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

Yi Wang¹, Junfu Chen², Benquan Fu³, Lei Zhang^{1*} , Heng Liu¹, Yanjun Huang¹ and Guangsen Song¹

Abstract

The disposal of waste oil, paint and coating barrels (WOPCBs) after use is an ongoing social and environmental problem. In this paper, a novel technological scheme using steel converter co-processing is proposed for the safe disposal and resource utilization of WOPCBs. The scheme is mainly composed of WOPCBs pretreatment and converter disposal, focusing on the impact of converter co-processing WOPCBs on the environment and production process. Compared to the traditional technology, the scheme 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, low cost, highly efficient and environmentally friendly. The industrial trials results show that after adding 180–540 kg WOPCBs to each furnace load (nominal capacity 250 t), the converter operation is safe and controllable, and all the pollutant emission indicators generated in the process meet the Chinese national standards. In addition, WOPCBs are suitable to be used as a supplementary material for scrap steel. Therefore, this study provides important insights for sustainable resource utilization from WOPCBs.

Keywords: WOPCBs, Hazardous materials, Co-processing, Steel converter

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 poses a great threat to environmental systems [1, 2]. For example, the disposal of the hazardous WOPCBs

generates oil containing wastewater, 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.

Generally, the disposal of WOPCBs should conform to the principles of safety, resource utilization, and simplification. At present, there are two main methods for industrial disposal of WOPCBs, the first is chemical cleaning and reuse, the second is oxygen cutting, then followed by high-temperature alkali washing, or simple incineration [3]. Both methods have their advantages, but they also have their fatal defects. The wet cleaning process can realize the recycling of resources. However, this method will discharge a large amount of cleaning waste liquid and strong alkali containing highly toxic organic substances. Existing incineration process is characterized by low incineration temperature (500–1000 °C), small disposal

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capacity (200–3000 t yr⁻¹), scattered distribution, and imperfect pollutant control measures, resulting in serious pollution to the surrounding atmospheric environment [4–6]. In contrast, pyrometallurgical processes have the potential to reduce environmental impact, are suitable for large scale applications, and have low capital cost [7]. Chen [8] 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 [9–12].

Inspired by the application of co-processing scrap in converter steelmaking, this paper intends to use the converter in steelmaking plant to dispose WOPCBs. In this study, industrial trials for co-disposing WOPCBs through well-developed high-temperature process were first conducted using the converter in steelmaking plant. The large-scale converter had a large co-disposal capacity, which was operated at high temperature ranges from 1600 to 1700 °C during the smelting process. In this paper, we designed a novel process scheme for comprehensive utilization and safe disposal of WOPCBs. The scheme not only realized the comprehensive recovery of valuable iron resource from WOPCBs, but also avoided the production of secondary pollution such as strong alkali containing oily wastewater and toxic gas. Moreover, the treatment cost was greatly reduced due to the decomposition and exothermic reaction of organic compounds in WOPCBs, combination of this system with a highly effective flue gas treatment facility and utilization of iron resources. Therefore, a win-win goal of simultaneously obtaining valuable iron resources and minimizing their impact on the environment can be achieved, which also makes the steel plant a potentially important sustainable and low ecological impact industry.

2 Experiment

2.1 Materials

WOPCBs were collected from WISCO Metal Resources Co., China. 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 were collected from Wuhan Iron and Steel Co. (WISCO, China). All the

solvents were of analytical grade, unless otherwise stated. The solvent such as H₂SO₄ (~98%), HNO₃ (~68%) were purchased from Shanghai Aladdin Biochemical Technology Co., China. Deionized water was obtained by a water purification system (Milli-Q Academic, Merck Millipore, Darmstadt, Germany), with the electrical conductivity less than 0.055 µS cm⁻¹. Other chemical reagents (Sinopharm Chemical Reagent, China) were of analytical reagent grade.

