



The regeneration of waste foundry sand and residue stabilization using coal refuse

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ABSTRACT

The processes for recycling waste foundry sand are divided between regeneration and beneficial reuse, and the potential for regeneration is higher than that of reuse. In this study, two processes for the recycling and residue stabilization of waste foundry sands were considered. One is the dry mechanical process for recycling, and the other is the stabilization process for powdered residue. The dry mechanical process of regeneration consists of crushing, grinding, separation, and classification. To stabilize the residues that were generated through the regeneration process, powdered residues were pelletized by a high-shear pelletizer, and the surfaces of the pellets were subsequently coated with coal refuse powders that contained sodium silicate as a binder. Coated pellets were sintered by a self-propagating combustion method. The refractory index of the recycled sands, as measured by the Seger cone method, was over -34 , and their SiO_2 contents of 94% was similar to that of green sand. The general conclusion that coal refuse and sodium silicate stabilize heavy metals better than other processes may lead to the development of a cost-effective solution for stabilizing heavy metals in residues.

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1. Introduction

Foundry sands are commonly used in casting processes by iron foundries to form molds in which molten iron is poured. After cooling, the sand molds are broken, and the finished iron products are removed. Either clay or chemically based binder is added to the sands to maintain the shape of the mold during pouring and cooling. Cast-iron foundries create a large quantity of waste: one quarter to one full ton of solid waste per casting [1], 30–60% of which is made up of core and molding sands. The recovery and reuse of wastes is an alternative that has the lowest environmental impact, resulting in a savings of raw materials and a reduction in manufacturing residues. Waste foundry sands (WFS) can be recycled or reused in several ways, through foundry manufacturing itself or via different industrial processes [2–5].

Many methods have been developed to recover green sands for the production of molds and cores [6–10]. Green sands can be continually reused for molding operations without any significant refinement. The sand is sieved to remove large particles, new sand is added to account for the lost sand, and, finally, the material is remolded for a different metal piece (mulling operation).

There are generally three methods for the recovery of green sand from WFS: thermal reclamation, wet reclamation and dry mechanical reclamation [2,4,5]. The thermal reclamation makes use of heat in a rotary kiln, a plural furnace, or a fluidized bed to combust binder and contaminants over the temperature of 700 °C. The gaseous emissions from the furnace, if not carefully controlled, often make a strong environmental impact. The wet reclamation uses a wet mechanical attrition phase that is performed using water and hydrochloric or sulfuric acid, and then is dried. A wet reclamation is the most effective for the recovery for the recovery of green sands. However, the wet reclamation involves a noticeable sludge, and therefore need an additional cost for the sludge treatment. The dry mechanical reclamation uses a particular attrition phase to remove the oolitic layer surrounding the silica grains and an air separation or a vibration screen to separate fine particles. This process is more economical and effective than the other methods, but produces large quantities of dust [3–6].

Despite the numerous efforts that have been undertaken regarding this question, few methods are economical in terms of efficiency and operational costs; hence, any successful stabilization process should be relatively cheap and sufficiently effective to ensure a sufficient advantage for its commercialization.

Two different treatment processes were used in this work for the regeneration of WFS. The first process is a dry mechanical process that involves comminution using an impact and friction

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Table 1
Chemical composition of green sand and waste foundry sand by XRF, wt%.

Sample	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	K ₂ O	Na ₂ O ₃	TiO ₂	MnO	P ₂ O ₅	L.O.I
Green sand	94.41	3.41	0.58	0.09	0.05	0.45	0.10	0.03	0.01	0.01	0.79
Waste sand											
YH	89.74	4.54	1.32	0.20	0.22	0.72	1.82	0.09	0.04	0.10	1.20
SS	90.88	3.83	2.26	0.16	0.27	0.57	1.00	0.08	0.05	0.10	0.64
DH	87.64	4.90	3.41	0.26	0.30	0.91	1.35	0.10	0.08	0.06	0.97

mechanism, magnetic separation to remove ferrous metals, and eddy current separation (ECS) to remove non-ferrous metals. The second process is a stabilization process for the residue that is generated by the first process. For the stabilization process, a self-propagating combustion method using coal refuse was used.

