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Review

Recycling of waste printed circuit boards: A review of current technologies and treatment status in China

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ABSTRACT

From the use of renewable resources and environmental protection viewpoints, recycling of waste printed circuit boards (PCBs) receives wide concerns as the amounts of scrap PCBs increases dramatically. However, treatment for waste PCBs is a challenge due to the fact that PCBs are diverse and complex in terms of materials and components makeup as well as the original equipment's manufacturing processes. Recycle technology for waste PCBs in China is still immature. Previous studies focused on metals recovery, but resource utilization for nonmetals and further separation of the mixed metals are relatively fewer. Therefore, it is urgent to develop a proper recycle technology for waste PCBs. In this paper, current status of waste PCBs treatment in China was introduced, and several recycle technologies were analyzed. Some advices against the existing problems during recycling process were presented. Based on circular economy concept in China and complete recycling and resource utilization for all materials, a new environmental-friendly integrated recycling process with no pollution and high efficiency for waste PCBs was provided and discussed in detail.

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1. Introduction

With the development of science and technology, the market demand for production of electric and electronic equipment (EEE) is increasing rapidly. EEE has become a necessary part of people's life. Printed circuit boards (PCBs), an essential part of almost all EEE, are widely subsistent in EEE. In recent years, the average rate of world-wide PCBs manufacture increases by 8.7%, and this number is much higher in Southeast Asia (10.8%) and mainland China (14.4%). In mainland China, the total production value of the PCBs manufacturing industry has already reached more than $10.83 billion in 2005, only next to Japan, and would reach more than $12 billion in 2006 by anticipating yields [1–2]. Meanwhile, both technological innovation and intense marketing continue to accelerate the update rate of EEE and shorten the average lifespan of EEE. As a result, the amounts of waste PCBs are dramatically increasing. The UN Environment Programme estimates that the world generates 20–50 million tones of waste electric and electronic equipment (WEEE) each year and amounts are rising three times faster than other forms of municipal waste [3]. Waste

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Fig. 1. Waste PCBs from all kinds of electronic equipments.

PCBs are from all kinds of electronic equipments as shown in Fig. 1.

A new environmental challenge is presented by waste PCBs, which contain plenty of toxic substances, such as brominated flame retardants (BFR), PVC plastic and heavy metals. They can cause serious environmental problems if not proper disposal. If they are discarded randomly in the opening or landfilled simply, the leachate may infiltrate into groundwater and soil. Uncontrollable incineration of waste PCBs also produces potentially hazardous byproducts (including mainly dioxins, furans, polybrominated organic pollutants and polycyclic aromatic hydrocarbons) caused by burning BFR, epoxy resins and plastics. The materials containing BFR are precursors to polybrominated dibenzo-*p*-dioxins and dibenzofurans (PBDD/Fs). These are classified as persistent organic pollutants (POPs) under the Stockholm Convention, a global treaty drawn up to protect human health and the environment [4–6]. Growing attention has been given to hazardous components in waste PCBs, which pose a severe threat to human health (inducing people's nervous system diseases or immune system diseases) and the sustainable economic growth as well.

Recycling of waste PCBs is an important subject not only from the treatment for waste but also from the recovery of valuable materials. In general, waste PCBs contain approximately 30% metals and 70% nonmetals. The typical metals in PCBs consist of copper (20%), iron (8%), tin (4%), nickel (2%), lead (2%), zinc (1%), silver (0.2%), gold (0.1%), and palladium (0.005%). The purity of precious metals in waste PCBs is more than 10 times higher than that of rich-content minerals [7–10]. So waste PCBs are considered as an "urban mineral resources". In addition, the nonmetal portions of PCBs consist of thermoset resins and reinforcing materials. They also can be reused as fillers in composite materials. Therefore, on the basis of the current situation of resources in China, all materials in waste PCBs are a kind of resources and needed to be recycled by a proper technology. Resource utilization of waste PCBs can protect environment and alleviate bottleneck for economic development constrained by resources shortage in China.

In this paper, current status of waste PCBs treatment in China was introduced, and several recycle technologies were analyzed. Some advices against the existing problems during recycling process were presented. Based on circular economy concept in China and complete recycling and resource utilization for all materials, a new environmental-friendly integrated recycling process with no pollution and high efficiency for waste PCBs was provided and discussed in detail.

2. Status of waste PCBs in China

China is now facing dual pressure of WEEE treatment both from domestic generation and illegal transboundary movement. The amounts of WEEE in China are increasing at higher rate of 5–10% annually. Additionally some developed and industrialized countries export considerable quantities of WEEE to China, even though the Basel Convention restricts transboundary trade of it. Fig. 2 indicates the main WEEE traffic routes in Asia. There are, however, no confirmed figures available on how substantial these transboundary WEEE streams are. From nonratifying countries, such as the USA, estimates have been made that 50–80% of the collected domestic WEEE is not recycled domestically but rather shipped to destinations such as China [11]. There were about 12.75 million retired computers in the USA in 2002, of which there were 10.2 million retired computers exported to Asia. According to an estimation of a research project, approximately 80% world-wide WEEE is transferred to Asia, of which about 90% is discarded in China [12–13]. So China has become the biggest dumping ground of WEEE, accommodating more than 70% WEEE all over the world annually.

