Municipal solid waste disposal by using metallurgical technologies and equipments

Jiuju Cai, Wenqiang Sun

State Environmental Protection Key Laboratory of Eco-industry, Institute of Thermal and Environmental Engineering, Northeastern University, Shenyang 110819, P. R. China.

Abstract
Pyrolysis of municipal solid waste can take full advantage of energy and resource and avoid producing hazardous material during this period. In combination with mature metallurgical technologies of coking by coke oven, regenerative flame furnace technology and melting by electric arc furnace, technologies of regenerative fixed bed pyrolysis technology for household waste, co-coking technology for waste plastic and blend coal, and incineration ash melting technology by electric arc technology for medical waste were respectively developed to improve current unsatisfied sorting status of waste. The investigation results of laboratory experiments, semi-industrial experiments and industrial experiments as well as their economic benefits and environmental benefits for related technologies were separately presented.

Keywords: Municipal solid waste; Household waste; Waste plastic; Medical waste; Metallurgical technology.

1. Introduction
Municipal solid waste (MSW) includes partial household waste (HW), waste plastic (WP), and medical waste (MW) which has complex components. With the rapid development of economy and raised people's standard of living, the quantity and lower heating value (LHV) of MSW has been rising year by year. It is shown that the LHV of HW in big cities such as Beijing and Shanghai, China has approached 6,200 kJ/kg, that the LHV of MW has already climbed to 10,000 kJ/kg due to packs of textiles, pledgets, mutilated bodies, etc., [1] and that WP has a higher average LHV of about 40,000 kJ/kg [2]. While the LHV of MSW is 2,980 kJ/kg in Delhi [3, 4], 18,800 kJ/kg in Mauritius [5], and 4,510∼12,662 kJ/kg in Havana city [6]. Many researchers have paid their attention to disposing MSW with high LHV. Wilson has developed a life cycle inventory model to compare the environmental burdens of different scenarios [7]. Ngnikam reported the emission of greenhouse gases resulting from MSW treatment. He pointed out that recuperation of methane is the major solution for reducing greenhouse gas which is supposed to reduce 1.7 ton of carbon dioxide equivalent per ton of MSW [8]. In addition, Chang evaluated the emission characteristics and removal efficiencies of heavy metals and represented that the removal efficiency of heavy metals could reach 95% or higher (except for Hg) for MSW incinerator [9]. There is no doubt that direct burning is the most convenient disposal method for pretreated MSW, since it has a sound volume-reducing effect of MSW and a complete hazardous and noxious substances eliminating effect. However, for untreated MSW, direct burning is not as reasonable as this attributable to the complex component, unsteady LHV, and incomplete combustion. Direct burning of untreated MSW has

a low thermal efficiency and releases secondary pollutants difficult to be solved in the incineration process.

In iron and steel industry, there are several mature high temperature metallurgical technologies, equipments, and well-found follow-up treatment systems, including coke oven, blast furnace and heating furnace, followed by coke oven gas (COG) treatment system, blast furnace gas purification system and waste heat recovery system of heating furnace flue gas, respectively. These technologies and equipments have function of both manufacturing steel products and absorbing bulks of wastes. By using advanced metallurgical crafts and post-treating systems, this paper summarizes several thermo-chemical MSW treatment technologies, i.e., regenerative fixed bed pyrolysis (RFBP) technology for HW originating from the combination of regenerative flame furnace, co-coking technology for WP and blend coal (BC) derived from coking process with byproducts recovery technology, and incineration ash melting technology by electrical arc furnace (EAF-IAM) for MW coming from flame furnace and EAF steelmaking. All the three technologies and equipments involved are respectively from steel rolling, coking, and steel making process in metallurgical industry, and they together constitute an integral and systematic MSW disposal technology related to metallurgical industry.

