Research Article

Physical, Thermal, and Morphology Characteristics of Waste Latex Rubber Glove-Modified Bitumen

Auni Diyana Fadzil,[1] Nur Izzi Md Yusoff [iD],[1] Shuhaida Harun,[2] Khairiah Haji Badri,[3] Iswandaru Widyatmoko [iD],[4] Faridah Hanim Khairuddin,[5] Abdalrhman Milad [iD],[6] Munder Bilema [iD],[7] and Naeem Aziz Memon[8]

[1]*Department of Civil Engineering, Faculty of Engineering and Built Environment, Universiti Kebangsaan Malaysia, Bangi 43600, Selangor, Malaysia*
[2]*Department of Chemical and Process Engineering, Faculty of Engineering and Built Environment, Universiti Kebangsaan Malaysia, Bangi 43600, Selangor, Malaysia*
[3]*Department of Chemical Sciences, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, UKM, Bangi 43600, Selangor, Malaysia*
[4]*Infrastructure and Transportation Materials R&D, Centre of Excellent for Asset Consultancy, AECOM, NGM, England, UK*
[5]*Department of Civil Engineering, Universiti Pertahanan Nasional Malaysia, Kuala Lumpur 57000, Malaysia*
[6]*College of Engineering and Architecture, University of Nizwa, P.O. 33 Postal Code, Nizwa 616, Oman*
[7]*Centre for Transportation Research, Universiti Malaya, Kuala Lumpur 50603, Malaysia*
[8]*Department of Civil Engineering, Mehran University of Engineering and Technology, Indus Highway, Jamshoro 76062, Sindh, Pakistan*

Correspondence should be addressed to Nur Izzi Md Yusoff; izzi@ukm.edu.my

Researchers across the globe have explored several alternatives to recycling natural rubber and have identified several challenges. Therefore, this study evaluates the feasibility of recycling waste latex rubber gloves (WLRG) as a bitumen modifier to enhance the bitumen's physical, thermal, and morphological characteristics. The study adds varying percentages of WLRG (3%, 5%, 7%, and 9%) to the 60/70 bitumen and analyzes them to determine the optimum WLRG percentage. The penetration, softening point, ductility, and viscosity tests of the modified binders show a consistent pattern. All WLRG-modified bitumens are stable storage blends at high temperatures. The thermal characteristics of the WLRG particles in the modified bitumen are examined through thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC). The ogive graph shows that the weight loss of the bitumen modified with 3%, 5%, 7%, and 9% WLRG occurred at 457.5, 464.3, 462.2, and 459.5°C. The maximum weight loss of the control sample occurred at 465.6°C when the environment switched from nitrogen (N_2) to air. The DSC graph reveals the changes in the structure or physiochemical processes of the WLRG. The melting point for the binders modified with 3%, 5%, 7%, and 9% WLRG is 133.6, 132.1, 103.5, and 133.2°C. The morphological characteristics were determined using atomic force microscopy (AFM). The bee structure gives a scientific explanation of the microstructural characteristics. A contact angle test was performed to identify the wettability of the sessile drop device by using three types of solvent, namely distilled water, formamide, and glycerol. The contact angle of water showed a decreasing trend, where the binder containing 9% WLRG had the lowest contact angle. For the control sample, the contact angles of formamide and glycerol are 73.95° and 71.85°, respectively. In summary, WLRG is a suitable bitumen modifier and can enhance the physical, thermal, and morphological characteristics of the asphalt binder.

1. Introduction

Waste rubber products, particularly gloves and tires, are commonly disposed of in landfills [1]. However, landfill disposal of waste rubber becomes impractical with the increasingly limited landfill sites. Disposing waste rubber at landfill is also the best unwelcome due to environmental difficulties for dispose of it. There is an urgent need to recycle as much natural rubber (NR) to preserve the environment. One way to minimize the problems created by illegal disposals is by

recycling rubber gloves. Dumping rubber gloves in landfills can cause the release of small-molecular-weight additives, which rise to the surface and enter the atmosphere. These small-molecular-weight additives are nonbiodegradable and kill some of the good soil bacteria. Disposal of unsorted wastes in landfills is harmful to the environment and costly because of the hidden costs of transporting the waste to landfill sites and preparing and maintaining the landfill sites.

Pavement deterioration, including cracking, rutting, and bleeding, results from increasing traffic volume and weather effects. Asphalt behavior is closely related to temperature and loading time [2]. The tropical climate in Malaysia means that asphalts are softer at average temperatures and flow like a viscous liquid at high temperatures [3–5]. In this condition, the low-temperature properties of asphalt can change into a rigid solid, causing instability and weakness crack disaster. Previous studies reported that conventional asphalt pavements sustained severe damage, especially fatigue failure and moisture damage [6, 7], and there were efforts to introduce alternative techniques in pavement rehabilitation, including hot in-plant recycling (HIP), hot in-place recycling (HIPR), and cold in-place recycling (CIPR) [8–10]. Unfortunately, some damage to the asphalt pavement occurs before the road reaches maturity, and, thus, it is essential to modify bitumen to produce asphalts with excellent performance. Researchers are exploring using renewable natural resources, such as NR latex, which has high elasticity due to its high stretch fraction and flexibility [11–14].

NR is one of the polymers of isoprene (methylbuta-1, 3-diene) that can transform into cis-polyisoprene through the enzyme rubber process called enzymatic polymerization. This enzyme is tied to the rubber particles in the latex serum. NR has a general bimodal molecular weight distribution [15–17] that can produce the end product effectively because of the descending molecular weight. The higher molecular weight fractions increase the tensile strength, abrasion resistance, and tear strength. The isopentyl pyrophosphate starting substance is used to create farnesyl pyrophosphate. The trans-farnesyl pyrophosphate produces a trans-polyisoprene called gutta-percha. Gutta-percha is an isomeric polymer with a trans configuration. This polymer is integrated from the biosynthesis of isopentenyl pyrophosphate [18–21]. NR spontaneously crystallizes at low temperatures or when stretched, thus losing its elasticity value. Table 1 presents the physical and chemical properties of NR latex [22, 23].

Previous studies have demonstrated that the disulfide bond in rubber limits its degree of freedom. The adhesives contracted rapidly, making the rubber more complex and less stretchable with greater elastic force. The characteristics of NR are influenced by modifiers and fillers such as chalk and carbon black. Binder attributes are the primary determiner of road asphalt performance. The crude oil base and its refinement process determine the complex characteristics of asphalts, and, thus, high-quality crude oil and refinement can ensure quality asphalts [24–28].

Bitumen modification can improve asphalt performance. Researchers have explored using various materials, such as NR and carbon black, in bitumen modification. NR is a

TABLE 1: The physical and chemical properties of NR latex.

