

THE EFFECT OF USING PYROLYSIS OILS FROM POLYETHYLENE AND DIESEL ON THE POLLUTANT EMISSIONS FROM A SINGLE CYLINDER DIESEL ENGINE

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ABSTRACT. The paper presents data related to the results obtaining following the testing of a commercial diesel fuel and a fuel obtained by chemical recycling of polymers waste based on polyethylene in a single cylinder diesel engine. The concentrations of CO, CO₂, hydrocarbons, NO_x and smoke have been determined in the exhausted gases. The experimental results showed that using synthesized polyethylene-based fuel, the emission of pollutants was smaller compared to the emissions when using commercial diesel fuel, except the concentration of hydrocarbons which was higher.

Keywords: *recycling, pyrolysis, polymers waste, fuel, pollutants, diesel engine.*

INTRODUCTION

The Directive 2008/98/EC on waste, provides a framework for the environmentally sound management of waste, including the preferred hierarchy of waste management options. Waste management strategies should focus both on reducing the amount of produced waste and its hazardousness [1]. The amount of used plastic materials (PM) continues to increase every year. Using and producing more PM involves the producing of higher quantities of waste. Because waste generation can not be avoided, waste should be reused as much as possible.

Quantitative targets for the recycling and recovery of packaging waste were set for 31 December 2008 (55 - 70 % recycling, 60 - 75 % recovery), but Romania is far from reaching this target. Directive 94/62/EC regulates the Packaging and Packaging Waste in European Union, while Government Decision

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no. 349/2002 revised and amended in 2004 (899/2004) is applied in Romania, resulting an imperative necessity for identification of an appropriate method for recycling of each type of waste, because new European regulators are setting increasingly stringent standards to reduce the impact of PM waste in the environment.

Current statistics for Western Europe estimate the annual consumption of plastic products at almost 100 kg/person for a total of over 39.1 million tons [2, 3]. Over 78 wt % of this total corresponds to thermoplastics (low and high density polyethylene LDPE-HDPE, polypropylene PP, polyvinyl chloride PVC and polyethylene terephthalate PET) and the remaining 22 wt % to thermosets (mainly epoxy resins and polyurethane) [2]. This generates a vast waste stream, estimated at around 21.5 million tons, that needs to be effectively managed. Today 61 wt % of all the waste plastics generated in Europe are still disposed of to landfill, a management alternative that generates serious environmental problems due to their low density, resistance to biological degradation and combustible nature.

Even though in European Union collection and recycling of waste is already a current practice; plastic materials recycling represents nevertheless an important objective of European and Romanian researches, in order to elaborate the most efficient technologies from economical end environmental point of view. In the Sectorial Operation Programs (POS) of Romania, the first priority is „The development of regional systems for water and waste management” [4].

One of the potential options for polyolefin waste management is chemical recycling for fuel production.

Plastic pyrolysis is a method for plastic materials recycling that can be applied to a variety of polymers. A wide range on degradation products can be obtained. The composition of degradation products depends on many factors such as: polymer type, temperature, catalysts.

Pyrolysis technology has reached by now a state of development that opens more favorable possibilities for waste disposal than some of the traditional techniques. Special complex waste materials which contain plastic, such as contaminated materials, can now be properly recycled by pyrolytic treatment.

Pyrolysis, also termed thermolysis, is a process of chemical and thermal decomposition of polyolefins, generally, leading to smaller molecules. In most pyrolysis processes, air is excluded, for reasons of safety, product quality and yield. Pyrolysis can be conducted at various temperature levels, reaction times, pressures and in the presence or absence of reactive gases or liquids, and/of catalysts. PE pyrolysis proceeds at low (<400°C), medium (400–600°C) or high temperature (>600°C). The pressure is generally atmospheric. The thermal decomposition of PE yields gases, distillates and char, albeit in widely variable relative amounts. These can be applied as fuels or petrochemicals.

Polyolefin resins contain only carbon, hydrogen and additives, such as antioxidants and UV stabilizers. Depending on the polymers or polymer mixtures fed and the operating conditions used, yields can vary widely. Both gaseous

and liquid products are mixtures of numerous different compounds. The problem of fractionating these effluents and upgrading to commercial specifications, while separating undesirable impurities must be investigated on a case-by-case basis, but until then, the developing of appropriate methods for using the pyrolysis product as is, can be of high interest.

