

RESEARCH ON THERMAL DECOMPOSITION OF WASTE PE/PP

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The most important and the most frequently used plastics are polyethylene (PE) and polypropylene (PP). They are characterised with high heating values (approximately 40 MJ/kg). Moreover, their chemical composition, based mainly on carbon and hydrogen, allows to use them in industrial processes. One of the methods of utilisation of plastic waste can be its use in the metallurgical industry. This paper presents results of thermal decomposition of waste PE/PP. Chemical and thermal analysis (TG) of studied wastes was carried out. Evolved gaseous products from the decomposition of wastes were identified using mass spectrometry (TG-MS). This paper also presents an application of plastic wastes as supplemental fuel in blast furnace processes (as a substitute for coke) and as an addition in processes of coking coal.

Keywords: plastic wastes, supplemental fuel, blast furnace process, coking of coal

1. INTRODUCTION

The growth in global steel production in 2001-2008 reached unprecedented amounts (Table 1). The production of crude steel increased during this period by 497.7 million t, i.e. by 58.6%, and pig iron by 370 million t, i.e. by 64.2% (Kosewska and Merta, 2009). This increase was mainly generated by China and India. The first symptoms of the economic crisis, which emerged in the fourth quarter of 2008, forced steel manufacturers to reduce production amounts. However, it is only a matter of time before the global economy comes out from recession.

Table 1. Crude steel production and blast furnace pig iron production in the world 2001-2008, mln t (Kosewska and Mertas, 2009)

Year	2001	2002	2003	2004	2005	2006	2007	2008
Crude steel production	850.3	903.8	968.2	1054.7	1130.8	1250.0	1344.3	1329.7
Blast furnace production of pig iron	578.5	611.1	658.5	717.8	788.9	868.8	946.3	927.1

Poland produced 9.7 million tonnes of crude steel in 2008, about 9% less than in the previous year. The share of steel smelted in the Bessemer process accounted for 53.7%, and steel smelted in the electric process – 46.3%. Poland belongs to the world's leaders in terms of the production of coke, used among others in the blast furnace process. Table 2 below shows the largest producers of coke in the world.

At the same time, over the last several years there has been a steady increase in the production of plastics. They are widely applied in many areas of life: as packaging, in construction, agriculture, the automotive industry or in electronic equipment. Polyolefin, and particularly polyethylene and

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polypropylene, account for 50% of all produced plastics. Their increasing production affects the increase of the amount of waste generated and the need for its use. Besides material recycling, of which the product is a recyclate for secondary production e.g. pots and garden furniture, chemical recycling is also used - mainly for polyethylene terephthalate (PET) bottles. Material recycling is possible only after the initial sorting of waste according to the types of plastic, and after removing impurities from them. Both actions limit the profitability of material recycling of waste products. In addition, the ongoing problem of segregation of municipal waste is responsible for the fact that a vast majority of such waste is sent to a landfills, where it decomposes for even to a few hundred years. This waste can be successfully used in the metallurgical industry. Plastic wastes can find application in the blast furnace process as a substitute of coke (Kim et al., 2002; Fink, 1999) and as an additive in the coking of coal. In the process of coke production, pyrolysis processes play a major role, whereas, in the blast furnace process - combustion and gasification.

Table 2. Crude steel production and blast furnace pig iron production in the world 2001-2008, mln t (Kosewska and Mertas, 2009)

Country	2007	2008
China	328.80	322.01
Japan	38.30	39.50
Russia	31.90	30.00
India	19.40	22.50
Ukraine	19.60	19.00
USA	15.00	14.80
South Korea	10.00	11.50
Poland	10.30	9.83
Brazil	8.60	9.50
Germany	8.50	9.20

The main sources of obtaining plastic waste include (Niesler, 2001):

- municipal waste,
- unutilised post-production waste,
- recycling of cars.

In recent years, remains from the process of automobile shredding, containing not only scrap metal, but also more and more plastic, have found application in blast furnace processes (Mirabile et al., 2002). Plastic recovered from shredded automobiles is very diverse in terms of assortment (more than 20 types) and contaminated. Therefore, the possibilities of its use in material recycling are very limited. Used electronic equipment also finds application in the blast furnace process.

