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Pyrolysis of plastic wastes as a way of obtaining valuable chemical raw materials

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ABSTRACT

This article presents a mini-review of the literature on the laboratory scale pyrolysis of selected plastic waste. Among the various types of polymer materials polyolefins (PE and PP), as well as polystyrene (PS), polyoxymethylene (POM), and polyvinyl chloride (PVC), were selected. The paper highlights valuable compounds and chemical raw materials that can be obtained by pyrolyzing the waste polymers. Pyrolysis of polyolefins can lead to the formation of liquid paraffins and olefins. By pyrolyzing aromatic polymers such as polystyrene, a valuable BTEX fraction can be obtained, the efficiency of which depends on the conditions of the process. Polyoxymethylene pyrolysis can be an alternative way to produce valuable formaldehyde. A short discussion based on the cited articles has been presented

1. Introduction

Plastics are present in every branch of industry nowadays. Polymers are used in heavy fuel and energy industries, in the production of machinery and apparatus, in electronics and electrical engineering, in the medical and pharmaceutical fields, as well as in the food and textile industries [1]. In 2014, the worldwide plastic production exceeded 300 million tons, and this number is increasing every year [2]. In 2018, nearly 360 million tonnes of plastics were produced worldwide, most of which were thermoplastics and polyurethanes. Since 2010, an annual increase of plastics production at the level of 9 million tons could be observed. After 2012, this production increased by an average of 11 million tons annually (Fig. 1). The observed trend proves the continuous increase in plastic used in the industry as well as in the consumption of these materials in everyday life [3].

Due to the increasing worldwide production of these materials, plastic waste management should become a priority for many economic entities around the world. There are many methods of plastic waste disposal. However, according to the five-step hierarchy of waste management, as a first step their generation should be prevented, and if this is not possible, then waste should be prepared for reuse or recycled as a second and third option respectively [4,5].

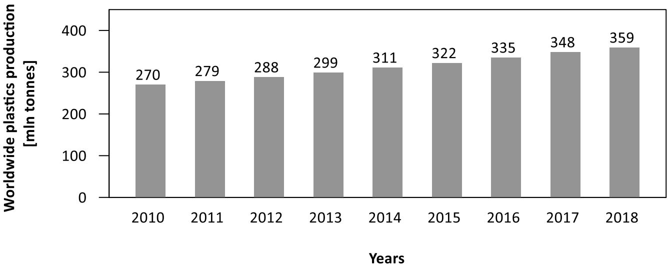

Figure 1. Worldwide plastics production in years 2010 - 2018 [3]

Due to the increasing worldwide production of these materials, plastic waste management should become a priority for many economic entities around the world. There are many methods of plastic waste disposal. However, according to the five-step hierarchy of waste management, as a first step their generation should be prevented, and if this is not possible, then waste should be prepared for reuse or recycled as a second and third option respectively [4,5]. The last two methods of waste management include methods like combustion or co-combustion (fourth step) and waste disposal, e.g. by landfilling as a last resort [4]. According to data on waste management in Europe, in 2018 as much as 45.8% of wastes were landfilled. However, there were significant

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differences in the application of waste treatment methods between EU members. For instance, Italy and Belgium showed a high recycling rate of waste (approx. 75%), whereas, in countries such as Greece and Bulgaria, more than 90% of waste was landfilled [6]. Plastic waste should not be landfilled at all because its biodegradation may take even hundreds of years [7]. The European Commission has adopted an ambitious and long-term waste management plan, demanding that by 2030 the percentage of municipal waste deposited does not exceed 10% [8]. However, the first two methods of waste management are often impossible to implement. There are wastes, the production of which cannot be prevented or cannot be reused, i.e. waste from craft plants or already processed waste. Most of the wastes that cannot be processed by the two first hierarchy stages can be pyrolyzed [9]. In the waste pyrolysis process, plastics are heated to temperatures mostly exceeding 300°C. It causes structure damage by breaking intramolecular bonds, and valuable organic products are produced such as syngas, liquid (oil), or solid (char). Depending on the temperature and contact time, slow and fast pyrolysis can be distinguished. Pyrolytic gases can be used to supply the energetic demand of the process and the surplus may be used for additional power generation [10]. The liquids from plastic pyrolysis can be used as alternative fuels or as a source of valuable chemicals (e.g. styrene, toluene). It is an interesting technology that allows the simultaneous disposal of plastic waste and the production of fuels and chemical raw materials [5]. The pyrolysis process is not yet as widely developed as other thermal waste degradation methods. However, the process is commonly studied by scientists because there is a necessity for the transformation of the economy to a circular type. Technological problems such as adjusting the appropriate reactor or process conditions depending on processing wastes have not been solved yet [11]. The use of a catalyst in the process is also being investigated. Despite the advantages of catalytic pyrolysis, the challenges, i.e. adjusting the cost and type of catalyst to pyrolyzed materials remain to be solved [12,13]. Therefore, plastic waste pyrolysis research are needed taking into consideration the amount of plastic waste growing every year.

