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# GASIFICATION OF CARBONEOUS WASTES USING THE HIGH TEMPERATURE REFORMER

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**Abstract** : Gasification of carbonaceous wastes such as shredded tire, waste lubricating oil, plastics, and powdered coal initiates a single-stage reforming reactor(reformer) without catalyst and a syngas burner. Syngas is combusted with $O_2$ gas in the syngas burner to produce $H_2O$ and $CO_2$ gas with exothermic heat. Reaction products are introduced into the reforming reactor, reaction heat from syngas burner elevates the temperature of reactor above 1,200°C, and hydrogen gas fraction reaches 65% of the product gas output. Reactants and heat necessary for the reaction are provided through the syngas burner only. Neither $O_2$ gas nor steam is injected into the reforming reactor. Multiple syngas burners may be connected to the reforming reactor in order to increase the syngas output, and the product syngas is recycled into syngas burner.

**Key Words** : Hydrogen fuel, Steam reformer, Carbonaceous waste, Gasification, Syngas

## INTRODUCTION

Carbonaceous wastes such as shredded tire, waste lubricating oil, waste organic solvents, plastics, and powdered coal have been disposed on landfill and incineration sites, but induce to release contaminants into the atmospheric, subsurface, and aquatic environment. Recently researchers in Korea consider the pyrolysis of wastes at the pyrolysis temperature of 400-800 °C for energy recovery (oil and gas with residues - char and tar) and environmental conservation.[1-3)]

Gasification process converts carbon and hydrogen contained in the organic materials into fuel gases, hydrogen and carbon monoxide gas (generally called syngas) at the higher temperature compared to pyrolysis process. When the gasification is considered for waste disposal, it has definite benefits over the conventional method of waste disposal by means of combustion or incineration.[4-6)]

In the conventional method of gasification, the gasification should be kept at a high temperature by means of combustion heat generated from the oxidation reaction of carbonaceous materials with oxygen. Further, in the state of high temperature sufficient for sustaining the gasification reaction, steam or water in the reduction reaction is additionally supplied to promote gasification and increase the concentration of hydrogen.

In this work the reactor was designed for prevention of the secondary pollution from the oxidation reaction through separation of the oxidation chamber from the reduction chamber. Using this reactor, it has been investigated on the gasification of carbornaceous wastes such as coal, shredded tire, waste oil, and plastic waste materials. We have examined (i) an optimum

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temperature, (ii) an efficiency of hydrogen fuel production, and (iii) the effect of additional water for hydrogen gas production on the high-temperature reformer.

## EXPERIMENTS

### Reactor Descriptions

Figure 1 shows schematically the constitution and mechanism of the gasification reactor. As shown in Figure 1, gasification reactor 1 is composed of two parts of the same shape and size which are connected to each other vertically. The lower end of reactor 1 is an oxidation reaction chamber while the middle portion of reactor 1 is a reduction reaction chamber. Reactor 1 is 60 cm of center diameter with 150 cm height. The reactor's inner cavity is aligned with 5 cm thick molded Cerak wool provided from Kum Kang Corp. in S. Korea and shielded with 5 cm thick blanket of the same and then 3 mm SS casing. In the reduction reaction chamber of reactor 1, a liquid feed nozzle (2) for spouting liquid waste such as waste oil into reactor 1, a solid waste supply nozzle (3) for supplying solid waste such as coal powder into reactor 1 using a screw feeder, and a steam supplier (4) for spouting steam into reactor 1 are equipped. A liquid feed heater (5) is connected with the liquid feed nozzle (2) for heating the liquid feed supplied into reactor 1, and a water heater (6) is connected with the steam supplier (4) for supplying water into reactor 1 as steam. An outlet (7) for discharging syngas produced from reactor 1 is provided in the upper end of reactor 1, and a produced syngas recycling tube (8) is installed to recycle a portion of produced syngas discharged from the outlet (7) into reactor 1. Close to the produced syngas recycling tube (8), an oxygen supplier (9) is equipped at the lower end of reactor 1 in the oxidation reaction chamber for supplying oxygen required to react with the produced syngas recycled into reactor 1.

