Chapter 1

Asphalt Modified with Biomaterials as Eco-Friendly and Sustainable Modifiers

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Additional information is available at the end of the chapter

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Abstract

High construction costs, when combined with awareness regarding environmental stewardship have encouraged the use of waste and renewable resources in asphalt modification. Increasing energy costs and the strong worldwide demand for petroleum have encouraged the development of alternative binders to modify or replace asphalt binders. The benefits of using alternative binders are that they can help save natural resources and reduce energy consumption while maintaining and in some cases improving asphalt performance. Common alternative binders include engine oil residue, bio-binder, soybean oil, palm oil, fossil fuel, swine waste, and materials from pyrolysis. Chemical compositions of the majority of these alternative binders are similar to those of unmodified asphalt binders (e.g. Resin, saturates, aromatics, and asphaltene). On the other hand, tests indicate the wide variability in the properties of alternative binders. Also, the chemical modification mechanism for asphalt with alternative binders depends clearly on the unmodified asphalt and is consequently not well understood. For energy sustainability, environment-friendly materials and an urgent need for infrastructure rehabilitation that more research is needed to evaluate the alternative binders for use in asphalt modification. The alternative binders should have moisture resistance and good aging characteristics.

Keywords: asphalt, sustainable, alternative binders, biomaterials, eco-friendly, bio-binder

1. Introduction

1.1. General introduction

Bio-materials that are used as binders in asphalt mixtures are termed “bio binders” [1]. The interest on using bio-binders in pavement engineering has significantly grown over the last decades due to the increasing scarcity of raw materials and environmental concerns about the use of non-recoverable natural resources.

Common alternative binders include engine oil residue, bio-binder, soybean oil, palm oil, fossil fuel, swine waste, and materials from pyrolysis [2]. Chemical compositions of the majority of these alternative binders are similar to those of unmodified asphalt binders (e.g. Resin, saturates, aromatics, and asphaltene) [3]. On the other hand, tests indicate the wide variability in the properties of alternative binders. Also, the chemical modification mechanism for asphalt with alternative binders depends clearly on the unmodified asphalt and is consequently not well understood [4, 5]. For energy sustainability, environment-friendly materials and an urgent need for infrastructure rehabilitation that more research is needed to evaluate the alternative binders for use in asphalt modification. The alternative binders should have moisture resistance and good aging characteristics [6].

2. Bio binder’s definition and resources

2.1. Definition

Bio-binder is an eco-friendly asphalt binder alternative obtained from non-petroleum-based renewable resources, which should not rival any food material. From the definition, bio-binder can be described as dark brown, high flow, organic liquids that are comprised mainly of highly oxygenated compounds [7–10].

2.2. Bio-binder material sources

A range of different vegetable oils has been investigated in recent times, through the application of scientific research and development, to determine their physical and chemical properties to study their applicability to be used as bio-binders in the pavement industry [11–13]. Bio-oils are produced from plant matter and residues, such as municipal wastes, agricultural crops, and byproducts from agricultural and forestry [8]. Other biomass sources include molasses and rice, sugar, potato starches and corn, gum resins and natural tree, vegetable oils and natural latex rubber, cellulose, lignin, waste oil of palm, peanut oil waste, coconut waste, potato starch, canola oil waste, dried sewerage effluent, and others.

Utilize this bio-binder can be a great potential as a modifier for asphalt binder because of similar chemical properties when compared with crude petroleum as shown in **Table 1**.

Currently, bio-binder is the second leading renewable energy in the nation after hydropower [14, 15].

Physical property	Value
Percent of moisture (wt %)	15–30
pH	2.5
Specific gravity	1.2
Elemental composition (H, C, O, N) (wt %)	(5.5–7.0, 54–58, 35–40, 0–0.2)
Distillation residual (wt %)	Up to 50
Viscosity @500°C (pa)	40–100

Table 1. Typical characteristics of wood-based bio-oils.

3. Materials used for the production of bio-binder

3.1. Bio-oil

Bio-Oil is the liquid produced from the rapid heating of biomass in a vacuum condition [17]. There are many advantages for bio-oils over asphalt from crude oil as they are environment-friendly, renewable, present a great economic opportunity, and provide energy security.