Parameters	Requirement
Color	Milky white
Total solids content (TSC)	Minimum of 61.5
Dry rubber content (DRC)	Minimum of 60
pH at 25.4°C	Maximum of 11
Potassium hydroxide (KOH) number	Maximum of 0.7
Alkalinity (as ammonia on total weight)	0.65–0.75
Nonrubber content	Maximum of 1.7
Volatile fatty acid (VFC) number	Maximum of 0.05
Mechanical stability time (MST)	1,000–1,200
Specific gravity at 25°C	0.94
Magnesium content	Maximum of 40

fascinating polymer, which can mix with asphalt efficiently [4, 29]. The distinct characteristics of NR-modified bitumen are high stability, high tear strength, fatigue resistance, and extended asphalt pavement durability [30, 31]. The outstanding dynamic properties of NR are low hysteresis loss, superior low-temperature properties, and strong NR-to-metal bond [5]. It is crucial to enhance bitumen quality to extend the service life of road pavements and minimize maintenance costs. However, there is a dearth of research on bitumen modified with waste latex rubber gloves (WLRG) since previous studies focused primarily on utilizing modifiers produced from synthetic polymers and powdered rubber. This study explored the outcomes of using varying percentages of WLRG (3%, 5%, 7%, and 9%) as a bitumen modifier. The results showed that the WLRG is the superior bitumen modifier to enhance bitumen characteristics in regions with tropical climates.

2. Materials and Methods

2.1. Raw Materials. This study added varying WLRG percentages of 3%, 5%, 7%, and 9% binder weight to 60/70 penetration grade bitumen. Table 2 presents the physical characteristics of the 60/70 bitumen. The NR latex was added to the preheated bitumen at 160°C and mixed in a high-shear mixer at 1,100 rpm for 30 min.

2.2. Blending Process. The WLRG-modified bitumen was produced by weighing 1,000 g of the base bitumen and heating it in a 150°C oven for 120 min, after which the base bitumen was removed from the oven and placed inside the cylinder metal by using gloves. Then, the samples were blend using a high-shear mixer machines and kept at a temperature of 150°C. The required amount of WLRG (3%, 5%, 7%, and 9%) was added gradually to the bitumen while stirring the bitumen manually using a glass rod to minimize lump and air bubble formation and ensure optimum blending to obtain a homogeneous blend with minimal air bubbles. The temperature needs to be checked until it reaches 150°C.

2.3. Storage Stability. The storage stability test to determine the stability of the WLRG-modified bitumen was conducted at high temperatures following ASTM D7173. This test was

Properties	Unit	Requirement	Result	Standard test
Penetration at 25°C, 100 g, 5 s, 0.1 mm	dmm	60–70	68.46	ASTM D5
Softening point	°C	46 min	>46	ASTM D36
Ductility	cm	100 min	>100	ASTM D113

conducted to examine the physical characteristics of materials. Since this study requires a 140-mm sample, the WLRG-modified bitumen was heated and put in an aluminum tube with a 25-mm diameter and 200-mm length and then heated at 163°C for 48 hr while ensuring the oven was constantly at the required temperature. After being cooled, the aluminum tube was cut into three pieces using a knife. The softening point difference (SPD) was used to identify the difference between the top and bottom samples [32]. For acceptable results, the dissimilarity must be less than or equal to 2.5°C.

2.4. Physical Test. This study conducted physical tests to determine the samples' penetration, softening point, and ductility. Bitumen with higher penetration values has a softer consistency. ASTM D5 requires pouring the melted bitumen sample into a cylindrical metallic dish with a flat bottom and a 55-mm diameter and 35-mm depth. The test measures the vertical penetration by a penetrometer needle with a 100-g load at 25°C for about 5 s. The penetration test requires a minimum of three determination points on the sample surface at least 10 mm from the side of the container and more than 10 mm apart.

The softening point test was conducted following ASTM D36 by immersing a ring and ball apparatus in distilled water. Two bitumen disks, cast in shouldered brass rings, were heated in a liquid bath at a controlled rate of 5°C/min. Each supporting steel ball has a 9.5-mm diameter and weighs 3.5 g. The softening point is the average temperature on the softened double disk sufficient to allow each ball to pass through the bitumen brass ring. The bottom of the shouldered rings in the ring holder must be 25 mm from the upper surface of the bottom plate. The distance from the top plate and the lower surface to the bottom of the bath is 16 mm. The thermometer must be able to take the temperature for every 1°C.

The ductility tests were conducted following the ASTM D113 to measure the bitumen's adhesive properties and the distance it stretches under any conditions. Bitumen ductility is the distance (in centimeters) it extends before splitting in half. The test was carried out at 25°C and a speed of 5 cm/min. The filled mold was allowed to cool to room temperature for 35 min before removing the bitumen sample from the water bath and immediately cutting off the excess material with a trimming tool.

2.5. Thermal Test

2.5.1. Thermogravimetric Analysis. Thermogravimetric analysis (TGA) identifies the amount of oil or polymer in a rubber compound by measuring the weight change. The TGA in this study followed ASTM D6370. The WLRG-modified samples were heated to a controlled temperature at a predetermined rate and environment, and the weight change was recorded as a function of temperature. The weight loss above the specified temperature range used for TGA is 25–600°C was recorded [33]. A TGA analysis is used to identify the preferred material, quality control, and material screening. The first step in TGA is placing 10–12 mg of the sample (small pieces) on the platinum pan of the calibrated TGA analyzer, followed by applying a 75-cm^3/min or the manufacturer-recommended flow of argon or nitrogen purge. After allowing the instrument to equilibrate, it was heated to 50°C for at least 2 min. The WLRG sample was heated from 50 to 500°C at a rate of 50°C/min, after which the temperature was allowed to equilibrate for 2 min and reduce to 300°C. The purge gas was changed to air or oxygen and purged at 75 cm^3/min. The transition was observed through the energy emission or absorption by the sample, which produced the corresponding endothermic or exothermic peaks in heating or cooling.

2.5.2. Differential Scanning Calorimetry. Differential scanning calorimetry (DSC) is a thermal analysis technique, which measures the heat flow in or out of a sample as a function of time or temperature. The glass transition is a step change in specific heat capacity and pivotal information on the thermal history, processing, stability, chemical reactions, and mechanical behavior of amorphous and semicrystalline materials. A change in the degree of heating and cooling can affect test results. Impurities that plasticize or form solid solutions or are miscible in the posttransition phase affect the glass transition. The samples should be compared to the same particle size if the particle size affects the detected transition temperature. Occasionally, the samples may interact with air during the transition temperature and cause misestimating of the glass transition.

