



Evaluation of the rheological effect of asphalt binder modification using *Linum usitatissimum* oil

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ABSTRACT

Warm mixtures are an essential alternative for paving as they minimize costs and environmental impacts, and their performance is similar to hot mixtures. This research evaluated the effect of adding linseed oil obtained from *Linum usitatissimum* in the contents of 1, 2, 3, 4, 5, and 6% by weight on the physical and rheological properties of the asphalt binder. The asphalt binder was evaluated through tests of penetration, softening point, rotational viscosity, performance grade (PG); these tests were performed before and after the short-term aging procedure; multiple stress creep and recovery (MSCR), and linear amplitude sweep (LAS). It has been verified that the mixing temperature could be reduced by up to 15 °C for the 6% linseed oil content, but it was lost in terms of performance, level of supported traffic, and fatigue life. In the so-called ideal content found, 4% linseed oil, there is a reduction in the mixing temperature of around 10 °C, as well as a maximum temperature of PG, 58 °C, it reduces by only one step compared to the pure binder, 64 °C, in addition to presenting the second-longest fatigue life of the grades studied, being classified as ideal for standard traffic by the MSCR test. Linseed oil proved to be an excellent green alternative for the production of warm mixtures.

Keywords: Rheology; Warm mixtures; Linseed oil; Alternative materials.

1. INTRODUCTION

Biobinders are renewable and ecologically correct materials that have been widely studied as additives to asphalt binders. These materials can soften the asphalt binder and improve its property at low temperatures [1-3]. PORTUGAL *et al.* [4] claim that vegetable oils are rich in unsaturated fatty acids and are resistant to high temperatures.

The use of oils in asphalt binders has a positive impact on the environment and can lower the viscosity of the asphalt binder, thus reducing the mixing temperature and leading to less energy consumption and greenhouse gas emissions [5]. On the economic side, low viscosity reduces mixing and compaction temperatures, leading to more economical pavement construction. This process brings cost savings and environmental benefits, reducing energy consumption during the production of the mixture [6].

Asphalt pavement can present many problems under the long-term effect of loads and environmental factors; fatigue cracking is one of the most common problems [7–11]. In addition, the asphalt binder with lower viscosity has a faster curing process than the more viscous asphalt binder [12].

The conventional asphalt mixture has an application temperature in the range between 160 and 175 °C, which generates a significant emission of gases that promote the greenhouse effect, putting the lives of workers who are performing the service at risk and making energy expenditure high to reach these temperatures in the mixing and compaction steps.

In this bias, warm asphalt mixtures (WMA) emerged, which are mixed and compacted at a temperature of up to 150 °C, generating savings in energy expenditure and reducing the emission of pollutants. Furthermore, the addition of organic/chemical additives, emulsions, and synthetic binders can lower mixing and field operating temperatures by approximately 40 °C. Lowering the temperature reduces energy costs, less smoke, and greenhouse gas emissions [13].

Among the researched organic additives, the use of oils stands out. SOUZA [14] investigated the incorporation of Castor Oil in Petroleum Asphalt Cement (PAC) 50/70 to manufacture warm mixtures with oil contents ranging from 2 to 9% by weight of the binder. The author found reductions of 10 °C in mixing and compaction temperatures when adding 5% of Castor oil without compromising the mechanical properties. RIBEIRO [15] evaluated warm mixtures with the addition of cashew nut liquid ranging from 0.5 to 2% by weight of the PAC. The author obtained more significant viscosity reductions for the addition of 2% of the liquid and found that the addition contributed to delay the aging of the PAC. LUCENA *et al.* [16] analyzed the consistency properties of the asphalt binder and the variation of mixing and compacting temperatures with the addition of Moringa Oleifera Lam oil at contents of 0, 0.5, 1, 1.5, 2, 2.5, and 3%. The authors pointed out that the use of this additive should be between 0.5 and 1%, as in these contents, there is no compromise in the performance of the asphalt binder, but it allows for a reduction in viscosity and in mixing and compaction temperatures.

ZENG *et al.* [17] investigated the use of castor oil as a rejuvenator and aged asphalt binder and verified that its incorporation causes a reduction in temperatures, a drop in resistance to permanent deformation, and again in fatigue resistance. However, the gain in fatigue strength is more significant than the loss of permanent deformation strength. GIRIMATH and SINGH [18] evaluated the incorporation of bio-oil derived from wood residue biomass on the physical and rheological properties of the asphalt binder. Amounts of 2, 4, 6, 8, and 10% of bio-oil by total weight of binder were used. The addition of this modifier provided an increase in penetration, a decrease in the softening point, and the asphalt binder's viscosity.

MELO NETO *et al.* [19] analyzed the effects of the incorporation of copaiba oil and refined cottonseed oil in the asphalt binder PG 64-XX at contents of 4 and 5% by weight of the asphalt binder. The results showed that the addition of 5% of copaiba oil resulted in a reduction of 9.6 °C in the mixing and compaction temperature. Asphalt binder samples with 4% refined cottonseed oil, 4% copaiba oil, and 5% copaiba oils reduced the PG of pure asphalt binder from 64 to 58 °C, classifying it as PG 58-XX. The addition of 5% refined cottonseed oil resulted in the greatest reduction in the PG value, from 64 to 52 °C, classifying it as PG 52-XX. Finally, PORTUGAL *et al.* [20] evaluated the behavior of an asphalt mixture with asphalt binder modified with waste and new soybean oil. The results showed that adding 1% of both oils in the base binder improved its physical and rheological characteristics, and a reduction of 2.7 °C in production temperatures was achieved.

