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Biopolymers as bitumen additives: a systematic quantitative literature review

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ABSTRACT

As sustainability gains importance, the search for eco-friendly alternatives to petroleum-based additives in bitumen modification has intensified. Biopolymers have been of great interest due to their positive impact on bitumen performance. This review analyzes studies on biopolymer-modified bitumen using the systematic quantitative literature review (SQLR) method. An article pool was prepared based on specific criteria, and the test results of the articles were quantified and compared. Biopolymers were categorised into five main groups according to their frequency of use in bitumen modification: lignin and derivatives, natural rubber and derivatives, natural oil-based polymers, bio-based polyurethanes and others. The physical, rheological, and chemical properties of the biopolymer-modified bitumen were analyzed, as well as the results of mechanical tests on asphalt mixtures prepared with the relevant bitumens. Findings reveal that the effects of biopolymers vary by type, highlighting common trends, controversies, and research gaps in the literature.

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KEYWORDS

Biopolymer; bitumen modification; lignin; natural rubber; biobased polyurethane; natural oil based polymers

1. Introduction

Nowadays, plastics have become an inseparable part of the daily life (Rahardiyan et al., 2023). Plastics are widely used in many industries, and their annual production constantly increases (Babaremu et al., 2023). Indeed, worldwide plastics production reached 400.3 million tons by the end of 2022 (Plastics Europe, 2023). Only 8.9% of this production is recycled plastics (Plastics Europe, 2023). 85% of plastic waste ends up in the oceans, causing serious damage to the environment and biodiversity (Khalid & Arif, 2022). Furthermore, most of the polymers used in the production of plastics are petroleum-based polymers (Kabir et al., 2020). Petroleum resources are not sustainable, and considering the current use and production amounts, researchers predict that petroleum reserves may be depleted soon (Penki & Rout, 2023). This situation causes petroleum prices to fluctuate worldwide, leading to increased production costs of petroleum-based synthetic polymers (Gaudenzi, Cardone, Lu, et al., 2023b; Mehta & Saboo, 2023; Penki & Rout, 2023; Yatish et al., 2024).

Biopolymers offer a sustainable alternative to petroleum-based polymers, reducing environmental impact (Juikar & Warkar, 2023; Khalid & Arif, 2022; Vinod et al., 2020). These polymers are biologically compostable in nature (biodegradable) and/or bio-based (Babaremu et al., 2023; Fredi & Dorigato, 2023; Juikar & Warkar, 2023). Due to their advantages, biopolymer-based plastics are increasingly used

in various sectors such as packaging, agriculture, textiles, and many others (Fredri & Dorigato, 2023; Vinod et al., 2020).

Bitumen, a key material in asphalt pavements, and most performance-enhancing additives are petroleum-based (Chen et al., 2023; Luo et al., 2024). This has led researchers to look for more sustainable solutions in asphalt pavement construction. Accordingly, researchers have conducted comprehensive studies on asphalt pavement construction using biobased materials (Abe et al., 2022; Bozdemir et al., 2023; Mehta & Saboo, 2023). In this regard, studies on partial or complete bitumen replacement with biobased materials are especially remarkable (Behnood & Modiri Gharehveran, 2019). As a result of these studies, the concept of bio-bitumen has been introduced. Depending on the amount of biobased material used, bio-bitumen can be categorised into three different categories: additives (< %10), extenders (25–75%), and alternative binders (100%) (Abe et al., 2022; Behnood & Modiri Gharehveran, 2019; Riccardi & Losa, 2024; Z. Zhang, Fang, et al., 2022). While bio-bitumen as an extender or binder is environmentally beneficial, its performance, particularly in high-temperature resistance and aging, remains inadequate (Mehta & Saboo, 2023; Z. Zhang, Fang, et al., 2022). Therefore, the most practical and beneficial application in the near future is using bio-based materials as bitumen additives.

Several narrative reviews examined bio-derived materials in bitumen modification. Also, there are reviews on the subject of the bio-bitumen which investigates the effect of bio-oil and waste oil on bitumen performance (Abdulnaser M. Al-Sabaeei et al., 2022; Behnood, 2019; Elahi et al., 2021; Fang et al., 2021; N. Xu et al., 2023; Z. Zhang, Fang, et al., 2022). These studies indicate that bio-oils and waste oils offer limited high-temperature performance. Therefore, the importance of polymeric materials has emerged to enhance the performance of bio-bitumen. Accordingly, lignin and natural rubber have emerged as biopolymers of interest in bitumen modification. Comprehensive studies have already been conducted on these materials, and narrative reviews have been included in the literature on lignin (Gaudenzi, Cardone, Lu, et al., 2023b; Yatish et al., 2024) and natural rubber (Ansari et al., 2021; Luo et al., 2024). However, given the diversity of biopolymers, focusing only on lignin and natural rubber is insufficient.

To the best of the authors' knowledge, no comprehensive review with comparative quantitative analysis of all biopolymers in bitumen modification exists. This study fills this gap by evaluating biopolymers' effects on bitumen using quantitative data. Thus, it is aimed to reveal the impact of biopolymers, as sustainable and eco-friendly materials, on bitumen modification.

In this study, a systematic quantitative literature review (SQLR) was conducted within specified limitations. Relevant studies were collected and analyzed bibliometrically to provide an overview. Then, additives and experimental methods were categorised. Experimental results were quantified to assess additive effect on neat bitumen. Thus, the impact of biopolymers on bitumen modification has been evaluated from different perspectives, identifying research gaps to guide future studies.

2. Biopolymers

Biopolymers are substances formed by living organisms and found in nature as macromolecules. Also known as natural and bio-based polymers, they can be derived from animals, plants, bacteria, fungi, and biological substances such as sugars, amino acids, and natural oils (Juikar & Warkar, 2023). Bioplastics, made from biopolymers, are defined by The European Bioplastics Organization as plastics that either biobased, biodegradable, or having both characteristics (European Bioplastics, 2024; Fredri & Dorigato, 2021; Pinto et al., 2021; Thomas et al., 2023). Biopolymers can also be considered as a subgroup of bioplastics (Pinto et al., 2021).

Materials must satisfy the definition of being biobased and biodegradable by meeting specific international standards (Fredri & Dorigato, 2023). For instance, ISO 17088:2021 (2021) and ASTM D6400-21 (2021) standards set out the period when a plastic must degrade under specified conditions to be considered compostable. ASTM D6400-21 (2021) defines compostability criteria for domestic and industrial aerobic composting. A product qualifies if (i) no more than 10% of its weight remains after

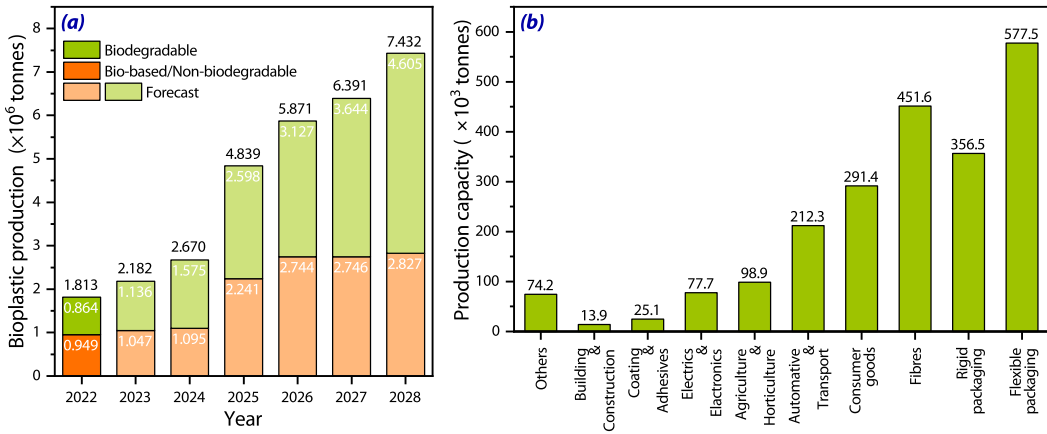


Figure 1. (a) Annual bioplastics production and 5-year forecast, (b) use of bioplastics in different market areas (Plastics Europe, 2023).

84 days in compost, (ii) 90% of its organic carbon converts to CO_2 within 180 days, and (iii) it supports plant growth comparable to control compost (Fredri & Dorigato, 2023). Furthermore, standards such as EN 16640 (2017) and ASTM D6866-22 (2022) define methods to measure renewable carbon content in bio-based plastics, independent of biodegradability, by techniques such as radiocarbon dating (Fredri & Dorigato, 2023; Niaounakis, 2019). Accordingly, it is possible to assess whether biopolymers are bio-based or not.

The European Bioplastics organisation, in partnership with the Nova-Institute, researches bioplastics market trends. They analyze annual production, estimate future growth, and assess sectoral distribution, publishing updates annually (European Bioplastics, 2024). The report published in 2023 stated that roughly only 0.5% of the total annual plastic production of 400 million tons worldwide was bioplastics (Plastics Europe, 2023), highlighting the need for further research to expand sustainable alternatives.

The annual production and 5-year forecast data published in the 2023 report are given in Figure 1(a). This figure shows that approximately 1.8 million tons of bioplastics were produced in 2022, about half of which were biodegradable, and the other half were bio-based bioplastics. It is estimated that this ratio will remain constant and that bioplastic production will be on a steady upward trend in the coming periods. Thus, it is predicted that by 2028, the current annual bioplastic production amount will increase approximately five times.

Furthermore, Figure 1(b) shows statistics on the distribution of annual production capacity in various market areas. When this figure is analyzed, it is clear that bioplastics are most commonly used in the packaging industry. The main reason for this situation is that, compared to other plastics, packaging wastes play a greater role in polluting the nature, which has forced countries to implement environmentalist policies in this regard. As a result of these environmental policies, the production of packaging products from biodegradable bioplastics has become widespread today. On the other hand, it is understood that only a minor share of bioplastics are used in the construction and building industry. When bioplastics are used in the construction sector, it is more important that they are bio-based rather than biodegradable in terms of sustainability. In the construction industry, biopolymers are generally used to enhance the strength of the soil and, in doing so, to allow plants to grow in the soil (Chang et al., 2020). Accordingly, various biopolymers such as natural rubbers, xanthan, chitosan, casein, starch are used in soil reinforcement applications (Chang et al., 2020). However, the use of biopolymers in asphalt pavements is a new topic that is still in the research stage.

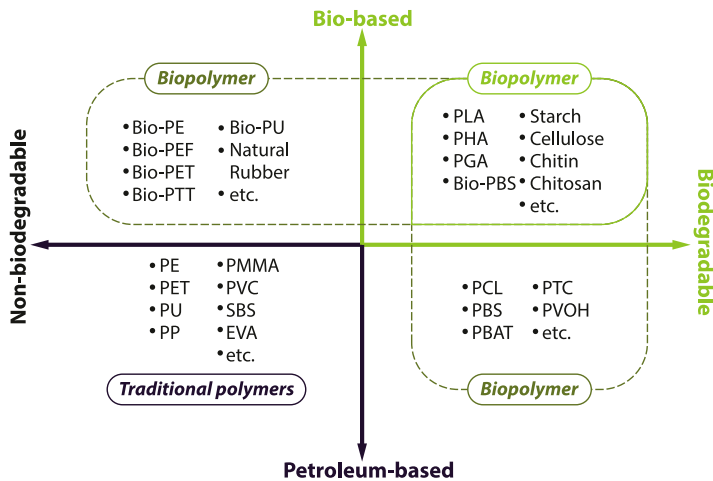


Figure 2. Material coordinate system for biopolymers (European Bioplastics, 2024).

2.1. Classification of biopolymers

The European Plastics organisation has developed a material coordinate system bioplastic classification (European Bioplastics, 2024), widely referenced in reviews (Fredri & Dorigato, 2021, 2023; Goel et al., 2021; Jeremić et al., 2020; Rahardiyani et al., 2023). Similarly, utilising this coordinate system as a general classification of biopolymers is possible. Accordingly, Figure 2 shows the coordinate system redrawn for this study based on the coordinate system prepared by the European Plastics organisation for the general classification of biopolymers. Additionally, for the convenience of readers, the abbreviations used for polymers in this study are listed in Table 1.

As can be seen from Figure 2, biopolymers cover a wide range of materials. Although this classification gives a general idea about biopolymers, a more detailed classification is needed. Therefore, researchers have proposed different classifications that consider various factors such as production method, source, and so on. When 26 different classification systems were reviewed in various review articles and books (Abdul Khalil et al., 2018; Al Mamun & Chen, 2020; Averous & Boquillon, 2004; Babaremu et al., 2023; Balart et al., 2021; Basavegowda & Baek, 2021; George et al., 2020; Gurunathan et al., 2015; Juikar & Warkar, 2023; Kabir et al., 2020; Kamarudin et al., 2022; Khalid & Arif, 2022; Madbouly & Zhang, 2021; Mangal et al., 2023; Niaounakis, 2015; Pinto et al., 2021; Rahardiyani et al., 2023; Rouf & Kokini, 2016; Samir et al., 2022; Shaikh et al., 2021; Sharma et al., 2018; Sid et al., 2021; Vinod et al., 2020; Yoruç & Uğraşkan, 2017; Younes, 2017; Zhong et al., 2020), it was observed that almost every classification has its unique differences. Therefore, a new classification system was organised within this study by combining the biopolymer classifications in the related studies. The related classification is given in Figure 3.

In Figure 3, biopolymers are primarily divided into two groups according to their biodegradability. If a polymer is not biodegradable, it must be synthesised using bio-based monomers to be considered a biopolymer. On the other hand, biodegradable polymers can be divided into two main groups: natural and synthetic. Natural biopolymers are polymers that are readily available in nature or can be simply obtained from nature. The most common examples are cellulose and starch. Furthermore, using PHAs derived from microorganisms is becoming widespread. On the other hand, synthetic biodegradable polymers are biopolymers produced by chemical synthesis from natural or fossil-based sources.

From the above information, it can be concluded that the concept of biopolymers covers a wide range of materials. Since each product has its own area of use and unique properties, examining the products in the classification in detail within the scope of this study would cause a distraction from the purpose of the study. Therefore, a simple grouping of biopolymers was made for this study, and the

Table 1. List of abbreviations used for polymers in this study.

Abbreviation	Definition	Abbreviation	Definition
AESO	Acrylated epoxidized soybean oil	BANR	Buton asphalt natural rubber
Bio-PBA	Bio-polybutyl acrylate	Bio-PBS	Bio-polybutylene succinate
Bio-PE	Bio-polyethylene	Bio-PEA	Bio-polyethyl acrylate
Bio-PEF	Bio-polyethylene furanoate	Bio-PET	Bio-polyethylene terephthalate
Bio-PMA	Bio-poly(methyl acrylate)	Bio-PP	Bio-polypropylene
Bio-PTT	Bio-polytrimethylene terephthalate	Bio-PU	Bio-based polyurethane
Bio-PVC	Bio-polyvinyl chloride	CH	Chemically crosslinked chitosan
CH-MA	Chitosan metakrylamide	CH-PA	Chitosan polianilin
CH-PM	Chitosan poly(2-acrylamido-2-methylpropane- sulfonic acid)	CH-PU	Oxetane substituted chitosan-polyurethane
CLR	Cup lump rubber	CLS	Calcium lignosulfonate
CN-CH	CaNiFe ₂ O ₄ @Chitosan	CO-PU	Castor oil based polyurethane
ECNSL	Epoxidized cashew nut shell liquid	EHL	Enzymatic hydrolysis lignin
EMS	Epoxidized methyl soyate	ENR	Epoxidized natural rubber
ERO	Epoxidized rapeseed oil	ESO	Epoxidized soybean oil
EUG	Eucommia ulmoides gum	EVA	Ethylene vinyl acetate
EWL	Eucalyptus wood lignin	FAL	Formic Acid Lignin
F-RME	Fatty acid methyl esters derived from rapeseed oil	GR	Guayule resin
KL	Kraft Lignin	KL-C	Kraft lignin based polycarboxylic acid
L	Lignin	LC	Lignocellulose
LNR	Liquid natural rubber	L-PR	Lignin synthetic phenolic resin
L-PU	Lignin-based polyurethane	L-TZ	Lignin coated Nano TiO ₂ -ZnO
L-V	Lignin-based vitrimer	LW	Lignin-containing waste/Waste lignin
MLS	Magnesium lignosulfonate	NR	Natural rubber
NRL	Natural rubber latex	OL	Organosolv lignin
OSB	Oxypropylated sugarcane bagasse	PAESO	Polyacrylated epoxidized soybean oil
PBAT	Polybutylene adipate-coterephthalate	PBS	Polybutylene succinate
PBSA	Polybutylene succinate-co-adipate	PCL	Poly-caprolactone
PDL	Partially depolymerised lignin	PE	Polyethylene
PES	Polyethylene succinate	PET	Polyethylene terephthalate
PGA	Polyglycolic acid	PHA	Polyhydroxyalkanoate
PHBV	Poly-3-hydroxybutyrate-co-3-hydroxyvalerate	PHF	Polyhydroxy fatty acid
PL	Pyrolytic lignin	PLA	Poly lactic acid
PLGA	Poly lactide-co-glycolide	PLNR	Pre-vulcanised liquid natural rubber
PMMA	Polymethyl methacrylate	POE	Polyortho ester
PO-PU	Palm oil based polyurethane	PP	Polypropylene
PPHO	Polyphosphazene	PS-PAESO	Polystyrene-acrylated epoxidized soybean oil
PU	Polyurethane	PVA	Polyvinyl acetate
PVC	Polyvinyl chloride	PVOH	Polyvinyl alcohol
PWL	Pinus wood lignin	RME	Oxidized rapeseed oil methyl ester
SA	Sodium alginate	SBS	Styrene-butadiene-styrene
SESO	Sub-epoxidized soybean oil	SL	Soda lignin
ST	Starch	TSNR	Technically specified natural rubber
VEUG	Vulcanised eucommia ulmoides gum	VNR	Vulcanised natural rubber
VO-PU	Vegetable oil based polyurethane	WL	Wood lignin
WLP	Waste leather powder		

main factor here was the frequency of biopolymers preferred in bitumen modification studies in the literature. Accordingly, one main group was formed for materials with more than 30 samples prepared in the literature. Thus, the biopolymers used in bitumen modification were grouped into five different groups, and the information about these groups is given in Table 2. For the integrity of this review study, the colouring and coding in Table 1 were used in all analyses and figures.

2.1.1. Lignin and derivatives

Lignin is the most abundant biopolymer on earth after cellulose and is found in all vascular plants (I. P. Pérez et al., 2019). Lignin-containing plant biomass can be described as a waste material from the

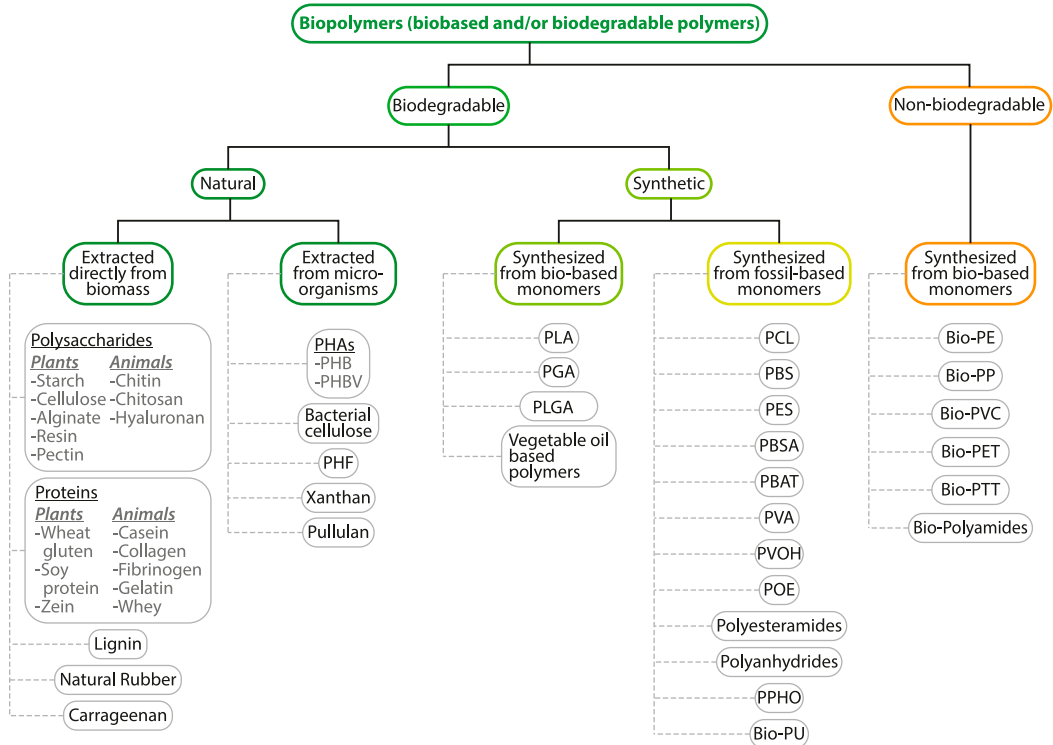


Figure 3. General classification of biopolymers.

paper, wood, and biomass industries or agricultural processes (I. P. Pérez et al., 2019). In other words, lignin can be classified as a waste biomass material (Fatemi et al., 2022a). Depending on the source of the biomass and the extraction method or industrial process used, lignins with different properties can be obtained. The main types of lignin are organosolv lignin, kraft lignin, Klason lignin, enzymatic hydrolysis lignin, sulfonated lignin, and calcium lignosulfonate (Fatemi et al., 2022a). Organosolv lignin is a by-product of the wood processing industry, while kraft lignin is obtained from the process of converting coniferous trees into pulp. Moreover, Klason lignin is obtained from the acid hydrolysis of wood, while enzymatic hydrolysis lignin is obtained from enzymatic hydrolysis to produce bioethanol. Furthermore, the source of sulfonated lignin is black liquor, while calcium lignosulfonate is obtained from the sulfite pulping process, producing paper from softwood.

