Combustion behaviour of plastic waste – A case study of PP, HDPE, PET, and mixed PES-EL

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ABSTRACT

Plastic production is skyrocketing due to convenience needs and thus, the amount of plastic waste is increasing. There are several different methods to handle plastic waste, within which waste-to-energy processes are viable options in certain settings. However, the combustion of materials is a complicated and complex process that can take place in a number of ways. When exposed to heat, different polymers behave differently (*e.g.* have different emission factors) and thus, not all types of plastic waste recommended to be handled through combustion. Therefore, it is vital to achieve a deeper understanding of these processes in order to decide on the end use of plastic materials. Four common types of polymers were studied: polypropylene (PP), high-density polyethylene (HDPE), polyethylene terephthalate (PET), and polyester-elastane (PES-EL) mixture. The combustion properties of the plastic materials were investigated. The experiments were carried out in a laboratory electric resistance tube furnace at two different temperatures (650 and 900 °C). The behavior of these mainly single-use plastic wastes and the changes in the concentrations of CO, CO_2, NO_x, and O_2 flue gas components generated during combustion was studied to determine the quality of potential waste-to-energy processes.

1. Introduction

In a consumer society, access to consumer goods and services is easier and more widely available (Cavaliere et al., 2020). With the increase in the needs of the consumer society, the intensity of the production of plastics has also increased. Around half of the plastic items produced are designed for single-use purposes, while the other half is intended for long-term use such as in consumer products or infrastructure projects (Hopewell et al., 2009; Kedzierski et al., 2020).

The majority of single-use plastics (SUPs) are utilized in packaging, agricultural films, and disposable consumer products (Hopewell et al., 2009). Among these applications, packaging accounts for the largest portion, comprising approximately 40% of total consumption. The most commonly used everyday plastics for SUPs are polyethylene (PE), which is used for films, bags, houseware, and other items, as well as

polypropylene (PP) for food packaging, microwave containers, pipes, and so on, and polyethylene terephthalate (PET) for water bottles (Chen et al., 2021). Thus, PP, PE, and PET polymers are considered the most prevalent materials used for everyday plastics (Matthews et al., 2021).

As part of the European Union's directives, a waste management pyramid was created, which organizes options to handle waste according to how polluting the material to be treated. According to the waste pyramid, recycling plastics would be recommended, however the most popular way to treat plastic waste is to landfill, which followed by incineration (Adu-Boahen et al., 2014). In 2015, worldwide 79% of plastic waste was landfilled and 12% was incinerated and only 9% was recycled (Geyer et al., 2017). In China the rates of incineration of plastics was higher, up to 30% in 2014 (Idumah and Nwuzor, 2019). Based on Eurostat data (Eurostat, n.d.), in the EU in 2020, the most used method of polymer waste management is recycling (71.20%), followed

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by energy recovery (23.18%). The remaining minimum amount is incinerated or landfilled. Direct combustion of polymers leads to a reduction in the volume of waste by more than 90%, which is beneficial for its disposal in landfills (Żukowski et al., 2021). The reduction of volume of plastic waste with incineration takes place in properly designed waste incinerators and under strict operating conditions. High-tech waste incinerators are designed to prevent or minimize emissions of toxic substances (Idumah and Nwuzor, 2019; Swiss Federal Institute of Aquatic Science and Technology, 2008). Although some plastics have a similar or even higher calorific value than fossil fuel (Chen et al., 2021), several toxic substances are released during combustion, which require the use of expensive flue gas treatment equipment. The heat released from combustion is used to produce steam, which is then applied to generate electricity or residential heat (Swiss Federal Institute of Aquatic Science and Technology, 2008).

Natural and synthetic polymers, when exposed to a source of sufficient heat, will decompose or 'pyrolyse' evolving flammable volatiles (Price et al., 2001). The combustion process of solid fuels is very complicated and consists of drying, devolatilization, volatiles combustion and char combustion (Křůmal et al., 2019). Thus, the plastic material-air mix will ignite if the temperature is high enough. In polymer combustion, the chemical and physical phenomena are interconnected across the condensed and gas phases where combustible volatiles are respectively generated by polymer pyrolysis and the issued volatiles are oxidized (Camino and Camino, 2019). The following typical stages can be distinguished: thermal decomposition of liquid and solid combustible components, gas-phase ignition and combustion of gas products, heterogeneous ignition in presence of embers, and combustion of the carbonaceous residue of the fuel (Vershinina et al., 2021). The combustion of thermoplastics can be basically divided into four phases: softening, melting, thermal decomposition, and combustion (Baron et al., 2006). In the melting process, the physical properties of the material change: it converts from a solid to a liquid. At higher temperatures, the molecular structure of the substance decomposes, smaller molecular structures are formed, which are created when the thermal energy is higher than the energy required to break the bond (Izdebska, 2016). These two processes are heat absorbing, *i.e.* endothermic. As a combined effect of thermal decomposition and oxidation, formations of, inter alia, hydroxyl, carbonyl, aldehyde groups, peroxides or hydroperoxides, along the polymer chain or at its ends (Izdebska, 2016; Hawkins, 1964). Thus, formed smaller molecules are further oxidized with reacting violently with oxygen (exothermic process), resulting mainly in CO_2 and H_2O at the end of the process. Smoke formation during diffusion combustion of polymers depends on the polymer structure, the mechanism of thermal decomposition and the conditions of the pyrolysis and oxidation processes (Levchik et al., 2011).