Linseed oil, extracted from linseed (*Linum usitatissimum*), has an orange color and a characteristic sweet odor, rich in fatty acids and protein, and is an excellent antioxidant. This oil has in its composition 9% saturated fat, 18% monounsaturated fat, and polyunsaturated fat (omega 3, 57% and omega 6, 16%). It is a commercially accessible compound used in construction, cooking, medicine, manufacturing linoleum, varnishes, dyes, and paints, acting as a drying agent. Linseed oil has favorable characteristics as it is an oil from a renewable and biodegradable source, has tensoactive properties, is an inert oil, antioxidant, easy to mix, and has a large amount of linoleic linolenic acids in its composition. It is promising to reduce the high viscosity of pure binders and polymer-modified binders, reducing costs in the plant and increasing storage stability and adhesiveness in mixtures with aggregates.

Thus, the following question arises: to study the influence on the rheological properties of the asphalt binder modified by linseed oil to enable its use as a temperature reducer for asphalt mixtures. Therefore, this research aimed to evaluate the rheological properties of a modified PG 64-XX asphalt binder in contents of 1, 2, 3, 4, 5, and 6% by weight of binder with linseed oil.

2. MATERIALS AND METHODS

2.1. Materials

In the production of the mixture, Petroleum Asphalt Cement was used with a 50/70 degree of penetration (PAC 50/70) granted by the processing plant of the company Rocha Cavalcante, located in the city of Campina Grande – PB, and *Linum usitatissimum* oil (linseed) obtained from local businesses in the same municipality. To carry out the research, contents of 1, 2, 3, 4, 5, and 6% of linseed oil were used as an additive to the pure asphalt binder (PAC 50/70). The contents established for addition to the binder were based on studies [6, 14, 16, 19, 21–23], which used contents ranging from 1 to 10% of oils as a modifying agent for the asphalt binder. Table 1 presents the description of the samples used in the research and their respective nomenclatures.

SAMPLES	NOMENCLATURE
Conventional pure binder	PAC 50/70
PAC 50/70 + 1% Linseed oil	1%OL
PAC 50/70 + 2% Linseed oil	2%OL
PAC 50/70 + 3% Linseed oil	3%OL
PAC 50/70 + 4% Linseed oil	4%OL
PAC 50/70 + 5% Linseed oil	5%OL
PAC 50/70 + 6% Linseed oil	6%OL

Table 1: Nomenclature of the samples.

2.2. Methods

In the mixing process (binder/bio-oil), a FISATOM mechanical stirrer, Model 722D, was used. To produce the mixture between the asphalt binder and linseed oil, the parameters consolidated in the literature for mixing the binder with organic oils were followed, as described by CAVALCANTE [24]. The shear mixer was used, where it indicates rotation speed and temperature in a digital device. The procedure was followed by heating the PAC 50/70 in an oven at a temperature of around 120 °C for about an hour and a half, ensuring that the material was fluid for the mixture, then the PAC 50/70 was placed in the beaker and took it to the mixer. In the mixer, the temperature used was 135 °C at a rotation of 400 rpm. After guaranteeing the stability of temperature and agitation, the linseed oil was added at the contents of 1, 2, 3, 4, 5, and 6% by weight of asphalt binder, leaving the mixture under agitation for 20 minutes. Once the mixing process was completed, the material was stored in suitable containers and allowed to cool.

After obtaining the modified asphalt binders, the physical and rheological characterization of the binders was carried out using Penetration (ASTM D5M [25]), Softening Point (ASTM D36M-14 [26]), Rotational Viscosity (ASTM D4402 [27]) tests, Performance Grade (PG) (ASTM D6373 [28]) – these tests were performed before and after the RTFO aging procedure (ASTM D2872 [29]), Multiple Stress Creep Recovery (MSCR) (ASTM D7405 [30]), and linear amplitude sweep (LAS) (AASHTO TP 101–12 [31]).

The rheological tests were performed according to the Superpave methodology for pure and modified asphalt performance analysis. From then on, an optimal content was indicated among those tested, which showed the best performance. The rheological characterization assays were performed with a hybrid Oscillatory Rheometer Discovery HR-1 rheometer at the Pavement Engineering Laboratory – LEP in the Federal University of Campina Grande – UFCG, Brazil.

The maximum performance of the binder was given by the temperature corresponding to the value of the parameter $G^*/sin\delta$ greater than 1.0 kPa (before short-term aging) and greater than 2.2 kPa (after the aging process in the Rolling Thin Film Oven – RTFO). ASTM specification D6373 [28] determines standard maximum temperatures for PG analysis of binders ranging from 46 °C to 82 °C. Tests were performed on samples before and after RTFO. The determination of continuous PG was performed in 1 °C steps in a temperature range of 46 °C to 82 °C.

The MSCR test was performed based on ASTM D7405 [30]. This test was performed in RTFO using their respective PG temperatures to obtain Jnr and R's parameters (%). In the test, 10 cycles were applied, each at two load levels (0.1 and 3.2 kPa). Each cycle consists of a 1-second creep load period, followed by a 9-second relaxation period.

The LAS test is governed by AASHTO TP 101–12 [31], which determines that samples must be aged in RTFO and PAV; however, as this was a laboratory limitation, the test was performed only for samples aged in RTFO. In the LAS test, the geometry of the rheometer had to be heated to 56 °C to guarantee the adherence of the sample to the geometries. Therefore, the samples in the DSR needed to be cooled to 25 °C in the LAS test. In this test, the samples underwent two steps: (i) frequency scanning, in which the sample was subjected to a shear load to obtain the rheological properties of the asphalt binders, carried out at a temperature of 25 °C, with frequencies from 0.1 to 30 Hz and the amplitude level of 0.1%, followed by (ii) amplitude sweep to measure the damage suffered by the binder, in which small torques were applied to the same sample under a constant frequency of 10 Hz and the amplitude level was varied from 0.1 to 30%.

