



Effects of polymer additives on bituminous mixtures



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HIGHLIGHTS

- Constructability and volumetric characteristics are not affected by polymers.
- Polymers are analogous (or better) to modified bitumen, in terms of modulus.
- Polymers reduce temperature and frequency susceptibility of bituminous mixtures.
- Bituminous mixtures fatigue life is improved by using polymer additives.
- The addition of polymers improves the rutting resistance of bituminous mixtures.

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ABSTRACT

The laboratory investigation described in this paper focuses on the effects of polymers as additives for bituminous mixtures. Three dosages (3%, 6% and 9% by weight of bitumen) of EVA and LDPE polymers were used. The investigation was divided into two main phases. The first one focused on a comparison between mixtures to which polymers were added and a modified bitumen-bound mixture. The comparison was performed by observing constructability, volumetric and mechanical performances (e.g. dynamic moduli). The second phase aimed at evaluating the effects of such polymers on the performance of the corresponding mixtures at in service temperatures by means of simulative tests concerning stiffness master curves, fatigue life and rut resistance. In the second phase, the modified bitumen-bound was not investigated. Both phases also include investigations on a standard bitumen-bound mixture. Results of the first phase revealed that polymers guarantee the same, if not better, performances than modified bitumen on the corresponding bituminous mixtures. Results of the second phase showed that polymers increase the mixtures' performances at in-service temperatures. In particular, polymers reduce mixtures' Stiffness at low temperatures, increase fatigue life at intermediate temperatures and reduce rutting deformations at high temperatures.

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1. Introduction

In recent decades, the ever-increasing traffic volume on transportation infrastructures (e.g. roads and airports) has brought about the need to produce high quality materials for construction, rehabilitation and maintenance operations. In the field of flexible pavements, in which bituminous mixtures are the most important material in terms of their structural and functional performance, this requirement is currently satisfied through the use of polymer-modified bitumen as the binder [1–6]. In other words, bitumen modification is a technique in which a certain type and dosage of polymers is added to the binder in order to improve its

viscoelastic characteristics. Modified bitumen is produced prior to the manufacturing of the bituminous mixtures. It is important to note that, as they are unstable materials, modified bitumens require refineries with high quality equipment for their manufacture. Therefore, it is clear that in some parts of the world, such as in developing countries, it would be very difficult to produce and use such modified binders, even though, in these countries, infrastructures are those most subjected to traffic volume growth.

For all these reasons, another technology, based on the use of additives, has been recently developed. Unlike modifier agents, additives aim to obtain the required pavement performance, working not on the binder but on the bituminous mixtures. In fact, polymers can be added directly during the production of the bituminous mixture. As for previous studies on the topic, over the last decade works of other researchers have been devoted to highlight the effects of additives on porous wearing courses [7] or on dense grade bituminous mixtures by focusing on a restricted number of

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performances, such as dynamic moduli [8,9], rutting [10,11], fatigue [12].

Within the framework described above, a research study was carried out in the Road Research Laboratory of Politecnico di Milano, with the aim of understanding the effects of polymers on the performances currently required to dense grade bituminous mixtures. To this end, the goals of the investigation were:

- comparing polymers-added bituminous mixtures with a modified bitumen-bound mixture;
- evaluating the effects of polymers on the performances of the corresponding mixtures at in service temperatures.

According to the goals described above, the investigation was divided into two main phases.

During the first phase, the comparative investigation was performed by measuring constructability parameters (e.g. self-compaction and workability), volumetric characteristics (e.g. air voids, voids in the mineral aggregates, and voids filled with bitumen), and mechanical performance (e.g. dynamic moduli). The second phase included simulative mechanical tests concerning stiffness master curves in a temperature ranging from -5°C up to 30°C , fatigue life at 20°C , and the rut resistance at 60°C .

This paper provides an overview of the results obtained and describes some details of the specific protocols followed during the research project.

2. Experimentation

2.1. Key materials

The composition of asphalt mixtures was determined according to the current specifications for binder courses provided by the Italian Road and Highways Administration (ANAS). Therefore, a single particle-size distribution of aggregates (Fig. 1) and a single value of bitumen content (%B equal to 4.0% by weight of mixture) for all the mixtures were chosen.