2. Experimental

2.1. Starting materials

In this study, three kinds of WFS were collected from major casting plants in South Korea. The chemical composition and heavy-metal contents of the samples were analyzed using XRF (XRF-1700, Shimadzu Co., Ltd., Japan) and ICP-MS (OPTIMA 5300DV, Perkin Elmer Co., Ltd., USA), respectively, to compare the WFS to green sand. All analyses were performed on dried samples that were obtained by heating the original samples for 2 h in a dry oven at 105 °C. The chemical compositions of the samples of the WFS and green sand are given in Table 1. As shown in Table 1, the SiO₂ content of the green sand that was used for the casting process in this study was 94%, whereas the SiO₂ contents of the waste foundry sands ranged from 87% to 90%. The major impurities of the WFS were Fe₂O₃ and Na₂O₃.

Coal refuse, which is gangue matter that has been separated from coal during the coal preparation process, was obtained from the Hwasoon coal mine in South Korea and was used for self-propagating combustion. The coal type of the Hwasoon coal mine is anthracite. The fixed-carbon content of the coal refuse was analyzed by proximate analysis on a dry basis and was observed to be less than 15%.

2.2. Experimental

The process of recycling the waste foundry sand consisted of two stages. The first stage involved a recycling process [Fig. 1(a)], whereas the second stage involved the stabilization of residue that was generated during recycling [Fig. 1(b)]. Comminution of the WFS was performed by a jaw crusher and gyratory crusher. A rotor mill was used to grind the WFS. The rotor mill used an impact mechanism through which collision and abrasion were induced by friction between fragments during grinding. We expected the rotor mill to employ effective grinding mechanisms to remove impurities (such as binder and heavy metals) from the surfaces of sand particles. After comminution, a magnetic separator was used to separate ferrous metals from the crushed sands, and an ECS was used to remove non-ferrous metals, such as Al, Mg, Mn, Cu, and Cr. The magnetic separator had two kinds of cross-belts: 3000-G and 6000-G cross-belts.

The ECS that was used in this study consisted of a short conveyor that was driven from the feed end. A rapidly rotating system of permanent magnets, the pole system, which generates high-frequency changing magnetic fields, was incorporated into the head drum. These fields create strong eddy currents in non-ferrous metals, which enhance the metals' opposing magnetic fields. The non-ferrous metal parts jump out of the remaining material flow. The

permanent magnet of the ECS that was used in this study consisted of Nd-Fe-B and pure iron.

After separation, the ground sample was subsequently sieved using a vibration screen with opening sizes of 0.075 mm and 3.35 mm. The particle size fractions within the range of 0.075–3.35 mm were considered to be regenerated green sand. Fine particles that were smaller than 0.075 mm were considered to be powdered residue, which required further stabilization processing because it was expected that most of the heavy metals and binders would be concentrated in the powdered residue.

A bottom-driven, high-shear granulator (HSG) was used to pelletize the powdered residue, and the experimental conditions of the pelletization process were as follows: the flow rate of air was 1 L/min, the rotation speed of the shopper was 1500 rpm, and the rotation speed of the rotor was 1000 rpm.

