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Laboratory Studies of the Effect of Recycled Glass Powder Additive on the Properties of Polymer Modified Asphalt Binders

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ABSTRACT

In this paper, the effect of adding recycled glass powder (RGP) as a third component in polymer modified bitumen is studied. RGP and styrene butadiene rubber (SBR) are added to the base bitumen in order to prepare modified binders and mixtures. The initial study is performed to determine physical properties of unmodified, SBR modified and RGP-SBR modified asphalt binders which prove RGP's compatibilizing effect on asphalt and polymer as well as high compatibility between RGP and polymer leading to better dispersion of the polymer in asphalt. The results of the study showed how final rheological properties of the mentioned combination could be affected by adding RGP and SBR. Modified and unmodified binders are used for preparing Marshall samples for the following tests: Marshall stability, indirect tensile strength (ITS), indirect tensile strength ratio (ITSR), and resilient modulus tests. The results also indicate that combination of 3% SBR plus 2% RGP can improve mechanical properties of asphalt mixture considerably.

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1. INTRODUCTION

Bitumen is one of crude oil derivatives, having black to dark brown color. Such organic material is solid in ambient temperature. By increasing the temperature of bitumen, it turns into plastic and liquid material. Major amount of bitumen produced worldwide are used in the road construction industry. Bitumen, as the main material for road construction, has been used in the industry for three centuries since it has a lot of advantages in comparison with other suggested alternative materials [1]. In addition, considering improvement in physical and chemical alternative materials may enhance efficiency and lifetime of asphalt. Every year, a great amount of money is spent on rehabilitation and reconstruction of roads and pavements in most countries [2]. One of the major disadvantages of using bitumen in road construction is its phase change as a function of temperature. Bitumen is brittle at low temperatures and turns into liquid at high temperatures, which is called temperature susceptibility. Base bitumen should be modified to reduce its temperature susceptibility. Different materials such as polymers or rubber could be used to improve bitumen quality and reduce its temperature susceptibility [3, 4].

SBR is one of the bitumen polymer modifiers. Using SBR modified bitumen for pavement construction results in more flexibility of pavement at lower temperatures and its more resistance to cracking [5, 6]. Application of SBR modified bitumen has other advantages like improving adhesion and cohesion of pavement and increasing its elasticity [7]; also, it has higher ductility compared with base bitumen [8]. Despite these advantages, SBR is an expensive polymer and material application is not beneficial for the environment while recycled glass powder (RGP) is an inexpensive recycled material and its application can reduce environmental pollution.

The aim of this study was to evaluate effects of adding RGP on properties of SBR modified bitumen. Nowadays, as a result of growth in the production of industrial materials all over the world, generation of different waste materials has been also increased. Therefore, it is essential to develop methods for recycling waste materials. There are different methods for dealing with waste materials such as landfill,

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incineration and material recycling. Material recycling is the best and most practical method from economical, technical and environmental points of view [9]. About ten million tons of waste and crushed glass (which is one of the waste materials) are annually produced worldwide [10]. This material can be recycled several times without any change in its properties. In road construction industry, only waste glass cullet has been used as an aggregate for glass recycling thus far. Results of previous studies in this regard have shown that combination of waste glass cullet and asphalt could improve some properties of pavements [10]. A recent study by Ghasemi and Marandi [11] which was done on the influences of RGP modifier on physical and mechanical properties of SBS modified asphalt binders and mixtures demonstrated increase of stiffness modulus in RGP-SBS modified pavements compared with the conventional asphalt mix. The main objective of this research was to investigate the influences of micro size crushed glass on the physical and mechanical properties of SBR modified asphalt binders and mixtures.

2. MATERIALS AND METHODS

2. 1. Aggregate The aggregate was obtained from an asphalt plant in Kerman, located in southeast of Iran. Figure 1 demonstrates the suggested gradation for the SMA mixtures developed by the National Asphalt Pavement Association (NAPA) [12]. Aggregate gradation of the mixture was in the middle of the limits. Properties of the aggregates such as specific gravities, values of water absorption, toughness and soundness

Loss of heating (%)

were specified; the test results are presented in Table 1.

The applied filler was calcium carbonate $(CaCO_3)$, which was brought from an asphalt plant. Calcium carbonate was passed through an ASTM #200 sieves and its specific gravity was 2.64.