3. RESULTS AND DISCUSSION

This section presents and discusses the results obtained in the experimental phase of asphalt binders modified by linseed oil.

3.1. Physical properties of asphalt binders

Table 2 presents the results of physical characterizations for the study samples.

It is possible to observe that the addition of this modifying agent provided a mass variation lower than that observed in the pure PAC. Also, the modified samples were less prone to mass loss with the increase of the linseed oil content in the composition. About the pure binder, the 6% content of linseed oil presents a 55% lower mass loss, which shows the antioxidant properties. Linseed oil is rich in omega 3, which is an important antioxidant agent, and around 6 to 11% of saturated fatty acids, 13 to 29% of oleic acid, 17 to 30% of linoleic acid, and 47 to 55% linolenic acid. Oils rich in oleic acid are more stable to oxidation both at ambient storage temperatures and at high temperatures (existing in the machining of asphalt binders), where these organic additives have long chain hydrocarbons that provide low viscosity values at high temperatures when added to asphalt binders [32].

All modified binders and the pure binder were consistent with what is prescribed in the DNIT 095 standard [33], where for PAC 50/70, the mass loss variation should not exceed 0.5%. After aging, the binders were submitted to the same characterization tests as virgin (non-aged) binders to verify the influence of oxidation on the properties of the binder modified with linseed oil.

It is noticeable that with the increase in the oil content in the mixture, the more significant the penetration, with the maximum value in the content of 6% of linseed oil, in the order of 135 tenths of a millimeter, which corresponds to an increase of 107% about pure binder before aging. Such behavior also occurs when studying the aged binder, increasing 102% of the penetration value.

SILVA [34] studied the effect of the addition of Sunflower oil and Moringa oil on the rheological properties of asphalt binder and found similar penetration values for the contents of 1 and 2% of addition of these oilseeds. According to ANP No. 19 [35], for an asphalt binder PAC 50/70 subjected to short-term aging in the RTFO, a minimum of 55% of retained penetration is admitted. Therefore, it is observed that the results are by the specification.

The results of retained penetration were at least 62.02% for 5% linseed oil content and at most 70.93% for 2% linseed oil content. All values obtained are forecast for a retained penetration of at least 55% [33]. It is known that the retained penetration is shown as an indicator of the susceptibility of asphalt binders to aging,

TESTS BEFORE RTFO

			ILSI	S BEFUR	E KIFU				
				RESULT	ſS				
		PAC 50/70	1%OL	2%OL	3%OL	4%OL	5%OL	6%OL	Limits
Penetration 0.1 (100 g, 5 s, 25		65	83	86	105	113	129	135	50 a 70
Mass variation	(%)	0.113	0.09	0.09	0.071	0.057	0.053	0.051	≤0.5
Softening Poin	nt (°C)	48	44	43	41	40	37	36	-
Rotational	135 °C	455.00	427.00	398.00	354.00	315.00	290.00	260.00	≥274
Viscosity	150 °C	221.50	235.00	195.00	179.00	164.00	152.50	139.00	≥112
(cP)	177 °C	79.00	72.00	70.50	65.50	63.25	60.00	56.25	57 a 285
TESTS AFTI	ER RTFO								·
Penetration 0.1 5 s, 25 °C)	mm (100 g,	46	52	61	67	70	80	93	-
Softening Poin	nt (°C)	53	52	51	51	47	46	45	_
Softening Poin Variation (°C)	ıt	5	8	8	10	7	9	9	≤8
Retained penet	tration (%)	70.77	62.65	70.93	63.81	61.95	62.02	68.89	≥55
Rotational	135 °C	640.00	592.50	534.50	487.00	466.25	450.00	400	_
Viscosity	150 °C	298.50	285.00	261.50	237.50	232.50	225.50	203	_
(cP)	177 °C	101.50	97.50	84.00	82.50	84.25	80.25	76.5	-

Table 2: Physical characterization of asphalt binders before and after RTFO.

thus indicating that the contents of 1, 4, and 5% are more sensitive to aging as they have lower values of retained penetration. CAVALCANTE [24], in research, carried out with linseed oil, found results of 63.3, 62.2, and 66.9% for the additions of 4, 5, and 6%, respectively, values similar to those found in this research.

In line with the penetration result, the softening point test shows that the binder consistency decreases with linseed oil in the binder. In addition, the results showed a decrease in the softening point of the binder modified with linseed oil about the pure binder, before and after aging, as expected.

For PAC 50/70, the maximum increase allowed is 8 °C; according to DNIT 095 [33] Standard, the only samples that exceeded this value were with the addition of 3, 5, and 6% of Linseed oil, not meeting the limit established by the standard. The decrease in the softening point may indicate minor aging of the asphalt binder, as the softening point is related to the material's stiffness. Asphalt binders after the aging procedure demonstrate greater stiffness and therefore have increases in softening point values [36].

The test of rotational viscosity of virgin binder and aged binder was carried out to measure the effect of oxidation on viscosity and the antioxidant effect of linseed oil.

According to the results obtained, it appears that with the increase in the content of linseed oil added to the asphalt binder, its viscosity continuously decreased. Comparing the results obtained with the pure binder to the results obtained for the binder with 6% linseed oil content, an average reduction of 36% in viscosity at all temperatures are observed, while the 3% linseed oil content is observed. In samples of asphalt binder with linseed oil, there was an average reduction of 19% in the viscosity value at all temperatures before the binder aging. It is essential to note the proportionality of the results, considering that the decrease in viscosity occurs uniformly with the increase in the contents of linseed oil present in the asphalt binder.

