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A new efficient method for the processing of post-consumer polypropylene and other polyolefin wastes into polar waxes

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ABSTRACT

This paper describes a new efficient method for the processing of post-consumer polypropylene (and other polyolefins) wastes into polar waxes that have potential applications. This technology includes two key steps. In the first step, the post-consumer polyolefins are purified mostly from oil which is used as a solvent in processing of polyolefin wastes (like metalized polyolefin films), and in the second step, the oxidative degradation process is initiated to obtain polar waxes or, alternatively, the purified polymer can be granulated. The results of laboratory research on both steps are presented, as well as a comparison of the obtained granulates and polar waxes with commercially available products.

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1. Introduction

Polyolefins (particularly polyethylene, polypropylene) are the most important group of plastics. The advantages of polyolefins include the following: they are odourless, they are non-toxic, and they have relatively good chemical resistance. Polyolefins, together with polyvinyl chloride (PVC), have the highest global production because of their ease of processing and affordable cost. In 2009, approximately 230 million tons of plastics were produced and approximately 25% of which were used in the European Union. This global market has been increasing at an average rate of 9% since 1950, reaching a peak of 245 million tons in 2008, after which there was a slight drop in production (PWE, 2011). The worldwide market for polypropylene (PP) will reach 62.4 million metric tons by 2020, driven by growth in the Asia Pacific region, the Middle East and Africa (Plastics News, 2013).

The demand for plastics – mainly used in the production of various types of packaging – results in the formation of a large quantity of post-consumer waste. Approximately 50% of plastics are used for single-use disposable applications, such as packaging, agricultural films and disposable consumer items (Hopewell et al., 2009).

As plastics manufacturing increases, the amount of waste also increases. There are many methods to manage or process such

waste (Pichtel, 2014). The simplest and most widespread method is to store the waste in a landfill. Unfortunately, the light weight of polymers together with their significant volume and a very long decomposition period (up to hundreds of years) results in the landfills filling quickly.

A better solution is to use material (mechanical) recycling. The waste polymers are sorted, grinding, sometimes washed and dried, and then granulated (Aznar et al., 2006). The sorting and identification have a key role in the recycling industry. Therefore, a number of new techniques have been recently developed in order to separate and sort plastic wastes, such as density sorting methods for polymer–electronic wastes (Kang and Schoenung, 2005; Murariu et al., 2005), electrostatic separation of plastic mixtures (Saekia, 2008; Park et al., 2007), sorting based on laser scanning (Ahmad, 2004), selective dissolution (Pappa et al., 2001) or gravity – sink/float separation in water to separate mainly polyolefins (PP, HDPE, L/LLDPE) or PET from other polymers (Pongstabodee et al., 2008; Wang et al., 2015). Mechanical recycling is difficult for complex and contaminated wastes, so it is usually performed on single-polymer plastic, e.g. polyurethane (Zia et al., 2007), PVC (Sadat-Shojai and Bakhshandeh, 2011), PE (Kartalis et al., 1999), etc. However, the obtained regranulate often has properties inferior to those of the primary polymer and can be used in combination with virgin polymer, with the exception of some applications, such as in the pharmacy or food processing industry.

The chemical recycling of polymers are also presently in use. Condensation polymers such as polyethylene terephthalate (PET)

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undergo degradation to produce monomer units (Yoshioka et al., 2004), while vinyl polymers (like polyolefins) produce a mixture containing numerous components for use as a fuel. During thermal processes (thermal cracking) of polyolefins at high temperature (400–700 °C) and under anaerobic conditions, hydrocarbon bonds break down. Depending on the temperature of process, numerous products can be obtained, such as gaseous C₁–C₄ hydrocarbons, liquid hydrocarbons (gasoline and oil fractions) and paraffin (Achilias et al., 2007, 2006; Kaminski et al., 1995). This process can be carried out as catalytic cracking (Butcher, 1994), which often occurs in fluidized bed reactors (Nishizaki, 1975; Kirkwood et al., 1994); or it can be carried out as hydrocracking (Venkatesh et al., 2001) or cracking in an aqueous medium as well (Hughes and Ramakumar, 1987).

Another methods of polymer wastes utilization (including polyolefin wastes) are using them in construction and building materials, such as polymer-bitumen mixtures (Vasudevan et al., 2012) or sand bricks and cement concentrates (Wahid et al., 2015; Suganthi et al., 2013), and different polymer-based composites (Cerruti et al., 2014; Hugo et al., 2011). Special attention is given also in relation to the recycling of medical plastic wastes (Lee et al., 2002).

If neither of recycling methods gives satisfactory results, the polymer (especially polyolefin) wastes can be combusted in specifically designed furnaces to recover the energy. The disadvantage of this method is the irretrievable loss of hydrocarbon material (Zucchelli and Rizzi, 2009).

