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Production of an alternative fuel by the co-pyrolysis of landfill recovered plastic wastes and used lubrication oils

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ABSTRACT

This work is a preliminary study for the development of a co-pyrolysis process of plastic wastes excavated from a landfill and used lubrication oils, with the aim to produce an alternative liquid fuel for industrial use. First, thermogravimetric experiments were carried out with pure plastics (HDPE, LDPE, PP and PS) and oils (a motor oil and a mixture of used lubrication oils) in order to highlight the interactions occurring between a plastic and an oil during their co-pyrolysis. It appears that the main decomposition event of each component takes place at higher temperatures when the components are mixed than when they are alone, possibly because the two components stabilize each other during their co-pyrolysis. These interactions depend on the nature of the plastic and the oil. In addition, co-pyrolysis experiments were led in a lab-scale reactor using a mixture of excavated plastic wastes and used lubrication oils. On the one hand, the influence of some key operating parameters on the outcome of the process was analyzed. It was possible to produce an alternative fuel for industrial use whose viscosity is lower than 1 Pa s at 90°C, from a plastic/oil mixture with an initial plastic mass fraction between 40% and 60%, by proceeding at a maximum temperature included in the range 350–400°C. On the other hand, the amount of energy required to successfully co-pyrolyze, in lab conditions, 1 kg of plastic/oil mixture with an initial plastic mass fraction of 60% was estimated at about 8 MJ. That amount of energy is largely used for the thermal cracking of the molecules. It is also shown that, per kg of mixture introduced in the lab reactor, 29 MJ can be recovered from the combustion of the liquid resulting from the co-pyrolysis. Hence, this co-pyrolysis process could be economically viable, provided heat losses are addressed carefully when designing an industrial reactor.

1. Introduction

The production and consumption of plastics is increasing worldwide since the first polymer was developed in the early 1900s (Wang et al., 2015). In 2013, 299 millions of tons of plastic were produced in the world, among which 20% in Europe. In addition, plastic products have a short useful lifetime, which is shorter than one month for those used for packaging (nearly 40% of all plastics) (Achilias et al., 2007). The combination of the increased production of plastics and their short useful lifetime results in an always increasing amount of plastic wastes to be managed each year (35 millions of tons in Europe in 2010, according to Panda et al. (2010)).

Four routes are known in order to deal with plastic wastes: landfilling, mechanical recycling, energy recovery (including incineration and co-processing) and chemical recycling.

In Europe, landfilling was the favored route of waste treatment thanks to its simplicity, when environmental awareness was low. But, today, the cost of the land becomes higher and higher, making landfilling economically non-profitable (Singhabhandhu and Tezuka, 2010). Moreover, landfills are sources of water pollution due to highly polluted leaches, greenhouse gas emissions and local inconvenience (Garforth et al., 2004; Krook et al., 2012). In addition, the cost of raw materials has increased. This is why EU regulations constraint heavily landfilling and encourage the recycling (Quaghebeur et al., 2013). In that context, in 2010, eight European countries have highly restricted landfilling, resulting in less than 10% of the municipal solid waste mass landfilled (European Environment Agency, 2013). Besides, some projects of so-called

Abbreviations: DSC, differential scanning calorimetry; HDPE, high density polyethylene; Hi-Res TGA, high resolution thermogravimetric analyses; LDPE, low density polyethylene; PET, polyethylene terephthalate; PP, polypropylene; PS, polystyrene.

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landfill mining have been developed in several countries (Belgium, Sweden, Germany, Finland, The Netherlands, the Baltic countries, etc.). They aim to valorize the excavated wastes from the landfills through energy or material production and then restore the land for other uses. For instance, the goal of the MINERVE project (Walloon Region, Belgium) is precisely to excavate wastes from old landfill sites and upgrade them. The work presented in this paper, taking place in the frame of the MINERVE project, on behalf of Holcim (a cement production company), contributes to the development of a new technology to valorize landfilled recovered plastic wastes.

Mechanical recycling of plastic wastes is the optimal way of valorizing them in order to reduce energy and resource consumption as well as to reduce CO_2 emissions (Melendi et al., 2011). However, the main problem recyclers face is the degradation and the heterogeneity of the plastic wastes. The more contaminated and complex the wastes, the more difficult is the recycling (Al-Salem et al., 2009). Because of the heterogeneity and the contamination of municipal and, especially, excavated plastic wastes, choosing mechanical recycling to valorize them is often not an appropriate solution from an environmental and economic point of view (Panda et al., 2010).

Incineration, which consists in plastic waste combustion, results in toxic and/or greenhouse gas emission (Paradela et al., 2009) and presents a low efficiency, especially regarding incineration plants that produce only electricity (Grosso et al., 2010). This is why this way of valorization should be considered as a last resort.

Another way of recovering energy is by replacing a fraction of the usual energy source in an industrial process by plastic wastes (i.e. co-processing). This technique is already used, but mainly with plastic films or plastic wastes possessing one small dimension.