Gasification reactor 1 has two parts of the same shape and size connected to each other vertically, which makes the manufacture and maintenance of reactor 1 easy. In the upper section of reactor 1, a tungsten grille (10a) is installed to promote the reaction of $H_2O$ and $CO_2$ with unreacted feed in the gas before being discharged from reactor 1. Also in the lower section of gasification reactor 1, another tungsten grille 10b is installed for supplying uniformly $H_2O$ and $CO_2$ produced in the oxidation reaction chamber into the reduction reaction chamber and supporting solid feeds in the reactor. Between the upper and the lower tungsten grilles (10a) and (10b), carbonaceous feed materials react with $CO_2$ and $H_2O$ to produce CO and $H_2$. There is no oxygen present in the reduction reaction chamber, since oxygen supplied through the oxygen supplier (9) is completely consumed in the oxidation reaction chamber. Under the oxidation reaction chamber of reactor 1, an ash trap (11) is installed for storing remained ash. Further, on the wall of the reactor, thermocouples are installed for measuring the temperature in reactor 1, and a view port (12) is also installed for viewing the state of the reaction carried out in reactor 1.

The following is the operation of gasification reactor 1:

(a1) The reactor at room temperature is preheated using a conventional fuel such as LPG or oil to a temperature sufficient to ignite a syngas burner. Typically, the temperature is above 600°C.

(a2) When the temperature of the reactor reaches above 600°C, syngas and oxygen are supplied into the oxidation reaction chamber in the lower end of the reactor through the syngas(or $H_2$ gas) recycling tube, and then the temperature of the reactor elevates to about 1,200°C. At this time, the reactor can be filled with combustion products, $CO_2$ and $H_2O$, produced from the reaction of the outside syngas fuel with oxygen.

(a3) When the temperature of the reactor is kept at 1,200°C, carbonaceous materials to be gasified are fed into the reduction reaction chamber through the waste feed nozzles. Then,

$CO_2$ and $H_2O$ are produced from the reactions of the syngas with oxygen enter into the lower section of the reactor and react with the organic feed materials.

(a4) Syngas produced during the gasification reaction is discharged through the upper end of the reactor.

(a5) When the syngas is discharged from the reactor, a portion of the syngas is supplied again into the oxidation reaction chamber in the lower end of the reactor through the syngas recycling tube, and then reacts with oxygen to produce $H_2O$ and $CO_2$ along with heat. At this time, the supply of the outside syngas has been cut off. The heat source required to maintain the reactor at high temperature is obtained by recycling a portion of the produced syngas which then reacts with oxygen. At this time, oxygen is supplied as the minimum amount required to maintain the reactor at about 1,200°C. The combustion products of the recycled syngas, $H_2O$ and $CO_2$, react with the feed materials to be gasified (reduction reaction) and produce syngas again. The recycled syngas, which remains unreacted after the reaction with oxygen, is discharged from the reactor with the rest of the produced syngas.

In this gasification reaction, combustion of syngas in the oxidation chamber provides both heat and reactants (i.e. $H_2O$ and $CO_2$) for reforming. Therefore, there is no need for the steam supplied from the outside. However, for some feedstock, the addition of outside steam enhances the reforming and increases the $H_2$ content in the syngas. By controlling the ratio of oxygen and recycled syngas, a desired reactor temperature is obtained, and oxygen should be completely consumed in the oxidation reaction chamber. There should be no oxidation reaction in the reduction chamber, and feedstock in the reduction chamber should react only with $H_2O$ and $CO_2$. Gasification reactor 101 as shown in Figure 2 has a simplified structure of reactor 1 shown in Figure 1, and has a syngas burner equipped in the body of the gasification reactor.

