The International Journal of Pavement Engineering and Asphalt Technology (PEAT) ISSN 1464-8164.
Volume:19, Issue:2, December 2018

INVESTIGATION OF THE EFFECTS OF ADDITIVES ON MOISTURE SUSCEPTIBILITY OF ASPHALT MIXES CONTAINING SULFUR-POLYMER

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doi: 10.1515/ijpeat-2016-0024

ABSTRACT

Large amounts of sulfur is produced in gas production refineries. From these, only small amounts are used in various applications and the rest remain unused. Sulfur deposits not only impose heavy economic costs, but these impart numerous adverse effects on the environment. The application of sulfur in asphalt mixes is rather simple. It requires minor changes to asphalt plants in order to produce a sulfur asphalt mix. However, according to previous researches, it was recognized that bituminous mixes containing sulfur are susceptible to moisture damage and show early stripping problems. Moreover, at mixing and laying stages of sulfur asphalt mixes, excessive H_2S emission could be harmful for the operators.

In this research a sulfur polymer additive, named Googas, was used to produce sulfur polymer mixes that had little H_2S emission and provided some plastic properties to mixes. In order to improve performance of asphalt mixes containing Googas, three additives, namely; Crumb Rubber, Zycotherm and Nano Clay were used in mixes and their properties were determined. All were added at mixing stage of aggregates, bitumen and sulfur polymer. With the purpose of evaluating performance of the above mixes, Indirect Tensile Strength and Marshall Tests were carried out. From these, Tensile Strength Ratio (TSR) and Marshall Residual Stability (MRSR) parameters were determined. The results indicated that the addition of Crumb Rubber, Zycotherm and Nano Clay to sulfur polymer mixes resulted in improved moisture resistance of mixes. The best improvement was achieved in mixes containing 40% binder content of sulfur polymer and 15% Crumb Rubber. This resulted in average 23% increase in moisture resistance of mixes.

Keywords**:** Crumb rubber; Moisture susceptibility; Nano clay; Sulfur polymer; Zycotherm.

1. INTRODUCTION

A significant amount of sulfur remains deposited near gas production refineries. This not only imposes heavy economic costs, it imparts also numerous environmental problems. Due to very low price of sulfur, its application in asphalt mixes is quite feasible. It requires simple changes in asphalt plants in order to produce sulfur asphalt mixes (Javadi, 2005 and Weidong, 2006). However, ordinary sulfur asphalt mixes produce harmful and unpleasant gases. This is why some producers have modified sulfur using some polymers (Timm et al, 2010). A product of these families, called Sulfur-Polymer (SPAM) or Googas, is produced in Iran and has been used in several projects. The weak point of mixes containing this product is their moisture susceptibility (Timm et al, 2010).

Since early 1970s researchers have concluded that moisture has sever damaging effects on asphalt pavements (Little and Epps, 2001 and Liu et al, 2011). This phenomenon resulted in damages in asphalt mixes in West and South East of USA and millions of dollars were spent to repair the damages (Little and Epps, 2001). Moisture susceptibility can be defined as loss of resistance and reduced durability of asphalt mixes, caused by adverse effects of moisture. Moisture susceptibility is the tendency of asphalt mixes to strip which will then lead to develop several other distresses, including cracking, rutting, raveling, and potholes (Sungun et al, 2014 and Ojum et al, 2017). In a study, it was determined that the damaging effects of moisture resulted in reduced asphalt modulus (up to 25%); increased rutting (up to 60%); and increased fatigue cracking (Mehrara and Khoadii, 2013). In this research, it was revealed that among asphalt mixes containing various additives, the ordinary sulfur asphalt mix was recognized to be the most moisture susceptible mix that requires further treatments before being laid on pavements.

In this research, with the aim of reducing moisture susceptibility of sulfur polymer asphalt mixes (SPAM), three additives, namely crumb rubber, Zycotherm and nano clay were used in SPAM mixes at various amounts. The effects of Zycotherm on moisture susceptibility of asphalt mixes was investigated by Ranka (2012). The results of this latter study showed significant effects of Zycotherm in improving moisture resistance of asphalt mixes. In fact, replacing 0.1 and 0.15% of the mix binder content with Zycotherm resulted in 14 and 7% increase in indirect tensile strength ratio of mixes, respectively.

