

Transformation of the polyethylene wastes into fuel products

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Introduction

Polyolefines (polyethylene - PE and polypropylene – PP) constitute the basic group of the synthetic polymers. They do not have any smell nor taste. They are not toxic, do not dissolve in water, are chemically resistant and easy in processing. These features redounded to a great popularity of polyolefines. The bulk production of the polyolefines outstrips all remaining polymers. In 2010 the total production volume of PE in Europe reached the level of 15.1 mln tons. The annual increase of the production is about 4%. In the intensively developing China the total production of PE in 2010 was to reach the volume of 11 mln tones and the estimated demand indicated the need of the additional import of a similar amount of this polymer. In the rapidly growing Middle East countries there is expected a deficit of the PE after the year 2015. The global consumption of the PE in 2010 was estimated to be 77 mln tones. Local production of the polyolefines in Bassel Orlen Polyolefines reached in 2010: LDPE - 96 000 tones, HDPE – 262 000 tones and PP – 327 000 tones. A continuous increase of the polyolefines production is still observed and similar increase tendencies of the polyolefine wastes production should be expected.

The lifetime of the polymer products is short or very short as for the plastic bags for instance. The lack of incineration plants in Poland indicates the landfills as the main direction of the final handling with these wastes together with the urban wastes. Polyolefines undergo very slow degradation and biodegradation and will remain in the landfill prisms for tens or hundreds of years. It seems that a selective collection of the polyolefines is a right way to achieve reduction of the amount of polyolefines reaching the landfills. The re-granulates made of waste polyethylene or polypropylene in general do not exhibit as good processing properties as fresh polymers. They suit but well for the production of goods of a lower quality requirements. Regranulates may be also used as an acceptable admixtures to the fresh polymer. In the long run the material recycling alone does not reduce the amount of the polyolefine wastes stream transferred into landfills.

If the pharaohs new plastics in their times it wouldn't for sure influence their decision on the choice of the construction material by the rising of the pyramids. Thanks to this pyramids reminded few thousands years. After our civilisation may remain rather inglorious garbage mountains containing polymer wastes. Strong pressure onto the natural environment protection including protection of the atmosphere begin to bring the results. The regulations are modified to strengthen pro-ecological actions. Nevertheless it seems rather impossible to solicit the reach societies to reduce the consumption level but it is quite real to convince them to increase the level of waste recycling and industrial reuse of the wastes. Polyethylene and polypropylene form around 7% of the urban waste. In Poland nowadays about 860 000 tones of waste polyolefines come down to the landfills annually. European Union recommends their members reduction of the waste storage down to 50% in 2013 under the threat of punishment. The wastes segregation itself will not solve the problem of the huge increasing the wastes amount. It is necessary to work out efficient methods of the waste management and technologies of their recycling.

Methods of the management of the waste polyolefines

There are various methods of rational management with the waste polyolefines which are alternative to their storage at landfills:

- co-incineration in the waste incineration plants
- energy production from the separated polyolefines in adjusted burners
- material recycling, re-granulation and reuse for new goods production
- chemical recycling, depolymerisation toward the starting-point ethylene and propylene
- separation from the wastes stream, purification and exporting
- fuels production

Polyolefine wastes are of high calorific value (Tab.1) and may be used directly as a fuel for the electricity and heat production. The PE combustion face but some technical problems. PE delivery to the burner requires special dispenser. Inside the burner the PE first softens, then melts down and before liquefies it forms a viscous mass hard to pump and to atomise. The oven fed with PE should be able to handle with the fuel which changes its state of matter and relatively slowly but finally turns into a light liquid. During the preheating it may arise the effect of formation of the molten polymer foam. New concept in EU regulations recommend limitation of the waste incineration in aid of material or chemical recycling. The mechanical contaminants, other polymers, non-homogeneity of the material and necessity of the change of the state of matter are serious impediments in conducting the burning process and in the automation of the work of the burner/furnace appliance.

Table I

Calorific values of various fuels

Fuel	Calorific value, MJ/kg
coal	25 - 28
wood	7 - 14
heating oil	40 - 48
polyethylene	44

All mentioned above methods of the recycling of the polyolefine wastes are used in various extent depending on the temporary or local economic trends and available technologies and raw material. The possibility of the application of a given technology depends also on the uniformity of the raw material, type of admixtures, colour, degree of initial degradation etc. In general the recycling of the polyolefines in Poland is not carried out on a large scale.

Fuels out of waste polyolefines.