3. The problems of current technologies for waste PCBs in China

There are huge amounts of illegal transboundary movements of WEEE along coast of southeastern China, such as Guangdong province and Zhejiang province. In April of 2004, China Central Television (CCTV) reported about environmental pollution from WEEE recycling in Guiyu, Guangdong province [14]. The report aroused the attention of government and strong reaction of the public. According to the report, the occurrence of recycling of WEEE by environmentally unfriendly, hazardous and primitive technologies in this village has increased over the past years. Massive amounts of dumping of imported computer waste took place along the riverways in Guiyu. Local residents adopted rude and rudimentary methods (such as the open dumping and burning, acid washing, etc.) to WEEE treatment as shown in Fig. 3. Such activities posed grave environmental and health hazards. For example, the deterioration of local drinking water can result in serious illnesses. The Basel Action Network (BAN) investigator took a river water sample from the Lianjiang river near this "recycling village". The sample revealed lead levels that were 2400 times higher than World Health Organization Drinking Water Guidelines. Sediment samples there showed lead levels 212 times higher than what would be treated as hazardous waste [13].

Open burning of waste PCBs is commonly used for the recovery of metals such as copper and iron as shown in Fig. 4. Dioxins and furans can be generated due to the combustion of BFR in the

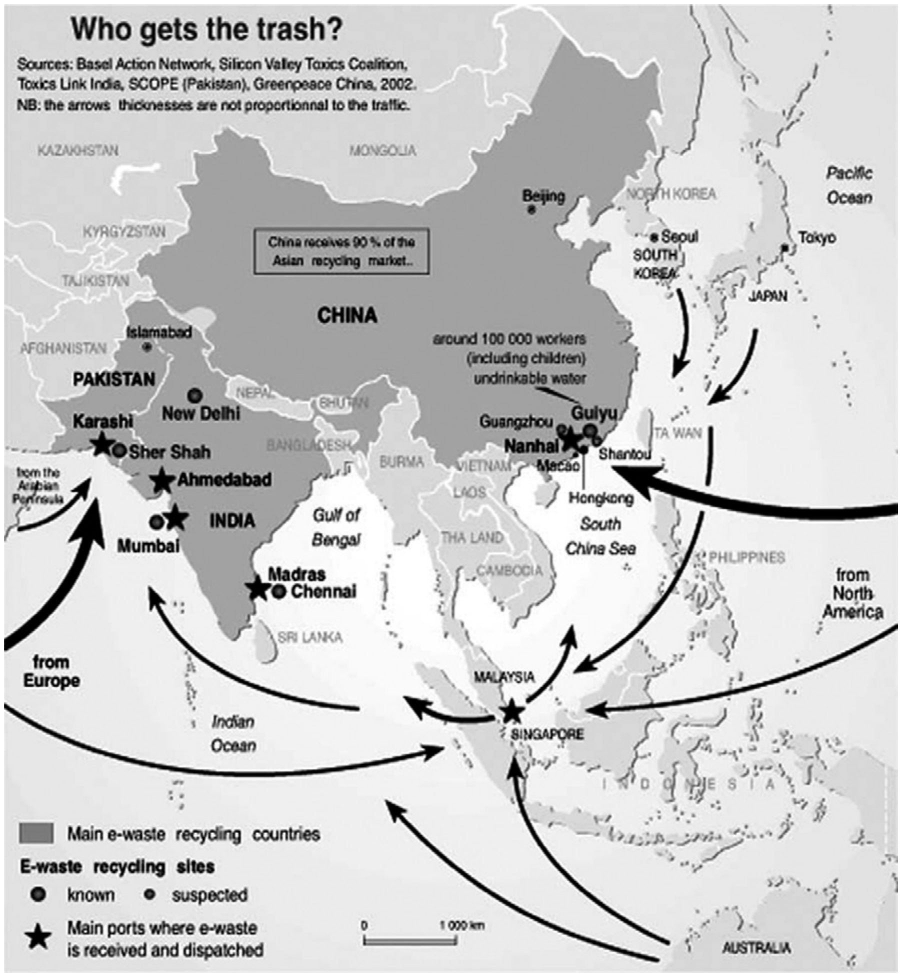


Fig. 2. Asia WEEE traffic routes [12].

PCBs. According to a report from Hong Kong Baptist University, the combustion of WEEE in the presence of copper from PCBs could be a major component of the global dioxin inventory and may lead to higher emissions of PCDD/Fs into the environment. The researchers found that polybrominated diphenyl ethers (PBDEs) levels in combustion residues from open burning in Guiyu were some of the highest ones found in any environmental medium (33 000–97 400 ng/g), more than 16 000 times higher than those found in soil samples in a distant reservoir that served as a control site [15].