The thermal behaviour of materials, as well as the gaseous and solid products produced during combustion or pyrolysis, are studied in a number of publications in the literature using laboratory-sized electric furnaces. For instance, Peng et al. (2016) looked into the PAH emission characteristics during the co-combustion of coal and MSW. In comparison to hard coal, Wielgosiński et al. (2017) studied the CO, NO_x, and TOC emissions from biomass burning at 5 different temperatures and 3 different air flow rates. Pingshuo bituminous was pyrolyzed in a nitrogen atmosphere by Xiao et al. (2020), and the impact of the pyrolysis temperature on the combustion properties and NO emission of Pingshuo semi-coke was also investigated. Furthermore, burned plastic waste had been pyrolyzed by Kiran et al. (2000).

The thermal degradation of plastic materials is in fact, in addition to processes such as chemical depolymerization, catalytic cracking, gasification, or partial oxidation, one of the most important types of recycling of raw materials (Izdebska, 2016). Therefore, the deeper understanding of such processes is important not only because of the recycling but for the proper storage and use of materials. Furthermore, it is important to know how the waste behaves during combustion, how much energy can be produced, and what emission indicators it has.

Despite the fact that a significant amount of plastic is thermally recovered in waste incineration plants, there is still insufficient information in the literature on flue gas concentrations from such processes. Therefore, the aim of this research is to find out the combustion behaviour of carefully selected mainly single-use plastic waste based on polypropylene (PP), high-density polyethylene (HDPE), polyethylene terephthalate (PET), and polyester-elastane (PES-EL) mixture, and to study the changes in the concentrations of CO, CO_2, NO_x and O_2 flue gas components generated during combustion, and thus, to determine the combustion quality.

2. Materials and methods

2.1. Materials

A primary consideration in the selection of plastics were what are those which can be most commonly found in the households. Households' plastics can be divided into two major groups: packaging materials, and textiles. Based on this the most popular plastics are polypropylene (PP), high-density polyethylene (HDPE), and polyethylene-terephthalate (PET).

The PP is the largest amount of plastic found in the households, which is mainly present as food packaging, sweet and snack wrappers (Chen et al., 2021). HDPE can be raw material of toys, milk bottles, shampoo bottles, pipes, houseware (Chen et al., 2021). Objects of use made of PET can be divided into two groups. It is called PET or PET resin when used as bottles, containers and other applications, while it is called polyester (PES) when applied as a fiber (Sarioğlu and Kaynak, 2017).

Thus, the selected materials were as follows.

(1) PP - plastic bowl, which is mainly used at food delivery,
(2) HDPE - cap of bottle,
(3) PET - mineral water bottle,
(4) and PES-EL - a textile containing 93% w/w polyester (PES) and 7% w/w elastane (EL) (Table S1).

It has to be noted, that PP and HDPE have a similar molecular structure, differing only in one methylene group (Table S1). These materials belong to the group of polyolefins (Troitzsch, 2004). Furthermore, the molecular structure of PET and the textile, which contain 93% w/w polyester, include aromatic hydrocarbons. In PES fibres, the molecules are mainly arranged in one direction, while in the case of PET they are in three directions ("The Essential Chemical Industry - Polymers," n.d.). The one dimensional structure of PET fiber achieved by using the spinning method (Sarioğlu and Kaynak, 2017), while blow moulding techniques are applied to stretch PET, which will lead to three dimensional structures, resulting in the rapid growth of light and unbreakable PET bottle manufacturers (Park and Kim, 2014). Like polyester, elastane contains an aromatic hydrocarbon and consists of soft and hard segments. The special elastic property and wide industrial application of elastane fibers are directly related to the chemical structure of the material which includes a multiblock copolymer comprising alternating soft polyester or polyether and hard polyurethane–urea segments (Otaigbe and Madbouly, 2009).

2.2. Methods

The following methods have been employed to investigate the combustion properties of the materials.

(1) Determination of the moisture content with Mettler Toledo HB43-S Halogen moisture measurement.
(2) Determination of ash content of samples by heating to 550 ± 15 °C to constant mass according to MSZ EN 14775: 2010.

(3) Carlo Erba EA1108 type ultimate analyzer was used for the measurement of the C, H, N, S, and O content in samples according to MSZ EN 15104:2011 standard.

(4) The calorific test was performed using a Parr 6200 type isoperibol oxygen bomb calorimeter according to MSZ EN 14918:2010 standard.

(5) Effect of the increasing temperature on the behaviour of the samples was examined with a SYNLAB IF2000G heating microscope. The heating rate was 5 °C/min in air atmosphere.

(6) The thermal behaviour of the materials was studied by using a MOM Derivatograph-C. For TG, specimens of about 28–35 mg were heated from 25 °C to 900 °C at a heating rate of 10 °C/min. Heating was performed in air atmosphere.

In this study the combustion process was tested in a Hőker Cső 350/900 type electric resistance tube furnace, which had a horizontal working chamber and a programmable temperature of the furnace temperature controller, thus allowing the temperature set at a given time to be maintained. The analysis of the concentration of the gases from the combustion process (CO_2, O_2, CO, NO_x) was performed with a Horiba PG 250 gas analyzer (Mentes and Póliska, 2020). Investigation of the combustion process happened at a combustion air volume flow of 160 dm^3/h and at a firebox temperature of 650 °C, and 900 °C. The appropriate flow rate was determined based on experimental experience and stoichiometric calculations, which enabled to safely investigate of the combustion behavior. To ensure accurate measurements, a higher amount of oxygen than stoichiometric combustion oxygen was introduced into the firebox. The flow rate of oxygen (in the form of air) was kept constant at both higher and lower temperatures.

