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The effect of silica nanoparticles and crumb rubber additives on chemical and physical properties of bitumen

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Bitumen modified by suitable additives enjoys many benefits such as increase in strength against rutting and fatigue, as well as against cryogenic cracks, reducing damage and thermal sensitivity. In this work, we investigated the effect of silica nanoparticles and crumb rubber additives to improve the quality of bitumen. The chemical and physical properties of bitumen were investigated after modification with silica nanoparticles and crumb rubber additives. The effects of silica nanoparticles in the range of 2-8% (w/w) and crumb rubber at ratios of 5-20% (w/w) on softening point, rheological properties, penetration, tensile strength, and weight loss of bitumen were investigated. The results exhibit an improvement in the quality of bitumen in the presence of silica nanoparticles and crumb rubber additives. According to the experimental data, the tensile strength of bitumen increased from 1000 mm to 1160 mm, and 113 mm after addition of 15% of crumb rubber and 6% of silica nanoparticles, respectively. In addition, the results showed an improvement in all properties of the bitumen in the presence of silica nanoparticles and crumb rubber additives.

Keywords: Silica nanoparticles, Crumb rubber, Bitumen, Modification process

INTRODUCTION

The use of industrial waste materials for engineering applications is considered as a useful tool for environmental protection. The high value of industrial waste materials is a major problem in environmental health. Therefore, various designs have been proposed for the use of industrial wastes. In between, using industrial wastes in bitumen is a useful and economic strategy for environmental protection and engineering applications. Due to its good properties and low cost, the crumb rubber was suggested as a modifier for bitumen since the 1930s and application of crumb rubber increased in the recent years with an increase in rubber waste in human and industrial societies. The swelling process of rubber particles helps to improve properties of bitumen. The swell up of crumb rubber in a bitumen matrix can be related to absorption of maltenes component [2, 3]. The scientific reports showed that crumb rubber could be used for modification of bitumen by dry or wet process as two different strategies. The scientific researchers showed that wet process could be useful for improving bitumen properties such as fatigue cracking, resistance of asphaltic, and resilience modulus compared to dye process [4].

Nano-materials with unique properties such as high thermal stability, high resistance, and good electrical conductivity were suggested as a new approach in engineering applications [5-10]. They improve the pavement durability, enhance the decrease in moisture susceptibility, and in aging, storage stability, and decreases maintenance costs which are the main advantages of bitumen modified with nano-materials [11]. The silica nanoparticles are a good choice for modification of bitumen due to their chemical purity, good dispersing ability, excellent stability, low cost, and strong adsorption [12-18]. Due to these properties, many researchers focused on the application of silica nanoparticles in modification of bitumen. As an example, Shafabakhsh and Ani used nano TiO_2/SiO_2 particles to improve properties of bitumen. They showed that the presence of nano TiO_2/SiO_2 particles could improve some properties of bitumen such as viscosity, adhesive bonding, and softening point [19]. Shi *et al.* investigated the role of nano-silica and rock asphalt as a high-quality additive on the rheological properties of bitumen. Results showed an improvement in G^* factor of bitumen [20]. In this research, the effects of silica nanoparticles and crumb rubber additives on the chemical and physical properties of bitumen were investigated.

EXPERIMENTAL

Materials

Bitumen 60/70 penetration grade was chosen as an unmodified bitumen for all of the investigations. Tetraethyl orthosilicate (TEOS), ammonium hydroxide, and ethanol were purchased from

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Sigma-Aldrich Company for synthesis of SiO_2 nanoparticles. SiO_2 nanoparticles were synthesized according to the following procedure: 100 mL of ethanol (4.0 M) was sonicated for 10 min. In continuation, 4 mL of TEOS (0.04 M) was added to the ethanol under sonication. After 20 min, ammonium hydroxide (14.0 M) was added to the solution under sonication, as a catalyst to promote the condensation reaction and sonication process was continued for 50 min. The obtained gel dried at 100 °C for 6 h.

