

Article

The Utilization of Waste Toner as a Modifier in Trinidad Asphalts

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Abstract: As more persons are adjusting to home working in light of the COVID-19 pandemic, there has been a significant increase in the use of technology. Trinidad and Tobago, like many other Small Island Developing States, began exploring strategies in the areas of recycling and reuse techniques to mitigate negative environmental impacts from the disposal of waste toners. The reuse of waste toners as a performance enhancer in bituminous materials has successfully been achieved in foreign jurisdictions; however, the lack of research on the utilization of the indigenous Trinidad Lake Asphalt (TLA) and Trinidad Petroleum Bitumen (TPB) has stymied the application of this strategy locally. The influence of four waste toners (A, B, C, and D) on the rheological properties of an unmodified TLA/TPB paving binder was measured using the dynamic shear rheology (DSR) testing technique. The addition of waste toners noted improvements in the rheological parameters of stiffness, elasticity, and viscosity, exhibiting superior temperature susceptibility. Of great interest was the observation at 90 °C, where the modified paving binders containing 5% Toner C and 20% Toner D were elastically superior to the world-renowned TLA. This study demonstrated the potential of utilizing waste toner as a bitumen modifier, providing an innovative, sustainable disposal option.

Keywords: waste toner; asphalt; rheology; recycling; e-waste



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1. Introduction

E-waste can be defined as an electronic product that has become unwanted, has stopped working, or is approaching the end of its “useful life” [1]. Electronic products include computers and associated accessories, such as printers and toner cartridges, batteries, chords, televisions, stereos, copiers, and fax machines. It was estimated that in 2019 alone, approximately 53.6 million metric tons (Mt) of e-waste was generated worldwide, projecting an increased generation rate of approximately 2 Mt per year and resulting in a yearly generated quantity of 74 Mt by 2030 [1]. The advent of the COVID-19 pandemic has resulted in a significantly higher level of e-waste generation as the global demand for electronics increased due to more people (professionals and students) functioning from home [2].

Waste toner generated from copiers and printers is a category of e-waste that has proven to be problematic due to its favored disposal techniques [1,3]. The plastic waste and toner dust are disposed of using methods such as incineration and landfilling, which adversely affect the environment and have associated health issues [4]. The plastic component, comprising of engineering grade polymer, takes several years to decompose in the natural environment and, if incinerated, can produce harmful dioxin gases; the toner dust can pose respiratory problems if inhaled in large quantities from exposure through leaking residual toner [5,6]. The key ingredients of the toner dust, which is described as a fine powder, are pigments and polymers; its primary constituents are binder or toner resin, colorants, such as dyes or pigments, charge control agents, releasing agents, and other additives [7].

Although many toner cartridge manufacturers have implemented cartridge return programs, the occurrence of improper disposal is still frequent, especially in developing countries and Small Island Developing States (SIDS) such as Trinidad and Tobago (TT), as these territories tend to lag in regard to the recycling of waste materials [8]. TT has made key advancements in the areas of policy development and action in waste minimization and recycling, which include the development of the National Environment Policy (4) [9]. TT is also a signatory to the Basel Convention Regional Centre for Training and Technology Transfer for the Caribbean Region (BCRC-Caribbean), which is principled in the idea that waste is a resource, and recycling and reusing are techniques to mitigate negative environmental impacts and stimulate economic development, innovative employment opportunities, and small business entrepreneurs [10].

A review of the literature has shown that the sustainable recycling of waste toner cartridges has the potential for the mitigation of the negative effects caused by the dumping of this e-waste [3]. Factors such as the practicality, cost, and benefits associated with the usage of the waste material in any application must be taken into consideration when exploring its versatility [11]. Research conducted on the incorporation of waste toner dust as an asphaltic binder has proven to be successful. In a study conducted by Solaimanian et al., 1997 [12], the impact on the complex shear modulus, phase angle, creep stiffness, logarithmic creep rate, and rotational viscosity parameters were analyzed upon the addition of different levels of waste toner to two binders across different temperatures. The results from this study showed that the toner-modified binder displayed improvements in resisting permanent deformation with increasing temperature; additionally, increasing the amount of waste toner added amplified the viscosity and complex shear modulus of the binder as the phase angle decreased. Through their work, Solaimanian et al., 1997 [12] were able to conclude that although the addition of the toner to the binders enhanced the high-temperature properties, resulting in improved resistance to the occurrence of permanent deformation, the stiffness at lower temperatures was compromised.

Despite the successes experienced in utilizing waste toner for asphalt modification in other countries, the wholesale adoption of these techniques in TT may not be feasible. This is due to the possibility that the degree of compatibility of the waste toner with the indigenous Trinidad Petroleum Bitumen (TPB) and Trinidad Lake Asphalt (TLA) binders may vary compared to the binders used in other studies. Asphaltic binders vary in composition, which is dependent on their source (as in the case of naturally occurring binders, such as TLA) and the manufacturing processes used (as in the case of refinery bitumens, such as TPB); this may result in differences in the rheological and performance characteristics of modified blends [13–18]. Simply stated, no two binders will interact the same with a particular additive. Because of the nature of waste toner, Yildirim et al., 2004 [11] found that agitation before mixing with the aggregates gave consistent results when analyzing blends within a 0–20 wt% range. The possibility of determining the benefits of utilizing waste toner as a modifier for TLA and TPB road paving binders would require scientific investigations incorporating the recommendations from the previous studies, as the literature has revealed no relevant studies in this area.

