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MAGNETIC SEPARATION OF ELECTRONIC WASTE AFTER THE COMBUSTION PROCESS IN THE FLUIDIZED BED

SEPARACJA MAGNETYCZNA ODPADÓW ELEKTRONICZNYCH PO PROCESIE SPALANIA W ZŁOŻU FLUIDALNYM

Abstract: The paper presents the results of magnetic separation of materials received after the thermal utilization process of mobile phones in a laboratory fluidized bed reactor. The starting material constituted ten mobile phones which were subjected to the combustion process receiving brittle, solid products. Next, the received materials were grinded to 1 and to 0.5 mm and after the magnetic separation was conducted using neodymium magnet, plate separator (three-phase) and disk (belt) separator. The received waste fractions were subjected to the analyze of content phases (XRD) and chosen chemical elements (ICP).

Keywords: WEEE, printed circuit board, magnetic separation

Introduction

Both in Europe and the world, there is a constant upward trend in the amounts of waste electrical and electronic equipment (WEEE). Each year in the European Union alone, approximately 8 million tons of electronic waste is produced, and the rate of such production increases annually by 3-5%. Every year, 20-50 million tons of this hazardous waste appear worldwide [1].

The most increasing tendency among WEEE constitute the information and telecommunication equipment such as mobile phones and personal computers due to the much shorter liveliness. These kind of equipment is exchanged almost every 2-5 years. At present it is estimated that every year about 17 mln of computers are thrown away because of improper work of these equipment, old technology or willing to possess equipment that is more functional. It is predicted that in 2020, the quantity of waste cell phones will increase sevenfold [2, 3].

Mobile phones, especially those of older generation, pose a significant risk to the environment due to the fact that they contain such elements as lead, cadmium or mercury. A printed circuit board with its electronic components alone contains numerous harmful elements, including noble metals (Au, Ag, Pt, Cu), coloured metals (Sn, Ni, Cr, Zn) as well as iron and silicon [4]. Therefore, unprocessed electronic waste which is stored together with domestic waste in landfills is a serious threat to human health and the environment, as it may lead to water and soil contamination and consequently to the contamination of agricultural produce as well [5].

In order to properly manage hazardous waste like electrical and electronic equipment, the European Union introduced the Directive 2002/96/EC of the European Parliament and of the Council of 27 January 2003, whose main purpose is to reduce the quantity of WEEE by proper reusing and recycling [6].

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In this paper the magnetic separation process of solid products received after combustion of mobile phones fragments in laboratory fluidized bed reactor were conducted. The received solid products were subjected to the grinding process to 1 and 0.5 mm. The neodymium magnet, plate separator, and disk separator were used for magnetic separation process. The received waste fractions were analyzed for particular phases by X-ray diffraction powder (XRD) and chosen chemical elements (ICP).

Experimental

In order to obtain material for the magnetic separation tests combustion process of mobile phones was conducted in the laboratory fluidized bed reactor which was previously used for the studies on selected aspects of fuel and waste burning [7, 8]. The process was proceeded in a hot fluidized bed, which constituted of glass sand with the grain size of 0.3-0.385 mm and weight of 413.6 g. The fluidized bed formed the environment with a homogeneous high temperature and considerable turbulence of the solid phase and gas phase, providing constant access of the oxidizer to the surface of the introduced samples. This access was necessary for the proper course of thermal treatment of the combustible components contained within the samples. Combustion was conducted with the use of oxygen from the air and propane-butane as auxiliary fuel assuring constant, high temperature.

As the starting material ten mobile phones were used. Initially, the phones were mechanically disassembled, the batteries were separated and the resulting material was divided into smaller parts with a guillotine. Next, the combustion process was conducted leading to receive brittle, solid products which were used to the magnetic separation. Solid products after combustion in the fluidized bed were crumbed and grinded. This way two materials with the grain size below 1 mm and more than 1 mm were received. Both materials were conducted the separation process, this over 1 mm was separated using the neodymium magnet. The sample below 1 mm was given to two-stage separation using plate and belt separator. After the separation processes of material with grain size below 1 mm was divided as two magnetic and one non-magnetic fractions. The last one was grinded to about 0.5 mm and the separation process on the belt separator was conducted again.

The first device used for the magnetic enrichment was plate separator with magnetic pole generated by alternating electric current. It is built with the flat plate made of transformer metal plates. In the trenches of plate there are windings connected to the three-phase electrical source (AC) producing three independent magnetic streams (similar to the stator of three-phase engine). Magnetic field which changes in time and space has waving character and moves along the plate with definite speed. The advantage of this separator is the opportunity of separating grids with strong magnetic properties due to the changing magnetic field.