On the contrary, the chemical recycling of plastic wastes gives the possibility to treat heterogeneous and contaminated plastic wastes avoiding long and costly pre-treatments (Al-Salem et al., 2009). Chemical recycling is based on thermal and/or chemical treatments producing either fuel or raw materials for petrochemical applications. The production of fuel seems to be appropriate for common plastic wastes since most of them possess a heating value as high as that of oil (around 40 MJ/kg) (Themelis et al., 2011).

In this work, chemical recycling is chosen in order to develop a new technology to valorize plastic wastes excavated from a landfill. The purpose is to produce an alternative fuel that could be used by Holcim in cement kilns. The selected chemical recycling process is the co-pyrolysis (i.e. the simultaneous thermal decomposition) of the plastic wastes and used lubrication oils, in the absence of oxygen. The use of oil in the process is expected to provide two main benefits. On the one hand, it should improve material and heat transfers and, hence, reduce the technical limitations that occur in pyrolysis of plastic wastes alone due to the high viscosity and the low thermal conductivity of the molten plastics (Scheirs and Kaminsky, 2006; Achilias et al., 2007; Kim et al., 2012). On the other hand, it has been reported that, when plastics are co-pyrolyzed with a solvent, the latter can act as a catalyst for the plastic decomposition, by providing hydrogenated radicals (Yoon et al., 1999).

The goal of the project partner Holcim is to obtain a profitable alternative fuel that can be easily stored, transferred and used in cement kilns. Therefore, the developed process should fulfill the following specifications, given by Holcim:

- the maximum temperature reached during the process should be lower than 500°C, in order to limit the total energy consumption of the process;
- the mass fraction of the plastic wastes in the mixture of plastics and oils should be preferably equal or higher than 50%, as used oil is a cost factor;
- the viscosity of the obtained fuel should be under 1 Pa s at 90°C. It guarantees that the fuel can be easily processed in a cement kiln.

Several studies highlighted synergistic effects between two components when they are co-pyrolyzed: plastic and biomass (Zhou et al., 2006; Paradela et al., 2009; Abnisa and Daud, 2014; Oyedun et al., 2014), plastic and oil shale (Aboulkas et al., 2012), plastic and cellulose (Suriapparao et al., 2014), polyethylene and waste newspaper (Chen et al., 2016), sub-bituminous coal and biomass (Krerkkaiwan et al., 2013), oil-palm solid wastes and paper sludge (Lin et al., 2014), oil shale and pine sawdust (Johannes et al., 2013), oily wastes and scrap tires (Siva et al., 2013), waste polyolefins and waste motor oil (Uçar et al., 2016), waste automobile lubricating oil and polystyrene (Kim et al., 2012). As far as we know, the works of Uçar et al. (2016) and Kim et al. (2012) are the only ones that report the interactions occurring during the co-pyrolysis of plastic wastes and waste motor oil. Although some of the above mentioned articles explain the nature of the synergistic effects between two components during their co-pyrolysis, the synergistic effects that take place during the co-pyrolysis of a plastic and an oil are still unclear.

In addition, Yoon et al. (1999) have studied, in a microreactor, the influence of the temperature, the reaction time and the mass fraction of the plastics into the mixture on the co-pyrolysis of waste plastics and waste motor oil. Kim et al. (2012) investigated the influence of the heating rate and the mass fraction of the plastics into the mixture on the co-pyrolysis of polystyrene and waste motor oil into a 1 L reactor. Uçar et al. (2016) showed that, in the co-pyrolysis of plastic and used motor oil mixture, the yield is a function of the plastic (PE, PP). Nevertheless, to the best of our knowledge, the influence of the temperature and the composition of the mixture on the viscosity of the liquid resulting from the co-pyrolysis of a plastic/oil mixture has never been studied.

As mentioned previously, this work is a preliminary study for the development of a co-pyrolysis process of landfill recovered plastic wastes and used lubrication oils in order to produce an alternative fuel according to the industrial specifications. Considering the literature review, three specific goals are defined:

- To highlight the synergistic effects between a plastic and an oil during their co-pyrolysis.
- To set up a co-pyrolysis process of plastic wastes and used oils at lab scale and to study the influence of two parameters on the viscosity of the liquid resulting from the process. These two parameters are the time evolution of the temperature during the process and the initial mass fraction of plastics into the plastic/oil mixture.
- To provide information concerning the energy consumption of this process and recommendation about its scale-up.

The first objective is achieved by TGA runs performed with plastics and oils individually as well as with plastic/oil mixtures. The second objective is achieved by performing co-pyrolysis tests of waste plastics and used lubrication oils in a sealed and stirred 5 L reactor. The results of these tests are analyzed using a model of the energy transport in the reactor in order to achieve the third objective.

2. Materials and methods

2.1. High resolution thermogravimetric analyses

2.1.1. Materials

The plastics used for this study are among the most commonly used plastics worldwide: HDPE, LDPE, PP and PS. These plastics