2. DISPOSAL OF PLASTIC WASTE

There are two methods of disposal of plastic waste in metallurgical processes. The first is utilisation of plastic waste in the blast furnace process. During this process plastic waste generates a lot of flammable gases, which are a precious fuel in blast furnace process. The second use of plastic waste is using it in the coking process (CARBOTHERM process). During this process plastic wastes are destroyed by the coke formation process. An addition of plastic waste during the coke formation process does not adversely affect it.

2.1. Application of plastic waste in the blast furnace process

Providing a blast furnace with plastic waste first begun in foundries in Germany and Japan in 1995 (Maki and Ariyama, 2003; Okuwaki, 2004). German steel manufacturers: Stahlwerke Bremen and Krupp Hoesch substitute fuel oil, used in the production of pig iron, with granular plastic. Industrial and municipal plastic waste is used, which does not meet the conditions for material recycling. These two German steelworks use up to 30% of fuel oil used in the production process of pig iron in blast furnaces with plastic. In Japan plastic waste in the blast furnace process began to be used more or less at the same time as in Germany. In the blast furnaces, the only exothermal reaction is the combustion of coke and substitute fuel. Generated heat is necessary to reduce iron oxides and melts created iron. Depending on the ironworks, fuel oil or coke is replaced with plastics in the amount of several dozen kilograms per tonne of crude steel. This method provides considerable savings, on the one hand, by reducing the purchase of expensive coke, and on the other hand, by not having to pay for the collection of waste.

Thin, lightweight foil is broken down, melted and granulated; granules that are produced are sent to a silo. Large, solid waste is first crushed in a two-step process; then granulated. The final product is sent by a conveyor pipe to the same silo. From there, they go to a container of a blower, which then blows them through nozzles into a blast furnace. Granules of a diameter of 6-10 mm are fed into a blast furnace by a device that also injects pulverised coal (PCI-pulverized coal injection) through nozzles. Hot air is injected into the bottom of the furnace at high speed, creating a combustion zone, in which plastics are subject to gasification (incomplete combustion) to CO and H₂. While pulverised coal or coke quickly undergo combustion immediately after entering the zone of oxidation, using up O₂ and generating CO₂ at a temperature above 2000 °C (Ogaki et al., 2001):



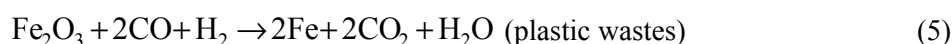
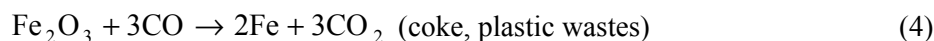
In the final combustion zone, once all the oxygen is consumed, CO is created as a result of the reaction of CO₂ with coke:



Plastic exposed to CO₂ degrade into CO and H₂ (as an example in the reaction polyethylene was used):



Hydrogen resulting in the process of degradation of plastic is a reducer in the blast furnace acting jointly with CO. The reaction of reduction with H₂ is less endothermal than a direct reaction with C, which is of course beneficial for the heat balance of the blast furnace. The reaction of reduction of iron oxides proceeds as follows:



Applying plastic waste reduces CO₂ produced in the blast furnace process by approx. 30% compared to using only coke and pulverised coal. In addition, resulting gases, which are recovered at the exit of a blast furnace, are enriched in hydrocarbons produced from the decomposition of plastic, have a high calorific value. In Poland, a significant part of generated plastic waste can also be utilised in blast furnaces. However, the cost of adapting nozzles providing air to delivering plastic wastes granules is a large barrier. Another method of recycling plastic waste in blast furnaces is their direct input in the blast furnace feedstock. This method would enable to use plastic waste almost without any investments. Slight financial outlay would be connected only to the process of grinding or granulating of polyethylene foils. In Poland, at the Institute of Iron Metallurgy, studies are being carried out on