A mixture of nitric acid and hydrochloric acid is also used for the recovery of precious metals from waste PCBs. A great deal of waste gases and acid solutions were produced in this process of "recycling" as shown in Fig. 5. Wastes were generated by every stage of the recycling process, and environment was contaminated with toxic heavy metals and POPs [13]. Furthermore, the untreated wastes were exposed to wind and rain, posing a serious threat to local water, air environment and human health. High levels of heavy metals were also found in soils from an acid-leaching site. In addition, workers engaging in waste PCBs recycling operations face dangerous working conditions, as they may be without protection (no masks or gloves) as shown in Fig. 6. Released gases, acid solutions, toxic smoke and contaminated ashes are the most dangerous threats for such people. According to a report from State Key Laboratory of Organic Geochemistry in China, severe PCDD/Fs, PBDD/Fs, PBDEs and organochlorine pesticides (OCPs) pollution in air were investigated in Guiyu. In this WEEE dismantling area, atmospheric PCDD/Fs abundances and toxic equivalent values were 64.9–2365 pg/m^3 and 0.909–48.9 pg of W–TEQ/m^3 respectively. These were the highest documented values of these compounds found in ambient air in the world. The total PCDD/Fs intake doses far exceeded the World Health Organization (WHO) 1998 tolerable daily intake limit of 1–4 pg of W–TEQ k/g d [16–17]. To make matters worse, the severe dioxin pollution present in Guiyu substantially influenced the adjacent area of Chendian, where atmospheric PCDD/F and 2,3,7,8-PBDD/F levels were higher than those of common urban areas in the world. Local residents were also exposed to these toxic pollutants through inhalation, dermal exposure, and oral intake [16–17].

Recycling of waste PCBs is a challenge due to the fact that PCBs are diverse and complex in terms of the type, size and shape of materials and components. In China, immature technologies are the main obstacle to recycling of waste PCBs comparing with developed countries. In fact, fluid bed separation was widely used for recycling of waste PCBs by private enterprises due to lack of effective technologies in China (Fig. 7). The main problems of this process are as follows: (1) Huge amounts of wastewater are generated during the process, which may contain heavy metals as shown in Fig. 8. Wastewater and residues will lead to more serious secondary pollution without proper treatment; (2) the separation efficiency for copper is low compared with other methods; (3) it is hard to recover other metals except copper, and nonmetal materials cannot be reused and recycled.

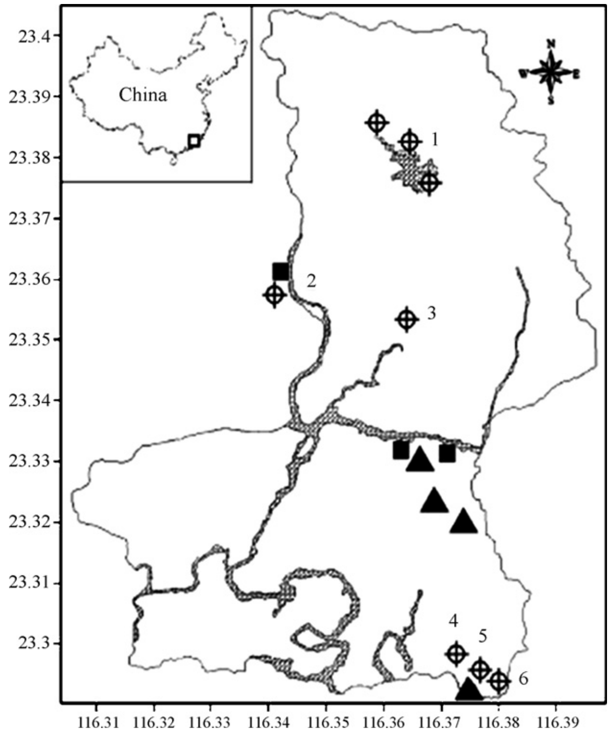


Fig. 3. Map of Guiyu and sampling locations: (circle with plus sign) sampling location [15]. ▲ WEEE dumping and open burning site and ■ acid leaching of waste PCBs site.

Fig. 5. Workers recover precious metals using acid washing.

Fig. 6. People make a living from salvaging WEEE, exposing themselves to health risks.

4. Improved integrated recycling process for waste PCBs

According to the previous analysis, it is urgent to develop a recycling technology without negative impact to the environment to resolve the problems of waste PCBs. Zhenming Xu of Shanghai Jiao Tong University in China, improved the existing recycling process and resolved the problems of secondary pollution and the reuse of materials. An integrated recycling process for waste PCBs was established by Xu [2], including two-step crushing, corona electrostatic separation, metallic materials recovery and reuse of nonmetallic materials as shown in Fig. 9.

4.1. Two-step crushing

The purpose of crushing is to strip metals from the base plates of waste PCBs. Crushing technology is intimately related to not only energy consumption of crushing equipment, but also further selective efficiency. Waste PCBs are comprised of reinforced resin and metal parts such as copper wires and joints. They have a high hardness and tenacity. In addition, most of base plates contain a