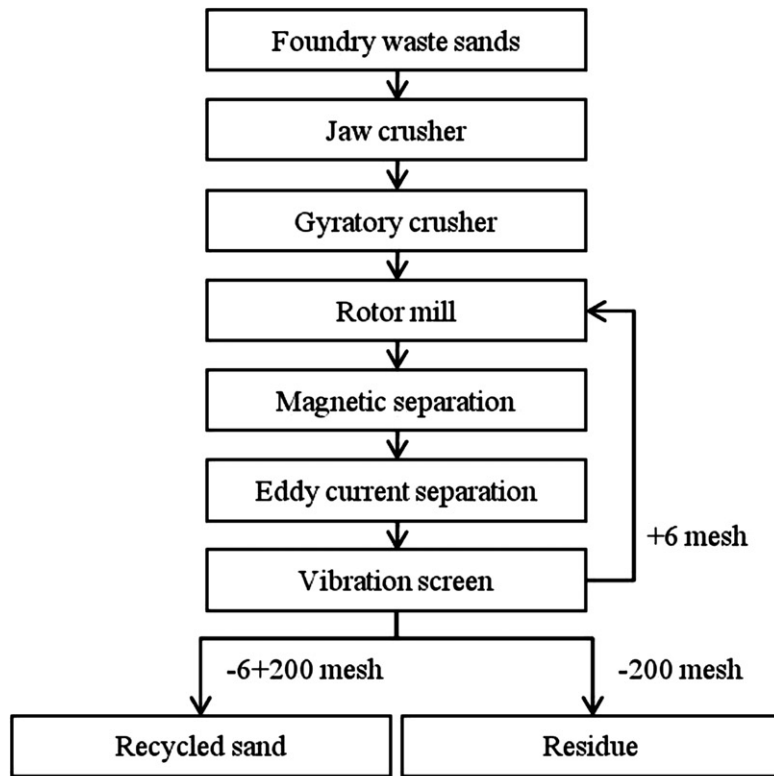
The pellets were coated with a mixture of coal refuse and sodium silicate using the HSG. Sodium silicate was considered to be a binding agent and was used to decrease the sintering temperature of the coated pellets. The coated pellets were sintered using a self-propagating combustion method using fixed carbon in the shell of the pellet. Fig. 2 depicts a schematic diagram of the self-propagating combustor and its method of operation. The self-propagating combustor consisted of a part for feeding green pellets, the vibrator for discharging sintered pellets, a heating coil for the initial ignition, a fan blower for the air supply, and three thermal couples for temperature control. The three thermal couples were placed at the feeding zone of the green pellets, the sintering zone, and the discharging zone, respectively. The heating coil was used to ignite the green pellets that were fed into the self-propagating combustor. The electric power that was supplied for heating was stopped after ignition. The fan blower supplied oxygen for the continuous combustion of the coal-fired pellets. Sintered pellets were discharged into the bottom of the combustor by vibration, while new green pellets were fed into the top of the combustor. The firing of pellets into the sintering zone re-ignited new green pellets such that a continuous process was maintained. The sintering temperature was controlled by the feeding rate of the green pellets, discharging rate of the sintered pellets, and the air flow rate.

The refractoriness index of the regenerated sand was measured via the Korean standard (KS) L-8001 method using Seger kegels (SK) and Seger cones [11]. A toxicity characteristic leaching procedure (TCLP) was used to determine the leaching characteristics of the samples. The TCLP test is the USEPA leaching procedure for determining the characteristics of hazardous waste [12–14].

