Recycling of banana production waste bags in bitumens: A green alternative.

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ABSTRACT

In tropical countries that produce bananas, the disposal of the bags that are used to cover the bananas is troublesome. Hundreds of tons of this material are disposed every year as a result of the banana production industry, with direct impact on the environment and the ecology. Costa Rica and Colombia are countries with high biodiversity and with great interest in protecting the environment. However, large percentage of their territories is cultivated with bananas and as a result of this industry, significant amounts of non-biodegradable waste from the fruits' protective bags is generated.

The use of modified bitumen is widespread. The modifiers are added to the bitumen with the goal of improving some of the bitumen's properties and its performance through its service life. As part of this research, a waste from the banana production industry is used to modify bitumen: polyethylene bags that are used to cover the fruit to prevent plagues and to increase the overall quality of the product. The bag is introduced to the bitumen as cutouts with 4 cm faces in a 2 hour homogenization process at 160°C. The homogenization process exhibited proper distribution of the modifier within the bitumen, as measured by the Raman spectroscopy. The properties of the bag were evaluated by means of physical-chemical and calorimetric tests, and the performance of the modified bitumen was analyzed by means of creep, multi stress creep recovery, and fatigue tests. It was observed that the bag initiates its degradation at 150°C and approximately at 450°C degrades completely. At 325°C chlorinated gases and sulfonated chloride are released.

The original bitumen corresponds to an AC-30 with a PG grading of PG 64-22. It was found that the modifier reduces the deformation by 50% after 250 loading cycles of repeated creep and is appropriate for use under normal traffic loading. With the addition of the banana bag to the bitumen, the solid residue can be used at a rate of approximately 4 kg per cubic meter of asphalt. Additionally, the release of toxic chlorinated gases and sulfonated chloride into the atmosphere is minimized.

Keywords: Modified binders, performance testing, banana bags

1. INTRODUCTION

The area of Costa Rica consists of 52.000 km^2. In 2010, the land destined for banana production reached 43.031 ha (CORBANA, 2011). Similar is the case of Colombia where 47.108 ha are destined for banana production (Ministerio de Agricultura de Colombia, 2011). At a global scale, in 2009 the largest producer of banana was India, followed by the Philippines, China, Ecuador, Brazil, Indonesia, Tanzania and Guatemala. Costa Rica ranks in 9th place in production of banana (2.365.471 million tons), while Colombia ranks 11th with a production of 2.020.390 million tons [1]. To ensure a better production of the fruit, the banana bunch is wrapped in a polyethylene bag. This practice ensures that the fruit grows in optimal sale conditions, since the bag allows the fruit to develop a microclimate that protects the fruit from the sun, pesticides, and the insects that affect the plantation.

The problem with this plastic is that it is not biodegradable, and as such cannot be disposed directly into the environment. Additionally, the bags are impregnated with chlorpyrifos to protect the fruit from insects. This constitutes a major drawback in the recycling of the bag for applications destined for human consumption [2]. Studies have indicated that a 1 ha of banana plantation produces 1,5 tons of non-biodegradable material. This is a serious problem for the banana industry and for the environment (CENIBANANO Colombia) since it is currently being disposed, with no previous treatment, in open pits, chemical reduction, reutilization, and recycling applications, or worst of all, incineration [3].

The bitumen is a material that is derived from petroleum with a large quantity of hydrocarbons. Normally, the hydrocarbons are heavy, dark colored, soluble, and may be liquid or solid. The bitumen is a material with viscous or viscoelastic behavior depending on the temperature: at low temperatures it behaves as a solid material (elastic) and as the temperature increases it behaves as a fluid (viscous). Due to this chemical complexity, currently the best model to describe it is a colloidal model where the solid particles (asphaltenes) are dispersed in an oily liquid matrix (maltenes) [4].

The composition of the bitumen can be explained in terms of relative fractions of saturates, aromatics, resins, and asphaltenes. Corbett [5] stated that the asphaltenes behave as thickeners, the fluidity is provided by the saturates and the aromatics, the resins provide the ductility to the binder, and the saturates and the aromatics, in combination with the asphaltenes, facilitate the flow of the bitumen. The properties of the bitumen affect directly the performance of the asphalt. Consequently, a low viscosity bitumen subject to high service temperatures can result in development of permanent deformation, while a high viscosity bitumen subjected to low service temperatures hardens the asphalt and promotes the formation of cracks (fatigue cracking). The bitumen has its deficiencies and limitations, which can be observed in the field as damage due to heavy loads, permanent deformation, and fatigue cracking. All of this highlights the importance of following adequate design requirements, establishing proper quality control procedures to the production of bitumen, and modifying its properties with additives.

There has been past research in the use of recycled polyethylene as a bitumen modifier, and there are several publications in the subject, mainly regarding the use of crumb rubber [6,7,8,9,10]. Nonetheless, it seems that only in few cases has the modification been implemented for field use, and mostly it has been with crumb rubber specifically. The authors believe that this has probably been the case because of the limited recycling applications associated with used tires, and more specifically in the case of bitumen modification, because of the workability difficulties associated with the modified material. However, as will be shown in the following paper, the incorporation process that was used with the proposed modifying material is relatively simple, should not require upgrades for mixing the material in an asphalt plant, and the performance shown by the modified bitumen and the asphalt produced with it is promising.

