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Enhancing the Functional and Environmental Properties of Asphalt Binders and Asphalt Mixture Using Tourmaline Anion Powder Modification

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Abstract: Due to its good piezoelectric and thermoelectrical properties, tourmaline anion powder (TAP) can be used as a potential modifier to improve the piezoelectric, thermoelectric, rheological, and mechanical properties of asphalt binders and asphalt mixtures, respectively. This study was conducted to investigate the functional, piezoelectric, and thermoelectric properties of a TAP-modified asphalt binder (TAPMA) and the corresponding asphalt mixtures. In the study, the TAPMA’s environmental friendliness, such as the volatile organic compound (VOC) adsorption and metal immobilization, were investigated. Compared to TAP at 3.95 pC/N, the piezoelectric constant of TAPMA was found to be 3.42 pC/N. In general, the results indicated that TAP could potentially improve the functional properties of asphalt binders and asphalt mixtures, including the piezoelectric and thermoelectrical properties. With respect to environmental enhancement, the asphalt binder VOC emission reduced to 50% after TAP addition. In terms of metal immobilization, the heavy metals Fe and Ti exhibited the best stability followed by the alkali metals Li, K and Na, and lastly, Ca and Mg, respectively. Nonetheless, the emission concentrations of all the metals were below the regulatory threshold. Furthermore, the study findings also indicated that TAPMA can potentially adsorb the tail gas emissions of vehicles and heavy metals.

Keywords: environmentally friendly material; piezoelectric property; volatile organic compound; metal immobilization; tourmaline anion powder; asphalt binder

1. Introduction

With the growing global awareness of environmental protection and conservatism, there has been a paradigm shift to not only focusing on the functional, structural, and durability aspects of the pavement design, but also on the intelligence (i.e., smart roads), safety, and environment-friendliness (i.e., green roads) aspects of the pavement [1,2]. In the recent years, the modification of asphalt binders using inorganic minerals has become one of the frontline research areas in pavement engineering [3,4]. The consumption of massive natural materials and the emission of global greenhouse gases during conventional road construction have been aggravating the environment issues [5]. It is worth noting that the environmental impact may change at different life-cycle stages, such as raw material processing, asphalt production or waste disposal [6]. To reduce the environmental impact, at present, it is mainly focused on that using reclaimed asphalt pavement, reducing the...
asphalt production temperature and lengthening the service life of the pavement during the life cycle of the asphalt pavement [7]. To better understand the environmental impact during the life cycle of the pavement, life cycle assessments (LCA) were employed in many studies [8,9]. Among other beneficial characteristics, inorganic modifiers have shown potential to improve the interface bonding between the asphalt binder and mineral aggregates, ultimately resulting in enhancing the pavement performance. This modification technology has the characteristics of a simple technical process, low production cost, and abundant reserves [10]. Inorganic modifiers, including carbon black, nanometer calcium carbonate, fiber, diatomite, and other traditional inorganic materials have been widely and successfully used in the modification of asphalt binders [11–14]. Nanoparticles, metal leaching, and polymers [15] showed different effects on fluid properties. Compared to the aged binders, the virgin binders had sparser and larger fibril microstructures as observed by Mikhailenko et al. with ESEM, which may be due to the saturate content [16]. However, most of these inorganic modifiers have unstable modification effects, with little to no environmental benefits [17]. This inevitably warrants the need to explore more stable and environmentally friendly modifiers [18,19].

1.1. Literature Review and Study Motivation

Tourmaline, which comprises widespread borosilicate minerals in nature, generally occurs in high-temperature gas-forming hydrothermal deposits [20]. As a family of the boron-bearing ring silicate minerals, it has a complex chemical composition mostly containing SiO$_2$, B$_2$O$_3$, Al$_2$O$_3$, MgO, CaO, MnO, FeO, Na$_2$O, Li$_2$O, and other substances. Its chemical formula can be generalized as $\text{XY}_3\text{Z}_6[\text{Si}_6\text{O}_{18}][\text{BO}_3]_3(O,\text{OH,F})_4$, where $\text{X}$ is Na$^+$, K$^+$, Ca$^{2+}$, or nothing; $\text{Y}$ is Mg$^{2+}$, Al$^{3+}$, Fe$^{2+}$, Mn$^{2+}$, and Li$^+$, respectively; and $\text{Z}$ is Al$^{3+}$, Fe$^{3+}$, Cr$^{3+}$, and Ti$^{4+}$, respectively [21,22]. Being a tourmaline that belongs to the trigonal system, it has a complex crystal structure, with the basic unit comprising of trigonal rings of silicon–oxygen tetrahedron, (SiO$_4$)$_6$ [23].

The anions released by the tourmaline-neutralizing free radicals have the same properties as those produced in nature. On this basis, tourmaline has been widely used in the fields of air purification, water purification, coating, indoor decoration, and refrigeration, respectively [24–26]. Similarly, tourmaline has been explored for its potential use in pavement materials because of its environmentally friendly properties. Based on the literature reviewed, however, its application as an asphalt binder modifier is still limited [20–28].