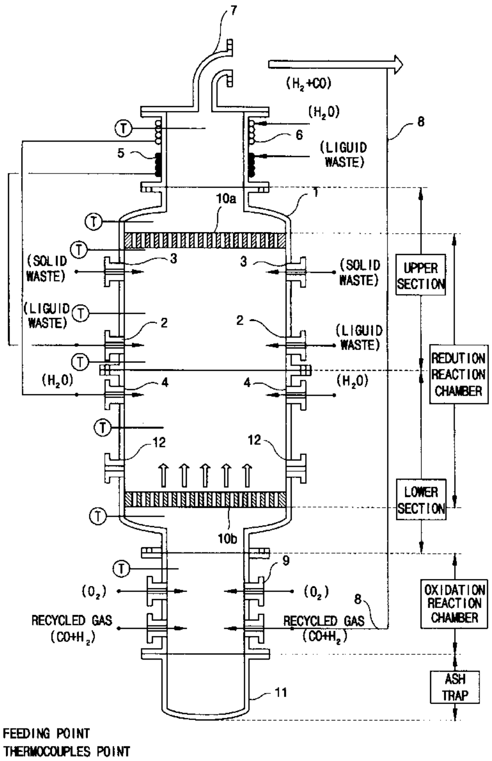


Figure 1. Gasification reactor (reactor 1).

Specifically, in the body of gasification reactor 101, a screw press feeder (102) is equipped at the bottom of reactor 101 for supplying solid feed into the reactor. An outlet (103) for discharging produced syngas from reactor 101 is provided in the upper end of reactor 101, and an ash trap (104) is provided in the lower end of reactor 101 for storing molten salt flowed out from the reactor. Ash trap (104) is interchangeable with solid feeder (102). Further, the body of reactor 101 is equipped with a syngas burner 105 which replaces the oxidation reaction chamber of reactor 1 in Figure 1. One syngas burner on each side, that is, two syngas burners are equipped in reactor 101. On the wall of reactor 101, thermocouples (106) are installed for measuring the temperature in reactor 101, and a heat exchanger (107) is installed for cooling the produced syngas discharged from the upper portion of reactor 101 to recover the heat. Further, a produced syngas recycling tube (not shown in Figure 2) is also provided in the body of reactor 101 for recycling the produced syngas

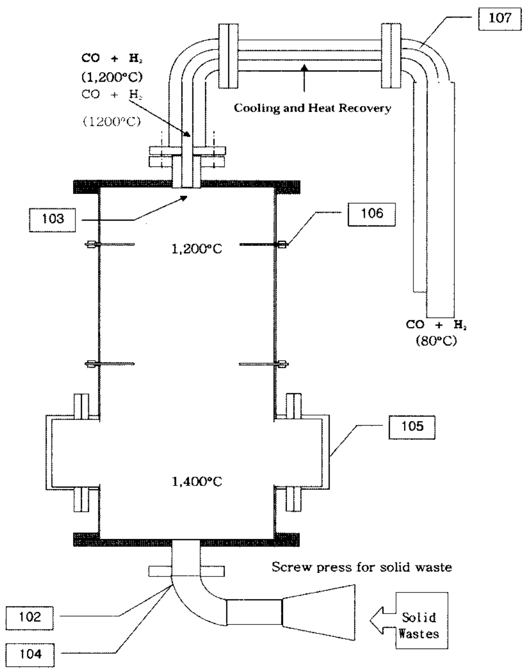


Figure 2. A high temperature reformer(reactor 101).

discharged from the outlet (103) and supplying it into the syngas burner (105).

In gasification reactor 101, in the syngas burner the recycled syngas reacts with oxygen to produce carbon dioxide and steam required for the reduction of feed materials as well as to maintain the temperature in the reactor at 1,200~1,400°C. The syngas burner in Figure 2 is the oxidation reaction chamber of reactor 1 shown in Figure 1.

The following is the operation of the gasification reactor shown in Figure 2:

(b1) the reactor at room temperature is heated slowly by burning syngas in the burner (105), preheating is not necessary as in the case of reactor 1 in Figure 1.

