Binder performance and testing

First experiences in Argentina to modify commercial bitumen with nanosilica

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Abstract

Nanomaterials are extensively used to modify and improve different properties of construction materials. The asphalt modification with nanocomposites for pavement purpose is not the exception and can be found researches in the literature about the topic. The incorporation of nanocomposites is used to improves the bitumen characteristics to obtain asphalt mixture with better performance against rutting, fatigue and cracking. The potential of nanocomposites can be evaluated studying the rheological behaviour of modified bitumen. Rheological measures related to the performance in mixture elaborated with these modified bitumen can be measured and result the best way to analyses them. In Argentina there are not investigations at the moment about bitumen modification with nanocomposites. In this work, the incorporation of nanosilica at commercial conventional bitumen was studied. These represent a preliminary study about the changes measured by the incorporation of nano silica in traditional properties as well as in the rheological measures like the performance grade and other performance related properties of bitumen (low shear viscosity, Multiple stress creep recovery test (MSCR), fatigue binder test, etc.). Additionaly, rutting and fracture performance test on asphalt mixture elaborated with these modified bitumens were done. The nanosilica improves traditional properties like viscosity, and softening point as was expected. The performance grade was modified, the high temperature was increased and better behaviour at low and intermediate temperatures was presented. The nanosilica modified bitumens shown lower permanent deformations in the MSCR test and also improvements in asphalt binders fatigue behaviour.

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1. INTRODUCTION

Asphalt pavements are one of the most expensive elements on road construction. In addition, is one more maintenance investment requires. Asphalt concrete supports traffic and weather conditions and its performance becomes crucial to offer resistance to vehicle loadings, reduce the stress to the lower layers and support the changes in temperature and weather conditions [1]. Thereafter, a proper and durable asphalt pavement is important. Obtain better asphalt concretes performance becomes crutial. A way to achieve this is enhancing the bitumen behaviour.

Nanomaterials are extensively used in engineering to improve the properties of traditional construction materials like steel and concrete [2-5]. The addition or incorporation of nanocomposites into bitumen is not the exception. Different researches were reported about modified bitumen with nanocomposites [6-12]. There, it was found that their addition improves bitumen and asphalt mixture performance against rutting, fatigue, cracking and moisture sensitivity. Besides, it was found that the addition of nanocomposites to the bitumen also improves ageing [8], asphalt self-healing [9] and rheological behaviour of bitumen [12].

Nanosilica is usually defined as an amorphous nano-sized silicon dioxide powder with high specific area. It is produced under different production methods, such us sol-gel, vaporization, precipitation or biological processes [13]. Nanosilica can be used as a powder or a colloidal suspension [14]. Yao et al. [15] and Zafari et al. [16] studied the addition of nanosilica in the bitumen and concluded that nanosilica can be used to improve anti-aging properties. Also, other researchers shown the beneficial effect of nanosilica in bitumen properties and asphalt behaviour.

The potential of nanocomposites can be evaluated studying the rheological behaviour of modified bitumen. The rheological properties of bitumen, which contribute significantly to the final performance of the asphalt pavement, theoretically could be related to asphalt structures at the micro and nano scales [17]. The Superpave bitumen specification is based on a similar premise. Rheological measurements related to the performance of asphalt with nanosilica modified bitumens can provide useful information to characterize it [18-19].

Currently, there is no research in Argentina regarding modified bitumen with nanocomposites. In this study, the incorporation of nanosilica to a commercial conventional bitumen was studied. The obtained results represent a preliminary study about the changes measured on the bitumen when are modified with nanosílica. Traditional and rheological properties (like the performance grade and other performance related properties of bitumen; Low Shear Viscosity, Multiple Stress Creep Recovery test (MSCR), fatigue binder test) were measured. Additionally, performance test, rutting and fracture on asphalt elaborated with these modified bitumens were done.

2. EXPERIMENTAL

2.1. Materials

The study was developed over a conventional bitumen (CA-30 of Argentinian specification) which was blended with nanosilica (NS) from Dalian Fuchang Chemical Group Co. Ltd, distributed with the trade name of FUSIL 200. Table 1 and 2 show the main characteristic of these materials. NS was incorporated to the bitumen in dosages of 1, 2 and 4 % by weight of bitumen by means of a high-Speed/high-Shear mixer Silverson L5M-A following the next procedure. The bitumen was heated in an oven up to 160 °C and then placed in a hot plate to maintain the temperature. A surfactant was added into the bitumen during a few minutes. The surfactant helps to obtain a proper NS dispersion into bitumen. Then, the NS was slowly incorporated to the bitumen and mixed during one hour at 3000 RPM.

