A Laboratory-based Investigation to Attain Longer-lasting Modified Binder with Crumb Rubber and Acid

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ABSTRACT: Increased traffic factors such as higher traffic volume, heavier loads, and higher tire pressure has lead to the need for higher performance pavements. Modification of bituminous binders can enhance the overall performance of a binder and therefore a pavement. Over the years several materials have been added to bituminous binder as modifiers, one of which is crumb rubber. It is more than 50 years that highway engineers around the world have tried to incorporate scrap tire rubber in bituminous pavements and it is less than 30 years that they have been successful in their efforts. Crumb Rubber Modifier (CRM) not only provides improved engineering characteristics in binder but also has the benefit of contributing to the reduction of waste tires and related environmental hazards. This paper presents the findings of a research project which was aimed at attaining the optimum composition of bitumen, crumb rubber and acid and comparing the physical and chemical properties and performance characteristics of this modified binder with neat bitumen. The tests used in this research are Rotational Viscometer (RV), Dynamic Shear Rheometer (DSR), and Bending Beam Rheometer (BBR). The paper includes analysis of the test results and explanation of sample preparation procedure.

KEY WORDS: Modified bitumen, crumb rubber, Acid, PPA.

1 INTRODUCTION

Although Hot Mix Asphalt (HMA) has many advantages, experience has shown there are shortfalls attributed to it that should be taken into account. The quality and durability of roads are becoming an increasingly important issue. The need for high-performance roads has lead researchers' attention to performance related properties and redirected the test and design methods to be based on fundamental properties. Also, several studies have been conducted to produce enhanced material by the use of additives, one of which is crumb rubber modified binder. Rubber modified binders have qualities similar to polymer modified binders while reducing the production costs.

Crumb rubber or Ground Tire Rubber (GTR) is a binder modifier that has significant potential to improve the rheological properties of bitumen. It is produced by grinding tires at either ambient or cryogenic temperatures to get a fine powder with particle sizes generally smaller than 2 mm (RMA, 2006). The first use of GTR in bitumen goes back to the mid-1960s and it has evolved since then.

There are two processes for blending Crumb Rubber Modifier (CRM) with bituminous binder: wet and dry. In the wet process, crumb rubber is mixed with and allowed to interact with binder at elevated temperatures prior to mixing with aggregate while, the dry process refers to any process that adds the rubber after or at the mixing time of binder and aggregate. In the dry process, coarser particles of CRM, acting as part of mineral aggregate, are used and limited reaction takes place between the rubber and binder. Conversely, in the wet process, a more complete reaction occurs between the two materials depending on temperature and blending time. Wet process is the most commonly used method and this research is designed based on it.

Acid is another additive type that has been used for chemical modification of bituminous binders since the 1970s. It is added to bitumen either alone or together with polymer or rubber. Different types of acids are mentioned in literature such as Polyphosphoric Acid (PPA), phosphoric acid and citric acid. Among the studied acids, PPA has shown more efficiency in improving bitumen's properties (Trakarnpruk and Chanathup, 2005). PPA is a viscous, hygroscopic and colorless liquid that exists in the form of a linear chain composed of phosphoric acid groups (Austroads, 2007).

PPA can increase the asphaltenes content by approximately 35 to 55% by mass depending on the crude source (Baumgardner et al. 2005). It also reduces the degree of asphaltene agglomeration, allowing individual asphaltenes to form a better dispersion in the maltene phase. Individual asphaltenes are more effective in contributing to elastic behavior. Moreover, a better dispersion of asphaltenes will increase the number of interactions and leads to an increase in the high temperature and a decrease in the low temperature susceptibility (Orange et al. 2004). In addition, the PPA modification can result in more stability during storage at high temperature (Daranga et al. 2009).

The objective of this study is to investigate the modification of the most commonly used Iranian bitumen with crumb rubber, VESTENAMER trans-polyoctenamer (TOR) and Acid to attain longer-lasting binder.

2 MATERIALS AND COMPOSTION

Neat bitumen selected for this study is 60-70 penetration grade bitumen provided by one of the Iran's refineries and commonly used for bituminous pavements in the country. The CRM is ambient ground rubber with the properties given in Table 1 as received from a local commercial supplier. Acid used in this research is Polyphosphoric Acid (PPA).