The application of Crumb Rubber in asphalt mixes has been studied by different researchers. In a research that was performed on mixes containing Crumb Rubber, it was shown that these mixes have better performance against fatigue cracking (Hainian et al, 2013). The effects of Crumb Rubber on rutting and moisture susceptibility of asphalt mixes was also investigated (Sungun et al, 2014). In this research, Crumb Rubber was used at 8, 10 and 12 percent of the binder content of the mix. The results showed significant improvement in rutting and moisture resistance of asphalt mixes.

In an experimental research, the effects of Nano Clay on moisture susceptibility of mixes was evaluated (Hossain et al, 2014). In this research, Nano Clay was used at 4 and 6 percent of the binder of the mix. Results showed increased tensile strength ratio and reduced moisture susceptibility of asphalt mixes. Similar results were achieved in another research, showing that Nano Clay resulted in improved moisture susceptibility of asphalt mixes (Shu et al, 2011). The effects of Nano Clay on fatigue and rutting performance of asphalt mixes was investigated in Iran too. Results showed significant improvements in various performance parameters of the mix (Ghaffarpour, 2011).

2. MATERIALS CHARACTERISTICS

In this research, in order to investigate the effects of the above three additives on sulfur polymer asphalt mixes; one source of aggregate was selected. This was from a quarry in Yazd province in central part of Iran. A 60/70 penetration grade bitumen, produced in Refinery of Isfahan, was also used in the whole research. All sulfur polymer mixes were continuously graded aggregates. Crumb Rubber modified binder was produced applying wet processing and using a high shear mixer. Major physical testing results of the aggregates and the bitumen binder are reported in Tables 1 and 2 respectively.

Table 1. Major physical testing results of the aggregates

Coarse Aggregates				
Test	**Standard Method**	**Iran Specification limits**		**Result**
		Minimum	Maximum	
Los Angeles Abrasion (%)	ASTM-C131	-	25	20
Sodium Sulfate Soundness (%)	ASTM -C88	-	8	8
Fracture Faces (%)	ASTM -D5821	60	-	100
Flaky and Elongated Particles (%)	ASTM -D4791	-	15	9
Fine Aggregates				
Plasticity Index (%)	ASTM -D4318	NP	NP	NP
Sodium Sulfate Soundness (%)	ASTM -C88	-	12	0.7
Sand Equivalent (%)	ASTM -D2419	50	-	70

Table 2. Testing results of the 60/70 pen bitumen used in mixes

Testing	Standard Method	Specifications limits		Result
		Minimum	Maximum	
Penetration (0.10 mm)	ASTM-D5	60	70	64
Softening Point (°C)	ASTM -D36	49	56	50.5
Ductility at 25°C (cm)	ASTM- D113	100	-	+100
Specific Weight (g/cm^3)	ASTM- D3289	1.013	1.017	1.018
Kinematic Viscosity at 135°C	ASTM- D2170	200	1000	326
Thin Film Oven Test (163°C for 5 h)	ASTM- D1754			
Change in Mass (%)		-	0.8	0.03
Penetration after TFOT (0.10 mm)		31	-	44
Ductility after TFOT (cm)		50	-	+100

3. TESTING RESULTS

Several testing methods can be performed on asphalt mixes in order to determine their moisture susceptibility. In this research, Marshall samples were prepared and advanced Marshall parameters and indirect tensile properties of the samples were determined. The results are reported in the following sections.

3.1. Marshall Residual Stability Ratio

Two different series of Marshall Samples were prepared. The first series were untreated dry samples. The second series consisted of samples that were saturated and were then subjected to one freeze-thaw cycle. In this latter treatment, the compacted samples were freeze treated at -18°C for two hours, as some researchers (Sengul et al., 2012) suggested that. These were then kept at room temperature for further two hours before being immersed in water for 24 hours at 60°C. Then were cooled to room temperature for two hours. Finally, the samples were placed in water at 60°C for 30 minutes before being subjected to Marshall testing at 60°C. From this, stability and flow values of both untreated and treated (i.e. samples subjected to the above mentioned conditions) were determined. The results are reported in Table 3. The results are the average of six samples for each series. From these results, a parameter named Marshall Residual Stability Ratio (MRSR) was determined. This is defined as the ratio of stability of treated samples to that of untreated ones. Fig. 1 reports MRSR results of samples containing various additives at different amounts. With reference to Table 3, it can be observed that the mix in that its binder content was consisted of 15% crumb rubber and 50% sulfur polymer resulted in greater Marshall Stability values (up to 23% increase). While the mix in that its binder content was consisted of

15% crumb rubber and 40% sulfur polymer resulted in 14% increase in Marshall stability.