According to the results obtained after aging, it is observed that the pure binder had an average increase in viscosity of 35% at all temperatures, while the binder modified with 3% linseed oil had an average increase of 32% and, for comparison, the modified binder with 6% linseed obtained an increase in average viscosity of 27% about the virgin binder. This is due to the antioxidant property present in linseed oil, which prevents the asphalt binder from losing its properties if there is an aging process.

The addition of linseed oil at the contents studied caused a decrease in viscosities compared to pure PAC, which consequently reflected in the compaction and mixing temperatures (CMT), which is related to the workability of the binder. ASPHALT INSTITUTE [37] points out that the ideal PAC temperature for carrying out the asphalt mixture should be at 0.17 ± 0.02 Pa.s when measured with a viscometer. The compaction temperature is that at which the asphalt binder has a viscosity of 0.28 ± 0.03 Pa.s. Table 3 presents the mixing temperatures found through interpolation for the addition of linseed oil.

From the results obtained, it is evident that linseed oil made the binder more fluid, that is, less viscous, which reduces the mixing and compaction temperatures of the mixture, generating energy savings during the execution of the work, in other words, making paving cheaper. The binder modified with 6% linseed oil presents an average reduction of 10% in the maximum and minimum mixing temperatures compared to the pure binder, which in practical numbers translates into a reduction of 15 °C, corroborating the results obtained by CAVAL-CANTE [24], verifying percentages of CMT reduction between 14 and 16% for the binder modified with 6% linseed oil.

SAMPLE	MIXING TEMPERATURE (°C)	REDUCTION (°C)	COMPACTION TEMPERATURE (°C)	REDUCTION (°C)
PAC 50/70	159.00	-	143.10	_
1%OL	157.05	1.95 (1%)	142.35	0.75 (1%)
2%OL	154.50	4.50 (3%)	139.75	3.35 (2%)
3%OL	151.85	7.15 (4%)	139.10	4.00 (3%)
4%OL	149.10	9.90 (6%)	137.50	5.60 (4%)
5%OL	146.50	12.50 (8%)	136.90	6.20 (4%)
6%OL	143.75	15.25 (10%)	136.20	6.90 (5%)

 Table 3: Mixing and Compaction Temperatures.

3.2. Rheological analysis of asphalt binders

3.2.1. Performance Grade (PG)

The PG test was performed both before and after short-term aging and provided information about the maximum use temperature of that asphalt binder, as well as deformability and stiffness parameters.

From this test, the Complex Modulus (G*) parameters are obtained, which is related to the stiffness of the binder, the delta angle (δ), which is related to the elasticity, the parameter G*/sin(δ), which reflects the deformability of the asphalt binder and, as the main result, delimits the minimum and maximum use temperature range of the binder. In this research, the test to determine the minimum Temperature of PG was dispensed with because, in Brazil, negative temperatures are not ordinary, stopping only at the maximum Temperature of PG. Figure 1 shows the result of the maximum use temperatures of the asphalt binder before and after the short-term aging process (RTFO).

An essential fact in the analysis of this result is that the pure asphalt binder, PAC 50/70, presents a reduction in the PG temperature after aging, decreasing from 64 to 58 °C, that is, it is susceptible to the effect of oxidation, and it loses its deformability and stiffness properties at high temperatures. Furthermore, it is observed that the addition of 1% linseed oil already had an antioxidant action, preventing the degenerative effect, keeping the PG temperature constant for both the virgin binder and the binder aged at 64 °C. However, as the content of linseed oil added to the binder increases, it is lost in PG temperature, a fact explained by the loss of viscosity and the consequent increase in deformability that occurs due to the increase in temperature.

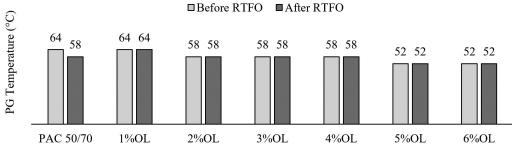
There is a significant loss in use temperature for the contents of 5% and 6%, stopping at 52 °C, a temperature that is easily reached by the asphalt pavement on hot days in northeastern Brazil, which makes its use unfeasible in this situation. Regarding the complex modulus (G^*), which is linked to the stiffness of the binder, Figure 2 shows that this parameter decreases with the increase in the addition of linseed oil; that is, the binder becomes less rigid, both before and after the short-term aging process.

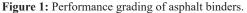
According to the results obtained, it can be seen that at a temperature of 46 °C before aging, the pure binder has a complex modulus higher than the other binders, with a G* value in the order of 19 kPa, a value that decreases as the add to and increases the linseed oil content to the asphalt binder reaching 4.2 kPa for the sample with 6%OL. Performing this same analysis for aged binders, it is noted that the pure binder and the binder modified with 1% linseed oil present practically equal values, with G* values of the order of 28 kPa, also decreasing with the increase in temperature. However, the modified binder with oil presents complex modulus values up to a temperature of 70 °C, which reinforces the idea that this substance confers antioxidant properties to the asphalt binder, allowing it to be better used at high temperatures without losing much of its performance.

In line with the analysis of the complex modulus (G*), Figure 3 shows the variation of the parameter $G^{*/}$ sin(δ) with the increase in temperature during the test, before and after aging, respectively.

The parameter $G^*/\sin(\delta)$ measures the resistance to permanent deformation of the asphalt binder and is established as a stopping criterion for the PG test. In the virgin binder, when $G^*/\sin(\delta)$ is less than 1.0, the test is terminated, as it is assumed that from then on, the binder is very deformable and does not meet the minimum performance criteria. The analysis is the same in the test with the aged binder, but the test stop criterion is when $G^*/\sin(\delta)$ is less than 2.0.