Table 1: Crumb rubber modifier properties

Gradation (Maximum size, mm)	0.4
Specific Gravity (g/cm^3)	1.11 to 1.15
Acetone Extract (%)	19 ± 2
Ash (Maximum, %)	15
Rubber Hydrocarbon Content (%)	48 ± 2
Mooney Viscosity	35 – 45
Tensile Strength (kg/cm^2)	50 ± 5
Carbon Black (%)	18

Sample binders modified with acid were prepared with 0, 0.5, 1, 1.5, and 2 percent PPA to find the optimum acid concentration. In the next stage, the rubber modified binders composed of the optimum acid content, 4.5% TOR by weight of ground rubber and different CRM percentages including 5, 8, 10, 12, and 15 were made to find the optimum CRM amount.

3 TESTING PROGRAM

Traditional testing on crumb rubber modified binders has mostly relied on measuring non-fundamental properties such as viscosity. However, performance related properties of bituminous pavement can be inferred from fundamental properties of the materials and processes used to create the final product. These properties can be divided to material and interaction variables. The former includes bitumen properties, that are stiffness and chemical composition, as well as CRM properties which are source and method of processing and concentration. The latter includes time and temperature (Jensen and Abdelrahman, 2006). In this research the Superpave tests were used to study the effects of rubber modification on bituminous binder.

The testing plan was subdivided into three parts: finding the optimum PPA concentration (A), the optimum crumb rubber percentage (B) and the best mixing time (C).

The tests carried out for part A were penetration and softening point. In part B, sample blend properties were measured according to Superpave binder tests. Both unaged and aged binders were tested to simulate critical stages of the binder's life. Aging was done by Rolling Think Film Oven (RTFO) and Pressurized Aging Vessel (PAV) tests to simulate the production and construction conditions, and years of in-service life respectively. Rotational Viscometer (RV), Dynamic Shear Rheometer (DSR) and Bending Beam Rheometer (BBR) tests were conducted to investigate the physical properties of binders according to the following plan:

- RV on unaged or Original Binder (OB)
- DSR on OB, RTFO and PAV aged samples
- BBR on PAV aged samples

Part C of testing plan was to determine the best mixing time. Rotational viscosity tests were performed for this purpose. The mixing times considered for this part were 1, 2, 3, 3.5, 4, 4.5, 5, 5.5, and 6 hours.

4 RUBBER MODIFIED BITUMEN PREPARATION

Each modified binder was produced in the laboratory using a mechanical mixer. Additives were added to the binder in three subsequent stages. First, the bitumen's temperature was raised to 140°C and PPA was gradually added to it at a speed of 400 rpm. Then, the temperature was elevated to 180°C and 4.5% TOR (by weight of CRM) as well as appropriate quantity of room temperature crumb rubber was continuously and gradually added to the binder. All of the rubber and TOR were added within the first few minutes of mixing and mixing continued for 45 minutes while the temperature of the binder was maintained at 180°C. After complete mixing of all material, the blending speed was reduced to 100 rpm and mixing continued for 4 hours and 45 minutes at the same temperature. As the last stage the binder was stirred in the L4RT high shear mixer at a speed of 2000 rpm for 15 minutes.

5 TEST RESULTS AND ANALYSIS

5.1 PPA Concentration

While the exact effect of PPA on binder is dependant upon origin and composition of the bitumen, generally it reduces the penetration values and increases the softening point. The best amount of PPA would be the one that increases the softening point and at the same time does not have a significant effect on the penetration values. Referring to the results of these tests for this project as reflected in Table 2, one can realize that 0.5% addition of PPA to bitumen has resulted in 14% increase in softening point and 5.9% decrease in penetration. By adding 0.5% more acid, i.e. having 1% acid in bitumen, the softening point has not changed notably (1.8% increase) but the penetration value is decreased 14.1%. Increasing the PPA content to 1.5% still does not lead to a big change in the softening point value however it causes a 12.7% decrease in penetration value as compared to 1% PPA. Therefore, the optimum PPA content for acid modification of the 60-70 grade bitumen would be 0.5%. Since the focus of this project is to modify the neat bitumen with both acid and CRM, it was decided to have a higher concentration of acid thus the PPA content was selected to be 1%.

Table 2: Test results of softening point and penetration on acid modified bitumen

Acid Content (%)	0	0.5	1	1.5	2
Penetration (mm)	68	64	55	48	42
Softening Point (oC)	50	57	58	59	61

5.2 CRM Content

After selecting the required PPA in the binder mix, samples were made with 1% acid, 4.5% TOR (by weight of CRM) and different rubber percentages using the mixing procedure explained earlier and tested as mentioned above. The results of these tests are summarized in Tables 3 to 5. The flash point test, conducted according to ASTM D92, was used to address safety concerns. This test that is only applied to unaged binders requires a minimum value of 230°C for all grades. All the modified binders meet this requirement with values above 300°C.

