Effect of using Nano-clay to improve Asphalt materials

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Abstract---

The objective of this study is to review existing literature in the area of Nano-modification of asphalt and proceed to apply Nano-materials to asphalt to improve the performance. This study integrates literature review, preparation and characterization of Nano-modified asphalt materials.

This study including aggregate tests to prove that these aggregate accepted to use it in asphalt binder, and tests of used bitumen samples.

In this study the experimental testing three types of Nano clay (A, B, C) at 2% and 4% by the weight of asphalt were blended in asphalt binder at high temperature to exfoliate the Nano clay within the asphalt. The asphalt binder was then characterized using the AASHTO viscosity, penetration test, softening point, flash point and Marshall Test.

These types of additives of Nano-clay are increasing the viscosity by increasing the percent of Nano clay

and decreasing the penetration by increasing the percent of Nano clay but not affected with softening point and flash point. About Marshall Stability there are many changes:

In case of adding Nano clay (A-2%) Marshall Stabilization decrease; although it increased again when adding Nano clay (A-4%) but not reached to the original sample stabilization.

In case of adding Nano clay (B-2% and 4%) it still increase.

In case of adding Nano clay (C-2%) it increased; although when adding Nano clay (C-4%) it decreases again (bell shape).

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I. Introduction

A. Asphalt materials:

In the United States, transportation infrastructure investments account for 7% of the gross domestic product (GDP) according to the National Asphalt Pavement Association [3]. Over 550 million tons of hot-mix asphalt (HMA) produced annually for construction projects. Increasing traffic loads and traffic volume, combined with the rising cost of asphalt, have led to an urgent need to improve the durability safety and efficiency of asphalt pavements through asphalt modification. Asphalt mixtures are composed of very irregular aggregates bound together with hydrocarbon-based asphalt, with a low volume fraction of voids dispersed within the matrix. Polymer-modified asphalts have increasingly been used over the last decade to minimize low-temperature cracking and high temperature rutting while improving the fatigue cracking resistance of asphalt concrete [4-5].

Polymer modifiers fall into one of two major elastomeric (rubber/polymer) categories: (1) modifiers including styrene/butadiene polymers, natural rubber and crumb rubbers (both virgin and recycled) [6-7]. (2) Blastomeric modifiers such as polypropylene and polyethylene [8]. In the experimental testing montmorillonite, Nano clay at 2% and 4% by weight of asphalt were blended in asphalt binder at a high temperature to exfoliate the Nano clay within the asphalt. The asphalt binder was then characterized using the super pave rotational viscosity, dynamic shear modulus and direct tension test. The rotational viscosity results indicate that the addition of the two types of Nano clay (Nano clay 1 and Nano clay 2) increased the



rotational viscosity by an average of 41% and 112% respectively. It was found that the dynamic shear complex modulus(G) value increases significantly across a range of testing temperature(from 13^{0} to 70^{0} c) and loading frequencies(0.01-25hz)by an average of 66% and 196%. In terms of direct tension strength the use of Nano clay A and Nano clay B reduced the strain failure rate of the original binder while the secant or direct tension modulus showed increase with the addition of the Nano clays [1].

B. Nanotechnology and the current development in pavement materials:

Nanotechnology is the creation of new materials, devices and systems at the molecular level as phenomena associated with atomic and molecular interactions strongly influence macroscopic material properties[9].Even though engineers are interested in material properties at the macro and miso scales, the Nano and micro scales provide fundamental insight for the development of science and technology. Although improvements in asphalt performance have been achieved through polymer modification, it will be interesting to explore what nanotechnology offers in improving asphalt pavement performance.

In August 2006, the NSF Workshop on Nano modification of Cementitiuos Materials was held to develop a "Roadmap for research" for Portland cement concrete and asphalt concrete using nanotechnology. The Nano science and nanotechnology may lead to progress in asphalt pavement technology [10]. Researchers [11] are planning to form an ad hoc consortium to study and developed tools to model asphalt's physio-chemical properties that effectively establish the field of "asphalt Nano-material science". Greenfield [12] has studied the molecular simulation of asphalt like materials. Birgisson [13] envisions that Nano studies are needed to develop safe and sustainable pavement infrastructure, starting that the future life of pavements could reach two to three times their current life. Other researchers have also initiated investigations into the use of nanotechnology in Portland cement concrete [14-15].