Since asphaltic binders exhibit viscoelastic properties, the use of the dynamic (oscillatory) shear rheology (DSR) testing technique has proven to be a successful technique for measuring the rheological properties of asphaltic materials, as well as blends modified with waste materials [15,18–24]. The effect of additives and modifiers on parent binders can be studied by measuring their effects on the rheological values: complex modulus, G^* (degree of stiffness); phase angle, δ (degree of elasticity); and viscosity, μ (resistance deformation). The complex modulus (G^*) represents the degree of stiffness of the material, while the phase angle (δ) is used to measure the degree of elasticity of the material [25,26].

The objective of this study was to measure the effect of several waste toner products (HP, Xerox, Sharp, and Konica Minolta) on the key rheological parameters, G^* , δ , and μ , of modified TLA and TPB using the small angle dynamic (oscillatory) shear rheology (DSR) testing technique. The results of this study were then used to evaluate the potential for

utilizing waste toner as a performance-enhancing additive for road paving applications in TT, while also providing a sustainable and environmentally friendly strategy for the disposal of waste toner material. This presents shortcomings for utilizing environmentally unfriendly waste toner as an asphalt modifier in Trinidad and Tobago; thus, the attempt to bridge the gap in empirical data for combining waste toner with Trinidad asphalts is addressed in this paper.

2. Results and Discussion

In order to study the viscoelastic properties of an asphalt material at different stress and strain levels, the behavior of the material must still be linear; that is, the relationship between stress and strain must only be affected by temperature and loading time and not by the magnitude of the stress or strain [27]. Previous studies have employed rheological analysis to gauge how well the new mixes performed after recycled materials were added as a binder [15,18–24,27]. The rheological characteristics of a specimen's complex shear modulus (G^*), phase angle (δ), and viscosity (μ) were measured using a dynamic shear rheometer. The viscoelastic behaviors of binders at moderate to high temperatures were assessed using a dynamic shear rheometer [28]. The complex shear modulus (G^*) is a representation of a material's total resistance to deformation when constantly sheared, while the phase angle (δ) represents the lag between the applied shear stress and the resulting shear strain; practically, a mainly viscous material will have $\delta = 90$ degrees, whereas a mainly elastic material will have $\delta = 0$ degrees [28]. Viscosity, according to Wang et al. [29], is the measure of a material's resistance to flow and deformation. An asphaltic binder with a high viscosity is more likely to uphold a certain standard of strength and stiffness without significant shear deformation at high temperatures [25].

Building the master curve at a specified reference temperature is the first step in modeling the viscoelastic behavior using the results of the dynamic shear rheometer tests; this is done in accordance with the time–temperature superposition principle, which involves correlating the equilibrium between time and temperature [30]. Master curves (the relationships between the complex moduli (G^*), phase angles (δ), and viscosity (μ) at different frequencies) were developed for the bituminous blends containing each of the four (4) toners. The temperature of 60 °C and the frequency of 1.59 Hz were selected for the discussion of results since the average road pavement temperature in the tropics ranges from 55 °C to 65 °C, and the 1.59 Hz frequency simulates the shear rate associated with the traffic speed of a vehicle traveling at 90 km/h (55 mph) [16,26].

Figures 1–3 depict the master curves representing the variations in the complex moduli (G^*), phase angles (δ), and viscosity (μ), respectively, with the addition of Toner D. The trends in the master curves were the same for all the four (4) waste toners (A to D), demonstrating that the frequency of loading had a significant influence on the rheological behavior of bituminous materials. Consistent with the findings of previous studies, the complex modulus increased, whereas the phase angle and viscosity decreased with the increase in the loading frequency at a specified temperature [16,19–21].

Other important observations from Figure 1 to Figure 3 are the trends that show that the rheological characteristics of complex moduli (G^*), phase angles (δ), and viscosity (μ) are dependent on the type of waste toner used in the modified blends, as well as the concentration of the added toner, providing supporting evidence that variations in the nature of the bituminous-based binders, as well as additives and modifiers, influence the rheological and performance characteristics of modified blends [11,31].

