



Pyrolysis of polyolefin plastic waste and potential applications in asphalt road construction: A technical review

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ABSTRACT

Pressures on the current plastic waste management infrastructure has made the growth of new sustainable recycling capacities crucial. Pyrolysis is an emerging thermochemical technology that may be utilised at a large scale to aid in reaching the EU 2030 targets for plastic waste. Plastic valorisation via this process could gain increased competitiveness with conventional methods through the use of concepts such as 'Design for Recycling', identifying further marketable applications for pyrolysis end co-products. This paper presents a review on the pyrolysis of the most abundant plastic waste polyolefins, low-density polyethylene (LDPE), high-density polyethylene (HDPE) and polypropylene (PP), with a focus on the heavy wax products. A sizeable research gap in its known applications outside of the petrochemical and chemical feedstock industries was identified. Furthermore, its potential use in the hot mix asphalt (HMA) layers of flexible roads as an alternative binder material and aggregate is discussed. A plastic-derived bitumen modifier could be a viable solution to the current limitations associated with plastic bitumen modifiers (PMB), while producing asphalt with enhanced rheological properties and failure resistances. Consequently, future trends in research may include obtaining a full understanding of the capacity for pyrolysis products from waste polyolefins in bitumen modification. The key relationships between pyrolysis process parameters and the subsequent product properties, modification mechanisms and binder performance may also be explored. This application pairing process for pyrolysis products from plastic wastes may also be more extensively adopted in sustainable infrastructure, as well as other industries.

Table of contents

1. Introduction	2
2. Plastic Waste Management	5
3. Pyrolysis of Polyolefin Plastic Wastes	6
3.1 Polyolefins in Plastic Solid Waste Streams	6
3.2 Polyolefin Plastic Recycling via Pyrolysis	8
3.3 Pyrolysis of Polyolefins for Oil and Wax Production	11
4. Polyolefin Plastics in Sustainable Road Construction	13

(continued on next column)

(continued)

4.1 Polyolefin Plastics as Flexible Road Materials	13
4.2 Polyolefin Plastics in Asphalt (Binder) Modification	14
4.2.1 Polyolefin Modified Asphalt Binders	16
4.2.2 Polyolefin Modified Asphalt Mixtures	17
4.2.3 Limitations of Polyolefin Modified Asphalt (Binders)	19
4.2.4 Life Cycle Analysis and Cost Assessment	20
4.3 Pyrolysis Polyolefins in Asphalt (Binder) Modification	21
4.3.1 Plastic Waxes as Flexible Road Materials	23

(continued on next page)

Abbreviations: PSW, Plastics Solid Waste; MPW, Municipal Plastic Waste; CEP, Circular Economy Package; OECD, Organisation for Economic Co-operation and Development; PO, Polyolefin; LDPE, Low Density Polyethylene; LLDPE, Linear Low-Density Polyethylene; HDPE, High Density Polyethylene; XLPE, sCross-linked Polyethylene; PP, Polypropylene; PVC, Polyvinyl Chloride; TCT, Thermochemical Technology; LCA, Life Cycle Assessment; GHG, Greenhouse Gas; TRL, Technology Readiness Level; HMA, Hot Mix Asphalt; WMA, Warm Mix Asphalt; SMA, Stone Mastic Asphalt; PMB, Polymer Modified Bitumen; PMA, Polymer Modified Asphalt; Wt%, Percentage weight of dry feedstock (pyrolysis) or base bitumen binder (unless specified); MW, Molecular Weight; MFI, Melt Flow Index; DSR, Dynamic Shear Rheometer; G^* , Complex Stiffness Modulus (Pa); δ , Phase Angle; Jnr, Non-recoverable Creep Compliance; IFT, Indirect Tensile Fatigue; ITS, Indirect Tensile Strength; TSR, Tensile Strength Ratio; EVA, Ethylene-vinyl Acetate; EPDM, Ethylene-propylene-diene; GMA, Glycidyl methacrylate; PA, Polyphosphoric acid; MA, Maleic anhydride; ASTM, American Society for Testing and Materials.

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(continued)

4.3.2 Plastic Pyrolysis Wax Applications	24
5. Discussion and Conclusion	26
6. Acknowledgements	26
7. References	28

1. Introduction

To increase Europe's sustainability and resource efficiency as part of the European Commission's Circular Economy Plan (CEP), concepts such as Industrial Symbiosis have recently been promoted. Industrial symbiosis is the process by which wastes, or by-products of an industry or industrial process become the raw materials to be used in a more sustainable way (European Commission 2010; European Commission, 2021). Moreover, the document published by the European Commission entitled 'Roadmap to a Resource Efficient Europe' emphasises the role that waste should play in boosting the economy and viewing it as a resource by the year 2020 (Al-Salem et al., 2020). Therefore, there is increasing importance in the investment of research examining the innovative ways of using wastes such as recycled plastics as a resource. In 2018, global plastics production reached 359 million tonnes, with Europe producing 61.8 million tonnes of that total. Correspondingly, of the 29.1 million tonnes of European post-consumer plastic waste collected in the same year, only 32.5% of the waste was sent to recycling facilities (M. D. PlasticsEurope Deutschland e.V., 2019). A major portion of the plastic produced each year is used to make disposable items of packaging or other short-lived 'single use' products that are discarded within a year of manufacture (Hopewell et al., 2009). Polyolefin (PO) plastics such as High-Density Polyethylene (HDPE), Low-Density Polyethylene (LDPE) and Polypropylene (PP) make up the majority of municipal plastic waste streams and have had a recent demand surge due to their versatility in single-use medical applications, mainly due to the COVID-19 pandemic (Dharmaraj et al., 2021). Plastic is a petroleum-based material, and its rising demand has led to the increased depletion of the non-renewable fossil fuel, indicating that our current

use of plastics is not sustainable (Anuar Sharuddin et al., 2016). With the rising demand and consumption of this inexpensive, non-biodegradable material and its recycling still at a low rate, the sheer accumulation and disposal of end-of-life plastic solid waste (PSW) is now a great problem. In 2019, the EU exported 1.5 million tonnes of plastic waste mostly to recycling facilities in Turkey and Asian countries. This share of exported plastic waste to countries such as China has recently fallen significantly due to the adoption of restrictions on the import of plastic waste in 2018 (European Commission, 2020). Additionally, as of 2021 the export of hazardous plastic waste and plastic waste that is hard to recycle from the EU to non-member countries of the Organisation for Economic Co-operation and Development (OECD) has been banned. Therefore, new sustainable recycling capacities must grow to accommodate the new tonnages of plastic waste, diverting from incineration and landfill and equally fulfilling the new EU recycling targets (Simon, 2021).

The potential of thermochemical technologies such as pyrolysis for the conversion of waste polyolefins to value-added products and feed-stocks/new materials for other industries makes it a suitable and a viable waste management route, as well as a possible contributor to achieving industrial symbiosis in the future for the plastic industry and others. A concept relevant to examining application routes for plastic wastes and advocated by recent review works is Design from Recycling (Delva et al., 2018; Lore et al., 2017). This concept was adapted from the Design for Recycling schematic promoted within the framework of the CEP and entails that the secondary raw materials originating from the recycled polymer waste of a previous products end of life (EoL) is the new starting point of new product development. Its key aspects are shown in Fig. 1, adapted from (Ragaert et al., 2017). Pyrolysis could become increasingly crucial in the future as a strategy for upgrading plastics to products that can be extensively characterised and matched with applications throughout different industries. On the other hand, following a recent push from governments and road authorities to become more sustainable, there has been an increasingly prominent trend within the pavement industry in using waste materials in hot mix asphalt (HMA) roading applications. This is in response to the need of reducing consumption of both costly virgin and increasingly scarce materials and

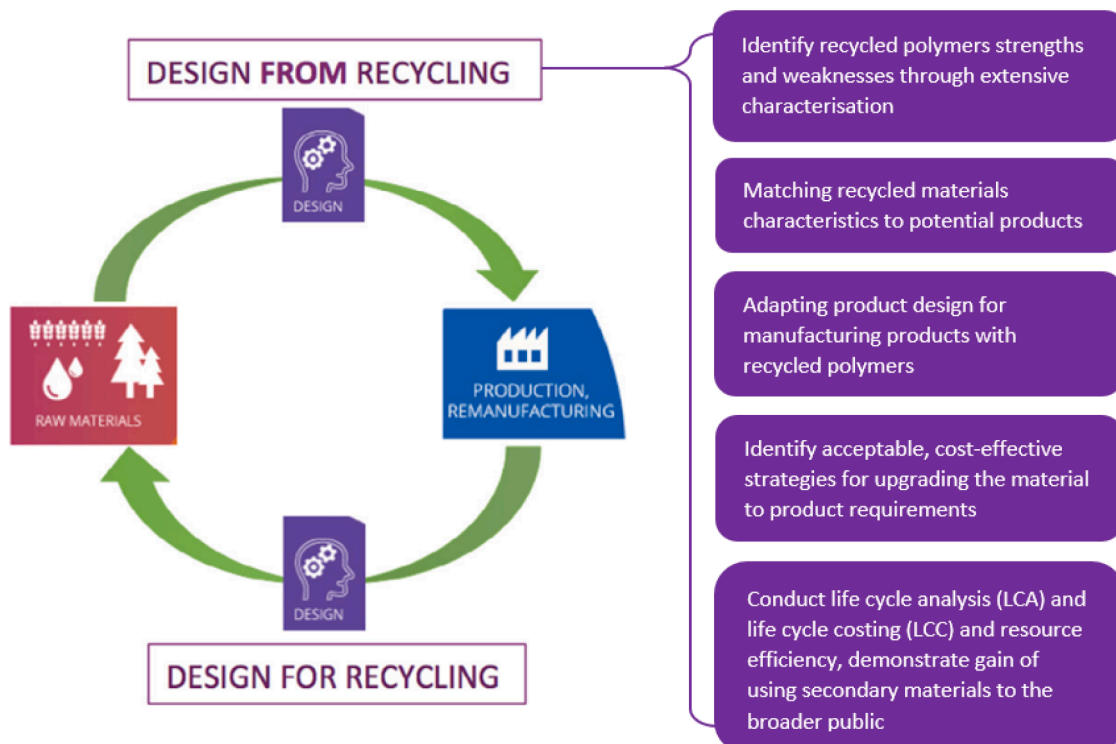


Fig. 1. Design from recycling concept key aspects, adapted from (Ragaert et al., 2017).

avoiding landfilling (Santos et al., 2021b). The use of waste polyolefin plastics in asphalt modification are well documented to impart enhanced service properties over a wide range of temperatures in road paving applications, especially improving rutting resistance at high in-service temperatures as well as temperature and stripping susceptibility (Kalantar et al., 2012, Yu et al., 2015). However, using these waste materials doesn't come without limitations, from both technical and environmental standpoints (Santos et al., 2021a).

A sizeable research gap has been identified by the authors in known applications for plastic pyrolysis heavy wax products, outside of petrochemical and chemical feedstock industries. This gap in literature can be attributed to a lack in the extensive characterisation for this product to match it with potential applications. Furthermore, there is a large absence of research knowledge in the sole capacity of fully degraded PO plastic pyrolysis products and their capacities as flexible pavement materials, such as in asphalt binder and mixture modification. Additionally, there has also been no comprehensive attempt to form relationships between the pyrolysis parameters used and the resultant effects of pyrolysis products on the asphalt binder performance, from both a chemical and rheological standpoint. Therefore, this review article presents the following research objectives:

- 1) Discuss the current plastic waste management methods and the need for new sustainable recycling capacities.
- 2) Investigate the thermal pyrolysis process for the most abundant plastic waste polyolefins, low-density polyethylene (LDPE), high-density polyethylene (HDPE) and polypropylene (PP), with a focus on the heavy wax products.
- 3) Explore the current use of recycled PO plastics in the hot mix asphalt (HMA) layers of flexible roads as an alternative binder material and aggregate. However, its use in binder modification via wet process manufacturing techniques is mainly considered.
- 4) The achievable property enhancements of PO plastic asphalt (binder) modification, the associated limitations from a technological and

environmental standpoint, as well as life cycle and economic assessment data will be examined.

