

Gasification characteristics of organic waste by molten salt

Kimihiko Sugiura^{a,*}, Keishi Minami^a, Makoto Yamauchi^a,
Shinsuke Morimitsu^b, Kazumi Tanimoto^c

^a Osaka Prefectural College of Technology, 26-12 Saiwai, Neyagawa, Osaka 572-8572, Japan

^b Takuma Co. Ltd., 1-2-1 Shinhama, Arai, Takasago, Hyogo 676-8540, Japan

^c National Institutes of Advanced Industrial Science and Technology-Kansai, 1-8-31 Midorigaoka, Ikeda, Osaka 563-8577, Japan

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Abstract

Recently, along with the growth in economic development, there has been a dramatic accompanying increase in the amount of sludge and organic waste. The disposal of such is a significant problem. Moreover, there is also an increased in the consumption of electricity along with economic growth. Although new energy development, such as fuel cells, has been promoted to solve the problem of power consumption, there has been little corresponding promotion relating to the disposal of sludge and organic waste. Generally, methane fermentation comprises the primary organic waste fuel used in gasification systems. However, the methane fermentation method takes a long time to obtain the fuel gas, and the quality of the obtained gas is unstable. On the other hand, gasification by molten salt is undesirable because the molten salt in the gasification gas corrodes the piping and turbine blades. Therefore, a gasification system is proposed by which the sludge and organic waste are gasified by molten salt. Moreover, molten carbonate fuel cells (MCFC) are needed to refill the MCFC electrolyte volatilized in the operation. Since the gasification gas is used as an MCFC fuel, MCFC electrolyte can be provided with the fuel gas. This paper elucidates the fundamental characteristics of sludge and organic waste gasification. A crucible filled with the molten salt comprising 62 Li₂CO₃/38 K₂CO₃, is installed in the reaction vessel, and can be set to an arbitrary temperature in a gas atmosphere. In this instance, the gasifying agent gas is CO₂. Sludge or the rice is supplied as organic waste into the molten salt, and is gasified. The chemical composition of the gasification gas is analyzed by a CO/CO₂ meter, a HC meter, and a SO_x meter gas chromatography. As a result, although sludge can generate CO and H₂ near the chemical equilibrium value, all of the sulfur in the sludge is not fixed in the molten salt, because the sludge floats on the surface of the carbonate by the specific gravity of sludge lighter than the carbonate, and is not completely converted into CO and H₂. Moreover, the rice also shows good characteristics as a gasifying agent. Consequently, there is high expectation to using the organic waste as a molten salt gasifying agent. However, this requires lengthening the contact time between the organic waste and the molten salt.

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1. Introduction

World energy consumption has continually increased since the Industrial Revolution, with a more dramatic increase in recent years. Fossil fuels make up 90% of this energy consumption, the increase of which is related to the economic development of developing countries and world population growth. Population growth and economic development not only increase energy consumption, but also dramatically increase the

output of garbage. In the past 20 years, household sludge has increased by as much as 280 million m³, and its disposal is a big problem [1]. A current disposal method is to reduce the sludge by incineration and reclaim or recycle the residue by processing it into tiles or some other product. However, as landfills become filled, there are few potential new landfill sites, and recycled tile does not sell well because the price is too high [2]. Organic waste from households and restaurants and other places is roughly 30 million tonnes a year. Food waste, including food which is past the expiration date or kitchen refuse, amounts to about 18 million tonnes a year and occupies 30% of community waste. While restaurants and convenience stores near livestock farms recycle some of their garbage as compost and domestic

* Corresponding author. Tel.: +81 72 820 8537; fax: +81 72 821 0134.

E-mail address: sugiura@sys.osaka-pct.ac.jp (K. Sugiura).

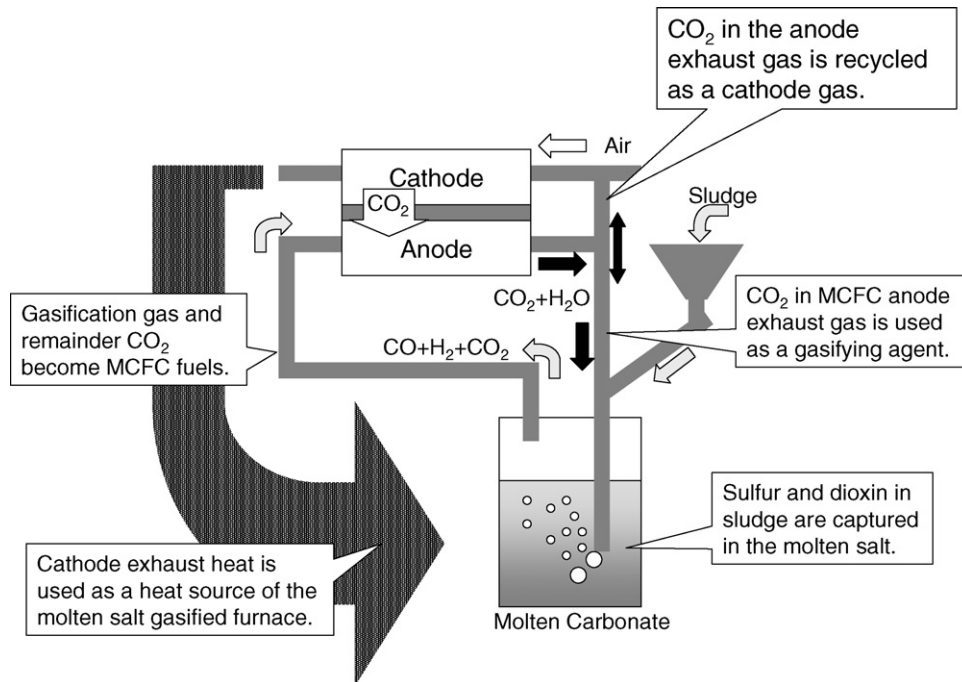


Fig. 1. Schematic diagram of the molten salt gasification.