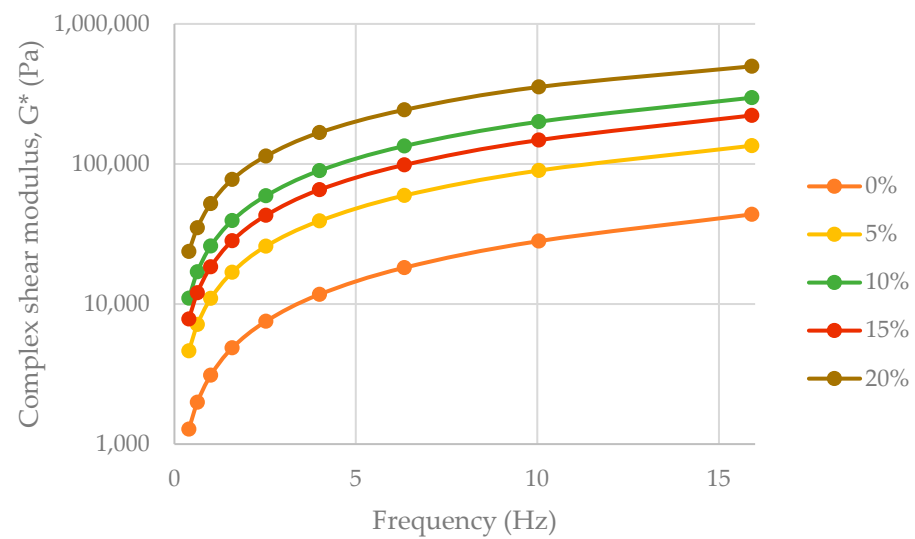


Figure 1. Representative plot of complex shear modulus (G^*) versus frequency at 60 °C at varying concentrations of TLA/TBP blend with toner D additive.

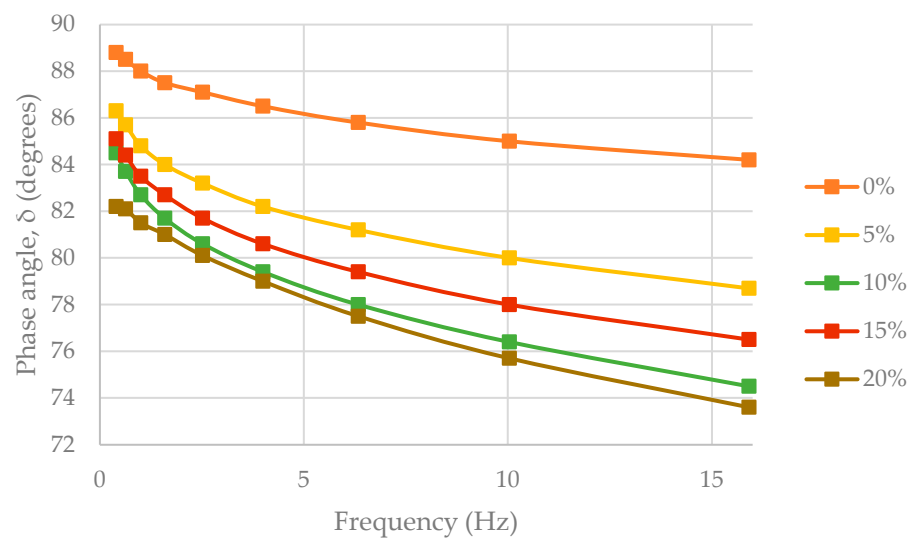


Figure 2. Representative plot of phase angle (δ) versus frequency at 60 °C at varying concentrations of TLA/TBP blend with toner D additive.

As seen in Figure 4, which depicts the variations of complex shear moduli (G^*) with the concentration of the added toners, the addition of waste toners generally resulted in an increase in G^* (stiffness) for all the modified bituminous blends, with the exception being the modified blend containing 5% of Toner B, which exhibited a 23% decrease in G^* . The maximum values of G^* (highest blend stiffness) for Toner A, B, C, and D were the 5%, 20%, 10%, and 20% blends, respectively. The greatest improvement in G^* was exhibited by Toner A; when the concentration was compared to the unmodified parent binder, the improvement observed at the 5% added toner was approximately 44%. The blend with the highest G^* (stiffness) was the blend containing 20% Toner B.

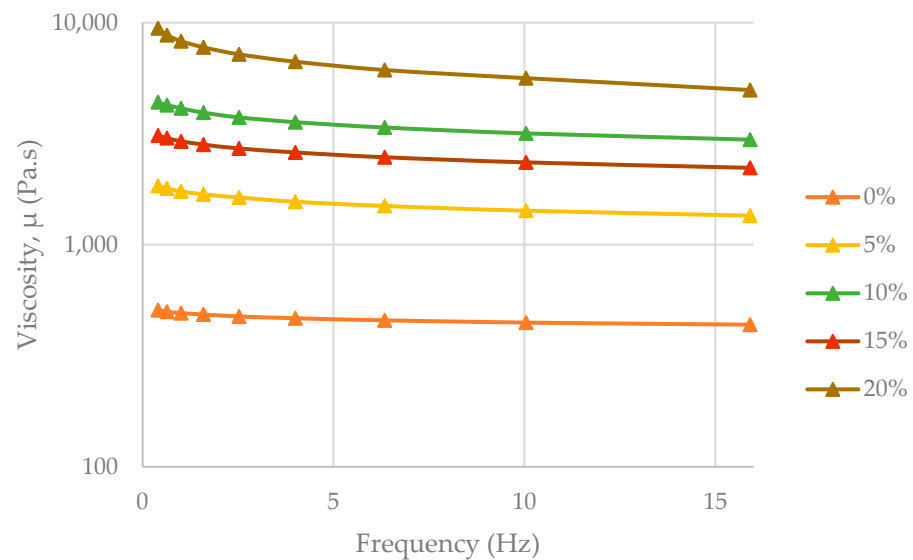


Figure 3. Representative plot of viscosity (μ) versus frequency at 60 °C at varying concentrations of TLA/TBP blend with toner D additive.

