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# Improving moisture sensitivity and mechanical properties of sulfur extended asphalt mixture by nano-antistripping agent



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# HIGHLIGHTS

• A new technique is introduced to mix the modified-sulfur (Googas) with asphalt binder.

- Sulfur extended asphalt (SEA) mixture can significantly improve resistance against rutting phenomenon.
- Nano-antistripping agent (NZ), improved tensile strength of SEA mixture.
- NZ improves moisture sensitivity and mechanical properties of SEA mixture.
- Fatigue life of SEA slightly improved due to NZ-modification.

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# ABSTRACT

Moisture damage and fatigue cracking are the most common defects of Sulfur Extended Asphalts (SEAs). Moisture damage in asphaltic mixtures occurs due to the weak cohesion and adhesion that causes the creation of different forms of pavement defects. Various methods have been employed in order to enhance the asphalt mixture's resistance to the moisture damage. One of the main methods is the addition of antistriping agents (ASAs) which reinforce the bonding between asphalt binder and aggregates. In this study, a series of laboratory testing has been performed to appraise the mechanical properties and moisture sensitivity of the SEA mixtures modified with ASA. In addition, Googas as a new generation of modified-sulfur-mix additive and ASA (nanotechnology Zycotherm) were employed to make warmmix asphalt (WMA) specimens, through modification of neat asphalt (PG 64-22). Furthermore, four types of mixtures with different additive proportions, containing ASA and sulfur, were prepared, and the moisture susceptibility, resilient modulus, rutting resistance, and fatigue behavior were measured. Obtained results, demonstrated the improvement of mechanical characteristics due to the implementation of the modified sulfur, exclusively for rutting phenomenon. Moreover, nanotechnology Zycotherm (NZ) as an ASA enhanced the adhesion between aggregates and sulfur-extended asphalt; thus, the resistance of SEAs against moisture damage and fatigue cracking improved.

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# 1. Introduction

Asphalt is a viscoelastic binder that experiences elastic behavior during rapid loading or at low environmental temperatures and viscous behavior during slow loading or at high temperatures. This temperature-dependent behavior causes a desire to improve the properties of asphalt to resist more against the rutting phenomenon, which usually appears at high temperatures, and against cracking phenomenon, which mostly happens at low temperatures

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[1,2]. On the other hand, temperature variations between warm and cold months, as well as, increased traffic volume of trucks, create critical pressures in the structure of pavements. Such conditions have caused an increased desire to improve asphalt properties. There are various methods to modify the asphalt properties [3]. One of the most popular and common procedures is using of modifier additives such as sulfur.

Since four decades ago, many studies have demonstrated that sulfur can modify and improve the properties of asphaltic pavements [4]. Application of sulfur as an asphalt binder performance extender is an economical solution to decrease virgin binder consumption and consequently reducing greenhouse gas emission. In addition, it was observed that Sulfur Extended Asphalts (SEAs)

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could improve pavement's mechanical properties such as stiffness compared to conventional asphalt mixtures [5]. The use of sulfur to improve the quality of asphalt began in 1970 and was commercially used during the 1970s and 1980s. In the past, environmental concerns including H<sub>2</sub>S generation and lack of appropriate corresponding technology were the primary issues of the implementation of molten sulfur in the asphalt industry. Therefore, there was an interruption period with using sulfur as an asphalt modifier in the 1980s [6]; however, the concept of SEA reappeared with the development of a new version of solid dust-free sulfur pellets. Solid dust-free sulfur facilitates the displacing of sulfur in a solid pellet shape. This new product was improved by using polymeric compounds in the sulfur pellets. By this change, it became possible to add a new product to the asphaltic mixtures while emission of hazardous gasses like H<sub>2</sub>S and annoying odors reduced considerably compared to the conventional liquid sulfur extended asphaltic mixture [7]. This new product reduced mixing temperature to about 135 °C while compaction temperature is reduced to about 90 °C. Many of the aforementioned safety problems seemed to be resolved, if the asphalt concrete was constructed at a mixing temperature of  $135 \pm 5$  °C. Moreover, the new sulfur pellets could be poured directly into the asphalt-aggregates mixture during the mixing operation [7,8]. As a result, there was no concern about difficulties of blending it with asphalt before adding the binder to the aggregates. Since mixing operation should be done in the temperature range lower than conventional hot mix asphalt (HMA), it should be considered as a Warm Mix Asphalt. The Mixing temperature of WMA is approximately 16-55 °C lower than conventional HMA, therefore, using the new sulfur extended asphalt as WMA can reduce required energy to produce and compact asphalt mixture [9].