By thermal degradation of HDPE and XLDPE, a significant quantity of waxes is formed, and the quantity of liquid products is smaller when using LDPE and LLDPE. This suggests that PE which contain lateral chains (such as LDPE and LLDPE, respectively), by pyrolysis process a higher quantity is transformed in to liquid products, comparing with HDPE and XLDPE (with linear chains). The use of catalyst leads to the increase of liquid and gaseous degradation products.

Catalytic degradation of LDPE, in controlled conditions, leads to the obtaining of valuable aromatic products (namely: benzene, toluene, xylene and feedstock for petrochemicals [5-7]). By pyrolysis of LDPE from plastic bags results a mixture of compounds, whose concentration differs depending on pyrolysis conditions (temperature, catalysts) resembling the composition of commercial gasoline [6].

Even if the studies regarding chemical recycling of plastic waste were systematically done, a few studies regarding the impact of using plastic fuels derived in engines are presented in the literature [8-14].

Moriya et al [13] shown that cracked PE (polyethylene) can be used for engine as a 30-40 % blended fuels with Diesel fuel. Other studies involved a thermal recycling system of plastic wastes. In this case, the plastic waste is melted and mixed with heavy oil resulting a fuel for Diesel engine generator systems [12,14].

Santaweesuk and Janyalertadun used for their study a 4-Stroke, compression ignition, water cooled, single cylinder engine. By changing the engine speed they concluded that compression ignition engine is able to run with distilled waste plastic oil, because physical characteristics of distilled waste plastic oil (DWPO) is similar to diesel oil. Engine fueled with distilled waste plastic oil exhibits lower brake thermal efficiency but increase average when setting nozzle pressure and results are higher than average in brake specific fuel consumption for distilled waste plastic oil (DWPO) but lower average when setting nozzle pressure. Exhaust gas emission increase average when setting nozzle pressure in distilled waste plastic oil (DWPO) compared to diesel operation and results are that the nozzle setting pressure can help reduce better the emissions [15].

Jani, Patel, Rathod also used a 4-Stroke, water-cooled diesel engine of 5 HP rated power and established that using the synthesized fuel, the best results have been obtained in CI engine with blends containing 30 % pyrolysis product regarding both performance and emission of diesel engine [16] .

The novelty of our researches consists in the study of a fuel obtained from polyolefin in our laboratory and testing of synthesized fuel in diesel engine air-cooled, without any modification, without blending with commercial diesel fuel. Our team conducted both studies regarding fuel production and testing.

RESULTS AND DISCUSSION

The yield in liquid degradation products of interest was up to 93 %, [17]. The composition of the liquid fuel resulted following the pyrolysis process has been determined using gas chromatography and revealed that the fuel contains both saturated and unsaturated compounds [18, 19]. The main difference between pyrolysis product and commercial products consists on the small quantities of light fraction (C₃-C₅) up to 4 %, and heavy fraction (C₂₃-C₃₅) up to 12 % [17].

The formation of the above presented hydrocarbons in the pyrolysis product can be explained by the mechanism of thermal cracking of PE.

Polyethylene is formed from $-(CH_2-CH_2)_n-$ monomer units (mers), containing also a small number of terminal methyl ($-CH_3$) groups. Thermal cracking of PE leads to saturated and α unsaturated hydrocarbons due to scissions of PE long chains following a multistep radical chain reaction with all the general features of such reaction mechanisms: initiation, propagation, branching, and termination steps [18].

Random chain scission takes place in the initiation stage, conducting to the formation of free radicals [18].



where: P_n is a polymer containing n mers; $\bullet R_r$ and $\bullet R_{n-r}$ are radicals containing r and respectively $(n-r)$ mers.

In the propagation stage the chain suffer other scission reactions due to intramolecular or intermolecular H transfer and unzipping depolymerization resulting new radicals, saturated and unsaturated (P_m , P_{n-j} , P_1) hydrocarbons.



Intramolecular H transfer reaction involves the transfer of a hydrogen atom within a single polymer chain forming an unsaturated (P_m) polymer and a new terminal radical (R_{r-m}).