The key materials used in the study were natural calcareous aggregates provided by a local contractor, a standard 70/100 penetration, unmodified bitumen [13], a 4% SBS modified bitumen (50/70–60 according to the European Specifications [13]) and a calcareous filler (filler/bitumen ratio equal to 1.1). In line with technical literature [14], preliminary viscosity tests were performed on the two bitumens and results showed a mixing/compaction temperature equal to $150/140^{\circ}\text{C}$ for the standard bitumen and $170/160^{\circ}\text{C}$ for the modified one, respectively.

During the experimentation, two polymers were utilized for comparison: the first one was made of amorphous polyolefin with a low molecular weight and a low fusion point, belonging to the family of EVA (ethyl-vinyl-acetate), herein named polymer A, while the second was mainly composed of LDPE (low density polyethylene), named polymer B. More specifically, polymer B is a compound made of LDPE, EVA and other polymers with a low molecular weight and a medium fusion point. Polymers are provided as small pellets, workable at room temperature, in order to be easily stored or added directly into the mixing chamber during the production of the bituminous mixture. Fig. 2 shows the flexible semi-soft granules at room temperature, while Table 1 reports the basic physical properties of both polymers.

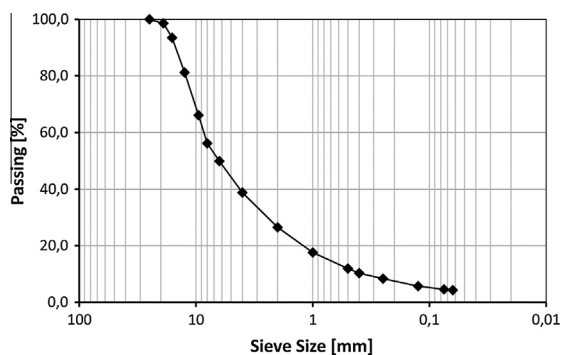


Fig. 1. Gradation of the aggregates.

2.2. Experimental program

As mentioned above, the investigation herein described consisted of two main phases.

Phase 1 involved eight bituminous mixtures prepared in the lab: a modified bitumen-bound mixture (named M-BM), a standard bituminous mixture made by using the 70/100 unmodified bitumen (named BM), three mixtures incorporating polymer A and three containing polymer B. Dosages were fixed at 3%, 6% and 9% by weight of 70/100 unmodified bitumen. In this paper, these mixtures are identified by an alphanumeric code formed by the acronym of the mixture (BM) followed by the polymer identification (A or B) and the dosage (3%, 6% or 9%); e.g., the code of a bituminous mixture containing 3% of polymer A will be BM-A-3%.

Mixing operations were performed using a lab mixer, obtaining 60 kg per time. Mixing procedures were performed and verified according to Authors' previous experience [15,16] in order to obtain a reasonable homogeneity of the mixtures.

Specimen compaction was carried out by using a Gyratory Shear Compactor (GSC), according to the protocol specifications defined within the Strategic Highway Research Program [17] (1.25° gyration angle, 30 rev min^{-1} gyration speed, 600 kPa vertical pressure, 150 mm mold diameter). A minimum of three cylindrical specimens for each mixture were compacted at 100 gyrations of GSC. During the GSC compaction, self-compaction C_1 and workability k parameters were measured. These parameters are currently used by pavement engineers to quantify the compaction properties and consequently to compare different mixtures, as proven by previous investigations [18,19]. Furthermore, all GSC compacted specimens were compared using both a volumetric characterization, including the analysis of voids (%v), voids in the mineral aggregate (VMA) and voids filled with bitumen (VFB), and the measure of the dynamic modulus at 20°C , according to the EN 12697-26 Annex C [20].

Based on the results obtained during Phase 1, which will be described in Section 3.1, Phase 2 aimed at evaluating the individual effects of each type and dosage of polymers on the corresponding mixtures, without considering the mixture bound with modified bitumen, investigating a total amount of seven mixtures. To this end, simulative mechanical tests were performed. Roller compactor slabs ($500 \times 260 \times 50\text{ mm}$) were prepared imposing a target air void equivalent to that obtained during the first phase. The equipment employed included a prismatic mold on which a series of metal plates was set. The compaction energy was transferred by means of two twin steel wheels moving horizontally over the plates (kneading compaction, EN 12697-33) [21]. The slabs were used both to measure rutting resistance (EN 12697-22 [22]) and to obtain beams (prismatic specimens) for four point bending tests (Stiffness according to EN 12697-26 Annex B [19] and fatigue resistance according to EN 12697-24 Annex D [23]).