An interesting feature of the polyolefinic polymers is their relatively easy decay at high temperatures. In the molten polymer in the temperature range 400°C (450°C) – 800°C occur the cracking of the chemical bonds and formation of low molecular products which distill off. As a result of this process four groups of products are obtained:

- Gas products which do not condense, they consist of simple saturated and unsaturated hydrocarbons C_1-C_5 such as: methane, ethane, ethylene, propane, propylene etc.
- Liquid, condensing hydrocarbons with the carbon chain in the range C_6-C_{20} , they are saturated, unsaturated, aliphatic and cyclic ones. At room temperature they form a liquid of specific smell of kerosene products and of a colour from light yellow to dark brown which tends to darken during storage due to slow oxidation in contact with air.
- Solid paraffin products $C_{20}-C_{40}$ (C_{60}). They distill off together with the lower boiling products and crystallize after cooling.
- Coke which is a solid, not distilling remaining of the reaction.

Figure 1 shows tentative fraction of the individual groups of products as a function of the applied temperature of the cracking process. Dashed line in the range of lower temperatures indicates that the cracking process goes very slowly or stops.

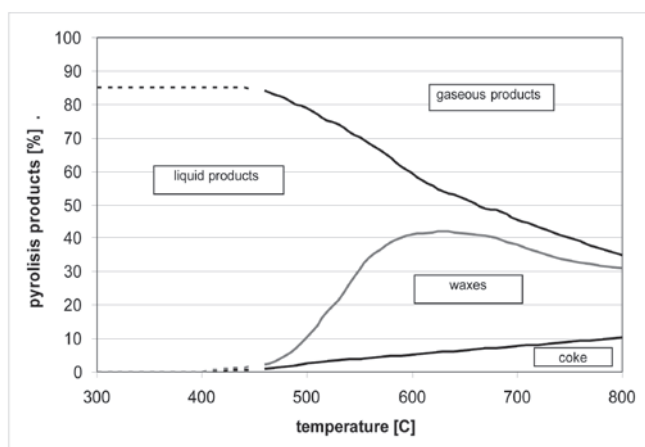


Fig. 1. Tentative changes of the fractions of the products of the polyolefines cracking reaction as a function of the applied temperature

During a mild pyrolysis of PE conducted at temperature 470°C it is possible to receive about 85% of liquid products, 10% of non-condensing gases and 5% of coke.

Thermic pyrolysis processes

Polyolefines subjected to high temperature undergo chemical degradation. Long carbon chains crack and arise products of shorter and shorter molecular mass. This process is hard to control precisely despite of the use of various catalysts and final product is a composition of numerous hydrocarbons, saturated and unsaturated, also cyclic with different number of carbon atoms in the main chain in the range from C_1 to app. C_{40} . In relation to the chosen technology different fractions of gaseous, liquid and solid products may be achieved. Many polyolefinic waste treatment technologies come from the petrochemical industry what seems to be understandable due to obvious chemical similarities of the transformed materials.

There are numerous techniques of PE cracking process. Further are shown the most known ones together with their descriptions and comments.

Thermal Cracking

Direct heating of polyolefines in the temperature range 380-480°C leads to a gradual cracking of the carbon chains in the polymer (thermal cracking reaction). The higher temperature applied the quicker this

process runs but it also change the proportions in the products stream. Degradation and the carbon chains destruction lead to formation of molecules of hydrocarbons boiling at lower temperatures which gradually evaporate from the reactor. The gas phase leaving the reactor consists of various gaseous hydrocarbons (C_1-C_6) like methane, ethane, ethylene, propane, propylene etc, and higher ones which can condense. The condensing fraction forming a liquid phase at room temperature consists of two parts: gasoline - with boiling temperature up to 200°C and kerosene part with the boiling temperature in the range 200°C - 340°C. Paraffin (wax) products distill off together and if are not separated tend to crystallize and sediment. In general the thermal cracking process comprise initial heating and melting of the waste and further begins carbon chains cracking in endothermic reaction accompanied by the evaporation of the products. The melting of the wastes itself is not easy because polyolefines have low coefficient of heat conduction and additionally after melting have a form of very viscous liquid difficult to handle. In few technologies the waste is initially molten and later introduced into the main reactor in other ones the polymer wastes are introduced directly into the reactor where undergo melting and cracking at the same time. The methods of the conduction of the polyolefines cracking process may be divided also in terms of different of ways of delivering of energy necessary for heating the waste and maintaining the endothermic cracking reaction.