Therefore, it is essential to conduct the test in a vacuum or inert gas atmosphere when this effect is present. It is crucial to differentiate degradation from glass transition because the degradation of some materials occurs close to the glass transition region. Since the samples are in milligram quantities, the specimen must be homogeneous to ensure accurate sampling. Instrument selection and uniform controlled sample heating (or cooling) are essential to achieve minimum DSC capability. Although ASTM E1356 specifies a temperature range of −120 to 500°C, the temperature range in this study was −50 to 350°C.

A temperature sensor ensures that the specimen temperature is 60.1°C. The variance sensor detects the difference in the heat flow between the specimen and the reference with a reactivity of 6 μW. The atmosphere of the purge gas test chamber is between 10 and 100 mL/min at a rate of 4 mL/min. A temperature sensor can be determined by operating the

furnace at a temperature rate from 20 to 60.5°C/min. All data are stored for later use. The minimum output signals required for a DSC are time, temperature, and heat flow.

2.6. Morphology Test

2.6.1. Atomic Force Microscopy. Atomic force microscopy (AFM) produces images of polymers, ceramics, composites, glass, and biological samples. This study used AFM to measure the WLRG's adhesion strength, magnetic force, and mechanical properties. AFM is an advanced technology widely used by nanotechnology researchers; it is an indispensable tool for visualizing and measuring structures at the nanoscale level following the ASTM E2859. The WLRG-modified bitumen was placed on a flat surface for AFM measurements, and the surface roughness was much smaller than the nominal nanoparticle size, ideally less than 5%, to ensure a uniform baseline for measuring height. Single-crystal silicon can be used as the substrate to minimize the surface roughness effects on the nanoparticle.

The process for depositing the sample was developed using negatively charged citrate-stabilized gold nanoparticles suspended in an aqueous solution at a mass concentration of $50\,\mu g/g$. The deposition process works with other nanoparticles with a negative surface charge or zeta potential but is not limited to commercially available citrate-stabilized colloidal gold. This process can be modified for positively or neutral-charged nanoparticles. Each approach may require optimization to ensure adequate deposition density and minimize artifacts, such as agglomerate formation on the substrate or organic film formation resulting from the additives present in the solution phase.

2.7. Contact Angle. Contact angle test is a measurement to identify the wettability of the binder surface by using different types of solvent such as distilled water, formamide, and glycerol. This study tested the WLRG samples at 23°C and a ≥50% relative humidity. The goniometer was arranged as instructed on a level stage. Use small weights or clips to hold it close to the stage and place the sample on the instrument. The contact angle drop was made 3 mm from the surface using the tip of a hypodermic needle. The recommended drop size is $5\,\mu L$ for manual equipment and $20\,\mu L$ for automated equipment. The drop size should be $\pm0.1\,\mu L$ to obtain the best results. The contact angle measured at this droplet was the advancing contact angle. The goniometer eyepiece and internal measuring mechanism were adjusted to ensure an accurate internal angle measurement for each contact point of the droplet. For every specimen, an average of three drops is taken. The test should be repeated if the contact angle values are significantly different. This study measured six contact angles. The water contact angle was measured quickly (within 30 s after depositing the droplet) to avoid a change in the contact angle as the water evaporated. According to Young's equation [34], the contact angle is determined by balancing the surface tension, as shown in Equation (1). Figure 1 shows the diagram of a drop of liquid on a solid surface under three interfacial tensions such as
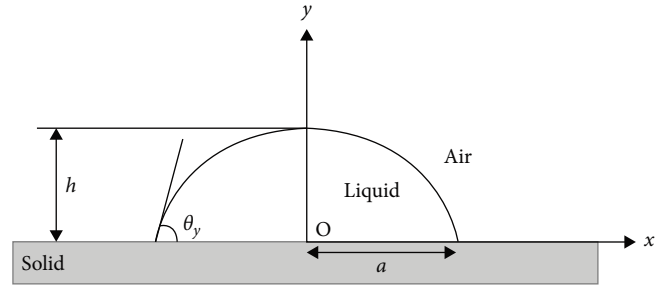

FIGURE 1: The diagram of a drop of liquid on a solid surface, with the radius (a), height (h), and the Young's contact angle (θ_y).

TABLE 3: Storage stability.

Sample	SPD temperature (°C)	Specification <2.5°C
Control sample	0.77	Pass
3% WLRG	1.80	Pass
5% WLRG	2.43	Pass
7% WLRG	2.37	Pass
9% WLRG	2.17	Pass

solid–air surface, solid–liquid interfacial, and liquid–air surface tension.

$$\gamma_{SV} = \gamma_{SL} + \gamma \cos\theta_y \qquad (1)$$

3. Results and Discussion

3.1. Storage Stability. This study evaluated the additive's storage stability to ensure that the modification did not adversely affect its effectiveness. Table 3 presents the difference in the SPD temperature of the top and bottom surfaces. The temperature difference between the bottom and top surfaces of the WLRG-modified bitumen should be less than 2.5°C [17]. The temperature difference after treating the samples for 72 hr at 163°C was between 1.8 and 2.43°C, indicating that the WLRG in the modified binder sample was stable under intense heat storage. The lowest temperature difference of 0.77°C for the control sample means that the temperature difference was more considerable with higher WLRG contents, indicating that the WLRG-modified bitumen did not separate or underwent phase modification during storage at high temperatures.

3.2. Physical Analysis. Figures 2 and 3 present the results of the penetration and softening point tests, and Table 4 presents the results of the ductility test. The penetration test was conducted to evaluate the consistency of the base bituminous binder and WLRG-modified bitumen. The softening point is the melting point where the material gradually softens and becomes less viscous with higher temperatures. The ductility test measures a material's ductility. The penetration values of the samples containing 3%, 5%, 7%, and 9% WLRG
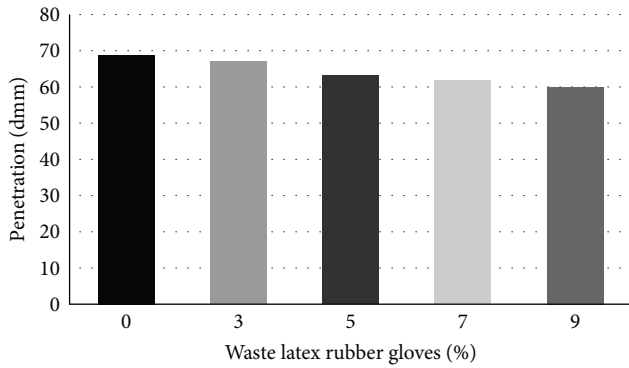
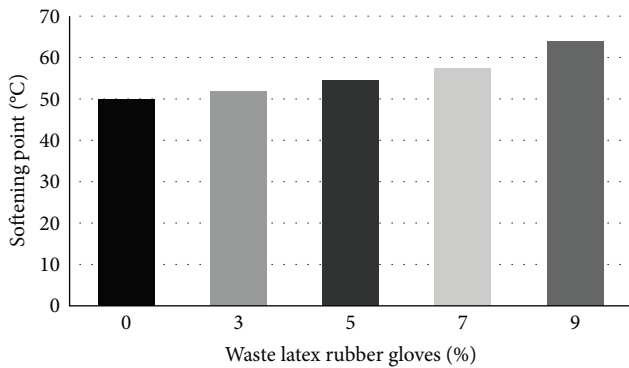FIGURE 2: The penetration value of the WLRG-modified bitumen.