This article demonstrates achievements from the laboratory pyrolysis of plastics such as PE, PP, PS, POM, and PVC. The commercialization of the pyrolysis process of these plastics should bring particular benefits to the circular economy, as these polymers are the most widely used or produced on a large scale.

2. Pyrolysis of polyolefins

Polyolefins are chemical compounds that contain only carbon and hydrogen atoms in the molecule and they are the lightest of all polymers. Polyolefins are commonly used for the production of bags, foils as well as pipes, and electronic devices [9]. These compounds are the most commonly used worldwide plastics and represent a majority of landfilled plastic waste. Santamaria et al. [10] investigated the influence of temperature on the pyrolysis process of resembled municipal plastic wastes. The experiments have been. carried out in a semi-batch reactor at temperatures 600–800°C. The authors confirmed that temperature has a significant effect on the composition of liquids pyrolysis products but a lesser one on solid and gas properties. Aromatic compounds were produced at relatively high temperatures due to the triggering of secondary reactions [9]. Obeid et al. [14] produced fuel-like products by thermo-catalytic pyrolysis of polyethylene bottles. The research has been carried out in a packed bed reactor at a temperature of approximately 450°C. Products i.e. alcohols (C16 and C17), paraffins (≤C44), aromatics (≤C14), and olefins (≤C22) were generated. The number of paraffins was mostly higher than 60%. They also investigated that parameters such as paraffins to olefins ratio could be changed by using different bed materials (white clay, silica, cement, and sand). The authors have noted that the use of zeolitic catalysts i.e. HUSY and HBeta zeolite increases the yield of aromatic compounds like D-limonene and naphthalene. Cheng et al. [14] investigated high-pressure pyrolysis conditions for polyethylene plastics waste. Their results showed that high pressure (21 bar) leads to the formation of mostly aromatic products, isoparaffins, and cyclic compounds such as cycloparaffins. Park et al. [16] carried out the pyrolysis of polypropylene in a fluidized bed reactor (FBR). Their research was based on the innovative two-stage process employing two reactor types: auger reactor and fluidized bed reactors connected in series. The auger reactor was employed in order to activate the vibrational states of molecules fed into the fluidized bed reactor. Such an organized process led to much faster material degradation and a higher yield of gases (81 $\%_{\text{wt.}}$) which were mainly hydrogen and methane. The same kind of system was applied for polyethylene waste degradation [17]. Auger reactor again led to increasing the yield of valuable pyrolysis gas. The highest gas (74.6 $\%_{\text{wt.}}$) and ethene (34.5 $\%_{\text{wt.}}$) yields were obtained when nitrogen was used as the fluidized agent. Kaminsky et al. [18] tested the pyrolysis process of polyolefins in the fluidized bed reactor with the additive of Lewis acids such as titanium chloride and aluminum chloride. The experimental results showed that the above-mentioned acids lead to the reduction of the temperature of the pyrolysis by approximately 100°C without changing the quantity and quality of the generated gas products. Colantonioet et al. [19] performed reducing the temperature process of plastic degradation by adding zeolites. The highest efficiency (29 $\%_{\text{wt.}}$) of monoaromatic compounds (mainly benzene, toluene, styrene, and ethylbenzene) at temperature 450°C was achieved when HUSY zeolite was used as a catalyst. In turn, HZSM-5 promoted the generation of gases

