Research on co-disposal and utilization of ferrous packaging containers contaminated with hazardous wastes by steel converter

Yi WANG
Wuhan Polytechnic University
Junfu CHEN
Guangdong Academy of Sciences
Benquan Fu
WISCO: Wuhan Iron and Steel Group Corporation
LEI ZHANG (zhanglei@whpu.edu.cn)
Wuhan Polytechnic University https://orcid.org/0000-0001-6374-0530
Heng LIU
Wuhan Polytechnic University
Yanjun HUANG
Wuhan Polytechnic University
Guangsen SONG
Wuhan Polytechnic University

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Abstract

The disposal of waste oil, paint and coating barrels (WOPCBs) after use is an ongoing social and environmental problem. A novel co-disposal and utilization method of WOPCBs has been proposed in this paper. Compared with the traditional methods, the method presented takes full advantages of the production process and environmental protection facilities of steel converter, and has many advantages, such as a large disposal capacity, high incineration temperatures, thorough decomposition of organic compounds, low cost, easy operation, highly efficient and environmentally friendly. Results of industrial tests revealed that the addition of 180–540 kg WOPCBs for each furnace load(250 ton capacity per batch), none of the assessed pollutant discharge indices deteriorated, meeting the relevant national standards. Furthermore, WOPCBs were suitable for use as supplementary materials for scrap steel. Therefore, this study provides important insights for sustainable resource utilization from WOPCBs.

1. Introduction

Wuhan is the largest industrial city in central China, dominating in industries such as shipbuilding, automobile, petrochemical, and steel. In these industrial processes, a large number of waste iron packaging containers contaminated with waste oil, paint and coating barrels (WOPCBs) and other hazardous materials, are produced annually. According to the Chinese “National Hazardous Waste List”, WOPCBs are recognized as HW49 (900-041-49, National Hazardous Wastes Catalogue of China, 2016 version) hazardous waste and therefore, improper disposal presents a hazard to the environment [1, 2]. For example, the disposal of the hazardous WOPCBs generates oil containing waste water, which must be adequately treated prior to discharge into the receiving water body to avoid negative effects such as toxicity to aquatic organisms, reduced oxygen penetration and deterioration of natural photosynthetic activity.

At present, the methods of disposal of iron packaging containers contaminated with hazardous waste mainly include cleaning, oxygen cutting, disintegration, processing and packaging for use in scrap steel production, or recycling by cutting and crushing, followed by high-temperature alkali washing, or simple incineration. The process is characterized by low incineration temperature (500–1000°C), small disposal capacity (200–3000 ton/a), scattered distribution, and imperfect pollutant control measures, resulting in serious pollution to the surrounding atmospheric environment [3–5]. In contrast, hydrometallurgical processes have the potential to reduce environmental impact, are suitable for large scale applications, and have low capital cost [6]. Chen et al [7] used a vertical melting pyrolysis furnace process to dispose of industrial slag steel and WOPCBs, using high-temperature melting treatment and slag iron separation. This method was shown to effectively recycle solid waste, generating good economic benefits .

Due to the comprehensive functions of steel plants (i.e broaden-manufacturing function of steel product, function of energy conversion and function of waste recycling), much research has focused on exploring new ways to recycle metallurgic solid waste, based on the guidelines for energy conservation, emissions reduction, circular economy development and ecological protection [8–11].

In this study, industrial trials for co-disposing WOPCBs through well-developed high-temperature process were first conducted using the converter currently existing in a steel plant. The large-scale converter had a large co-disposal capacity and operated at high temperature ranges from 1600 to 1700 ℃ during the smelting process and was assessed for its ability to fully decompose organic compounds in packaging containers contaminated with hazardous waste. Furthermore, in order to achieve comprehensive utilization of iron resources, the potential for combination of residual scrap steel with molten iron was assessed, to form an effective scrap steel supplement. Finally, the combination of this system with a highly effective flue gas treatment facility was estimated, to ensure that the co-incineration process did not increase environmental pollution due to harmful atmospheric emissions. The aim of this study was to reduce the social impact of contaminated waste packaging container disposal, achieving the reuse of valuable resources and reduction in energy requirements, allowing steel plants to become a potentially important sustainable and low-ecological impact industry.