animal fodder, the recycled garbage only accounts for about 9% of total garbage produced [3]. Most garbage is incinerated by entrepreneurs, placing a large responsibility on the entrepreneur because of high cost and environmental burdens such as dioxin emissions, serious issues for restaurants and convenience stores in urban areas. Increased energy consumption is also serious in urban areas, making it necessary to solve the two issues of stable energy supply and processing of organic waste in urban areas.

Although new energy developments such as fuel cells have recently been promoted as potential solutions to the question of the urban energy supply, there is still insufficient promotion to develop fuel production methods. Though the main current of a hydrogen fuel cell production method is the fossil fuel production of hydrogen by steam reforming, a method which only decreases the consumption speed of the fossil fuel, but cannot solve the problem of fossil fuel dryness. Recently, gasification of garbage has been highlighted, with methane fermentation comprising the main gasification system used by the organic waste as fuel [4–6]. However, the methane fermentation method takes a long time to obtain the gasification gas, and the quality of the obtained gas is made unstable by the activity of the methane bacterium. Moreover, because the gasification gas obtained by the methane fermentation method contains sulfur as H₂S, sulfur removal equipment is required to use the methane fermentation gas as fuel [7]. Therefore, a gasification system, which uses molten salt, is proposed because the gasification rate is fast and the molten salt absorbs impurities, such as ash and sulfur in the gasification gas. Generally, molten salt gasification has been researched by many researchers for the gasification of coal [8]. However, molten salt gasification was not easily adopted in spite of its good reactivity, because the mist of the molten salt is included in the gasification gas, corroding metallic pip-

ing and the heat exchanger, etc. This is due to poor matching of the molten salt gasification and equipment, which uses gasification gas, the adoption of MCFC is proposed as equipment that uses the gasification gas as shown in Fig. 1, because the MCFC used in operation requires molten salt as a decreasing electrolyte (molten salt). Moreover, this combined system has the merits of a gasifying agent use in anode exhaust gas, a gasification gas and residual CO₂ as an MCFC fuel and the use as a heat source of molten salt gasified furnace of cathode exhaust heat, etc. Combined MCFC and molten salt gasification solves the problem of organic waste and energy supply. Therefore, this paper elucidates the fundamental characteristics of the gasification of sludge and the organic waste. In this instance, rice is selected as the organic waste.

2. Chemical equilibrium calculation of the sludge

In order to elucidate the fundamental characteristics of sludge gasification, the chemical equilibrium calculation of the sludge is carried out by the thermodynamic database software MALT-2. The components of the sludge used for the chemical equilibrium calculation and the gasification experiment are shown in Table 1. Here, the sludge is a sample supplied by the sewage plant of city A. Moreover, the input data for the chemical equilibrium calculation is rearranged from Table 1 as shown in Table 2. Here, the molten salt is 62 Li₂CO₃/38 K₂CO₃ for replenishing volatilizing and dispersing molten salt to MCFC as an electrolyte. Moreover, CO₂ and N₂ are verified as gasifying agents to examine the gasification characteristics.

Fig. 2 shows the results of calculating the chemical equilibrium of sludge using CO₂ and N₂ as the gasifying agents. CO₂ was selected as a gasifying agent because the anode-exhaust gas

Table 1
Components of the sludge

Water (H ₂ O)	0.6%	0.333 mol kg ⁻¹
Ash	17%-dry	
Volatile matter	73.5%-dry	
C	40%-dry	33.133 mol kg ⁻¹
H	6%-dry	59.64 mol kg ⁻¹
O	33.2%-dry	20.626 mol kg ⁻¹
N	3.5%-dry	2.485 mol kg ⁻¹
Total-S	0.21%-dry	0.066 mol kg ⁻¹
Total-Cl	0.35%-dry	0.098 mol kg ⁻¹
Si	21,000 mg kg ⁻¹	0.75 mol kg ⁻¹
Al	13,000 mg kg ⁻¹	0.481 mol kg ⁻¹
Na	1,320 mg kg ⁻¹	0.057 mol kg ⁻¹
Mg	3,550 mg kg ⁻¹	0.146 mol kg ⁻¹
K	1,820 mg kg ⁻¹	0.047 mol kg ⁻¹
Ca	19,900 mg kg ⁻¹	0.498 mol kg ⁻¹
Zn	1,070 mg kg ⁻¹	0.016 mol kg ⁻¹
Cu	230 mg kg ⁻¹	0.004 mol kg ⁻¹
Fe	8,670 mg kg ⁻¹	0.155 mol kg ⁻¹
Hg	0.71 mg kg ⁻¹	0 mol kg ⁻¹
Cd	1 mg kg ⁻¹	0 mol kg ⁻¹
L.H.V.	16,600 kJ kg ⁻¹ -dry	