Tourmaline can be categorized as tourmaline powder (TP) or tourmaline anionic powder (TAP) based on its chemical composition. TP is predominantly composed of SiO$_2$, Al$_2$O$_3$, and Fe$_2$O$_3$ whilst TAP has components such as CaO, MgO, and SiO$_2$, respectively [25]. There are two main preparation methods for tourmaline-modified asphalt binder, denoted herein as TMA. Compared to the surface modification method, TMA prepared using the mechanical method, namely, the high-speed shear blending method, are susceptible to interfacial separation between the asphalt binder and the tourmaline during storage and construction [27,29].

From the literature, the addition of TP has been proved to be an efficient way to enhance the performance of asphalt binders and asphalt mixtures, respectively. Based on chemical composition analysis, the colloidal structure of the asphalt binder remains as a sol–gel type with TP [30]. From the study [30], the OAC of the three different modified asphalt mixtures with TP dosages of 14%, 17%, and 20% were 5.45%, 5.53% and 5.60% respectively. Hu et al. [31] observed the aging properties of the modified asphalt binder with TP after the thin film oven test (RTFOT). Mozaffari et al. [32] reported that asphaltene aggregates and aggregate clusters formed continuously decreased the viscosity of diluted bitumen with time. The introduction of MnO$_2$ to Fe$_3$O$_4$/CuO nanoparticles was found to enhance the surface area and pore structure to achieve color removal of the fluid [33]. Ye et al. [29] studied the effects of tourmaline on the zero-shear viscosity (ZSV) of modified asphalt binders. Furthermore, it was observed from the study [29] that the quantitative value of ZSV increased as the dosage of TP increased. From the study findings, it was also
observed that TP and TAP modification had a comparable influence on the asphalt binders and asphalt mixtures, respectively. Zhao et al. [10] systematically studied the high- and low-temperature properties of the modified asphalt binders with TAP using three basic asphalt binders based on the bending beam rheometer (BBR) test. The low-temperature property of the base asphalt binder was enhanced to some extent due to the addition of TAP. Hu et al. [31] studied the different anti-aging effects on asphalt binders modified with TP and TAP using the RTFOT test based on the ASTM D 2872 [34] standard specification.

Asphalt binders and asphalt mixtures modified with TAP and TP are considered to have a purification effect. Wang et al. [35] also studied the automobile exhaust-purifying performance of tourmaline-modified asphalt mixtures, using exhaust-purifying tests with an exhaust-gas analyzer and a self-made air purifier chamber. Celikbilek et al. [36] evaluated the performance of TMA mixtures using the Grey multi-criteria evaluation model, whilst Chen et al. [37] made similar evaluations using the Grey target decision method.

From the perspective of pavement performance and purification effect, TAP has a benefit on both the asphalt binder and the corresponding asphalt mixture, respectively, whilst acting as a filler. However, TAP contains B, Li\(^{+}\), Mn\(^{2+}\), Cr\(^{3+}\) and other heavy metal or alkali metal ions that may be potentially harmful to plants, animals, and human health [25,38]. When an asphalt mixture modified with TAP is used in the pavement, these ions have the potential to slowly leach into the soils below or on either side of the road when rain falls on the pavement surface, and possibly contaminate the surrounding soil and groundwater. The net result could be environmental pollution and an undesired threat to human health, animal safety, and plant life. Thus, as the global awareness for environmental protection and conservatism keeps on gaining momentum, so is the quest for the use of more environmentally friendly, resilient, and smart material (i.e., additives, TAP modifiers, etc.) pavement engineering applications, including asphalt binders and asphalt mixtures, respectively. However, the literature reviewed is fairly limited in this area, and hence, there is a need for continued exploration of environmentally friendly and superior-performing tourmaline (TAP) asphalt binder modifiers.

1.2. Study Objectives and Scope of Work

Based on the above stated motivation and as a supplementary enrichment to the literature, the primary goal of this study was to investigate and characterize the functional, piezoelectric, and thermoelectric properties of TAP-modified asphalt binder (TAPMA) and the corresponding asphalt mixtures. For environmental friendliness quantification, this study determined the volatile organic compound (VOC) adsorption and the immobilization of metals in TAPMA and the corresponding asphalt mixtures. In the study, the rheological properties of TAPMA were comparatively measured and quantified using the dynamic shear rheometer (DSR) test device. Additionally, the morphology and microstructure of the asphalt binders were studied and characterized using a scanning electron microscope (SEM) device.

In the subsequent sections of the paper, the study plan and materials are discussed, followed by the laboratory experimentation and test methods. The laboratory test results are thereafter presented, analyzed, and synthesized. The paper then concludes with a summary of the key findings and recommendations.

