Effects of mixing procedures on the properties of polymer modified bitumen

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ABSTRACT: In this study two types of polymer pellets, Styrene-Butadiene-Styrene (SBS) and Ethylene Vinyl Acetate (EVA), were used to modify a 70/100 pen grade bitumen. Mixing variables such as, mixing speeds, mixing times and maturation times were considered in the polymer modification process. The influences of the mixing variables on the polymer modified binder were evaluated using conventional binder test methods and morphology study using florescence microscopy. The microscopy and conventional binder test result analysis in general showed the strong influence of mixing speed on polymer dispersion. The effects of mixing times on the binder test results were not distinct for both EVA and SBS polymers. The effect of pre/post stirring procedures on the binder test results was also found minimal, only the maximum speed used in the maturation process seem to influence the test results.

KEY WORDS: Polymer, SBS, EVA, microscopy, bitumen

1 INTRODUCTION

Road networks are nowadays experiencing an ever-increasing traffic volume and axle loads. Because of this reason demand for long performing pavements is increasing. The limited availability of raw materials combined with strict pavement performance requirements prompted the use of innovative materials for construction. To enhance the performance of paving materials, various research on the use of new materials and/or modifications are being carried out. For example, with respect to improving the performance of the asphalt concrete mixtures, polymer modification is widely practiced. The effectiveness of polymers in improving the performance of asphalt concrete (AC) mixtures, especially at high temperature, is widely reported in literature (Airey 2004, Isacsson et.al 1995, Lesueur 2009). Its effectiveness, however, depends on various factors. One of the factors that determine the effectiveness of the polymer modification is mixing procedure. If polymers are not mixed very well, the morphology of the resulting binder tend to show a distinct difference in phase. This means the network-like morphology within the binder essential for storing deformation energy with subsequent recovery especially at high temperature will not be obtained. Other factors that influence the polymer modified bitumens (PMBs) morphology include the type and origin of the crude, chemical composition of the bitumen and the polymer and thermal history (Lesueur 2009). Depending on these factors, several types of interactions between the different components occur influencing the final bitumen –polymer system.

For quality control performance related binder tests are carried out in the laboratory. This means it is of paramount importance that the mixing process in the laboratory at best reproduces conditions in the field. In the laboratory the morphology of PMBs differs with mixing temperatures (Soenen et.al. 2009). There are also indications that the morphology of the PMBs in the laboratory differs from the morphology of the PMBs in Asphalt mixtures (Soenen et.al.2009, Wegan et.al. 1999, Wegan et.al. 2001). The change in morphology could be the result of chemical interactions that develop when the binder is mixed with fillers, sand and aggregates. The implications of the change in morphology on the performance of the mixture are not yet known. While this is the case for binders and asphalt mixtures prepared in the laboratory in a controlled environment, the approach used in practice may yet introduce other variables. Hence, for the mixing procedure that is being practiced in asphalt plants, it is important to get insight on the mixing levels of the PMBs.

The polymers commonly used in road engineering are usually available either as pre-blends or in the form of small pellets. In this study, the term pre-blend refers to polymer modified bitumen as supplied by bitumen producers in hot bulk. Although pre-blends are preferred for use, their usage in practice is not always feasible. When used on irregular basis, pre-blends have both adverse logistical and qualitative consequences. The latter is caused by too long storage at the asphalt plant. From practical point of view the use of polymers in the form of pellets is more attractive. This is because their usage does not require additional measures to be taken when producing asphalt. However, when considering the mixing time used in an asphalt plant, which usually vary from 30 seconds up to a maximum of 90 seconds, questions may arise in relation to the extent to which the polymer pellets are well blended in the asphalt mixture.

The work presented in this article is performed to obtain insight into the mixing process for two polymers in the form of pellets. Hence, the variables that could affect the polymer pellets mixing process; such as mixing speeds and mixing time are considered. For quantification, conventional binder test methods, such as pen and ring-and-ball tests, are utilized. In addition to these, morphology study using fluorescence microscopic is also conducted. The paper presents details on the materials, laboratory tests, results, analysis and discussions.

2 MATERIALS AND EXPERIMENTS

To obtain quantitative information with respect to the mixing of polymer pellets, a laboratory study was conducted using two types of polymer pellets: one SBS and one EVA. Using a Silverson LM5A high shear laboratory mixer, several blends were prepared using a pen 70/100 base bitumen under varying conditions. The properties of the bitumen used in this study are summarized in Table 1 and 2. The amount of PMB modification was kept constant at 6% by mass for all cases. A mixing temperature of 180°C was used. The properties of the used materials in this study are summarized in the tables below.