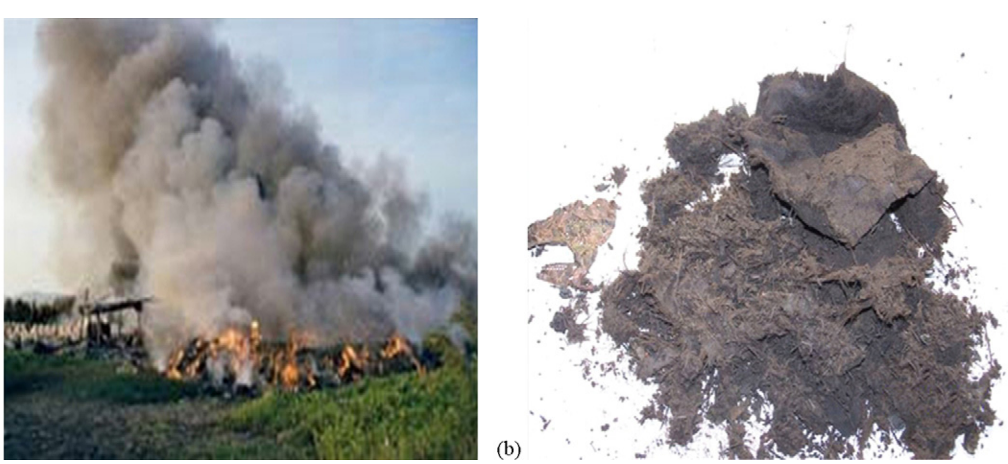
(a)  (b)

Fig. 4. Open burning of waste PCBs in Guiyu, Guangdong province. (a) Generated huge amounts of hazardous gases and (b) burning residues.

Fig. 7. Fluid bed separation in waste PCBs treatment in Kunshan, Jiangsu province.

Fig. 8. Huge amounts of wastewater generated during the process of fluid bed separation.

fiber structure (glass cloths) that is easier to break under shearing action. So the hammer grinder, whose main acting force is a shear force, is suitable for crushing waste PCBs. Additionally single crusher cannot satisfy the conditions. In order to optimize crushing effect, two-step crushing was used in their study as shown in Fig. 10 [2]. A high-speed shearing machine was used as the crude crusher. The shearing action generated by the rotor cutters and stator cutters crushed PCBs plates to small particles. A hammer grinder specially designed for waste PCBs was employed as the second crusher. The materials were stroked and milled by high-speed hammerheads. The action of hammerheads not only promoted metals to be completely stripped from base plates but also decreased the opportunity of wires wrapping around the tool tips [2]. The results indicated that the diameter of screen holes in the hammer grinder was 1 mm to warrant an excellent grade of metal stripping from the base plates. The diameter of circular velocity and rotor radius in shearing machine is 1440 rpm, 0.25 m respectively. The diameter of circular velocity and rotor radius in hammer grinder crusher is 2000 rpm, 0.2 m respectively [18].

4.2. Corona electrostatic separation

Corona electrostatic separation (CES) was an environmental-friendly (no wastewater or gas during the process) and efficient way for the recovery of metals from waste PCBs and widely studied by Xu [19–29]. CES (Fig. 11) is based on the extreme differences in density and electric conductivity of metallic and nonmetallic materials from pulverized PCBs. According to a report from the Shanghai team, effects of operating parameters of CES and properties of metallic metals (such as particle shapes and particle sizes) on

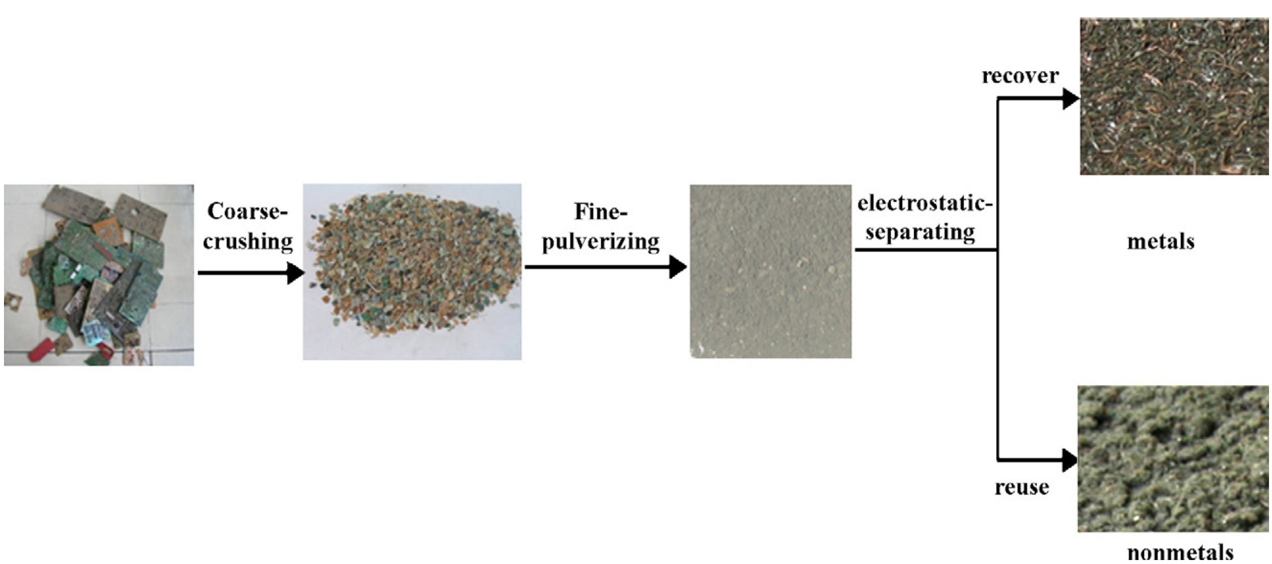


Fig. 9. Whole process of integrated recycling for waste PCBs.