utilising plastic waste in blast furnaces (Niesler, 2001; Niesler and Stecko, 2000). Industrial trials were carried out with the addition of waste in quantities of about 7 kg/t of pig iron, for a period of 12 days. Applying plastic waste not containing silica of carbon content approx. 78% and hydrogen approx. 12%, affects the economy of coke in the blast furnace process. However, the use of waste containing glass fibres with a high content of silica can have an impact on increasing the share of raw flux in the feedstock and on an increase in the consumption of coke associated with this. Chlorine content in blast furnace dust was approximately 0.6%. This is not a value exceeding the limits of chlorine content in blast-furnace dust, which is approximately 0.3-0.75%. Due to the possibility of dioxin formation and the corrosive properties of chlorine compounds arising from the degradation of polyvinyl chloride (PVC) should be removed from the waste stream fed into a blast furnace. The limit of chlorine content in raw material should not exceed 1% by weight. For waste with a high content of chlorine - gas treatment installations must be based on chemical absorption and catalytic or adsorption treatment and devices of blast furnace should be made of materials with high resistance to corrosion. In order to use waste with a high chlorine content (more than 2%), a dechlorifying technology developed in Japan can be used. It consists in heating raw materials containing chlorine at a temperature of 300-400°C, in an oxygen-free atmosphere. These raw materials are subject to thermal decomposition with a release of hydrogen chloride. The substance that has been dehydrochlorinated, after cooling and granulating, can be used in a blast furnace. Hydrogen chloride is absorbed in water, refined by distillation and recycled as hydrochloric acid (HCl). Owing to high construction costs of a dehydrochlorination station PVC is most often removed from the waste stream fed into a blast furnace.

2.2. Applying plastic waste in the coking process

Waste plastic can be used directly in coke chambers, where at high temperatures, together with coal, it is subject to thermal decomposition to: coke (20%), oils (40%) and coke gas (40%) (Nomura et al., 2003; Okuwaki, 2004).

2.2.1. Direct addition to the coke feedstock

An addition of plastic waste at the level of 1% to coke coal does not cause changes to the properties of coke (Niesler, 2001). Adding plastic at the level of 2%, containing aliphatic polymers such as: polyethylene (PE), polypropylene (PP) and polyvinyl chloride (PVC) has only a minor impact on the deterioration of coke's durability. In some cases, the addition of polyethylene even caused an increase in coke's durability. Figure 1 shows the effect of plastic addition on coke strength. Coke drum strength increases slightly in the case of PE addition, but the addition of PS and PET decreases coke drum strength to a great extent. The drum strength of PET addition coke is lower than that of PS addition coke because PET addition decreases the coal caking property more than PS addition. Figure 2 shows the effect of the addition of plastic waste on the index CRI (coke reactivity index). The addition of plastic to coal, in all cases, causes CRI to rise. This is due to the increase in the porosity of coke in all cases for which the addition of plastic waste was used (Figure 3). The cause of this phenomenon is the emergence of empty spaces in coke, resulting from the thermal process of decomposition of plastic particles. Figure 4 shows the influence of the plastic waste addition on CSR (coke strength after reaction). In the case of applying polyethylene an increase in CSR has been observed. This is caused by the growth of DI (Dilatation Index). For PS and PET addition, CSR decreases due to both a lower DI and a growth of CRI. The impact of plastic waste on the properties of coke is different depending on the material used. These differences are caused by the mutual effects between thermal products of plastic waste degradation and hydrogen from coal. Radicals resulting from the thermal process of degradation of PS and PET remove hydrogen from carbon, which may result in the deterioration of properties of coals of carbon. PE shows a trend of "transferring" hydrogen from plastic to carbon (Nomura, 2003). A direct addition of plastic waste to the coke feedstock has been used widely in Japan.

Since 2000, in Japan, in coking plants in Nagoya and Kimitsu, and since 2002 in coke plants in Yahata and Muroran, plastic waste has been dosed into the coke feedstock. The size of the addition amounts to 1%, which allows to dispose of 140 000 Mg plastic waste/year. Attempts are also being made to directly dose plastic waste (up to 3%) to the coke feedstock in Ukraine. Studies on the application of direct addition are also being conducted in China.

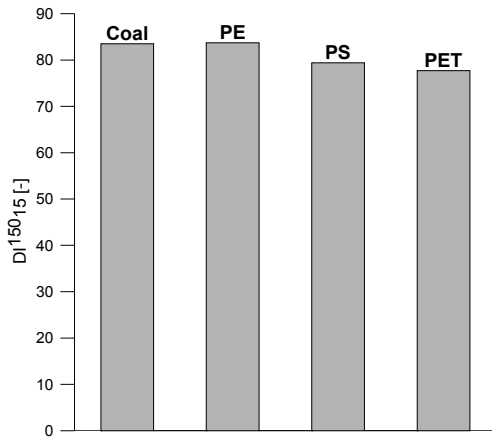


Fig. 1. Effect of plastic addition on coke strength (Nomura et al., 2003)