Lignin is a highly cross-linked and three-dimensional macromolecule chemically composed of three substituted phenols: sinapyl, coniferyl, and p-coumaric alcohols (Al-falahat et al., 2024). Moreover, lignin resembles bitumen resin due to the large rings joined by alkyl chains in its structure (Cheng et al., 2020). Therefore, it is a material compatible with bitumen and is widely used in bitumen modifications in studies in the literature.

2.1.2. Natural rubber and derivatives

Natural rubber is a biopolymer extracted by tapping from the rubber tree and is chemically a dispersion of cis-1,4-polyisoprene in the water phase (Yong Wen et al., 2017). Raw NR is generally not used directly in practice due to its high water content and low resistance to bacterial attack (Yong Wen et al., 2017). The raw NR latex is mixed with ammonia and centrifuged until the dry rubber content is at least 60% to obtain concentrated NR latex (Hoy et al., 2024). On the other hand, cup lump rubber is obtained by coagulation of raw NR latex by bacterial action without a special production process (Mustafa Kamal et al., 2022). In addition to the mentioned NR types, NR-derived additives such as epoxidized natural

Table 2. Grouping of biopolymer types in the context of this study.

Group name	Code	Colors	References in the article pool
Lignin and derivatives	L	Brown and its shades	(Adwani et al., 2023, 2024; Al-falahat et al., 2024; Andiyappan & Kuna, 2023, 2024; Andiyappan et al., 2024; Arafat et al., 2019; Batista et al., 2018; Cai et al., 2022, 2023; Z. Chen et al., 2019; Cheng et al., 2020; de Lima Neto et al., 2023; Duarte Mendonça et al., 2023; Fakhri & Norouzi, 2022; Fatemi et al., 2022a, 2022b, 2021; Fayzrakhmanova et al., 2016; Gao et al., 2020; Gaudenzi, Cardone, Lu, et al., 2023a; Gaudenzi et al., 2022; Ghabchi, 2022; Guo et al., 2020; D. Hu et al., 2022; Kalampokis et al., 2022; Khadim & Ahmad, 2024; R. Li et al., 2020; Yiming Li & Lv, 2023; Y. Li et al., 2023; Luz et al., 2021; Ma et al., 2022; McCready & Williams, 2008; Miao et al., 2019; Nahar et al., 2023; Norgbey et al., 2020; I. Pérez et al., 2020; I. P. Pérez et al., 2019; Qiu et al., 2024; Rachman et al., 2023; Ren et al., 2021; Rezazad Gohari et al., 2023; X. Shi & Xu, 2023; Song et al., 2023; Su et al., 2023; Sun et al., 2023; Sundstrom et al., 1983; D. Wang et al., 2019; K. Wang et al., 2023; J. Wu et al., 2021; Xie et al., 2017; Xin et al., 2016; C. Xu et al., 2021; G. Xu et al., 2017; Xue et al., 2022; Yilmaz & Ugurlu, 2023; Yu et al., 2021; Zabelkin et al., 2019; Zahedi, Zarei, & Zarei, 2020; Zahedi, Zarei, Zarei, et al., 2020; M. Zarei, Abdi Kordani, ghamarimajd, et al., 2022; M. Zarei, Abdi Kordani, & Zahedi, 2022; E. Zhang et al., 2024; R. Zhang et al., 2021; Y. Zhang et al., 2019; Y. Zhang et al., 2020; Y. Zhang, Liu, et al., 2022; Y. Zhang, Si, et al., 2023; S. Zhou et al., 2023; Ziaee et al., 2023)
Natural rubber and derivatives	NR	Green and its shades	(Abd El Rahman et al., 2012; Abdulrahman et al., 2021; Al-Mansob et al., 2014, 2016, 2022; Al-Mansob, Ismail, Rahmat, et al., 2017; Al-Mansob, Ismail, Yusoff, et al., 2017; Al-Mutlaq & Mahal, 2021; Abdunaser M. Al-Sabaeei et al., 2020; Albuaymi et al., 2023; Ansari et al., 2023; N. M. Azahar et al., 2021; Norfazira Mohd Azahar et al., 2019; Begam Rasheda et al., 2022; Hadiwardoyo et al., 2016; Hazoor Ansari et al., 2022; Hoy et al., 2024; X. Hu et al., 2020; Ibrahim et al., 2024; Ismail et al., 2012; Jitsangiam et al., 2021; F. Liu et al., 2023; Mustafa Kamal et al., 2022; Nair et al., 1998; Poovaneshvaran et al., 2020, 2023; Ramadhan et al., 2020; Ramadhani et al., 2023; Safaeldeen et al., 2022; Sani et al., 2020, 2023; Sani, Mohd Hasan, et al., 2021; Sani, Shariff, et al., 2021; Saowapark et al., 2019; E. Shaffie et al., 2018; Shafii et al., 2017, 2018; Subagio et al., 2021, 2022; Suwanto et al., 2023; Usman et al., 2019; Yong Wen et al., 2017; Wittitanapanit et al., 2021; Y. Yan et al., 2022, 2024; Yousefi, 2002a)
Natural oil-based polymers	NO	Yellow and its shades	(Arabzadeh et al., 2022; Caputo et al., 2018; Chen et al., 2017, 2018, 2020; C. Chen et al., 2019; Fuhaid et al., 2018; Hrynchuk et al., 2019; Iwański et al., 2022, 2023; Kowalski et al., 2017; Król et al., 2016; Król et al., 2017; J. Lu et al., 2023; Q. Lu et al., 2021; Nykypanchuk et al., 2013; Joseph H. Podolsky, Saw, et al., 2021; Quan et al., 2024; Radziszewski et al., 2023; Starchevskyy et al., 2021; Uchoa et al., 2021)
Biobased polyurethanes	PU	Blue and its shades	(Cuadri et al., 2013, 2014a, 2014b; Gong, Liu, Wan, et al., 2023; Gong, Liu, Wang, et al., 2023; Kazemi et al., 2024; Kazemi, Goli, et al., 2022; Kazemi, Mohammadi, et al., 2022; Kök et al., 2021; Meng et al., 2022; Peng et al., 2020; Shirzad et al., 2019; K. Wei et al., 2024; L. Xia et al., 2023, 2016)
Other biopolymers	Others	Gray and its shades	(Ahmed et al., 2022; Ai et al., 2011; G. Airey et al., 2008; Airey et al., 2016; G. D. Airey et al., 2008; A. I. Al-Hadidy & Tan, 2009; A. I. Al-Hadidy & Yi-qiu, 2010; Araújo et al., 2017; Caputo et al., 2023; Cui et al., 2023; Hemida & Abdelrahman, 2021b; Jailani et al., 2021; N. Li, Hu, et al., 2022; N. Li, J. Xu, et al., 2021; Malinowski et al., 2023a; Malinowski et al., 2023b; Malinowski et al., 2022; Porto et al., 2019; Tabaković et al., 2023; H. Wang et al., 2022; C. Xia et al., 2022; S. Yan et al., 2023)

rubber (ENR) (Abd El Rahman et al., 2012; Al-Mansob et al., 2014, 2022; Al-Mansob, Ismail, Yusoff, et al., 2017; Ismail et al., 2012) and vulcanised natural rubber (VNR) (F. Liu et al., 2023; Y. Yan et al., 2024) have been used in bitumen modification.

2.1.3. Natural oil-based polymers

Natural oils such as soybean oil, canola oil, sunflower oil, palm oil, and rapeseed oil are widely used as modifiers, rejuvenators, and extenders in bitumen, as well as monomers in polymerizations (Quan et al., 2024). These oils, known as triglyceride oils, are a vital renewable resource for biopolymers. Triglyceride oils enable the production of biopolymers with rubbery and flexible properties due to the functionality of their long fatty acid chains (Chen et al., 2018). Most of the studies using natural oil-based polymers for bitumen modification used polymers containing rapeseed methyl esters (Iwański et al., 2022, 2023; Kowalski et al., 2017; Król et al., 2016; Radziszewski et al., 2023) and epoxidized soybean oils (Caputo et al., 2018; Chen et al., 2018, 2020; Fuhaid et al., 2018; Tabaković et al., 2023). It should be noted here that natural oils are a very broad concept and the use of bio-oils in bitumen modification is quite common. However, within the scope of this study, in order to be considered as a polymer and to include the relevant study in the article pool, it was taken care that the natural oils should have undergone a polymerisation process.

2.1.4. Biobased polyurethanes

Polyurethanes are supramolecular polymers with main components such as isocyanate and polyol (Kazemi, Goli, et al., 2022). The ability of supramolecular polymers to form hydrogen bonds more than once gives them the ability to cure more than once at room temperature (Kazemi, Goli, et al., 2022). Therefore, the use of polyurethane in bitumen modifications for bitumen recovery has been of interest. However, the fact that conventional polyester polyols and polyether polyols used in polyurethane production are mostly produced from non-renewable petrochemical sources has encouraged researchers to produce biobased polyurethanes (Gong, Liu, Wan, et al., 2023). Accordingly, biobased polyurethanes based on castor oil (Cuadri et al., 2013, 2014a, 2014b; Kazemi, Mohammadi, et al., 2022; K. Wei et al., 2024; L. Xia et al., 2016), palm oil (Kök et al., 2021), vegetable oil (L. Xia et al., 2023) and lignin (Peng et al., 2020) were produced and used in bitumen modification.

2.1.5. Other biopolymers

The main biopolymers used in bitumen modification are mentioned above. However, different biopolymers have been used in some studies other than the mentioned polymers. One of these is *Eucommia ulmoides* gum (EUG), also known as gutta-perka and balata gum. EUG has the main composition of trans-1,4-polyisoprene (TPI) and is an isomer of natural rubber (Cui et al., 2023). In some studies on bitumen modifications (Cui et al., 2023; N. Li, Hu, et al., 2022; N. Li, J. Xu, et al., 2021; H. Wang et al., 2022), vulcanised EUG (VEUG) obtained by improving the properties of EUG by vulcanisation has been used. Another biopolymer used in bitumen modification is starch. Starch is commonly found in plants and contains the carbohydrate polymers amylose and amylopectin (A. I. Al-Hadidy & Tan, 2009). Starch is a free-flowing powder form material and can be used directly in bitumen modifications (Ai et al., 2011; A. I. Al-Hadidy & Tan, 2009; A. I. Al-Hadidy & Yi-qiu, 2010; Porto et al., 2019). Another polymer used in bitumen in a limited number of studies is chitosan. Chitosan is a derivative of chitin and chemically contains β -(1 \rightarrow 4) 2-acetamido-2-deoxy-D glucose units (Malinowski et al., 2023a; Malinowski et al., 2023b). Apart from the mentioned biopolymers, biopolymers such as cellulose, pectin, carrageenin, Arabic gum, corn flour, guar gum, and xanthan gum have also been used in bitumen modification (Porto et al., 2019).

3. Methodology

3.1. Literature review and data organisation

This review adopts the Systematic Quantitative Literature Review (SQLR) method to provide a structured, replicable analysis. Unlike traditional narrative reviews, SQLR minimises author bias and enables

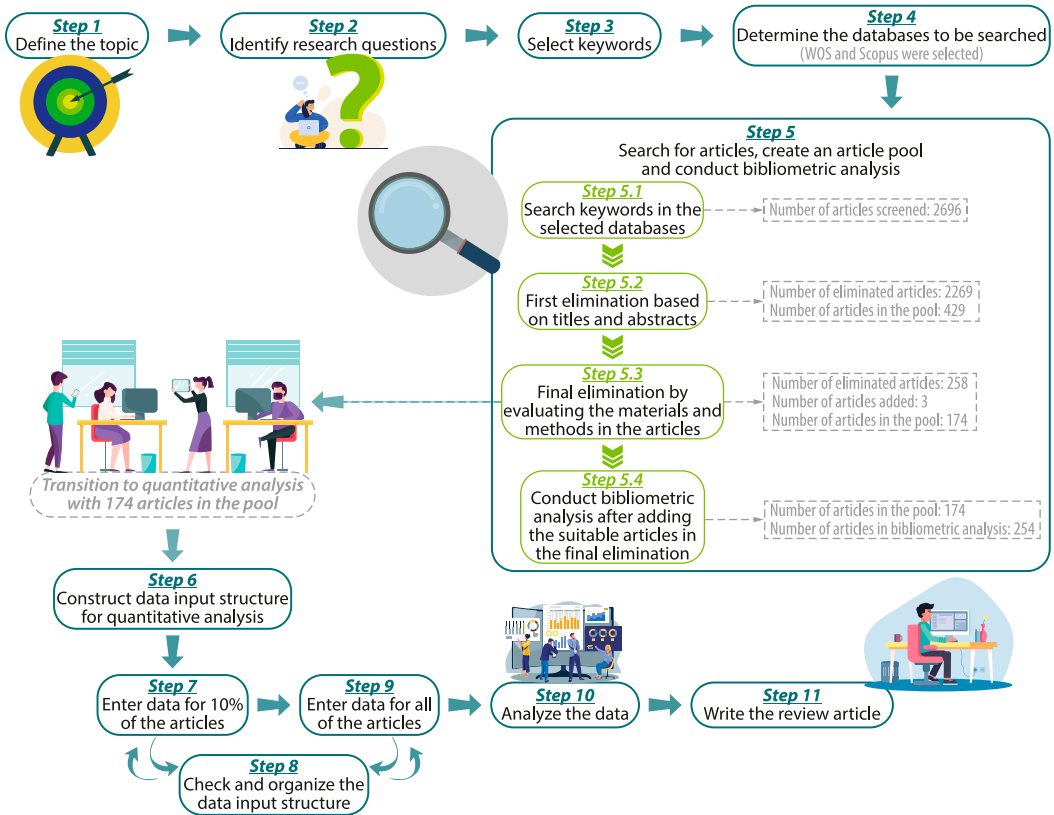


Figure 4. Methodology of this study.

quantitative evaluation, helping to identify research gaps (Borenstein et al., 2021; Petticrew, 2001; Pickering & Byrne, 2014; Şahan et al., 2023). It also offers insights into study locations, methodologies, and findings, guiding future research directions.

Pickering and Byrne (2014) outlined a 15-step SQLR method for social sciences. Şahan et al. (2023) updated and adapted it for bitumen modification studies. In this study, a new, more advanced review methodology is prepared by combining the methodology described by Şahan et al. (Şahan et al., 2023) with bibliometric analysis. Accordingly, the steps of the review methodology proposed in this study are given in Figure 4.

The first step in the literature review is to identify the topic and draw certain boundaries for this topic. Accordingly, the topic of the study was determined as ‘the use of biopolymers in bitumen modification’. As the principal limit of the topic, especially considering the studies in which quantitative analysis will be carried out, it was stipulated that the article to be selected for the article pool must have added a biopolymer to a base bitumen using a wet method. It should be noted that the addition of biopolymers, particularly lignin fibre, calcium alginate, and PVA fibre, to asphalt mixtures via the dry method is a widely used approach. However, since this method primarily impacts asphalt mixture performance rather than bitumen performance, studies utilising this approach were excluded from this review. After that, the second step was to determine the research questions. While selecting articles for the article pool, the questions expected to be answered in an article are given below:

- Where and when was the study conducted?
- Within the context of the study, was bitumen performance or asphalt mixture performance analyzed?

Table 3. Keywords used in the literature review.

Keyword Group	Keywords
KWG1	'biopolymer*'; 'bio*' AND 'polymer*'; 'bio*' AND 'plastic*'; 'bio*' AND 'modif*'
KWG2	'lignin*'; 'ligno*'
KWG3	'natural rubber'; 'cup lump rubber'; 'crepe rubber'; 'ribbed smoked sheet'
KWG4	'epoxid*' AND 'oil'; 'bio*' AND 'polyurethane'; 'oil*' AND 'polyurethane'
KWG5	'polysaccharide*'; 'starch'
KWG6	'alginate'; 'gum'; 'agar'; 'chitin'; 'chitosan'; 'pectin'; 'hyaluronan'; 'casein'; 'collagen'; 'fibrinogen'; 'gelatin'; 'whey'; 'carrageenan'; 'xanthan'; 'pullulan';
KWG7	'polylactic acid'; 'poly lactic acid'; 'PLA'; 'PHB'; 'PHBV'; 'PGA'; 'polycaprolactone'; 'PCL'; 'PBS'; 'poly butylene succinate'; 'PES'
KWG8	'polyvinyl acetate'; 'poly vinyl acetate'; 'poly vinyl alcohol'; 'polyvinyl alcohol'; 'PVA'; 'PVOH'

- Which biopolymers were added to the bitumen and which additive ratios were selected?
- What are the preparation conditions for biopolymer modified bitumen?
- Which experimental methods were utilised in the study?
- What are the quantitative results obtained from the study?

In the next step, the keywords for the literature review were determined. A total of 94 keyword combinations were generated by combining the biopolymer types indicated in Figure 3 with the words 'asphalt*' and 'bitumen*'. The keywords were used by grouping, and the related groups are given in Table 3.

In the fourth step, Scopus and Web of Science, the most commonly used engineering databases, were selected for the literature search. The fifth step involves keyword-based searching for articles in the identified databases. Limitations used in the databases are essential for the general validity of the study. While searching both Scopus and Web of Science databases, the document type was limited to 'article', the document language to 'English' and the occurrence of specific keywords to 'abstract, keywords, and title'. Therefore, it is possible to access articles that are more relevant to the subject. The results obtained from the searches (Step 5.1) according to keyword groups and databases and the number of articles eliminated after title and abstract reading are given in Table 4.

From 2696 screened articles, 429 were initially selected based on title and abstract relevance (Step 5.2). A final screening of materials and methods reduced this to 174 articles (Step 5.3). The methodology then followed two paths: bibliometric analysis (Step 5.4) and data structuring for collection (Step 6).

In order to conduct the bibliometric analysis (Step 5.4), the VOSviewer (VOSviewer, 2024) application and Scopus list analysis were used. Accordingly, some of the eliminated papers (Abdulrahman et al., 2023; Aguirre et al., 2019, 2020; A. Al-Hadidy et al., 2008; Arabzadeh et al., 2021; Bindu et al., 2020; Das & Panda, 2020; Deng et al., 2017; Gaudenzi, Ingrassia, Cardone, et al., 2023; He et al., 2018, 2022, 2023; He, Yu, Du, et al., 2019; He, Yu, Hu, et al., 2019; Hemida & Abdelrahman, 2019, 2021a, 2023; Huang et al., 2022; Ibrahim et al., 2020; Jiang et al., 2023; Jimenez del Barco-Carrion et al., 2017; Jimmyanto et al., 2024; Jovanović et al., 2019; Kök & Gürçay Özdemir, 2023; Kou et al., 2020, 2022; Kuang et al., 2018; Kuanusont et al., 2021; C. Li et al., 2023; J. Li, F. Meng, et al., 2021; J. Li, Z. Su, et al., 2021; J. Li, Wang, et al., 2022; J. Li et al., 2020; N. Li, H. Zhan, et al., 2021; Q. Li, H. Zhang, & Z. Chen, 2021; Q. Li, H. Zhang, C. Shi, et al., 2021; Yunyu Li et al., 2024; Z. Li et al., 2021; Z. Li & Li, 2013; Z. Li et al., 2022; Z. Li et al., 2020; Lin et al., 2023; J. Liu et al., 2021; Pan et al., 2023; Perumal et al., 2023; Joseph H. Podolsky, Chen, et al., 2021; Joseph H. Podolsky et al., 2020; Joseph H. Podolsky et al., 2018; Puculek et al., 2021; Putra et al., 2017; Roziafanto et al., 2020; Ekarizan Shaffie et al., 2016; K. Shi et al., 2022; Shu et al., 2018; Staver et al., 2021, 2022; S. Wang, Liu, et al., 2021; T. Wang, Xu, et al., 2021; W. Wang et al., 2020; C. Wei et al., 2020, 2022; Yalu Wen et al., 2024; M. Wu, Cai, et al., 2022; M. Wu et al., 2014; M. Wu, Zhao, et al., 2022; Xiao et al., 2024; Xing et al., 2022; S. M. Yan et al., 2020; Y. Yan et al., 2023; Yang et al., 2024; Yin et al., 2013; Yousefi, 2002b; Yuniarti, 2019; A. Zarei et al., 2019; M. Zarei et al., 2021, 2023; F. Zhang, Liu, et al., 2023; L. Zhang, Hoff, et al., 2022; X. Zhou et al., 2015; Zhu et al., 2022) were included in the bibliometric

Table 4. Number of screened, eliminated and selected articles according to databases.