Ideal asphalt [16] should possess both: (1) high relative stiffness at high services temperatures (summer) to reduce rutting and shoving. (2) Increased adhesion between asphalt and aggregate in the presence of moisture to reduce stripping. The project team conducted preliminary tests, blending small percentages of Nano clay-composites into virgin asphalt with the hope of producing a binder that is less susceptible to high-temperature rutting and low-temperature cracking. The motivation is to significantly reduce the temperature sensitivity of the binder at service temperature while maintaining workability at construction temperatures. Nanomodified asphalt may potentially improve the rutting, crack and fatigue resistance of asphalt mixtures.

II. Literature review:

A. Asphalt chemical components and modification efforts:

Most asphalt molecules, primarily consisting of carbon and hydrogen; also contain one or more of the elements sulfur, nitrogen and\or oxygen [16]. Within asphalt, heteroatoms typically replace carbon atoms in the asphalt molecule; the interaction of heteroatoms and the hydrocarbons lead to the unique chemical and physical properties of various asphalt mixtures [17].

Asphalt molecules are classified in two major fractions based on solubility. These fractions are the asphaltenes and maltenes[17]. Both the asphaltenes and maltenes constitute the non-volatile, highmolecular-weight fractions of petroleum but the maltenes constitute the fraction of asphalt which is soluble in n-alkane solvents such as pentane and heptane. The three basic types of molecules in asphalt are aliphatic, cyclic and aromatics.

Research conducted in the Netherlands suggests Nano clay modification improves some characteristics of asphalt binders and asphalt mixtures, but more research is required before it can



be applied on a large scale [18]. The major reason for additional research is that while the Nano clay increased the rutting resistance little is known about how it mitigates the fatigue problem of asphalt. In china, similar research has been conducted on Nano calcium carbonate (Nano caco₃) modified asphalt [19-20]; it was found that the nano-caco₃ can enhance asphalt's rutting-resistance as-well-as improve its low-temperature toughness. It was found that the mixture of Nano caco₃ and asphalt forms a uniform and steady system which improves temperature susceptibility of asphalt at high temperatures. However, the mechanism of asphalt material behavior with this modification is not well understood.

In china, Yu has studied the effect of Nano clay (montmorillonite) on properties of styrenebutadiene- styrene (SBS) copolymer modified asphalt [21] by melt blending with different contents of sodium montmorillonite (Na-MMT) and organophilic montmorillonite (OMMT). It was found that the addition of Na-MMT and OMMT increases the viscosity of SBS-modified asphalt. In addition, the Nano clay\SBS modified asphalt gained a higher complex modulus and lower phase angle, implying stiffer and more elastic asphalt. Therefore, the Nano clay-modified asphalt was determined to have good rutting resistance compared to the original asphalt or the SBSmodified asphalt. It was found that MMT-modified asphalt may form an intercalated structure, whereas the OMMT modified asphalt may form exfoliated structure based on the X-ray diffraction (XRD) results [22-23].

B. Nano clay, montmorillonite, and organic modification of Nano clay:

Layered silicates (Nano clay) are widely used in the modification of polymer matrices to realize significant improvement in mechanical, thermal and barrier properties. One of the most frequently used layered silicates is montmorillonite (MMT), which has a 2:1 layered structure with two silica tetrahedron sand wishing an alumina octahedron. Roy et al. [24-25] enhanced the compressive and shear strength of thermoplastic polymers using only a small weight percent of Nano clay reinforcement.

When the polymer penetrates between the adjacent layers of the Nano clay, the gallery spacing is increase and the resulting morphology is an intercalated structure [26]. An exfoliated morphology occurs when the clay platelets are extensively delaminated and completely separated due to thorough polymer penetration by various dispersion techniques. In combination with Nano clay, coupling agents such as saline are used to disperse the Nano clay in a stable manner [MMT surface modification]. Also saline coupling agents have been observed to enhance the bonding of Nano clay to the polymer matrix.

III. Laboratory testing and results:

A. Laboratory program design:

The research team used the information gathered from the literature review and evaluations conducted to shape the direction of the laboratory program. (1) The US-127 asphalt binder modified with 2% and 4% of first Nano clay (Nano clay A) by weight of asphalt binder. (2) The US-127 asphalt binder modified with 2% and 4% of second Nano clay (Nano clay B) by weight of asphalt binder.

