UNIVERSITY OF PALERMO

DEPARTMENT OF ENGINEERING

Doctoral course in Advances in Modelling, Health-Monitoring, Infrastructures, Geomatics, Geotechnics, Hazards, Engineering Structures, Transportation (AIM HIGHEST)

Scientific-Disciplinary Sector: ICAR/04 – Roads, Railways and Airports
Curriculum: Infrastructure, Geomatics and Transport Engineering

A thesis submitted to the University of Palermo in fulfilment of the requirements for the degree of European Doctor of Philosophy

The Frontiers of Multi-Recyclability of Bituminous Mixtures with Recycled Plastic under the Scope of Circular Economy

Ph.D. Candidate
Vineesh Vijayan

Ph.D. Coordinator
Prof. Giuseppe Campione

Advisor
Prof. Gaetano Di Mino

Co-Advisors
Prof. Evangelos Manthos
Dr. Ing. Konstantinos Mantalovas

Cycle XXXVI
Academic Year 2022-2023

UNIVERSITÀ DEGLI STUDI DI PALERMO

DIPARTIMENTO DI INGEGNERIA

Corso di Dottorato di ricerca in Advances in Modelling, Health-Monitoring, Infrastructures, Geomatics, Geotechnics, Hazards, Engineering Structures, Transportation (AIM HIGHEST)

Settori Scientifico-Disciplinari: ICAR/04 - Strade, ferrovie ed aeroporti
Curriculum: Infrastrutture, Geomatica and Ingegneria Trasporti

Tesi presentata all'Università degli Studi di Palermo per il conseguimento del titolo di Doctor Europaeus

La Frontiere della Multiriciclabilità dei Conglomerati Bituminosi con Plastica Riciclata nell'ambito dell'Economia Circolare

Dottorando
Vineesh Vijayan

Coordinatore
Prof. Giuseppe Campione

Relatore
Prof. Gaetano Di Mino

Co-Relatore
Prof. Evangelos Manthos
Dr. Ing. Konstantinos Mantalovas

Ciclo XXXVI
Anno Accademico 2022-2023

"Dream is not what you see in sleep, dream is the thing that doesn't let you sleep."

-Dr. A.P.J. Abdul Kalam

Acknowledgements

First and foremost, I am deeply grateful to my esteemed advisor, Prof. Gaetano Di Mino, for his unwavering support, encouragement, and guidance, which have been invaluable throughout my doctoral journey. The trust he bestowed upon me, and his continuous intellectual mentorship have significantly shaped my personal and professional development. I am greatly appreciative of the myriads of opportunities he has provided and his genuine commitment to nurturing my academic growth.

My co-advisor, Dr. Konstantinos Mantalovas, a postdoctoral researcher at the University of Palermo, deserves my sincere thanks for his ongoing support, insightful suggestions, and friendship throughout this Ph.D. journey. Further, I am indebted to Prof. Evangelos Manthos for offering me the invaluable opportunity to conduct a part of my research activities at the Aristotle University of Thessaloniki. His unwavering support and collaboration have been pivotal in acquiring invaluable and significant experience.

I wish to extend my heartfelt gratitude to Prof. Clara Celauro, Prof. Laura Inzerillo, and Prof. Davide Lo Presti for their invaluable support and advice during my time at the university. I acknowledge the Ph.D. committee members and the professors of the Department of Engineering of the University of Palermo for their support and the precious opportunities they have given me. I am also grateful to Prof. Claudio Lantieri from the University of Bologna for his unwavering support and motivation throughout my studies.

My gratitude extends to Iterchimica S.p.A. for supplying the research materials and for giving me the opportunity to conduct a part of the research activities at their laboratory. Their contribution has significantly enriched my research journey.

I extend my special thanks to Mr. Antonello Lorello, the technical officer of the road laboratory at the University of Palermo, for his support and patience during the experimental campaign. I am immensely grateful to all my colleagues who have accompanied me on this journey; their encouragement and support over the past few years have been invaluable. I also extend my thanks to my friends and the professors at the University of Bologna.

My utmost gratitude goes to my family for giving me the freedom to pursue my dreams and for the trust they have placed in me. I am delightfully inspired by you and proud to have become who I am now. Last, but certainly not least, I want to express my heartfelt thanks to Hanna Baby, whose genuine friendship, continuous encouragement, and unwavering support throughout this journey have been an enduring source of strength.

Abstract

The road engineering sector is increasingly embracing the principles of circular economy and sustainability, leading to the introduction of alternative materials in asphalt pavement. In this regard, the use of recycled plastic as a modifier in asphalt mixtures provides an eco-friendly and cost-effective alternative that improves pavement performance while addressing environmental concerns over plastic waste to a certain extent. While recycled plastic-modified asphalt pavements demonstrated enhanced performance, their recyclability at the end of their service life remains under-explored, which significantly influences the implementation of a circular economic approach. To address this issue, the current thesis aims to investigate the multi-recyclability of asphalt mixtures modified with recycled plastic over two life cycles. To achieve a controlled environment for all the desirable parameters while considering the novelty of the recycled plastic used, the reclaimed asphalt pavement (RAP) was produced using an artificial laboratory ageing protocol, which served as a basis for assessing multiple life cycles. To achieve the main goal of this thesis, a comprehensive experimental campaign was performed to evaluate the binder-scale and mixture-scale performance properties. Additionally, a novel analytical circular economic model was developed, along with a novel indicator, the Product Multi-Recyclability Index ($PMRI_n$). This way, it became possible to quantify the product circularity over multiple life cycles for the asphalt mixtures that not only contain recycled materials in an open-loop approach but also over multiple life cycles. The findings revealed that the asphalt mixtures modified with recycled plastic performed comparably, or in some cases, outperformed the conventional mixture over two cycles at a recycling rate of 50%. Overall, it can be concluded that the asphalt mixture containing the recycled plastic modifier can be recycled for multiple life cycles without compromising its mechanical and performance characteristics. The introduced $PMRI_n$ can be used as a metric to assess the potential recyclability of asphalt mixtures, thereby enabling a plethora of opportunities for the involved stakeholders and the relevant decision-makers towards the actual implementation of the circular economy in the field of bituminous materials.

Publications included in the current thesis

All the chapters of this thesis have been published as parts or slightly modified parts of the following international peer-reviewed publications:

- **"Evaluating the Ageing Sensitivity of the Asphalt Binder via Distinct Ageing Methods,"** *published in the international open access journal "Infrastructures" of MDPI under the terms and conditions of the Creative Commons Attribution (CC BY 4.0) license, has thus been reproduced here with the mission of the copyright holder [https://doi.org/10.3390/infrastructures8110163].*

- **"Investigating the Multi-Recyclability of Recycled Plastic-Modified Asphalt Mixtures,"** *published in the international open access journal "Infrastructures" of MDPI under the terms and conditions of the Creative Commons Attribution (CC BY 4.0) license, has thus been reproduced here with the mission of the copyright holder [https://doi.org/10.3390/infrastructures8050084].*

- **"A Top-Down Approach Based on the Circularity Potential to Increase the Use of Reclaimed Asphalt,"** *published in the international open access journal "Infrastructures" of MDPI under the terms and conditions of the Creative Commons Attribution (CC BY 4.0) license, has thus been reproduced here with the mission of the copyright holder [https://doi.org/10.3390/infrastructures8050083].*

- **"Multi-Recyclability of Asphalt Mixtures Modified with Recycled Plastic: Towards a Circular Economy,"** *which has been accepted for publication in the international open access journal "Results in Engineering" of Elsevier on July 5, 2024, under the terms and conditions of the Creative Commons Attribution (CC BY-NC-ND 4.0) license, has thus been reproduced here with the mission of the copyright holder.*

Contents

List of Figures

List of Tables

List of Abbreviations

AASHTO: American Association of State Highway and Transportation Officials

ABS: Acrylonitrile Butadiene Styrene

ATR: Attenuated Total Reflectance

BTSV: Binder Fast Characterization Test

CAI: Climatic Ageing Index

CE: Circular Economy

DSR: Dynamic Shear Rheometer

EAPA: European Asphalt Pavement Association

EBA: Ethylene-Butyl Acrylate

EEA: European Environmental Agency

EICM: Enhanced Integrated Climatic Model

EMF: Ellen MacArthur Foundation

EU: European Union

EVA: Ethylene-Vinyl Acetate

FHWA: Federal Highway Administration

FTIR: Fourier Transform Infrared

GHGs: Greenhouse gases

GMA: Glycidyl Methacrylate

GRP: Graphene-enhanced Recycled Plastic

HDPE: High-Density Polyethylene

HMA: Hot Mix Asphalt

ITFT: Indirect Tensile Fatigue Test

ITS: Indirect Tensile Strength

ITSM: Indirect Tensile Stiffness Modulus

ITSR: Indirect Tensile Strength Ratio

LCA: Life Cycle Assessment

LDPE: Low-Density Polyethylene

LLDPE: Low-Linear Density Polyethylene

LTA: Long-Term Ageing

LWARB: London's Waste and Recycling Board

MAH: Maleic Anhydride

NCHRP: National Cooperative Highway Research Program

PAV: Pressure-Ageing Vessel

PC: Polycarbonate

PE: Polyethylene

PET: Polyethylene Terephthalate

PMB: Polymer-Modified Bitumen

PMRI: Product Multi-Recyclability Index

PP: Polypropylene

PS: Polystyrene

PU: Polyurethane

PVC: Polyvinyl Chloride

RA: Reclaimed Asphalt

RAP: Reclaimed Asphalt Pavement

RTFO: Rolling Thin Film Oven

SARA: Saturate, Aromatic, Resin, and Asphaltene

SBR: Styrene-Butadiene-Rubber

SBS: Styrene-Butadiene-Styrene

SEBS: Styrene-Ethylene/Butylene-Styrene

SHRP: Strategic Highway Research Program

SIS: Styrene-Isoprene-Styrene

STA: Short-Term Ageing

US: United States

UV: Ultraviolet

VAPro: Viennese Ageing Procedure

WMA: Warm Mix Asphalt

WTS: Wheel Tracking Slope

Chapter 1
Introduction

1.1 Background

The road pavement infrastructure plays an integral role in a country's economic and social development. It improves geographical accessibility and enables the mobility of people, thereby creating job prospects and fostering wealth [1]. Asphalt pavements, in particular, play a substantial role in global road networks since they are the most widely used paving solution due to their multiple benefits, including cost-efficiency, noise reduction, durability, and a comfortable ride experience [2]. However, the production of asphalt mixtures significantly impacts the environment by exploiting natural resources, emitting greenhouse gases (GHGs), and generating waste. In addition to these environmental challenges, the increasing resource scarcity and the rising cost of crude oil have forced the road industry to focus on the implementation of sustainability and circular economy principles [3], [4]. Over 80 percent of Europe's road networks are constructed using asphalt concrete, which accounts for nearly 950 billion tonnes. The global use of bituminous binders for flexible pavements exceeds 100 million tonnes per year, resulting in a considerable exploitation of non-renewable resources and energy [5].

To address these environmental concerns, the European Commission has initiated the European Green Deal, a unified strategy aimed at fostering a climate-neutral, resource-efficient, and competitive economy. The implementation of this strategy within the road construction sector could substantially reduce GHGs emissions and consumption footprints by transitioning towards a more regenerative growth model [6]. However, the crucial challenge for road agencies is to deal with the waste generation during the management of existing pavements. In alignment with the European Green Deal, the road construction agencies are mandated to adopt more sustainable practices and find alternative materials for pavement construction. Asphalt recycling is considered an effective solution for embedding sustainability in the asphalt industry, which reuses or recycles valuable materials to cope with both environmental and economic challenges. The use of reclaimed asphalt pavement (RAP) has been proven to be a promising solution in asphalt production that allows for minimising the consumption of non-renewable resources, waste generation, environmental pollution, and the cost of production [7], [8]. Nevertheless, the amount of RAP incorporation in new asphalt mixtures is still limited due to several reasons, such as technical issues related to production, performance properties, and durability. The issues are mainly related to the presence of aged binder in the RAP, which increases the stiffness of the resulting asphalt mixture and increases the cracking susceptibility of the pavement. Many studies have reported that the use of rejuvenators can significantly increase the reusing/recycling of RAP, with performance properties similar to or even superior to those of the reference asphalt mixture without RAP [9], [10].

Besides, unpredictable weather conditions and increased traffic volume have influenced the service life and performance of asphalt pavements, leading to pavement deterioration such as rutting, fatigue cracking, and thermal cracking [11]. Consequently, polymer modification has been adopted as a feasible approach to address these premature pavement distresses since the 1980s [12]. The polymer incorporated in the asphalt modification can be categorised mainly into plastomers and elastomers. The plastomers such as polyethylene (PE), polypropylene (PP), and ethylene-vinyl acetate (EVA) and the elastomers such as styrene-butadiene-styrene (SBS) and styrene-butadiene-rubber (SBR) are the most used polymers. Depending on the type and mode of use, the use of polymers could enhance either the rheological properties of asphalt binder or the mechanical and performance properties of the asphalt mixtures [13]. There are two methods to incorporate polymers in asphalt mixtures: the wet method and the dry method. In the wet method, polymers are introduced into the bitumen and blended using a mechanical mixer, producing polymer-modified bitumen (PMB). On the other hand, in the dry method, the polymer is directly added to the hot aggregates before mixing it with the bitumen during the mixture production. Depending on the type of polymer, in this case, the polymer could be considered an alternative to aggregate, filler, or an asphalt binder modifier, improving its properties [14].

The growing demand for sustainability in asphalt pavement construction has prompted the exploration of cost-effective and eco-friendly alternatives that embrace circular economy principles [15], [16]. Meanwhile, the pervasive issue of plastic waste poses a significant threat to the environment due to its massive volume, which is capable of inflicting harm on ecosystems and their inhabitants. Annually, approximately 300 million tonnes of plastic waste are generated worldwide due to its widespread use in sectors like automotive, manufacturing, packaging, and healthcare. Furthermore, the inherently low biodegradability of plastic poses considerable challenges to recycling and disposal [17], [18]. To overcome this challenge, it is essential to explore innovative applications that effectively repurpose plastic waste, providing sustainable solutions for its management. Consequently, asphalt practitioners and researchers have adapted their emphasis towards incorporating waste plastic into asphalt concrete, thereby addressing the issue of plastic waste to some extent while also enhancing the performance of asphalt concrete via plastic modification. The literature shows that the incorporation of some of the recycled plastics could enable promising results as modifiers for asphalt binders or mixtures [19]. It has been reported that the addition of some of the recycled plastic modifiers improves the performance-related properties of asphalt mixtures, such as resistance to moisture and rutting [20]. In this regard, a study conducted by Poulikakos et al. reported that the dry addition of 1.5% polyethylene waste (by weight) in the asphalt mixture showed improved resistance to permanent deformation, higher elastic behaviour, a low creep rate, and a higher creep modulus. However, the increase in the percentage of PE to 5% exhibited an increase in permanent deformation [21]. The increased stiffness and resistance to permanent deformation have been studied both for the mixtures modified via wet and dry methods. D'Angelo et al. studied the performance of asphalt mixtures modified using the wet method with two types of recycled plastics [22]. The outcome of their research demonstrated that the plastic-modified binder blends performed better than the neat bitumen at high temperatures during the rheological analysis, which shows improved rutting resistance. They also concluded that the modified blends exhibited comparable results in terms of bond strength.

While the mechanical and performance properties of asphalt mixtures modified with recycled plastics are very clear at medium and high temperatures, the properties at low temperatures and fatigue are less investigated. A few of the available studies on fatigue and low-temperature properties demonstrated that the asphalt mixture modified by recycled plastic has similar or slightly superior performance compared to control asphalt mixtures [23]. Looking from the Life Cycle Assessment (LCA) perspective, the asphalt mixture containing the recycled plastic modifier at the end of the life cycle was also a matter being investigated. In this regard, a study conducted by Lu et al. concluded that the investigated recycled plastic-modified asphalt mixtures are recyclable and provide similar properties compared with the reference (unmodified) RAP. According to their findings, incorporating 30% RAP-containing recycled plastic in new asphalt mixtures did not have any adverse effects on their cracking and rutting performances [24].

However, despite the scientifically proven benefits of asphalt recycling and the implementation of various best practices, further study is required in specific and specialised areas. One such scenario involves investigating whether plastic-modified asphalt pavement can be recycled at the end of its service life, similar to RAP from conventional asphalt pavement. While the technical performance properties of recycled plastic-modified asphalt binders and mixtures have been researched and presented in several studies, there are still some aspects that have received less attention. Considering the increasing trend towards high RAP reusing and recycling, along with the importance of life cycle assessment, a clear understanding of RAP-containing recycled plastic modifiers is inevitable. The failure to achieve this could undermine the initial performance benefits obtained from incorporating waste plastic into asphalt concrete. Currently, research on the recyclability of plastic-modified asphalt concrete is limited by the absence or unavailability of RAP from such plastic-modified pavement sources with 10 or more years of field service. Further studies are imperative to ascertain the recyclability of plastic-modified asphalt mixtures and to devise effective recycling techniques. This is crucial for ensuring circularity and sustainability when employing these innovative alternative materials in pavement construction.

1.2 Scope and Objectives of the Thesis

The scope of this thesis is to scientifically investigate the feasibility of recycling asphalt mixtures modified with recycled plastic, evaluate their mechanical performance throughout multiple life cycles, and establish guidelines for their processing and application. This approach intends to contribute to a better understanding of characterising plastic-modified asphalt mixtures at the end of their service life, potentially promoting the incorporation of waste plastic in asphalt modification. Ultimately, this could improve the circularity of materials through innovation and optimised recycling. Considering these crucial dimensions, the research aims to achieve the following key objectives:

- To introduce the concept of circular economy and a synthesis of existing literature on asphalt modification using waste plastics, as well as asphalt recycling.
- To develop and validate a laboratory protocol to simulate the ageing behaviour and end-of-life scenarios for asphalt mixtures for multiple life cycles.
- To explore methodologies for developing a cohesive rejuvenator blend design for multi-recycling asphalt mixtures with and without a recycled plastic-modifier.
- To assess the impact of ageing and rejuvenation on the repeated recycling of asphalt mixtures via physical, chemical, and rheological characterization.
- To investigate the multi-recyclability potential of asphalt mixtures, both with and without a recycled plastic modifier, using a comprehensive evaluation of mechanical and performance characteristics.

- To develop and validate a circular economic model that quantifies the material circularity of distinct asphalt mixtures through multiple life cycles.

These objectives aim to provide a scientific background on the multi-recyclability of plastic-modified asphalt, addressing crucial sustainability concerns and proposing feasible solutions for enhanced performance within the framework of a circular economy.

1.3 Significance of this research

The study contributes to understanding the potential recyclability of asphalt mixtures containing recycled plastic, which is crucial for promoting circular economy practices in the road engineering sector. This approach creates confidence and opportunities for stakeholders and decision-makers to implement closed and open-loop recycling strategies, integrating more waste alternatives into the road industry. Consequently, this reduces the use of virgin materials, their costs, and energy consumption while allowing products to remain in the market for an extended period without losing their value or performance.

1.4 Structure of the Thesis

This thesis is structured in eight chapters, as follows:

Chapter 1: Introduction, presents the background and context, identifies the problem statement and the research gaps, defines the scope and objectives of this thesis.

Chapter 2: Literature review provides a theoretical background and elaborates the state-of-the-art knowledge on the concept of circular economy, asphalt modification via waste plastics, asphalt recycling and their challenges.

Chapter 3: Research Methodology provides an overview of the adopted procedures for preparing and testing the mechanical and performance characteristics of asphalt binders and mixtures.

Chapter 4: Simulation of ageing behaviour in asphalt mixtures, focuses on the simulating the ageing behaviour and end-of-life scenario of asphalt mixtures using different laboratory ageing protocols.

Chapter 5: Multi-recyclability study: From a binder-scale perspective, aims to investigate the multi-recyclability of asphalt mixtures, with and without the recycled plastic via physical, chemical, and rheological characterization.

Chapter 6: Multi-recyclability study: From a mixture-scale perspective, focuses on investigating the multi-recyclability of asphalt mixtures, with and without the recycled plastic using comprehensive volumetric and mechanical performance evaluation.

Chapter 7: A proposed model to evaluate the multi-recyclability potential of asphalt mixtures under the scope of circular economy, which presents a novel methodology for quantifying the material circularity of various asphalt mixtures across multiple life cycles.

Chapter 8: Conclusions and Future perspectives, reports the summary and key findings of this thesis along with few recommendations for future researchers.

Chapter 2
Literature Review

2.1 Circular Economy and Asphalt pavements

2.1.1 Concept of Circular Economy

The world is shifting towards a circular economy (CE) due to its regenerative growth model, which focuses on solving global environmental, climatic, and economic challenges. The aim of the CE is to maintain a resource loop and to retain the resources in the system for as long as possible at the highest utility level [3]. In light of its future aspects, the European Union (EU) and other policymakers are promoting circular strategies, which accelerate the transition from the linear economic model to the circular economic model to guarantee resource availability and put an end to waste formation through design optimisation and recycling [25]. According to the new circular economy action plan released by the European Commission, the transition to a CE contributes to a future-oriented scheme for achieving a cleaner and more competitive Europe in co-creation with economic actors, consumers, citizens, and civil society organizations [6]. Moreover, it has been acquiring traction with businesses and policymakers, with remarkable opportunities recognised for both groups of stakeholders. Even though it is not easy to construct such an economic system, it requires utilising associated technologies, restructuring products, creating new business models, and creating changes in consumer behaviour to develop considerable customer value [26].

The notion of CE is not new, but it has gained increased attention in recent years due to its competitive approach to eliminating the 'end-of-life' concept. The concept of 'Circular Economy' made its first appearance in the late 1970s, mainly due to the concern raised by Carson and Boulding about climate change and the increasing price of resources [3], [27]. The Club of Rome's 'Limits to Growth' theory, Braungart and McDonough's cradle-to-cradle concept, Stahel's 'performance economy', and Lyle's 'regenerative design' model were some of the few that established the deep roots of CE concepts [28]. During this period, Europe and the United States (US) have formed policies to control environmental impacts due to the increase in waste disposal, which has gained much attention globally to the 3R concept of reduce, reuse, and recycle [29]. Ellen MacArthur Foundation (EMF) effectively demonstrated the framework of CE for systems-level redesign, which offers creativity, innovation, and a restorative economy. Later, their definition of circular economy was widely accepted, and it states that CE is an, "Economy that is restorative and regenerative by design and which aims to keep products, components, and materials at their highest utility and value at all times, distinguishing between technical and biological cycles [30]." Over decades, the global economic sector has been dominated by a linear economic model, which follows a 'take-make-use-dispose' pattern in that products are manufactured by using extracted raw materials, then sold to consumers, and after that, consumers discard them as waste when they no longer serve their purpose [31]. On the contrary, CE follows a cradle-to-cradle approach that focuses on the quality of products, where materials are circulating in a way that guarantees safety and health for both humans and the environment [26].

The notion of the circular economy offers various methods to create value without relying on the consumption of limited resources [32]. As shown in Figure 2.1, the material flow can be divided into two resource loops: technical and biological resource cycles. In a true circular economy, consumption should only take place within efficient biological cycles, and usage takes the place of consumption in other areas [31]. The resources are either regenerated in the biological cycle through natural processes or recovered and restored in the technical cycle using circular economy approaches and business models that focus on maximising the value derived from finite stocks of technical assets and materials [33]. To be more precise, within the biological cycle, the highest priority is given to products that are renewable and bio-based, which can be regenerated and reintegrated into the environment through composting or anaerobic digestion [28]. Hence, the bioeconomy is considered a promising sector that ensures reductions in resource exploitation, energy consumption, and waste generation. On the other hand, considering the technical cycle, the products can be designed in such a way that when they reach at the end of their service life and cannot be repaired or reused for their original purpose, their components can be extracted, repurposed, or refurbished into new products [3]. This approach ensures the value of the product is retained, prevents waste from being sent to landfills, and establishes a closed-loop system. Thus, the focus on the circular economy is essential, as it can create various business and economic opportunities while also producing positive environmental and social impacts, establishing a beneficial development cycle [25].

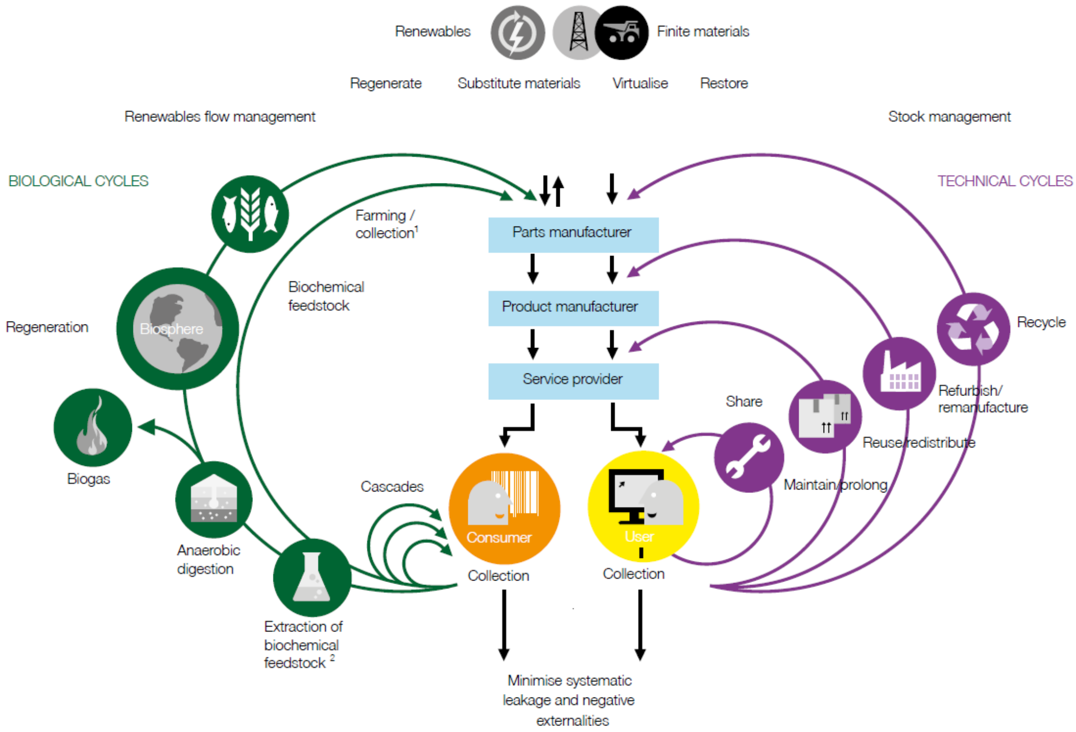

Figure 2. 1 Outline of a circular economy (ARUP, 2016) [28], [31].

Basically, the idea of circular economy relies on three principles: foster system effectiveness, optimise resource yields, and regenerate natural systems [34]. Fostering system effectiveness means revealing and designing out the negative externalities of economic activity that can cause damage to human health and the ecosystem by avoiding toxic substances, greenhouse gas emissions, and other environmental pollution. Optimising resource yields means customising the entire design for re-use, remanufacturing, and recycling of components and materials to retain their circularity within the system or cascading to an external system. Such an economic system maximises the use of bio-based materials to cascade them in various applications. Finally, regenerating natural systems intensifies the natural capital by maintaining nutrient flow within the system for regeneration [15]. The study conducted by EMF provides new evidence that CE, facilitated by the technological revolution, helps Europe to improve its resource productivity by up to 3 percent annually. It will generate a primary resource benefit for the European economy of more than 0.6 trillion euros per year by 2030. Moreover, it has the capability to generate 1.2 trillion euros through non-resource and external benefits, bringing the annual total benefits to approximately 1.8 trillion euros compared with the current situation [33], [35]. Later, based on case studies and interviews with several experts, a 'ReSOLVE' framework was formulated to help governments and businesses to adapt circular strategies and growth initiatives. The framework consists of six actions [36] and is briefed below:

- **Regenerate:** It helps to shift to renewable energy resources and aims to reclaim, retain, and restore the health of the ecosystem.
- **Share:** It means to increase the utilisation of products by sharing, designing to reuse them during their technical lifetime, and maintaining their life through proper maintenance, repair, and design for better durability.
- **Optimise:** It maximises the performance of the product, removing waste in production and leveraging big data, automation, remote sensing, and steering.
- **Loop:** The components and the materials need to be in closed loops and prioritized inner loops, which means remanufacturing products or components by recycling materials in the case of finite materials and extracting bio-chemicals from organic waste.
- **Virtualize:** The utilities need to be delivered virtually, for instance, books or music, online shopping, and so on.
- **Exchange:** The application of new technologies, replacing old materials with advanced non-renewable materials, and choosing new services or products are the scope of this action.

Furthermore, the report published by the European Environmental Agency (EEA) suggests that achieving a higher level of circularity will necessitate significant changes in all aspects of material use, including the practices related to how products are produced and consumed [37]. Thus, a circular economy utilises reuse, refurbishment, remanufacturing, and recycling to establish closed-loop systems by integrating circular principles at the design stage. This approach minimises resource inputs and reduces waste, pollution, and carbon emissions. Hence, within this system, 'waste' from one process serves as the input for another [37].

2.1.2 Circular Economy and its benefits

The circular economy promotes alternative energy and material flow strategies within the economic framework, aiming to reduce reliance on non-renewable resources by extending the service life of each material, service, and product [25]. This approach also aims to minimise waste generation by returning materials back into the product cycle at the end of their service life through recycling [38]. The successful implementation of CE can stimulate economic growth and environmental prosperity while creating new job opportunities in sectors such as recycling and remanufacturing, contributing to societal well-being alongside its economic and environmental benefits. The report on ‘Delivering the Circular Economy: A Toolkit for Policymakers’ published by EMF clearly demonstrated the potential of a circular economy, which can bring the long-lasting benefit of a more innovative, resilient, and productive economy [31]. In accordance with their report, the key benefits of CE are as follows:

- **Substantial net material savings and reduced exposure to price volatility:** According to the study, the EMF estimated that the adoption of a CE within medium-lived product industries could generate total annual net material cost savings of up to $630 billion at the EU level and $700 billion globally.
- **Increased innovation and job creation potential:** The initial evidence shows that the powerful circularity frame can create solutions and innovation. Further exploration of the circular industrial model and labour markets results in more positive impacts. The desire to replace one-way products with goods that are ‘circular by design’ and the potential to develop reverse logistics networks and other systems to aid the circular economy is an impactful motive for innovations.
- **Increased resilience of systems and economies**: It helps in higher land productivity, less waste generation, and reinstating nutrients to the soil by enhancing the value of land and soil. Moreover, the use of more biological material in each sector reduces the need for replenishment with additional nutrients and retains biodiversity.

2.1.3 Importance of Circular Economy in road construction sector

The road construction industry is a major contributor to rising environmental issues such as global warming, resource depletion, and pollution. It consumes a considerable amount of non-renewable resources and energy for the construction and maintenance of road infrastructure, resulting in a significant carbon footprint. Consequently, the heavy reliance on non-renewable resources has led to resource scarcity and a substantial rise in material costs [39]. However, the adoption of circular economy principles in the road engineering sector can provide a solution to these challenges [40]. By embracing these principles, the road industry can significantly reduce its dependence on natural resources through reusing and recycling materials. This approach, as shown in Figure 2.2, can limit resource depletion and environmental pollution to a certain extent while also reducing production costs and fostering benefits for both construction firms and government entities [16], [41]. For instance, the use of reclaimed asphalt pavement (RAP) in new asphalt mixtures has reduced the demand for crude oil and virgin aggregates, thereby reducing the environmental impact associated with raw material extraction while cutting down on related extraction and transportation costs [42].

Figure 2. 2 Circular Economy of asphalt pavement (EAPA, 2022) [40].

The concept of a circular economy has created new opportunities for the road construction industry to incorporate recycled materials, industrial by-products, and other discarded materials from an open-loop system into asphalt production. This potential collaboration with different industries or businesses through a collaborative approach could be more beneficial to various stakeholders. For instance, utilising waste engine oil or waste cooking oil as a recycling agent for incorporating a high amount of RAP in asphalt mixtures substantially reduces the cost of purchasing commercial recycling agents while minimising waste generation [43], [44]. Such application enables the transformation of waste from one system into a valuable resource for another system, in this case, the road construction industry. Furthermore, waste plastics are being incorporated into asphalt mixtures as a modifier to enhance the mechanical and performance properties of the pavement [45], [46]. In this way, a crucial environmental concern related to waste plastic is being addressed through repurposing and resource optimization. Also, in this manner, the expensive polymers are replaced with recycled plastics, which not only improves the resourcefulness of the product over time but also greatly cuts down on costs related to polymer modification.

2.2 Asphalt recycling

2.2.1 Reclaimed asphalt pavement: General overview

The recycling of asphalt pavement is a sustainable approach to reusing valuable material to cope with resource scarcity and the rising price of asphalt binder. The use of reclaimed asphalt pavement, also known as reclaimed asphalt (RA) in Europe, provides a promising alternative to asphalt production by reducing the exploitation of natural resources, waste generation, environmental pollution, and production costs [42], [47]. In general, RAP refers to removed and/or reprocessed asphalt pavement containing bitumen and aggregates or asphalt material deriving from reject and surplus production [48]. Asphalt pavements are typically removed for resurfacing or reconstruction when they exhibit severe distress or at the end of their service period to enhance the comfort of the road user. The conscious use of such material in the production of asphalt ensures the sustainability of pavement construction. Furthermore, asphalt pavement typically consists of about 93–95% aggregates by weight and 5–7% bitumen by weight, which can be reused in new asphalt mixtures to promote the circularity of valuable materials over the long term rather than being used for landfilling or downgrading applications [49]. Nevertheless, the regulations and specifications of several road agencies have limited its use in surface applications due to concerns that the high content of RAP may not perform as well as traditional mixtures [42]. However, incorporating RAP into the surface and intermediate layers of asphalt pavement is considered a cost-effective approach, as the binder present in the RAP can replace a portion of the expensive virgin binder. Zaumanis et al. [50] have demonstrated that asphalt mixtures containing 100% RAP can be utilised in asphalt production, and such mixtures can perform similarly or even better than traditional hot mix asphalt.

The recycling of asphalt pavements has a long history, with its primary application being recorded in 1915. During the 1970s, the cost of crude oil increased dramatically due to the Arab oil embargo [51]. At that time, most of the road agencies and paving contractors faced resource scarcity and huge budget constraints for producing asphalt pavements. Basically, asphalt production costs comprise four major categories: material costs, asphalt production plant costs, transportation costs, and laydown costs [52]. Consequently, 70 percent of the total asphalt production cost is conferred by the cost of virgin materials. The price volatility of bitumen and aggregates compelled the road agencies to reclaim the valuable asphalt pavement materials at the end of their service life and use them in hot mix asphalt (HMA) production [53]. This shift eventually helped the asphalt industry to adopt a more feasible approach with multiple benefits for the economy, environment, and society. In the late 1990s, the implementation of the Superpave mix design by the Strategic Highway Research Program (SHRP) limited the use of RAP due to a lack of specific provisions for its integration. However, later on, the Federal Highway Administration's (FHWA) Superpave mixtures expert task group developed guidelines for incorporating RAP into the Superpave mix design method [47], [54]. Their major findings revealed that the performance of hot mix asphalt (HMA) depends on the incorporated RAP content. A low RAP content has minimal effect on its performance as it acts like a "black rock" and mainly influences the volumetric properties. On the other hand, an increase in RAP content affects HMA performance as it breaks down this black rock phenomenon and requires evaluation of the final blend properties when aged binder blends with virgin binder [55]. Consequently, the use of higher percentages of RAP (more than 25%) in HMA was limited due to the lack of guidelines for RAP processing, mix design, and best practices for RAP handling.

Besides, most research has focused on increasing the amount of RAP in recycled mixtures without compromising their mechanical performance compared to conventional mixtures. The most common methods of asphalt recycling include hot recycling in an asphalt plant, hot-in-place recycling, cold-in-place recycling, and full-depth reclamation. Globally, the commonly used rate of RAP incorporation is typically in the range of 10 to 30% in asphalt mixtures, as it performs similarly to traditional mixtures [56]. However, various studies have shown that the use of recycling agents like rejuvenators or softeners can potentially increase the rate of RAP in HMA [57]. Meanwhile, many factors, such as legislative limitations, recycling methods, available asphalt plant production technology, variability of RAP materials, mix design, type of recycling agents, and mixing time, still restrict the use of high RAP content in asphalt recycling in most countries [58]. The prime issue regarding the high RAP incorporation is the presence of aged RAP binder, which is very stiff and can result in cracking failures for asphalt mixtures. The stiffness of the aged binder influences the workability and compactability of mixtures containing a high RAP content [59]. Therefore, it is crucial to select the type and dosage of recycling agent during the mix design procedure. The improper type or dosage of recycling agents results in inferior performance in recycled mixtures [60].

2.2.2 State of Practice of RAP in Europe

Hot mix asphalt is widely used as a paving material, but it deteriorates over time due to climate and repeated loading cycles [61]. It needs to be removed from the pavement through milling or full-depth removal. The recycling of such pavements is considered a structural rehabilitation technique that reduces the use of virgin materials, construction costs, and environmental impacts [62]. Additionally, global consumption of natural resources like biomass, fossil fuels, metals, and minerals are expected to double in the next 40 years. This increase in raw material consumption will lead to a predicted 70% rise in annual waste generation globally by 2050 [6]. The 2022 annual report from the European Asphalt Pavement Association (EAPA) reveals that a total of 280 million tonnes of hot mix asphalt (HMA) and warm mix asphalt (WMA) were produced in European countries, as shown in Figure 2.3 [63]. Over the past five years, there has been consistent and significant usage of HMA/WMA in the surface layers of asphalt pavement by European countries. It is important to note that these surface layers are the costliest due to their high-quality aggregates and expensive binders. Figure 2.4 demonstrates the trend in HMA/WMA application across various flexible pavement layers from 2018 to 2022, highlighting consistently high demand for producing asphalt mixes for wearing courses, accounting for approximately 59% of total HMA/WMA production each year within Europe.

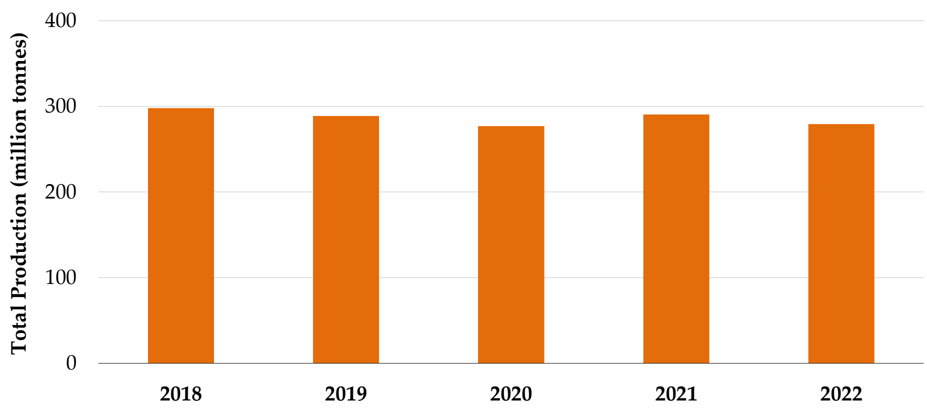

Figure 2. 3 Total production of HMA and WMA (EAP, 2024) [63].

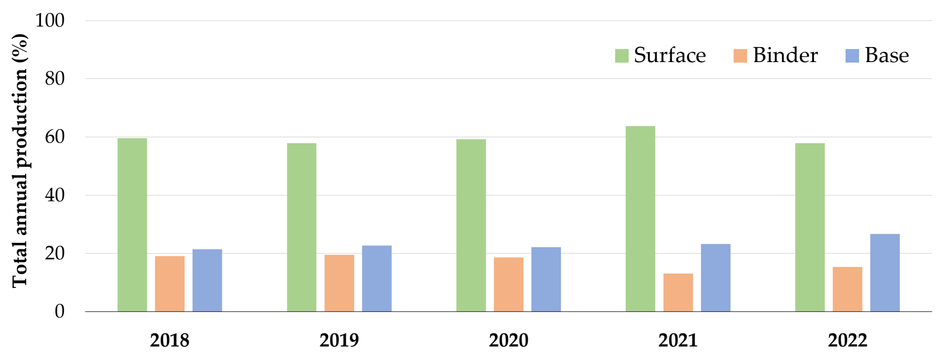

Figure 2. 4 HMA/WMA application in various layers of asphalt pavement in Europe (EAPA 2020-2024) [63]–[66].

Figure 2.5 shows the availability of RAP and its usage in the new production of HMA/WMA from 2018 to 2022. In Europe, the availability of RAP was nearly 31.4 million tonnes in 2022, with only 15.3 million tonnes used in HMA/WMA applications. The remaining quantity of RAP has been either used in unbound layers or landfill applications. The improper practices and degraded application of RAP usage in Europe have wasted approximately 55% of available RAP over the past five years, despite waste management being a key goal for the European Union's sustainable future [6]. A strategic approach to enhancing resource value and reducing the environmental impact caused by road construction is critical to maximising the utilisation of RAP in pavement construction. Recycling asphalt material becomes unsustainable when it is degraded and used in lower-value applications like unbound base layers or road shoulders [53].

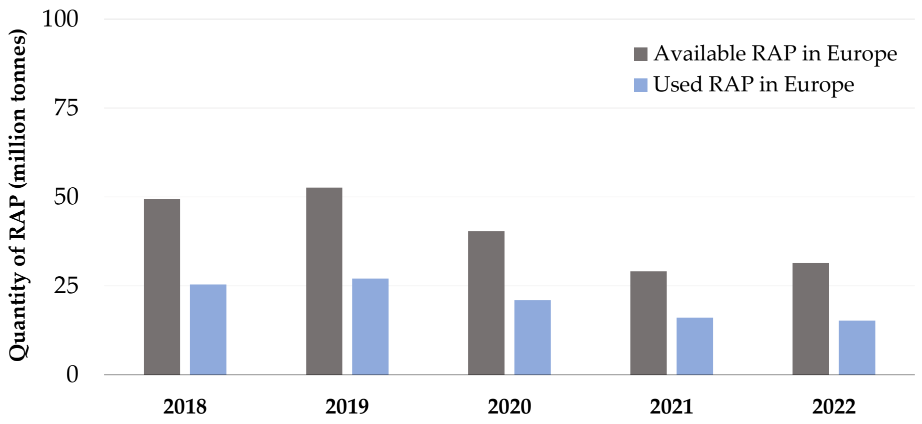

Figure 2. 5 Availability of RAP and its usage in asphalt production (EAPA 2019-2023) [63]–[66].