1.1 Objective

The first step in the current research project consisted of envisioning the use of polyethylene bags that are used in the production of banana as a possible modifier for conventional bitumen LPI-03 (AC-30). This paper discusses the properties of the material and the possibility of re-using and re-incorporating it in the production of a new material. Additionally, the changes in performance of the bitumen as a result of incorporating the modifier are evaluated to assess the feasibility of using the modified bitumen in road applications.

2. MATERIALS AND ANALYSIS METHODS

The tests to characterize the materials were performed at the National Laboratory for Materials and Structural Models (LanammeUCR, Universidad de Costa Rica), in collaboration with the National Nanotechnology Laboratory (LANOTEC) and the Polymers Laboratory at the National Learning Institute (INA).

The bitumen that was used is an AC-30 obtained from RECOPE (Costa Rican Petroleum Refinery). This is the most common bitumen used in Costa Rican roads. The waste polyethylene bags were provided by CORBANA (Costa Rica's National Banana Corporation).

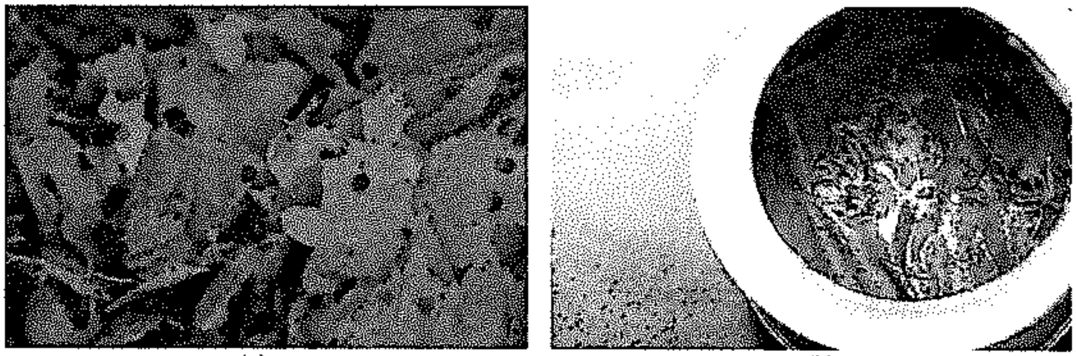
2.1 Banana Bag Preparation and Incorporation

The experiment consisted in preparing modified bitumen using banana bags. However, note that because of the presence of insecticides (eg. chlorpyrifos), the process of modifying the bitumen with this material is more complex than if standard polyethylene bags were used. This is the case since special care has to be placed in the mixing process.

Large doses of chlorpyrifos can be highly toxic to humans. Consequently, the bags that were recovered from the banana plantations and brought to the laboratory were initially blown with air to remove solid particles attached to the surface of the bags. After removing the solid particles, the bags were washed three times with tetrahydrofuran initially and then acetone for eliminating as much of the organic compounds as possible from the treated bags. After each of the wash cycles, the bags should be allowed to drain until the solvent is released from the bags. Then these organic compounds (chlorpyrifos) can be recovered from the solvent by simple distillation.

An additional step in the treatment process can correspond to washing the bags with soap and water to ensure any remains of the tetrahydrofuran is removed from the bags.

Finally, the bag samples were cut using a guillotine into squares with sides measuring 4 cm. The recommended size of the particles was found by trial and error since it was found that if large particles are used, some of the polyethylene will form capsules that develop a rigid outer crust that will difficult the mixing process with the bitumen (Figure 1).



(a) (b)

Figure 1. (a) Processed Banana Bags and (b) Formation of Polyethylene Capsules (due to Large Bag Particles)

During the modification process, the polyethylene was added, using a 3% dosage, by means of a low shear stirrer at 160°C for 2 hours. The 160°C is optimal for the mixing process since, as will be shown later, it was found that any remaining traces of chlorpyrifos will be degraded at approximately 150°C ensuring that the substance will not be present in the modified asphalt.

Special care needs also be placed in the previous modification process. Because polyethylene is less dense than the bitumen (polyethylene density is approximately 0,92 g/cm^3, while bitumen density was 1,03 g/cm^3), and because the cut bags are flat in one dimension, the shear blade used in the low shear stirrer has to be placed very close to the surface of the bitumen so that the vortex that is generated can be capable of introducing the plastic material in the bitumen. This is important since the polyethylene will tend to float over the surface of the bitumen.

2.2 Material Analysis

Finally, both the polyethylene from the bag and the modified bitumen were evaluated. The bitumen was characterized using the SUPERPAVE methodology, based on the PG grading of the material before and after the modification. In the case of the polyethylene, the material was analyzed based on its calorimetric curve, thermal transition, distribution in bitumen after modification, overall quality of the material, and the possibility of recycling the material to obtain other products.

The analysis of the polyethylene bag was performed by means of Raman Spectroscopy, Thermogravimetric Analysis (TGA), Differential Scanning Calorimeter (DSC) Sweep, and Fourier Transform Infrared (FTIR) Spectrometry. In the case of the material composition analysis and performance of the original and modified bitumen, Iatroscan Chromatography, Atomic Force Microscopy (AFM), and Fourier Transform Infrared (FTIR) Spectrometry were used. For analyzing the performance of the bitumen during its service life, the bitumen was aged using the Rolling Thin Film Oven (RTFO) and the Pressure Aging Vessel (PAV), and was analyzed using a Dynamic Shear Rheometer (DSR) under repeated creep (NCHRP 459, Appendix 4), Multiple Stress Creep Recovery (MSCR) (AASHTO TP70-07), and fatigue under controlled displacement.