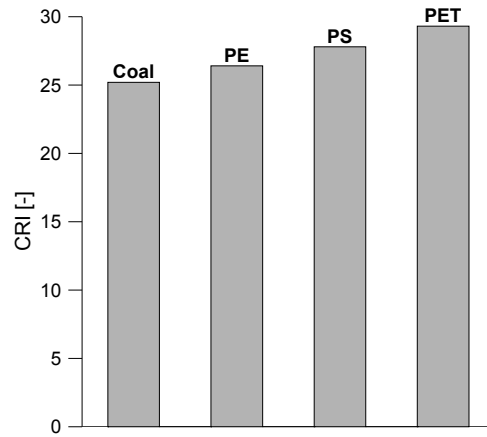


Fig. 2. Effect of plastic addition on CRI (Nomura et al., 2003)

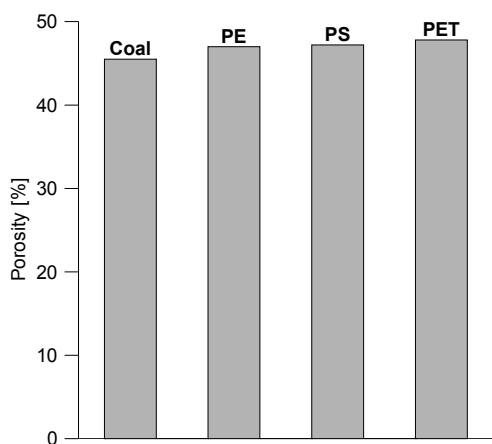


Fig. 3. Effect of plastic addition on porosity (Nomura et al., 2003)

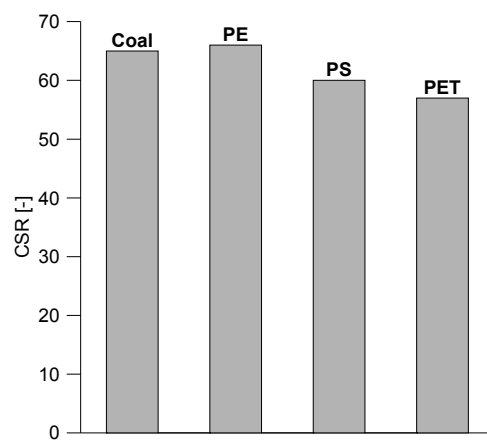


Fig. 4. Effect of plastic addition on CSR (Nomura et al., 2003)

2.2.2. Application of the CARBOTHERM process

A direct addition of split plastic waste to carbon mixture, in theory the simplest way of their use in the coke industry, is limited by a number of phenomena. This solution affects negatively the quality of coke (increased reactivity and reduced strength), as well as technological process (excessive degassing in the first phase of coking causing increased pressure in the coke chamber). Additionally, due to the formation of hydrogen chloride, it is necessary to separate PVC from the waste stream. These problems can be eliminated through the application of the CARBOTHERM process of plastic waste combined with partial degassing and structural destruction of polymers (thermolysis). The method was developed jointly by the Institute of Chemical Processing of Coal in Zabrze, Poland, and the Institute of Industrial Chemistry in Warsaw (Kijęński and Polaczek, 2005; Sobolewski and Wasilewski, 1998; Sobolewski

and Wasilewski, 2005). The CARBOTHERM process (Figure 5) leads to obtaining a component of the carbon feedstock of full value for the coking industry. This allows to both carry out the process of deep dechlorinating of plastic waste, and eliminate technological difficulties concerned with the process of cooling and solidification of the product of plastic waste thermopreparation. Carbotherm process also bypasses the operation of its grinding to a particle size that is compatible with technological requirements of the process of coking coal. In order to bring the physico-chemical properties of processed waste closer to the coal mixture, plastic waste is subjected to the process of thermopreparation.