Furthermore, the complete advantages of RAP materials can be achieved only if they are utilised in the surface layers or intermediate layers, as those are the expensive layers in the pavement structure. The legislative norms imposed by various countries limit the incorporation of high RAP content in asphalt mixtures due to their uncertainty in mechanical performance in the surface course and intermediate course [67]. According to the data provided by EAPA, countries such as Austria, Denmark, Finland, France, Germany, Ireland, and Spain consider more than 70% of their available RAP to produce hot and warm mix asphalt. In that, Finland and Ireland are utilising 100% of RAP in their asphalt production, which depicts the exact potential of RAP in regenerating the road sector with zero waste [63]. A considerable number of studies have shown a positive impact on the performance of asphalt pavements containing RAP of up to 50% under different climatic conditions. Besides, the selection of recycling techniques plays a crucial role in the implementation of quality recycled mixtures with high RAP content [68].

2.2.3 Environmental and Economic benefits

The environmental and economic benefits of using reclaimed asphalt in new asphalt mixtures have been recognised for decades. Maximising the utilisation of RAP in pavement construction is a crucial strategy to enhance the value of resources and reduce the environmental impact caused by the road construction sector [69]. Furthermore, integrating reclaimed asphalt into new mixtures can lead to significant environmental advantages, such as reduced greenhouse gas emissions and reduced fuel consumption for extracting, processing, and transporting virgin materials [49]. The study conducted by Zaumanis et al. has demonstrated that a 100% RAP mixture results in an 18 kg or 35% reduction in CO2 equivalent emissions as well as saving 20% energy per tonne of paved mixtures produced with 100% RAP compared to conventional mixes containing only virgin materials [53].

The life cycle assessment study by Aurangzeb et al. on asphalt mixtures containing high RAP found a significant reduction of up to 28% in both greenhouse gas emissions and energy consumption. In addition, the study recorded a 40% reduction in energy requirement for the mixture containing 50% RAP compared to conventional mixtures, showcasing the efficient use of rejuvenators in recycled hot mix asphalt that reduces mixing temperature without compromising workability [70]. The advances in asphalt plant technology have enabled the industry to incorporate a high rate of RAP in asphalt mixtures, resulting in a remarkable reduction in emissions. Notably, improvements in drum mixer technology have minimised the direct interaction of RAP with flames and enhanced the production rate of recycled mixes when compared to virgin mixtures [71].

The road construction sector started incorporating RAP into asphalt mixtures to reduce production costs and address environmental concerns. Furthermore, the use of RAP reduces the need for virgin aggregates and bitumen, thus lowering the initial cost of road construction and enabling road agencies to manage maintenance within their budgets. In 2019, a survey conducted by NAPA reported that using RAP in asphalt mixtures saved a significant 3.29 billion dollars compared to using virgin materials [72]. The survey clearly showed the economic benefits of recycling by comparing the savings in production costs with previous years. The greatest advantage of RAP as a material is that it can be fully utilised when it is used in the wearing course or binder course layers [73]. Typically, binders used in surface courses or binder courses are expensive. However, if RAP is incorporated into such layers, it reduces binder costs by replacing them with less expensive RAP binders. The cost of extracting and transporting virgin aggregates is another important factor that affects the total production cost of asphalt pavement [73]. A study reported on material-related costs in the production of asphalt mixtures ranging from 0 to 100% RAP content revealed that mixtures containing 100% RAP reduced material costs by around 50–70% per tonne compared to virgin asphalt mixtures [53], [74]. As mentioned earlier, one effective solution for addressing rising bitumen costs and diminishing availability of natural minerals is integrating high proportions of RAP into asphalt mixtures using efficient technologies.

2.2.4 Mix design and performance of high RAP mixtures

The mix design method for HMA with a high content of RAP aims to achieve similar performance characteristics as traditional asphalt mix designs, particularly in terms of optimum binder content [67]. Indeed, incorporating a high RAP content in the new asphalt mixture requires modifying the mix design methodology to account for the material's gradation, mixture volumetrics, and aged binder properties [57]. Moreover, for the implementation of a quality recycled mix design, it is important to have information about the source of RAP. Using RAP from various stockpiles is not recommended because of its variability in terms of aged binder content and aggregate properties [75]. The mix designer must determine the binder content and its properties in order to calculate the total binder content in the final asphalt mixture. Therefore, extracting aged binder from RAP is an essential process for evaluating its properties, as it significantly influences both rheological properties and the total binder content of the final blend of asphalt mixture [49].

According to a report published by the National Cooperative Highway Research Program (NCHRP), certain tests need to be conducted on binder and aggregate for designing HMA with RAP [54]. The properties of RAP that need to be determined include RAP binder content, RA aggregate gradation, specific gravity of RAP aggregate, angularity of the coarse aggregate particles in the RAP, flat and elongated particles for the coarse aggregate in the RAP, and angularity of the fine aggregate in the RAP [76]. The ignition method is more accurate than the solvent extraction method for determining binder content in RAP. However, both methods are suitable for finding out specific gravity, aggregate gradation, and other consensus properties.

When the content of RAP in the new mix design is equal to or less than 15% of the total weight of the mixture, there is no need to evaluate the aged binder properties. However, if more than 15% of RAP is used in the recycled mixture, the aged binder affects the binder grade and must be evaluated using blending charts [77]. The addition of a high rate of RAP in an asphalt mixture depends on various factors, such as the available quantity of RAP, hot mix plant capability, specification limits in that country, the gradation and variability of RAP, and the properties of the aged binder [78].

The performance of HMA is influenced by the composition of the mixture and its density, while performance-based mix design strictly relies on binder properties, aggregate properties, and volumetric mixture properties [54]. The performance of asphalt mixtures with RAP generally depends on the percentage of RAP in the total mixture. According to the literature, an asphalt mixture containing less than 15% RAP exhibits similar properties compared to traditional asphalt mixtures. However, a high RAP content in recycled mixtures has shown increased stiffness properties [79] and decreased fatigue compared to asphalt mixtures without RAP [80]. The most important characteristic of a recycled asphalt mixture is the stiffness of its resulting binder blend, which typically tends to be stiffer than virgin binder blends [55]. The reason for this is the ageing of RAP binder, which results from chemical changes due to high production temperatures and oxidation during service. Therefore, when designing recycled HMA, it is important to consider the effects of ageing on its binder properties in order to achieve a mix with good performance in terms of rutting, fatigue, thermal resistance, and durability [81].

Numerous studies have shown that high-RAP mixtures exhibit good resistance to rutting compared to conventional asphalt mixes [82]. The presence of aged binder in RAP increases the stiffness of the final binder blend, resulting in improved rutting resistance. However, it's crucial to carefully control stiffness, as it directly impacts the overall performance of the mixture. To mitigate the effects of ageing, rejuvenating agents have been introduced to restore and enhance the lost properties of the RAP binder [83]. Zhang J. et al. conducted a study on the mechanical performance of recycled asphalt mixtures with two self-developed rejuvenators having different viscosities [84]. Their results indicated improvements in cracking behaviour, fatigue life, resistance to permanent deformation, and moisture durability. On the contrary, West R. et al. reported that high RAP contents in the mixture have shown low fatigue resistance but better moisture resistance than conventional ones [56]. Essentially, the aged binder exhibits high stiffness, leading to stiffer mixtures, which result in fatigue cracking and low-temperature brittleness [85].

Furthermore, studies have demonstrated that mixtures containing up to 40% RAP exhibit improved fatigue life compared to virgin asphalt mixtures [86]. Suo Z. et al. conducted research on rejuvenating aged bitumen with vegetable oil, studying its rheological properties, high temperature performance, and fatigue resistance of the restored binder [86]. The findings of their study validated an enhancement in fatigue resistance and low-temperature cracking resistance. Zaumanis et al. [9] investigated the effect of six rejuvenators on a 100% recycled asphalt mixture to evaluate their performance properties and concluded that all mixtures with different rejuvenators at their optimal dosage ensured excellent rutting performance and extended fatigue life compared to virgin mixtures, although they required higher compaction energy. Moreover, other studies have shown that asphalt mixtures containing 50% RAP content improve dynamic stability and resistance to rutting [82], [87].

2.2.5 High RAP incorporation: Challenges and limitations

The use of a high rate of reclaimed asphalt is limited by the available asphalt plant production technology. Conventionally, superheated virgin materials indirectly heat the RAP without exposing them to direct flames, imposing limitations on incorporating a high amount of RA in asphalt mixtures [88]. The normal continuous drum mixer can accommodate RAP up to a rate of 50% when compared to the batch plant. Hence, producing a high RAP mixture using conventional asphalt plants demands an unrealistically high superheating temperature for the virgin materials, leading to high volatilization of the RAP binder and emitting blue smoke [67]. In most countries, the use of RAP on asphalt pavements is typically restricted to 20–30%. Some previous studies have indicated that asphalt mixes with a high RAP content often show poor resistance to fatigue and thermal cracking [69]. Moreover, another issue is the unpredictable blending and diffusion between aged RAP binder and virgin binder. The high rate of RAP incorporation is mainly limited by the specifications or regulations of road authorities in different countries due to uncertainties about the durability and mechanical performance of recycled mixtures [57].

2.3 Asphalt pavement modification

2.3.1 General overview

Asphalt binders are viscoelastic materials that have a crucial role in asphalt pavement as they act as an adhesive, which binds the aggregates together. However, they are significantly influenced by several factors, including service temperature, climatic conditions, increased traffic load, and volume, which can lead to deterioration of the pavement such as rutting, fatigue cracking, and thermal cracking [89]. Consequently, polymer modification has been widely adopted as a feasible approach to address these premature pavement distresses. It involves the incorporation of various polymers into asphalt binders or mixtures to enhance its properties, such as durability, elasticity, resistance to ageing and deformation, thus contributing to the overall quality and extended service life of the pavement structure [90], [91]. To date, several polymers have been used to enhance the properties of asphalt concrete based on specific needs or target performance requirements. These polymers are typically classified into three categories: elastomers, plastomers, and reactive polymers. Elastomers such as styrene-butadiene-styrene (SBS), styrene-isoprene-styrene (SIS), styrene-butadiene-rubber (SBR), and styrene-ethylene/butylene-styrene (SEBS) have been widely used to improve the elastic properties of asphalt concrete, thereby resisting deformation, and ensuring quick recovery when the stress is removed [92]. Plastomers such as polyethylene (PE), ethylene-vinyl acetate (EVA), polypropylene (PP), ethylene-butyl acrylate (EBA), and polyolefins have been employed to enhance the high-temperature performance of asphalt concretes to resist permanent deformation [93]. Furthermore, reactive polymers such as random terpolymers of ethylene, maleic anhydride (MAH), glycidyl methacrylate (GMA), etc. could improve the bonding with asphalt binder molecules with their reactive functional groups, leading to superior storage stability, enhanced temperature susceptibility, excellent ageing resistance, and improved rheological behaviour [94], [95].

Although polymer modification offers several benefits, it also poses certain challenges. Over time, certain polymers may undergo phase separation, causing a non-uniform distribution within the asphalt binder [96]. It is crucial for the polymer to be uniformly dispersed within the binder to ensure proper adhesion of the asphalt binder, as any incompatibility can lead to weak bonding, which can result in poor pavement performance and limited durability [97]. Furthermore, the cost of polymers and their modification methods can be a significant concern, as high-quality polymers and their modification processes tend to be more expensive, thereby contributing to the overall expenses of road construction [98].

Meanwhile, the growing concern over plastic waste generation, estimated at approximately 300 million tonnes annually worldwide, along with its inherently low biodegradability, has prompted the exploration of innovative applications for effectively repurposing plastic waste [17], [18]. Consequently, asphalt practitioners and researchers have shifted their emphasis towards incorporating waste plastic into asphalt concrete, thereby addressing the issue of plastic waste to some extent while also enhancing the performance of asphalt concrete through plastic modification.

2.3.2 Waste plastic as Asphalt modifier

Over the past 65 years, there has been a substantial increase in plastic production, rising from 2 million metric tonnes in 1950 to a staggering 380 million tonnes in 2015 [99]. It is expected to double again over the next 20 years as plastics become increasingly used in many applications [100]. Consequently, the rapid increase in production has led to a significant rise in plastic waste. According to a study conducted by Geyer et al., as of 2015, only 9% of total plastic waste had been recycled, 12% was incinerated, and 79% ended up being accumulated in landfills or in the ocean [99]. In 2018, China implemented a ban on importing plastic waste from foreign nations as part of their national policy known as "National Sword." Prior to this decision, China was handling an estimated 45% of the world's plastic waste, equivalent to about 106 million tonnes annually [101]. The global ramifications of this move have underscored the urgent need for international solutions to address plastic waste, such as reducing non-recyclable material production, redesigning products, and enhancing domestic waste management. It is estimated that by 2030, approximately 111 million tonnes of displaced plastic waste will result from China's new policy [102].

In 2022, a report revealed that global annual plastic production reached 390.7 million tonnes, with only 32.5 million tonnes coming from post-consumer recycled plastic (as shown in Figure 2.6). Furthermore, Figure 2.7 illustrates the annual worldwide production of plastic in 2022, categorised by its type. It is evident from Figure 2.2 that only 8.3% of post-consumer plastics were recycled in the year 2021. In response to this challenge, there has been a significant increase in global efforts to address plastic waste.

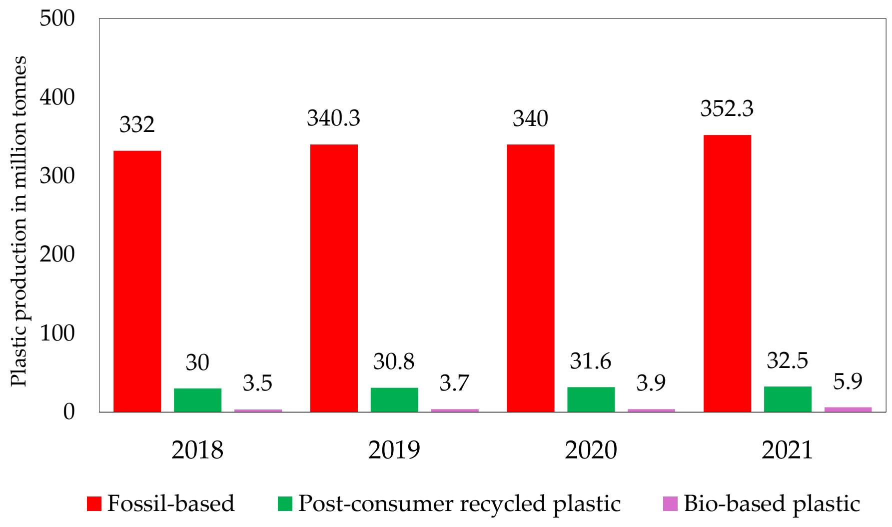

Figure 2. 6 Global plastic production from the year 2018 to 2021 (Plastics Europe, 2022) [103].

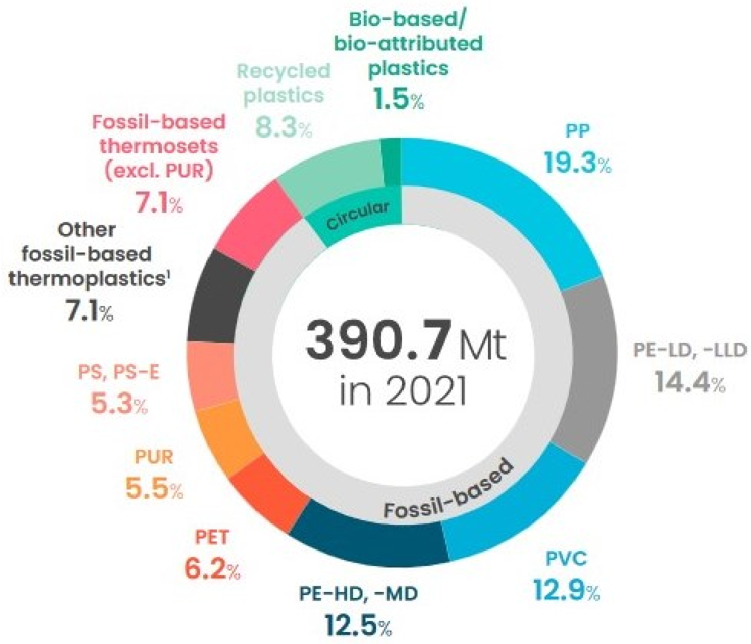

Figure 2. 7 Global production of plastic in 2021 based on their type (Plastics Europe, 2022) [103].

The growing concern over plastic waste has led to its incorporation in road pavements as a significant substitute for expensive virgin polymers, thereby improving the mechanical performance of the pavement. The types of waste plastic commonly incorporated in asphalt pavements include high-density polyethylene (HDPE), low-linear density polyethylene (LLDPE), low-density polyethylene (LDPE), polypropylene (PP), polystyrene (PS), polyvinyl chloride (PVC), ethylene vinyl acetate (EVA), polyethylene terephthalate (PET), acrylonitrile butadiene styrene (ABS), polyurethane (PU), and polycarbonate (PC) [104], [105]. Table 2.1 summarises the source of these waste plastics, their corresponding melting points, and their benefits in asphalt modifications. The selection of these waste plastics depends entirely on the target needs, performance criteria, and method of application. The recycled plastics are available in various forms such as shredded flakes, chips, pellets, powders etc [13], [106]. Moreover, the current trend involves producing recycled plastic pellets by mixing various waste plastics together with specific additives aimed at enhancing particular properties of asphalt [107]. There are typically two methods for incorporating recycled plastic into asphalt pavement: the wet method and the dry method. Figure 2.8 shows the graphical representation of wet and dry methods in an asphalt plant.

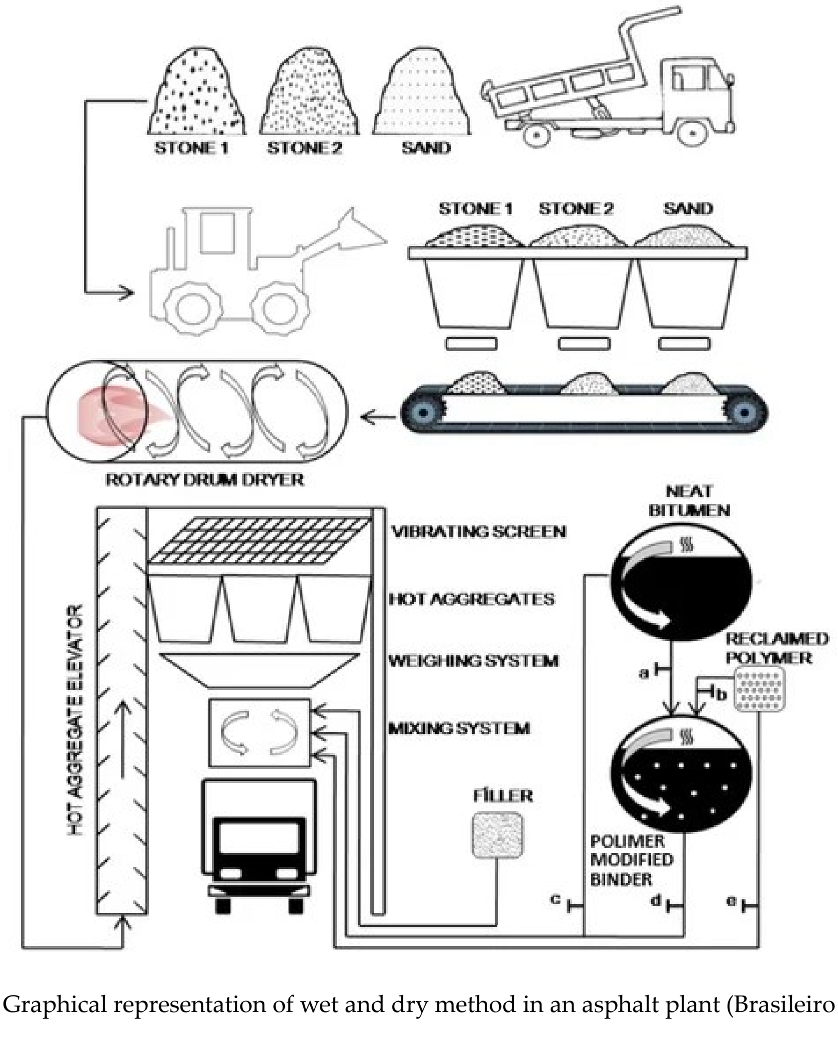

Figure 2. 8 Graphical representation of wet and dry method in an asphalt plant (Brasileiro et al.) [108].

Table 2. 1 Types of waste plastic and their benefits in asphalt modification. [97], [106], [109]–[112].

Type of waste plastic	Source	Melting point (°C)	Performance benefit in asphalt binder/mixtures
Polyethylene Terephthalate (PET)	Food packaging containers, soft-drink bottles, beverage glasses, jars, ropes	250–260	• Improved resistance to rutting and fatigue [113]. • Improvement in elastic properties [113]. • Resistance against moisture induced damage [114].
High-density polyethylene (HDPE)	Shopping bags, rigid pipes, toys, shampoo bottles, freezer bags, milk bottles, detergent bottles, buckets, crates, fencing, housewares	130–149	• Improved stiffness modulus at 25 °C [115]. • Enhanced resistance to permanent deformation [116]. • Higher resistance to moisture susceptibility [117].
Polyvinyl chloride (PVC)	Plumbing pipes and fitting, wire jacketing, blister, garden hoses, shower curtains, packaging, cooking oil bottles	100–260	• Improved tensile strength. • Improved ageing resistance [118]. • Increased resistance to rutting, fatigue [94]. • Improved storage stability [111].
Low-density polyethylene (LDPE)	Bubble wrap, films, sheeting, squeezable bottles, shopping bags, heavy duty sacks, general packaging	110–135	• Improved stiffness modulus [116]. • Improved resistance to cracking, thermal susceptibility [119]. • High resistance to rutting, fatigue [116]. • Good workability. • Improved Marshall stability [111].
Polypropylene (PP)	Industrial fibre, film, carpet fibre, appliances, automotive, toys, housewares, crates, palls, bottles caps, furniture, rigid packaging	130–171	• Improved high temperature performance [120]. • Increased resistance to rutting, moisture susceptibility [121].

Polystyrene (PS)	Trays, toys, medical disposals, package, hangers, CD cases, food container, transparent cups, diary containers, egg cartons	210–249	• Improved stiffness. • Improved resistance to deformation [120]. • Increased elastic recovery and storage stability [122]. • High resistance to rutting, thermal susceptibility [122].
Other plastics such as acrylonitrile butadiene styrene (ABS), acrylic, polycarbonate (PC), polyurethane (PU)	Automotive parts, aircraft parts, furniture, electrical appliances, CD/DVD, trolley bag etc.	depends on the type	• Increased resistance to rutting, moisture susceptibility [45], [104]. • Partial aggregate substitutes

2.3.3 Wet method

In the wet method, recycled plastic is directly introduced to the bitumen and blended using a high shear mixer (typically 1500 to 3500 rpm), typically at high temperatures ranging from 160 to 180 °C. The recycled plastic can be added in any form, such as shredded flakes, granules, or powders; however, the melting point of the recycled plastic is an indispensable factor [109]. The process of modifying bitumen in this way is commonly referred to as polymer-modified bitumen or plastic-modified bitumen. In asphalt plants, as shown in Figure 2.8, the neat bitumen and recycled plastic can be blended with a high shear mixer within a modification chamber when lines a and b are open, prior to entering the mixing phase [108]. Generally, an enhancement in elasticity, adhesion, and stiffness properties are identified, leading to increased mechanical performance and extended service life [110]. The crucial challenge in this method is to identify compatible recycled plastics that ensure certain properties with bitumen. Therefore, commonly used recycled plastics in this approach include polyethylene, polypropylene, and polyvinyl chloride. The rheological properties of the bitumen can be improved using a typical dosage of 1–8% recycled plastic, based on the weight of the bitumen, and thereby it could enhance the pavement performance at high, intermediate, and low temperatures [19], [109]. Several studies reported that the addition of recycled polyethylene terephthalate (PET) via wet method could increase the rutting resistance, bonding strength [123], moisture susceptibility, low temperature properties [124] and cracking resistance [125]. In addition, binder modification with low density-polyethylene (LDPE) has showed an enhanced impact on workability, resistance to rutting, antistripping and adhesive strength [126], [127]. However, this modification method is confined to recycled plastics with a relatively low melting point. A few studies have indicated that while the addition of recycled plastics improves binder properties, certain types, like PVC, may release harmful chloride emissions, rendering them unsuitable for modifying binders [128], [129].

2.3.4 Dry method

The dry method modification involves adding recycled plastic directly to the preheated aggregates before they are mixed with bitumen during asphalt mixture production [106], [130]. This approach can eliminate the need for sophisticated equipment and reduce high energy consumption, making it easier to integrate into any asphalt plant. As shown in Figure 2.8, in an asphalt plant, if the line e opens prior to the line c, the recycled plastic can be added to the mixing system along with the hot aggregates prior to mixing it with the neat bitumen [108]. In this method, recycled plastic can be introduced either as a partial substitute for the aggregate or as a modifier to the asphalt binder or mixture, depending on the type of plastic and its characteristics [14]. The recycled plastic with a high melting point is considered for aggregate replacement. Some studies have reported that improper distribution of recycled plastic in the asphalt mixture can negatively impact pavement performance; however, an enhanced implementation of this approach can substantially improve pavement performance in a sustainable manner.

The dry method allows for the incorporation of a wide range of recycled plastics in asphalt concrete, regardless of their melting point, when compared to the wet method. In most studies, recycled polyethylene (including LDPE and HDPE) is commonly employed using this approach. A study investigated asphalt mixtures with LDPE with varying contents of 0.5% and 1% on the weight of the aggregates using the dry method, and its results indicated an improvement in tensile strength, permanent deformation, and resistance to moisture damage [131]. The study by Poulikakos et al. reported that adding 1.5% polyethylene (PE) waste to asphalt mixtures resulted in improved resistance to permanent deformation, higher elastic behaviour, a low creep rate, and a higher creep modulus. However, increasing the percentage of PE to 5% led to an increase in permanent deformation [21]. In another study, comparing two distinct recycled plastic modifiers using the dry method showed comparable stiffness modulus values, rutting resistance, and fatigue cracking properties between both modified asphalt mixtures when compared with a reference SBS-modified mixture [132]. Moreover, studies have also shown effective integration of recycled PET, PP, and PS into asphalt concrete through this method [101]. Some previous studies have also suggested that the addition of recycled plastic through this method could significantly affect the compactability of the modified mix, depending on factors such as dosage, type, size, shape, and characteristics of the recycled plastic [133].

2.3.5 Plastic-modified asphalt pavements: Challenges and limitations

The previous sections have highlighted the advantages of incorporating recycled plastics into asphalt pavement. Many studies have shown that recycling plastic can offer numerous environmental and economic benefits over virgin plastic. However, there are currently no technical regulations or standards in place to determine the appropriate dosage of recycled plastic, its type, size, or the right method of incorporation [133]. While most studies have reported an increase in stiffness properties resulting from the addition of recycled plastic and its positive impact on rutting resistance [12], [134], only a few studies have investigated fatigue and low-temperature performance characteristics [23]. These aspects are crucial for ensuring the durability of the pavement. From an environmental perspective, concerns have been raised about the health and safety of road workers due to the incorporation of recycled plastic. Asphalt workers may be exposed to hazardous air pollutants when heating recycled plastics during recycled plastic-modified asphalt production and construction, posing significant health and safety concerns. The recycled plastics can release hazardous air pollutants such as benzene, toluene, and polycyclic aromatic hydrocarbons at elevated temperatures [101].

In Europe, strict regulations are in place for incorporating waste products, particularly plastics, due to concerns about the health and safety of the workers when handling secondary waste in asphalt production. Moreover, there is uncertainty about the environmental impacts and technical performance of recycled plastic-modified asphalts compared to traditional asphalt, leading to a lack of confidence within the road industry [135]. In regard to this, a study [136] examined the release of carcinogenic compounds from asphalt modified with various recycled plastics, including LDPE, PE, PET, and ABS. Their findings suggested that recycled plastic-modified asphalt reduces non-carcinogenic and carcinogenic risks compared to conventional asphalt. However, there is still limited research on the emissions and leaching effects of plastic-modified asphalts concerning the safety of workers and road users.

Furthermore, there is a lack of studies on the impact of recycled plastics on the recyclability of plastic-modified asphalt at its end-of-service life, which poses a crucial concern in the asphalt industry. If this is not feasible, incorporating plastic into asphalt may offer initial economic and performance improvements but could raise potential challenges at the end of its service period. There is only limited research regarding the impact of recycling agents on reclaimed asphalt pavements derived from plastic-modified asphalt sources, raising concerns about their high rate of recycling in asphalt mixtures.

Chapter 3
Research Methodology

3.1 Methodology

This chapter provides a comprehensive overview of the research methodology adopted in this thesis. The research was conducted in collaboration with multiple institutions, including the University of Palermo, Iterchimica S.p.A., and Aristotle University of Thessaloniki. In order to achieve the goals of this thesis, the research is structured into four tasks, as shown in Figure 3.1. The tasks are designed in a sequential manner, where each task acts as a driver for the next task, eventually contributing to the main objectives of this study. The primary task involves simulating the ageing behaviour of asphalt mixtures, which is crucial to replicate the end-of-life scenario and simulate the multiple life cycles. The next task deals with studying the multi-recyclability of asphalt mixtures, both with and without a recycled plastic modifier, for three life cycles through binder scale characterization. This task is pivotal as it involves the creation of multiple life cycle scenarios and assessing the influence of repeated ageing and rejuvenation within the asphalt mixtures. The third task of this research involves evaluating the multi-recyclability at the mixture scale, as it is significant to understand the mechanical and performance characteristics of the asphalt mixtures. The results of this provide valuable insights into the practical implications of the recycling process. Ultimately, developing a circular economic model to quantify material circularity for multiple life cycles enables us to identify and serve as a tool to assess the multi-recyclability potential of asphalt mixtures.

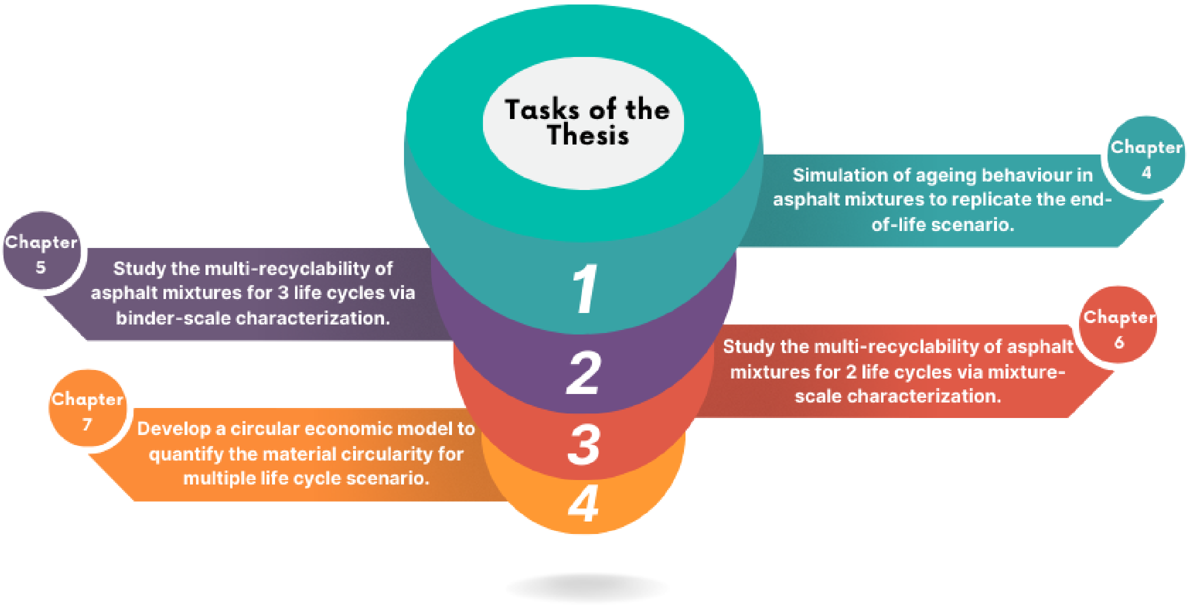

Figure 3. 1 Research methodology.

3.2 Testing methods

In order to accomplish the objectives of each task, various experimental methods were employed to study the binder-scale and mixture-scale characteristics. The following sections provide detailed insights into their significance, the procedures for preparing the specimens, and how the rheological, chemical and mechanical performance of asphalt binders and mixtures were evaluated.

3.2.1 Penetration test

The penetration test is a traditional method used to evaluate the consistency of the asphalt binders. The test was performed on the asphalt binder samples conditioned in a water bath at a temperature of 25 °C using a standard penetration needle according to EN 1426 [137]. The needle has a load of 100 g, and the loading duration lasts for 5 seconds. The vertical penetration of the needle is measured in tenths of millimeters, where lower values indicate high stiffness and higher penetration values represent softer consistency of asphalt binders.

3.2.2 Softening point test

The softening point test is a conventional method used to evaluate the thermal sensitivity of an asphalt binder at elevated service temperatures. The test involves casting the asphalt binder in two shoulder brass rings with metal balls on top of each and then heating them at a consistent rate of 5 °C/min in a liquid bath as per EN 1427 standards [138]. Subsequently, the softening point was determined by measuring the temperature at which the binder cast in the two brass rings becomes sufficiently soft to allow the enveloped metal balls to fall from a height of 25mm. In this study, softening point values were used to characterise the binders to study the ageing and rejuvenating effects.

3.2.3 Dynamic viscosity test

The viscosity of the asphalt binder reflects its resistance to flow and deformation. It is defined as the ratio of shear stress to shear rate and was determined at specific temperatures using a Brookfield rotational viscometer following EN 13302 [139]. The test involved measuring the torque needed to rotate a cylindrical spindle submerged in the binder at a constant speed using a thermoset temperature system. The resultant torque was then converted into viscosity, which is temperature dependent. As the temperature rises, the dynamic viscosity of asphalt binder decreases. In this study, dynamic viscosity of the asphalt binders was investigated at various high temperatures such as 135, 150, 160 and 180 °C.

3.2.4 Temperature sweep test

The temperature sweep test was performed on a dynamic shear rheometer (DSR) in order to investigate the behaviour of the asphalt binders within the temperature range of 25 °C to 90 °C. The test was carried out on a plate geometry of 25 mm with a gap of 1 mm between the plates and a constant shear stress of 500 Pa at a frequency of 10 rad/s with an increase in temperature of 1 °C/min. The loss factor of the asphalt binder is determined by the loss modulus (G'') and the storage modulus (G'). The G'' indicates the lost deformation energy of the material and serves as the viscous component of the material, whereas the G' represents the energy stored in the system and reflects the elastic part of the material. The parameter G'' is directly proportional to the loss factor that often depicts material losses, and which indicates the softness or hardness of the asphalt binder. Since G'–G'' equilibrium indicates the structure of a material, a rheological investigation employing this approach is regarded as valid for evaluating the behaviour of the ageing of asphalt binders. The loss factor also referred as tan δ can be calculated as the ratio of G'' to G'. In this study, loss factor curves and isochronal plots in terms of complex shear modulus were considered to assess the rheological behaviour of the binders.

3.2.5 Performance Grading

The upper-performance grading of the asphalt binders was determined using a DSR. The test was performed at a frequency of 1.59 Hz using a 25 mm plate geometry and a 1 mm gap in accordance with EN 14770:2012 [140]. The test started at 46 °C, and the temperature gradually increased at a rate of 6 °C until the test met the criterion of G*/Sin δ, which equals 1 kPa. The upper critical temperature refers to the failure temperature at which the test complies and G*/Sin δ equals to 1 kPa.

3.2.6 Binder Fast Characterization test

The Binder Fast Characterization Test, or BTSV (Bitumen-Typisierungs-Schnell-Verfahren), was used to evaluate the deformation behaviour of the asphalt binder at high temperatures using a DSR. The test determines the equi-shear modulus temperature (T_{BTSV}) and the corresponding phase angle (δ_{BTSV}) at which the asphalt binder exhibited a complex shear modulus (G*) of 15 kPa under a stress-controlled oscillation mode at a gradually increasing temperature as per EN 17643 [141]. The test involves a steady increase in the test temperature within the range of 25 °C to 90 °C, while the specimen was subjected to a constant shear stress of 500 ± 5 Pa under the oscillation at a frequency of 1.59 Hz. Nevertheless, in the scenario where there is no measured value within the range of complex shear modulus, G* = 15 ± 0.05 kPa, T_{BTSV} and δ_{BTSV} were calculated by interpolating the nearest values using an exponential value for T_{BTSV} and a linear function for δ_{BTSV}. The viscoelastic properties of asphalt binders can be investigated depending on the equi-shear modulus temperature, which correlates with the hardness of the binder and its corresponding phase angle to assess the degree of alteration.

3.2.7 Fourier Transform Infrared Spectroscopy

Fourier Transform Infrared (FTIR) spectroscopy examines the infrared light absorbed by bonds in molecules when it has the same frequency as the vibration frequency of the bonds, enabling the identification of different chemical functionalities based on the wavenumber. The analysis of the spectrum was carried out considering the intensity of the peaks, which represents the concentration of the bonds or functional groups. The wavelength of the spectral peaks corresponding to the Sulfoxide group (around 1030 cm^{-1}) and the Carbonyl group (around 1700 cm^{-1}) were considered in this study. The spectral peaks corresponding to the wavelength around 1460 cm^{-1} and 1376 cm^{-1} were considered as baseline for the peak analysis.

These spectral peaks are referred to aliphatic groups (CH_2 - CH_3) and they are not affected significantly during the oxidation process.

A Bruker Tenson 27 FTIR spectrometer with an Attenuated Total Reflectance (ATR) diamond crystal was used to investigate the chemical changes that occur due to the ageing and rejuvenation in an asphalt binder. The test was conducted by placing approximately 10 mg of asphalt binder directly on to the diamond crystal and the spectra were recorded within the wavelength range of 600 – 4000 cm^{-1} with a resolution of 4 cm^{-1} and an average of 32 repetitive scans. The analysis of the spectrum was carried out by calculating the Sulfoxide index and Carbonyl index, using the formula below:

$$\text{Sulfoxide Index (SI)} = \frac{\text{Area of the sulfoxide band centred at 1030 cm}^{-1}}{(\text{Area of the CH}_2\text{ centred at 1460 cm}^{-1} + \text{Area of the CH}_3\text{ centred at 1376 cm}^{-1})} \quad (3.1)$$

$$\text{Carbonyl Index (CI)} = \frac{\text{Area of the carbonyl band centred at 1700 cm}^{-1}}{(\text{Area of the CH}^2\text{ centred at 1460 cm}^{-1} + \text{Area of the CH}^3\text{ centred at 1376 cm}^{-1})} \quad (3.2)$$

3.2.8 Volumetric properties

The volumetric properties of the asphalt mixtures were evaluated in accordance with EN 12697-8 guidelines [142]. For this purpose, three specimens with a diameter of 100 mm from each mixture were compacted using a gyratory compactor up to 210 gyrations, as stipulated by the Italian technical specification. The gyratory compaction was performed by applying a constant vertical stress of 600 ± 3 kPa during each gyratory cycle. The data obtained from the compaction process was used to create the densification curves for the asphalt mixtures, a typical example depicted in Figure 3.2, indicating the relationship between the applied number of gyrations and the relative density of the compacted specimen. As per the technical specifications, determining air voids content at specific gyration levels such as 10, 120, and 210 is crucial for approving mixtures intended for use on highways.

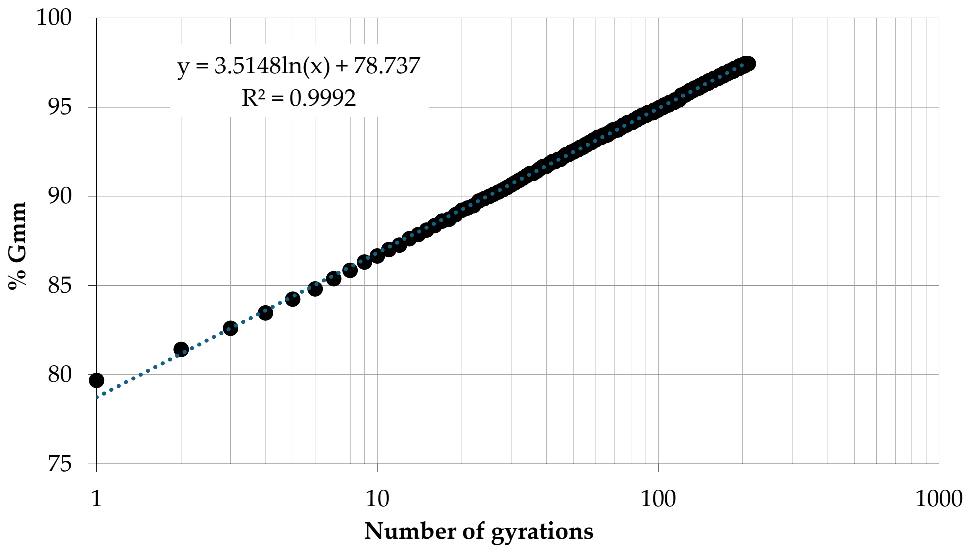

Figure 3. 2 Typical representation of densification curve.

3.2.9 Indirect Tensile Strength

The purpose of this test was to determine the indirect tensile strength (ITS), which refers to the maximum tensile stress calculated from the peak load applied to a cylindrical specimen loaded diametrically at a rate of 50 ± 2 mm/min until it fails. It clearly provides information on the cohesion and tensile strength of the asphalt mixtures. In accordance with EN 12697-23 [143], three specimens of 100 mm diameter from each mixture compacted at 120 gyrations were tested after being conditioned at 25 °C for 4 hours. The ITS can be calculated using the formula below:

$$ITS = \frac{2\,.\,P}{\pi\,.\,D\,.\,h}\,.\,1000 \qquad (3.3)$$

In the equation, ITS in kPa, P is the peak load in N, D is the diameter of the specimen in mm, and h is the thickness of the specimen in mm.