2. RFBP technology for HW
2.1 Flowsheet and technical features
The schematic diagram of RFBP technology for HW is shown in Figure 1. The pyrolysis chamber and combustion chamber are barrier-wall type, being arranged in turn. The pyrolysis chamber is similar to carbonization chamber of coke oven, and its width is an important structural parameter the pyrolysis device. Pretreated HW was fed into pyrolysis chamber, where the pyrolysis process was finished under conditions of closing and high temperature, accompanied by pyrolysis gas, tar, char and clinker. Partial pyrolysis gas was sent into combustion chamber after being cooled and cleaned to supply energy for HW's pyrolysis, and the other part was provided as town gas. High temperature air combustion (HTAC) technology was applied to recover waste heat of flue gas at the maximum limit, and then the temperature of preheated air and flue gas was controlled at about 1000 and 200ºC, respectively. Tar after extraction could be served as material for fuel oil or other industrial chemicals. After the pyrolysis process, solid products — char and clinker — left in the combustion chamber were pushed out of the pyrolysis chamber, cooled, sorted and reused.
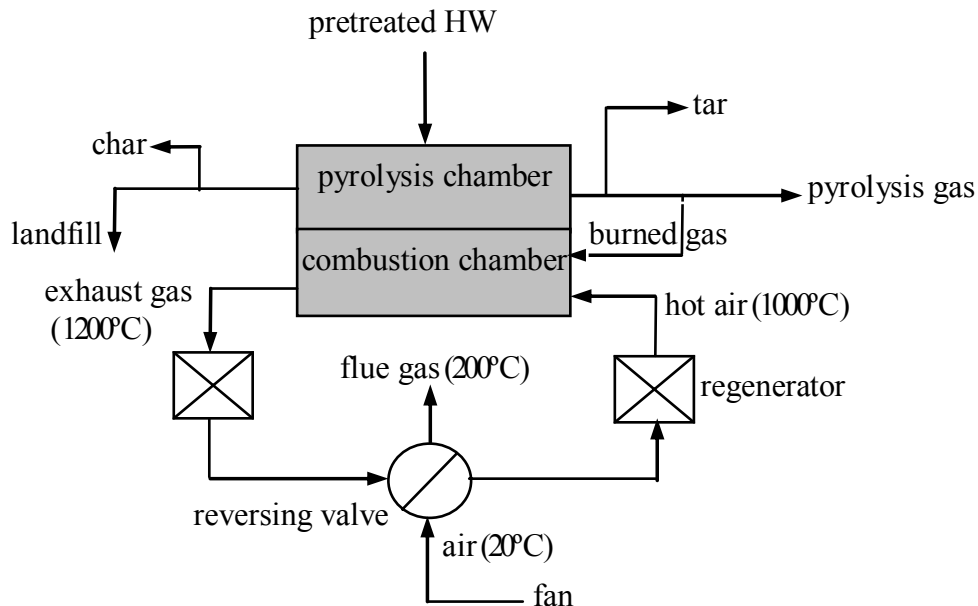

Figure 1. Regenerative fixed bed of HW pyrolysis system

The RFBP technology has three main features: (1) It is suitable for non-sorted HW with unsteady LHV and has strong adaptability to HW with complex and fluctuant components. (2) The combustion chamber operates on the principle of HTAC so that the low calorific pyrolysis gas may combust at ultra-high

temperature, the waste heat of flue gas can be recycled to the utmost, and the conversion efficiency and thermal efficiency of HW pyrolysis process are both improved. (3) In metallurgical industry, many medium- and small-sized coke ovens are to be disused which, however, still have well-found profiles, constructions and operating parameters, which can be used for HW disposal after little modification. And the above practice can make the best use of obsolete metallurgical equipments and reduce investment in HW disposal equipments as well.

2.2 Experimental & simulation

The advantages of RFBP technology can be evaluated by pyrolysis characteristic and production indices, such as pyrolysis time, products yield and quality. The pyrolysis characteristic is related to structural parameters, operating parameters and packing state of HW in the fixed bed. The structure and permeability of fixed bed are of prime importance when apparatus' configuration and process parameters are given. Thus, experimental and simulation studies on pyrolysis characteristics of HW are conducted at three levels: particle, fixed bed, and pyrolysis equipment.

2.2.1 Pyrolysis characteristic of bio-particle

Because pyrolysis of bio-particle is a complex physic-chemical phenomenon involving heat transfer, mass transfer, fugitive constituent flow, reaction kinetics, and structural transformation, this paper uses numerical simulation to discuss the pyrolysis characteristic of bio-particle based on theory of porous medium's heat and mass transfer, according to which particle size has much effect on pyrolysis characteristic. The physical parameters of bio-particle in the simulation are chosen from Di Blasi [10]. To discuss the influence of particle size, the pyrolysis temperature is set as 1300 K. The influence of bio-particle size on pyrolysis time and products distribution was simulated, as shown in Figure 2 [11]. Figure 2a shows that pyrolysis time increases in a parabola form when bio-particle size multiplies which reflects, in the reaction system, particle size has to be managed within an allowed range for smoothly running of the process. With the rising of bio-particle size, the gaseous yield decreases, and tar and char yield increases, according to Figure 2b.
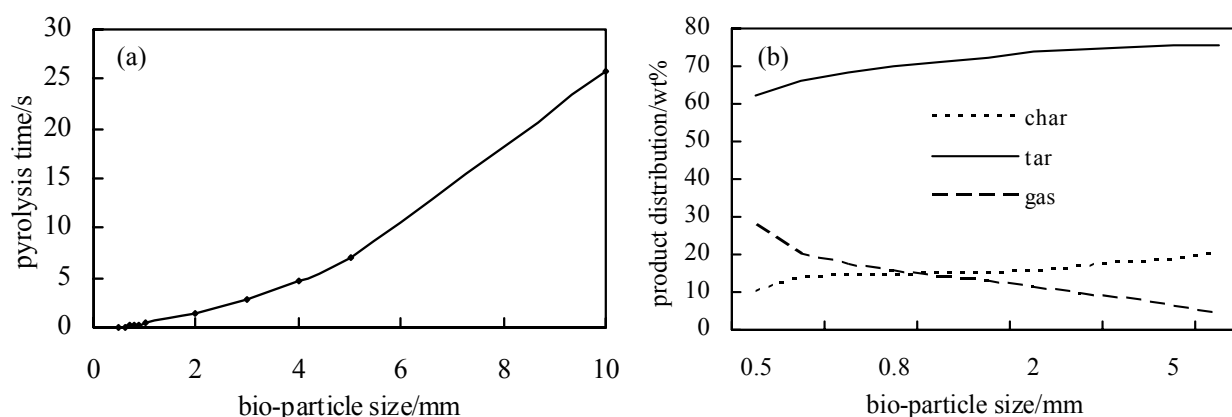