Intermolecular H transfer involves the transfer of one H atom from a inner carbon atom (not a terminal one), between neighbors polymer chains forming a new saturated polymer (P_r) and a new inner located radical that breaks up into an unsaturated polymer (P_{n-j}) and a new radical.

The last propagation mechanism (unzipping depolymerization) that involve the depolymerization with the formation of monomers and smaller chain polymers has no important impact in the specific case of polyethylene since the yield in liquid fraction is so high, being specific for polymers with functional groups such as polystyrene and polymethyl methacrylate [19] that suffer depolymerization reactions conducting to monomers with a high yield.

The termination stage occurs due to recombination of monomers or to disproportionation. While in the case of recombination two radicals react forming a new polymer, in the case of disproportionation one of the radicals donate a hydrogen atom, forming an unsaturated hydrocarbon (P_n), while the other radical accept the hydrogen atom forming a saturated hydrocarbon (P_s).



The mechanism of polyethylene degradation is not so simple since the unsaturated hydrocarbons can participate to polymerization reactions conducting to the formation of higher molecular weight hydrocarbons in tar or coke [20, 21].

For the limitation of the re-polymerization reactions, the gaseous product resulted following the pyrolysis was rapidly cooled using a Low Temperature Ethylene Glycol/Water Recirculation System. The rapid cooling conducted to the increasing of the yield of liquid pyrolysis product reducing the quantity of solid residue.

One can conclude that thermal cracking of polyethylene occurs by random chain scissions conducting to a broad spectrum of saturated and unsaturated hydrocarbons.

Both breaking and re-polymerization takes places due to free radicalic reactions [22].

For the investigation of thermal degradation of PE, FT-IR spectra for polyethylene and the pyrolysis product were recorded (fig. 1).

In polyethylene spectrum one can observe only vibrations due to metylene groups [23].

The absorption band cantered at 2916 cm^{-1} can be assigned to C-H asymmetric stretch, while the one from 2848 cm^{-1} to C-H symmetric stretch from metylene groups. Around 1467 cm^{-1} can be observed a doublet due C-H bends for metylene groups.

In the spectrum of the fuel the asymmetric/symmetric C-H stretch due to metylene groups are shifted towards higher wavenumbers 2922 cm^{-1} (compared to 2916 cm^{-1}) and 2853 cm^{-1} (compared to 2848 cm^{-1}) respectively, while C-H bends can be observed at 1460 cm^{-1} . The presence of metyl groups can be clearly observed at 2957 cm^{-1} (C-H asymmetric stretch) and 2873 cm^{-1} (C-H symmetric stretch), 1379 cm^{-1} (C-H symmetric bend), proving the formation of new CH_3 terminal groups. The presence of unsaturated compounds can be observed due to the presence of alkenyl absorption band (C=C stretch at 1643 cm^{-1}), vinyl terminal stretch at 3075 cm^{-1} , vinyl C-H out of plane bend at 989 and 908 cm^{-1} and vinylidene C-H out of plane bend.

No oxidation processes took places since the C=O absorption band is missing (around 1700 cm^{-1}).

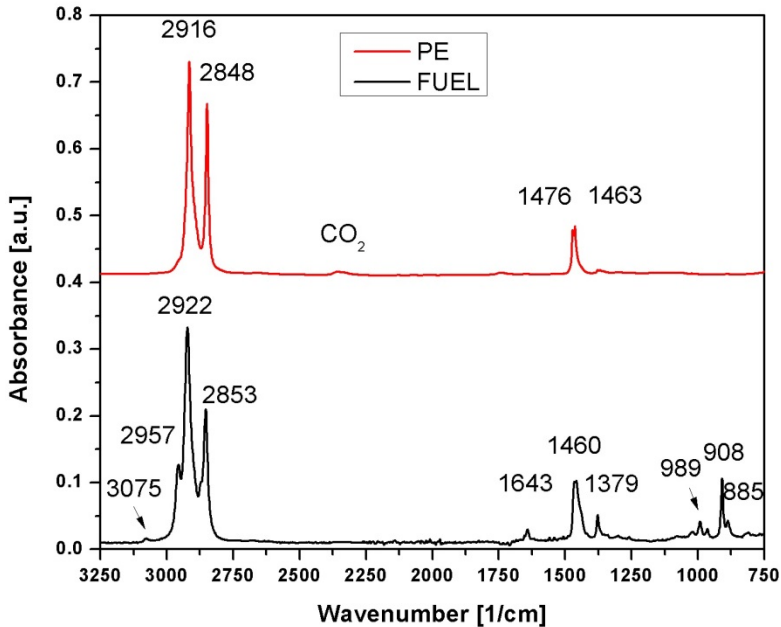
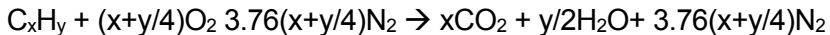