Table 3: Test results of unaged modified and unmodified binder

		OB							
Binder Grade	CRM (%)	Flash Point (°C)	Viscosity @ 135°C (Pa.s)	$G^*/\sin\delta$ (kPa)				
				@ 58°C	@ 64°C	@ 70°C	@ 76°C	@ 82°C
PG 58-22	0	320	0.425	2.55	**1.04**	0.50	0.26	0.14
PG 76-28	5	306	1.067	14.28	5.96	3.47	**1.64**	0.92
PG 82-28	8	308	1.235	15.58	7.64	3.57	2.34	**1.23**
PG 82-28	10	310	1.625	18.74	8.91	4.04	2.62	1.35
PG 82-28	12	312	1.845	22.13	9.86	4.89	2.80	1.44
PG 82-28	15	316	2.028	27.14	13.84	7.08	3.60	1.76

Table 4: Test results of RTFO-aged modified and unmodified binder

		RTFO				
Binder Grade	CRM (%)	$G^*/\sin\delta$ (kPa)				
		@ 58°C	@ 64°C	@ 70°C	@ 76°C	@ 82°C
PG 58-22	0	**4.17**	1.78	0.85	0.80	0.21
PG 76-28	5	23.40	11.90	4.71	**2.70**	1.33
PG 82-28	8	46.38	23.17	11.93	6.60	**3.53**
PG 82-28	10	32.57	16.38	8.56	4.89	**2.83**
PG 82-28	12	32.40	14.94	8.35	4.66	**2.57**
PG 82-28	15	30.89	12.10	8.14	4.37	**2.35**

Table 5: Test results of PAV-aged modified and unmodified binder

Binder Grade	CRM (%)	PAV						
		G^*.sin δ (kPa)			Stiffness (MPa)		m value	
		@ 22°C	@ 28°C	@ 31°C	@ -12°C	@ -18°C	@ -12°C	@ -18°C
PG 58-22	0	**4890**	2451	1532	122	208	**0.337**	0.293
PG 76-28	5	3137	**1753**	1165	79.1	202	0.355	**0.306**
PG 82-28	8	2639	1234	**665**	76.4	189	0.368	**0.316**
PG 82-28	10	3200	1237	**765**	72.6	156	0.371	**0.327**
PG 82-28	12	2258	1045	**695**	63.2	133	0.382	**0.329**
PG 82-28	15	1411	994	**681**	61.2	122	0.388	**0.331**

5.2.1 Rotational Viscosity

The rotational viscosity test, normally referred as Brookfield viscosity, was used to evaluate the change in consistency of binders or the workability at high temperatures. The test was performed according to ASTM D 4402. Since this test is a measure to ensure sufficient fluidity during pumping and mixing, it is performed on unaged bitumen. According to the Superpave mix design, the RV at 135°C should not exceed 3 Pas. All the rubber modified binders had viscosity values less than this limit and therefore meet this guideline.

5.2.2 Dynamic Shear Rheometer

The Dynamic Shear Rheometer (DSR) was used to characterize the viscous and elastic behavior of bituminous binders. In this test the complex shear modulus (G*), a measure of the total resistance of a material to deformation under repeated shear that consists of an elastic (recoverable) and a viscous (non-recoverable) part; and the phase angle (δ) of binder, an indicator of the relative amounts of recoverable and non-recoverable deformation, are measured.

Permanent deformation is controlled by limiting G*/sin δ at the test temperatures to the minimum value of 1.0 kPa for unaged binders and 2.2 kPa for RTFO-aged samples. Also, fatigue cracking is controlled by limiting G*.sin δ at the test temperature to the maximum value of 5000 kPa for PAV-aged samples. The results of G*/sin δ on original binder and RTFO-aged binders, given in Tables 3 and 4, show that by adding 5% CRM, the high temperature properties of the binder is improved such that the high temperature grade of binder is increased three grades. The addition of 8% rubber has shifted the binder grade even higher to four high temperature grades. However, adding more ground rubber will not improve the bitumen's high temperature properties. In addition, all the binders meet the maximum 5000 kPa requirement for testing on PAV-aged samples. Therefore, with respect to permanent deformation criteria, 8% CRM would be the optimum concentration.

5.2.3 Bending Beam Rheometer

The Bending Beam Rheometer (BBR) was used to measure the stiffness of binders at low temperatures. In the BBR test the following two parameters are evaluated: creep stiffness and m-value. Creep stiffness is a measure of how the bitumen resists constant loading. The binder will behave in a brittle manner if creep stiffness is too high, and as a result cracking will be more likely. The Superpave mix design has set a maximum limit of 300 MPa for creep stiffness to prevent cracking.