Figure 2. Regenerative fixed bed of HW pyrolysis system. (a) Pyrolysis time vs. bio-particle size; (b) Product yield vs. bio-particle size

2.2.2 Pyrolysis characteristic of reaction bed

Due to the large quantity of water in HW, the heat efficiency of pyrolysis process is affected. As a countermeasure, blending coal is adopted instead of mechanical dewatering because of its advantages [12]. HW in the experiment consists of 30% wastepaper, 15% plastic, 15% wood block and 40% kitchen waste. The apparent density of coal is 1.25 kg/cm^3 which far outweigh that of HW, 0.2 kg/cm^3. The coal particles will be dispersed in the spaces of HW once they are mixed with HW. To study the influence of amount of mixed coal on pyrolysis performance, the mixture height is 300 mm in our experiment, whether the amounts of mixed coal are 100 g or 200 g. Partial experimental results were shown in Figure 3 and Table 1 [13]. Blending coal may change the HW reaction bed performance in reactor. The porosity of bed increases with rising amount of BC, which may increase heating transfer speed, gas yield and decrease pyrolysis time, tar yield. The gas's LHV is also increased.
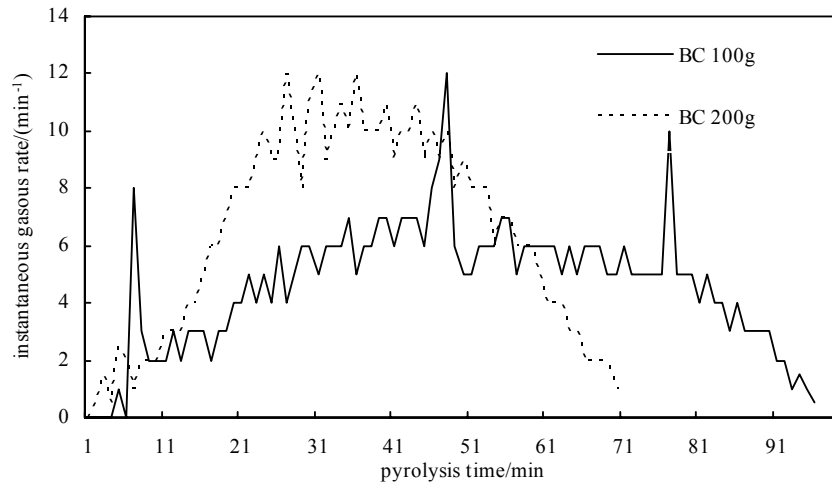
Figure 3. Instantaneous gaseous rate profile with heating power of 28.8 kW

Table 1. Component and characteristic of pyrolysis gas

gas component [vol%]	BC 100 g		BC 200 g	
	19.6 kW	28.8 kW	19.6 kW	28.8 kW
CO_2	21	19	16	15
C_nH_m	8	5	7	3
O_2	3	3	2	2
C_O	14	19	13	14
CH_4	19	17	20	18
H_2	35	37	44	48
calorific value [kJ/m^3]	18255	17372	19830	17836

2.2.3 Structure and operating optimization of HW pyrolysis equipment
Given the disposal time in practice, fuels are not yet effective during vacant combustion period, which causes the decrease of disposal intensity of single chamber and the increase of specific heat consumption. So, different widths have different influences on pyrolysis indices. HW in the experiment is also comprised of 30% wastepaper, 15% plastic, 15% wood block and 40% kitchen waste. The relationship between pyrolysis cycle and heating intensity of the HW pyrolysis equipment was shown in Figure 4a [11]. It can be seen that pyrolysis cycle decreases distinctly with higher heating intensity under given conditions. However, the principle only works within a special range. The relationship of disposal intensity of single chamber and pyrolysis chamber's width was given in Figure 4b. It is shown that different heating intensities correspond to different optimum widths of pyrolysis chamber. Optimum widths are 0.3, 0.4 and 0.5 m when heating intensities are 9.8, 12.6 and 14 kW/m^2, respectively.
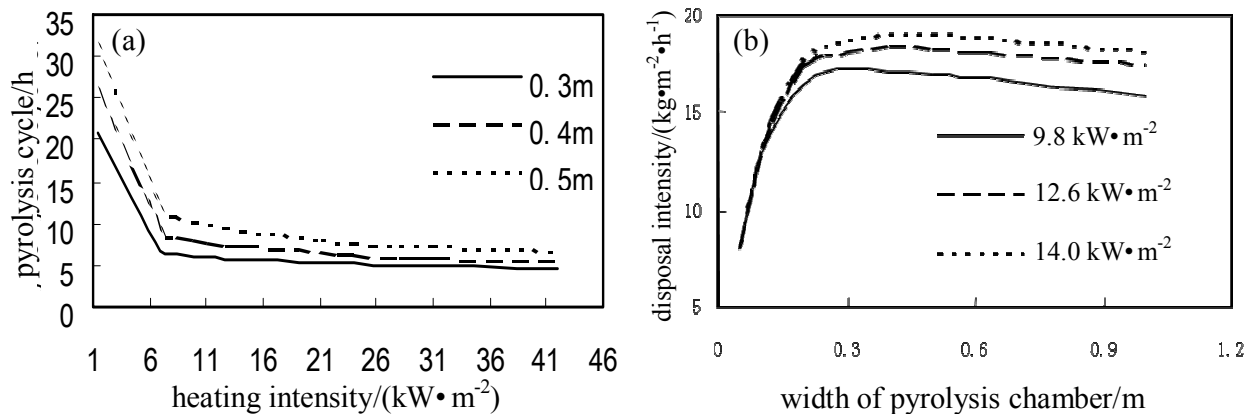