(b2) Syngas (or $H_2$ gas, if there is no produced syngas available) is introduced into the syngas burner through the produced syngas recycling tube, and oxygen is also introduced. Syngas is ignited by introducing a pilot light through pipe (112). The temperature rises suddenly with the ignition of syngas (or $H_2$ gas). The temperature is controlled by adjusting the oxygen intake while monitoring oxygen detector at the gas outlet. At the end of the operation, the supply of oxygen gas is turned off first and then syngas is turned off as the temperature of reactor falls.

In this manner, when the reactor temperature reaches to 1,200~1,400°C by controlling the amount of oxygen intake, the reactor becomes filled with $CO_2$ and $H_2O$, produced from the reaction of syngas with oxygen.

(b3) While the temperature was kept at 1,200°C, solid waste materials, which was compressed, degassed and dried previously, are supplied into the reactor through the solid waste feeder. Under the condition of about 1,200°C, solid feeder material reacts rapidly with $CO_2$ / $H_2O$ supplied from the syngas burner to produce syngas (reduction reaction indicated as Reactions 3 to 6).

(b4) Syngas produced during the gasification reaction is discharged through the upper end of the reactor. The syngas discharged from the reactor at about 1,200°C is cooled up to 100°C or below through a heat exchanger and then stored in a storage tank(not shown in Figure 2).

(b5) When the syngas is discharged from the reactor, a portion of the syngas is supplied again into the syngas burner connected with the reactor through the produced syngas recycling tube(not shown in Figure 2), and then reacts with oxygen in the syngas burner to produce steam and $CO_2$ along with heat. That is, the heat source required to maintain the reactor at high temperature is obtained by recycling a portion of the produced syngas which then reacts with oxygen. At this time, the temperature in the reactor is adjusted by controlling the supply of oxygen. The combustion products of the recycled syngas, $H_2O$ and $CO_2$, react with waste materials to be gasified and produce again syngas. The recycled syngas, which remains unreacted after the syngas burner, is discharged from the reactor with the rest of the produced syngas.

Figure 3 shows schematically the constitution of the syngas burner which is connected with the gasification reactor shown in Figure 2. As

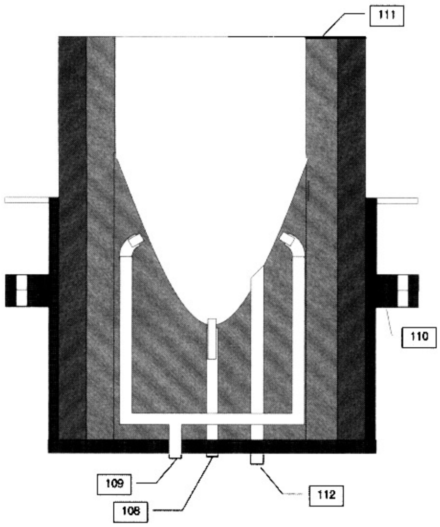


Figure 3. Syngas burner.

shown in Figure 3, in the syngas burner (105), a tube 108 for supplying syngas or hydrogen gas and a tube (109) for supplying oxygen are fixed by a flange 110, and the tubes are surrounded by an insulating material (111) and (111B)(cerak wool mold and blanket).[7] Pipe (112) is placed so that a stick of pilot light is inserted for ignition. The schematic diagram of experimental apparatus is shown in Figure 4.

# EXPERIMENTAL RESULTS

The reactor shown in Figure 2 has been tested with variety of feedstocks such as shredded tire, waste oil, waste plastics, both liquid and solid forms. They are converted readily into syngas as the reactor temperature rises up to about 1,200°C as shown in Table 1. Feedstocks were placed in the reactor when it was cold, and the top closed, and then the syngas burner was turned on. Initially we have used hydrogen gas instead of syngas. By controlling an input of oxygen gas to the syngas burner, reactor temperature was slowly raised to the 1,200°C mark. The output gas flowrate versus the reactor temperature is shown in Figure 5. Surprisingly, different feedstocks show pretty much the same output pattern with respect to the temperature. There is a sharp change with syngas outputs at about 1,200°C. This experiment was carried out with the external steam injection. There were a plenty of steam in the reactor coming from the syngas burner.