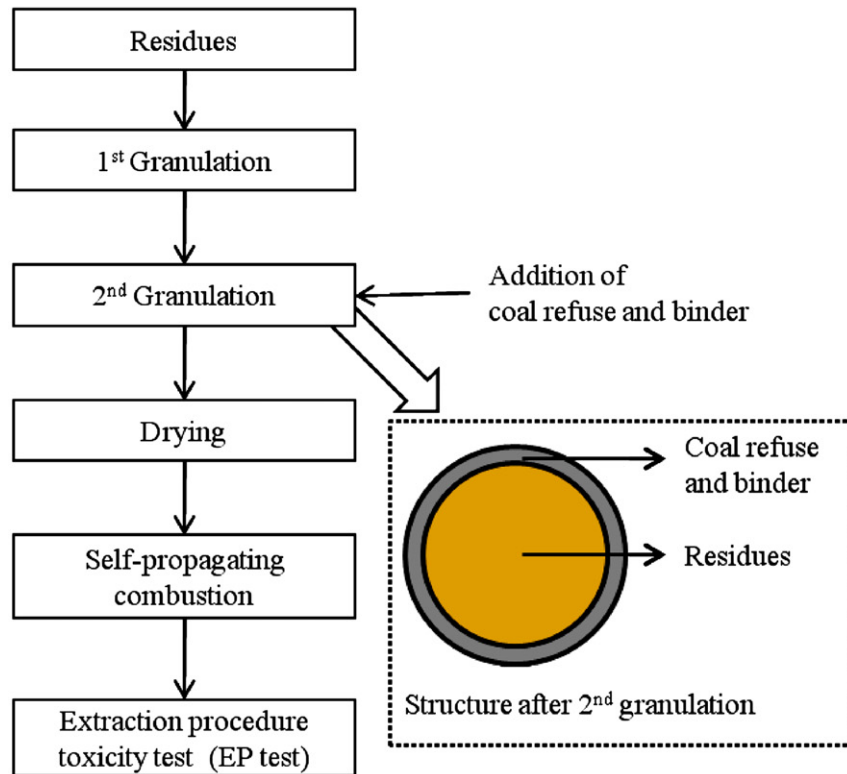
3. Results and discussion

3.1. Comminution

The particle size distribution of the samples of WFS after being crushed by the jaw crusher and the gyratory crusher is depicted in Fig. 3. The weight percent of particles ranging from 0.075 mm to 3.35 mm in size was over 90%. This range is the typical particle size distribution of foundry sand that is used in the casting industry [15]. Ries and Conant also reported that foundry sand is usually observed in sizes that are coarser than 0.25 mm [16]. Thus, the

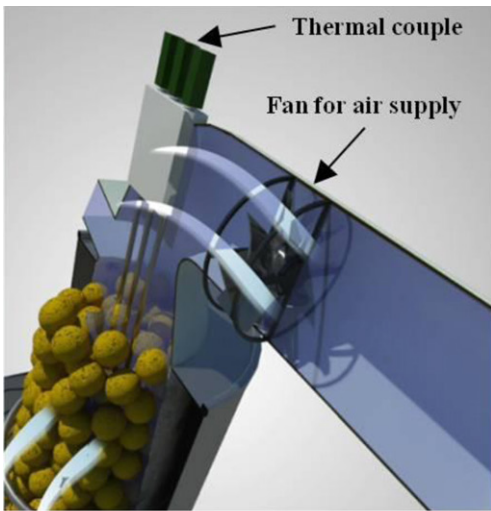


(a)

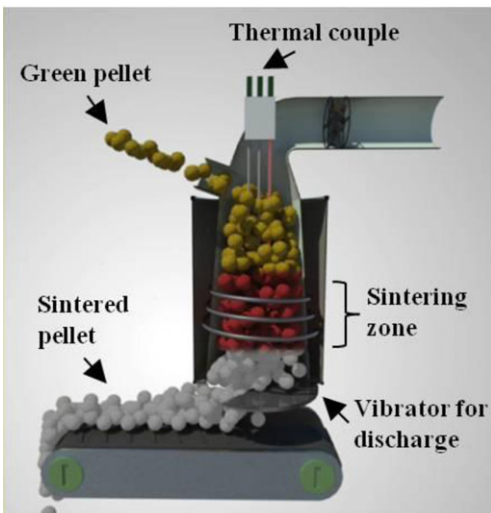


(b)

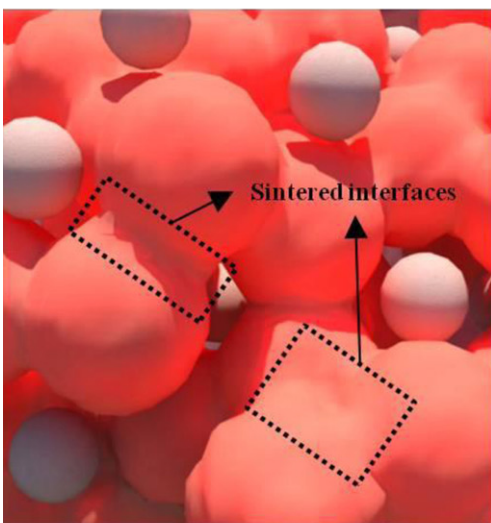
Fig. 1. Flowcharts describing the regeneration of waste foundry sand and residue stabilization. (a) Regeneration process. (b) Stabilization process.



(a)



(b)



(c)

Fig. 2. Schematic diagram of the self-propagating combustor and its method of operation. (a) Schematic diagram. (b) Fan blower for the air supply. (c) Sintering.