2. 2. Bitumen In this investigation, 60–70 penetration asphalt cement was used, which was obtained from Isfahan Mineral Oil Refinery. Engineering properties of the asphalt cement are presented in Table 2.

2. 3. SBR SBR, containing 27.3 wt% styrene, 0.64 wt% soluble water and 0.37 wt% volatile fraction with 48–55 viscosity was produced in Kumho Petrochemical Co., Ltd., Korea.



Figure 1. Envelope of SMA gradation and design gradation.

0.03

| Property | Coarse aggregate | Fine aggregate | Filler | ASTM |
|---------------------------|------------------|----------------|--------|---------------|
| Bulk specific gravity | 2.73 | 2.66 | 2.48 | C-127 & C-128 |
| Apparent specific gravity | 2.77 | 2.70 | 2.64 | |
| Water absorption (%) | 0.28 | 1.49 | - | |
| Toughness (%) | 23.09 | - | - | C-131 |
| Soundness (%) | 1.87 | 1.21 | - | C-88 |

| TABLE 2. Conventional Rheological | Properties of Asphal | t Concrete |
|---|----------------------|------------|
| Test | Standard ASTM | AC 60-70 |
| Penetration (100g, 5sec., 25°C) (0.1 mm) | D5-73 | 64 |
| Ductility (25°C, 5 cm/min) (cm) | D113-79 | 100+ |
| Ductility after loss of heating test (cm) | D113-79 | 100+ |
| Solubility in trichloroethylene (%) | D2042-76 | 98.8 |
| Softening point (°C) | D36-76 | 47.4 |
| Flash point (°C) | D92-78 | 280+ |

D1754-78

| TABLE 3. | Particle Size Distribution of RGP |
|----------|-----------------------------------|
| | |

| Size (nm) | 458.7 | 396.1 | 342.0 | 295.3 | 255.0 | 220.2 | 190.1 |
|---------------------|-------|-------|-------|-------|-------|-------|-------|
| Percent passing (%) | 100 | 99.6 | 88 | 52.4 | 14.3 | 0.6 | 0 |

2. 4. RGP The recycled glass was first crushed and ground by a jaw crusher and a ball mill, respectively, for 10 min in order to obtain RGP, which was then passed through an ASTM #200 sieve having the specific gravity of 2.47. The RGP particle size distribution was determined by a laser particle analyzer, as given in Table 3.

The microscopic morphology of the RGP, measured by Scanning Electron Microscopy (SEM) is shown in Figure 2. SEM examinations revealed that glass powders mainly consisted of coarse and angular flaky particles with a broad range of particle size. Furthermore, to verify RGP absorption degree, the specific surface area test was implemented. According to ASTM C204, the specific surface area of the recycled glass powder was 467 m²/kg. Thus, the recycled glass powder had high absorption.

2. 5. Asphalt Mixture All the modified bitumen samples were prepared using a high shear mixer with shearing speed of 3000 rpm for 60 min at $165-175^{\circ}$ C. First, 1000 g bitumen was heated in an iron container to turn into fluid; then, the modifier (RGP, SBR-RGP) was added to the bitumen while reaching the temperature around 175 °C.

The Marshall asphalt mixture design method (ASTM D1559) was used in this study. The optimum asphalt content was selected to have maximum stability, maximum unit weight and median allowed limits for percent air voids. After selecting these three values, the content of the average asphalt cement (AC) was chosen and checked for satisfying the AV, VMA, stability and flow specification limits. Then, the optimum bitumen content for the control mixtures was obtained as 6.1% which was used for preparing all other modified mixes for maintaining consistency throughout the research. Stone matrix asphalt (SMA) is a type of asphalt mixture used for pavements surface courses in many countries. Due to its excellent performance characteristics, especially in freeways with heavy traffics and hot environment, SMA application has been gradually considered by many road builders around the world; hence, this type of mixture was used in this research [13]. In the SMA mixture design, the Marshall method of mix design is usually applied to confirm satisfactory voids in SMA mixtures. Laboratory specimens were also prepared by fifty blows of the Marshall hammer per side. It should be noted that SMA mixtures are more easily compacted to the desired density on the roadway compared with the effort required for the conventional HMA mixtures [14].