Table 3. Marshall Stability, flow and MRSR values of the various samples

Mix samples with various additive compositions	**Before Conditioning (Dry)**		**After Conditioning (moisture and freeze-thaw)**		
	Stability (Kg)	**Flow (mm)**	**Stability (Kg)**	**Flow (mm)**	**MRSR (%)**
30% Sulfur-Polymer	1330	1.96	1077	1.56	81
40% Sulfur-Polymer	1440	1.86	1193	1.50	83
50% Sulfur-Polymer	1497	1.80	1203	1.36	81
30% Sulfur-Polymer – 1.5% Nano Clay	1460	2.67	1371	2.46	94
40% Sulfur-Polymer – 1.5% Nano Clay	1591	2.70	1484	2.30	93
50% Sulfur-Polymer – 1.5% Nano Clay	1613	2.76	1520	2.40	94
30% Sulfur-Polymer – 15% Crumb Rubber	1443	2.53	1337	2.10	92
40% Sulfur-Polymer – 15% Crumb Rubber	1561	2.36	1479	1.60	95
50% Sulfur-Polymer – 15% Crumb Rubber	1598	2.35	1478	1.67	93
30% Sulfur-Polymer – 0.15% Zycotherm	1346	3.15	1260	2.80	93
40% Sulfur-Polymer – 0.15% Zycotherm	1457	3.10	1373	2.60	94
50% Sulfur-Polymer – 0.15% Zycotherm	1515	3.00	1417	2.50	93

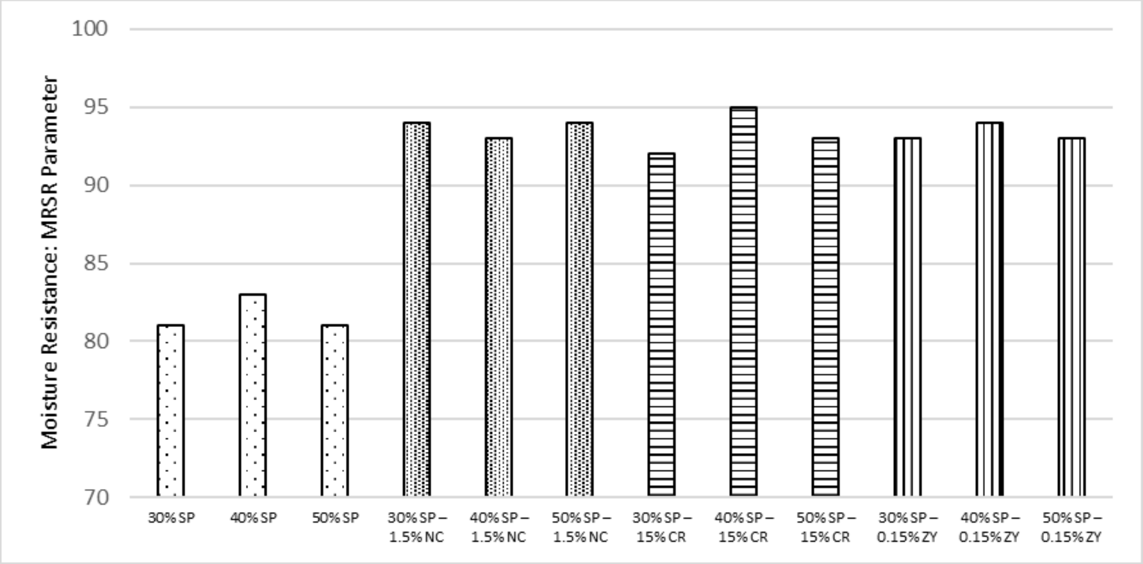

Fig. 1. Marshall Residual Stability Ratio (MRSR) of mixes containing various additives

3.1.1. Moisture Resistance

Samples consisting of the combination of various additives were compacted applying Marshall Hammer. Additives in various samples were consisted of the following combinations:

30% Sulfur-Polymer
40% Sulfur-Polymer
50% Sulfur-Polymer
30% Sulfur-Polymer – 1.5% Nano Clay
40% Sulfur-Polymer – 1.5% Nano Clay
50% Sulfur-Polymer – 1.5% Nano Clay
30% Sulfur-Polymer – 15% Crumb Rubber
40% Sulfur-Polymer – 15% Crumb Rubber
50% Sulfur-Polymer – 15% Crumb Rubber
30% Sulfur-Polymer – 0.15% Zycotherm
40% Sulfur-Polymer – 0.15% Zycotherm
50% Sulfur-Polymer – 0.15% Zycother

With reference to Fig. 1, it can be seen that with adding nano clay to sulfur polymer asphalt mixes (SPAM), MRSR values of mixes were increased greatly. This shows the positive role of Nano Clay in increasing moisture resistance of SPAM mixes.

When crumb rubber (CR) was added to both control samples and those containing SP additive, MRSR values of the samples were increased significantly. For instance, MRSR values of three asphalt mix compositions that contained 30% SP-15% CR, 40% SP-15% CR and 50% SP-15% CR were increased by 13, 17 and 15% respectively.

Similar results were obtained from samples containing Zycotherm additive. The extent of increase for mix samples that contained 3% SP-0.15% Zycotherm, 40% SP-0.15% Zycotherm and 50% SP-0.15% Zycotherm were 16, 15 and 15%.

3.2. Tensile Strength Ratio

With reference to Marshall testing and after achieving the optimal bitumen content of 4.3%, samples containing various additive combinations were prepared for evaluation of their moisture susceptibility. Indirect Tensile Strength (ITS) testing was performed and Tensile Strength Ratio (TSR) of the samples were determined. ITS testing was recognized by other researchers to be a suitable testing method for evaluating the effectiveness of polymer additives (e.g. Shuklamanoj et al, 2014). The summary results of these parameters are reported in Table 4. Fig. 2 shows comparison of TSR results of SP asphalt samples containing various combinations of additives.

Table 6. ITS testing results of sulfur polymer mixes containing various additives

Additive combinations in mixes	**Indirect Tensile Strength, dry condition (kPa)**	**Moisture susceptibility**	
		Indirect Tensile Strength, saturated condition(kPa)	**TSR Ratio (%)**
30% Sulfur-Polymer (30% SP)	728	532	73
40% Sulfur-Polymer (40% SP)	692	492	71
50% Sulfur-Polymer (50% SP)	558	373	68
30% Sulfur-Polymer – 1.5% Nano Clay (30% SP-1.5% NC)	948	777	82
40% Sulfur-Polymer – 1.5% Nano Clay (40% SP-1.5% NC)	898	727	81
50% Sulfur-Polymer – 1.5% Nano Clay (50% SP-1.5% NC)	890	703	79
30% Sulfur-Polymer – 15% Crumb Rubber (30% SP-1.5% CR)	943	886	94
40% Sulfur-Polymer – 15% Crumb Rubber (40% SP-1.5% CR)	862	784	91
50% Sulfur-Polymer – 15% Crumb Rubber (50% SP-1.5% CR)	798	702	88
30% Sulfur-Polymer – 0.15% Zycotherm (30% SP-1.5% ZY)	780	647	83
40% Sulfur-Polymer – 0.15% Zycotherm (40% SP-1.5% ZY)	820	656	80
50% Sulfur-Polymer – 0.15% Zycotherm (50% SP-1.5% ZY)	850	662	78