FIGURE 3: The softening point of the WLRG-modified bitumen.

TABLE 4: The ductility of the WLRG-modified bitumen samples.

Experimental	0%	3%	5%	7%	9%
Ductility at 25°C	>150	134.5	127.5	116.0	90.5

decreased with higher WLRG percentages at a mixing rate of 1,100 rev/min.

The histogram graph, as shown in Figure 1, presents the frequency distribution of the data points, where the average penetration value decreased by about 2.14% with every 2% increase of the WLRG added to the bitumen. The higher penetration values indicate softer texture consistency. The reduced penetration also means higher bituminous binder hardness [35, 36]. Relative to the base bituminous binder, the sample containing 3% and 9% WLRG had an average penetration of 66.92% and 59.92%, respectively. The difference in the penetration values showed that the higher percentages of WLRG have lower penetration, which means it is harder than bituminous binder and suitable to be used in hot climates and it shows more durability exposing to heavy traffic loads.

Figure 3 shows that the 9% WLRG-modified bitumen sample had a maximum softening point of 63.6°C, and the 3% WLRG-modified bitumen had the lowest softening point of 51.7%. The softening point of the bitumen modified with 3% WLRG at a high shear mixing rate of 1,100 rev/min was similar to the softening point of the base binder. In comparison, 7% and 9% of WLRG-modified bitumen had higher
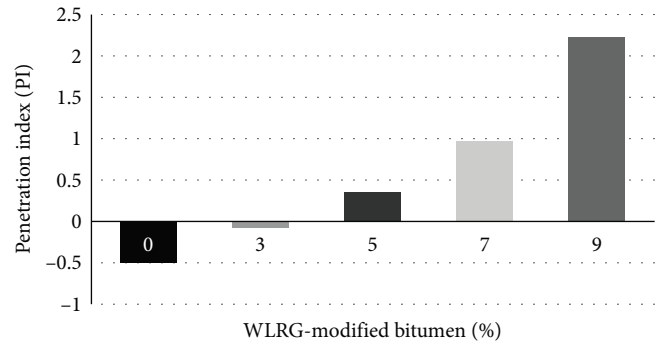

FIGURE 4: The penetration index of the WLRG-modified bitumen samples.

values than the control sample. The resulting soft areas were due to the elastic properties of the WLRG and its concentration. The higher stiffness of the WLRG-modified bitumen can be attributed to the stiffening effect of adding WLRG to the control sample. Therefore, this study has shown that WLRG-modified bitumen has a more significant impact than the control sample. The penetration index (PI) was calculated using the penetration test result at 25°C and the softening point test. Figure 4 shows the PI values of the WLRG-modified bitumen samples. The PI standards range from −3 to +7 for highly blown, low-temperature susceptible bitumen [37]. The PI for all modified bitumens was within this acceptable range and consistent with the increased stiffness of the modified bitumen, where the stiffness increased with lower softening points. The higher stiffness of the modified bitumen reduced its temperature susceptibility.

The ductility test was conducted following the ASTM D113 at 25°C for varying WLRG contents. Table 4 shows a marked decrease in ductility values from 150 to 90.5. The ductility test measures the length a standard specimen elongates in centimeters before breaking [35, 38, 39]. The average decrease in the ductility of the WLRG-modified bitumens and the control sample was 14.67% and 46%. However, the bitumen modified with 9% of WLRG showed the smallest decrease in ductility. The ductility of the control sample exceeded 150 cm and remained unchanged. The bitumen modified with 3%, 5%, and 7% WLRG had ductility values exceeding 100 cm. The ductility of the bitumen modified with 9% of WLRG was lower than the minimum standard required by the ASTM D113, and 3%, 5%, and 7% may perform better than 9% of WLRG-modified bitumen. ASTM D113 allows WLRG-modified bitumens to have a ductility value higher than 100 cm [40]. Furthermore, a low ductility indicates that the modified bitumen is more complex after modification and is prone to become brittle.