All in all, six experiments were performed in each setting. In the case of combustion experiments, the evolution of gaseous flue gas components emitted during the combustion of plastics was investigated. The experimental system is shown in the Supplementary Material (Fig. S1) and described in previous studies (Mentes et al., 2022a).

2.3. Emission value calculation

In order to compare the emissions of gaseous pollutants from different experimental materials, an emission factor was calculated. This parameter can be used to eliminate the differences in sample weights and experimental times. According to the EPA ("US EPA - Unites States Environmental Protection Agency," n.d.) 'an emission factor is a representative value that attempts to relate the quantity of a pollutant released to the atmosphere with an activity associated with the release of that pollutant. Emission factors usually are expressed as the weight of pollutant divided by the unit weight, volume, distance, or duration of the activity that emits the pollutant.'

The emission factors can be relative to time (Berdowski et al., 1999), to fuel energy, (Win et al., 2012; Nuutinen et al., 2014), and also to fuel mass (Wielgosiński et al., 2017). In this research, the emission factor based on the mass of the fuel was used. The emission factor for each pollutant can be determined by the following equation (Wielgosiński et al., 2017):

$$w_z = \frac{Q \times c_{z,average} \times \tau}{m_{sample}} \left[\frac{mg\ gas\ pollutant}{g\ combusted\ sample} \right] \qquad (1)$$

where Q is the combustion air flow rate [m^3/s], $C_{z,\ average}$ is z average emission concentration of the flue gas component [mg/m^3], τ is sampling time [s], m_{sample} is the mass of the sample placed in the boat [g].

3. Results and discussion

3.1. Combustion properties of plastics

For all materials intended for combustion, it is important to know their combustion properties (Table S2). The two materials with the highest C content are PP and HDPE, which are associated with the highest heat of combustion. The combustion parameters of PP and HDPE are similar, as their molecular structure differs only in one methyl group. The heat of combustion of the materials with aromatic structure (PET, PES-EL) lags behind that of polyolefins, as the resolution of aromatic rings requires much more energy than that of a chain structure (Mark, 2007; Huang et al., 2018).

Some polymeric materials may contain a certain amount of water, which is mainly adsorbed from the environment. Polyolefins such as PE and PP do not contain easily hydrolysable chemical bonds and thus have very low water adsorption. However, certain types of PET tend to have higher water adsorption due to their hygroscopic properties (Baschek et al., 1999; Stan, 2020).

The ash content of the artificial material is minimal when compared to natural materials, but can be attributed to the presence of inorganic substances, either as contaminants from previous use or incorporated into the molecular structure of the sample (e.g. paint substances). It is worth noting that PP, HDPE, and PET were thoroughly washed prior to the experimental phase, and the textile materials were also freshly washed to minimize the presence of possible contaminants.

3.2. Thermal analysis of plastics

Thermoanalytical examinations were carried out by using a derivatograph, which was supplemented with examinations performed by a heating microscope. Microscopic examination can also be used to trace visually the behaviour of the tested plastics under the influence of temperature increase. PP and HDPE have similar derivatograms due to their previously mentioned similar molecular structure (Fig. 1.). The major changes on the derivatograms are determined and the corresponding temperatures were collected (Table S3).

PP completely melts at 261 °C, but the melting process reach its maximum intensity at 159 °C which is in agreement with the literature (melting point in the temperature range of 160–170 °C) (Akay, 2012; Dalhat and Al-Abdul Wahhab, 2017; Phulkerd et al., 2014). PP thermally degrades to volatile products above 261 °C and completely in a single step finishing at about 483 °C with a maximum rate at 465 °C (Yang et al., 2013; Zanetti et al., 2001). Lighter molecules released from PP by thermal decomposition burn at 427 °C, and at this point more than 50% of the sample weight was lost (Fig. 1). Above this temperature, no significant change is seen on the heating microscopy images. According to previous studies, PP decomposes between 330 °C and 410 °C, and its inflammation occurs in the temperature range of 390 °C–410 °C (D. Price et al., 2001). In another work, a slightly different temperature range was determined, and according to this, the thermal decomposition of PP occur in the range of 290–390 °C (Ramazanov et al., 2010). Furthermore, in argon atmosphere the thermal decomposition happened above 400 °C (Jakab et al., 2000), while in nitrogen atmosphere the same process occurred between 310 °C and 475 °C (Longo et al., 2011).

In the case of HDPE, the mass of the sample remained almost stable till 340 °C (Fig. 1). Above this temperature, mass loss started and increased abruptly above 340 °C up to 519 °C (Durak and Durusoy, 2012). The most intense part of the melting process takes place at a lower temperature (143 °C). From 340 °C, the process of thermal decomposition of HDPE begins, during which the material begins to intensively fragment, which lasts up to 519 °C. In the case of HDPE, there are examples in the literature of derivatographic experiments mainly in a nitrogen atmosphere, where the thermal decomposition is in the range of 375–510 °C (Kumar and Singh, 2013; Banat and Fares, 2015; Eyubova et al., 2011), and the intense thermal decomposition temperature is between 460 °C and 478 °C (Kumar and Singh, 2013; Banat and Fares, 2015; Kumar and Singh, 2014). In this case, it is no need to consider a combustion phase.