3.1.1. Bio-oil sources and production

3.1.1.1. Thermochemical liquefaction

To obtain more gasoline and other liquid fuels, the quality of the bio-oil is improved by using processes such as thermal cracking and hydrogenation [16, 17]. Upon fractionation, the light hydrocarbon fraction can be used as fuel. The remaining heavy residue called bio-binder can be used as an asphalt binder modifier.

3.1.1.2. Pyrolysis process

Fast pyrolysis is a thermal decomposition process that requires a high heat transfer rate to the biomass particles and a short vapor residence time in the reaction zone [8], which is a high-temperature process for the production of vapors, aerosols and some coal-like char where biomass is quickly heated in the vacuum and then decomposes. The dark brown mobile fluid (bio-oil) is formed after cooling and condensation of these vapors and aerosols. When organic matter is biomass, consisting of biopolymers (such as cellulose, hemicelluloses, and lignin), the oils produced are called bio-oils. Generally, fast pyrolysis is used to obtain high-grade bio-oil. Fast pyrolysis processes produce 60–75 wt% of liquid bio-oil, 15–25 wt% of solid char, and 10–20 wt% of non-condensable gases. Fast pyrolysis initially starts with slow heating rates, and then involves a rapid heating rate of the biomass, that can reach up to 300°C/min, but not as fast as flash pyrolysis.

Fast pyrolysis design variables include, but are not limited to the following ones reported by [8]: feed moisture content, particle size, pretreatment, reactor configuration, heat supply, heat transfer, heating rates, reaction temperature, vapor residence time, secondary cracking, char separation, ash separation, and liquid collection.

3.1.2. Importance of using bio-oil in the modification of asphalt

The bio-oil obtained from waste biomass at low cost is an environmentally friendly material containing the natural antioxidant lignin, bio-renewable asphalt modifier and asphalt substitutes potential to be successfully applied as an antioxidant additive in asphalt pavements. The other various chemicals that using as antioxidant additives are not environmentally and economically preferable [18].

3.2. Biopolymers

Bio-plastics or organic plastics are a form of plastics derived from renewable biomass sources, such as vegetable oil, corn starch, pea starch, or microbiota. Some of its advantages are often biodegradable, not toxic to produce and alternative to traditional plastics this is included in the definition of biodegradable polymers mentioned in [19]. Biopolymers are an alternative to petroleum based polymers produced by living organisms. The field of biopolymers is still in its early stage but is growing in popularity every day. Biodegradable polymers are produced by using micro-organisms, plants and animals (biological systems), or biological starting materials, which have been synthesized chemically from (e.g. starch, sugars, oils or natural fats, etc.). The biopolymers are biodegradable most of them and water-soluble some of them. Most of the biopolymer are compostable or will decompose in landfills, but the time can vary from a few days to even years, and will eventually decompose [20]. The natural rubber is the best example classified as a biopolymer. The natural rubber is the most common elastomer in almost all human activities due to its unique characteristics, raw or combined with synthetic elastomers. This elastomer has the ability to improve pavements performance and durability for being applied in road pavements as a recycled material from tires [21].

3.2.1. Importance of using the biopolymers

The bitumen degree controls the performance of the pavement mixture during the service temperature. In many cases, bitumen properties need to change to enhance their flexible characteristics at low temperatures to withstand adequate cracking and increase shear resistance through high-temperatures and continuous loads to resist corrosion. With the addition of SBS polymers, the physical properties of polymers are usually modified to produce an improved asphalt grade that improves the performance of hot mix asphalt. In 2008, there was a shortage of type spherical polymers for the asphalt industry forcing asphalt mixing producers and owner/agencies to search for different products that could be used as bitumen [21]. With expectations of increased demand from soft asphalt in the next time, the need for viable asphalt is cost-effective that can be used instead of typical spike-type rates will still strong. Of asphalt-modified polymer mixture, nearly 80% of the polymer modified asphalt uses the SB type polymers. Thus there is a great market opportunity to create new polymers that can complement and/or replace the type SBS polymers used in asphalt paving. The researchers of chemistry and engineering are currently working to obtain chemicals and polymers from

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renewable raw materials, composite materials and manufactured products known as "biological materials." When deliberating biomaterials, it is noteworthy that there are no direct negative effects caused by plants on the ecosystem, they can recycle carbon dioxide for the Earth's crust, grow in different climatic zones, and much work on soil fertility improvement [22].