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 0.6–0.8 µm micropore membrane. The leachates were digested in a mixture of H₂SO₄ and HNO₃. 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 leachates were measured by using the standard “Water Quality - Determination of Chromium (VI) – 1,5-Diphenylcarbohydrazide Spectrophotometric Method” (GB7467–87, China). Elemental analyzer (Elementar/UNICUBE, Germany) was used for quantitative analysis and determination of C, H, N, S, O elements in organic solids, highly volatile and sensitive substances. Volatile organic compounds (VOCs) and dioxins in flue gas were analyzed by HP 6890 Gas Chromatography (GC) equipped with HP5975B Mass Spectroscopy (MS) Detector (Agilent, USA). All 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 (nominal capacity 250 t) basic oxygen furnace, No. 3 steelmaking plant at WISCO, Wuhan, China. According to the steel-making production plan, after cleaning, extrusion and packing the WOPCBs were transported and used as a raw material in the scrap steel room in No. 3 steelmaking plant of WISCO. 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 180–540 kg per furnace load. During the test period, the concentrations of nonmethane hydrocarbons (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 d.

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 plant. It should be noted that the residue in WOPCBs must be less than 5 wt% of the total mass. 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 t. Generally, 1–3 WOPCBs pressed blocks were first moved into the converter hopper by the magnetic crane, and then poured into the converter ladle together with the scrap in the hopper for smelting. 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 a ± 50 degree angle after the addition of WOPCBs pressed blocks and scrap steel. The disposal process should be immediately stopped if any spray explosion occurred. Under the high temperature and strong oxidation conditions of the converter system, the inorganic components in the WOPCBs are melted into molten iron or steel slag, and the organic components in the WOPCBs are decomposed into flue gas, which is sent to the converter exhaust gas purification and recovery system. The technical process of comprehensive utilization and disposal of WOPCBs is shown in Fig. 1 with the practical disposal process shown in Fig. 2.

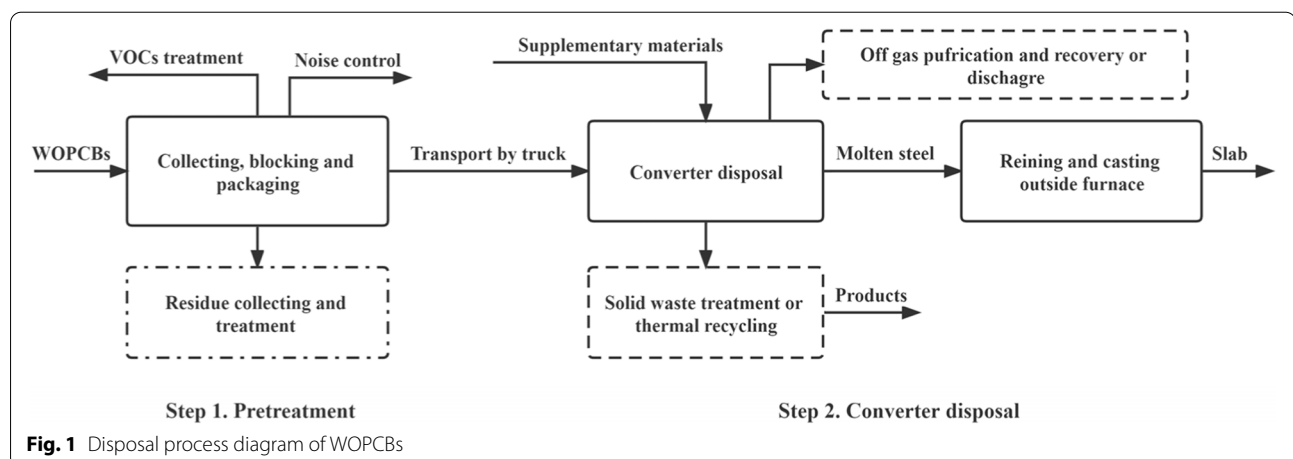
3 Results and discussion

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 [13]. 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 Fig. 3.

3.2 Analysis of influence on steelmaking production

Three pieces of WOPCBs (~540 kg per batch) pressed block scrap steel were added, the converter was shaken 4 times at ± 50 degree angle. 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 are 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 and 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.