Comparing two samples of binder that have the same PG value, 3 and 4% for example (58 °C), it is observed that 4%OL presents a 20% reduction in the value of G*/sinδ over 3%OL, making the binder more deformable, highlighting that the increase in oil is crucial for the performance of the binder in terms of permanent deformation at the same temperature of use. Thus, the optimum content of linseed oil addition must be





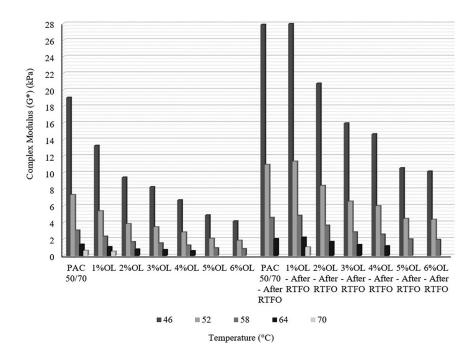


Figure 2: Complex Modulus Parameter (G*) as a function of temperature before and after RTFO.

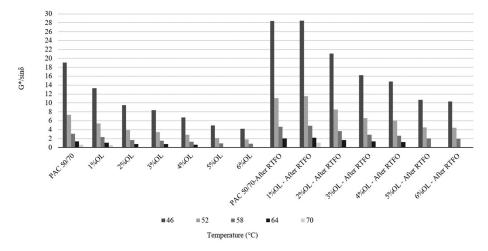


Figure 3: Parameter G*/sino as a function of temperature before and after RTFO.

meticulous, considering that it gains in some properties, but there is a loss in others. The 3%OL sample was less susceptible to permanent deformation, but this data was also analyzed in the MSCR test to confirm the statement.

3.2.2. Multiple Stress Creep Recovery (MSCR)

The MSCR is, as a rule, carried out only with the aged asphalt binder. The test makes it possible to obtain the level of traffic supported by the binder and the percentage of recovery of the binder when subjected to two load levels (100 and 3200 Pa). The higher the value of Jnr, the material becomes more susceptible to permanent deformation. On the other hand, smaller values of Jnr indicate the binder's resistance to this effect [36]. The results are shown in Figure 4, representing the value of the non-recoverable compliance, Jnr, under loads of 100 and 3200 Pa.

According to AASHTO M320 [38], the relation between the values obtained for Jnr at 3200 Pa and the traffic class in which the binder is found can be made. Table 4 presents this classification.

According to the AASHTO M320 [38] classification, shown in Table 4, the pure binder is suitable for heavy traffic; it has several requests above 10 million if used at the PG temperature, 58 °C. The binder modified with 5% linseed oil appears at the maximum demand limit of heavy traffic, but its PG is 52 °C, making its use very dependent on temperature changes. The binder modified with 1% linseed oil is not suitable for paving, as it is far above the maximum limit established by the standard, which is 4.0 kPa⁻¹. This fact is because the test was carried out at a temperature of 64 °C, which makes the binder very fluid and deformable, and these results

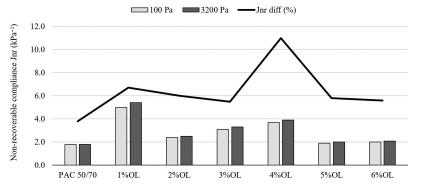


Figure 4: Values of Jnr under loads of 100 and 3200 Pa.

PROPERTY	JNR (KPA-1)	TYPE OF TRAFFIC	NUMBER OF PASSES ON A STANDARD AXIS
Jnr at 3200 Pa at the	2.0 - 4.0	Standard (S)	<10 millions
maximum temperature	1.0 - 2.0	Heavy (H)	>10 millions
of PG	0.5 - 1.0	Very heavy (V)	>30 millions
	0.0-0.5	Extremely heavy (E)	>100 millions

Source: AASHTO M320 [38].

are corroborated by CAVALCANTE [24]. The other binders with added linseed oil fall into the standard traffic category, with several requests less than 10 million. The 4% content of linseed oil stands out as possible ideal content for the research because it fits like a warm mixture; since its mixing and compaction temperature are below 150 °C, its maximum PG did not show significant reduction compared to the pure binder and still classified as adequate for standard traffic.

Another essential parameter highlighted in the analysis of the MSCR assay is the Jnr differential (Jnr diff), which measures the difference between the Jnr at 100 and at 3200 Pa, expressed as a percentage. According to SOBREIRO [39], the difference between Jnr under the tension of 100 and 3200 Pa must be less than 75% to attest that the property of the binder is not overly sensitive to changes in loading. In this way, all the binders were researched to meet the established criteria and are suitable for paving. The MSCR test also provides the elastic recovery result, expressed as a percentage, which measures the elastic properties of the asphalt binder. The pure binder presented, as the standard, practically zero elastic recoveries, in the order of 1.90%. This value did not change as linseed oil is added at the levels studied because it does not have elastic properties.

3.2.3. Linear Amplitude Sweep (LAS)

Figure 5 shows the results of the evaluation of parameters A and B in the resistance to damage obtained by the linear amplitude sweep (LAS) test for samples of PAC 50/70 and samples modified with linseed oil at the levels under study.

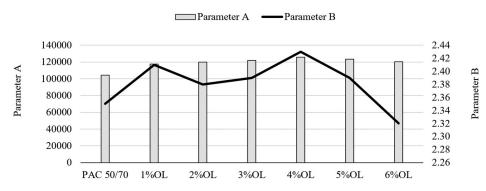


Figure 5: Results of Parameter A and Parameter B from the LAS tests.

STRAIN	1.0%	2.5%	5.0%	10.0%
PAC 50/70	63370	12082	2364	463
1%OL	67145	12899	2400	496
2%OL	69134	13181	2517	511
3%OL	69433	13238	2553	557
4%OL	69709	13528	2603	600
5%OL	69332	13151	2453	568
6%OL	67981	13249	2332	526

Table 5: Estimated values for fatigue life (Nf) of the binder under study.