Keyword Group	Data-base	Screening Date	Number of Scanned Articles	Number of Duplicates	Number of Articles		Number of Selected Articles	Total Number of Selected Articles
					Duplicates Removed	Total Number of Scanned Articles		
KWG1	Scopus	5.05.2024	1013	0	1013	1013	110	110
KWG1	WOS	5.05.2024	1022	646	376	1389	15	125
KWG2	Scopus	7.05.2024	415	75	340	1729	113	238
KWG2	WOS	9.05.2024	530	329	201	1930	6	244
KWG3	Scopus	9.05.2024	106	11	95	2025	62	306
KWG3	WOS	9.05.2024	108	71	37	2062	4	310
KWG4	Scopus	9.05.2024	102	32	70	2132	28	338
KWG4	WOS	9.05.2024	115	82	33	2165	2	340
KWG5	Scopus	10.05.2024	73	20	53	2218	8	348
KWG5	WOS	10.05.2024	71	52	19	2237	0	348
KWG6	Scopus	10.05.2024	238	48	190	2427	61	409
KWG6	WOS	10.05.2024	239	174	65	2492	2	411
KWG7	Scopus	10.05.2024	29	2	27	2519	1	412
KWG7	WOS	10.05.2024	41	27	14	2533	0	412
KWG8	Scopus	10.05.2024	153	11	142	2675	14	426
KWG8	WOS	11.05.2024	72	51	21	2696	3	429

analysis to assess the general information on the topic more accurately. For example, some studies did not include base bitumen but directly examined composite modification. Although such studies are beyond the boundaries of this review in terms of conducting quantitative analyses, they were included only at this stage to make the bibliometric analysis more meaningful. Accordingly, the bibliographic information of 254 articles was inputted into the WOSviewer, and a list was created for these articles in Scopus. Subsequently, bibliometric analyses were conducted, and the general evaluation of the studies was based on these analyses.

In Step 6, the data input structure for quantitative analyses was created. The data input structure was prepared by creating different tables. In the first table, each row represents an article in the article pool, and the columns describe the details of the materials in the article. Accordingly, data on the types and utilisation ratios of biopolymers were collected in the articles. Then, in the second large table, research was conducted on the experimental methods in the articles. In this table, the columns represent different experiments. The presence or absence of experimental methods in an article was recorded by writing '1' or '0' in the intersection cell. Thus, a general overview of the materials and experimental methods in the literature is provided. As shown in Figure 4, after the data entry structures are prepared in Step 6, Steps 7–9 form a loop. In Step 7, 10% of the articles in the article pool are first entered into the table as data. During data entry, if the columns of the tables are insufficient, for example, if an experimental method needs to be added to the table, the data entry structure must be reorganised (Step 8). This situation should be checked repeatedly to ensure that the overall data for all articles is entered (Step 9).

After all data has been entered into the main data tables, the analysis process can be initiated by passing on to Step 10. At this point, it is possible to evaluate an overview of the studies in the literature with the data already available in the main data tables. Indeed, it is possible to gather much information such as in which studies, which types of biopolymers were used, how often, in which ratios, and which experimental methods were preferred and how often. On the other hand, a more detailed analysis has been undertaken to make an advanced quantitative assessment.

3.2. Quantitative comparative performance evaluation

In order to compare the studies in the literature with a more detailed quantitative data, the analyses were first categorised according to the experiments. For each experiment, the results of the experiments were collected from the studies that included the relevant experiment in the article pool by taking into account the criteria specified for the experiments. While collecting the experimental results, WebPlotDigitizer (Rohatgi, 2024) programme was used to digitise the graphs in the studies. In order to ensure cross-validation, data extraction was performed independently by two authors, and discrepancies were assessed for consistency. A threshold of $\pm 2\%$ was established, and if deviations exceeded this limit, the readings were repeated using the software to enhance data reliability.

However, directly comparing the collected test results may lead to misunderstandings. Indeed, the influence of the characteristics of base bitumen on the numerical results of each experiment is too significant to be ignored. Considering this, percentage change (% change) values were used to normalise the data obtained from a study. When calculating the % change values, the experimental result of the base bitumen in the article from which the data was taken and the result of the experiment to be transformed (the result of the bitumen modified by adding a certain amount of biopolymer) were used. Accordingly, calculations were made with the help of Equation (1).

$$\%Change = \frac{\text{Modified bitumen test result} - \text{Base bitumen test result}}{\text{Base bitumen test result}} \times 100 \quad (1)$$

Within the context of this study, box plots were utilised for the visual evaluation of all the data obtained. Accordingly, an example of a box plot for a normally distributed dataset is given in Figure 5. This figure illustrates the meaning of the elements in the box plot. Thanks to this type of graph, it is

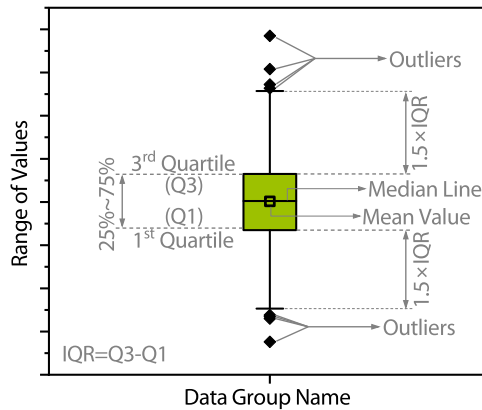


Figure 5. Anatomy of a typical box plot.

possible to have much information about a dataset simultaneously. Thus, it is possible to observe the values of the 25–75% range with the borders of the box, the median value of the data with the line in the centre of the box, and the mean value of the dataset with the small square in the centre of the box. Additionally, outliers are the values in the dataset that are above the limit of $1.5 \times \text{IQR}$ above the third quartile value and below the limit of $1.5 \times \text{IQR}$ below the first quartile value. Outliers can be considered as values that are outside the general distribution of the dataset. Dataset of the experimental results obtained using Equation 1 will be evaluated comparatively on box plots. Therefore, it is aimed to provide the reader with a more detailed, comprehensive, and understandable presentation of the analysis results.

3.3. Experimental methods chosen for performance evaluation

Many standardised tests and experimental setups for bituminous binders and asphalt mixture samples help to investigate and determine the physical, chemical, morphological, and rheological properties of the binders. In this study, as will be discussed in section 4.1.5, the most frequently chosen experimental methods in the literature were analyzed, and the quantitative analysis was carried out according to the results obtained. However, the relevant experimental methods are briefly mentioned in this section so the reader can get brief information about the analyzed test methods. Furthermore, in this study, all experimental data from the articles reviewed in the literature were analyzed with certain limitations. These restrictions were sometimes based on a specific temperature value in the tests, sometimes by selecting a specific parameter for that test to make a more accurate comparison and quantitative evaluation between the articles under the same conditions.

3.3.1. Bitumen tests

For the determination of consistency, which is one of the physical properties of bituminous binders, the penetration test (ASTM D5/D5M-20, 2020), and the softening point test (ASTM D36/D36M-14, 2020), are frequently applied. Furthermore, the ductility value (ASTM D113/D113M-17 (2023)e1, 2023) gives an idea about flexibility of the bitumen, while the elastic recovery test (ASTM D6084/D6084M-21, 2021), gives an idea about the degree of its elasticity. Additionally, the workability of bitumen is critical in practice, and viscosity measurements evaluate this property. Accordingly, the rotational viscosity test (D4402/D4402M-23, 2023) is frequently used in studies.

The chemical structure of bitumen is quite complex and various test methods are preferred to investigate this structure. Among these methods, Fourier transform infrared (FTIR) analysis has been

frequently used in studies due to its practicality and accuracy. Thanks to this test method, it is possible to quantitatively evaluate the types and strength of bonds occurring in the internal structure of bitumen.

The rheological properties of bitumen are of vital importance in the evaluation of its performance. Thanks to the rheological properties determined using dynamic shear rheometry (DSR), parameters representing the asphalt pavement performance of bitumen under direct field conditions, such as rutting resistance and fatigue resistance at high and medium temperatures, are obtained (ASTM D7175-23, 2024). By using these parameters, performance grading (PG) of bitumens is made (ASTM D6373-23, 2023). In addition, due to the controversial usability of the relevant parameters specifically for modified bitumens, it is possible to determine the rutting resistance of modified bitumens more reliably thanks to the multiple stress creep and recovery (MSCR) (ASTM D7405-24, 2024) test developed. Furthermore, thanks to this test, the PG classification was developed and associated with traffic loads, and a specification was developed for the classification of bitumens according to the traffic levels they can serve (ASTM D8239-23, 2023). On the other hand, the performance of bitumens at low temperatures is also essential in the classification specifications. In this regard, the bending beam rheometer test (BBR) (ASTM D6648-08, 2018), in which the strength of a bitumen beam under bending load at low temperatures is determined, is widely used.

3.3.2. Asphalt mixture tests

In order to evaluate the performance of bituminous binders on a macro-scale, asphalt mixture samples are prepared by mixing bitumen and aggregate with specific procedures, and the prepared macro-sized samples are subjected to various tests. Thanks to these tests, it is possible to observe how the asphalt pavement will perform in service based on the performance of bituminous binders in the asphalt mixture. Accordingly, many standardised tests are used to determine the different properties of asphalt mixtures. One of the most essential properties of asphalt mixtures is deformation resistance. Accordingly, Marshall stability (MS) and flow values (ASTM D6927-22, 2022), based on the strength and deformation values of asphalt mixture specimens under constant load, have been among the basic properties used in asphalt pavement design for many years. Additionally, with the advancement of technology, it is possible to design experimental equipment that more closely simulates the in-service condition of asphalt pavements. Accordingly, the rutting test (EN, 12697-22 + A1, 2023), which directly simulates the in-service wheel loading of the pavement, makes it possible to evaluate the rutting resistance of asphalt mixture specimens.

Another vital characteristic of asphalt mixture samples is their tensile strength. An indirect tensile strength test (EN, 12697-26 + A1, 2022) makes determining the tensile strength of asphalt mixture samples possible. On the other hand, the resilient modulus test (ASTM D7369-20, 2020), which was determined after the development of the apparatus used in the indirect tensile strength test to be able to perform repeated loading, has become the most critical parameter of asphalt pavement design today. Thanks to a similar test setup, it is also possible to observe the fatigue strength of asphalt mixture specimens with the indirect tensile fatigue test (EN, 12697-24, 2018).

The extreme climatic conditions exposed to asphalt pavements during service directly affect their performance. Therefore, asphalt pavements must have high durability. Accordingly, it is a frequently used research method to determine the resistance of asphalt mixture samples to different climatic effects by subjecting them to ITS and MS tests after conditioning with different types of conditioning. Among the conditioning types, the most commonly used methods are dry-wet and freeze-thaw conditioning. After producing two mixture samples with the same design, one sample is conditioned, and the other is left in its original condition. The two specimens are then subjected to ITS or MS tests, and the indirect tensile ratio (TSR) or residual Marshall stability (RMS) values are determined by comparing the values between the two specimens. These values make it possible to evaluate the durability of asphalt mixture samples.

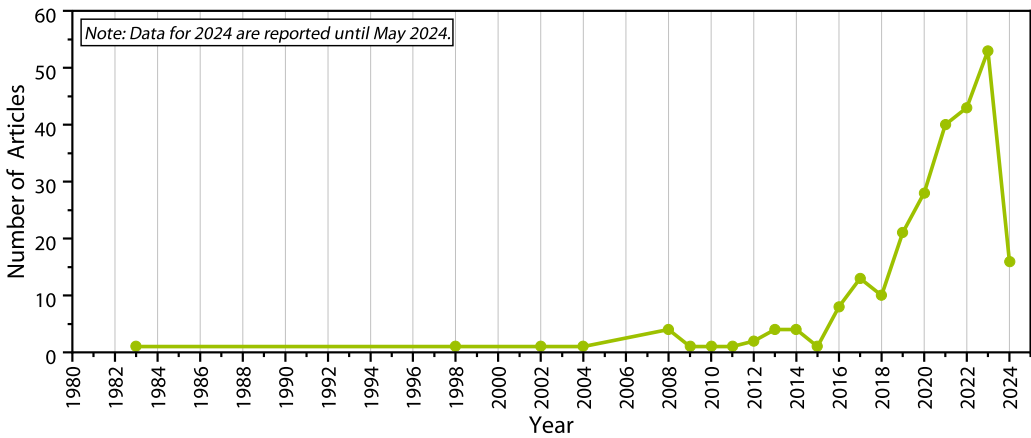


Figure 6. Number of articles by years.

4. Results and discussion

4.1. Overview

In this section, it is aimed to compile general information about the studies before proceeding to the analysis of the experimental results carried out in the studies in the literature. Accordingly, bibliometric analysis was used to produce relationship networks between the countries where the studies were conducted, authors, keywords, and journals in which the studies were published. Furthermore, the basic parameters of bitumen modification studies, such as neat bitumen, biopolymer types, modified bitumen preparation conditions, and experimental methods used in the studies, are also evaluated in this section.

4.1.1. Bibliometric analysis

Bibliometric analysis is a research method that can summarise the current state of knowledge in a particular field, discover new trends, and quantify the number of publications and research (Arias-Cárdenas et al., 2024; Marathe et al., 2024). Thanks to this method, it is possible to examine the studies on a specific topic in the literature under different topics such as year, country, author, keyword, and journal. A more detailed evaluation of the relationship networks between authors, countries, and keywords can also be made.

In the bibliometric analysis of this study, the publication years of the articles in the pool were examined first, and the result obtained is given in Figure 6. Accordingly, the first study in the literature within the boundaries of this study was conducted by Sundstorm et al. (Sundstrom et al., 1983) in 1983. However, only 17 academic studies on biopolymer-modified bitumen were published between 1983 and 2014. The main reason for this may be that biopolymers had yet to gain popularity at that time. However, since 2016, there has been a significant increase in the number of studies on biopolymer-modified bitumen, and this rise has been maintained constantly. Although only the studies published in the first five months of 2024 could be analyzed in this study, 16 articles on the subject have already been published in 2024. In line with all these results, it is possible to say that the use of biopolymers in bitumen modification is a quite new topic that has attracted the interest of researchers.

Another element of bibliometric analysis is the distribution of studies by countries and interaction network analysis based on the countries where the institutions of the responsible authors of the studies are located. The results obtained from the analysis are presented in Figure 7. When the number of articles by country in Figure 7 (a) is examined, it is understood that most of the articles in the pool are conducted in China. It is followed by the USA and Malaysia, respectively. These results prove the high level of support given to studies on this topic in the countries directly involved. On the other hand,

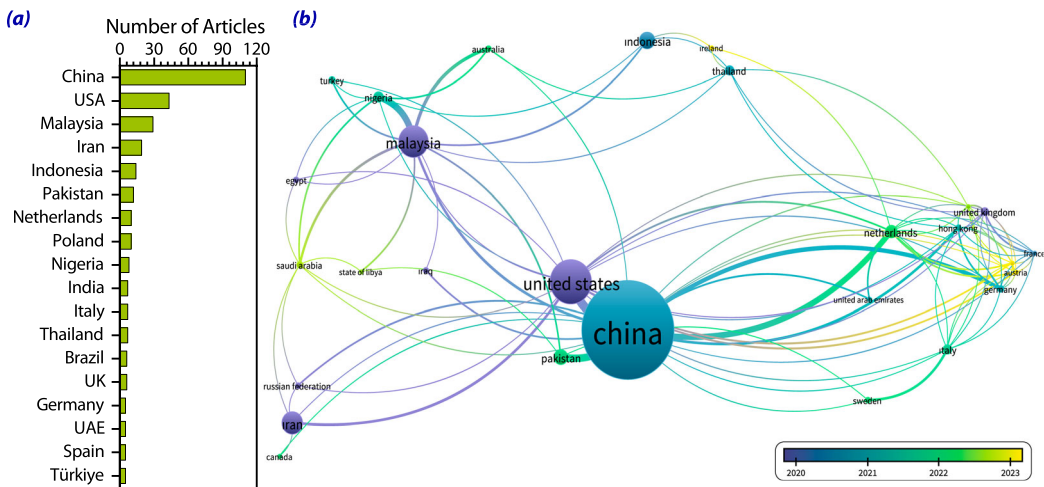


Figure 7. Distribution of studies by countries: (a) number of articles and (b) relationship network between countries.

evaluating several pieces of information simultaneously from the relationship map in Figure 6(b) is possible. The size of the circle in a network of relationships indicates the number of occurrences of the word to which the circle belongs (in this case, the country where the study was conducted) in the article pool. Furthermore, the fill colour of the circle means the average publication year of the articles belonging to the circle. Finally, the lines between the circles indicate the relationship between the circles, such as citing and collaborating, and the thicker the line, the stronger the relationship. In light of all this information, Figure 7(b) shows that China is at the centre of the network map due to the high number of articles. Additionally, the USA is located next to China. The average publication years of the studies conducted in the USA and Malaysia are older than those of the other studies. This indicates that these countries are among the leading countries in studies on using biopolymers in bitumen modification. On the other hand, the countries on the right side of the network map have lighter-colored circles, and considering that the majority of these countries are European countries, researchers in European countries have just started to work in this field. Finally, considering that studies are being conducted in many different countries, biopolymers in bitumen modification are a topic of worldwide interest.

The results of the bibliometric analysis conducted on the authors who wrote articles on the use of biopolymers in bitumen modification are given in Figure 8. When the number of articles by authors in Figure 8(a) is analyzed, it is seen that 'William, R. C.' is the author with the highest number of articles on the subject, followed by 'Podolsky, J. H.' and 'Cochran, E. W.'. Therefore, it is possible to consider these authors as the pioneers in the related field. On the other hand, in the relationship network between authors in Figure 8(b), there is a clustering of Chinese authors on the left side of the figure. This shows that another reason for the high number of articles written in China is the high number of researchers in that country. However, in the upper part of the figure, 'Williams, R. C.' and his team are clustered, while in the right part of the figure, 'Yusoff, N. I. Md.' and 'Al-Mansob, R. A.' are grouped with mostly Malaysian authors. These clusters and groupings indicate a close relationship between these authors in terms of co-authorship and co-citation. Furthermore, 'Chen, C.' appears to be a bridge between US and Chinese authors, and 'Mohd Hasan, M. R.' and his team appear to be a bridge between Malaysian and Chinese authors. This result implies that these authors provide a link between the two groups through both citations and collaborations.

4.1.2. Base bitumen used in the studies

The selection of unmodified (base) bitumen grades is of vital importance both in asphalt pavement applications and scientific research. In field applications, the choice of base bitumen grade is usually

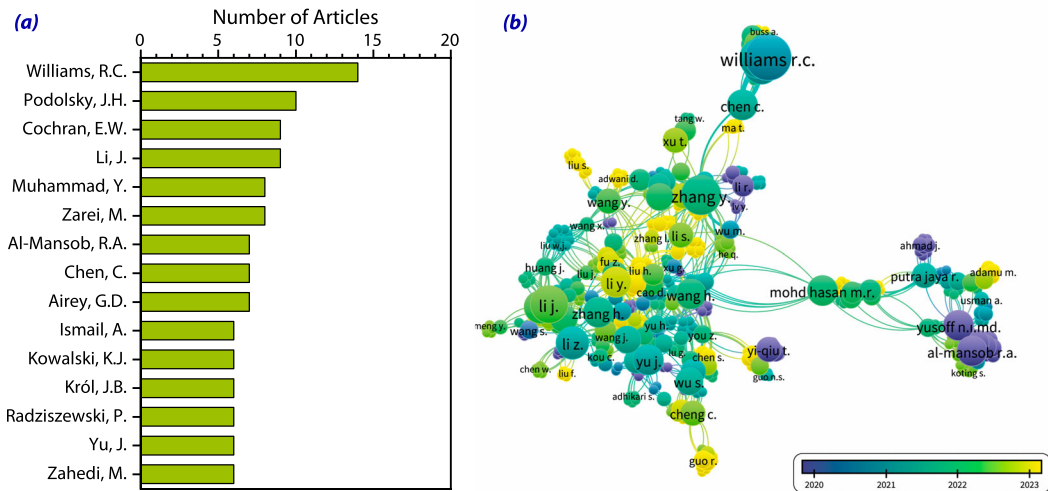


Figure 8. Distribution of studies by authors: (a) number of articles by authors and (b) relationship network between authors.

directly related to the climatic conditions of the country where the application will be carried out and the conventional bitumen grades used in existing applications in the country. On the other hand, in academic studies, a researcher who will choose a base bitumen grade can benefit from base bitumen grades used in similar studies on the subject of the study. Therefore, in this section of the study, the preferred base bitumen grades are compiled to provide information for researchers who intend to conduct a study using biopolymers in bitumen modification.

Two different types of classification were considered for the classification of base bitumen: the traditional penetration (Pen) classification and the relatively more recent performance grading (PG). While organising the PG data, only high-temperature PGs were compiled to ensure a standard data organisation between studies since some articles only consider the high-temperature grade. The results obtained from the analysis are given in Figure 9. When the distribution of base bitumen according to penetration grades is analyzed in Figure 9(a), it is seen that in 46.1% of the studies, Pen 50/70 penetration grade base bitumen was preferred. This indicates that Pen 50/70 is the most preferred bitumen penetration grade for biopolymer-modified bitumen. Additionally, the Pen 70/100 penetration grade is the second most frequently used base bitumen penetration grade, with a rate of 25.7%. Moreover, it is seen that only PG is used in 16.2% of the articles. On the other hand, when the distribution of articles by PG in Figure 9(b) is analyzed, it is observed that the PG of base bitumen is not mentioned in 62.3% of the studies. This situation is related to penetration classification being chosen more frequently in practice worldwide because it is more practical and economical, although the PG system is a superior classification system designed to provide higher quality asphalt pavements based on performance. On the other hand, it is understood that PG64 high-temperature grade base bitumen was used most frequently in the studies in the article pool, with a rate of 22%.