Finally, to complement the bitumen analysis, initial performance testing on the modified and non modified asphalt was performed. The properties that were measured were Indirect Tensile Strength (ITS) with and without conditioning, resilient modulus, and rutting resistance as measured by the Asphalt Pavement Analyzer (APA). All the samples were prepared according to Superpave design and compaction procedures.

3. RESULTS

3.1 Characterization of the banana plastic bags

The region shown on the figure has been analyzed by means of Confocal Raman, with an alpha300AR, WITEC Raman spectrometer. The difference on the color intensity corresponds with only one chemical component; the colors of the spectra correspond with the color of the image.

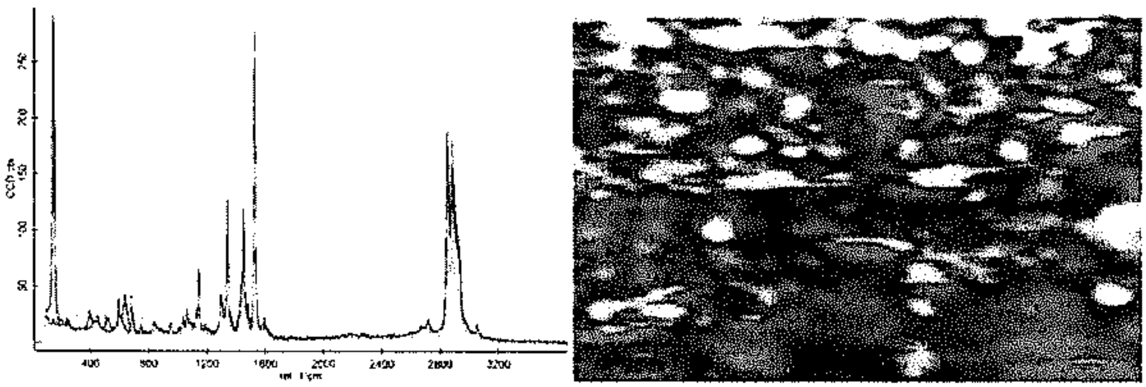


Figure 2. Average Raman Spectrum for the Banana Bags

The Raman Spectrum on Figure 2 is typical for the [-CH2- CH2-]n group which might suggest that the material is a simple polymer. The most important bands are the ones located at the spectrum at 1450 1/cm and at 2850-3000 1/cm where the flexures for the saturated -CH_2 groups are found at 1450 1/cm and the -CH tensions of the saturated -CH_2 and -CH_3 are found at 2850-3000 1/cm.

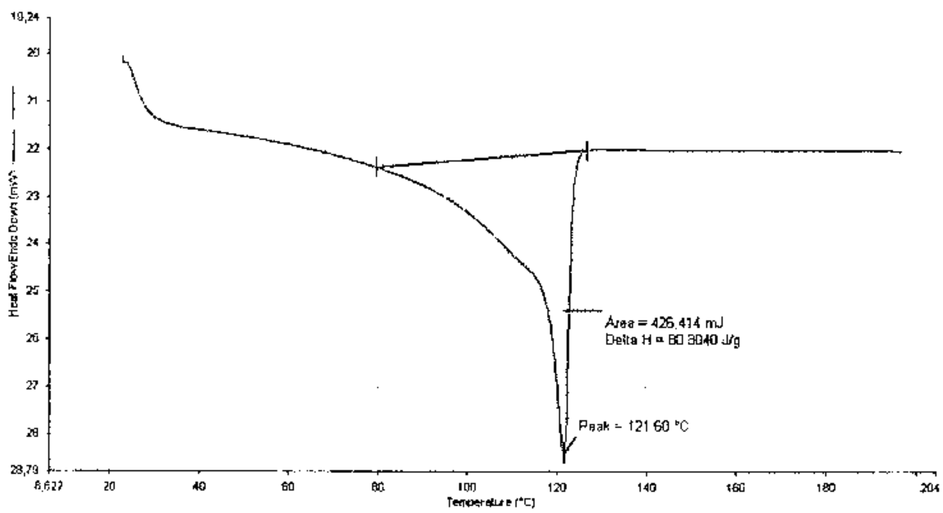


Figure 3. DSC Analysis for the Banana Bags

According to the Differential Scanning Calorimetry Analysis, performed on a Jade Perkin Elmer DSC, by observing the sample behavior due to the change on temperature from 25°C up to 200°C, it was identified that the material that composes the bag is a high density polyethylene, because its fusion point is found between 120-136° C (Figure 3); although initially it was thought to be a low density material, as commented on Figure 2. To confirm that it is a polyethylene, a thermogravimetry test was performed to measure the effects of material degradation due to heat. The analysis was run with a TA-Instruments Q5000 TGA, performed at temperatures ranging from 20 to 800°C, shows that the material initiates its degradation at 150°C. Between this temperature and 325°C, a 6.6% loss of an initially unidentified material was observed.

Additionally, Figure 4 indicates that total degradation of the material occurs at 450°C. The material degradation curve follows the same behavior of other polyethylenes evaluated with this same technique.