Figure 4. Influence of structural and operating parameters. (a) Pyrolysis cycle vs. heating intensity; (b) Disposal intensity of single chamber vs. width of pyrolysis chamber

3. Co-coking technology for WP and BC

3.1 Flowsheet and technical features

Co-coking technology for WP and BC assumes three steps. Firstly, WP was crushed to a given granularity (hereafter WP1). Secondly, WP1 was uniformly mixed and bonded with coal. Then, the mixture was coated and compressed into plastics-formed-coal (hereafter WP2). Finally, WP2 was fed into coke oven together with BC; and the resulting coke, tar and COG could be disposed and recycled through conventional coking process directly.

Co-coking technology for WP and BC shows four features as following: (1) WP1 is pre-melted in the coal to briquette WP2, which solves both the technical problem of segregation in the mixing of WP and BC and the tickler of dust raising in conventional technology of preheating coal. (2) Additional equipments are just equipments for sorting, crushing, mixing and forming which enormously reducing original investment and operation expense with simple craftwork and short construction cycle. (3) The solid residue of WP charring, coke, can be severed as blast furnace fuel saving the resource and stock cost of coking coal. And the resulting liquid product can be used as liquid fuel and chemical material that yields a high added value, the resulting gas as high LHV fuel. (4) The developed environmentally beneficial win-win technology — new disposal technology of WP — opens a new approach to disposing white pollution through coking process.

3.2 Experimental

The experimental materials were comprised of WP from HW of Shijingshan District, Beijing, and BC from Shougang Group, China. The BC, WP1, and WP2 were crushed into 90-mesh by a universal crusher to be used as experimental sample. And its proximate analysis and ultimate analysis are listed in Table 2. Experimental equipments like thermo gravimetric (TG) analyzer , Fourier transform infra-red (FTIR) spectroscopy, 2 kg coke oven, and 200 kg coke oven as well as No. 4 coke oven in Coke Plant of Shougang Group, China [14] were employed in the work. Systemic experiment was conducted on key issues of co-coking technology for WP and BC, range from charge ratio, mixing mode, pyrolysis regime, to industrial application.

Table 2. The proximate and ultimate analysis of samples

sample	proximate analysis [wt%]			ultimate analysis [wt%]				
	V	A	FC	C	H	O	N	S
BC	24.11	9.69	66.20	83.69	5.43	8.68	1.32	0.89
WP	96.11	3.84	0.05	84.62	14.12	1.09	0.12	0.05

3.2.1 TG experiment

Thermo gravimetric and differential thermo gravimetric (TG/DTG) curves of sample of 2 wt% WP1 and 98 wt% BC were plotted in Figure 5 [15]. It is found that there is an overlapped decomposition temperature region during the pyrolysis of single WP1 and BC where synergism occurs because different radicals from BC and WP interact with each other. Additionally, the synergism of co-coking of WP1 and BC can increase oil and reducing water, which may be caused by hydrogen's combining with radical to produce oil instead of combining with oxygen to form water.

It is defined that $(dv/dt)_{max}$ is the maximum release rate of fugitive constituent, t_{max} is the corresponding temperature of $(dv/dt)_{max}$, $\Delta t_{1/2}$ is the half-peak width, α is the coefficient of temperature region, V and V_{calc} are the actual and calculated percentage of liberated fugitive constituent, and E, E_{calc} are the actual and calculated average activation energy, respectively. According to different pyrolysis synergism indices (σ) of co-pyrolysis can be calculated, depending on the percentages of WP1, WP2, and BC, as listed in Table 3 [15].

$$\sigma = \frac{(dv/dt)_{max}}{t_{max} \cdot \Delta t_{1/2}} \cdot \sum_{i=1}^{n}\left(\alpha_i \cdot \left| \frac{E}{V_i} - \frac{E_{calc}}{V_{i,calc}} \right| \right) \tag{1}$$