With the objective of better characterization, a sample of neat bitumen without NS (the control bitumen) was mixed in the Silverson with the surfactant during one hour to expose it to the same procedure and potential aging that the bitumens with NS. The different bitumens were submitted to RTFOT (EN 12607-1) and PAV (AASHTO R28) aging process to further characterization.

Table 1. Main characteristics of base bitumen.

Penetration at 25 ºC (0.1mm)	EN 1426	58
Softening point (ºC)	EN 1427	51.8
Viscosity at 60 ºC (Pa.s)	EN 12595	304.5
performance Grade (PG)	AASHTO M320	64-22

Table 2. Physical and chemical properties of nanosilica

Specific Surface BET (m2/g)	200±15
pH pH Value in 4% dispersion	3.8-4.2
Loss on Drying (105°C, 2h) (wt%)	≤ 1.5
Loss on ignition (1000°C, 2h) (wt%)	≤ 2
SiO2 (wt%)	99.9
Apparent Density (g/l)	40-46

The performance studies on asphalt concretes were done on a coarse dense asphalt (CAC19 in Argentina. AC22 in Europe). The aggregate proportion and principal properties of this asphalt are shows in Table 3.

Table 3. Asphalt concrete characteristics.

Asphalt Proportions		**CAC19 (AC22)**
Coarse aggregate 6-20 mm (Granitic)	[%]	25.7
Coarse aggregate 6-12 mm (Granitic)	[%]	11.4
Fine aggregate 0-6 mm (Granitic)	[%]	55.2
Filler (Limestone)	[%]	2.86
Asphalt binder	[%]	4.80
Main asphalt properties		
Bulk density	[g/cm^3]	2.420
Air voids	[%]	3.9
Mineral Air Voids	[%]	15.7
Stability	[kN]	17.3
flow	[mm]	3.0

The bitumen and asphalts were identified according to next nomenclature, C for control bitumen (without NS) and NS1, NS2 and NS4 in reference to the dosage of NS (1, 2 and 4 % respectively).

2.2 Test procedures

The main objective was to evaluate the potential improvement in a conventional bitumen by the addition of a nanocomposite like the NS. Several traditional and rheological tests were performance on the bitumens. Additionally, performance tests on asphalt concretes with these bitumens were done. Table 4 shows the testing plan followed.

Table 4. Testing plan.

Traditional test on bitumen	**Standard**	**T (°C)**
Penetration	EN 1426	25
Softening point	EN 1427	-
Ductility	ASTM D113	25
Viscosity	EN 12595	60-135-150-170
Rheological test on bitumen		
Superpave Performance Grading (PG)	AASHTO M320	-
Multiple Stress Creep Recovery (MSCR)	[19]	60
Low Shear Viscosity (LSV)	[20]	60
Frequency sweep at different Temperatures	[21]	10 to 70
Time sweep	[21]	20 and 30
Performance test on asphalt		
Wheel tracking	EN 12697-22 B	60
Fracture	[22]	0
Indirect Tensile Stress (ITS)	EN 12697-23	25

3. RESULTS

3.1 bitumen with NS properties

The objective of this work was to observe the impact on several properties of the bitumen by the addition of NS in different dosage. Initially the traditional properties characterization was investigated. Table 5 shows the main properties results. The incorporation of NS decreases the penetration and increases the softening point as well as the viscosity at 60 °C. The viscosity was increased in all temperatures measured; it seems to be notably increased whilst the dosage of NS increases. This offer advantages in front rutting at high temperatures. The ductility of original bitumens are not affected with dosages of 1 and 2 % of NS. After RTFOT, ductility of NS bitumens were affected but met the specification requirements. The ductility of NS4 was affected for both states, original and RTFOT. A minor loss of mass was observed in asphalt with NS. An interesting aspect is observed with the durability index, it remains the same for all the bitumens studied despite the increase in viscosity. It can be observed that the addition of NS increases the elaboration temperature of asphalt. In the case of NS4 this temperature could result disadvantageous.

Table 5. Traditional properties results.