Figure 2. SEM micrograph of RGP

Asphalt mixtures are complex materials. Their behavior is strongly dependent on temperature, strain rate and stress conditions [15]. Hence, in the present research more than 280 samples of bitumen and asphalt were examined through several laboratory tests to study influence of RGP for bitumen modification completely.

2. 6. Tests for Bitumen ASTM D36, ASTM D5 and ASTM D113 are used for measuring physical properties of the modified bitumens, including softening point, penetration and ductility, respectively.

Nowadays, dynamic mechanical methods using oscillatory type testing are the best methods for evaluating fundamental rheological characterization of bitumen. These tests are contracted using Dynamic Shear Rheometers (DSRs). The DSR test was performed on unmodified, SBR and SBR-RGP modified bitumens. The primary viscoelastic parameters obtained from the DSR consist of the magnitude of the complex shear modulus (G*) and the phase angle (δ). G* is defined as the ratio of maximum stress to maximum strain and provides a measure of the entire resistance to deformation when bitumen is subjected to shear loading. According to ASTM D7175, the G* and δ of binders were determined by a DSR using parallel plate geometry at 10 rad/s (1.59Hz). According to the standard test method for high temperature tests (46-82 °C), specimens were 25 mm in diameter by 1 mm thick.

Storage stability of SBR-RGP modified bitumen was determined in accordance with regulations [16]. The specimen was placed in an aluminum foil tube of 32

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mm diameter and 160 mm height. The tube was packed completely without even any air outlet and placed vertically in an oven with a temperature of 163°C. After 48 h, the tubes containing the modified bitumen were cooled down to the room temperature and cut horizontally into three equal parts. The upper and lower parts were melted separately and maintained in small containers with names T and B, respectively. When the difference between T and B softening point became less than 2.5°C, the sample reached good storage stability [17, 18].

2. 7. Marshall Stability Test Marshall stability and flow tests were performed on compacted specimens with various binder contents according to ASTM D1559. In Marshall test, as an empirical test, cylindrical compacted specimens (100 mm in diameter by approximately 63.5 mm in height) were immersed in water at 60 $^{\circ}$ C for 30–40 min.; then, they were loaded to failure using curved steel loading plates along the diameter at constant compression rate of 51 mm/min.

The Marshall quotient (MQ, calculated as the ratio of stability (kN) to flow (mm)) which represents an approximation of the ratio of load to deformation under particular conditions of the test may be used as a measure of the material's resistance to permanent deformation in service [19].

2. 8. Indirect Tensile Strength and Indirect Tensile Strength Ratio In this research, 24 h soaking method was chosen as the pre-conditioning process. The procedure of this process was in the following way: The specimens of modified mixes were placed in 25 °C water bath and soaked for 24 h. Then, they were taken out and their surfaces were dried before the tests. In order to investigate moisture effect on strength of the modified mixes, the results of indirect tensile strength (ITS) after no-conditioning and 24 h soaking conducted in accordance with the procedures specified in the protocol of AASHTO T283. ITS tests were done at 25 °C with the loading rate of 51 mm/min.

2. 9. Resilient Modulus In recent years, major progress has been made in pavement design using the mechanistic approach based on elastic theory instead of more empirical approaches for pavement design. This mechanistic approach is being applied by many highway agencies recently. It should be mentioned that design methods based on elastic theory need the elastic properties of pavement materials as their main inputs. Resilient modulus of asphalt mixtures (measured in the indirect tensile mode (ASTM D4123)) is the most popular form of stress–strain measurement which could be used for evaluating elastic properties. The resilient

modulus as well as other useful parameters are applied as the inputs to the model of elastic theories in order to generate an optimum thickness design. Therefore, effectiveness of the thickness design procedure is directly related to the accuracy and precision of measuring resilient modulus of the asphalt mixture. Accuracy and precision are also important in the areas in which resilient modulus is used as an index for evaluating stripping, fatigue and low temperature cracking of asphalt mixtures [20]. In this research, indirect tensile tests were applied for both conventional and modified mixtures. Temperature which applied in the experiments was 25°C. Applied load was nearly 20% of the indirect tensile strength test at 25°C. The pulse time was chosen 1000 ms for high trafficked volume roads and 3000 ms for low trafficked volume roads.