(3) The US-127 asphalt binder modified with 2% and 4% of Third Nano clay (Nano clay A) by weight of asphalt binder. -Viscosity Grade bitumen's are specified by the methods described in ASTM Standard Viscosity test according to (AASHTO T201-10). applied Viscosity Graded specifications covers bitumen (asphalt) graded by Viscosity at 60 C (140 °F). -Penetration test (Rheological properties) of asphalt binder using penetration test according to (AASHTO T49-07).

-Softening point test according to (AASHTO_T53-09) -

Flash point test according to (AASHTO T 73 and ASTM D 93).

Marshall TEST according to (AASHTO_T48-06).



B. Preparation of the Nano clay particles:

• Chemical composition:

 $TABLE \ (1): NANO \ CLAY \ TYPE \ (A) \ ALUMINUM \ OXIDE \ (XRDML)$

Pos. [°2Th.]	Height [cts]	d- spacing [Å]	Rel. Int. [%]	Crystall ite Size only [Å]	Micro Strain only [%]
20.2849	19.68	4.37794	8.83	77.15447	2.837126
20.8988	43.84	4.25069	19.67	316.0828	0.672402
26.6757	222.86	3.34184	100.00	1230.287	0.135815
27.9631	44.90	3.19084	20.15	237.5981	0.671478
29.6044	25.83	3.01756	11.59	118.0452	1.278139
35.6577	24.11	2.51796	10.82	59.79444	2.105515
+50.199	43.63	1.81743	19.58	343.1427	0.264821
60.0521	26.16	1.54066	11.74	266.4789	0.289077
67.4105	25.06	1.38927	11.25	54.72719	1.269272

Table (2): Pattern List of Nano clay (A):

Visi ble	Ref. Code	Sco re	Compo und Name	Displace ment [°2Th.]	Scale Factor	Chemic al Formul a
*	01-075- 1568	42	Magnesi um Aluminu m Silicon Oxide	0.854	0.996	Mg.6 Al1.2 Si1.8 O6
*	04-009- 8364	15	Magnesi um Aluminu m Silicate	0.178	0.071	Mg3 Al2 (Si O4)3

Table (3) shows the chemical composition of Nano clay type (B).

Table (5) shows the chemical composition of Nano

clay type (B).

Table (6) shows the pattern list of Nano clay type (B).

TABLE (5): NANO CLAY TYPE (C) ALUMINUM OXIDE (XRDML)

Visi ble	Ref. Code	Sco re	Compo und Name	Displac ement [°2Th.]	Scale Factor	Chemic al Formul a
*	04-012-	30	Magnesi	0.919	0.057	Mg0.2
	5002		um			Al0.4
			Aluminu			Si0.6 O2
			m			
			Silicon			
			Oxide			
*	01-084-	8	Magnesi	0.076	0.128	Mg2 Al4
	1222		um			Si5 O18
			Aluminu			
			m			
			Silicate			

Table (4) shows the pattern list of Nano clay type (B)

.TABLE (3): NANO CLAY TYPE (B) ALUMINUM OXIDE (XRDML)

TABLE (4): PATTERN LIST OF NANO CLAY (B):

DOCUMENT HISTORY:

Pos. [°2Th.]	Height [cts]	d-spacing [Å]	Rel. Int. [%]	Crystall e Size only [Å	Stra	ain
20.8998	96.79	4.25049	12.60	685.7489	0 0.30	9916
26.7400	768.17	3.33395	100.00	691.8370	00 0.24	0949
29.5396	63.05	3.02404	8.21	695.6282	20 0.21	7360
31.6411	230.99	2.82783	30.07	698.8600	00 0.20	2317
35.4393	15.09	2.53298	1.96	59.75792	20 2.11	9365
44.7036	22.98	2.02722	2.99	335.8832	20 0.30	1775
50.2234	88.72	1.81660	11.55	743.6500	00 0.12	2141
60.0701	34.81	1.54024	4.53	781.8296	50 0.09	8502
61.1995	6.17	1.51450	0.80	44.06921	10 1.71	8319
68.1643	14.16	1.37574	1.84	137.8500	00 0.49	8998
Pos. [°2Th.]	Height [cts]	d- spacing [Å]	Rel. Int. [%]	Crystall ite Size only [Å]	Micro Strain only [%]	
20.9600	92.81	4.23842	18.69	685.7994	0.309013	
26.7543	3 496.49	3.33219	100.00	691.8549	0.240816	
29.5820) 55.55	3.01980	11.19	695.6901	0.217036	
35.4067	34.00	2.53524	6.85	95.72578	1.324219	
39.6102	2 26.05	2.27535	5.25	714.3006	0.159271	
50.2780		1.81476	9.70	743.8302	0.121987	
54.8663	6.40	1.67335	1.29	42.73775	1.957701	