Table 2
Input data for the chemical equilibrium calculation

H ₂ O	0.0333 mol
C	3.3133 mol
H	5.964 mol
O	2.0626 mol
N	0.2485 mol
Total-S	0.0065 mol
Total-Cl	0.0098 mol
Na ₂ O	0.0057 mol
MgO	0.0146 mol
K ₂ O	0.0047 mol
CaO lime	0.0498 mol
ZnO	0.0016 mol
CuO	0.0004 mol
Fe ₂ O ₃	0.0155 mol
CO ₂	3.3133 mol
Na ₂ CO ₃	1.9282 mol
Li ₂ CO ₃	2.0889 mol
Sludge	0.1 kg h ⁻¹
Molten carbonate	
Na ₂ CO ₃	160.04 mol g ⁻¹
Li ₂ CO ₃	139.96 mol g ⁻¹
Total	300 mol g ⁻¹
Gasifying agent	
CO ₂	0.0742 m ³ h ⁻¹
N ₂	0.0423 m ³ h ⁻¹

of MCFC is mainly composed of CO₂ and steam. Moreover, N₂ was selected for reference data. The general molten salt gasification reactions of sludge are shown below. Most reactions are solution loss reactions that use the carbon in the sludge:



When the gasifying agent is CO₂, the gasifying gas is almost composed of CO and H₂. Because CO is produced by reactions (r.1) and (r.2), CO₂ is the best for use as the gasifying agent. H₂ decreases a little with an increased reaction temperature because the reverse reaction of reaction (r.3) is caused by a temperature-

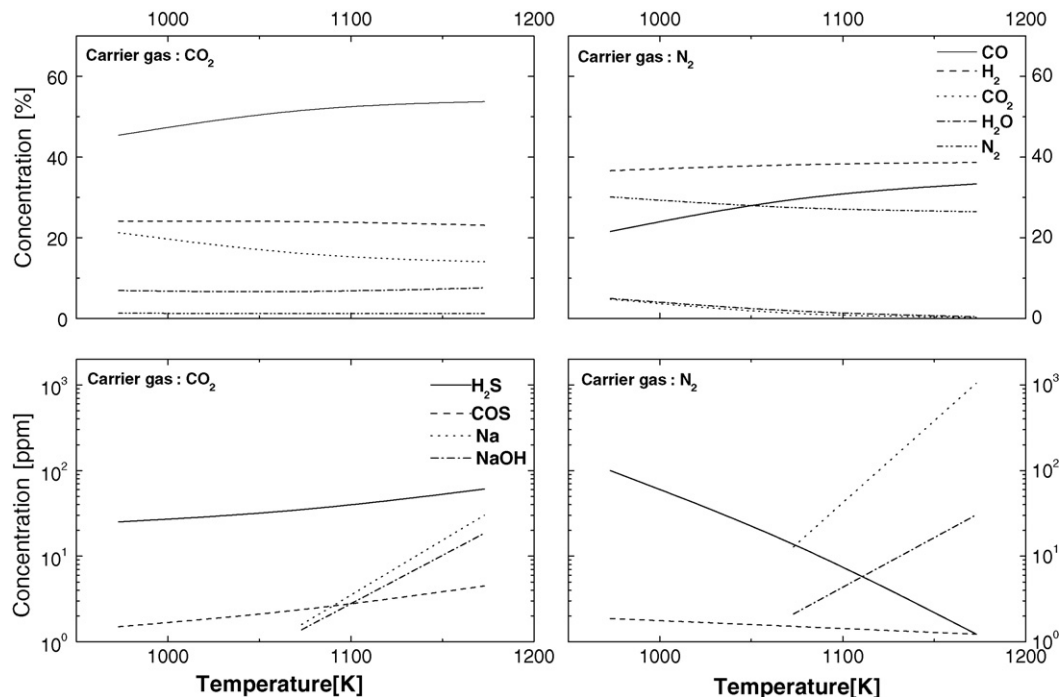


Fig. 2. Results of calculating the chemical equilibrium of sludge.

rise. Although the sulfur concentration is low, it is emitted as H_2S . Generally, because the molten salt is assumed to absorb sulfur, the sulfur concentration in the gasification gas by the gasification experiment can be expected to decrease below the chemical equilibrium computation. Under N_2 of the gasifying agent, the CO concentration is lower than H_2 because there is no CO_2 necessary for reaction (r.1). On the other hand, gasification has been carried out by the carbonate's decomposing from the discharge of Na, which is one of main components of the molten salt, as Na and NaOH. By this influence, it is understood that sulfur decreases by the reaction of the decomposed component of molten salt and sulfur in the sludge.

From the above-mentioned results, gasification of sludge is effective as fuel for MCFC, given that the main components are CO and H_2 , CO_2 being the best as the gasifying agent.