- 5) Provide a discussion on plastic wax-based warm mix asphalt (WMA) technologies and the potential use of PO plastic-derived wax products from pyrolysis as a modifier with similar properties in HMA.

2. Plastic waste management

Landfilling plays an important role in current PSW management strategies; however, this method is a non-sustainable and questionable option as the plastics remain as a lasting environmental burden. The continuous disposal of PSW and its accumulation in landfill sites has been associated with significant effects that are detrimental to both environmental quality and human health (Bale, 2011). Recycling and re-use play considerable roles in the 2019 Zero Waste Europe hierarchy for achieving circularity and sustainability goals (Simon, 2019). Recycling is an important PSW management method as it avoids the environmental burdens associated with landfill, while minimizing the consumption of energy and finite resources as well as reducing emissions associated with plastic production (Al-Salem et al., 2009). There are currently four recycling methods outlined for PSW, which are shown in Fig. 2 as part of the plastic production chain, adapted from (Brems et al., 2013; Vilaplana and Karlsson, 2008).

Re-extrusion (primary) recycling is a closed loop recycling scheme that is considered as an efficient solution for industrial PSW process scrap (Al-Salem et al., 2010). The scrap is mechanically reprocessed into a product with equivalent properties through its re-introduction into the heating cycle of the processing line, in order to increase production (Hopewell et al., 2009). This type of recycling requires semi clean, non-contaminated waste which is the same type as the virgin resin to which the degraded plastic will partially substitute. Increasing the recycled plastic fraction may save on market prices compared with the virgin counterparts as well as reduce processing costs and waste generated by plastic converters, however, the feed mixture decreases the

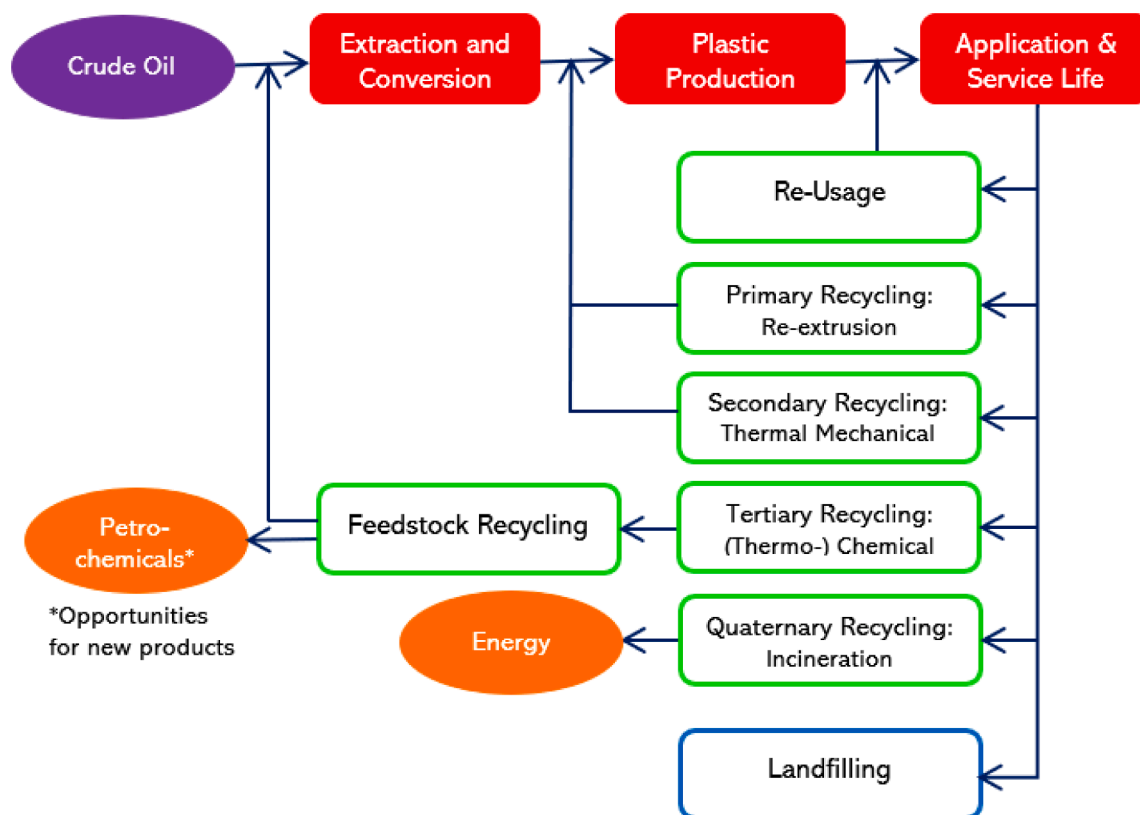


Fig. 2. Plastic solid waste management methods including four valorisation routes, adapted from (Brems et al., 2013; Vilaplana and Karlsson, 2008).

quality of the overall product (Antelava et al., 2019). Mechanical (secondary) recycling is the most common method for the recycling of plastic wastes. This is an open-loop recycling scheme that involves the mechanical reprocessing of PSW into new lower-value products. The mechanical recycling of PSW can only be performed on single-polymer waste, thus needing an extensive number of pre-treatment and preparation steps (collection, sorting, washing, grinding) that are discussed in detail elsewhere (Ragaert et al., 2017). This creates the challenge of high energy demand and utilities associated with process operation. Multiple re-processing steps (heating and mechanical shearing) also result in molecular damage mechanisms, reducing the integrity of the final product (Delva et al., 2018; González-González et al., 1998). Open-loop recycling can be considered as a form of cascading into ever lower-valued applications and the consumer demand for high end products makes investment in these types of recycling schemes non-profitable (Al-Salem et al., 2017). Alternatively, energy recovery via incineration (quaternary recycling) is a well-supported treatment method for municipal solid waste with high PSW content in Europe by associations such as ISOPA (European Diisocyanate & Polyol Producers Association). The high calorific value (CV) of plastics match that of conventional fuels and therefore PSW can be used in incinerators to give energy via electricity generation, district heating or combined heat and power schemes (Williams and Williams, 1997). This method is an effective way to reduce the volume of PSW, however, results in the loss of valuable raw materials as all waste is converted to energy. It is also considered ecologically unacceptable due to the production of airborne highly toxic substances (furans, dioxins, acid gases) and groundwater pollution as a result of the plastic content in the processed waste and process conditions (Verma et al., 2016). The EU has established, directives (2000/76/EC) for emission limits and guidelines for incineration plants, however, reducing hazardous components dramatically increases operation costs (Antelava et al., 2019).

The discussed challenges faced by the current PSW recycling methods lead to them being unacceptable to the principles of sustainable development and consequently, there has been a large growing research interest surrounding chemical (tertiary) recycling. Chemical recycling is the process in which polymers are converted into smaller hydrocarbon molecules via a change to their chemical structure using a chemical reaction or heat. It is based on a number of technologies that can generate pure value-added products for various industrial and commercial applications (Ragaert et al., 2017). These technologies include both chemical and thermo-chemical techniques, such as catalytic cracking, glycolysis, gasification, hydrogenation, methanolysis, pyrolysis, amongst others. Pyrolysis and gasification are thermochemical methods that are especially under extensive research and development for the establishment of suitable conditions due to recent works demonstrating the potential of the products from PSW as alternative fuels (Patni et al., 2013; Wong et al., 2015; Kunwar et al., 2016). The increased global effort in this area of research has particularly been demonstrated in recent bibliometric analysis (Khatun et al., 2021). Monomer recovery is another product destination that has received research interest from these techniques, involving feedstock recycling in closed cycle material flows for certain plastic-wastes, thus reducing the demand on petroleum for virgin plastic production (Donaj et al., 2012; Díaz-Silvarrey et al., 2018). Chemical recycling has a high potential for heterogeneous and contaminated PSW when separation is not economical or technically feasible. Furthermore, the depolymerisation technologies utilised in chemical recycling can result in a very profitable and sustainable industrial scheme, providing high product yields with minimum residual waste. The research and development of such technologies would increase the incentive to recycle polymers, as well as decouple the plastic industry from fossil feedstock (Brems et al., 2013; Horvat and Ng, 1999).

3. Pyrolysis of polyolefin plastic wastes

3.1. Polyolefins in plastic solid waste streams




Thermoplastics are materials that can be melted when heated and hardened when cooled. Due to this susceptibility to change and molecular reformation with heat exposure, currently about 80% of consumed plastics in western Europe are thermoplastics, especially polypropylene (PP), low-density polyethylene (LDPE), high-density polyethylene (HDPE), polyvinyl chloride (PVC), polyethylene terephthalate (PET) and polystyrene (PS) (Al-Salem et al., 2017). Polyethylene alone makes up around 40% of the plastic total in plastic waste streams, thus the majority of solid plastic wastes are of polyolefin (PO) nature. In fact, due to their versatility towards many applications (packaging, bags, foils, toys, storage, etc.), within municipal solid waste streams; LDPE, HDPE, and PP account for around 65% of waste stream PSW (Onwudili et al., 2009). Waste plastics have been thoroughly studied over the years with major research focus on their valorisation through thermochemical processes from low grade recycled plastics into petrochemical feedstock. More recently, pyrolysis particularly has been extensively applied on various scales and has been presented as a promising technique for such purposes. The suitability of plastic waste as a feedstock has also been demonstrated and its viability is clear, especially for polyolefins such as polyethylene, polypropylene, and polystyrene (de Marco et al., 2009). This is due to their capability for the production of somewhat clean distillates by comparison to other organic feedstock materials. As well as this, unlike biomass, polyolefins in particular have little oxygen content, thus higher carbon efficiency as well as higher gross margins can be obtained (Ragaert et al., 2017). This study has focused its scope on the pyrolysis of polyolefins namely LDPE, HDPE, and PP, as they are most abundant in plastic waste streams and have very similar composition, such that the same valorisation routes may be applied to them (Lopez et al., 2017). They are suitable as a feedstock with high carbon and volatile matter contents, and low ash content. Due to these properties, the capacity for production of valuable pyrolysis tar products has been demonstrated extensively in literature, making pyrolysis a favoured treatment and method of choice for the thermochemical treatment (TCT) of plastics (Anuar Sharuddin et al., 2016; Al-Salem et al., 2017; Abnisa and Wan Daud, 2014). The resin codes, properties and examples of each polyolefin is shown in Table 1, collated from (Plastics Europe, 2021; American Chemistry Council, 2021).