The Stiffness of the mixtures was determined by performing four point bending tests (4-PBT) with a minimum of three prismatic specimens for each mixture. Each specimen was tested in a controlled-strain mode, imposing a strain level of 50 micro-strain at four different temperatures ($-5, 10, 20, 30^{\circ}\text{C}$) and seven frequency levels (0.5, 1, 2, 4, 6, 8, 10 Hz). The applied load was a symmetric sinusoid in order to impose equal deformations on the upper and lower faces of the prismatic specimen.

The values collected at all temperatures and different loading frequencies were used to obtain the master curves of the dynamic modulus by following the numerical method proposed by AASTHO [24], which applies temperature shift factors (a_T) based on the theory of Arrhenius.

In this regard, a nonlinear least squares regression was performed in order to minimize the summed square error (SSE) between the estimated values (taken from the model) and the experimental ones. The calibration of the model base was carried out using the Solver function of a self-made spreadsheet.

In order to quantify the fatigue resistance, the samples were tested under four-point bending (the same applied to determine dynamic modulus) in a strain-controlled mode at a strain level of 300 micro-strain, imposing a haversine wave at a frequency of 10 Hz. All tests were carried out at 20°C using a minimum of three specimens. Two failure criteria were established: the number of cycles after which a 50% reduction of the initial dynamic modulus occurs (recorded after one hundred cycles) or, if necessary, a maximum of 1.5 million cycles.

Wheel tracking tests were performed to determine the rutting resistance of mixes at 60°C . According to this method, a rubber wheel moves across the $500 \times 260 \times 50\text{ mm}$ slab at speed of 37 passes/min, applying a repetitive load (700 N) for 10,000 passes. The final Rut Depth (RD) in dry conditions and the WTS (Wheel Tracking Slope) were measured according to the European Standard EN 12697-22 part B, in air [23].

3. Results and discussion

3.1. Phase 1: comparative investigation

In Figs. 3–5 the average results related to constructability parameters, volumetric characteristics and mechanical performances are shown as a function of the type and dosage of polymers, including those of the two reference mixtures (BM and

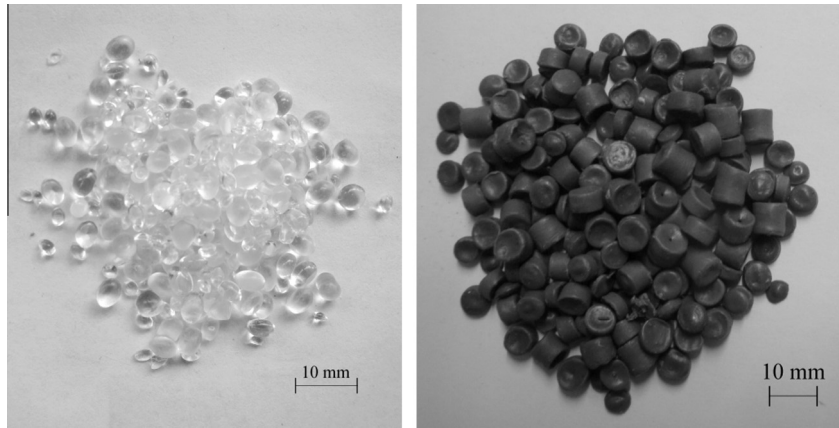


Fig. 2. Polymer pellets at room temperature: polymer A – EVA (left), polymer B – LDPE (right).

Table 1
Basic physical properties of polymers.

Polymer code	Main component	Aspect	Softening point (°C)	Fusion point (°C)	Melt index
A	EVA	Neutral granules	100	120	<10
B	LDPE	Black/gray granules	150	160	1–5