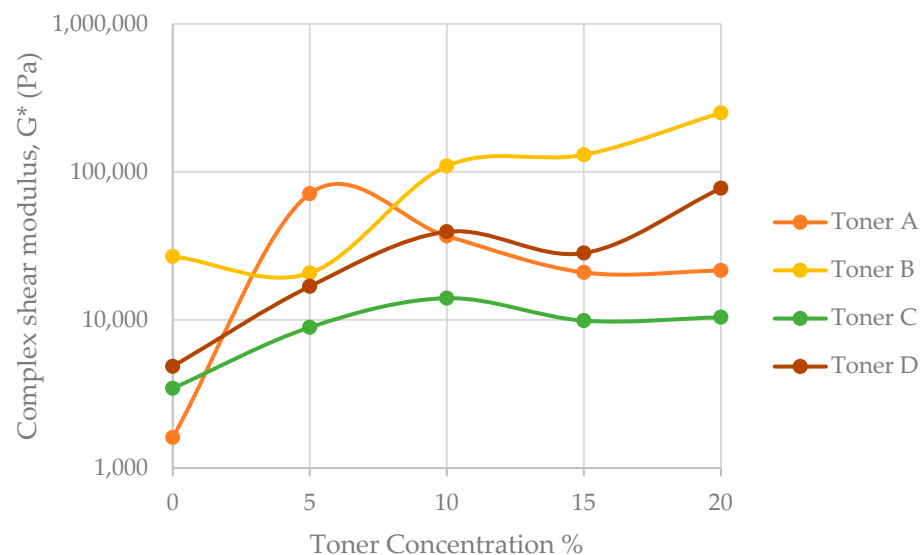


Figure 4. The influence of % toner concentration on complex shear modulus (G^*) for waste toners A, B, C, and D, measured at 1.59 Hz and 60 °C.

Figure 5 shows the variations of the phase angle (δ) with the concentration of the added toners measured at 1.59 Hz and 60 °C. The results indicate that all the toner-modified blends exhibited a higher degree of elastic properties compared to the unmodified TLA/TPB paving binder, as reflected by their relatively lower values of δ . The minimum values of δ (highest blend elasticity) were observed for Toner A, B, C, and D at 5%, 15%, 5%, and 20%, respectively. The greatest improvement in δ was exhibited by Toner B at the 15% added toner concentration, which was recorded and compared to the unmodified TLA/TPB paving binder, which showed a 78% decrease in δ . This blend exhibited the best elastic properties (lowest δ) of all the samples analyzed.

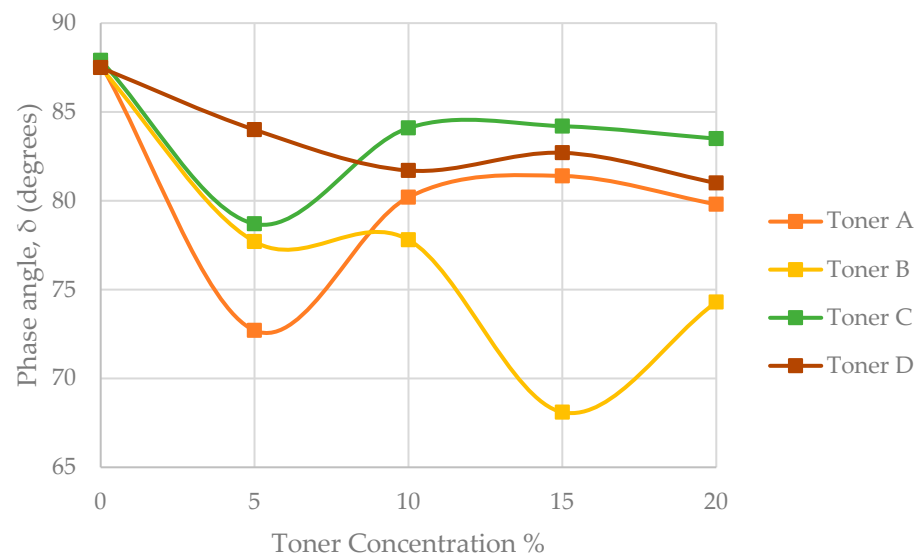


Figure 5. The influence of % toner concentration on phase angle (δ) for waste toners A, B, C, and D, measured at 1.59 Hz and 60 °C.

In Figure 6, the variations in the viscosity of the blends (μ) with % toner concentration of the four added waste toners measured at 1.59 Hz and 60 °C can be seen. The results indicate that all the toner-modified blends exhibited higher viscosities compared to the unmodified bituminous binders, except for the modified blend containing 5% Toner B, which exhibited a 23% lower viscosity. The minimum values of μ for Toner A, B, C, and D were the 5%, 20%, 10%, and 20% blends, respectively. The greatest improvement in μ was exhibited by Toner B at the 15% added toner concentration; when compared to the unmodified bituminous binder, a 23-fold increase in μ was recorded. This blend was the most viscous blend of all the samples analyzed.