4.1.3. Types of biopolymer additives and frequency of use

It is essential to examine the frequency of bitumen modification with which biopolymers are used in the literature to evaluate which material is popular and to see the gap in the literature. Accordingly, the modified bitumen prepared in each article in the pool was analyzed by biopolymer type and addition rate. Each additive ratio in a study was counted as one sample, and the number analysis was carried out based on the number of samples. In other words, in a study (Gao et al., 2020) where lignin was added to base bitumen at rates of 2, 4, 6, and 8%, it was assumed that four lignin-modified bitumen samples were produced. Accordingly, the results obtained from the analyses are given in Figure 10. When the distribution of sample numbers according to additive types in Figure 10(a) is examined,

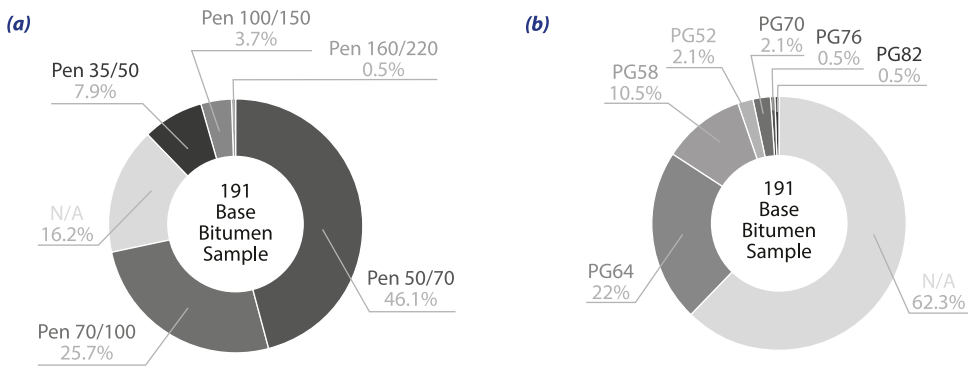


Figure 9. Distribution of base bitumen grades in the studies: distribution (a) by penetration grades and (b) by PGs.

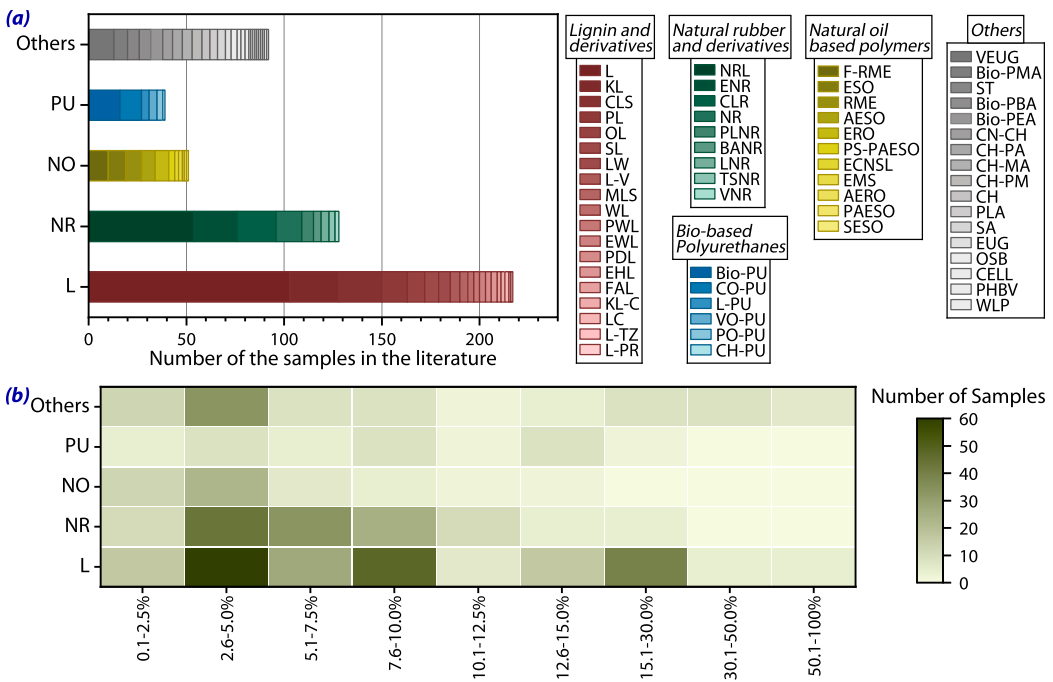


Figure 10. Frequency of biopolymer use in bitumen modification: (a) distribution by type of biopolymers and (b) distribution by addition ratios.

it is seen that 217 samples modified with lignin and its derivatives (L) are included in the literature. Subsequently, the second most frequently used biopolymer type in bitumen modification is natural rubber and its derivatives (NR) with 128 samples. After that, according to the biopolymer groups in this study, 92 samples were in the Others group, 51 in the NO group, and 39 in the PU group. It should be noted here that the number of samples in the subgroups belonging to the Others group is 13 only for VEUG, while the number of samples belonging to the other subgroup members is not more than 10. Therefore, the subgroups of the Others group are not included in the main groups of the study.

When the distribution of biopolymer main groups according to subgroups is analyzed, it is seen that the most frequently used lignin type in the subgroups belonging to the Lignin and derivatives group is 'L' with 103 samples. This sub-group includes studies in which the material added to bitumen is directly defined as lignin without giving details of the lignin type (Andiyappan & Kuna, 2023, 2024; Andiyappan

et al., 2024; Arafat et al., 2019; Batista et al., 2018; Cai et al., 2022, 2023; Z. Chen et al., 2019; Cheng et al., 2020; Duarte Mendonça et al., 2023; Gao et al., 2020; Gaudenzi et al., 2022; Gaudenzi, Cardone, Lu, et al., 2023a; Ghabchi, 2022; Guo et al., 2020; D. Hu et al., 2022; Yiming Li & Lv, 2023; Ma et al., 2022; McCready & Williams, 2008; Miao et al., 2019; I. P. Pérez et al., 2019; Rachman et al., 2023; Ren et al., 2021; Su et al., 2023; Sun et al., 2023; Sundstrom et al., 1983; K. Wang et al., 2023; J. Wu et al., 2021; C. Xu et al., 2021; Xue et al., 2022; Zahedi, Zarei, & Zarei, 2020; Zahedi, Zarei, Zarei, et al., 2020; E. Zhang et al., 2024). The subgroups with more than 10 sample repeats in the L main group are KL (25 samples) (Adwani et al., 2023, 2024; Al-falahat et al., 2024; de Lima Neto et al., 2023; Fakhri & Norouzi, 2022; Kalampokis et al., 2022; Nahar et al., 2023; Rezazad Gohari et al., 2023; Xie et al., 2017; R. Zhang et al., 2021), CLS (22 samples) (Fatemi et al., 2022a, 2022b; Fatemi et al., 2021; Qiu et al., 2024; M. Zarei, Abdi Kordani, Ghamarimajd, et al., 2022; M. Zarei, Abdi Kordani, & Zahedi, 2022; Ziaee et al., 2023), and PL (13 samples) (Chen et al., 2017; Fayzrakhmanova et al., 2016). Similarly, when analyzed for the NR main group, it is understood that the most frequently used subgroup is NRL, with 54 samples (Al-Mutlaq & Mahal, 2021; Abdunaser M. Al-Sabaei et al., 2020; Begam Rasheda et al., 2022; Hoy et al., 2024; Jitsangiam et al., 2021; Poovaneshvaran et al., 2020, 2023; Sani et al., 2020, 2023; Sani, Mohd Hasan, et al., 2021; Sani, Shariff, et al., 2021; E. Shaffie et al., 2018; Shafii et al., 2017, 2018; Suwanto et al., 2023; Usman et al., 2019; Yong Wen et al., 2017; Y. Yan et al., 2022). This is followed by ENR (Abd El Rahman et al., 2012; Al-Mansob et al., 2014, 2016, 2022; Al-Mansob, Ismail, Rahmat, et al., 2017; Al-Mansob, Ismail, Yusoff, et al., 2017; Ismail et al., 2012; Safaeldeen et al., 2022), CLR (Abdulrahman et al., 2021; Albuaymi et al., 2023; Ansari et al., 2023; N. M. Azahar et al., 2021; Norfazira Mohd Azahar et al., 2019; Hazoor Ansari et al., 2022; Mustafa Kamal et al., 2022), and NR (X. Hu et al., 2020; F. Liu et al., 2023; Ramadhan et al., 2020; Saowapark et al., 2019; Wititanapanit et al., 2021; Y. Yan et al., 2024; Yousefi, 2002a) subgroups with 22, 30, and 13 samples, respectively, while the other subgroups have less than 10 replicates. For the NO main group, the subgroup with the highest number of samples is F-RME (Iwański et al., 2022; Kowalski et al., 2017; Radziszewski et al., 2023), with 10 samples, while the number of occurrences of other NO biopolymers is less than 10. The most frequently used subgroups for the PU main group were Bio-PU (Gong, Liu, Wan, et al., 2023; Gong, Liu, Wang, et al., 2023; Kazemi, Goli, et al., 2022; Kazemi et al., 2024; Meng et al., 2022) and CO-PU (Cuadri et al., 2013, 2014a, 2014b; Kazemi, Mohammadi, et al., 2022; K. Wei et al., 2024; L. Xia et al., 2016) groups, with 16 and 11 samples, respectively. In the other subgroups in the PU group, the number of samples is less than 10. All these results confirm that lignin and natural rubber are the most frequently used biopolymer types for bitumen modification in the literature. Furthermore, there is a gap in the literature for the subgroups where the number of samples is less than 10, and further studies are needed to provide a comprehensive understanding of the interaction of these materials with bitumen.

Researchers who modify bitumen must determine the addition ratios when using any material. Determining these ratios through preliminary experiments is the most accurate approach, but it is challenging and time-consuming. Therefore, researchers usually select additive ratios by referring to other studies in the literature with relevant or similar materials. Accordingly, to help researchers who will conduct studies on biopolymers, the additive ratios and their usage frequencies were analyzed to determine the ratios at which the main biopolymer groups were added to bitumen. The results are shown in Figure 10(b), and it is clear from this figure that the additive rates are grouped at specific intervals. The main reason for this is that the researchers in each study used unique additive ratios, making it difficult to evaluate on a regular scale. Accordingly, when grouping the additive ratios, the usage ratios that can be considered as modifiers were grouped in the range of 0–15% in small increments of 2.5%, while the additive ratios that can be considered as extenders and substitutes were grouped in the ranges of 15–30%, 30–50%, and 50–100%, respectively. This grouping was considered in all numerical analyses within the scope of this review.

When the additive ratio is evaluated over the biopolymer main groups, it is seen that most samples for the L main group were prepared in the range of 2.6–5.0%. This result indicates that the biopolymers in the L group may be suitable for use in this range. The use of 7.6–10.0% and 15.1–30.0% additive ratio ranges in the L group is also widely preferred. On the other hand, when evaluated for the NR

group, it is understood that biopolymers in the NR group are mostly added to bitumen in the range of 2.6–5.0%. Moreover, 5.1–7.5% and 7.6–10.0% ranges can also be preferred for this type of additive. As for the NO group, the 2.6–5.0% additive range was used relatively more frequently than the other ranges. However, in the studies conducted for the PU group, it is impossible to state that any particular additive range is dominant.

4.1.4. *Biopolymer-modified bitumen preparation conditions*

Another critical decision-making process for researchers conducting studies on modified bitumen in the laboratory environment is the mixing conditions they will prefer for mixing base bitumen and additives. Accordingly, to help researchers consider modifying bitumen with biopolymers, the mixing conditions were analyzed according to the main groups of biopolymers in the studies in the article pool. The effect of the type of mixer on bitumen modification was taken into account when conducting the analyses. Thus, mechanical mixers, which have a relatively simple mechanism, have impellers of different geometries at the end of the mixer. In contrast, shear mixers with a more complex structure usually have stator-rator and saw-tooth blades (Dalhat & Al-Adham, 2023). Shear mixers disintegrate the additive into smaller particles, allowing them to be easily dispersed in the base bitumen. An additive (especially polymers) that can be mixed with a shear mixer is passed through a high-shear disintegrator after pre-mixing in the asphalt plant (Kök et al., 2024). On the other hand, mechanical mixers do not affect the additive directly but only ensure that it is mixed with bitumen at a certain speed. As it can be understood, the mechanisms of the two main types of mixers in bitumen modification are rather different. Therefore, while analyzing the parameters of biopolymer-modified bitumen modification, the evaluations for mechanical and shear mixers were carried out separately.

The results obtained from the analysis are given in Figure 11. When the mixing speeds in Figure 11(a) are examined, it is understood that the mean speeds preferred in high-shear mixers are much higher than those preferred in mechanical mixers, regardless of the biopolymer type. The main reason behind this is that the motor power in the high-shear mixers is higher, so the mixing speed can be selected higher. Most mechanical mixers do not have the power to reach these high mixing speeds. For the L group, the mean mixing speed in the shear mixer is around 4000 rpm, while this value is around 750 rpm in the mechanical mixer. When the evaluation is made for the NR group, it is noticeable that the mixing speeds, especially when using the shear mixer, are distributed in a wide range. Furthermore, while the mean mixing speed for the shear mixer in the NR group is around 3500 rpm, it is around 1600 rpm for the mechanical mixer. However, in the NO group, the mean mixing speed is around 2800 rpm for the shear mixer, while it is 900 rpm for the mechanical mixer. Finally, when the mixing speeds are analyzed in terms of the PU group, it is observed that the mean mixing speeds are around 2500 rpm when the shear mixer is used, while this value is around 1800 rpm when the mechanical mixer is used.

When the mixing times in Figure 11(b) are analyzed, it is observed that the most frequently used mixing time is 60 min, regardless of the biopolymer group. Furthermore, when evaluated for the L main group, it is understood that this time was never exceeded, and the mixing process was carried out for an average of 40–45 min regardless of the mixer type. When the NR main group is examined, it is seen that the studies were carried out in a very wide mixing time range, especially when using shear mixers. Additionally, while the mean mixing time for the NR main group is approximately 90 min for the shear mixer, this value is approximately 30 min for the mechanical mixer. This is an unexpected result. Indeed, since using shear mixers provides better homogenisation, mixing times are expected to be shorter in shear mixers. This unexpected result may be attributed to the compatibility of different NR types with bitumen in the subgroups of the NR main group. Continuing the analysis for the NO main group, the mean mixing time for the NO group is 110 min for the shear mixer and 60 min for the mechanical mixer. Finally, when the mixing times for the PU main group are evaluated, it is understood that an average mixing time of 50 min is preferred for both shear and mechanical mixers.

The final evaluation of the mixing conditions based on the mixing temperatures in Figure 11(c) reveals that the mean mixing temperature for biopolymer-modified bitumen studies in the literature

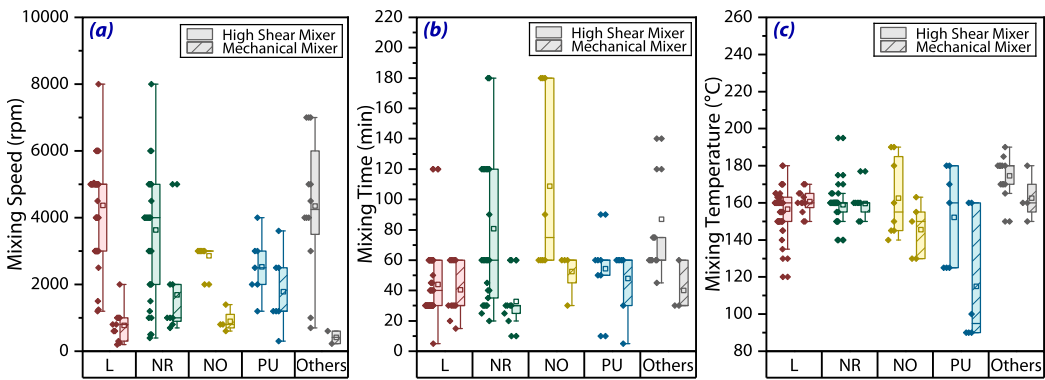


Figure 11. Selected mixing parameters for biopolymer modification by mixer type: (a) mixing speed, (b) mixing time, and (c) mixing temperature.

is approximately 160°C, irrespective of the biopolymer group and mixer type. It is possible to observe that the values of L and NR main groups are distributed very near to this temperature, while a wider temperature range was preferred for NO and PU main groups.

Above, different mixing conditions for different biopolymer groups have been examined and evaluated in detail. The mean values of each parameter of the mixing conditions for different biopolymer groups and mixer types can be considered reference values for researchers in this field. However, it should be noted here that the optimum mixing conditions for each additive type may vary. Therefore, researchers should always consider that the effect of mixing conditions other than the reference values here may be different.

4.1.5. Experimental methods chosen in the studies

In studies on bitumen modification, it is possible to investigate the properties of a modified bitumen with different experimental methods. However, considering the abundance of experimental methods and the fact that each method is carried out for a different aim, it is critical for researchers to choose the experiment to be carried out in their study. Accordingly, although the purpose of the study is of great importance, it is possible to utilise various experimental methods for the same objective. Therefore, researchers should follow the current and popular experimental methods used in the literature. Considering this situation, the articles in the pool were analyzed in terms of the experimental methods used in the studies in order to determine the experimental methods to be analyzed in the quantitative analysis of this study. Accordingly, Figure 12 was prepared to examine which experimental methods were used and at which frequency. First of all, in Figure 12(a), the articles in the pool are generally evaluated in terms of the scope of experimental methods. Out of 174 articles in the article pool, 125 of them conducted only micro-scale investigations on bitumen tests. In contrast, 16 of them conducted only macro-scale experimental studies on bituminous hot mixture samples. However, in 33 studies, comprehensive research was conducted using both micro- and macro-scale experimental methods to investigate biopolymer modification. In line with these results, it is possible to say that most of the existing biopolymer-modified bitumen studies have been conducted with micro-scale bitumen experiments. However, on these results, it can also be said that there are still not enough studies in the literature on the effects of biopolymer-modified bitumen on asphalt mixture performance.

Since almost all of the studies in the article pool included more than one experimental method, a more detailed frequency analysis was carried out by grouping the experimental methods according to which properties of the bitumen or asphalt mixture were investigated. The results obtained in this respect are given in Figure 12(b). 34.8% of the total 1050 experiments carried out in the studies were conducted to determine the physical properties of bitumen. This is mainly because the physical experiments are accessible and practical methods that can be carried out with relatively low-cost equipment.

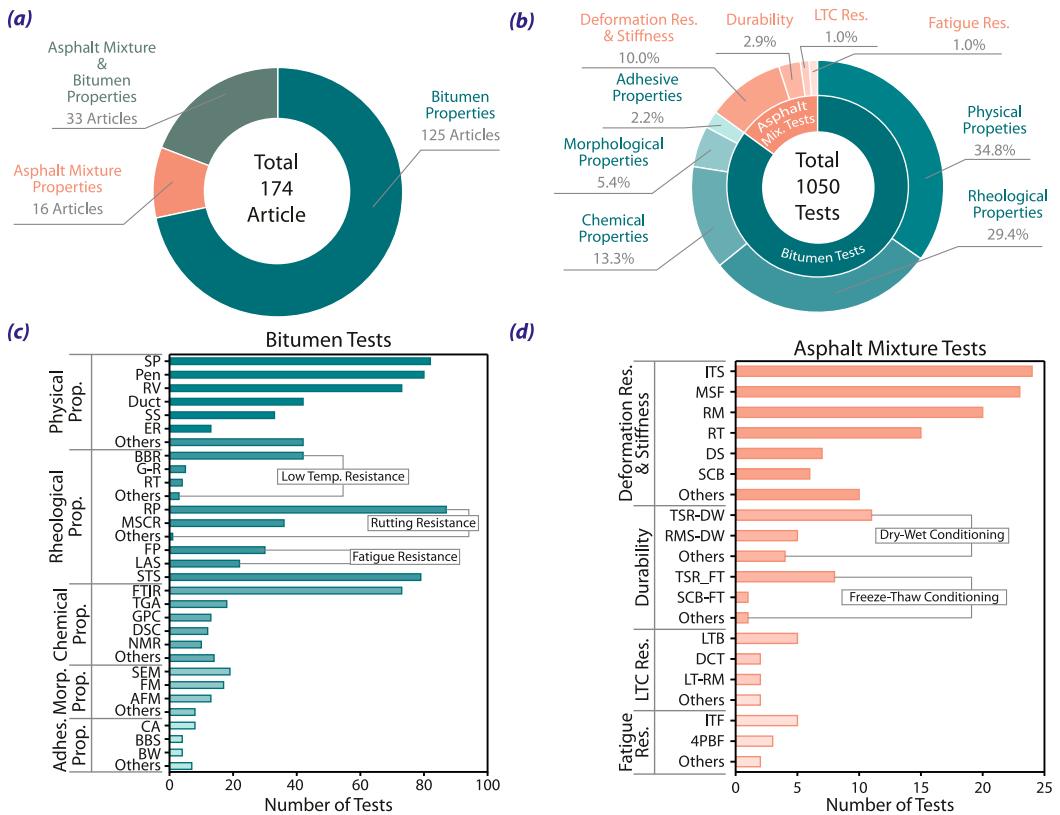


Figure 12. Distribution of experimental methods used to investigate biopolymer-modified bitumen: **(a)** scope of experimental methods chosen in the articles, **(b)** distribution of experimental methods according to the properties investigated, **(c)** the preference frequency of bitumen experiments, and **(d)** the preference frequency of asphalt mix experiments.