Table 1: Properties of the neat bitumen used in this study

Reference	***70/100 bitumen***	
Test no.	Pen (dmm)	Tr&b (°C)
1	72	46,5
2	73	46,4
3	72	

Table 2: SARA analysis of the bitumen used in this study

Fraction	***Content (%)***
Saturates	4,0
Aromatics	57,5
Resins	21,2
Asphaltenes	17,3

The polymers used in this study appear in pellet form. The properties, as supplied by the producers, are summarized in Table 3 and Table 4.

Table 3: Properties of the EVA polymer pellets

Density (g/cm³)	0.949
Melt Index (g/10 min)	1.8
Vinyl Acetate Content (wt%)	24.0
Peak Melting Temperature (°C)	77

Table 4: Properties of the SBS polymer pellets, supplied by producer

Bitumen blend	***1***	***2***
Pen 30/45 (%)	100	93
SBS pellet (%)		7
Properties		
Tr&b (°C)	52	59
Penetration 25°C,100g, 5s (dmm)	42	37
Viscosity at 180°C (cP)	123	201

An laboratory high shear mixer using a specially designed head was used to achieve optimal blending and dispersion of the polymer pellets, see Figure 1. After placing the tin of an weighed amount of bitumen on the bottom plate, which is temperature controlled and part of the mixer, the temperature was set to 180°C and the bitumen was left for conditioning for at least 2 hours. Hereafter an fixed amount of 6 % by mass of the polymer pellets was added gradually into the hot bitumen.

Figure 1: Laboratory high shear mixer used in this study (left), and specially designed head for blending polymer pellets (right)

To investigate the effect of mixing time, speed and maturation time, the following three cases were considered:

- Case 1: Effect of mixing time
  - Constant mixing speed of 5000 rpm and mixing times of 0.5, 1 and 2 hours.
- Case 2: Effect of mixing speed
  - Constant mixing time of 2 hours and mixing speeds of 2500, 5000 and 7000 rpm.
- Case 3: Influence of maturation time:
  - 1 hour of pre-stirring at 2500 rpm followed by 1 hour stirring at 7000 rpm
  - 1 hour of pre-stirring at 5000 rpm followed by 1 hour stirring at 2500 rpm
  - 1 hour stirring at 5000 rpm followed by 2 hour of stirring at 2500 rpm

For the above cases, the polymer blends were characterized using empirical bitumen test methods. Hence Penetration and Softening point tests according to the European standards EN 1426 and EN 1427 were made. In order to get insight on the morphology, more fundamental understanding was generated by examining the several blends by means of fluorescent microscopy.

3 RESULTS & DISCUSSION

3.1 Conventional binder test results

3.1.1 Case 1: Effect of mixing time

In case 1, the effect of mixing time was investigated using a constant mixing speed of 5000 rpm. Penetration and ring-and-ball test results for samples that were taken at 0.5h, 1h, and 2h are presented in Figure 2. It can be seen from Figure 2 that for SBS modifications the effect of mixing time was not clearly seen in the pen and softening point results. However, the presence of EVA polymer in the binder is reflected through the reduction of pen and an increase of the softening point values. For the SBS, a slight reduction in pen value is observed, however, the softening point result showed no clear sign of the polymer presence in the binder. The individual test results are summarized in Table 5 and Table 6.

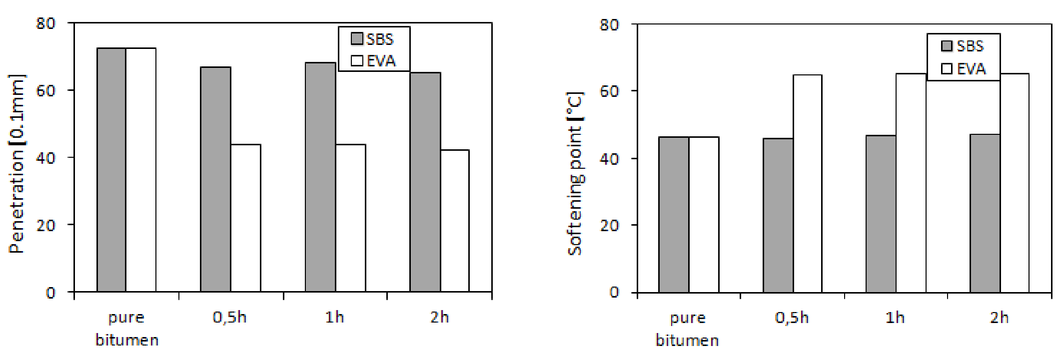


Figure 2: Effect of mixing time on empirical properties of PMB (the mixing speed is constant at 5000rpm)