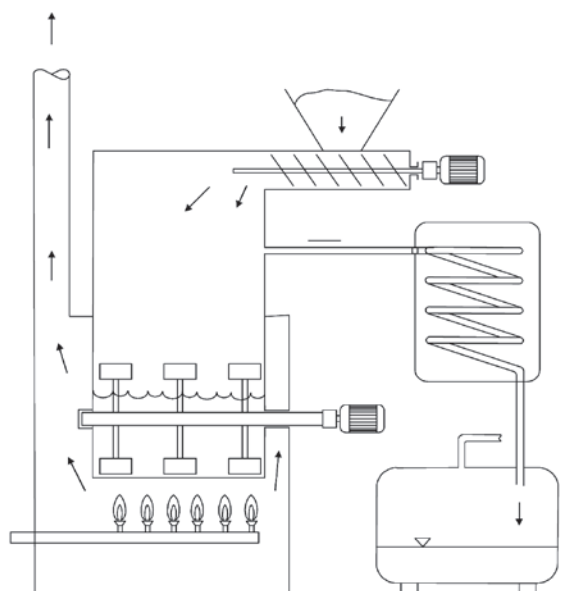


Fig. 2. Cracking reactor with indirect heating - scheme

The most frequently described in the literature are indirect techniques of the polyolefines heating (Fig.2). Steel reactors of various shapes filled with wastes melting and cracking at high temperature are heated electrically or with the use of gas burners. The cracking reaction of PE/PP wastes is being conducted at the oxygen-free atmosphere of course. Various horseshoe and helical agitators ensure mixing of the melted wastes inside the reactor, remove coke sediments and dirt collecting on the heating surfaces of the reactor. Reactors of this type work periodically but most of the inventors expects additional continuous and automatic withdrawing of the forming coke and other inert dirt out of the reactor in order to prolongate the working time of the reactor. It generates some technical problems. First of all it requires confirmation of good sealing of the installation to avoid possible leakages and burning of flammable content. Besides the solid sediments saturated with molten polymers tend to solidify while cooled and this complicates secure lock construction. Together with the dirt extracted from the reactor some polymer is extracted too and lost. This leads to decreasing of the efficiency of the reactor. The examples of the reactors with indirect heating are described in [7÷9, 16÷19]. In the

case when the main expected products are the liquid fractions, the arising gases are often used in place and burned under the reactor or used for initial melting of the applied polyolefine wastes [22].

Catalytic cracking

The use of catalysts in the cracking reaction enables achievement of additional benefits. The temperature of the process may be decreased and the reaction products become more homogenous and of better quality. Various zeolites are often used as the catalysts and introduced into the reactor in a powder form. The catalyst particles are kept dispersed in the molten polymer due to the applied agitators. In [20] there are proposed aluminosilicates and activated dolomite as catalysts. In the different publications are mentioned almost all types of catalysts applied in petrochemical industry. Even Portland cement was suggested as a right catalyst in one patent. Sometimes reduced or increased pressure: 0.02 – 0.20 MPa is recommended. Besides the free suspension of the catalyst particles in the molten polymer sometimes the inner walls of the reactor are additionally covered with the catalyst layer [22]. To enhance the outgoing of the products out of the reactor it may be sometimes blown with the inert gas like nitrogen or hydrogen. In [24] numerous types of catalysts are suggested together with the use of water vapour as a carrier gas. Water vapour is recommended as a cheaper solution for local installations. The application of copper catalyst made by the solution of Cu₂O in molten NaOH and KOH is recommended [30]. The use of catalysts gives also the possibility of effective alkylation of the PE cracking reaction products with the cracking products of polystyrene (PS) and other hydrocarbons [3]. The catalysts applied in cracking reaction but undergo gradual deactivation, they are covered with the carbonaceous products layer (carbon fouling) and require supplement and exchange. It is always recommended to compare the advantages of the use of catalyst, accompanying costs and technical complications.

Liquid metal reactors

A very interesting method of the heat transfer to the treated waste polyolefines is the use of a molten metal (e.g.: lead m.p. 327°C; zinc m.p. 419°C) as a heat carrier [1]. The pyrolytic reactor is filled partially with the molten metal and the waste polymers are introduced into the reactor (with the help of a feeder) on the surface of the liquid metal. Additional agitator intensively increase the heat transfer. Volatile products of the pyrolysis leave the reactor and are condensed in an outer condenser. The gaseous products are used in place to heat the reactor. Such a construction ensure very effective melting of the polyolefines which are difficult to melt because of their low heat conductivity.