Examination of data clearly indicates that polyolefines degrade in a single stage process. The thermal stability of PP is lower than that of PE.
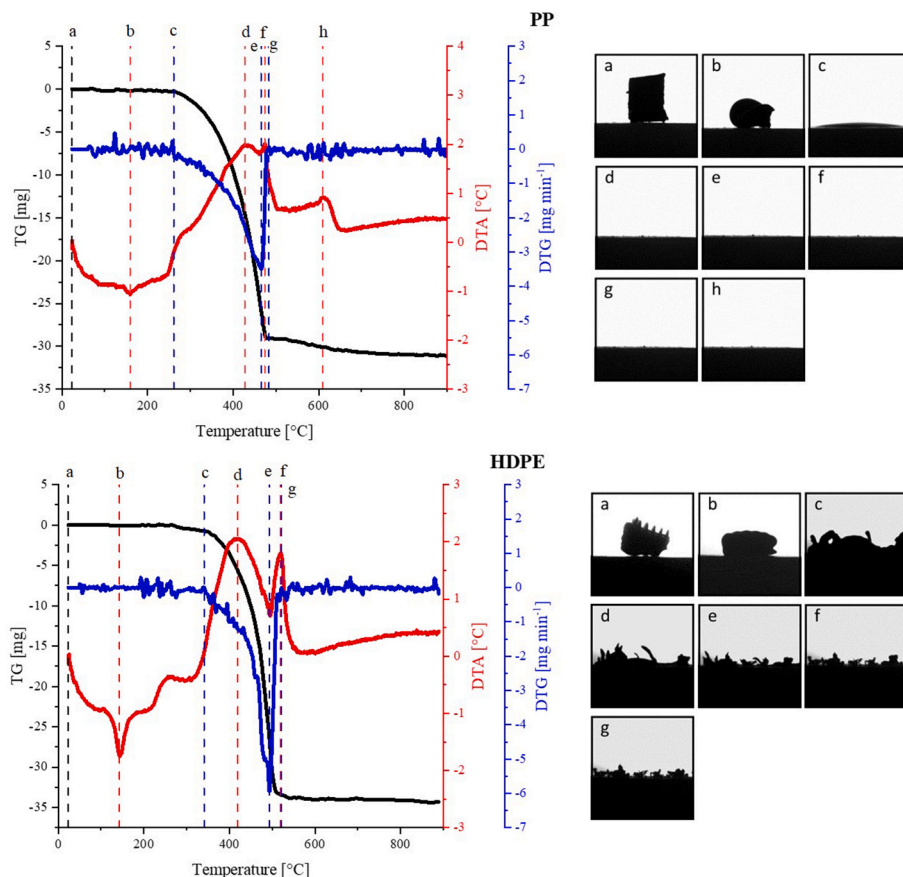
Fig. 1. Derivatograms and heating microscopy images of polypropylene (PP), and high-density polyethylene (HDPE).

In contrast to PP, the thermal decomposition of HDPE more spectacular, macro decomposition is also visible. The difference between their molecular structure significantly influences the degradation process. The presence of the methyl group in all alternating carbon atoms in the main PP backbone allows the formation of more unstable secondary free radicals, thereby favouring intramolecular hydrogen transfer reactions as well as interaction with oxygen, resulting in lower thermal stability for hydroperoxide groups (Ray and Cooney, 2018).

The thermal degradation of polyolefines was described previously by random chain scission and a radical chain mechanism (Yang et al., 2013) which include chain fission, radical recombination, allyl chain fission, intermolecular hydrogen abstraction, midchain β-scission, end-chain β-scission, intramolecular hydrogen transfer, and dehydrogenation (Ray and Cooney, 2018; Tarani et al., 2017). Polyolefins exposed to heat melt with a sudden emission of gaseous hydrocarbons (Żukowski et al., 2021). The decomposition starts with the removal of volatile parts through a radical chain process propagated by carbon centered radicals originated from carbon–carbon bond scission at weak links (Nie et al., 2014; Bernstein et al., 2008).

In the case of PP the branching of the chain leads to a decrease in the thermal stability of the polymer (Żukowski et al., 2021). The pyrolysis products of PP at 470 °C are methyl-propylene, pentene, and dimethyl-pentene (Nie et al., 2014) which can be formed by following a secondary radical degradation pathway (Ray and Cooney, 2018). Furthermore, other major volatile products are found during the degradation, such as dimethyl-heptene, propylene, methyl-pentene, and in much smaller amount, isobutene (Troitzsch, 2004).

The main products observed during the degradation of PE include ethylene and higher oligomers, which arise from hydrogen transfer to different positions along the polymer chain, thus, one observes propylene, butene, pentene, hexene, and so forth, as well as a series of alkanes

(Troitzsch, 2004). In air, the degradation is strongly enhanced by the presence of oxygen, which begins at 345 °C. A small increase in the sample mass by about 0.5% occurs just prior to degradation due to oxygen absorption followed by hydroperoxides formation (Ray and Cooney, 2018). At low retention time, gases and volatile products such as CO_2, acetaldehyde and low molecular mass products (the monomer propylene) are formed (Tarani et al., 2017).