3.2.10 Indirect Tensile Stiffness Modulus

The indirect tensile stiffness modulus (ITSM) test was conducted to measure the resilient stiffness of the asphalt mixtures. The test was performed on a servo pneumatic load machine as shown in Figure 3.3, which applies 5 load pulses on the cylindrical specimen at a rise time of 80 ms to achieve the specified target horizontal deformation (7 ± 2 μm based on test temperature) according to the Annex C of EN 12697-26 [144]. In this study, in order to investigate the linear viscoelastic behaviour of the asphalt mixtures, three test temperatures of

10 °C, 20 °C and 30 °C were carried out. The stiffness modulus of each pulse can be calculated using the following equation.

$$E = \frac{F.(\nu + 0.27)}{(z.h)} \qquad (3.4)$$

In the Equation, E is the stiffness modulus in MPa, ν is the Poisson's ratio (assumed as 0.35), F is the applied load in N, z is the amplitude of the resilient horizontal deformation and h is the thickness of the specimen in mm.

Figure 3. 3 ITSM test setup.

3.2.11 Indirect Tensile Strength Ratio

The moisture susceptibility of the asphalt mixtures was determined using the indirect tensile strength ratio (ITSR), defined as the ratio of the indirect tensile strength of wet specimens to dry specimens. In accordance with EN 12697-12 [145], three specimens from each asphalt mixture were conditioned in a water bath at 40 °C for 72 hours followed by conditioning in an air chamber at 25 °C for 4 hours. The ITSR values can be calculated using the following equation:

$$ITSR = \frac{ITS_W}{ITS_D}.100 \qquad (3.5)$$

In the equation, ITS_W and ITS_D are the indirect tensile strength of wet and dry specimens in MPa, respectively.

3.2.12 Wheel tracking test

The wheel tracking test was conducted to evaluate the rutting resistance of the asphalt mixtures under repetitive wheel loads. The samples were prepared in accordance with EN 12697-22 [146], which involved producing two slabs measuring 305 x 305 x 50 mm for each mixture using a roller compactor. To ensure consistency, all specimens from each mixture were compacted to achieve a targeted air void range of 4 ± 0.5%. Prior to the test, the samples underwent conditioning at the test temperature of 60 °C for 4 hours within the chamber of the wheel tracking device. The test involved applying a load of 700 N to the specimen using a solid rubber tyre that reciprocated over it, covering a distance of 230 ± 10 mm. The frequency of the wheel was maintained at 26.5 cycles per minute while traversing across the specimen. The test was conducted under the condition that it would terminate when it reached 20000 passes or a maximum rut depth of 20mm. A typical rutting profile of an asphalt mixture is presented in Figure 3.4. Additionally, the wheel tracking slope (WTS_{Air}) can be calculated using the following equation to see the trend of rutting among the asphalt mixtures.

$$WTS_{Air} = \frac{(d_{10000} - d_{5000})}{5} \tag{3.6}$$

In the equation d_{10000} is the rut depth after 10000 cycles and d_{5000} is the rut depth at 5000 cycles.

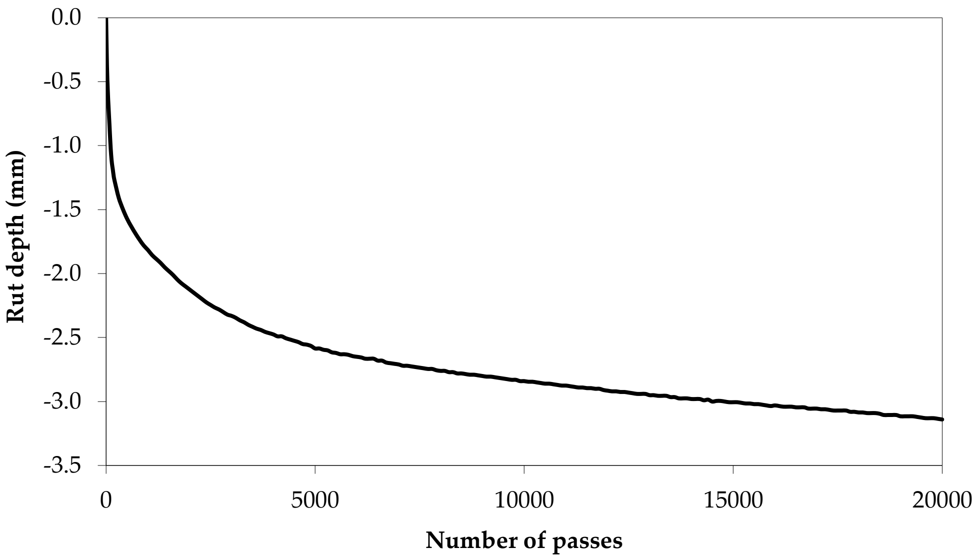

Figure 3. 4 Typical rutting profile of an asphalt mixture.

3.2.13 Indirect Tensile Fatigue Test

The indirect tensile fatigue test (ITFT) as shown in Figure 3.5 was conducted to evaluate the fatigue life of asphalt mixtures by applying repeated load pulses in a constant stress-controlled mode using an indirect tensile method according to EN 12697-24 [147]. A servo-pneumatic loading machine applied repeated haversine compressive load pulses along the vertical diameter of the specimen with a 0.1 s load time and 0.4 s rest time, inducing uniform tensile stress perpendicular to the applied load and ultimately causing specimen failure. The study included three stress levels (150, 250, and 350 KPa) for each mixture. A minimum of three cylindrical specimens of 100 mm diameter were prepared for each stress level, resulting in a total of nine specimens tested per mixture to assess the fatigue life. The fatigue life refers to the total number of load cycles that lead to the failure of the specimen and can be calculated using the following equations:

$$\varepsilon_0 = \left(\frac{2\,\Delta H}{\Omega}\right) . \left[\frac{1+3\nu}{4+\pi.\nu-\pi}\right] . 10^6 \tag{3.7}$$

$$N_f = k_\varepsilon . \left(\frac{1}{\varepsilon_0}\right)^{n_\varepsilon} \tag{3.8}$$

In the equations, ε_0 is the initial horizontal strain in μm/m, ΔH is the initial deformation in mm, ν is the Poisson's ratio, Ω is the diameter of the sample in mm, N_f is the number of load cycles, and k_ε, n_ε are the regression parameters of the fatigue line. Figure 3.6 shows a graphical representation of the fatigue line based on the aforementioned method.

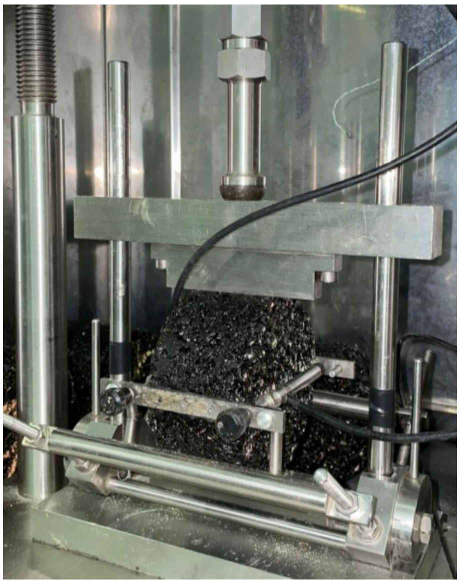

Figure 3. 5 ITFT test setup.

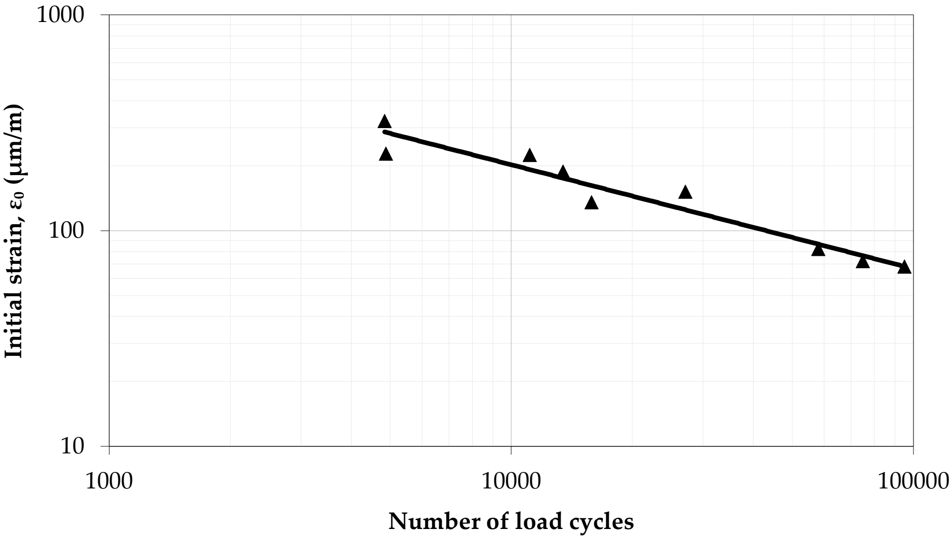

Figure 3. 6 Graphical representation of fatigue line.

Chapter 4
Simulation of ageing behaviour in asphalt mixtures

4.1 Introduction

Bitumen, often referred to as asphalt binder, is a viscoelastic material derived from the distillation of crude oil. It serves as a binding component in asphalt pavements, holding together other constituents and ensuring their structural integrity [148], [149]. However, over time, asphalt pavements undergo oxidative ageing processes due to climatic volatility and chemical reactions. Consequently, the asphalt binder becomes increasingly stiff and brittle, leading to higher susceptibility to cracking and degradation of the pavement [150], [151]. It is crucial to identify the ageing mechanism of asphalt binders to implement strategies that can extend the service life of the pavement. In the context of a circular economy, where resource efficiency and sustainability are of utmost importance, investigating the ageing mechanism becomes indispensable for adopting methodologies that enhance pavement performance, optimise resource utilisation, and minimise waste generation [152], [153]. Several studies have been conducted to understand the factors that influence the ageing of asphalt pavements and their impact on pavement performance. It has been observed that the effects of ageing have a significant impact on the long-term performance of pavements, which is crucial in the field of road engineering to provide durable and comfortable road surfaces for users [154]–[156].

In general, the ageing of asphalt mixtures can be categorised into two stages: short-term ageing (STA) and long-term ageing (LTA). The impact of high processing temperatures during the production and construction phases of asphalt mixtures results in the volatilisation of bitumen, known as short-term ageing. The evaporation of volatiles such as saturated and aromatic components is directly proportional to the production temperature of the asphalt mixture. Some previous studies reported that a rise in the production temperature increases the amount of loss of volatility, which has a linear influence on the imbalance of saturate, aromatic, resin, and asphaltene (SARA) fractions. Hence, it increases the stiffness, hardness, viscosity, and fragile nature of the bitumen [157], [158]. Simultaneously, long-term ageing occurs over the service life of the pavement due to climatic fluctuations, ultraviolet (UV) radiation, oxygen, and vehicle-induced loads. In this regard, oxidation is one of the key factors influencing the change in the chemical properties of bitumen as it gets exposed to oxygen. The oxidation influences the polar components in bitumen, such as causing asphaltenes to increase, and consequently, they stiffen the binder more, whereas nonpolar components like aromatics that control the flexibility of the binder get reduced due to ageing. Eventually, such ageing stiffens asphalt mixtures and leads to pavement distress [159], [160]. The oxidation rate generally depends on factors such as the bitumen content, access to oxygen by the pavement layer, void content, temperature, thickness of the layer, and the existence of cracks [161]. Based on this understanding, researchers have made numerous attempts to simulate field ageing phenomena in the laboratory using various accelerated ageing techniques to evaluate and anticipate the long-term performance of the pavement. Most studies have focused on analysing the influence of temperature, oxygen pressure, or a combination of both parameters to correlate with field performance. Among the ageing techniques, the most widely used laboratory protocol is the asphalt binder ageing, which utilises a rolling thin film oven (RTFO) and a pressure ageing vessel (PAV) to simulate the short-term and long-term ageing mechanisms of asphalt binders, respectively [162], [163]. These techniques are often employed in the Superpave performance grading system and serve as valuable tools for comparing and establishing benchmarks for various binders. However, this simulation approach fails to account for the influence of mineral aggregates, which can significantly impact the ageing simulation of asphalt pavement [164]. Although the use of high temperatures and pressure allows for the quick oxidation of bitumen, it cannot effectively replicate field conditions. To address the effects of mineral aggregates in the ageing mechanism, the American Association of State Highway and Transportation Officials (AASHTO) developed a standard asphalt mixture ageing protocol. This protocol can simulate long-term ageing effects in a compacted specimen with a diameter of 100 mm in an oven at 85 °C for 120 h. According to AASHTO R30, this accelerated ageing method simulates a long-term ageing level corresponding to five to ten years of service life. The significance of this method lies in its ability to simulate LTA effects within a reasonable time frame using standard laboratory equipment, making it cost-effective and easier compared to other approaches. However, this method has been criticised for the non-uniformity of ageing throughout the specimen, geometry distortion, and its inability to simulate ageing under various climatic conditions [165], [166]. Another advanced ageing technique is the Viennese Ageing Procedure (VAPro), which was developed in Austria. VAPro is considered an extension of the ageing procedure outlined in the SHRP-A-383 report. It utilises a unique configuration consisting of a triaxial cell with a forced flow of highly oxidative gaseous agents such as ozone and nitric oxide over the specimen, at a rate of 1 litre per minute. This approach reportedly simulates a long-term ageing condition closely resembling the field within an efficient timeframe of 4 days [167]. However, this ageing approach utilises an intricate setup for the ageing process, demanding costly configurations.

Furthermore, to address the limitations of previous ageing protocols on compacted specimens, researchers have made efforts to implement accelerated ageing methods for loose asphalt mixtures. The aim is to replicate more realistic field ageing mechanisms and better simulate the characteristics of reclaimed asphalt pavement (RAP). As mentioned earlier, the oxidative ageing mechanism of asphalt binder depends on various contributing factors, particularly temperature and ultraviolet (UV) radiation [168], [169]. Several studies have attempted to incorporate UV light into ageing techniques to investigate its influence on the behaviour of bitumen. They have identified that the energy from UV radiation can break down molecular bonds, leading to the surface ageing of the binder [170], [171]. Acknowledging the significance of temperature, it is a crucial factor considered in the laboratory simulation of long-term ageing characteristics of asphalt mixtures. The rate of oxidation increases with temperature, and particularly at high temperatures, the ageing may not accurately reflect real-field-aged asphalt binder [172]. A study conducted by the National Cooperative Highway Research Program (NCHRP) reported that long-term ageing can be simulated on a loose asphalt mixture at 95 °C inside a forced-draft oven. The duration of the conditioning period is calculated based on the climatic ageing index (CAI), which depends on the pavement temperature histories at different locations and depths. Data from the Enhanced Integrated Climatic Model (EICM) were used to predict pavement temperature by considering its relationship with pavement depth [166]. However, this approach, while considering pavement temperature histories at distinct locations, still neglects significant dynamic factors such as moisture and traffic conditions during the ageing simulation. This limitation could impact its applicability and its ability to accurately reflect real-field ageing phenomena, which remains a topic of ongoing discussion among researchers.

4.2 Scope and Objectives of the chapter

The literature reveals that several accelerated ageing techniques have been implemented to study the ageing mechanism and long-term performance of asphalt binders. However, there is still no consensus on the most effective approach for simulating ageing behaviour, leading to discrepancies in outcomes among research studies and making it difficult to compare and generalise the findings. In view of these challenges, in this chapter, the study aims to investigate and compare the ageing behaviour of asphalt binder using two different thermo-oxidative ageing approaches, one focusing on the binder itself and the other on the loose asphalt mixtures, with varying ageing durations. Although UV radiation is also a triggering factor for ageing, thermo-oxidative ageing was considered a vital cause, and that accounts for more than 80% of the total life cycle ageing of the bitumen [173]. The ageing behaviour was analysed extensively through physical, chemical, and rheological characterisation, and an ageing sensitivity index was employed to correlate their trends and provide a broader understanding of the ageing approaches. Further, the obtained results are compared with the field-aged RAPs collected from the same geographical region to assess the relevance of these ageing approaches under real field conditions.

4.3 Materials

In this study, a bitumen with a penetration grade of 50/70 was chosen considering the local availability and as the commonly used asphalt binder to produce asphalt mixtures on the highways of the southernmost region of Italy. The loose asphalt mixtures were prepared using limestone aggregates according to the Italian technical specifications for the highways [174], and their particle size distribution is reported in Table 4.1. Furthermore, to have extensive knowledge regarding the correlation of the considered artificial ageing methods with field ageing mechanisms, reclaimed asphalt pavements (RAPs) from two different sources were included. RAP-1 was milled from the surface layer of a pavement with a known history of 12 years of service life, and RAP-2 was from an unknown origin. The particle size distribution of the RAPs is shown in Figure 4.1.

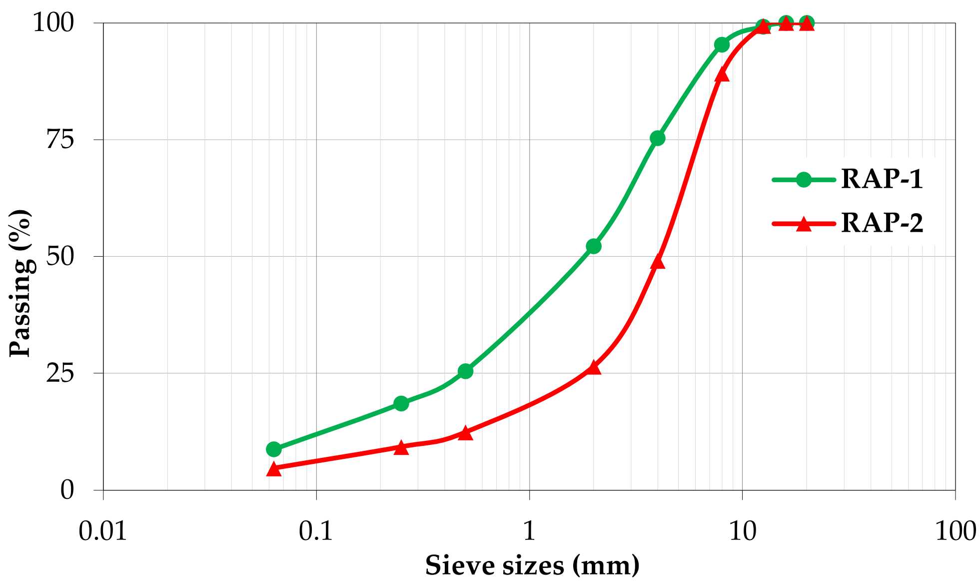

Figure 4. 1 Particle size distribution of RAPs.

Table 4. 1 Particle size distribution of aggregates.

Sieve Size	Passing Percent (%)			
	Fraction 6/12	Fraction 2/6	Fraction 0/4	Filler
16	100	100	100	100
12.5	100	100	100	100
8	81.3	100	100	100
4	5.3	38.5	94.2	100
2	0.6	6.2	68.6	100
0.5	0.6	3.8	38.3	100
0.25	0.6	3.6	27.1	100
0.063	0.1	2.2	10.3	70.7

4.4 Methods

4.4.1 Accelerated ageing techniques

The study consists of mainly two ageing approaches, one on the binder and another on the loose asphalt mixtures, to simulate the long-term ageing mechanism of asphalt binder that occurs over the service life of a pavement. Figure 4.2 illustrates the comprehensive research plan adopted in this study. The extensive details of the ageing methods are described in the following sections:

4.4.1.1 Ageing of asphalt binder

In this method, the asphalt binder was aged using the combination of a rolling thin film oven (RTFO) and a pressure ageing vessel (PAV) to simulate the long-term ageing mechanism. This approach was chosen since it is widely used and is the standard protocol for binder ageing. The RTFO is used to simulate the short-term ageing mechanism of asphalt binder in accordance with EN 12607-1 [175]. For this purpose, 35 g of bitumen was placed in a glass bottle and kept in the RTFO setup for 75 min at a temperature of 163 °C with an airflow of 4 L/min. Subsequently, after the completion of short-term ageing using RTFO, the binder residual of 50 g was placed directly into each of the circular pans and kept in the PAV for 20 h with an air pressure of 2.1 MPa according to EN 14769 [176]. The ageing temperature of 100 °C was chosen based on the grade of the binder used in the study. Furthermore, to have an extensive understanding of ageing behaviour with the increase in duration, the PAV-aged binder was repeatedly aged for an extra ageing duration of 20 h within the PAV under the same defined condition.

4.4.1.2 Ageing of loose asphalt mixtures

In this approach, the loose asphalt mixtures were prepared according to an Italian technical specification [174] for a dense-graded wearing course, and the mix design was optimized using the volumetric method. The grading distribution of the aggregate mix used is summarised in Table 4.2, with a binder content of 5% on the weight of aggregates determined as optimum to achieve an air void level of 4% on compacted specimens at the recommended design gyration level. All loose asphalt mixtures were produced according to this defined mix design and placed in a square tray (460 mm × 460 mm × 50 mm) with a maintained height of 25 mm, as depicted in Figure 4.3. This thickness was selected to align with the typical surface layer thickness of the pavement, which usually ranges from 25 mm to 50 mm.

Subsequently, the loose asphalt mixtures were short-term aged at 135 °C for 4 hours. Afterwards, for the purpose of long-term ageing, the trays of asphalt mixtures were kept in a non-ventilated oven at 95 °C for various ageing durations: 3 days, 5 days, 7 days, and 9 days. This was done to investigate the ageing behaviour with respect to the increment in duration. The ageing temperature was selected based on the NCHRP report as stated in the literature.

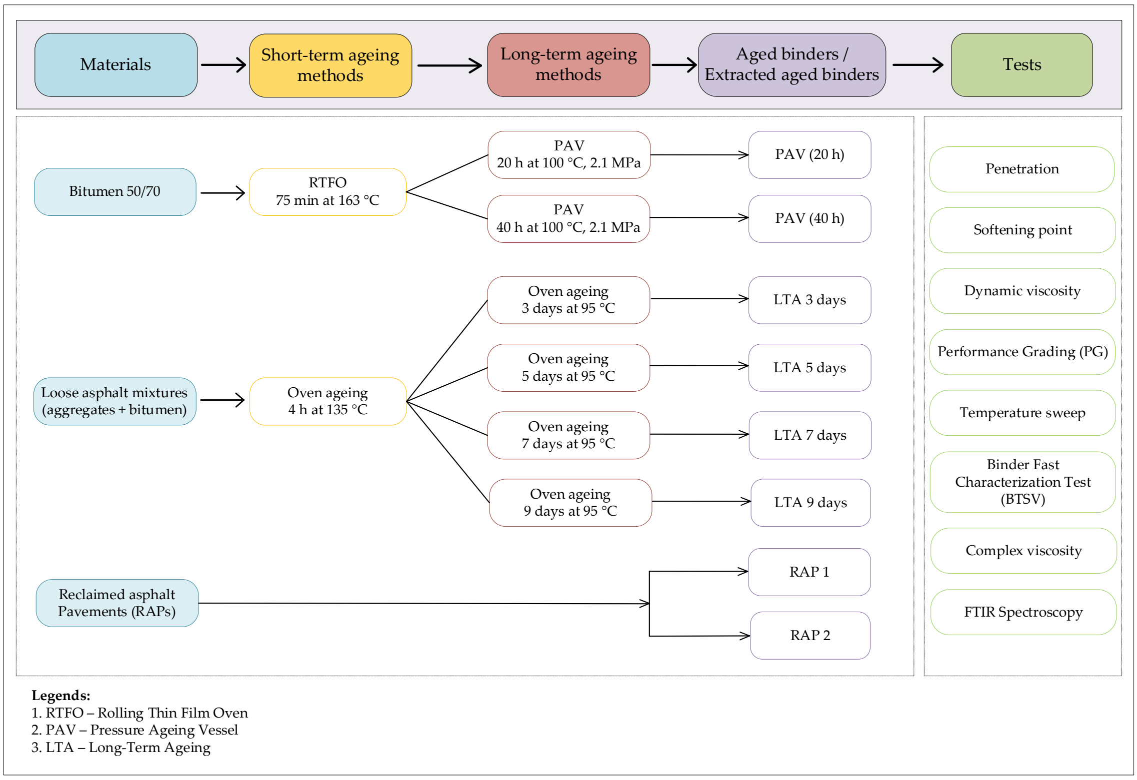

Figure 4. 2 Research plan.

Table 4. 2 Grading distribution of the mixture.

Sieve Size (mm)	Passing Percent (%)	
	Optimised Mix Gradation	Specification Limits
16	100	100–100
12.5	100	100–100
8	95.3	90–100
4	53.9	44–64
2	32.6	28–42
0.5	20.6	12–24
0.25	16.4	8–18
0.063	8.1	6–10

Figure 4. 3 Loose asphalt mixture ageing.

4.4.2 Extraction and recovery of aged binders

The binder was extracted from loose asphalt mixtures aged at discrete ageing durations and from two RAP sources using the standard binder extraction procedures prescribed in EN 12697-1 [177]. The hot extractor method with a wire mesh was used, and tetrachloroethylene served as the extraction solvent. Subsequently, a centrifuge was employed to remove the fine particles from the extracted solution, followed by binder recovery using a rotavapor in accordance with EN 12697-3 [178].

4.4.3 Testing methods

4.4.3.1 Physical properties

The physical properties of the aged binders were assessed using traditional methods, including the penetration test, softening point test, and dynamic viscosity test. The penetration test compared the consistency of unaged and aged binders at 25 °C. The softening point test evaluated the temperature sensitivity of asphalt binder aged through different ageing approaches. Additionally, the dynamic viscosity test examined the flow and deformation behaviour of the aged binders at various temperatures.

4.4.3.2 Rheological properties

The rheological characteristics of the binders aged via two different approaches and the field-aged binders were investigated with a dynamic shear rheometer. A temperature sweep test was performed in the range of 25 to 90 °C to evaluate parameters such as complex shear modulus (G^*) and phase angle (δ). Additionally, a comprehensive rheological analysis was conducted using test parameters including upper-performance grading, loss factor (tan δ), complex viscosity, equi-shear modulus temperature, and its corresponding phase angle.

4.4.3.3 Chemical properties

In this study, Fourier transform infrared (FTIR) spectroscopy was used to examine the chemical characteristics of aged binders. The analysis employed a Bruker Tensor 27 FTIR with an attenuated total reflectance diamond crystal to investigate the changes in the chemical functional groups of the aged binders.

4.5 Results and discussion

4.5.1 Penetration test

Figure 4.4 shows the results of the penetration tests. The aged binders, regardless of their ageing methods, exhibited a significant reduction in penetration value compared to the virgin bitumen 50/70 and RTFO binder. This reduction clearly quantifies the effect of oxidative ageing on asphalt binder. The PAV (40 h) and LTA 9 days binders showed relatively lower and comparable penetration values despite the ageing techniques, indicating the influence of ageing duration on the consistency of asphalt binder. Additionally, it was observed that PAV (20 h) and LTA 5 days aged binders demonstrated a similar hardening effect. Comparing field-aged binders' penetration values, RAP-1 correlated with LTA 3 days, while RAP-2 showed results comparable to PAV (20 h), LTA 5 days, and LTA 7 days aged binders. A previous study reported that two distinct RAP sources field-aged for twenty years had respective penetration values of 26.6 and 29.7 [179]. In comparison to these reported values, all investigated aged binders in our study demonstrated lower penetration values during analysis.

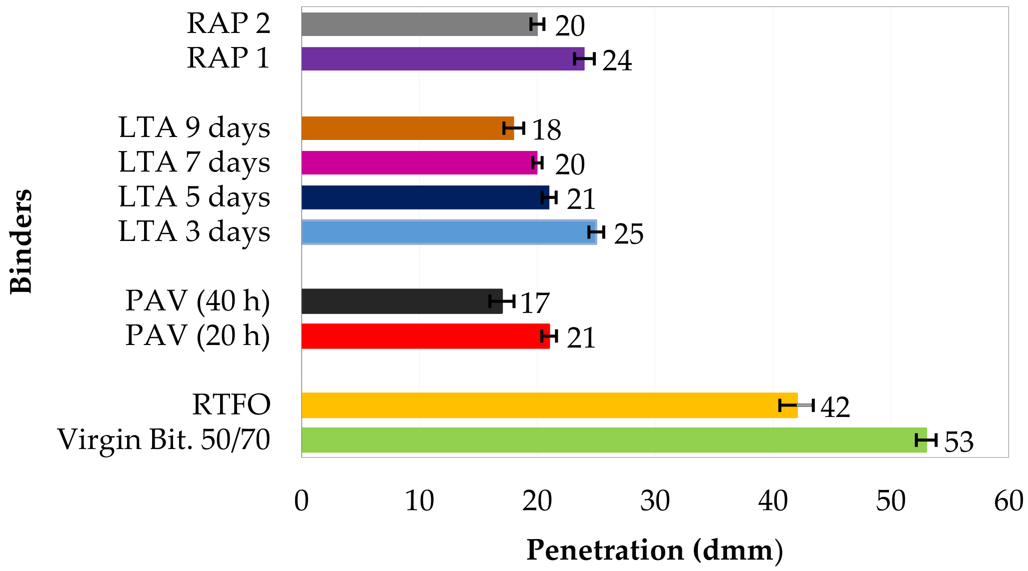

Figure 4. 4 Penetration values.

4.5.2 Softening point test

Figure 4.5 shows the softening point of all the binders studied. The results clearly demonstrate that different ageing techniques had varying effects on the softening points of the asphalt binder. In comparison, the PAV (40 h) binder exhibited a higher softening point of 65.6 °C, representing a 32.3% increase compared to unaged bitumen. It's important to note that both PAV (20 h) and LTA 5 days binders showed similar softening point temperatures, with an approximate increase of 22.6% compared to virgin bitumen 50/70. However, the extended ageing durations in loose asphalt mixtures did not show comparable high temperature sensitivity as seen in PAV (40 h) aged binder. The softening point values for LTA 7 days and LTA 9 days only increased slightly by 1.2% and 2.6%, respectively, when compared to PAV (20 h) and LTA 5 days. The thermal conductivity of aggregate could affect oxidation levels in the loose asphalt mixture approach, resulting in slowed oxidation rates with longer ageing durations. Considering the field-aged binders, RAP-1 showed a softening point similar to LTA 7 days and comparable to LTA 5 days and PAV (20 h). Conversely, the results of RAP-2 demonstrated a value comparable to LTA 9 days. According to the literature [179], two RAP sources with an age of 20 years had softening points of 61.4 and 61.7 °C, consistent with the values observed in this study for RAP-1.

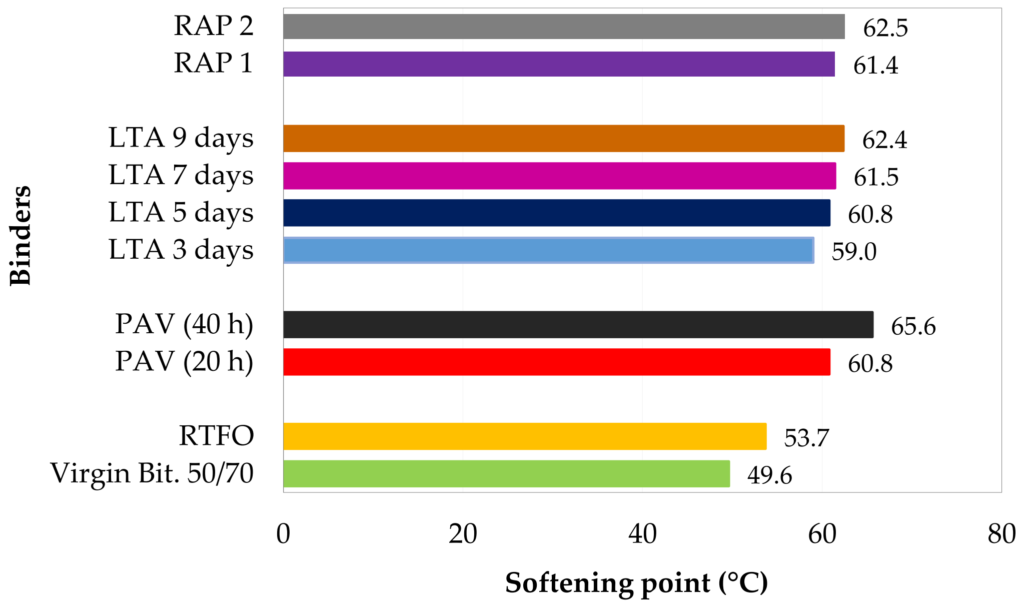

Figure 4. 5 Softening point values.

4.5.3 Dynamic viscosity test

In this research, a series of temperatures (135 °C, 150 °C, 160 °C, and 180 °C) were considered to evaluate and compare the dynamic viscosity of the aged binders. The results obtained are presented in Figure 4.6. According to the findings, PAV (40 h) exhibited the highest viscosity across all the tested temperatures, followed by the LTA 9 days aged binder. All the extracted aged binders from loose asphalt mixtures demonstrated a similar viscosity property to PAV (20 h) at the tested temperatures without showing a consistent trend of ageing severity for PAV (40 h). It is worth mentioning that both LTA 5 days and PAV (20 h) showed comparable dynamic viscosity at all performed temperatures. In terms of field-aged binders, both RAP-1 and RAP-2 displayed lower dynamic viscosities at all tested temperatures compared to PAV (40 h).

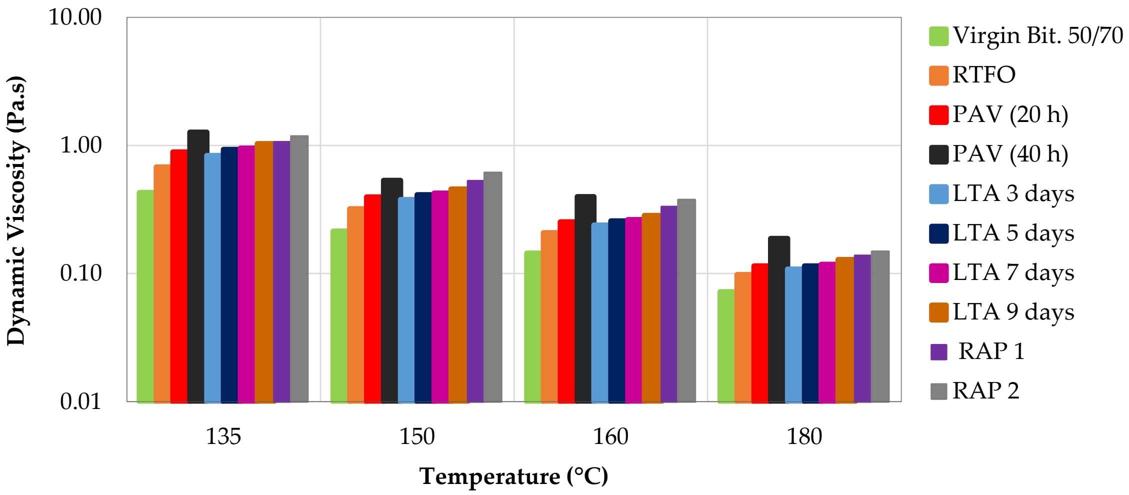

Figure 4. 6 Dynamic viscosity of the aged binders over various temperatures.

4.5.4 Performance Grading

In this study, the test was conducted using a DSR at a frequency of 1.59 Hz with a 25 mm plate geometry and 1 mm gap following EN 14770 standards. The test commenced at 46 °C, and the temperature gradually rose by 6 °C until reaching the criterion of G*/Sin δ = 1 kPa. The same testing conditions were applied to all aged binders and virgin bitumen since the primary focus was evaluating ageing correlations. It is anticipated that during binder ageing, their complex shear modulus (G*) should increase as phase angle (δ) decreases, resulting in increased binder stiffness. Consequently, asphalt binders aged under various conditions and methods exhibit variations in PG values at high temperatures; higher degrees of ageing correspond to higher upper PG values. According to the results presented in Table 4.3, the effect of ageing is clearly observed by the increase in upper PG values compared with virgin bitumen. The study mainly considered the upper critical temperature for comparison and found that the standard upper PG of aged binders was similar. The upper critical temperature refers to the failure temperature at which G*/Sin δ equals 1 kPa. Notably, among all studied ageing methods, PAV (40 h) showed the highest PG and upper critical temperature values. LTA 9 days exhibited the highest upper critical temperature among loose asphalt mixture ageing methods. However, the loose asphalt mixture ageing mechanism showed a deceleration in ageing level as duration increased after reaching a particular state compared to the binder ageing technique.

The PAV (40 h) depicted an increase of 6.5% and 26.2% of upper critical temperature compared to the PAV (20 h) and the virgin bit. 50/70, respectively. On the other hand, considering the loose asphalt mixture ageing technique, the LTA 5 days demonstrated a similar PG and upper critical temperature compared to the PAV (20 h). The binder LTA 9 days showed a 2.3% and 21.2% increase in upper critical temperature compared to LTA 5 days and virgin bit. 50/70, respectively. Accordingly, it is worth mentioning that the aged binders, PAV (20 h), and LTA 5 days demonstrated similar PG and upper critical temperatures during the analysis. Meanwhile, the field-aged binder RAP-1 showed a comparable result with LTA 5 days, LTA 7 days, and PAV (20 h). The results of RAP-2 were relatable to LTA 9 days, and both the field-aged binders exhibited a relatively lower critical temperature than PAV (40 h).

Table 4. 3 Results of the Upper Performance Grading.

Binder	Original Upper PG (°C)	Upper Critical Temperature (°C)
Virgin Bit. 50/70	64	67.5
RTFO	70	72.0
PAV (20 h)	76	80.1
PAV (40 h)	82	85.2
LTA 3 days	76	78.4
LTA 5 days	76	80.0
LTA 7 days	76	80.6
LTA 9 days	76	81.8
RAP-1	76	80.9
RAP-2	82	82.4

4.5.6 Temperature sweep test

The test results, shown in Figures 4.7 and 4.8, demonstrate the loss factor and isochronal plot of the complex shear modulus (G*), respectively. The findings clearly indicate that all ageing methods increased the stiffness of the binder compared to the virgin bit. 50/70. A lower value for the loss factor and a higher value for the complex shear modulus represent higher stiffness properties of the asphalt binder. Based on different ageing approaches, it is evident that PAV (40 h) aged binder exhibited the highest level of ageing, as observed in both plots. It's worth noting that LTA 5 days binder and PAV (20 h) aged binders showed similar curves in both plots, indicating a comparable stiffness effect during both ageing processes. Among the aged binders in the loose asphalt mixture, the LTA 7 days aged binder showed a curve very close to that of PAV (20 h) and LTA 5 days. It is important to note that the stiffness of LTA 9 days aged binder increased compared with other loose mixture aged binders; however, the ageing effect on stiffness remains less intense compared to PAV (40 h). The loss factor and isochronal curves of field-aged binder RAP-1 were comparable with those of LTA 5 days, PAV (20 h), and LTA 7 days. The isochronal curve of RAP-2 showed a similar trend to LTA 9 days. However, during the loss factor analysis, it was observed that the loss factor of RAP-2 decreased comparatively after reaching 60 °C compared to that of LTA 9 days.

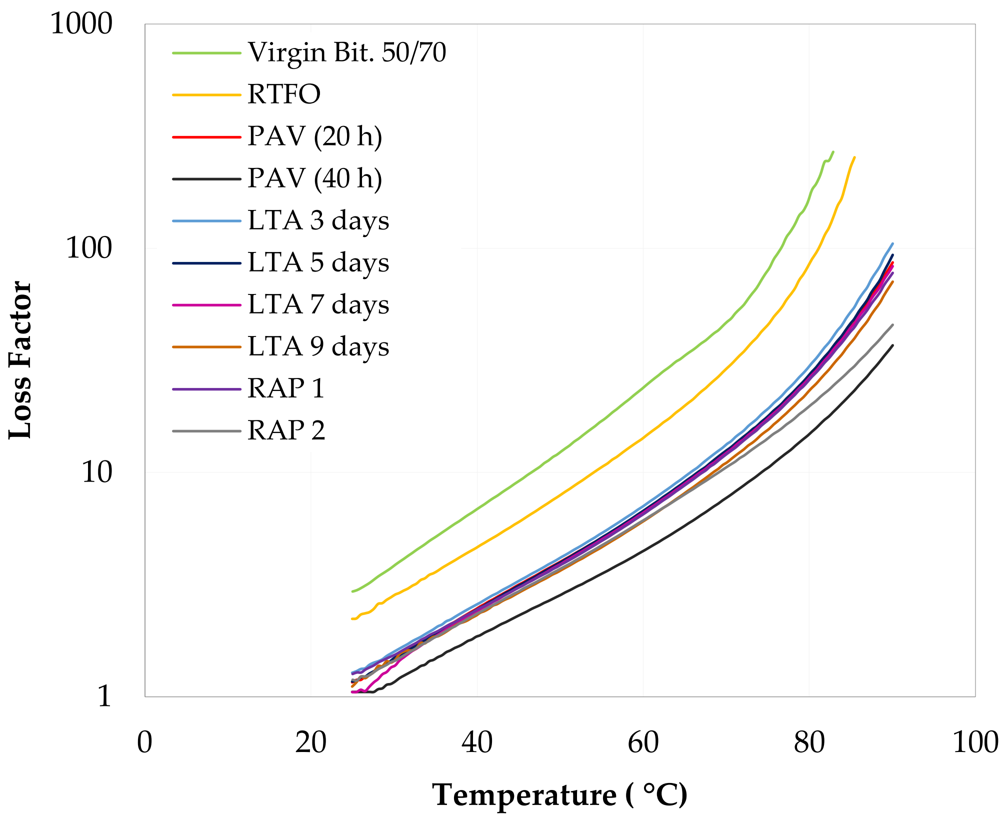

Figure 4. 7 Loss factor versus temperature curve.