2. Experiment

2.1. Materials

WOPCBs were collected from WISCO Metal Resources Co. Ltd.. After manual oil cleaning and liquid leaching, these WOPCBs and packaging materials were sent to the packing machine for flattening and compression into blocks. Steel slag, dust, mud cake and washing water of converter gas used in this work was collected from Wuhan Iron and Steel Company Limited (Wuhan, China). All the solvents were of analytical grade, unless otherwise stated. The solvent such as H_2SO_4 (≥99.0%), CH_3COOH (≥99.0%), HCl (≥99.0%) were purchased from Shanghai Aladdin Biochemical Technology Co., Ltd.. Deionized water was obtained by a water purification system, which the electrical conductivity was less than 0.055 μS/cm. The type of water purification machine was Milli-Q Academic, Merck Millipore, Darmstadt, Germany.

2.2. Analytical methods

The heavy metals were leached from steel slag, dust and mud cake following the Chinese Standard Procedure HJ/T299-2007. The leachates were filtered with a 0.45μm micropore membrane. The leachates were digested in a mixture of hydrochloric acid (HCl) and nitric acid (HNO_3). The concentrations of heavy metals and element analysis of P, Si and Mn in samples were analyzed by inductively coupled plasma optical emission spectroscopy (ICP-OES; Thermo Fisher scientific, ICAP 6500, USA). Moreover, the chromium (VI) concentrations in these leachate were measured by using the standard “Water Quality - Determination of Chromium (VI) - 1.5 Diphenylcarbohydrazide Spectrophotometric Method” (GB7467-87, China). The CS-800 carbon sulfur analyzer (Eltra, Germany) was equipped with four independent infrared detection cells to obtain accurate analysis of carbon and sulfur content in a single measurement process.Volatile organic compounds (VOCs), polycyclic aromatic hydrocarbons (PAHs) and dioxins in flue gas were analyzed by HP 6890 gas chromatography equipped with HP5975B mass spectroscopy detector (Agilent Co., USA). Sampling and analytical procedure for PAHs were adopted by Chen et al [12]. For details, see Zhong et al [13] and Gan et al [14]. All of the analyses were performed in duplicate.

2.3. Disposal process

The procedures of co-disposal of WOPCBs by converter steelmaking had been tested in a commercial (250 ton capacity per batch) basic oxygen furnace (BOF), No.3 steelmaking plant at Wuhan Iron and Steel Company Limited (WISCO), Wuhan, China. According to the steelmaking production plan, after cleaning, extrusion and packing the WOPCBs were transported and used as a raw material in the scrap room in No.3 steelmaking Plant of WISCO in time. It was preliminarily determined that the load of WOPCBs added was 180 kg per furnace, accounting for 0.072% of the total capacity. If the addition of 180 kg per furnace load was confirmed to present no safety problems, the adding amount could be adjusted to 180-540kg per furnace load. During the test period, the concentrations of PAHs, NMHC, VOCs and dioxins in the converter flue gas and the content of heavy metals in the scrap steel, gas washing water and mud cake, were monitored. The experimental process was carried out continuously for 90 days.

2.4. Disposal method

The collected and stacked WOPCBs in the factory were weighed and transported by truck to the WOPCBs packaging operation area of WISCO Metal Resources Co. Ltd. After degreasing and cleaning, these WOPCBs and packaging materials were sent to the packing machine for flattening and compression into blocks. The final products were transported to the product stacking area via a conveyor belt and fork-lift truck. The liquid generated during the compression process was collected and transported to the leachate storage area.

A piece of pressed block of WOPCBs was about 180 kg and the total amount of pressed block of WOPCBs was approximately 3.24 tons. Generally, 1-3 pieces of WOPCBs pressed block scrap were drawn by magnetic crane into each furnace load. Calculation of the variation in the main raw material content and flux in the test furnace efficiency were performed. In order to prevent excessive dust when pouring melted iron, the test furnace was shaken 4 times at ±50° after the addition of scrap steel. The disposal process should be immediately stopped if any spray explosion occurs. Under the high temperature and strong oxidation conditions of converter systems, The ferrite content of these WOPCBs was melted into the molten iron or steel slag, while oil compounds in WOPCBs are decomposed into flue gas emissions, which enter the converter off gas purification system via the dust removal system, before being discharged. The technical process of comprehensive utilization and disposal of WOPCBs was shown in Figure 1 and the practical disposal process was shown in Figure 2.