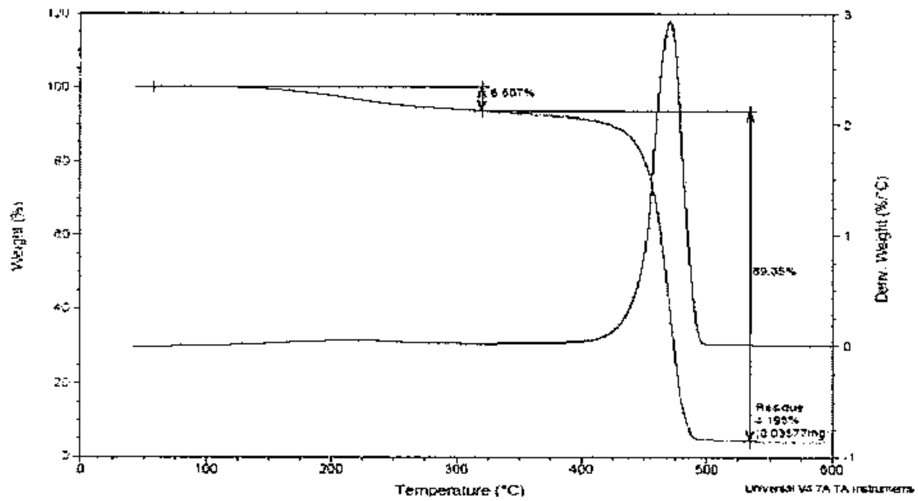


Figure 4. TGA Analysis for Banana Bags

With the objective of identifying the degraded material between 150°C and 325°C on the TGA analysis, another test was performed with a Pyris 6 Perkin Elmer TGA, connected to a Spectrum One FT-IR Perkin Elmer Gas Spectrometer, which was used to analyze de gases produced by the polyethylene degradation. Figure 5 shows the FT-IR Spectrum which consists mostly of [-CH_2- CH_2-] and CH_3 groups. The most important bands are the ones located at the spectrum 1000 1/cm and at 2850-3000 1/cm where the flexures for the saturated -CH_2 groups are found at 1000 1/cm and the -CH tensions of the saturated -CH_2 and -CH_3 are found at 2850-3000 l/cm (Figure 5). As a result of the decomposition, the spectrum was used to identify chloride and chloro-sulphide groups, which are added also at the fumigation process at the plantation. This are the compounds released to the environment when the incineration of the polyethylenes is performed.

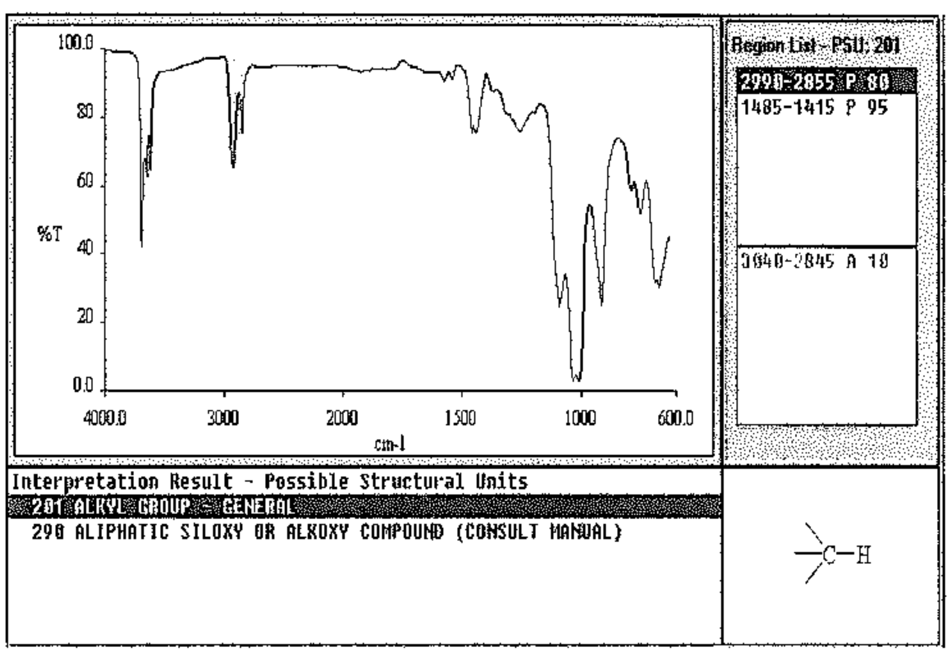


Figure 5. Gas Spectrum Produced During the Decomposition of Banana Bags

3.2 Characterization of the Virgin and Modified Bitumen

The results of the chromatography made with the IATROSCAN MK-6 chromatographer show that the SARA fractions of the virgin bitumen are distributed as follows: Saturated 5.4%, Aromatics 35.3%, Resins 41.4% and Asphaltenes 17.9%. According to this information, the stability index is 0.3. The binder characterization was performed by the SUPERPAVE methodology for the original bitumen; the short term aged bitumen (RTFO) and the long term aged bitumen (RTFO+PAV). Table 1 shows the viscosity of both the original and modified bitumen. Additionally, Table 2 shows the performance results of the non-modified bitumen, the bitumen modified with banana bags, and modified with other materials for the 3 different aging conditions. These tests were performed on a TA- Instruments DSR, AR-G2 model.