2. Study Matrix Plan and Materials

The research methodology and study matrix plan incorporated the following key work activities: (a) material procurement and sample preparation (i.e., surface modification); (b) laboratory experimentation and testing; and (c) data analysis and synthesis to draw conclusions and recommendations. The materials and sample preparation are discussed below, followed by the rest of the other work activities in the subsequent sections.
2.1. Base Asphalt Binder

The base asphalt binder used in this study was the AH-70 petroleum asphalt binder. The basic properties of the asphalt binders were evaluated according to the Chinese specification JTG E20-2011 [39], and are listed in Table 1.

Table 1. Technical indices of AH-70 petroleum asphalt binder.

| Technical Indices                  | Unit | Specification | Results |
|-----------------------------------|------|---------------|---------|
| Penetration (25 °C, 100 g, 5 s)   | mm   | 60–80         | 61      |
| Penetration index, PI             | –    | –1.5 ~ +1.0   | –1.42   |
| Softening point, T<sub>R&B</sub>  | °C   | ≥46           | 48.5    |
| Ductility (15 °C, 5 cm/min)       | cm   | ≥100          | >100    |
| Ductility (10 °C, 5 cm/min)       | cm   | ≥15           | 18.3    |
| Density @ 15 °C                   | g/cm³| /             | 1.044   |
| Flash point                       | °C   | ≥260          | 336     |
| Dynamic viscosity @ 60 °C         | Pa·s | ≥180          | 202     |
| Kinematic viscosity @ 135 °C      | Pa·s | /             | 0.418   |
| Mass change After RTFOT           | %    | –0.8 ~ +0.8   | 0.092   |
| Penetration ratio @ 25 °C         | %    | ≥61           | 72      |
| Ductility (10 °C, 5 cm/min)       | cm   | ≥6            | 6.2     |
| Ductility (15 °C, 5 cm/min)       | cm   | /             | 49.4    |

2.2. Tourmaline Anionic Powder (TAP)

The TAP used in this study was yellow in color and had an anionic release rate of over 8000 ions cm⁻³ with a particle size comprising of a 2000 mesh. Its chemical composition and constituent components are listed in Table 2.

Table 2. The chemical composition of TAP.

| Chemical Components | Al₂O₃  | SiO₂   | B₂O₃  | Fe₂O₃  | MgO   | FeO   | Na₂O  | Other |
|---------------------|--------|--------|-------|--------|-------|-------|-------|-------|
| Contents/%          | 35.1   | 34.8   | 11.0  | 10.2   | 4.7   | 1.4   | 0.9   | trace |

2.3. Surface Modification Process

TAP has the properties of both hydrophilicity and lipophobility [40,41]. These characteristic properties are partially responsible for its poor compatibility and dispersibility with the petroleum base asphalt binder, ultimately making the storage stability of TMA problematic. To improve the compatibility and dispersibility between the powder and asphalt binder, a surface modifier (SCA KH-550, Nanjing, China) was used in this study to modify the TAP surface. As exemplified in Figure 1 and discussed in the subsequent text, the TAP surface modification process, which was based on a wet mixing method, comprised of a 4-step procedure.

Figure 1. Surface modification process of TAP.
As aforementioned, a 4-step procedure, namely, dilute solution preparation, material mixing, filtration, and grinding, was adopted for the TAP surface modification in this study. The dilute solution (malcohol:msurface modifier = 90:10) was prepared using anhydrous ethanol and was evenly mixed by stirring. The weighed TAP (~50 g) was then added into the ethanol solution whilst stirring, followed by stabilization/curing of the blended solution at room temperature for 2 h. The surface-modified TAP was thereafter filtered to remove the excess coupling agent and alcohol solution, respectively, followed by vacuum drying of the powder at 60 °C for 4 h. Lastly, the dry powder cake was ground into a powdery material and sealed for subsequent use.

3. Laboratory Experimentation and Testing

As discussed in the subsequent text, the laboratory experimentation conducted in this study included the following: (a) DSR testing for quantifying the rheological properties; (b) VOC measurements; (c) leaching test for the heavy metals; (d) SEM for morphological and microstructure characterization; (e) evaluation of the piezoelectric and thermoelectric properties; (f) semi-circular bending (SCB) test; and, (g) the four-point bending fatigue tests. For each respective laboratory test, a minimum of three samples or test replicates were performed per asphalt binder per TAP dosage per test condition.

3.1. Measurement of the Volatile Organic Compounds (VOC)

TG-MS (toxic gas monitoring system, Liuzhou, China) was used in this study to analyze the absorption effects of different TAP dosages on the VOCs emission from the petroleum base asphalt binder. The TG-MS test was conducted firstly to achieve thermal decomposition, and thereafter, the specimens were exposed to MS to separate out the pure substances using the deflecting magnetic field. For the TG conditions, the initial temperature was 25 °C, which was increased to 600 °C at a rate of 10 °C/min and maintained for 30 min to accelerate the VOC emission. The thermal decomposition products containing VOC were blown into MS with the carrier gas, helium (He), at a flow rate of 1.0–2.0 mL/min under TG conditions. The purity of helium is 99.9% and the helium was bought from Nanjing Hongfa Industrial Gas Co. Ltd.