3. Results And Discussions

3.1 Principle of utilization and disposal of WOPCBs

The purpose of converter systems in steelmaking is to produce steel with optimal physical, chemical and mechanical properties, by oxidizing carbon, silicon, manganese, and other impurities in pig iron under pure oxygen conditions. Converter systems complete the process using the physical heat generated from molten iron and the chemical heat formed from the reaction of inner components. Hot metal, scrap steel and ferroalloys are the main raw materials in this process. The melting point temperature of pig iron ranges from 1100 to 1250°C, while the temperature of molten steel ranges from 1450 to 1700°C [15]. Oxygen is lanced to the furnace to oxidize the impurities in the hot metal. After the blowing, smelting temperatures can reach levels of up to 1700°C, the impurities (C, Si, S, etc.) present in steel is utilized in reducing the iron oxide contained in the WOPCBs and melting as Fe or slag; thereafter, metal and slag are collected separately and the iron of WOPCBs provides an effective supplement to scrap steel. The remaining oil in WOPCBs is fully decomposed, the emissions from this process entering the converter off gas purification system through the dust removal system and finally being recovered by gas holder or discharged via a chimney. The possible chemical reactions during disposal process are shown in Figure 3.

3.2. Analysis of influence on steelmaking production

3 pieces of WOPCBs (~540kg per batch) pressed block scrap steel was added, the converter was shaken 4 times at ±50°. The flame and dust concentration at the furnace mouth were observed to be normal. Two minutes later, iron mixing was initiated for a duration of 4 min. The smoke and flame levels at the furnace mouth were increased slightly, although no splashing or shooting was observed during the process. Blowing was performed from the 7 to 16 min, with no splashing or abnormal noises observed in the furnace. At the 31 min, the process was returned to normal functioning and steel was discharged. The chemical components of hot metal were presented in the Table 1.

These results illustrate that it is feasible to use scrap steel from WOPCBs pressed blocks as the test raw material. Compared with the disposal process for ordinary scrap steel, the modified scrap steel disposal process required 4-cycles of shaking, prolonging the production cycle time. According to the presented data, the WOPCBs pressing block scrap steel could be used in ladle furnace desulfurization steel. There were no obvious differences observed in the converter blowing process and both production and safety conditions could be controlled, which did not affect the quality of steel products and allowed control of the sulfur content of molten steel.

3.3. Analysis of influence on the environment

3.3.1 Waste gas emission

For the converter off gas generated in the converter refining process, the dedusting technique of oxygen converter gas recovery(OG) is used. Dust is then scrubbed out of the off-gas from the process through wet venturi scrubber systems before the off-gas is suitable to either collect for gas recovery or to flare to the atmosphere. The flue gas escaping from the converter smelting process, as well as the flue gas produced by slag stripping and desulfurization in ladle argon blowing station, adopts the type of smoke hood in front of the furnace to collect, and then sends it to the bag filter for purification, and then discharges it to the atmosphere through the exhaust pipe. After dosing WOPCBs in the converter, VOCs, PAHs, dioxins and other pollutants may be generated in the incineration process. The environmental indicators during the WOPCBs disposal process were commissioned by a third party company.