Table 1. DSR Viscosity for Modified Bitumen at 60°C

Bitumen type	Viscosity (Poises)
LPI-03 (AC-30)	2854
SBR(A)	4312
Banana Bag	5039
SBR(B)	5698
SBR(C)	5542
SBS(A)	8054
SBS(B)	6069

Table 2. Rheology Analysis of the Modified Bitumen

G*/senδ ≥ 1 kPa, Original Bitumen			
Test Temperature	64 °C	70 °C	76 °C
AC-30	1.78	0.89	0.47
SBR(A)	2.79	1.42	0.75
Banana Bag	3.11	1.53	0.80
SBR(B)	3.72	1.97	1.06
SBR(C)	3.00	1.52	0.81
SBS(A)	5.39	2.72	1.44
SBS(B)	3.60	2.04	1.20
G*/senδ ≥ 2,2 kPa, RTFO-aged Bitumen			
Test Temperature	64 °C	70 °C	76 °C
AC-30	5.97	2.94	1.47
SBR(A)	9.74	4.92	2.55
Banana Bags	11.31	5.55	2.77
SBR(B)	-	-	-
SBR(C)	9.23	4.87	2.60
SBS(A)	18.80	10.02	5.24
SBS(B)	13.84	8.13	4.89
G*·senδ ≤ 5 MPa, PAV+RTFO-aged Bitumen			
Test Temperature	Intermediate Temp. (°C)		MPa
AC-30	22		4.14
SBR(A)	22		4.60
Banana Bags	22		4.14
SBR(B)	-		-
SBR(C)	13		4.20
SBS(A)	25		4.06
SBS(B)	22		4.89

The performance of the modified binder was also evaluated by means of the repetitive creep test; the results are shown on Figure 6. This test was performed at 70°C and the sample was subjected to 250 load cycles. The applied shear stress was 100 Pa, the defined creep time was 1,0 s and the recovery time was 9,0 s according to specifications.

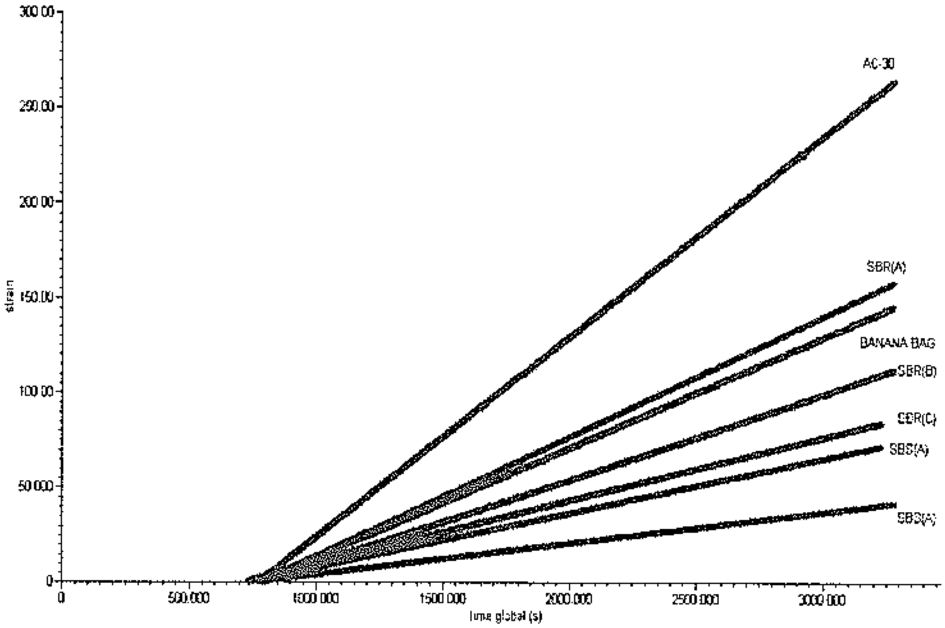


Figure 6. Repeated Creep Comparison for the Different Modified Bitumens

Table 3 shows the performance of the bitumen modified with the different materials, when analyzed with the Multiple Stress Creep Recovery (MSCR) test for permanent deformation.

Table 3. Results of the MSCR (ASTM – D7405) for the Different Bitumens

Polymer	$J_{NR@3.2KPa}$	$(J_{NR@3.2KPa}-J_{NR@0.1KPa})/J_{NR@0.1KPa}$	Condition
LPI-03 (AC-30)	7.344	0.22	NA
SBR (A)	3.896	0.32	Standard Traffic
BANANA BAG	3.180	0.26	Standard Traffic
SBR (C)	3.284	0.37	Standard Traffic
SBS (A)	1.575	0.26	High Traffic
SBS (B)	0.510	0.33	Very High Traffic

Fatigue analysis was also performed using the DSR. The analysis was performed at 22°C using an angular frequency of 10 rad/s, under strain controlled mode (10%). It was found that the dissipated energy of the modified material seems to be more stable than that of the virgin material. Although at the beginning of the lifespan the modified material shows less dissipated energy, after 12.000 cycles the dissipated energy for both the virgin and the modified bitumen is almost equal (Figure 7). This indicates that in the long term, fatigue resistance of both the materials should be fairly similar. This is consistent with the information that was obtained from the RTFO+PAV aged samples (Table 2) that indicates that the $G^{*}\cdot sen\delta$ for both materials are relatively close and therefore fatigue performance should also be similar for both materials.