60.0780	19.45	1.54006	3.92	359.3476	0.214285
61.8054	12.35	1.50110	2.49	44.20813	1.697764
65.6516	40.98	1.42217	8.25	809.3126	0.087863
68.3197	33.35	1.37299	6.72	376.5443	0.182314

TABLE (6): PATTERN LIST OF NANO CLAY (C):

Visi ble	Ref. Code	Sco re	Compo und Name	Displac ement [°2Th.]	Scale Factor	Chemic al Formul a
*	00-014- 0249	21	Magnesi um Alumin um Silicate	0.632	0.048	Mg2 Al4 Si5 O18
*	01-089- 8922	11	Magnesi um Alumin um Silicate	0.291	0.068	Mg3 Al2 (Si O4)3

• Fabrication of asphalt Nano composite:

The addition of a silane coupling agent was first carried out using the procedure outlined by Qian [27]. The surfactant modified Nano clay was dispersed in organic solvent (isopropanol0 and the desired amount of silane coupling agent was added with continuous stirring in a water bath until the solvent was completely evaporated. The products were then dried in a vacuum at approximately 80 C, wetted, and filtered using a 280 mesh. The surfactant-modified Nano clay in conjunction with the coupling agent was then dispersed in the asphalt. The asphalt Nano composite was fabricated using a high-shear mixer. The asphalt was first heated to 160 C to a fluid state and the surfactant-modified Nano clay, A, B or C, was added to the system and mixed at 2500 rpm for 3 h to disperse the intercalated MMT Nano clay. The detailed parameters such as temperature, speed, and mixing duration were adjusted to control the quality of the modified asphalt. Nano clay A and B are similar in terms of the structure.

c. Experimental results and analysis:

• Aggregate tests:

To make sure that this aggregate can be used in the asphalt mix or not; it should apply aggregate tests. -ASTM C131 Standard Test Method for Resistance to Degradation of Small Size Coarse Aggregate by Abrasion and Impact in the Los Angeles Machine.

Tests of aggregate1:

Total Specific Weight	2.554gm\cm3
Saturated Specific Weight	2.596gm\cm3
% Absorption	1.6%
%Degradation	0.6%

1-LOS ANGELES TEST:

No; of r.p.m	Retained weight(gm)	Losses weight(gm)	%losses
100	4685	315	6.3
500	3597	1403	28.06
600	3568	1432	28.64

2-SPECIFIC WEIGHT, ABSORPTION AND DEGRADATION:

Total Specific Weight	2.459gm\cm3
Saturated Specific Weight	2.537gm\cm3
%Absorption	3.2%
%Degradation	0.4%

Tests of aggregate2:

1-LOS ANGELES TEST:

No; of r.p.m	Retained weight(gm)	Losses weight(gm)	%losses
100	4754	246	4.92
500	3828	1172	23.44
600	3807	1193	23.86

2-SPECIFIC WEIGHT, ABSORPTION AND DEGRADATION:

Coverage of test procedures related to the characteristics of asphalt mixes and finished asphalt pavements. Tests include measurement and evaluation of performance parameters. The viscosity, penetration and Marshall test of bitumen improved by adding Nano clay.

The asphalt viscosity, penetration and Marshall differ after adding the Nano clay according to percentage; but softening point and flash point didn't affected. Table(7)and chart(1,2) show the results (viscosity-penetration) after adding Nano clay (A), table (8) and chart(3) show the results before and after adding Nano clay (A) in Marshall test ,table (9) and chart(4-5) show the results after adding Nano clay (B).Table(10) and chart(6) show

the results of Marshall test before and after adding Nano clay(B). Table (11) and chart (7-8) show the results after adding Nano clay (C).Table (12) and chart (9) show the results of Marshall Test before and after adding Nano clay(C).

W1=Weight of sample in air.

W2=Weight of sample after submerged in water for 2 minutes.
W3=Weight of sample in water.
V=Volume of sample.
G.S=Specific gravity of sample.
G.S avg= Average specific gravity of sample.
e=percent of air voids.
R1=Marshall Stability reading.
F=Failure load (bound).
C.F=Correction factor.