3. Experimental apparatus and procedures

Fig. 3 shows a schematic diagram of an experimental apparatus. The reactor is maintained at a preset temperature by a PID controlled electric furnace. A crucible filled with 300 g of (62 + 38) mol% (Li + K)/ CO_3 is installed in the reactor, and the temperature of the molten salt is measured by an alumina covered thermocouple. The reactor has an observation window and an irradiation window at the top of the reactor, the appearance of gasification in a crucible is captured by a high spatial resolution video camera (12 bit: 1300 pixel \times 1030 pixel) and metal halide illumination light (180 W). The sludge and CO_2 are supplied as the gasifying agent into the molten salt in a crucible through an Al_2O_3 tube (i.d.: 20 mm). Sludge is supplied from the funnel installed in the upper part of the alumina tube in the fundamental gasification characteristics experiment. In this instance, sludge hardened like a cake is crushed with the hammer, and crushed sludge only of a size of 1–2 mm is extracted with a sieve and used as an experimental sample. Moreover, the amount of sludge

Table 3
Experimental conditions

Molten salts	62 mol% Li_2CO_3 /38 mol% K_2CO_3
Mass of molten salts	300 g
Temperature of molten salts	500, 600, 700, 750 °C
Feed rate of sludge	0.667 g min^{-1} (3, 5, 10 min)
Rice	0.3 g (one unit)
CO_2 as gasifying agent	1.0 L min^{-1}
N_2 for dilution	4.0 L min^{-1}

supply is controlled by a screw feeder in the gasification characteristic experiment by continuous feed. CO_2 is controlled by a flow meter in each experiment. CO, CO_2 , Hydrocarbon and SO_2 in the gasified gas are analyzed by a CO/ CO_2 /HC meter (HORIBA Co. Ltd.; MEXA-554J) and an SO_x meter (Kyoto Electronics Manufacturing Co. Ltd.; SL-02), respectively, and the data are recorded online by a computer. H_2 is gathered in a sample pack every three minutes, and analyzed by gas chromatography (Shimadzu Co. Ltd.; GC-8A). The gasified gas is diluted with N_2 of 4 L min^{-1} at the exhaust pipe because the analytical critical value of each analyzer is low and each analyzer requires a large amount of sample gas. Moreover, the flow rate of N_2 for the dilution is confirmed with a soap film flow meter in each experiment. Therefore, each gas concentration of the experimental results is corrected, excluding the dilution nitrogen.

The experimental conditions are shown in Table 3. The temperature of the molten salt varies from 500 to 750 °C to verify the optimum temperature. Organic waste comprising sludge and rice is fed into a reactor in units of 0.3 g or is continuously fed by the feeder (the feed rate of 0.667 g min^{-1} at 3, 5 and 10 min). CO_2 is supplied as the gasifying agent by 1 L min^{-1} .

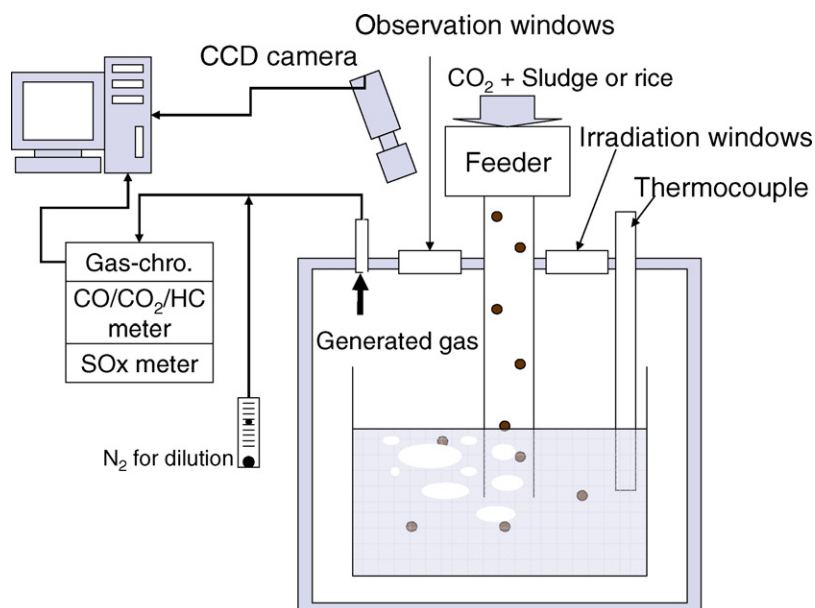


Fig. 3. Schematic diagram of the experimental apparatus.

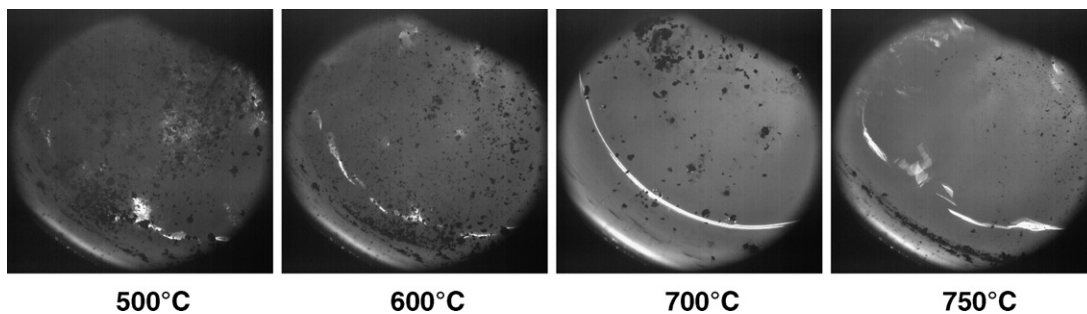


Fig. 4. Measurement image of the reaction phenomenon.