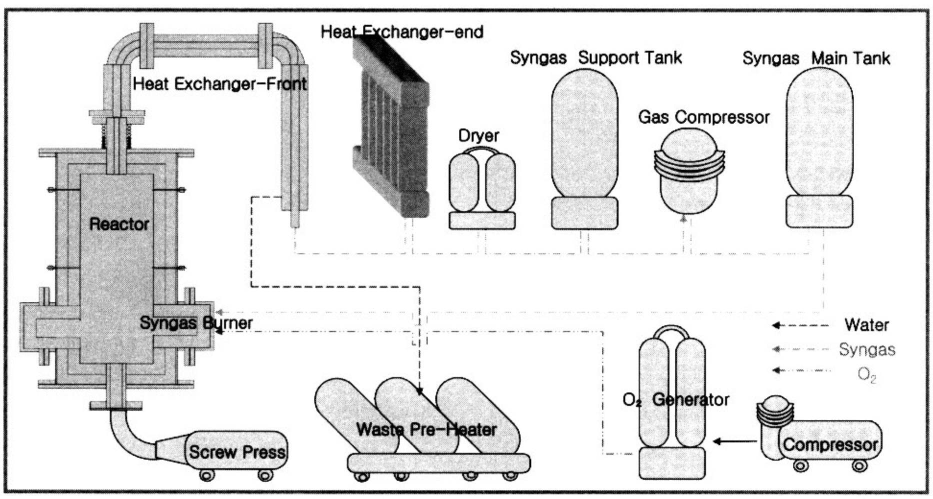


Figure 4. The schematic diagram of experimental apparatus.

Table 1. Total gas output measured with a gas flowrate meter

| Temperature (°C) | 1,000 | 1,100 | 1,150 | 1,180 | 1,200 | 1,220 | 1,300 | 1,400 | 1,500 |
|---|---|---|---|---|---|---|---|---|---|
| Flowrate (l/min) | 12 | 20 | 43 | 65 | 92 | 125 | 103 | 87 | 810 |

With $H_2$ gas analyzer and CO gas analyzer(AD systems, S. Korea), we measured percent compositions of each gas in the output gas. We have used waste auto oil having the weight percent composition of 65%C, 15%H, 16%O, 2%N and 2%S. The waste oil was fed into the reactor at the rate of 10kg per hour. Figure 5 represents $H_2$ production efficiency of the output gas versus temperature, when the syngas [(a) 50% $H_2$ and 50% CO and (b) pure $H_2$ gas] made up through the gasification process is recycled for supporting a heat source. It has been investigated that the $H_2$ production efficiency reaches their maxima at about 1,200°C.

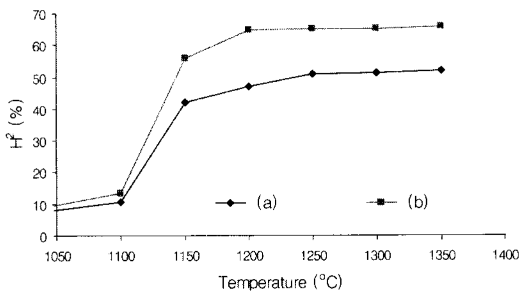


Figure 5. The $H_2$ production efficiency of the syngas output: (a) when syngas (50% $H_2$ and 50% CO) made up through the gasification process is recycled to syngas burner; (b) when syngas (100% $H_2$) is recycled to syngas burner for supporting a heat source.

Data shown in Table 1 and Figure 5 suggest that the gasification reaction reaches a optimum peak at about 1,200°C. In a conventional reactor where the supplied oxygen reacts with the feedstock to provide the heat for the reforming reaction, the temperature is not uniform within the reactor. And the reaction reaches sporadically maximum production of output at the 1,200°C temperature within the reactor. Therefore, the effect of reactor temperature on the reaction rate was not measurable. Studies have been continuing to separate component gases and also to identify minor gas components such as SOx, NOx, $H_2S$, dioxins etc. In this process, $O_2$ gas is used to only burn syngas only and none reacts directly with feedstocks. And the reactor is not opened into atmosphere as is the case with combustion and incineration.