3.2. Polyolefin plastic recycling via pyrolysis

Pyrolysis is a thermochemical conversion technology that can be considered a 'feedstock recycling' process, in which the thermal degradation of long chain organic materials in an inert atmosphere occurs at temperatures of around 500 °C (Aguado et al., 2008; Butler et al., 2011). The hydrocarbon content in the waste is converted into gases, oil and chars of different proportions and compositions depending on multiple process operational factors that have been comprehensively studied throughout plastic pyrolysis literature. These commonly include the feedstock used, e.g. co-pyrolysis of plastic mixtures and the use of virgin or real-life plastic waste (Westerhout et al., 1998; Adrados et al., 2012), the reactor technology (Al-Salem et al., 2017; Csukás et al., 2013; Qureshi et al., 2019), variable reactor operating parameters such as the temperature (Williams and Williams, 1997; Onwudili et al., 2009; Kumar and Singh, 2011), reaction or residence time (Anuar Sharuddin et al., 2016; McCaffrey et al., 1995; Papuga et al., 2016), as well as the presence of chemical agents (Oh et al., 2007; Artetxe et al., 2013; Quesada et al., 2020; Kumar and Singh, 2013), or catalysts (Artetxe et al., 2013; Quesada et al., 2020; Kumar and Singh, 2013; Sakata et al., 1999; López et al., 2011), used in the reaction. The process is considered flexible such that it is capable of treating many different solid hydrocarbon wastes, producing clean fuel gas of high calorific value (Kodera et al., 2006). The solid char contains both carbon and mineral content of

Table 1

Resin codes, properties, and examples of applications for polyolefins HDPE, LDPE, and PP, collated from (Plastics Europe 2021; American Chemistry Council 2021).

Waste Plastic	Resin Identification Code	Properties	Applications
High Density Polyethylene (HDPE)		Melting range: 250–260 °C Softening range: 90 °C (approx.) Clear, tough, solvent resistant, barrier to gas and moisture	Shopping bags, household articles, bottle caps
Low Density Polyethylene (LDPE)		Melting range: 120 °C (approx.) Softening range: 80–90 °C (approx.) Soft and flexible, translucent, easily scratched	Shopping bags, bin bags, cosmetic and detergent bottles, milk pouches
Polypropylene (PP)		Melting range: 140–160 °C (approx.) Softening range: 95–110 °C (approx.) Hard, translucent, versatile, solvent resistance	Detergent wrapping, bottle caps and fasteners, food and vapour packaging, microwave food trays

the original feed material, while the tar fractions consist of a complex of organic compounds and can be very rich in aromatics (S.M. Al-Salem et al., 2010). The pyrolysis process varies between authors in terms of cracking technologies, conditions, catalysts, and units used, depending largely on the selected feedstock and targeted products. However, the principle depolymerisation cracking reactions and subsequent recovery of valuable hydrocarbon products remain analogous. An example pyrolysis system is the cylindrical screw reactor depicted in the schematic shown in Fig. 3. During operation, feedstock continuously enters the system via a screw feeder and conveyed through the reactor via an inner screw conveyor. The pyrolysis vapour leaves the reactor and passes through a shell and tube condenser, where the pyrolysis vapour is condensed to form pyrolysis liquid. The permanent gases pass through an electrostatic precipitator system for aerosol removal. After further fibre cartridge filtration, the final cleaned gas is sent to a flare system. Char from the pyrolysis is collected in a char pot (Yang et al., 2018b).

Pyrolysis has long been investigated as a viable recycling route for wastes such as biomass and rubber and more recently has received increasing attention for PSW treatment. Surveying the literature reveals

an extensive number of studies on virgin and PSW polyolefin pyrolysis at micro-, bench and pilot scales, some articles are summarised in Table 2 for example. It is important to note that pyrolysis experiments are dominantly operated at laboratory scale (micro- and bench scale). Availability of information is more limited for larger scale processes; however, many countries and business entities are developing processes for waste polymer pyrolysis (Csukás et al., 2013).

So far, pyrolysis has been developed to a commercial scale (Antelava et al., 2019). In 2018, there were 15 companies operating 87 plants worldwide to produce petrochemical and chemical feedstocks from plastics (Stclair-Pearce and Garbett, 2018). For example, the Recycling Technologies pilot plant located in the UK operating an advanced fluidised reactor system that converts non-recycled plastic, such as those commonly used in the packaging industry for film, into the low sulphur hydrocarbon Plaxx™. This can be refined into a low sulphur alternative to heavy fuel oil, slack wax or as a feedstock for more plastics. The system is being upscaled to process 7000 tonnes per annum (tpa) of plastic waste (Stclair-Pearce and Garbett, 2018). In the US, Renewology pyrolyzes mixed plastics and packaging materials (grade 3–7) in a continuous auger kiln reactor calibrated to produce naphtha or a middle distillate suitable for diesel use at refineries. With current 10 tpd modules, the company will be commencing ‘Renew Pheonix’ in 2021 for a 6000,000 lbs annual target capacity commercial plant (Qureshi et al., 2019; Renewology 2018; finnCap, 2019). In Europe, Plastic Energy’s recycling plant in Spain has a processing capacity of 5000 tonnes of plastic waste (including PE, PP and PS) per annum. The PSW is recycled via their thermal anaerobic conversation (TAC) process in a continuous stirred-tank reactor at moderate temperatures to produce TACCOIL™ chemical feedstock, sold to the petrochemical industry to convert to virgin plastic, oil, or transportation fuels (Qureshi et al., 2019; Plastic Energy, 2020). Further information on other commercial pyrolysis processes and advanced technologies can be found elsewhere (Al-Salem et al., 2010; Aguado et al., 2008).

Numerous authors have published articles for the purpose of presenting various thermochemical recycling technologies for the conversion of PSW, with a number of technologies including pyrolysis concluded to be robust enough to warrant further research and development (Al-Salem et al., 2017; Nanda and Berruti, 2021; Soni et al., 2021). Such review articles frequently offer similar analyses of PSW recycling technologies, both conventional and thermochemical, with a research focus on comparisons of environmental impacts, such as greenhouse gas (GHG) emissions, energy consumption and waste reduction between technologies (Ragaert et al., 2017; Al-Salem et al., 2017; Lopez et al., 2017; Pickering, 2006; Al-Salem et al., 2010). Other authors have utilised specific waste management strategies using evaluation tools for comparison, such as life-cycle assessments (LCA) and Technology Readiness Levels (TRL) (Antelava et al., 2019; Solis and Silveira, 2020). A common trend in these research analyses is the comparison of current waste management in Europe with the pyrolysis

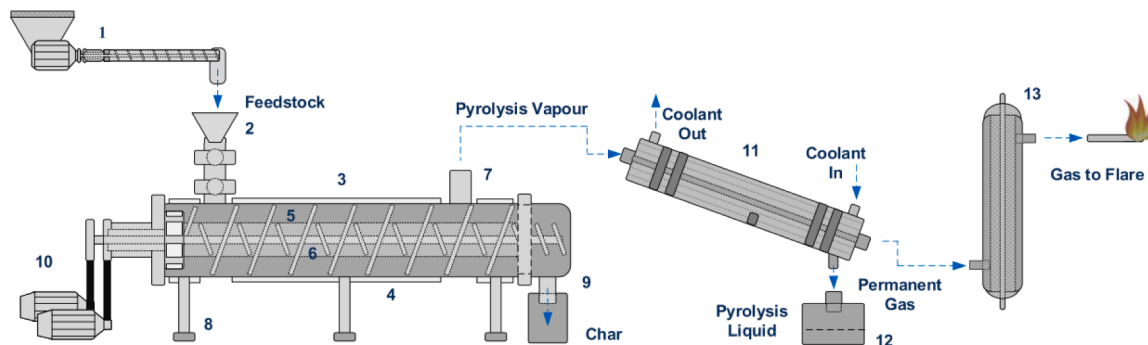


Fig. 3. Schematic diagram of the intermediate pyrolysis system (1) Feeder; (2) Feed Inlet; (3) The Pyrolysis Reactor; (4) Heating Jackets; (5) Outer Screw; (6) Inner Screw; (7) Vapour Outlet; (8) Stands; (9) Char Outlet and Char Pot; (10) Motor; (11) Shell and Tube Condenser; (12) Liquid Vessel; (13) Electrostatic Precipitator (Yang et al., 2018b).

Table 2

Summary of example studies investigated as a chemical treatment for virgin/waste polymers in laboratory micro-, bench and pilot scale.

Author	Scale	Feedstock	Process Conditions	Products	Summary
Al-Salem et al. (Al-Salem et al., 2018)	Micro	LLDPE	20–800 °C, heating rates of 5, 10, 15, 20, 25 °C/min	Gases	Thermal degradation kinetics and stability investigated via non-isothermal thermogravimetric analysis.
Al-Salem et al. (Al-Salem, Jul. 2019)	Micro	HDPE	500–800 °C, heating rate 5 °C/min, nitrogen gas flow rate 20 ml/min	Oil, waxes, gases, solid chars	Isothermal pyrolysis including a comprehensive gas chromatography (GC) analysis of both liquid and gaseous products. Proposed model for degradation kinetics of parallel first order reactions.
Horvat et al. (Horvat and Ng, 1999)	Bench	HDPE, LDPE, waste PE	400–460 °C, 4-hour reaction time, nitrogen flowrate 5–75 mL/min	Oil, polymer residue (waxes), gases, coke	Pyrolysis in a semi-batch pyrolysis reactor, followed by hydrogenation of product oils into liquid fuel fractions.
Mastellone et al. (Mastellone et al., 2002)	Bench	Recycled PE	300–450 °C, residence time 60–180 s	Gases, Liquid, Solid (not degraded PE), Solid (tar)	Pyrolysis in a fluidised bed reactor. Process yields with differing fluidizing velocities, bed temperatures, residence times were reported.
Miskolczi et al. (Miskolczi et al., 2009)	Pilot	Waste LDPE	250–400 °C, 60 min reaction time	Liquid oil, gases, solid residue	Pyrolysis using a ZSM-5 catalyst, feed capacity 9.0 kg/hr. The composition of product gasoline and light oil fractions produced using the catalyst were advantageous for fuel-like applications.
Csukás et al. (Csukás et al., 2013)	Pilot	Waste HDPE	465–545 °C	Gases, Naphtha, middle distillates, heavy oil	Pyrolysis in a tubular reactor with distillation unit, feed capacity range 0.04–1.2 kg/h. Dynamic simulation model developed for pyrolysis of plastics wastes based on a four-step degradation scheme.

of waste plastics to produce petrochemicals including propane, butane, and gasoline (Benavides et al., 2017). Table 3 presents key advantages and limitations for the pyrolysis of PSW, gathered from the analyses mentioned.

3.3. Pyrolysis of polyolefins for oil and wax production

Polyolefin pyrolysis proceeds via a random degradation mechanism, yielding a broad spectrum of products (C₁–C₃₀), yet the primary devolatilization reaction yields mainly wax-like materials with a typical carbon number range of C₂₀–C₅₀ (Aguado et al., 2002b). Wax is the main product obtained by thermal pyrolysis of polyolefins at lower temperatures and its formation is an attractive route to minimise energy requirements. The pyrolytic waxes from polyolefin feedstocks are aliphatic, mostly long-chain unbranched alkanes, with a chemical composition that is very similar to that obtained for commercial paraffinic waxes, as demonstrated by (Chaala et al., 1997). However, significant differences include the olefinic groups present in pyrolytic waxes due to radical degradation mechanisms, as well as the alkyl chains being much less branched (Chaala et al., 1997; Arabiourrutia et al., 2012b).

A prominent trend in polyolefin pyrolysis literature is the thermal and catalytic pyrolysis of PE and PP into oils with heating values similar to conventional diesel and high aromatic content (BTX-benzene, toluene, and xylene aromatics) to be used as a petrochemical feedstock

Table 3

Key advantages and limitation for the pyrolysis of PSW.