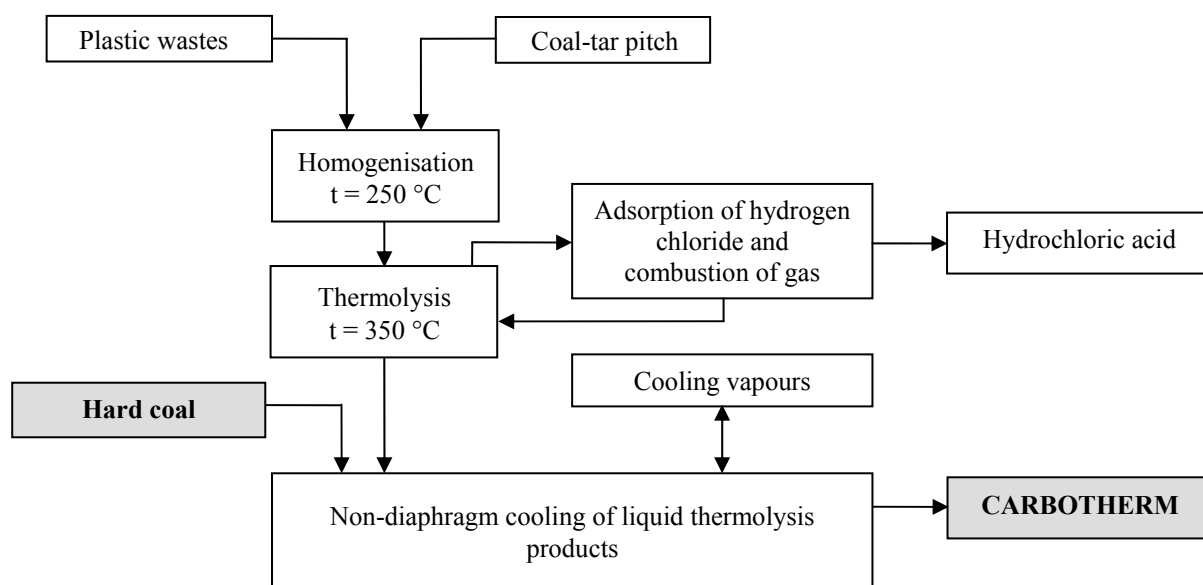


Fig. 5. Block diagram of CARBOTHERM technology (Sobolewski and Wasilewski, 2005)

Plastic waste generated from the entire stream of municipal waste, after mechanical reduction, is subjected to the two-stage process of thermolysis in coal or petroleum bitumen. To this end, plastic waste is mixed with hot, liquid coal-tar pitch the recommended mass ratio plastic waste/bitumen = 4/1, not exceeding the temperature of 250°C, without extracting volatile products. After initial homogenisation, the substrates are heated in the thermolysis reactor up to the temperature of 350 °C and are subjected to shear stress, which breaks causes the breaking of polymer chains (Sobolewski and Wasilewski, 2006). Small quantities of raw gas extracted during this process is subject to treatment from hydrogen chloride (originating from the decomposition of polyvinyl chloride). After the absorption of hydrogen chloride, raw gas combusts, and the resulting heat is used to conduct the thermolysis process. The hot, liquid product of thermolysis is mixed with culm, causing its cooling and solidification on the surface of coal grains. The resulting solid product which has been called CARBOTHERM - is loose, thermomechanically stable, and does not change its properties during storage. It can constitute both a coal feedstock for coking, and a substitute for solid fuel with a higher calorific value. In Table 3 a comparison is shown of the physico-chemical properties of hard coal and the finished product CARBOTHERM. The results of the analyses indicate a very large similarity of the properties of hard coal and CARBOTHERM.

First laboratory tests and then industrial tests were carried out in the Coking Plants "Wałbrzys" S.A. (Wasilewski and Sobolewski, 1998). Below (Table 4) the properties of coke obtained in industrial tests are shown. In assessing the results of determination of coke, it can be claimed that an addition of 5% of a thermolysis product to a coal mixture did not have a great influence on the parameters of coke strength nor on its reactivity (all results are within the limits of determination error). The chlorine content in coke produced from the coke mixture with the addition of a thermolysis product increased slightly, without exceeding the value normally obtained in classic coke.

