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Gasification of municipal solid waste in a pilot plant and its impact on environment

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Abstract–Municipal solid waste from three cities was gasified in a 3 ton/day capacity gasification/melting pilot plant based on Thermoselect at a temperature of around 1,200 °C using double inverse diffusion flame burner. The synthesis gas (syngas) obtained from gasification contains 25-34% CO and 28-38% of H_2. The high heating value of syngas was in the range of 10.88-14.65 MJ/Nm^3. Volatile organic compounds like furan, dioxin, and other organics in gaseous and liquid phase were effectively destroyed because of the high temperature of the high temperature reactor and shock cooling of syngas. Pollutants in exhaust gases were also found to be satisfying the Korean emission standard. Leaching concentration of heavy metals in the melted slag (vitrified mineral aggregate), fly ash, and treated water was much less than the Korean regulatory limit values due to high melting temperature (1,600 °C). The vitrified slag was of dark brown color. The glassy and amorphous nature of the vitrified mineral aggregate was further confirmed from SEM micrograph and XRD spectra of slag. The vitrified mineral aggregate could be used as natural raw material in cement and construction industry.

Key words: Municipal Solid Waste, Thermoselect Process, Gasification, Synthesis Gas, Heavy Metals

INTRODUCTION

Due to industrialization, the quantity of MSW has increased in Korea raising the question of its sustainable disposal management [Sakai et al., 1996; Tchobanoglous et al., 1993]. Waste management options include waste collection and sorting followed by one or more of the following options: resource recovery through recycling [Park et al., 2002, 2003; Kaminsky 1992], biological treatment of biomass, i.e., production of marketable compost [Goldstein et al., 1996], thermal treatment, i.e., incineration to recover energy in the form of heat and electricity and landfilling [Lee et al., 2000]. Landfilling of MSW release green house gases (GHGs) [IPCC, 2001; Park and Shin, 2001], volatile organic compounds and leachable toxic heavy metals to the surrounding environment [Christensen, and Christensen, 2000; Hur and Kim, 2000; Hur et al., 2001; Kim and Kim, 2002; Kjeldsen et al., 2002; Sanin et al., 2000]. Over the years, incinerating waste to generate energy has become the most common method of dealing with combustible waste efficiently as it decreases the volume and mass of MSW [Park and Heo, 2002]. But, incineration has drawbacks as well as particularly hazardous emissions and harmful process residues [Chandler et al., 1997]. Moreover, incineration of MSW generates fly and bottom ashes which release leachable toxic heavy metals, dioxin, furans and volatile organic compounds [Li et al., 2003a; Hyun et al., 2004; Park et al., 2005]. Stringent environmental regulations are being imposed to control the environmental impact of MSW and incinerator residues [MOE report, 2000].

Furthermore, the experiences of the waste incineration industry driven in the past by regulatory as well as technical issues may facilitate their commercial potentials outside the common market especially in highly populated developing countries like Korea with scarce landfill sites. The total amount of MSW generated by Korea was 50,763 tons per day in the year 2003 [MOE report, 2005]. About 4.6 million tons of total waste is being incinerated per year that leads to the generation of a large amount of solid residues including fly ash and bottom ash and also emits hazardous gases to the environment [Kim, 2003]. Moreover, thermal waste disposal can no longer be seen as a process for the reduction of the amount of MSW by weight and volume with the disposal of the generated ashes and Air Pollution Control residues on landfill sites or their application in cement and construction industry. Thus, there is a need to consider MSW as a valuable indigenous source of fuel abundance especially in consumer-oriented societies able to substitute fossil fuels in power generation and other industrial processes. Increasing space constraints for landfilling of MSW and public opposition to new incinerators for waste disposal has effectively eliminated this as a future option in many countries.

In recent years, several new technologies which involve gasification or combinations of pyrolysis, combustion and gasification processes are being brought into the market for energy efficient, environment friendly and economically sound methods of thermal processing of wastes [Choi et al., 2006; Lee et al., 2006; Fung and Kim, 1990; Ko et al., 2001; Malkow, 2004; Yun and Yoo, 2001; Yun and Ju, 2003]. Gasification of MSW yields syngas that has a potential to be used as a fuel in fuel cells or fuel cell vehicles [Björklund et al., 2001; Jun et al., 2004; Kim et al., 2002].