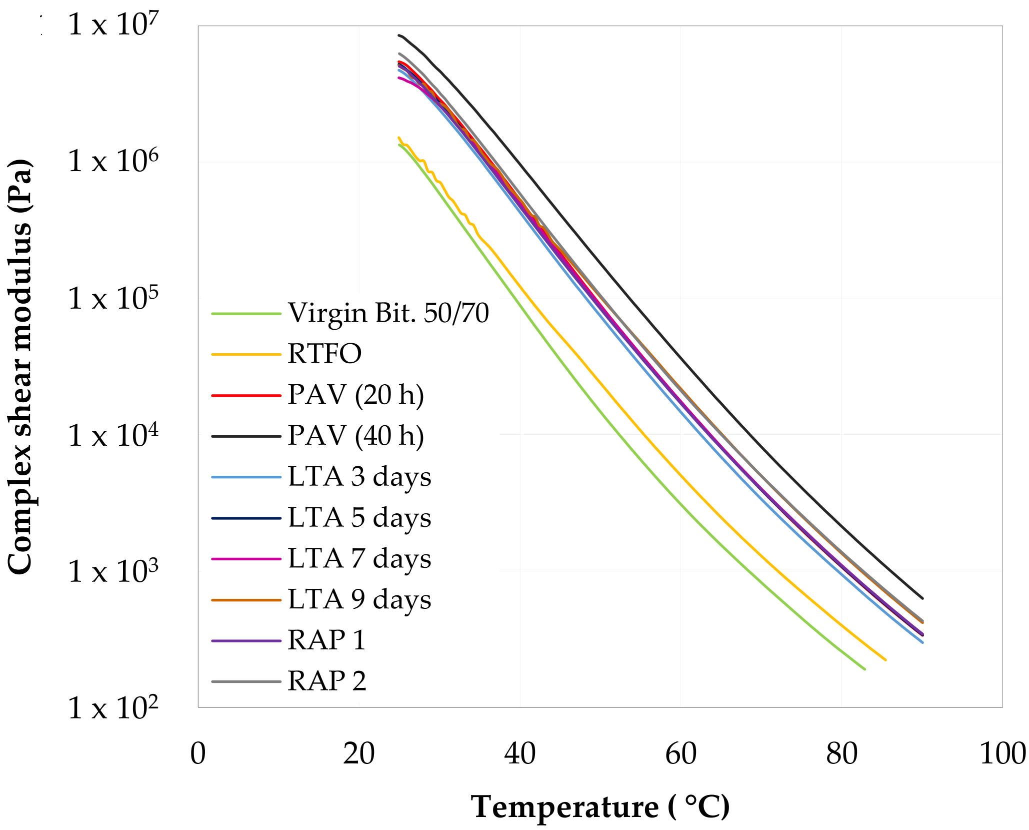

Figure 4. 8 Isochronal plots of the complex shear modulus (G*).

4.5.7 Binder Fast Characterization Test

The results of the BTSV test are summarised in Table 4.4. According to the obtained results, the equi-shear modulus temperature of all the aged binders increased and their corresponding phase angle decreased compared to that of virgin bit 50/70 and RTFO aged binder. Significantly, PAV (40 h) binder showed the highest equi-shear modulus temperature (T_{BTSV}) and lowest phase angle (δ_{BTSV}) among other aged binders. However, different ageing approaches led to different ageing levels of the binders. It is noteworthy that the results of the BTSV test also affirm a similar trend between PAV (20 h) and LTA 5 days aged binders, which demonstrated a comparable T_{BTSV} and δ_{BTSV}. The LTA 7 days and LTA 9 days aged binders showed a slight increase in the test parameters compared to the LTA 5 days and PAV-binders. However, all the binders demonstrated comparable T_{BTSV} values related to their respective softening point temperatures.

Table 4. 4 BTSV test results.

Binder	T_{BTSV} (°C)	δ_{BTSV} (°)
Virgin Bit. 50/70	49.9	85.3
RTFO	52.9	83.9
PAV (20 h)	61.0	81.9
PAV (40 h)	65.8	80.6
LTA 3 days	59.8	82.3
LTA 5 days	60.9	81.9
LTA 7 days	61.4	81.8
LTA 9 days	62.4	81.6
RAP 1	61.5	81.7
RAP 2	62.3	81.5

4.5.8 Complex viscosity

The complex viscosity is a parameter that can be calculated as the ratio of the complex shear modulus to its corresponding angular frequency. Figure 4.9 illustrates the complex viscosity curve of the aged binders within the temperature range of 25 to 90 °C. It is evident that all the long-term aged binders, despite their ageing methods, exhibited high complex viscosity compared to virgin bit. 50/70 and RTFO. In order to compare the effect of ageing, the complex viscosity at 60 °C was considered on the basis of its significance in the performance evaluation. It is worth mentioning that the PAV (20 h), LTA 5 days, and LTA 7 days exhibited comparatively similar and nearly 5.6 times higher viscosity than that of the virgin bit. 50/70. It was observed that PAV (40 h) showed the highest complex viscosity at 60 °C, which is almost 12 times higher than that of the virgin bitumen. The extended duration of 20 h with the combination of an induced pressure of 2.1 MPa and a temperature of 100 °C after the standard PAV could significantly escalate the degree of ageing, which results in a high stiffness of the binder. The complex viscosity of LTA 3 days and LTA 9 days was 4.8 times and 7 times higher than that of the virgin bit. 50/70, respectively. These results demonstrate that long-term ageing methods on loose asphalt mixtures could not drastically increase the degree of oxidative ageing similar to that of PAV (40 h) by extending the ageing duration. It was observed that the complex viscosity curve of RAP-1 was comparable with LTA 5 days, LTA 7 days, and PAV (20 h). On the other hand, RAP-2 exhibited a relatable curve with the LTA 9 days aged binder.

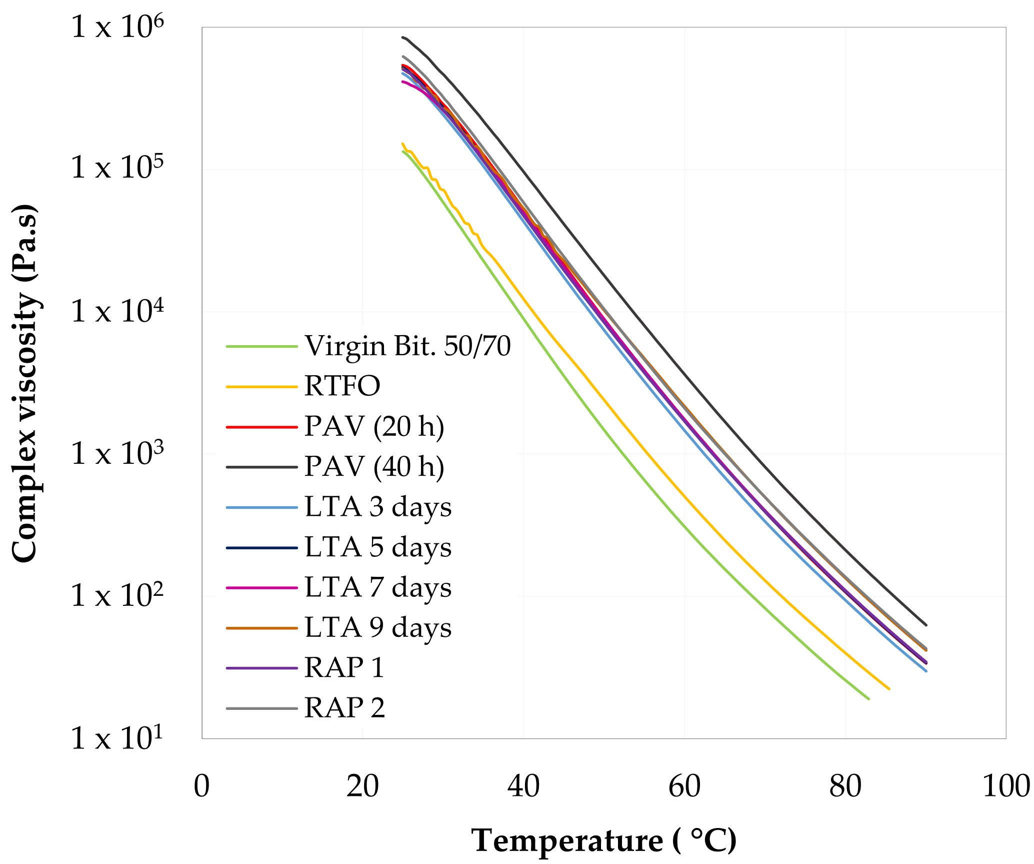

Figure 4. 9 Complex viscosity curve.

4.5.9 Fourier Transform Infrared spectroscopy

In this study, the wavelengths of the spectral peaks around 1030 cm^{-1} (sulfoxide group) and around 1700 cm^{-1} (carbonyl group) were analysed as they are the spectral peaks of the functional groups significantly affected during the ageing of asphalt binder, which were reported in many studies. Figure 4.10 shows the average obtained spectra of the aged binders and the virgin bit. 50/70. The spectral peaks corresponding to the sulfoxide band and carbonyl bands of all the aged binders increased during the oxidation phase compared to the virgin bit. 50/70. It is worth mentioning that the spectral peaks of PAV (40 h) were the highest among the aged binders, followed by PAV (20 h). A similar intensified trend was not observed among other long-term-aged binders. However, the oxidative ageing could be identified by the increase in the peaks of sulfoxide and carbonyl bands. This was also observed in the case of field-aged binders.

The calculated sulfoxide and carbonyl indices are summarised in Table 4.5. According to the calculated indices, the binders aged using PAV exhibited higher indices than the loose mixture ageing method carried out in the oven. Overall, it can be deduced that the application of air pressure during the PAV ageing method accelerated the chemical changes in the significant functional groups in the bitumen. In contrast, due to the absence of air pressure and the influence of the mineral aggregate composition in the loose asphalt mixture ageing method, the intensity of the chemical changes was comparatively slower than that of the binder ageing method. The spectrum analysis provided clear knowledge on the impact of ageing in the chemical functional group; however, a comparable trend could not be observed with this method. The calculated indices of RAP-1 and RAP-2 showed less ageing severity compared to PAV (20 h) and PAV (40 h). For instance, the severity of PAV (20 h) and PAV (40 h) was 1.2 and 1.6 times greater, respectively, than that of the investigated field-aged RAPs. The sulfoxide indices of both RAP binders exhibited comparable values to those of aged binders from loose asphalt mixtures, regardless of the ageing durations.

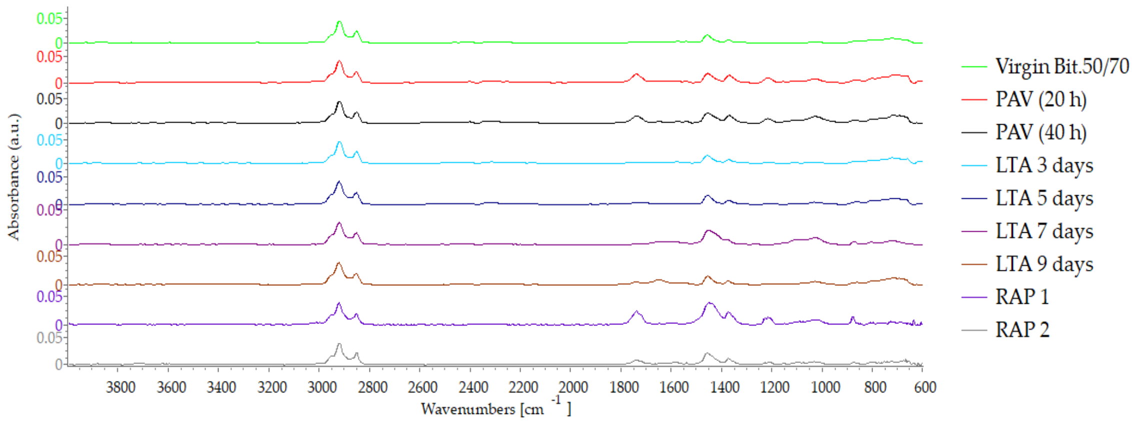

Figure 4. 10 Average obtained spectra of the investigated binders.

Table 4. 5 Calculated values of sulfoxide and carbonyl indices.

Binder	Sulfoxide Index (SI)	Carbonyl Index (CI)
Virgin Bit. 50/70	0.12	0.08
PAV (20 h)	0.31	0.48
PAV (40 h)	0.58	0.54
LTA 3 days	0.20	0.17
LTA 5 days	0.24	0.23
LTA 7 days	0.25	0.26
LTA 9 days	0.27	0.31
RAP 1	0.24	0.45
RAP 2	0.27	0.40

4.5.10 Comparative evaluation based on the ageing sensitivity index

The aim of this study was to investigate and compare the effect of the long-term ageing sensitivity of asphalt binder using two thermo-oxidative ageing approaches. More specifically, based on the results of the tests performed, it was shown that both ageing approaches were able to accelerate ageing to a certain extent. In order to meticulously evaluate the intensity of the investigated ageing methods, an ageing sensitivity index was deployed. This can be used to assess the severity of ageing with respect to the original unaged properties of the asphalt binder. This index tool can be utilised to identify the ageing sensitivity of each ageing approach with the increase in ageing duration. The ageing sensitivity index can be calculated according to the following equation:

$$\text{Ageing Sensitivity Index} = \frac{|(\text{Evaluated parameter after ageing} - \text{Evaluated parameter before ageing})|}{\text{Evaluated parameter before ageing}} \quad (4.1)$$

Figures 4.11 and 4.12 depict the calculated ageing sensitivity index for all the assessed parameters. The increase in the ageing sensitivity index signifies the severity of ageing with respect to the unaged condition and its resultant impact on each test parameter. Based on the observed results, among the various ageing methods examined, PAV (40 h) demonstrated the highest ageing sensitivity index. It is worth noting that the standard binder ageing protocol, PAV (20 h), and the loose asphalt mixture aged for 5 days exhibited a very similar ageing index during the rheological evaluation. Significantly, it is noted that the degree of ageing of the loose asphalt mixture approach was slower as the ageing duration increased. According to the literature, the surface of the aggregates can adsorb the most polar compounds of bitumen in the fractions of asphaltenes and resins [180], [181]. The adsorption, along with the thermal conductivity of limestone aggregates, could decelerate the oxygen diffusion in the studied loose asphalt mixtures. On the other hand, extending the PAV duration to 40 hours considerably accelerated the ageing level due to the repeated application of 2.1 MPa pressure and a temperature of 100 °C. It is essential to highlight that such an extension in ageing duration did not align with the ageing of the studied field-aged RAP binders in both rheological and chemical analysis. Consequently, the extension of PAV duration demonstrated a more pronounced ageing effect compared to all the investigated binders. Considering the sum of all the calculated ageing sensitivity indices for the physical and rheological parameters, PAV (40 h) showed 2.1 times the severity of PAV (20 h) and 1.7 times with LTA 9 days. Since data on the unaged properties of field-aged binders were unavailable, RAP binders were excluded from this specific analysis.

In the chemical analysis, the carbonyl and sulfoxide indices of PAV (20 h) and PAV (40 h) were relatively higher than those of the aged loose asphalt mixtures, indicating a high sensitivity to ageing among the investigated aged binders. In particular, LTA 5 days and PAV (20 h) showed mostly identical results from the physical and rheological analyses. The FTIR spectroscopy analysis, on the other hand, demonstrated that the ageing sensitivity index for the chemical parameters of PAV (20 h) was 2.3 times greater than that of LTA 5 days, indicating a varying trend among the rheological and chemical behaviours of the asphalt binder.

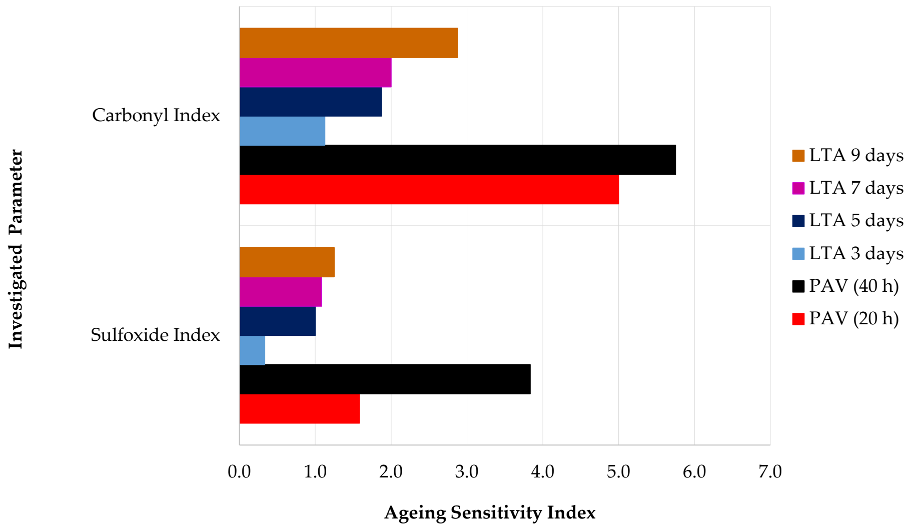

Figure 4. 11 Ageing sensitivity index of the investigated chemical parameters.

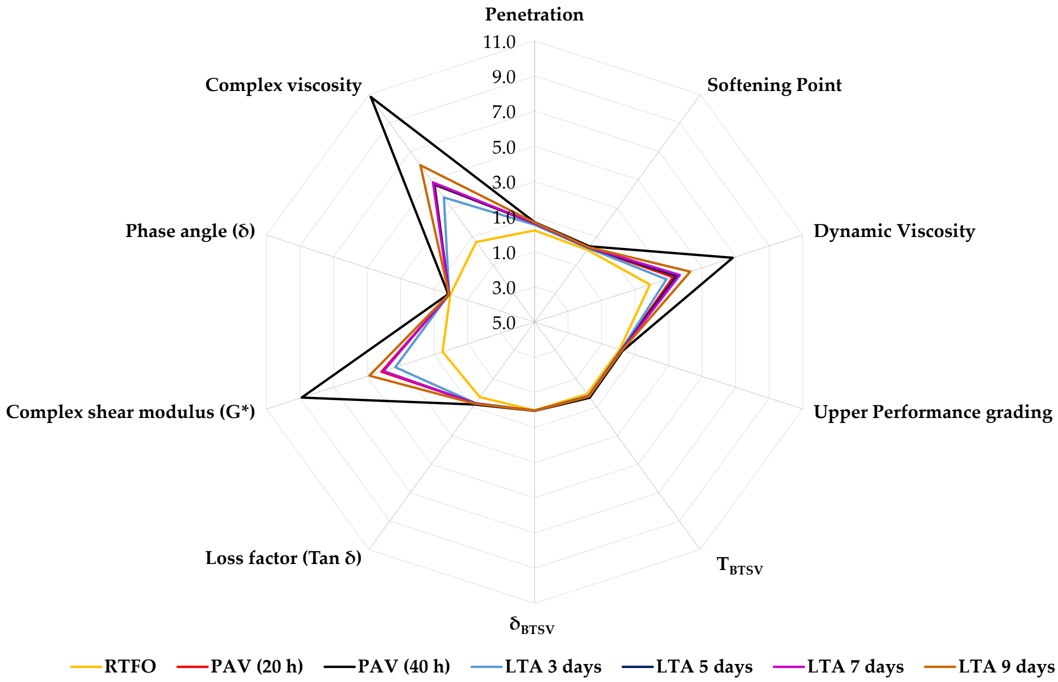

Figure 4. 12 Ageing sensitivity index of the investigated physical and rheological parameters.

4.6 Summary and Conclusions

This study addressed a comprehensive evaluation of the effect of thermo-oxidative ageing methods on asphalt binder, their correlation, and their relevance to field-aged binders. For this purpose, two artificial ageing approaches were employed in the study, one on the binder and another on the loose asphalt mixtures with distinct ageing durations. The impact of these ageing methods on the behaviour of asphalt binder was investigated using physical, rheological, and chemical characterization. The ageing degree caused by these ageing methods was meticulously evaluated using an ageing sensitivity index.

Based on the results, it was observed that both ageing approaches for the asphalt binder could significantly accelerate the ageing degree to a certain level with an increase in the duration. The ageing sensitivity analysis signified that PAV (20 h), which is the standard binder ageing protocol, and the loose asphalt mixture aged for 5 days demonstrated a very similar ageing trend among the investigated rheological parameters. Alongside that, the intensity of oxidative ageing in the loose asphalt mixture decreased after a certain level of ageing, and further increments in the ageing duration did not escalate ageing severity compared to that of PAV ageing. The adsorption of the polar compounds of bitumen on the aggregate surface, along with the thermal conductivity of limestone aggregates, could account for decelerating the oxygen diffusion in the loose asphalt mixtures. In this regard, a decline in the oxidation rate was noticed in the cases of the LTA 7 days and LTA 9 days aged binders. The highest ageing degree was observed in the PAV (40 h) aged binder, which was higher than that of all the studied approaches and field-aged binders, showing the severity of such an ageing procedure. On the other hand, the rheological and chemical properties of the loose asphalt mixture ageing method provided an analogous relationship with the studied field-aged RAPs. However, these observed trends might not be evident enough to predict the ageing degree of certain years of field ageing phenomena and require further investigation. The FTIR spectroscopy analysis revealed that the ageing methods influenced the functional groups associated with ageing in distinct ways, even though they exhibited similar rheological behaviour. The studied chemical indices of binder aged in the PAV for 20 and 40 h exhibited significantly high values, which were not consistent with the field-aged binders.

Overall, it can be inferred that this study could yield significant insights into various accelerated ageing methods for asphalt binders and their correlation with the standard ageing protocol. Thermo-oxidative ageing methods are consistently employed to assess the long-term performance and ageing characteristics of asphalt binders, whether they include modifiers or not. Contrastingly, several factors influence the ageing behaviour of asphalt pavement; oxidative ageing remains a crucial aspect. Based on the results, it becomes evident that in order to evaluate the ageing behaviour or long-term performance of any asphalt binder, it is essential to consider the thermal properties of the mineral aggregates, as well as their composition within the mixture, to gain a comprehensive understanding. Additionally, due to the absence of a standardised ageing protocol for loose asphalt mixtures, this study opens significant possibilities for comprehending the ageing mechanisms within these asphalt mixtures. This comparative assessment of the loose asphalt mixture ageing protocol could lead to a way to implement this approach to evaluate the ageing mechanism and long-term performance of dry method modification in asphalt mixtures, which was considerably challenging to evaluate. It provides a cohesive evaluation of the expected life cycle of a binder and thus allows us to understand the service life and the time window of the life cycle of a binder due to its ageing. However, the presented results are only valid for the specified type of bitumen, and further investigation is recommended with modified binders and other mineral aggregates in loose mixture ageing to gain a prolonged knowledge of the ageing mechanism and its associated mixture-level performance. Further study on the influence of UV rays, along with thermo-oxidative ageing, on loose asphalt mixtures is also recommended to gain a broader understanding.

Chapter 5

Multi-recyclability study: From a binder-scale perspective

5.1 Introduction

Asphalt pavement, a crucial component of modern infrastructure, has undergone substantial evolution over decades [182]. Advancements in material composition, design approaches, and construction methodologies have transformed traditional asphalt into a highly engineered product capable of addressing the diverse requirements of modern transportation systems [183]. In this context, asphalt modifications play a significant role in enhancing the performance and durability of the pavement. Several polymers have been introduced in the binder or asphalt mixture and successfully implemented to improve the elasticity and resistance to deformation of the pavement [184]. Subsequently, the growing trend towards sustainability has led to the incorporation of alternative materials in the modification process. Considering the increasing issue of plastic waste and policies aimed at reducing the reliance on virgin materials, waste plastic has been repurposed for use in asphalt pavement as a modifier or aggregate replacement [185], [186]. Technological advancements have made it less challenging to integrate recycled plastic into asphalt mixtures using either dry or wet methods [125]. Several studies have demonstrated that the incorporation of recycled plastic in asphalt mixtures can improve mechanical performance while reducing the overall carbon footprint involved in the production process [187], [188]. Furthermore, the incorporation of recycled plastic as a potential modifier has reduced the need for extracting and processing costlier virgin materials, thereby improving resource efficiency [189]. However, within the context of the circular economy of asphalt pavements, the recyclability of recycled plastic-modified pavement is also a vital factor to be considered.

5.2 Scope and Objectives of the chapter

The technical performance of recycled plastic-modified asphalt binders and mixtures has been extensively studied. However, there are still some aspects that need to be addressed, such as the recycling of reclaimed asphalt pavements containing asphalt modifiers like recycled plastic. With the increasing trend of high RAP recycling, the importance of understanding the characteristics of RAPs containing recycled plastic remains crucial. Currently, limited research exists on this topic; therefore, this study aims to investigate the characterization and viability of multi-recycling recycled plastic-modified asphalt mixtures at a 50% recycling rate for three life cycles through a binder-scale study.

5.3 Materials and methods

5.3.1 Materials

In this study, limestone aggregates were used to produce a dense graded asphalt mixture for a wearing course. The design gradation used in this study was in accordance with an Italian technical specification for the highways [174]. As for the asphalt binder, a paving grade bitumen 50/70 was used in the study. The bitumen was selected based on firstly, the local availability and secondly, as the commonly used binder in the production of asphalt mixtures for the highways of the Sicily region in Italy.

Considering the main objective of this study, a recycled plastic asphalt modifier was used for the modification of the asphalt mixture. The additive shown in Figure 5.1 was made of recycled hard plastics and graphene nano materials so-called Graphene-enhanced Recycled-Plastic additive, hereinafter (GRP). Some of the given technical properties are summarized in Table 5.1. It is worth mentioning that the waste plastics used to produce this asphalt modifier are hard plastics, such as toys, bins and baskets, plastic tables, etc., and not softer ones like plastic bags and water bottles.

Figure 5. 1 Graphene-enhanced recycled plastic modifier.

Table 5. 1 Technical properties of the Graphene-enhanced recycled plastic.

Properties	Unit	Value/Aspect
Material State	-	Granules
Colour	-	Black
Apparent density at 25 °C	g/cm³	0.4–0.6
Softening Point	°C	160–180

In addition, this study employed a bio-based rejuvenator, hereafter (R), composed of vegetal derivatives as the recycling agent, as shown in Figure 5.2, considering the high recycling rate during each cycle. The choice of the rejuvenator was based on material screening according to the proven performance of the commercially available rejuvenators on the market. Among many available rejuvenators, the quality and performance of this rejuvenator have been examined in several broad research works that ascertained its choice. Some of the technical properties of the rejuvenator are presented in Table 5.2.

Table 5. 2 Technical properties of the rejuvenator.

Properties	Unit	Value/Aspect
Material State	-	Liquid
Colour	-	Dark brown-purple
Density at 25 °C	g/cm³	0.85–0.95
Viscosity at 25 °C	cP	50–150
Flash point	°C	≥ 200

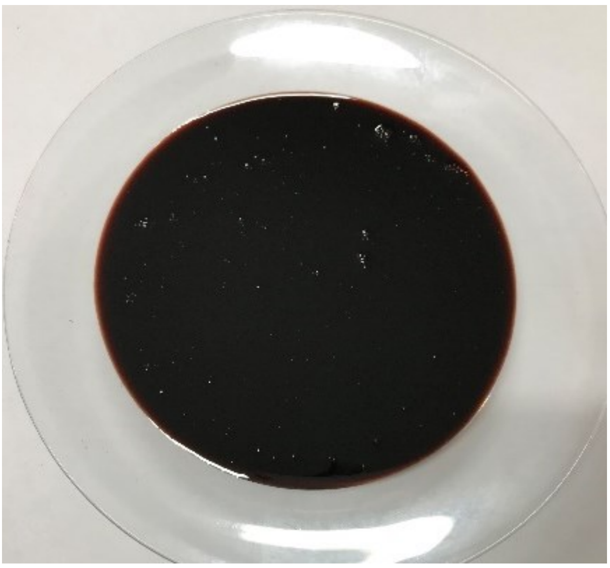

Figure 5. 2 Bio-based rejuvenator.

5.3.2 Research plan

Considering the objective of this research, two sets of asphalt mixtures were investigated: i) unmodified asphalt mixtures as the control mixtures without any asphalt modifier, and ii) recycled plastic-modified asphalt mixtures. The multi-recyclability of the asphalt mixtures was evaluated by applying a defined research plan, shown in Figure 5.3. According to the defined plan, a 50% recycling rate was considered at each recycling cycle. It is worth mentioning that in addition to the binders extracted from the mentioned mixtures, as reference, bitumen 50/70 at unaged and aged states was also considered during the experimental work and the data analysis. This study aimed to investigate the viability of multi-recycling via a comprehensive binder-scale characterization. According to the literature, high RAP incorporation is restricted due to the high stiffness of the aged binder, which is one of the prime reasons. Within this context, the properties of RAP from recycled plastic-modified asphalt mixtures are still unknown or not yet broadly researched. Responding to the lack of knowledge about the circular economy of plastic-modified asphalt mixtures, the study of RAP from such a modified asphalt mixture is inevitable. Currently, according to most of the Italian technical specifications, the maximum allowable rate of RAP inclusion is 15-20% in the wearing course. The choice of 50% RAP incorporation in the study was based on the foreseen level of recycling in the Italian asphalt industry and the feasibility of the recycling approach in many existing asphalt plants.

The abbreviations used for this study, along with their descriptions, are provided below:

- Mix-CA^n: long-term aged control asphalt mixture of the recycling cycle n; where n = 0, 1, 2.
- Mix-$GRPA^n$: long-term aged GRP-modified asphalt mixture of the recycling cycle n; where n = 0, 1, 2.
- CA^n: extracted aged binder from Mix-CA^n, where n = 0, 1, 2.
- $GRPA^n$: extracted aged binder from Mix-$GRPA^n$, where n = 0, 1, 2.

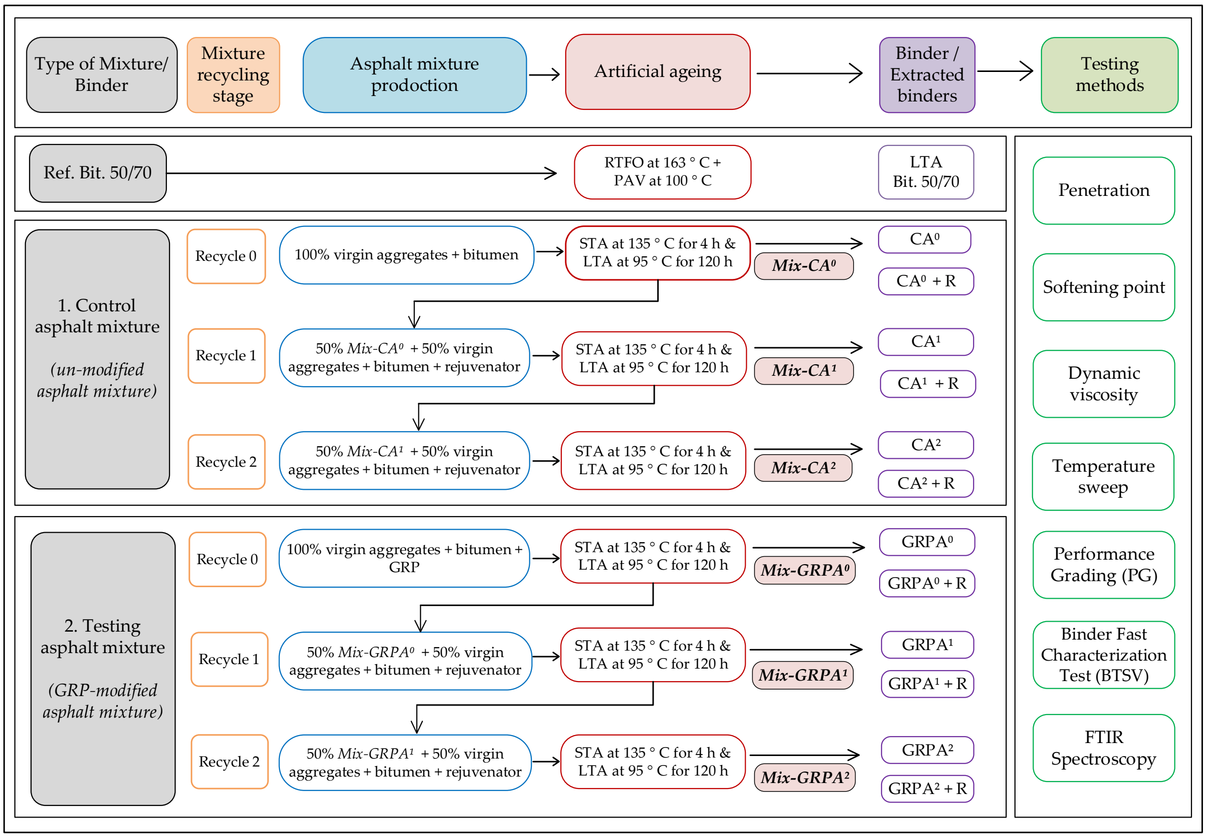

Figure 5. 3 Research plan.

5.3.3 Mix design

The mix design used in the study was according to an Italian technical specification [174] for wearing courses with a nominal maximum aggregate size of 12.5 mm. The grading distribution curve of the designed mix is demonstrated in Figure 5.4. The control mixture was optimized using both the Superpave and Marshall methods. The volumetric characteristics of the control mix were evaluated using the gyratory compacted cylindrical specimens up to a gyration level of 210 gyrations. As per the technical specifications, determining air voids content at specific gyration levels such as 10, 120, and 210 is crucial for approving mixtures intended for use on highways. Based on this, as shown in Figure 5.5, the optimum binder content was identified as 5% by the weight of the aggregates, which exhibited air voids content within the recommended limits at these specified gyrations. Additionally, indirect tensile strength (ITS) test was also performed on these specimens; the results of the test were within the prescribed limit of the technical specification, which is presented in Figure 5.6. Moreover, the Marshall stability of the control mixture with different binder contents was also studied using Marshall specimens compacted with 75 blows at each side according to EN 12697-34, and the results are shown in Figure 5.7. The analysis of the volumetric characteristics of Marshall compacted specimens with different binder contents is illustrated in Figure 5.8. Hence, all these studied factors affirmed the quality of the mix design.

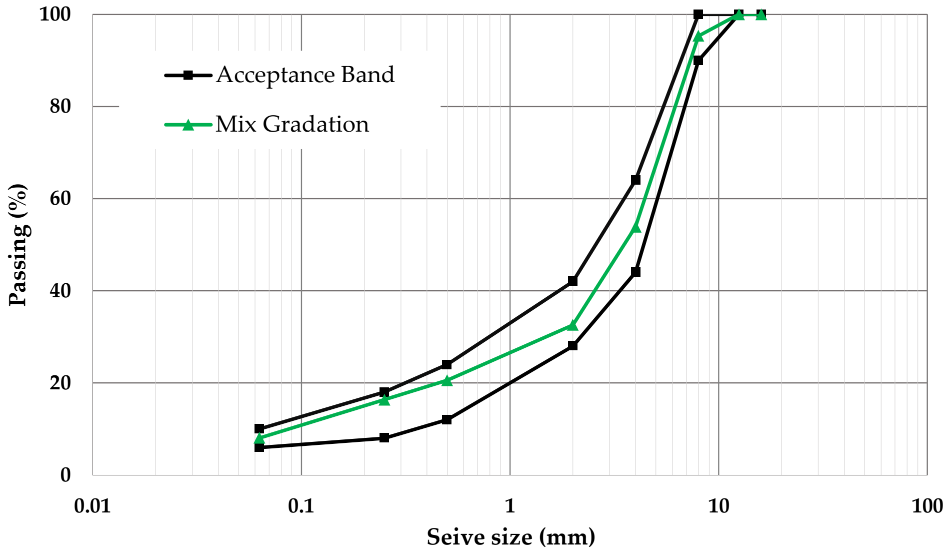

Figure 5. 4 Mix design gradation.

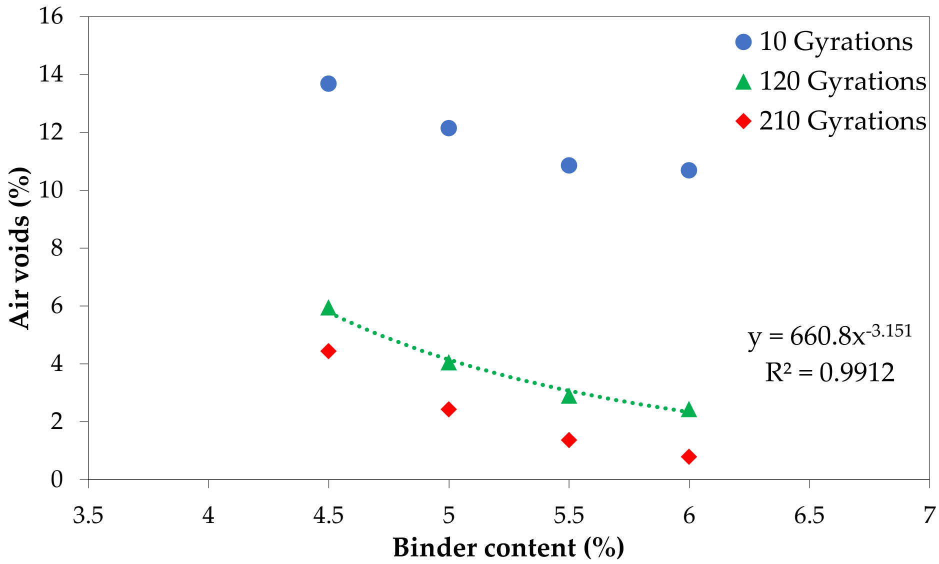

Figure 5. 5 Volumetric characteristics based on Superpave method.

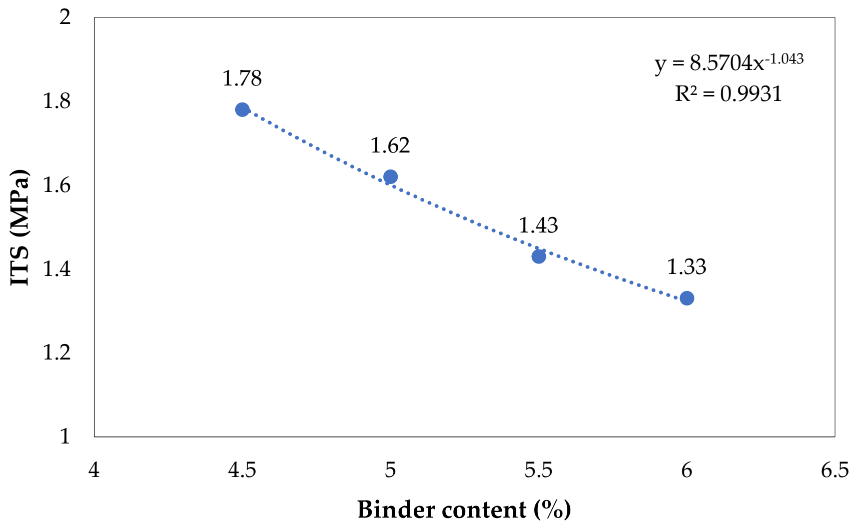

Figure 5. 6 ITS of control asphalt mixture with different binder content.

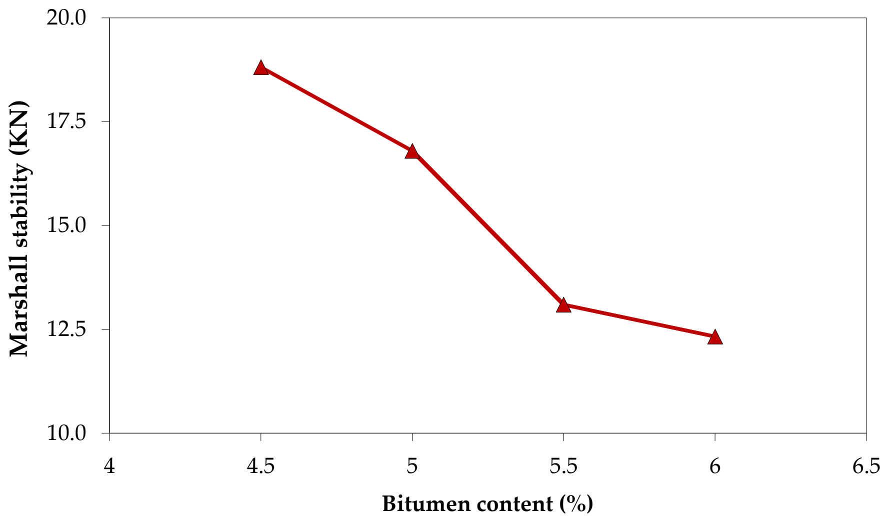

Figure 5. 7 Marshall Stability.

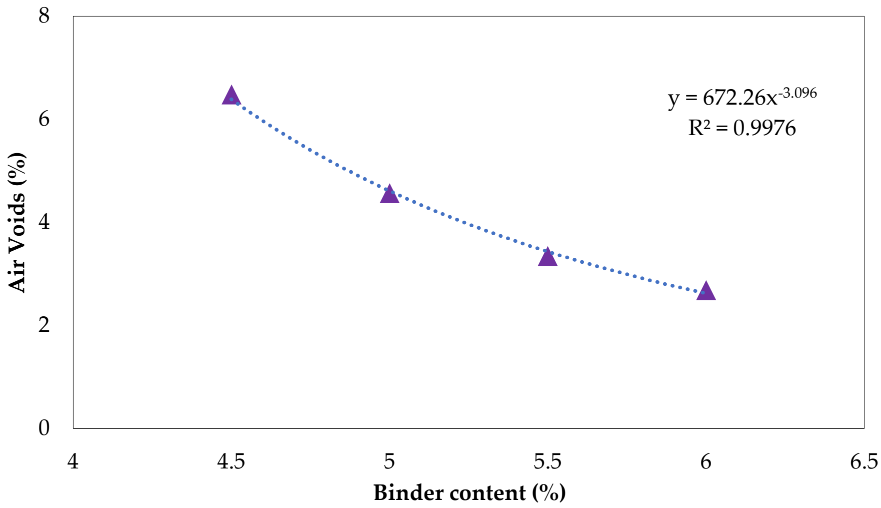

Figure 5. 8 Air voids content based on Marshall mix design.

Furthermore, for the mix design of the asphalt mixture containing recycled plastic asphalt modifier, 6% GRP (on the weight of the bitumen) was added to the mixture using the dry method, maintaining a total binder content similar to that used in the control mixture. During mixing, as shown in Figure 5.9, the GRP granules were added to hot aggregates and maintained in an oven at a temperature of 170 ± 5 °C for approximately 1 hour until they softened before adding hot bitumen maintained at 160 °C.

Figure 5. 9 GRP granules placed over the hot aggregates.