3.2.2. Bio-elastomers

Elastomers materials, or rubber materials, have a cross-linked structure. Natural and synthetic rubbers are both common examples of Elastomers [23]. Elastomer is a polymer with viscoelasticity property in general, especially at low Yung's modulus, and high yield strain compared to other materials. The plastics are very flexible and flexible, which means they can undergo large elastic deformities without rupture and greatly restore the shape and size after removal has been removed. Plastics are usually resistant to oil and fuel, not permeable to liquids and gases, but tend to deteriorate by oxidation [23]. Furthermore, plastics are either thermoplastic (can be thawed) or thermoplastic (which cannot be melted). The use of plant-derived triglycerides is a substance used in the production of plastics because it provides two interactive sites, the double bond in the series of unsaturated fatty acids, and the ester group [24].

3.2.3. Using of bio-elastomers in asphalt modification

The introduction of crumb rubber in the production of asphalt-rubber mixtures for road pavement should be considered a sustainable technology that turns unwanted residues into a new mixture with high resistance to fatigue and breakage. According to ASTM D 6814–02, rubber is a synthetic or natural synthetic rubber that can be chemically cross-linked/vulcanized to enhance its useful properties. Rubber or elastic joints across the link are three-dimensional molecular networks, with long molecules held together by chemical bonds. It absorbs solvents and swells but does not melt. Moreover, it cannot be reprocessed simply by heating [25–27].

3.3. Waste cooking oil

The amount of waste cooking oil collected every year from food shops and restaurants is found to be 3 billion gallons; this is according to the U.S. Environmental Protection Agency (2011) [28]. This can be treated by polymerization and used as an alternative to bitumen [29].

The researchers investigated the potential use of cooking oil waste as an approved bitumen rejuvenation agent [30, 31]. The result was very promising because of the successful application of waste cooking oil with bitumen as an activated agent for used bitumen or older leads to an economical and environmentally friendly solution. More modification and research is needed to obtain more efficient and effective results.

3.3.1. Problems caused by waste cooking oil and its treatment methods

In general, the dumping of untreated waste into a land fill or river leads to a negative environmental impact. One of the main environmental issues arises the enrichment process that occurs when there is an obstacle to sunlight to penetrate the surface of the river caused by the blocking of a thin layer of oil. Ultimately, oxygen supply to aquatic life is disturbed when nutrient enrichment occurs in the river [32, 33]. The balance of the water ecosystem balance in the lake or river has also affected water quality. Engine oil from vehicles or cooking oil waste from residential areas contributes to the main source of river pollution. The responsibility to overcome high construction costs and reduce waste disposal issues has begun to practice the recycling of waste as an alternative and an alternative way to prevent these problems [32]. According to Hamad et al. [33] and Singhbando and Tezuka [34], only the designated licensed companies responsible for collecting cooking waste World Customs Organization (WCO) from the food industry and especially the fast food restaurant before being sent to the recycling centers.

3.3.2. Production of waste cooking oil

Waste cooking oil originates from frying activity at high-temperatures during food preparation, usually in the food industry, restaurants, hotels, and residences. The application of global customs organizations is diverse, for example, the use of yellow grease [35, 36], potential as a fuel source in biodiesel production [37], animal food [38], and making soap production [37]. Despite efforts to collect up to 15 million tons of WCO annually, only a small amount of cooking waste is properly managed through the recycling process [38]. Regular monitoring and management of the WCO creates a challenge to be addressed and becomes the primary consideration for overcoming serious dumping problems that eventually lead to serious water pollution [39]. **Figure 1** shows the total production of the WCO by States. The production of 10 million tons of WCO per year, accounting for 55%, makes the United States the highest producer of the WCO.

A large amount of vegetable oil consumption about 17 million tons causes a huge amount of waste that has reduced the production of oil resources, and this amount increases about 2%