Figure 1. FT-IR spectra of PE and PE derived fuel

Other properties of the fuels, determined according to ISO 3104 and ASTM D445, revealed the similarity of the pyrolysis product properties with the property of commercial fuel [24] and the similar dependence of density, kinematic and dynamic viscosity on the temperature [25].

The reactions for the formation of unburned hydrocarbons (UBHC), CO₂, CO, NO_x following fuel burning in internal combustion engine are briefly presented subsequently [26].

General exothermal reaction of fuel burning in the presence of oxygen from air leads to the formation of CO₂:



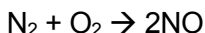
The presence of unburned hydrocarbons (UBHC) in the exhausted gas can be explained by the incomplete burning reaction. The temperature in the cylinder is not uniform, since the temperature at the time of combustion is 2500-3000°C, while the walls of cylinder are around 200°C. The exhaust starts at 1000 °C, but cools quickly after leaving cylinder and UBHC are formed in the quench zone.

The formation of CO can be explained both by the incomplete burning of hydrocarbon and the combustion in the condition of deficit of oxygen. Even if there is sufficient oxygen for total combustion of fuel, the formation of CO is possible because of the fact that fuel injected in later part of the injection does

not find enough oxygen due to local depletion in certain parts of the combustion chamber. More than that, when the temperature reaches certain values the dissociation of CO₂ can occur:

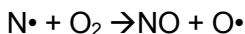
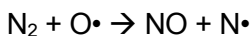


Nitrogen oxides are formed due to the presence of nitrogen from the air and nitrogen compounds from some fuels. Generally the used fuels have small concentration of nitrogen compounds. NO forms when fuel combustion takes places at high temperatures leading to the oxidation of nitrogen molecules following the general reaction:

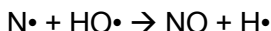


This reaction is strongly endothermic, having the free enthalpy of $\Delta H=90,4$ KJ/mol. The quantity of the formed NO markedly decreases with the decrease of the combustion temperature.

The reaction is much more complex taking places due to the formation of free radical of oxygen in the flame by the dissociation of oxygen molecules. The following reaction takes places initiated by a radicalic mechanism, as described by Yakov Borisovich Zeldovich:



An important role is played by the reaction with hydroxyl radicals (HO•) that can be formed at high temperatures from combustion chamber:



The smoke is a direct product of burning process while it forms as a pollutant in the process of burning propagation and quantitatively intensify on the path of gases evacuation.

Engine tests

One concludes that the properties of the oil obtained by thermal degradation of PE are close enough (in accordance with EN 590 regulation) with the properties of commercial diesel fuel [25], leading to the conclusion that the synthesized fuel can be used as it is or as mixtures with commercial diesel fuels, in diesel engines. During tests, the hourly fuel consumption and the concentration of pollutants from exhausted gasses were determined as a function of engine's speed.

Hourly fuel consumption

The determination of hourly consumption (figure 2) has been made by weighting the quantity of consumed fuel in an establish time period and a specific speed rate. The test was timed, obtaining the time for fuel consumption, and in the final, the hourly consumption (c_h) has been calculated with the relation:

$$c_h = \frac{\tau \cdot m_\tau}{3600} \quad [g / h]$$

where: τ is the time, in seconds, during which m_τ fuel has been consumed; m_τ - the mass, in grams, of the fuel consumed in the time interval τ