Methods

Process of addition of silica nanoparticles. 200 g of bitumen 60/70 was placed in a metal container and heated at 190 ^{0}C for 30 min. After melting the bitumen, the obtained sample was homogenized by a stirrer for 10 min. In continuation, we added silica nanoparticles at different ratios (2%; 4%; 6 and 8% w/w) at a stirring speed of 900 rpm for 15 min.

Process of addition of crumb rubber. 200 g of bitumen 60/70 was placed in a metal container and heated at 190 ^{0}C for 30 min. After melting the bitumen, the obtained sample was homogenized by a stirrer for 10 min. In continuation, we added the crumb rubber at different ratios (5.0%; 10%; 15.0% and 20% w/w) at a stirring speed of 900 rpm for 15 min.

Softening point determination. For the determination of softening point, we used the standard test ASTM D36 for unmodified and modified bitumen. The proposed test method covers the determination of the softening point of bitumen in the range from 30 to 157°C [86 to 315°F] using a ring-and-ball apparatus immersed in distilled water [30 to 80°C] or USP glycerin (above 80 to 157°C).

Bitumen weight loss test. We used the standard test ASTM D6 to investigate bitumen weight loss; the results of this test were reported according to weight ratio for total samples according to the following equation:

$$\text{Loss of weight} = (W_a - W_b/W_a) \times 100 \qquad \text{(eq. 1)}$$

where W_a is the weight of the sample before placing it in air furnace, and W_b is the weight of the sample after removing it from the furnace.

Tensile test. For studying the tensile factor, we used standard test ASTM D113 for the unmodified and modified bitumen. The present test method describes the procedure for determining the ductility of an asphalt material measured by the distance to which it will elongate before breaking when the two ends of a briquet specimen of the material, described in Experimental, are pulled apart at a specified speed and temperature. Unless otherwise specified, the test shall be carried out at a temperature of 25-60.5 °C [77-60.9 °F] and at a speed of 5 cm/min 65.0 %. The speed for other temperatures should be specified.

DSR Test. Viscoelastic behavior of bitumen-modified samples was investigated by a dynamic shear rheometer (DSR) test by employing standard test ASTM D-7175. The present test determines the dynamic shear modulus and phase angle of asphalt binders when tested in dynamic (oscillatory) shear using parallel-plate geometry. It is applicable to asphalt binders having dynamic shear modulus values in the range of 100 Pa to 10 MPa.

RESULTS AND DISCUSSION

Characterization of SiO_2 nanoparticles and bitumen morphology

The synthesized silica nanoparticles were characterized by SEM method. As can be seen in Figure 1a, silica nanoparticles with spherical shape were synthesized by the procedure recommended above. The SEM image of the unmodified bitumen (Figure 1b) and bitumen modified with silica nanoparticles (Figure 1c) and crumb rubber (Figure 1d) are presented. As can be seen, the silica nanoparticles and crumb rubber additives were successfully dispersed in the bitumen matrix. Due to the high adhesion strength of the additives, as well as their placement in the porous space of the bitumen, the cracks of the bitumen after their increase were significantly reduced.

Effect of silica nanoparticles and crumb rubber additives on softening point

The softening point of bitumen was investigated as an important factor in the preparation of asphalt. The results showed a softening point of ~51 ^{0}C for unmodified bitumen. After modification of bitumen with silica nanoparticles and crumb rubber, the softening point of asphalt changed. We detected an increase in the softening point of the modified bitumen in the range of 2.0-6.0 % of silica nanoparticles (Figure 2A).