Table 3 indicates that maximum pyrolysis synergism index σ under different WP1 which changed by 1 wt% more WP1. The experiment presented negative relationship of WP1 and synergism while σ increases with the increasing percentage of WP2; so was yield synergism β_T which is defined as the rate

of real yield of one product to the sum of separate yield of the product; it reflects the effect of synergism of co-coking of WP and BC on the yield of pyrolysis product. The reason for difference of pyrolysis behavior between WP1 and WP2 is probably that the pretreated WP is uniformly melt and attached to the surface of coal or filled in the pore of coal granule, which can change the physical and chemical function deriving from action of WP's pyrolysis radicals and BC's pyrolysis.
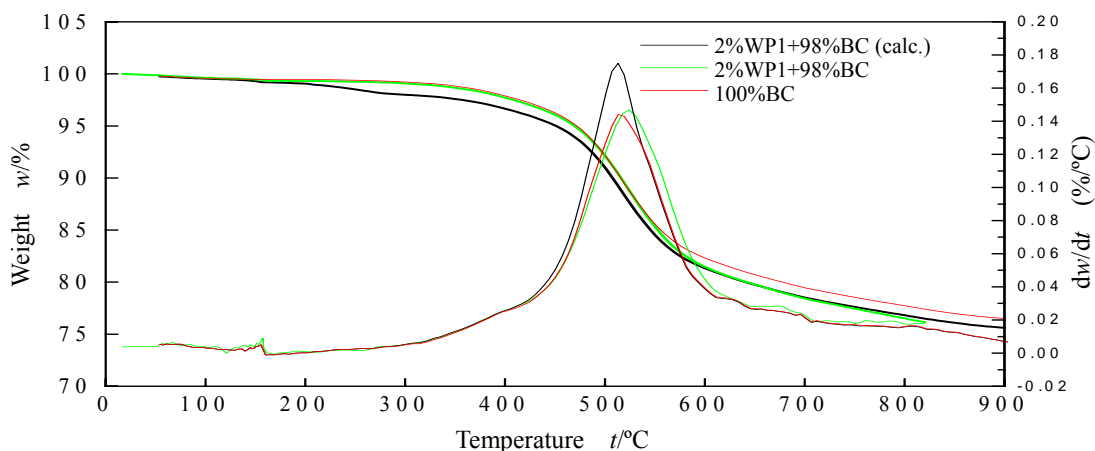

Figure 5. TG/DTG curves of sample of 2 wt% WP1 and 98 wt% BC

Table 3. The pyrolysis characteristics of sample

Project	1%WP1+ 99%BC	2%WP1+ 98%BC	3%WP1+ 97%BC	4%WP1+ 96%BC	1%WP2+ 99%BC	2%WP2+ 98%BC	3%WP2+ 97%BC	4%WP2+ 96%BC
σ	86.824	72.3000	60.242	31.681	70.857	102.279	109.140	114.889
β_T	2.380	1.085	1.024	0.202	3.569	8.255	10.886	12.741

3.2.2 FTIR Experiment

FTIR absorbance curves of chlorine release as HCl (2500 ∼ 3100 cm^{-1}) during 2 wt% WP1 and 98 wt% BC pyrolysis at different temperature were depicted in Figure 6 [15]. The theoretical curve in Figure 6 is a curve for single pyrolysis of containing WP.

It can be seen from Figure 6 that HCl release temperature rises to 320 °C from the theoretical temperature 240 °C, that the maximum release value occurs at 350 °C, and that the end temperature is 420 °C for chlorine release. The maximum release temperature for HCl is about 13 °C later than the corresponding temperature of the first zero-gravity peak, when the released HCl accounts for 22.57% of the total; while it accounts for 91.04% at 400 °C. About 74% of HCl is released during the range of 340 to 400 °C. The findings show that different radicals coming from WP1 and BC pyrolysis interact with each other distinctly which cause massive release of HCl within a small temperature region. Comparing the theoretical and actual release amount, it can be found that the 85.04% of theoretical release value is released for 2 wt% WP1 and 98 wt% BC pyrolysis. This indicates that, for 2 wt% WP1 and 98 wt% BC pyrolysis, 14.96% of HCl is not released, i.e., 14.55% of Cl is remained in the solid pyrolysis products. The remaining chlorine in coke will not erode blast furnace facilities because its existing approaches are: (1) to form compound with metal ions, existing in the ash of coke; (2) to form stable high molecular polymer by replacing chloride ion with hydroxyl, existing in the coke.