(41 % wt.) at 650°C. Butler et al. [20] performed a literature review and stated that it is not possible to generate a well-defined transport grade fuel from pyrolysis of polyolefins without using catalysts. The authors of the review declared that catalyst coking is becoming a significant problem. The presence of such a phenomenon suggests the pyrolysis process should be organized in a fluidized bed, in which it is easy to burn coke. Elordi et al. [20] investigated the incorporation process of polyolefins pyrolysis with an additional separation system to a refinery plant. It can be summed from the energy balance that the aforementioned solution can be energetically beneficial and only about 5% of the input mass flow rate is needed to meet energy requirements. Ali et al. [22] performed research on the validity of using the polypropylene pyrolysis doped with coal and petroleum residues for transport fuel production. The research showed that the addition of the above-mentioned mixtures has a positive effect on the yield of the produced liquid fuels (efficiency exceeding 80%). The obtained results may be the basis for further work related to the co-processing of polymer and petroleum waste, the number of which is growing in the world every year [2]. Ahmad et. al. [23] carried out polypropylene and polyethylene pyrolysis in the temperature range of 250 and 400°C. The experimental tests showed thatliquid products of the process consist mainly of C1 - C16 hydrocarbons. The percentage distribution of paraffins and olefins from polypropylene pyrolysis were respectively 66,55 % wt. and 25,70 % wt. Pyrolysis of polyethylene resulted in the production of 59,70 % wt. of paraffins and 31,90 % wt. of olefins.

3. Pyrolysis of PS, PVC and POM

Polystyrene (PS) is a type of vinyl polymer derived from a phenyl-containing monomer. It is widely used in various industries e.g. in construction as a foamed form and also for the production of everyday life objects like compact boards, toothbrushes, toys, or jewelry. Due to the aromatic ring in its structure, PS degradation can lead to valuable liquid products such as oils and gasoline additives [24]. Park et. al. [25] investigated the polystyrene pyrolysis process in a fluidized bed reactor with an auger reactor. The experiment aimed to find out the possibility of producing oils rich in BTEX (benzene, toluene, ethylbenzene, xylene) fractions. The laboratory test showed that the highest efficiency of the oil produced was achieved at 780°C. Moreover, the conducted fuel tests confirmed the possibility of using the produced oil as a replacement for diesel fuel by mixing with other liquids of low aromatic content. Prathib et al. [26] tested two types of polystyrene pyrolysis, i.e. conventional and microwave heating pyrolysis in the presence of activated carbon. The influence of the carbon to polystyrene ratio on the quality of the products and the amount of energy needed to carry out the pyrolysis process was investigated. The research showed that the use of microwaves can reduce the reaction time and additionally lowers the pyrolysis temperature by over 100°C. Discussed research has become an important factor in favor of using microwaves in terms of the amount of energy converted. Moreover, it has been found that the most suitable ratio of activated carbon to PS is 1:10 % wt. as this results in a liquid product mixture that can be used in the production of liquid fuels. Razzaq et al. [27] investigated the effect of the biomass and metal-modified zeolite catalysts on the PS pyrolysis [26]. The experiments were carried out in a stationary bed reactor in the temperature range of 500-650°C. The use of zeolites increased the efficiency of produced liquids and resulted in an increase in selectivity to monoaromatic hydrocarbons. Ferrosilicate (Fe-ZSM-5) turned out to be the most effective. At the same time, the coke yield in the process was reduced by more thanhalf when modified zeolites were used.

Polyvinyl chloride (PVC) is the world's third-most commonly used polymer, synthesized by polymerizing vinyl chloride. This material is widely used as a component for the production of carpets, skirting boards, windows and doors. It is also used in the medical and pharmaceutical fields for the production of syringes, probes, and tubing [21]. Due to the chlorine atoms present in the PVC molecule, a significant amount of hydrogen chloride and organic halogen derivatives are produced during the pyrolysis of PVC, which makes the process much more difficult to carry out [5]. Ji et al. [28] investigated the pyrolysis of PVC in the presence of transition group metal oxides. The addition of oxides significantly accelerated the decomposition of the material and reduced the amount of produced organic compounds, i.e. benzene, naphthalene, toluene, and anthracene. It also has been shown that the additives had a significant influence on the process temperature, which was increased and the reaction time was lower. Moreover, it was observed that the oxides present in the reactor absorb the released hydrogen chloride and increase the amount of residual carbon in the system. The oxides of vanadium and copper were the most effective. Torres et al. [29] developed a method of absorptive removal of chlorine and hydrogen chloride generated during the pyrolysis of PVC. The retention possibilities of typical absorbents, i.e. zeolites and aluminum oxide, as well as other substances, for instance, sodium bicarbonate, sodium