Recently, Daewoo Engineering & Construction Corporation installed a high temperature recycling pilot plants for waste of any kind

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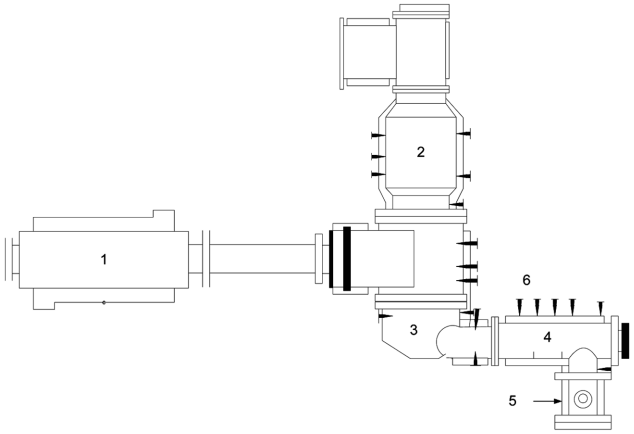

Fig. 1. Schematic diagram of gasification/melting pilot plant.

1. Degassing facility
2. High temperature reactor
3. High temperature reactor bottom
4. Homogenization reactor
5. Granule cooling tank
6. Double inverse diffusion flame burner

with a patented process called Thermoselect with the collaboration of THERMOSELECT, Switzerland. The process consists of compression, degassing with fixed bed oxygen blown gasification, and melting of mineral residues [Kiss et al., 1994]. In our previous paper [Kwak et al., 2005], we studied the gasification of municipal solid waste using double inverse diffusion flame burner and double normal diffusion flame burner. The goal of this paper is to study the environmental aspects of gasification of MSW in this pilot plant with double inverse diffusion flame burner.

MATERIALS AND METHODS

1. Gasification of MSW

MSW from three cities (S-3, S-4, S-5) was used for gasification in 3 ton/day capacity pilot plant which is shown schematically in Fig. 1. The flow diagram of the gasification process is shown in Fig. 2. In this process, pyrolysis and gasification processes are carried out in a single unit [Stahlberg, 1992; Feuerriegelet al., 1994]. The stages of waste preparation and sorting are eliminated. The wastes are compressed to about one fifth of their initial volume by using a hydraulic press in a long canal heated from outside and maintained at temperatures higher than 600 °C. The high degree of compaction greatly reduces the residual air content, nitrogen does not need to be heated and subsequently cleaned, and heat conductivity is significantly improved. Liquids which escape during compaction flow into the remaining cavities. The compression enables the canal to be airtight. As wastes move through the canal they are heated, dried and nearly completely pyrolyzed by the time they reach the end of the canal. The products of pyrolysis then enter into the gasification zone where the materials are gasified with oxygen at a temperature of around 1,200 °C. A high quality synthesis gas and a molten byproduct are formed. The gas is rapidly cooled from 1,200 °C to 70 °C by spraying water through the gas. This rapid temperature reduction, i.e., shock cooling of gases combined with an absence of available oxygen avoids the reformation of dioxins, furans and other organic compounds from elementary molecules in the syngas due to the de novo synthesis back reactions [Huang and Buekens, 2001]. Thereafter, cleaned and made available for use either in power generation or as a raw material for chemical processes. The molten byproduct flows to the combustion zone where, with a supply of oxygen and propane gas, combustion takes place at a temperature in excess of 1,600 °C ensuring thermal destruction of all chlorinated carbon and stabilization of heavy toxic metals.