However, since performance evaluations of modified bitumen are generally based on rheological properties, 29.4% of the experiments in the literature are rheological property experiments. Following that, chemical properties, morphological properties, and adhesive properties of biopolymer-modified bitumen were the most frequently investigated features on the bitumen scale, respectively. When the experiments conducted with biopolymer-modified bitumen at the asphalt mixture scale are examined, it is understood that the most frequently examined properties are deformation resistance and stiffness. Furthermore, asphalt mixture properties such as durability, low-temperature cracking (LTC) resistance, and fatigue resistance were also investigated.

When choosing the experimental methods to be used in the quantitative comparative analysis, the most frequently used experimental methods in the literature were preferred. In the results of the analysis carried out in this respect, the bitumen and asphalt mixture test frequencies in the article pool are given in Figure 13(c and d), respectively. When the experiments performed on the bitumen scale for biopolymer-modified bitumen were examined, it was seen that softening point, penetration, rotational viscosity, storage stability, and elastic recovery experiments were used to determine the physical properties, while rutting parameter, sweep (amplitude, frequency, time, etc.), multi-stress creep and recovery, bending beam rheometer, fatigue parameter and linear amplitude sweep tests were used to determine the rheological properties. Furthermore, it is understood that Fourier transform infrared (FTIR) spectroscopy is used to determine chemical properties, and scanning electron (SEM), fluorescence (FM), and atomic force (AFM) microscopy are the most preferred experimental methods to determine morphological properties. On the other hand, when the experiments carried

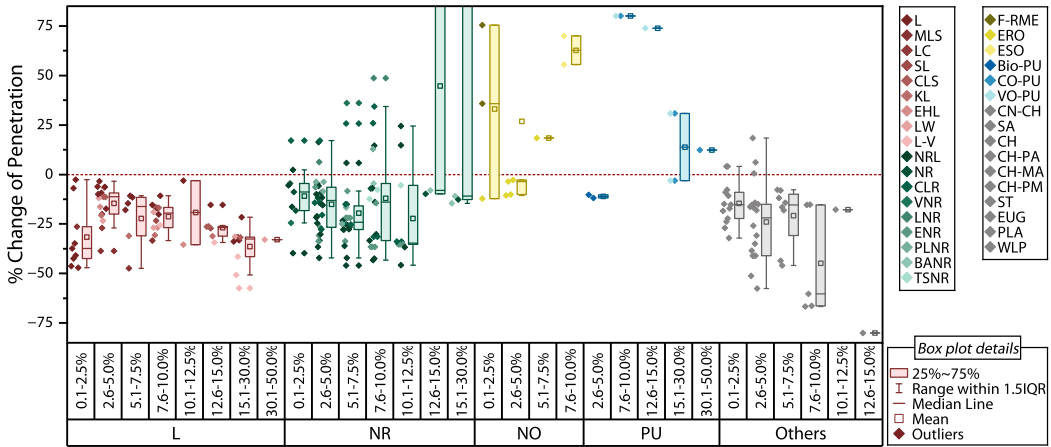


Figure 13. Effect of biopolymer addition on penetration value of base bitumen.

out for biopolymer-modified bitumen at asphalt mixture scale are examined, it is observed that for deformation resistance and stiffness properties, indirect tensile strength, Marshall stability and flow, modulus of elasticity and rutting tests; for durability properties, dry-wet conditioning and freeze-thaw conditioning methods; and for fatigue resistance properties, indirect tensile fatigue test are the most frequently used experimental methods. In line with these results, the experimental methods that will be used in the quantitative analysis of this study have been preferred. However, researchers who want to study the effect of different biopolymers on bitumen modification can design their experiments using these methods.

4.2. Effect of biopolymers on bitumen performance

In this section of the study, the effect of biopolymer addition to base bitumen on various properties is comparatively evaluated based on the results of studies in the literature. Accordingly, the physical, chemical, and rheological properties of biopolymer-modified bitumen are discussed and analyzed in detail.

4.2.1. Physical properties

The effect of various biopolymers on the penetration value of base bitumen is given in Figure 13. In general, it is possible to say that adding biopolymers decreases the penetration value of base bitumen. However, it is also understood that this effect may change depending on the type of biopolymer. Therefore, it would be more reasonable to evaluate based on the main groups of biopolymers. Accordingly, when the effect of the L main group on the base bitumen is analyzed, it can be clearly stated that the addition of L decreases the penetration value of the bitumen, considering the consensus of all studies. Furthermore, in the 2.6%–30.0% addition ratio range, the effect of decreasing the penetration value with increasing addition ratio is more pronounced. However, some studies (Z. Chen et al., 2019; K. Wang et al., 2023; Yilmaz & Ugurlu, 2023) using 0.1–2.5% L addition ratios show a very high penetration reduction effect. It is thought that these results may be related to the type of L used in these studies. On the other hand, in general, it can be said that biopolymers in the L main group further reduce the penetration of bitumen with increasing additive ratio.

When evaluated for the NR main group, although the majority of the studies showed that the addition of NR decreased the penetration value of the base bitumen, some studies also showed that the addition of NR increased the penetration value. Therefore, it would not be accurate to mention a certain overall effect in the NR main group. However, considering that the majority of the data is below

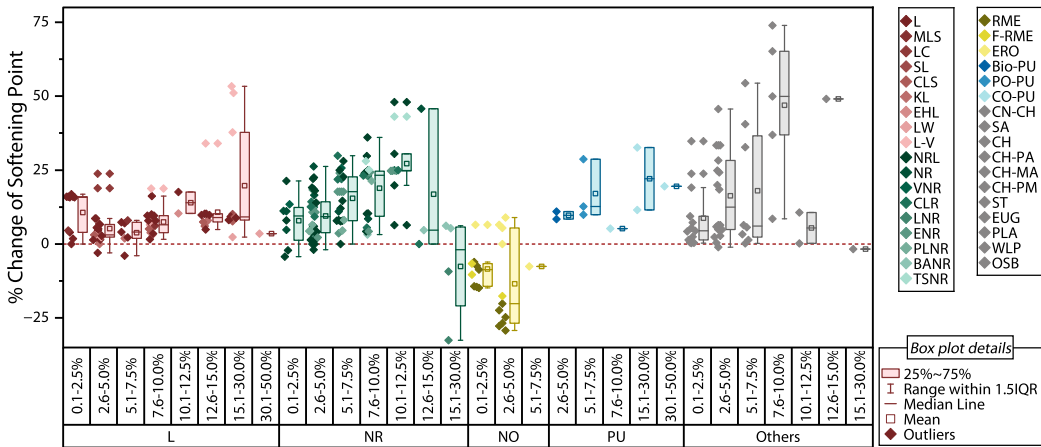


Figure 14. Effect of biopolymer addition on the softening point value of base bitumen.

the zero baseline, it can be assumed that the addition of NR will decrease the penetration of the base bitumen. Also, it can be assumed that the penetration decrease effect will increase with increasing addition ratio. On the other hand, whether NR will increase or decrease the penetration of bitumen will be directly related to the type of NR used.

Finally, when the penetration test results of the NO and PU main groups are analyzed, it is evident that the data distribution for the two biopolymer groups prevents a definite conclusion. Indeed, some studies show that bitumen modification with NO increased the penetration value (Fuhaid et al., 2018; Hrynychuk et al., 2019; Iwański et al., 2022) and decreased it (Hrynychuk et al., 2019; Q. Lu et al., 2021; Nykypanchuk et al., 2013). Similarly, PU addition increased (L. Xia et al., 2016, 2023) and decreased (Cuadri et al., 2014a; L. Xia et al., 2016, 2023) the penetration of base bitumen. For both biopolymer groups, this effect depends on the preferred conditions of biopolymer production and the amounts added to bitumen. However, it is understood that the penetration test is relatively underutilised in determining the physical properties of biopolymers in the NO and PU groups.

The results of softening point tests, another essential physical property determination method for biopolymer-modified bitumen, were analyzed, and the findings are given in Figure 14. When the results of softening point values are compared with the penetration value results generally, it can be observed that the softening point test results are distributed in a very close range and harmony with each other compared to the penetration value results. This is mainly because, compared to the penetration test, the softening point test is less influenced by the test operator. On the other hand, except for the results of only one study in the L main group, all studies show that adding L biopolymer to bitumen will increase the softening point value of base bitumen. The magnitude of this increase varies depending on the addition ratio, except the 0.1–2.5% range, which shows that the softening point can be increased more with increasing addition amount. Furthermore, when the NR main group is analyzed, it is seen that the results obtained have a much more precise meaning compared to the penetration test results. Indeed, the addition of NR increased the softening point of base bitumen in most of the studies, and the effect increased significantly with increasing additive content.

The number of softening point tests for biopolymers in the NO main group is considerably higher than the number of penetration tests, which makes it possible to evaluate the effect of biopolymers in the NO main group on the softening point of base bitumen in a reliable way. Accordingly, the addition of NO decreases the softening point of base bitumen in most studies, and the magnitude of the decrease is directly proportional to the increasing additive content. On the other hand, it is difficult to say that there is a sufficient number of softening point test data for the PU main group. However, all PU studies in the literature have a typical result of increasing the softening point of base bitumen.

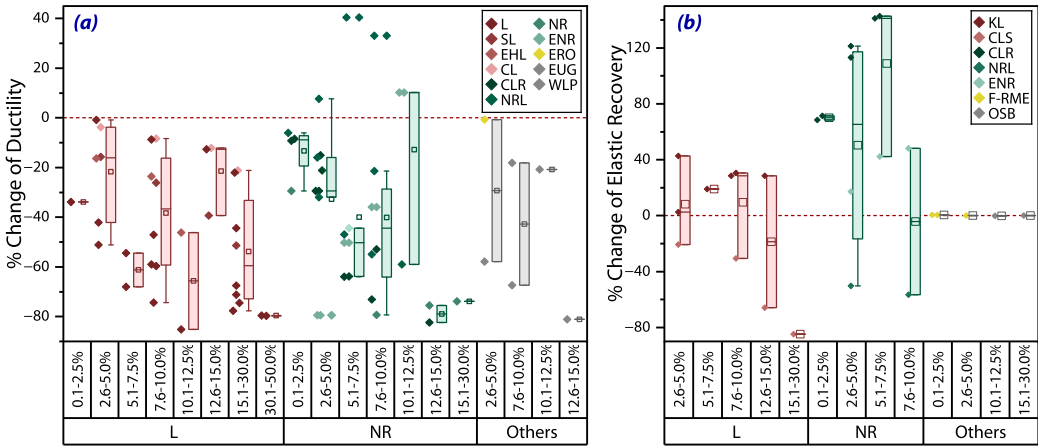


Figure 15. Effect of biopolymer addition on (a) ductility and (b) elastic recovery values of base bitumen.

In general, it is possible to get a general idea about the effect of various biopolymer groups on the consistency of base bitumen by the combined evaluation of penetration and softening point tests. Accordingly, adding L, NR, and PU biopolymers to the base bitumen generally hardens the base bitumen and increases its consistency, whereas NO biopolymers generally soften the base bitumen and decrease its consistency.

Other important physical properties of modified bitumens include their ability to elongate without breaking and recover from deformations. Accordingly, the results of ductility and elastic recovery tests applied to biopolymer-modified bitumen were analyzed, and the results of the analysis are given in Figure 15. From this figure, it is possible to say that ductility and elastic recovery tests are much less included in the literature than other physical tests. In fact, it is possible to access only the ductility value of ERO biopolymer and the elastic recovery value of F-RME biopolymer from PU and NO main groups. Therefore, only L and NR main biopolymer groups were used in the analysis. Therefore, there is a gap in the literature on the ductility and elastic recovery values of different biopolymers.

When the ductility values in Figure 15(a) are analyzed for the L main biopolymer group, it is understood that all the test results in the literature show that the addition of L jointly decreases the ductility value of the base bitumen. However, due to the small number of studies, a trend in the ductility change related to the addition ratio could not be identified. On the other hand, for the NR main group, except for some studies (Al-Mansob et al., 2014, 2016; Abdalnaser M. Al-Sabaei et al., 2020), most articles showed that adding NR biopolymer to the base bitumen decreased the ductility of the base bitumen. It is believed that the main reason for obtaining a different result compared to the general trend in the relevant studies is the type of NR used. However, it is understood that the magnitude of the decrease in ductility value with the addition of NR varies proportionally to the increasing addition ratio. When the change in elastic recovery values in Figure 15(b) is examined, it is observed that data on very few biopolymer types are available in the literature. Furthermore, the effect of the L main group on the elastic recovery varied entirely according to the L type; while the addition of KL (de Lima Neto et al., 2023; Kalampokis et al., 2022) caused an increase in the elastic recovery, the addition of CLS (Kalampokis et al., 2022) caused a decrease. Additionally, when the elastic recovery values of the NR main group are analyzed, it is understood that the results similarly depend on the NR type. Indeed, while the use of NRL (Poovaneshvaran et al., 2020) decreased the elastic recovery of base bitumen, the use of CLR (Abdulrahman et al., 2021; Albuaymi et al., 2023) and ENR (Al-Mansob et al., 2022) increased it. According to these results, when the effect of biopolymers on ductility and elastic recovery properties is generalised, it is observed that biopolymers generally decrease the ductility values and increase or

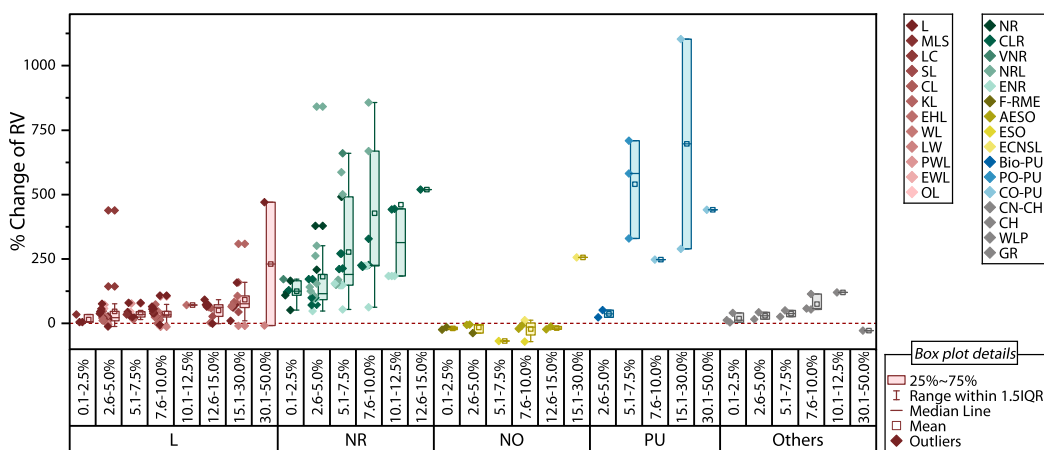


Figure 16. Effect of biopolymer addition on the rotational viscosity of base bitumen at 135°C.

decrease the elastic recovery values depending on the type of biopolymer. Accordingly, it is understood that adding biopolymers to base bitumen will limit the ability of base bitumen to elongate without breaking and, in some cases, will negatively affect the elastic recovery ability.

The rotational viscosity test is the most technological method used to evaluate the physical properties of bitumen. In this test, the viscometer can measure the resistance of the bitumen to flow very precisely and is minimally affected by operator experience or environmental conditions compared to other physical property tests. Furthermore, the viscosity values of bitumen are critical values to evaluate its usability in asphalt mixtures in terms of workability. Accordingly, the results of the analyses on the viscosity values of biopolymer-modified bitumen are given in Figure 16. When this figure is examined, it is observed that the experimental results in the literature, especially for the L main group, are clustered in a small range and highlight a consensus result. Accordingly, most of the studies in the literature indicate that adding a biopolymer in the L group to the base bitumen will increase the viscosity value. Moreover, the level of enhancement of viscosity will increase in direct proportion to the increasing additive ratio. On the other hand, although the data for the NR main group is distributed in a wide range, all studies show that biopolymers in the NR group will increase the viscosity values. Also, for the NR group, there is a linear relationship between the additive ratio and the enhancement effect. The effect of biopolymers in the NO group on viscosity differs from the other biopolymer types, and adding biopolymers in the NO group to the base bitumen decreases the viscosity value of the base bitumen. Finally, when the biopolymers in the PU main group are evaluated, it is generally understood that a PU biopolymer increases the viscosity of the base bitumen in all studies, although it is not possible to talk about a trend between the data and additive ratios within the group.

In general, when the effect of biopolymers on viscosity values is analyzed, it is understood that except for the biopolymers in the NO main group, all other biopolymers will increase the viscosity of bitumen. The main reason for this different effect of the NO main group can be considered to be the natural oil origin of these biopolymers. In fact, the softening effect of oils on bitumen is well known (Çavdar et al., 2024; Kumandaş et al., 2022, 2024). Therefore, it is understandable that this group of biopolymers polymerised from natural oils has such an effect on the viscosity of bitumen.

4.2.2. Chemical properties

FTIR is one of the most widely used methods to investigate the chemical properties of bitumen by characterising the bonds between bitumen molecules. FTIR can be used to identify the functional groups, bonds, and their bonding positions in bitumen. On the other hand, based on the IR spectra of additives, base bitumen, and modified bitumen made with FTIR, it can be determined whether there is a

Table 5. Characteristic FTIR peaks of base bitumen.

Wavelength (cm ⁻¹)	Description	References
2930–2900, 2858–2800	Stretching vibrations of C-H bonds of CH ₂ and CH ₃ groups	(Al-Mansob, Ismail, Rahmat, et al., 2017; Andiyappan et al., 2024; Gong, Liu, Wan, et al., 2023; Gong, Liu, Wang, et al., 2023; Hazoor Ansari et al., 2022; Hemida & Abdelrahman, 2021b; Jitsangiam et al., 2021; Król et al., 2017; N. Li, J. Xu, et al., 2021; Yiming Li & Lv, 2023; F. Liu et al., 2023; Malinowski et al., 2023a; Malinowski et al., 2023b; Malinowski et al., 2022; Peng et al., 2020; Saowapark et al., 2019; X. Shi & Xu, 2023; D. Wang et al., 2019; K. Wei et al., 2024; C. Xia et al., 2022; G. Xu et al., 2017; Yilmaz & Ugurlu, 2023; Yu et al., 2021)
1730–1698	Peak attributed to carbonyl group (C = O)	(Gaudenzi, Cardone, Lu, et al., 2023a; Yiming Li & Lv, 2023; Peng et al., 2020; G. Xu et al., 2017)
1630–1570	Stretching vibrations of the conjugated double bond (C = C) of the aromatic ring	(Al-Mansob, Ismail, Rahmat, et al., 2017; Andiyappan et al., 2024; N. M. Azahar et al., 2021; Hemida & Abdelrahman, 2021b; Jitsangiam et al., 2021; Król et al., 2017; N. Li, J. Xu, et al., 2021; Yiming Li & Lv, 2023; F. Liu et al., 2023; J. Lu et al., 2023; Malinowski et al., 2023a; Malinowski et al., 2023b; Malinowski et al., 2022; X. Shi & Xu, 2023; K. Wei et al., 2024; C. Xia et al., 2022; G. Xu et al., 2017; Yu et al., 2021)
1460–1450, 1377–1373	Symmetric and asymmetric deformation vibrations of methyl and methylene groups	(Al-Mansob, Ismail, Rahmat, et al., 2017; Andiyappan et al., 2024; Gaudenzi, Cardone, Lu, et al., 2023a; Gong, Liu, Wan, et al., 2023; Gong, Liu, Wang, et al., 2023; Hazoor Ansari et al., 2022; Hemida & Abdelrahman, 2021b; D. Hu et al., 2022; Jitsangiam et al., 2021; N. Li, Xu, et al., 2021; Luz et al., 2021; Malinowski et al., 2023a; Malinowski et al., 2022; Saowapark et al., 2019; X. Shi & Xu, 2023; K. Wei et al., 2024; C. Xia et al., 2022; Yilmaz & Ugurlu, 2023; Yu et al., 2021)
1035–1000	Peak attributed to the sulfoxide group (S = O)	(Al-Mansob, Ismail, Rahmat, et al., 2017; Andiyappan et al., 2024; Gaudenzi, Cardone, Lu, et al., 2023a; Hemida & Abdelrahman, 2021b; Jitsangiam et al., 2021; N. Li, J. Xu, et al., 2021; Yiming Li & Lv, 2023; Malinowski et al., 2023b; G. Xu et al., 2017)
900–700	C-H vibrations of groups in aromatic rings	(Al-Mansob, Ismail, Rahmat, et al., 2017; N. M. Azahar et al., 2021; Hazoor Ansari et al., 2022; Hemida & Abdelrahman, 2021b; Jitsangiam et al., 2021; N. Li, J. Xu, et al., 2021; F. Liu et al., 2023; Luz et al., 2021; Malinowski et al., 2023a; Malinowski et al., 2023b; Malinowski et al., 2022; X. Shi & Xu, 2023; C. Xia et al., 2022; Yilmaz & Ugurlu, 2023; Yu et al., 2021)

chemical interaction between bitumen and additive. When the article pool of this study was examined, it was seen that the chemical properties of additives and bitumen were frequently investigated by FTIR analysis. Therefore, to investigate the chemical properties of base bitumen and additives and determine the interaction between them, the FTIR analyses performed in these articles were analyzed. The characteristic peaks seen in the IR spectrum of the base bitumen and their counterparts can be compiled as shown in Table 5. As seen in this table, there are characteristic peaks at approximately 2920 and 2850 cm⁻¹ in the base bitumen spectra, which are caused by the stretching vibrations of the C-H bonds of the CH₂ and CH₃ groups. The studies observed these peaks in 2930–2900 cm⁻¹ and 2858–2800 cm⁻¹, respectively. Bitumen shows a peak at approximately 1700 cm⁻¹, which is attributed to the carbonyl (C = O) group, and this peak is observed in the wave number range of 1730–1698 cm⁻¹ in the studies. It is stated that the characteristic peak of bitumen at about 1600 cm⁻¹ is due to the stretching vibrations of the conjugated double bond (C = C) of the aromatic ring, and this peak is observed in the range of 1630–1570 cm⁻¹. Two characteristic peaks near 1460 and 1375 cm⁻¹ are attributed to the symmetric and asymmetric deformation vibrations of methyl and methylene groups. It can be said that these peaks are observed in the wave number range 1460–1450 cm⁻¹ and 1377–1373 cm⁻¹, respectively. Another characteristic peak of bitumen, the peak near 1030 cm⁻¹, is attributed to the sulfoxide (S = O) group, and it can be said that this peak was observed in the wave number range of 1035–1030 cm⁻¹ in the studies. Furthermore, when the other peaks seen in the bitumen spectra in the studies are compiled, it can be said that bitumen shows peaks at wavelengths ranging from 900 to 700 cm⁻¹, and these peaks are due to the C-H vibrations of the groups in the aromatic rings contained in bitumen.