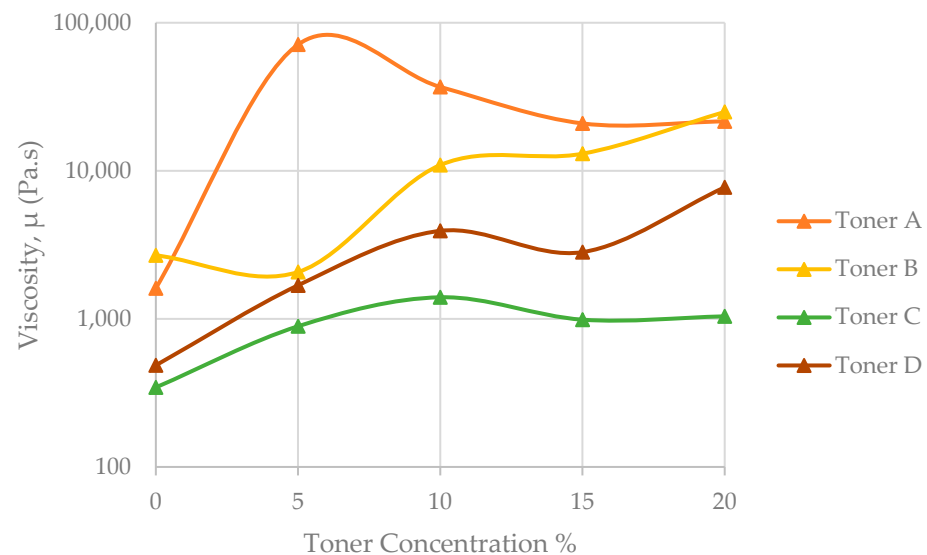


Figure 6. The influence of % toner concentration on viscosity (μ) for waste toners A, B, C, and D, measured at 1.59 Hz and 60 °C.

The temperature dependence of the various blends which exhibited optimized rheological performance of G^* , δ , and μ are shown in Figures 7–9, respectively. In each case, the performance of the optimized blend was compared with the unmodified TLA/TPB paving binder, as well as with pure TLA and TPB.

As can be seen in Figure 7, with regards to G^* , the pure TLA exhibited the best temperature susceptibility while pure TPB exhibited the worst. Compared with the unmodified TLA/TPB paving binder, all the modified TLA/TPB paving binders containing various amounts of toners A, B, C, and D exhibited superior temperature susceptibility. Of all the toner-modified blends, the blend containing 20% Toner B exhibited the best temperature susceptibility at temperatures below 75 °C, while the blend containing 20% Toner D exhibited the best temperature susceptibility at temperatures above 75 °C. All the samples analyzed followed the identical trend of a decrease in G^* as the temperature increased.

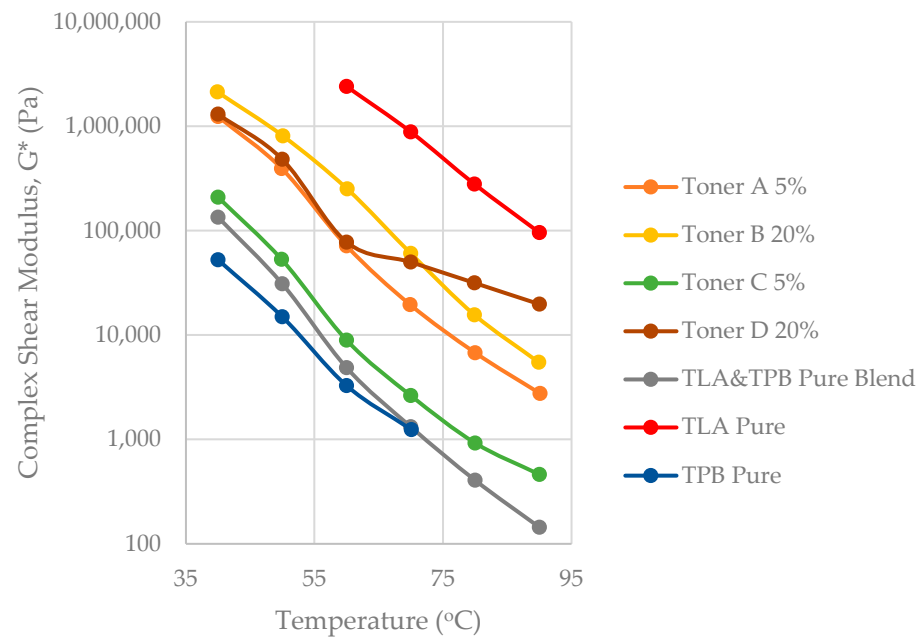


Figure 7. The effect of temperature on the complex shear modulus (G^*) of the various optimized toner blends, unmodified TLA/TPB paving binder, and pure TLA and TPB.