3. RESULTS AND DISCUSSIONS

3. 1. Conventional Tests for Bitumen The effect of modification agents SBR and RGP on the properties of the base bitumen is presented in Table 4. The results show that due to increase in RGP and decrease in SBR, the penetration and softening point are decreased and increased, respectively. However, using 5% RGP modifier showed maximum decrease of 37.5% in penetration and maximum increase of 21.4% in softening point.

Decreasing and increasing in penetration and softening point demonstrate the enhancing hardness of the modified bitumen. Based on the test results; it can be concluded that RGP had significant effect on improving the stiffness and elastic properties of the bitumen. It seems that during bitumen interaction process with modifiers, the variation in bitumen properties is due to internal bitumen structure. Also, the swelling of SBR and RGP particles absorbed by bitumen oil is very good, these phenomena may lead to increase in softening point and decrease in penetration of the modified bitumen.

The bitumen modification causes reduction in temperature susceptibility. Generally, lower penetration index (PI) indicates higher thermal sensitivity. Thus, asphalt mixtures containing bitumen with higher PI have higher resistant to low temperature cracking and permanent deformation [21].

Figure 3 shows that SBR-RGP modified bitumen has less temperature susceptibility with increase in RGP and decrease in SBR to optimum PI. It is shown that 2% SBR plus 3% RGP modification provides the maximum penetration index, which indicates less temperature susceptibility. Therefore, with addition of RGP, thermal sensitivity of modified bitumen decreased and resistance to low temperature, rutting and permanent deformation increased. 1187

Also, with increase in PI the pavement rheological performance is improved for hot weather condition, and bitumen modified with RGP-SBR may be able to withstand cold temperatures. The storage stability test showed that the maximum tolerance in softening points is 1.6 which means that the presence of RGP has improved the storage stability of SBR modified bitumen significantly. This may be caused by proper dispersion of RGP in SBR modified bitumen which has led to a homogenous blend and consequently improved the storage stability. In addition, the softening point of top section of RGP-SBR modified bitumen is higher than that of the bottom section at all combinations and is increased with increase in RGP content. This can be due to the difference in densities of RGP, SBR and bitumen which leads to separate and float the SBR and RGP particles on the top of the bitumen. On the other hand a network structure is formed between the accumulated upper modifier phases which increase the softening point of the upper section.

 $G^*/Sin\delta$, the rutting parameter, at 64°C for the base and modified bitumens is shown in Figure 4. The effect

of using RGP and SBR can be easily observed by noticing that rutting parameter of the base bitumen is significantly lower than that of the modified binders. In addition, the rutting parameter decreased as the result of increasing the RGP content and decreasing the SBR content. In other words, the DSR test result indicates that at higher temperatures, the performance of SBR modified bitumen decreases with addition of RGP. As a result, use of RGP decreases SBR modified bitumen strength against rutting resistance.

3. 2. Marshall Stability Test Asphalt concrete mixtures with 5, 5.5, 6, 6.5, 7, 7.5 and 8% binder content by mass of aggregate were prepared in order to determine optimal binder content. The optimal binder content for the unmodified and modified mixtures is given as 6.1%. Results of Marshall design were obtained from compacted specimens at the optimal binder content of each bitumen type and each result is obtained from an average of three test specimens which are given in Table 5.

| Property | 5%SBR + 0%RGP | 4%SBR + 1%RGP | 3% SBR + 2% RGP | 2% SBR + 3% RGP | 1% SBR + 4% RGP | 0% SBR + 5% RGP |
|---------------------------|------------------|------------------|--------------------|--------------------|--------------------|--------------------|
| Penetration, (d-mm) | 53 | 52 | 49 | 46 | 44 | 40 |
| Ductility (cm) | 100+ | 100+ | 100+ | 100+ | 100+ | 100+ |
| Storage Stability | | | | | | |
| Top ring and ball (°C) | 53.4 | 53.7 | 55.7 | 57.9 | 58.0 | 58.6 |
| Bottom ring and ball (°C) | 50.0 | 52.1 | 54.2 | 56.9 | 57.2 | 56.5 |
| Difference | 3.4 | 1.6 | 1.5 | 1.0 | 0.8 | 2.1 |
| Penetration Index (PI) | -0.65 | -0.41 | -0.08 | 0.29 | 0.23 | 0.0003 |