bicarbonate with zinc oxide calcium oxide were investigated. The test results showed much weaker retention of typical absorbents. In the case of zeolites and Al_2O_3, maximum chlorine retention of 20% at 550°C was achieved. Much better results were obtained with sodium bicarbonate and zinc oxide, as gas retention was then over 70 % wt. Gui et al. [30] investigated the PVC pyrolysis to find the most favorable temperature of the process. Various temperatures were considered. The heating rate of 1000 K/s resulted in the presence of mainly aromatic compounds involving three rings in products. Increasing temperature from 500 to 800°C resulted also in increased efficiency of aromatics compounds involving two rings of about 25%. During longer holding, time of 300 s obtained 42% higher yield of 2 rings group compounds in the temperature equal to 500°C.

Polyoxymethylene (POM) is a polymer that is widely used in engineering, construction, and automotive applications. It is mainly used for the production of materials with high strength requirements such as shafts, gears, or bearings and the production of fittings and piping. POM is obtained by polymerization of formaldehyde [21]. Berkowicz et al. [31] investigated the pyrolysis of POM in a fluidized bed made of cenospheres. Nitrogen and carbon dioxide were used as the fluidized agents. Depending on the fluidizing gas, the monomer recovery of the CO-rich gas was obtained. Under CO_2 atmosphere, the yield of formaldehyde was 90% at 400°C. Above 600°C, carbon incorporation from the fluidizing gas (CO_2) into the process products was observed. The amount of carbon in the products of the process was even doubled compared to carbon in the degraded polymer. That kind of process is characterized by a negative emission of CO_2. Hasegawa et al. [32] performed the pyrolysis process of POM by a combination of adding zeolites to the layout and using irradiated polymer material. The effect of zeolite on the pyrolysis temperature of irradiated POM had been estimated. The initial and end temperatures of the process were respectively 280°C and 312°C. Also, the use of only 320HOA or 320NAA (type Y) zeolite decreased the end temperature of the process compared to the pyrolysis without additives. It had been shown that pyrolysis of irradiated POM is faster with the presence of a special type of zeolite. It is suggested that the Y structure of the zeolite is a crucial factor in decreasing the end temperature of POM pyrolysis.

This mini literature review shows that pyrolysis processes can be carried out in various types of reactors. Rotary kilns and fluidized bed reactors seem to be future-oriented because of the continuous movement of wastes in both reactors. Examples of these types of solutions are shown in Figure 2. The most important advantage of carrying out pyrolysis in a fluidized bed is the low emission of harmful gases into the atmosphere, a large surface area of interfacial contact, and small temperature gradients. In the rotary kilns, wastes are heated and melted in a cylindrical combustion chamber [33]. The materials are mixed by gravity, and the main advantage of this technology is good aeration.

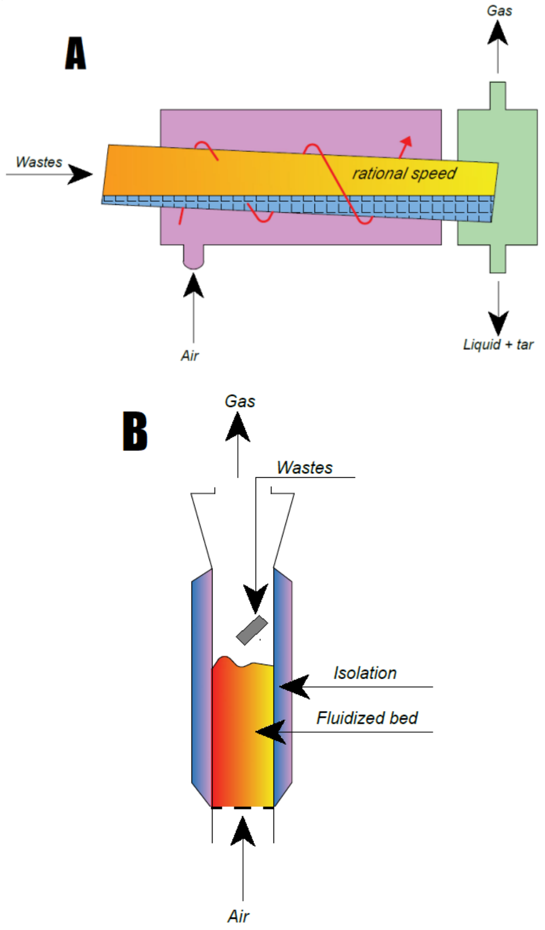

Figure 2. Examples of pyrolysis reactors schemes - rotary kiln (A), fluidized bed reactor (B)

Some of the studies discussed above are summarized in Table 1, paying attention to the process conditions and the efficiency of individual product groups.