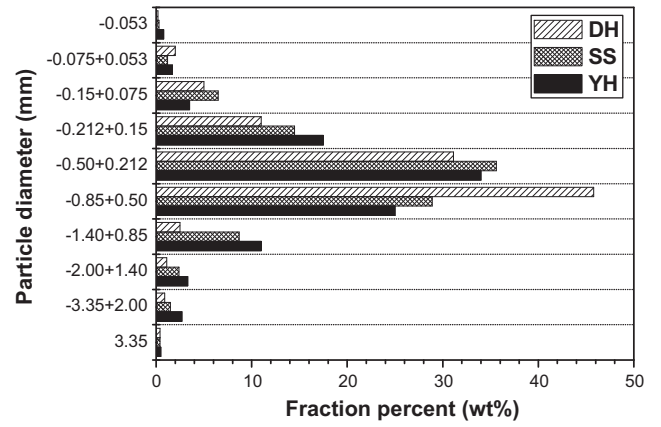


Fig. 3. Particle size distribution of samples crushed by a jaw crusher and a gyratory crusher.

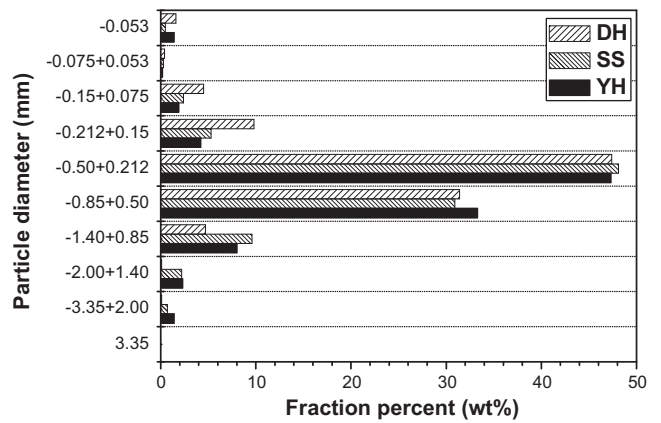


Fig. 4. Particle size distribution of samples ground by a rotor mill.

comminution process in this study was performed to focus the particle size distribution around particle sizes ranging from 0.15 mm to 2.00 mm.

Fig. 4 depicts the variation in the particle size distribution of samples that were ground using a rotor mill. The overall particle size distribution shown in Fig. 4 is finer than that shown in Fig. 3. Also, the weight percent of fine particles that were smaller than 0.15 mm in size increased after grinding but was below 10% compared with all of the samples.

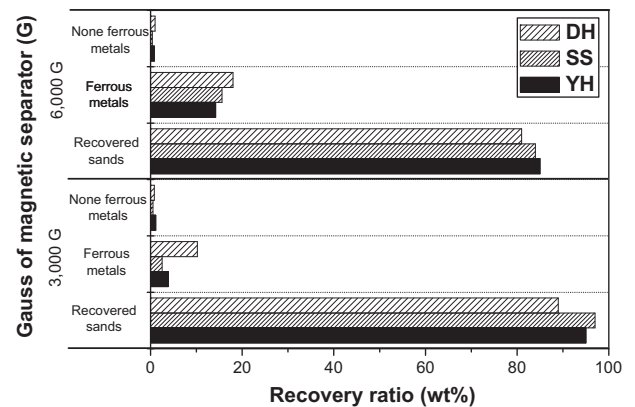


Fig. 5. Comparison of the ratio of recycled sand recovered by magnetic and eddy current separation.

Table 2
Chemical composition of regenerated sands at 3000 G by XRF, wt%.

Sample	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	K ₂ O	Na ₂ O ₃	TiO ₂	MnO	P ₂ O ₅	L.O.I
Green sand	94.41	3.41	0.58	0.09	0.05	0.45	0.10	0.03	0.01	0.01	0.79
Regenerated sand											
YH	94.39	3.32	0.32	0.12	0.08	0.51	0.22	0.08	0.03	0.05	0.81
SS	94.47	3.30	0.34	0.11	0.05	0.51	0.16	0.05	0.03	0.04	0.76
DH	94.34	3.49	0.30	0.10	0.06	0.50	0.33	0.07	0.01	0.02	0.95

3.2. Separation

To remove ferrous metals from the ground WFS, magnetic separation was performed under applied magnetic fields of 3000 G and 6000 G. Next, eddy current separation was performed to remove any non-ferrous metals from the samples. Fig. 5 depicts the difference between the ratio of recovered recycled sand and the ratio of sand removed by magnetic and eddy current separation. The recovery ratio of recycled sand decreased as the magnetic force increased. All of the samples that were processed at 3000 G had recovery ratios that were greater than 90%, and the recovery ratios of all of the samples that were processed at 6000 G were over 80%.