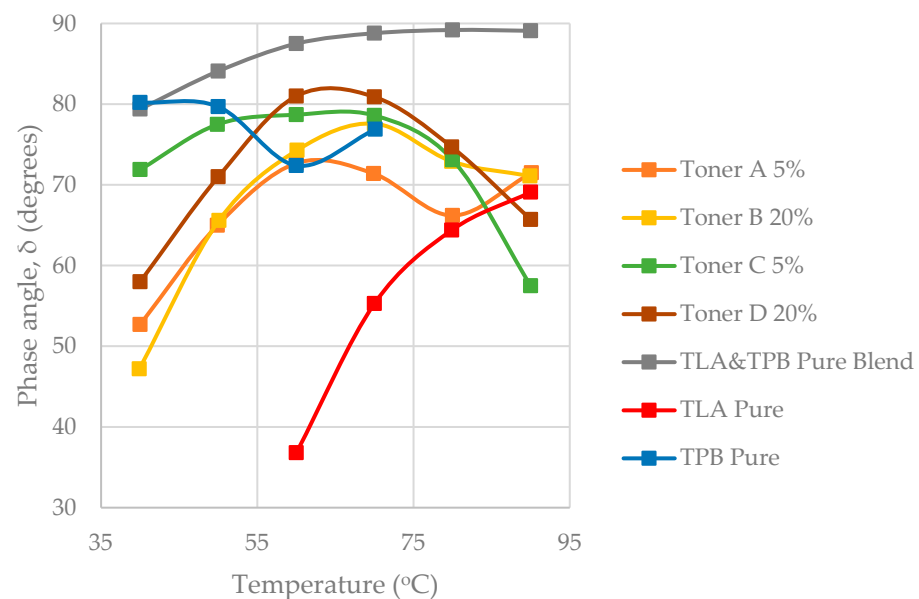


Figure 8. The effect of temperature on the phase angle (δ) for the various optimized toner blends, unmodified TLA/TPB paving binder, and pure TLA and TPB.

With regards to δ , and as shown in Figure 8, the toner-modified blends exhibited superior temperature susceptibility characteristics compared to the unmodified TLA/TPB

paving binder. The superior performance of the pure TLA was again observed; the material exhibited significantly lower δ values, except at 90 °C, at which the modified TLA/TPB paving binders containing 5% Toner C and 20% Toner D were elastically superior to the pure TLA.

The trends observed for the temperature susceptibility of the μ of the various samples were similar to those obtained for the G^* and are shown in Figure 9. However, in the case of viscosity (μ), the blend containing 20% Toner B exhibited the best temperature susceptibility at temperatures below 70 °C, while the blend containing 20% Toner D exhibited the best temperature susceptibility above 70 °C.

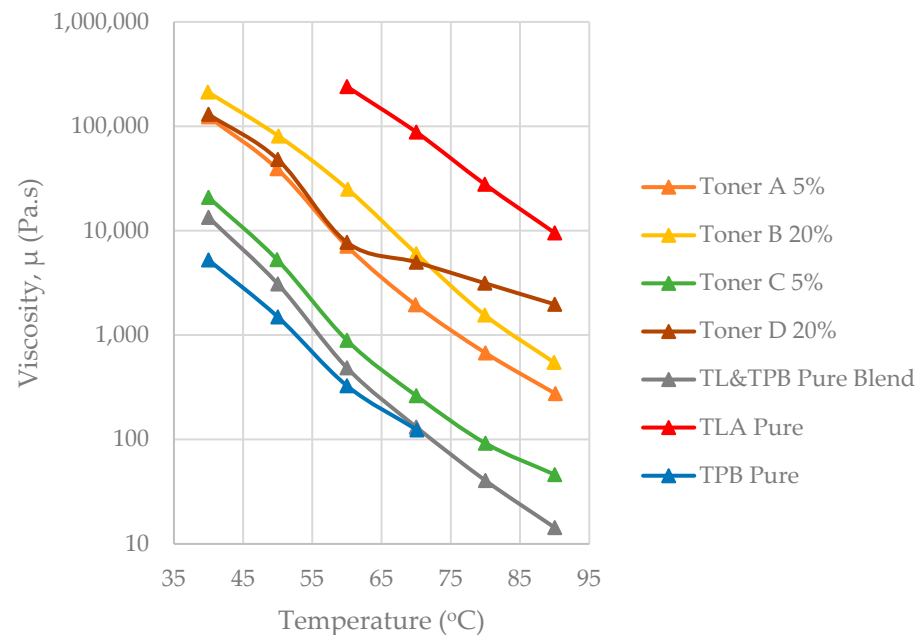


Figure 9. The effect of temperature on viscosity (μ) of the various optimized toner blends, unmodified TLA/TPB paving binder, and pure TLA and TPB.

The variations observed in the rheological values of G^* , δ , and μ when different types and quantities of toners were added to unmodified TLA/TPB paving binder can be attributed to compositional differences of both the parent bituminous base binder materials and the various toners used. The performance of a modified blend is dependent on the dispersion of the additive (toner) within the bituminous matrix; the degree of dispersion is dependent on the maltenes content and the chemical composition of the bituminous matrix, both of which are critical factors in the determination of the eventual additive-bitumen compatibility [14,32]. The superior physical properties of G^* , δ , and μ of TLA compared to the TPB and the other blends are indicative of the superior stiffness (rigidity) and elasticity of TLA; this can be attributed to the unique chemical composition of TLA [14,33]. TLA is renowned internationally as a superior quality asphalt (due to its consistent properties, resistance to cracking, stability, and durability), and it is considered a mandatory ingredient for paving in high-demand situations, such as those encountered in airport runways [13].