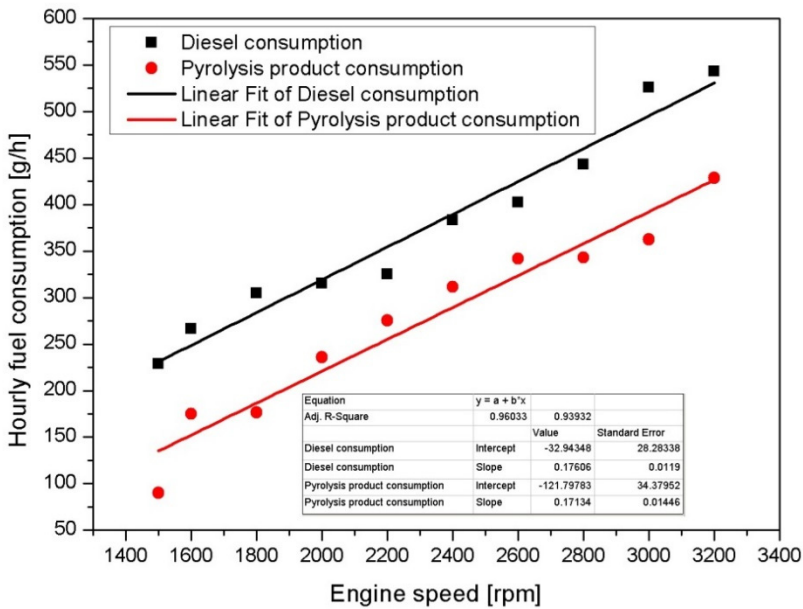


Figure 2. Comparison between the hourly consumptions of the two fuels

From the figure 2 it can be seen that at all engine speeds the hourly fuel consumption achieved from polyethylene-based fuel is significantly lower than of diesel.

Carbon monoxide emissions

Carbon monoxide forms when the fuel burnig process is incomplete and it is considerate a toxic gas due to his capacity to react with hemoglobin from the blood. The reduction of CO from exhausted gases is important both for human and environment protection.

Figure 3 presents the concentration of CO measured from the exhausted gases as a function of engine speed. The plot contains the values obtained following the measurements and the curve obtained applying second order polynomial regression. As can be seen from the figure 3, for engine speed smaller than 2400 rpm, the concentration of carbon monoxide is almost the same for experimental PE-based fuel and the commercial fuel.

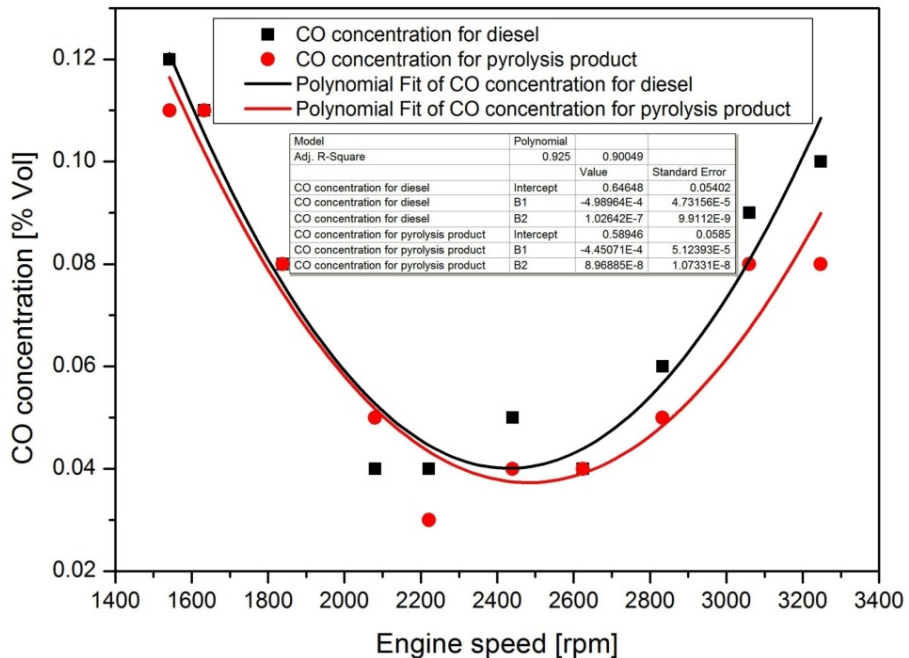


Figure 3. The concentration of CO from the exhausted gas as a function of engine speed

Increasing the speed engine, the concentration of CO resulted when pyrolysis product was used as fuel suffered a slightly reduction comparing with the concentration of CO resulted when commercial diesel fuel was used. The reduction of CO emissions when plastic derived fuels was used in a four stroke, compression ignition, water cooled, single cylinder, change speed diesel engine has been observed also by Santaweek Chumsunti, and Adun Janyalertadun [15]. In our experiment, the smallest CO emissions can be observed for both fuels when the engine speed is about 2400 rpm. The highest CO concentration was attained for engine speed of 1400 rpm. The increasing of engine speed at values higher than 2400 rpm leads to an increase of CO emission concentration.