4. Results and discussion

4.1. Optimization of gasification temperature

In order to examine the gasifying characteristics of the sludge, the reaction temperature is optimized by observing the reaction phenomenon using image measurements. Fig. 4 shows the measurement image of the reaction phenomenon after 5 min from the sludge supplied at each reaction temperature. The sludge is floated in the molten salt at a reaction temperature of 500 °C, and does not disappear with the passage of time. A reaction temperature of about 600 °C shows some improved reactivity, and unreacted sludge remains on the molten salt. Therefore, there is little gasification reaction at reaction temperatures of 500 and 600 °C. On the other hand, 80% of the sludge can be gasified at a reaction temperature of 700 °C, and all the sludge can be gasified at 750 °C in a few minutes. Moreover, it is confirmed that the suspended matter on the molten salt in 750 °C is ash. Therefore, the gasification temperature is established at 750 °C.

4.2. Fundamental gasification characteristics of the sludge

To evaluate the ability as the gasification fuel of the sludge, the gasification gas composition is measured with a small amount of sludge (0.3 g) and the change of the CO₂ concentration of the gasifying agent. In this instance, the CO₂ concentration is varied as 100%, 80%, 50% and 20%, and the remainder is balanced by N₂. The temperature of the molten salt is 750 °C. The change of concentration of CO, SO_x and HC are shown in Fig. 5. Here, Fig. 5 also shows the measurement image of gasification under conditions of 20 CO₂/80 N₂ after 15 and 300 s from supplying the sludge. The sludge gasifies when supplied into the molten salt and almost finishes gasifying in less than 30 s because the peak of the CO and HC concentration is about 15–30 s. As shown in the measurement image of Fig. 5, in spite of a low CO₂ concentration of 20% (with few gasifying agents), most of the sludge finished gasifying by 300 s. Therefore, it is understood that the reactivity of the molten salt gasification is excellent. On the other hand, the peak of SO_x concentration is about 80–100 s, and SO_x is gradually emitted until reaching a concentration of 200–300 ppm. Generally, though the molten salt is assumed to absorb sulfur, SO_x is emitted. It is understood that because the sludge is floating on the molten salt as shown in the measurement image of Figs. 4 and 5, sulfur emitted as SO_x before being

absorbed into the molten salt. Moreover, because the SO_x generation does not depend on the CO₂ concentration, it is understood that the contact time of the sludge and the molten salt is related to the control of the sulfur concentration in the gasification gas. Kawase et al. said that the solubility of H₂S by the molten salt in MCFC relates to partial pressure of gas composition according to reaction (r.4) and the current density [9]. Because this molten salt gasification uses CO₂ as gasifying agents, the sulfur, which dissolved to the molten salt, is emitted as H₂S by the reverse reaction of four expressions. Moreover, because Kawase et al. also said that reaction (r.4) is slowly caused, the contact time of

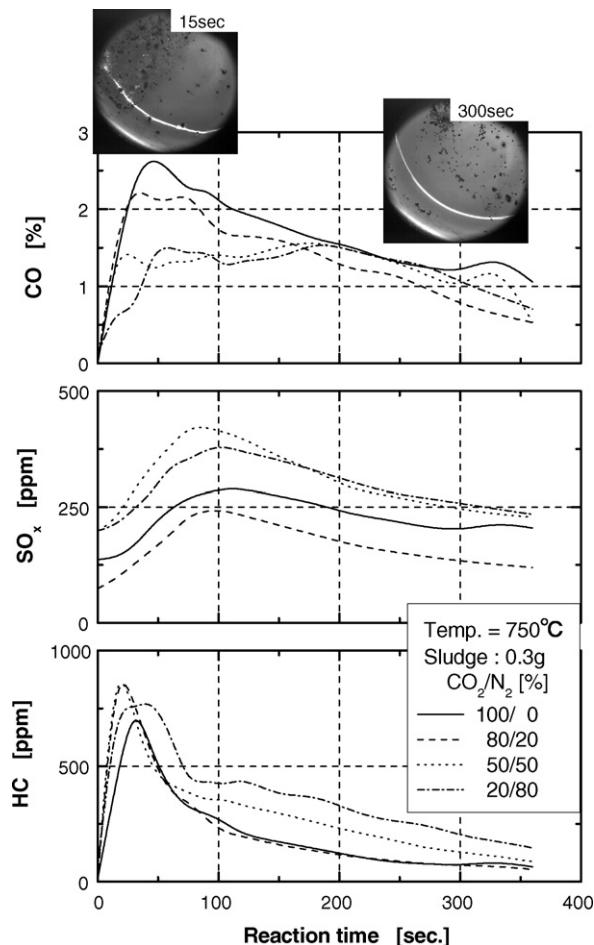
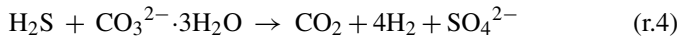


Fig. 5. Gasification characteristics of the sludge.

the sludge and the molten salt should be longer for absorbing the sulfur into the molten salt:



The above results confirm that the gasification of sludge is effective as a fuel for MCFC, given that main components are CO and H₂, as well as from the chemical equilibrium calculation. However, because sulfur causes the anode electrode and the MCFC electrolyte deteriorates, it is necessary to avoid the inclusion of sulfur in the MCFC fuel. Because high H₂O concentration promotes the solubility of H₂S by the molten salt from reaction (r.4) and promotes the gasification too from reaction (r.2), H₂O should be added as the gasifying agent, and we understand that the sludge of high water content is more desirable for this gasification.