MS can potentially be used to detect changes in the chemical characteristics of an asphalt binder system using different deflecting radii. In this study, the adopted MS conditions comprised of using an electron beam energy with a 65 eV ion source to achieve electron bombardment. The mass scanning temperature was like that used for the TG conditions, with a mass scanning range of 0 m/z to 300 m/z and a scanning time-period of 0.1 s. The VOC concentration was tested by a gas detector (ADSK-4) after heating to the aimed temperature.

3.2. The Leaching Test of Heavy Metals

The heavy metal leaching test used in this study conformed to the existing national guidelines and environmental protection industry standards of China, GB 5085.3-2007, HJ/T 299-2007, and GB/T 30810-2014, respectively. The sulfuric–nitric acid method was used for the leaching of the modified asphalt mixture. This method can simulate the leaching process of leachable components from the modified asphalt binder with TAP under the influence of acidic rainfall, which is a relatively conservative leaching method.

During the leaching test, the specimens of the modified asphalt binder were dried and broken into small pieces, and thereafter, they were grinded using the agate ball mill followed by sieving with a square hole sieve. Particles with a dimensional size of 0.125–0.25 mm were collected as the test samples. Secondly, 10 g samples were weighed and put into a 1 L beaker. After the addition of 500 mL water, a magnetic stirrer was used for stirring for up to 3 h. Thirdly, the extraction solution (Vwater:Vsulfuric acid:Vnitric acid = 3:2:1, 450 mL in total) was added until the pH = 7.0 ± 0.5 and then stirred for 2 h.

After the leaching solution was collected by filtering with a 0.45 µm filter membrane, the used filter membrane and sample residues were put into a beaker. Fourthly, after
500 mL water was added, the leaching solution was continuously added until the pH value was $3.2 \pm 0.5$, and thereafter, the solution was stirred continuously for 7 h. A 0.45 µm filter membrane was used for filtration and sample residue collection. Thereafter, the two leaching solutions were transferred into a 2 L volumetric flask for constant volume measurements. Finally, the concentration of the heavy metals was determined using graphite furnace atomic absorption spectrometry, also known as inductively coupled plasma–atomic emission spectrometry (ICP–AES).

3.3. Morphological and Microstructure Characterization

A field emission scanning electron microscope (SEM, Zeiss, Sigma HD, Carl, Germany) was used to investigate and characterize the morphology of the asphalt binders, the base asphalt binder and TAPMA, respectively. The SEM voltage and current used were 35 kV and 10 mA, respectively. To avoid magnetic effects, the distance between the probe and sample was kept above 10 mm. Energy dispersive spectroscopy (EDS) was used to evaluate and quantify the microscopic agglomerations of the elemental components within the asphalt binder and TAPMA, respectively.

3.4. Measuring the Piezoelectric and Thermoelectric Properties

The purification effect of TAP mainly originates from its piezoelectric and pyroelectric effects. When the pressure or temperature changes, positively charged particles can be absorbed and sedimented with the free electrons that are released. The piezoelectric and pyroelectric properties of TAP are very critical in enhancing its modification effects and chemical blending with the base asphalt binder. In this study, piezoelectricity and pyroelectricity testing was conducted for both the TAP materials and modified asphalt binders, respectively.

In the study, the physical indices, namely, the piezoelectric and dielectric constants were used to evaluate and quantify the piezoelectric properties using the static piezoelectric constant tester for piezoelectric materials. Likewise, the following physical indices, conductivity, Seebeck coefficient, thermal conductivity, and power factor were used to evaluate and quantify the pyroelectric properties using the multifunctional conductivity tester.

3.5. The Semi-Circular Bending (SCB) Test

In this study, the universal testing machine (UTM, Branchburg, NJ, USA) was used for performing the SCB test within 80% of the machine loading capacity to quantify the fracture strength of the TAP-modified asphalt mixtures. The 3-point loading mode comprised of a bottom beam support and a cylinder support with a 1 cm diameter top pressure head and lower support. The contact area between the lower support and specimen had plastic paper coated with oil to reduce sliding resistance. The test was conducted in a temperature-controlled chamber at 0 °C. Standard SCB specimens, comprising of 150 mm diameter and 40 mm thickness, were used [42].

After SCB testing, the maximum vertical load, $F$, and vertical displacement, $d$, at the peak load were used to compute the material failure strength, $\delta_t$, using Equation (1) [43,44] below:

$$\delta_t = \frac{4.976F}{BD}$$

where $\delta_t$ is the specimen failure strength; $F$ is the maximum vertical load; $B$ is the thickness of specimen; and $D$ is the specimen diameter, respectively.