The derivatograms of PET and PES-EL are also recorded (Fig. 2), and the major changes on the are determined, while the corresponding temperatures were collected (Table S4). Unlike polyolefins, materials with an aromatic ring melt at higher temperatures. PET melting was the most intense at 255 °C. From 377 °C, the thermal decomposition of PET started, and coupled with strong gas evolution. During the thermal decomposition process, the spread material inflated to a spherical shape (Fig. 2). Thermal decomposition process finished at 484 °C. From 484 °C, an intense macro decomposition (fragmentation) begins, which last up to 593 °C. This process can be considered as a second stage of thermal decomposition, during which the intense gas formation is indicated by the suddenly created extremes observed in the DTG curve (Fig. 2).

For PES-EL, the most intense point of the melting process is experienced at 251 °C. From 356 °C, the thermal decomposition process starts with intense gas formation like in case of PET (Fig. 2). At the beginning of the thermal decomposition phase, the material inflated to spherical shape, because of the gas formation, then it started the macro decomposition which lasts up to 502 °C. After the stage of the first thermal decomposition, fragmentation continues with less intensity, and at 545 and 875 °C two combustion peaks can be seen.

For PET and PES, experiments in air have shown that thermal decomposition starts at different temperatures (325 °C (Jabarin and Lofgren, 1984) and 380 °C (Miranda Vidales et al., 2014)), but
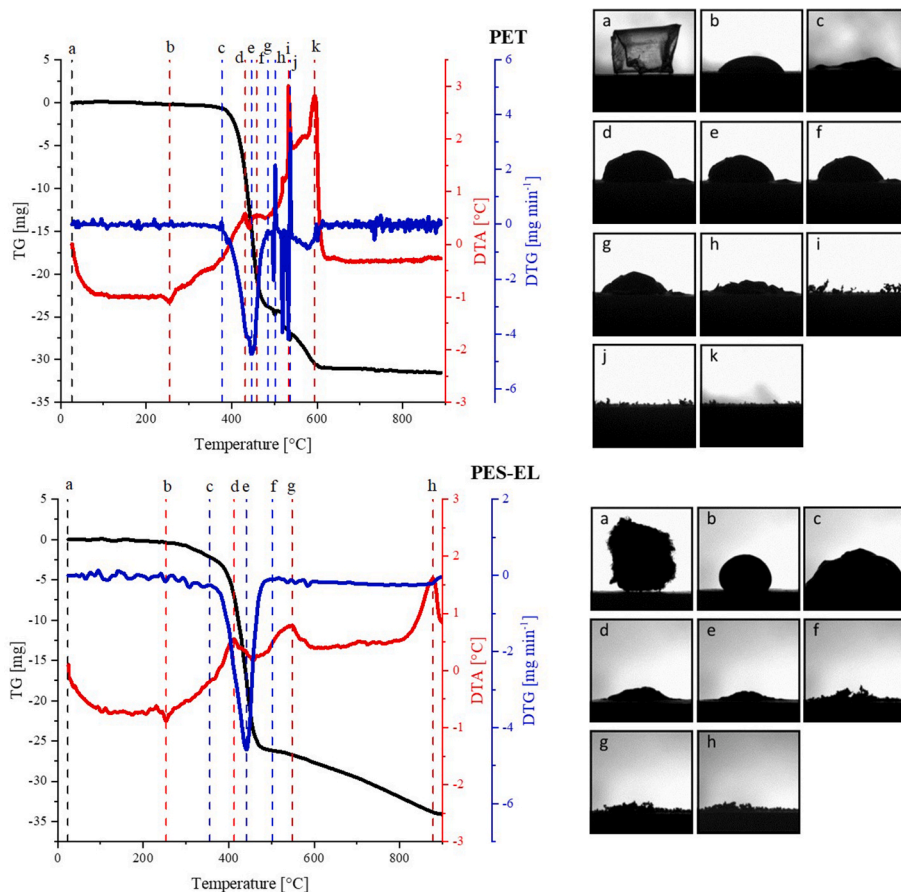
Fig. 2. Derivatograms and heating microscopy images of polyethylene-terephthalate (PET), and polyester (PES) - elastane (EL) mixture (PES-EL).

regardless of the nature of the atmosphere, the thermal decomposition end up to about 500 °C (Jabarin and Lofgren, 1984; Miranda Vidales et al., 2014; Faraj et al., 2011; Pires et al., 2015). There are several exothermic peaks in the DTA curves, regardless of the material, which can be explained by the fact that the products which continuously formed during thermal decomposition gradually ignite, thus, heat is released.

In the case of PET, and PES-EL two stage degradation profiles were noticed (Ganeshan et al., 2018). Pyrolysis of PET, and PES involves a network of heterolytic and homolytic chain cleavage reactions (Hujuri et al., 2013). Thermal decomposition of PET initiates by scission of an alkyl-oxygen bond following random-chain scission (Ray and Cooney, 2018). Kinoshita et al. (1992) found that at 400 °C the CO_2 ratio was higher than at 420 °C. Moreover, the presence of phthalic acid dimethyl ester and terephthalic acid diethyl ester as well as alkyl vinyl esters were detected at the temperature range of 400–500 °C. The gases, which are formed at temperatures above 500 °C with the proportion of benzoic acid, are bigger than in the initial stage of decomposition. Studies by Sovová et al. (2008) have shown that the products of PET decomposition at 500 °C are methane, ethane, ethyne, formaldehyde, carbon dioxide, carbon monoxide and water. In addition at 800 °C (*i.e.* above the inflammation point) a more efficient burning process can be achieved and the main combustion products are carbon oxides and water.