Carbon dioxide emissions

Carbon dioxide forms when the combustion of fuels is complete. When the speed rate of the engine is smaller than 2200 rpm, the difference between the emissions of CO₂ is insignificant for the two fuels.

The increasing of engine speed determined the increase of the concentration of CO₂ for both fuels, but the concentration was higher when commercial diesel fuel was used. One can conclude that a positive effect on the concentration of CO₂ in the exhausted gases can be observed in the case of pyrolysis product (figure 4).

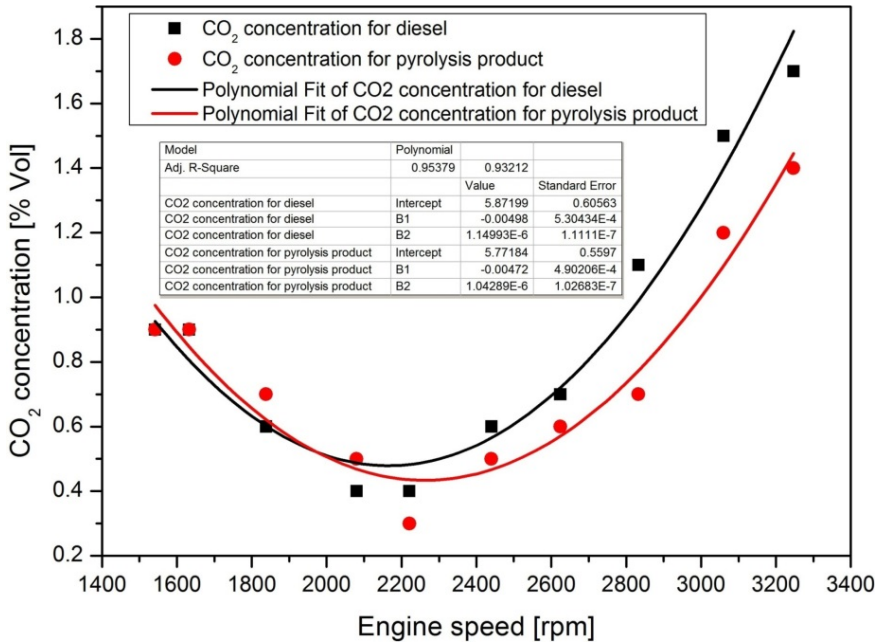


Figure 4. The concentration of CO₂ from the exhausted gas as a function of engine speed

Smoke's opacity emissions

The formation of smoke in the exhaust gases of the engine is related to the incomplete burn of the fuel, when particles of unburned carbon are present into the gases.

The smoke's opacity resulted following the combustion suffered an important decrease when the PE-based fuel was used (figure 5), for all speed rates. The reduction of the smoke concentration can be explained by the composition of the plastic derived oil, that contains only hydrocarbons with C and H, and the combustion took places in better conditions.