According to MARINHO FILHO [40], parameter A is related to the variation in the integrity of the material due to accumulated damage; higher values of A mean that the sample maintained its initial integrity. Therefore, it was noted with the help of the graph that the sample that obtained the highest initial integrity was 4% linseed oil, with an increase of 20% compared to the pure binder. It is also noticeable that there was a gradual increase in this pattern until reaching the peak in the 4% sample. In the case of the pure binder, this presents a parameter A 12.60% lower than the binder with a lower linseed oil content, 1%. This fact attests that, even in a small proportion, linseed oil acts by increasing the deformability of the asphalt binder.

Parameter B, indicated by the slope of the frequency line versus storage modulus (G*), demonstrated that despite the 4% linseed oil sample having more excellent resistance to accumulated damage, it was more sensitive to strain levels. According to MARINHO FILHO [40], this sensitivity is not desired for asphalt binders, as it indicates an easy susceptibility to deformations under temperature variations. Considering this, the 3% sample obtained the best parameter B, as it had practically no variation about the 2% sample.

Also, from the LAS test, it is possible to estimate the fatigue life of the asphalt binder (Nf) as a function of strain levels. Table 5 was originated from the use of fatigue life estimate of binders modified with linseed oil for strain levels of 1, 2.5, 5, and 10%.

NASCIMENTO [41] states that deformation levels of up to 3% are characteristic of pavements with low deflection levels and have a behavior similar to a rigid pavement. In contrast, high deflectometry levels are better compared to flexible pavements. Analyzing the data in Table 5, it can be seen that the Nf for linseed oil contents has a gradual increase as the percentages of the modifier are added. However, from the sample with the addition of 5% of linseed oil, a decrease in this parameter was noted, that is, the addition of contents greater than 5% of linseed oil did not promote improvements in fatigue resistance. Therefore, for high deflectometry levels in the pavement, it is suggested to use the contents with the most extended fatigue lives, which in this research are 3% and 4%.

Another important fact when analyzing the data in Table 5 is that between 2 and 5% of linseed oil added to the asphalt binder, changes in fatigue life are practically nil. The initial increase in fatigue life, on the average of 5%, of the binder modified with 1% linseed oil compared to the pure binder reinforces the idea that linseed oil acts as an agent that prevents premature aging binder asphalt.

4. CONCLUSIONS

The results of this research allowed us to conclude that:

- The physical characterization tests confirmed that linseed oil gives the asphalt binder loss of viscosity and consistency; however, as it has antioxidant properties, it provided more excellent resistance to aging, as evidenced by the result of loss of mass, which undergoes a gradual reduction in as the oil content in the binder is increased.
- A reduction in mixing and compaction temperatures were observed from the rotational viscosity test at all contents studied. However, the most significant reduction was observed in the binder modified with 6% linseed oil, with its ideal mixing temperature was reduced by 15 °C compared to the pure binder, representing a percentage of 10.20%. In addition, the compaction temperature decreased by 5 °C.
- When analyzed about the degree of performance, the modified binder managed to maintain the same temperature class of 64 °C as the pure binder at 1% linseed oil content. From the binder with 2% content to 4% linseed oil, the modified binder had a drop in its strength to the point of decreasing its PG at a temperature of 58 °C and, finally, 52 °C at contents of 5 and 6% oil. The antioxidant effect of linseed oil, evidenced in the

short-term aging test (RTFO), is reinforced in the PG test of the samples after aging, considering that the pure binder suffered a PG reduction of 64 °C when the virgin, to 58 °C when aged. In samples modified with oil, there was no drop in PG comparing virgin and aged samples.

- The MSCR test showed that with the increase in the oil content in the binder, the level of traffic supported by it decreases. In this way, it is necessary to compensate the extent to which it is advantageous to lose mixing and compaction temperature, gain in resistance to aging, and lose in performance and applicability of the asphalt binder.
- The parameter A found in the linear amplitude sweep test showed that the modified binders had greater integrity of their characteristics and the 3% sample had zero sensitivity to other strain levels. It was also noted that the modified samples had a longer fatigue life, especially for pavements with high deflection levels.
- The results showed that the sample with 4% linseed oil has the best rheological performance without a sudden reduction in the strength of the binder. Using this content, the mixing temperature can be reduced by 10.8 °C, with values on the order of 145.2 °C, thus classifying it as a warm mixture. Furthermore, its maximum Temperature of PG, 58 °C, is reduced by only one step compared to the pure binder, 64 °C. This grade also had the second-longest fatigue life of the studied binders and is ideal for standard traffic.

Thus, the addition of linseed oil proved to be a good alternative as a green modifier for incorporation into the asphalt binder. Due to the reduction in the softening point temperature and PG, it is advisable that these modified binders are applied in regions that do not have such high temperatures in the summer, such as the southern region of Brazil, which generally does not exceed 40 °C. Studies investigating the use of linseed oil-modified asphalt binder in recycled asphalt mixtures are encouraged.

5. ACKNOWLEDGMENTS

The authors would like to acknowledge the financial support of the Brazilian National Council for Scientific and Technological Development (CNPq) and the Coordination for the Improvement of Higher Education Personnel (CAPES) for the research scholarships of the authors. The authors also acknowledge the support of the Pavement Engineering Laboratory (LEP) of the Federal University of Campina Grande.

