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Effect of SBR/Waste Ceramic Powder Nanocomposite on High Temperature Performance of Asphalt Binder

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Abstract

Modifying asphalt binder with styrene butadiene rubber (SBR) improves low temperature performance (resistance to cracking), elastic recovery, and ductility of binder. However, some of difficulties were found at high temperatures susceptibility (Rutting), and low storage stability (compatibility). Therefore, the aim of this study is to resolve these defects (rutting and compatibility) by formulating SBR/ waste nanoceramic powder (WNCP) nanocomposites. Three nanocomposites formulations are fabricated from SBR/WNCP with mass ratios (90:10), (80:20), and (70:30) respectively. The prepared nanocomposites are assessed by XRF, XRD, and DLS. Asphalt binder is modified by 5wt% from each fabricated nanocomposite. Physical tests of modified asphalt binder including softening point, penetration, and penetration index are measured. The high temperature performance; rutting factor (G*/sindelta) is studied with DSR before and after exposure to short term aging. The compatibility of modified binder was studied by conducting storage stability test. Results confirmed that fabrication of SBR/WNCP nanocomposite enhance the compatibility, storage stability, and stiffness of blended binder. Also, DSR results approved that the rutting resistance of binder is enhanced.

Keywords: Asphalt binder; SBR; Ceramic powder waste; Rutting; storage stability; Nanocomposites.

1. Introduction

The high viscoelastic properties and good services performance of asphalt binder allow it to be widely used in pavement construction. However, various defects such as rutting at high temperatures and cracking at low temperatures are usually appeared in asphalt pavement due to the high sensitivity to temperature, climatic conditions changes, and high traffic loads [1]. Hence modifying asphalt binder had to be performed to develop high-quality, safe, reliable, and environment friendly pavement materials [2,3]. Various modifiers were used to enhance the rheological characteristics of asphalt binder; however almost previous research studies confirmed that the best results were obtained for polymers [4, 5,6, 7]. Between all of these polymer modifiers, SBR (Styrene butadiene rubber) is one of the most commonly used polymers for modifying asphalt [8,9]. According to an engineering report that is available at the US Federal Aviation Administration website [10], adding SBR to bitumen is beneficial and improves properties like low-temperature ductility and elastic recovery in asphalt. Also, it was found that SBR polymers increase flexibility and ductility of asphalt binder, which enhances crack resistance at low temperatures [11]. However, like other polymer modifiers, some of disadvantages have been reported for using SBR. It is easily oxidized after short-term aging due to a great amount of butadiene in SBR molecule [12]. Also, it has low compatibility with asphalt binder and easily separates from it when stored at high temperatures [13]. As a result, a third component is required to make more stable system, including clays such as Montmorillonite (MMT), organobentonite, organic

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Table (1) Physical properties and chemical constituents of asphalt (60/70)

Asphalt characteristics	Results	Standard
Physical properties		
Penetration (@ 25ºC, 100g,5s)0.1mm	62	ASTM D5
Softening point (ring and ball) °C	50	ASTM D36
Kinematic viscosity (@135 °C) cSt.	380	ASTM D2170
Chemical constituents		
Oils	45	ASTM D3297
Resins	32	
Asphaltenes	23	