Monitoring results showed that the concentrations of benzene, toluene, xylene, NMHC, VOCs and dioxins in emissions were slightly increased from 0.024 mg m^{-3}, 0.207 mg m^{-3}, 0.0235 mg m^{-3}, 0.44 mg m^{-3}, 0.841 mg m^{-3} and 0.032 (ng-TEQ m^{-3}) prior to testing, to 0.326 mg m^{-3}, 0.378 mg m^{-3}, 0.0455 mg m^{-3}, 2.22 mg m^{-3}, 2.966 mg m^{-3} and 0.039 ng-TEQ m^{-3} during testing, as indicated in Table 2. However, the concentrations of all pollutants met the limits of Chinese standards "Integrated Emission Standard of Air Pollutants" in China (GB16297-1996) [16] and the "Emission Standard of Air Pollutants for Steel Smelt Industry" in China (GB28664-2012) [17]. It should be pointed out that it is unnecessary too much concerned about the increase of dioxin concentration when the WOPCBs disposal process. The boiling points of the dioxins ranged between 421 and 447 ℃ and the decomposition temperature was above 800 ℃ [18]. A key characteristic of steelmaking converter systems was that the instantaneous temperature of added WOPCBs was over 1300 ℃, while the smelting temperature can reach above 1600 ℃. Therefore, the temperature conditions required for the formation of dioxins are not formed during the smelting process. Subsequently, the converter off gas was cooled and atomized efficiently to make the flue gas rapidly cool from above 1000 ℃ to 170~200 ℃. This ensured that dioxins can be completely decomposed and would not be generated again during cooling [19, 20].

3.3.2 Waste water emission

The waste water mainly comes from the water for primary converter off gas cooling and dust removal, which is recycled and not discharged. The process flow of circulating water treatment system can be described as: firstly, the backwater of circulating water entered the coarse particle separator through the elevated flow tank, then after separating the coarse particles, the supernatant was pumped into the distribution tank, and then coagulated sedimentation in radial-flow sedimentation tank. At last, the clear water was cooled to room temperature and sent back to the circulating water system for reuse. ICP-MS analysis in circulating water showed that the concentrations of seven heavy metals such as mercury (Hg), cadmium (Cd), nickel (Ni), lead (Pb), chromium (Cr), arsenic (As) and copper (Cu) in circulating water were found to be close to detection limits, and all the concentrations of heavy metals from washing water could meet the corresponding standard requirements of the "Discharge standard of water pollutants for iron and steel industry of China (GB13456-2012)". The results are shown in Table 3.

3.3.3 Solid waste emission

The main solid waste produced in the disposal process of WOPCBs was steel slag, mud cake produced by circulating water treatment system and dust collected by converter off gas. Ministry of Ecology and Environment of China has developed a regulatory definition and process that identifies specific substances known to be hazardous and provides objective criteria for "Identification standard for hazardous wastes-Identification for extraction procedure toxicity"(GB 5085.3-2007). The sequential leaching test were performed to investigate the leachability of heavy metal ion from the original steel slag, mud cake and dust in aqueous solution according to the Chinese Standard Procedure HJ/T299-2007. The leaching results for these heavy metals were shown in Table 4.

The results in Table 4 indicate that the concentrations of heavy metal leaching solution of all metallurgical solid wastes were far lower than the standard limits of “Indentification standards for hazardous wastes indentification for extraction toxicity of China"(GB5085.3-2007). This means that all metallurgical solid wastes can generally be categorized as a non-hazardous by-product from the steel industry and could potentially be recycled for use elsewhere, which is beneficial to the environmental protection and resource saving. Currently, mud cake and dust were used as raw materials of sintering, and steel slag was used as cement aggregate [10, 21]. WOPCBs contained a small quantity of inorganic substances, mainly Ca and Si compounds and trace amounts of heavy metals, such as barium, lead, antimony, cadmium, chromium, strontium and other heavy metals or other harmful metal elements, which can be reacted with lime under high temperatures and strong liming effects to form a very stable silicate which eventually combined with the steel slag [22, 23]. Meanwhile, The precipitation of calcium carbonate on the surface of the steel slag particles blocked the liquid phase from reaching the surface of the soluble mineral phase, and this action caused a decrease in the dissolution rate of the mineral phases, affecting the leachability of heavy metal ions in the encapsulated crystal phase. Because of the stable properties of silicates, their use did not lead to environmental concern [24]. Meanwhile, Silicates and steel slag can be comprehensively reused as a building material such as cement, avoiding further contributions to overall emissions .