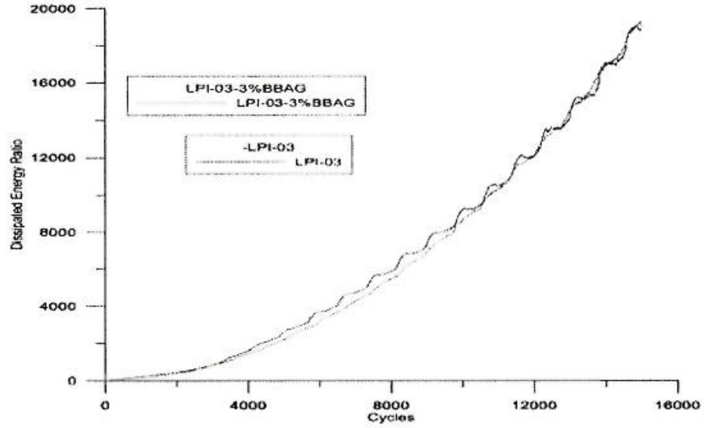
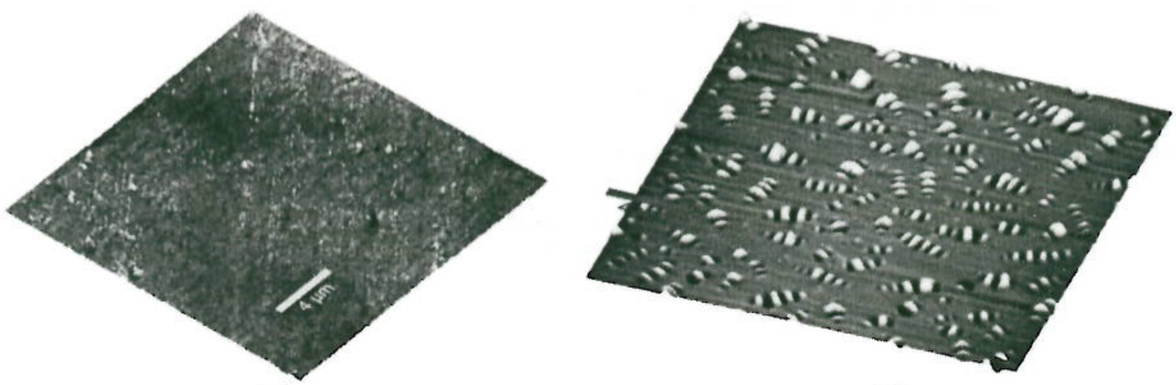


Figure 7. Dissipated Energy for the Virgin and Modified Bitumen under Fatigue

The analysis of the modifier distribution on the bitumen was performed using Atomic Force Microscopy (AFM). This test shows information on the topography and the tridimensional environment of the sample, and also on the roughness of the material. Figure 8(a), shows the topography of the non-modified AC-30 binder and Figure 8(b), shows the same bitumen modified with the SBS compound. The images of the distribution of the banana bag modified bitumen are shown on Figure 9.



(a) **(b)**

Figure 8. Bitumen AFM Topography for (a) AC-30 and (b) AC-30 modified with SBS(A)

Figure 9. AFM Topography for AC-30 Modified with Banana Bags

Finally, infrared spectroscopy was performed with a Thermo FT-IR Spectrophotometer. For the FT-IR bitumen spectrum with no modification, the characteristic bands are: 2851-2920 cm^{-1} high intensity CH_2 bands which come from the saturated hydrocarbons, 1455 cm^{-1} medium intensity and 1375 cm^{-1} medium intensity bands can be ascribed to CH_2 and CH_3 bending frequencies, respectively (Figure 10).

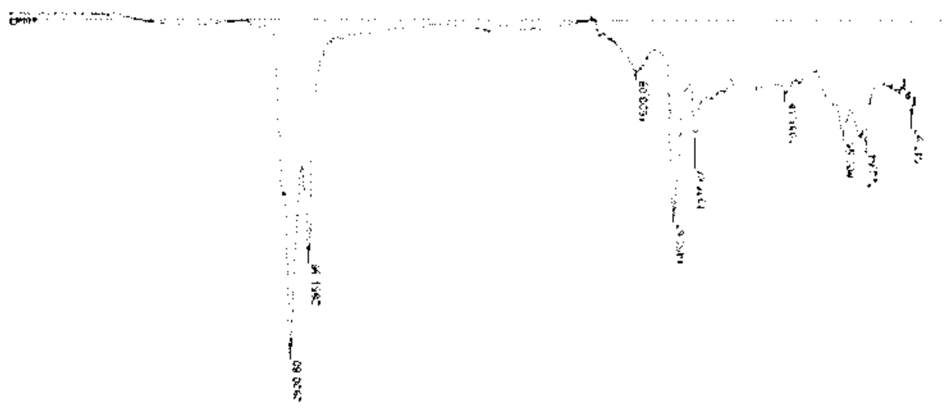


Figure 10. IR Virgin AC-30 Bitumen Spectrum

The IR spectrum corresponding to the banana bag modified bitumen exhibits the characteristic bands same as the ones for the virgin bitumen but with an intensity increment as follows: 2851-2920 cm^{-1} CH_2 bands featuring saturated hydrocarbons, and the corresponding bands being assigned to CH_2 and CH_3 bending frequencies (Figure 11).

Interestingly, when compared to the unmodified bitumen (Figure 11), the banana bag modified bitumen results in small increases to the 2851-2920 cm^{-1} intensity CH_2 band. However, when the results are compared to the rheology analysis, it is clear that the viscosity of the original and modified bitumen is significantly different (77% increase in viscosity at 60°C). This finding can in part also be attributed to the change in the 1600 cm^{-1} medium intensity C=C from highly substituted aliphatic rings present as resins and asphaltenes; and consequently it is expected that an increase in these components be associated with an increase in the viscoelastic response of the bitumen.