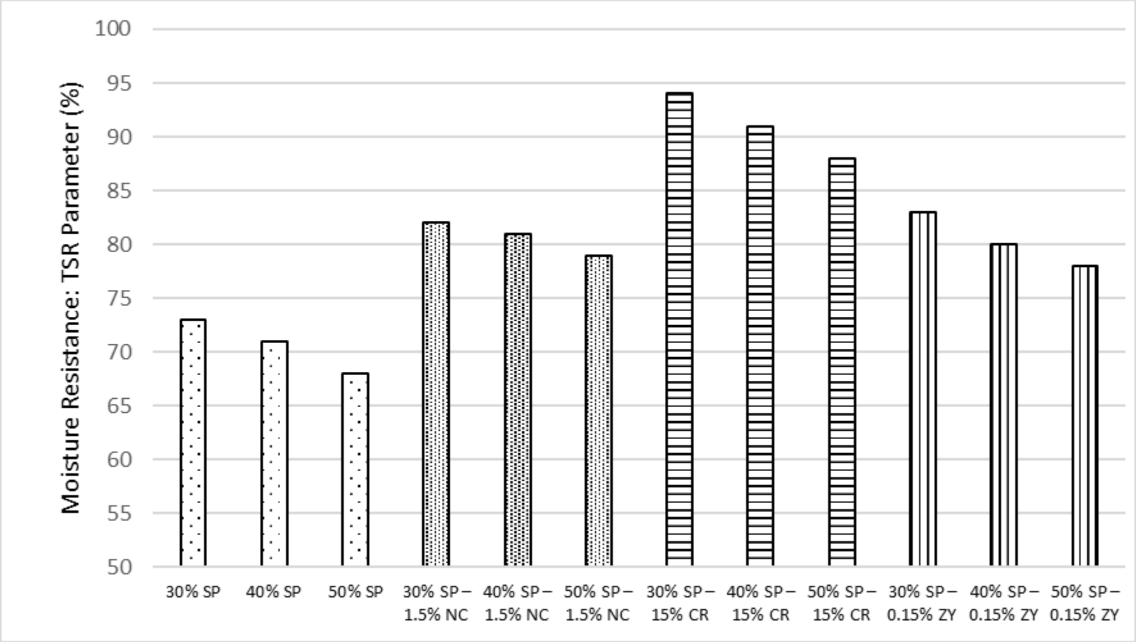

Fig. 2 Comparison of TSR results of sulfur polymer mixes containing various additives

3.2.1. Moisture Damage Resistance

The addition of Nano Clay to SP asphalt mixes resulted in increased indirect strength resistance of both dry and saturated samples. TSR parameter of asphalt mixes containing 30% Sulfur-Polymer-1.5% Nano Clay, 40% Sulfur-Polymer-1.5% Nano Clay and 50% Sulfur-Polymer-1.5% Nano Clay showed TSR increases of 16, 14 and 11%, respectively.

It was also observed that with adding crumb rubber to asphalt mixes containing SP, the indirect tensile strength resistance of both dry and saturated samples were increased significantly (compared with the ordinary SP mixes). TSR parameter of asphalt mixes containing 30% Sulfur-Polymer-15% Crumb Rubber; 40% Sulfur-Polymer-15% Crumb Rubber; and 50% Sulfur-Polymer-15% Crumb Rubber resulted in TSR increased values of 32, 28 and 24% respectively.

Similar results were achieved with adding Zycotherm to SP asphalt mixes, resulting in increased TSR values. The extent of TSR increase in mixes containing 30% Sulfur-Polymer-0.15% Zycotherm; 40% Sulfur-Polymer-0.15% Zycotherm; and 50% Sulfur-Polymer -0.15% Zycotherm were 17, 13 and 10%, respectively.

4. CONCLUSIONS

From the experimental works conducted on asphalt mixes containing Sulfur-Polymer and with using three different additives, the following conclusions can be drawn:

1- Although SP containing bituminous mixes have good mechanical properties, these lack moisture resistance, making them prone to stripping.

2- In order to overcome the stripping susceptibility of SPAM bituminous mixes, three additives, namely Nano Clay, Zycotherm and Crumb Rubber were tested. These, when applied at certain conditions, were considered to be effective in reducing moisture susceptibility of SPAM mixes.

3- All the above additives resulted to increase moisture resistance of SPAM mixes. However, the extent of increase varied according to the additive type and amount used in mixes.

4- Tensile Strength Ratio, determined in Indirect Tensile Test and Marshall Residual Stability Ratio, determined in Marshall Test were considered effective parameters in quantifying moisture susceptibility of SP mixes.

5- ITS testing results indicated that mixes in those the bitumen binder content consisted of 40% sulfur polymer and 15% crumb rubber resulted in 23% increase in TSR value. Marshall Retained Stability Results (MRSR) of mixes containing 40% sulfur polymer and 15% crumb rubber showed 17% increase in MRSR value.

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