Table 5: Individual test results, bitumen with EVA pellets

Mixing time	***0,5h***		***1h***		***2h***	
Test no.	Pen (dmm)	Tr&b (°C)	Pen (dmm)	Tr&b (°C)	Pen (dmm)	Tr&b (°C)
1	44	64,6	44	65,3	42	65,3
2	44	64,8	43	65,3	42	65,3
3	43		44		43	

Table 6: Individual test results, bitumen with SBS pellets

Mixing time	***0,5h***		***1h***		***2h***	
Test no.	Pen (dmm)	Tr&b (°C)	Pen (dmm)	Tr&b (°C)	Pen (dmm)	Tr&b (°C)
1	67	45,7	68	46,8	65	47,3
2	67	46,2	68	46,8	65	47,3
3	67		68		64	

3.1.2 Case 2: Effect of mixing speed

In this case for a constant mixing time of 2 hours, mixing speeds were varied. The penetration and softening results corresponding to the various mixing speeds are presented in Figure 3. The individual results are summarized in Table 7 and Table 8.

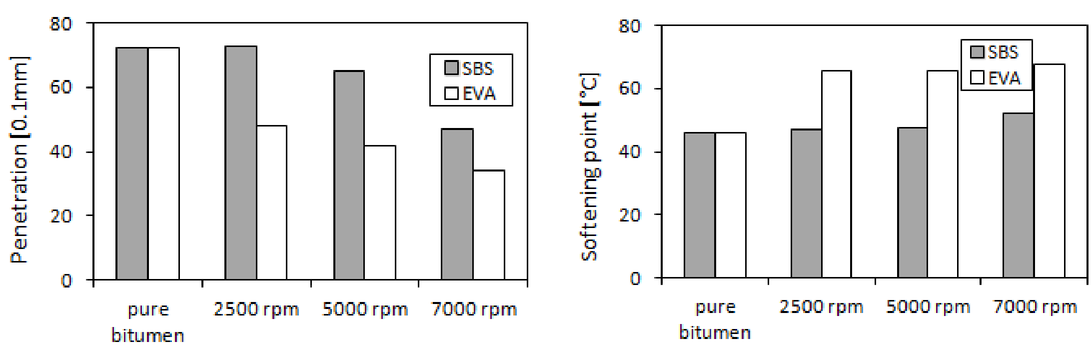


Figure 3: Effect of mixing speed on empirical properties of PMB (the mixing time is constant at 2 hours)

Table 7: Individual test results, bitumen with EVA pellets

Mixing speed	***2500 rpm***		***5000 rpm***		***7000 rpm***	
Test no.	Pen (dmm)	Tr&b (°C)	Pen (dmm)	Tr&b (°C)	Pen (dmm)	Tr&b (°C)
1	47	65,2	42	65,3	33	67,4
2	48	65,3	42	65,3	33	67,5
3	48		43		35	

Table 8: Individual test results, bitumen with SBS pellets

Mixing speed	***2500 rpm***		***5000 rpm***		***7000 rpm***	
Test no.	Pen (dmm)	Tr&b (°C)	Pen (dmm)	Tr&b (°C)	Pen (dmm)	Tr&b (°C)
1	72	46,7	65	47,3	46	52,0
2	74	47,1	65	47,3	47	52,0
3	74		64		47	

Figure 3 shows the effect of mixing speed on pen values for both SBS and EVA binders. The effect of mixing speeds is clearly observed for both PMBs on the Pen results. The effect of speed on the Softening point results is not clearly reflected.

3.1.3 Case 3: Effect of pre/post stirring procedures

The results presented in Figure 4 are obtained from the tests carried out to investigate the effect of slow, and fast, pre/post stirring procedures. From the results in Figure 4 no distinct difference has been observed on the softening point results. On the Pen values, the result for case 3-1 has shown slightly lower pen values than case 3-2 and 3-3. The maximum speed used for case 3-1, 7000 rpm, is higher than the maximum speed, which is 5000 rpm, used other two cases. From Section 3-1 and Section 3-2, it was observed that mixing speed has more influence than mixing time on Pen values. The observed difference in Pen values in this case is, therefore, most likely a result of the maximum mixing speed used rather than the pre/post stirring effects. Individual test results can be found in Table 9 and Table 10.