In summary, that in the case of the PET carbon dioxide and aromatic esters were detected in the initial stage of decomposition. The benzoic acid and esters are formed during thermal degradation, while benzoic acid, anhydrides, aromatic and aliphatic acids, and alcohols are formed by thermo-oxidative degradation (Ray and Cooney, 2018; Kinoshita et al., 1992).

3.3. Evolution of concentration of flue gas components during combustion

From a combustion technology viewpoint, the combustion of plastics can be divided into two main stages: ignition and combustion phase. During the ignition stage, air and fuel will interact, CO is formed by a rapid exothermic reaction. The subsequent reaction of CO to CO_2 is slow and requires adequate residence time to achieve completion. (Ndiema et al., 1998). The other flue gas components, the maximum concentration of CO_2 and the minimum concentration of O_2 indicate the intense combustion phase (Mentes et al., 2022b), while the NOx concentration reaches its maximum when the firebox temperature is also at its maximum. In general, all the factors that increase the temperature of the flame also increase the amount of NOx too. NOx is a product of high-temperature combustion processes, and it is further oxidized to NO_x at temperatures below 650 °C (István, 2001). Since the temperature of the combustion chamber was set to less than 1300 °C during the experiments, only NOx formation from the nitrogen content of the fuel should be taken into account (Mitchell et al., 2016; Monedero et al., 2018).

To compare the combustion stages of each material, it was necessary to determine the time, which is assigned to the appearance of the CO flue gas component. While at 650 °C the ignition and intense combustion phases can be clearly distinguished, at 900 °C the firebox temperature was high enough that the combustion of the material took place so fast that the ignition and intense combustion phases coincided (Fig. 3). The evolution of flue gas concentrations at a jacket temperature of 650 and 900 °C was recorded for both studied plastics (Fig. 3). At lower temperatures (650 °C) in the case of materials which consist of aromatic structure (PET and PES-EL), the CO and CO_2 emissions are zero or very minimal. This can be explained by the fact that the temperature of the firebox is lower than the jacket temperature of 650 °C, and the boat
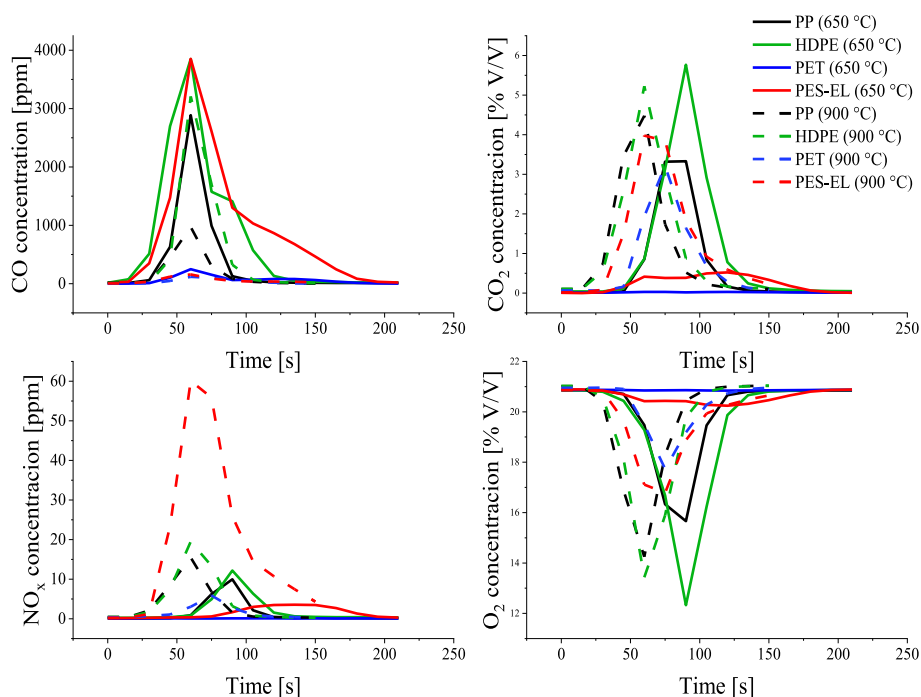
Fig. 3. Evolution of flue gas component concentrations at a jacket temperature of 650 and 900 °C during the combustion of polypropylene (PP), high-density polyethylene (HDPE), polyethylene-terephthalate (PET), and polyester (PES) - elastane (EL) mixture (PES-EL).

pushed into the firebox with the plastic sample further reduces it. As a result, the temperature of the firebox becomes so low that the input energy is used to break down the bonding energies. According to previous studies, in case of PET the level of oxygen concentration does not alter the mode of the pyrolysis process, although it accelerates it (Senneca et al., 2004). The pyrolysis process is followed by the char combustion. A particular type of combustion that occurs without the generation of flames is smouldering, or glowing combustion (Rosace et al., 2015). At low temperatures in the presence of sufficient oxygen, incandescence occurs (D. Price et al., 2001) and the oxygen directly attacks the surface of a condensed-phase fuel (Rosace et al., 2015). This phenomenon is an induction period (involving smouldering) before flaming ignition. The plastic waste could ignite and burn strongly if the temperature on the polymer surface is hotter than 200 °C (Hull and Kandola, 2008) and at the centre of polymer is 220 °C, meanwhile white strong malodorous smoke appears (Kamiya et al., 2010).