The Information gap due to the lack of scientific studies and data regarding the possibility of reusing waste toner in road paving applications in TT was addressed in this study. The information derived suggests dosages of specific waste toner products that, when added to the TLA/TPB paving binder, will produce modified blends of specific rheological characteristics that can suit road paving requirements and other specialized applications.

3. Materials and Methods

The asphaltic materials utilized in this study were obtained from the Lake Asphalt Company of Trinidad and Tobago (TLA) and the Petroleum Company of Trinidad and Tobago (TPB), with their properties listed in Table 1. The four (4) printer toners under investigation were obtained from the University of Trinidad and Tobago.

Table 1. Source and specifications of Trinidad Lake Asphalt (TLA) and Trinidad Petroleum Bitumen (TPB) samples utilized in this study [14].

	TLA	TPB
Source	Natural product mined from the Pitch Lake. Obtained from the Lake Asphalt of Trinidad and Tobago Limited	By-Product of the Petroleum Fractionation Process. Obtained from the Petroleum Company of Trinidad and Tobago Limited
Packing	Drum	Drum
Penetration at 25 °C (ASTM D5)	0–5	60–70
Specific Gravity (ASTM D70)	1.3–1.5 g/cm ³	1.00–1.06 g/cm ³
Softening Point (ASTM D36)	89–99 °C	225 °C
Flash Point (ASTM D92)	255–260 °C	49–56 °C

3.1. Materials and Preparation of Samples

The sample blends were prepared for rheological measurements in accordance with the recommended procedure by Polacco et al., 2004 [34]. In TT, as outlined by Charles, 2013 [35], in the Road Paving Inspection and Evaluation Report, the formulation utilized for road paving consists of limestone or gravel aggregate (92%) mixed with a bituminous material binder (8%). The bituminous material binder is a combination of an emulsified soft bitumen, such as TPB, and powdered TLA in a ratio of 3:1. For applicability with road paving procedures in TT and to obtain more meaningful results, a 3:1 ratio of TPB:TLA was utilized for the blending of the four (4) waste toners A, B, C, and D, which were added in the range 0–20 wt%; a graphic representation of the waste toners used in the study is shown in Figure 10. The unmodified TLA/TPB paving binder, consisting of a mixture of 25% TLA and 75% TPB relevant to road paving applications, was utilized as the base sample for this study [35]. Approximately 6g of the unmodified TLA/TPB paving binder was measured and transferred into a 50 cm³ aluminum can, where the temperature was raised to 200 °C using a thermoelectric heater, Thermo Scientific Precision (Model 6555). A high shear digital mixer, IKA (Model RW20D), was submerged in the can and set at a speed of 3000 rpm. The temperature was maintained at 200 ± 1 °C, while Toner A was gradually added by weight % measure. The percentages and concentrations analyzed were selected based on the study conducted by Yildirim et al., 2004 [11]. These steps were repeated for the other three (3) toners under analysis. A breakdown of the concentrations of the modified sample blends can be seen in Table 2.

After homogeneity of the blend mixtures was obtained, each was stored in a desiccator under static conditions, with an oxygen-free environment, for 24 h of curing. Prior to the rheological analysis, the cans were removed from the desiccator, remixed with the high shear mixer, and cast into ring stamps of dimensions 25 mm in diameter and 1 mm. The temperature of the samples was reduced to room temperature and stored in a Fisher Isotemp freezer at −20 °C before testing began.