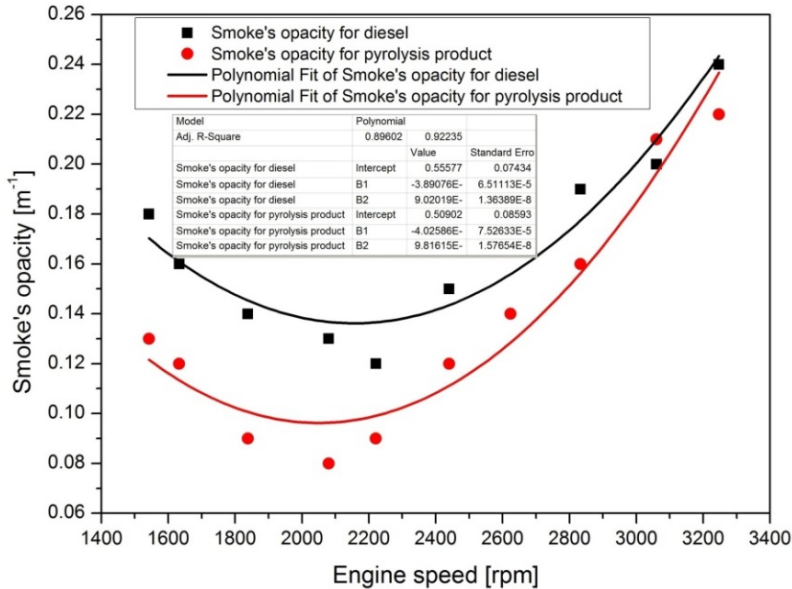


Figure 5. The smoke's opacity from the exhausted gas as a function of engine speed

Unburned hydrocarbons emissions

The pyrolysis product contains a fraction of hydrocarbons (HC) with high molecular weight, higher than the heavy fraction from the commercial fuel.

The existence of the high molecular pyrolysis products conducted to the increased of UBHC concentration in the exhausted gases compared to the concentration resulted when commercial diesel fuel was used (figure 6). Probable, heavy chains of pyrolysis product can not be broken in the engine combustion process.

Nitrogen oxides emissions

The results obtained for nitrogen oxides (NO_x) emissions are presented in figure 7. It can be seen that, when the engine is powered with fuel obtained by pyrolysis, the concentration of NO_x suffered a significant decrease comparing to the concentration resulted by burning the commercial diesel fuel.

One can explain this situation through the composition of pyrolysis product that is formed only by saturated and unsaturated hydrocarbons, without nitrogen products, compounds that can suffer proper combustion reactions inside the internal combustion engine.

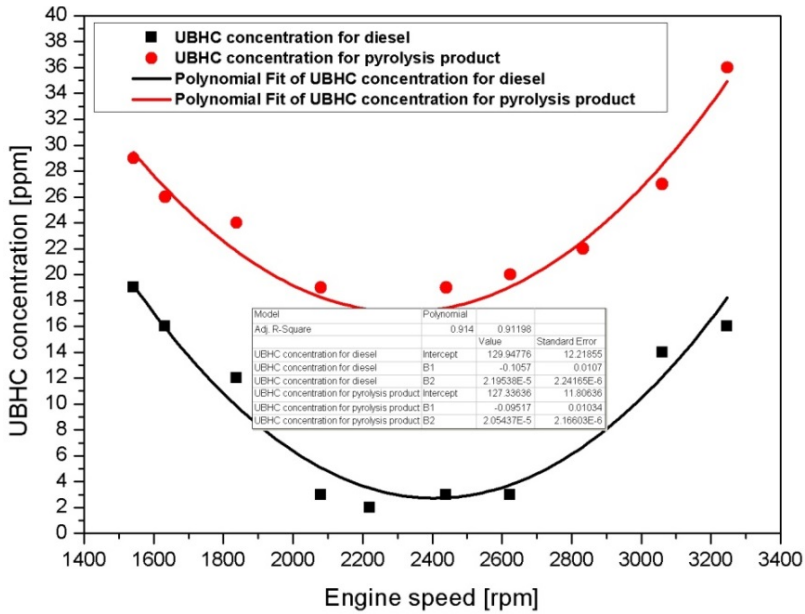


Figure 6. The concentration of HC from the exhausted gas as a function of engine speed