		C	**NS1**	**NS2**	**NS4**	**Argentinian Specification**
Penetration	(0.1 mm)	51	50	47	45	-
Softening point	(ºC)	52.8	55.6	56.4	56.0	-
Ductility	(cm)	> 150	> 150	> 150	110	> 100
Viscosity	(Pa.s)					
T (ºC)						
60		371.9	406.0	497.0	644.0	240 – 360*
135		0.61	066	0.85	1.14	> 0.35*
150		0.30	0.35	0.42	0.56	
170		0.14	0.16	0.19	0.25	
Mixing temperature	(ºC)	166	169	174	182	
Compaction temperature	(ºC)	154	157	161	168	
RTFOT						
Loss of mass	(%)	0.19	0.19	0.14	0.13	-
Ductility	(cm)	> 150	100	72	85	> 50
Viscosity @ 60 ºC	(Pa.s)	744.0	786.0	1024.0	1374.0	
Durability Index**		2.0	1.9	2.1	2.1	< 3.5

*This range of viscosity is for the original bitumen. Here C was submitted to the same ageing that suffer the modified bitumens to compare.
**Ageing Viscosity (60 °C) to Original viscosity (60 °C) ratio.

According to the Superpave Performance grading, all bitumen with NS were ranked as PG 64-22 like the former neat bitumen. However, the NS influence the behaviour of base bitumen. Figure 1 shows what is called the high temperature of "true PG". This is the maximum temperature at which the thresholds values of Superpave standard ($G^*/sen\delta$) are reached for both original and RTFOT states. It can be observed how the addition of NS improved the behaviour of bitumen, increasing the windows of high temperature in which the asphalt can be exposed without suffering rutting. It is interesting to observe the delta of temperature (ΔT) between both states, original and RTFOT. It can be seen that C experiments a higher ΔT (4 °C). Meanwhile, in NS bitumens, both temperatures are closer (ΔT around 1.6 to 2.4). It is important to remember that, the minor of both temperatures rule the final PG grade at high temperature. The bitumen C shown more impact after the RTFOT than bitumens with NS. This is related with the RTFOT is highly depending on the viscosity of the material used. However, according to PG standard this will be the true high temperature of bitumen.

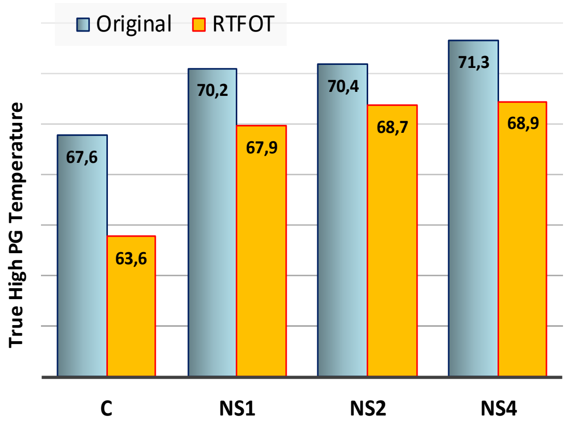

Figure 1. True high temperature of the PG.

Regarding fatigue behaviour of bitumens, Table 6 shows the G*.senδ values measured at different temperatures to define the performance grading. It can be seen how the NS bitumen has lower values than C at all temperature studied. In consequence modified bitumens, and indirectly the asphalts, should have better fatigue behaviour. This improvement was observed in time sweep tests (Figure 2). It can be observed in Figure 2-above how the NS bitumens endure more loading cycles than C until G* start to drop. When analysing the fatigue behaviour in terms of Dissipated Energy Ratio (DER), Figure 2-below, the NS bitumens accumulate less DER than C during the test. In other words, less damage occurs in the bitumen for the same type of loading cycles. It is important to mention that curve DER-Cycles of C is hidden by the curve of NS1 in the graph at T = 20 °C.

Table 6. G*.senδ at different temperatures of bitumens studied.