4.3. Gasification characteristic by continuous feed of the sludge

The ability as the gasification catalyst of the molten salt is evaluated by continuously supplying sludge. In this instance, the supply time of the sludge is 3, 5 and 10 min, and the total sludge mass becomes 2.0, 3.3 and 6.7 g, respectively. Fig. 6 shows the lapse of time of the CO and H₂ concentration with the change of the amount of supplied sludge. Here, the vari-

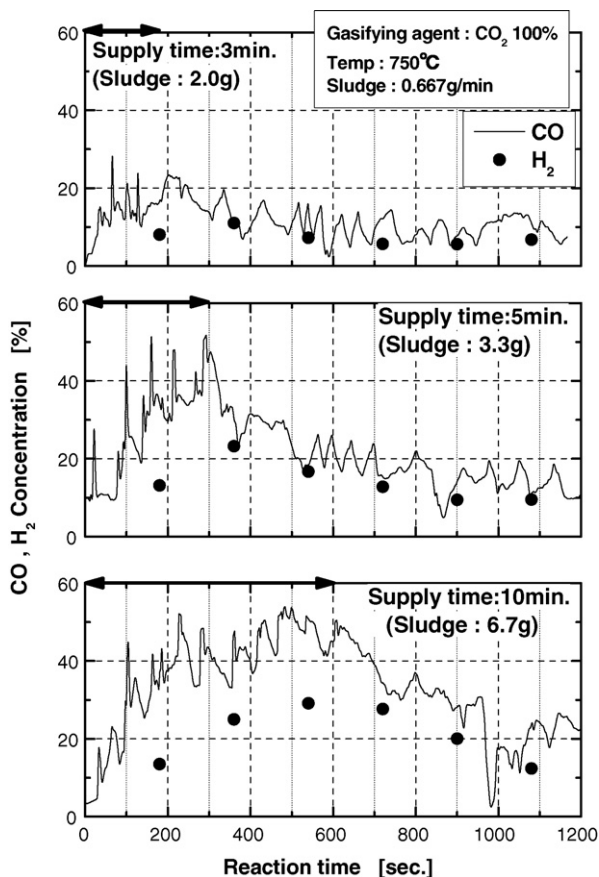


Fig. 6. Lapse of time of the CO and H₂ concentration with the change of the amount of supplied sludge.

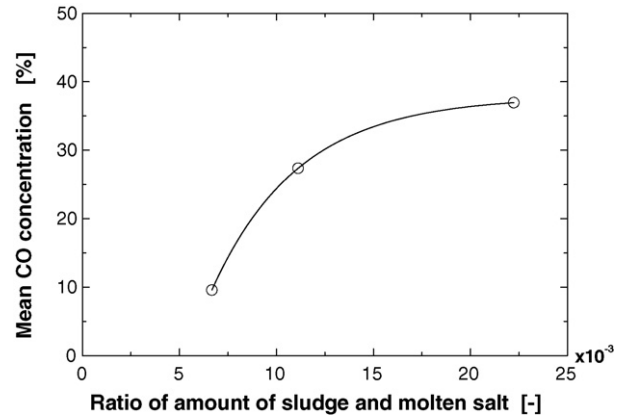


Fig. 7. Influence on gasification characteristic of the sludge amount supplied.

ation of CO concentration is large because sludge is supplied in mass, and gasifies as mass decomposing in the molten salt. The CO and H₂ concentration increases while supplying the sludge under all conditions, and are saturated when the supply time exceeds 300 s. At a supply time of ten minutes, the CO concentration becomes constant at about 42%, and H₂ becomes about 30%, respectively. As the CO and H₂ concentration of the chemical equilibrium calculation at 750 °C are about 50% and 24%, respectively, the experimental value of the CO is a little low, and that of the H₂ is a little high. It is understood that this also originates in the poor contact of the molten salt and the sludge caused by floating the sludge on the molten salt. Moreover, much soot is collected by the filter located upstream of the analyzer while experimenting, and an especially great amount of soot is generated with an increased supply of the sludge. It is understood that there is a lot of supplied sludge compared with the amount of the molten salt or the few gasifying agents. Therefore, the mean CO concentration, which is main component of gasification gas, is rearranged to compare the ratio of the sludge amount supplied and the amount of molten salt as shown in Fig. 7. Here, the mean CO concentration is defined, by which the CO concentration is integrated when supplying the sludge, and is divided by the supply time of the sludge. The mean CO concentration increases logarithmically with the increase of the sludge amount supplied, and is almost saturated at a lower value than the chemical equilibrium calculation value of under 2.5% of the ratio of the amount of sludge and molten salt. However, the sludge is floated on the molten salt from the measurement image of Figs. 5 and 6. Because the molten salt does not create enough contact between the sludge and the gasifying agent, it is necessary that the molten salt gasification of sludge cause the sludge to stay in the molten salt for a long time. In the future, it will be necessary to examine the influence of steam on the gasification characteristics.