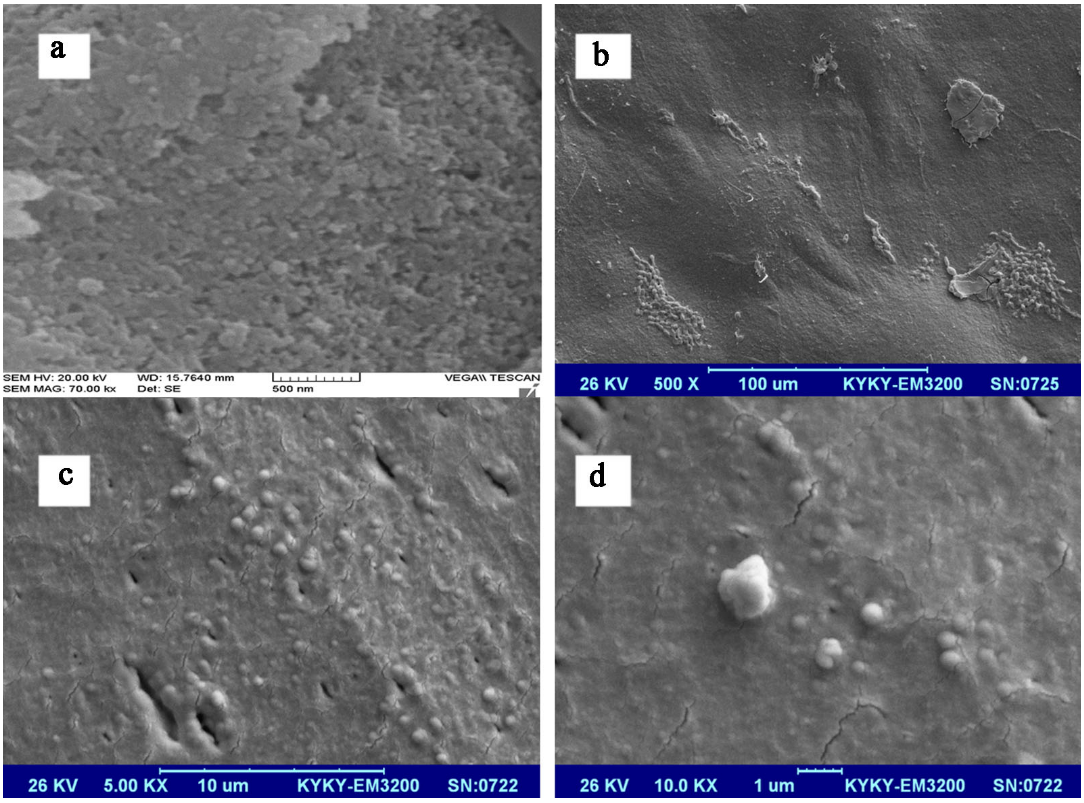

Figure 1. SEM image of a) SiO_2 nanoparticles; b) bitumen; c) bitumen after addition of silica nanoparticles; and d) bitumen after addition of crumb rubber

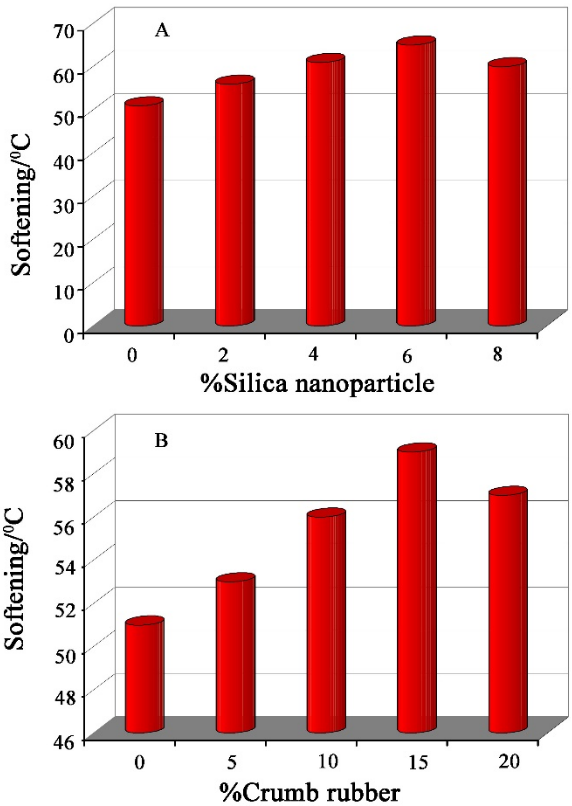

Figure 2. Softening point diagram of bitumen after addition of silica nanoparticles (A) and crumb rubber (B).

After this value, the softening point of the modified bitumen decreased due to a decrease in the homogeneity of nanoparticles, which might decrease the softening point of the whole composite. The softening point of bitumen increased in the presence of 5%-15% of crumb rubber and after that value it decreased (Figure 2B). With an increase in the amounts of the additives, their aggregation created a major problem, decreasing the softening point of the modified bitumen.