In order to explore the chemical structure of the biopolymer materials, the specific peaks seen in the FTIR spectra of the articles in the pool are categorised for each biopolymer main group and given in Table 6. When the characteristic peaks in the L main group are analyzed, the peak in the range 3470–3290 cm^{-1} is attributed to O-H or N-H vibrations. The prominent peaks in the ranges 2938–2920 cm^{-1} and 2847–2840 cm^{-1} of the biopolymers in the L group were associated with C-H stretching vibrations. These biopolymers showed a characteristic peak in 1709–1699 cm^{-1} associated with the carbonyl group. Significant peaks in the range of 1614–1565 cm^{-1} , 1517–1506 cm^{-1} , and 1470–1409 cm^{-1} were attributed to the stretching vibrations of the aromatic rings contained in the L group. The peak near 1600 cm^{-1} can be indicated as the specific peak corresponding to the aromatic C = C bond stretching. For the biopolymers in the L group, the prominent peaks in the range 1270–1115 cm^{-1} and 880–800 cm^{-1} were characterised as peaks corresponding to Guaiacyl units of lignin. The peaks in the 1335–1325 cm^{-1} and 1129–1100 cm^{-1} range were characterised as peaks corresponding to Syringyl units. The peak in the 1035–1030 cm^{-1} range of this group was attributed to the in-plane bending of aromatic C-H bonds in some studies (D. Hu et al., 2022; Sun et al., 2023; R. Zhang et al., 2021), while in others (Andiyappan et al., 2024; Gaudenzi, Cardone, Lu, et al., 2023a; D. Wang et al., 2019), it was attributed to the sulfoxide group. This difference in evaluation is thought to be related to the types of lignin used in the studies. It is thought that in sulfonated lignins and similar types, the peak near 1030 cm^{-1} can be associated with sulfoxide. However, this peak can be associated with the in-plane bending of aromatic C-H bonds in other sulfonate-free lignins. The characteristic peaks of the materials in the L main group are remarkably similar to those of the base bitumen. As a result of the structural similarity of lignin and bitumen, it can be said that the characteristic peaks, especially in 2920, 2850 cm^{-1} , 1700 cm^{-1} , 1600 cm^{-1} , and 1600–1400 cm^{-1} range, are observed at close wave numbers in the spectra of both. On the other hand, when the IR spectra of lignin-modified bitumen were analyzed, it was reported that although the intensity of the peaks changed compared to the base bitumen, no new peaks were observed that could be attributed to chemical reactions except for the peaks from lignin and bitumen content (Andiyappan et al., 2024; Cheng et al., 2020; Rezazad Gohari et al., 2023; X. Shi & Xu, 2023; Yu et al., 2021; Y. Zhang et al., 2020; Y. Zhang, Liu, et al., 2022; Y. Zhang, Si, et al., 2023). Therefore, it can be concluded that the additives in the L main group generally do not undergo any significant chemical reaction with the bitumen but are mainly mixed physically.

For the NR group, the peaks were characterised by limited data since few studies involving NR included FTIR analysis. In these studies, the peak at 3038 cm^{-1} was assigned to the stretching vibration of the C-H bond related to the C=C bond, the peaks at 1020, 1220, and 1083 cm^{-1} were assigned to the C-O stretching vibrations of vinyl ether and aliphatic ether, the peak at 842 cm^{-1} was assigned to the absorption of cis 1,4-polyisoprene associated with C-H out-of-plane bending, the peak at 693 cm^{-1} was assigned to the absorption of cis 1,4-polyisoprene absorption associated with C-H out-of-plane bending, the peak at 693 cm^{-1} is attributed to the toluene group and the peaks in the range 619–775 cm^{-1} are attributed to out-of-plane absorption from the toluene group. Additionally, it can be said that the peaks at 2960, 2920, 2850 cm^{-1} , 1642–1662, and 1375 cm^{-1} are similar to the characteristic peaks of bitumen. When the IR spectra of NR-modified bitumen were analyzed, it can be said that no new peak was formed in the spectra of bitumen with the addition of NR and that NR and bitumen were only physically mixed (Al-Mutlaq & Mahal, 2021; F. Liu et al., 2023; Saowapark et al., 2019).

For the NO group, the peak at 3385 cm^{-1} is associated with the stretching of the -OH bond of the phenolic group, the peaks at 1436 and 1195 cm^{-1} with the O-CH₃ stretching and -CH₃ asymmetric bending characteristic for methyl esters, respectively, the peaks in the range 995–910 cm^{-1} with C-H stretching, and the peaks in the range 914–823 cm^{-1} with C-O-C (oxirane ring). In terms of the bands at 2926 and 2852 cm^{-1} attributed to asymmetric and symmetric vibrations of methyl and methylene groups and the peak at 1593 cm^{-1} attributed to the stretching of the C = C bond of the aromatic ring, it can be stated that the NO group shows characteristic peaks similar to bitumen. On the other hand, in one of the two studies in which the FTIR spectra of bitumen modified with NO group biopolymers were examined (Iwański et al., 2023), it was reported that the ester absorption peak at 1741 cm^{-1} appeared

Table 6. Characteristic FTIR peaks of biopolymer groups.

Wavelength (cm ⁻¹)	Description	References
<i>Lignin and derivatives</i>		
3470–3290	O-H or N-H stretching vibration	(Andiyappan et al., 2024; Andiyappan & Kuna, 2024; Cheng et al., 2020; Gaudenzi, Cardone, Lu, et al., 2023a; Rachman et al., 2023; X. Shi & Xu, 2023; D. Wang et al., 2019; Xin et al., 2016; Yu et al., 2021; S. Zhou et al., 2023)
2938–2920, 2847–2840 1709–1699	C-H stretching vibrations Peak attributed to carbonyl group (C = O)	(Andiyappan et al., 2024; Rachman et al., 2023; Xin et al., 2016; Yu et al., 2021) (Arafat et al., 2019; Gaudenzi, Cardone, Lu, et al., 2023a; Rachman et al., 2023; X. Shi & Xu, 2023; Xin et al., 2016; Yu et al., 2021)
1614–1565, 1517–1506, 1470–1409	Stretching vibrations of aromatic rings	(Andiyappan et al., 2024; Rachman et al., 2023; Rezazad Gohari et al., 2023; X. Shi & Xu, 2023; Xin et al., 2016; Yu et al., 2021; R. Zhang et al., 2021; S. Zhou et al., 2023)
1600	Stretching of aromatic C = C bond	(Andiyappan et al., 2024; D. Hu et al., 2022; Sun et al., 2023)
1270–1115, 880–800	Guaiacyl units	(Andiyappan et al., 2024; Andiyappan & Kuna, 2024; Arafat et al., 2019; Rachman et al., 2023; Yu et al., 2021; R. Zhang et al., 2021)
1335–1325, 1129–1100	Syringyl units	(Andiyappan et al., 2024; Andiyappan & Kuna, 2024; Rachman et al., 2023; Yu et al., 2021; R. Zhang et al., 2021)
1035–1030	In-plane bending of aromatic C-H bonds or sulfoxide group (S = O)	(Andiyappan et al., 2024; Gaudenzi, Cardone, Lu, et al., 2023a; D. Wang et al., 2019)
<i>Natural rubber and derivatives</i>		
3038	Stretch vibration of the C-H bond connected to the C=C bond	(F. Liu et al., 2023)
2960 and 1375, 2920 and 2850 1642–1662	Presence of methyl -CH ₃ and methylene CH ₂ - C = C bond or molecular vibrations of amide and amine in natural rubber	(F. Liu et al., 2023) (F. Liu et al., 2023; Mustafa Kamal et al., 2022)
1020, 1220 and 1083	C-O stretching vibrations of vinyl ether and aliphatic ether	(Al-Mutlaq & Mahal, 2021)
842	Absorption of cis 1,4-polyisoprene associated with C-H out-of-plane bending	(Albuaymi et al., 2023)
693 619–775	Toluene group Out-of-plane absorption from the toluene group	(N. M. Azahar et al., 2021) (Albuaymi et al., 2023)
<i>Natural oil based polymers</i>		
3385	Stretching of the -OH bond of the phenolic group	(Uchoa et al., 2021)
3008, 2926 and 2852	C – H stretching of the inner unsaturated part, and asymmetric and symmetric vibrations of the -CH ₃ and -CH ₂	(Uchoa et al., 2021)
1741	C = O stretching in the ester functional group	(Iwański et al., 2023)
1593 and 1454	Stretching of the C = C bond in the aromatic ring	(J. Lu et al., 2023; Uchoa et al., 2021)
1436 and 1195	O-CH ₃ stretching and -CH ₃ asymmetric bending, characteristic for methyl esters	(Iwański et al., 2023)
995–910 914–823	C-H C-O-C (oxirane ring)	(Uchoa et al., 2021) (Uchoa et al., 2021)

(continued).

Table 6. Continued.

Wavelength (cm ⁻¹)	Description	References
<i>Bio-based polyurethanes</i>		
3338–3288	Stretching vibration of -NH bonds	(Gong, Liu, Wan, et al., 2023; Gong, Liu, Wang, et al., 2023; Meng et al., 2022; K. Wei et al., 2024; L. Xia et al., 2016)
2926 and 2854	C-H in-phase symmetric stretching and out-of-phase asymmetric stretching vibrations of methylene groups	(L. Xia et al., 2016)
2325–2275	Stretching vibration of the -NCO group	(Gong, Liu, Wan, et al., 2023; L. Xia et al., 2016)
1741–1745	C = O stretching vibration	(Gong, Liu, Wan, et al., 2023; Gong, Liu, Wang, et al., 2023; Meng et al., 2022)
1800–1600	Urethane and urea bonds	(Kazemi, Goli, et al., 2022; Kazemi, Mohammadi, et al., 2022; Kök et al., 2021; Meng et al., 2022)
1597–1595, 1536–1525, 1450–1411	Stretching vibration of the benzene ring frame	(Gong, Liu, Wan, et al., 2023; Gong, Liu, Wang, et al., 2023)
1243	C-O vibration of the carbamate group	(Gong, Liu, Wan, et al., 2023)
1070–1104	-CO-C- formation	(K. Wei et al., 2024)
970	C-H vibration of the PU molecule	(Gong, Liu, Wan, et al., 2023)
720	-NHCOO- group	(L. Xia et al., 2023)

in the modified bitumen. However, the same peak was also observed in the spectrum of the NO additive used, indicating that this peak came from the NO component. In the aforementioned study, since no new peak was observed in the IR spectra of the modified bitumen except the peaks coming from the base bitumen and NO additive, it can be said that there is no chemical interaction between NO additive and bitumen. In the other study (Uchoa et al., 2021), the NO group additive used was bio-epoxidized, and the band at 914 cm⁻¹ due to the oxirane group in the spectra of modified bitumen remained after modification, indicating that these groups did not react completely. These two studies suggest that there is no significant chemical interaction between the bitumen and the NO group additive used. However, since the NO group has a very wide scope and natural oils are not directly biopolymers but are used as monomers for biopolymers with different properties, it is not possible to make a definite or general judgment about the chemical interaction of NO group additives with bitumen.

When the characteristic peaks of the PU group were analyzed, the peaks seen in the range 3338–3288 cm⁻¹ were attributed to the stretching vibrations of -NH bonds, while the prominent peaks seen in the range 2325–2275 cm⁻¹ were attributed to the stretching vibrations of the -NCO group. Peaks in the 1800–1600 cm⁻¹ range were associated with urethane and urea bonds. In another study, the peak at 1243 cm⁻¹ was attributed to the C-O vibration of the carbamate group, the peak in the range 1070–1104 cm⁻¹ to the formation of -CO-C- by synthesising bio-based polyurethane, the peak at 970 cm⁻¹ to the C-H vibration peak of the PU molecule and the peak at 720 cm⁻¹ to the formation of -NHCOO-. Moreover, the biopolymers in the PU group showed similar characteristic peaks at 2926, 2854, and 1745–1741 cm⁻¹ with bitumen. In addition, it can be said that the prominent peaks in the range of 1597–1595 cm⁻¹ and 1536–1525 cm⁻¹ are similar to the peaks in bitumen due to vibrations in aromatic rings. When the effect of biopolymers in the PU group on the chemical structure of bitumen was examined, it was stated in some studies (Gong, Liu, Wan, et al., 2023; Meng et al., 2022) that the PU biopolymer used caused a change in the chemical structure of the base bitumen and reactions between the polyurethane and the active groups in the bitumen occurred during the mixing process with bitumen. In another study (Gong, Liu, Wang, et al., 2023), it was reported that only some physical cross-linking points were observed between one of the two different PU biopolymers used and bitumen and that there was no chemical bonding. However, in the same study (Gong, Liu, Wang, et al.,

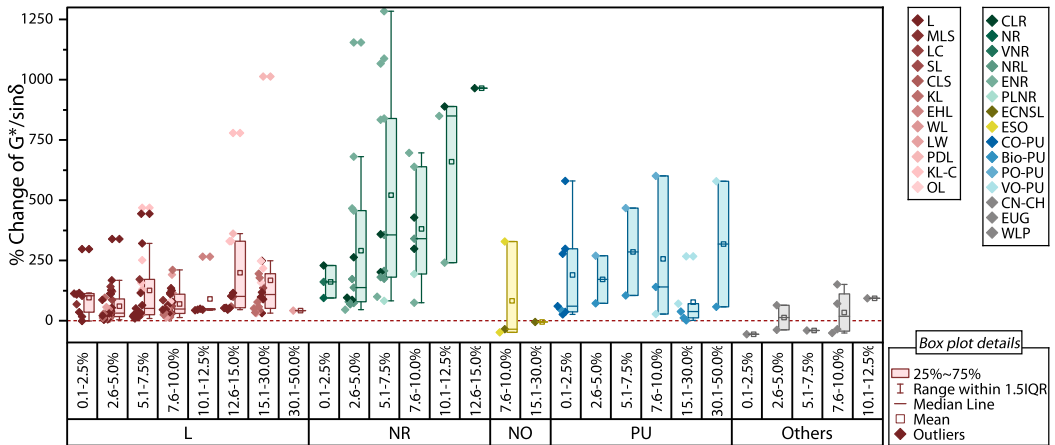


Figure 17. Effect of biopolymer addition on the rutting parameter of base bitumen at $64 \pm 6^\circ\text{C}$.

2023), it was determined that depending on the curing time of the modified bitumen of another PU biopolymer used, the -NCO groups in PU reacted with active hydrogen groups in bitumen to form carbamate or urea groups. Based on these findings in the studies, it can be said that, in general, a chemical reaction may occur between the active groups of PU and bitumen, and the PU group may cause a change in the chemical structure of bitumen. However, it can be stated that this interaction varies depending on the properties of the PU biopolymer synthesised in the study and the curing time of the modified bitumen.

4.2.3. Rheological properties

Currently, the evaluation of bitumen has been based on performance-based rheological properties. Therefore, the articles in the pool were analyzed to clearly assess the effect of the biopolymer on the rheology of the base bitumen. Initially, the rutting parameter ($G^*/\sin\delta$), which is the most preferred rheological criterion, was analyzed, and the results are given in Figure 17. When the effect of biopolymers in the L main group on the base bitumen is analyzed, it is understood that adding L increases the rutting parameter of the base bitumen as a unified result in all studies in the literature. Moreover, it can be said that there is a linear relationship between the increasing amount of L and the rutting parameter. On the other hand, when the effect of the NR main group on the rutting parameter of base bitumen is analyzed, it is possible to say that the addition of NR has a similar effect on the rutting parameter. However, the level of increase in the related parameter of the biopolymers in the NR group is relatively higher than that of the biopolymers in the L group. It can also be considered that for the NR group, increasing the additive ratio increases the rutting parameter more effectively.

It can be said that there are not enough articles in the literature on the rutting resistance of biopolymers in the NO main group. Moreover, the results of the existing studies are not in harmony regarding their effects. In some studies (Fuhaid et al., 2018; Uchoa et al., 2021), NO addition decreased the rutting parameter of bitumen, while a sample in (Fuhaid et al., 2018) increased it. The main reason for the increase in the rutting parameter of this sample could be the curing process of the ESO sample. Therefore, the effect of biopolymers in the NO group on the rutting parameter of base bitumen is closely related to the production process of the respective biopolymer. On the other hand, when the effect of the PU group on the rutting of bitumen is analyzed, it is observed that the addition of PU increases the rutting parameter as a unified result in all studies.

The results obtained by analyzing the articles in the pool that examined the performance of biopolymer-modified bitumen under repeated loadings by MSCR test are given in Figure 18. From Figure 18(a), it is possible to evaluate the effect of biopolymer addition on the $J_{nr,3,2}$ values of the

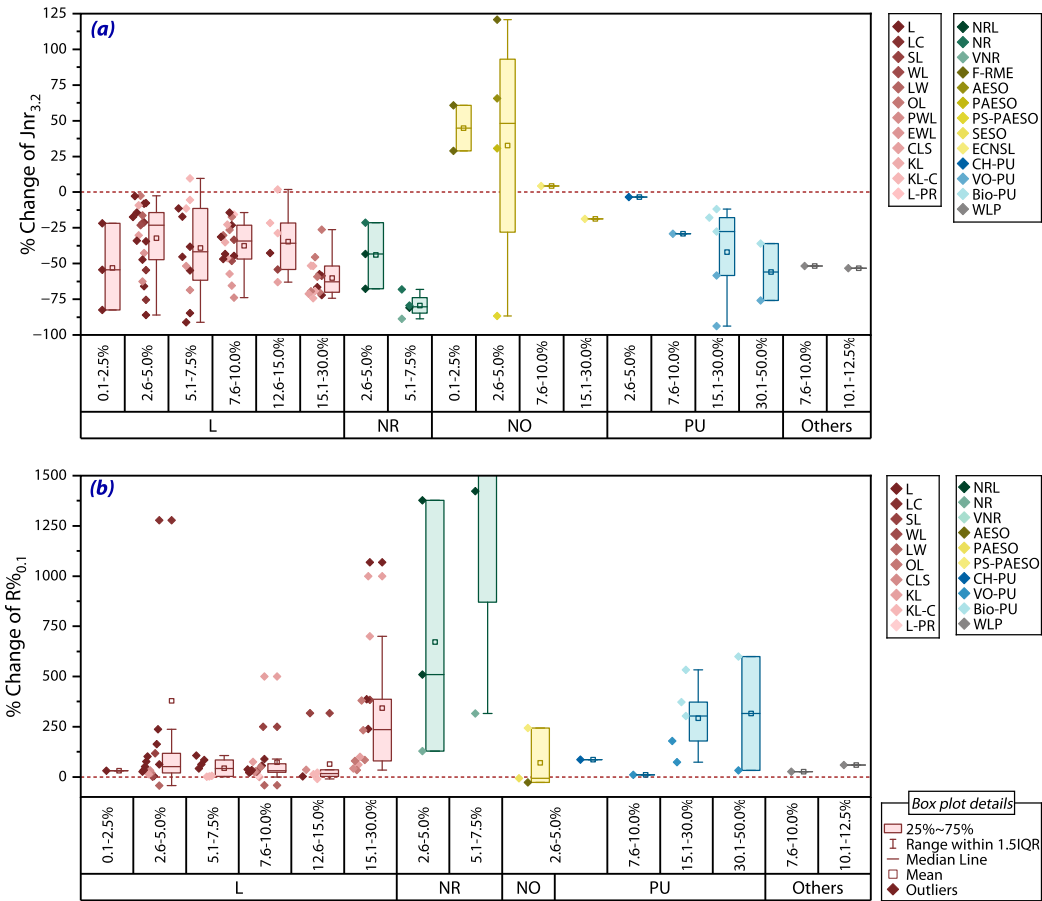


Figure 18. Effect of biopolymer addition on (a) $Jnr_{3,2}$ and (b) $R\%_{0,1}$ values of base bitumen at $64 \pm 6^\circ\text{C}$.

base bitumen. Accordingly, when the effect of biopolymers in the L main group on $Jnr_{3,2}$ values is analyzed, it is understood that in most studies, the reduction of the $Jnr_{3,2}$ value is observed. However, similar to the penetration results, in the range of 2.6%–30.0% addition ratio, the reduction effect on the $Jnr_{3,2}$ value became more pronounced with increasing addition ratio. In contrast, an unexpectedly more severe effect was observed at low ratios such as 0.1–2.5%. On the other hand, when the effect of biopolymers in the NR main group on the $Jnr_{3,2}$ value is examined, it is understood that the $Jnr_{3,2}$ value decreased in all studies. However, the number of studies examining the effect of NR biopolymers on base bitumen over $Jnr_{3,2}$ values is quite limited in the literature. Therefore, there is a gap to be filled in the literature on this subject. Similarly, the number of studies in the literature is very limited for the biopolymer in the NO and PU main group.