4.4. Fundamental gasification characteristics of organic waste

The treatment of organic waste is a serious problem in urban restaurants and convenience stores, etc. It is understood that the

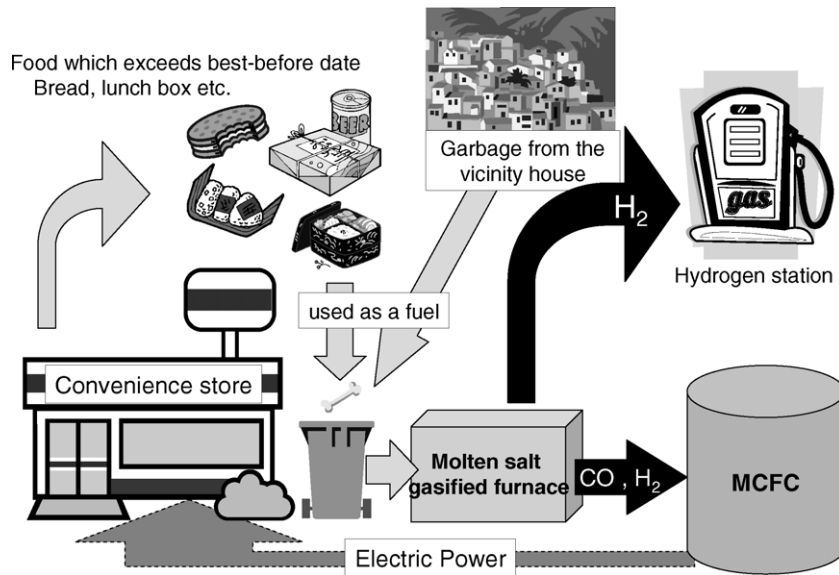


Fig. 8. Schematic diagram of molten salt gasification system with the organic waste.

combined MCFC and molten salt gasification solves the problem of the organic waste and the energy supply. Therefore, a combined system such as that shown in Fig. 8 is proposed. This system is located near a convenience store, and uses garbage as a molten salt gasification fuel. The garbage used as fuel is food, which have past the expiration date, such as bread and lunch box meals, etc., and it is gathered from closely proximate houses. The gas, which gasifies using the molten salt, is used as an MCFC fuel, or the fuel of a hydrogen station. If the gasification gas is used for the MCFC, electricity and hot water generated by the MCFC are supplied to the convenience store. The fundamental gasification characteristics of the organic waste were evaluated for this system. There are a lot of lunch boxes and rice balls in the garbage thrown out by the convenience store. Because the main component is rice, the gasification characteristics of the rice on this paper was evaluated, including rice supplied into the molten salt a funnel as well as the fundamental gasification characteristics of the sludge. The optimized temperature of the rice gasification was evaluated by changing the temperature of the molten salt. The results are shown in Fig. 9. In this

Table 4
Components of the rice

Water (H ₂ O)	13.16%
Ash	0.39%-dry
Volatile matter	89.46%-dry
C	43.25%-dry
H	6.36%-dry
O	1.39%-dry
N	48.53%-dry
Fixed carbon	10.15%-dry
Total-S	0.05%-dry
Inflammability-S	0.05%-dry
Total-Cl	<0.05%-dry
Inflammability-Cl	<0.05%-dry
Na	120 mg kg ⁻¹
K	660 mg kg ⁻¹
Ca	91 mg kg ⁻¹
Mg	230 mg kg ⁻¹
Fe	32 mg kg ⁻¹
Zn	21 mg kg ⁻¹
Si	<500 mg kg ⁻¹
H.H.V.	17,300 kJ kg ⁻¹ -dry

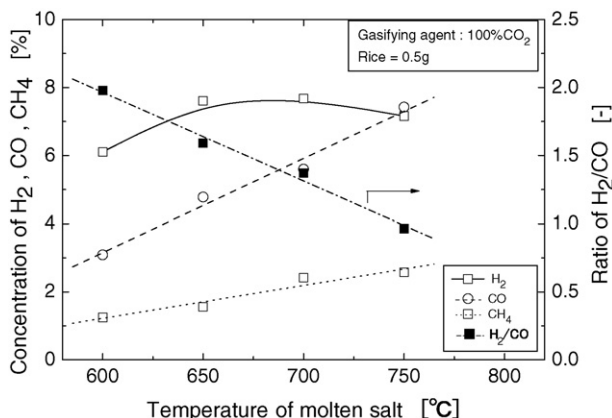


Fig. 9. Influence on gasification characteristic of the molten salt's temperature.

instance, experimental conditions are as follows. The gasifying agent is 100% CO₂ (1 L min⁻¹). The temperature of the molten salt is changed at 600, 650, 700 and 750 °C. Rice, which is cleaned rice, and not cooked rice, is supplied at 0.5 g. Though CO and CH₄ increase with a temperature increase of the molten salt, H₂ is saturated from 700 °C. Because the reverse reaction of reaction (r.3) is caused by a temperature-rise, H₂ decreases and CO increases. Because the quality of the gasification gas is good when the temperature of the molten salt is 750 °C, the gasification temperature of the rice is established at 750 °C. Fundamental gasification characteristics are evaluated by supplying 1.0 g of rice. The moisture amount of the rice is 13.16%, and is larger than that of the sludge, as shown in Table 4. Though the carbon and the hydrogen content of rice are almost same as the sludge, the oxygen content of the rice is less than that