Additionally, the fracture energy of the specimens was used to analyze and compare the amount of energy needed to produce cracking in the different asphalt mixtures when subjected to SCB testing. Equations (2) and (3) were used for computing the fracture energy [45].

$$A = \sum_{i=0}^{n} [(x_{i+1} - x_i)y_i + 0.5(x_{i+1} - x_i)(y_{i+1} - y_i)]$$

$$G_f = \frac{A}{I \times h} \times 10^6$$
where $A$ is the area under the load-mid-span deflection curve; $x_i$ is the vertical displacement of point $i$; $x_{i+1}$ is the vertical displacement at point $i + 1$; $y_i$ is the load at point $i$; $y_{i+1}$ is the load at point $i + 1$; and $h$ is the height of the SCB specimen.

3.6. The 4-Point Flexural Bending Beam Fatigue (FBBF) Test

A constant strain-controlled fatigue test, which was a 4-point flexural bending beam fatigue (FBBF) test with a loading span of 118.5 mm, was conducted on the designed asphalt mixtures using the Chinese standard specification T 0739 2011 [39]. An Australian UTM-100 machine was used for conducting the FBBF tests. Typically, vertical loading causes tension in the bottom central zone of the beam specimen. The induced mid-span deflection and the maximum measured tensile strain recorded are the key output parameters used to characterize the fatigue behavior of a specimen subjected to FBBF testing.

Theoretically, the load spectrum of an in-service pavement is assumed to be close to a haversine waveform. So, a haversine repeated loading waveform was selected for the FBBF tests conducted in this study. The input haversine waveform was thus formulated as illustrated in Equation (4) below:

$$Y = \frac{A}{2} [1 - \cos(2\pi ft)]$$

where $A$ is the amplitude, $f$ is the loading frequency, and $t$ is the loading time. Four replicate beam specimens were tested per mix-design per test condition. Prior to FBF testing, the beam specimens were preconditioned in an environmental chamber for 12 h. In this study, the FBBF testing was conducted at 15 °C in accordance with the Chinese standard specification T 0739 2011 [39]. Parameters recorded during testing included the phase angle and stiffness. A reduction of 50% in the initial stiffness of the beam was used as the fatigue failure criterion [46]. In the study, the FBBF test was automatically set to terminate when the initial stiffness of the beam specimens decayed to 50% of the initial value.

4. Laboratory Results, Analysis, and Discussions

The laboratory test results, including the TAPMA rheology, VOC emissions, heavy metal leaching, morphological structure, piezoelectric, and thermoelectric properties, are presented and synthesized in this section of the paper. The fracture properties and cracking characteristics of the corresponding TAP-modified asphalt mixture, tensile strength, fracture energy, flexural stiffness, and fatigue life are also presented and discussed in this section as well.

4.1. DSR Rheological Properties and Synthesis

The complex shear modulus ($G^*$) represents the ability of asphalt binders to resist permanent deformation at high temperatures [47]. In this study, the rheological properties, namely, the complex shear modulus ($G^*$) and phase angle ($\delta$) of the asphalt binders and TAPMA were measured using the DSR test device. The corresponding DSR test results are plotted in Figure 2.

The relationship between the complex shear modulus ($G^*$) and the TAP contents for all the asphalt binders evaluated is shown in Figure 2a. As shown in Figure 2a, the complex shear modulus decreases significantly with an increase in temperature. However, as indicated from the increasing magnitude of the $G^*$ value as a function of TAP dosage, adding TAP into the base asphalt binder can potentially improve the high-temperature tolerance and stiffness of the asphalt binder. This indicates that the addition of TAP improves the high-temperature deformation resistance of the asphalt binder. This is mainly because TAP has a large specific surface area and strong adsorption effects on the asphalt binder. Due to these characteristic properties, it has the potential to effectively improve the high-temperature rheological properties of the asphalt binder.
As theoretically expected and considering the viscoelastic nature of asphalt binders, the greatest stiffness with the highest G* value was recorded at 58 °C, followed consecutively by the G* values at 64 °C and 70 °C (i.e., lowest values), respectively. However, the difference in the G* value is quite significant for 58 °C in comparison to the other points. This response behavior is consistent with viscoelastic materials such as the asphalt binder, which can change from a high elastic state to a viscoelastic state with an increase in temperature. In general, as the elastic ratio of the asphalt binder decreases with an increase in temperature, its viscous ratio increases with a corresponding decay in its ability to resist deformation.

The phase angle (δ) illustrated in Figure 2b represents the material performance in terms of the viscous or elastic response behavior. This was experimentally determined by measuring the time lag between the applied sinusoidal stress and the corresponding strain response. The phase angle reflects the elastic and viscous properties of asphaltic materials. The larger the δ value is, such as 90°, the stronger the viscous response behavior and the weaker the elastic property of the asphalt binder. As evident in Figure 2b, the phase angle generally increased up to a 15% TAP content, and thereafter, decreased. TAP is an elastic material and its phase angle is zero. For TAP dosages over 15%, the influence on the TAPMA gradually increases and suppresses the influence of the asphalt binder viscosity.