When the effect of biopolymers in the NO group on the $Jnr_{3,2}$ value of base bitumen is examined, it is generally understood that it increases the $Jnr_{3,2}$ value. However, a decreasing effect was also observed in some studies (J. Li, Z. Su, et al., 2021; Uchoa et al., 2021). When the related studies are examined for this reason, it is understood that both the additive ratio and the biopolymer production process are effective. Finally, when the analysis results of the biopolymers in the PU main group are examined, it is seen that the addition of PU biopolymer decreases the $Jnr_{3,2}$ value of the base bitumen as a unified result in all studies.

In order to examine in general terms how the addition of biopolymers affects the rutting resistance of base bitumen, it would be helpful to consider $G^*/\sin\delta$ and $Jnr_{3,2}$ values in combination. Accordingly,

it is possible to say that a similar effect on rutting resistance is obtained for the results of both parameters. In fact, considering the mean values for each main biopolymer group and the results shown by the majority of the studies, it is possible to say that L, NR, and PU biopolymer groups increase the rutting resistance of the base bitumen, while NO biopolymer group decreases it.

In Figure 18(b), it is possible to observe the effect of biopolymer addition on the elastic recovery ability of the base bitumen under repeated loads through the change in $R\%_{0.1}$ values. Accordingly, it is observed that the majority of biopolymers in the L main group increase the $R\%_{0.1}$ value of the base bitumen, and it is not possible to establish a correlation between the additive ratio and the level of increase in the $R\%_{0.1}$ value based on the available data. On the other hand, in limited studies on biopolymers in the NR main group, a very high increase in the $R\%_{0.1}$ of base bitumen has been observed with the addition of NR. It is helpful to point out here that since the $R\%_{0.1}$ value is a percentage expression, the fact that the value of the base bitumen is very close to zero causes the result of the modified bitumen to be misleadingly high in percentage change calculations. Here, the exceptionally high values in the NR group are due to this reason and may be misleading. However, it is not wrong to say that biopolymers in the NR group increase the $R\%_{0.1}$ of the base bitumen. Nevertheless, more MSCR test data on bitumen modified with NR biopolymers are needed in the literature to quantitatively evaluate this increase more precisely.

When the effect of NO on the $R\%_{0.1}$ value is analyzed, it is understood that the increase or decrease depends on the type of biopolymer in the NO group and the production process. On the other hand, adding biopolymers in the PU main group increases the $R\%_{0.1}$ value of the base bitumen, a common result obtained in the studies conducted with the related material.

The rheological properties of bitumen at low temperatures are also crucial for evaluating their performance. Accordingly, the effects of biopolymers on the creep stiffness and m -value parameters of the base bitumen, which are the results of the BBR test, were analyzed through the studies in the article pool, and the results obtained are given in Figure 19. When the results of the creep stiffness analyses in Figure 19(a) are examined, a unified result could not be observed in the studies regarding the effect of biopolymers in the L main group on base bitumen. In fact, in some studies (Z. Chen et al., 2019; D. Wang et al., 2019; K. Wang et al., 2023; J. Wu et al., 2021; C. Xu et al., 2021; G. Xu et al., 2017; R. Zhang et al., 2021), a decrease in the creep stiffness of the base bitumen was observed with the addition of biopolymers in the L group, while in other studies (Al-falahat et al., 2024; Batista et al., 2018; Fakhri & Norouzi, 2022; R. Li et al., 2020; Norgbey et al., 2020; X. Shi & Xu, 2023; G. Xu et al., 2017; Xue et al., 2022; Yu et al., 2021; R. Zhang et al., 2021; Y. Zhang, Liu, et al., 2022; Y. Zhang, Si, et al., 2023; S. Zhou et al., 2023) an increase was observed. When the related studies are examined, it is understood that the effect of biopolymers in the L main group on the creep stiffness varies according to the type of biopolymer and the addition ratio. Therefore, it would not be correct to generalise creep stiffness for the L main group. On the other hand, when the effect of biopolymers in the NR main group on creep stiffness is examined, it is observed that the biopolymers in the NR main group reduce creep stiffness, which is a unified result in all studies. Furthermore, it is understood that the effect of decreasing the creep stiffness becomes more pronounced as the NR content increases.

The effect of biopolymers in the NO main group on the creep stiffness of base bitumen also varies. However, most studies (Arabzadeh et al., 2022; Chen et al., 2018; Quan et al., 2024) stated that biopolymers in the NO group decreased the creep stiffness of the base bitumen, while only one study (Chen et al., 2018) mentioned an increasing effect. When the relevant study is examined, it is understood that the reason for this effect is the type of material used. In fact, in the same study, an AESO produced by the laboratory researchers and a commercial AESO product were used. The related study results show that the commercial product AESO increases the creep stiffness (Chen et al., 2018). Therefore, although it is possible to say that most NO biopolymers reduce the creep stiffness, this effect may differ depending on the type of NO. On the other hand, it is understood that very few BBR tests have been conducted in the literature for biopolymers in the PU main group. When evaluated based on the available data, it is understood that biopolymers in the PU group reduce the creep stiffness of base bitumen in all studies.

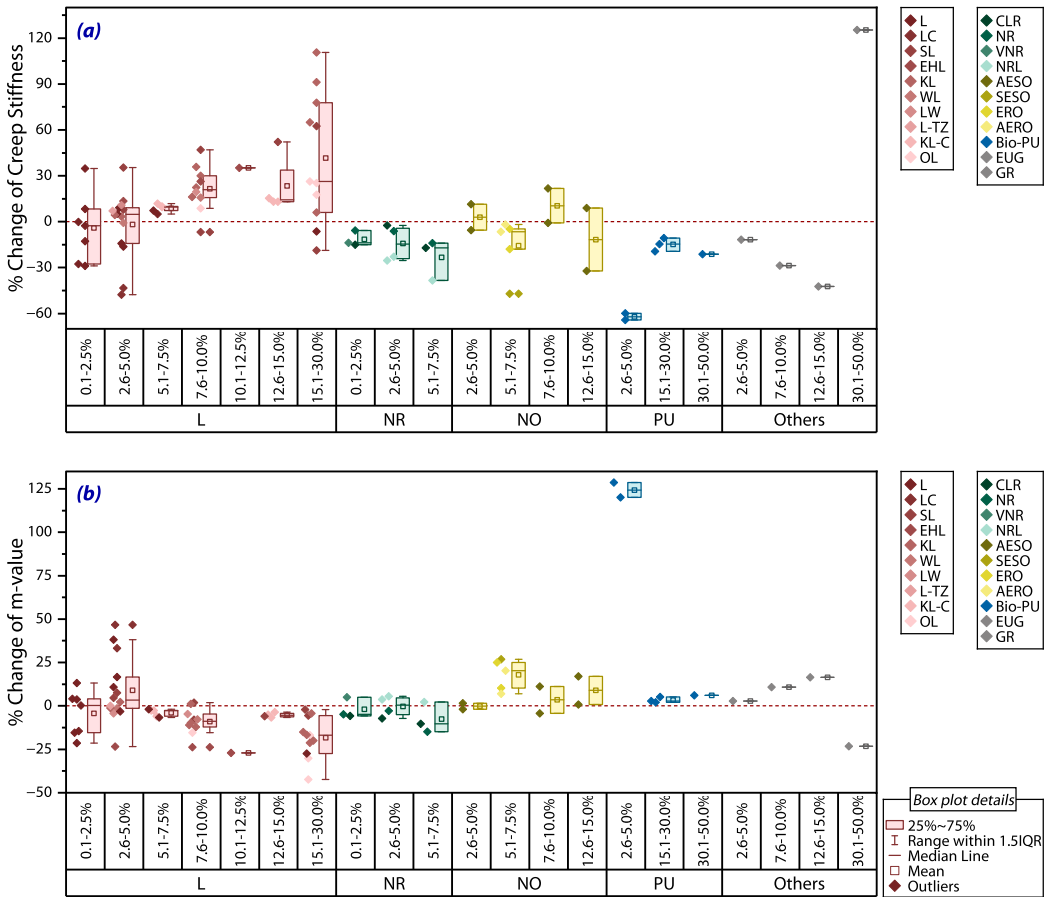


Figure 19. Effect of biopolymer addition on (a) creep stiffness and (b) m-value of base bitumen at $-18 \pm 6^\circ\text{C}$.

When the effect of biopolymers on the m-value of base bitumen is analyzed in Figure 19(b), no unified result can be observed for the biopolymers in the L main group. It is understood that some biopolymers in the L group increase the m-value, and some decrease it. On the other hand, when the NR main group was examined, it was observed that the common result obtained in creep stiffness was not obtained here, and the studies in the literature diverged from each other. In fact, it was stated that the m-value increased in some studies (F. Liu et al., 2023; Yong Wen et al., 2017) and decreased in others (Albuaymi et al., 2023; Wititanapanit et al., 2021) by adding biopolymers in the NR group to the base bitumen. The main reason for this situation can be considered as the type of NR biopolymer used. The addition of NRL (Yong Wen et al., 2017) and VNR (F. Liu et al., 2023) showed an enhancement effect, while the addition of NR (Wititanapanit et al., 2021) and CLR (Albuaymi et al., 2023) showed a reduction effect. Therefore, it would not be correct to make a definite generalisation about the effect of the NR main group on the m-value based on the available data. When the effect of biopolymers in the NO group on the m-value is evaluated, it is understood that they generally show an increasing effect. However, a decreasing effect was observed in one study (Chen et al., 2018), and the reason for this was previously mentioned when evaluating the creep stiffness. On the other hand, when the effect of biopolymers in the PU main group on the m-value is examined, although there are limited studies, all studies stated that PU biopolymers increase the m-value as a unified result.

When the creep stiffness and m-value parameters of the BBR test are evaluated in combination, it is possible to understand the overall effect of biopolymers on the low-temperature rheology of the base

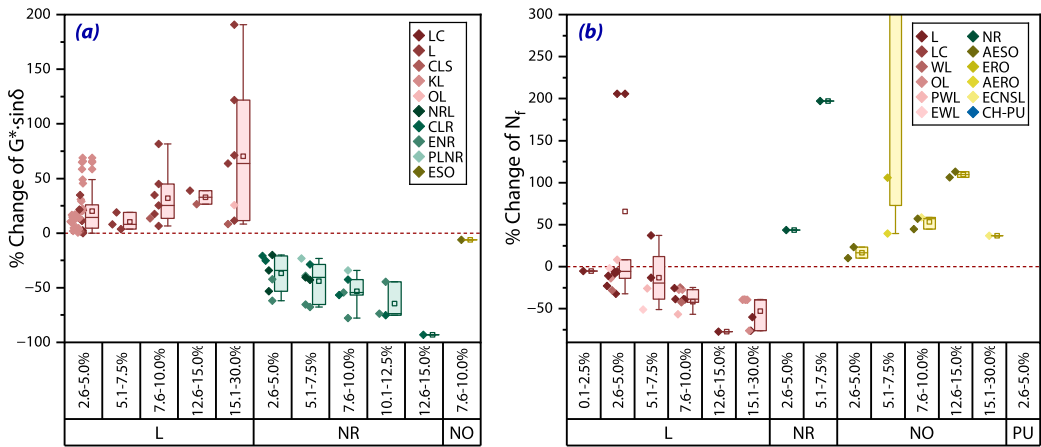


Figure 20. Effect of biopolymer addition on fatigue performance of pure bitumen: (a) $G^* \cdot \sin \delta$ and (b) N_f .

bitumen. Accordingly, it could not be said that there is a common effect for the biopolymers in the L main group in terms of both creep stiffness and m-values. Therefore, the effect of biopolymers in the L group on the low-temperature performance of the base bitumen depends solely on the L type. When the performance of biopolymers in the NR main group is considered, although the studies show a common effect of reduction in creep stiffness, the results of the studies diverge for the m-values results. Therefore, while NR biopolymers will benefit and improve performance in terms of creep stiffness, the benefit in terms of m-value will vary depending on the type of NR. Biopolymers in the NO main group generally improve the low-temperature performance of the base bitumen, except for a few of them. Finally, biopolymers in the PU group are expected to impact low-temperature performance positively. Additionally, it should be added that considering the limited number of studies in which BBR tests have been conducted, it is obvious that the effect of different types of biopolymers on the low-temperature performance of base bitumen still needs to be investigated.

Another critical performance characteristic determined by utilising the rheology of the bitumen is fatigue resistance. The $G^* \cdot \sin \delta$ parameter of the DSR test and the N_f parameter of the LAS test are frequently used to evaluate the fatigue resistance of bitumen. Accordingly, to examine the effect of biopolymer addition on the fatigue performance of base bitumen, articles in the pool that include the relevant tests in their methodology were analyzed, and the results obtained are given in Figure 20. When the $G^* \cdot \sin \delta$ parameters in Figure 20(a) are evaluated, it is understood that the biopolymers in the L main group increase the $G^* \cdot \sin \delta$ value of the base bitumen. Furthermore, the magnitude of the enhancement effect is directly proportional to the increasing additive ratio. On the other hand, when the effect of biopolymers in the NR main group on $G^* \cdot \sin \delta$ was analyzed, it was determined that the addition of NR biopolymer to the base bitumen decreased the $G^* \cdot \sin \delta$ value of the base bitumen as a common result of all studies. Moreover, the magnitude of the decrease in the $G^* \cdot \sin \delta$ value increases as the addition rate increases. On the other hand, only one study (Fuhaid et al., 2018) investigated the $G^* \cdot \sin \delta$ parameter for the NO main group, while $G^* \cdot \sin \delta$ values for the PU main group could not be found in the existing article pool. The only study result for ESO biopolymer in the NO group shows that adding this material to the base bitumen reduces the $G^* \cdot \sin \delta$ value by a small amount.

When the results of the analysis of N_f parameters in Figure 20(b) are examined, it is seen that the biopolymers in the L main group reduce the N_f parameter of the base bitumen as a unified result in other studies, except for the results in only two studies (Sun et al., 2023; D. Wang et al., 2019). The reason for the increase in the related studies can be considered to be related to the type of L biopolymer used. On the other hand, the N_f parameters of the biopolymer in the NR main group were investigated in only one study (Wititanapanit et al., 2021), and according to the results of the relevant study,

the addition of NR biopolymer increases the N_f value of the base bitumen. In addition to these, only one study includes the $G^* \cdot \sin \delta$ value of biopolymers in the NO main group, while three studies (Chen et al., 2018; Quan et al., 2024; Uchoa et al., 2021) examined the N_f values. The results obtained from the related studies show that the biopolymers in the NO group will increase the N_f value of the base bitumen. Finally, it is seen that fatigue parameters for bitumen modified with biopolymers in the PU main group have not been investigated.

A general evaluation of the effect of biopolymers on the fatigue resistance of base bitumen shows that biopolymers in the L main group negatively affected the fatigue resistance of base bitumen, while biopolymers in the NR and NO main groups increased it. However, the number of studies that investigated the fatigue resistance of biopolymer-modified bitumen in detail is very limited in the literature. Therefore, in order to fill this gap, studies must be conducted examining the performance of different types of biopolymers.

4.3. Effect of biopolymers on asphalt mixture performance

In order to evaluate the performance of asphalt pavement upon modification of base bitumen with biopolymers, asphalt mixture samples prepared with biopolymer-modified bitumen should be subjected to macro-scale asphalt mixture tests. However, since the use of biopolymers as bitumen additives is a very recent subject, as can be seen from Figure 11(b), micro-scale bitumen experiments were mostly included in published articles. It is expected that macro-scale tests will be conducted once the benefits of biopolymer addition have been proven by micro-scale tests in the literature. However, the scarcity of macro-scale mixture tests in the existing studies shows a significant gap in the literature on this subject.

In this section, studies that investigated the performance of biopolymer-modified bitumen at the asphalt mixture scale were analyzed. Accordingly, considering the most frequently used tests, samples prepared with biopolymer bitumen versus base bitumen were compared in terms of deformation resistance, stiffness, fatigue resistance, and durability properties.

4.3.1. Deformation resistance and stiffness

One of the most frequently used methods for evaluating the stability of asphalt mixtures is the Marshall stability and flow test. Most studies in the article pool that conducted asphalt mixture tests included this method. Therefore, to evaluate how biopolymer modification affects the stability of the asphalt mixture, the Marshall stability and flow values in the relevant studies were analyzed, and the results obtained are given in Figure 21. When the effect of biopolymers on Marshall stability values in Figure 21(a) is analyzed, it is seen that Marshall stability increases as a unified result in the studies conducted with biopolymers in the L main group. However, it is impossible to establish a relationship between the L additive ratio and stability increase based on the available data. On the other hand, when the biopolymers in the NR main group are evaluated, it is seen that the biopolymers in the NR group generally increase the stability of the asphalt mixture, except for the findings in two studies (Hadiwardoyo et al., 2016; Shafii et al., 2018). It should be noted here that the reason for the different results obtained in some studies from the trend in the literature may be that many factors other than bitumen properties, such as aggregate gradation in the mixture and mixture voids, may affect the experimental results. Therefore, in general terms, it is not controversial to say that biopolymers in the NR group increase Marshall stability values. Finally, when the effect of biopolymers in the NO group on stability is examined, it is understood that different effects were observed in different studies. While in one study, a slight decrease in the Marshall stability value was observed with the addition of ESO in (Fuhaid et al., 2018), in the other study, an increase in the stability value was observed in (Q. Lu et al., 2021). Especially in the study (Q. Lu et al., 2021), the fact that asphalt mixture samples were prepared by adding bitumen modified with additives in equal ratios to the samples produced with different gradations and that significant differences in stability values occurred even among these samples reveals the effect of gradation selection. Therefore, the few studies in the literature on asphalt mixtures prepared

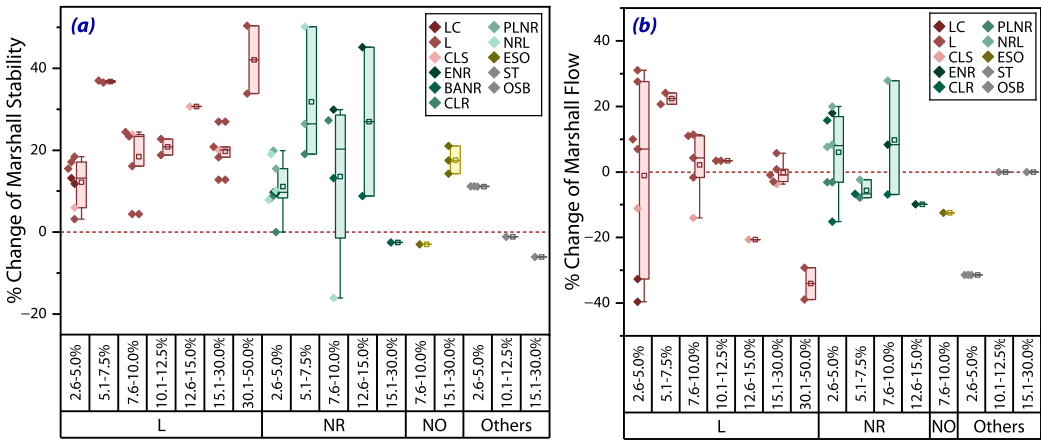


Figure 21. Effect of biopolymer modification on Marshall (a) stability and (b) flow values of asphalt mixture.

with biopolymer bitumen in the NO group prevent a general evaluation. Furthermore, no Marshall stability values were found for biopolymers in the PU main group in the article pool.