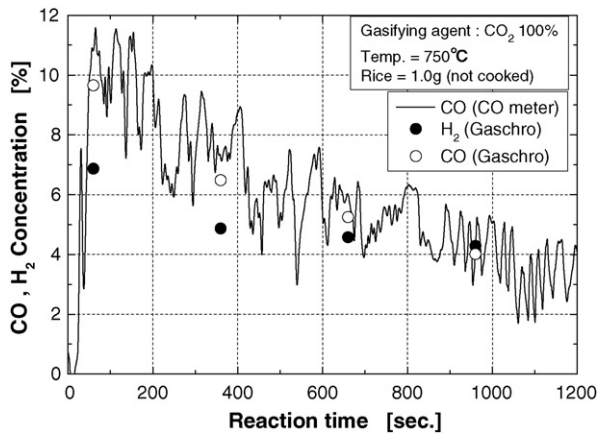


Fig. 10. Gasification characteristics of the rice.

of the sludge. Generally, rice has about 0.05% of inflammable sulfur, present as sulfur amino acids such as methionine, cysteine, glutathione, etc. [10]. CO concentrations obtained by a CO meter and gas chromatography are shown in Fig. 10. Because the results of two analyzers are almost the same, the result of the CO meter can be ascertained from the results of gas chromatography. Although the CO and H₂ concentration are higher than that of sludge because the supplied amount of rice is greater than that of the sludge, the soot generated from the rice is less than that of the sludge. When the rice supplied is 0.3 g as well as the supplied amount of the sludge, the gasified gas composition can be compared with the sludge. Fig. 11 shows the concentra-

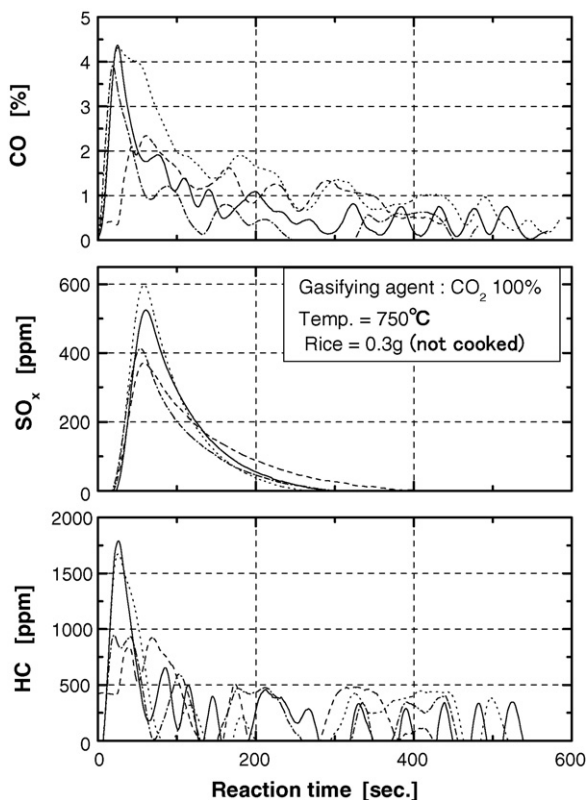


Fig. 11. Lapse of time of the gasification characteristics of the rice.

tion of CO, SO_x and HC when supplying the rice. Experiments were carried out four times to achieve reproducibility. Each data shows a similar tendency though there is some dispersion. The CO of the rice is about 4%, greater than that of the sludge. Moreover, though the oxygen content of sludge is 30 times that of rice, the gasified gas such as CO is low. Therefore, it is understood that because the moisture of the rice is larger than that of the sludge, the moisture promotes greater reactivity than does the oxygen content.

On the other hand, sulfur is generated immediately when the rice is supplied into the molten salt. The peak of the sulfur concentration is about 500–600 ppm. This concentration means that most of the sulfur in the rice is expelled. Therefore, the contact time of the rice and the molten salt is very short as well as with the sludge case because the specific gravity of the rice is lower than that of the molten salt. The same explanation is provided regarding the HC concentration. HC of the rice is about 1500–1700 ppm, and is larger than that of the sludge. It is understood that as the rice has more moisture than the sludge, carbon that was not able to react to CO becomes HC when reacting with moisture. Therefore, though the gasification of the sludge easily causes a carbon deposition, the gasification of rice causes little carbon deposition. If the contact time of the rice and the molten salt is enough and the rice has more moisture, most HC will react to CO.

Consequently, although it is necessary that the gasification of the organic waste by molten salt cause the organic waste to stay in the molten salt for a long time, the gasification of the rice is promising.

5. Conclusions

The objective of this study is to elucidate the fundamental characteristics of the gasification of sludge and the organic waste. The obtained results in this study are summarized as follows:

- (1) The gasification of sludge is effective as an MCFC fuel given that the main components are CO and H₂, and CO₂ is the best when used for the gasifying agent as determined by the chemical equilibrium calculation.
- (2) The molten salt temperature needs to be 750 °C for gasifying the sludge and the organic waste.
- (3) The gasification of sludge is effective as the MCFC fuel given that the main components are CO and H₂.
- (4) Sulfur cannot be removed by molten salt during the gasification process because the contact time of the sludge and the molten salt is very short.
- (5) CO and H₂ of gasified gas are lower than the chemical equilibrium calculation, and the sludge is floated on the molten salt because the specific gravity of the sludge is lower than that of the molten salt.
- (6) Although the gasification of rice is effective as an MCFC fuel given that the main components are CO and H₂ as well as the case of sludge, a lot of SO_x and HC are generated in a short contact time between the rice and the molten salt.

- (7) Moisture in the organic waste promotes gasification reactivity.
- (8) The sludge and the rice are promising the gasification fuel if the molten salt can absorb sulfur enough.

Acknowledgement

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