In this study, the rutting temperature of TAPMA was determined at a rutting factor (G*/Sin (δ)) of 1.0 kPa. The results of these rutting factors as a function of TAP dosage at different temperatures are shown in Figure 3.

As theoretically expected of viscoelastic materials, the rutting factor of the asphalt binders in Figure 3 exhibited a decreasing trend with an increase in temperature. The rutting factor (G*/Sin (δ)) is a rheological parameter that is quantitatively used to characterize the rutting resistance of asphalt binders. In general, an asphalt binder with a higher G*/Sin (δ) value is theoretically considered to have better rutting resistance at high temperatures, and vice versa for asphalt binders with lower G*/Sin (δ) values.

Figure 3 shows that the G*/Sin (δ) of the TAP-modified asphalt binder increased with an increase in the TAP content. This observation indicates that the addition of TAP has the potential to improve the rutting resistance of asphalt binders at high temperatures. With an increase in the temperature, however, the (G*/Sin (δ)) of the TAP-modified asphalt binder decreased, with the (G*/Sin (δ)) differential decreasing as the TAP dosage increased. This response was not unexpected and was theoretically attributed to the viscoelastic nature of the asphalt binder itself.
Asphalt binder is a complex material composed of different molecular weight organic hydrocarbons, with the potential to easily produce VOCs at high or normal temperatures and pressures, respectively. The VOCs of TAPMA mainly included C\textsubscript{14}H\textsubscript{30}, C\textsubscript{18}H\textsubscript{38}, C\textsubscript{18}H\textsubscript{39}N. As shown in Table 3, the VOCs concentration of TAPMA increased with an increase in temperature, and vice versa as the TAP content increased.

### Table 3. VOC emissions of TAPMA (ppm).

| Type          | 160 °C VOCs Concentration | 180 °C VOCs Concentration |
|---------------|---------------------------|---------------------------|
| Base asphalt-binder | 253.1                     | 696.0                     |
| 5% TAPMA      | 139.3                     | 235.1                     |
| 10% TAPMA     | 112.2                     | 189.2                     |
| 15% TAPMA     | 90.4                      | 124.9                     |
| 20% TAPMA     | 87.0                      | 123.5                     |
| 25% TAPMA     | 71.4                      | 105.1                     |

As exemplified in Table 3, the VOCs emission of the asphalt binder reduced by over 50% when TAP was added into the base asphalt binder. Moreover, the VOCs concentration of TAPMA is lower than that of the base asphalt binder. Overall, the results indicated that a high temperature could promote the VOCs emission and that TAP can potentially inhibit the VOCs emission. When compared to the VOCs concentration at 15% TAP, the decline in the VOCs concentration is minimal for TAP contents greater than 15%. Therefore, it is tentatively reasonable to propose 15% as the optimum TAP dosage.

### 4.3. Heavy Metal Leaching Results and Synthesis

The heavy metal leaching results for both the base asphalt binder and TAPMA are listed in Table 4. As shown in the table, the heavy metal leaching of TAPMA included Fe, Mg, Al, Si, Na, and La, respectively.

### Table 4. Heavy metal leaching results (leaching concentration (µg/L)).

| Type                | Al   | Si   | Mg   | Fe   | Na   | La  |
|---------------------|------|------|------|------|------|-----|
| Base asphalt binder | 0.15 | 0.24 | 0.23 | 0.58 | 0.62 | 0.01|
| 5% TAPMA            | 0.19 | 0.28 | 0.45 | 1.25 | 0.63 | 0.02|
| 10% TAPMA           | 0.23 | 0.31 | 0.52 | 1.46 | 0.64 | 0.04|
| 15% TAPMA           | 0.27 | 0.35 | 0.61 | 1.58 | 0.65 | 0.05|
| 20% TAPMA           | 0.31 | 0.38 | 0.68 | 1.63 | 0.65 | 0.05|
| 25% TAPMA           | 0.35 | 0.42 | 0.72 | 1.72 | 0.65 | 0.06|
As evident in Table 4, the leaching concentrations of the different heavy metals are different. From the table, the leaching concentrations of Fe and Mg were comparably higher than the other metals, with La having the least concentration. In general, the heavy metal leaching concentration increased with an increase in the TAP content, with TAPMA exhibiting higher concentrations than the base asphalt binder. Overall, the results indicate that TAP can potentially promote heavy metal leaching and that the TAP dosage has a significant effect on the heavy metal leaching of asphalt binder.

4.4. SEM Morphological and Microstructure Results

The micro-morphological results of both the base asphalt binder and TAPMA from the SEM testing are shown in Figure 4. As shown in the figure, the particles increased as a function of the TAP content without any aggregation. In fact, the TAP appears to be evenly distributed within the asphalt binder. By comparison, Figure 4 shows that TAP can potentially distribute well into the asphalt binder matrix when the TAP content is less than 25%. On this basis and similar to previous results (i.e., Table 4), 15% would be recommended as the optimum TAP dosage.