3.2.3 Pilot experiment

The influences of co-pyrolysis of WP2 and BC upon the main industrial indices — abrasion resistance index (M10), anti-broken strength (M40), coke reactivity index (CRI), and coke strength after reaction (CSR) — were illustrated in Figure 7 [15]. Results show that co-coking of WP2 and BC makes coke and water yield lower, tar and gas yield higher, compared with coking of single BC. When co-coked with BC, the level of WP2 can rise to 4 wt% without obvious negative influence on its co-coking; the quality of coke can be improved without using any other organic additive as if the percentage of WP2 addition is limited lower than 2 wt%, which have reversed current status in the world that higher percentage of WP

co-coking with BC cannot co-exist with longer coke strength. Additionally, the tar from co-coking of WP2 and BC has a palpable methylation; the aromatic hydrocarbon and alkane in the tar present a distinct feature of lightweight, and this is beneficial to upgrade tar. The COG's components derived from co-coking of WP2 and BC was optimized, light combustible components increased, LHV increased which then was translated into higher economic benefit.
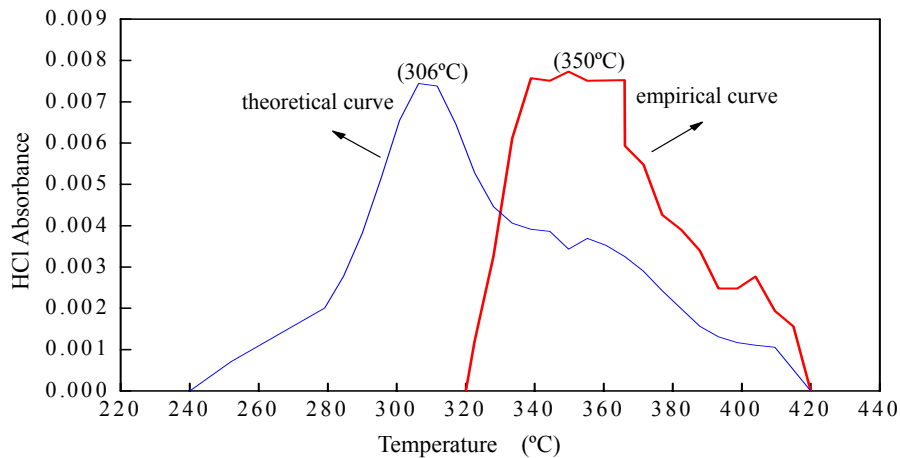

Figure 6. FTIR absorbance curves of chlorine release as HCl for 2 wt% WP1 and 98 wt% BC pyrolysis

3.2.4 Industrial experiment
The results of Industrial experiment employing No. 4 coke oven in Coke Plant of Shougang Group, China indicate that the industrial application of WP disposal technology by coking craft has no impact on the normal operation of coke plant. Compared with single coking of BC, coke from co-coking of 1 wt% WP2 and 99 wt% BC causes an insignificant change in M40, a M10 reduction of 1.6, a CRI reduction of 10.0, and a CSR rise of 18.0 and the quality of coke is improved consequently. In addition, the new technology not only increases coal capacity per carbonization chamber by 3% but also shortens coking time by 17 minutes, and hence it may improve productivity of coke oven and cut coking duration. It is estimated that 164 US$ or more will be risen per ton WP with the technology. Therefore, the plastic-formed technology has notable prospect for application in disposing WP.
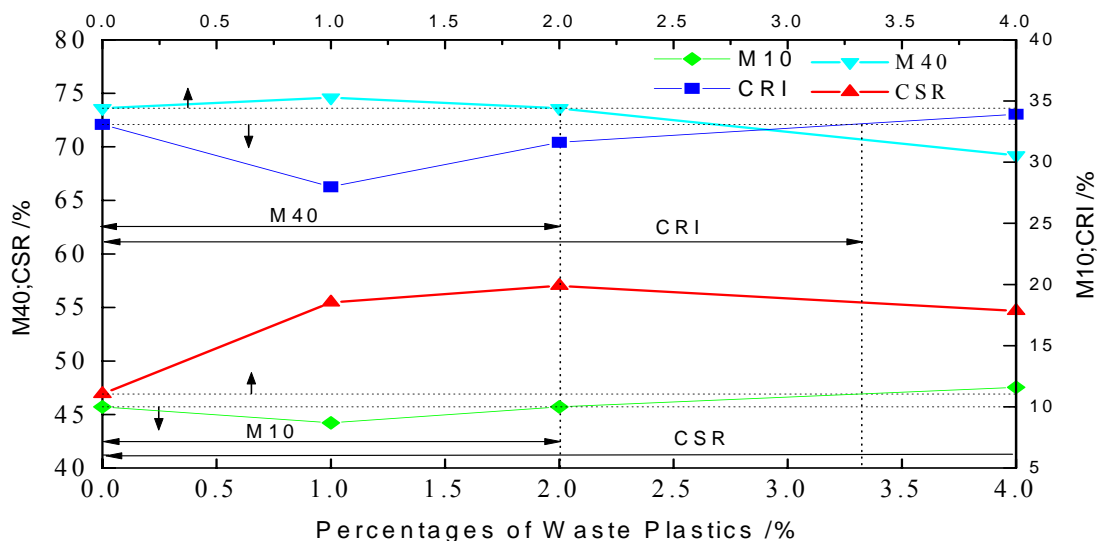