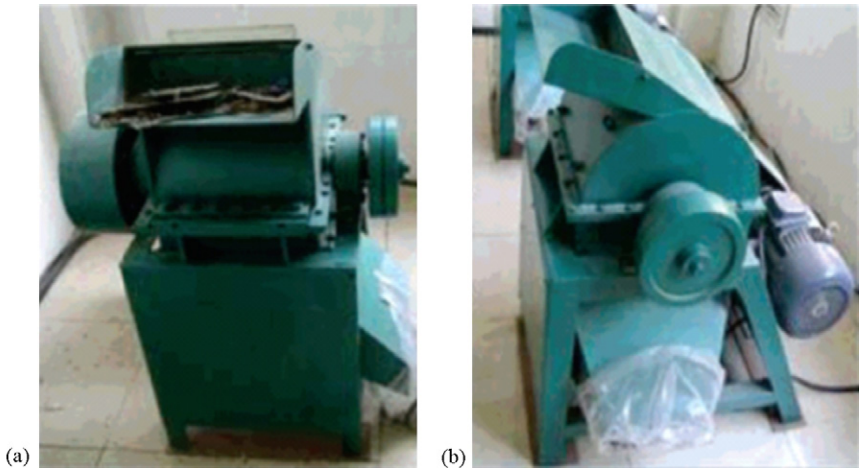


Fig. 10. Crushing equipments: (a) shearing machine and (b) hammer grinder [2].

the efficiency of CES were widely studied by Xu [19–22]. The results indicated that CES was an important technology suitable for separating metals from nonmetals and particle sizes between 0.6 and 1.2 mm were most feasible for separation in industrial application [19].

A computer program was employed for analysing the behavior of spherical metallic particles in cylinder electrostatic separators. The results revealed that the particle's motion depends on its radius and density and amplitude of the applied voltage [20]. From both a computational and an experimental point of view, particle shape factor (sphere, cylinder and flake) affected the efficiency of CES and movement behavior of three shape particles was distinguishing: the sphere particles could be projected farther, subsequently cylinder particles, flake could be projected closer [21].

The computer simulation of electric field was for optimizing the process of recycling waste PCBs by CES [23] and some models were established [24–26]. The software MATLAB was used to simulate the distribution of electric field in separating space as shown in Fig. 12. It was found that the variations of parameters of electrodes and applied voltages directly influenced the distribution of electric field. Through the correlation of simulated and experimental results, the good separation results were got under the optimized operating parameter: $U = 20–30$ kV, $L = L_1 = L_2 = 0.21$ m, $R_1 = 0.114$, $R_2 = 0.019$ m, $\theta_1 = 20^\circ$ and $\theta_2 = 60^\circ$ [23]. The models guided the definition of operator parameter and offered a possible for designing of CES [24–25]. Furthermore, theoretic model for computer simulation of separating metallic particles mixture could be used to simulate separating other metallic (tin, zinc, etc.) particles during the process of recycling waste PCBs by CES [26].

Regarding to the limitations of the one-roll CES, such as the impurity of nonconductive products and the stability of the separation process, a laboratory two-roll CES was built as shown in Fig. 13 [28–29]. Compared with one-roll CES, the conductive products increase 8.9%, the middling products decrease 45% and the production capacity increases 50% in treating comminuted PCBs wastes by two-roll CES. In addition, the separation process in two-roll CES is more stable. Therefore, two-roll CES is a promising separator for recycling comminuted PCBs wastes.

4.3. Metallic materials recovery

In developing new technology for waste PCBs, most researchers have focused on the technology by which the valuable metals in PCBs can be separated and recovered. The major economic driver for recycling of waste PCBs has been from the recovery of metals for a long time. Initially, the simple incineration (uncontrollable incineration, open burning, etc.) was adopted to recover metals from waste PCBs. As this process lacked of effective environmental pro-

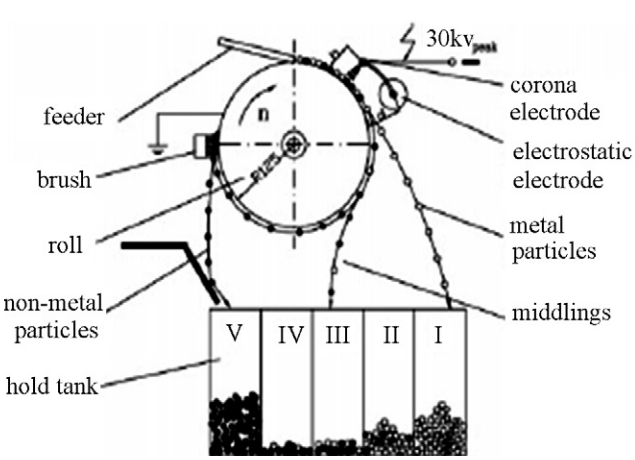


Fig. 11. Diagram of CES [19].