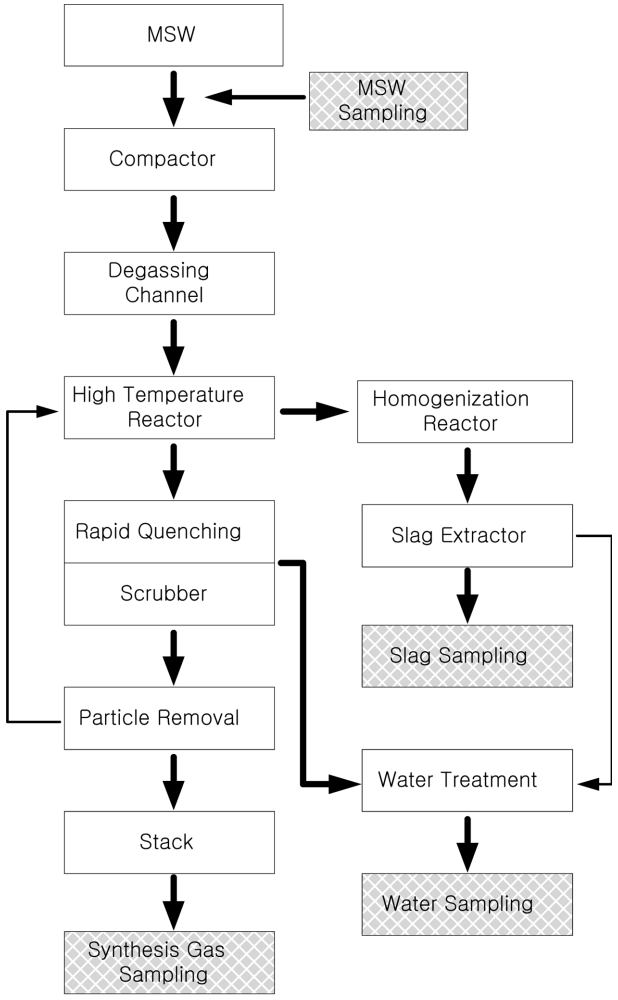

Fig. 2. Block flow diagram for 3 ton/day gasification/melting furnace.

Following the shock cooling, the synthesis gas flows through an acidic scrubber where further HCl and HF acids are removed. The acid content of the gas depresses the flushing liquid of these scrubbers to pH~3, which results in the volatilized heavy metals and their compounds dissolving as metal ions. Weaker acid formers such as H_2S, SO_2, and CO_2 do not dissolve at this pH value. The acid scrubber is followed by an alkaline scrubber which uses NaOH in solution to knock out any residual acid liquid droplets.

The process water originates from the condensed water vapor inherent in the processed waste and from the reaction products of the gasification process. Other small process water streams are generated from the gas scrubbing processes. The water treatment process occurs in batches, which is a reflection of the relatively small quantity of water undergoing treatment. The water from the quench circuit is settled; solids are removed. The water from the alkaline scrubber, with traces of hydrogen sulfide dissolved in it, is fed into

vessels and oxidized by using hydrogen peroxide. Soluble sulfate is formed and prevents the evolution of H_2S gas in later processing stages. A two stage precipitation then takes place. In the first stage NaOH is added and in the neutralization step in which the pH of the water is reduced to 7 with the addition of HCl acid.

2. Analysis of MSW and Slag

The composition of MSW was characterized by proximate and ultimate analysis. The MSW ash, fly ash, and melted slag were dried at a temperature of 110 °C for 24 h. After drying, these were pulverized to a size of <300 μm and were screened through a size of 300 μm. Chemical compositions of these samples were analyzed with X-ray fluorescence spectroscopy (XRF, PW 2400, Philips, The Netherlands).

The heavy metal content of these samples was analyzed by inductively coupled plasma atomic emission spectrometry (ICP-AES) except for Pb, Hg, and Cd for which atomic absorption spectroscopy (AAS) was used. The morphology of slag was examined via X-ray diffraction (XRD, PW 1710, PHILIPS, The Netherlands) using Cu-K radiation and scanning electron microscopy (SEM, S-3500N, Hitachi).

The chemical stability of the fly ash and melted slag was determined by Korean Standard Testing Method for Solid Waste (KS F 2563(1997)). The Korean Standard Leaching Test (KSLT) uses a leachate value of pH 5.8-6.3, solid weight and leachate volume ratio is 1 : 10 (W : V); the leaching time is 6 h and it agitates horizontally with 200 rpm. The leachate is filtered with 1-μm glass fiber filter.