Figure 7. Effect of WP2 additions on quality of the coke from co-coking

4. EAF-IAM technology for MW
4.1 Flowsheet and technical features
For EAF-IAM, the pyrolysis and gasification of MW were finished in primary combustion chamber with anoxic and moderate temperature. The resulting gas, together with auxiliary fuels, was burned in the

secondary combustion chamber whose temperature reaches 1100ºC or higher. High temperature exhaust gas went into waste heat boiler to produce steam for heating. The air pollution control equipments are quench tower, device for injecting activated carbon, semi-dry deacid system, and bag-type dust collector in turn. After mixed, the resulting fly ash (FA) and bottom ash (BA) then moved into an EAF, where they were melt into liquid phase under high temperature resulting from graphite electrode. Molten metals and vitreous slag could be separated and recycled at the outlet.

Recently, MW pyrolysis and incineration technology and air pollution control technology have been relatively mature. But the incineration ash disposal technology is still immature and it becomes the key to the development of technology of MW pyrolysis and incineration. The features of EAF-IAM are: (1) The electric arc is encircled by clinkers and has a higher energy density, a less radiation heat loss, a less flue gas emission and hence a higher thermal efficiency. (2) The technology has a low requirement of feedstock, which needs only crush and dryness without requirement of additives. (3) The technology can decompose organic poisons like dioxin and solidify heavy metals in molten slag. (4) The molten slag and metals flow out stratified so that metals may be recycled; whilst the vitreous slag may be widely used in building materials with high-added value, such as glass ceramic, refractory, and soil cement.

4.2 Experimental

Thermal processing including vitrification of incineration ash gains some advantages over other treatment options, namely, it leads to a volume reduction, a separation or stabilization of heavy metals, a destruction of dioxins/furan as well as material recovery and reuse. Considering the negative effects of other treatments such as solidification of the bag fly ash using as cement, the melting treatment of incineration ash using small-sized metallurgical electric arc furnace about to be disused was studied in the paper. By employing X-ray fluorescence (XRF), X-ray diffraction (XRD), scanning electron microscopy (SEM) and atomic absorption spectrometry (AAS), traits of incineration ash of MW such as composition, phase, micro-geometry, heavy metals strength and its leachate behavior were analyzed. Also, a 2 kg EAF was employed in the experiment.

4.2.1 Chemical composition of incineration ash of MW

Main chemical components of FA, BA and slag after they mixed and melted were listed in Table 4. It can be drawn that the content of SiO_2 in BA reaches 40.90%, higher than that in FA. Both BA and FA are heavily charged with CaO. The content of SO_3 and Cl in FA accounts for 12.10% and 12.31%, respectively. And there are a lot of SiO_2, CaO, and Al_2O_3 in slag which hold round 75% in total.

Table 4. Chemical components of ash and slag [wt%]

sample	SiO_2	CaO	Na_2O	Al_2O_3	Fe_2O_3	MgO	K_2O	TiO_2	P_2O_5	SO_3	Cl	F
FA	20.60	15.30	11.00	6.54	6.73	3.43	2.97	1.50	0.94	12.10	12.31	1.31
BA	40.90	22.10	5.30	9.75	3.11	3.15	0.97	1.12	1.16	8.53	2.06	0.18
slag	45.40	19.10	6.82	10.89	7.80	2.33	1.02	1.29	0.486	0.335	1.01	0.6

4.2.2 Phase and micro-geometry of incineration ash of MW

The XRD analysis results of FA and BA were given in Figures 8a and 8b, respectively. FA abounds in $CaSO_4$ and NaCl crystal compounds; whilst main phases of BA are complex silicate, such as Ca_2SiO_4, Al_2SiO_5, $CaMgSi_2O_6$, and a small quantity of $CaCO_3$. The XRD and SEM analysis results of slag were illustrated in Figures 8c and 8d, from which it can be seen the palpable glass phase of slag.