5.3.4 RAP manufacturing

Although evaluating the properties of in situ aged RAP is vital in any recycling study, due to the theme of the research, which deals with multiple recycling and controlling the consistency of the RAP, artificial RAP was manufactured. In addition, the introduced GRP modifier is a novel product, yet the corresponding RAP was not available. Therefore, such modified asphalt mixtures need to be produced in the laboratory to simulate the properties of RAP.

For this purpose, to produce RAP, both sets of asphalt mixtures, the control and testing asphalt mixtures, were artificially aged following the loose asphalt mixture ageing protocol based on the findings of Chapter 4. The unmodified asphalt mixture and the GRP-modified asphalt mixture were prepared according to their respective mix designs. After mixing, both asphalt mixtures were placed in separate trays with a maximum thickness of 25 mm. The trays were then placed in an oven at 135 °C for 4 hours to simulate short-term ageing. Subsequently, they were cooled down and kept at 95 °C for 5 days to simulate long-term ageing. This ageing technique was utilised because it can deliver a more accurate simulation of the ageing happening in the viscoelastic component of the asphalt mixtures, i.e., the binder, and thus provide an enhanced understanding of the ageing mechanisms. The same procedure was applied at each ageing cycle for producing the RAP of the recycled mixtures for the next recycling cycles. At the end of the ageing procedure, the aged binder was extracted by the standard binder extraction and recovery method. Tetrachloroethylene was the solvent used for the extraction, and the distillation was done by means of rotavapor.

5.3.5 Binder ageing

The properties of bitumen 50/70 were considered as the reference during the optimisation of the rejuvenator and the recycling process. As indicated in Figure 5.3, in order to have the aged reference properties, a widely accepted protocol that simulates short-term ageing and long-term ageing was also considered. The short-term ageing (STA) was performed using a Rolling Thin Film Oven (RTFO) according to EN 12607. Afterward, the long-term ageing of bitumen was carried out using a pressure ageing vessel (PAV) according to EN 14769. For this purpose, 50 g of STA binder was placed on a circular tray and kept in the PAV at a conditioning temperature of 100 °C with a pressure of 2.1 MPa for 20 hours.

5.3.6 Multiple recycling

The multi-recycling of the asphalt mixtures containing 50% RAP was simulated by using multiple ageing and recycling of the mixtures using the loose mixture ageing protocol. During each cycle of asphalt mixture recycling, 50% RAP from the prior cycle and 50% virgin materials were used. It is worth mentioning that since the study was developed at the binder scale, the extracted binders were recycled three cycles after each cycle of mixture ageing.

The required properties of the mixtures were achieved using the rejuvenator optimised for each cycle of recycling. The rejuvenator was added to the hot unaged (virgin) bitumen at 160 °C based on the weight of the aged binder content. The asphalt binder blend was then stirred manually for approximately 1 minute. Finally, to provide consistency and homogeneity, the blend was kept in the oven for 10 minutes. The rejuvenator dosage was optimised based on the equi-shear modulus temperature (T_{BTSV}) using the Binder Fast Characterization Test (BTSV), along with a complementary upper critical temperature (T_C) to ensure that the optimised binder's upper PG range falls within the target range. The innovative rejuvenation scheme used in the study is presented in Figure 5.10. In addition, the dosage was additionally controlled by other tests, such as standard penetration and softening point temperature, as a commonly used approach for the optimisation of the rejuvenators in Europe. For this purpose, initially, different dosages of rejuvenator were added to the extracted aged binders, and the blends were tested and compared with the reference bitumen values. Table 5.3 shows the optimized dosages of the rejuvenator used at each cycle of recycling.

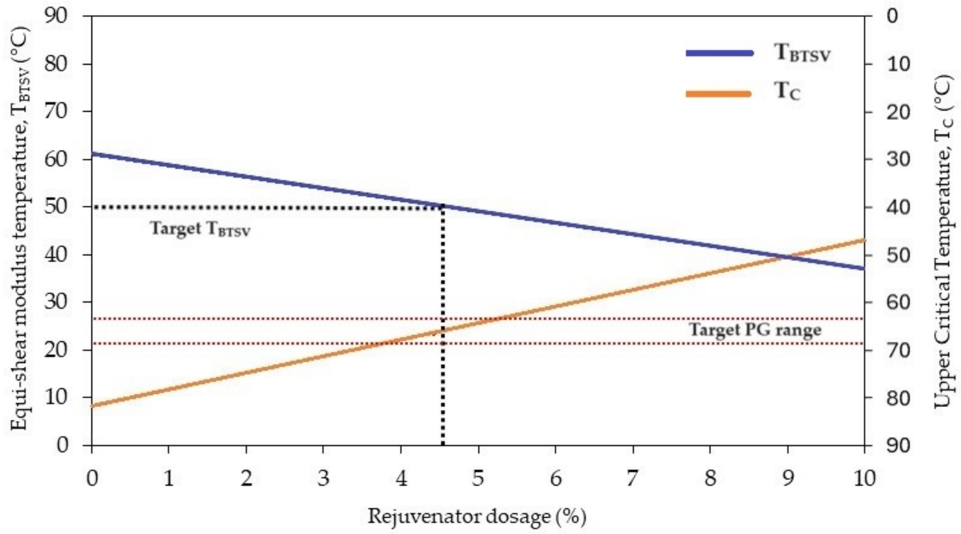

Figure 5. 10 Rejuvenation scheme used in the study.

Table 5. 3 The optimum dosage of the rejuvenator for each cycle of recycling.

Binders from the Control mix	Cycle of Rejuvenation	Optimum dosage* (%)	Binders from the Testing mix	Cycle of Rejuvenation	Optimum dosage * (%)
CA^0 + R	1	4.5	$GRPA^0$ + R	1	5.5
CA^1 + R	2	4.5	$GRPA^1$ + R	2	4.5
CA^2 + R	3	4.4	$GRPA^2$ + R	3	4.4
*The dosage of rejuvenator is on the weight of the aged binder					

5.3.7 Testing methods

According to the objectives of this research, the experimental plan shown in Figure 5.3 consisted of a full binder scale characterization by means of both conventional tests and complementarily rheological analysis using a dynamic shear rheometer (DSR). In addition to the physical and rheological analysis, the binders were chemically characterised using Fourier transform infrared (FTIR) spectroscopy. All the tests were carried out according to the European standard testing methods.

5.4 Results and discussion

5.4.1 Penetration test

During the penetration test, the average of five readings was considered to determine the penetration values, which are shown in Figure 5.11. According to the results of the penetration test, the binders extracted from both the long-term aged control asphalt mixture Mix-CA^0 and the long-term aged GRP modified mixture Mix-GRPA^0 have shown similar penetration values. These values were also similar to the reference aged (RTFO + PAV) Bit. 50/70. Although, during the first cycle of rejuvenation, different dosages were used (optimized), it can still be seen that the penetration values of the extracted binder from GRP-modified asphalt mixtures are comparable with the binder extracted from the control mixture and the reference Bit. 50/70. In the further cycles of ageing and recycling, the dosages used for rejuvenations were the same, and the obtained values were comparatively similar to the extracted binders from the control mixture and reference Bit. 50/70. Having such values, it can be stated that except for the first-round results that are impacted by the modification, the extracted binders of the further cycles are similar.

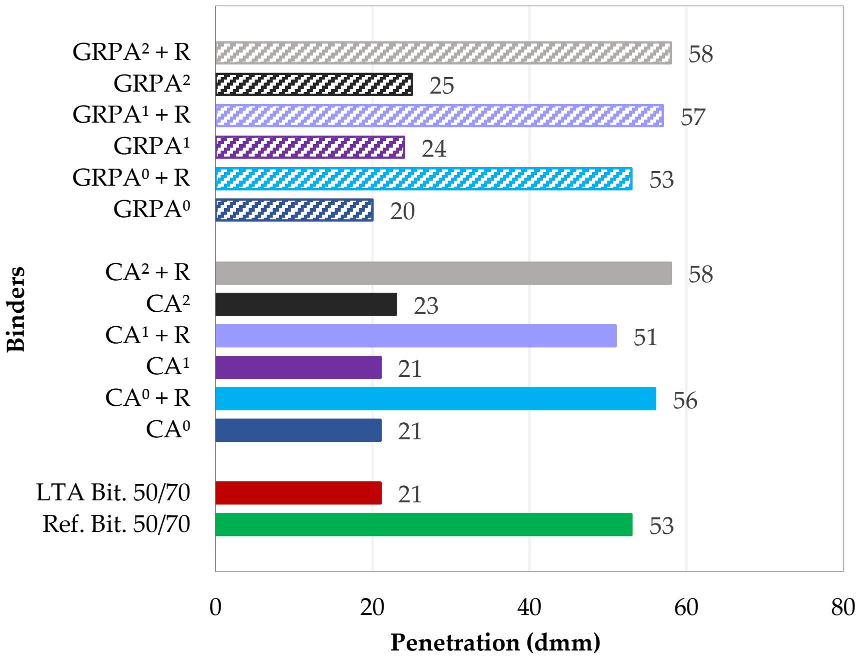

Figure 5. 11 Penetration values.

5.4.2 Softening point test

The results of the softening point test are summarised in Table 5.4. Considering the obtained values, the extracted binder from the long-term aged GRP-modified asphalt mixture, Mix-GRPA^0, showed a higher softening point compared to the extracted binder from the control mixture, CA^0, and the LTA Bit. 50/70. It is noteworthy that the higher softening point was also recorded for the binder extracted from the GRP-modified asphalt mixture immediately after the production, where 55.3 °C was obtained compared to 51.7 °C for the control mixture. The increase in the softening point of GRPA^0 was expected due to the addition of the GRP additive. This higher performance with the GRP additive, in fact, would allow a higher resistance to permanent deformation of the mixture. Considering the target of this research to bring back the properties to the original one, at the first cycle of recycling, a comparatively higher dosage of rejuvenator, shown in Table 5.3, was used for the binder GRPA^0. Accordingly, during the further phases of ageing and recycling, the used rejuvenator dosage (optimised dosage) and softening point values of the extracted binders were comparable. From another point of view, it was noted that the mixtures containing rejuvenator from the last cycle show less susceptibility to aging. This can be seen where, for both sets of the extracted binders, the softening point values are less than the cycle before.

Table 5. 4 Softening points of the binders.

Reference binders	Softening Point (°C)	Binders from the Control mix	Softening Point (°C)	Binders from the Testing mix	Softening Point (°C)
LTA Bit. 50/70	60.8	CA^0	60.8	GRPA^0	62.5
Ref. Bit. 50/70	49.6	CA^0 + R	49.7	GRPA^0 + R	50.2
LTA Bit.50/70	60.8	CA^1	60.6	GRPA^1	59.4
Ref. Bit. 50/70	49.6	CA^1 + R	50.3	GRPA^1 + R	49.3
LTA Bit. 50/70	60.8	CA^2	58.6	GRPA^2	58.5
Ref. Bit. 50/70	49.6	CA^2 + R	49.2	GRPA^2 + R	48.8

5.4.3 Dynamic viscosity test

According to the results shown in Figures 5.12 and 5.13, the extracted binders of both sets of testing and control asphalt mixtures showed the same trend after each ageing and recycling cycle. The trend recorded was very similar to the penetration and softening point tests. It is noteworthy that, as it was expected due to the presence of GRP, the binder extracted from the first round aged modified mixture, GRPA⁰, showed a higher viscosity compared to the control mixture. However, after the first cycle of recycling, the extracted binder from both control and GRP-modified mixtures performed comparably, and the same optimum dosage of the rejuvenator was optimised for recycling the aged binders at recycling cycles 2 and 3.

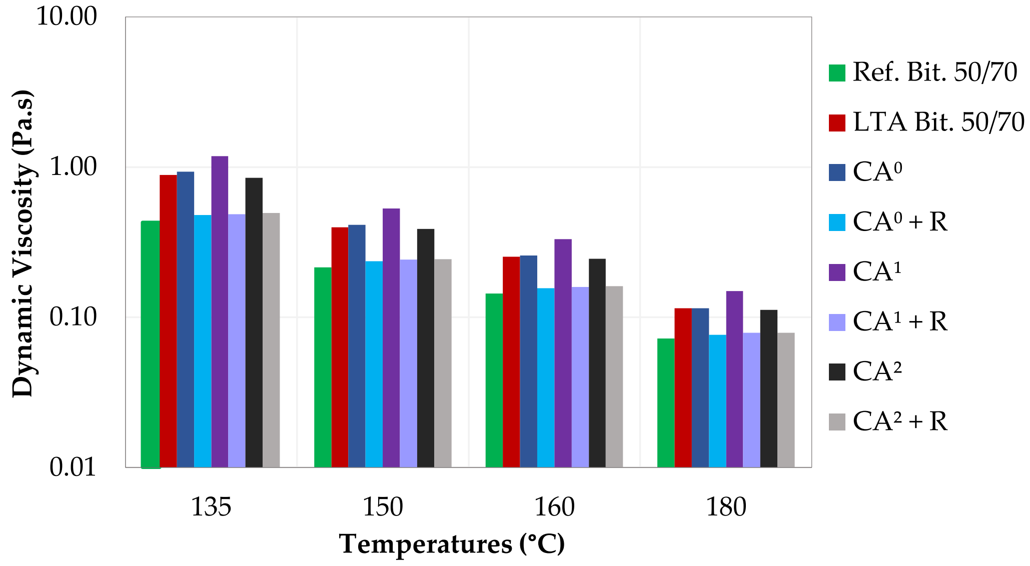

Figure 5. 12 Dynamic viscosity of the binders from the control asphalt mixture.

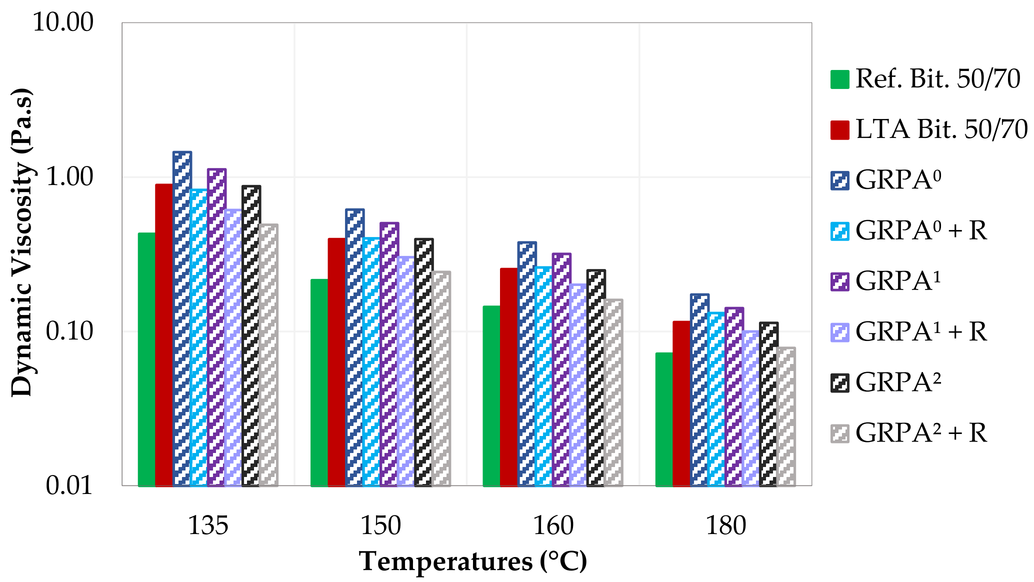

Figure 5. 13 Dynamic viscosity of the binders from the GRP-modified asphalt mixture.

5.4.4 Temperature Sweep test

Figures 5.14 and 5.15 display the loss factor curves of the binders, while Figures 5.16 and 5.17 illustrate the isochronal plots of the binders in terms of the complex shear modulus. According to the obtained results, the following observations can be drawn:

- As expected, both short-term and long-term ageing increased the stiffness of the reference Bit. 50/70; however, within the loss factor figure, the significance of short-term ageing is more visible compared to other testing methods.
- None of the recycled binders, neither the control mix binders nor the GRP mix binders were shown to have the same loss factor plots compared to the Ref. Bit. 50/70 virgin bitumen. It is worth mentioning that if a higher dosage of rejuvenator had been added during the recycling cycles, the binders would become excessively soft, and the corresponding mixtures would be susceptible to rutting. This fact is also attested by the loss factors at high temperatures, where the recycled binders showed similar loss factors at these temperatures.
- It is noteworthy that despite the difference between the recycled binders' loss factor curves and Ref. Bit. 50/70 at unaged (virgin) state, the aged binders showed very similar curves after long-term ageing with the PAV aged bitumen 50/70 (LTA Bit. 50/70).
- Regardless of the Ref. Bit. 50/70, as it was observed via other testing methods, the extracted binders from the control and the GRP-modified asphalt mixtures after the first cycle of recycling showed comparative performance.
- Not similar to the loss factor plots, the G* isochronal plots of the extracted binders from the aged/recycled control and testing asphalt mixtures were comparable to the reference bitumen 50/70 in its un-aged state. This could assure the accuracy of the optimised rejuvenator dosage.

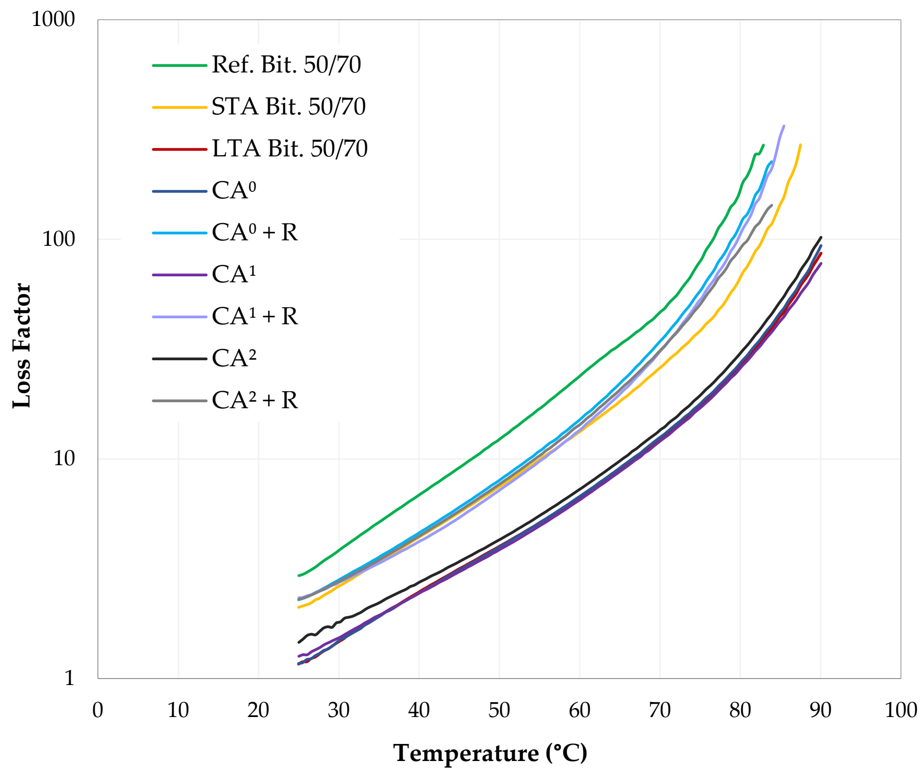

Figure 5. 14 Loss factor versus temperature curve of the binders from the control asphalt mix.

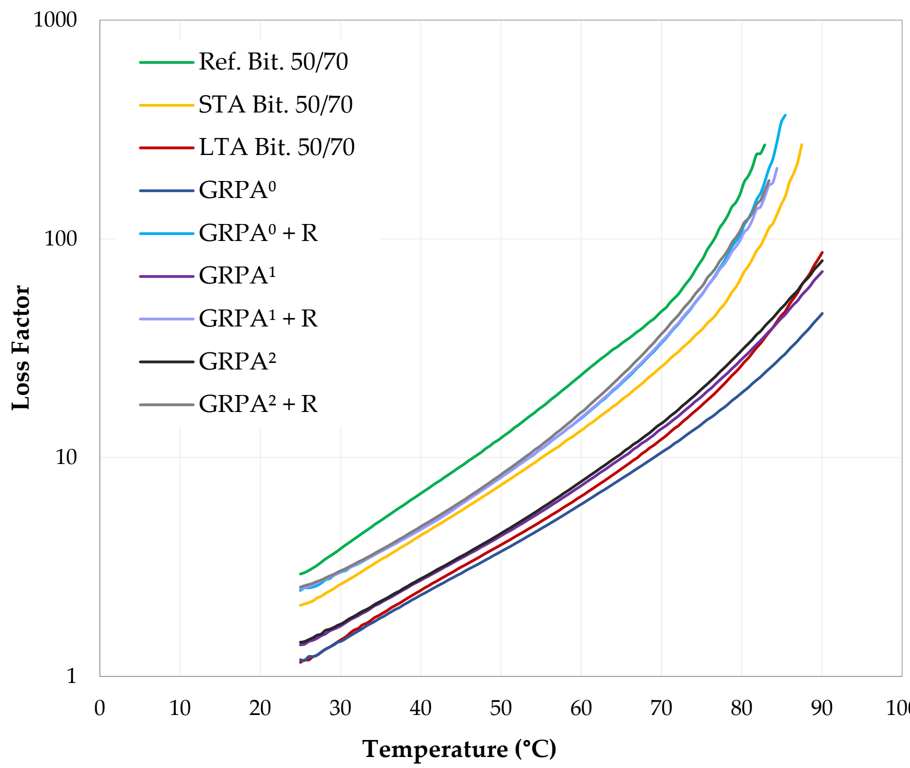

Figure 5. 15 Loss factor versus temperature curve of the binders from the GRP-modified asphalt mix.

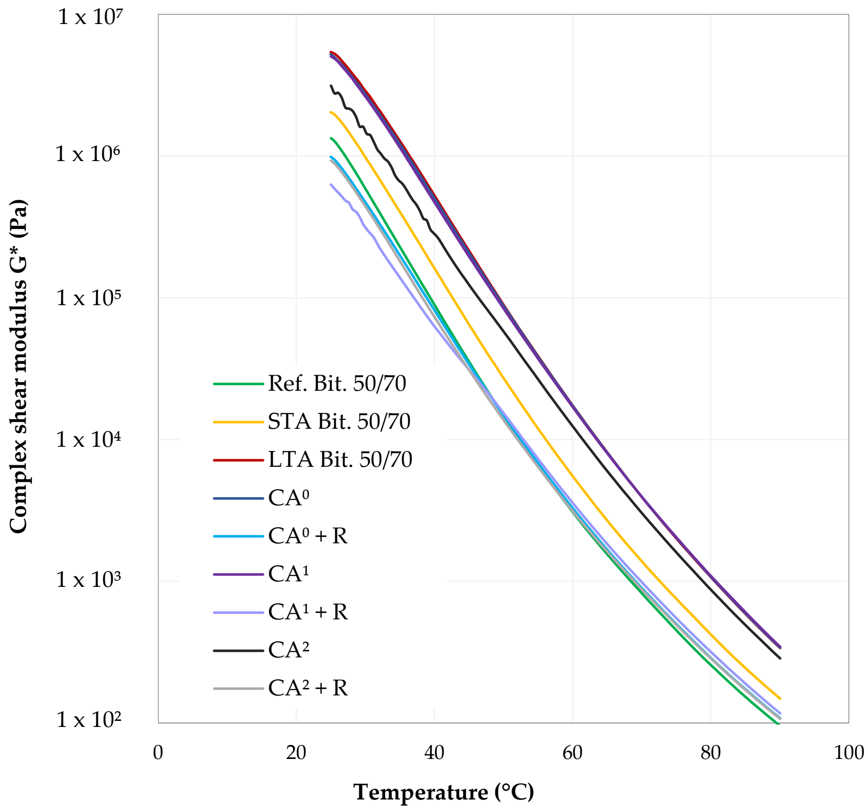

Figure 5. 16 Isochronal plots of the binders from the control asphalt mix.

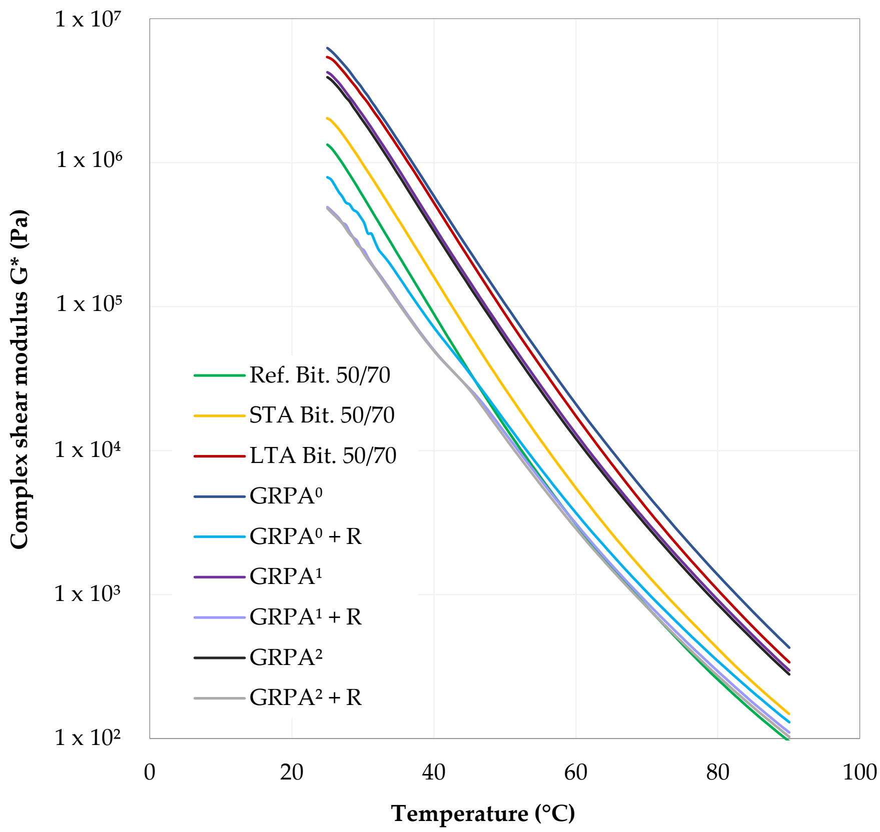

Figure 5. 17 Isochronal plots of the binders from the GRP-modified asphalt mix.

5.4.5 Upper Performance Grading

The upper-performance grading of the samples was tested to confirm the performance of the binders at high temperatures. The test results are summarized and compared in Table 5.5. According to the obtained results, the upper PGs complied with the trend obtained via softening points and loss factors. Similar to the other tests' results, the PG values showed that the presence of the GRP increased the upper PG value as a modifier. At each recycling cycle, the addition of the rejuvenator reduced the PG of the binders extracted from the control and the testing mixture to the target value of reference bitumen 50/70.

Table 5. 5 Upper Performance Grading.

Reference binder	Upper PG (°C)	Upper Critical Temp. (°C)	Binders from the Control mix	Upper PG (°C)	Upper Critical Temp. (°C)	Binders from the Testing mix	Upper PG (°C)	Upper Critical Temp. (°C)
LTA Bit.50/70	76	80	CA^0	76	80	$GRPA^0$	82	82.2
Ref. Bit. 50/70	64	67.5	CA^0 + R	64	68.5	$GRPA^0$ + R	70	70.6
LTA Bit.50/70	76	80	CA^1	76	80.1	$GRPA^1$	76	78.4
Ref. Bit. 50/70	64	67.5	CA^1 + R	64	69.4	$GRPA^1$ + R	64	68.0
LTA Bit.50/70	76	80	CA^2	76	78.2	$GRPA^2$	76	78.4
Ref. Bit. 50/70	64	67.5	CA^2 + R	64	68.3	$GRPA^2$ + R	48.8	68.2

5.4.6 Binder Fast Characterization test

The BTSV test results in terms of equi-shear temperature and phase angle are summarised in Table 5.6. As it can be seen, not only the equi-shear temperatures but also the respective phase angles obtained at the optimum dosage of the rejuvenator were comparable. This could also confirm the quality and right dosage of rejuvenator in recovering the rheological parameters that have been considered during the rejuvenator optimisation in this study. Moreover, the obtained equi-shear modulus temperature of all the binders was similar to their corresponding softening point values.

Table 5. 6 BTSV results.

Reference binder	T_{BTSV} (°C)	δ_{BTSV} (°)	Binders from the Control mix	T_{BTSV} (°C)	δ_{BTSV} (°)	Binders from the Testing mix	T_{BTSV} (°C)	δ_{BTSV} (°)
LTA Bit. 50/70	61.0	81.9	CA^0	60.9	82	$GRPA^0$	62.2	81.7
Ref. Bit. 50/70	49.9	85.3	CA^0 + R	49.9	82.8	$GRPA^0$ + R	50.4	83.2
LTA Bit. 50/70	61.0	81.9	CA^1	60.8	81.7	$GRPA^1$	59.1	81.9
Ref. Bit. 50/70	49.9	85.3	CA^1 + R	50.1	82.2	$GRPA^1$ + R	49.2	82.7
LTA Bit. 50/70	61.0	81.9	CA^2	58.7	81.6	$GRPA^2$	58.6	82
Ref. Bit. 50/70	49.9	85.3	CA^2 + R	49.4	82.3	$GRPA^2$ + R	48.7	82.7

5.4.7 Fourier Transform Infrared spectroscopy

The obtained spectra of all binders are shown in Figures 5.18 and 5.19. In addition, the calculated values of sulfoxide and carbonyl indices are summarised in Table 5.7. As shown in the literature, comparing the spectra of the reference bitumen unaged and LTA aged, it can be clearly seen how the oxidation is represented by increasing the peaks of the sulfoxide and carbonyl bands. This phenomenon was also observed for the binders of both control and testing mixtures to different extents. Adding the rejuvenators at each cycle of recycling could also restore some of the chemical properties, especially at the first cycle of recycling, where both oxidation representative bands were reduced compared to the original state. However, the same trend was not observed during all the recycling cycles. This could be due to, firstly, the chemical components and nature of the rejuvenators and the recycled plastic modifier, and secondly, the dosage of the rejuvenator, which has been reported in several studies. From another point of view, the significance of the rejuvenation impact was not the same for both sets of binders, but still, the same trend can be observed considering each cycle of ageing and rejuvenation. Overall, it is worth mentioning that for further information and quantifying the impact of the recycled plastic modifier, particularly the dry method, a further chemical investigation should be carried out, i.e., different dosages of the asphalt modifier, which was out of the scope of this research.

Table 5. 7 Calculated values of sulfoxide and carbonyl indices.

Binders	Sulfoxide Index (SI)	Carbonyl Index (CI)
Ref. Bit. 50/70	0.16	0.08
LTA Bit.50/70	0.58	0.36
CA^0	0.60	1.13
$CA^0 + R$	0.15	0.08
CA^1	0.36	0.19
$CA^1 + R$	0.31	0.26
CA^2	0.29	0.59
$CA^2 + R$	0.25	0.62
$GRPA^0$	0.36	0.26
$GRPA^0 + R$	0.33	0.21
$GRPA^1$	0.31	0.37
$GRPA^1 + R$	0.19	0.41
$GRPA^2$	0.23	0.32
$GRPA^2 + R$	0.23	0.34

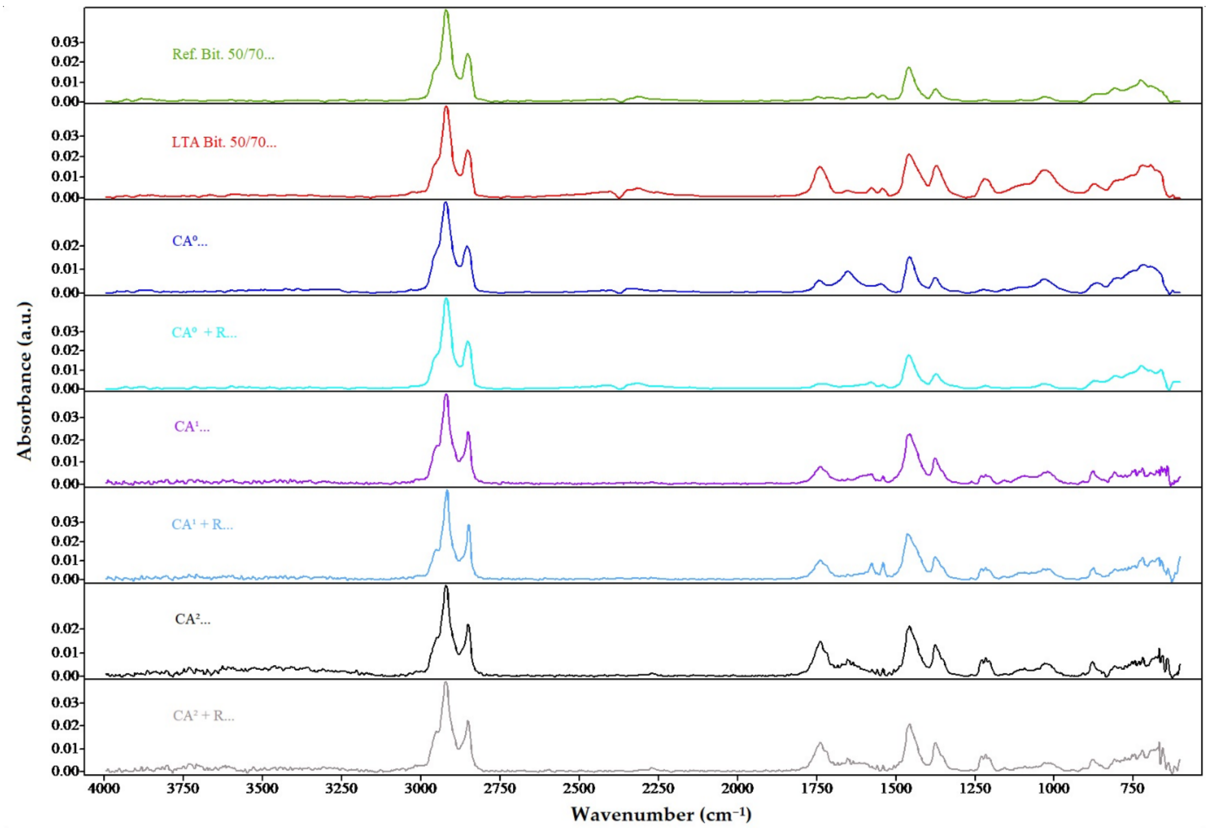

Figure 5. 18 Average obtained spectra of binders extracted from the control asphalt mixtures.

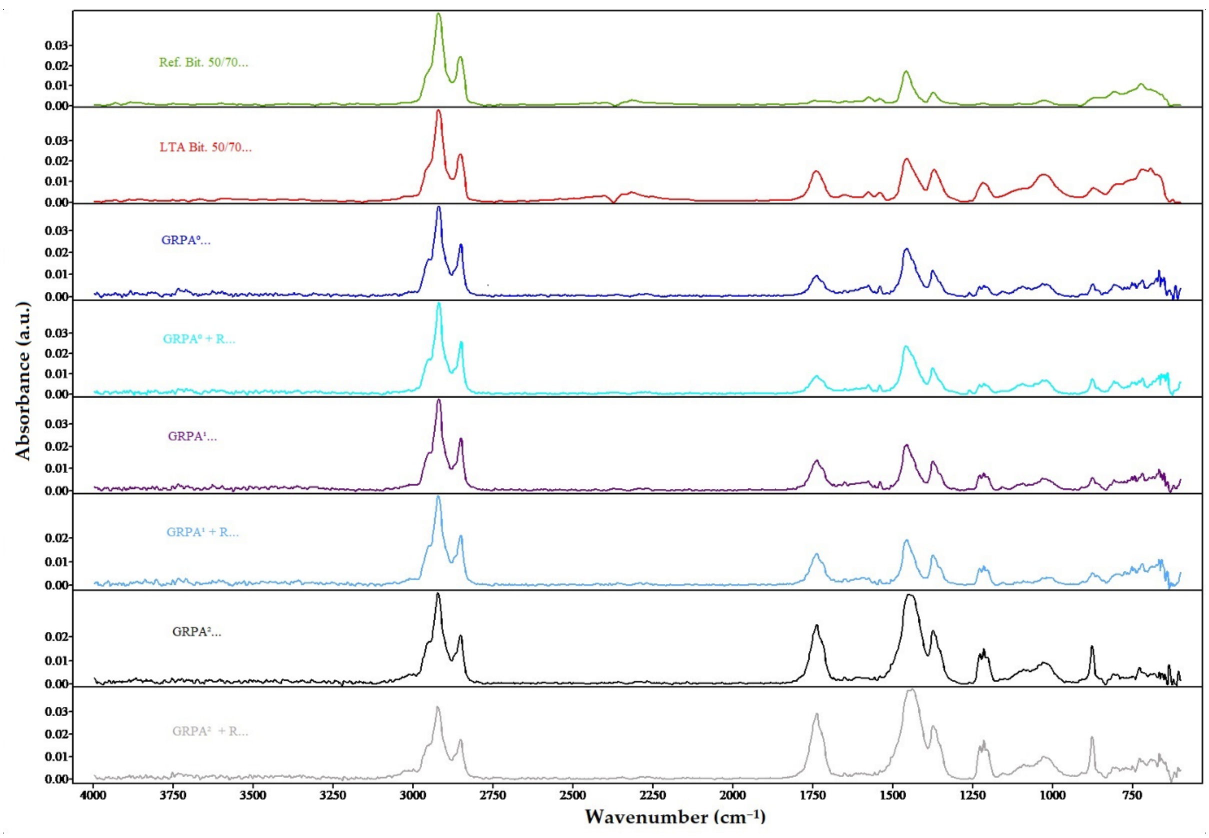

Figure 5. 19 Average obtained spectra of binders extracted from the GRP-modified asphalt mixtures.

5.5 Summary and Conclusions

In this study, the potential multi-recyclability of asphalt mixtures was investigated via binder-scale characterization, where three sets of data were compared and analysed. These three sets comprise i) a reference bitumen 50/70, ii) the extracted binders from a control mixture without any asphalt modifier, and iii) the extracted binders from a mixture containing recycled plastic asphalt modifier. The recycling rate was 50%, and the mixtures were designed and optimised based on the Italian specifications for a dense-graded wearing course. The study was structured to include three recycling cycles of the aforementioned materials. In this case, analysing the obtained results, it can be seen that, as a modifier, the recycled plastic asphalt additive increases the softening point, upper PG, and equi-shear modulus temperature. However, in this study, it should be considered that since the method of modification was the dry method (process), the asphalt mixture mixing and manufacturing process could also impact the mentioned properties. Moreover, at the first cycle of recycling, a slightly higher dosage of rejuvenator was found to be optimal for the binder extracted from first-cycle aged GRP-modified asphalt mixtures. This was because the modifier enhanced the properties of the mixture. During the following cycles, the same dosages of rejuvenator were found to be optimal for both control and testing mixtures. Furthermore, the rheological analysis has shown that optimising the rejuvenator is a critical factor in asphalt recycling, especially for mixtures with high RAP content. The non-optimal dosage can lead to an adverse effect on the properties of the asphalt binder and the produced asphalt mixture. Accordingly, a complete rejuvenator optimisation should encompass the properties of bitumen from different aspects and at different temperatures. In addition to the physical and rheological properties, the extracted binders with and without the modifier, at different cycles of ageing and rejuvenation, were chemically characterised by means of FTIR spectroscopy. Although the technique provided a clear image of the ageing impact on the reference bitumen, the same impact was not observed at all three cycles of rejuvenation for the extracted binders, but the influence of the rejuvenator was clearly observed by the reduction in the carbonyl and sulfoxide bands. Finally, the binders corresponding to the reclaimed asphalt containing the introduced recycled plastic modifier showed similar or comparable properties to the binders of the control mixtures without asphalt modifier. This has been recorded at all three recycling cycles of the extracted binders. Overall, through the obtained results and analysis, it is possible to immediately understand that the partial substitution of virgin materials with recycled ones can provide end products that have similar, or in some cases, depending on the investigated property, better performance than the conventional ones.

Now, bearing in mind the importance of the sustainability and circular economy implications of the pavement engineering sector, the relevance of this study becomes obvious. The pavement engineering industry exploits tremendous amounts of natural, non-renewable resources during the extraction of the materials, their transport, the production of binders, and, of course, asphalt mixtures. The impacts even extend up to the construction of pavements and eventually their end of life. Keeping this in mind, and in order to shift towards a more circular and sustainable way of conducting business, the potential multi-recycling of materials is of paramount importance. It is worth reiterating that in this study, a novel ageing technique was utilised because it can deliver a more accurate simulation of the ageing happening in the viscoelastic component of the asphalt mixtures, i.e., the binder, and thus provide an enhanced understanding of the ageing mechanisms. Hence, overall, it is proven that for the asphalt binders investigated, it is possible to expect at least 3 life cycles of service life with a performance that falls well inside the acceptable window of the performance of conventional binders. In other words, any asphalt binder can have a tremendous potential for multi-recyclability, given that the optimal amount and appropriate type of rejuvenator are selected and used according to a proper rheological characterization. This paradigm can enable a plethora of opportunities for the involved stakeholders and the relevant decision-makers towards the actual implementation of the circular economy by cutting down on the usage of virgin materials, transport costs, and distances, and allowing a product to be present in the market for a longer period of time without losing its value or performance. Obviously, this is a lab-scale investigation, proving the ability to recycle conventional and plastic-modified asphalt mixtures for at least 3 life cycles based on a binder-scale investigation without any mechanical performance degradation.

Chapter 6

Multi-recyclability study: From a mixture-scale perspective

6.1 Introduction

The global shift towards a circular economy (CE) is driven by its regenerative growth model, aimed at addressing environmental, climatic, and economic challenges. Its goal is to expedite the transition from the traditional linear economic model to a CE that ensures resource availability while minimising waste generation through sustainable practices [29], [152]. In particular, CE promotes the recirculation of materials and energy within the same (closed-loop) or alternative product system (open-loop) [153]. The significance of the CE in reflecting on society's development is also evidenced by the activities and outcomes of the technical committee of ISO/TC 323 "Circular Economy" [190]. Furthermore, the implementation of circular economy principles and sustainability in asphalt pavement construction has prompted exploring cost-effective and eco-friendly alternatives. Asphalt recycling is one such promising alternative, as the use of reclaimed asphalt pavement (RAP) in new asphalt production reduces the demand for non-renewable resources, energy consumption, environmental pollution, waste accumulation, and production costs [15], [41], [153]. However, technical issues concerning production, mechanical performance, and durability have restricted the high rate of RAP incorporation in asphalt mixtures, even though it is beneficial [10].