The chemical composition of the sand that was recycled at 3000 G, as determined by XRF, is summarized in Table 2. Most literatures reported that the dry mechanical process for WFS reclamation is ineffective in recovering initial chemical composition of the green sand [2–6]. Moreover, Andrade et al. reported that the SiO₂ of 86.3–88.5% can be recovered by the reclamation method using the blast furnace and acid treatment [4]. However, the results of Table 2 demonstrate that all of the recycled sands had similar chemical compositions in comparison to that of new green sand. The SiO₂ contents of all of the recycled sands were higher than 94%, and, moreover, the recycled sands had almost the same SiO₂ contents in comparison to the green sand. The Fe₂O₃ and Al₂O₃ contents of the recycled sand were lower than those of the green sand. From the results depicted in Table 2, we expect that the recycled sand is commutable to green sand as regeneration sand. Therefore, if the rejected weight percents of both the fine particles (residue) obtained by screening and impurities obtained by separation are considered, the ratio of regeneration will be greater than 80%.

3.3. Refractoriness index

The refractoriness index of foundry sand is an important factor. Regenerated sand has to maintain sufficient refractoriness during casting. In order to be used as foundry sand, the refractoriness index of the regenerated sand should be equivalent to that of an SK of 33. An SK of 33 has a melting temperature of 1730 °C, whereas an SK of 34 melts at 1750 °C. Table 3 summarizes the refractoriness index of the regenerated sand as measured by the KS L-8001 method. The refractoriness index of all of the WFS before regeneration were below that of an SK of 32, and the SK value of the green sand was –34, meaning that the sample melted at 1750 °C but not at 1730 °C. Two samples (YH and DH) exhibited the same refractoriness index (an SK of –34) as that of the green sand, and the other sample (SS) exhibited the refractoriness index of an SK of 34.

Table 3
Refractoriness of regenerated sands measured by the KS L-8001 method.

Sample	Waste foundry sands	Recycled sands
Green sand	–	–34
Regenerated sand		
YH	30	–34
SS	32	34
DH	30	–34

This means that all of the regenerated sands can be used as foundry sand.

3.4. Stabilization of residue

The heavy metals on the surfaces of the particles of the WFS were removed from the particles by friction and abrasion during comminution. The sizes of the separated heavy metal particles were finer than those of the raw particles of the WFS. Therefore, the residue that passed the 100-mesh screen (an opening size of 0.15 mm) was a concentrate of heavy metals. The contents of heavy metals in the residues and untreated samples of the WFS were analyzed by ICP-MS and are summarized in Table 4. The Cr⁺⁶ ion was not detected in all of the samples. Table 4 demonstrates that the residue samples contained large amounts of Cu, Zn, Pb, and Cr. Although the heavy metals were concentrated by the regeneration process illustrated in Fig. 1(a), the concentrations of each element were not higher than that found in the WFS. This result is because the heavy metals that were separated from the WFS were removed by magnetic and eddy current separation. Thus, the separation process in the regeneration of WFS is an effective process. Although large amounts of heavy metals were removed, the residue still contained toxic elements, and the Cu and Cr contents of the residues significantly increased. Therefore, the stabilization of toxic elements was considered to prevent their leaching and elution.

Many methods have been developed to stabilize heavy metals. In 1990, Holmes reported a method to quantify leachability with respect to the binders used in a comparative study of three separate solidification and stabilization (S/S) treatment technologies [13]. In 1993, Cocke and Mollah reviewed the binding chemistry and leaching mechanisms of hazardous substances, particularly heavy metal pollutants, in cementitious S/S systems in terms of the surface and solution chemistries of the pollutants [17]. In 1993, Butler et al. and Ivey et al., among several others, reported the results of using experimental techniques, such as electron microscopy and NMR spectroscopy, to characterize cement-solidified hazardous wastes. In addition, the American Foundry Society recently studied a number of applications for foundry sands, including geotechnical fill applications, such as drainage aggregate, filtration media, slurry and utility trench back fill, final landfill cover, Portland cement, and concrete [18–20].