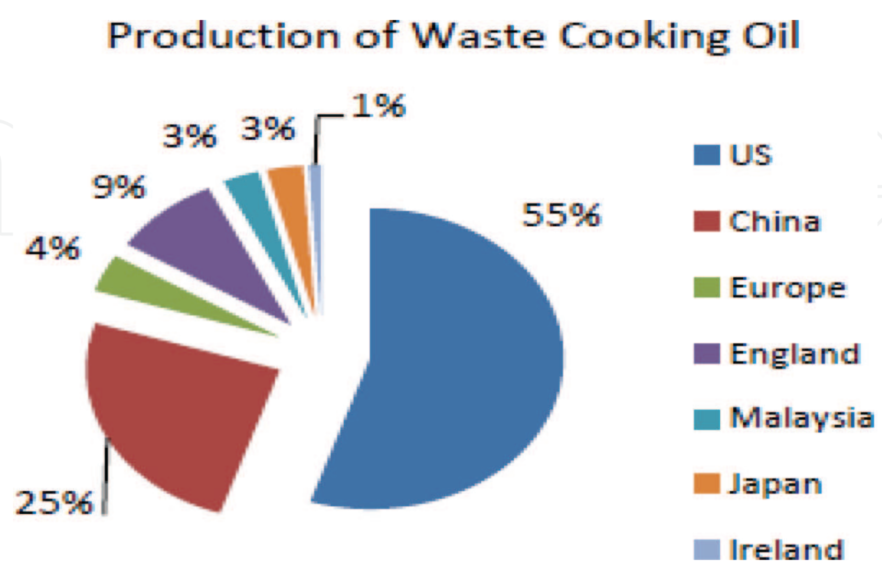

Figure 1. Production of cooking oil based on the country [36, 40, 41].

every passing year [42]. According to the food and drug administration (FDA) report [42, 43], 40% of the sewage system is blocked with frying oil which is cast in the kitchen sink.

3.3.3. Chemical properties and characterization of waste cooking oil

Chemical properties gas chromatography–mass spectrometry test (GCMS) is one of the chemical tests that are used for the purpose of the identification of chemical compound in unknown waste cooking oil. Based on the observation, the major chemical compound identified in the WCO is oleic acid, which represents 43.67% of the entire compound. Meanwhile, palmitic acid represents for 38.35% whereas 11.39% is recorded linoleic acid as shown in **Table 2**.

Due to long chain of palmitic acid and oleic acid [44], the potential of being cracked is high by thermal cracking or catalytic cracking process. Based on the research conducted by Zahoor et al. [45], the properties of unused oil is differ from used cooking oil especially in term of density, kinematic viscosity, and moisture content which presented in **Table 3**.

3.3.4. Experimental application of bio-binders

The main field of application of bio-binders is in the paving industry. However, market opportunities exist in housing products via roofing shingles and sealants [46]. There are three ways that a bio-binder reduces the use of bitumen from fossil fuels (as shown in the percentage) [8].

1. Directly alternative (75–100% bitumen substitute.
2. Bitumen extender (10–75% bitumen substitute)
3. Bitumen modifier (<10% bitumen substitute)

Free fatty acid's type	Waste cooking oil %
Heneicosanoic acid	0.08
Cis-11-Eicosenoic acid	0.16
Linolenic acid	0.29
Palmitic acid	38.35
Linoleic acid	11.39
Stearic acid	4.33
Myristic acid	1.03
ɣ-Linolenic acid	0.37
Lauric acid	0.34
Oleic acid	43.67
Total	100

Table 2. Chemical properties of waste cooking oil [44].

Properties	Used cooking oil values	Unused cooking oil values
Acid value (mg KOH/gm)	4.03	0.3
Calorific value (J/gm)	39,658	—
Saponification value (mg KOH/gm)	177.97	194
Peroxide value (meq/kg)	10	< 10
Density (gm/cm^3)	0.9013	0.898
Kinematic viscosity (mm^2/s)	44.956	39.994
Dynamic viscosity (mpa.s)	40.519	35.920
Flash point (°C)	222–224	161–164
Moisture content (wt. %)	0.140	0.101

Table 3. Comparison between properties of unused oil and used cooking oil.

In almost, all the road construction has been used modifiers in conventional bitumen binder. To decrease the demand for petroleum-based bitumen research is being done on bitumen extender as 100% replacement of bitumen. Additives are the resin, emulsions, crumb rubber, polymer and so on. Waste materials like waste cooking oil, waste engine oil and so on, may be the promising alternatives [47, 48].

4. Overview of the results of using of bio-modified asphalt

4.1. Predictable overall specifications

Asphalt is a mixture of moderately molecular weight hydrocarbons, aliphatic, and aromatic hydrocarbons containing moderate amounts of sulfur, small amounts of oxygen and nitrogen, and other rare elements including transitional metals. The physical and chemical properties of asphalt are results of their chemical composition. Large changes in chemical composition may lead to changes in unexpected physical properties. Bio-binders are at best linked to performance, so important chemical changes may give a false test. There are many noticeable, tacit assumptions about asphalt to consider:

- Expected aging characteristics.
- It is expected to have rheological properties.
- Will have the characteristics of the expected adhesion to aggregates.
- Has the expected coating behavior in the mixing plant.
- Have predictable flow characteristics throughout construction.