3.4. Analysis of material balance

Most of the coating residue of waste paint barrels is formed of organic substances (about 10wt.%), containing the elements C, H and O, which was reduced in the smelting process to a coating residue proportion of less than 0.004%. The converter smelting temperature was about 1600 ℃, with smelting being an oxidation process in an oxygen environment. All organic substances were decomposed under the high temperature conditions, which did not affect the safety or quality of converter smelting products [25]. Through decomposition and transformation under high temperatures in the converter, the content of N, S and other heavy metals in WOPCBs were lower the limit of emission standard in Chinese iron & steel industry. The inorganic substances component contained the elements Ca and Si in the coating, which reacted with lime at high temperatures to form very stable silicate materials, which entered the steel slag but do not affect the molten steel products. Ferrite also entered the molten iron in the converter, becoming a partial substitute of scrap steel [26].

4. Conclusions

The procedures of co-disposal of WOPCBs by converter steelmaking carried out for this study were proved to be successful and feasible. This novel procedures allow both the valuable ferrous metal to be used and the hazardous substance to be accessible for control in a more efficient way. As shown by these results, with the addition of 180–540 kg of WOPCBs for each furnace load, the developed smelting steel converter work effectively in conducting the disposal of WOPCBs. The product quality and production process were both found to be safe and controlled, emission of pollutants can meet related standards and requirements of user, and the leaching of heavy metals in solid waste are far lower than the requirements of the emission standard, which has been shown to be an economically feasible alternative. Therefore, when co-disposing WOPCBs through converter steelmaking process, adding 180–540 kg of WOPCBs for each furnace load was recommended, and suggest that the disposal process of WOPCBs exhibited no environmental risks, which was within safe limits.

Declarations

Availability of data and materials

All data generated or analyzed during this study are presented within the submitted manuscript.

Competing interests

The authors declare they have no competing interests.

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Authors' contributions

Yi Wang: provided conceptualization, methodology, writing-review & editing.Junfu Chen: provided conceptualization, methodology,writing-original draft preparation and writing-review & editing. Benquan Fu: provided resources, project administration and formal analysis. Lei Zhang: provided conceptualization,writing-original draft, and acquisition of funding. Heng Liu:processed investigation, validation and formal analysis. Yanjun Huang: provided visualization, investigation and methodology. Guangsen Song: provided supervision and resources. All authors read and approved the final manuscript..

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Tables

Table 1

Chemical components of hot metal in co-disposal process [a]

No.	Dosage Kg per furnace	C (wt.%)	S (wt.%)	P (wt.%)	Mn (wt.%)	Si (wt.%)
1	180	0.0775	0.0025	0.012	1.5232	0.1674
2	180	0.0779	0.0026	0.012	1.5106	0.1492
3	360	0.0777	0.0012	0.0112	1.5635	0.1463
4	540	0.0616	0.0023	0.011	1.3865	0.1508
5	540	0.0535	0.0068	0.011	1.2135	0.1575
Standard [b]		0.06-0.10	<0.008	<0.025	1.2-1.65	0.1-0.4

a The unit of elemental analysis is weight percent.

b The element content standard of desulfurization steel in WISCO.

Table 2

Characteristics of the exhaust gas for co-processing of the WOPCBs in converter

Compounds	Practical average value		Standard limits
	Before test (mg Nm^{-3})	In the process (mg Nm^{-3})	(mg Nm^{-3})
Benzene	0.024	0.326	12 [a]
Toluene	0.207	0.378	40 [a]
Xylene	0.0235	0.0455	70 [a]
NMHC	0.44	2.22	120 [a]
VOCs	0.841	2.966	-
Dioxin	0.032 (ng-TEQ m^{-3})	0.039 (ng-TEQ m^{-3})	0.5 [b] (ng-TEQ m^{-3})

a "Integrated Emission Standard of Air Pollutants" in China (GB16297-1996)

b "Emission Standard of Air Pollutants for Steel Smelt Industry" in China (GB28664-2012)

Table 3

The concentrations of heavy metals from washing water of converter gas. (unit: mg L^{-1})