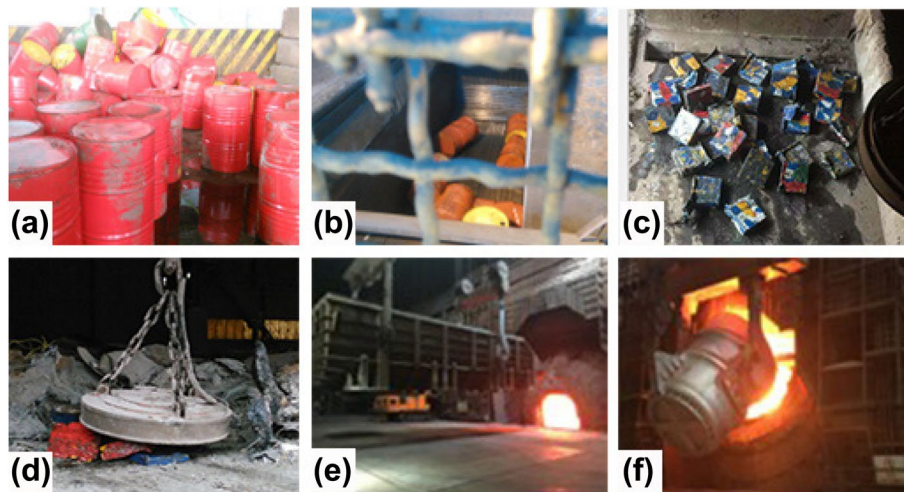


Fig. 2 On-site test process: **a** barrels collected; **b** pressed block; **c** block product; **d** drawn by magnetic crane into feed hopper; **e** feeding into converter; **f** co-disposal in converter

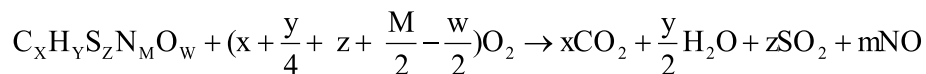
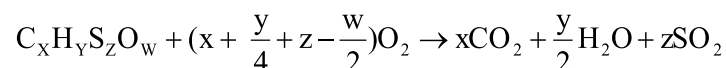
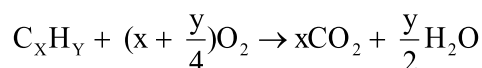


Fig. 3 Chemical reactions in disposal of WOPCBs

Table 1 Chemical components of hot metal in co-disposal process

No.	Dosage kg per furnace	C (wt%)	S (wt%)	P (wt%)	Mn (wt%)	Si (wt%)
1	180	0.078	0.0025	0.012	1.52	0.17
2	180	0.078	0.0026	0.012	1.51	0.15
3	360	0.078	0.0012	0.011	1.56	0.15
4	540	0.062	0.0023	0.011	1.39	0.15
5	540	0.054	0.0068	0.011	1.21	0.16
Standard ^a		0.06–0.10	< 0.008	< 0.025	1.20–1.65	0.1–0.40

^a The element standard of desulfurization steel in WISCO

3.3 Impact on the environment

3.3.1 Flue gas purification, recovery and treatment

For the converter off-gas generated in the converter refining process, the dedusting technique of oxygen converter gas recovery 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. There is a general concern about problems related to the odor, toxic gases and particles during the WOPCBs disposal. The rigorous tests of off-gas generated in the process have been detected by a third party, and VOCs, dioxins, benzene, toluene, xylene, NMHC and other organic pollutants were analyzed by HP 6890 GC equipped with HP5975B MS detector.