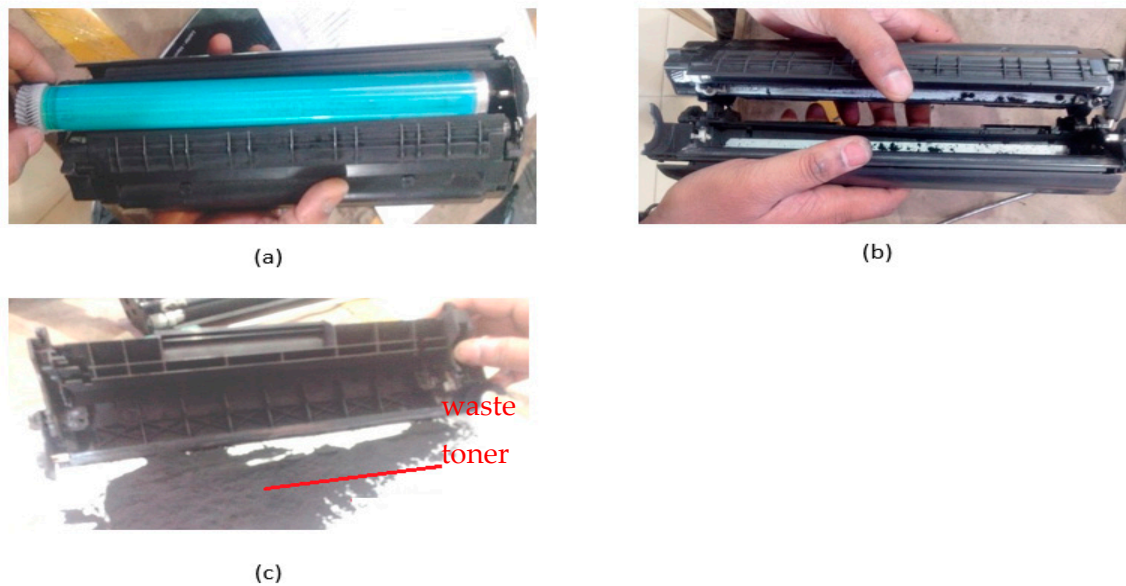


Figure 10. The following depicts the process, which was adopted from a previous study, that was undertaken to obtain the waste toners that were added to the TLA/TPB paving binder. (a) Image of a sample printer cartridge; (b) Deconstruction of printer cartridge to access waste toner powder; (c) Example of the waste toner product [36].

Table 2. Concentration of sample blends utilized in this study.

Mass of Sample 6 g						
% Toner Required in Blends	Asphalt Blend					
	Toner A			Toner B		
	Actual Mass of TLA/TPB Added (g)	Actual Mass of Toner A Added (g)	Actual % Toner A	Actual Mass of TLA/TPB Added (g)	Actual Mass of Toner B Added (g)	Actual % Toner B
0.00	6.0064	0.0000	0.00	0.0000	0.0000	0.0000
5.00	5.7067	0.3061	5.01	5.7022	0.3121	5.20
10.00	5.4168	0.6076	10.13	5.4057	0.6009	10.01
15.00	5.1128	0.9052	15.01	5.1070	0.9034	15.06
20.00	4.8056	1.2075	20.13	4.8046	1.2059	20.10
% Toner Required in Blends	Toner C			Toner D		
	Actual Mass of TLA/TPB Added (g)	Actual Mass of Toner C Added (g)	Actual % Toner C	Actual Mass of TLA/TPB Added (g)	Actual Mass of Toner D Added (g)	Actual % Toner D
	Actual Mass of TLA/TPB Added (g)	Actual Mass of Toner C Added (g)	Actual % Toner C	Actual Mass of TLA/TPB Added (g)	Actual Mass of Toner D Added (g)	Actual % Toner D
0.00	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
5.00	5.7031	0.3010	5.02	5.7005	0.3082	5.14
10.00	5.4059	0.6039	10.07	5.4042	0.6016	10.03
15.00	5.1064	0.9023	15.04	5.1073	0.9108	15.18
20.00	4.8030	1.2036	20.06	4.8097	1.2007	20.01

3.2. Sample Characterization

The ATS RheoSystems Dynamic Shear Rheometer (Viscoanalyzer DSR) was used to determine the rheological properties of the asphaltic blends, which were tested under the strain-control mode to ensure that the parameters were kept within the linear viscoelastic range [15,18–24]. The DSR testing was software-controlled using plate-plate configuration test geometry. Experiments were conducted within a 0.1 to 15.91 Hz frequency and a

40 °C and 90 °C temperature range. The data obtained at the different oscillating shear frequencies and temperatures were stored and analyzed using the Viscoanalyzer software (RheoExplorer V540P9, ATS RheoSystems, New Jersey, NJ, USA).

4. Conclusions

The results of this study demonstrated that the addition of these waste materials to a TLA/TPB paving binder resulted in improvements in the rheological values of G^* (stiffness), δ (elasticity), and μ (viscosity). These improvements are summarized as follows:

- The toner-modified blend containing 20% Toner B exhibited the highest G^* , while the blend containing 15% Toner B exhibited the highest elasticity and viscosity;
- All of the modified TLA/TPB paving binders exhibited superior temperature susceptibility;
- The blend containing the 20% Toner B exhibited the best temperature susceptibility at temperatures below 75 °C, while the blend containing 20% Toner D exhibited the best temperature susceptibility at temperatures above 75 °C;
- The same two modified blends (20% Toner B and 20% Toner D) exhibited the best temperature susceptibility for viscosity (μ) at temperatures below 70 °C and above 70 °C, respectively;
- In terms of elasticity (δ), at 90 °C the modified paving binders containing 5% Toner C and 20% Toner D were elastically superior to the world-renowned TLA.

This study offers evidence that blending waste toner with TLA/TPB paving binders is an innovative and sustainable option for disposal. Adoption of this strategy will also facilitate associated positive impacts, such as cost savings on asphalt, improvement of local road surfaces, innovation and entrepreneurship, and increased employment opportunities with potential foreign income.

Author Contributions: Conceptualization, R.M.; methodology, S.R., R.M., S.M. and N.S.; software, S.R., R.M., S.M. and N.S.; validation, R.M. and N.S.; formal analysis, S.R., R.M., S.M. and N.S.; investigation, S.R., R.M., S.M. and N.S.; resources, S.R., R.M., S.M. and N.S.; data curation, S.R., R.M., S.M. and N.S.; writing—original draft preparation, S.R. and S.M.; writing—review and editing, R.M. and S.M.; visualization, S.R. and S.M.; supervision, R.M. and N.S.; project administration, R.M. and N.S. All authors have read and agreed to the published version of the manuscript.

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