![Morphological results](image)

Figure 4. Morphological results: (a) base asphalt, (b) 5%, (c) 10%, (d) 15%, (e) 20%, and (f) 25%.
Figure 5a shows the SEM imaging of the tourmaline powder. It can be seen from the figure that TAP exits in the shape of lamellar structures, and each layer of the lamellar structure are stacked on top of each other, which inherently forms clusters of distribution structures with a large specific surface area. As shown in Figure 5b, TAP is dispersed in the base asphalt binder with irregular particles, and a three-dimensional network structure, which is “spongy”, can be obtained/observed. Under this morphological condition, the TAP is essentially wrapped with the base asphalt binder, and there is no obvious boundary between them, thus forming a stable matrix with the base asphalt binder.

![Figure 5. SEM imaging: (a) TAP and (b) TAPMA with 15% TAP content.](image)

### 4.5. Piezoelectric and Thermoelectric Properties

The laboratory results of the piezoelectric and thermoelectric properties of TAP and TAPMA are listed in Table 5. The results include the piezoelectric, dielectric, electrical, and Seebeck constants, respectively.

| Type   | Piezoelectric Constant (pC/N) | Dielectric Constant (10–12 F/m) | Electrical Conductivity (S/m) | Seebeck Constant (µV/K) |
|--------|------------------------------|---------------------------------|-------------------------------|-------------------------|
| TAP    | 3.98                         | 3.54                            | 4.9                           | 172.2                   |
| 5% TAPMA | 3.29                        | 3.30                            | 4.5                           | 174.4                   |
| 10% TAPMA | 3.32                      | 3.45                            | 4.8                           | 176.2                   |
| 15% TAPMA | 3.41                      | 3.51                            | 5.6                           | 179.8                   |
| 20% TAPMA | 3.45                      | 3.56                            | 5.7                           | 180.1                   |
| 25% TAPMA | 3.48                      | 3.59                            | 5.8                           | 180.3                   |

As shown in Table 5, the Seebeck constant indicates that the thermoelectric properties of TAPMA increased with an increase in the TAP content. However, the table also shows that the introduction of asphalt binder to generate the TAPMA matrix could slightly decrease the piezoelectric, dielectric, and electrical constants of TAP, for dosages less than 15%. In particular, the piezoelectric constant of TAPMA decreased with the addition of TAP through to 10%, and thereafter, increased for TAP dosages of 15% and higher. On this basis, any TAP dosage equal to or higher than 15% would be proposed as the optimum. Overall, the results in Table 5 indicate that TAP can potentially enhance the piezoelectric properties of the asphalt binder and that TAP content has a significant effect on the piezoelectric properties of TAPMA.
4.6. Fracture Properties and Fatigue Life of the TAPMA Mixture

The low-temperature tensile strength results from the SCB splitting tests are shown in Figure 6. As shown in the figure, the SCB results of the half-circle splitting tests show that the tensile strength exhibited an increasing trend as a function of the TAP dosage. In terms of the low-temperature SCB tensile strength, Figure 6 shows that all TAPMA mixtures had a higher tensile strength than the base asphalt mixture with 0% TAP. Compared to the base asphalt mixture, when the TAP contents were 5%, 10%, 15%, 20% and 25%, the SCB tensile strength of the TAPMA mixtures increased by 3.65%, 6.44%, 19.65%, 21.70%, and 22.33%, respectively.

Figure 6. The SCB tensile strength results.

In general, the larger the proportion of fine aggregates, the larger the specific surface area of the aggregates. This characteristic property is conducive for the formation of the structural asphalt binder. This increase in the amount of structural asphalt binder is conducive for improving the cohesion and stability of the asphalt mixture, ultimately leading to the enhancement of the low-temperature tensile strength observed in Figure 6, and the overall performance of asphalt mixtures under low-temperature conditions. This performance enhancement can be attributed to the filling effects of the TAP micro-aggregates in the asphalt mixture, leading to an improvement in the pore structure and making the asphalt mixture more compact. Additionally, TAP has a large specific surface area, and its particles exhibit strong physical and chemical interactions with the asphalt binder, thus increasing the overall strength and stiffness of the asphalt mixture.

Compared to 5% and 10%, Figure 6 shows a significant improvement in the tensile strength for 15% TAP. However, the tensile strength gain for a TAP dosage greater than 15% is very marginal. Using the above theory, these results infer that a dense structure within the asphalt mixture occurred at 15% TAP content. Thus, based on the SCB results in Figure 6, 15% would be suggested as the optimum TAP dosage.