Advantages	Limitations
Technical	
Simple Technology	Insufficient understanding of reaction pathways
Suitable for lower quality/mixed wastes	Subsequent problems with scaling up reactions
High efficiency and product yield	Requires handling of char co-product
High capacity for electricity and valuable products from waste	Sensitivity to feedstock
Capable of continuous operation	
TRL range 7–9 for production of liquid fuels	
Environmental	
Lower GHG emissions and consumption of fossil fuel resources (compared to other TCTs and conventional recycling methods)	Landfilling of residues: char, silica (sand) and bottom ashes
Dioxins not formed due to inert reaction atmosphere	
Cost	
Few pre-treatment steps required	High energy requirement
Lower operating temperatures compared to other TCTs	Products often need upgrading

or fuel oil (Jung et al., 2010; Miandad et al., 2017). Others have also considered the light and heavy wax co-products for their suitability as feedstock for steam and catalytic crackers to be upgraded into products for both chemical and petrochemical industries (Predel and Kaminsky, 2000; Arabiourrutia et al., 2012a). Pyrolysis experiments are dominantly operated at laboratory scale, however, the range of reactor technologies used with a focus on polyolefin wax production, or the wax co-product analysis is diverse. Prevalent technologies include fluidised bed and conical spouted bed (CSBR) reactors. Kaminsky et al. pyrolysed pure and mixed polyolefins (PE, PP and PS) in a fluidised bed reactor at 510 °C to produce aliphatic waxes. The wax product fractions were categorised as light waxes (bp between 300 and 500 °C, *n*-C_{18–37}, and *i*-C_{21–50}) and heavy waxes (bp > 500 °C, > *n*-C₃₇ and > *i*-C₅₀) to give a total wax yield of 84 wt% for PE and 57.5 wt% for PP. No interaction between the two polymers PE and PP was indicated in fluidised bed pyrolysis (Predel and Kaminsky, 2000). Williams et al. pyrolysed LDPE between 500 and 700 °C in a fluidised bed reactor, obtaining a 45.3 wt% wax yield (Williams and Williams, 1999). It should be noted that this yield in comparison to results from other authors highlights the inconsistency that can be found in literature regarding wax product categorisation and methods of reporting yield data (Predel and Kaminsky, 2000). In this instance, for conditions below 600 °C, the distinction between the oil and wax phases were less clear and the product oil was classed as a low viscosity waxy material.

Alternatively, in more recent works Aguado et al. utilised a conical spouted bed reactor (CSBR) for the selective obtaining of wax by polyolefin (HDPE, LDPE, and PP) pyrolysis. The design of this alternative technology negates the limitations of the fluidised bed reactor that can lead to bed defluidisation, such as particle agglomeration which can be serious when handling solids of irregular texture and/or sticky nature and with size distribution. No differences were observed between the results for the two types of PE, both producing very similar wax products, with a maximum yield of 80 wt% at the lowest temperature of 450 °C (Aguado et al., 2002a). Arabiourrutia et al. received very similar results using the same technology, obtaining a maximum wax yield of 80 wt% at 450 °C for both HDPE and LDPE. However, a notable trend in these studies is the highest wax yield coming from PP, obtaining 92 wt% at 450 °C. Higher wax yields at lower temperatures for PP were deduced to be due to a more branched structure in PP compared to that of PE (Arabiourrutia et al., 2012a). Other authors have utilised vacuum pyrolysis in a batch reactor, in which the energy of evaporation of the primary macromolecules formed in the radical initiation reactions is lower than the activation energy required to trigger further radical initiation reactions, yielding fewer light organics and gases. Chaala et al. obtained a 71.3 wt% wax yield in a bench scale reactor at 20 kPa

reduced total pressure from electric cable wastes (Chaala et al., 1997). Additional pyrolysis reactor technologies to note include batch and auger reactors, in which authors have previously obtained > 75 wt% and 93.2 wt% wax yields respectively from thermal pyrolysis at lower to moderate process temperatures (Al-Salem et al., 2020; Ademiluyi et al., 2013). Table 4 can be referred to for further comparison of pyrolysis technologies in literature and resultant wax yield capacities and further technologies and their analysis can be found throughout review articles in current literature (Anuar Sharuddin et al., 2016; Lopez et al., 2017).

In terms of pyrolysis parameters, the wax yield decreases dramatically at higher process temperatures. The temperature is the main parameter that controls the cracking reaction of the polymer chain and high temperatures support the cleavage of C–C bonds, resulting in the increased cracking of macromolecules into lighter organic oil fractions, and at higher temperatures to gases (Westerhout et al., 1998; Williams and Williams, 1999). An increase in pyrolysis time will also decrease the yield of wax due to the increased conversion of the primary products (Anuar Sharuddin et al., 2016; Jixing, 2003). Furthermore, studies have been carried out with the focus to produce wax from plastic waste, investigating the effect of these pyrolysis parameters on the resultant wax quality and comparing the wax products to industrial standards. This was done using indicators such as wax melting point and penetration degree and clearly demonstrated that with careful control of both temperature and pyrolysis time, waxes with the properties of both commercial paraffinic and microcrystalline wax can be produced. It has

also been indicated that plastic waste dyes do not affect the quality indicators of the PE pyrolysis wax (Ademiluyi et al., 2013; Jixing, 2003). Additionally, at low pressures with short residence time of vapours in the reactor, it has been seen in the pyrolysis of mixed PE wastes that the major reaction to take place is the disproportionation of the primary macroradicals, forming n-paraffins and olefins at moderate temperatures (Murata et al., 2004). Or when operating under vacuum conditions, low cleavage of the macromolecular chain ends occurs, resulting in lower yields of light organics and gases, with increased soft and hard wax products (Chaala et al., 1997).

4. Polyolefin plastics in sustainable road construction

4.1. Polyolefin plastics as flexible road materials

Hot mix asphalt (HMA) is a composite material that consists largely of mineral aggregates bound by a bituminous binder. The bitumen binder is a by-product of vacuum distillation in petroleum oil refineries and is a viscoelastic, rheological, and non-magnetic material (Aziz et al., 2015). In order for an application to be viable for the reuse of plastic waste, it would need to have the capacity for incorporating high volumes of plastic. As well as this, very little cost should be involved in the preparation. Using waste plastic directly in HMA pavement structures is considered as a practical application with the large volumes of both plastics and roads produced (Grady and Brian Grady, 2021). In a

Table 4

Pyrolysis technologies and parameters used in literature for polyolefin pyrolysis with resultant product wax yields.

Author	Type of Pyrolysis	Reactor	Polyolefins	Pyrolysis Temperature (°C)	Pyrolysis Time (s)	Wax Yield (wt%)	
Arabiourrutia et al. (Arabiourrutia et al., 2012a)	Thermal Pyrolysis	Conical Spouted Bed (CSBR)	LDPE	450	–	80	
				500		69	
				600		51	
			HDPE	450		–	80
				500		68	
				600		49	
			PP	450		–	92
				500		75	
				600		50	
Chaala et al. (Chaala et al., 1997)	Vacuum Pyrolysis	20 kPa Bench	Electric cable waste (mainly XLPE)	450	–	71.3	
Predel et al. (Predel and Kaminsky, Jan. 2000)	Thermal Pyrolysis	Fluidised Bed	PE	510	–	84	
				–		83.7	
				–		76	
			PE/PP (60:40)	510		–	71.5
				510		–	56.2
				510		–	57.5
Williams et al. (Williams and Williams, 1999)	Thermal Pyrolysis	Fluidised Bed	LDPE	500	–	45.3	
				550		35.4	
				600		24.8	
			650	12.1			
				700		4.0	
				–		–	
Aguado et al. (Aguado et al., 2002a)	Thermal Pyrolysis	Conical Spouted Bed (CSBR)	LDPE	450	1–1080	80	
				500		69	
				550		57	
				600		51	
				–		–	
			HDPE	450	1–1080	80	
				500		68	
				550		61	
				600		49	
				–		–	
			PP	450	1–1080	92	
				–		–	
				500		75	
				550		67	
				600		50	
Al-Salem et al. (Al-Salem et al., 2020)	Thermal Pyrolysis	Auger	PSW (PE majority)	500	–	93.2	
Ademiluyi et al. (Ademiluyi et al., 2013)	Thermal Pyrolysis	Batch	Waste PE sachets	130–150	18,00–2400	>75	

* Excluding results for 'Oil and Wax' product mixtures.

literature analysis conducted by Zhao et al., out of the articles evaluated between 2014–2019 studying plastics as a waste solid material in pavement engineering, polyolefins PE and PP were two of the most studied plastics (Zhao et al., 2020). It is noted that the plastics are predominantly evaluated in literature for the application as a modifier or ‘reinforcement’ in the pavement material, more specifically via incorporation into the bituminous binders. Throughout its service life, HMA requires durability over a range of climate conditions and traffic loads. The engineering performance of its constituent materials such as the neat bitumen binders may not be satisfactory under all possible climates. Therefore, cost effective and alternative binder materials such as waste plastics are increasingly being investigated to boost binder performance and extend service life, while decreasing the use of bitumen (Porto et al., 2019; Lesueur, 2009b).

Waste polyolefin (PO) plastics are good candidates as a cheaper material for asphalt binder modification. The melting point ranges of polyolefin plastics are below the temperature range that is commonly used in the production of HMA (usually varying between 150 and 180 °C); therefore, these materials can be readily incorporated into the bitumen (Brasileiro et al., 2019). Plastics may be directly introduced via the wet process into the hot asphalt binder (polymer modified bitumen, PMB.) This is done prior to being used in the production of modified asphalt mixtures (polymer modified asphalt, PMA (Radeef et al., 2021).) Alternatively, the plastics can be added via the dry process to the hot mineral aggregates before the asphalt binder as another aggregate. The dry process promotes simplicity and requires no process modification within the asphalt plant, facilitating the use of waste plastics via this modification technique (Brasileiro et al., 2019). In this way, authors have investigated varying percentage replacements of mineral aggregates with waste polymeric materials in different asphaltic mixture fractions. Many have reported improved asphalt mixture properties such as Marshall stability, indirect tensile strength (ITS), indirect tensile strength ratio (TSR), resistance to permanent deformation and fatigue (Lastra-González et al., 2016b; Awwad and Shbeeb, 2007; Tapkin, 2008; Ahmadinia et al., 2011; Zoorob and Suparma, 2000). This method can utilise a higher amount of waste plastic in comparison to the wet process (by weight of the total mix) and result in a reduction in the bulk density of the compacted mix, proving advantageous in terms of haulage costs (P. Lastra-González et al., 2016). Additionally, upon adding the plastics directly with the hot aggregates, they can soften and coat the aggregates, improving aggregate linkage as well as the adhesiveness between the binder and aggregates. However, the dry process has been reported to have the limitation of performing inconsistently when using high plastic content in the asphalt mixtures (Radeef et al., 2021; Lastra-González et al., 2016b). The majority of published studies prefer the use of the wet process, as this leads to a larger degree of modification and therefore to a better use of the waste plastics for property improvement (Brasileiro et al., 2019). Due to this, studies modifying bitumen binders and subsequent asphalt with waste plastics via the wet process will be primarily reviewed in Section 4.2.