Monitoring results showed that the concentrations of benzene, toluene, xylene, NMHC, VOCs and dioxins in emissions were slightly increased from 0.024, 0.21, 0.024, 0.44, 0.84 mg m^{-3} and 0.032 (ng-TEQ m^{-3}) prior to testing, to 0.33, 0.38, 0.046, 2.22, 2.97 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) [14] and the “Emission Standard of Air Pollutants for Steel Smelt Industry” in China (GB28664–2012) [15]. It should be pointed out that it is unnecessary for much concern about the increase of dioxin concentration when the WOPCBs disposal process. The boiling points of the dioxins ranged between 421 and 447°C and the decomposition temperature was above 800°C [16]. A key characteristic of steelmaking converter systems was that the instantaneous temperature of added WOPCBs was over 1300°C, while the smelting temperature can reach above 1600°C. 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°C. This ensured that dioxins can be completely decomposed and would not be generated again during cooling [17, 18].

3.3.2 Circulating cooling water treatment and reuse

The wastewater 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 wastewater 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 Hg, Cd, Ni, Pb, Cr, As and Cu in circulating water were found to be close to the limit of detection, 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 treatment and utilization

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 treatment. 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 metals from the original steel slag, mud cake and dust in aqueous solution according to the Chinese Standard

Table 2 Characteristics of the exhaust gas for co-processing of the WOPCBs in converter

Item	Average value					
	Benzene (mg Nm^{-3})	Toluene (mg Nm^{-3})	Xylene (mg Nm^{-3})	NMHC (mg Nm^{-3})	VOCs (mg Nm^{-3})	Dioxin (ng-TEQ m^{-3})
Before test	0.024	0.21	0.024	0.44	0.84	0.032
In the process	0.33	0.38	0.046	2.22	2.97	0.039
Standard	12 ^a	40 ^a	70 ^a	120 ^a	–	0.5 ^b

^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

^a total content of Cr; ^b content of Cr (VI)**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	0.04	0.05	0.04
Dust	0.03	0.04	0.21	0.37	0.41	0.04	0.03	0.06	0.43
Mud cake	0.04	0.05	0.30	0.45	0.39	0.04	0.05	0.05	0.59
Chinese National Standard (GB5085.3–2007)	0.1	1	5	5	15	5	5	100	100

^a total content of Cr; ^b content of Cr (VI)

Procedure HJ/T299–2007. The leaching results for these heavy metals are 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 Chinese Standards (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 used directly in environmental applications. Currently, mud cake and dust were used as raw materials of sintering, and steel slag was used as cement aggregate [11, 19]. WOPCBs contained a small quantity of inorganic substances, mainly Ca and Si compounds and trace amounts of heavy metals, such as Hg, Cd, Ni, Pb, Cr, As, Zn, and other heavy metals or other harmful metal elements, which can be eventually reacted under high temperatures with strong liming effects to form a very stable silicate which eventually combined with the steel slag [20, 21]. Steel slag is a residual product from the steel smelting process, consisting of oxides such as calcium oxide, silicon oxide, and iron oxide. It should be noted that 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 [22]. Meanwhile, Silicates and steel slag can be comprehensively reused as a building material such as cement.

3.4 Material balance

Materials balance principle is the balance of the materials between inputs, desirable outputs and undesirable outputs in the production progress [23]. The process diagram (see Fig. 1) shows the inputs and outputs of steel converter. The mass balance of the process is presented in the Table 5. It is worth noting that this calculation does not take into account the withdrawal of molten steel samples, losses of hot metal splash and flue gas leakages.

Table 5 shows that the molten steel would have increase 8.5%, this is attributed to the addition of scrap steel, ferroalloys and WOPCBs in the steelmaking process. As usual, the proportion of scrap steel added is 16.5–18.8%, and the increase proportion of molten steel is within the

Table 5 Mass balance of the process

Input item	Mass percentage (%)	Output item	Mass percentage (%)
Molten iron	71.7	Molten steel	80.2
Steel scrap	15.7	Steel slag	7.5
WOPCBs	1.3	Metal iron bead	0.2
Lime	3.0	Dust	0.9
Dolomite	1.4	Flue gas	9.8
Fluorite	0.1	Residual slag in furnace	0.2
Erosion of furnace lining	0.2	Leakage	1.3
Ferroalloy	0.05		
Oxygen blowing	6.5		
Total	100	Total	100