Table 1. Composition of MSW

Property	Component	S-3	S-4	S-5
Physical composition[a] (%)	Plastics	20.1	20.7	22.5
	Paper	17.3	16.5	15.3
	Food	48.9	50.3	47.2
	Wood	3.2	3.9	4.9
	Fiber	4.3	3.2	5.1
	Metal	2.5	2.0	2.7
	Non-metal	3.7	3.4	2.3
Proximate analysis (%)	Moisture	52.7	50.5	34.2
	Volatile matter	32.4	36.4	48.2
	Fixed carbon	7.1	6.5	7.9
	Ash	7.8	6.6	9.7
Ultimate analysis[b] (%)	C	46.2	49.4	51.7
	H	6.1	4.1	5.1
	O	34.8	30.8	27.7
	N	1.3	0.9	1.4
	S	0.1	0.4	0.2
	Cl	0.5	1.7	1.4
	Ash	11.0	12.7	12.5
Apparent density (kg/m^3)		358	350	365
LHV (MJ/kg)		9.31	9.88	13.42

RESULTS AND DISCUSSION

MSW collected from three cities in Korea was used for gasification. The composition of MSW was characterized by proximate and

Table 3. Composition (%, dry volume basis) of synthesis gas along with high heating value (HHV)

MSW sample	HHV (MJ/Nm^3)	CO	H_2	CO_2	O_2	CH_4
S-3	10.88	34.23	37.54	27.43	0	0
S-4	11.30	24.80	32.70	23.30	0	0.9
S-5	14.65	31.41	27.66	28.60	0.07	2.23

Table 4. Exhaust gas composition of synthesis gas combustor

Phase	Component		Korean emission standards	Average detected value S-3	S-4	S-5
Gas phase (ppm)	CO		<600(12)	30	27	32
	SOx		<300(12)	12	10	14
	HCl		<50(12)	1.4	1.7	1.1
	NOx		<200(12)	28	35	31
	NH_3		<100	34	36	29
	H_2S		<15(12)	0.5	0.7	0.4
	Cl		<10	N.D.	N.D.	N.D.
	Carbon disulfide		<30	N.D.	N.D.	N.D.
	Hydrogen cyanide		<10	1.1	0.9	1.6
	F complex		<3.0(12)	N.D.	N.D.	N.D.
	B complex		<5	N.D.	N.D.	N.D.
	Benzene complex		<50	N.D.	N.D.	N.D.
	Phenol complex		<10	N.D.	N.D.	N.D.
	Formaldehyde		<20	0.5	0.7	0.6
	Hg complex		<5.0(12)	N.D.	N.D.	N.D.
	As complex		<3.0(12)	N.D.	N.D.	N.D.
Particulates (mg/Sm^3)	Dust		<100(12)	29	34	33
	Smoke		<2	0.7	0.9	1.0
	Heavy metal	Cd	<1.0(12)	N.D.	N.D.	N.D.
		Pb	<5.0(12)	0.25	0.22	0.27
		Cr	<1.0(12)	0.11	0.14	0.13
		Cu	<10	0.32	0.35	0.38
		Zn	<10	0.30	0.32	0.31
		Ni	<20	0.02	0.02	0.02

N.D. not detected

Table 2. Operating conditions for gasification of MSW

MSW sample	MSW input (kg/h)	LPG (L/min)	Oxygen flow rate (Nm^3/h)	Furnace pressure (kg/cm^2)	HTR temp. (°C)	Homogenized furnace temp. (°C)	HTR exit temp. (°C)
S-3	128.7	260	70	0.01-0.2	1,260-1,340	1,320-1,550	>1,200
S-4	128.7	257	92	0.01-0.2	1,280-1,390	1,270-1,580	>1,200
S-5	128.7	201	97	0.01-0.2	1,250-1,350	1,400-1,550	>1,200

ultimate analysis and the results are shown in Table 1 along with its low and heating value (LHV, HHV). In order to study the gasification characteristics of MSW in this system, the test results obtained from this pilot plant were analyzed.