4.2. Polyolefin plastics in asphalt (Binder) modification

Polymers are one of the most important and widely used additives for the improvement of asphalt properties. Synthetic and virgin polymers have been used in asphalt as a binder modifier as early as 1843 for improving asphalt resistance against permanent deformation related to temperature changes (Alghrafy et al., 2021c). However, they are well documented to impart enhanced service properties over a wide range of temperatures in road paving applications (Kalantar et al., 2012; Yu et al., 2015). The use of virgin polymers is uneconomical due to their high cost, thus can only be used in small amounts to enhance asphalt binder properties. With the current challenge of minimizing plastic waste and finding sustainable routes for its valorisation and re-use, a large number of research works have emerged for the incorporation of waste plastics into bitumen and studying the effect of these additives on the properties

of the modified asphalt. It has been shown throughout numerous studies that the modification of bitumen with recycled polymers gives analogous results in enhancing the properties of bitumen as compared to virgin polymers (Zakaria, 2020; Murphy et al., 2000; Attaelmanan et al., 2011). This can be seen further in Table 5, which details the studies that have used both virgin and waste plastics as binder modifiers, obtaining similar conclusions that will be further discussed in Sections 4.2.1–4.2.3. The percentage of polymer added to the binder can vary between 1–10 wt%, the most common being between 3–5 wt% (Brasileiro et al., 2019). The majority of authors employ the wet process to incorporate plastic additives into bitumen, using high shear mixing at a particular temperature until a homogenous blend is achieved. However, this mixing can be done using a variation of process parameters. In Table 5 it can be seen that both virgin and waste PO plastics are typically added at temperatures of 150–190 °C, blended for 0.5–2 h at a blending speed of 200–8200 rpm. The structure and properties of PMBs can be a function of blending conditions, mainly the temperature and blending time. Optimum blending parameters are chosen by evaluating certain blend characteristics including the waste plastic dispersion and storage stability, microstructure, and performance properties of the PMBs (Kalantar et al., 2012; R. Yu et al., 2015).

4.2.1. Polyolefin modified asphalt binders

A wide application of standard rheological parameter tests such as Penetration, Softening Point, Ductility and Viscosity (typically used to characterise neat bitumen grades) have been employed to characterise the performance of waste PO modified bitumen mixtures in literature. A prominent trend in bitumen modification with PO plastics is the decreasing penetration point (increasing stiffness) and increasing softening point of the binder as the proportion of polymer is increased (Brasileiro et al., 2019; Murphy et al., 2000; Attaelmanan et al., 2011; Abdullah et al., 2017). However, it is noted that some authors such as Casey et al. have observed lower variations of penetration and softening point values for PP than those for PE (Casey et al., 2008). Moreover, a substantial rise in viscosity has been observed by authors once more with increasing plastic dosage. HDPE modified asphalt binders have relatively higher viscosities than its LDPE and PP counterparts and have been seen to exceed the Superpave Performance Grade (PG) defined limit of 3000 cP at 135 °C at lower percentage addition. This can be attributed to its more crystalline and higher density structure, with strong forces of attraction between molecules (Alghrafy et al., 2021b; Dalhat and Al-Abdul Wahhab, 2015; Alghrafy et al., 2021). Additionally, the ductility of the PO modified binders can remain at a minimum range (100 cm) of the specification up to certain percentages, which is important at low winter temperatures (Al-Hadidy, 2006). Al-Ghannam et al. observed this up to 8% addition of polyethylene, after which the ductility sharply decreased at further higher percentages (Al-Ghannam, 1996). From the observed trends in standard rheological parameters, it has been established that the addition of PO plastics and their wastes improves thermal susceptibility as well as the resistance to permanent deformation (rutting) at high temperatures (Polacco et al., 2005). Therefore, the use of waste polymers could help to extend the service life of pavements with high traffic loads and those placed in hot climates (Brasileiro et al., 2019; Abdullah et al., 2017; Cardone et al., 2014). Naskar et al. explored the enhancement of binder thermal stability through a number of thermal degradation characteristics, observing enhancements up to 5 wt% waste plastic addition (Naskar et al., 2010). This was attributed to better swelling of the waste plastic by bitumen light components (paraffinic and aromatic compounds), disrupting the crystallinity of the polymer and improving interaction between plastic and bitumen matrix.

The viscoelastic properties of PO modified asphalt binders can additionally be characterised by the complex (stiffness) modulus (G^*) and phase angle (δ), which are obtained from a dynamic shear rheometer (DSR). Yu et al. modified bitumen using 4 wt% waste LDPE and LLDPE and when compared with the original base binder, the G^* of the

Table 5

Existing studies using virgin or waste Polyolefins HDPE/LDPE/PP in Polymer-Modified Bitumen Binders (PMBs) and Asphalts (PMAs.).

Author	Base Binder (pen. grade)	Plastic Modifier	Blending Method	Conclusions
Attaelmanan et al. (Attaelmanan et al., 2011)	80/100	<ul style="list-style-type: none"> HDPE: 1,3,5,7 wt% 	Mixed at 170 °C using a high-speed stirrer, 3000 rpm, 2 h	<ul style="list-style-type: none"> Raised softening point and decreased penetration Modified binders less susceptible to temperature and more favourable for hot climates (higher rutting resistance) Increase in adhesion between aggregate and bitumen, decrease stripping of HMA and moisture susceptibility
Casey et al. (Casey et al., 2008)	200 and 150	<ul style="list-style-type: none"> Waste PP, LDPE, HDPE powder: 2,3,4,5 wt% Waste PP mulch: 2,3,4,5 wt% Blends 4 wt% HDPE/LDPE with 2wt% DETA/ 0.8 wt% PPA Additives: Diethylenetriamine (DETA) 	Heat the base binder to 160 °C, add additive and mix for 60 min, add the plastic, mix for a further 90 min	<ul style="list-style-type: none"> Storage stability not achievable from PP powder and mulch blends 4% LDPE/HDPE concentrations minimises mixture disassociation with improved performance Addition of 0.8 wt% PPA further enhanced performance levels while improving storage stability of blends
Ait-Kadi et al. (Ait-Kadi et al., 1996)	159/200	<ul style="list-style-type: none"> HDPE: 1,3,5 wt% Blends HDPE/EPDM (90/10): 1,3,5 wt% Additives: Ethylene-Propylene-Diene terpolymer (EPDM) 	Prepared at 170 °C using high shear mixing element in jacketed reactor, stored in metallic containers at -4 °C	<ul style="list-style-type: none"> HDPE and HDPE/EPDM results in increased low frequency viscosity and storage modulus The presence of chemical additives like EPDM result in a more stable emulsion
Hinislioglu et al. (Hinislioglu and Agar, 2004)	AC-20*	<ul style="list-style-type: none"> Waste HDPE: 4–6 and 8 wt% 	Vertical shaft mixer, 200 rpm, 145–155 and 165 °C, 5–15 and 30 min	<ul style="list-style-type: none"> 4% HDPE binder prepared at 165 °C for 30 min has the highest Marshall Stability HDPE mixes highly resistant to permanent deformation (rutting) in asphalt
Fang et al. (Fang et al., 2014)	90A#*	<ul style="list-style-type: none"> Waste PE (WPE): 4 wt% 	High shear emulsifier, 3750 rpm, 150, 170, 190 °C, 0.5, 1, 1.5, 2 h, particles added over 5 min	<ul style="list-style-type: none"> Optimum WPE dispersion in the mixture is achieved at 150 °C after 1.5 h of shearing Relatively good storage stability achieved Decreased temperature sensitivity of modified bitumen

* Sinopec Xi'an Petrochemical Company, Styrene-butadiene rubber (SBR) Modified Asphalt.

modified binders was increased, showing that the non-deformability of modified asphalt binders is enhanced. The phase angle (δ) of the modified binders was smaller than that of the base binders, which, combined with the enhanced G^* parameter, the storage modulus (G' , equals $G^* \cdot \cos\delta$) of the modified binders increased, indicating an increase in the elastic component (Yu et al., 2015). Cardone et al. also observed that the presence of a polymer network in 2–6 wt% plastic modified bitumen leads to an increase in stiffness (G^*) and the elastic properties of the material (lower phase angle (Cardone et al., 2014).) Abdullah et al. blended 1.5–6 wt% waste plastic with a 60/70 bitumen binder and focused on the rutting factor $G^*/\sin(\delta)$ obtained from DSR, finding that a higher percentage of plastic waste will result in a higher rutting resistance (Abdullah et al., 2017). To further compare the rutting behaviour of short-term aged binders, the recoverable strain (R) and non-recoverable creep compliance J_{nr} are also measured from the Multiple Stress Creep and Recovery (MSCR) test. Authors have demonstrated a reduced total strain with higher dosages of PO plastics. The PMBs have higher R values and lower J_{nr} values with increased dosages, as the inclusion of polymeric chains in the asphalt binder restricts and thus improves resistance to permanent deformation (Moghadas Nejad et al., 2015; Roja et al., 2021; Amirkhani, 2018).

An asphalt binder will have higher resistance to fatigue cracking if it is more elastic and not too stiff (Ghuzlan et al., 2022). Multiple authors have conducted recent popular tests (the Linear Amplitude Sweep test, for example) to determine the fatigue life of long-term aged, modified binders by calculating the damage accumulation. In a study conducted by Amirkhani et al., it was shown that the addition of PE improves binder fatigue life in comparison to the base binders, regardless of the PE type, content and applied shear strain (Amirkhani, 2018). Roja et al. showed that a 3–5 wt% addition of lower molecular weight (M_w) LDPE will improve the fatigue life of the binder, due to increased chain mobility (Roja et al., 2021). Despite this, some authors have alternatively found that PO plastics fail to improve the elastic recovery and stress relaxation of the binder, indicating the potential for thermal and

fatigue cracking at low in-service temperatures (Zhang et al., 2022). Isacson et al. found that the addition of PE failed to significantly improve the low-temperature flexibility of bitumen (Isacson and Lu, 1995). This was attributed to its regular long chain structure, the PE molecule is prone pack closely and crystallize, which could lead to a lack of interaction between the bitumen and polymer and result in instability of the modified bitumen.

4.2.2. Polyolefin modified asphalt mixtures

The stability of an asphalt mixture verifies its performance under loading. A prominent trend in literature is that the addition of recycled PO plastic can substantially increase the stiffness and thus stability of the asphalt mixture, but only up to certain percentages, after which the stability will decrease. For example, authors have determined the highest Marshall stability values to be at a 4% dosage for HDPE (Hinislioglu and Agar, 2004; Abdullah et al., 2022). Hinislioglu et al. saw a stability increase in the range of 3–21% using 1–4 wt% HDPE in asphalt concrete formulated via the wet process, with other authors seeing similar results (Hinislioglu et al., 2005; Tapkin et al., 2009). The higher stabilities have been suggested to be attributable to higher cohesion of the binder and internal friction due to the plastic addition, indicated by lower flow values for modified specimens (Hinislioglu and Agar, 2004). In comparison, Zoorob et al. applied the dry method for the formulation of 'Plastiphalt', replacing 29.7 wt% of aggregates with LDPE and found a Marshall stability of approximately 2.5 times the control mix (Zoorob and Suparna, 2000). The recorded flow values were also higher, indicating that the mixes were both stronger and more elastic. The Marshall Quotient (MQ) is largely used as an indicator of the resistance against deformation of asphalt. Higher stability and lower flow values of Marshall specimens modified with PO plastics seen in various studies supports the increase in MQ values. This has been used to conclude that mixes blended with PO plastics are highly resistant to rutting at high temperatures (Hinislioglu and Agar, 2004; Hinislioglu et al., 2005; Tapkin et al., 2009; Tapkin et al., 2009).