Table 6 Cost-effectiveness of converter co-processing technology

Description	Unit cost	Amount	Total cost (USD)
Materials cost			241,448
Waste oil & oily wastewater treatment	769 USD t ⁻¹	250 t	192,250
Waste activated carbon treatment	923 USD t ⁻¹	20.4 t	18,829
Fresh activated carbon	923 USD t ⁻¹	20.4 t	18,829
PE packaging film	2308 USD t ⁻¹	5 t	11,540
Fuel and power cost			45,623
Diesel fuel	977 USD m ⁻³	0.5 m ³	489
Electricity	0.1 USD (kWh) ⁻¹	450,000 kWh	45,000
Tap water	0.54 USD m ⁻³	249 m ³	134
Labor service cost			230,800
Depreciation and maintenance costs			113,700
Total cost			631,571
Co-processing cost per ton			126.31 (A)
Chemical cleaning process per ton			242.85 (B)
Cost saving per ton			116.54 = (B) - (A)

normal range (i.e., WOPCBs as scrap steel substitutes). This can be explained in two ways: firstly, most of the coating residue of WOPCBs is formed of organic substances (below 10 wt%), which was reduced in the smelting process to a coating residue proportion of less than 0.004% [18]. In the steelmaking converter, all organic substances are completely decomposed as volatile gases, such as carbon monoxide, carbon dioxide, sulfur dioxide, vapour, and various other gases under the high temperature (above 1500°C) and oxidation atmosphere, which does not affect the safety and quality of the products smelted by the converter [24]. Secondly, the inorganic substances components in the WOPCBs, such as silicon, manganese, phosphorus, and sulfur, etc., react with lime at high temperatures to form very stable silicate materials, which becomes part of the steel slag. Ferrite in the WOPCBs also enters the molten iron in the converter and becomes a partial substitute of scrap steel [25].

3.5 Economic benefits of the treatment of WOPCBs in converter steelmaking

The operational costs for the process tested at industrial trials were estimated and results summarized in Table 6. Based on the results of the industrial trials and the design of the integrated treatment flow sheet, the key operation factors for the developed process were estimated and an economic feasibility study was carried out. The operating cost is mainly composed of four parts: materials, fuel and power, labor service and depreciation and maintenance. Calculation of the co-processing cost is based on a WOPCBs capacity of 5 kt yr⁻¹. As shown in Table 6, the total cost per ton for this novel technology is 48% lower than that of the traditional WOPCBs treatment and disposal

methods (\$126.31 vs. \$242.85 USD) [3]. Furthermore, the WOPCBs can be sold as steel products for steel scrap substitutes and earn an extra profit (about \$230 USD t⁻¹ WOPCBs obtained as end product). Therefore, we can conclude that this proposed novel technology is promising from an economic perspective.

4 Conclusions

The novel technology of co-disposal of WOPCBs by converter steelmaking carried out for this study is proven to be successful and feasible. The industrial trial results indicate that co-disposal of WOPCBs by converter steelmaking is more efficient than the traditional chemical cleaning process, and reduce treatment costs per ton by about 48%. With the addition of 180–540 kg of WOPCB per furnace load, both the steel product quality and the production process are proven to be safe and controllable. This technology is economical and environmental friendly and can solve WOPCBs disposal problems, and recover WOPCBs as steel scrap substitutes. Flue gas, wastewater generated in this process after treatment can meet related standards of China, and the leachability of heavy metals in metallurgical solid wastes is far lower than the standard threshold. Therefore, this novel technology can be a total solution for the emerging WOPCBs disposal problem with resource utilization, environmental protection and security concern. Meanwhile, due to the lack of information on WOPCBs disposal in the literatures, this study can be used as a reference for future research work on this topic. Further work is necessary to address more detailed study for the dioxin distribution characteristics in flue gas along the purification system and the migration and diffusion mechanism of heavy metals in this process.

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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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Availability of data and materials

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

Declarations

Competing interests

The authors declare they have no competing interests.

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