The second used device was disk separator to which the amount of raw material is given through the hopper on the conveyor belt rolled on the drum and it leads material into the slit under the plate that is turning around. The magnetic ingredient is caught by the turning around plate and is drained outside the operation range of magnetic field where it mechanically comes off on the sharp edge putted to the plate. The non-magnetic ingredients come under the surface of plate and it is drained on the moving conveyor belt to the

container of non-magnetic product. The operating slit of separator constitutes magnetic system wedge-surface. Inside the plate there are installed magnetic pole pieces producing magnetic field [9].

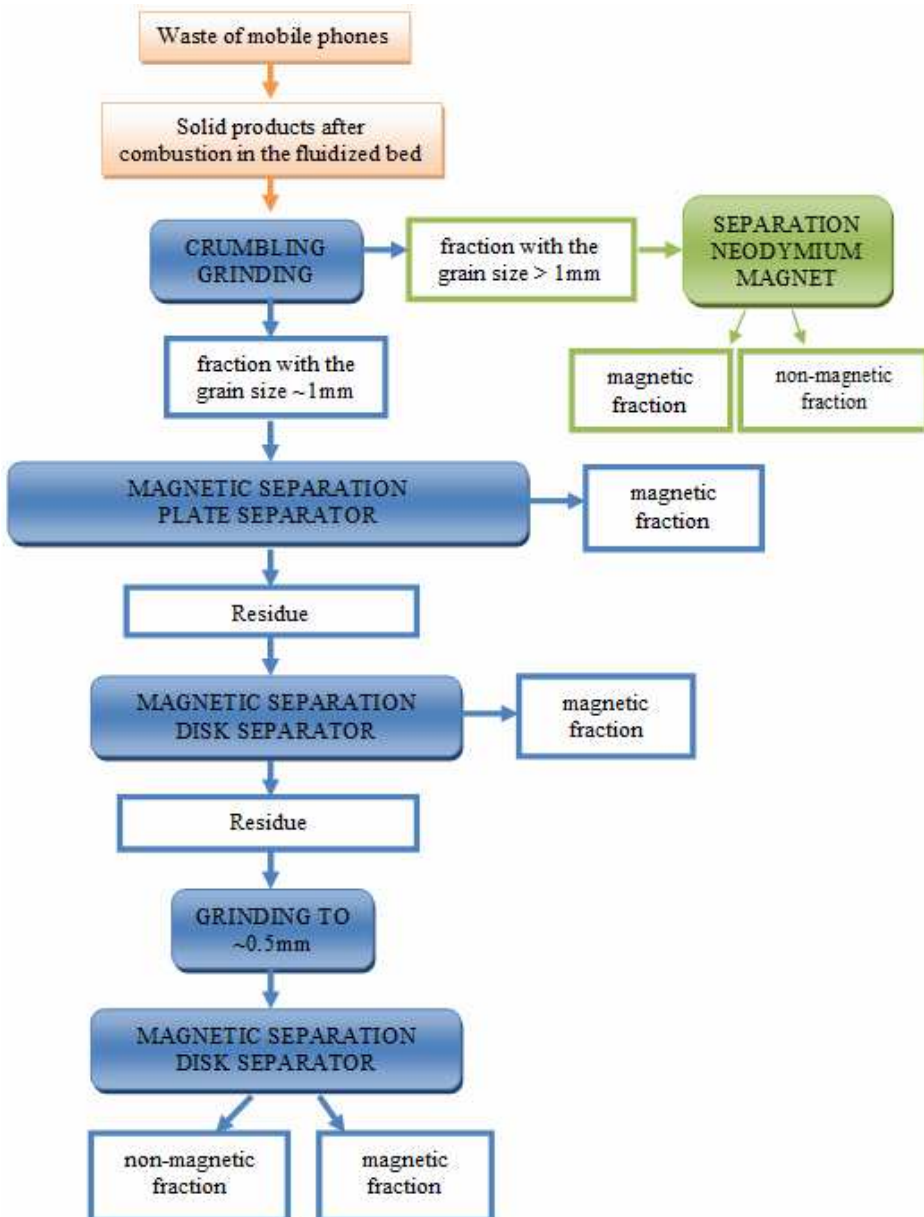


Fig. 1. The scheme of magnetic enrichment process of solid products after the combustion of used mobile phones









In Figure 1 there is presented the whole scheme of magnetic enrichment of solid products after the combustion process of used mobile phones.