Moreover, m-value is used to control the rate at which the binder stiffness changes with creep load. A high m-value is desirable because it means that as the temperature changes and thermal stresses accumulate, the stiffness will change rather fast. A relatively fast change in stiffness means that the binder will tend to shed stresses that would otherwise build up to a level where low temperature cracking would occur. The minimum value of 0.3 is considered for m-value by the Superpave binder specification.

The BBR testing reveals that the rubber modification yields a one grade improvement in the low temperature grade of the bitumen. In order to find the optimum crumb rubber content, the results of BBR testing on modified binders at the test temperature (-18^{o}C), are plotted in Figure 1. It can be seen that by adding up to 10% CRM to the bitumen, the m-value increases notably, however, more addition of rubber modifier does not have a significant effect on m-value. Also, the creep stiffness curve shows that for rubber modifiers more than 10%, the stiffness values decrease with a lower rate. In other words, 10% CRM is a turning point in this curve. Hence, 10% CRM would be a better amount for an enhanced performance against cracking.

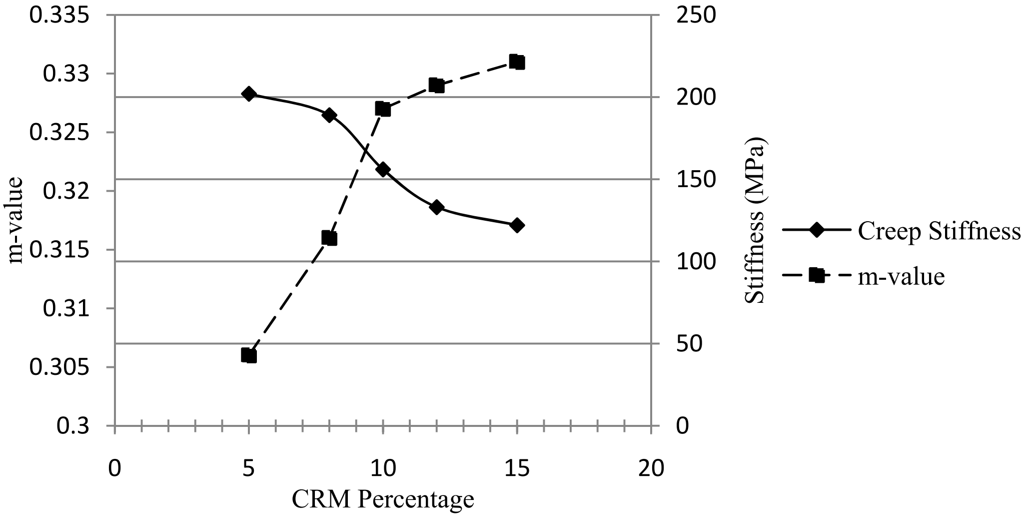

Figure 1: Creep stiffness and m-value of rubber modified binders

5.3 Interaction Time

Viscosity is a measure to find the best interaction time. During the interaction process, the rubber particles absorb oils or volatile fluids; therefore, they swell and form a viscous gel. Swelling is dependant on interaction time and temperature and causes reduction in the interparticle distance, and thus increases viscosity. If temperature is maintained too high or for too long a period after the rubber is swelled, it will lead to partial depolymerization and disintegration of rubber. This depolymerization causes a reduction in viscosity (Jensen and Abdelrahman 2006). Viscosity is an important property for pumpability and mixability of binder and workability of HMA. The compatibility of bituminous binder and CRM is controlled by the increase in viscosity versus time during blending.

A hypothesis was made by Huffman (1980) that under the effect of temperature, time or both, bitumen and rubber particles will chemically exchange their components. Based on this concept, both time and temperature must be carefully controlled to minimize this exchange of components from the rubber to the bitumen. Therefore, controlling time and temperature is critical for rubber to keep it close to its maximum possible volume and have a stiffer binder.

Abdelrahman and Carpenter (1999) showed that at low interaction temperatures (160^{o}C) swelling is continuous over the entire period, however, at intermediate temperatures (200^{o}C) the swelling is complete in the first hour and after that the swollen rubber particles start to depolymerize.

Rotational viscosity tests were carried out on the modified binder containing 1% PPA, 10% CRM and 4.5% TOR (by weight of CRM) with different interaction times. Samples were tested using two mixing methods that are blending with low shear and a combination of low and high shear mixers. In the high shear mixing, binder is stirred with a high shear mixer in the last 15 minutes of blending. The results of these tests are shown in Table 6.