palygorskite [14], weathered coal [15], and carbon black [16]. Recently, nanomaterials have been used to improve various properties of polymer modifiers including nanotubes [17], nanofibers [18], nanohydrated lime [19], nanosilica [20], polymerized powders, nanosized plastic powders, and nanoclay [21]. Fabrication of nanocomposite polymers donate unique mechanical properties in comparison to pure polymers or micro and macro composites [22, 23]. Behnam Amini approved that the existence of nanoclay (NC) in SBR improves storage stability, and increases the indirect strength of asphalt mixture. Zhang Baochang also used SBR/montmorillonite (MMT) nanocomposite for improving the viscoelastic properties of asphalt binder and its high resistance to rutting. The main objective of this study is to use waste nano ceramic powder (WCP) as filler for SBR instead of high cost modified nanoclay. The study aimed to save the environment from such wastes and produce low cost composites exhibiting attractive mechanical properties. Approximately 30% of products in the ceramic industry are regarded as waste [24]. To date, the utilization of these wastes as modifier for bitumen binder is rarely studied. Moreover, few studies used different industrial waste materials as fillers in hot mix asphalt [25,26]. Muniandy [27] reported the improvement of stiffness and rutting resistance by replacing 10% of conventional limestone filler with ceramic waste filler. Also Ahmed Hussein [28] studied the behavior of bitumen modified with varying concentrations of nanoceramic powder (NCP), they approved that the physical, chemical, and rheological properties of bitumen are improved. In this study,

SBR/WNCP nanocomposites were fabricated to modify asphalt binder performance at high temperature. The use of fabricated SBR/WNCP nanocomposites will offer a double advantage: (i) improving the high temperature performance of asphalt binder (rutting factor) due to the effect of WNCP and improving the low temperature performance of asphalt binder due to the effect of SBR. (ii) Compatibilizing SBR with asphalt binder due to the presence of NCP. (iii) reducing the cost of added polymer by replacing 30wt% of the SBR content with WNCP waste and consuming less time and temperature during the production. Three nanocomposite formulations of SBR/WNCP were prepared with mass ratios (90:10), (80:20), and (70, 30) respectively. From each formulated nanocomposites 4wt% and 6 wt% are blended to asphalt binder. The morphology of WNCP is determined by transmission electron microscopy (TEM). Also X-ray diffraction (XRD) is used for structural analysis. Particle size of WNCP is measured using dynamic light scattering (DLS). All physical tests of asphalt binder are performed including penetration, softening point, penetration index, storage stability, and viscosity. Furthermore, the rutting factor G*/sindelta is measured by using dynamic shear rheometer (DSR) before and after exposure to short term aging by applying rolling thin film oven test (RTFOT).

2. Experimental

2.1. Materials

In this study, asphalt penetration grade 60/70 is selected as the base binder provided from El-Nasr Petroleum Co. SEUZ. The main properties of base asphalt binder are illustrated in Table (1). SBR rubber is purchased from Chinese Plastic Company, waste ceramic powder (WCP) is supplied from the waste of brick and tiles manufacturing industries.

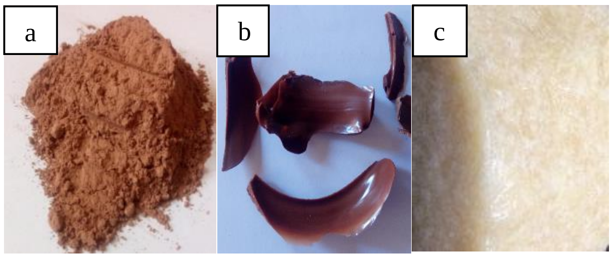

Fig. 1. Photograph of (a) waste nanoceramic powder, (b) SBR/WNCP nanocomposite, (c) SBR rubber.

2.2. Methods of preparation

2.2.1. Fabrication of SBR/WNCP NanoComposites

The process of formulating SBR/WNCP nanocomposite is depending on a research conducted by Ahmed Mansourian [29]. Firstly, WCP were ground to nanosized WNCP using a bowl mill machine. The ground WNCP was sonicated in toluene at 60^{o}C for 15 min then SBR was dissolved in it for 1 h using mechanical stirring. Three polymer nanocomposites were prepared from different SBR/WNCP mass ratios (90, 10) and (80, 20) and (70, 30) respectively. The obtained nanocomposites were dried for 24 h at ambient temperature. After complete evaporation of the solvent, the polymer nanocomposites were obtained as shown in Fig. 1.