3.3. Thermal Analysis

3.3.1. Thermogravimetric Analysis. TGA ensures uniform heating or cooling of the WLRG-modified bitumen between 30 and 500°C. TGA is an analytical method for determining the sample's physicochemical characteristics, thermal stability, and volatile components fraction [33, 41, 42]. Figure 5(a)–5(e)
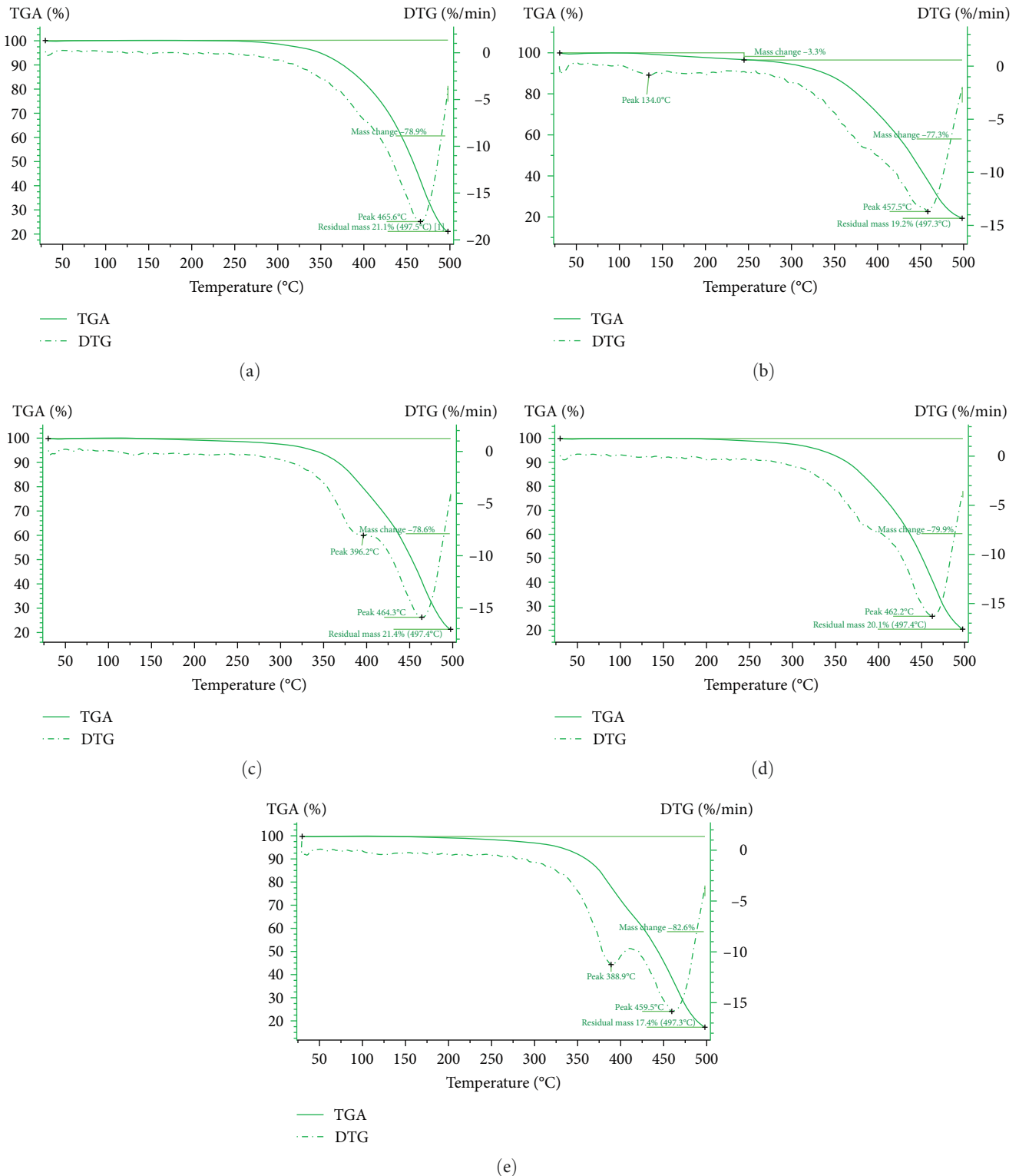
(a)

(b)

(c)

(d)

(e)

FIGURE 5: (a) The TGA and DTG of the control sample. (b) The TGA and DTG of the sample with 3% WLRG. (c) The TGA and DTG of the sample with 5% WLRG. (d) The TGA and DTG of the sample with 7% WLRG. (e) The TGA and DTG curves of the sample with 9% WLRG.

shows that the temperature of the WLRG samples was +1°C. The temperature controller executed a specific temperature program and operated the furnace within the set temperature range with a heating rate of 10–100°C/min and the required time of at least 100 min.

The graph was plotted based on the weight change while heating the samples. The TGA was interpreted to determine polymer degradation during heating between 30 and 500°C and 50°C/min heating rate. Figure 5(a)–5(e) shows the TGA and DTG for the samples containing 0%, 3%, 5%, 7%, and 9%
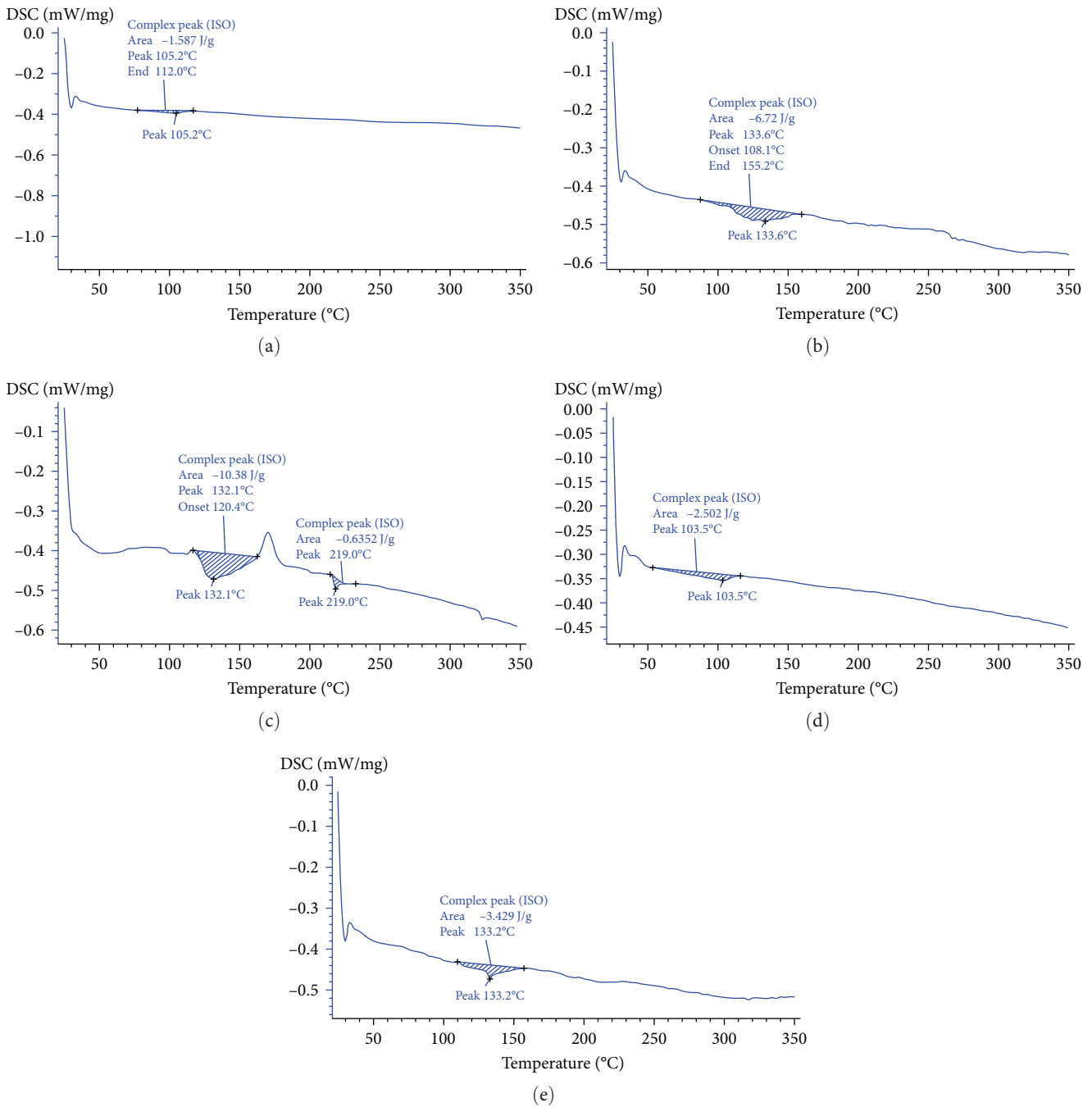
FIGURE 6: (a) The DSC of the base bitumen. (b) The DSC of the sample with 3% WLRG. (c) The DSC of the sample with 5% WLRG. (d) The DSC of the sample with 7% WLRG. (e) The DSC of the sample with 9% WLRG.