T (°C)	G*.senδ (MPa) < 5 MPa [AASHTO M 320]			
	C	NS1	NS2	NS4
22	3.14	1.90	2.75	2.15
25	2.13	1.31	1.91	1.38
28	1.44	0.90	1.33	0.94

Regarding the MSCR test, the addition of NS improves the behaviour of bitumens. The NS bitumens shown a reduction in the final strain and $Jnr_{3.2}$ values in comparison with C (Table 7). NS only affected the stiffness of bitumen; it did not affect the elastic recovery at all. Obviously, C, being a conventional bitumen, does not show an elastic recovery in the MSCR. The NS bitumen maintain this behaviour as it was expected. It is important to mention that the MSCR performed is an implementation developed in our laboratory and is similar to the MSCR test of ASTM D7405. The main differences are in the load-rest period with 2-18 s instead of the 1-9 s of ASTM standard and the number of cycles applied in each load level is 5 instead of 10 cycles of the ASTM standard. This changes response to resolution issues of our DSR to carried out the MSCR test with load-rest periods of 1-9 s.

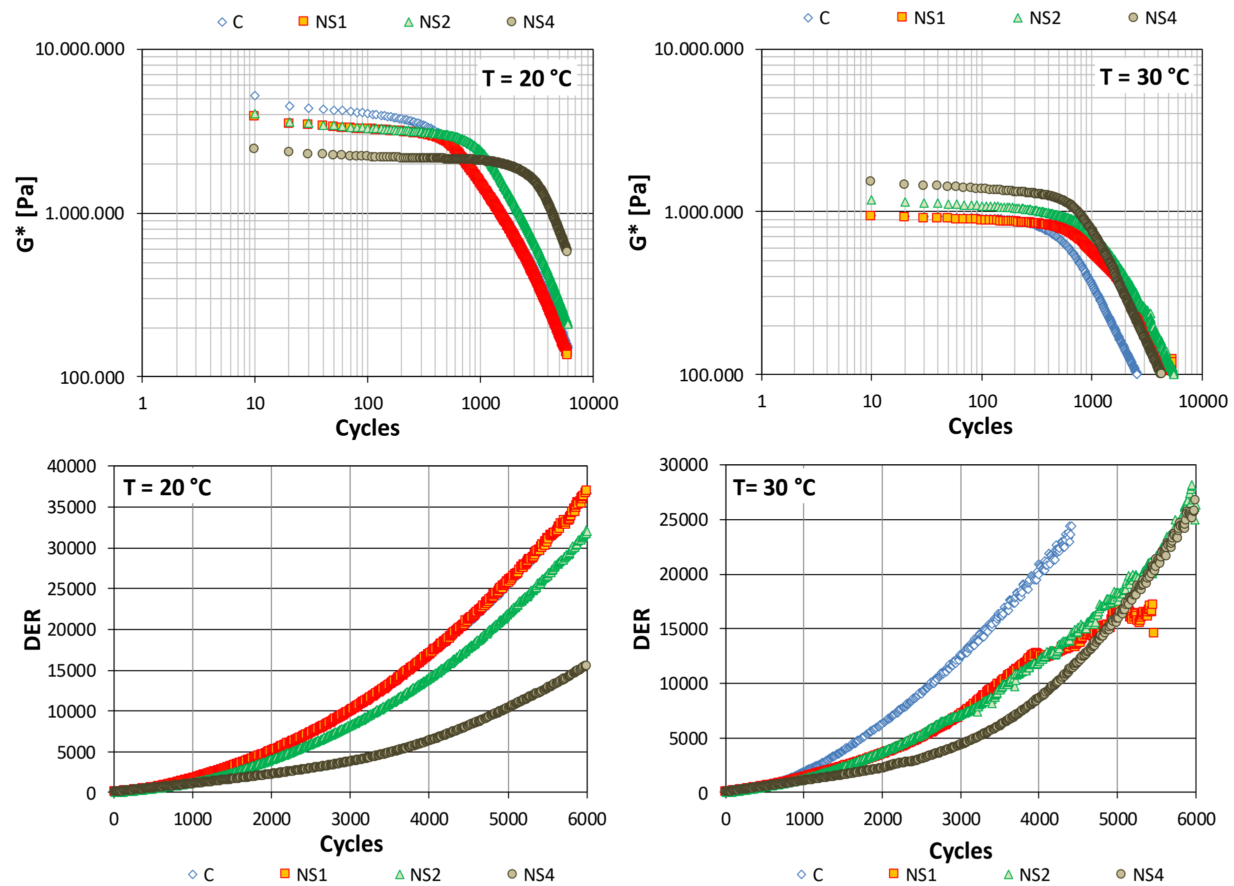

Figure 2. Result of time sweep tests. Above: G* vs Cycles. Bellow: Dissipated Energy Ratio (DER) vs cycles.