Despite numerous research studies, few researchers have addressed methods that are economical with respect to efficiency and operational costs. Therefore, any successful stabilization process should be relatively cheap and sufficiently effective to ensure an advantage for its commercialization.

In this study, a self-propagating combustion method was used to stabilize heavy metals. We previously reported that self-propagating combustion is an effective sintering method for clay minerals [21]. This method consists of three stages. The first stage involves the pelletization of powdered residue, while second stage consists of a coating process using a mixture of coal refuse and sodium silicate, and the last stage involves sintering via the combustion of carbon in coal refuse. Sodium silicate was used to decrease the sintering temperature of the layer coated with coal refuse and to increase the bonding strength between carbon in the

Table 4

Heavy metal contents of the waste foundry sands and the residues after regeneration by ICP-MS, ppm.

Sample	Cu	Zn	Pb	Cr	As	Cd	Cr ⁺⁶
Waste foundry sand							
YH	60	107	40	188	<1.0	<1.0	0.0
SS	40	70	30	100	<1.0	<1.0	0.0
DH	70	120	60	230	<1.0	<1.0	0.0
Residue after regeneration							
YH	69	22	13	388	<1.0	<1.0	–
SS	48	30	<5	87	<1.0	<1.0	–
DH	121	180	7.4	389	<1.0	<1.0	–

Table 5

Change of carbon content of DH pellet by coated coal refuse of 25 wt% versus sintering temperature by proximate analysis.

Temperature	Weight (g)	Moisture (%)	Volatile (%)	Ash (%)	Fixed-carbon (%)
1100°C	0.9911	0.01	0.02	99.78	0.18
1000°C	1.0583	0.02	0.04	99.75	0.19
900°C	1.0843	0.02	0.04	99.77	0.17
Green pellet	1.0425	0.43	4.98	86.67	7.92

coal refuse and the pelletized residue. The use of coal refuse as an energy source has two advantages: a decrease in the operational costs and the recycling of mine waste.

To prepare the mixture, coal refuse was ground to a particle size of less than 0.075 mm, and the resultant pelletized residue was coated with sodium silicate (0.03–1.08% of coal refuse). Approximately 20–30% of the pelletized residue was used in the mixture.

Table 5 depicts the change in the carbon content of the DH pellet at various sintering temperatures. This result was measured by proximate analysis. The content of fixed carbon after sintering decreased from 7.92% to less than 0.19%. The amount of fixed carbon that was consumed during sintering was almost the same as that at all sintering temperatures above 900 °C. This finding means that all of the fixed carbon in the DH pellet was oxidized at a temperature below 900 °C.

The relationship between the compressive strength (CS) and added amount of sodium silicate at various sintering temperatures was investigated without coating the coal refuse. The point-load test was used to measure the CS of each sintered pellet. The point-load test is an effective method to determine the compressive strengths of irregularly shaped pellets [22]; thus, it was well suited to analyze the irregularly shaped sintered pellets that were obtained in this study. The compressive strength can be calculated using the following equation:

$$CS \approx I_{s(50)} \times A$$

where CS is the compressive strength, $I_{s(50)}$ is the point-load strength index of the sample, and A is a conversion factor. The value of A generally ranges between 20 and 25, and CS was calculated as the lowest value of A .

Fig. 6 demonstrates that the CS increased as the added amount of sodium silicate increased. Also, the CS increased as the sintering temperature increased from 900 °C to 1100 °C.

Fig. 7 summarizes the change in the CS versus different sintering temperatures by the self-propagating combustion method using 25% coal refuse with 1.08% of added sodium silicate. The CS increased as the sintering temperature increased but rapidly decreased at temperatures above 1000 °C.