Results and discussion

Few-stages process of magnetic enrichment allowed to receive particular fraction with various masses as presented in the Table 1. From the material with the grain size of < 1 mm after separation process on the plate and belt separator received three magnetic fractions with the weight of 44.9 g and non-magnetic fraction with the weight of 207.7 g.

Table 1

Received fractions after magnetic enrichment processes

No.	Separation	Fraction	Grain size [mm]	Mass [g]	Picture
1	The sample after crumbling and separating on sieves		> 1	70.4	
1a	Neodymium magnet	Magnetic		27.2	
1b		Non-magnetic		43.2	
2	The sample after crumbling and separating on sieves		~ 1	263.2	
2a	Plate separator	Magnetic		2.5	
2b	Disk separator	Magnetic		19.8	
2c	Disk separator	Magnetic	< 0.5	22.6	
		Non-magnetic		207.7	

Moreover, despite of repetition of crumbling and grinding processes of the initial material the fraction with the grain size > 1 mm was also received which mass was 70.4 g. This coarse fraction after the separation process with the use of neodymium magnet divided to the magnetic material with the mass of 27.2 g and non-magnetic material with the mass 43.2 g.

Table 2

The contents of selected chemical elements and their separation degree in each fraction

No.	Element	Element content in fraction [mg]					Separation degree in fraction [%]			
		0	I	II	III	IV	I	II	III	IV
1	Cu	44846	77.6	3010	4048	37710	0.17	6.71	9.03	84.1
2	P	13022	18.1	851	1146	11008	0.14	6.53	8.80	84.5
3	Fe	3139	1425	1416	135	164	45.4	45.1	4.29	5.23
5	Ni	2215	285	992	179	759	12.9	44.8	8.09	34.2
6	Ba	2021	75.7	1025	173	747	3.75	50.7	8.54	37.0
7	Al	1719	0.001	230	256	1233	0.00	13.4	14.9	71.7
8	Pb	1190	6.96	115	139	929	0.59	9.70	11.7	78.0
9	Sn	876	3.23	32.2	1.18	839	0.37	3.68	0.13	95.8
10	Zn	686	74.0	134	84.2	394	10.8	19.5	12.3	57.4
11	Nd	506	338	159	5.22	3.99	66.8	31.4	1.03	0.79
12	Ir	447	77.8	244	32.8	92.4	17.4	54.5	7.35	20.7
13	B	344	13.0	25.1	41.8	264	3.78	7.30	12.2	76.8
14	Au	188	0.876	15.3	7.19	164	0.47	8.16	3.83	87.5
15	Sr	82.3	3.63	35.9	6.28	36.57	4.41	43.5	7.62	44.4
16	Cr	81.4	20.9	49.1	3.25	8.09	25.7	60.4	3.99	9.94
17	Mn	67.5	4.19	51.8	4.45	7.05	6.22	76.7	6.59	10.4
18	Ti	45.7	7.94	8.15	3.82	25.8	17.4	17.8	8.37	56.4
19	La	26.8	0.001	22.4	0.732	3.72	0.00	83.4	2.73	13.9
20	Co	26.7	16.9	4.71	1.29	3.80	63.3	17.6	4.82	14.2
21	Ag	11.6	0.179	1.51	0.470	9.47	1.54	13.0	4.04	81.5

0 - initial material received after combustion of mobile phones

I - magnetic fraction (1 mm) after enrichment plate separator

II, III - magnetic fraction 1 mm (II), 0.5 mm (III) after enrichment on the disk separator

IV - non-magnetic fraction (0.5 mm) after enrichment on the disk separator

Table 3

Crystalline phases in particular fractions determined by the x-ray diffraction

Fraction	Crystalline phase identified				
	Si	Cu	Fe	Sn	Others
I Magnetic 1 mm Plate separator	-	-	Fe ₁₄ Nd ₂ B - 31.4% Fe ₃ O ₄ - 25.7%	SnO ₂ - 5.7%	Ni - 37.2%
II Magnetic 1 mm Disk separator	SiO ₂ - 62.3%	Cu ₂ O - 8.9% CuO - 4.8% Cu - 5.6%	CuFe - 3%	SnO ₃ Sr - 2.7% SnO ₂ - 2.2%	BaTiO ₃ - 10.6%
III Magnetic 0.5 mm Disk separator	SiO ₂ - 61%	Cu ₂ O - 25.3% Tl _{0.01} Cu _{3.99} - 6.7% CuO - 3.6% CuGaTe ₂ - 1.4%	-	SnO ₂ - 2.2%	-
IV Non-magnetic 0.5 mm Disk separator	SiO ₂ - 59.8%	Cu ₂ O - 29.4% Tl _{0.01} Cu _{3.99} - 5.1% CuO - 2.1%	-	SnO ₂ - 3.5%	-

Next, to determine the chemical and elementary composition and the separation degree of chosen elements for each fraction the analysis using the method of spectrometry mass

with the inductively coupled plasma and x-ray diffraction were conducted. The results are presented in the Tables 2 and 3.