A substantial amount of information about the valorization or recycling and recovery routes of plastic solid waste can be found in numerous review articles (Al-Salem et al., 2014, 2009; Panda et al., 2010; Hopewell et al., 2009; Siddique et al., 2008). Recent works reported the mechanical and chemical recycling of most important polymers, like polylactic acid (PLA), PVC, PE, PP, polystyrene (PS), PET, acrylonitrile–butadiene–styrene copolymer (ABS) and polycarbonate (PC) together with their systems including blends and composites have been also described in detail (Hamad et al., 2013).

A special type of polymer wastes is metalized polyolefin films (mainly polypropylene), which consist of a polyolefin material coated with a thin layer of metal, such as zinc, copper, aluminum, silver or gold. These films are used primarily in the food packaging industry, as well as in the electronic and electrical industry.

The problem of aluminum and polyethylene separation is well known with regard to recycling of composite packaging material (paper–plastic–aluminum) Tetra Pak and the existing technologies include wet process in aqueous solution (Yan et al., 2001) or dry process like pyrolysis or plasma treatment (Korkmaz et al., 2009; Alcoa, 2005).

The method of metallic dust concentrate recovery from metalized polyolefin films has been described (Nolte and Zieba, 2008). In this process the metalized polypropylene films are placed in metal basket inside reactor and dissolved in the hydrocarbon oil fraction (boiling between 50 and 280 °C) in mass ratio 1:5–20. The dissolving process is carrying out for 2–30 min at 50–110 °C. According to the patent description, the polymer remains in the basket, and the metal particles are suspended in oil. After sedimentation and centrifugation, the oil–metal concentrate is obtained. The rest of oil can be re-used for the demetallization process or for complete dissolution of the pure polymer film, which is carried out at 120–160 °C. The mixture polypropylene in oil can be subjected to catalytic cracking at 350–500 °C to obtain fuel fractions (no information about catalyst type). Alternatively, the polymer soaked with oil can be drained on a mechanical press and combusted (energy recovery). The interesting idea in this process is to use biodiesel, which can be considered as a solvent obtained from renewable sources.

This article presents the results of laboratory trials on the development of a new method for the processing of polymer wastes (post-consumer polyolefins), especially from the metallic dust recovery process described above (Nolte and Zieba, 2008). Particular attention was given to polypropylene waste, because oxidized waxes obtained from the oxidation of polypropylene can become new products on the market. Moreover, this method can be applied also to post-consumer HDPE and LDPE (high-density and low-density polyethylenes).

2. Material and methods

2.1. Materials

The post-consumer polypropylene (in the paste form) containing approximately 70% by weight of oil (biodiesel), which was used as the raw material in this work, was obtained from the processing of metalized films, performed by PIW ARCADIA Sp. z o.o. company, according to patent (Nolte and Zieba, 2008). The acid and saponification numbers of that polypropylene containing biodiesel were 35.5 and 188.6 mg KOH/g, respectively. Post-consumer high-density and low-density polyethylenes containing about 60% by weight of mineral oil were also obtained and provided by ARCADIA company according to patent (Nolte and Zieba, 2008).

The analytical-grade solvents used were as follows: methanol, ethanol, n-propanol, isopropanol, xylene and toluene, which were purchased from Avantor Performance Materials Poland S.A. The catalyst and initiators: manganese(II) acetylacetonate (97%), dicumyl peroxide (98%) and 1,1'-azobis(cyclohexanecarbonitrile) (98%), were purchased from Sigma–Aldrich and Rokanol NL6 was purchased from Rokita S.A.

2.2. Purification of post-consumer polyolefin wastes

Preliminary experiments of post-consumer polyolefin wastes purification refinement were performed in a 750 mL glass reactor. The polymer (50 g) was mixed with solvent in an amount of 100–300 g and stirred with a mechanical stirrer (400 rpm) for 2 h at 20 °C.

The main studies on the purification refinement of post-consumer polypropylene were carried out using a cylindrical column with a diameter of 6 cm and a height of 40 cm and a sintered glass disc G4 on the bottom. Between 400 and 500 g of polymer containing approximately 70 wt% of oil was placed inside. Then, the solvent (1000–1100 g) was introduced from the top. The solvent passed through the polymer bed, either by means of gravity (for approximately 3 h) or by applying 0.06 MPa of vacuum at the bottom of the column (for ca. 1 h). The process was performed at approximately 20 °C. Similar procedure was applied also for some trials with post-consumer HDPE and LDPE.

The purified polypropylene was dried at 70–105 °C to a constant weight. Then, the acid and saponification numbers and the melting point were determined. The oil–solvent mixture was distilled at 215 °C. The solvent was collected as the distillate, while the oil remained in the flask.

2.3. Oxidation of purified post-consumer polyolefins

The oxidation processes were carried out in a Parr Reactor model 5103 (made of Hastelloy C-276) with a volume of 600 mL and equipped with a magnetically coupled double pitched blade stirrer, a heating jacket, a temperature sensor, a reflux condenser, and a bubble tube.

Powdered purified polymer, additives (initiator, catalyst, and surfactant), and water were introduced into the autoclave. Next,

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