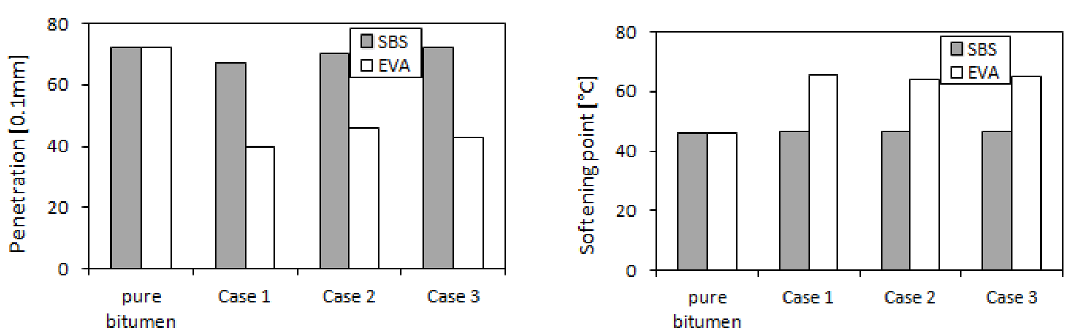


Figure 4: Effect of pre/post stirring procedures on empirical properties of PMB

Table 9: Individual test results, bitumen with EVA pellets

Case no.	***3-1***		***3-2***		***3-3***	
Test no.	Pen (dmm)	Tr&b (°C)	Pen (dmm)	Tr&b (°C)	Pen (dmm)	Tr&b (°C)
1	40	65,4	45	64,1	43	65,0
2	40	65,4	47	64,2	44	65,1
3	40		47		43	

Table 10: Individual test results, bitumen with SBS pellets

Case no.	***3-1***		***3-2***		***3-3***	
Test no.	Pen (dmm)	Tr&b (°C)	Pen (dmm)	Tr&b (°C)	Pen (dmm)	Tr&b (°C)
1	67	46,6	72	46,6	72	46,6
2	67	46,8	72	46,7	71	46,6
3	67		72		72	

For the combination of mixing times and speeds used in this work, results were analyzed. Based on the conventional binder test results, the following conclusions can be drawn:

- Test results partially reflected the presence of polymers in the binder. Modification of the bitumen with EVA resulted in a lower Pen value and higher softening point. For the case of SBS modification, the polymer presence was reflected only in the Pen values.

- Effect of mixing speed was reflected in Pen values. Higher mixing speeds resulted in lower pen values. This may be a result of good blending.

- Effect of mixing time and pre/post stirring procedures were not observed in the test results

For each of the cases investigated in the previous sections, morphology study was also conducted using Fluorescence Microscopy. The obtained results are presented in the following section.

3.2 Fluorescence Microscopy Analysis

3.2.1 Case 1: Effect of mixing time

Figure 5 and Figure 6 show results from microscopy analysis for EVA and SBS binders, respectively. The three images in each figure correspond to the three different mixing times. The figures show a slight change in the morphology between the different mixing times. In Figure 5, light spheres with a diameter of about 0.5 mm to 1 mm can be observed after 1h of mixing. This is believed to be the swelling of the EVA pellets. These spheres grow bigger after 2h of mixing. The change in morphology between the different mixing times presented in Figure 5 were however not detected in the pen and softening point results presented in Figure 2.

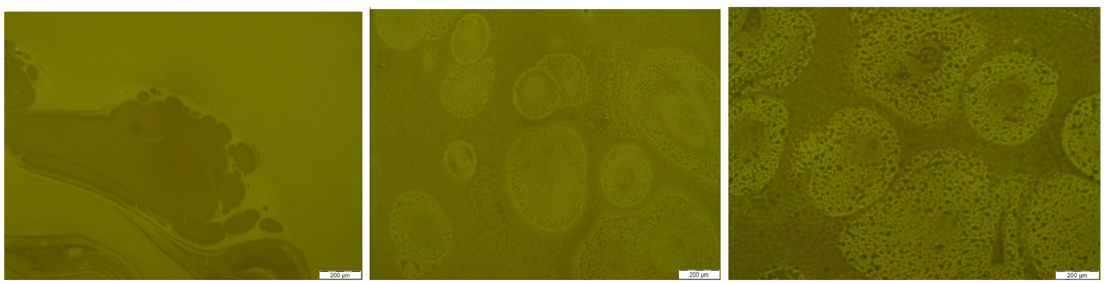


Figure 5: Effect of mixing time of 0.5h (left), 1h (middle) and 2h (right) on morphology of EVA modified bitumen (the mixing speed is constant at 5000rpm)