Besides, unpredictable weather conditions and increased traffic volume have influenced the service life and performance of asphalt pavements, leading to pavement deterioration such as rutting, fatigue cracking, and thermal cracking. Consequently, polymer modification has been adopted as a feasible approach to address these premature pavement distresses [12]. It involves the integration of various polymers into either asphalt binders or mixtures to enhance its properties, such as durability, elasticity, resistance to ageing and deformation, thus contributing to the overall quality and extended service life of the pavement structure [90], [91]. In asphalt modification, polyethylene (PE), ethylene-vinyl acetate (EVA), polypropylene (PP), styrene-butadiene-styrene (SBS), styrene-isoprene-styrene (SIS), styrene-butadiene-rubber (SBR), and styrene-ethylene/butylene-styrene (SEBS) are the most widely employed polymers [191]. The application of these polymers has the potential to improve either the mechanical performance of the asphalt concrete or the rheological characteristics of the asphalt binder, depending on their type and method of application [22]. However, the cost of polymers and their modification methods can be a significant concern, as high-quality polymers and their modification processes tend to be more expensive, thereby contributing to the overall expenses of road construction [98].

Meanwhile, the increasing concern over plastic waste generation, estimated at approximately 300 million metric tonnes annually worldwide, along with its inherently low biodegradability, has prompted the exploration of innovative applications for effectively repurposing plastic waste [17], [18], [192]. Consequently, asphalt practitioners and researchers have shifted their emphasis towards incorporating waste plastic into asphalt concrete, thereby addressing the issue of plastic waste to some extent while also enhancing the performance of asphalt concrete through plastic modification. The recycled plastics can be incorporated into asphalt mixtures via two methods: the wet method and the dry method [193]. In the wet method, recycled plastic is directly introduced to the bitumen and blended with the aid of a high-shear mixer, typically at high temperatures ranging from 160 to 180 °C. The rheological properties of the bitumen can be improved using a typical dosage of 1–8% recycled plastic, based on the weight of the bitumen, and thereby it could enhance the pavement performance at high, intermediate, and low temperatures [19], [109]. Several studies indicated that the addition of recycled polyethylene terephthalate (PET) via wet method can increase the rutting resistance, bonding strength [123], moisture susceptibility, low temperature properties [124] and cracking resistance [125]. In addition, binder modification with low density-polyethylene (LDPE) has showed an enhanced impact on workability, resistance to rutting, antistripping and adhesive strength [126], [127]. However, this approach of modification is limited to recycled plastics with a considerably low melting point. Some studies reported that although adding recycled plastics enhanced binder qualities, some, such as PVC, could emit harmful chloride emissions, making them unsuitable for binder modifications [128].

While in the dry method, recycled plastic can be introduced directly to preheated mineral aggregates prior to being mixed it with bitumen in the production of asphalt concrete. On the basis of the characteristics and type of recycled plastic, such modifications can be served as either a partial substitute for aggregate or as a modifier for asphalt binder or mixture [14]. It has been stated that incorporating recycled plastic using the dry method could also effectively enhance the mechanical characteristics of asphalt concrete. There are several recycled plastics being incorporated using this approach despite of their melting point [130]. For instance, several studies indicated that the addition of recycled polyethylene via the dry method to the asphalt mixture showed improvements in compactability, rutting resistance, and fatigue cracking resistance [21], [134], [194]. In a study, two different recycled plastic modifiers were studied using the dry method. The findings of their study showed that the stiffness modulus, rutting resistance, and fatigue cracking properties of both the asphalt mixtures modified with recycled plastics were similar to those of the reference SBS-modified mixture [132]. From an environmental perspective, recycled plastic incorporation raised a concern over health and safety of the road workers. In response to this, Boom et. al conducted a study investigating the release of carcinogenic compounds from asphalt modified with recycled plastic additives. The findings of their study suggested that recycled plastic-modified asphalt reduces the non-carcinogenic and carcinogenic risk compared to the conventional asphalt [136].

The potential recyclability of plastic-modified asphalt is worth investigating from the perspective of a circular economy. In this regard, Lu et al. studied the recyclability of plastic-modified asphalt mixtures without a rejuvenator and found that asphalt mixtures containing 30% RAP from plastic-modified asphalt exhibited no detrimental effects on rutting and cracking performances [24]. However, the increasing prevalence of incorporating waste plastics into asphalt as modifiers necessitates further research to better comprehend the potential recyclability of pavements modified using such alternatives.

6.2 Scope and Objectives of the chapter

The literature extensively explored the efficiency of various recycled plastics in enhancing the rheological and mechanical performance of asphalt binders and mixtures. However, certain aspects, such as the characteristics of RAPs containing recycled plastics and their recyclability, are some of the missing information that needs to be addressed. It is crucial to gain a comprehensive understanding of RAP-containing recycled plastic modifiers, considering the increasing trend of incorporating recycled plastic as an asphalt modifier and the significance of RAP recycling. Currently, limited research exists on this topic; meanwhile, the multi-recyclability of asphalt mixtures has not been properly investigated. Therefore, the present study focuses on investigating the feasibility of multi-recycling the asphalt mixtures, with and without a recycled plastic modifier, at a 50% recycling rate for two recycling cycles using a comprehensive volumetric and mechanical performance evaluation.

6.3 Materials and methods

6.3.1 Materials

In this study, limestone aggregates in various fractions were used based on local availability. It was observed that these aggregates are commonly utilised for highway construction in the southern region of Italy. Table 6.1 summarises the characteristics of the aggregates and the particle size distribution of the aggregates can be found in Appendix A. Moreover, in this research, a paving grade bitumen with a penetration of 50/70 was utilised. Table 6.2 provides some of the physical properties of the bitumen.

Table 6. 1 Characteristics of the aggregates.

Properties	Unit	Value	Standard
Apparent density of aggregates	g/cm³	2.80	EN 1097-6
Resistance to fragmentation	%	20	EN 1097-2
Flakiness index	%	7	EN 933-3
Sand Equivalent	%	88	EN 933-8
Polished stone value	PSV	46	EN 1097-8

Table 6. 2 Properties of the bitumen.

Properties	Unit	Value	Standard
Density at 25 °C	g/cm³	1.03	EN 15326
Penetration	dmm	53	EN 1426
Softening point	°C	49.6	EN 1427
Dynamic viscosity at 135 °C	Pa.s	0.43	EN 13302
Fraass breaking point	°C	-6	EN 12593

Furthermore, the study used an innovative recycled plastic modifier, as shown in Figure 5.1 in the previous chapter, to modify the asphalt mixture. The modifier, referred to as graphene-enhanced recycled plastic (GRP), was already described in detail in the previous chapter, which comes in pellet form, typically ranging from 3 to 5 mm in size. In this study, a bio-based commercial rejuvenator, referred to as R, was used as the recycling agent, chosen based on its proven performance in previous research. The technical properties of the rejuvenator were already described in the previous chapter.

6.3.2 Experimental plan

The experimental plan aimed to assess the feasibility of multi-recycling the asphalt mixture modified with recycled plastic, as compared to the conventional mixture. Figure 6.1 presents the experimental plan for this research, which included two sets of asphalt mixtures: unmodified and GRP-modified asphalt mix. The study focused on evaluating the recyclability of these mixtures, at a recycling rate of 50% for two recycling cycles through comprehensive volumetric and mechanical performance characterization. The abbreviations used for the studied mixtures, along with their descriptions, are provided below:

- **CA-0:** unmodified asphalt mixture (conventional asphalt mix), consisting of 100% virgin materials.
- **CA-1:** First-cycle recycled mixture of the conventional mix, consisting of 50% RAP of CA-0 and 50% virgin materials, along with a rejuvenator.
- **CA-2:** Second-cycle recycled mixture of the conventional mix, consisting of 50% RAP of CA-1 and 50% virgin materials, along with a rejuvenator.
- **GRPA-0:** asphalt mixture modified with recycled plastic, consisting of 100% virgin materials and GRP modifier.
- **GRPA-1**: First-cycle recycled mixture of the recycled plastic-modified asphalt mix, consisting of 50% RAP of GRPA-0 and 50% virgin materials, along with a rejuvenator.
- **GRPA-2:** Second-cycle recycled mixture of the recycled plastic-modified asphalt mix, consisting of 50% RAP of GRPA-1 and 50% virgin materials, along with a rejuvenator.

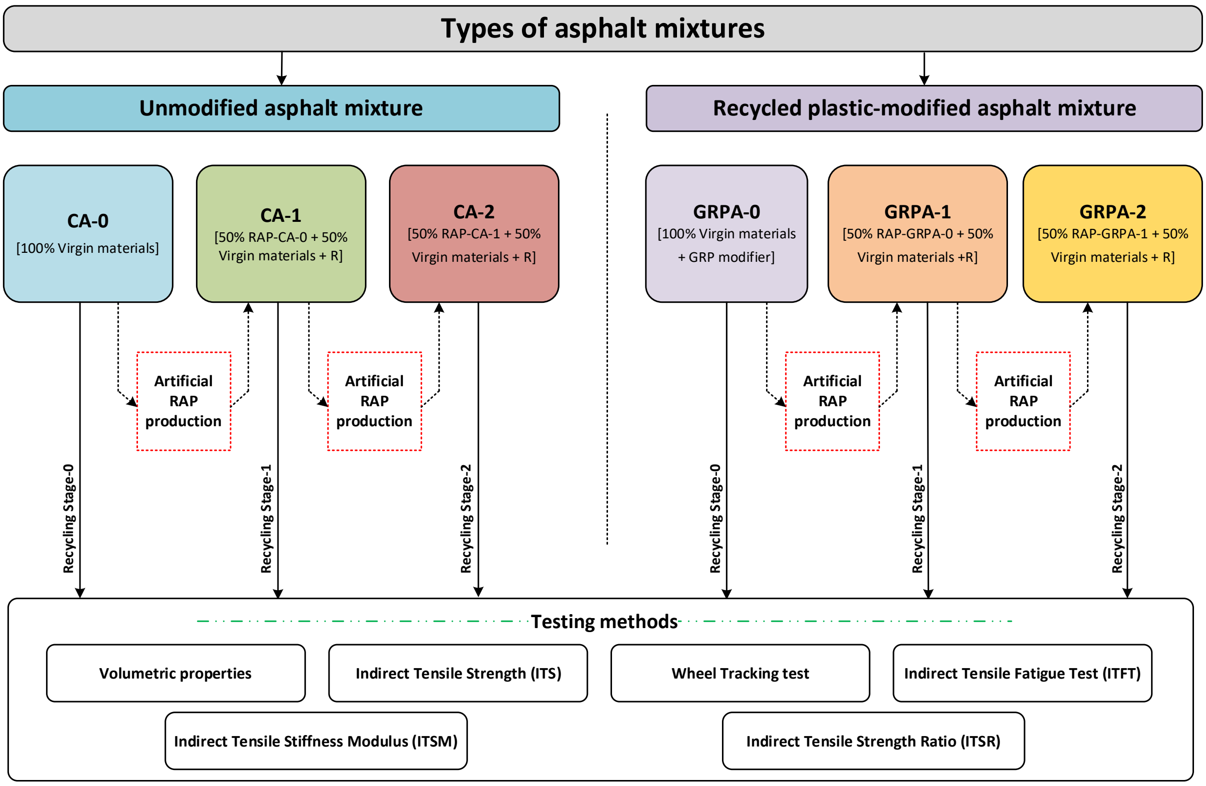

Figure 6. 1 Experimental plan.

6.3.3 Mix design and sample preparation

The mix design for unmodified and GRP-modified asphalt mixtures was carried out in accordance with the Italian technical specifications for highways [174]. In this study, both asphalt mixtures were prepared for a dense-graded wearing course with a nominal maximum aggregate size of 12.5 mm. Figure 6.2 presents the grading distribution curve of the designed mix. For the unmodified asphalt mixture, the binder content was optimised based on the Superpave volumetric method, which resulted in an identified binder content of 5% by weight of the aggregates. The specimens prepared using the gyratory compactor demonstrated 4 ± 0.5% air voids at 120 gyrations, adhering to the recommended limit prescribed by the technical specification.

Meanwhile, the recycled plastic-modified asphalt mixture was prepared using the same mix gradation as previously stated. The GRP modifier was incorporated into the asphalt mixture via the dry method; specifically, 6% GRP modifier by weight of bitumen was added to the hot aggregates approximately 1 hour prior to the mixing phase with bitumen. This process enables the recycled plastic granules to soften, enhancing the workability of the mixture. Furthermore, a total binder content of 5% by weight of aggregates was used for producing the GRP-modified asphalt mixture, and a mixing temperature of 170 ± 5 °C was employed based on the softening point of the GRP granules. The detailed descriptions of the mix design and sample preparation for recycled mixtures are reported in the subsequent section.

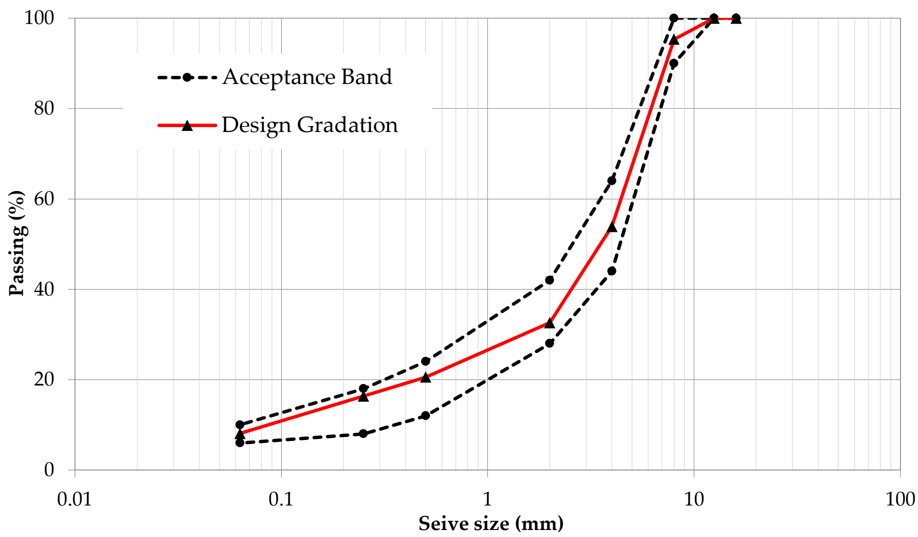

Figure 6. 2 Grading distribution curve of the mix.

6.3.4 Artificial RAP production

Considering the crucial aspect of this research regarding the recyclability of recycled plastic-modified asphalt mixtures, it is essential to study the characteristics of the RAP coming from plastic-modified asphalt pavements. In this study, an innovative recycled plastic, specifically the GRP modifier, was utilised for the modification of the asphalt mixture. However, RAP sources from GRP-modified asphalt pavements were unavailable, so the study necessitated manufacturing such RAP within the laboratory. Moreover, this research utilised the dry method approach for incorporating the GRP modifier into the asphalt mixture, as there is no established standard protocol for ageing loose asphalt mixtures. Although multiple factors influence the ageing process of asphalt concrete, thermo-oxidative ageing is one of the predominant factors among them. Therefore, this study employs a loose asphalt mixture ageing protocol based on the findings of Chapter 4 to simulate the effects of ageing and the end-of-life scenarios on both the asphalt mixtures, unmodified and GRP-modified, to provide a comprehensive understanding of their characteristics.

In order to prepare the RAPs, the unmodified asphalt mixture and the GRP-modified asphalt mixture were produced according to their defined mix design. After the mixing process, these two asphalt mixtures were placed in separate trays with a maximum thickness of 25 mm. The trays were then kept in the oven at 135 °C for 4 hours to simulate the short-term ageing. Afterwards, the trays were taken out to cool, and to simulate the long-term ageing effect, the trays were again kept at 95 °C for 5 days in the oven. The procedure was repeatedly applied to produce RAPs of both the recycled mixtures for their further recycling stages.

6.3.5 Simulation of multiple recycling

The research comprehensively evaluates the viability of recycling asphalt mixtures, with and without a recycled plastic modifier, to ascertain if these mixtures can be effectively recycled multiple times in a comparable manner. To achieve this goal, as shown in Figure 6.1, a systematic approach was formulated to assess their recyclability, which involves multiple ageing and recycling phases with a consistent recycling rate of 50% at each cycle. Initially, both asphalt mixtures were prepared according to their defined mix designs. Subsequently, the subsets of these mixtures were subjected to the accelerated ageing process explained in the former section to produce their respective RAPs. The recycling process involves systematically using 50% RAP from the preceding cycle along with 50% virgin materials (aggregates and bitumen) to produce their subsequent recycled mixtures. For instance, GRPA-1, the first recycling cycle for the plastic-modified asphalt mixture, was produced by combining 50% virgin materials with 50% RAP derived from the GRPA-0 mix, along with an optimised rejuvenator dosage. The process is repeated for its second recycling cycle, where, in this case, the recycled mixture is composed of 50% virgin materials and 50% RAP derived from the GRPA-1, along with an optimised dosage of rejuvenator. The same methodology was applied to recycle the conventional mixture as well.

The mix design for all the recycled mixtures, regardless of the recycling cycle, was kept similar to that of the conventional asphalt mix. This approach was chosen for comparative analysis with the conventional asphalt mix and to better understand the activation level of aged binders in the RAPs during recycling. Consequently, an optimal binder content of 5% by weight of the aggregates was maintained for all recycled mixtures at both cycles. Considering a 50% recyclability rate, 2.5% virgin bitumen (based on the weight of the total mixture) along with an optimised dosage of rejuvenator was added at each stage of recycling under the assumption that the remaining portion of binder from RAP would fully activate upon the mixing phase and blend with bitumen. Furthermore, before the mixing phase, the optimised rejuvenator dosage was added to the pre-heated virgin bitumen at 160 °C. Subsequently, the bitumen-rejuvenator blend was manually stirred for about 1 minute and then placed in an oven at the mixing temperature of 160 °C for 10 minutes to ensure consistency. Table 6.3 summarises the optimised dosages of rejuvenator used in the study to attain the targeted characteristics for the recycled mixtures at each cycle, based on the findings from the previous chapter.

Table 6. 3 The dosage of rejuvenator at each recycling cycle.

Recycled mixture	**Recycling cycle**	**Optimum dosage***	**Recycled mixture**	**Recycling cycle**	**Optimum dosage ***
CA-1	1	4.5%	GRPA-1	1	5.5%
CA-2	2	4.5%	GRPA-2	2	4.5%

*The dosage of rejuvenator is on the weight of the aged binder in RAP

6.3.6 Testing methods

The test methods used in the study were performed in accordance with the European standard testing methods and adhered to the Italian technical specification for highways. The following laboratory tests were conducted to achieve the goal of this study:

- Volumetric properties (EN 12697-8)

- Indirect Tensile Strength (ITS) at 25 °C, to evaluate the strength and cohesion of the mixture (EN 12697-23)
- Indirect Tensile Stiffness Modulus (ITSM), to assess the stiffness modulus of the mixtures at temperatures of 10 °C, 20 °C, and 30 °C (EN 12697-26)
- Indirect Tensile Strength Ratio (ITSR), to evaluate the moisture susceptibility of the mixtures (EN 12697-12)
- Wheel Tracking test at 60 °C, to assess the rutting resistance of the mixtures (EN 12697-22)
- Indirect Tensile Fatigue Test (ITFT), to evaluate the fatigue resistance of the mixtures at 20 °C (EN 12697-24).

6.4 Results and discussion

6.4.1 Volumetric properties

The volumetric properties of the asphalt mixtures were evaluated in accordance with EN 12697-8 guidelines [142]. To achieve this, three specimens with a diameter of 100 mm from each mixture were compacted using a gyratory compactor up to 210 gyrations, as stipulated by the Italian technical specification. As per the technical specifications, determining air voids content at specific gyration levels such as 10, 120, and 210 is crucial for approving mixtures intended for use on highways. Moreover, other volumetric parameter such as voids in mineral aggregates (VMA), voids filled with binder (VFB) and the bulk density (G_{mb}) of each mixture were also studied.

Table 6.4 provides a summary of the determined volumetric characteristics of the asphalt mixtures. The obtained results indicate that all the studied mixtures comply with recommended technical limits, affirming the validity of the mix design. It is worth noting that, at 120 gyrations, all mixtures exhibited an air voids content within a range of 4–5%. The data obtained from the compaction process was used to create the densification curves for the asphalt mixtures, depicted in Figure 6.3, indicating the relationship between the applied number of gyrations and the relative density of the compacted specimen. In this analysis, a slightly higher air voids content was observed in the curve of the recycled plastic-modified mixture (GRPA-0) compared to the conventional one. However, this trend did not persist across subsequent cycles of recycling, as evidenced by GRPA-1 and GRPA-2. The densification curve significantly illustrates the compaction trend of all the mixtures, and from that, it is clear that the dry method modification with recycled plastic did not adversely affect the compactability of the mixture. Notably, all the recycled mixtures at both recycling cycles demonstrated comparable densification curves with those of the conventional mix, indicating that the rejuvenator reactivates the aged binder during mixing phases and leads to workability comparable to that of a conventional mix. In addition, the mixture GRPA-0 showed a slightly lower bulk density among others, as the incorporation of the modifier lowers the density of the modified mixture. However, other volumetric parameters, such as VMA and VFB, of all the studied mixtures are within the acceptable threshold, ensuring good volumetric performance.

Table 6. 4 Volumetric properties of the gyratory compacted specimens.

Mixture	**Air voids content** (%)			**VMA**	**VFB**	**G_{mb}**
	10 gyrations	120 gyrations	210 gyrations	(%)	(%)	(kg/m³)
CA-0	13.3	4.4	2.6	15.8	72.0	2461
CA-1	13.5	4.2	2.2	16.2	74.0	2467
CA-2	13.1	3.9	2.1	15.9	75.1	2476
GRPA-0	13.7	5.1	3.3	17.8	71.4	2452
GRPA-1	13.6	4.3	2.4	16.2	73.7	2465
GRPA-2	13.0	4.1	2.1	16.0	74.4	2462

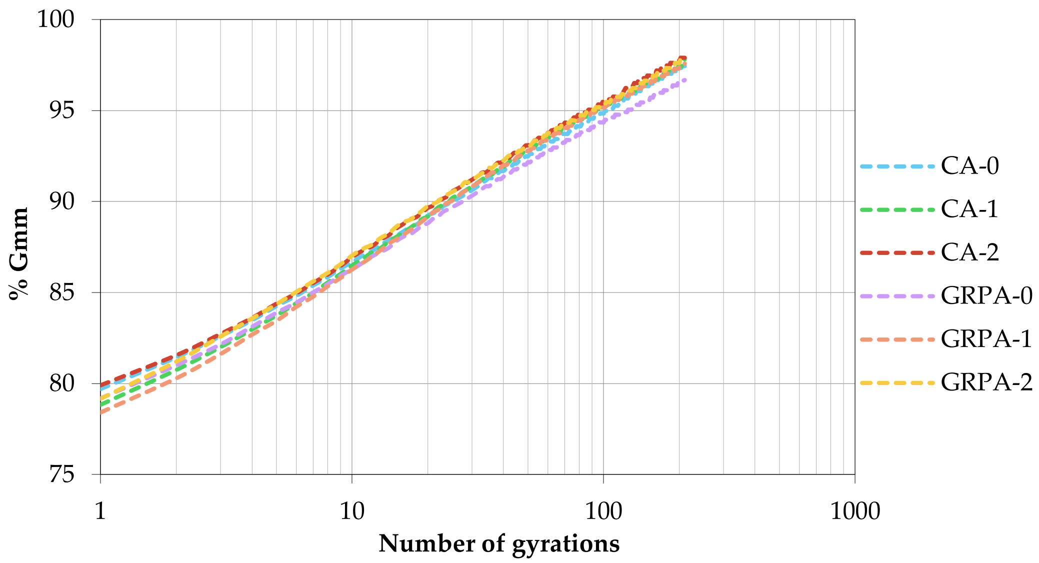

Figure 6. 3 Densification curves of the mixtures.

6.4.2 Indirect Tensile Strength

In accordance with EN 12697-23 [143], three specimens from each mixture compacted at 120 gyrations were tested after being conditioned at 25 °C for 4 hours. The results of the ITS test are shown in Figure 6.4. The findings indicate that the ITS value of the recycled plastic-modified asphalt mixture is significantly higher than that of the unmodified mixture, with a

29% increase. It signifies that the addition of the GRP modifier improved the cohesion and tensile strength of the mixture, which resulted in a high ITS value. In particular, in first-cycle recycled mixtures, CA-1 and GRPA-1 showed superior performance in ITS compared to the reference conventional mix (CA-0), consistent with previous studies suggesting that incorporating RAP into asphalt mixtures increases their tensile strength [50]. However, as anticipated, there was a slight decline in the ITS values for GRPA-1 (3.6%) and GRPA-2 (7.3%) compared to GRPA-0 because rejuvenator dosage was optimised to achieve comparable performance to conventional asphalt at each cycle; however, these recycled mixtures outperformed the reference mixture. Overall, all results for the recycled mixtures at both recycling stages comply with the technical limits (ITS = 0.90–2.2 MPa) suggested by the Italian highway authority. Based on this, it is obvious that modifying the asphalt mixture with recycled plastic did not have a negative impact on its tensile strength throughout the recycling cycles.

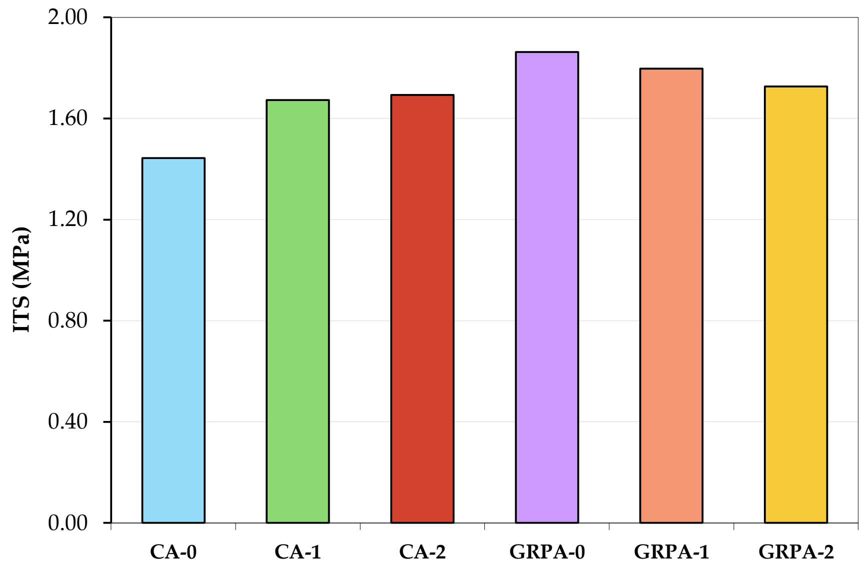

Figure 6. 4 ITS of the mixtures.

6.4.3 Indirect Tensile Stiffness Modulus

In this study, in order to investigate the linear viscoelastic behaviour of the asphalt mixtures, three test temperatures of 10 °C, 20 °C and 30 °C were carried out following the Annex C of EN 12697-26 [144]. Figure 6.5 illustrates the average ITSM values for the tested mixtures at three different temperatures. The asphalt mixture modified with recycled plastic, GRPA-0, consistently exhibited a significantly higher stiffness modulus compared to the unmodified mix, CA-0 across all the test temperatures, confirming the substantial impact of the GRP modifier on mixture stiffness. In the first recycling phase, both CA-1 and GRPA-1 demonstrated a marginal reduction in ITSM values compared to the conventional mix. This minor decline in stiffness modulus could be attributed to the effects of rejuvenation and aggregate distribution within the recycled mixtures. Despite the varying dosages of the rejuvenator in CA-1 and GRPA-1, both exhibited comparable ITSM values throughout all tested temperatures, affirming that recycling asphalt mixtures containing recycled plastic performs similarly to that of the conventional asphalt. Furthermore, this trend of a slight decrease in stiffness modulus was also observed in subsequent recycling phases. CA-2 and GRPA-2 showed comparable ITSM values among themselves and with their prior cycles. It is worth noting that the rejuvenated mixtures did not undergo severe ageing compared to their predecessors during multiple ageing cycles, which can be identified by the similar dosage of rejuvenation used for both CA-2 and GRPA-2 compared to the first-cycled recycled mixtures. It is obvious that if severe ageing occurred, it would have substantially influenced the stiffness modulus of the recycled mixtures and could be identified with a higher magnitude; however, this trend was not observed in this study. This suggests that, with the right selection and dosage of rejuvenators, recycled mixtures exhibit less severity of ageing compared to non-recycled mixtures. Besides, the Italian technical specification requires the ITSM value at 20 °C to be within the range of 3000-8000 MPa for a conventional asphalt mix. The obtained results showed that all the recycled mixtures at both recycling cycles are within these recommended limits, indicating that the recycled plastic modifier has no detrimental effect on recycling asphalt mixtures containing such a modifier.

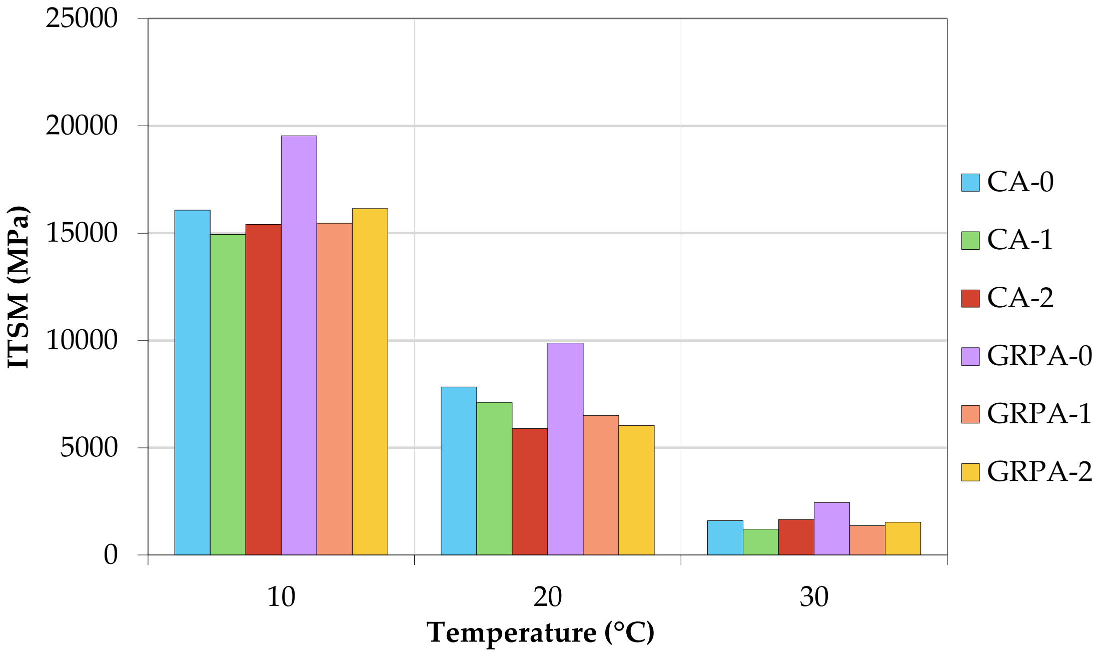

Figure 6. 5 ITSM results of the mixtures.

6.4.4 Indirect Tensile Strength Ratio

The moisture susceptibility of the asphalt mixtures was determined using the ITSR, in accordance with EN 12697-12 [145], three specimens from each asphalt mixture were conditioned in a water bath at 40 °C for 72 hours, followed by conditioning in an air chamber at 25 °C for 4 hours. The average ITSR results are presented in Figure 6.6. The results showed that all mixtures demonstrated significantly high ITSR values, exceeding the recommended threshold limit of 75% suggested by the Italian technical specification. In this assessment, it was observed that the conventional mix performed better than the recycled plastic-modified asphalt mixture. The effect of recycled plastic in GRPA-0 mixture slightly reduced the ITSR value, which is 16% less than that of CA-0. Furthermore, the first-cycle recycled mixtures, CA-1 and GRPA-1, demonstrated a slight decline in ITSR values, i.e. 8.9% and 13.5% respectively, compared to CA-0, while the second-cycle recycled mixtures outperformed among all other mixtures. However, an increasing trend of resistance to moisture susceptibility was observed in the recycled plastic-modified mix over two recycling cycles. It is important to note that limestone aggregates, which are known for being less susceptible to moisture impact, were utilised in this study. Consequently, the adhesive bonding strength between the aggregates and the binder remains unaffected by moisture, and wet conditioning may contribute to increased stiffness, ultimately resulting in a higher ITS of the wet subsets. This could potentially contribute to the overall superior performance observed during the moisture susceptibility evaluation.

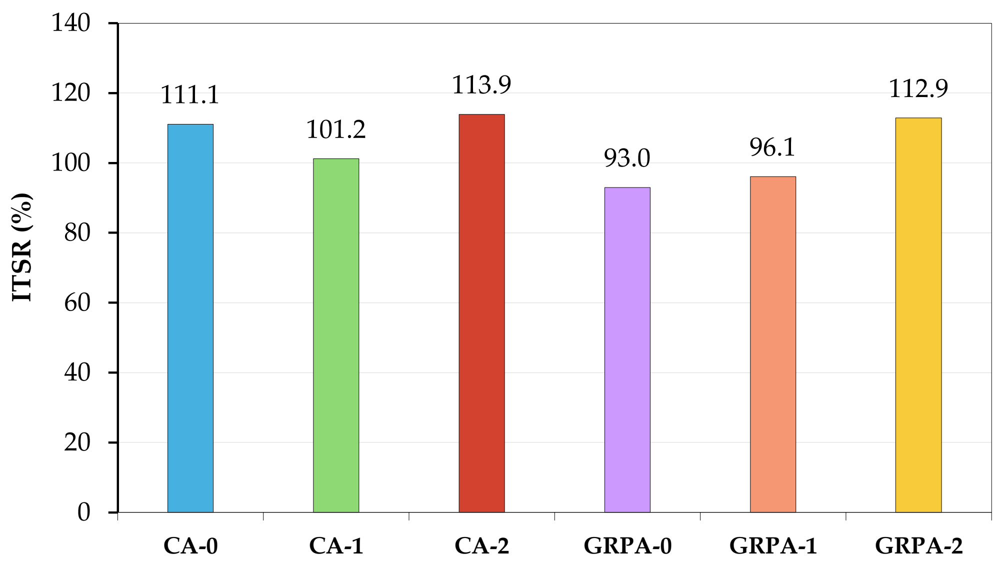

Figure 6. 6 ITSR results of the mixtures.

6.4.5 Wheel tracking test

The wheel tracking test was conducted to evaluate the rutting resistance of the asphalt mixtures under repetitive wheel loads. The samples were prepared in accordance with EN 12697-22 [146], which involved producing two slabs measuring 305 x 305 x 50 mm for each mixture using a roller compactor. To ensure consistency, all specimens from each mixture were compacted to achieve a targeted air void range of 4 ± 0.5%. The results presented in Figure 6.7 shows the evolution of rut depth obtained from the wheel tracking test over the load cycles, while Table 6.5 provides a summary of rutting parameters such as rut depth, WTS, and PRD for all mixtures. The results indicate significant differences in these parameters between the asphalt mixtures containing recycled plastic and the conventional mixture. Specifically, the recycled plastic-modified mixture (GRPA-0) exhibited a reduced rut depth of 2.9 mm compared to the reference unmodified mixture (CA-0), which had a substantially higher rut depth of 5.2 mm. Furthermore, comparing other rutting parameters between these two mixtures reveals that GRPA-0 showed an 82.6% reduction in wheel tracking slope and a 44.8% reduction in proportional rut depth compared to CA-0. The improved resistance to rutting observed with the recycled plastic-modified mix demonstrates its efficacy for high-temperature performance, which aligns with existing literature [195]. Furthermore, improved resistance to rutting was also evident in all the recycled mixtures during two recycling cycles when compared to the conventional mix. It is worth noting that during wheel tracking slope evaluation, GRPA-1 showed a marginally steeper slope compared to that of CA-1; this could be attributed to different rejuvenator dosages (1% more) used during the recycling stage of the GRPA-1 mix. However, this variation in wheel tracking slope did not affect their final rut depth, which was identical. Some previous studies have reported that a higher dosage of rejuvenation can lead to increased rutting depth due to excessive softening of the binder [9]. Among all the evaluated mixtures, GRPA-2 exhibited the highest resistance to rutting, with a minimal rut depth of 2.3 mm and a proportional rut depth of 4.6%. The higher resistance to moisture susceptibility identified in the ITSR test for the second-cycle recycled mixtures is consistent with the results of the rutting test, indicating their improved high-temperature performance.

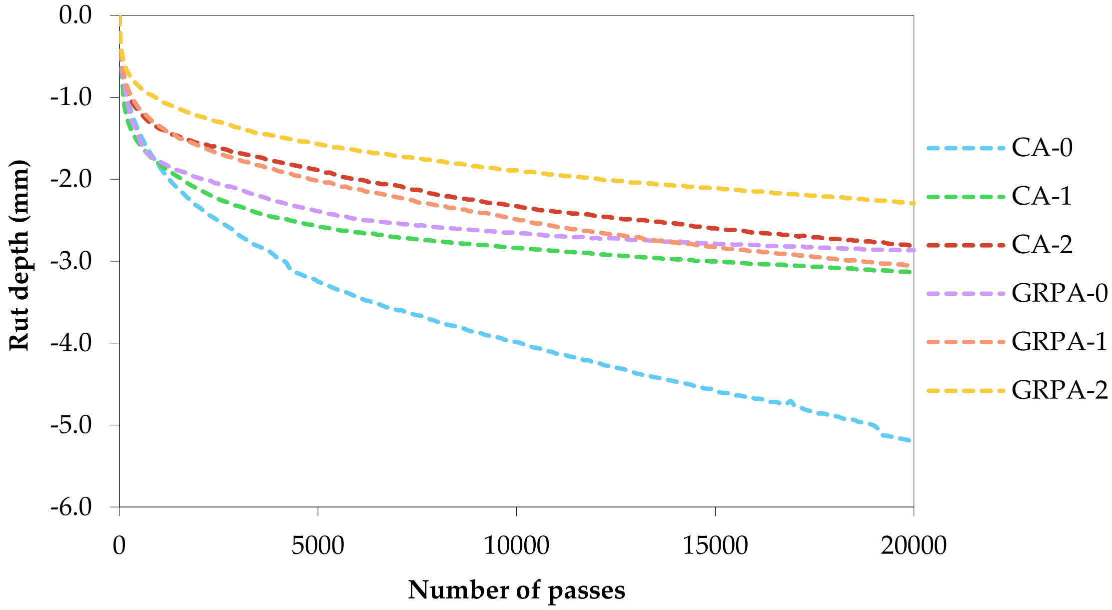

Figure 6. 7 Results of the wheel tracking test.

Table 6. 5 Evaluated rutting parameters.

Mix	CA-0	CA-1	CA-2	GRPA-0	GRPA-1	GRPA-2
Rut depth (mm)	5.2	3.2	2.8	2.9	3.1	2.3
WTS$_{Air}$ (mm/10^3 cycles)	0.24	0.07	0.10	0.04	0.11	0.08
PRD$_{Air}$ (%)	10.4	6.3	5.6	5.7	6.1	4.6

6.4.6 Indirect Tensile Fatigue Test

The study included three stress levels (150, 250, and 350 KPa) for each mixture. A minimum of three cylindrical specimens were prepared for each stress level, resulting in a total of nine specimens tested per mixture to assess the fatigue life. The fatigue life refers to the total number of load cycles that lead to the failure of the specimen. Figure 6.8 illustrates the fatigue life of asphalt mixtures in relation to their initial tensile strain and the total number of load cycles until failure. The fatigue life equations along with the co-efficient of determination are included in the figure. It is significant that that all the fatigue lines have co-efficient of determination higher than that of 0.90, which indicate a good statistical correlation within the obtained results to predict the fatigue behaviour. Figure 6.9 demonstrates the fatigue behaviour of the studied mixtures over various applied stress levels. The results indicate that incorporating recycled plastic via dry method improved the fatigue life of the modified asphalt mix, potentially extending its service life compared to conventional pavements, aligning with previous research. The enhancement in fatigue life can be identified by the lower slope of the fatigue line. Conversely, the conventional mix (CA-0) exhibited higher initial strain at all stress levels compared to other mixtures, leading to inferior fatigue performance. In contrast, GRPA-0 showed lower initial strain across all stress levels than other mixes, although it showed higher stiffness modulus at the test temperature. Notably, the recycled mixtures from both plastic-modified and conventional mixtures showed higher fatigue performance at each recycling cycle compared to the conventional mix, which can be observed from the fatigue model. This could be attributed to the influence of rejuvenation, which altered the viscoelastic behaviour of these mixtures, which was also evident by the lower stiffness modulus obtained in the ITSM test at 20 °C. The same enhanced trend was also observed in the second-cycle recycled mixtures (CA-2 and GRPA-2). Furthermore, in order to compare the fatigue life within the investigated mixtures, an initial strain of 75 μm/m has been considered. The comparison with this initial strain revealed that the asphalt mixtures such as GRPA-0, GRPA-1, GRPA-2, CA-1, CA-2 have a fatigue life more than 74%, 67%, 36%, 41%, 23% respectively to that of conventional mix. In this regard, it is evident that the influence of recycled plastic enhanced the fatigue life of the mixtures at each recycling cycle nevertheless fatigue life over each cycles decline compared to their predecessors. This was expected, as during the repeated recycling at a 50% rate, the content of the recycled plastic-modified mixtures gets reduced over the recycling phases, along with the rejuvenation could impact the behaviour of the mixture.