The MSW was compacted and gasified at a high temperature of about 1,250 °C. The amount of oxygen supplied through the burner set in the upper reactor was adjusted to make the temperature about 1,200 °C at the exit of high temperature reactor. The summary of operational results is shown in Table 2 and the synthesis gas composition is shown in Table 3. It contains 25-34% CO and 28-38% of H_2. Table 4 illustrates the composition of exhaust gases from the synthesis gas combustor. The Korean emission standards shown in Table 4 were applicable to a incinerator with capacity under 200 kg/h until 2004. Looking at the concentration of pollutants, all values of pollutant gases were found to be much below the regulation level. Also, concentrations of toxic heavy metals emission level were below the Korean emission standard. The level of dioxin concentration. which is highly concerned in incinerator installation or operation resulting in only 0.03 ng-TEQ/Nm^3, was lower than the concentration allowed in Korea's large incinerator of 0.1 ng-TEQ/Nm^3. Acid gas scrubbing and particle removal which was adopted in this plant were composed of far more simpler installation than the normal technologies used in incineration installations. However, it was confirmed that dioxin level was much lower, which confirms one of the main advantages of employing the gasification melting process for MSW [Calaminus and Stahlberg, 1998; Stahlberg, 1992].

Table 5. Composition of melted slag and basicity (CaO/SiO_2)

MSW sample	Slag composition (wt%)									
	SiO_2	Al_2O_3	CaO	MgO	Na_2O	K_2O	P_2O_5	TiO_2	T-Fe	Basicity
S-3	37.59	18.37	10.37	2.97	1.11	0.65	2.15	0.93	13.82	0.28
S-4	39.84	17.84	9.93	3.78	1.28	0.51	2.21	0.72	14.23	0.25
S-5	38.43	16.73	11.58	3.17	1.32	0.49	2.41	1.05	14.95	0.30

Table 6. Heavy metal concentrations in MSW ash, slag, fly ash and waste water (ppm) along with their leaching concentrations (mg/L) for MSW sample S-3, S-4 and S-5

Metal	Ash of MSW		Slag		Fly ash		Waste water	Korean limit
	ppm	mg/L	ppm	mg/L	ppm	mg/L	mg/L	mg/L
MSW S-3								
Cr	1002	15.44	4125	0.042	1071	0.004	0.004	1.5
Mn	174	1.126	3952	0.023	104	0.896	0.012	10
Cu	24500	0.897	2584	0.010	4448	0.024	0.014	3
Zn	350	6.138	580	0.026	14150	3.26	0.520	5
As	138	0.006	87	0.001	62	0.001	0.005	0.05
Cd	11	0.004	0.42	0.002	29	0.178	N.D	0.3
Hg	2.4	0.002	0.48	0.001	0.72	0.001	N.D	0.005
Pb	911	3.505	3.45	0.003	1675	0.002	N.D	3
MSW S-4								
Cr	955	14.37	4518	0.042	1210	0.003	0.004	1.5
Mn	205	1.376	3950	0.022	130	0.813	0.016	10
Cu	25475	1.217	2420	0.010	4024	0.022	0.015	3
Zn	426	6.734	505	0.019	14140	3.04	0.658	5
As	125	0.006	99	0.001	62	0.001	0.005	0.05
Cd	12	0.004	0.55	0.002	35	0.185	N.D	0.3
Hg	2.7	0.003	0.45	0.001	0.81	0.001	N.D	0.005
Pb	915	3.456	3.2	0.001	1675	0.001	N.D	3
MSW S-5								
Cr	1155	15.85	4312	0.042	1125	0.005	0.002	1.5
Mn	158	1.150	3896	0.036	135	0.758	0.021	10
Cu	23586	1.195	2345	0.024	4350	0.021	0.009	3
Zn	375	5.980	475	0.029	15018	3.15	0.478	5
As	135	0.008	99	0.001	59	0.001	0.005	0.05
Cd	14	0.004	0.49	0.002	28	0.158	N.D	0.3
Hg	2.2	0.002	0.45	0.001	0.73	0.001	N.D	0.005
Pb	895	3.306	3.2	0.001	1515	0.001	N.D	3

N.D. not detected

The mineral components of the waste were transformed into inert and non-toxic primary materials in the form of slag. The composition of slag is illustrated in Table 5. It was mainly composed of SiO_2, Al_2O_3, CaO and Fe. Fly ash was actually the solid material of the cleaning water which was separated and dried. The heavy metal concentrations in MSW ash, melted slag, fly ash and waste water along with their leaching concentrations are shown in Table 6. Cr, Mn, Cu and Zn were the main heavy metals within the slag and fly ash. The higher content of some of the metals in melted slag and fly ash might be due to the refractory wears which was composed of materials like Cr corundum, bauxite, mullite etc [Ecke et al., 2001].