When the Marshall flow values in Figure 21(b) are analyzed, the effect of biopolymer modification on the deformation capacity of the asphalt mixture can be examined. Accordingly, when examined for the biopolymers in the L main group, it is observed that the Marshall flow values decreased in some studies (Fatemi et al., 2022a; Sundstrom et al., 1983; D. Wang et al., 2019) while they increased in some studies (Sundstrom et al., 1983; C. Xu et al., 2021; Zahedi, Zarei, & Zarei, 2020; Zahedi, Zarei, Zarei, et al., 2020). This situation prevents a general evaluation of the effect on Marshall flow values for the L main group. However, it is possible to say that the effect on Marshall flow values varies depending on the type and utilisation rate of biopolymers in the L main group. Additionally, a similar result is encountered when evaluations are made regarding the biopolymers in the NR main group. Indeed, while the use of bitumen modified with biopolymers in the NR group increased the Marshall flow values in some studies (Abd El Rahman et al., 2012; Abdulrahman et al., 2021; Hoy et al., 2024; Shafii et al., 2018), in other studies (Abd El Rahman et al., 2012; Ansari et al., 2023; Norfazira Mohd Azahar et al., 2019; Begam Rasheda et al., 2022; Subagio et al., 2021), these values decreased. Therefore, although it is not possible to reach a clear conclusion for the NR main group, it is understood that the effect of NR addition varies according to the type of biopolymer. Finally, when the results of the only study (Fuhaid et al., 2018) with ESO, one of the biopolymers in the NO main group, on Marshall flow values are examined, it is seen that the addition of ESO decreases these values.

When the effect of biopolymers on Marshall stability and flow values is evaluated in general terms, it can be said that biopolymers in the L and NR main group will generally increase Marshall stability values. However, since the effect of biopolymers on Marshall flow value varies, it is not possible to make a precise assessment based on the available data in the literature. Considering that no study in the article pool conducted Marshall flow and stability tests with biopolymers in the PU main group, more research is needed to clarify how different biopolymers affect the relevant parameters.

One of the most critical distresses that develop in asphalt pavements is rutting. Therefore, the rutting resistance of asphalt mixtures is vital for their performance in service. In this direction, in order to evaluate the rutting performance of biopolymer modification, the rut depths obtained in the studies that conducted rutting tests in the article pool were analyzed, and the results are given in Figure 22. When this figure is analyzed regarding biopolymers in the L main group, it is observed that the L group increases the rutting resistance as a common result of all studies in the article pool. On the other hand, when the results of the NR main group are evaluated, it is understood that, in general, mixtures prepared with bitumen modified with NR biopolymers have lower rutting values compared to base

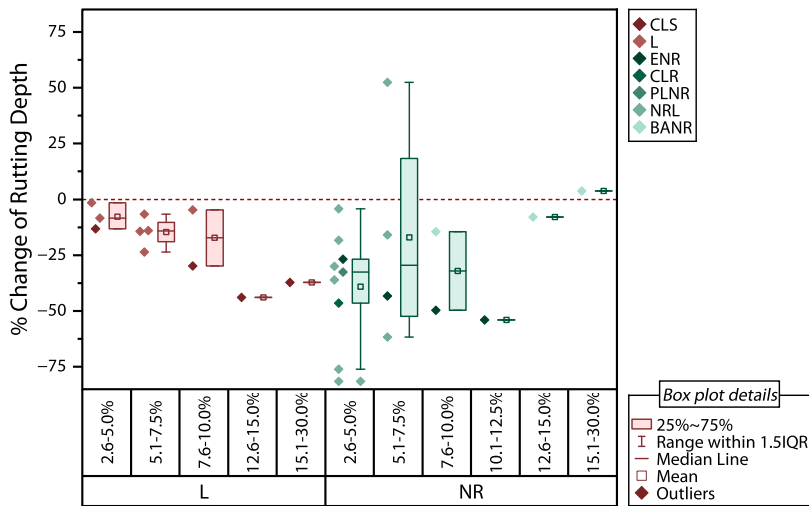


Figure 22. Effect of biopolymer modification on the rutting value of asphalt mixture.

bituminous mixtures, except for the results of some samples in two studies (Hadiwardoyo et al., 2016; Hoy et al., 2024). In those two studies where the results differed from the general trend, it was stated that the reason for the increase in rutting was the addition of NR at high ratios. As a result, when a general evaluation is made regarding rutting resistance, it is possible to say that biopolymers in both L and NR groups will generally have an increasing effect on rutting resistance. However, it is also worth mentioning that using extremely high amounts of NR will negatively affect the rutting resistance. It should be noted here that very few studies in the literature have conducted macro-scale rutting tests to evaluate the performance of biopolymer-modified bitumen. Data for the NO and PU groups are not available in the article pool of this study. Therefore, there is a significant gap in the literature regarding evaluating such a critical property, and more studies need to be conducted to reach a general conclusion.

Indirect tensile strength and resilient modulus of asphalt mixture samples are among the critical parameters in the design of asphalt pavements. In order to evaluate the effect of biopolymers on the performance of base bitumen at the asphalt mixture level, the articles in the pool that included indirect tensile strength and resilient modulus tests were analyzed. The results obtained from the analysis are given in Figure 23. When the effect of biopolymers on the indirect tensile tests in Figure 23(a) is examined, it is understood that the biopolymers in the L main group generally have an increasing effect on indirect tensile strength, except for the results in some studies (Duarte Mendonça et al., 2023; I. P. Pérez et al., 2019). It is thought that the reason for the exception of the general effect in the related studies may be related to the biopolymer type and biopolymer addition ratio. On the other hand, a similar result is also valid for biopolymers in the NR main group. Although it is stated that the addition of NR in most studies has an increasing effect on the indirect tensile strength, a decreasing effect was also observed in some studies (Abdulrahman et al., 2021; Norfazira Mohd Azahar et al., 2019; Hoy et al., 2024).

Among the NO main group biopolymers, ESO was investigated for indirect tensile strength only in one study. As a result of this study (Q. Lu et al., 2021), it was stated that ESO increased the indirect tensile strength. Finally, when the results were analyzed in terms of biopolymers in the PU main group, it was observed that only one study was conducted for Bio-PU. In the related study (Kazemi et al., 2024), different aggregate types were investigated, and it is understood that adding Bio-PU increases the indirect tensile strength regardless of the aggregate type.

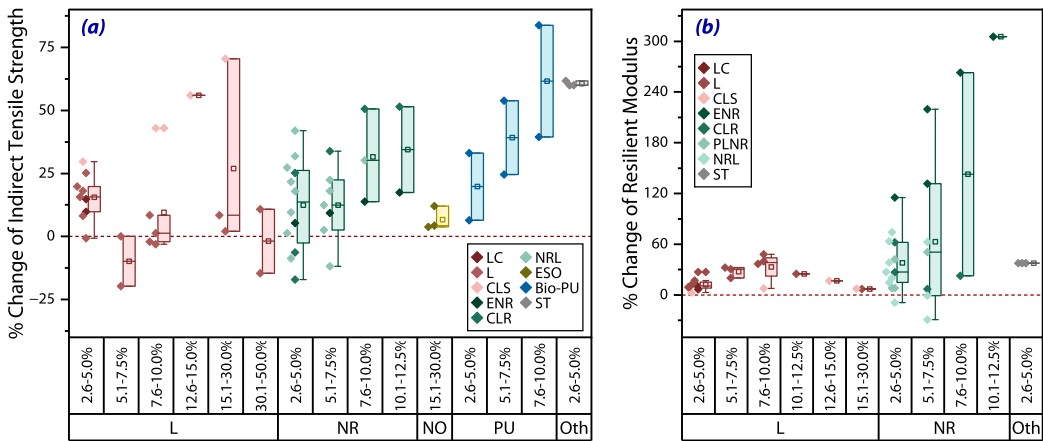


Figure 23. Effect of biopolymer modification on (a) indirect tensile strength and (b) resilient modulus of asphalt mixture.

When the effect of biopolymers on the resilient modulus is analyzed in Figure 23(b), it is observed that the biopolymers in the L main group have an increasing effect on the resilient modulus as a unified result of the studies. Furthermore, it is understood that the addition ratio of more than 10% for L biopolymers reduces the enhancement effect on the resilient modulus. Therefore, this ratio can be considered the optimum ratio for resilient modulus. On the other hand, when the resilient modulus results of the biopolymers in the NR main group are evaluated, it is observed that an increase in the values occurs generally, except for the results of some samples in a study (Hoy et al., 2024). When the related study is examined, it is understood that the NRL ratio used according to different aggregate types provides optimum benefit at certain ratios and decreases the resilient modulus at certain ratios. Therefore, the effect of biopolymers in the NR group may vary in some cases according to the aggregate type and utilisation rate.

In general, when indirect tensile strength and resilient modulus are evaluated, it is possible to say that biopolymers in the L and NR groups generally have an increasing effect on these parameters. However, resilient modulus values of asphalt mixture specimens prepared with bitumen modified with biopolymers in the NO and PU main groups were not found in the studies in the article pool. Therefore, studies need to be conducted in which modulus of elasticity experiments are determined with the relevant biopolymer groups and contribute to the literature.

4.3.2. Fatigue resistance

Additives can increase the fatigue resistance of asphalt mixtures and extend their service life. Accordingly, it would be helpful to evaluate the effect of biopolymers on fatigue life through indirect tensile fatigue (ITF) values of asphalt mixture specimens prepared with biopolymer-modified bitumen. Among the studies in the article pool, analyses were carried out on the ones that included the ITF test, and the results obtained are given in Figure 24. However, it should be noted that the number of studies in which the relevant test was carried out is very limited, and there is a significant gap in the literature at this point. However, when the only study (Zahedi, Zarei, & Zarei, 2020) that examined how the addition of L biopolymer affects the fatigue performance of base bitumen on asphalt mixtures is examined, it is understood that L biopolymer increases the fatigue life at 3% and 6% and decreases it at 9% and 12%. Accordingly, it can be concluded that using L biopolymer in proportions greater than 6% is not recommended for the fatigue performance of the asphalt mix. However, more studies need to be conducted in the literature to reach a more precise and generalised conclusion about the fatigue performance of biopolymers in the L main group. On the other hand, when the results for biopolymers in the NR main group are examined, it is seen that NR biopolymers increase fatigue life, except for only one sample in

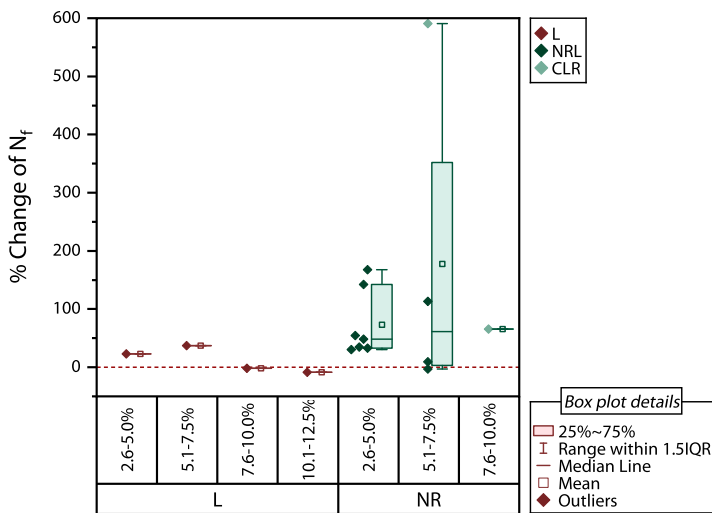


Figure 24. Effect of biopolymer modification on the fatigue life of asphalt mixture.

one study (Hoy et al., 2024). The decreasing effect in that study is related to the type of aggregate used and the biopolymer addition ratio. Therefore, NR biopolymers can generally be considered to increase fatigue life. Furthermore, no data on ITF experiments for biopolymers in the NO and PU main groups were found in the articles in the pool. Therefore, investigating the effect of different biopolymer types on the fatigue performance of asphalt mixtures is a topic that will contribute to the literature.

4.3.3. Durability

Whether asphalt pavements are resistant to environmental impacts such as rain and freeze-thaw significantly influences the quality of the road. Therefore, researchers can test asphalt mixture samples subjected to different conditions and compare them with their unconditioned counterparts to get a prediction about the durability of the samples. Accordingly, to evaluate the effect of biopolymers on the durability of asphalt mixtures, the articles in the pool were analyzed, and the results are given in Figure 25. The evaluation of the indirect tensile strength ratio (TSR) values in the dry-wet condition in Figure 25(a) shows that the L biopolymer generally decreases the TSR. This indicates that the durability of the asphalt mix specimen prepared with this biopolymer is lower than that of the base bituminous mixture specimen. However, in one study (I. P. Pérez et al., 2019), it was found that some samples would increase the TSR value. When the related study is analyzed, it is understood that the bitumen content of the asphalt mixture sample and the amount of L added are significant to TSR values. Therefore, although most studies indicate that L biopolymer harms TSR values, selecting the appropriate additive ratio and bitumen content can reverse this adverse effect. On the other hand, when the effect of biopolymers in the NR main group on dry-wet condition TSR values is examined, it is seen that the use of NR increases the TSR value.

When Figure 25(b) and Figure 25(c) are examined, it is understood that biopolymers in both L and NR main groups increase the residual Marshall stability value in dry-wet conditioning and biopolymers in L, NR, and PU main groups increase the TSR values in freeze-thaw condition. Therefore, it can be concluded that biopolymers can improve the durability of asphalt mixtures. However, the small number of studies investigating the effect of biopolymers on the durability of asphalt mixtures prevents a definitive and generalised conclusion. Therefore, more studies are needed to fill the gap in the literature on this subject.

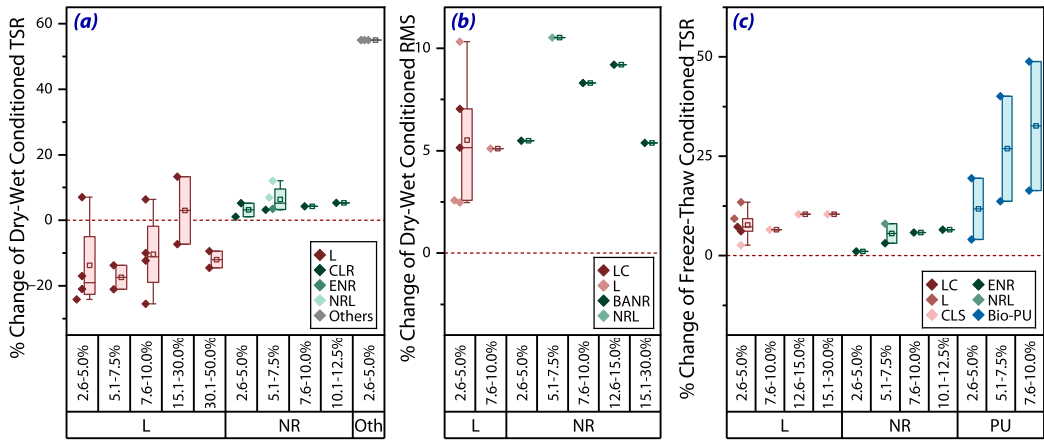


Figure 25. Effect of biopolymer modification on asphalt mixture durability: (a) dry-wet conditioned TSR, (b) dry-wet conditioned RMS, and (c) freeze-thaw conditioned TSR.

Table 7. Summary of the effect of biopolymers on bitumen and asphalt mixture performance according to the main biopolymer groups in this study.

Parameters	Main biopolymer groups in this study			
	Lignin and derivatives	Natural Rubber and derivatives	Natural oil based polymers	Bio-based polyurethanes
Effect on the bitumen properties				
Penetration	▼*	▼	▼▲	▼▲
Softening point	▲**	▲	▼	▲
Ductility	▼	▼	N/A***	N/A
Elastic recovery	▼▲	▲	N/A	N/A
Rotational viscosity	▲	▲	▼	▲
$G^*/\sin\delta$	▲	▲	▼▲	▲
$J_{nr3,2}$	▼	▼	▼▲	▼
$R_{0.1}$	▲	▲	▼▲	▲
Creep stiffness	▼	▼	▼	▼
m-value	▼	▼▲	▲	▲
$G^* \cdot \sin\delta$	▲	▼	N/A	N/A
N_f	▼	▲	▲	N/A
Effect on the asphalt mixture properties				
Marshall stability	▲	▲	▲	N/A
Marshall flow	▼▲	▼▲	▼	N/A
Rutting depth	▼	▼	N/A	N/A
Indirect tensile strength	▲	▲	▲	▲
Resilient modulus	▲	▲	N/A	N/A
N_f	▼▲	▲	N/A	N/A
Durability	▼▲	▲	N/A	▲

*▼: Decrease, **▲: Increase, ***N/A: Not Available.

4.4. Summary of the biopolymer's effect on bitumen

In this section, the findings of the previous sections on the effect of biopolymer modification on bitumen and asphalt mixture performance are summarised in Table 7. Thus, a general insight into the effect of biopolymers can be obtained. Furthermore, it is possible to observe the gaps in the literature in this table.

5. Conclusions

In this review study, the use of biopolymers derived from sustainable resources in bitumen modification was examined through a detailed analysis of the studies in the literature. Accordingly, firstly, the types of biopolymers used in bitumen modification were grouped, and five main groups were formed: lignin and its derivatives, natural rubber and its derivatives, natural oil-based polymers, biobased polyurethanes, and others. The articles that investigated the physical, chemical, and rheological properties of bitumen modified with biopolymers in these main groups were analyzed. Furthermore, studies examining the properties of macro-scale asphalt mixture samples modified with biopolymers were also analyzed. The results obtained from all the analyses carried out within the scope of the study are given below:

- When the results of penetration and softening point tests in the literature are evaluated, biopolymers in the L and NR main groups increase the consistency of the base bitumen and harden it. On the other hand, the effect of biopolymers in the NO and PU main groups on the hardness of the base bitumen varies depending on the type of biopolymer in the group, and some of them cause an increase in hardness while others cause a decrease.
- When the ductility and elastic recovery test results are analyzed, it is seen that the L and NR main groups increase the ductility of the base bitumen. Moreover, the NR main group increases the elastic recovery ability of the base bitumen, while no definite conclusion can be reached for the L main group. However, there are no studies where relevant tests have been carried out for the NO and PU main groups.
- When the effect of biopolymer additives on the high-temperature performance of base bitumen is analyzed by considering the rheological test results, it is understood that the biopolymers in the L, NR, and PU main groups increase the rutting resistance and high-temperature performance of the base bitumen. On the other hand, it is not possible to reach a clear conclusion for the biopolymers in the NO main group.
- The effect of biopolymers on the low-temperature performance of the base bitumen is analyzed according to the rheological test results, and it is understood that the biopolymers in the NO and PU main groups increase the low-temperature performance of the base bitumen. In contrast, those in the L main group decrease it. However, although it is observed that the biopolymers in the NR main group reduce the creep stiffness, it is not correct to make a general conclusion due to the lack of clear effects on the m -value.
- Micro-scale bitumen tests on the effect of biopolymers on the fatigue life of base bitumen show that biopolymers in the NR and NO main groups increase the fatigue life of base bitumen, while those in the L main group decrease it. On the other hand, no data on the relevant experiment for the PU main group was found in the literature.
- When the deformation resistance and stiffness of asphalt mixture specimens prepared with biopolymer-modified bitumen are compared with asphalt mixture specimens prepared with base bitumen, it is generally understood that biopolymers in the L, NR, and NO main groups increase the stiffness and deformation resistance of asphalt specimens. However, it is not possible to conclude on the deformation resistance of PU main group biopolymers since the number of studies in which PU main group biopolymers were used as modifiers in asphalt mixture specimens is very limited.
- Evaluating the fatigue resistance of biopolymer additives by macro-scale asphalt mixture tests shows that asphalt mixture samples prepared with bitumen modified with biopolymers in the NR main group have higher fatigue life than those prepared with base bitumen. On the other hand, it was not possible to reach a unified conclusion from the studies conducted with biopolymers in the L main group. In contrast, no studies were found for the biopolymers in the NO and PU main groups.
- When the effect of biopolymers on the durability of asphalt mixture samples is analyzed, it is understood that the biopolymers in the NR and PU main group increase the durability, while it

is not possible to reach a unified conclusion from the existing studies in the literature for those in the L main group. Furthermore, no literature study investigates the durability of asphalt mixtures prepared with bitumen modified with biopolymers in the NO main group.

This study was conducted to examine the effect of biopolymers on the properties of bitumen in general, but the study has certain limitations. In fact, while creating the article pool to be used in the analyses of the study, other types of studies, such as conference proceedings and thesis, were excluded. Although this is to ensure that the articles in the pool have preferably been through an academic peer review process, it is possible that there are new and current studies that could be included in the analysis of this study. On the other hand, the authors of this study are of the opinion that the results of other types of studies that could be included would not affect the overall conclusion of the analyses.

As a result of this study, it is seen that the number of studies examining the properties of bitumen modified with biopolymers, especially at the asphalt mixture scale, is very limited in the literature. Therefore, there is a need for researchers to conduct many more scientific studies to fill this gap in the literature. Furthermore, considering the broadness of the biopolymer concept and the diversity of these polymers, it is thought that it would be interesting to examine the effect of different biopolymers on bitumen performance, which have not yet been tried in bitumen modification. Beyond bitumen performance studies, research on the life cycle assessment (LCA) of biopolymer modification and field applications of biopolymer-modified asphalt pavements remains limited. Further studies in these areas would provide valuable contributions to the literature.

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