6. **BIBLIOGRAPHY**

- [1] CAI, X., ZHANG, J., XU, G., *et al.*, "Internal aging indexes to characterize the aging behavior of two bio-rejuvenated asphalts". *Construction & Building Materials*, v. 220, pp. 1231–1238, 2019.
- [2] MENDONÇA, A.M.G.D., MELO NETO, O.M., RODRIGUES, J.K.G., et al. "Análise física-reológica de ligantes asfálticos modificados com óleo de algodão refinado para uso em misturas asfálticas mornas", *Revista Cubana de Ingeniería*, v. 13, n. 2, pp. e325, 2022.
- [3] MENDONÇA, A.M.G.D., MELO NETO, O.M., RODRIGUES, J.K.G., et al., "Characterisation of modified asphalt mixtures with lignin of pinus and eucalyptus woods", *Australian Journal of Civil Engineer*ing, 2022. In press.
- [4] PORTUGAL, A.C.X., LUCENA, L.C.F.L., LUCENA, A.E.F.L., *et al.*, "Rheological properties of asphalt binders prepared with maize oil". doi: http://dx.doi.org/10.1016/j.conbuildmat.2017.07.077. *Construction* & *Building Materials*, v. 152, pp. 1015–1026, 2017.
- [5] PODOLSKY, J.H., BUSS, A., WILLIAMS, R.C., et al., "Effect of bio-derived/chemical additives on warm mix asphalt compaction and mix performance at low temperature". doi: http://dx.doi.org/10.1016/j. coldregions.2017.02.003. Cold Regions Science and Technology, v. 136, pp. 52–61, 2017.
- [6] NOGUEIRA, R.L., SOARES, J.B.S., SOARES, S.A., "Rheological evaluation of cotton seed oil fatty amides as a rejuvenating agent for RAP oxidized asphalts". doi: http://dx.doi.org/10.1016/j.conbuildmat.2019.06.128. Construction & Building Materials, v. 223, pp. 1145–1153, 2019.
- [7] ZHANG, H., CHEN, Z., XU, G., et al., "Physical, rheological and chemical characterization of aging behaviors of thermochromic asphalt binder". doi: http://dx.doi.org/10.1016/j.fuel.2017.09.111. Fuel, v. 211, pp. 850–858, 2018.
- [8] ZHANG, R., YOU, Z., WANG, H., et al., "The impact of bio-oil as rejuvenator for aged asphalt binder". doi: http://dx.doi.org/10.1016/j.conbuildmat.2018.10.168. Construction & Building Materials, v. 196, pp. 134–143, 2019.
- [9] MENDONÇA, A.M.G.D., MELO NETO, O.M., RODRIGUES, J.K.G., et al., "Physicochemical and rheological effects of the incorporation of micronized polyethylene terephthalate in asphalt binder". doi: http://dx.doi. org/10.1080/10916466.2021.2007124. Petroleum Science and Technology, v. 40, n. 7, pp. 822–838, 2022.

- [10] CRUZ, G.K.A., MELO NETO, O.M., ARRUDA, S.M., *et al.*, "Influence of particle size selection methods on asphalt mixtures produced with lateritic aggregates". doi: http://dx.doi.org/10.1016/j.conbuildmat.2021.125201. *Construction & Building Materials*, v. 314, pp. 125201, 2022.
- [11] CARVALHO, F.S.S., LUCENA, A.E.F.L., MELO NETO, O.M., et al., "Análise dos parâmetros mecânicos das misturas asfálticas com adição de óxidos metálicos", *Revista Matéria*, v. 26, n. 3, pp. e13020, 2021.
- [12] HE, M., TU, C., CAO, D.W., et al., "Comparative analysis of bio-binder properties derived from different sources". doi: http://dx.doi.org/10.1080/10298436.2017.1347434. The International Journal of Pavement Engineering, v. 20, n. 7, pp. 792–800, 2017.
- [13] SHU, X., HUANG, B., SHRUM, E.D., et al., "Laboratory evaluation of moisture susceptibility of foamed warm mix asphalt containing high percentages of RAP". doi: http://dx.doi.org/10.1016/j.conbuildmat.2012.02.095. Construction & Building Materials, v. 35, pp. 125–130, 2012.
- [14] SOUZA, J.L.S., "Estudo das propriedades mecânicas de misturas asfálticas com cimento asfáltico de petróleo modificado com óleo de mamona", Tese de M.Sc., UFCG, Campina Grande, PB, Brasil, 2012.
- [15] RIBEIRO, E.A., "O efeito da modificação de ligante asfáltico com o liquido da castanha de caju (LCC) na resistência ao dano por umidade em misturas asfálticas", Tese de M.Sc., UFC, Fortaleza, CE, Brasil, 2012.
- [16] LUCENA, L.C.F.L., SILVEIRA, I.V., COSTA, D.B., "Avaliação de ligantes asfálticos modificados com óleo da Moringa Oleífera Lam para uso em misturas mornas". doi: http://dx.doi.org/10.1590/S1517-707620160001.0007. *Revista Matéria*, v. 21, pp. 72–82, 2016.
- [17] ZENG, M., LI, J., ZHU, W., et al., "Laboratory evaluation on residue in castor oil production as rejuvenator for aged paving asphalt binder". doi: http://dx.doi.org/10.1016/j.conbuildmat.2018.10.204. Construction & Building Materials, v. 193, pp. 276–285, 2018.
- [18] GIRIMATH, S., SINGH, D., "Effects of bio-oil on performance characteristics of base and recycled asphalt pavement binder's", *Construction and Building Materials*, v. 227, pp. 116684, 2019.
- [19] MELO NETO, O.M., MENDONÇA, A.M.G.D., RODRIGUES, J.K.G., et al. "Rheological study of asphalt binder modified by cotton and copaiba oils", *Revista Cubana de Ingeniería*, v. 13, n. 1, pp. e315, 2022.
- [20] PORTUGAL, A., LUCENA, L., LUCENA, A., et al., "Rheological performance of soybean in asphalt binder modification". doi: http://dx.doi.org/10.1080/14680629.2016.1273845. Road Materials and Pavement Design, v. 19, n. 4, pp. 768–782, 2017.
- [21] SUN, Z., YI, J., HUANG, Y., et al., "Properties of asphalt binder modified by bio-oil derived from waste cooking oil". doi: http://dx.doi.org/10.1016/j.conbuildmat.2015.10.173. Construction & Building Materials, v. 102, pp. 496–504, 2016.
- [22] PRADHAN, S.K., SAHOO, U.C., "Performance assessment of aged binder rejuvenated with Polanga oil", Journal of Traffic and Transportation Engineering, v. 6, pp. 608–620, 2019.
- [23] MELO NETO, O.M., "Viabilidade de misturas asfálticas recicladas com ácido graxo da borra do óleo de soja", Tese de M.Sc., UFCG, Campina Grande, PB, Brasil, 2022.
- [24] CAVALCANTE, F.P., "Efeito da adição dos óleos de ricinus communis e linus usitatissimum nas propriedades reológicas do cimento asfáltico de petróleo puro e modificado", Tese de D.Sc., UFCG, Campina Grande, PB, Brasil, 2016.
- [25] AMERICAN SOCIETY FOR TESTING AND MATERIALS, ASTM D5M Standard Test Method for Penetration of Bituminous Materials, West Conshohocken, ASTM, 2020.
- [26] AMERICAN SOCIETY FOR TESTING AND MATERIALS, ASTM D36M-14 Standard Test Method for Softening Point of Bitumen (Ring-and-Ball Apparatus), West Conshohocken, ASTM, 2020.
- [27] AMERICAN SOCIETY FOR TESTING AND MATERIALS, *ASTM D4402 Standard Test Method for Viscosity De-termination of Asphalt at Elevated Temperatures Using a Rotational Viscometer*, West Conshohocken, ASTM, 2015.
- [28] AMERICAN SOCIETY FOR TESTING AND MATERIALS, ASTM D6373 Standard Specification for Performance-Graded Asphalt Binder, West Conshohocken, ASTM, 2021.
- [29] AMERICAN SOCIETY FOR TESTING AND MATERIALS, ASTM D2872 Standard Test Method for Effect of Heat and Air on a Moving Film of Asphalt (Rolling Thin-Film Oven Test), West Conshohocken, ASTM, 2019.