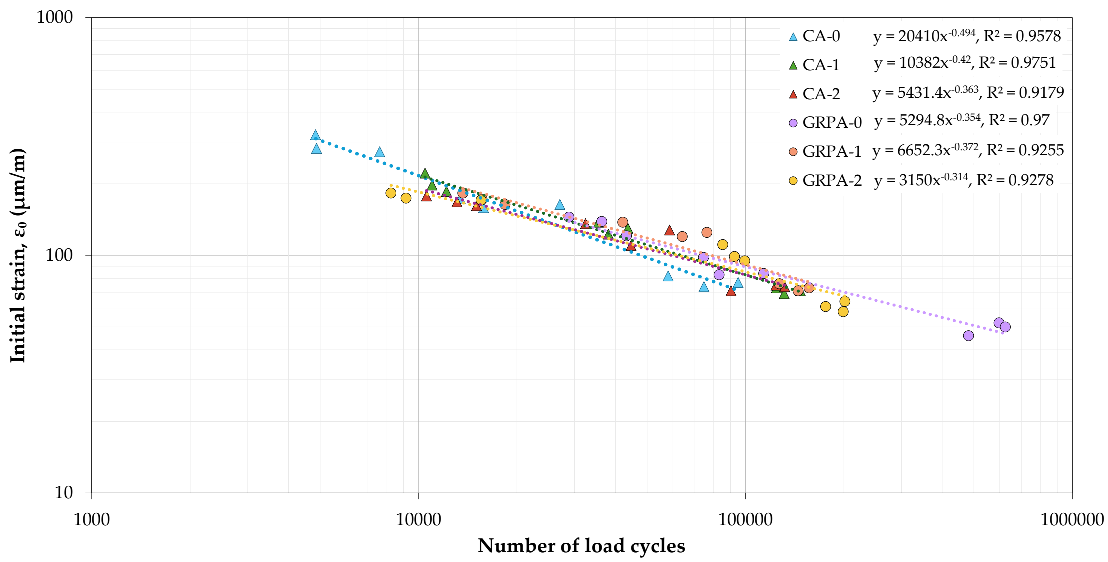

Figure 6. 8 Fatigue line of the mixtures.

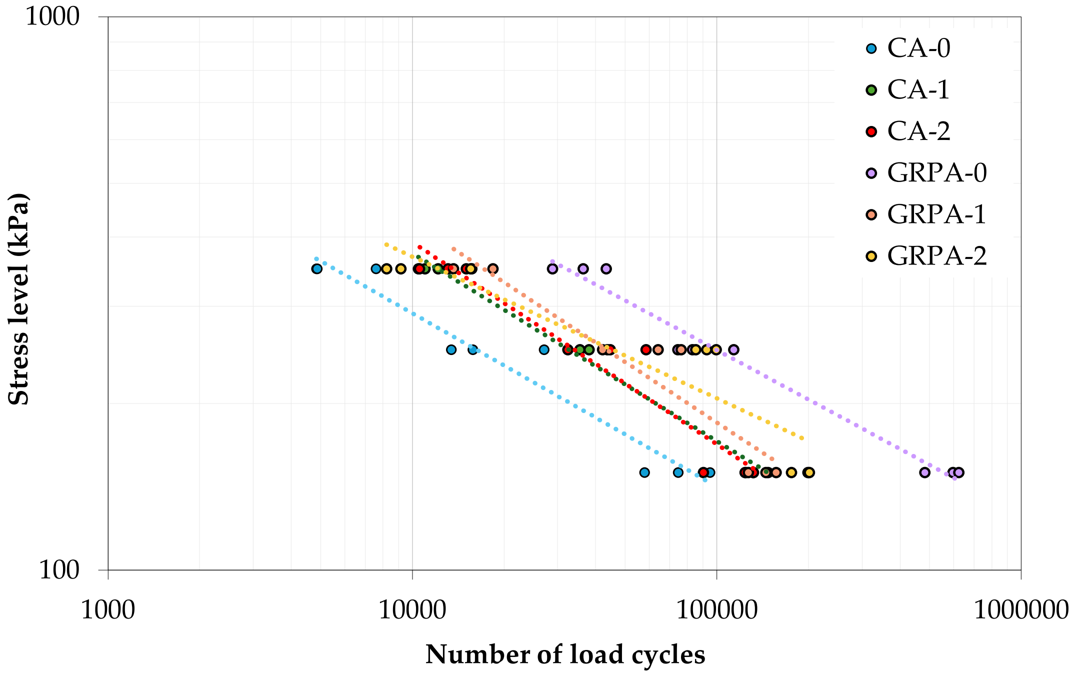

Figure 6. 9 Fatigue life over various stress levels.

6.5 Summary and Conclusions

In this chapter, the study investigated the feasibility of multi-recycling asphalt mixtures modified with and without a recycled plastic modifier at a recycling rate of 50% for two recycling cycles. An innovative recycled plastic, the GRP modifier, was used in this study to modify the asphalt mixture using the dry method. The viability of recycling these asphalt mixtures for two cycles was comprehensively evaluated through volumetric and mechanical characterization. The following conclusions can be drawn based on the findings:

- The incorporation of the innovative recycled plastic modifier in the asphalt mixture via the dry method significantly improved the volumetric and mechanical performance of the asphalt mixture, as evident when compared to the unmodified mixture.
- The influence of the rejuvenator did not have a negative effect on the compactability and mechanical behaviour of the recycled mixtures over two recycling cycles. However, a slightly lower stiffness modulus was observed at 20 °C compared to the conventional one, although the results are within the recommended limits of Italian technical specifications.

- The asphalt mixtures, both modified with and without the recycled plastic, at a recycling rate of 50% over the two cycles demonstrated higher tensile strength and improved resistance to moisture compared to the recommended technical limits.
- All recycled mixtures, regardless of their origin and recycling cycle, exhibited substantially higher resistance to rutting compared to the conventional mixture.
- The analysis of fatigue life indicated that the asphalt mixture modified with recycled plastic has superior fatigue resistance compared to the conventional mixture. Additionally, all the recycled mixtures with a 50% recycling rate also showed significant improvement in the fatigue life cycle. This implies that an effective recycling approach could enhance the fatigue resistance of mixtures containing a high RAP content, regardless of their origin.

Overall, based on the findings, it is evident that the asphalt mixture, when modified with a recycled plastic modifier, can be effectively recycled over multiple life cycles without compromising their volumetric and mechanical characteristics, which are comparable to conventional asphalt. The study contributes to understanding the potential recyclability of asphalt mixtures containing recycled plastic, which is crucial for promoting circular economy practices in the road engineering sector. This approach creates confidence and opportunities for stakeholders and decision-makers to implement closed and open-loop recycling strategies, integrating more waste alternatives into the road industry. Consequently, this reduces the reliance on virgin materials, thereby lowering costs and energy consumption while enabling products to maintain their value and performance in the market for an extended period. Further research is underway to explore a wide range of recycled plastics and incorporate them into asphalt via both wet and dry methods to gain a broader perspective.

Chapter 7

A proposed model to evaluate the multi-recyclability potential of asphalt mixtures under the scope of circular economy

7.1 Introduction

The civil engineering industry, particularly the pavement engineering sector, is a major consumer of natural resources, which are extensively used in the production, construction, and management of transportation infrastructure. Natural aggregates are one of the most highly exploited resources to produce asphalt mixtures and even unbound layers and earthworks, which in turn compose the main components of an asphalt pavement infrastructure. The use and maintenance phases of pavement assets also involve the extraction and utilisation of significant amounts of raw materials. Hence, the implementation of circular economy principles in the road engineering industry can lead to substantial reductions in energy and material consumption [196]. One notable example is the use of reclaimed asphalt pavement (RAP), which, despite being considered waste, holds considerable market value, and offers an excellent opportunity to minimise resource depletion through reuse and recycling.

Circular Economy, as stated previously, is restorative and regenerative and aims to keep products, components, and materials at their highest utility and value at all times. Specifically, it supports the "re-circulation" of materials and energy within the same (closed-loop process) or alternative product systems (open-loop process) and, thus, leads to the elimination of avoidable wastes [197]. In this context, RAP is perceived as an ideal material that can re-enter the cycle of asphalt mixture production [198]. Although the concept of CE is not new, it has not been widely and formally implemented in transportation infrastructure projects, specifically in asphalt pavements. The concept of CE first emerged as a proactive policy goal for many businesses and political agendas in the late 1970s, driven primarily by concerns about climate change and rising resource prices raised by Carson and Boulding [15]. It encompasses principles from multiple schools of thought, including industrial ecology, performance economy, biomimicry, cradle-to-cradle design, blue economy, regenerative design, cleaner production, and natural capitalism [3], [199]. However, when it comes to transport infrastructure, specifically asphalt pavements and RAP, it becomes complex to integrate all aspects of CE into their life cycles. As a result, there are no records of studies or approaches relevant to quantifying their circularity. Highways England, London's Waste & Recycling Board (LWARB), Ellen MacArthur Foundation (EMF), and Opportunity Peterborough are among the institutions and companies that are driving towards introducing CE principles and its metrics within their agenda with hopes of influencing a wider decision-making audience [31], [200]. Highways England, in collaboration with two of the most reputable global construction companies, published an "Approach and Route Map" detailing how to achieve CE implementation through their initial strategies. LWARB recently released London's CE Route Map to accelerate the growth of CE across London [153]. The roadmap includes an ambitious plan of action for the built environment and transportation infrastructure. "Cities in the Circular Economy: An Initial Exploration," a report by the Ellen MacArthur Foundation, highlights the challenges of the linear economy and promotes urban-scale implementation benefits within built environments [34]. In 2018, Opportunity Peterborough published its "Circular City Roadmap," providing a resourceful plan and performance monitoring framework for 2021 that outlines next steps towards realising circular infrastructure goals, ultimately aiming for a "circular city [201]." Some attempts have also been made by companies trying to specifically implement the principles of CE in the production of asphalt mixtures. Tarpaper recycling, along with Super Asfalt, have proposed the production of REC100. Additionally, SYLVAROAD™ RP1000 is an additive derived from crude tall oil that can increase levels of RAP incorporated into asphalt mixtures [153].

Although many researchers and companies have shifted their focus to implementing circular economic principles in the road engineering sector, ongoing debates persist on how to measure and identify the circularity value of a product [202]. Such measurement is imperative in many applications, such as product design, material selection, etc [203]. In this regard, the EMF has proposed a model called the Material Circularity Indicator (MCI), a methodology to measure the material circularity of products, mainly designed for technical cycles [204], [205]. This model can be significant in understanding the relative transition of any product from a linear economy to a circular economy. Later, Mantalovas and Di Mino [3] developed an analytical model named the Product Material Circularity Index (MCI_{MRA}), as shown in Figure 7.1, specifically tailored for road pavements. This index enables the determination of the circularity potential of an asphalt mixture containing RAP and provides a comprehensive assessment of the product by evaluating its environmental and mechanical performance.

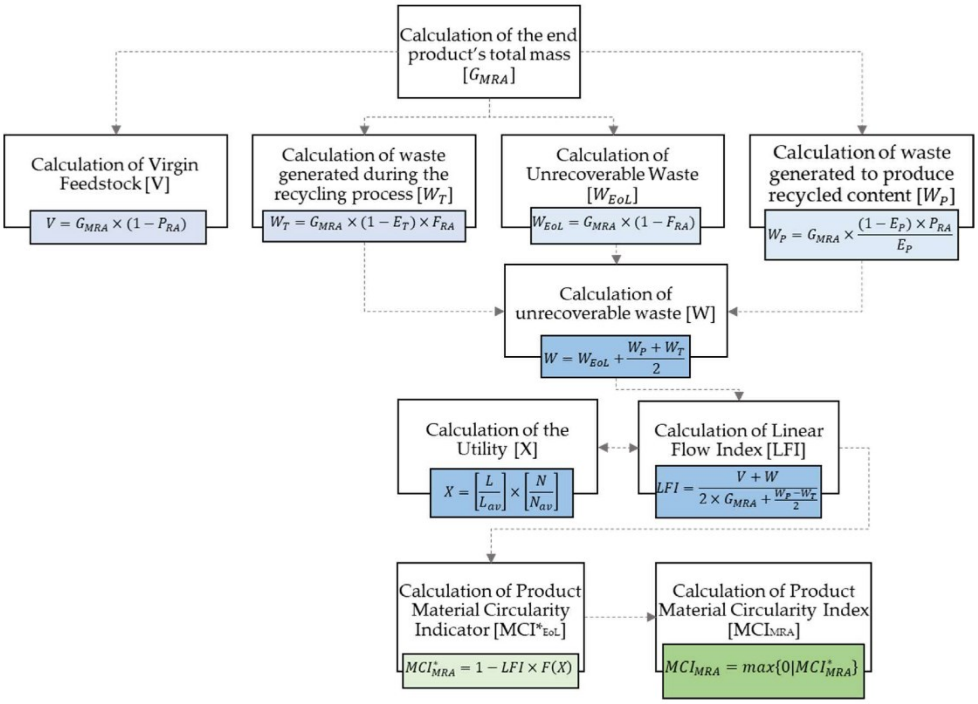

Figure 7. 1 Analytical model developed by Mantalovas and Di Mino to quantify the product circularity [3] .

7.2 Scope and Objectives of the chapter

The adoption of circular economy principles in the road construction industry is vital for addressing the escalating environmental concerns related to climate change, resource depletion, and pollution. The recycling of asphalt pavement has been recognised as a significant solution to tackle these challenges, as implementing CE in this way can reduce the reliance on virgin materials, leading to lower costs and energy consumption while maintaining product value and performance over an extended period in the market. However, the increasing use of alternative materials like waste plastic via an open-loop recycling approach in asphalt pavement has raised concerns about their recyclability. Therefore, it is crucial to assess the circularity of such asphalt mixtures across multiple lifecycles within a circular economy framework. With the growing interest in implementing circular economic strategies in the road engineering sector, the metric to identify product circularity has become increasingly significant. Currently, there is limited research on material circularity at the product level, while focusing on the significance of implementing product circularity indicators for multiple cycle scenarios, which could be instrumental in promoting both open-loop and closed-loop approaches for a long-term strategy. Hence, this chapter aims to develop a novel methodological approach for quantifying product circularity for asphalt mixtures, both with and without a recycled plastic modifier, over two life cycles.

7.3 Methodology

In line with the circular economy framework, this study solely focuses on the technical cycles and evaluates the recirculation capability of asphalt mixtures. The study considered both scenarios of asphalt mixtures, with and without recycled plastic modifier, using a circular economic model for multiple life cycles. The goal is to assess their recyclability potential at the end of their service period. To quantify the product's circularity for multiple life cycles, an analytical model was developed, as represented in equation 7. 1. To formulate this circular economic model, certain key parameters were defined and are described in general terms as following.

- $PMRI_n$ – Product Multi-Recyclability Index at n cycle
- α_n – performance factor at n cycle
- ϑ – recycling rate
- S_n- sum of the series of n cycle.
- P_r^{-1} – Mechanical performance in terms of rutting
- P_f - Mechanical performance in terms of fatigue
- P_m - mechanical performance in terms of moisture susceptibility.

$$\mathrm{PMRI_n} = \alpha_n . \vartheta . S_n \qquad (7.1)$$

In the equation, $PMRI_n$ represents the product multi-recyclability index for n life cycles. It directly depends on the performance factor (α_n), the recycling rate (ϑ), and the sum of the series (S_n). To better understand the framework's scheme and the definition of S_n, a table has been provided to demonstrate the circularity of a product at a constant recycling rate of 50% over 7 typical recycling cycles. In this specific case, V represents the virgin materials' rate at each recycling cycle, set constantly as 50%, and ϱ_n represents the multi-recycling rate of the product. The table clearly shows that with increasing recycling cycles, there is a gradual decrease in the multi-recycling rate of the product due to both virgin material and recycling rates being kept constant. Therefore, in this specific instance during cycle 7, we observe that multi-recycling rate tends to be less than 1%. Thus, we set the multi-recyclability concept as a sum of the fraction replacing a part of recycled product.

Table 7. 1 Scheme of the product circularity over multiple recycling cycles.

Cycle	V (%)	ϱ1 (%)	ϱ2 (%)	ϱ3 (%)	ϱ4 (%)	ϱ5 (%)	ϱ6 (%)	ϱ7 (%)
0	100	0						
1	50	50						
2	50	25	25					
3	50	25	12.5	12.5				
4	50	25	12.5	6.25	6.25			
5	50	25	12.5	6.25	3.125	3.125		
6	50	25	12.5	6.25	3.125	3.125	1.56	
7	50	25	12.5	6.25	3.125	3.125	1.56	0.78

Considering this scheme, S_n is defined as the sum of a series representing the product circularity for n recycling cycles. Since the series is geometric, having a constant ratio between successive terms, equal to ϑ. Thus, Sn can be formulated as follows:

$$S_n = \sum_{j=0}^{n} \left(\frac{1}{\vartheta}\right)^j \quad (7.2)$$

As geometric series, the S_n can be expressed as follows

$$S_n = \frac{1-\vartheta^n}{1-\vartheta} \quad (7.3)$$

Furthermore, the performance factor (α_n) is a crucial parameter in assessing the product multi-recyclability index ($PMRI_n$), as it is directly proportional to this index. The performance factor (α_n) is defined as the ratio between product performance at n cycle to that of the reference/original product, as shown in equation 7.4. The significance of this parameter is that it determines the product's efficiency for multiple life cycles, which depends on various factors such as the method of recycling, recycling agents, the recycling rate, and the quality of materials used. In this study, the performance factor (α_n) is calculated as the ratio of product of the evaluated performance parameters of the recycled mixture to those of the reference mixture, as seen in equation 7.5. The performance factor can also be interpreted as the normalised value of overall recycled product performance to the overall performance of the reference one. Considering the main goal, performance parameters such as resistance to rutting, fatigue and moisture susceptibility were considered; these are significant in identifying durability and mechanical performance of asphalt mixtures.

$$\alpha_n = \prod \left[\frac{P_{n\,recycled}}{P_{reference}}\right] \quad (7.4)$$

$$\alpha_n = \frac{\prod[P_r^{-1}, P_f, P_m]_{n\ recycled}}{\prod[P_r^{-1}, P_f, P_m]_{reference}} \tag{7.5}$$

Where, P_r^{-1} is the resistance to rutting, evaluated according to EN 12697-22, is represented here by the inverse of rut depth of the asphalt mixture in mm. A higher value indicates better resistance to rutting. P_f represents the number of cycles to fatigue failure at a controlled stress level of 250 kPa, chosen according to EN 12697-24. P_m represents resistance to moisture and is evaluated via indirect tensile strength ratio measured in % as per EN 12697-12.

7.4 Results, analysis and discussion

After establishing the methodological framework for the product multi-recyclability index, a case study was structured and carried out to validate the framework. Initially, an analysis of the performance of the product - bituminous mixtures with and without a recycled plastic modifier at a 50% recycling rate over two life cycles - was conducted. To evaluate overall product performance, resistance to rutting, fatigue, and moisture susceptibility were taken into consideration. The values of these evaluated performance parameters for both reference mixtures and their corresponding recycled mixtures across two life cycles are presented in Tables 7.1 and 7.2 respectively. The results presented here were the outcomes of the study discussed in the previous chapter.

Table 7. 2 Evaluated performance parameters of the reference mixtures.

Mixture	P_r^{-1} (mm^{-1})	P_f (cycles)	P_m (%)
CA-0	0.19	18773	111.1
GRPA-0	0.35	90475	93.0

Table 7. 3 Evaluated performance parameters of the recycled mixtures.

Mixture	P_r^{-1} (mm^{-1})	P_f (cycles)	P_m (%)
CA-1	0.32	39115	101.2
CA-2	0.36	48603	113.9
GRPA-1	0.33	72214	96.1
GRPA-2	0.44	109105	112.9

Table 7. 4 Case-1: Product multi-recyclability index of asphalt mixtures with reference to their non-recycled counterparts.

Mixture	n	ϑ	S_n	α_n	$PMRI_n$
CA-1	1	0.5	1	3.14	1.57
CA-2	2	0.5	1.5	4.91	3.68
GRPA-1	1	0.5	1	0.77	0.39
GRPA-2	2	0.5	1.5	1.83	1.37

Table 7. 5 Case-2: Product multi-recyclability index of asphalt mixtures with reference to the conventional asphalt.

Mixture	n	ϑ	S_n	α_n	$PMRI_n$
CA-1	1	0.5	1	3.14	1.57
CA-2	2	0.5	1.5	4.91	3.68
GRPA-1	1	0.5	1	5.65	2.81
GRPA-2	2	0.5	1.5	13.41	10.06

The results in Table 7.4 indicate an increasing trend in the performance factor (α_n) for both asphalt mixtures, with and without a recycled plastic modifier, over two life cycles. However, there is a difference in the magnitude of this factor between the two types of mixtures. This disparity can be attributed to the fact that for the recycled plastic-modified asphalt mixture, the product multi-recyclability index was calculated based on its original performance (100% virgin materials with a recycled plastic modifier). As expected, the performance factor at recycling cycle 1 for this mixture was lower than that of the non-recycled one; however, in subsequent cycle, it demonstrated superior performance compared to their original mixtures and contributed to better $PMRI_n$ relative to their predecessor. Furthermore, since the recycling rate of these mixtures over two cycles remained constant, their other circularity parameters also remained unchanged between them. In contrast, conventional asphalt mixtures exhibited a different trend: as recycling continued over multiple cycles, the performance factor (α_n) improved significantly, leading to higher $PMRI_n$ values compared to those achieved by recycled plastic-modified mixtures at each cycle. The higher $PMRI_n$ value indicates its high multi-recyclability potential, which aligns with the principles of the circular economy. Obviously, this is preliminary evidence, and further investigation is needed to confirm that as a rule, which requires a higher number of recycling cycles than two.

However, considering the main objective of the thesis, its significance is to investigate whether asphalt mixtures modified with recycled plastic can be recycled like conventional mixes. Therefore, this aspect can be explored using the proposed circular economic model. In this regard, the performance factors of the recycled mixtures from the plastic-modified mixture were calculated with reference to conventional asphalt, as shown in Table 7.5. The analysis revealed a significant improvement over each cycle, indicating a higher performance factor compared to the conventional mix. This suggests that both asphalt mixtures, with or without a recycled plastic modifier, have potential for re-circulation within the closed loop using optimised recycling technologies.

Based on the proposed novel approach, it is evident that the product multi-recyclability index ($PMRI_n$) could increase for any asphalt mixtures if their recycled mixtures at n cycles have a performance factor greater than 1. Taking into consideration the scheme shown in Table 7.1, one can determine the maximum recycling cycle that contributes to circulating the product at its highest utility. Based on these parameters, the product multi-recyclability index for any asphalt mixture can be quantified.

7.5 Summary and Conclusions

This chapter dealt with the development of a novel circular economic framework to quantify the product circularity across multiple life cycles. The study introduced a mathematical model, the product multi-recyclability index ($PMRI_n$), and employed it to assess the multi-recyclability potential of various asphalt mixtures, including those with and without recycled plastic modifiers, over two life cycles at a 50% recycling rate. Certain key parameters were identified for this evaluation, including the performance factor which plays a crucial role in determining $PMRI_n$. The study analysed different case studies within the scope of the thesis. Based on this analysis, the following conclusion can be drawn:

- The product multi-recyclability index, $PMRI_n$ significantly depends on the performance factor of the recycled mixtures at their respective recycling cycles. An improvement in the performance factor directly contributes to a higher potential for multi-recyclability of the mixture.
- The case study of $PMRI_n$ of asphalt mixtures with and without a modifier at a recycling rate of 50% for two life cycles revealed that the recycled plastic-modified mix showed comparatively lower $PMRI_n$ than the unmodified one. This can be attributed to two factors: firstly, the performance was compared with its respective non-recycled mix, which served as a baseline for this case study; and secondly, the performance evaluation on these asphalt mixtures showed a significant improvement in resistance to moisture for the unmodified asphalt mix.
- The case study of $PMRI_n$ in asphalt mixtures, with and without a modifier, at a recycling rate of 50% for two life cycles compared to the conventional mix revealed

that the PMRI of the recycled plastic-modified mix showed significantly higher performance than the unmodified one. In this case, performance was significantly improved, as evidenced by a higher performance factor compared to its counterpart.

Based on the findings, it can be deduced that assessing the multi-recyclability potential of a product within the context of a circular economy relies significantly on factors such as recycling rate (ϑ) and product performance (α_n), as seen in asphalt mixtures. Therefore, the performance factor of any recycled product after n cycle demonstrates a value greater than or equal to 1, contributes to an increase in $PMRI_n$. In this respect, the findings indicate that recycled plastic-modified asphalt mixtures can be recycled multiple times, exhibiting its higher recyclability potential in terms of maintaining durability and mechanical performance. A higher $PMRI_n$ value indicates a high potential for multiple recycling, aligning with the principles of the circular economy. However, this is preliminary evidence and further investigation is needed to confirm this trend, which requires evaluating several recycling cycles.

Chapter 8
Conclusion and Future Perspectives

The current thesis has addressed a significant research question related to the recyclability of recycled plastic-modified asphalt mixtures. Understanding this aspect is crucial for promoting alternative materials like waste plastics in modifying asphalt pavement within the context of the circular economy. To achieve this, the study scientifically investigated the feasibility of recycling asphalt mixtures modified with recycled plastic and assessed their physical, rheological, chemical, and mechanical performance over multiple life cycles. It ultimately developed and validated a circular economic model that quantifies material circularity over multiple life cycles, which can be used as a tool for stakeholders and decision-makers looking to implement circular economy principles in asphalt pavement. This chapter concludes by presenting the major findings from this research, along with recommendations and future perspectives.

In this thesis, after introducing the scope and objectives, the research was structured into four significant tasks aimed at achieving the main goal of this study. The study initially identified the urge to establish an operational method to simulate the ageing behaviour of asphalt mixtures within the laboratory. Several accelerated ageing methods were attempted on both asphalt binder and mixture. Consequently, a specific ageing protocol has been implemented, which served as a significant driver for simulating end-of-life scenarios across multiple life cycles for the investigated asphalt mixtures, as well as for the properties of recycled material. The findings suggest that oven ageing of loose asphalt mixtures at 95 °C for 5 days following short-term ageing could simulate similar behaviour observed in the existing standard binder ageing method. Additionally, it highlighted that the ageing severity occurring within the asphalt mixture and binder as independent components becomes distinct with the increasing duration of ageing. However, this ageing protocol is useful for comprehending the ageing mechanisms of dry method modified asphalt mixtures specifically.

Considering the main goal of this study, after establishing the ageing protocol, the multi-recyclability of asphalt mixtures modified, with and without a recycled hard plastic modifier was evaluated preliminarily on the basis of binder-scale evaluation for three recycling cycles via physical, rheological, and chemical characterisation. Furthermore, the impact of recycled plastic as a modifier, incorporated via dry method in the asphalt mixture, was found out and it indicated an improvement also in the rheological properties of the subjecting binder. The characteristics of RAP derived from both conventional and recycled plastic-modified asphalt mixtures, as well as RAP from their recycled counterparts, were investigated. As the study considered a recycling rate of 50% over each cycle, the rejuvenator was employed and the impact of rejuvenation over each recycling stage was investigated according to the objectives of the thesis. The findings suggest that the rejuvenation effectively worked on both asphalt mixtures, with and without a recycled plastic modifier. However, interestingly over multiple recycling cycles, it was found that the rejuvenated mixtures were marginally less subjected to ageing compared to the virgin mixtures. However, the choice of type and dosage of rejuvenator is crucial in asphalt recycling, specifically for high recycling rate. The binder-scale evaluation clearly showed no adverse effects on recycling the recycled plastic-modified asphalt mixture, which performed similarly to unmodified mixtures. To comprehensively investigate the multi-recyclability of recycled plastic-modified asphalt compared to conventional mix at a 50% recycling rate over two life cycles, volumetric and mechanical performance properties were evaluated. The mixture-scale performance evaluation at each recycling cycle is crucial for understanding the feasibility of incorporating high RAPs in the asphalt mixtures, especially in the case of RAPs from plastic-modified asphalt pavement. The experimental results clearly exhibited its potential recyclability and its benefits in ensuring superior performance compared to virgin asphalt mixtures.

Furthermore, within the context of research on circularity metrics, the ultimate goal of this research was to develop a circular economic model for quantifying material circularity over multiple life cycles. The study introduced a novel analytical formulation called the Product Multi-Recyclability Index ($PMRI_n$), which served as a tool for quantifying the multi-recyclability of asphalt mixtures, both with and without a recycled plastic modifier for two life cycles. The $PMRI_n$ depends significantly on the recycling rate and performance factor of recycled mixtures over multiple life cycles. Consequently, $PMRI_n$ was observed to increase with each subsequent recycling cycle, highlighting that maintaining the performance of recycled mixtures through optimised techniques is crucial to increase $PMRI_n$. Overall, based on the findings of the experimental campaign and the novel formulation introduced, it is evident that recycled plastic-modified asphalt mixtures can be recycled multiple times without compromising rheological and mechanical performance properties compared to conventional asphalt. In this regard, reclaimed asphalt pavement derived from such a plastic-modified mixture can be treated similarly to conventional RAP.

The research contributes to advancing insights into the potential recyclability of asphalt mixtures modified with recycled plastic, which is important for promoting sustainable practices in road engineering. This approach foster confidence and provide opportunities for stakeholders and decision-makers to implement recycling strategies, reducing reliance on new materials and cutting costs while extending product performance in the market. Further study is recommended to better understand the influence of UV rays on ageing behaviour. Additionally, it would be beneficial to explore recyclability from the perspective of wet method modification and considering a wider range of recycled plastic. Finally, the future work will be focused on considering a range of recycling rates and several cycles of recycling to establish a consistent trend for $PMRI_n$ and its sensitivity to each driver.

References

[1] E. Santolini, G. Tarsi, D. Torreggiani, and C. Sangiorgi, "Towards more sustainable infrastructures through circular processes: Environmental performance assessment of a case study pavement with recycled asphalt in a life cycle perspective," *J. Clean. Prod.*, vol. 448, no. February, p. 141380, 2024, doi: 10.1016/j.jclepro.2024.141380.

[2] European Asphalt Pavement Association, "Asphalt the 100% recyclable construction product: EAPA Position paper," *Eur. Asph. Pavement Assoc.*, pp. 1–21, 2014.

[3] K. Mantalovas and G. Di Mino, "The sustainability of reclaimed asphalt as a resource for road pavement management through a circular economic model," *Sustain.*, vol. 11, no. 8, 2019, doi: 10.3390/su11082234.

[4] M. Manosalvas-Paredes, R. Roberts, M. Barriera, and K. Mantalovas, "Towards more sustainable pavement management practices using embedded sensor technologies," *Infrastructures*, vol. 5, no. 1, 2020, doi: 10.3390/infrastructures5010004.

[5] European Asphalt Pavement Association, "EAPA'S Position Statement on the use of secondary materials, by-products and waste in asphalt mixtures," *Eur. Asph. Pavement Assoc.*, 2017.

[6] European Commission, "Circular economy action plan," 2020.

[7] A. Bonicelli, P. Calvi, G. Martinez-Arguelles, L. Fuentes, and F. Giustozzi, "Experimental study on the use of rejuvenators and plastomeric polymers for improving durability of high RAP content asphalt mixtures," *Constr. Build. Mater.*, vol. 155, pp. 37–44, 2017, doi: 10.1016/j.conbuildmat.2017.08.013.

[8] M. Elkashef, R. C. Williams, and E. Cochran, "Investigation of fatigue and thermal cracking behavior of rejuvenated reclaimed asphalt pavement binders and mixtures," *Int. J. Fatigue*, vol. 108, no. June 2017, pp. 90–95, 2018, doi: 10.1016/j.ijfatigue.2017.11.013.

[9] M. Zaumanis, R. B. Mallick, L. Poulikakos, and R. Frank, "Influence of six rejuvenators on the performance properties of Reclaimed Asphalt Pavement (RAP) binder and 100% recycled asphalt mixtures," *Comput. Chem. Eng.*, vol. 71, pp. 538–550, 2014, doi: 10.1016/j.conbuildmat.2014.08.073.

[10] I. Camargo, B. Hofko, A. Graziani, and V. Grilli, "Dilauryl thiodipropionate as a regeneration agent for reclaimed asphalts," *Constr. Build. Mater.*, vol. 365, no. December 2022, p. 130049, 2023, doi: 10.1016/j.conbuildmat.2022.130049.

[11] F. Russo, S. Eskandarsefat, L. Venturini, and N. Viscione, "A complete study on an asphalt concrete modified with graphene and recycled hard-plastics: A case study," *Case Stud. Constr. Mater.*, vol. 17, no. June, p. e01437, 2022, doi: 10.1016/j.cscm.2022.e01437.

[12] J. Zhang, Z. Yao, T. Yu, S. Liu, and H. Jiang, “Experimental evaluation of crumb rubber and polyethylene integrated modified asphalt mixture upon related properties,” *Road Mater. Pavement Des.*, vol. 20, no. 6, pp. 1413–1428, 2019, doi: 10.1080/14680629.2018.1447505.

[13] I. B. Joohari and F. Giustozzi, “Chemical and high-temperature rheological properties of recycled plastics-polymer modified hybrid bitumen,” *J. Clean. Prod.*, vol. 276, p. 123064, 2020, doi: 10.1016/j.jclepro.2020.123064.

[14] N. Viscione, D. Lo Presti, R. Veropalumbo, C. Oreto, S. A. Biancardo, and F. Russo, “Performance-based characterization of recycled polymer modified asphalt mixture,” *Constr. Build. Mater.*, vol. 310, no. September, p. 125243, 2021, doi: 10.1016/j.conbuildmat.2021.125243.

[15] K. Mantalovas and G. Di Mino, “Integrating circularity in the sustainability assessment of asphalt mixtures,” *Sustain.*, vol. 12, no. 2, pp. 9–12, 2020, doi: 10.3390/su12020594.

[16] A. Lachat *et al.*, “From buildings’ end of life to aggregate recycling under a circular economic perspective: A comparative life cycle assessment case study,” *Sustain.*, vol. 13, no. 17, 2021, doi: 10.3390/su13179625.

[17] P. O. Awoyera and A. Adesina, “Plastic wastes to construction products: Status, limitations and future perspective,” *Case Stud. Constr. Mater.*, vol. 12, p. e00330, 2020, doi: 10.1016/j.cscm.2020.e00330.

[18] D. Lazarevic, E. Aoustin, N. Buclet, and N. Brandt, “Plastic waste management in the context of a European recycling society: Comparing results and uncertainties in a life cycle perspective,” *Resour. Conserv. Recycl.*, vol. 55, no. 2, pp. 246–259, 2010, doi: 10.1016/j.resconrec.2010.09.014.

[19] G. M. Duarte and A. L. Faxina, “Asphalt concrete mixtures modified with polymeric waste by the wet and dry processes: A literature review,” *Constr. Build. Mater.*, vol. 312, no. July, 2021, doi: 10.1016/j.conbuildmat.2021.125408.

[20] A. Diab, M. Enieb, and D. Singh, “Influence of aging on properties of polymer-modified asphalt,” *Constr. Build. Mater.*, vol. 196, pp. 54–65, 2019, doi: 10.1016/j.conbuildmat.2018.11.105.

[21] L. D. Poulikakos *et al.*, “RILEM interlaboratory study on the mechanical properties of asphalt mixtures modified with polyethylene waste,” *J. Clean. Prod.*, vol. 375, no. August, 2022, doi: 10.1016/j.jclepro.2022.134124.

[22] S. D’angelo, G. Ferrotti, F. Cardone, and F. Canestrari, “Asphalt Binder Modification with Plastomeric Compounds Containing Recycled Plastics and Graphene,” *Materials (Basel).*, vol. 15, no. 2, 2022, doi: 10.3390/ma15020516.

[23] G. White, “Evaluating recycled waste plastic modification and extension of bituminous binder for asphalt,” *Proc. Eighteenth Annu. Int. Conf. Pavement Eng. Asph. Technol. Infrastruct.*, vol. 1, no. February, pp. 1–14, 2019.

[24] D. X. Lu, M. Enfrin, Y. J. Boom, and F. Giustozzi, "Future recyclability of hot mix asphalt containing recycled plastics," *Constr. Build. Mater.*, vol. 368, no. January, p. 130396, 2023, doi: 10.1016/j.conbuildmat.2023.130396.

[25] Association of Nordic Engineers, "Towards a Circular Economy. Skills and competences for STEM professionals," *Assoc. Nord. Eng.*, no. November, 2021, [Online]. Available: https://nordicengineers.org/wp-content/uploads/2022/11/ane-report-towards-a-circular-economy.pdf.

[26] Ellen MacArthur Foundation, "Towards a Circular Economy: Business Rationale for an Accelerated Transition," *Ellen MacArthur Found.*, p. 20, 2015.

[27] N. Gregson, M. Crang, S. Fuller, and H. Holmes, "Interrogating the circular economy: the moral economy of resource recovery in the EU," *Econ. Soc.*, vol. 44, no. 2, pp. 218–243, 2015, doi: 10.1080/03085147.2015.1013353.

[28] ARUP, "The Circular Economy in the Built Environment," *ARUP*, pp. 1–93, 2016.

[29] D. Reike, W. J. V. Vermeulen, and S. Witjes, "The circular economy: New or Refurbished as CE 3.0? — Exploring Controversies in the Conceptualization of the Circular Economy through a Focus on History and Resource Value Retention Options," *Resour. Conserv. Recycl.*, vol. 135, no. February 2017, pp. 246–264, 2018, doi: 10.1016/j.resconrec.2017.08.027.

[30] Ellen MacArthur Foundation, "Towards the Circular Economy," *Ellen MacArthur Found.*, vol. 1, 2013, doi: 10.1596/37331.

[31] Ellen MacArthur Foundation, "Delivering The Circular Economy- A Toolkit for Policymakers," *Ellen MacArthur Found.*, no. June, 2015, doi: https://www.ellenmacarthurfoundation.org/publications/delivering-the-circular-economy-a-toolkit-for-policymakers.

[32] V. H. S. De Abreu, M. G. Da Costa, V. X. Da Costa, T. F. De Assis, A. S. Santos, and M. de A. D'Agosto, "The Role of the Circular Economy in Road Transport to Mitigate Climate Change and Reduce Resource Depletion," *Sustain.*, vol. 14, no. 14, 2022, doi: 10.3390/su14148951.

[33] Ellen MacArthur Foundation, "Achieving ' Growth Within ,'" *Ellen McArthur Found.*, 2017.

[34] Ellen MacArthur Foundation, "Cities in the Circular Economy: An initial exploration," *Ellen MacArthur Found.*, p. 16, 2017.

[35] Bocconi University, Ellen MacArthur Foundation, and Intesa Sanpaolo, "The circular economy as a de-risking strategy and drive of superiot risk-adjusted returns," *Ellen MacArthur Found.*, 2021, doi: 10.36661/2596-142x.2019v1i1.10902.

[36] M. & C. Ellen MacArthur Foundation, "Growth Within: a circular economy vision for a competitive Europe," *Ellen MacArthur Found.*, 2015.

[37] European Environment Agency, "Accelerating the circular economy in Europe," 2024. doi: 10.2800/055236.

[38] European Circular Economy Stakeholder Platform, "Towards a biodiversity - Inclusive and Climate- friendly circular economy," 2024.

[39] F. Russo, C. Oreto, and R. Veropalumbo, "A practice for the application of waste in road asphalt pavements in an eco-friendly way," *Appl. Sci.*, vol. 11, no. 19, 2021, doi: 10.3390/app11199268.

[40] Euopean Asphalt Pavement Association, "Asphalt-A Key Construction Product for the European Circular Economy," 2022. doi: www.eapa.org.

[41] D. Vandewalle, V. Antunes, J. Neves, and A. C. Freire, "Assessment of eco-friendly pavement construction and maintenance using multi-recycled rap mixtures," *Recycling*, vol. 5, no. 3, pp. 1–21, 2020, doi: 10.3390/recycling5030017.

[42] V. Antunes, J. Neves, and A. C. Freire, "Performance assessment of reclaimed asphalt pavement (RAP) in road surface mixtures," *Recycling*, vol. 6, no. 2, pp. 1–17, 2021, doi: 10.3390/recycling6020032.

[43] A. Eltwati, A. Mohamed, M. R. Hainin, E. Jusli, and M. Enieb, "Rejuvenation of aged asphalt binders by waste engine oil and SBS blend: Physical, chemical, and rheological properties of binders and mechanical evaluations of mixtures," *Constr. Build. Mater.*, vol. 346, no. April, p. 128441, 2022, doi: 10.1016/j.conbuildmat.2022.128441.

[44] N. Bardella, M. Facchin, E. Fabris, M. Baldan, and V. Beghetto, "Waste Cooking Oil as Eco-Friendly Rejuvenator for Reclaimed Asphalt Pavement," *Materials (Basel).*, vol. 17, p. 13, 2024, doi: https://doi.org/10.3390/ma17071477.

[45] H. Wu, M. Yang, W. Song, Z. Wu, D. Chen, and X. Chen, "Mechanical and rheological properties of polyurethane-polyurea (PU-PUa) modified asphalt binder," *Constr. Build. Mater.*, vol. 411, no. December 2023, p. 134798, 2024, doi: 10.1016/j.conbuildmat.2023.134798.

[46] K. Li, J. Xie, Y. Liu, Y. Tan, and Y. Pan, "Development and evaluation of roadway-oriented epoxy-modified asphalt binders and mixtures," *Constr. Build. Mater.*, vol. 418, no. December 2023, p. 135398, 2024, doi: 10.1016/j.conbuildmat.2024.135398.

[47] Audrey Copeland, "Reclaimed Asphalt Pavement in Asphalt Mixtures: State of the Practice," *Rep. No. FHWA-HRT-11-021*, no. FHWA, p. McLean, Virginia, 2011.

[48] CEN, "EN 13108-8 Bituminous Mixtures-Material Specifications-Part 8: Reclaimed Asphalt." 2016.

[49] G. Tarsi, P. Tataranni, and C. Sangiorgi, "The Challenges of Using Reclaimed Asphalt Pavement for New Asphalt Mixtures: A Review," *Materials (Basel).*, 2020, doi: doi:10.3390/ma13184052.

[50] M. Zaumanis, R. B. Mallick, and R. Frank, “100% recycled hot mix asphalt: A review and analysis,” *Resour. Conserv. Recycl.*, vol. 92, pp. 230–245, 2014, doi: 10.1016/j.resconrec.2014.07.007.

[51] M. Rathore and M. Zaumanis, “Impact of laboratory mixing procedure on the properties of reclaimed asphalt pavement mixtures,” *Constr. Build. Mater.*, vol. 264, p. 120709, 2020, doi: 10.1016/j.conbuildmat.2020.120709.