Elementary analysis showed that the plate magnet separated the most effectively valuable rare earth element, which is neodymium. The results showed in this magnetic fraction the separation degree of 66.8% neodymium but also 63.3% cobalt and 45.4% iron.

As a result of second stage magnetic enrichment on the disk separator received the separation degree in the fraction for iron 45.1% and neodymium 31.4%. Total degree of two-stage magnetic enrichment of sample with the grain size of 1 mm was 80.9% for Co, 90.5% for Fe and 98.2% for Nd. In addition, the analyses of powder x-ray diffraction (XRD) showed total separation of neodymium as an alloy $\text{Fe}_{14}\text{Nd}_2\text{B}$, whose content in the magnetic fraction after the plate separation is 31.4% and also iron as Fe_3O_4 (25.7%) and CuFe (3%). The received results allow to conclude that the separation of such kind of materials with the grain size of 1 mm is sufficient and additional grinding to 0.5 mm is not necessary.

The received crystalline phases indicate the presence of cassiterite SnO_2 in each fraction which content is in the range from 2.2% (magnetic fractions after disk separator) to 5.7% (magnetic fraction after plate separator). Also in each fraction, apart from the magnetic after the plate separator, was shown the presence of quartz SiO_2 in the range from 59.8 to 62.3%.

The copper which was identified in the obtained fractions occurred in the metallic form Cu (5.6% in the fraction II), as oxide (cuprite Cu_2O from 8.9% in the fraction II to 29.4% in the fraction IV, tenorite CuO from 2.1% in the fraction IV to 4.8% in the fraction II), $\text{T}_{10.01}\text{Cu}_{3.99}$ in the range from 5.1% (fraction IV) to 6.7% (fraction III) and CuGaTe_2 in the magnetic fraction III (1.4%).

Non-magnetic fraction received after three-stage magnetic enrichment showed the highest separation degrees for tin 95.8%, gold 87.5%, copper 84.1% and silver 81.5%.

Conclusions

The combustion process of used mobile phones in the fluidized bed reactor allow to receive a starting material for magnetic enrichment which was a brittle material, so it was possible to grind it to the intended grain size. However, about 21% of subjected mass was not possible to grind to the expected size what was due to presence of the larger metal fragments coming out from cases of mobile phones.

Magnetic enrichment of material with the grain size with about 1 mm by plate separator (three-phase) and by disk separator allowed to receive in the magnetic fractions total separation degree 80.9% for cobalt, 90.5% for iron and 98.2% for neodymium. It can be conclude that to the achieve an effective magnetic enrichment of such type of materials the grain size of 1 mm is sufficient. As a result neodymium is separated in the form of alloy $\text{Fe}_{14}\text{Nd}_2\text{B}$ which constitutes one of the best materials used for producing permanent magnets.

Non-magnetic fraction, received as a result of magnetic enrichment, contain tin (95.8%), iron (87.5%), copper (84.1%), silver (81.5%) compared to the initial material.

The magnetic separation with the use of plate separator (three-phase) and disk separator allows the effective separation of ferromagnetic elements (Co, Fe, Nd) from non-magnetic (Cu, Sn, Au, Ag).

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Abstrakt: W artykule przedstawiono wyniki badań separacji magnetycznej materiałów uzyskanych po procesie termicznej utylizacji telefonów komórkowych w laboratoryjnym reaktorze fluidyzacyjnym. Materiał wyjściowy stanowiło 10 telefonów komórkowych, które poddano procesowi spalania, uzyskując kruche produkty stałe. Następnie otrzymane materiały zmielono do uziarnienia 1 oraz 0,5 mm oraz przeprowadzono separację magnetyczną, wykorzystując magnes neodymowy, separator płytowy (trójfazowy) oraz separator talerzowy (taśmowy). Uzyskane frakcje odpadów poddano analizie na zawartość faz (XRD) oraz wybranych pierwiastków (ICP).

Słowa kluczowe: ZSEE, obwód drukowany, separacja magnetyczna