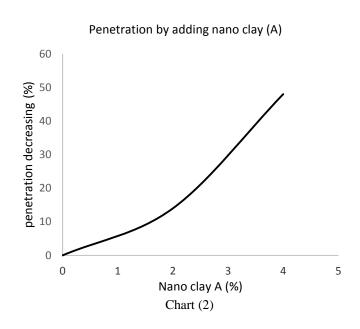


TABLE (7): RESULTS OF ADDING NANO CLAY (A) OF (2%-4%):

sample	%Additives (A2%)	%Additives (A4%)
viscosity	Increase11%	Increase70%
penetration	Decrease14%	Decrease48%
Softening point		

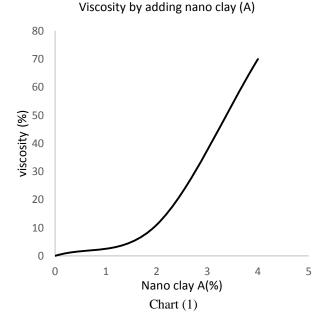


TABLE (8): RESULTS OF ADDING NANO CLAY (A) OF (2%-4%): (MARSHALL TEST)

%Nano clay	Orig sam			Adding(A) 2%		ng(A) %
Sample No;	1	2	3	4	5	6
W1	1198	1200	1204	1198	1200	1197
W2	1208	1211	1210	1209	1212	1209
W3	663	665	668	669	671	666
V	545	545	542	540	541	543
G.S	2.198	2.19	2.221	2.219	2.218	2.204
G.S avg		2.19		2.220		2.211
% e	8.4	8.4	12.0	12.2	11.0	11.4
R1	0.9	1.02	1	1	1.1	1.1
F	2354	2666	2615	2615	2875	2875
C.F	0.93	0.93	0.93	0.93	0.93	0.93
Stabilization(Ib)	2189	2185	2432	2432	2674	2674
Stabilization avg(Ib)	2187		2432		2674	
% stabilization		0%		11.2%		22.3%
Flow(1/100")	9	16.5	10	10	9	11



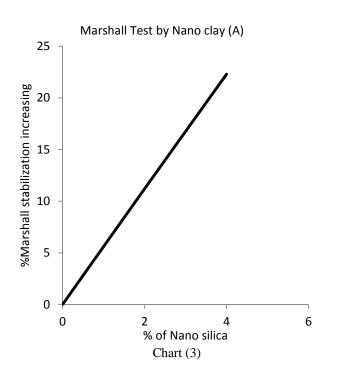
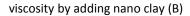
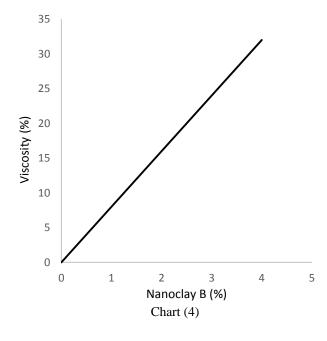
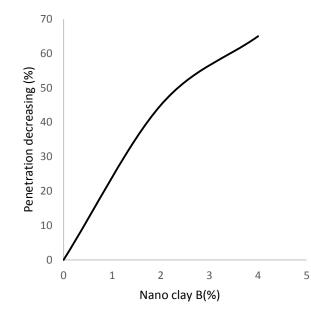


TABLE (9): RESULTS OF ADDING NANO CLAY (B) OF (2%-4%):

sample	%Additives (B2%)	%Additives (B4%)
viscosity	Increase16%	Increase32%
penetration	Decrease45%	Decrease65%
Softening point		







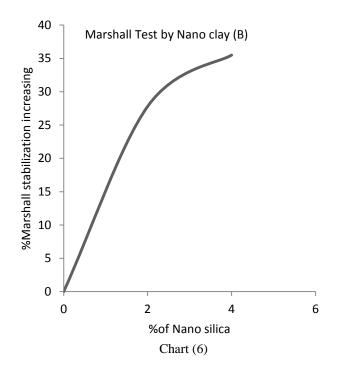
Penetration by adding nano clay (B)

Chart (5)

TABLE (10): RESULTS OF ADDING NANO CLAY (B) OF (2%-4%): (MARSHALL TEST)