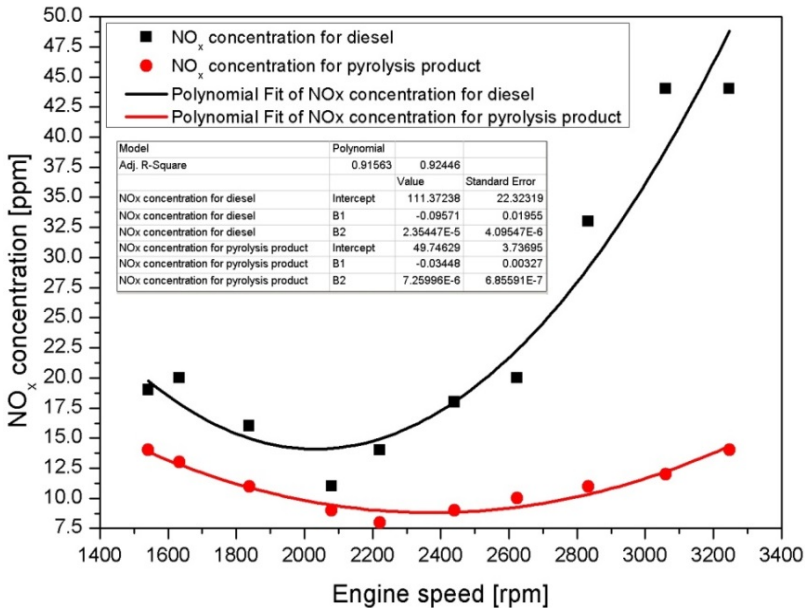


Figure 7. The concentration of NO_x from the exhausted gas as a function of engine speed

CONCLUSIONS

A comparative study regarding the emissions of CO, CO₂, smoke's opacity, hydrocarbons and NO_x in the exhausted gases, when a commercial diesel fuel and an experimental fuel obtained from polyethylene by pyrolysis, was made.

One concluded that pollutant emissions with the exception of hydrocarbons is smaller when the experimental PE-based fuel was used, leading to the conclusion that PE plastic waste can be successfully converted into a fuel with good properties for using as an alternative fuel in diesel engines with no other additives or blending with commercial diesel fuel.

At all engine speeds the hourly fuel consumption achieved from polyethylene-based fuel is significantly lower than of diesel.

For engine speed smaller than 2400 rpm, the concentration of carbon monoxide is almost the same for experimental PE-based and the commercial fuels, respectively. Increasing the speed engine, the concentration of CO resulted when pyrolysis product was used as fuel suffered a slightly reduction comparing with the concentration of CO resulted when commercial diesel fuel was used.

The increasing of engine speed determined the increase of the concentration of CO₂ for both fuels, but the concentration was higher when commercial diesel fuel was used.

The smoke's opacity resulted following the combustion suffered an important decrease when the PE-based fuel was used, for all speed rates.

The high molecular pyrolysis products from PE-based fuel conducted to the increased of the unburned HC concentration in the exhausted gases from the engine.

The concentration of NO_x suffered a significant decrease in the case of pyrolysis product comparing to the concentration resulted from commercial diesel fuel.

Another important advantage of the experimental fuel is the reduced hourly consumption comparing to the consumption of diesel commercial fuel.

Large scale application of chemical recycling of PE for fuel production can be a good alternative for solving the problems related to the impact of plastic waste on environment, leading to fossil fuel saving in conditions of proper engine efficiency, due to the high quality of pyrolysis product regarding combustion behavior.

EXPERIMENTAL SECTION

The obtaining of the fuel

A bench scale installation has been used for the obtaining of fuel starting from PE. Details regarding the pyrolysis process applied for fuel production were presented in our previous studies [17, 24, 25, 27, 28].

IR spectra of polyethylene and pyrolysis product were recorded with a Spectrum BX II FT-IR spectrophotometer provided with an Attenuated Total Reflectance (ATR) accessory.

For fuel testing, a single cylinder diesel engine model Kipor KM 186FAG, has been used. The main specifications and technical data for the diesel engine are presented in table 1. All tests were conducted on the engine running without load.

A gas analyzer AGS 688 Brain Bee was used to determine the concentration of different pollutants gases. For the determination of the opacity from the exhaust gases, a DSS 2 opacimeter from the diagnosis equipment SMP 4000 was used.

Following the experiments the concentration of pollutants from exhausted gasses was determined as a function of the engine speed.

Table 1. Technical specification of the diesel engine used for tests

Item	Technical specifications and data
Model	KM186FA
Type	In line, 4-stroke, single cylinder, air-cooled, direct injection, diesel engine
Cylinder bore x stroke (mm)	86 x 72
Displacement (cc)	418
Rated speed (rpm)	3000
Rated power kW	5.7
Compression ratio	19:1
Starting system	Recoil and electric starter
Rotating direction (viewed from flywheel)	Clockwise

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