TABLE 4. Basic Properties of SBR-RGP Modified Asphalt Binders

TABLE 5. Marshall Design Results

| | 1 | 2 | 3 | 4 | 5 | 6 | 7 |
|---|-----------------|--------------------|--------------------|--------------------|--------------------|--------------------|--------------------|
| Property | Base Bitumen | 5% SBR + 0% RGP | 4% SBR + 1% RGP | 3% SBR + 2% RGP | 2% SBR + 3% RGP | 1% SBR + 4% RGP | 0% SBR + 5% RGP |
| A.BSG1.(g/cm ³) | 2.643 | 2.643 | 2.643 | 2.643 | 2.643 | 2.643 | 2.643 |
| M.BSG ² (g/cm ³) | 2.389 | 2.392 | 2.385 | 2.395 | 2.388 | 2.390 | 2.387 |
| Air void, (%) | 2.99 | 2.56 | 2.94 | 2.65 | 2.49 | 2.87 | 2.79 |
| VMA ³ (%) | 13.66 | 13.51 | 13.59 | 13.65 | 13.43 | 13.49 | 13.19 |
| Marshall stability (kN) | 7.61 | 9.92 | 8.53 | 10.17 | 10.0 | 8.00 | 8.41 |
| Flow (mm) | 2.95 | 2.82 | 2.79 | 2.77 | 2.81 | 2.92 | 2.85 |
| MQ (kN/mm) | 2.58 | 3.52 | 3.06 | 3.67 | 3.56 | 2.74 | 2.95 |
| Drain down (%) | 0.287 | 0.274 | 0.272 | 0.269 | 0.268 | 0.263 | 0.261 |

¹ Aggregate bulk specific gravity, ² Mix bulk specific gravity, ³Voids in mineral aggregate



Figure 3. Penetration index for the base and modified bitumen



Figure 4. The rutting parameter at 64 °C for the base and modified bitumens



Figure 5. ITS results of unmodified and modified SMA mixes



Figure 6. TSR of unmodified and modified SMA mixes

The important criteria for Marshall stability and flow tests are the combination of high stability and low flow values. As shown in Table 5, the best results of Marshall stability and flow values were obtained with the mixture modified by 3% SBR plus 2% RGP. The increasing of stability indicates that this mixture is stronger than the unmodified mixtures. Since MQ is an indicator of the resistance against the deformation of the asphalt concrete, MQ values are calculated to evaluate the resistance of the deformation of all mixtures. The 3% SBR plus 2% RGP mixture has higher MQ value than other ones. A higher value of MQ indicated a stiffer mixture; as a result, the mixture was likely more resistant to permanent deformation.

In addition, MQ for all modified mixtures is higher than the control mixture. Consequently, it can be concluded that an improvement occurred in the Marshall properties of asphalt concrete mixtures using RGP modifier. This can be due to the absorption of bitumen oil by SBR-RGP particles.

3. 3. ITS and TSR In this study, six types of modified SMA mixes are provided in order to compare ITS results of unmodified mixes with those of SMA mixes. ITS results of each group under the dry and soaked conditions were averaged based on results of three specimens. ITS results are depicted in Figure 5. It can be found that ITS roughly increases under both dry and soaked conditions with modified mixes. As it is shown in Figure 5 the best results of ITS values were obtained with the mixture modified by 1% SBR plus 4% RGP. The measured tensile strength of all modified samples was higher than that of the control sample. Moreover, the tensile strength of SBR-RGP samples was about 13% and 31% higher than SBR and unmodified samples, respectively. Indirect Tensile Strength Ratio (ITSR) is the ratio of soaked ITS to dry ITS. The average values of the wet samples including RGP and SBR are slightly lower than those of dry samples.

Figure 6 depicts TSR results of all unmodified and modified SMA mixes. It can be seen clearly from this figure that modification of binders significantly affected TSR. TSR of unmodified mixes was at almost the smallest level (0.72) which implies that modified binders help SMA mixes to improve their moisture susceptibility. These findings indicate that use of RGP with the SBR modified bitumen can increase the tensile strength and moisture resistance of mixtures.