The dynamic creep has been widely used to characterise the permanent strain behaviour of paving mixtures at relatively high temperatures where the flexible pavements are more susceptible to rutting distress (Goh and You, Nov. 2009). The influence of PO plastics on the Marshall properties of asphalt provides an increase of the service life under repeated creep testing (Napiyah et al., 2014; Abdullah et al., 2016). Nejad et al. saw such results with the addition of 5 wt% HDPE into the bitumen binder (Moghadas Nejad, Azarhoosh, and Hamed, 2014). It was also observed that moisture increased the rutting potential the least for the modified binders, indicating a decreased moisture susceptibility. Tapkin et al. observed a substantial increase in the service life of PP fibre-reinforced asphalt specimens under repeated loading, to the extent that the control specimens entered the tertiary stage of creep at the loading rate corresponding to the primary stage of creep for the modified mixes (Tapkin et al., 2009). However, it is importantly noted by some authors that due to the stiffening effect of the polymers, mixtures with the highest polymer percentage are rather incompressible and have lower workability (Tapkin, 2008; Napiyah et al., 2014). This is an important consideration in terms mixture pumpability and the potential increase in energy and emissions required for the PMA formulation. This also may present a limit on the amount of each plastic that can be added to the flexible pavement via this production technique. Especially when plastics used in the 'wet' process already account for a very small percentage of the overall mass, given that bitumen is approximately 5% of the total mix (Santos et al., 2021a). Care must also be exercised with very high stiffness mixes due to their lower tensile strain capacity to failure. Such mixes are more likely to fail by cracking, particularly when laid over foundations which fail to provide adequate support (Zoorob and Suparna, 2000). Regardless, multiple authors have agreed in fatigue life studies for PO modified asphalt that for a thick pavement that is evaluated in the constant stress mode of testing, increasing stiffness of the asphalt binder causes a better fatigue life in the asphalt mixture (Moghadas Nejad, Azarhoosh, and Hamed, 2014; Roberts et al., 2009). Nejad et al. saw the fatigue life of 5 wt% HDPE asphalt mixtures to be 1.16 times the control mixture when conducting Indirect Tensile Fatigue (IFT) tests (Moghadas Nejad, et al., 2014). Other authors have found an improvement in tensile strength and related properties of mixtures containing polymer fibres, especially polypropylene. Tapkin et al. (Zoorob and Suparna, 2000). found that a 0.3–1% addition of polypropylene fibres into asphalt concrete in a dry bases prolonged the fatigue life with less reflection cracking. Flexible pavements are additionally susceptible to the damaging effects of moisture, causing the displacement of the bitumen film from the aggregate surface in the presence of water. In literature it has been demonstrated via various techniques (Indirect Tensile Strength (ITS) and Tensile Strength Ratio (TSR), Marshall Stability ratio of 'wet and 'dry' specimens) that PO plastics improve binder-aggregate adhesion as higher moisture resistance is gained. Therefore, PO modified PMAs will have higher durability and service life with potentially lower construction and maintenance costs (Attaelmanan et al., 2011; Moghadas et al., 2014; Köfteci, 2016; Abdel-Wahed et al., 2020). In addition, Haider et al. found that PE modified asphalt mixtures through the wet process showed better adhesion and moisture resistance than those modified through the dry process (Haider et al., 2020). However, plastic coated aggregates have too been demonstrated to favour the impact, crushing, stripping and abrasion resistance of aggregates (Bajpai et al., 2017).

4.2.3. Limitations of polyolefin modified asphalt (Binders)

Modifying bitumen binders and the subsequent asphalt with waste PE and PP plastics is an attractive option for the low-cost improvement of asphalt concrete mixes at 3–5 wt% of bitumen (Ibrahim, 2019). However, despite the advantages of PO modification, there are several potential challenges that could limit its future development and commercialisation. Some are related to the chemical compatibility between waste plastics and bitumen, leading to phase separation and poor storage stability that has been reported in various works (Casey et al., 2008;

Pérez-Lepe et al., 2007; Fang et al., 2014). The tendency for long chain PO plastics to pack closely and crystallise can lead to a lack of interaction with the bitumen and result in the instability of the modified binder. Compatibility can also be poor due to the non-polar nature of the polymers, making them completely immiscible with the bitumen (Zhu, 2016; Polacco et al., 2006). Other factors such as viscosity and density differences, the molecular weight distribution, melt flow index (MFI), concentration, and the degree of branching of the polymers as well as the aromaticity and asphaltene content of the base binders have additionally been linked to determining the compatibility of PMB mixtures (Pérez-Lepe et al., 2007; Masad et al., 2020; Lesueur, 30, 2009a). A popular solution is the use of polyolefin-based copolymers that can have polar components and are either inert or reactive with the asphalt binder. Elastomers such as ethylene-vinyl acetate (EVA) and ethylene-propylene-diene (EPDM) have received much research attention for steric emulsion stabilization and can be effective (as seen in some of the studies included in Table 5.) The copolymers can additionally enhance binder properties that the polyolefin plastics cannot improve alone (Murphy et al., 2000; Ait-Kadi et al., 1996; Polacco et al., 2005; García-Morales et al., 2006). However, as reported by Pérez-Lepe et al., upon mixing various (EPDM/PE) blend compositions, these materials are not always efficient due to not being able to entangle sufficiently with the polyolefins in the absence of cross-linking promoters, therefore the stabilising effect is not fulfilled (Pérez-Lepe et al., 2007). Alternatively, reactive compatibilizer agents that modify the olefinic chain with additional functional groups can improve the miscibility of the polymer and bitumen and enhance binder characteristics further, for example maleic anhydride (MA), glycidyl methacrylate (GMA), polyphosphoric acid (PA) and reactive ethylene terpolymers (RET) (Brasileiro et al., 2019; Polacco et al., 2005; Giavarini et al., 1996; Yeh et al., 2005). Despite this, the use of compatibilizers could limit the economic benefits of using recycled materials and in some cases, reactive additives do not always result in sufficient enhancements to obtain a homogeneous and stable mix (Polacco et al., 2005). Another unknown is whether the enhanced material properties e.g., increased rutting resistance, less temperature susceptibility, and elevated adhesion, will remain being enhanced when the PSW mixing weight is at a higher percentage (greater than 10% of bitumen.)

Other authors have focused more on the environmental aspects that mixing bitumen with waste plastics could pose, such as fuming, emissions during production, contamination and microplastics (Santos et al., 2021). If waste plastics or other organic recycled material will be used in asphalts, more research is needed to assess their effects on workers' exposure and health, due to mutagenic potency. The sorting of plastics before possible use in asphalt may also be important in order to remove plastics that decompose to hazardous compounds in heating, for example the noxious HCl gas from Polyvinyl Chloride (PVC) (Chakraborty and Mehta, 2017; Heikkilä et al., 2003). The potential of waste plastic fragmentation into microplastics upon introduction in asphalt pavement layers and their role as a pollutant with potentially serious ecological impacts is also in need of further research (Andrady, 2017). Additionally, there are rising concerns regarding the future recyclability of the asphalt and whether this could be hindered by the incorporation of waste materials (Rodríguez-Fernández et al., 2020). The increased viscosity of PMB blends unfortunately results in the decreased workability of the binder, needing to increase the mixing and compaction temperatures for manufacturing. This increase in energy consumption is undesirable and creates environmental concerns about emissions generated (Köfteci et al., 2014).

4.2.4. Life cycle analysis and cost assessment

The potential environmental effects of using waste materials in road pavement applications must be quantitatively evaluated and numerous life cycles assessment (LCA) studies have been conducted in literature for this purpose (Santos et al., 2014; Praticò et al., 2020). Santos et al. conducted a LCA investigating the impact of recycling waste plastics

from local processing facilities in asphalt mixes when incorporated via the wet and dry method (Santos et al., 2021a). It showed that for the conditions and assumptions considered in the study, recycling PE plastics (as a substitute of virgin plastics) using the wet method (up to 8% addition) can be beneficial for the environment by reducing greenhouse gas (GHG) emissions up to approximately 16%. However, the use of PMB is more environmentally burdensome than the use of standard bitumen due to the fact that asphalt binder modification implies the consumption of at least one additional material. However, as discussed by Zhu et al., environmental benefits are expected to be observed in the other pavement life cycle phases in terms of reduced maintenance costs as a result of increased pavement failure resistances and greater adhesion between asphalt binder and aggregates over the service life (Zhu et al., 2014). On the other hand, the production of asphalt mixes via the dry method consumes a larger quantity of recycled plastic, however, the environmental benefits of this technique are diminished. The GHG emissions increased by 160% when 20% recycled plastic was included in the mixes as a replacement of virgin quarry aggregate. The increase in certain impact categories is due to a greater energy demand required to produce the recycled plastic pellets. Though, it was seen that using a lower percentage aggregate replacement can still use a sizable amount of waste plastic without significantly increasing the number of emissions generated (Santos et al., 2021a). The LCA conducted by Oreto et al. also concluded that wet and dry plastic modification of asphalt mixtures does entail additional environmental burdens compared to the traditional HMA binders (Oreto et al., 2021). This was attributed to factors such as the plastic pelletization process and long transportation distances. However, human carcinogen toxicity was lowered through the reduction of particulate matter and polycyclic aromatic hydrocarbons emitted during the recycling and production of plastic pellets compared to industrial modification of bitumen with virgin polymers. The predicted service life (using the Boussinesq theory and the calculation of the stress-strain state of each pavement structural layer, in compliance with the fatigue damage and rutting accumulation laws) demonstrated the use of recycled polymers in HMA binder and base layers improved the service life compared to traditional HMA without significantly affecting the environmental burdens.

In terms of cost effectiveness, binder modification using recycled plastic in PMA is considered cost effective as this is a cheaper resource than virgin polymers, while reducing bitumen use. However, the cost of recycled plastic aggregates needs further evaluation as it could potentially be much higher than that of quarry aggregate due to pelletization production processes required (Santos et al., 2021a). Alghrafi et al. conducted an economic study in terms of initial cost and performance for various recycled PE modified binders (Alghrafi et al., 2021a). The authors found that in general, 2% and 1% in the initial cost of binder can be saved when using 6% LDPE and 2% HDPE to modify virgin asphalt. Furthermore, the cost could be further substantially decreased up to 29% when modifying sulphur extended asphalt with recycled PE plastics. Furthermore, Souliman et al. divided the expected performance (predicted number of fatigue cycles to failure) by the mixture unit cost of one mile for unmodified and polymer modified asphalt (Souliman et al., 2016). The cost effectiveness value of the PMA was 105.2 cycles per cost of one mile in comparison to 41.2 cycles for the unmodified mixture.

4.3. Pyrolysis polyolefins in asphalt (Binder) modification

As discussed in Section 3, waste PO plastics can be thermally cracked into smaller molecular products via thermochemical technologies such as pyrolysis. It has been common in the past for authors to use pyrolysis for waste material treatment prior to the preparation of bio-binders/bio-asphalt, especially for biomasses, crumb rubbers and cooking oils (Dong et al., 2019; Kolokolova, 2014). Pyrolysis could easily be applied in the same way as an acceptable strategy for upgrading waste plastics for this product application, through the concept of Design from Recycling. In this way, PO pyrolysis products could have large potential as a plastic

derived additive for HMA. The idea of using pyrolysis plastics in asphalt binder and subsequent mixture modification is not entirely new but is currently not widely researched in literature. Al-Hadidy et al. proposed the use of pyrolysis PP as a new modifier material, which was thermally degraded at 308 °C and grinded into powder before being incorporated into 40/50 grade bitumen at 1–8 wt% (Al-Hadidy, 2006). The pyrolysis plastic was mixed for 7 min at 150 °C, which is a substantially smaller time in comparison to raw recycled PP, as can be seen in Table 5. It was determined that the plastic is well dissolved into the binder and compatible to an extent. Similar to PP modification, the penetration point decreases while the binder stiffness, softening point and resistance to temperature susceptibility increases. The modified asphalt mixture showed an increase in Marshall stability and decrease in flow up to 2% PP content. From the indirect tensile strength test, it was concluded that the addition of pyrolysis PP increases the adhesion between aggregate and asphalt, indicating increased stripping resistance. The pyrolysis PP enhanced the resistance to permanent deformation and rutting. Later, Al-Hadidy et al. conducted a similar study using pyrolysis PP with an aim to produce a homogeneous a storage stable PMB (Al-Hadidy and Yi-qiu, 2009). The PP was pyrolysed at 400 °C for 2 h, producing a brown coloured monomer material with a lower density and melting point range of 153–157 °C. This was mechanically ground and incorporated at 1–11 wt% into 50/60 grade bitumen at 160 °C for 5 min by slow speed shearing. The study produced analogous conclusions to (Al-Hadidy, 2006).