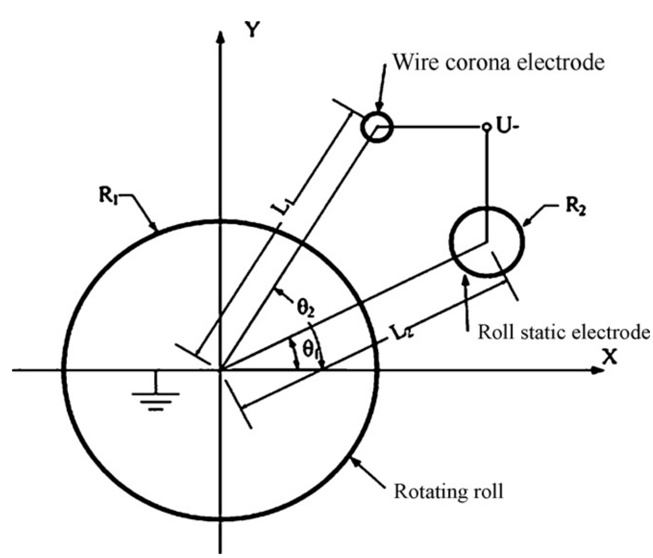


Fig. 12. The geometric model used in electrostatic field simulation.

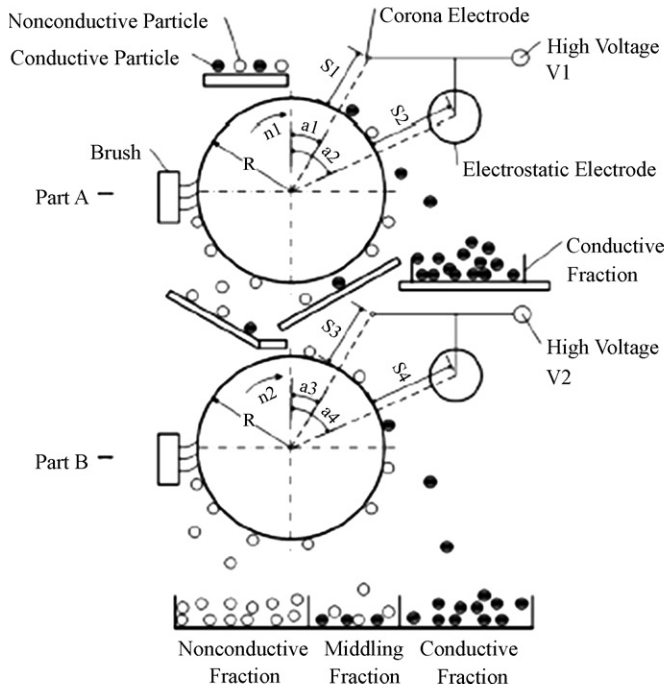


Fig. 13. Schematic representation of the two-roll CES.

tection and posed a threat to human health, it was banned in China. Consequently, pyrometallurgy and pyrolysis were developed based on this thermal process.

Pyrometallurgy is a traditional technology for recovery of non-ferrous metals as well as precious metals from waste PCBs. Pyrometallurgy included incineration, smelting in a plasma arc furnace or blast furnace, drossing, sintering, melting and reactions in a gas phase at high temperatures [30]. There were some applications to recover metals from WEEE by pyrometallurgy, such as at the Boliden Ltd. Rönnskär Smelter in Sweden [9,31–32] and the Noranda process at Quebec in Canada [33]. However, state-of-the-art smelters are highly depended on equipments investments and waste air processing. To our knowledge, large-scale recovery of metals from waste PCBs by pyrometallurgy do not appear in China.

Pyrolysis is the chemical decomposition of organic materials by heating in the absence of oxygen or any other reagents. Pyrolysis of organic materials contained in waste PCBs leads to the formation of gases, oils, and chars which can be used as chemical feedstocks or fuels [6]. At present, there are some pilot-scale studies on the recovery of metals from waste PCBs by pyrolysis in China. According to a report from Huazhong University of Science and Technology in China, pyrolysis was adopted to recover valuable materials from waste PCBs. The results indicated that liquid yield of 15–21%, gas yield of 15–20% and solid yield of about 60% were obtained in the nitrogen atmosphere as shown in Fig. 14 [34–36]. The liquid products had high gross calorific values that might be recycled as fuel oils after simple treatment. However, pyrolysis is also to be highly dependent on equipments investments and the residues are a mixture of organic and various metals which need to be further separated. Therefore further studies on the commercially viable option and separation of mixed metals should be developed.