There are many other less obvious predictions:

- Predictable outflow characteristics.
- Expectable water solubility.
- Expected interactions with fuels, oils, and so on.
- Predictable environmental characteristics.
- It will have a predictable smell.
- It will have a predictable mixing with a virgin binder as a reclaimed asphalt pavement
- It will have a predictable interaction with contiguous mixtures.
- Will be available in large quantities upon request.

4.2. Aging

Rolling thin film oven test (RTFO) and pressure aging vessel (PAV) may not sufficiently represent plant and field aging because an alternative binder may have significantly different aging characteristics. Start by identifying an aging index to compare with unmodified asphalt is suggested in experimental respects. RTFO It should be operated over a variety of times and temperatures to see if normal temperature correspondence is still present. The same is true for PAV. Use PAV at 60°C for excessive times and compare output performance with results in standard conditions.

4.3. Rheological properties of bio-modified asphalt

Bio-binder may it may completely break down or give suggestively different time-temperature relationships. A clear example is an alternative bond with a fine melting point. A binder with a melting point at 73°C may be well suited for use in heavy road traffic in the PG 64 environment, but the dynamic shear rheometer (DSR) test at a specific grade of 76°C will give inappropriate results.

4.4. Strength properties and cracking

While strength characteristics and cracking are precarious to pavement performance, they are controlled by the total and the gradient and the effect of the alternative binder properties is not easily expected. Unlike low-temperature characteristics, it may be necessary to reduce the mixture test to explain the effect of a new material on cracking and strength properties.

4.5. Rutting performance at high-temperature

As illustrious above, bio-binder may have a different shape to the master curve. They may also have varied stress tolerance. For more than a few reasons, the creep compliance is unrecoverable, Jnr, from the multiple creep and recovery creep (MSCR), is a better choice than the G */sin δ of the oscillating DSR to describe the performance of high-temperatures. MSCR is run at the expected high pavement temperature and therefore, does not depend on the time-overlay overlap. Jnr has been shown to be well correlated with the actual progressive performance of a larger group of substances from G */sin δ. Finally, stress tolerance is already a unit for MSCR testing. However, it would be practical to check the range of temperatures and pressures to look for unusual behavior that may affect progressive performance. The mixture test is also strongly recommended.

4.6. Cracking in cold climates

Cold climates tensile and crack the characteristics of a bio-binder may differ from conventional asphalt, so testing the bending beam rheometer (BBR) may not be sufficient. A fracture test such as a direct tensile test or asphalt cracking will provide information on low-temperature break characteristics. If there is a significant difference in expected temperatures, the fracture test results can still be used to re-evaluate the BBR results to simplify the test over the long term. One more caution, these still only characterize one single thermal cracker. May not adequately address fatigue at low-temperatures.

4.7. Fatigue performance

There is currently a no good way to characterize a binder for fatigue performance, although there are some talented tests in development. The G * sin δ from DSR has been included in the superpave specification to control the shape of the main curve, and as mentioned above, it may or may not be suitable for a substance different from asphalt. There are differences in mixing tests that are related to fatigue performance, and at least today, this is the only correct position to handle fatigue performance.

5. Comparison between bio-modified asphalt binders and unmodified ones

The temperature range of viscous behavior for bio-oils may be lower than the virgin asphalt rate, by about 30–40°C. The rheological properties of the original bio-binder differ from those of the asphalt, but the rheological properties of these modified biological bonds vary greatly when polymer rates are added. For the developed bio-binders the high-temperature performance grade may not vary significantly from that of the asphalt binders, nevertheless, the low-temperature performance grade may vary significantly [6].

5.1. Structure of bio-oil compared with asphalt

The chemical properties indicated that the amount of furfural and phenols were varying due to the different aging processes and intervals.

The chemistry of bio-oils is complex, similar to asphalt; thus, a complete chemical characterization is difficult or practically impossible. The complication of chemical characterization or analysis resulted from the attendance of high molecular weight of phenolic species [4]. In addition, the fragmented oligomeric products occur with different numbers of phenolic and carboxylic acids, and hydroxyl groups as well as aldehyde, alcohol, and ether purposes. Thus, phenolic species occur as different hydrogen-bonded aggregates, micelles, droplets, and gels.