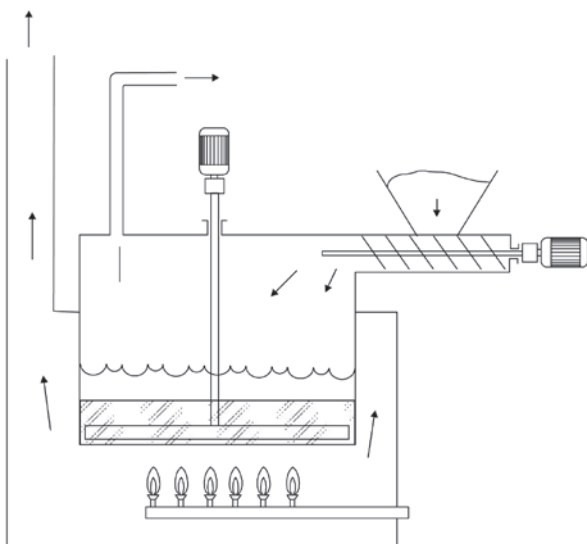


Fig. 3. Liquid metal reactor – scheme

The application of other metals as a heat carriers: tin, bismuth, antimony, alloys of Wood and Rose and inorganic salts like sodium and lithium chloride is recommended in patent [12]. There is also mentioned even sodium nitrate though it is obviously strong oxidizer and if applied would lead to explosion of the reactor. In [12] it is recommended in general inorganic heat carrier with the melting temperature below 500°C. The inventors do not explain if and in which extent the proposed metals evaporate and contaminate the products and how to separate from the molten metal other impurities getting into the reactor together with the treated polymeric wastes.

Reactors with fluidized bed

In the literature are also described apparatuses for pyrolysis in fluidized bed. Molten polymer is introduced onto the fluidized bed consisting of sand particles of diameter 0.2 mm and hold in the fluidic state with the help of inert gas (nitrogen). The temperature of the bed is 500 °C and is controlled with the help of outer heat source. It is also suggested to generate the energy necessary to keep the pyrolysis reactions going due to burning of the part of the input polymer inside the bed with the oxygen admixed to the carrier gas on the level 3-5% in relation to the mass of introduced polymer. According to the inventors the efficiency of the liquid products reaches level 89-92%. The gaseous products (7.8-10.5%) and coke (0.2-0.5%) are formed as the byproducts. Coke and other solid impurities remain inside the bed and need to be periodically removed together with the part of the bed [10]. In [13] it is recommended the use of fluidized bed formed by the particles of silica, ceramics, coal or aluminosilicates with the diameter in the range from 0.1 mm to 2 mm. The fluidized bed is being kept in the temperature between 450°C and 600°C with the help of hot carrier gas. The waste polyolefines are introduced into the bed from the bottom side in the form of aerosol but direct introducing on the top of the bed is also acceptable. Syngas and gaseous aliphatic hydrocarbons C_n-C_m are also suggested as a possible carrier gases. To avoid the coagulation of the bed particles in [14] is recommended addition of carbonates or oxides of calcium, barium and magnesium. The performance of the polyolefines cracking in this technology is a natural extension of the general application possibilities of the fluidized bed and became a popular subject of the experimental research [25].

Hot gas as a carrier

Besides the indirect methods of heating of cracking reactor also direct heat introduction is considered. In [5] an inert gas, water vapour or exhaust gases heated initially to 850°C-1100°C are mentioned as the possible heat carrier media. Hot gas gets into the pipe reactor together with the stream of molten polymer. Inside the reactor follows the atomisation of the polymer and heating to high temperature necessary to run the reaction. The time of the remaining of the polymer in the hot zone of the reactor is 10 to 100 milliseconds. After leaving the reactor the products are cooled and separated into desirable fractions.

Cracking in water

The complications in heating and melting of the raw polymer wastes and difficulties with further pumping of the melt because of its high viscosity and the risk of solidification when cooled inspired researchers to find a cracking technique in water medium. Shredded waste polyolefines are blended with water and further heated up to 300°C-500°C under high pressure from 560 to 840 atm in an autoclave. In such conditions follows the cracking of the polyolefinic wastes. After reduction of the pressure and cooling the products of the reaction may be separated [11]. The use of the water facilitates the separation of hydrocarbons. The inventor does not explain if and in which extent water takes part in the chemical reactions with the products.

Hydrocracking

The process of the thermal cracking may also be conducted in the presence of hydrogen gas. The presence of the hydrogen enables saturation of double bonds and receiving products which are saturated hydrocarbons having better chemical stability. Hydrocracking is conducted under increased pressure and because of the use of the hydrogen which forms explosive mixtures with the air is potentially dangerous. The cracking products achieved earlier in any method may themselves undergo hydrogenation reaction. Hydrogenation is performed catalytically at the temperature 450°C under pressure 14 atm [23]. To reduce the viscosity it is suggested to conduct initial hydrogenation of hot solutions of polyolefines in mineral oils in the presence of H₂ at higher temperature and increased pressure.