%Nano clay	Orig	inal	Adding	g(B) 2%	Adding	(B) 4%
	sam	ple				
Sample No;	1	2	7	8	9	10
W1	1198	1200	1202	1201	1203	1198
W2	1208	1211	1208	1210	1209	1203
W3	663	665	670	671	673	672
V	545	545	538	539	536	531
G.S	2.198	2.19	2.234	2.228	2.244	2.256
G.S avg		2.19		2.231		2.250
% e	8.4	8.4	9.0	9.0	9.5	9.1
R1	0.9	1.02	1.1	1.2	1.2	1.2
F	2354	2666	2875	3136	3136	3136
C.F	0.93	0.93	0.93	0.93	0.93	0.96
Stabilization(Ib)	2189	2185	2674	2916	2916	3011
Stabilization avg(Ib)		2187		2795		2964
% stabilization		0%		27.8%		35.5%
Flow(1/100")	9	16.5	9	10	10	11





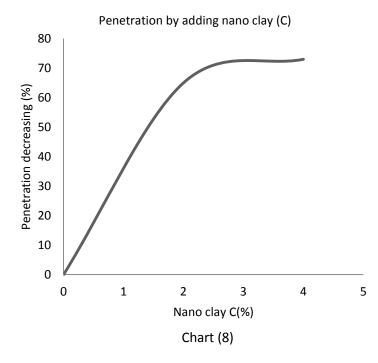
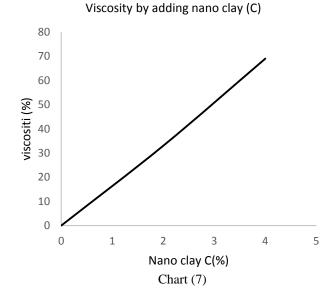


TABLE (12): RESULTS OF ADDING NANO CLAY (C) OF (2%-4%): (MARSHALL TEST)

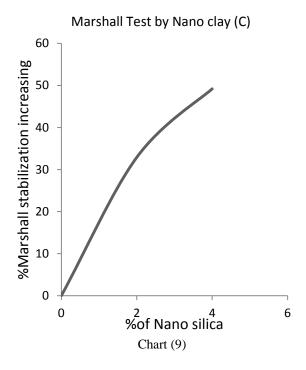
%Nano clay	Orig sam		Adding	(C) 2%	Adding	(C) 4%
Sample No;	1	2	11	12	13	14
W1	1198	1200	1199	1197	1197	1206
W2	1208	1211	1204	1207	1203	1209
W3	663	665	660	650	670	675
V	545	545	548	557	533	534
G.S	2.198	2.19	2.188	2.149	2.246	2.258
G.S avg		2.19		2.168		2.252
% e	8.4	8.4	8.4	8.5	12	12.2
R1	0.9	1.02	1.2	1.3	1.3	1.3
F	2354	2666	3399	3136	3399	3399
C.F	0.93	0.93	0.89	0.89	0.96	0.96
Stabilization(Ib)	2189	2185	3025	2791	3263	3263
Stabilization avg(Ib)		2187		2908		3263
%stabilization		0%		32.9%		49.2%
Flow(1/100")	9	16.5	9	10	12	13

sample	%Additives (C2%)	%Additives (C4%)		
viscosity	Increase33%	Increase69%		
penetration	Decrease65%	Decrease73%		
Softening point				

TABLE (11): RESULTS OF ADDING NANO CLAY (C) OF (2%-4%):







Conclusion:

These types of additives of Nano-clay improved the viscosity, penetration and Marshall Stabilization of bitumen according to the type and percent:

-Nano clay (A-2%) increased viscosity by 11%, decrease penetration by 14% and decrease Marshall Stabilization by 9.56%.

- Nano clay (A-4%) increased viscosity by 70%, decrease penetration by 48% and decrease Marshall Stabilization by 3.16%.

- Nano clay (B-2%) increased viscosity by 16%, decrease penetration by 45% and increase Marshall Stabilization by 5.07%.

- Nano clay (B-4%) increased viscosity by 32%, decrease penetration by 65% and increase Marshall Stabilization by 6%.

- Nano clay (C-2%) increased viscosity by 33%, decrease penetration by 65% and increase Marshall Stabilization by 10.6%.

- Nano clay (C-4%) increased viscosity by 69%, decrease penetration by 73% and decrease Marshall Stabilization by 2.01%.

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