Next, we analyzed the leachability of heavy metals in the slag, fly ash and waste water. Table 6 illustrates the results of a leaching test of all streams which are also shown in Figs. 3-5 along with Korea's regulatory limit values [MOE, 2000]. Looking at the leaching level, it was found that the measured levels of all the heavy metals in all these streams were far below the Korea's regulatory limit value [MOE, 2000]. The leaching of heavy metals was reported to be proportional to the basicity (CaO/SiO_2) of slag [Maken et al., 2005; Takaoka et al., 1997] and a basicity of 0.24-1.24 was found good for effective stabilization of heavy metals [Li et al., 2003]. Thus, in our case, low basicity (0.25-0.30) of slag might be the reason for effective stabilization of heavy metals where SiO_2 acts as a glass former and CaO as a stabilizing agent [Barbieri et al., 2000]. It was reported by Ecke et al. [2001] that melting of MSWI ash at high temperature reduces the availability of Cr, Cu, Zn, and Pb for leaching as formation of glassy material decreases the surface area and traps the metal in the amorphous glassy matrix composed of Si, O, Al, and Ca. Characteristics of melted slag from MSW incineration & gasification/melting were also studied earlier, and it was found that during the melting treatment the emission of lead and zinc under the reducing condition was larger than that under the oxidizing condition; also under oxidizing conditions, phosphorous in ash promoted the vitrification of ash and suppressed the emission of volatile metals even at higher temperatures [Yoshiie et al., 2000, 2002; Jung et al.,

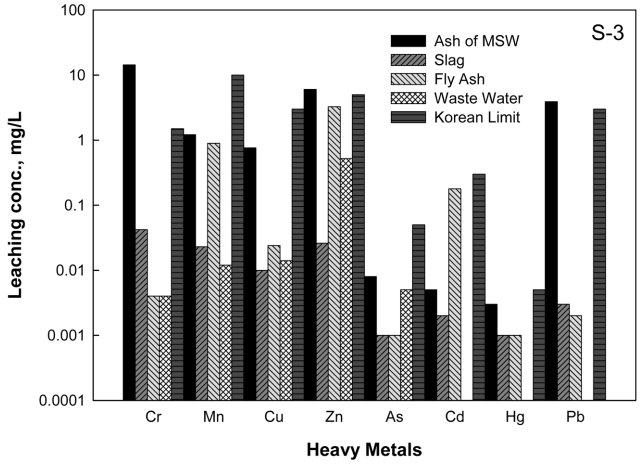

Fig. 3. Leaching concentration of heavy metals in different streams during the gasification using double inverse diffusion flame (DIDF) burner (MSW sample S-3).

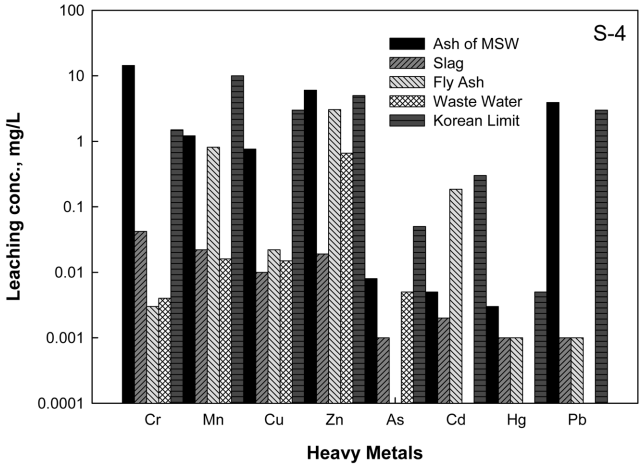

Fig. 4. Leaching concentration of heavy metals in different streams during the gasification using double inverse diffusion flame (DIDF) burner (MSW sample S-4).