The bonding between asphalt and aggregates is considerably important as it affects the integrity of asphaltic pavement and supports the asphalt structure stresses associated with traffic loading and temperature changes. The formation of bonding between the aggregates and asphalt binder initiates during the mixing process when asphalt coats aggregates. DiVito et al. [10] showed that pavement strength depends on (1) the asphalt's cohesive resistance. (2)the resistance of adhesive bond between the asphalt and the surface of aggregate, and (3) the interlock between the aggregates particles. ASAs are substances designed to improve chemically the adhesion between the asphaltic binder and the aggregates. They are available in both liquid and solid forms. One of the most common ASAs which has been extensively used to improve the HMA's resistance against moisture damages is hydrated lime. Other common conventional ASAs are fly-ash and Portland cement. In addition, liquid ASAs in the form of cationic surface-active agents (principally amines) have become popular in recent years [11]. In this study, a liquid ASA commercially named Zycotherm was used to make the aggregates hydrophobic while providing improved bonding with the sulfur-extended asphalt binder.

#### 1.1. Literature review

Bencovitz and Boe demonstrated that sulfur could be mixed with asphalt as an extender and make asphalt cement which can modify the asphalt rheological characteristics [12]. For the first time, SEA was constructed by Kennepohl et al. [13] and practically applied in asphaltic pavements in the early 1980s as an asphalt extender to decrease asphalt consumption. Some case studies by Beatty et al. [14] showed that SEA has better function in comparison with conventional HMA.

In recent studies performed on SEAs by Strickland et al. [15] and Timm et al. [16], sulfur was applied in the form of a solid pellet which could be melted at a temperature around 120 °C and added as asphalt extender with the ratio between 30% and 40%. These pellets were pretreated to decrease toxic gas emissions like hydrogen sulfide and also to lower the temperature of mixing and compaction operations. At mixing operation, a portion of the sulfur homogenizes with the asphalt at 120 °C and decreases the asphalt viscosity. As the asphalt cools down, the rest portion of the sulfur crystallizes and covers the aggregates. They also showed that dissolved, and crystallized sulfur makes the mixture stiffer and leads to a more resistant asphalt mixture at high temperatures, and consequently less vulnerable to permanent deformation [17]. Strickland et al. [18] suggested keeping the volume ratio of the total binder phase (asphalt and sulfur) constant at asphaltic mixture before sulfur extending and after that. This was recommended since the density of sulfur pellets is different from the base asphalt. They established a relationship between asphalt and sulfur content in the SEA based on the following equation:

Sulfur + asphalt % = 
$$\frac{100AR}{100R - P_S(R - G_{bitumen})}$$
 (1)

In which:

Sulfur + asphalt % = sulfur-extended binder content by the weight of WMA sample; A = asphalt content by the weight of conventional HMA sample (%); R = specific weight ratio (sulfur to asphalt); Ps = sulfur content in extended binder; and  $G_{bitumen}$  = specific weight of the asphalt.

Strickland et al. [18] did an experimental investigation on the functional properties of SEA in the laboratory. Results indicated deteriorated tensile-strength ratio (TSR) but improved dynamic modulus in various ranges of temperature and frequency.

In another research conducted by Cooper et al. [19], using SEA improved the predicted rutting and fatigue resistance and the overall pavement service life compare to unmodified HMA at various traffic intensities. It was also shown that SEA has the potential to reduce construction and maintenance costs in comparison with a conventional HMA produced with the same binder grade. In a study conducted by Timm et al. [16], dynamic moduli and moisture susceptibility of SEA were evaluated. Results indicated that SEA was more vulnerable against moist condition as exhibited lower TSR in comparison with conventional asphalt mixture: however, the SEA dynamic modulus increased.

### 1.2. Research objective

Mechanical properties and moisture sensitivity of antistripping agent-modified SEA were evaluated in this paper by the experimental method. A Nano-based antistripping agent named commercially zycotherm and petroleum base sulfur named commercially Googas were used respectively to replace a portion of asphalt and strengthen the bond between the binder (sulfur and asphalt) and aggregate's surface. In this study, moisture sensitivity, rutting performance, resilient modulus and fatigue behavior were evaluated for the proposed samples. Asphalt mixture tests were carried out on four different mixtures including one conventional HMA (control sample) and three modified WMA (in different combinations of sulfur and ASA additives).

#### 2. Test materials and mixing

#### 2.1. Materials

#### 2.1.1. Asphalt binder and aggregates

Materials used in this experimental investigation included a PG 64-22 virgin asphalt binder, with the physical properties that are shown in Table 1. Used aggregates have desired mechanical properties such as enough strength, toughness, and hardness. Also, crushed aggregates were used to make higher stability. The Super-

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