4.2.3 Heavy metals strength and leach concentration

Leachate test results of heavy metals in incineration ash and slag were listed in Table 5. The contents of volatile metals such as Zn, Pb, and Cd in FA are more than those in BA, and the leach concentrations of all heavy metals in FA are higher than those in BA. Leach concentrations of Pb, Cd and Zn in FA far exceed the hazardous waste landfill regulatory thresholds. However, the obtained molten slag is a single-phase amorphous, glassy product, so heavy metals in slag are combined into the Si-O network by the melting process and hence the molten slag contributes very little to leachate concentration and meets environmental requirement.
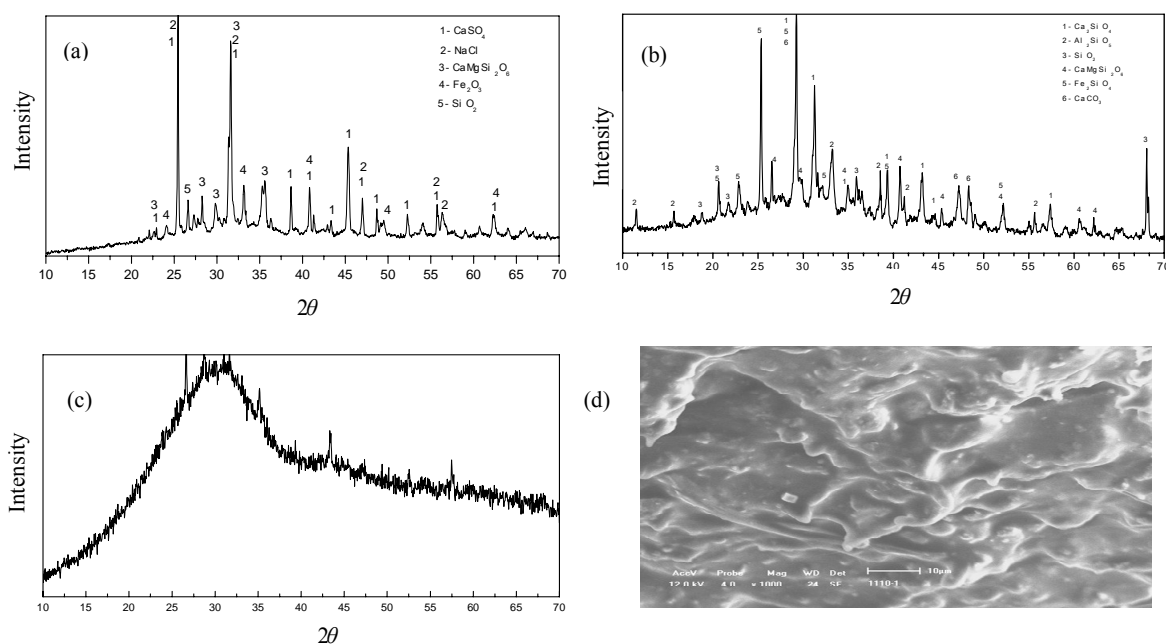
Figure 8. XRD and SEM analysis. (a) X-Ray diffractogram of FA; (b) X-Ray diffractogram of BA; (c) X-Ray diffractogram of slag; (d) SEM photographs of slag

Table 5. Heavy metals strength and leach concentration of ash and slag

samplecontent		Cu	Pb	Zn	Cd	Cr	Ni	Hg	As
FA	total strength [mg/kg]	898.5	1147.1	10761.2	47.1	110.3	1427.5	7.7	18.4
	leach concentration [mg/L]	1.06	6.50	377.93	2.96	0.09	1.11	-	0.01
BA	total strength [mg/kg]	788.7	40.0	2788.3	14.9	174.7	1176.5	0.6	2.8
	leach concentration [mg/L]	0.02	0.03	0.20	0.02	0.07	0.04	-	-
slag	total strength [mg/kg]	494.7	73.6	8611	12.9	124.8	318.7	-	-
	leach concentration [mg/L]	0.01	0.02	0.04	0.004	0.01	-	-	-
standard for appraisal of leach toxicity [mg/L]		50	3	50	0.3	10	10	0.05	1.5
the hazardous waste landfill regulatory threshold [mg/L]		75	5	75	0.5	12	15	0.25	1.5

5. Conclusion
The paper introduces a new system, RFBP technology for HW, by combining out-heating pyrolysis technology with regenerative combustion technology. On the background of this system, the basic researches are involved with three aspects: particle, reactive bed and pyrolysis device. The results show that the particle size, the amount of mixed coal, and the width of pyrolysis chamber are main influencing factors on pyrolysis characteristic of HW.

In this paper, through introducing experiments of thermal balance, TG-FTIR, 2 kg and 200 kg experimental coke ovens, and No. 4 commercially industrial coke oven of Shougang Group, the Co-coking technology for WP and BC was investigated from fundamental theory to industrial application. The new technology has been proved not only economically and environmentally friendly but also has notably industrial application to deal with 'white pollution'.

EAF-IAM technology for MW is also introduced in this paper. The characteristics of three samples from different positions in the incinerator were investigated with XRF, XRD, SEM and AAS for their chemical composition, mineralogy, morphology, metal contents and leaching behavior of heavy metals, aiming at finding out a safe and efficient disposal method. The results indicate that BA and FA are mainly consisted of silicates; they have low leachability and could be classified as non-hazardous wastes.

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Jiuju Cai is Professor in the Institute of Thermal and Environmental Engineering, Northeastern University, China. He is also the Director of State Environmental Protection Key Laboratory of Eco-industry, and the Director of Energy and Thermal Committee of CSM. He received his Ph.D. Degree on Thermal Power Engineering from Northeastern University (1986). His research focuses on systems energy conservation, industrial emissions reduction and eco-industry. Professor Cai has more than 200 scientific papers published in the above mentioned related fields.

Wenqiang Sun reveived his BS Degree (2007) in Thermal Energy and Power Engineering and MS Degree (2009) in Thermal Power Engineering from Northeastern University, China. His work covers topics in systems energy conservation for process industry and energy flow network for metallurgical industry.
E-mail address: neu20031542@163.com