2.2.2. Blending Nanocomposites to Asphalt Binder

5wt% from each SBR/WNCP nanocomposite with mass ratios (90, 10) and (80, 20) and (70, 30) samples is blended with asphalt binder constituting Blend 10, Blend 20, and Blend 30 respectively. Initially asphalt binder is preheated at 160°C, then each mass ratio of SBR/WNCP composites is mechanically blended under high-speed shear 3000rpm for 1 hr. Finally, the mixes are stirred with a low speed 500rpm for 10 minutes to remove the bubbles in mixture [30].

2.3. Characterizations

2.3.1. X-ray Fluorescence (XRF)

The chemical constituents of WNCP were analysed by Xray fluorescence technique (XRF) using Axios (PW4400) WD–XRF Sequential Spectrometer (Panalytical, The Netherlands), using (Rubidium) Rb ka radiation tube at 50 kV and 50 mA.

2.3.2. X-ray diffraction (XRD)

The crystalline structure of WNCP and prepared nanocomposites were verified using X-ray diffraction (wide angle XRD) analysis (Panalytical-X'pertPRO) with Cu K- alpha source radiation (k = 1.54 Å) operated at 40 kV and 40 mA, and the scanning angle 2h with a rate 1°/min ranged from 4 ^{o}C to 80 ^{o}C [19].

2.3.3. Dynamic laser scattering (DLS)

Particle size distribution for WNCP was statistically investigated through Malvern dynamic laser scattering (DLS). The specimen was firstly well dispersed and sonicated in deionized-water for 10min. The obtained dispersed solution was then subjected to DLS at 23 ^{o}C with a scattering angle of 90^o.

2.3.4. Thermal gravimetric analysis (TGA)

TGA-Mettler Toledo was used to study the effect of WNCP on thermal stability of prepared composites using (TGA) by heating from 25 °C to 600 °C at a rate of 5 °C/min.

2.4. Tests on asphalt binder

2.4.1. Physical tests

The physical properties were studied including penetration at 25 ^{o}C according to ASTM D 5, softening point test according to ASTM D 36, and dynamic viscosity as recommended by ASTM D4402.

2.4.2. Rheological properties test

The rheological properties are assessed by using dynamic shear rheometer (DSR), in which the plate geometry with a 25-mm diameter plate and 1-mm gap. The temperature sweeps from 50°C to 80 °C with 5°C increments were applied at a fixed frequency of 10 rad/sec. The resistance of the asphalt binder to permanent deformation at high temperatures (rutting resistance) was measured by factor $G^*/\sin\delta$ according to ASTM D7175 where G^* is the complex shear modulus and δ is the phase angle. The test is firstly conducted on the unaged asphalt binders and then carried out on aged binders subjected to RTFO test.

2.4.3. Short-term aging test

Asphalt is subjected to short term aging process using RTFO according to ASTM D2872. Asphalt sample is placed on thin film oven which was equipped with a thermostat and a rotating disc rack at 163 ^{o}C for 5 h.

2.4.4. High temperature storage stability test

The stability of polymer modified asphalt binder during long term storage at high temperature is examined according to ASTM D 7173. The sample of modified binder is placed in an aluminum tube (32 mm in diameter and 160 mm in height). The tube is vertically heated and held at 163°C for 48 h, and then left to cool. After cooling, the samples are cut into three equal sections. The sample taken from the top and the bottom sections are applied to evaluate storage stability by measuring their softening points. If the difference of the softening points between the bottom and the top sections was less than 2.5°C, the sample is considered to have allowable high temperature storage stability [31].

3. Results and discussion

3.1. Assessment of WNCP

Table 2 shows the chemical composition of WNCP. It is obvious that silica is a major component constituting 65% from total weight. Al_2O_3 represents slightly fourth of the material by weight (23%). Fe_2O_3,

TiO_2, and Cr_2O_3 are shown as traces in WNCP. Chemical analysis is in good agreement with X-ray diffraction (XRD) analysis of WNCP that is shown in Fig. 2 where the major height at $2\theta = 26$ which is corresponded to silica [32]. The particle size distribution of grinded waste ceramic powder is analyzed by DLS as shown in Fig. 3, where particle size is distributed through 100 to 200nm.