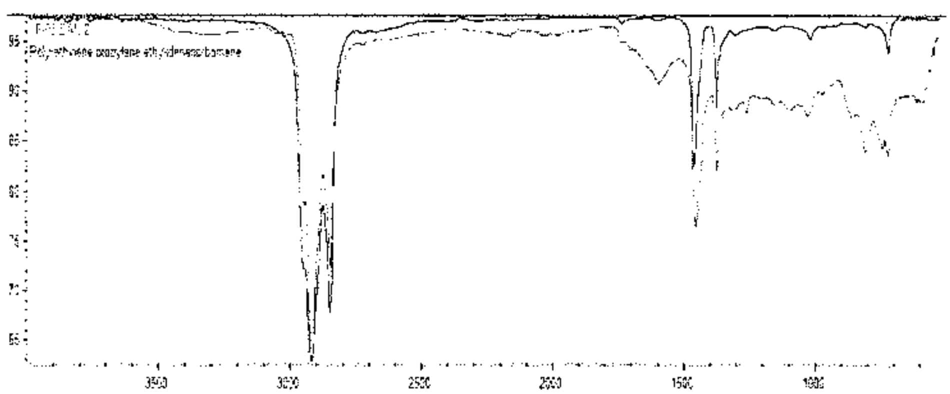


Figure 11. IR Spectra Comparison for Virgin Bitumen and the Banana Bag Modified Bitumen

3.3 Asphalt Performance

In order to assess the effect of the bitumen modification with banana bags on the asphalt performance, samples were prepared following the Superpave mixture design methodology. The aggregate gradation to be used in the design is the one shown in Figure 12, and corresponds to a typical plant gradation used in Costa Rica. Only one asphalt mixture was designed: the one for which the unmodified bitumen was used. It was found the optimum bitumen content corresponded to 6,6% (percentage of total aggregate weight). In order to evaluate the effect of the modification on the asphalt performance, the asphalt that was produced with the modified bitumen was also prepared using 6,6% of bitumen.

It is clear that adding a modifier to the bitumen will most likely shift the optimum bitumen content. However, all mixtures were designed to one binder content to control for every factor and isolate the effect of the modifier on asphalt performance.

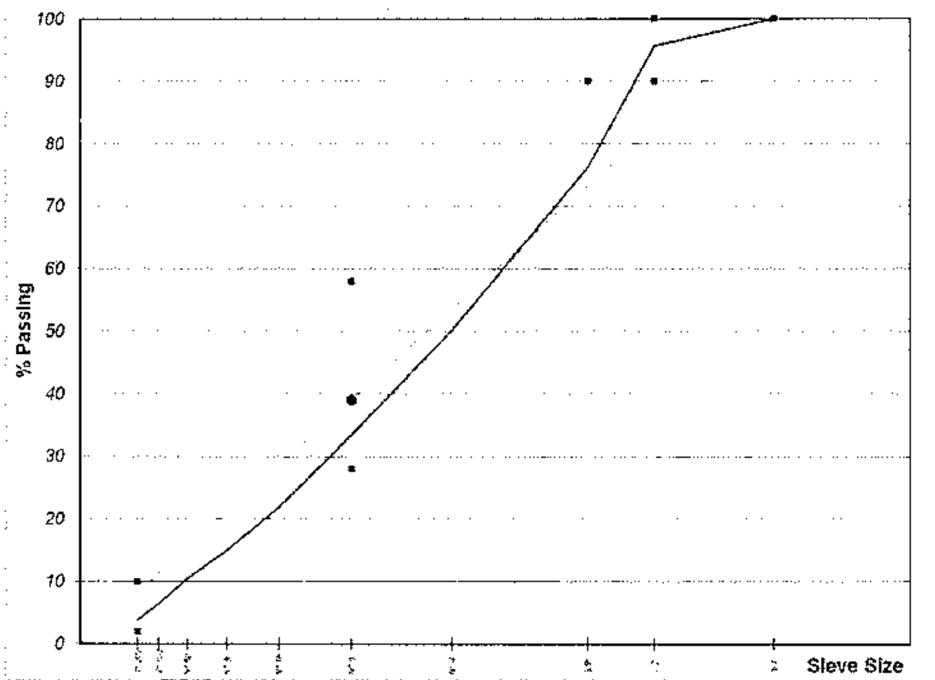


Figure 12. Aggregate Gradation used for Asphalt Preparation

Because the most common cause of pavement failures in Costa Rica is associated with moisture damage, Indirect Tensile Strength (ITS) was calculated for both the modified and non modified asphalt. The results are shown in Table 4. It can be observed from the table that the binder modification increases moisture damage resistance as measured by the Tensile Strength Ratio (TSR). When comparing the asphalt modified with banana bags and the asphalt modified with SBR, very small difference in retained strength were observed; both retain most of the strength from the dry condition after conditioning.

Table 4. Tensile Strength Ratio results

Bitumen Type	ITS at 25°C (N)		TSR
	Dry	Conditioned	
AC-30	11660	10459	96,07%
SBR(A)	16215	15036	99,27%
BANANA BAGS	16446	16428	100,00%

In order to assess the rigidity of the asphalt, the resilient modulus was measured for all of the evaluated asphalts. Testing was performed at 25°C. The results are shown in Table 5 and indicate that the resilient modulus increases by almost 65% when comparing the unmodified asphalt to the asphalt modified with banana bags. The increase is significantly larger than the one observed when modifying the asphalt with SBR polymer.