The same authors then proposed to use pyrolysis LDPE as a modifier material in the stone mastic asphalt (SMA) layer of flexible pavements (Al-Hadidy and Yi-qiu, 2009). The LDPE was subjected to thermal degradation at 406 °C in a stirred batch reactor set-up, depicted in Fig. 4. The reactor was heated using an internal heater connected to a temperature controller with corresponding temperature thermocouples. Upon heating, the feedstock was stirred, and upon cracking, left the system and entered a storage tank for collection. The pyrolysis LDPE was incorporated into bitumen at 2–8 wt% using a high-speed stirrer at 160 °C and 1750 rpm for 3–5 min. It was demonstrated that the pyrolysis LDPE can be well dissolved in the asphalt matrix and that it was compatible to an extent. The modified binders were less susceptible to temperature and had improved resistance to deformation. The Marshall stability increased with a decreasing flow up to 6 wt% LDPE addition. It was shown that the modified SMA specimens did not weaken when exposed to moisture and that the pyrolysis LDPE was effective in preventing excessive drain down of the mixtures (bleeding phenomenon.) It was speculated that after thermal degradation, LDPE contains a larger number of activated groups which made the chemical reaction with bituminous molecules easier and contributed to the indicated improvements in resistance to deformation. In addition, the pyrolysis LDPE reduced cracking potential at low temperatures (–10 °C.)

So far, pyrolysis PO plastics in asphalt binder and mixture modification has resulted in very similar results to raw recycled plastic modification (see Section 4.2.1–4.2.2.) From the publications reviewed, an issue arises being whether the additional cost and further process environmental burdens associated with using pyrolysis plastics outweigh the advantages of using this new modifier material. A clear advantage is the ability to use lower mixing times and temperatures while achieving seemingly good mixture homogeneity, as the lower density and molecular weight additives can be completely dissolved in the bitumen. Despite this, rising mixture viscosities with an increase in pyrolysis plastic addition could still be a concern, as this suggests potential issues associated with blend workability or a limit on the amount of pyrolysis plastic that can be used. It is noted that the pyrolysis temperatures used in the works discussed are much lower than those commonly used in plastic pyrolysis literature (Section 3.) Looking at the thermogravimetric curves for the PO plastics, they typically completely thermally degrade at higher temperatures (Mofokeng et al., 2012; Tajeddin and Abdulah, 2010). This serves as explanation for the monomer materials produced with a melting point range of 153–157 °C

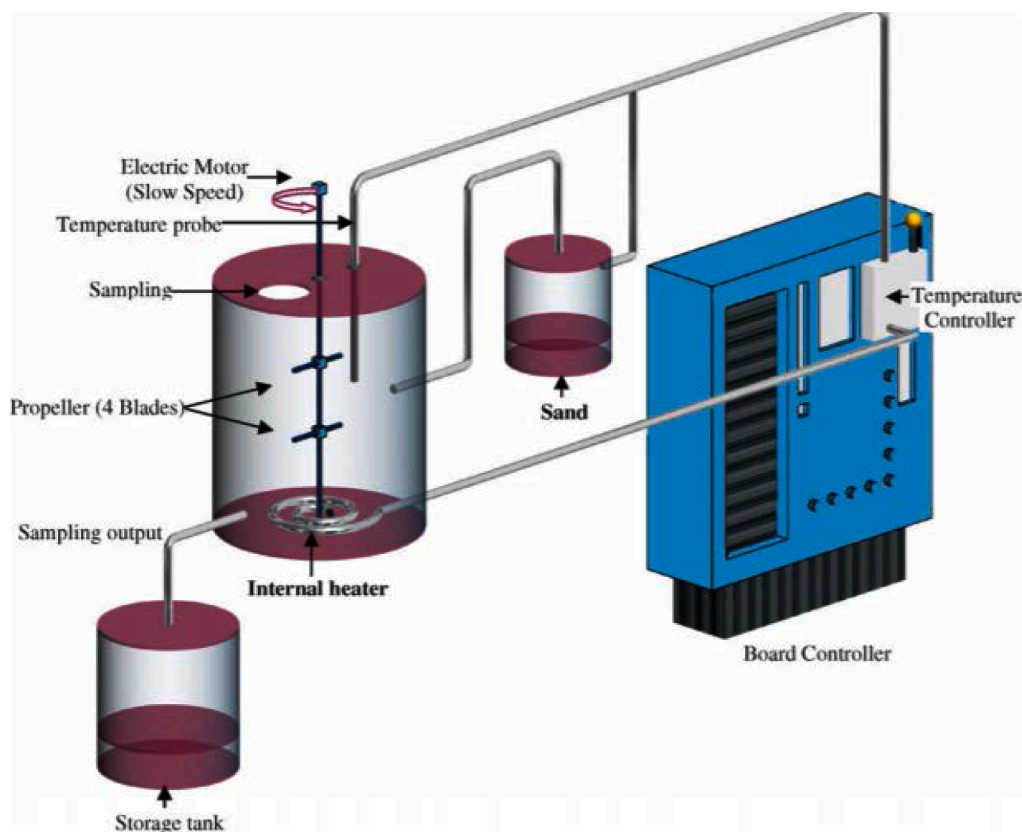


Fig. 4. Thermal degradation stirred reactor system for pyrolysis PP, used by Al-Hadidy et al. (Al-Hadidy and Yi-qiu, 2009).

that could be mechanically ground in certain studies (Al-Hadidy, 2006). Therefore, the extent of cracking of the plastics may not have been enough to allow for any real changes to be seen, in comparison to the parent polymers. More research in this area that can provide more detailed pyrolysis processes and degraded product characterisation, as well as exploration into the modification mechanism between this new modifier and bitumen is needed. Additionally, further studies into the extent of compatibility of pyrolysis plastics and binders are needed to determine whether there are outstanding concerns with the storage stability of plastic-derived modifiers. Furthermore, LCA and cost assessments including the pyrolysis process to upgrade recycled plastics for use in HMA production will be beneficial in the future.

Considering the uses of pyrolysis products from other waste materials and blends in flexible pavements, multiple authors have noted the potential of pyrolysis oils as a rejuvenator for aged bitumen to reduce stiffness (Yang et al., 2018a; Ling et al., 2019). Due to their softening effect, bio-oils have been widely studied in the recycling of end-of-life reclaimed asphalt pavement (RAP.) They offer a double advantage of greater RAP amount in the asphalt mix and superior recycled asphalt functioning. It is observed that the majority of literature surrounding this topic utilises oils largely derived from sources such as biomasses and cooking/vegetable oils (Gong et al., 2016; Raouf and Williams, 2011). This may be attributed to the large potential of PO plastic pyrolysis oils as alternative fuels, diverting research attractiveness. Further research into the effectiveness of PO plastic pyrolysis oils as sole rejuvenators for RAP could be conducted, apart from works that have previously studied combined polymer modified bio-derived rejuvenators (Wang et al., 2021). Alternatively, recent publications have been conducted demonstrating the viability of biochar (derived from biomasses) for use in asphalt mixtures as bitumen modifiers and cement-based composites as nanoparticle fillers (Kumar and Choudhary, 2020; Cosentino et al., 2018; Yang et al., 2013). Such investigations are again limited for such plastic pyrolysis products, highlighting a research gap in the full

understanding of plastic pyrolysis products as flexible road materials.

4.3.1. Plastic waxes as flexible road materials

Recently it has been suggested that due to the drawbacks associated with the waste plastic modification of asphalt binders, they instead be incorporated into warm mix asphalt (WMA) technologies. WMA mixtures have lower production temperatures of 100–140 °C (as opposed to HMA > 150 °C) and therefore reduce the required energy, cost, fuel consumption and hazardous fumes, without compromising the performance of the mix (Zaumanis, 2010). Use of higher amounts of RAP is reported to be feasible in WMA mixes, with ongoing research into new additives and rejuvenators (Guan et al., 2016). There are various additives and foaming technologies that have been developed for WMA, one of which is organic additives. Organic additives function by reducing the viscosity of the base asphalt binder due to the presence of waxes and have been widely applied to improve the workability of high viscosity asphalt binders such as rubberised asphalt (Ling et al., 2019; Sukhija and Saboo, 2021). A popular wax additive applied in literature from wax studios such as Sasol is the “Fischer-Tropsch” (Sasobit) paraffin waxes, produced synthetically from syngas (Desidery and Lanotte, 2021). While they reduce the viscosity of the bitumen at mixing and compaction temperatures, depending on the wax chain length, they can also generate stiffer materials at in-service conditions. Studies have shown an improved resistance of the mixtures against permanent deformation as well as improved interaction between modifiers (such as crumb rubber) and base asphalt (Fazaeli et al., 2012; Sasol, 2020; Yu et al., 2016).

Polyethylene wax is a lower molecular weight by-product of the polymerization process and is also widely studied for its application as a WMA additive (Nakhaei et al., 2016; Edwards, 2022). Authors have reported its incorporation in the form of fine-grained powder or granules into asphalt binder for mixture formulation (Valentová et al., 2017). The waxes have been seen to improve the moisture resistance of warm asphalt mixes; however, it is recommended by some authors to be used

as a warm mix additive at lower compaction temperatures and implemented in colder regions due potential rutting distress (Nakhaei et al., 2016). However, PE wax-based WMA additives have additionally been studied for its influence on polymer and rubber modified WMA mixtures, that provide high rutting resistance yet require higher mixing and compacting temperatures due to higher viscosities. Kim et al. compared polymer modified, stone mastic HMA and WMA (with 1.5% polyethylene wax additive) and concluded that the polymer modified WMA with the wax additive had better performance characteristics (Kim et al., 2013). The modified WMA had superior moisture, rutting, fatigue, and low temperature crack resistance than the modified HMA. A publication to note is that by Shang et al., using pyrolysed wax from recycled crosslinked polyethylene as a WMA additive for crumb-rubber modified asphalt (Shang and Wang, 2011). The pyrolysed wax was incorporated into the asphalt binder at 1–7% at 150 °C for 30 min. Its addition increased the softening point as well as the penetration point of the asphalt, while reducing the ductility negligibly. The addition of the wax also improved the rutting resistance parameter.