Table 7. MSCR results

	C	NS1	NS2	NS4
Final strain	98.1 ± 6.3	91.9 ± 4.0	73.4 ± 1.9	57.1 ± 0.5
$Jnr_{3.2}$	6.03 ± 0.43	5.70 ± 0.29	4.57 ± 0.13	3.52 ± 0.04

Other rheological test carried out on the bitumens was the frequency sweep test at different temperatures. Figure 3 shows these results in a form of Black diagram. As it can be seen, all the bitumens present similar behaviour at low temperatures or high frequencies (upper part of the graph with high values of G*). The influence of NS can be observed at high temperatures or low frequencies range. The presence of NS makes the behaviour of NS2 and NS4 more Newtonian. However, these changes are slightly significant.

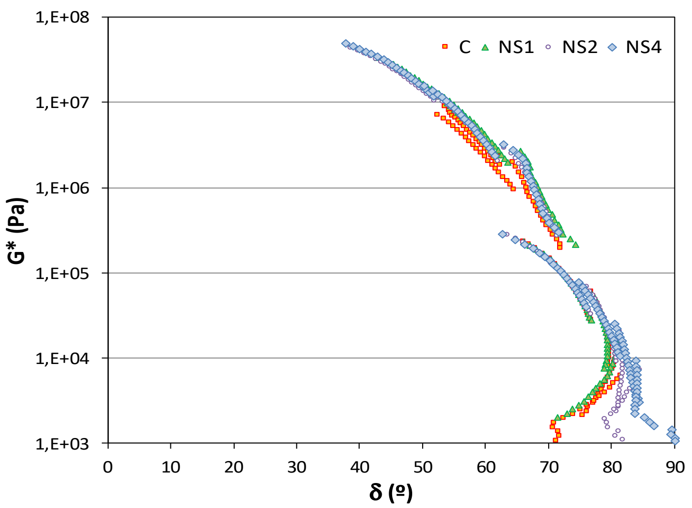

Figure 3. Black diagram of bitumen studied from frequency sweep test results.

From the frequency sweep test the Low Shear Viscosity (LSV) of bitumens can be calculated at different temperatures [20]. Table 8 shows the LSV values at the reference temperature of 60 °C. This values shows a similar trend that bitumens viscosities at 60 °C shown in Table 5.

Table 8. LSV results

		C	NS1	NS2	NS4
LSV @ 60 °C	**(Pa.s)**	414.1	349.9	418.9	630.8
Standard deviation	**(Pa.s)**	20.0	15.1	22.1	30.1

3.2 Asphalt performance results

One of the objectives of this research was to verify the behaviour of NS bitumen in asphalts to see the potential improvement trough out some performance tests. Figure 4 shows the result on rutting tests. It can be observed how the asphalts with NS bitumens shows notable reduction in rut depth. Since the point of view of Wheel Tracking Slope (WTS), the asphalts have values of 0.12, 0.059, 0.042 and 0.034 mm/10^3 cycles for C, NS1, NS2 and NS4 respectively. The WTS was reduced by more than a half in asphalts with NS bitumens. In fact, the result of NS2 and NS4 asphalts are comparable to results obtained for the same asphalt gradation elaborated with SBS polymer modified bitumen in our laboratory. In reference to the wheel tracking limits of Argentinian standard [23] or PG 3 of Spain [24], it can be said that NS asphalts competently meet their more rigorous requirements. Regarding the Proportional Rut Depth (PRD) a similar trend like WTS was observed.