Table (2) Chemical composition of WNCP from XRF

Chemical Components	Abundance %
Silicon Oxide SiO_2	65
Aluminium Oxide Al_2O_3	23
Calcium Oxide CaO	2.9
Ferric Oxide Fe_2O_3	5.3
Magnesium Oxide MgO	0.5
Potassium Oxide K_2O	1.6
Sodium Oxide Na_2O	1.7

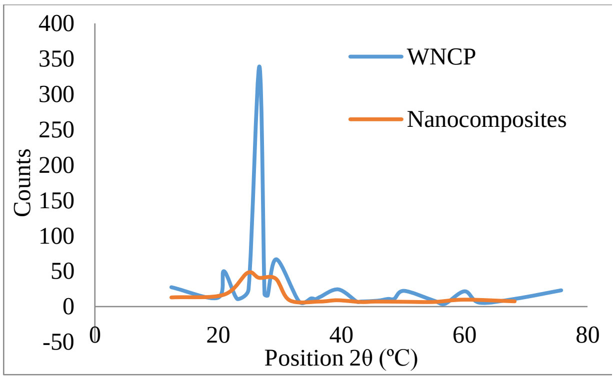

Figure (2) XRD of WNCP and SBR/WNCP nanocomposites

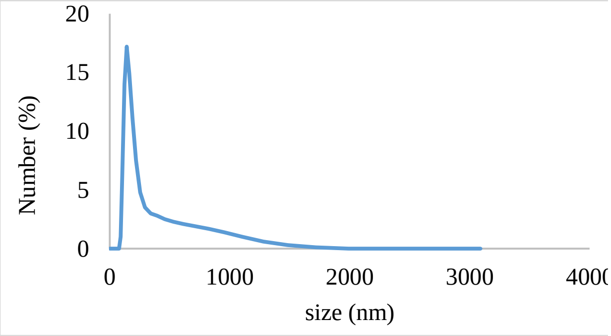

Figure (3) Particle size distribution of grinded nanoceramic powder

3.2. XRD of nanocomposites

WNCP mainly consists of silicate layers that are separated by Van der Waals gaps forming interlayers. The silicate layers are approximately 100-200 nm in length as assessed by DLS. The wide-angle X-ray diffraction curves of pure WNCP and SBR/WNCP nanocomposites are shown in Fig. 2. As can be seen SBR/WNCP nanocomposites show differences in X-ray patterns from that of pure WNCP. The appearance of a higher diffraction peak at $2\ \theta = 26$ for the pure WNCP indicated that the intergallery d-spacing between silicates was about 3.3 Å. For the SBR/WNCP nanocomposites show a broad diffraction peak shifted at $2\ \theta = 24$ with d-spacing 3.5 Å. The decrease of intensity of order peaks indicated that the ordered structure of the layers is disrupted. The broadening of the peaks suggests that the intercalation of SBR into the layered structure has occurred, leading to a disordered structure of the layers.

3.3. Thermal stability of nanocomposite

Fig.4 illustrates thermal degradation curves of pure SBR, SBR/WNCP nanocomposites, pure asphalt, and blended asphalt samples. It was noticed that thermal degradation of pure SBR occurred via two-stages due to the presence of styrene and butadiene monomer units. The first stage of degradation from 320 °C to 460°C which is mainly attributed to the degradation of butadiene, whereas the second stage of degradation from 460°C to 564°C that is mainly due to the degradation of styrene monomer units. Also, it can also be noticed that the thermal degradation curve of SBR/WNCP nanocomposite sample shift towards higher temperature by delaying the first degradation temperature to 380°C, and second degradation temperature to above 500°C with more stable, and more residues were retained. By comparing thermal degradation curves for pure asphalt and asphalt blended with SBR/ WNCP nanocomposites, it was observed that modified asphalt exhibits higher thermal stability and lower rate of mass loss (residue about 30%).