The production of warm mix asphalt (WMA) has been a topic of discussion for many years within the paving industry yet has not become the standard for types of rolled asphalt mixes. The complete estimation of the potential economic benefits of using WMA technologies still needs to be explored and may not be feasible if not jointly evaluated with environmental regulations and additional costs and offsets of WMA production (Ling et al., 2019; Diab et al., 2016). Furthermore, at HMA production temperatures, Roja et al. demonstrated that a 1% addition of PE wax to LDPE modified binders improved the dispersion of low melt flow index polymers (Roja et al., 2021). The addition of PE wax reduced viscosity of the blends as well as the strain response and non-recoverable creep compliance of the mixtures (however, not to the extent of the binders with only LDPE additives.) The addition of the PE wax alone deteriorated the fatigue life of the binders, except when blended with low molecular weight LDPE with increased polymer chain mobility. Zhang et al. reported an important relationship between the molecular weight (M_w) of LDPE and HDPE waxes and the modifying process (Zhang et al., 2021). As the M_w of the waxes increases, the viscosity, moisture and low temperature resistance capacity of the wax modified asphalt mixtures increases, while the storage stability decreases. The high temperature resistance capacity of the PE wax modified mixtures was improved at higher M_w waxes also. Ling et al. (Ling et al., 2019). produced blends of pyrolysis oils and waxes from waste rubber and plastic feedstock (WRP) that increased both the workability and mechanical properties including rutting resistance, low-temperature, and fatigue life of the asphalt binder. A clear benefit of using plastic waxes in both hot and warm mix asphalt binders and subsequent mixes is the significantly lower mixing times (10–30 min (Ling et al., 2019; Shang and Wang, 2011)) and/or temperatures (reduced from 20 to 60 °C (Diab et al., 2016).) Therefore, there is a lower cost of manufacturing, especially in comparison to raw plastic modification blending methods (see Table 5.)

4.3.2. Plastic pyrolysis wax applications

It is typical in the pyrolysis of municipal plastics wastes (MPWs) that contain a high fraction of polyolefin polymers that aliphatic waxes are the main product at temperatures of around 500 °C and that the pyrolytic oil obtained also has a high content of wax in the form of heavy hydrocarbons. The undesirable qualities of this low-grade wax-oil mixture have attracted research attention around its potential as a cracking feedstock, upgrading the pyrolysis wax-oil into high-value fuels and petrochemicals (Quesada et al., 2020; Arabiourrutia et al., 2012a). Currently, catalytic degradation using solid acid catalysts such as zeolites, silica-alumina, fresh FCC catalysts and MCM-41 is the most commonly used method. HZSM-5 catalysts especially are well documented for the upgrading of pyrolytic wax-oils (Artetxe et al., 2013; Lee and Oh., 2012). Furthermore, it has been established in literature the suitability of different pyrolytic wax fractions for different upgrading

processes. Light product waxes with a boiling point below 500 °C are typically planned for use as a (co-) feed in a steam cracker to yield monomers for polymer production, whereas the waxes with a boiling range above 500 °C are planned for use in fluid catalytic crackers (FCC-process) to yield petrochemical products, as aforementioned (Predel and Kaminsky, 2000). A commercial example of using paraffinic wax as a feed for refineries or steam crackers is the BP Polymer Cracking Process pilot plant, utilising a feedstock of mixed plastic wastes (Tukker and Picard, 1999). The higher heating value (HHV) analysis of pyrolytic waxes produced from cracking at 600 °C have been determined as comparable to original polymers and not much lower than gasoline or natural gas. Although an increase in olefin nature has been observed at higher processing temperatures, the HHV values are in the same order as standard fuels (Arabiourrutia et al., 2012a).

Alternatively, the direct combustion of pyrolytic waxes for energy is a suggested option if there is no commercial interest for their use as raw materials for production of fuels or chemicals. It has also been suggested that the product waxes could be gasified to hydrogen to make them more commercially beneficial (Onwudili et al., 2009). Additionally, properties such as the freezing point of polyethylene (PE) wax from degradation do not in some cases make it feasible for gasoline and it would be more profitable to convert to wax compositions alike to commercial PE wax. Commercial PE wax has a wide application field including lubricants, additives in polyolefin processing, dyes, inks, antioxidants, candles, toys. Most commercial waxes such as this are produced from PE resins, making production cost high and the world market in short supply. Therefore, a technology such as pyrolysis has large potential and very strong competitive capability for the production of PE wax especially (Jixing, 2003). Multiple studies have been conducted for the investigation of pyrolysis parameters influence on waste polyethylene, to determine the applicability of this thermochemical technology for the production of waxes of ASTM commercial standard. With the careful control of parameters, waxes within the same melting point ranges as paraffinic and microcrystalline waxes and with penetration points close to the material standard range can be achieved (Ademiluyi et al., 2013; Jixing, 2003). Compositional analysis has shown that degradation waxes are less branched and more olefinic than commercial waxes. Few authors have demonstrated that the application properties of waxes are only influenced to a minor extent by the olefinic groups, yet perhaps further investigation into their suitability for certain commercial applications may be necessary (Aguado et al., 2002b; Chaala et al., 1997). In terms of commercialisation, the first waste packaging plastic conversion pyrolysis facility opened in 2019 by Trifol in Ireland for the target production of 'Envirowax'. This product is estimated to remove up to two tonnes of plastic waste with each tonne of wax produced and can be used for the production of cosmetics, candles, lubricants and chewing gum (BaleBid, 2019). This commercialisation is significant as the technology is readily available for scaling up when further applications within other industries are identified for pyrolytic polyolefin wax. All established and on-going researched applications for pyrolytic waxes from waste polyolefin pyrolysis within the petrochemical and chemical industries have been summarised in Fig. 5. With the current commercialisation of plastic pyrolysis waxes for use in typical commercial PE wax applications, as well as research and pilot plant-based investigations into the production of feedstocks for the petrochemical and chemical industries, it is yet still noted that this trend in research is less prominent than that for the production of pyrolysis oils from waste plastics for use in fuels. As well as this, outside of the applications stated, there is a general lack in knowledge of the known applications of pyrolysis wax outside of these industries.

5. Discussion and conclusion

An extensive review of the pyrolysis of the abundant polyolefins found in municipal waste streams for the production of heavy wax products has been conducted. Additionally, the use of these plastics as

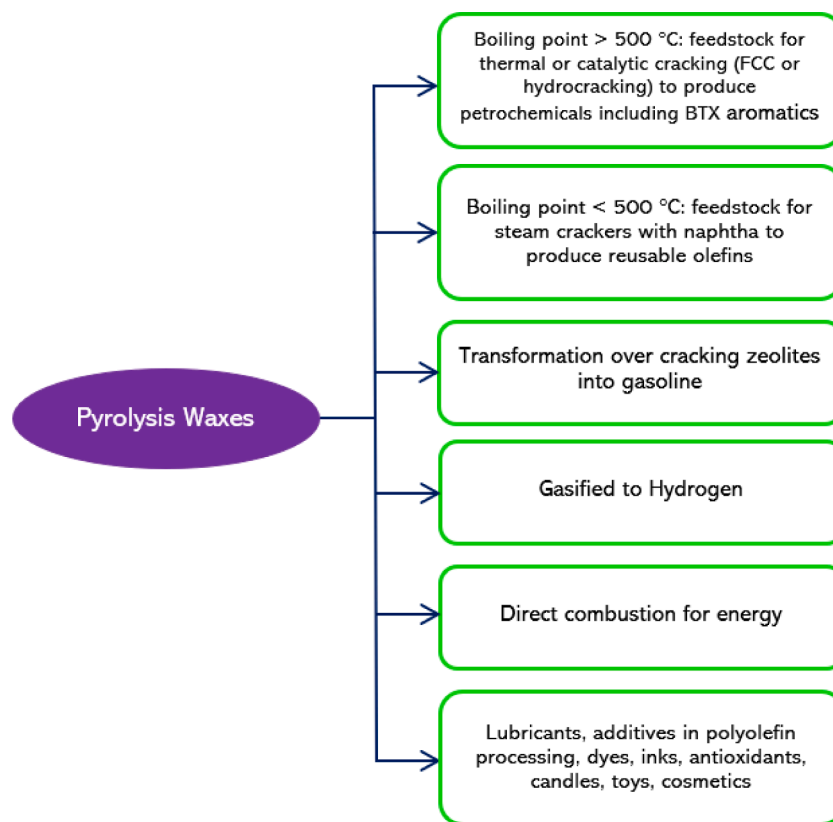


Fig. 5. Established and on-going researched applications/upgrading routes for plastic pyrolytic waxes within the petrochemical and industries.

alternative materials in flexible pavements was investigated, specifically in asphalt binder and mixture modification. Furthermore, the use of plastic wax-based technologies in WMA and as modifiers in HMA was presented, providing discussion for the potential future use of PO plastics-derived wax products from pyrolysis as a modifier material in HMA. The key points of discussion and resultant conclusions of this review article are as follows.

- Pyrolysis is a promising thermochemical technology for the treatment of co-mingled plastic wastes that still largely go to landfill. From an environmental perspective, a multitude of author reviews and assessments indicate its advantage when compared to conventional recycling methods. It additionally has the capacity for much future research and development to overcome its technological limitations.
- The current commercialisation of pyrolysis for waste plastic treatment to fuel oils and waxes implies that the technology is readily available for adaptation and scaling up. However, in terms of the heavy wax fraction that is a main product of polyolefin thermal pyrolysis, a general lack in knowledge of its known applications outside of the fuel and chemical feedstock industries was identified.
- The use of recycled polyolefin plastics in flexible pavements is a practical route as it has the capacity for incorporating high volumes of plastics, as well as reducing the cost of virgin materials. Their use in the modification of asphalt binders and subsequent mixtures, manufactured via the wet process, was mainly evaluated due to its prevalence in literature. These recycled modifiers offer a multiplicity of enhanced service properties over a wide range of temperatures, especially improving rutting resistance at high in-service temperatures as well as temperature and stripping susceptibility.
- LCA and cost assessment data from literature also support its use as opposed to the use of virgin plastics. However, it is not indicated as beneficial when compared to the traditional use of virgin binders due

to the associated cost and environmental burdens with extra process steps and materials. It is recommended that extended investigation into the environmental and cost savings over the service life of the modified pavements needs to be conducted in the future, considering the increased service life and lesser maintenance costs.

- The technical limitations with storage stability of polymer modified blends as well as environmental concerns of contamination, microplasticity and fuming remain as major hindrances to its future development and commercialisation. Such problems bring restrictions to the overall amount of waste plastic that can be used in this particular application, using either manufacturing method.
- Alternatively, the use of polyolefin pyrolysis products was considered in asphalt modification. From the available publications that were reviewed, a query arises being whether the additional cost and further process environmental burdens associated with using pyrolysis plastics outweigh the similar results obtained for this new modifier material in comparison to raw plastic modifiers.
- Future research is important to provide more detailed pyrolysis product characterisation and exploration into the modification mechanism between this new modifier and bitumen. Additionally, further studies into the extent of compatibility of pyrolysis plastics and binders are needed to determine whether there are outstanding concerns with the storage stability of plastic-derived modifiers.
- Furthermore, LCA and cost assessments including the pyrolysis process to upgrade recycled plastics for use in HMA production will be beneficial in the future.
- The use of plastic-based waxes in warm mix technologies are seen to be beneficial from the perspective of performance, as well as the cost and time savings associated with manufacturing. A wax-based additive derived from the pyrolysis of recycled PO plastics for possible use in unmodified and modified HMA, WMA and RAP recycled asphalt applications could potentially be a viable solution to the current limitations associated with raw plastic modifiers. This may

also offer environmental and economic benefits that are associated with wax-based modification and the increased use of recyclable materials in the flexible roads.

- At the moment, there is little comprehensive understanding of the full capacity for this pyrolysis product (and others) in industries such as infrastructure and construction and is worth more research, as part of Design from Recycling concepts.

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.

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