Hydrometallurgy is another traditional technology for the recovery of precious metals from waste PCBs. The main steps in hydrometallurgy consist of a series of acid or caustic leaches (cyanide leaching, halide leaching, thiourea leaching, and thiosulfate leaching, etc.) of solid materials. The solutions are then subjected to separation and purification procedures such as precipitation of impurities, solvent extraction, adsorption and ion-exchange to isolate and concentrate the metals of interest. Consequently, the solutions are treated by electrorefining process, chemical reduction, or crystallization for metal recovery [30]. In fact, the domestic universities and research institutes have done some pilot-scale research on the treatment for waste PCBs by hydrometallurgy in China. According to a report from Donghua University in China, twice oxidizing-acid leaching pretreatment was adopted to recover the base metals in waste PCBs, and then acid thiourea leaching and Zn/Fe exchange were used for the separation of gold and silver [37]. The result showed that twice oxidizing-acid leaching pretreatment was less hazardous air pollutants of nitrogen oxides than traditional oxidizing-acid leaching one. The leaching rate of copper, gold and silver is 97.6%, 95.1%, 80.5% respectively [37]. Renping Cao of South China University of Technology in China adopted calcine-leaching technology to recover gold, palladium and silver from the waste PCBs which contained in the mobile phones. The result showed that the recovery rate of gold, palladium and silver was over 95% respectively [38].

Mechanical-physical recycling process for waste PCBs is based on the differences of materials in physical characteristics (including density, magnetic susceptibilities, electric conductivity, etc.). Due to its better environmental property (such as less wastewa-

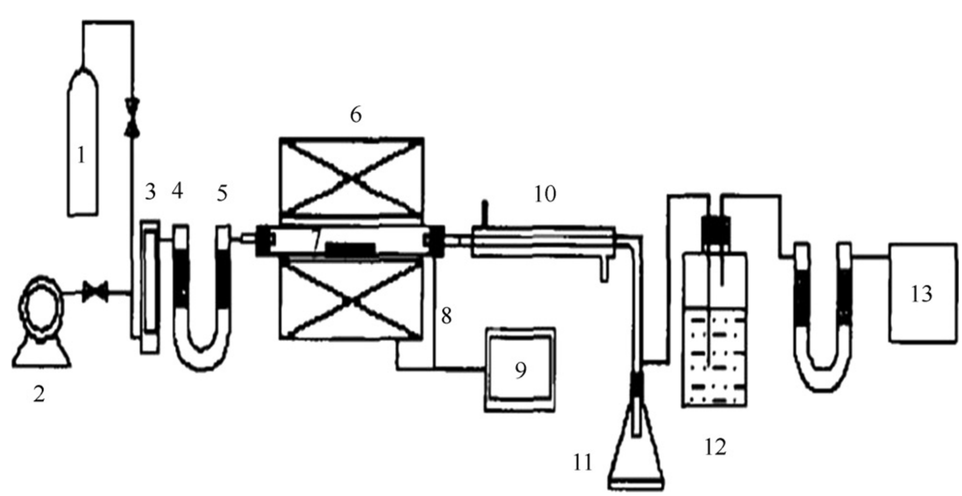


Fig. 14. Experimental apparatus for pyrolysis [34]. (1) Nitrogen cylinder, (2) air pump, (3) gas flow meter, (4) drying tube, (5) quartz tube, (6) electric furnace, (7) quartz board, (8) thermocouple, (9) temperature controller, (10) cooling tube, (11) collecting flask, (12) alkaline solution, and (13) gas bag.

Fig. 15. The vacuum distilling equipment.

ter), high efficiency and easier operability, additionally non-ferrous metals and precious metals contents have gradually decreased in concentration in PCBs. Mechanical-physical recycling process is drawing more attention by the researchers in recent year in China [2,18–29,39–41]. In the above-integrated recycling process for waste PCBs, the materials coming out of separators are metallic and nonmetallic. There are about 30 wt.% metallic materials after separation. These metallic materials are hard to recover, because the fractions concentrated on metallic materials obtained from these processes are still a mixture of various metals (copper, aluminum, lead, zinc, etc.). Until now, there are not proper methods to separate the various metals or recover them yet. The existing mechanical technologies (pneumatic separation, gravity concentration, etc.) focus on recover the copper, but the studies on further separation of the mixed metals are relatively fewer in China [39–41]. In order to further separate the concentrated fraction in metals and increase the copper content in the metallic mixture, vacuum metallurgy separation method (VMS) was presented in their studies [42–46] as shown in Fig. 15.

The principle of separating pure metals from mixed metallic particles by vacuum metallurgy is that the vapor pressures of various metals at the same temperature are different. As a result, the metal with high vapor pressure and low boiling point can be separated from the mixed metals through distillation or sublimation, and then it can be recycled through condensation under a certain condition. Therefore VMS can recover metals with high vapor pressure such as lead and cadmium, which would be wasted during smelting. Furthermore, VMS will reduce pollution dramatically and prevent formation of oxides because VMS processes are under vacuum in which there is no atmosphere engaging. However, VMS can only recover metals with high vapor pressure; other metals such as precious metals cannot be recovered efficiently. In order to recover all metallic materials in waste PCBs, VMS should be combined with other recycle technologies (hydrometallurgy, biometallurgy, etc.).

Biometallurgy has been used for recovery of precious metals and copper from ore for many years [47–48], but biometallurgy used for recycling waste PCBs is still in its infancy in China. There are two main areas of biometallurgy for metals recovery, namely bioleaching and biosorption. Compared with other methods, biometallurgy offers a number of advantages including low operating costs, less pollution, minimization of the volume of chemical and/or biological sludge to be handled and high efficiency in detoxifying effluents [30]. However, the known bacterium which is suitable for the treatment for waste PCBs is seldom and hard to culture, and the cycle for biometallurgy in recovery of precious metals is too long. Therefore, further work should be done for seeking and modifying a biomass to have a high uptake capacity and good biosorption characteristics. Further studies on the VMS and biometallurgy are being investigated by Xu [42–46].