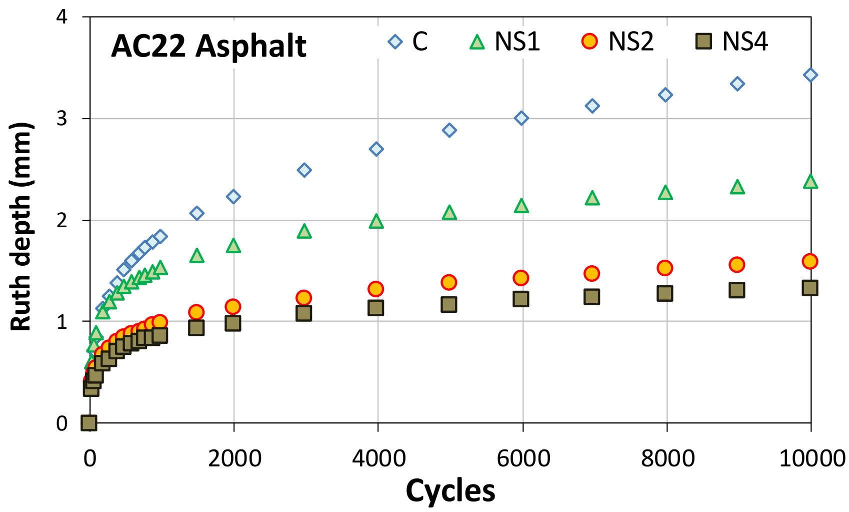

Figure 4. Rutting results in Wheel Tracking test.

Table 8 shows the fracture test result at 0 °C obtained in the Notched beam bending test [22]. Table 8 shown the peak stress and toughness results of different asphalts studied. These are the main fracture test values. The peak stress is the maximum tensile stress that generate the crack initiation on the notch of the beam. Then the test continues until the failure controlled by the Crack Opening Displacement (COD) at a constant rate of 1 mm/s up to 4 mm of crack opening. The toughness represents the area bellow the curve stress-COD up to a COD of 3 mm. As can be seen, the NS bitumens increase the values of peak stress of asphalts, resulting in fracture resistance improvements (24, 30 and 38 % for NS1, NS2 and NS4 respectively in comparison with C asphalt). Furthermore, these improvements do not impact unfavourably in the toughness of asphalt that remains similar to the C asphalt. In general terms it could be expected that an increase in peak stress will be traduced in a more fragile response (a minor toughness). It is important to note the trend is a reduction of toughness with the increase of NS for NS1 to NS2 while NS4 show a higher toughness. However, these changes are slightly significant. Figure 5 shows the results obtained in the ITS at 25 °C. It is clear that NS improves the tensile strength of asphalt.

Table 8. Fracture test results

	Peak Stress	Toughness
	(Mpa)	(J/m^2)
C	6.63 ± 0.19	658.6 ± 49.0
NS1	8.23 ± 0.43	610.7 ± 53.8
NS2	8.65 ± 0.34	588.0 ± 68.1
NS4	9.15 ± 0.26	678.0 ± 60.0

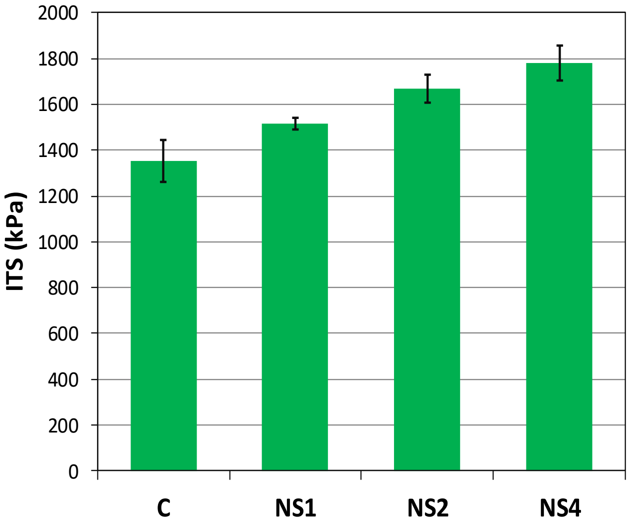

Figure 5. Indirect Tensile Stress (ITS) at 25 °C results.

4. CONCLUTIONS

This research studies the addition of nanosilica in conventional bitumen. Traditional and rheological properties of bitumens were analysis as well as the impact of these bitumens in the performance of a dense asphalt concrete. The main conclusions are:

- The addition of nasosilica had a positive impact in the main properties of the bitumen studied. Reduction in penetration, increases in viscosity and softening point. Other properties like ductility, durability Index or loss of mass were less affected by the additive used.

- The performance grading of asphalts with nanosilica were enhanced at high and medium temperatures. Also, rheological test shown these improvements with better behaviour in rutting and fatigue rheological test like Multiple Stress Creep Recovery and time sweep tests.

- Regard asphalt performance, the addition of nanosilica improves the rutting behaviour, the fracture resistance (with less effect on toughness) and increases the indirect tensile stress at 25 °C of the dense asphalt concretes studied.

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