5.2. Temperature of performance

In general, the mixing and compaction the temperature range for bio-oils may be lower than bitumen inhibitors at about 30–40°C.

- The flow characteristics, that is, the temperature and shear, of the biomedical biomaterials of the bio-oils differ from those of the asphalt, but when adding the polymer hosts, the rheological properties of these modified biomarkers change dramatically.
- Polymers should be carefully chosen because the temperature range of the bio-groups established differs from the polymers used extensively in the asphalt industry.
- The high-temperature performance of biologically determined groups may not change significantly from sediment deposits. However, the low-temperature performance level may change significantly because of the high oxygen content in biomaterials compared to typical asphalt volumes.

5.3. Comparison from viewpoints of environment, economy, and energy

Significantly, The United States is working to create a biobased economy that generates energy from renewable organic matter rather than fossil fuels. Because of access to large amounts of vital sources such as triglycerides, proteins, starch, and other carbohydrates from various plant sources, there are interesting technical and economic forecasts for their use to produce vital bonding materials. At present, research on the application of bio-oils has focused on their use as bio-defense fuel to replace fossil fuels. Based on the findings of these surveys, the use of bio-oils as asphalt metal is very promising.

On the other hand, no research has yet been conducted on the feasibility of using bio-oils as an alternative to asphalt (replacing 100%) for use in the paving industry. As a result, there is a lack of data demonstrating the development of biomaterials from essential oils. Biomass boxes (artificial bonding materials) can be used in three different ways to reduce the demand for fossil-based asphalt compounds.

6. Conclusion

High construction costs, when combined with awareness regarding environmental stewardship have encouraged the use of waste and renewable resources in asphalt modification.

Increased energy costs and strong global demand for oil have encouraged the development of alternative bonding materials to modify or replace asphalt bonding materials. The benefits of using alternative bonding materials are their ability to help conserve natural resources and reduce energy consumption while at the same time improving asphalt performance.

The use of alternative (or secondary) materials in asphalt mixtures may be one of the most complex methods of highway use. It is not a matter of throwing alternative materials into the mixture and coating them with cement. The best use should be designed and used, including the design of the mixture itself, and the effects of alternative materials on the behavior of the asphalt binder, and the pavement to be incorporated into it. It is necessary to know how to test the resulting mixture in order to conform to the specifications; in some cases, this knowledge is lacking. Also, the expected finding from this chapter is as follows:

- Bio-Binder modified asphalt is capable to change rheological properties of the asphalt binder which will improve the performance against pavement distress
- The bio-binder will increase viscosity of the asphalt binder at high service temperature
- The binder test will show that the addition of bio-oils is expected to improve the rutting performance
- Most of the bio-oil modified asphalt mixture has higher fatigue lives than the control asphalt mixture.
- The addition of bio-modified asphalt with asphalt binder reduced the PG of the base binder, representing an increased resistance to thermal cracking, nevertheless reduced resistance to rutting.
- The adding of bio-modified asphalt reduced the crack energy at middle temperatures when compared to unoriginal asphalt under monotonic loading, representing reduced resistance to fatigue cracking.
- The dynamic modulus of hot mix asphalt (HMA) reduced with addition of bio-modified asphalt.
- The flow numbers of the bio-modified asphalt HMA mixes were lower than those of unoriginal mixes. This finding indicated that bio-modified asphalt HMA mixes are more disposed to rutting than unoriginal mixes.
- The cracking energy reduced with addition of bio-modified asphalt in the HMA and crack work obtained from IDT testing at intermediate temperatures, indicating that the bio-modified asphalt mixes are disposed to fatigue failure as soon as compared to control HMA.
- Thermal cracking energy increased with addition of bio-modified asphalt in the HMA, which indicating that an improved resistance to low-temperature thermal cracking. Bio-modified asphalt mixes are highly elastic even at lower temperatures than unoriginal mixes.
- Overall bio-modified asphalt can be a promising applicant for low-temperature behavior. However, further modification of bio-modified asphalt for higher temperature is essential to increase its performance against rutting.

It will be a positive step in the direction of achieving mixture modified with bio-binder that has similar or improve performance when compare to conventional mixtures.

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