- [30] AMERICAN SOCIETY FOR TESTING AND MATERIALS, ASTM D7405 Standard Test Method for Multiple Stress Creep and Recovery (MSCR) of Asphalt Binder Using a Dynamic Shear Rheometer, West Conshohocken, ASTM, 2020.
- [31] AMERICAN ASSOCIATION OF STATE HIGHWAY AND TRANSPORTATION, AASHTO TP 101–12 Standard Method of Test for Estimating Fatigue Resistance of Asphalt Binders Using the Linear Amplitude Sweep, Washington, D.C., AASHTO, 2018.
- [32] FERROTTI, G., RAGNI, D., LU, X., et al., "Effect of warm mix asphalt chemical additives on the mechanical performance of asphalt binders". doi: http://dx.doi.org/10.1617/s11527-017-1096-5. Materials and Structures, v. 50, n. 5, pp. 226, 2017.
- [33] DEPARTAMENTO NACIONAL DE INFRAESTRUTURA DE TRANSPORTES, DNIT 095 Cimentos asfálticos de petróleo Especificação de material, Rio de Janeiro, DNIT, 2006.
- [34] SILVA, G.G., "Estudos reológicos de aditivos utilizados na fabricação de misturas mornas", Tese de M.Sc., UFCG, Campina Grande, PB, Brasil, 2016.
- [35] AGÊNCIA NACIONAL DO PETRÓLEO, GÁS NATURAL E BIOCOMBUSTÍVEIS, ANP Nº 19: Regulamento Técnico para Investimentos em Pesquisa e Desenvolvimento e Elaboração do Relatório de Demonstração, Brasil, ANP, 2005.
- [36] MELO NETO, O.M., FERREIRO, A.A., FREIRE, T.S., et al., "Rheological analysis of asphalt binders modified with hydrated lime and titanium dioxide nanoparticles". doi: http://dx.doi.org/10.31686/ijier. vol8.iss11.2787. International Journal for Innovation Education and Research, v. 8, n. 11, pp. 579–598, 2020.
- [37] ASPHALT INSTITUTE, "Superpave Level 1: Mix Design", In: Superpave Series, n. 2 (SP-2), Lexington, Asphalt Institute, 1995.
- [38] AMERICAN ASSOCIATION OF STATE HIGHWAY AND TRANSPORTATION, AASHTO M320 Standard specification for performance-graded asphalt binder, Washington, D.C., AASHTO, 2021.
- [39] SOBREIRO, F.P., "Efeito da adição de ácidos fosfóricos no comportamento reológico de ligantes asfálticos puros e modificados com copolímero SBS". doi: http://dx.doi.org/10.11606/T.18.2014.tde-28052014-100359. Tese de D.Sc., USP, São Carlos, SP, Brasil, 2014.
- [40] MARINHO FILHO, P.G.T., "Avaliação reológica de ligantes asfálticos modificados com nanopartículas de dióxido de titânio". Tese de M.Sc., UFCG, Campina Grande, PB, Brasil, 2017.
- [41] NASCIMENTO, T.C.B., "Efeito do envelhecimento termo-oxidativo e foto-oxidativo nas propriedades reológicas do ligante asfáltico modificado", Tese de M.Sc., USP, São Carlos, SP, Brasil, 2015.