The SCB fracture energy results are shown in Figure 7. At low temperature, the fracture energy of the base asphalt mixture was found to be smaller than that of the TAPMA mixtures. This suggests that TAPMA mixtures can endure more stress loading in low-temperature environments prior to fracturing or crack damage.
Due to the influence of the fine-aggregate ratio and the asphalt binder-aggregate ratio on the cohesion and stability of the asphalt mixture, the fracture energy will be different because of the different energy storage capacities prior to the fracturing of the asphalt mixture. The diameter of TAP is about 8 µm, which is much smaller than that of the mineral powder. The proportion of the fine aggregates in TAPMA mixtures is considerably higher and so is the proportion of the asphalt binder in asphalt mixtures. In the asphalt mixture, asphalt acts as an adhesive agent to glue the aggregate particles together, and the cohesion of the asphalt mixture is mainly provided by the asphalt binder. Therefore, when the TAP content is smaller than 20% as shown in Figure 7, the greater the asphalt binder-aggregate ratio in the TAPMA mixture is, the stronger the cohesion generated, and the greater the fracture energy. When the TAP content exceeds 20%, the asphalt binder-aggregate ratio remains the same due to an increase in the fine-aggregate proportion. Therefore, the adhesion between the asphalt binder and aggregate becomes insufficient and affects the overall fracture strength of the mixture.

The FBBF test results, namely, the fatigue life and initial stiffness modulus, are shown in Figure 8. Compared to the base asphalt mixture with 0% TAP, Figure 8a shows that the fatigue life of the TAPMA mixtures improved with the addition of TAP and increased almost linearly as the TAP content was increased. Within the range of the FBBT stress level, it is also worthwhile to note that the fatigue life of all asphalt mixtures exhibit a linear relation on the y-axis.

**Figure 7.** The SCB fracture energy results.

**Figure 8.** FBBF test results: (a) fatigue life numbers, and (b) initial stiffness modulus.
From Figure 8a, the TAPMA mixture with a 25% TAP content quantitatively exhibited the highest fatigue life, whilst the base asphalt mixture with 0% TAP recorded the poorest performance. As can be observed in the figure, the improvement effect of TAP is more significant at the lower FBBF stress ratio. When the stress ratio is 0.3, the fatigue life of the TAPMA mixture with a 25% TAP content was about four times that of the base asphalt mixture versus about 1.5 to 2.0 times at 0.7. In general, the fatigue life of all the asphalt mixtures decreased with an increase in the stress ratio, as would be theoretically expected. This is because an increase in the stress load can potentially lead to deformation of the beam specimens, and thus reducing the overall fatigue life of the asphalt mixture.

Theoretically, flexible materials are typically considered to have a superior fatigue life. Compared to the base asphalt mixture with 0% TAP, the TAPMA mixtures have higher densities due to the fine TAP material. In Figure 8b, the initial stiffness modulus of the TAPMA mixtures is lower than that of the base asphalt mixture at all the stress ratios. This means that the TAP-modified asphalt mixtures are more flexible with a better potential to absorb the accumulated stresses, and thus leading to their longer fatigue lives. Additionally, the fatigue life is also a function of the mixture porosity and density. With the TAMPA mixtures, TAP helps to fill in the porosity and density of the mixture, leading to more uniform stress distribution within the beam specimen, and ultimately contributing to a better fatigue life for the TAMPA mixtures.

TAP has a high specific surface area and many pores inside it can readily absorb the parts of the oil components of the asphalt binder that partially govern the asphalt binder viscosity. Furthermore, the surface modification agent has the tendency to potentially open up the TAP molecular chain, which inherently combines with the asphalt binder macro-molecular chain. This phenomenon partially hinders the movement of the asphalt binder molecular chain segments and effectively improves the overall stability and performance of the TAMPA mixture.

5. Conclusions and Recommendations

This study was conducted to investigate and characterize the functional, piezoelectric, and thermoelectric properties of tourmaline anionic powder (TAP)-modified asphalt binder (TAPMA) and the corresponding asphalt mixtures. The following conclusions were made:

- The addition of TAP improved the temperature tolerance, complex shear modulus, and rutting resistance of the TAP-modified asphalt binders;
- Whilst high temperature promoted VOCs emission, TAP exhibited the potential to inhibit VOCs emission;
- TAP indicated a potential to promote heavy metal leaching and that the TAP concentration had a significant effect on the heavy metal leaching of the asphalt binder;
- TAP indicated a potential for even distribution within the asphalt binder for TAP contents less than 25%. Thus, to optimize homogeneity within the asphalt binder matrix, the optimum TAP dosage should be less than 25%;
- TAP-modified asphalt binder indicated a potential to improve the low-temperature fracture properties and performance of the corresponding asphalt mixture, with the tensile strength results suggesting 15% as the optimum TAP dosage. Further, TAP reduced the initial stiffness modulus of the modified asphalt mixtures with a corresponding gain in the fatigue life.

Whilst the results were plausible, more asphalt binders, including laboratory testing for mixture rutting and moisture evaluation, along with field verification, is warranted in future studies to supplement the findings reported in this paper. However, the study beneficially contributes to enriching the literature through the provision of a reference datum for using TAP as an environmentally friendly asphalt binder modifier. The intelligent pavement will be a major research direction of TAP in the future, and the authors should find a more environmentally friendly method to use TAP. For example, TAPMA pavement can be used to absorb automobile exhaust.
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