



Fuel properties of the oils produced from the pyrolysis of commonly-used polymers: Effect of fractionating column

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ABSTRACT

The effects of using a packed column as a fractionating system on thermal degradation of commonly-used polymers have been studied. The polymers investigated were virgin polyethylene (LDPE and HDPE), polypropylene (PP), and polystyrene (PS), as well as two post-consumer polymer mixtures, namely municipal plastic waste (MPW) and scrap tire. The experiments were carried out under atmospheric pressure in a 6L semi-batch stainless steel vessel equipped with the fractionating system. The system led to total elimination of wax formation, color improvement, and reduction in the density of the resultant pyrolysis oils. The oils were analyzed according to ASTM D86, the standard test method for boiling range distribution of petroleum fractions. The obtained values were compared to the Iran standard limits established for diesel (ISIRI4903) and gasoline (ISIRI4904). Furthermore, Fourier transform infrared spectroscopy (FT-IR) was utilized to determine the chemical composition of the oils. Finally, the spectra of the generated tire and MPW-derived oils were compared with the spectra of conventional fuels, gasoline and diesel.

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1. Introduction

After population explosion and the introduction of modern lifestyle in the 20th century, plastics and rubbers have become one of the most widely used commodity materials known to humankind. The total global production of plastics has increased 183 times, rising from 1.7 Mt in 1950 to 311 Mt in 2015 [1]. It is also predicted that if the current rate of plastic manufacture continues, there will have been another 33 billion tons in circulation by 2050 [2]. Among different types of plastics, polyethylene (PE) and polypropylene (PP) constitute nearly 50% of the total plastic production, and in terms of usage, packaging applications together with construction sector are the two major consumers, nearly 60% of plastic market [1].

Similarly, the production of rubber-made goods, particularly automobile tires, has seen substantial growth in the last half century. The annual supply of tire is reported almost 1.6 billion units [3] and projected to reach 2.9 billion units in 2020 [4]. The overall demand for natural and synthetic rubber is estimated at 28.75 million metric tons [5,6], of which nearly 60% is used for tire production.

Polymeric products have different lifespans ranging from a couple of months for packaging applications to as long as 50 years for underground and exterior cables, and even 100 years for plastic pipes in construction sector. However, as the yearly production of polymers is not expected to drop in the foreseeable future, it can be inferred that the life service of polymeric products will not be a deciding factor in determining the yearly amount of polymeric waste. In other words, regardless of widely different lifespans of polymeric products, the total amount of polymeric waste will become equal to the overall production of polymeric products in the not distant future. This means that in each year, the amount of past long-service life products (60 wt.% of total production [7]) is added to the amount of yearly short-service life products. Therefore, ultimately, the total annual amount of polymeric waste will reach every year's production now estimated at nearly 340 million tons [1,5,6].

This massive amount of polymeric waste has caused, and will continue to cause, numerous environmental problems such as water/soil contamination, ocean pollution, as well as energy/financial loss. Hence, not only is it vitally necessary to mitigate polymeric waste's negative environmental and health consequences but also it is equally important to utilize its vast energy content.

To address this problem, many methods have been employed and tested. However, because of two major reasons, namely high capital investment and low quality of the eventual products, many

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of such practices were proved to be unviable. As a result, many countries have decided to adopt mechanical recycling and incineration as the main approaches to reduce the amount of polymeric waste being sent to landfills.

Soon after, new problems emerged. It has become obvious that mechanical recycling is only applicable to clean and pure polymeric waste. Notwithstanding, due to polymer ageing, the practicality of this method even for pure waste is also limited. This means that the products made from recycled materials are inferior and of low quality, and re-recycling would lead to the production of extremely low-grade products with highly limited market.

Similarly, using incineration technologies entails undesirable consequences. Employing these processes not only does demand for costly off-gas treatment, but also leads to mass production of greenhouse gases (GHG) mainly composed of carbon dioxide and nitrous oxide. Regarding the recent Paris pact, now almost all countries are obliged to move towards low-carbon economy. Therefore, incineration processes, despite their high maturity level, have again come under fierce attacks not only from public and environmentalists but from global organizations as well. Experts have been struggling for years to optimize incineration technology by combining it with electricity production. However, the impact of this modification on the mitigation of GHG has proved to be insignificant, and this method is still responsible for producing the largest amount of GHG in comparison with the other two main waste management strategies known as landfilling, and composting [8].