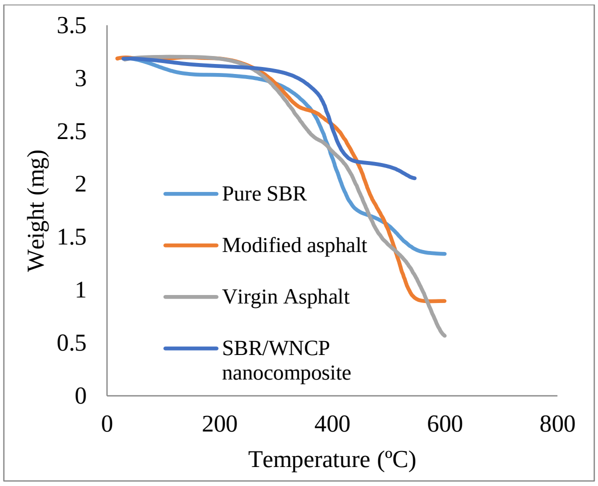

Figure (4) Effect of WNCP on thermal stability of SBR and effect of nanocomposite on thermal stability of asphalt binder

3.4. Physical properties of modified asphalt

As demonstrated in Fig.5, blending asphalt binder with 5wt% SBR/WNCP nanocomposites is followed by a decrease of penetration reached to 38 which is approximately 61% lower than that of control sample 62, and an increase in softening point temperature reached to 70 °C that is 40% higher than control binder which is may be due to the formation of exfoliated structure in SBR/WNCP modified asphalt [9]. As a result, the formulated nanocomposites exhibited a remarkable improvement on penetration and softening point of asphalt binder. Accordingly, blended binders led to higher resistance against rutting (permanent deformation) at higher temperatures.

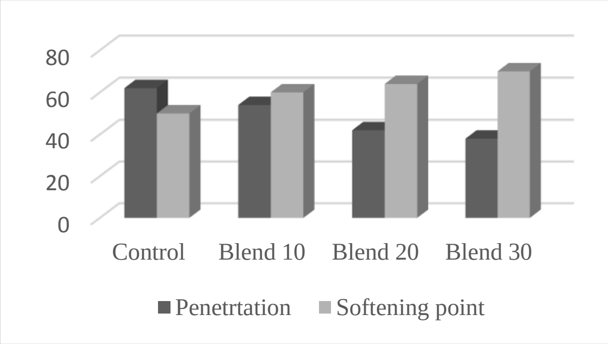

Figure (5) Penetration values and softening point temperature of control and 5wt% of blended asphalt binder.

Another factor which is considered the simplest factor for evaluating blended binder performance against temperature variation is the penetration index (PI). Where PI values for asphalt cement ranges between (-2 to +2), and binders with higher PI values (more positive) consider having lower susceptibly to temperature variations and resulted in favorable characteristics [33]. As shown in Fig.6, addition of SBR/WNCP nanocomposite to asphalt binder is responsible for decreasing the temperature susceptibility of blended binders. Where PI values increase from - 0.5 for control sample to + 2 for blended samples.