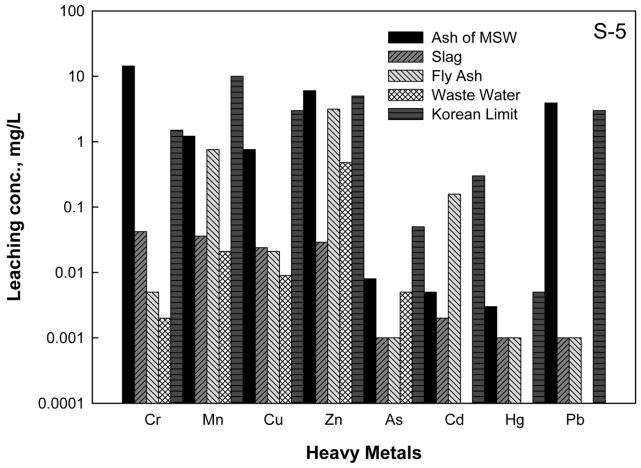

Fig. 5. Leaching concentration of heavy metals in different streams during the gasification using double inverse diffusion flame (DIDF) burner (MSW sample S-5).

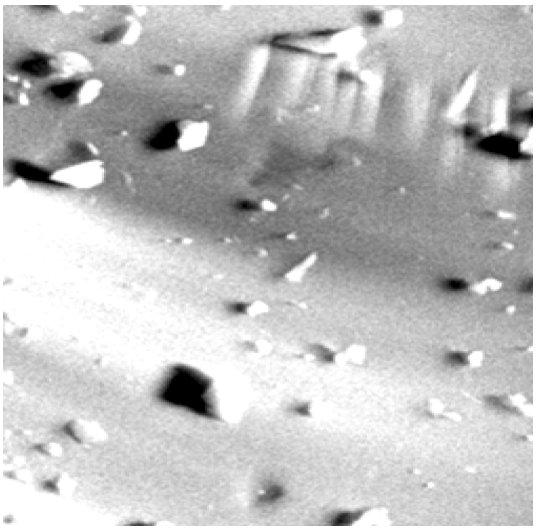

Fig. 6. Scanning electron microscopy (SEM) micrograph of melted slag (1,500× magnification) of MSW sample S-3.

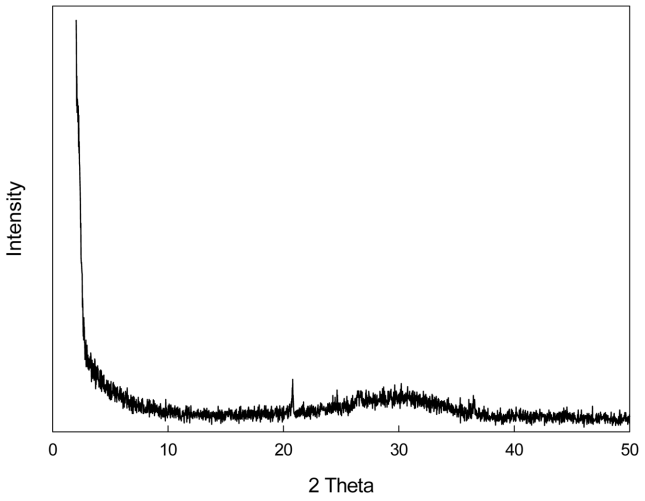

Fig. 7. X-ray diffractometry (XRD) spectra of melted slag MSW sample S-3.

2005].

The slag obtained from the gasification/melting process was of very dark brown color which might be due to the higher silica content (37%) [Li et al., 2003] and had a glassy surface. The glassy and amorphous nature of melted slag was further confirmed from SEM micrographs and XRD spectra of slag shown in Fig. 6 and Fig. 7, respectively. This confirms the slag's environmental friendliness and possibility of its reuse. The homogenized molten mineral material is like natural stone and may have the potential to replace sand and gravel-like granulates as a raw material for concrete or cement [Lin et al., 2004]. Its suitability as raw material for making filling materials, elements and pre-forms would be studied in our future work.

CONCLUSIONS

The MSW from three cities in Korea was gasified at ~1,200 °C in a gasification/melting pilot plant (3 ton/day) based on the Thermoselect process by using a double inverse diffusion flame burner. The produced synthesis gas contained 25-34% CO and 28-38% of H_2. The high heating value of syngas varies from 10.88-14.65 MJ/Nm^3. Moreover, the process was completely environment friendly as the concentrations of toxic heavy metals in melted slag, fly ash, and treated water and other pollutants in exhaust gas were found to be much below the Korean regulatory limits. During melting of slag in homogenization reactor at 1,600 °C, the mineral components of the MSW converted into inert and non-toxic primary materials in the form of vitrified mineral aggregate that can be used in cement and construction industry.

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