# DISCUSSION

In conventional gasification methods, oxygen is supplied with carbonaceous compounds (-$CH_2$) to the gasification reactor, thereby inducing the oxidation of carbon and hydrogen components in the carbonaceous feedstocks and producing combustion heat from the oxidation to maintain the high temperature required for the gasification in the reactor. The oxidation reaction is indicated as follows:

| | ΔH(25°C) kJ/mol | ΔH(1,300°C) kJ/mol | |
|---|---|---|---|
| $C + O_2 \rightarrow CO_2$ | -393.51 | -378.70 | (1) |
| $(-CH_2-) + 3/2\ O_2 \rightarrow H_2O + CO_2$ | * -802.61 | * -800.11 | (2) |

('*' = in the case of $CH_4$)

Reaction 1 indicates the combustion reaction usually occurs in coal whose main component is carbon, and Reaction 2 is the main combustion reaction in large molecular weight waste organic materials such as waste oil.

The requirement of oxygen, which depends on whether coal (C) or waste oil (-$CH_2$) is supplied into the reactor, amounts to 0.5~1.0 times the weight of the coal or waste oil. The oxygen supplied into the reactor is consumed according to the Reactions 1 and 2 to increase the temperature in the reactor and produce combustion products, $H_2O$ and $CO_2$.

The combustion products undergo gasification reactions with carbon, which is the main component of the organic materials, as indicated in Reactions 3 and 4. The gasification reactions require longer reaction time and higher temperature as compared with the combustion reactions. The gasification reactions of organic materials such as waste oil (-$CH_2$) are indicated by Reactions 5 and 6.

| | ΔH(25°C) kJ/mol | ΔH(1,300°C) kJ/mol | |
|---|---|---|---|
| $C + H_2O \rightarrow CO + H_2$ | 131.28 | 158.19 | (3) |
| $C + CO_2 \rightarrow 2\ CO$ | 172.45 | 193.02 | (4) |
| $(-CH_2-) + H_2O \rightarrow CO + 2H_2$ | *205.80 | *238.84 | (5) |
| $(-CH_2-) + CO_2 \rightarrow 2CO + H_2$ | *246.97 | *273.67 | (6) |

('*' = in the case of $CH_4$)

While the Reactions 1 and 2 are oxidation reactions, the Reactions 3 to 6 are reduction reactions. The gas produced from the reactions is syngas whose main components are CO and $H_2$. Mechanisms for the formation of syngas($H_2$ and CO) in the high-temperature reformer is indicated in Figure 6.

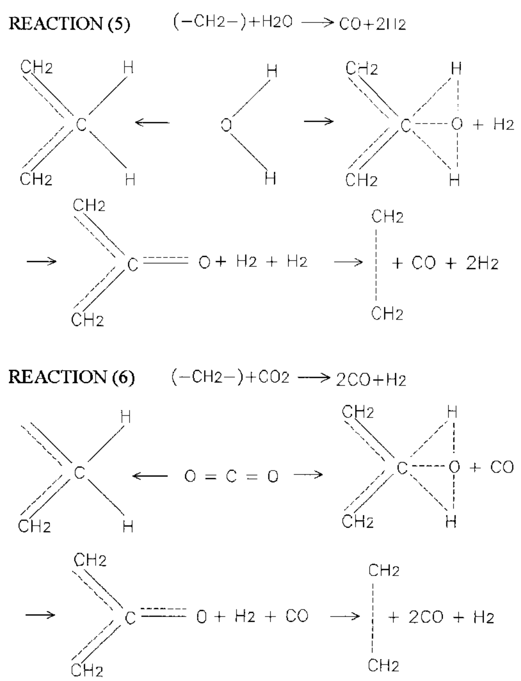


Figure 6. Mechanism for the formation of syngas ($H_2$ and CO) by the gasification of carboneous Wastes.