Table 6: Rotational viscosity test results on the modified binder

Mixing time (hr)	1	2	3	3.5	4	4.5	5	5.5	6	6.5
Viscosity @ 135°C (Pa.s) (Low Shear Mixing)	1.61	1.95	1.8	2.8	2.98	2.86	3.29	3.41	3.53	3.72
Viscosity @ 180°C (Pa.s) (Low Shear Mixing)	0.35	0.37	0.39	0.43	0.43	0.47	0.48	0.49	0.49	0.53
Viscosity @ 135°C (Pa.s) (High Shear Mixing)	1.48	1.49	1.20	1.28	1.23	1.29	1.48	1.51	1.46	1.5

In this study the mixing temperature was 180°C, a value between the low and intermediate temperatures as used in Abdelrahman and Carpenter's (1999) study. Therefore, a continuous increase in viscosity was expected. The changes of viscosity versus mixing time are plotted in Figure 2. The viscosity at 180°C shows a linear increase that agrees with the abovementioned expectations. The data related to tests performed at 135°C with low shear mixing method, also, illustrates the same increasing linear trend. However, mixing times more than 5 hours lead to viscosity values above the 3 Pa.s limitation. Higher viscosity blends may be employed if their workability and pumpability are demonstrated. The maximum 4 Pa.s viscosity for modified binders has also been mentioned in literature.

The results of tests carried out at 135°C with high shear mixing method indicate a decrease in the viscosities corresponding to 3 to 4.5 mixing hours. This may indicate that the depolymerization begins earlier as compare to low shear mixing. Further analysis is required to clarify this finding.

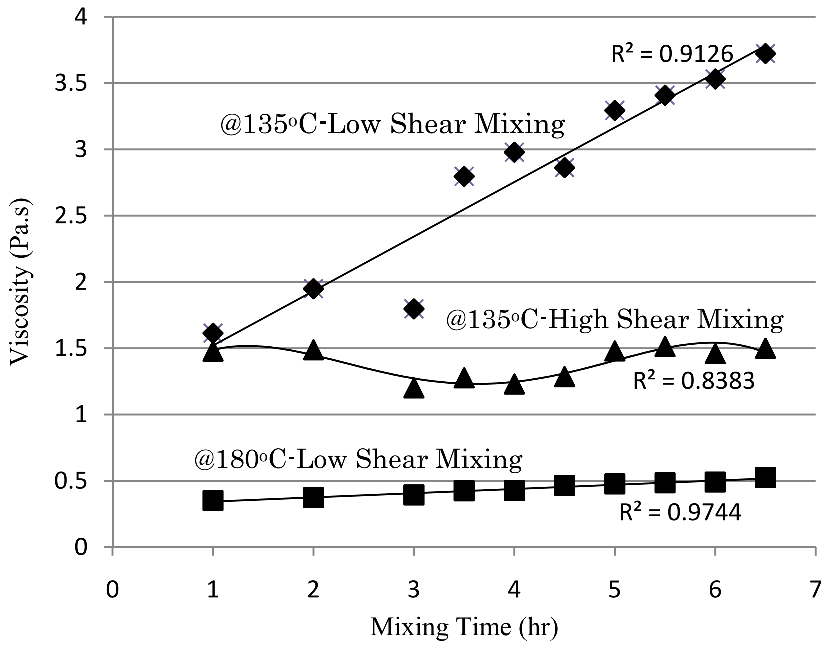

Figure 2: Viscosity changes versus interaction time

6 SUMMARY AND CONCLUSIONS

This paper described a research conducted to investigate the effect of crumb rubber and PPA modification on an Iranian bituminous binder. Five PPA concentrations and five CRM content were studied using Superpave testing methods. The following were found from these tests:

- Addition of CRM to the neat bitumen increased the high temperature grade up to four levels. The maximum high temperature grade was obtained by using 8% CRM. No more high temperature enhancement was achieved when higher amounts of rubber were added.
- The low temperature grade was improved one grade by the addition of CRM. The BBR testing revealed that for a better performance against cracking, higher CRM amounts (10%) can be used.

Moreover, the interaction time was studied using RV test on samples prepared with the optimum PPA and CRM percentages. Two mixing methods, i.e. blending with either low shear or a combination of low and high shear mixers, were used. The following were understood from this part of the study:

- The increasing trend of changes in viscosity, at 180^{o}C mixing temperature, versus interaction time implied that the swelling takes place continuously during blending.
- The results of viscosity on samples mixed using a high shear mixer showed a reduction at mixing times more than 3 hours. This may suggest that depolymerization begins earlier as compared to low shear mixing. However, further analysis is required to clarify this finding.

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