Table 1. Examples of the conditions and process products during PE, PP, and PS pyrolysis

Plastic type	Temp. [°C]/ Pressure [bar]	Reactor type +bed material	Yield of products [%]			Ref.
			Gas	Liquid	Wax	
PE	450/1	packed bed + cement powder	17	82 mainly paraffins and olefins	1	[11]
		packed bed + cement powder + HBeta zeolite	30	67 mainly paraffins, and olefins	3	
		packed bed + clay + HBeta zeolite	60	35 mainly paraffins, and olefins	5	
		packed bed + silica sand + HBeta	81	15 mainly paraffins, and olefins	4	
PE	380/1	batch reactor	4	18 mainly paraffins and α-olefins	78	[12]
	340/21		3	96 mainly paraffins	1	

				and α-olefins		
PE	298 and 736^{1}/1	auger reactor with FBR (N$_2$ as an agent) + sand	75 mainly ethene, propene, 1,3-butadiene	23 mainly aromatic hydrocarbons	2	[14]
PP	293 and 712^{1}/1	auger reactor with FBR (N$_2$ as an agent) + sand	81 mainly hydrogen, methane, ethane	18 mainly aromatic hydrocarbons	1	[13]
PP	300/1	tubular reactor	70 mainly hydrocarbon gases and hydrogen	29 mainly aromatic hydrocarbons	1	[20]
PS	300 and 780^{1}/1	auger reactor with FBR (N$_2$ as an agent) + sand	13 mainly methane and ethane	86 BTEX fraction with 26% yield	1	[21]
PS	418/1	semi-batch reactor	9	83 paraffins, methyl styrene, condensed ring and benzene derivatives	8	[22]

Higher temperature refers to FBR and lower to auger reactor.

4. Discussion

The scientific literature shows that the distribution of pyrolysis products depends on the process conditions. The commercialization of the plastic waste pyrolysis would require maintaining homogeneous conditions of the process. Among the various pyrolysis systems, the fluidized bed seems to be the best option because it is characterized by small temperature gradients, large interface area, proper particle mixing and easy control over melting wastes. Recent studies on the degradation of polymer materials in the fluidized bed reactor indicate that it is very important to adjust the density of the fluidized bed to the density of the processing material. For this purpose, Żukowski et al. [34] propose the use of an innovative bed based on cenospheres. Due to the low density of the fluidized bed material, polymer waste freely sinks into the bed volume and the process takes place in the entire volume of the bed. So far, the authors have used the cenospheric fluidized bed only for POM pyrolysis, focusing the process on monomer recovery or production of CO-rich gas depending on the fluidizing medium used. It is suggested to use low-density fluidized bed to process other polymer materials. On the other hand, there are some ideas for increasing the efficiency of the pyrolysis process e.g. by adding a catalyst. The combination of these two facts leads to the idea of using catalytic cenospheres. In the scientific literature are publications on the cenospheres coating with catalytic materials such as Fe_2O_3 [35] or Ni [36] but in this topic, more research is needed.

5. Conclusion

Scientific literature has shown that plastics waste can be a valuable source of chemical raw materials. The pyrolysis process was tested in various reactor types, the influence of temperature, pressure, and the addition of an active substance was investigated. Depending on the process conditions, different yields of gas and liquid fractions were obtained. The composition of the received fractions differs depending on the type of processed polymer material. High efficiencies indicate the possibility of effective waste utilization with the simultaneous production of valuable semi-finished products. Depending on the plastic used, the liquid fractions are rich in paraffins, olefins, and even valuable aromatic compounds. More research on polymer pyrolysis are needed to deal with the enormity of plastic waste. It is believed that the interest in the process of pyrolysis of plastics will increase due to the necessity to transform the economy under the European Union policy as part of abandoning the use of non-renewable energy sources and the development of the circular economy.

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