In conventional gasification methods, gasification reaction (Reactions 3 to 6) uses oxidation reaction (Reactions 1 and 2) which is induced by oxygen supplied with coal or waste oil for increasing the temperature of the gasification reactor. Further, an additional supply of steam is required to increase the concentration of hydrogen through water gas shift reaction (Reaction 9). The steam is acquired by means of heat exchange with the output syngas in the boiler installed for cooling the syngas in the gasification reactor.

| | ΔH(25°C) kJ/mol | ΔH(1,300°C) KJ/mol | |
|---|---|---|---|
| $H_2 + 1/2\ O_2 \rightarrow H_2O$ | -241.81 | -251.03 | (7) |
| $CO + 1/2\ O_2 \rightarrow CO_2$ | -282.98 | -285.86 | (8) |
| $CO + H_2O \rightarrow H_2 + CO_2$ | -41.21 | | (9) |

As described above, in conventional gasification methods, oxidation reactions (Reactions 1 and 2), reduction reactions (Reactions 3 to 6) and water gas shift reaction (Reaction 9) occur concurrently in the same reactor, and therefore, the production of hydrogen gas is low and secondary pollution usually occurs.

In this work the reactor was designed for prevention of the secondary pollution from the oxidation reaction through separation of the oxidation chamber from the reduction chamber, and in which the reaction heat is provided from the combustion of recycled syngas, thus no outside energy is needed for sustained reaction. Using this reactor, we have gasified variety of carbonaceous wastes such as coal, shredded tire, waste oil, and plastic waste materials without creating secondary pollution. With an appropriate amount of steam introduced into the reactor, better than 65% hydrogen gas output from the reactor was obtained.

Reforming reaction for a gasification of carbonaceous wastes such as shredded tire, waste lubricating oil, plastics, and powdered coal is initiated with heat and reactants(steam and carbon dioxide) generated from combustion of syngas($H_2$+CO). syngas is the product of the gasification reaction, and a portion of syngas produced is recycled into the syngas burner which is an integral part of the gasification reactor. The advantage of burning syngas to provide the heat for the reformng reaction is that the flame temperature of syngas combustion with oxygen gas is >2,000°C while the flame temperature of hydrocarbon combustion is below 1,000°C. With syngas burner, temperature of gasification reactor reaches readily up to the 1,400°C. Carbonaceous wastes are all gasified at about 1,200°C, and hydrogen gas fraction reaches 65% of the product gas output. With syngas burner, all the part of reactor is heated uniformly, and temperature control of the reactor is possible. Hydrogen fuel production at minimal cost is possible with this gasification technology using the high-temperature reformer. All forms of carbonaceous material including wastes are gasified, even hazardous wastes using this high temperature reformer.

# CONCLUSIONS

This present work relates to an apparatus in which all carbonaceous material such as coal, lubricant oil, plastics and any substance including carbon are reformed(gasified) into syngas at temperature above 1,200°C. It comprises a single-stage reforming reactor without catalyst. The method comprises the steps of supplying syngas and oxygen gas into syngas burners of the gasification reactor to produce steam and carbon dioxide gas, which in turn react with organic materials fed into the reactor to produce syngas. Carbon monoxide and hydrogen gas are discharged from the top of the reactor. And a portion of the discharged carbon monoxide and hydrogen gas is recycled into the syngas burner and produce more steam and carbon dioxide gas to react with the organic materials. Thus carbon monoxide and hydrogen gas is continuously produced as the product gas is recycled into the reactor to provide heat sources and reactants (steam and carbon dioxide).

This method facilitates the control of uniform temperature in the gasification reactor as well as produces syngas of high quality by increasing the concentration of hydrogen. At the reactor temperature above 1,200°C, the reformation reaction proceeds rapidly with the high production of hydrogen gas. The proposed reformer could be, with further studies for developing optimizing this reactor, not only suitable for production of hydrogen gas fuel at a minimal cost, but also a powerful tool to dispose of many types of carbonaceous wastes, which generate secondary pollution when disposed by means of incineration.

# ACKNOWLEDGEMENT

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