For materials with an aromatic ring, at a jacket temperature of 650 °C, yellowish-white smoke can be detected in the quartz tube, resulting from flameless combustion. According to the observation of Hull et al. (2007), this flameless combustion is a non-flammable oxidative pyrolysis, which products are rich in organic and partially oxygenated species. It is found that very high level of organic species found in the flue gas from combusted PS (which consist of aromatic rings too) at 350–400 °C, suggesting that flaming combustion did not occur. According to the literature, similar pale yellow smoke detected during PET destruction at the temperature range of 400–700 °C. In the investigation of Dzięcioł és Trzeszczyński (Dzięcioł and Trzeszczyński, 2001), the analyses have shown that the emitted smoke is a complex mixture of aromatic compounds: terephthalic acid and its esters, benzoic acid, *p*-substituted benzoic acid, low oligomers of PET, and acetylated and polycyclic aromatic hydrocarbons. During the pyrolysis of PET at 200 °C (*i.e.* at lower temperatures), the formation of fractions with larger C number (C16-C22 and C24-C32) is relatively higher than that of the lighter components (Hujuri et al., 2013). The negligible evolution of the C5-C10 fraction is due to the extra stability of the polymer, as there are resonance-stabilized structures in the backbone of the chain that require more energy to overcome the attractive forces. At temperatures above

300 °C, the intensity of cracking reactions increases, which increases the amount of lighter hydrocarbons.

The difference in CO and CO_2 concentration between PET and PES-EL is due to the difference in the orientation of the molecules (1D, and 3D) and the elastane content. The binding energy of PES-EL arranged in a 1D structure is easier to decompose, and thus, a small part of the materials leaving during thermal decomposition is oxidized to CO_2. This exothermic process reaches its maximum temperature later (120 s), which is indicated by the change in the concentration of the NOx component (Fig. 3). If the amount of energy input is not sufficient to end the oxidation of C, the energy released by the formation of CO_2 cannot catalyze the formation of NOx. Since the elastane part of the material produces a minimal amount of CO_2 in a prolonged time interval, N will also be released from the material later in the form of NOx. In contrast, polyolefins are burnt at a jacket temperature of 650 °C, the firebox reaching the maximum temperature in 90 s (Fig. 3). Decomposition is a multi-step process that involves the breakdown of molecules and reaction with oxygen, and at the end of the process leads to the generation of CO. However, in the case of PET, decomposition results in larger molecules that do not further decompose into CO at this temperature. In the case of PES-EL at the end of a completed decomposition process, there is not enough heat to oxidize CO further. Additionally, Hull and Kandola (2008) explained that a well-ventilated environment leads to a high CO ratio.

At a jacket temperature of 900 °C, the materials burn faster. In the case of HDPE combustion, the maximum concentrations of the flue gas components (excluding NO_x) are close to the values emitted at a jacket temperature of 650 °C. Irrespective of the jacket temperature, it can be said that reaching the maximum of the CO_2 concentration curve depends on the molecular structure of the plastic and the energy required to break the bonds. The maximum CO_2 concentrations vary as a function of the decomposition temperatures determined from the DTG curve during thermoanalytical studies (Table S5). The easier the resolution of the binding energies in the polymer, the faster the CO_2 component concentration reaches its maximum. It can be seen that aromatic compounds require more energy to break the bonds than aliphatic materials (Mark, 2007; Huang et al., 2018).

The emission factor values from the combustion of the polymers were also determined (Table S6). As it has already been observed for the evolution of concentrations, CO and CO_2 emissions are minimal for PET at low temperatures. It has to be noted that by increasing the temperature usually the CO emission factor values decreased while the CO_2 emission factor values increased except in case of HDPE.

The combustion quality of materials can be determined from the ratio of the CO_2/CO emission factor. If the CO_2/CO ratio is low, the process generates large amounts of CO, which indicates poor combustion conditions. If this ratio is high, a significant amount of CO_2 is generated, indicating favourable combustion conditions. It was found that the C/O and C/H element ratios of plastics relate to the ratios of CO_2/CO emission factors emitted at low and high temperatures (Fig. 4). In case of the aromatic materials (PET and PES), the C/O ratio is small, which can be mainly explained by the ester groups found in their molecular structure. In contrast, the aliphatic C/O ratio is about 7.5 or greater. Examining the C/H ratio, it can be said that this value is higher for aliphatic compounds than for materials with an aromatic structure.

At a jacket temperature of 650 °C, polymers with an aliphatic structure burn better than those with an aromatic one. Raising the jacket temperature to 900 °C, this trend changes, the combustion of aromatic materials becomes of better quality. The combustion quality of aliphatic compounds is less affected by the increase in temperature. In the case of HDPE, the quality of combustion does not change, the CO_2/CO emission ratio is around 20, regardless of the temperature, while in the case of PP emission ratio triples. In contrast, the CO_2/CO ratio of aromatic compounds increased 60-fold–70-fold with increasing jacket temperature.

4. Conclusions

The aim of this study is to describe the characteristic phenomena that accompany the combustion of carefully selected plastic waste. Four different types of materials, polypropylene (PP), high-density polyethylene (HDPE), polyethylene-terephthalate (PET), and polyester (PES) - elastane (EL) mixture (PES-EL), were selected and their combustion was studied. The changes at notable temperatures during the thermoanalytical studies were visually illustrated. Looking for similar

relationships between thermoanalytical studies and combustion experiments, it was found that the chemical structure of the materials strongly determines the combustion properties and the flue gas concentration from combustion.