3. 4. Resilient Modulus The average results of the resilient modulus are shown in Figure 6. Modified mixtures show more low-temperature cracking and rutting performance. As it is shown in Figure 7, the best results of resilient modulus values were obtained by the mixture modified by 5% RGP. All of modified mixtures had higher stiffness modulus than the control mixture,

resulting in the pavement to have less strain at lower temperatures. The results show that the use of RGP with SBR modified bitumen can significantly increase the resilient modulus of mixtures.



Figure 7. Average resilient modulus for unmodified and modified SMA mixes

3. 5. Economic Analysis The unit cost of RGP and SBR modifiers depends on local conditions such as: availability, shipping distance, applicable taxes and other custom charges. Hence, a justification on the amount of saving made by replacement of SBR with RGP is suggested to be based on the proportion of unit costs for specific locations where these materials are to be provided. An investigation on the cost of these materials shows that the unit price of SBR is nearly 20 times of RGP. Thus, by replacement of 2% SBR by RGP the total cost of modifier will be reduced by almost 38%. However, it is suggested that, an enormous experimental program should be furnished to consider all possible combinations of SBR as well as RGP towards establishing the optimum combinations of RGP and SBR in bitumen and determining properties of RGP-SBR modified binder and mixture.

4. CONCLUSION

In present research, more than 280 samples of bitumen and asphalt mixtures were examined through a laboratory test program. Based on the tests, the results can be summarized as follows:

- 1. Data of penetration and softening point of the SBR-RGP modified binder demonstrated that RGP had significant effect on stiffness and elastic properties of the bitumen. By adding RGP to the mixtures, thermal sensitivity of modified bitumen was reduced and the low-temperature cracking resistance and permanent deformation increased.
- 2. The combination of high stability with low flow and

high MQ values indicated high stiffness mix with greater ability to spread the applied load and resist creep deformation. Additionally, values of MQ for all modified mixtures were higher than those of the control mixture. Therefore, improvement occurred in the Marshall properties of asphalt concrete mixtures using RGP modifier and modified mixtures were probably more resistant to permanent deformation.

- 3. The tensile strength of all modified samples was higher than the control sample. Therefore, RGP modified mixtures had more resistance to formation and propagation of cracks and moisture damage.
- 4. Mixtures modified by more RGP had higher stiffness modulus than the control mixture; therefore, the mixture should have less strain at lower temperatures.

Overall assessment of performance of asphalt mixtures showed that, by using RGP in the mixtures, improvement was observed in Marshall stability and mechanical properties. It could be also inferred that stiffness and thermal sensitivity improved. Moreover, Marshall stability, tensile strength and stiffness modulus of asphalt mixtures increased with increase of RGP content. Optimal modification was attained with 3% SBR and 2% RGP. So, RGP can be partly substituted by SBR in the bitumen modification. Application of RGP in asphalt mixtures may also have many environmental benefits and prevent accumulation of waste glass in the natural environment.

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Keywords: Recycled Glass Powder Styrene Butadiene Rubber Bitumen Modificatin Indirect Tensile Strength Resilient Modulus Stone Matrix Asphalt در این مقاله اثرات پودر شیشه بازیافتی (RGP) بعنوان جزء سوم افزودنی در قیر پلیمری مورد مطالعه آزمایشگاهی قرار گرفته است. جهت تهیه قیرها و مخلوط های اصلاح شده به قیر پایه RGP و استایرن بوتادین رابر (SBR) افزوده شدند. تحقیقات اولیه که برای تعیین خواص فیزیکی قیرهای اصلاح نشده، اصلاح شده با SBR و اصلاح شده با RGP-SBR انجام شدند، اثر سازگار کردن RGP را ثابت کردند که سازگاری بالا بین RGP و پلیمر سبب توزیع بهتر پلیمر در قیر میگردد. نتایج این تحقیق نشان دادند که خواص رئولوژیکی نهایی ترکیب مذکورتحت تاثیر افزودن RGP و RGP قرار می گیرد. قیرهای اصلاح شده و اصلاح نشده برای تهیه نمونه ها و برای انجام آزمایشات استقامت مارشال، مقاومت کششی غیرمستقیم، نسبت مقاومت کششی و مدول سختی کششی غیرمستقیم مورد استفاده قرار گرفتند. نتایج نشان داده است که ترکیب سه درصد SBR و دو درصد RGP می تواند خواص مکانیکی مخلوط آسفاتی را بطور قابل توجهی بهبود بخشد.

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چكيده