4.4. *Resource utilization of nonmetallic materials*

There are also about 70 wt.% nonmetallic materials after separatiion. The nonmetallic materials of PCBs mainly consist of thermoset resins and glass fibers. Thermoset resins cannot be remelted or reformed because of their network structure. Incineration is not the best method for treating nonmetallic materials because of inorganic fillers such as glass fiber which significantly reduces the fuel efficiency. Disposal in landfill is the main method for treating nonmetallic materials of PCBs, but it may cause secondary pollution and resource-wasting. In order to take full advantage of nonmetallic materials of waste PCBs, nonmetallic materials were used to produce modified phenolic molding compound (PMC) [7,49] and nonmetallic plate (NMP), achieving complete recovery of reusable resources.

Nonmetallic materials were used to replace wood flour in the production of modified PMC as shown in Fig. 16. This process can protect timber resources and reduce the cost of PMC. Table 1 shows effects of contents of nonmetallic materials on properties of PMC. The results indicated that filling of nonmetallic materials in PMC improved the charpy notched impact strength and heat deflection temperature (HDT), reduce flexural strength and rasching fluidity. Rasching fluidity reduces dramatically with the increase of the content of nonmetallic materials [49].

A schematic illustration of preparation of NMP was shown in Fig. 17. The preparation of NMP was divided into two stages: the first stage was the preparation of nonmetallic dough. Nonmetallic dough was produced in the double Z-kneader. Different components such as nonmetallic materials, glass fibers and $CaCO_3$ can be well coated with liquid resin paste through the shear force of Z-kneader; the second stage was molding process of the NMP. Nonmetallic dough was molded into NMP in the pressurized and heated conditions through the crosslinking of unpolymerized resin

Fig. 16. Schematic illustration of nonmetallic materials of waste PCBs filling in the PMC [7,49].

Table 1
Effects of contents of nonmetallic materials on properties of PMC [49]

Properties	Standard	0	10%	20%	30%	40%
Relative density	≤1.45	1.40	1.40	1.39	1.39	1.38
Impact strength (notched, kJ/m^2)	≥1.5	2.1	2.4	2.3	2.4	2.7
HDT (°C)	≥140	154	157	168	164	169
Flexural strength (MPa)	≥70	71	64	70	70	71
Dielectric strength (90 °C, mV/m)	≥3.5	3.9	3.3	3.9	3.9	5.0
Rasching fluidity (mm)	≥100	135	132	103	75	62

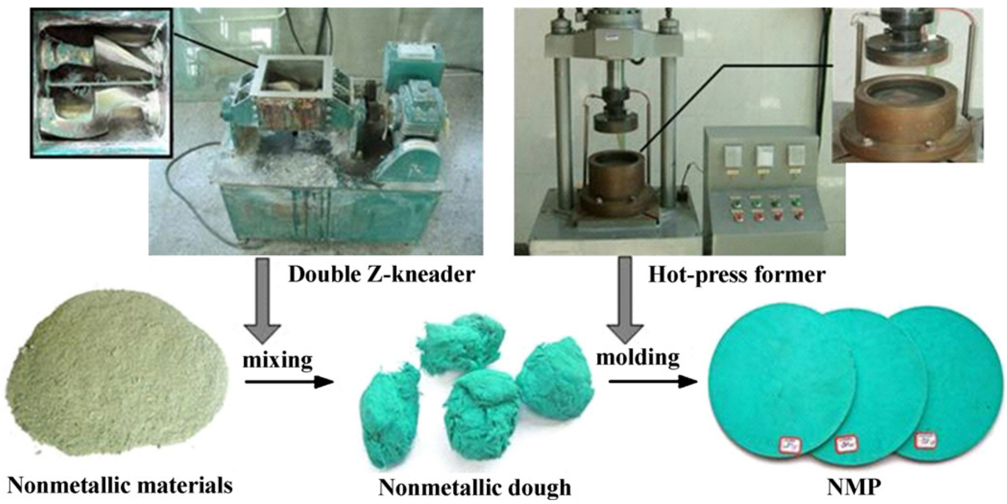


Fig. 17. Schematic illustration of production of NMP.

with various additives using a self-made hot-press former [50]. Nonmetallic materials of waste PCBs in NMP may attain 40 wt.%. NMP could be used as building materials (tiles, partitions, insulating boards, etc.). This novel technique offers another method for recycling of nonmetallic materials of PCBs.

5. Conclusion

Recycling of waste PCBs is an important subject not only from the treatment of waste but also from the recovery of valuable materials. In China, in order to accelerate the pace of waste PCBs treatment, it is vital to introduce and develop cost-effective and environmentally friendly recycling technologies for waste PCBs. The improved integrated recycling process is a new technology without negative impact to the environment. It is also in accordance with the principle of the circular economy in China and provides a new method for recycling of waste PCBs.

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