Effect of silica nanoparticles and crumb rubber on weight loss

The effect of silica nanoparticles and crumb rubber on the weight loss of bitumen was investigated. To this goal, we compared the weight of bitumen before and aftr its placement in a furnace. As can be seen, the value of bitumen weight before placing in a furnace is very similar to its weight after its removal from the furnace, which exhibits good stability of silica nanoparticles and crumb rubber as additives in bitumen modification (see Tables 1 and 2). According to the data in the Tables, the silica nanoparticles and crumb rubber are stable in the bitumen matrix.

Table 1. Data obtained for the effect of the addition of silica nanoparticles on bitumen weight loss

Percentage of silica particles (w/w)	Weight (g) of bitumen before furnace	Weight (g) of bitumen after furnace
2	70	69.6
4	70	69.3
6	70	69.1
8	70	68.7

Table 2. Data obtained for the effect of the addition of crumb rubber on bitumen weight loss

Percentage of crumb rubber (w/w)	Weight (g) of bitumen before furnace	Weight (g) of bitumen after furnace
5	70	64.4
10	70	62.1
15	70	62.0
20	70	61.8

Penetration investigation

The penetration properties of bitumen in the presence of silica nanoparticles and crumb rubber were investigated and the obtained data are presented in Figures 3A and 3B, respectively. The results showed a decrease in penetration after the addition of silica nanoparticles and crumb rubber, which means an increase in the binder hardness.

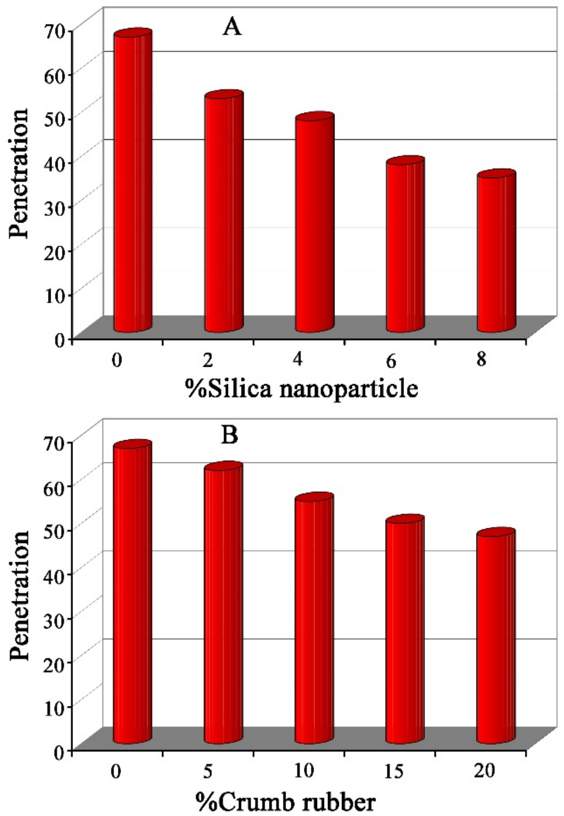

Figure 3. Penetration diagram of bitumen after the addition of silica nanoparticles (A) and crumb rubber (B).

Tensile strength investigation

The tensile factors of bitumen after the addition of silica nanoparticles and crumb rubber are presented in Figures 4A and 4B. As can be seen, the values of tensile strength in the presence of 6% of silica nanoparticles and 15% of crumb rubber showed good stability and after these values the tensile factor was stable.