WLRG. The graph shows the heat transformation of the WLRG, where the bitumen modified with 3% and 5% WLRG lost weight in two stages, while those modified with 7% and 9% WLRG lost weight in four stages. Figure 4(a) shows the TGA and DTG curves for the control bituminous binder sample. The weight loss for the bitumen modified with 3%, 5%, 7%, and 9% of WLRG occurred at 457.5, 464.3, 462.2, and 459°C, where the bitumen modified with 3% WLRG showed the lowest weight loss. The average weight loss for 3% and 9% of WLRG-modified bitumen compared to base bituminous binder was about 1.74% and

1.31%. The highest weight loss for the bituminous binder occurred at 465.6°C when the environment switched from nitrogen (N_2) to air reacting with carbon black.

3.3.2. Differential Scanning Calorimetry. The DSC graph shows the structural changes (or physiochemical processes), such as the glass transition (T_g), crystallization (T_c), melting point (T_m), and oxidation phase (T_{ox}). DSC is a thermal analysis technique for determining the direct absorption of thermal energy that describes the physical change from liquid to solid [42]. Figure 6(a)–6(e) shows the DSC for the
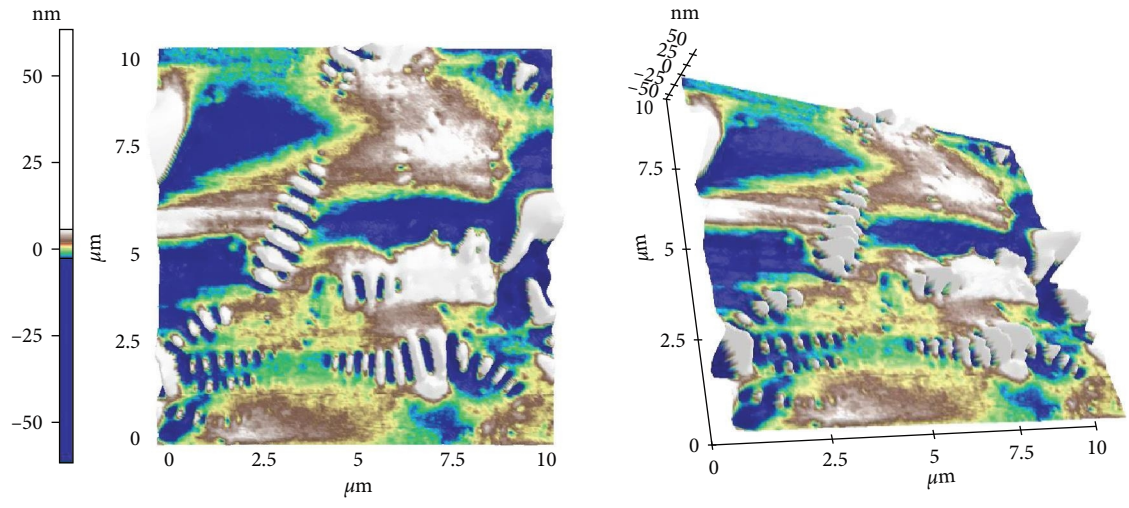
(a)
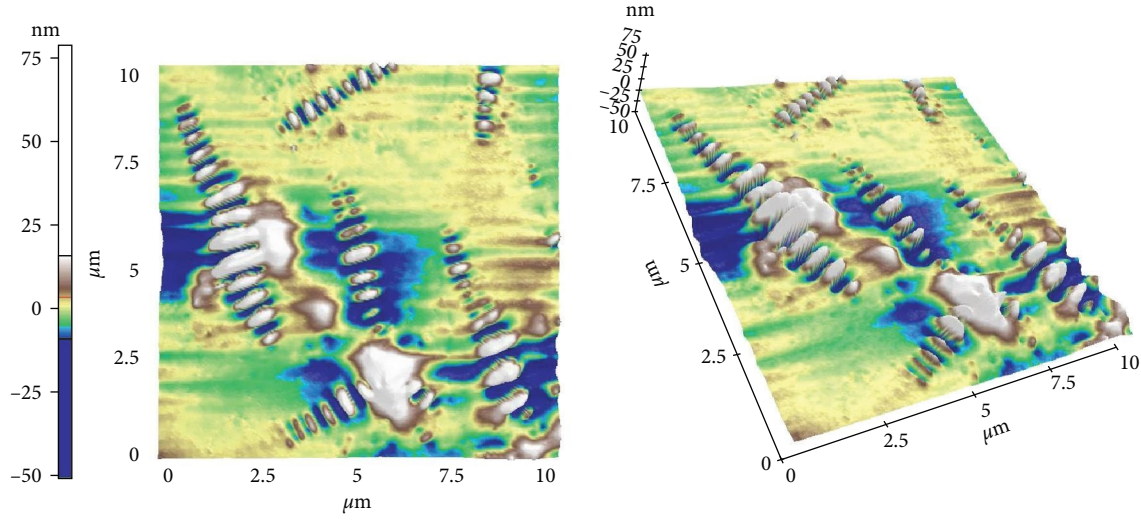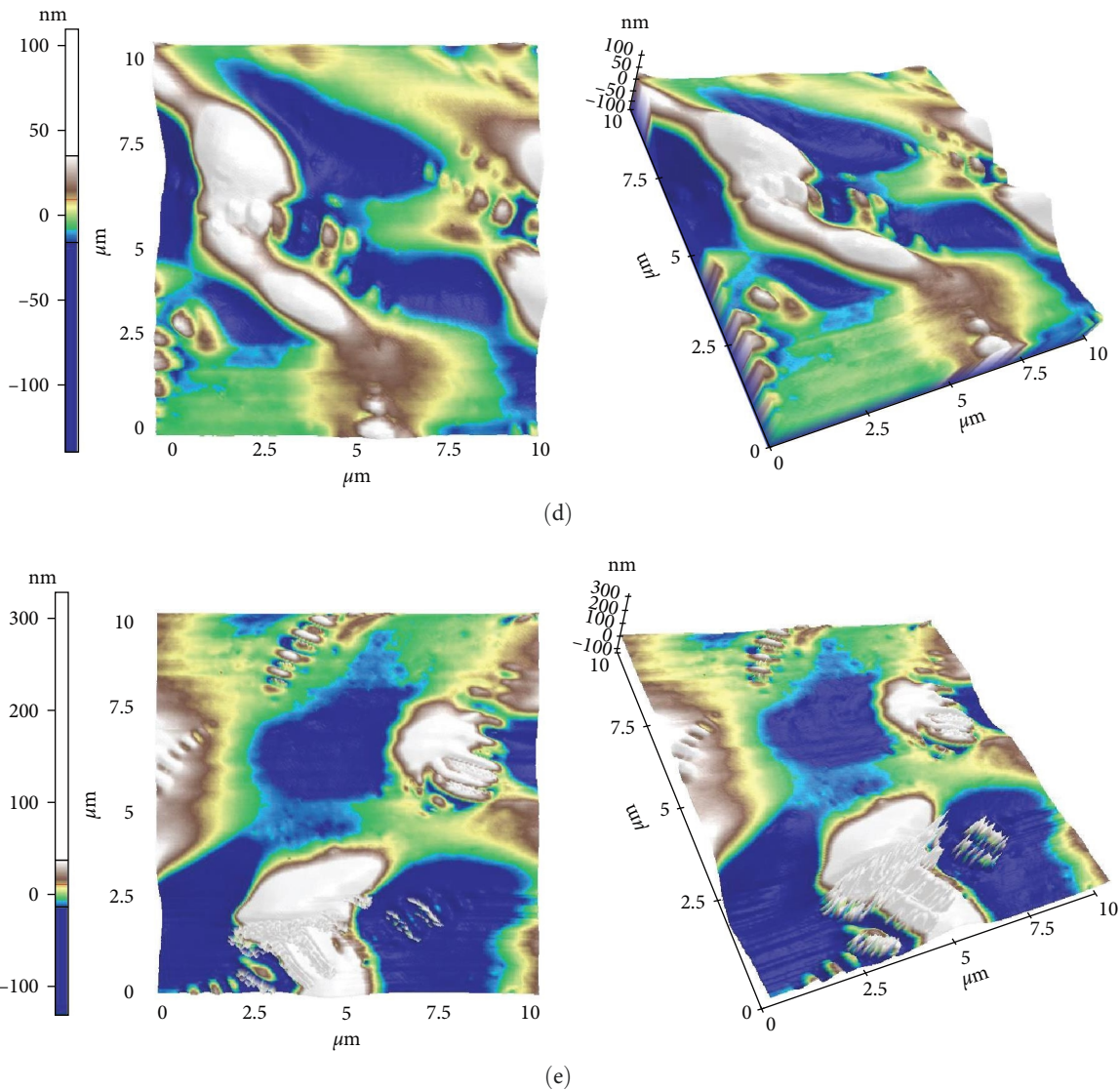