[52] P. Rath, J. E. Love, W. G. Buttlar, and H. Reis, “Performance analysis of asphalt mixtures modified with ground tire rubber modifiers and recycled materials,” *Sustain.*, vol. 11, no. 6, p. 1792, 2019, doi: 10.3390/su11061792.

[53] M. Zaumanis, R. B. Mallick, and R. Frank, “100% Hot Mix Asphalt Recycling: Challenges and Benefits,” *Transp. Res. Procedia*, vol. 14, pp. 3493–3502, 2016, doi: 10.1016/j.trpro.2016.05.315.

[54] R. McDaniel and R. Michael Anderson, *NCHRP Report 452 - Recommended Use of Reclaimed Asphalt Pavement in the Superpave Mix Design Method: Technician's Manual TRANSPORTATION*. 2001.

[55] I. L. Al-Qadi, Q. Aurangzeb, and S. H. Carpenter, “Impact of High RAP Content on Structural and Performance Properties of Asphalt Mixtures,” *Illinois Cent. Transp.*, p. 107, 2012.

[56] J. R. Willis and M. Marasteanu, *Improved Mix Design, Evaluation, and Materials Management Practices for Hot Mix Asphalt with High Reclaimed Asphalt Pavement Content*. 2013.

[57] M. Zaumanis, L. D. Poulikakos, and M. N. Partl, “Performance-based design of asphalt mixtures and review of key parameters,” *Mater. Des.*, vol. 141, pp. 185–201, 2018, doi: 10.1016/j.matdes.2017.12.035.

[58] M. Zaumanis, M. Arraigada, and L. D. Poulikakos, “100% Recycled High-Modulus Asphalt Concrete Mixture Design and Validation Using Vehicle Simulator,” *Constr. Build. Mater.*, vol. 260, p. 119891, 2020, doi: 10.1016/j.conbuildmat.2020.119891.

[59] R. Izaks, M. Rathore, V. Haritonovs, and M. Zaumanis, “Performance properties of high modulus asphalt concrete containing high reclaimed asphalt content and polymer modified binder,” *Int. J. Pavement Eng.*, vol. 23, no. 7, pp. 2255–2264, 2022, doi: 10.1080/10298436.2020.1850721.

[60] M. C. Cavalli, M. Zaumanis, E. Mazza, M. N. Partl, and L. D. Poulikakos, “Aging effect on rheology and cracking behaviour of reclaimed binder with bio-based rejuvenators,” *J. Clean. Prod.*, vol. 189, pp. 88–97, 2018, doi: 10.1016/j.jclepro.2018.03.305.

[61] S. Mo, Y. Wang, F. Xiong, C. Ai, D. Wang, and G. Y. A. Tan, “Changes of asphalt fumes in hot-mix asphalt pavement recycling,” *J. Clean. Prod.*, vol. 258, p. 120586, 2020, doi: 10.1016/j.jclepro.2020.120586.

[62] H. P. Torres, S. R. M. Fernandes, C. D. A. Loureiro, C. F. N. Moura, H. M. R. D. Silva, and J. R. M. Oliveira, "Use of polymer modified binders as rejuvenators in recycled asphalt mixtures," *Int. J. Pavement Res. Technol.*, vol. 13, no. 6, pp. 654–664, 2020, doi: 10.1007/s42947-020-6006-5.

[63] European Asphalt Pavement Association, "Asphalt in Figures 2022," *Eur. Asph. Pavement Assoc.*, pp. 1–13, 2024.

[64] European Asphalt Pavement Association, "Asphalt in Figure 2020," *Eur. Asph. Pavement Assoc.*, pp. 1–13, 2021, [Online]. Available: https://096.wpcdnnode.com/eapa.org/wp-content/uploads/2021/12/asphalt_in_figures_2020.pdf.

[65] European Asphalt Pavement Association, "Asphalt in Figures 2019," *Eur. Asph. Pavement Assoc.*, vol. 2020, pp. 1–13.

[66] European Asphalt Pavement Association, "Asphalt in Figures 2018," *Eur. Asph. Pavement Assoc.*, pp. 1–13, 2020.

[67] R. Izaks, V. Haritonovs, I. Klasa, and M. Zaumanis, "Hot Mix Asphalt with High RAP Content," *Procedia Eng.*, vol. 114, pp. 676–684, 2015, doi: 10.1016/j.proeng.2015.08.009.

[68] M. Zaumanis, R. B. Mallick, and R. Frank, "Evaluation of different recycling agents for restoring aged asphalt binder and performance of 100 % recycled asphalt," *Mater. Struct. Constr.*, vol. 48, no. 8, pp. 2475–2488, 2015, doi: 10.1617/s11527-014-0332-5.

[69] V. Antunes, A. C. Freire, and J. Neves, "A review on the effect of RAP recycling on bituminous mixtures properties and the viability of multi-recycling," *Constr. Build. Mater.*, vol. 211, pp. 453–469, 2019, doi: 10.1016/j.conbuildmat.2019.03.258.

[70] Q. Aurangzeb, I. L. Al-Qadi, H. Ozer, and R. Yang, "Hybrid life cycle assessment for asphalt mixtures with high RAP content," *Resour. Conserv. Recycl.*, vol. 83, pp. 77–86, 2014, doi: 10.1016/j.resconrec.2013.12.004.

[71] H. M. R. D. Silva, J. R. M. Oliveira, and C. M. G. Jesus, "Are totally recycled hot mix asphalts a sustainable alternative for road paving?," *Resour. Conserv. Recycl.*, vol. 60, pp. 38–48, 2012, doi: 10.1016/j.resconrec.2011.11.013.

[72] National Asphalt Pavement Association (NAPA), "Annual Asphalt Pavement Industry Survey on Recycled Materials and Warm-Mix Asphalt Usage 2019," 2020. doi: www.AsphaltPavement.org.

[73] M. Muñoz *et al.*, "Environmental impact of rejuvenators in asphalt mixtures containing high reclaimed asphalt content," *Road Mater. Pavement Des.*, vol. 23, no. 6, pp. 1400–1414, 2022, doi: 10.1080/14680629.2021.1891129.

[74] R. K. Veeraragavan, R. B. Mallick, M. Tao, M. Zaumanis, R. Frank, and R. L. Bradbury, "Laboratory comparison of rejuvenated 50% reclaimed asphalt pavement hot-mix asphalt with conventional 20% RAP mix," *Transp. Res. Rec.*, vol. 2633, no. 1, pp. 69–79, 2017, doi: 10.3141/2633-09.

[75] S. Purohit, M. Panda, and A. Kumar Das, "Performance of waste polyethylene modified bituminous paving mixes containing reclaimed asphalt pavement and recycled concrete aggregate," *Constr. Build. Mater.*, vol. 348, no. July, p. 128677, 2022, doi: 10.1016/j.conbuildmat.2022.128677.

[76] X. Li, M. O. Marasteanu, R. C. Williams, and T. R. Clyne, "Effect of reclaimed asphalt pavement (proportion and type) and binder grade on asphalt mixtures," *Transp. Res. Rec.*, no. 2051, pp. 90–97, 2008, doi: 10.3141/2051-11.

[77] I. L. Al-Qadi, M. Elseifi, and S. H. Carpenter, "Reclaimed asphalt pavement - A literature review," 2007.

[78] A. Abed, N. Thom, and D. Lo Presti, "Design considerations of high RAP-content asphalt produced at reduced temperatures," *Mater. Struct. Constr.*, vol. 51, no. 4, pp. 1–16, 2018, doi: 10.1617/s11527-018-1220-1.

[79] W. Mogawer, T. Bennert, J. S. Daniel, R. Bonaquist, A. Austerman, and A. Booshehrian, "Performance characteristics of plant produced high RAP mixtures," *Road Mater. Pavement Des.*, vol. 13, no. SUPPL. 1, pp. 183–208, 2012, doi: 10.1080/14680629.2012.657070.

[80] T. Ma, H. Wang, X. Huang, Z. Wang, and F. Xiao, "Laboratory performance characteristics of high modulus asphalt mixture with high-content RAP," *Constr. Build. Mater.*, vol. 101, pp. 975–982, 2015, doi: 10.1016/j.conbuildmat.2015.10.160.

[81] B. Tran and R. Hassan, "Performance of hot-mix asphalt containing recycled asphalt pavement," *Transp. Res. Rec.*, no. 2205, pp. 121–129, 2011, doi: 10.3141/2205-16.

[82] S. K. Pradhan and U. C. Sahoo, "Influence of softer binder and rejuvenator on bituminous mixtures containing reclaimed asphalt pavement (RAP) material," *Int. J. Transp. Sci. Technol.*, vol. 11, no. 1, pp. 46–59, 2022, doi: 10.1016/j.ijtst.2020.12.001.

[83] P. Slabonski, B. Stankiewicz, and D. Beben, "Influence of a rejuvenator on homogenization of an asphalt mixture with increased content of reclaimed asphalt pavement in lowered technological temperatures," *Materials (Basel).*, vol. 14, no. 10, 2021, doi: 10.3390/ma14102567.

[84] J. Zhang *et al.*, "Evaluation on the mechanical performance of recycled asphalt mixtures incorporated with high percentage of RAP and self-developed rejuvenators," *Constr. Build. Mater.*, vol. 269, p. 121337, 2021, doi: 10.1016/j.conbuildmat.2020.121337.

[85] C. Yang *et al.*, "Multi-scale performance evaluation and correlation analysis of blended asphalt and recycled asphalt mixtures incorporating high RAP content," *J. Clean. Prod.*, vol. 317, no. July, p. 128278, 2021, doi: 10.1016/j.jclepro.2021.128278.

[86] R. McDaniel, A. Shah, and G. Huber, "Investigation of Low and High-Temperature Properties of Plant-Produced RAP Mixtures," 2012.

[87] P. Yin and B. Pan, "Effect of RAP content on fatigue performance of hot-mixed recycled

asphalt mixture," *Constr. Build. Mater.*, vol. 328, no. 2, p. 127077, 2022, doi: 10.1016/j.conbuildmat.2022.127077.

[88] H. Sivilevičius, J. Bražiunas, and O. Prentkovskis, "Technologies and principles of hot recycling and investigation of preheated reclaimed asphalt pavement batching process in an asphalt mixing plant," *Appl. Sci.*, vol. 7, no. 11, 2017, doi: 10.3390/app7111104.

[89] J. Lin, J. Hong, J. Liu, and S. Wu, "Investigation on physical and chemical parameters to predict long-term aging of asphalt binder," *Constr. Build. Mater.*, vol. 122, pp. 753–759, 2016, doi: 10.1016/j.conbuildmat.2016.06.121.

[90] K. Błazejowski, M. Wójcik-Wiśniewska, H. Peciakowski, and J. Olszacki, "The Performance of a Highly Modified Binders for Heavy Duty Asphalt Pavements," *Transp. Res. Procedia*, vol. 14, pp. 679–684, 2016, doi: 10.1016/j.trpro.2016.05.331.

[91] C. Yan, J. Xi, C. Ai, and Z. Leng, "Investigating the unique entropy-elasticity of polymer modified asphalt," *Clean. Mater.*, vol. 11, no. December 2023, p. 100216, 2024, doi: 10.1016/j.clema.2023.100216.

[92] G. Polacco, S. Filippi, F. Merusi, and G. Stastna, "A review of the fundamentals of polymer-modified asphalts: Asphalt/polymer interactions and principles of compatibility," *Adv. Colloid Interface Sci.*, vol. 224, pp. 72–112, 2015, doi: 10.1016/j.cis.2015.07.010.

[93] A. I. Al-Hadidy and T. Yi-qiu, "Mechanistic approach for polypropylene-modified flexible pavements," *Mater. Des.*, vol. 30, no. 4, pp. 1133–1140, 2009, doi: 10.1016/j.matdes.2008.06.021.

[94] D. Mehta, N. Saboo, S. M. Abraham, and U. Diwaker, "A review on the use of waste plastics in hot mix asphalt," *Mech. Time-Dependent Mater.*, 2023, doi: 10.1007/s11043-023-09622-y.

[95] J. J. Benvenuta-Tapia, E. Vivaldo-Lima, A. Martínez-Estrada, M. Hernández-Valdez, S. Paredes-Castañeda, and C. Ramos-Valdes, "Enhanced asphalt performance upon addition of RAFT-synthesized reactive multi-block copolymers," *Mater. Chem. Phys.*, vol. 227, no. October 2018, pp. 269–278, 2019, doi: 10.1016/j.matchemphys.2019.02.004.

[96] A. Behnood and M. Modiri Gharehveran, "Morphology, rheology, and physical properties of polymer-modified asphalt binders," *Eur. Polym. J.*, vol. 112, no. September 2018, pp. 766–791, 2019, doi: 10.1016/j.eurpolymj.2018.10.049.

[97] M. R. Mohd Hasan *et al.*, *A Review on Utilization of Electronic Waste Plastics for Use Within Asphaltic Concrete Materials: Development, Opportunities and Challenges for Successful Implementation*, vol. 1–5. Elsevier Ltd., 2018.

[98] X. Liu and X. Wei, "Research development of polymer modified asphalt," *J. Phys. Conf. Ser.*, vol. 1759, no. 1, 2021, doi: 10.1088/1742-6596/1759/1/012011.

[99] R. Geyer, J. R. Jambeck, and K. L. Law, "Production, use, and fate of all plastics ever

made," *Sci. Adv.*, vol. 3, no. 7, pp. 25–29, 2017, doi: 10.1126/sciadv.1700782.

[100] Ellen MacArthur Foundation, "The New Plastics Economy: Rethinking the Future of Plastics & Catalysing Action," *Ellen MacArthur Found.*, 2017, [Online]. Available: https://www.ellenmacarthurfoundation.org/publications/the-new-plastics-economy-rethinking-the-future-of-plastics-catalysing-action.

[101] R. Willis, F. Yin, and R. Moraes, "Recycled Plastics in Asphalt Part A: State of the Knowledge.," 2020. [Online]. Available: www.AsphaltPavement.org.

[102] A. L. Brooks, S. Wang, and J. R. Jambeck, "The Chinese import ban and its impact on global plastic waste trade," *Sci. Adv.*, vol. 4, no. 6, pp. 1–7, 2018, doi: 10.1126/sciadv.aat0131.

[103] Plastics Europe, "Plastics – the Facts 2022," *Plast. Eur.*, no. October, p. 81, 2022, doi: plasticseurope.org.

[104] G. Huang, T. Yang, Z. He, L. Yu, and H. Xiao, "Polyurethane as a modifier for road asphalt: A literature review," *Constr. Build. Mater.*, vol. 356, no. September, 2022, doi: 10.1016/j.conbuildmat.2022.129058.

[105] N. Ahmed, "Utilizing plastic waste in the building and construction industry: A pathway towards the circular economy," *Constr. Build. Mater.*, vol. 383, no. March, p. 131311, 2023, doi: 10.1016/j.conbuildmat.2023.131311.

[106] C. Chin and P. Damen, "Viability of Using Recycled Plastics in Asphalt and Sprayed Sealing Applications," 2019.

[107] G. M. Duarte and A. L. Faxina, "Asphalt concrete mixtures modified with polymeric waste by the wet and dry processes: A literature review," *Constr. Build. Mater.*, vol. 312, no. April, 2021, doi: 10.1016/j.conbuildmat.2021.125408.

[108] L. Brasileiro, F. Moreno-Navarro, R. Tauste-Martínez, J. Matos, and M. del C. Rubio-Gámez, "Reclaimed polymers as asphalt binder modifiers for more sustainable roads: A review," *Sustainability*, vol. 11, no. 3, pp. 1–20, 2019, doi: 10.3390/su11030646.

[109] S. Wu and L. Montalvo, "Repurposing waste plastics into cleaner asphalt pavement materials: A critical literature review," *J. Clean. Prod.*, vol. 280, p. 124355, 2021, doi: 10.1016/j.jclepro.2020.124355.

[110] T. L. X. Wong, M. R. Mohd Hasan, and L. C. Peng, "Recent development, utilization, treatment and performance of solid wastes additives in asphaltic concrete worldwide: A review," *J. Traffic Transp. Eng. (English Ed.*, vol. 9, no. 5, pp. 693–724, 2022, doi: 10.1016/j.jtte.2022.06.003.

[111] Y. Ma *et al.*, "The utilization of waste plastics in asphalt pavements: A review," *Clean. Mater.*, vol. 2, no. July, p. 100031, 2021, doi: 10.1016/j.clema.2021.100031.

[112] Thermofisher Scientific, "Labware physical properties table," *Thermo Fish.*, 2021, doi:

thermofisher.com/nalgene.

[113] N. Mashaan, A. Chegenizadeh, and H. Nikraz, "Laboratory Properties of Waste PET Plastic-Modified Asphalt Mixes," *Recycling*, vol. 6, no. 49, p. 10, 2021, doi: https://doi.org/10.3390/recycling6030049.

[114] R. Li *et al.*, "Innovative application of waste polyethylene terephthalate (PET) derived additive as an antistripping agent for asphalt mixture: Experimental investigation and molecular dynamics simulation," *Fuel*, vol. 300, no. February, p. 121015, 2021, doi: 10.1016/j.fuel.2021.121015.

[115] S. Hassanpour-Kasanagh, P. Ahmedzade, A. M. Fainleib, and A. Behnood, "Rheological properties of asphalt binders modified with recycled materials: A comparison with Styrene-Butadiene-Styrene (SBS)," *Constr. Build. Mater.*, vol. 230, p. 117047, 2020, doi: 10.1016/j.conbuildmat.2019.117047.

[116] S. Ullah, M. Raheel, R. Khan, and M. Tariq Khan, "Characterization of physical & mechanical properties of asphalt concrete containing low- & high-density polyethylene waste as aggregates," *Constr. Build. Mater.*, vol. 301, no. June, p. 124127, 2021, doi: 10.1016/j.conbuildmat.2021.124127.

[117] S. Haider, I. Hafeez, Jamal, and R. Ullah, "Sustainable use of waste plastic modifiers to strengthen the adhesion properties of asphalt mixtures," *Constr. Build. Mater.*, vol. 235, p. 117496, 2020, doi: 10.1016/j.conbuildmat.2019.117496.

[118] P. Mahardi, Y. Risdianto, and M. I. Ramli, "Characteristics of asphalt binder modified with waste vegetable oil and waste plastics Characteristics of asphalt binder modified with waste vegetable oil and waste plastics," *IOP Conf. Ser. Mater. Sci. Eng.*, p. 10, 2020, doi: doi:10.1088/1757-899X/737/1/012126.

[119] M. Nouali, Z. Derriche, E. Ghorbel, and L. Chuanqiang, "Plastic bag waste modified bitumen a possible solution to the Algerian road pavements," *Road Mater. Pavement Des.*, vol. 0, no. 0, pp. 1–13, 2019, doi: 10.1080/14680629.2018.1560355.

[120] F. Xu, Y. Zhao, and K. Li, "Using waste plastics as asphalt modifier: A review," *Materials (Basel).*, vol. 15, no. 1, 2022, doi: 10.3390/ma15010110.

[121] A. M. Azam, S. M. El, and B. Rufak, "Evaluation of asphalt mixtures modified with polymer and wax," *Innov. Infrastruct. Solut.*, vol. 4, no. 1, pp. 1–12, 2019, doi: 10.1007/s41062-019-0230-3.

[122] R. K. Padhan, A. Sreeram, and A. Gupta, "Evaluation of trans-polyoctenamer and cross-linking agents on the performance of waste polystyrene modified asphalt," *Road Mater. Pavement Des.*, vol. 0, no. 0, pp. 1–13, 2018, doi: 10.1080/14680629.2018.1533490.

[123] X. Xu *et al.*, "Chemical upcycling of waste PET into sustainable asphalt pavement containing recycled concrete aggregates: Insight into moisture-induced damage," *Constr. Build. Mater.*, vol. 360, no. November, p. 129632, 2022, doi: 10.1016/j.conbuildmat.2022.129632.

[124] S. Baradaran, J. Rahimi, M. Ameri, and A. Maleki, "Mechanical performance of asphalt mixture containing eco-friendly additive by recycling PET," *Case Stud. Constr. Mater.*, vol. 20, no. August 2023, p. e02740, 2024, doi: 10.1016/j.cscm.2023.e02740.

[125] N. S. Mashaan, A. Chegenizadeh, H. Nikraz, and A. Rezagholilou, "Investigating the engineering properties of asphalt binder modified with waste plastic polymer," *Ain Shams Eng. J.*, vol. 12, no. 2, pp. 1569–1574, 2021, doi: 10.1016/j.asej.2020.08.035.

[126] G. Hao, M. He, S. M. Lim, G. P. Ong, A. Zulkati, and S. Kapilan, "Recycling of plastic waste in porous asphalt pavement: Engineering, environmental, and economic implications," *J. Clean. Prod.*, vol. 440, no. October 2023, p. 140865, 2024, doi: 10.1016/j.jclepro.2024.140865.

[127] S. Yu, J. A. Musazay, C. Zhang, P. Hu, and S. Shen, "Workability of low-density polyethylene modified asphalt mixtures: A statistical analysis of particle kinematics," *J. Clean. Prod.*, vol. 447, no. February, p. 141564, 2024, doi: 10.1016/j.jclepro.2024.141564.

[128] J. N. Hahladakis, C. A. Velis, R. Weber, E. Iacovidou, and P. Purnell, "An overview of chemical additives present in plastics: Migration, release, fate and environmental impact during their use, disposal and recycling," *J. Hazard. Mater.*, vol. 344, pp. 179–199, 2018, doi: 10.1016/j.jhazmat.2017.10.014.

[129] L. M. B. Costa, H. M. R. D. Silva, J. R. M. Oliveira, and S. R. M. Fernandes, "Incorporation of waste plastic in asphalt binders to improve their performance in the pavement," *Int. J. Pavement Res. Technol.*, vol. 6, no. 4, pp. 457–464, 2013, doi: 10.6135/ijprt.org.tw/2013.6(4).457.

[130] M. Ranieri, L. Costa, J. R. M. Oliveira, H. M. R. D. Silva, and C. Celauro, "Asphalt Surface Mixtures with Improved Performance Using Waste Polymers via Dry and Wet Processes," *J. Mater. Civ. Eng.*, vol. 29, no. 10, pp. 1–9, 2017, doi: 10.1061/(asce)mt.1943-5533.0002022.

[131] H. R. Radeef *et al.*, "Enhanced Dry Process Method for Modified Asphalt Containing Plastic Waste," *Front. Mater.*, vol. 8, no. July, pp. 1–14, 2021, doi: 10.3389/fmats.2021.700231.

[132] F. Cardone, S. Spadoni, G. Ferrotti, and F. Canestrari, "Asphalt mixture modification with a plastomeric compound containing recycled plastic: laboratory and field investigation," *Mater. Struct. Constr.*, vol. 55, no. 3, 2022, doi: 10.1617/s11527-022-01954-4.

[133] F. Giustozzi, D. L. Xuan, M. Enfrin, H. Masood, R. Audy, and Y. J. Boom, "Use of Road-grade Recycled Plastics for Sustainable Asphalt Pavements Towards the Selection of Road-grade Plastics – An Evaluation Framework and Preliminary Experimental Results-Research Report AP-R663-21," 2021. doi: www.austroads.com.au.

[134] I. A. Abdalfattah, W. S. Mogawer, and K. Stuart, "Recycled polyethylene (RPE) modified asphalt mixtures: Performance predictions using pavement Mechanistic-Empirical design and evaluation of return on investment," *Constr. Build. Mater.*, vol. 356,

no. September, p. 129164, 2022, doi: 10.1016/j.conbuildmat.2022.129164.

[135] EAPA, "The use of secondary materials, by-products and waste in asphalt mixtures.," *Eur. Asph. Pavement Assoc.*, pp. 1–10, 2020, doi: www.eapa.org.

[136] Y. J. Boom, M. Enfrin, S. Grist, and F. Giustozzi, "Analysis of possible carcinogenic compounds in recycled plastic modified asphalt," *Sci. Total Environ.*, vol. 858, no. November 2022, p. 159910, 2023, doi: 10.1016/j.scitotenv.2022.159910.

[137] CEN, "EN 1426 Bitumen and bituminous binders — Determination of needle penetration." CEN, 2015.

[138] CEN, "EN 1427 Bitumen and bituminous binders — Determination of the softening point — Ring and Ball method." 2015.

[139] CEN, "EN 13302 Bitumen and bituminous binders— Determination of dynamic viscosity of bituminous binder using a rotating spindle apparatus," *CEN*, p. 16, 2022.

[140] CEN, "EN 14770 Bitumen and bituminous binders — Determination of complex shear modulus and phase angle — Dynamic Shear Rheometer (DSR)." 2012.

[141] CEN, "EN 17643:2022 Bitumen and bituminous binders — Determination of equi-shear modulus temperature and phase angle using a Dynamic Shear Rheometer (DSR) — BTSV test," *CEN*, 2022.

[142] CEN, "EN 12697-8 Bituminous mixtures - Test methods - Part 8: Determination of void characteristics of bituminous specimens," *CEN*, 2018.

[143] CEN, "EN 12697-23 Bituminous mixtures - Test methods - Determination of the indirect tensile strength of bituminous specimens," *CEN*, 2017.

[144] CEN, "EN 12697-26 Bituminous mixtures - Test methods - Part 26: Stiffness," *CEN*, 2018.

[145] CEN, "EN 12697-12 Bituminous mixtures - Test methods - Part 12: Determination of the water sensitivity of bituminous specimens," *CEN*, 2018.

[146] CEN, "EN 12697-22 Bituminous mixtures - Test methods - Part 22: Wheel tracking," *CEN*, 2020.

[147] CEN, "EN 12697-24 Bituminous mixtures - Test methods - Part 24: Resistance to fatigue," *CEN*, 2018.

[148] L. Ma, A. Varveri, R. Jing, and S. Erkens, "Comprehensive review on the transport and reaction of oxygen and moisture towards coupled oxidative ageing and moisture damage of bitumen," *Constr. Build. Mater.*, vol. 283, p. 122632, 2021, doi: 10.1016/j.conbuildmat.2021.122632.

[149] J. Mirwald, S. Werkovits, I. Camargo, D. Maschauer, B. Hofko, and H. Grothe, "Investigating bitumen long-term-ageing in the laboratory by spectroscopic analysis of the SARA fractions," *Constr. Build. Mater.*, vol. 258, p. 119577, 2020, doi:

10.1016/j.conbuildmat.2020.119577.

[150] G. Di Mino, V. Vijayan, S. Eskandarsefat, L. Venturini, and K. Mantalovas, "Investigating the Multi-Recyclability of Recycled Plastic-Modified Asphalt Mixtures," *Infrastructures*, vol. 8, no. 5, pp. 1–18, 2023, doi: 10.3390/infrastructures8050084.

[151] M. Pazzini, G. Tarsi, P. Tataranni, C. Lantieri, and G. Dondi, "Mechanical Characterization of Thin Asphalt Overlay Mixtures with 100% Recycled Aggregates," *Materials (Basel).*, 2023, doi: . https://doi.org/10.3390/ma16010188.

[152] K. Mantalovas *et al.*, "European national road authorities and circular economy: An insight into their approaches," *Sustain.*, vol. 12, no. 17, pp. 1–19, 2020, doi: 10.3390/su12177160.

[153] K. Mantalovas, I. P. Dunn, F. Acuto, V. Vijayan, L. Inzerillo, and G. Di Mino, "A Top-Down Approach Based on the Circularity Potential to Increase the Use of Reclaimed Asphalt," *Infrastructures*, vol. 8, no. 5, 2023, doi: 10.3390/infrastructures8050083.

[154] X. L. U and U. Isacsson, "Effect of ageing on bitumen chemistry and rheology," no. July 2001, pp. 15–22, 2002.

[155] Q. Liu, B. Yu, A. Cannone, D. Wang, J. Liu, and W. Bo, "Characterization and molecular mechanism of the thermal-oxidative gradient aging behavior in asphalt films," vol. 199, no. March, 2022, doi: 10.1016/j.measurement.2022.111567.

[156] Y. Guo, P. Tataranni, and C. Sangiorgi, "The use of fibres in asphalt mixtures : A state of the art review," vol. 390, no. December 2022, 2023, doi: https://doi.org/10.1016/j.conbuildmat.2023.131754.

[157] M. C. Cavalli, M. Zaumanis, E. Mazza, M. N. Partl, and L. D. Poulikakos, "Effect of ageing on the mechanical and chemical properties of binder from RAP treated with bio-based rejuvenators," *Compos. Part B Eng.*, vol. 141, no. January, pp. 174–181, 2018, doi: 10.1016/j.compositesb.2017.12.060.

[158] E. Prosperi and E. Bocci, "A review on bitumen aging and rejuvenation chemistry: Processes, materials and analyses," *Sustain.*, vol. 13, no. 12, 2021, doi: 10.3390/su13126523.

[159] H. Zhang, Z. Chen, G. Xu, and C. Shi, "Evaluation of aging behaviors of asphalt binders through different rheological indices," *Fuel*, vol. 221, no. January, pp. 78–88, 2018, doi: 10.1016/j.fuel.2018.02.087.

[160] D. Ragni, G. Ferrotti, X. Lu, and F. Canestrari, "Effect of temperature and chemical additives on the short-term ageing of polymer modi fi ed bitumen for WMA," *Mater. Des.*, vol. 160, pp. 514–526, 2018, doi: 10.1016/j.matdes.2018.09.042.

[161] R. Tauste, F. Moreno-Navarro, M. Sol-Sánchez, and M. C. Rubio-Gámez, "Understanding the bitumen ageing phenomenon : A review," *Constr. Build. Mater.*, vol. 192, pp. 593–609, 2018, doi: 10.1016/j.conbuildmat.2018.10.169.

[162] L. Figueiredo, M. Leite, P. H. Osmari, F. Thiago, and S. Arag, "Rheological indexes for asphalt binders considering different aging conditions : Evaluation and correlations with performance," vol. 338, no. March, 2022, doi: 10.1016/j.conbuildmat.2022.127549.

[163] S. Xu, H. Wu, W. Song, and Y. Zhan, "Investigation of the aging behaviors of reclaimed asphalt," *J. Clean. Prod.*, vol. 356, no. February, p. 131837, 2022, doi: 10.1016/j.jclepro.2022.131837.

[164] J. Miguel, L. Crucho, L. G. De Picado-santos, J. Manuel, and S. Dias, "The TEAGE ageing method for asphalt mixtures," *Transp. Eng.*, vol. 2, no. October, p. 100030, 2020, doi: 10.1016/j.treng.2020.100030.

[165] N. F. Saleh *et al.*, "Effects of aging on asphalt mixture and pavement performance," *Constr. Build. Mater.*, vol. 258, p. 120309, 2020, doi: 10.1016/j.conbuildmat.2020.120309.

[166] Y. R. Kim *et al.*, *Long-Term Aging of Asphalt Mixtures for Performance Testing and Prediction*. National Academies Press, 2017.

[167] D. Steiner *et al.*, "Towards an optimised lab procedure for long-term oxidative ageing of asphalt mix specimen," *Int. J. Pavement Eng.*, vol. 17:6, no. February, pp. 471–477, 2016, doi: 10.1080/10298436.2014.993204.

[168] F. Xiao, S. N. Amirkhanian, M. Karakouzian, and M. Khalili, "Rheology evaluations of WMA binders using ultraviolet and PAV aging procedures," *Constr. Build. Mater.*, vol. 79, pp. 56–64, 2015, doi: 10.1016/j.conbuildmat.2015.01.046.

[169] Y. Li, J. Feng, S. Wu, A. Chen, D. Kuang, and T. Bai, "Review of ultraviolet ageing mechanisms and anti-ageing methods for asphalt binders," *J. Road Eng.*, vol. 2, no. 2, pp. 137–155, 2022, doi: 10.1016/j.jreng.2022.04.002.

[170] V. Mouillet, F. Farcas, J. Sauger, and E. Chailleux, "Study of UV rays effects on the evolution of bituminous mix behaviour," *6th Eurasphalt Eurobitume Congr.*, no. June, 2016.

[171] W. Zeng *et al.*, "Research on Ultra Violet (UV) aging depth of asphalts," *Constr. Build. Mater.*, vol. 160, pp. 620–627, 2018, doi: 10.1016/j.conbuildmat.2017.11.047.

[172] F. Fallah, F. Khabaz, Y. Kim, S. Reddy, and H. F. Haghshenas, "Molecular dynamics modeling and simulation of bituminous binder chemical aging due to variation of oxidation level and saturate-aromatic- resin-asphaltene fraction," *Fuel*, vol. 237, no. September 2018, pp. 71–80, 2019, doi: 10.1016/j.fuel.2018.09.110.

[173] W. Zhang, L. Zou, Z. Jia, F. Wang, Y. Li, and P. Shi, "Effect of Thermo-Oxidative Ageing on Nano-Morphology of Bitumen," *Appl. Sci.*, 2019, doi: https://doi.org/10.3390/app9153027.

[174] ANAS Gruppo FS Italiane, "Capitolato Speciale di Appalto, Norme Tecniche per l'esecuzione del contratto Parte 2. Pavimentazioni Stradali," *ANAS S.p.A.*, 2021, [Online]. Available: www.stradeanas.it.

[175] CEN, "EN 12607-1:2014 Bitumen and bituminous binders- Determination of the resistance to hardening under influence of heat and air- Part 1: RTFOT method." 2014.

[176] CEN, "EN 14769:2012 Bitumen and bituminous binders — Accelerated long-term ageing conditioning by a Pressure Ageing Vessel (PAV)." 2012.

[177] CEN, "EN 12697-1:2020 Bituminous mixtures – Test methods-Part 1: Soluble binder content." 2020.

[178] CEN, "EN 12697-3:2005 Bituminous mixtures — Test methods for hot mix asphalt-Part 3: Bitumen recovery: Rotary evaporator." 2005.

[179] K. Schwettmann, N. Nytus, M. Radenberg, and D. Stephan, "Bitumen reuse: physical and chemical approach to investigate the effectiveness of rejuvenators," *Road Mater. Pavement Des.*, vol. 24, no. 4, pp. 1130–1157, 2023, doi: 10.1080/14680629.2022.2060129.

[180] C. Clopotel and H. Bahia, "The effect of bitumen polar groups adsorption on mastics properties at low temperatures," *Road Mater. Pavement Des.*, vol. 14, pp. 38–51, 2013, doi: http://dx.doi.org/10.1080/14680629.2013.774745.

[181] G. Fritschy and E. Papirer, "Interactions between a bitumen , its components and model fillers," *Fuel*, vol. 57, no. 11, pp. 701–704, 1978, doi: https://doi.org/10.1016/0016-2361(78)90025-X.

[182] Y. Liu, P. Su, M. Li, Z. You, and M. Zhao, "Review on evolution and evaluation of asphalt pavement structures and materials," *J. Traffic Transp. Eng. (English Ed.*, vol. 7, no. 5, pp. 573–599, 2020, doi: 10.1016/j.jtte.2020.05.003.

[183] M. C. Cavalli *et al.*, "Review of advanced road materials, structures, equipment, and detection technologies," *J. Road Eng.*, vol. 3, no. 4, pp. 370–468, 2023, doi: 10.1016/j.jreng.2023.12.001.

[184] Y. Yildirim, "Polymer modified asphalt binders," *Constr. Build. Mater.*, vol. 21, no. 1, pp. 66–72, 2007, doi: 10.1016/j.conbuildmat.2005.07.007.

[185] G. White, "A synthesis on the effects of two commercial recycled plastics on the properties of bitumen and asphalt," *Sustain.*, vol. 12, no. 20, pp. 1–20, 2020, doi: 10.3390/su12208594.

[186] V. Beghetto *et al.*, "By-products from mechanical recycling of polyolefins improve hot mix asphalt performance," *J. Clean. Prod.*, vol. 318, no. August, p. 128627, 2021, doi: 10.1016/j.jclepro.2021.128627.

[187] Greg White and Connor Magee, "Laboratory Evaluation of Asphalt Containing Recycled Plastic as a Bitumen Extender and Modifier," *J. Traffic Transp. Eng.*, vol. 7, no. 5, pp. 218–235, 2019, doi: 10.17265/2328-2142/2019.05.004.

[188] J. Cardoso, A. Ferreira, A. Almeida, and J. Santos, "Incorporation of plastic waste into road pavements: A systematic literature review on the fatigue and rutting

performances," *Constr. Build. Mater.*, vol. 407, no. January, 2023, doi: 10.1016/j.conbuildmat.2023.133441.

[189] M. T. Rahman, A. Mohajerani, and F. Giustozzi, "Recycling of waste materials for asphalt concrete and bitumen: A review," *Materials (Basel).*, vol. 13, no. 7, 2020, doi: 10.3390/ma13071495.

[190] ISO/TC 323, "ISO 59004-2024 Circular economy — Vocabulary, principles and guidance for implementation," *ISO*, 2024. .

[191] C. Yan *et al.*, "Screw extrusion process used in the polymer modified asphalt field: A review," *J. Clean. Prod.*, vol. 448, no. December 2023, p. 141592, 2024, doi: 10.1016/j.jclepro.2024.141592.

[192] P. Singh and V. P. Sharma, "Integrated Plastic Waste Management: Environmental and Improved Health Approaches," *Procedia Environ. Sci.*, vol. 35, pp. 692–700, 2016, doi: 10.1016/j.proenv.2016.07.068.

[193] W. He, Z. Zhao, J. Yuan, and F. Xiao, "Recent development of ethylene–vinyl acetate modified asphalt," *Constr. Build. Mater.*, vol. 363, no. November 2022, p. 129800, 2023, doi: 10.1016/j.conbuildmat.2022.129800.

[194] C. L. Chiang, M. Mivehchi, and H. Wen, "Towards a use of waste polyethylene in asphalt mixture as a compaction aid," *J. Clean. Prod.*, vol. 440, no. November 2023, p. 140989, 2024, doi: 10.1016/j.jclepro.2024.140989.

[195] D. Movilla-Quesada, A. C. Raposeiras, L. T. Silva-Klein, P. Lastra-González, and D. Castro-Fresno, "Use of plastic scrap in asphalt mixtures added by dry method as a partial substitute for bitumen," *Waste Manag.*, vol. 87, pp. 751–760, 2019, doi: 10.1016/j.wasman.2019.03.018.

[196] A. Barragán-Escandón, J. Terrados-Cepeda, and E. Zalamea-León, "The role of renewable energy in the promotion of circular urban metabolism," *Sustain.*, vol. 9, no. 12, pp. 1–29, 2017, doi: 10.3390/su9122341.

[197] P. Swagemakers, M. D. D. Garcia, and J. S. C. Wiskerke, "Socially-inclusive development and value creation: How a composting project in Galicia (Spain) 'Hit the Rocks,'" *Sustain.*, vol. 10, no. 6, 2018, doi: 10.3390/su10062040.

[198] Y. Huang, Y. Ning, T. Zhang, and J. Wu, "Measuring carbon emissions of pavement construction in China," *Sustain.*, vol. 8, no. 8, pp. 1–13, 2016, doi: 10.3390/su8080723.

[199] G. Clark, J. Kosoris, L. N. Hong, and M. Crul, "Design for sustainability: Current trends in sustainable product design and development," *Sustainability*, vol. 1, no. 3, pp. 409–424, 2009, doi: 10.3390/su1030409.

[200] LWARB, "London's Circular Economy route map," *LWARB*, pp. 1–60, 2017.

[201] Opportunity Peterborough, "Circular Peterborough Circular City Roadmap—An

Ambitious Plan & Performance Monitoring Framework Towards 2021," *Oppor. Peterbrgh.*, 2018.

[202] P. Dräger, P. Letmathe, L. Reinhart, and F. Robineck, "Measuring circularity: evaluation of the circularity of construction products using the ÖKOBAUDAT database," *Environ. Sci. Eur.*, vol. 34, no. 1, 2022, doi: 10.1186/s12302-022-00589-0.

[203] V. Van Hoof, C. Maarten, and A. Vercalsteren, "Indicators for a Circular Economy," 2018. doi: https://ce-centre.be.

[204] Eellen MacArthur Foundation, "Material Circularity Indicator (MCI)," *Ellen MacArthur Found.*, pp. 1–64, 2019, doi: https://www.ellenmacarthurfoundation.org/material-circularity-indicator.

[205] L. Rocchi, L. Paolotti, C. Cortina, F. F. Fagioli, and A. Boggia, "Measuring circularity: an application of modified Material Circularity Indicator to agricultural systems," *Agric. Food Econ.*, vol. 9, no. 1, 2021, doi: 10.1186/s40100-021-00182-8.

List of Publications

- Vijayan, V.; Mantalovas, K.; Acuto, F.; Di Mino, G. Evaluating the Ageing Sensitivity of the Asphalt Binder via Distinct Ageing Methods. *Infrastructures* **2023**, *8*, 163. https://doi.org/10.3390/infrastructures8110163.
- Di Mino, G.; Vijayan, V.; Eskandarsefat, S.; Venturini, L.; Mantalovas, K. Investigating the Multi-Recyclability of Recycled Plastic-Modified Asphalt Mixtures. *Infrastructures* **2023**, *8*, 84. https://doi.org/10.3390/infrastructures8050084.
- Mantalovas, K.; Dunn, I.P.; Acuto, F.; Vijayan, V.; Inzerillo, L.; Di Mino, G. A Top-Down Approach Based on the Circularity Potential to Increase the Use of Reclaimed Asphalt. *Infrastructures* **2023**, *8*, 83. https://doi.org/10.3390/infrastructures8050083.
- Di Mino, G.; Mantalovas, K.; Vijayan, V. Investigating the Viability of Multi-Recycling of Asphalt Mixtures through a Preliminary Binder Level Characterization. *Eng. Proc.* **2022**, *17*, 20. https://doi.org/10.3390/engproc2022017020
- Mantalovas, K.; Acuto, F.; Vijayan, V.; Inzerillo, L.; Di Mino, G. Exploring the material circularity index of motorways: A case study over Europe, Bituminous Mixtures and Pavements VIII **2024**, https://doi.org/10.1201/9781003402541.
- Vijayan, V.; Manthos, E.; Mantalovas, K.; Di Mino, G. "Multi-Recyclability of Asphalt Mixtures Modified with Recycled Plastic: Towards a Circular Economy," has been accepted for publication in the international peer-reviewed journal "Results in Engineering" of Elsevier on July 5, 2024.