica sand. The experimental results are shown in **Fig. 7**.

The gasification rate was found to increase in the order of silica sand, Fe_3O_4 , FeO, and Fe_2O_3 . However,



Fig. 6 Catalytic behavior of iron oxide



Fig. 7 Effect of catalyst on carbon to gas conversion



Fig. 8 EDX-Mapping of OG dust



Fig. 9 Structure of OG dust

the gasification rate was lower than when using γ -alumina, even when using Fe₂O₃. Therefore, to elucidate the mechanism, cross-sectional SEM observations of the OG dust were conducted. The results, visualized as 2D images, are shown in **Fig. 8**.

OG dust was found to have a metal-Fe (M-Fe) core, which is surrounded by iron oxide, and most of the surface is covered by calcium. The structure of OG dust, as inferred from the XRD (X-Ray Diffraction) analysis results, is shown in **Fig. 9**.

From the innermost layer outward, OG dust was estimated to have a three-layer structure consisting of M-Fe, FeO or Fe₃O₄, and Ca (OH)₂. The Ca (OH)₂ was believed to be derived from steel slag, and the outermost layer of this Ca (OH)₂ undergoes thermal decomposition, which produces CaO. In addition, CO₂ was present in the generated gas, indicating that CaCO₃ was formed through the reactions. It was therefore believed that the catalytic activity of the OG dust decreased due to the formation of a CaCO₃ shell.

According to Knozinger, γ -alumina had been reported to exhibit catalytic activity due to its ability to exchange oxygen in the presence of high-temperature steam¹⁵⁾. Furthermore, the advantage of γ -alumina over OG dust in terms of the gasification rate may be attributable to the numerous micropores on the alumina surface, resulting in a large specific surface area of 200 m²/g and high contact efficiency with tar, leading to enhanced catalytic effects¹⁶. Based on these reports, the presence of oxygen-containing gases such as CO₂ and CO is thought to promote an increase in H₂ and light hydrocarbons through the decomposition of aromatic hydrocarbons. Indeed, when the experimental results of γ -alumina were examined, increased levels of CO₂, H₂, CO, and CH₄ could be observed compared to the results obtained with silica sand and OG dust, confirming the expected mechanism.

Therefore, like γ -alumina, OG dust also showed the potential to improve catalytic performance by enhancing contact efficiency.

4.2 Future Plans

This study revealed that high-calorific gas can be generated by gasification of used plastics at low temperatures using a fluidized bed reactor with a catalyst as the fluid medium.

Since γ -alumina is expensive, costing over 20 000 yen/kg, we would like to explore the possibility of improving gasification efficiency by using OG dust. In the future, we aim to enhance the catalytic performance of OG dust in order to improve gasification efficiency.

Furthermore, for commercialization of this process, it is necessary to scale up the fluidized bed reactor to increase its used plastic processing capacity. Because heat transfer with external heaters becomes insufficient when the diameter of a fluidized bed reactor is increased, it will be necessary to develop a method for circulating and heating the fluid medium externally. Therefore, in the future, we will investigate efficient circulation and heating methods for the fluid medium.

5. Conclusion

An experimental investigation was conducted with the aim of developing a technology for gasification of used plastics, and the following findings were obtained:

 This study demonstrated that hydrocarbons can be produced and high-calorific gas (LHV >5 000 kcal/ Nm³) can be generated by controlling the decomposition temperature to 600°C to 650°C using a fluidized bed and introducing a gasifying agent obtained by shift conversion of converter gas during the gasification of the used plastics. 2) When OG dust was used as a catalyst for gasification of used plastics in the fluidized bed, it was found that gasification efficiency improved in comparison with use of silica sand as the catalyst. Investigation of the cause revealed that the OG dust was coated with a calcium shell, leading to a decrease in catalyst activity.

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