(b)
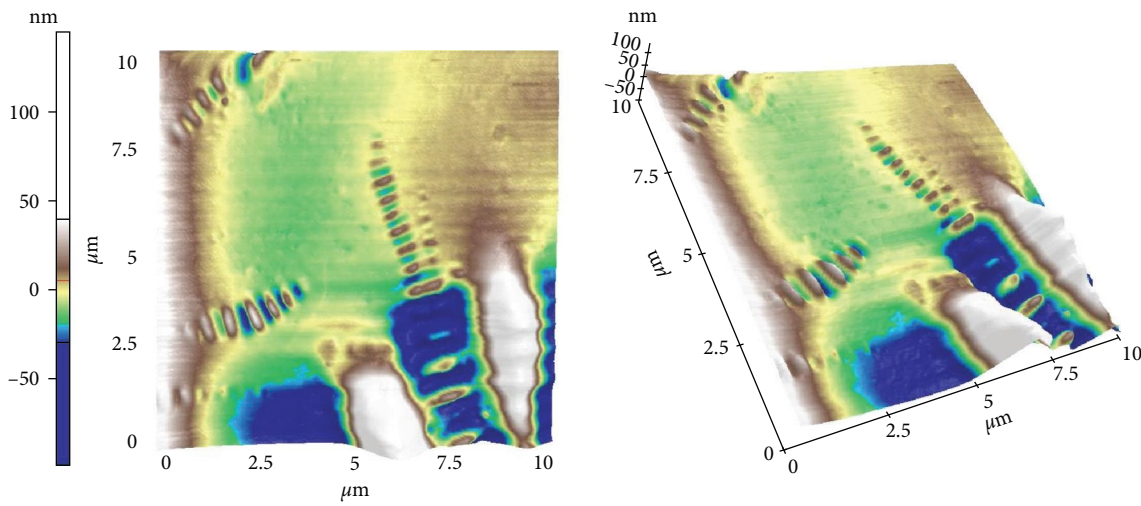
(c)

FIGURE 7: Continued.

(d)

(e)

FIGURE 7: (a) The 2D and 3D topography images of the base bitumen. (b) The 2D and 3D topography images of the bitumen with 3% WLRG. (c) The 2D and 3D topography images of the bitumen with 5% WLRG. (d) The 2D and 3D topography images of the bitumen with 7% WLRG. (e) The 2D and 3D topography images of the bitumen with 9% WLRG.

samples containing 0%, 3%, 5%, 7%, and 9% WLRG. Between 5 and 20 mg, WLRG-modified bitumen sample was used in this test. Figure 6(a) shows the DSC curves for the control bituminous binder. The furnace provided a uniform controlled heating of the WLRG-modified bitumen at a constant temperature range between 25 and 350°C.

The temperature controller implemented the specified temperature program and operated the furnace within the selected temperature range and a heating rate of up to 20°C/min. The maximum average percentages of melting point temperature for 3% and 9% of WLRG-modified bitumen compared to base bituminous binder were 27.0% and 26.62%. The melting point of the bituminous binder was 105.2°C when the environment switched from nitrogen (N_2) to air, where the air content was 78% nitrogen and 21% oxygen. The melting point for samples modified with 3%, 5%, 7%, and 9% WLRG was 133.6, 132.1, 103.5, and 133.2°C, where the sample with 7% WLRG had the lowest melting temperature.