As shown in Figure 6, the morphology of the SBS modified binder did not show significant changes between 0.5h and 2h of mixing. This is in agreement with the results obtained from the conventional tests, which also showed no significant change with mixing time. The shining spots observed in the morphology are believed to be the SBS molecules. The size of the spot is around 10 micron. A slight cluster of these spots was observed with 0.5h of mixing. After 2h of mixing the spots were dispersed. The morphology of the SBS pellet modifiers in bitumen is very different comparing with the typical morphology of the SBS modified bitumen (pre-blend), where the SBS molecules form a network-like morphology in bitumen. This could also explain why the SBS modifiers used did not show any difference in terms of conventional test results.

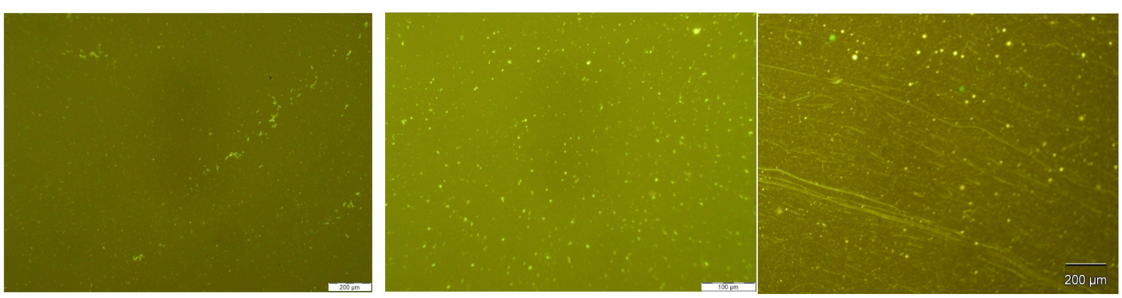


Figure 6: Effect of mixing time on morphology of SBS modified bitumen (the mixing speed is constant at 5000rpm,0.5h (left), 2h (middle)) and the typical morphology of an pre-blend SBS modified bitumen (right)

3.2.2 Case 1: Effect of mixing speed

Figure 7 and Figure 8 show results for EVA and SBS binders, respectively. The three images in each figure correspond to the three different mixing speeds. In Figure 7, there exist distinct changes in the morphology of the binder at high speeds. This is also supported from the pen and softening tests presented in Figure 3. For SBS modification, the morphology changes are not distinct. This can be seen from Figure 8.

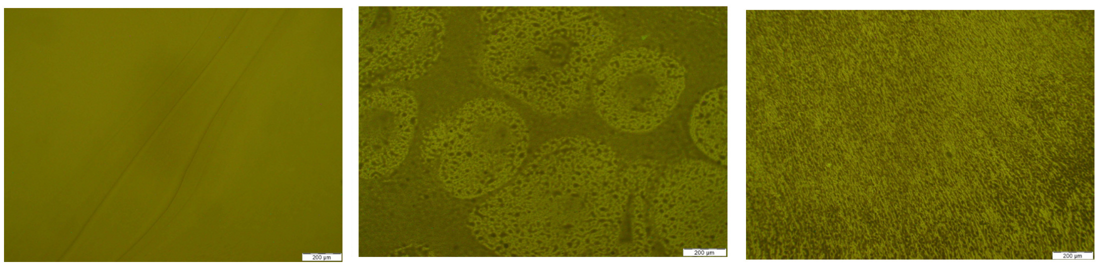


Figure 7: Effect of mixing speed of 2500rpm (left), 5000rpm (middle) and 7000rpm (right) on morphology of EVA modified bitumen (the mixing time is constant at 2 hours)