Table 3. Physical and chemical properties of coal and CARBOTHERM product (Sobolewski and Wasilewski, 2005)

Parameter	Hard coal	CARBOTHERM
Elemental analysis, wt. %		
C	84	83.05
H	7	7.95
O	7.9	7.92
N	0.4	0.34
S	0.5	0.45
Cl	0.2	0.21
Bulk density, kg/m ³	850	900
Calorific value, MJ/kg	27	33
Mechanical properties	brittle	Brittle

Table 4. Properties of cokes obtained in the industrial tests (Wasilewski and Sobolewski, 1998)

Parameter	First industrial tests		Second industrial tests	
	coal mixture	addition of 5% of a thermolysis product	coal mixture	addition of 5% of a thermolysis product
A^a , wt. %	11.8	11.8	11.8	11.8
Cl^a , wt. %	0.2	0.236	0.2	0.236
CRI , %	52.5	49.2	52.5	49.2
CRS , %	29	33.8	29	33.8
Coke strength, %	83.7	83.5	83.7	83.5

3. EXPERIMENTAL

In order to determine the suitability of plastic waste for use in blast furnace process it is necessary to carry out a series of studies, among others, in the scope of elemental analysis, the process of thermal decomposition or analysis of chemicals produced in the process of decomposition. The results are very important for blast furnace technology. Especially the composition of gases generated in the process of thermal decomposition is significant for DRI (Direct-reduced iron) process. In this work the results of the thermal analysis are presented.

3.1. Materials

Three samples of plastic waste from a polyolefin group were studied: 2 samples came from packaging for cosmetics and cleaning products containing high density polyethylene (PE-HD) and polypropylene (PP), the third sample came from the agricultural and horticultural industries and this sample was used for covering tunnels containing low density polyethylene (PE-LD).

3.2. Instrumental methods

For test samples an elemental analysis was conducted - the chemical composition, ash content and calorific value were examined. The elemental analysis was carried out by the elemental analyser Leco SC 132 and Leco CR 12. The calorific value was calculated on the basis of measurements made using a calorimetric bomb.

Thermogravimetric analysis is one of the instrumental methods used to analyse plastic wastes. In the present study thermogravimetry (TG) was carried out on the tested samples. Thermogravimetric analysis was carried out using a Mettler Toledo TGA/SDTA 851° apparatus. The TGA instrument was calibrated with indium, zinc and aluminium. Its accuracy was equal to 10^{-6} g. For the thermogravimetric analysis (TG) the samples were placed in alumina crucible. C.a. 2 mg of the sample was heated from an ambient temperature up to 1000°C at a constant rate: 10 °C/min in 3 dm³/h flow of air. The measurements for each sample had to be done under exactly the same conditions including e.g. the range of temperature, atmosphere, heating rate, etc. to determine the most repeatable and precise results. The experiments were replicated to determine their reproducibility, which was found to be very good. TG curves for each fuel sample were obtained as an output. TG curves were used in order to assess the thermal characteristics of plastic waste.

Evolved gaseous products from the decomposition of studied samples were identified on-line, using connected to ThermoStar GSD300T Balzers quadruple mass spectrometer (QMS). The mass spectrometer was operated in electron impact mode (EI) using channeltron as a detector. Screening analyses were performed in the selected-ion monitoring (SIM) mode. The following ion characteristic of each molecule, such as: 16, 18, 28, 29, 39, 40, 41, 43, 44 and 78 for CH₄, H₂O, C₂H₆, C₃H₈, C₄H₆, C₃H₄, C₃H₆, C₄H₁₀, CO₂ and C₆H₆. were monitored. It is important to notice that the QMS spectrum of mass 28 can represent not only C₂H₆ but also C₂H₄, and 29 can represent also CH₂O and C₂H₄O.

4. RESULTS AND DISCUSSION

The results of the elemental analysis indicate clearly a very similar chemical composition of the tested materials (Table 5). The carbon content in the analysed waste (82.8-83.1%) is very high, while the sulphur content in the materials (0.03-0.1%) is low, which has a substantial impact on reducing emissions of SO_x. It is worth noting a very low ash content and a very high calorific value of the tested samples, which significantly raises the value of the waste used as fuel in the blast furnace process.