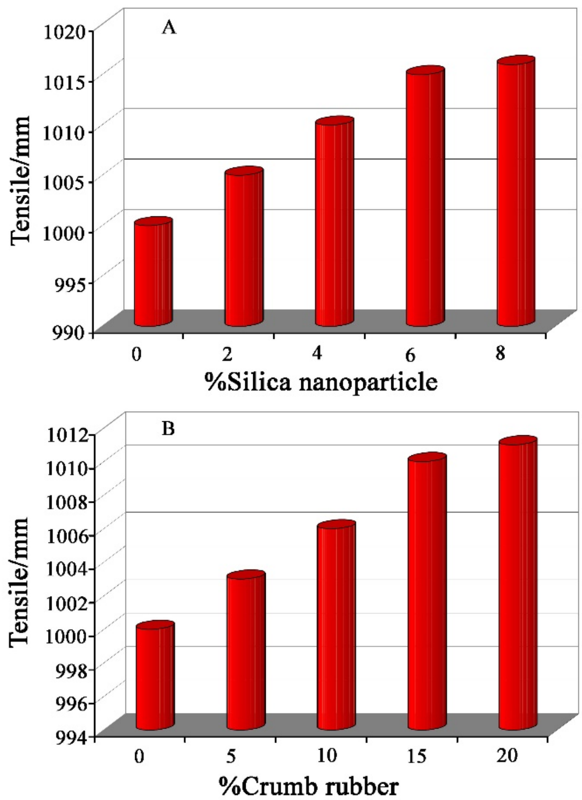

Figure 4. Tensile diagram of bitumen after the addition of silica nanoparticles (A) and crumb rubber (B).

DSR test results

The G* values of unmodified bitumen and bitumen modified with silica nanoparticles and crumb rubber at 40 °C are shown in Figure 5. As can be seen, the value of G* increased with an increase in the percentages of silica nanoparticles and crumb rubber that can be related to improved bitumen elasticity and viscoelasticity for the high amounts of modifiers. The good bond between bitumen and silica nanoparticles and crumb rubber with high surface area is the main cause of improving G* of bitumen after modification with modifiers. Due to good compatibility of bitumen and silica nanoparticles, we detected an increase in G* for all of the percentages of nanoparticles. The little decrease in G* after addition of 15% of crumb rubber can be related to low compatibility of bitumen and crumb rubber that changes the nature and rheological behavior of bitumen.

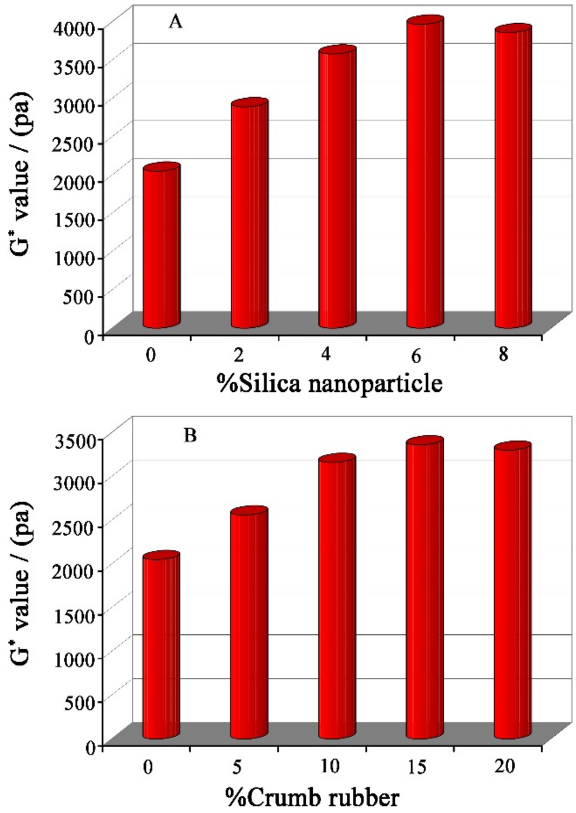

Figure 5. G^* diagram of bitumen after the addition of silica nanoparticles (A) and crumb rubber (B).

CONCLUSION

The present work revealed the significance of the chemical composition of base bitumen for its modification. The present study suggested new types of modifiers for improving the bitumen quality. The silica nanoparticles and crumb rubber showed very interesting properties for the modification of bitumen. The effect of silica nanoparticles in the range of 2-8% (w/w) and crumb rubber at ratios of 5-20% (w/w) on softening point, rheological properties, penetration, tensile strength, and weight loss of bitumen was investigated. The softening point, rheological properties, penetration and tensile properties of bitumen were improved in the presence of 6% of silica nanoparticles and 15% of crumb rubber.

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