Table 5. Resilient Modulus results

Bitumen Type	MR at 25°C (MPa)
AC-30	3515
SBR(A)	4210
BANANA BAGS	5792

Testing with the Asphalt Pavement Analyzer (APA) was also performed on the different asphalt mixtures to assess rutting resistance since the bitumen results indicated that modifying the bitumen with banana bags increased rutting resistance as measured by repeated creep and MSCR testing. The APA results are shown on Figure 13. It is interesting to note that the relative performance ranking that was observed at the bitumen level remains at the asphalt level since the banana modified bitumen outperforms the SBR modified bitumen, which in turns performs better than the unmodified bitumen (in terms of rutting resistance).

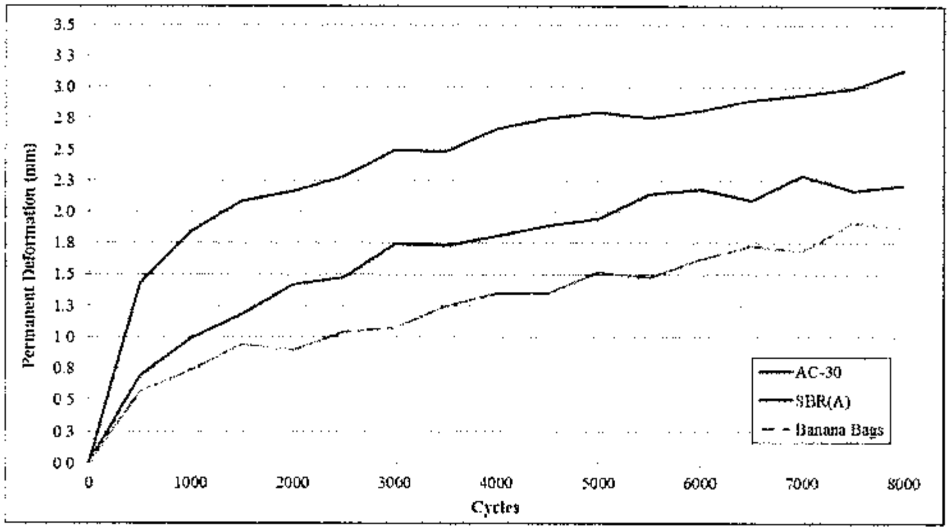


Figure 13. Permanent Deformation on APA Results

4. DISCUSSION

Banana bags are a material that can be used as a bitumen modifier due to the fact that it showed good affinity with the bitumen during the tests performed. The mixing process showed in fact an adequate distribution of the modifier on the bitumen, which was verified on the AFM image (Figure 9). This behavior is very similar to one of the best performing modifier that is used in Costa Rica: the SBS(A). Additionally, the calorimetry tests show that the banana bag fusion point is 121°C, at 150°C the chlorpyrifos is degraded, and the degradation of the polyethylene occurs at 425°C. The mixing of the materials was made at 160°C (normal temperature for the asphalt production), which allowed an excellent homogenization and also contributed to the removal of any traces of the chlorpyrifos, like the chlorides and chloro-sulphides, observed on the correspondent gas spectrum of a bitumen sample where no chlorpyrifos removal treatment was preciously applied.

With regards to the performance of the modified bitumen, the modification increases the upper limit of the PG grade from 64°C to 70°C in un-aged condition, and the short term aged bitumen temperature is also increased from 70°C to 76°C. This indicates better resistance to permanent deformation, even when compared to bitumen modified with SBR(A), a commercially sold bitumen modifier that was also evaluated as part of this study. When performing the repeated creep test, the improvement in performance when modifying the bitumen with waste banana cover bags was verified. This was evidenced by a 50% increase in resistance to permanent deformation at 70°C. However, the Multi Stress Creep Recovery (MSCR) analysis did not show any significant differences when comparing the bag modified bitumen with bitumen modified with other commercially sold additives. This is the case since the bag modified bitumen should be limited to normal loading conditions, as opposed to SBS(A) modifier which is designed for heavy loading applications.

5. CONCLUSIONS

The banana bag is a material that can be used as bitumen modifier since it increases the PG grade by 6°C, and reduces permanent deformation by up to 50% under normal traffic loading conditions. Based on the previous, the modified bitumen can be used in low volume roads, or roads without excessive heavy vehicle traffic.

The same performance trend that was observed at the bitumen level carries to the asphalt. This was confirmed since the rutting resistance of the banana modified asphalt performed significantly better that the asphalt containing the unmodified bitumen. Similar results were observed in the case of TSR, where the banana modified bitumen exhibited almost no reduction in tensile strength after a conditioning cycle. This was not the case with the asphalt containing the unmodified bitumen, which indicates that the polyethylene from the banana bags tends to improve resistance to moisture damage.

Consequently, the paper shows an effective method of not only improving bitumen performance, but most importantly has a positive effect on the environment. The use of the waste banana cover bags, instead of dumping the material in open pits or incinerating it, can result in a disposal of approximately 4 kg of material per cubic meter of asphalt and traps or eliminates the toxic gases, and consequent acid rain, that would be otherwise expelled into the atmosphere as a result of the incineration of the material.

6. ACKNOWLEDGEMENT

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7. FUTURE WORK

Given the results from the study, the next step consists in evaluating the performance of the asphalt with regards to rutting resistance, aging, and fatigue resistance in field test sections.

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