Reactors with radiant heating

The delivery of the energy necessary to maintain cracking reaction with the use of infrared radiators was proposed in [13]. The waste polyolefines pass along on the conveyer in the reactor chamber under the set of radiators which emit the energy for melting and destruction of the polymer (Fig.4).

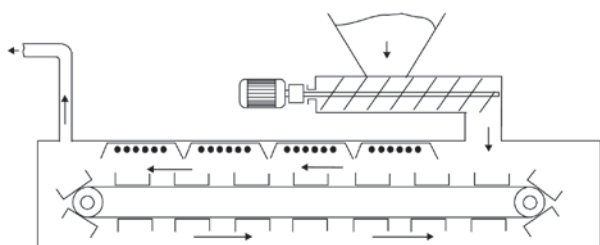


Fig.4. Reactor for pyrolysis of polyolefines with the use of infrared radiators [13]

In this time the radiation is absorbed in the material which melts and undergoes cracking reactions. In the reactor chamber there are oxygen-free conditions and slight under pressure. The gaseous products are being sucked off and condense in the outer condenser. A greater amount of gaseous products is expected and more paraffinic products which are driven away in the gas stream together with the coke particles. The dirt present in the raw waste collect in the reactor and are taken away through the lock in the bottom of the reactor chamber not shown at the scheme (Fig.4).

Pyrolysis in plasma

The radical destruction of the polymer carbon chains may be conducted also in a very high temperature of plasma reactor. In [29] is described proposition to introduce the molten polyethylene directly into the hydrogen plasma reactor. There exists the temperature of the range of 2500°C and under the pressure of 0.05-0.5 MPa follows the destruction of polymer molecules into gaseous products such like: H₂, C₂H₂, C₂H₄ etc. Later solid products (coke) are separated from the gas stream and the gases undergo purification and fractionation.

Syngas production

Polyolefinic wastes consisting of carbon and hydrogen may be transformed into a syngas (mixture of H₂ and CO). According to the propositions presented in [4] the polymer waste undergoes first hydrocracking in the hydrogen atmosphere in a periodical reactor for 30 minutes under the pressure of 70 MPa and temperature 430°C. Later the liquid products of the hydrocracking contact at 1315°C and pressure 3.5 MPa with the gas containing oxygen and water vapour as a moderator. In such condition the already degraded waste undergo partial oxidation producing energy necessary to maintain the endothermic synthesis reaction of the syngas out of the remaining part of the used polymer waste.

The use of microwaves

Another method of heating of the degraded polymer wastes is application of electromagnetic waves. In [6] is postulated the use microwaves of the frequency 2450 MHz and ultrasounds to conduct the reaction of cracking reaction of various waste polymers. The treated wastes are admixed with coal to increase the microwaves absorption. The whole process is conducted under the vacuum and the treated wastes reach the temperature only 93°C. According to the author [6] this process enables production of greater amounts of gaseous products (50%), oleic (25) and carbon (25%). According to the inventor, the coincidental use of the ultrasounds is to improve desorption of the gaseous products. The chemistry of the reactions is not explained.

Introductory irradiation of the wastes

It is also taken into consideration the introductory irradiation of polyethylene wastes with the use of gamma radiation in order to conduct tentative degradation of the carbon chains of the polymer. Polyethylene treated in this way undergoes much easier further thermal degradation and cracking. The descriptions of the experiments on this process are presented in [26 ÷ 28]. Introductory irradiated polyethylene undergoes the thermal cracking two times quicker.

Summary

The polyolefine wastes are very attractive raw material for various chemical transformation routes thanks to the bulk production and relatively uniform chemical composition. Such treatment of these wastes leads to the reduction of the waste amount stored at the landfills. The research on the polyolefine waste recycling is continued already since over forty years. Practical implementations in this area arise since about twenty years. A good atmosphere is formed around the problems of wastes handling and the local and European regulations evolve toward solutions which favour development of new recycling technologies. The practical applications find but these technologies which are worthwhile. The worthwhileness of a waste treatment technology, besides investment and operating costs including energy needs, depends on accessibility of possibly clean and cheap raw material. Nowadays when the oil prices go up the circumstances arise favouring transformation of polyolefine wastes into fuels. Developing waste segregation net and generation of a stream of selected polyolefine wastes trigger searching for good, sure, attractive and possibly already tested methods of the waste treatment. Still in the sector of the regular fuels producers are certain predispositions which may discourage potential investors. For the scientists and constructors but always is the right time to create new processes of waste treatment and new technological solutions.

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