There are no examples in the literature for the combination of heating microscopy and derivatography investigation. Visual tracing of the behaviour of the sample under the influence of temperature increase has been shown to be as important as the quantification of physical and chemical processes by MOM-type derivatography. Thermoanalytical studies have shown that materials composed of aliphatic compounds (PP, HDPE) decompose at lower temperatures than those composed of aromatic compounds (PET, PES-EL). Aliphatic compounds melt with increasing temperature and then, begin intense fragmentation. In contrast, aromatic compounds are inflated after melting. During that intense gas evolution is observed when lighter volatiles are removed from the samples. As the intense gas formation is completed, they are further pyrolyzed by fragmentation (macro-decomposition), similar to aliphatic compounds.

During the combustion experiments, it was found that at a jacket temperature of 650 °C, after inserting the boat containing the sample, the firebox cooled so much that it resulted a flameless process during the combustion of aromatic materials. Furthermore, a significant amount of organic matter was emitted, which appeared as white-yellow smoke. The flue gas contained little or no CO and CO_2 components. At a jacket temperature of 900 °C, besides low CO emissions the aromatic compounds burned with flames. The decomposition temperature detected by thermoanalytical tests strongly determines when the flue gas reaches its maximum CO_2 content. The higher the temperature of thermal decomposition, the later it reaches the stage of intense combustion (maximum CO_2 concentration). The quality of combustion was determined by measuring the ratio of CO_2/CO emission factors which is introduced in this study. Combustion of aliphatic plastic waste at a jacket temperature of 650 °C was ideal, while combustion of aromatic compounds was better at 900 °C. With the increase of the jacket temperature, the combustion of aliphatic compounds improved only slightly, although the combustion quality of aromatic plastic waste increased significantly. The low C/H ratio of the substances is aliphatic, while the low C/O ratio
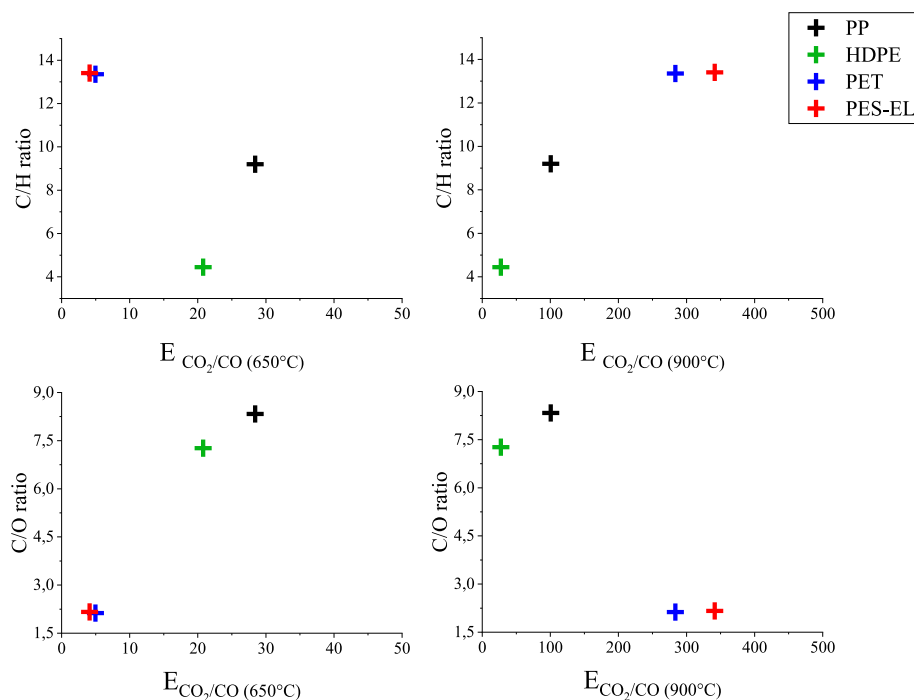
Fig. 4. Relationship between the element content of studied plastics, polypropylene (PP), high-density polyethylene (HDPE), polyethylene-terephthalate (PET), and polyester (PES) - elastane (EL) mixture (PES-EL), and the CO_2/CO emission rate.

is an indicator of the aromatic compounds. Overall, the combustion temperature, the C-, H-, O-content and the molecular structure of plastic wastes strongly determine the quality of combustion.

The results from the incineration experiments provide guidance on how to thermally recover different types of waste. In the first approximation, for thermal combustion (heat generation), the utilization of polymers with aliphatic chain containing molecular structure (PP, HDPE) is recommended, while the polymers with aromatic structure (PET, PES) are suggested to be used as secondary raw materials for pyrolysis due to their strong gas formation properties. However, any recovery method will be chosen, the material should be recycled for as long as possible, and it should be disposed of as a last resort.

Another area for future research is the examination of solid particles and PAHs identified during combustion that were not included in this study. Additionally, our observation of white smoke calls for further investigation, which we plan to conduct using GC-MS in the future.

CRediT authorship contribution statement

Dóra Mentes: Conceptualization, Experiments, Formal analysis, Writing – original draft, Supervision. **Gábor Nagy:** Experiments, Formal analysis, Writing – review & editing. **Tamás J. Szabó:** Formal analysis, Writing – review & editing. **Enikő Hornyák-Mester:** Writing – review & editing. **Béla Fiser:** Writing – review & editing. **Béla Viskolcz:** Writing – review & editing. **Csaba Póliska:** Investigation, Conceptualization, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgments

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2023.136850.

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