Table 5. Elemental analysis of polyolefine wastes

	PE-LD	PE-HD	PP
Elemental analysis, wt. %			
H	14.93	15.1	15.26
C	82.8	83	83.1
S	0.07	0.1	0.03
N	1.93	1.21	1.46
Ash, wt. %	0.01	0.7	0.06
Calorific value, MJ/kg	42.23	42.78	43.34

On the basis of the results of the TG-MS analysis both the process of thermal decomposition (TG curves) was conducted and the presence of chemical compounds in waste gases, depending on the

temperature was determined. The shapes of TG curves are very similar, and the residue after incineration is very small (Figure 6). Thermal decomposition begins at a temperature of approx. 250 °C and ends at approx. 550 °C, except the temperature at the end of decomposition of polypropylene, which is approx. 400 °C. It is easy to notice that the maximum speed of the decomposition of polyethylene takes place at a temperature of approx. 400 °C, while that of polypropylene at approx. 300 °C. The beginning of the process of decomposition can be caused by the combustion of compounds from an alkanes group, whose temperature of spontaneous combustion is lower than 250 °C (e.g. n-hexane and n-heptane, n-octane, n-nonane, n-decane, n-undecane). These compounds are products of the main chain of polymer splitting, therefore, they arise in relatively low temperatures (they do not require a large amount of energy to a possible recombination). Further stages in the process of decomposition are associated with the process of compound combustion, which includes: n-butane, 1,3-butadiene, ethane and ethylene, and these gases occur at a temperature of approx. 350 °C, 400 °C and 450 °C. Mass spectrometry analysis also revealed the presence of combustible compounds with higher (than 450 °C) ignition temperature such as: CH₄, CO and C₆H₆. During the identification of certain chemical compounds the database of NIST (National Institute of Standards and Technology) was used.

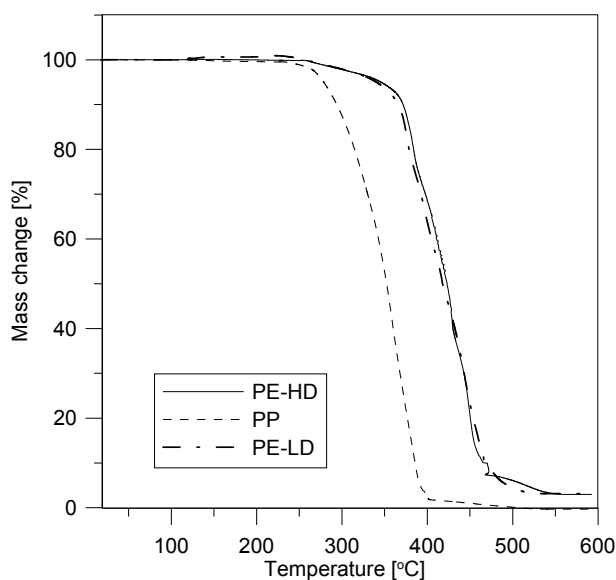


Fig. 6. TG curves for PP, PE-HD and PE-LD

Both chemical composition (high content of carbon and hydrogen), high calorific value and the composition of gases generated in the process of decomposition, make the tested polyolefin waste constitute a valuable substitute for coke in the blast furnace process. Feeding waste together with a feedstock generate gases educed in the thermal process of decomposition. In addition, hydrocarbons generated in the first phase of decomposition increase the calorific value of the blast-furnace gas. Due to a very similar composition of hard coal used in the coking process (Table 3), polyolefin waste can be used as an additive in coking coal.

5. CONCLUSIONS

One of the methods of utilisation of plastic waste can be its use in the metallurgical industry. This waste is used as a substitute for coke in the blast-furnace process or as an additive to coal in the coking process. In blast furnace process plastic waste can be fed via nozzles supplying air, at the bottom of a furnace or at the top together with the feedstock. In the coking process an addition of plastic can be applied directly to the coke feedstock (this method, however, has many defects) or after a prior completion of the process of CARBOTHERM (thermopreparation). In order to determine the suitability

of different groups of plastic to the above mentioned processes, it is necessary to carry out a series of tests of thermal analysis. Due to the chemical composition and the quality of the products generated in the process of thermal decomposition (n-hexane and n-heptane, n-octane, n-nonane, n-decane, n-undecane, n-butane, 1,3-butadiene, ethane, ethylene, CH₄, CO and C₆H₆), waste from the polyolefin group constitutes a very attractive group of materials used in blast furnace process. CO from the thermal decomposition of waste is both a fuel and a reducer. However, hydrocarbons produced in decomposition processes significantly increase the calorific value of the gas furnace.

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Received 23 April 2012

Received in revised form 11 February 2013

Accepted 14 February 2013