From the results depicted in Figs. 6 and 7, the optimum conditions were considered to be a sintering temperature of 1000 °C, 25% coal refuse, and 0.51–0.54% sodium silicate. An extraction procedure toxicity test was performed under the above conditions, and the results are presented in Table 6. The concentrations of extracted elements except Cr were less than 0.02 ppm. The concentration of extracted Cr was less than 0.1 ppm. This result means

that the surface of the stabilized pellet under the optimum conditions was almost perfectly melted, and the heavy metals within the pellet could not be extracted from the surface of the pellet. Therefore, self-propagating combustion using coal refuse is an effective

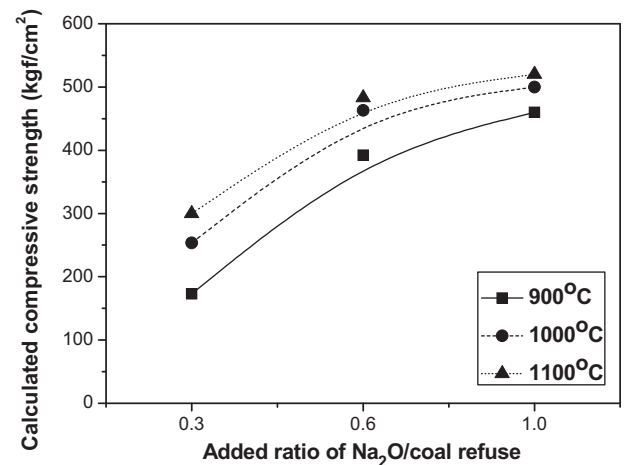


Fig. 6. Compressive strength versus different ratios of added Na₂O at various sintering temperatures. The weight ratio of coated coal refuse was fixed at 25 wt%.

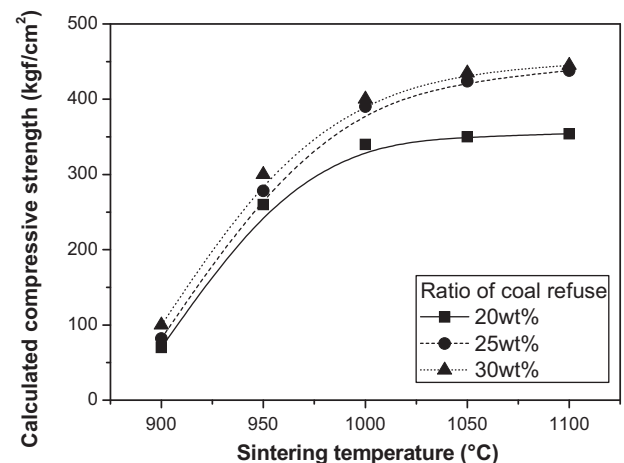


Fig. 7. Compressive strength versus different sintering temperatures under the self-propagating combustion method at various coal refuse weight ratios. The weight ratio of Na₂O was fixed at 0.6 wt%.

Table 6

Result on the TCLP test of stabilized pellet by self-propagating combustion using coal refuse under the optimum condition.

Sample	Extracted concentrate (ppm)					
	Cu	Zn	Pb	Cr	As	Cd
YH	<0.02	<0.02	<0.02	<0.1	<0.02	<0.02
SS	<0.02	<0.02	<0.02	<0.05	<0.02	<0.02
DH	<0.02	<0.02	<0.02	<0.1	<0.02	<0.02

stabilization method for residue generation from regenerated WFS.

4. Conclusions

The regeneration process and the stabilization process were considered as a suitable method for recycling of WFS: the regeneration process by comminution (crushing and grinding) and separation (magnetic separation at 3000 G and eddy current separation), and the stabilization process of the residue generated by screening during regeneration. For economical stabilization, the self-propagating combustion method using coal refuse was used.

The sand regenerated under optimum conditions had a SiO₂ content greater than 94%, and the recovery ratio was in excess of 80%. Also, the refractoriness index of the regenerated sand was greater than 1730 °C. The optimum conditions for the stabilization of the residue generated from the regeneration process was identified to include a coating mixture of 25% coal refuse, 0.51–0.54% sodium silicate, and a sintering temperature of 1000 °C. All of the heavy metal elements except Cr were found in concentrations of less than 0.02 ppm, and the Cr concentration was lower than 0.1 ppm.

In conclusion, it is adequately possible to replace green sand by the regenerated sand in the foundry industry and the self-propagation combustion could be used as an effective method to stabilize the residue.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.jhazmat.2011.11.100.

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