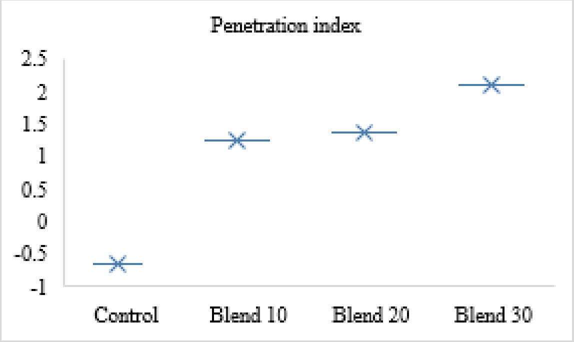

Figure (6) Penetration index of control and 5wt% blended asphalt binder

3.5. Rheological properties of modified asphalt

The visco-elastic behavior of modified asphalt binder is characterized by DSR at high temperatures. The test of resistance to permanent deformation (rutting resistance) is obtained from DSR measurements by the factor G*/sin δ where G* is the complex shear modulus, and δ is the phase angle. Fig. 7,8 showed that modifying asphalt binder with SBR/WNCP nanocomposites increases rutting resistance that implies a more viscoelastic behavior and more stiffness than that of control asphalt. This trend is mainly due to the fine intercalation and penetration of WNCP into SBR matrix structure [34]. Also, it was noticed that blend 30 exhibited the highest rutting resistance that may be due to the high stiffness of WNCP.

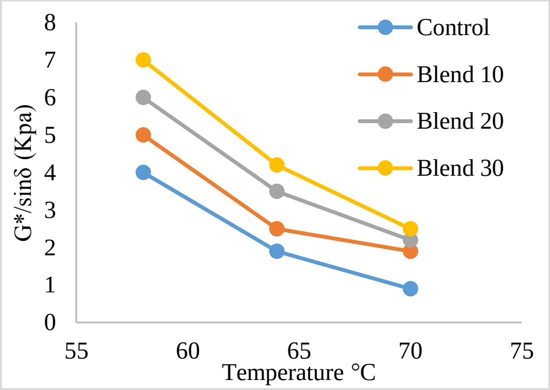

Figure (7) Rutting factor (G*/sinδ) for control asphalt binder and blended samples

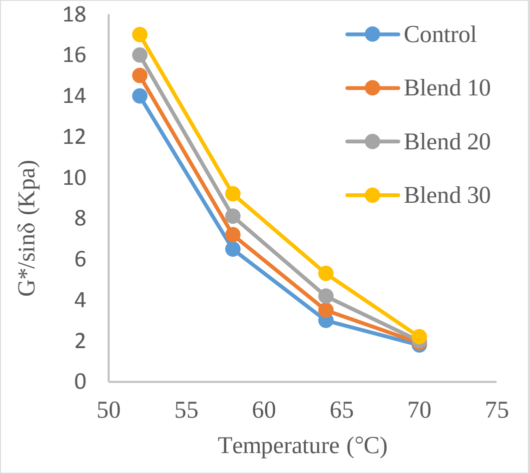

Figure (8) Rutting factor (G*/sinδ) for control binder and blended asphalt samples with SBR/WNCP nanocomopsites after RTFO

3.6. Storage Stability.

The phase separation of polymer modified asphalts usually takes place during storage, pumping and in the pavement processes [35]. The investigation of the storage stability of modified asphalt is an important property. The high-temperature storage stability test can determine whether the additives during mixing is strong enough to resist phase separation in the condition in which it is stored, which is realized by investigating the softening point variation between the top and the bottom for modified asphalt in the tube described in Section 2.4.4. If this variation was less than 2.5 ºC, the modified asphalt was considered to have good high temperature storage stability. The high-temperature storage stabilities of the SBR/WNCP blended asphalt were measured and the results are presented in Table 3. Clearly, the differences in the softening points are more than 2.5ºC for SBR modified asphalt binder, which indicated that the phase separation of the SBR/asphalt composite is certain and SBR modified asphalt binder is unstable. However, when blend 10, blend 20, and blend 30 were tested, the storage stability of blended asphalt was significantly improved and the difference between the softening points of the top and the bottom are less than 2.5 ºC. This can be explained due to equalize the differences in densities between asphalt and SBR in the presence of WNCP [9]. It is known that the density of the asphalt is 1.02 g/cm^3 and that of SBR is 0.91 g/cm^3 as well as the density of WNCP is around 2.7 g/cm^3. When the WNCP attached to SBR to form intercalated composite, the density of SBR/WNCP was more than the one of SBR and the density difference between asphalt and SBR/ WNCP was less in some SBR/WNCP content range, which resisted to phase separation of the SBR/WNCP modified asphalts.