In the light of these facts, the need for developing a third solution to fill the gap between practicality and morality has become vital even more. To do so, de-polymerization (monomer recovery), gasification, and pyrolysis processes are suggested. De-polymerization requires extremely clean and pure feed, while gasification involves huge investment, not to mention constant inspection and high operational costs of gas storage and transportation. These drawbacks have made the pyrolysis process an attractive option for developing one sustainable solution to meet both the needs of waste disposal and energy supply in an environmentally friendly manner. Pyrolysis process or thermal cracking has been the center of attention for many years. This process is versatile and lies between mechanical recycling and incineration in terms of the intensity of feedstock pretreatment. Pyrolysis can be utilized for producing chemical feedstock, synthetic crude, adsorbent as well as liquid fuel. Notwithstanding different pyrolysis's products, researchers' major focus have been on producing and optimizing pyrolysis oils as the most valuable product of pyrolysis. Despite this, extensive research has also been conducted for finding usage of the char by-produced during the process. these research is also vitally important to guarantee the feasibility of the process, particularly in the case of tire pyrolysis [9–16].

In the scientific literature, pyrolysis process is studied via different experimental and theoretical techniques. These methods include investigating the effects of temperature and pressure [17–21], utilizing the design of experiments (DOE) methodology and statistical analysis [22–25], using different reactor types and catalysts [26–34], co-pyrolysis with other materials [35–38], and examining novel approaches such as microwave assisted pyrolysis [39–42], molten metal bed [43–45], steam thermolysis [46], and catalytic hydro-reforming [47–52]. Regardless of all these pyrolysis methods, this process can be categorized into two main groups known as thermochemical decomposition and catalytic cracking, both of which suffer from various drawbacks. On the one hand, the oil produced from thermal degradation does not meet the physical and chemical requirements determined by fuel standards [53–56]. On the other hand, direct use of catalysts for polymer cracking would lead to rapid deactivation of catalysts due to both coke deposition and catalyst poisoning [31,32,57–60]. Further-

more, bulky nature of polymers is reported as one of the main causes of steric and diffusional restrictions for polymeric molecules in the porosity of zeolites [60]. Hence, as new findings imply, the most efficient approach would be through utilizing a two-stage pyrolysis process by which, in the first stage, waste are treated thermally resulting in the production of a suitable pyrolysis vapor. Subsequently, the vapor is upgraded by catalysts ending in the production of a higher value product. The idea behind employing two stage method which was first practiced by Bagri and Williams [61], is to simultaneously avoid excessive catalyst deactivation and obtain marketable products. Utilizing two-stage method is proven to improve the overall efficiency of pyrolysis process. This can be explained by the fact that the method provides the option of optimizing both the operating conditions of thermal cracking and catalytic upgrading stages separately [31,62–66]. Nevertheless, catalyst deactivation is still problematic due to coke deposition in the upgrading stage.

Thermal cracking of plastics in the first stage ends in the formation of long-chain heavy hydrocarbons [67,68]. In the second stage, catalytic upgrading stage, the waxes with large molecular weights form a yellow waxy coke on the surface of catalysts blocking zeolite pores and deactivating acid sites [69].

Castano et al. [70] observed that there were two types of coke formation: Type (I), amorphous long molecules of aliphatic nature deposited on the exterior of the zeolite crystals, and Type (II), molecules of aromatic nature deposited on both the interior and exterior of the catalysts.

Type I coke are formed by random scission mechanisms, whereas bimolecular reactions such as olefin oligomerization, aromatization, and condensation are attributed to type II coke formation [31].

Ibáñez et al. [63] reported the two mechanisms for the formation of coke in the catalytic upgrading stage: (I) initiation mechanism leading to degradation of the paraffin wax produced in the thermal cracking stage, and (II) steady coke formation mechanism leading to the condensation of light olefins also produced in the thermal cracking stage. They suggested that the rate of deactivation in the catalytic upgrading stage is dependent upon concentrations of the wax and light olefins both produced in the thermal cracking stage.

Deactivation of catalysts due to coke deposition can significantly shorten the useful life of catalysts resulting in the need for frequent catalyst regeneration. This drawback can make the upgrading stage quite costly and thus impractical for scale-up [68].

Eliminating wax formation in pyrolysis through the use of fractionating system, first introduced and practiced by Undri et al. [42]. The researchers studied the effects of using a fractionating system in microwave assisted pyrolysis of HDPE, PP, and tire [41,42]. They observed that the fractionating unit had two valuable functions: (I) Condensation and reflux of high boiling fractions of the generated pyrolysis vapor, and (II) Avoidance of micro-particle transport. The two characteristics resulted in the production of clear, and transparent pyrolysis oils with lower density and viscosity compared with the oils produced from classic pyrolysis process.

This paper investigates the effects of using an insulated column packed with stainless steel pall rings as a fractionating system. The effects of employing the fractionating unit were studied for a classic thermal cracking process which is still the most used process in the scientific literature. The fractionating unit was utilized for increasing the residence time of heavy components existed in pyrolysis oils. The process was designed to eliminate wax formation while maximizing oil yield in order to produce the pyrolysis oils suitable to be upgraded in the catalytic upgrading stage. This paper aims to provide a criterion for comparing the physical and chemical characteristics of pyrolysis oils obtained from commonly-used polymers utilizing a fractionating system.

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