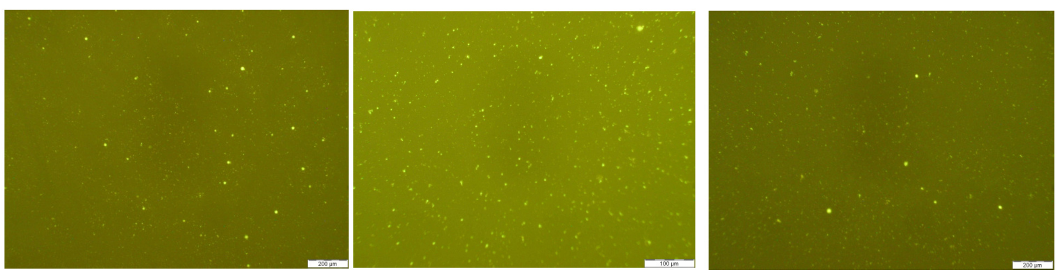


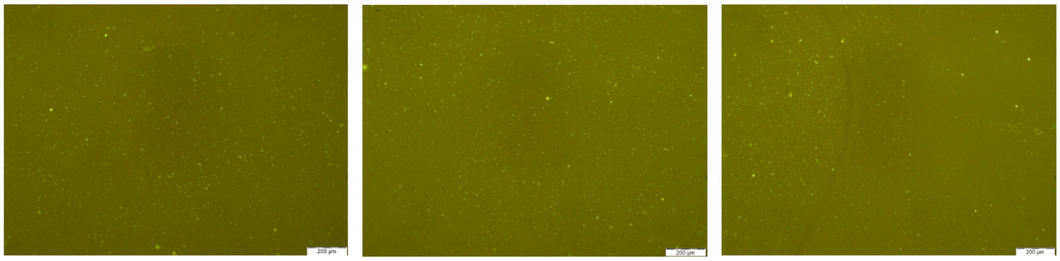
Figure 8: Effect of mixing speed of 2500rpm (left), 5000rpm (middle) and 7000rpm (right) on morphology of SBS modified bitumen (the mixing time is constant at 2 hours)

3.2.3 Case 3: Effect of pre/post stirring procedure

The effect of pre/post stirring, are presented in Figure 9 and Figure 10. Similar to the results from Pen and softening point tests, no distinct difference in morphology is observed. In the case of the EVA modifications, it was expected similar morphology as Figure 7 would be obtained. However, the spheres observed in the morphology are missing. This was believed to be a result of possible inconsistencies in sampling procedures.

Case 3-1 Case 3-2 Case 3-3

Figure 9: Effect of pre/post stirring procedures on morphology of EVA modified bitumen



Case 1 Case 2 Case 3

Figure 10: Effect of pre/post stirring procedures on morphology of SBS modified bitumen

4 CONCLUSIONS & RECOMMENDATIONS

Taking into account practical considerations, the aim of this study was to develop a better understanding on the mixing process of polymer pellets with bitumen. For this purpose, two kinds of polymer pellets were used to modify a pen 70/100 conventional binder. Mixing variables such as speed and time were varied, and effects were evaluated based on conventional binder test methods and morphology study using fluorescence microscopy. Based on the obtained results, the following conclusions are drawn:

- Both the morphology and the penetration test results show the significant effect of mixing speeds. Higher mixing speeds seem to result in lower pen values.
- The effect of mixing time was not observed in the conventional binder test results. Slight changes were however observed in the morphology.
- The effect of pre/post stirring was also minimal. Only the maximum speed utilized in the process seem to influence the polymer blending.
- For the EVA pellets: When adding EVA pellets to bitumen a direct influence on conventional bitumen test results was observed. Through all the studied cases, a constant increase of softening point around 20°C was observed while the decrease of the penetration values remained constant from 20 to 30 dmm. A higher mixing speed results in better blending as observed from the morphology results.
- For the SBS pellets: No distinct influence of the SBS was observed on the results obtained from the conventional binder tests. Only a slight reduction in pen value was observed. No change in blending can be observed from the morphology results.

Although conventional penetration and softening point tests were not sensitive enough to demonstrate differences within all the studied process parameters, the results from this study have shown that mixing speed is important in determining the extent of polymer dispersion. For the range of times used in this study, mixing times was found to have limited effect on dispersion. In practice, a much shorter mixing time of 30s to 60s is used in mixing polymer pellets in asphalt plants. Considering practical situations, further studies will be performed on mastics, mortars levels and also mixture level in order to better understand the influence of polymer dispersion on the mechanical behavior, e.g. by utilizing more fundamental test methods, such as dynamic shear Rheometer (DSR).

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