Table (3) Effect of SBR/WNCP on high temperature storage stability of modified asphalt

Additives	Softening point (ºC)		Difference (ºC)
Pure asphalt (control)	56	52	4.0
Blend 10	60	58.4	1.6
Blend 20	64	62.8	1.2
Blend 30	70	69.2	0.8

4. Conclusions

The aim of this study is to evaluate the effect of blending of SBR/ WNCP nanocomposites on high temperature performance, and storage stability of asphalt binder. The prepared nanocomposites are chemically characterized. Also, the physical, viscoelastic properties, and storage stability of blended asphalt were studied. Based on the results of this study, the following conclusion is drawn.

1. The Chemical analysis including XRD, XRF, DLS were successfully investigated to assess the prepared SBR/WNCP nanocomposites.
2. According to TGA results, it can be stated that fabrication of SBR/WNCP nanocomposites enhances thermal stability of SBR copolymer. the thermal resistance of blended asphalt binder is also improved.
3. Since SBR modified binder doesn’t have enough compatibility and high storage stability, it can be stated that the presence of SBR /WNCP nanocomposites in asphalt binder improve SBR modified binder compatibility and storage stability.
4. Based on the results of physical tests for blended binder, it can be concluded that SBR/WNCP increases the stiffness of binder, improves the strength, and decreases the temperature sensitivity.
5. The results of DSR test showed that adding SBR/WNCP nanocomposites to asphalt binder enhances its rheological properties and highly increase rutting resistance compared to control binder.

4. Conflict of Interest

“There are no conflicts to declare”.

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تحضير الكمبوزايت المكون من مطاط الاستيرين بيوتادايين مع بودره السيراميك ودراسه تاثيره على كفاءه الاسفلت فى درجات الحراره المرتفعة

الملخص العربى:

من المعروف ان معالجه الاسفلت باستخدام مطاط الاستيرين بيوتادايين يعطى نتائج مرضيه جدا فى كفاءه الاسفلت فى درجات الحراره المنخفضه فنجد انه يقلل من حدوث الشروخ والتشققات للاسفلت كما يزيد من ليونته. ولكن مع ارتفاع درجه حراره الاسفلت تبدا تظهر بعض العيوب مثل التخدد وفصل المطاط من الاسفلت اثناء التخزين. فكان الهدف من البحث وهو تحسين كفاءه الاسفلت المعالج بمطاط الاستيرين بيوتادايين عند درجات الحراره المرتفعه وذلك بتحضير الكمبوزايت المكون من مطاط الاستيرين بيوتادايين مع بودره السيراميك بعد طحنها الى حجم النانو. تم تحضير ثلاث تركيبات من الكمبوزايت باوزان نسبيه مختلفه 90:10و 80:20و70:30 من مطاط الاستيرين بيوتادايين وبودره السيراميك فى حجم النانو. تم تقييم مطحون بودره السيراميك كيميائيا باستخدام XRD&XRF و قياس حجمها بعد الطحن باستخدام (DLS). ثم تم معالجه الاسفلت المصرى باضافه 5% من الكمبوزيتات المحضره وقياس كفاءه الاسفلت عند درجات الحراره المرتفعه وقياس مدى ثباتيه المطاط المحضر اثناء التخزين وايضا قياس الخواص الفيزيائيه والريولوجيه للاسفلت المعالج قبل وبعد تعرضه لعملية تقادم الاسفلت. وقد اثبتت النتائج ان الكمبوزيتات المحضره كانت قادره على تحسين كفاءه الاسفلت عند درجات الحراره المرتفعه حيث عالجت مشكله التخددات كما قللت من قابليه المطاط من الفصل اثناء التخزين فى درجات الحراره المرتفعه.