3.4. Morphological Analysis

3.4.1. Atomic Force Microscopy. This study used AFM to investigate the form, shape, and structure of the WLRG-modified bitumen. The morphology and topography of the samples were observed on the *z*-axis because it gives images with the highest resolution. Figure 7(a)–7(e) shows the 2D and 3D topography images of the samples modified with 3%, 5%, 7%, and 9% WLRG. They show that the morphology of the control sample and WLRG-modified bitumens comprise interlaced light and dark stripes that present as a bee-like structure or catana phase. All images show the catana phase intermediate and their lower darker vicinity where the inhomogeneity of WLRG-modified bitumen surfaces, the imaged area may not represent the overall surface characteristics.
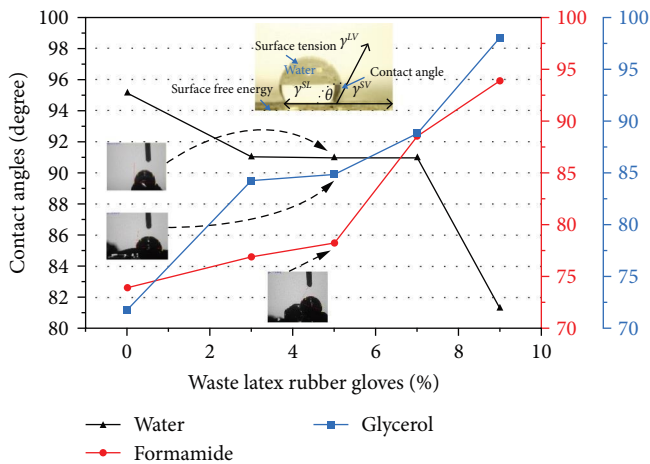
FIGURE 8: The contact angle of the WLRG-modified bitumen.

The distinct characteristics in the 2D and 3D images indicate that the WLRG influenced the morphologies of the bituminous binders. From the 2D and 3D images captured, it can be seen that the different weight of addition will give different amounts of bee's images [43].

The control sample showed a significant catana phase. Adding varying percentages of WLRG to the base bitumen resulted in the modified bitumen having fewer bee structures, as shown in Figure 7(b). The higher WLRG contents reduced the fluctuation in the topography images and made the dispersed zone more apparent. Figure 7(c) shows that the bee structure images were less apparent for the sample modified with 5% WLRG than the bitumen modified with 3% WLRG. The bitumen modified with 5% WLRG also showed a clustered bee structure and the catana phase expanded. Figure 7(d) shows that the catana phase in the sample modified with 7% WLRG blended with the dispersed area. In summary, adding higher percentages of WLRG reduced the number of the bee structure, indicating that the amount of WLRG used to modify the base bitumen influenced the morphologies of the WLRG-modified bitumen binders.

3.5. Contact Angle. This study determined the contact angle of each sample using distilled water, formamide, and glycerol. Distilled water is the most popular probe liquid for measuring contact angles, which shows whether a surface is hydrophilic or hydrophobic. A contact angle is also used to categorize the wetting behavior of a material's surface [44]. The liquid droplet deposited on a solid substrate at three phase boundaries where a liquid, gas, and solid intersect [34]. The benefit of using distilled water to determine the moisture susceptibility of modified bitumen is that water is the primary cause and contributor to asphalt deterioration. Figure 8 shows a decreasing contact angle for distilled water for the WLRG-modified bitumen. The contact angle was smaller when the probe liquid was water, where the sample modified with 9% WLRG had the smallest contact angle. The control binder had a contact angle of 95.15°, which decreased by 4.1% to 91.05° when adding 3% WLRG to the bitumen. The bitumen modified with 5% WLRG had a 4.15% smaller

contact angle of 91°, while the bitumen modified with 7% WLRG had a contact angle of 90.95°, which is a 4.4% difference from the base bitumen. Adding higher percentages of WLRG to the bitumen, from 0% to 3% to 5% to 7% and 9%, resulted in smaller contact angles, which means better substrate wetting.

Figure 8 shows that the contact angle decreased with higher percentages of WLRG. The contact angles of formamide showed a similar trend as distilled water. The contact angle was 73.95° of the base bitumen. The sample modified with 5% WLRG had a contact angle of 88.55°, which was 5.81% higher than the base bitumen. Adding 9% WLRG to the base bitumen resulted in a 26.4% higher contact angle than the base bitumen. The largest contact angle of 93.95° was achieved by the sample modified with 9% WLRG, while the base bitumen had the smallest contact angle of 73.95°. High contact angles of more than 90° indicate poor wetting, while contact angles of less than 90° indicate good wetting [44]. The surface with a 0° contact angle is wet. The contact angle is a critical parameter for applications related to adhesion, binding, adsorption, and intermolecular forces of interfaces.

Unlike water, the contact angles for formamide and glycerol increased after adding WLRG to the modified bitumen. The contact angle of the control sample was 71.85°, and adding 3% WLRG to the base bitumen increased the contact angle to 84.3°, an increase of 17.3%. The contact angle of the sample modified with 5% WLRG was slightly higher (0.9%) than that modified with 3% WLRG. The contact angle for the bitumen modified with 7% WLRG was 88.8°, an increase of 23.6%. In summary, the contact angle increased with higher WLRG contents, where the increase in contact angle was higher than when using glycerol as the liquid probe.

4. Conclusions

The penetration test showed that using WLRG as a modifier resulted in higher binder stiffness, softening point, and ductility. The TGA curve revealed that the first decomposition during the thermal treatment began at a low temperature and continued as the temperature increased, causing the binders to lose weight. The DSC patterns showed that the WLRG-modified bitumen did not affect the crystallization structure after the glass transition of the binders. Therefore, the heat can be measured from DSC machines. The contact angles of water–glycerol and water–formamide increased with higher WLRG contents, but the number of bee structures decreased, showing that the percentage of WLRG influenced the morphology of the bituminous binders. This study has demonstrated a considerable improvement in binder characteristics when using WLRG as a modifier. Bitumen modification with WLRG improved the workability of asphalt binders at low temperatures. Given the volatile rubber price in the global market, using crumb rubber in road pavement constructions could support the economic growth of the Malaysian rubber industry, particularly considering its cost-effectiveness and practicality compared to other materials.

Data Availability

The data will be made available upon request to the corresponding author.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this article.

Authors' Contributions

Auni Diyana Fadzil: conceptualization, formal analysis, and writing—original draft. Nur Izzi Md Yusoff: supervision, project administration, and funding acquisition. Shuhaida Harun: investigation, supervision, and writing—original draft. Khairiah Haji Badri: investigation, formal analysis, and writing—original draft. Iswandaru Widyatmoko: formal analysis, validation, and writing—original draft. Faridah Hanim Khairuddin: investigation, supervision, and writing—review and editing. Abdalrhman Milad: conceptualization, methodology, and writing—review and editing. Munder Bilema: methodology, validation, and writing—review and editing. Naeem Aziz Memon: project administration, investigation, and writing—review and editing. All authors have read and agreed to be accountable for all aspects of the work in ensuring that questions related to the accuracy or integrity of any part of the work are appropriately investigated and resolved.

Acknowledgments

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