Metal element	**Hg**	**Cd**	**Ni**	**Pb**	**Cr^a**	**Cr^b**	**As**	**Cu**	**Zn**
washing water of converter gas	0.028	0.050	0.50	0.45	0.47	0.080	0.060	0.071	0.78
Discharge standard of China (GB13456-2012)	0.05	0.1	1.0	1.0	1.5	0.5	0.5	0.5	2.0

Note: Cr^a mean the total content of Cr; Cr^b mean the content of Cr□□□

Table 4

Mean values of the leaching concentrations of studied metals in solid waste. (unit: mg L^{-1})

Metal element	**Hg**	**Cd**	**Ni**	**Pb**	**Cr^a**	**Cr^b**	**As**	**Cu**	**Zn**
Steel slag	0.03	0.03	0.38	0.12	0.30	0.03	0.040	0.050	0.035
Dust	0.03	0.04	0.21	0.37	0.41	0.04	0.031	0.061	0.43
Mud cake	0.04	0.05	0.30	0.45	0.39	0.04	0.05	0.047	0.59
Chinese National Standard (GB5085.3-2007)	0.1	1	5	5	15	5	5	100	100

Note: Cr^a mean the total content of Cr; Cr^b mean the content of Cr□□□

Figures

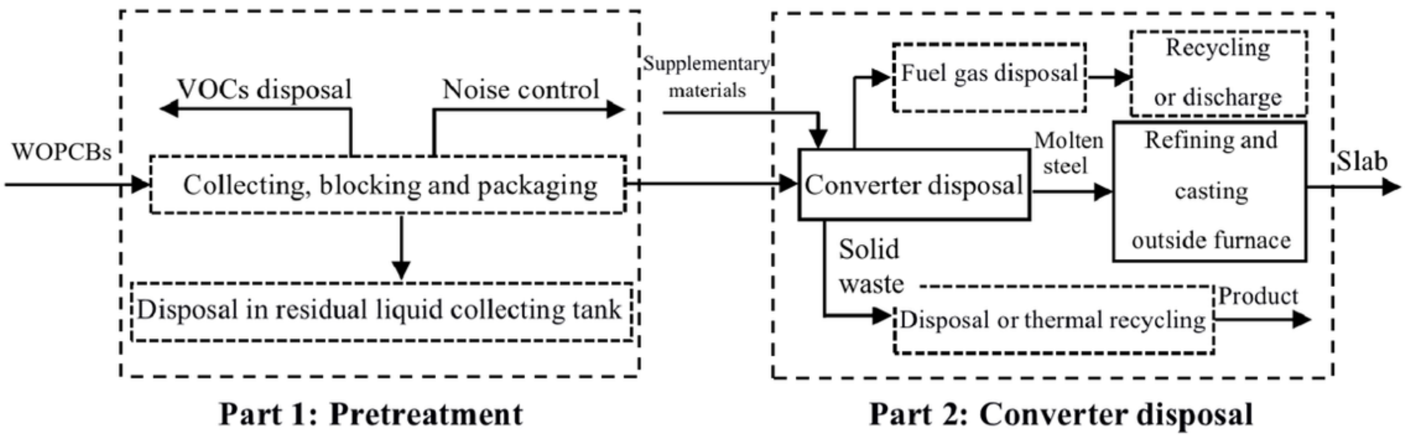

Figure 1

Disposal process of WOPCBs

Figure 2

On-site test process: (a) barrels collected; (b) pressed block; (c) block product; (d) drawn by magnetic crane into feed hopper; (e) feeding into converter; (f) co-disposal in converter

$$C_XH_Y+(x+\frac{y}{4})O_2 \rightarrow xCO_2+\frac{y}{2}H_2O+Q$$

$$C_XH_YS_ZO_W+(x+\frac{y}{4}+z-\frac{w}{2})O_2 \rightarrow xCO_2+\frac{y}{2}H_2O+zSO_2+Q$$

$$C_XH_YS_ZN_MO_W+(x+\frac{y}{4}+z+\frac{M}{2}-\frac{w}{2})O_2 \rightarrow xCO_2+\frac{y}{2}H_2O+zSO_2+\mathrm{m}NO+Q$$

Figure 3

Chemical reactions in disposal of WOPCBs