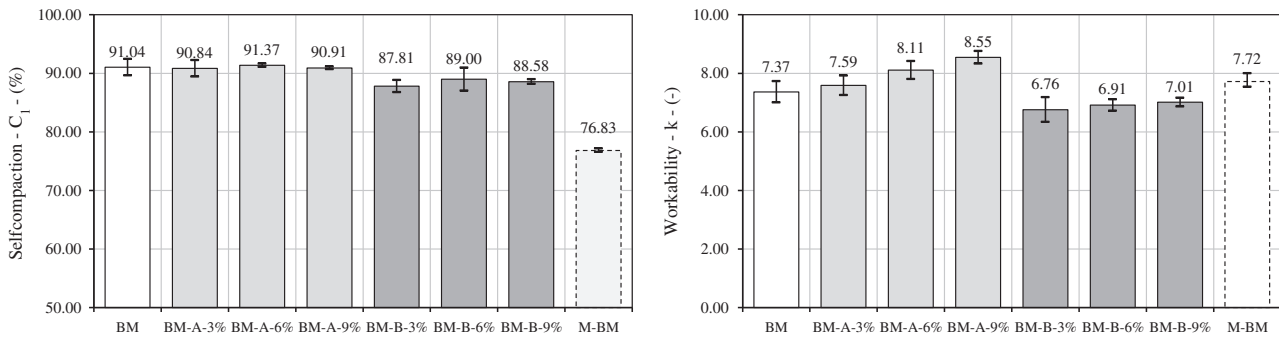


Fig. 3. Constructability parameters.

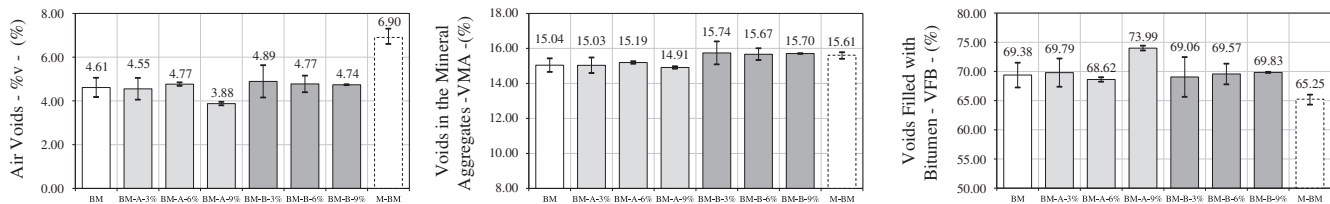


Fig. 4. Volumetric characteristics.

M-BM). Both average values and dispersion bars, displaying the maximum and minimum result of each mixture, are given in the bar charts.

As regards the constructability parameters C_1 and k (Fig. 3), the presence of polymer A does not significantly affect such parameters, although, by increasing the dosage, a slight increase in k can be noticed. However, it is possible to observe a slight decrease in both the investigated parameters for mixes with polymer B, regardless of the dosage of polymers. It probably depends on the fact that polymer B has a fusion point (160 °C as reported in Table 1) 10 °C higher than the mixing temperature (150 °C). Such difference in temperature did not allow the complete blending of the polymer into the mixture. Moreover, modified bitumen-bound mixture (M-BM) reveals a substantial drop in C_1 .

As regards the volumetric characteristics, the test results reported in Fig. 4 shows that polymers do not significantly affect either the air voids (between 4% and 5% for all the investigated mixtures) or the VMA (in the range of 14–16%). The same holds true for VFB, except for those mixes containing 9% of polymer A, which cause an increase in VFB. This is probably due to the fact that polymer retains the bitumen, thus increasing the quantity of binder present in the mixture. This proves to be consistent with the workability findings, in which the same mixture exhibits the maximum degree of k . Moreover, comparing these mixtures with the mixture bound using modified bitumen (M-BM), it is possible to observe that the latter is characterized by an increase in voids content and a decrease in VFB, a trend consistent with the constructability results, which show a reduction in the C_1 parameter.

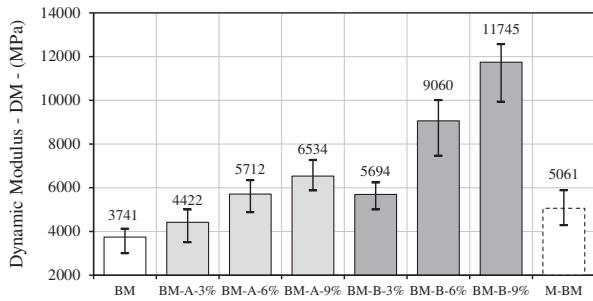


Fig. 5. Dynamic modulus.

In other words, modified bitumen appears to be detrimental for the mixture's constructability.

Mechanical results in Fig. 5 shows that dynamic moduli increase with an increase in the polymer dosage, and this is particularly evident when using high dosages (6% and 9%) of polymer B. Moreover, the mixture containing modified bitumen as binder (M-BM) exhibits the same performance as the mixtures with low dosages of polymer (3%). Based on the results in Fig. 5, it is possible to claim that polymers grant the same benefits as modified bitumen, or even better ones, when high dosages are used.

3.2. Phase 2: simulative mechanical tests

In Figs. 6–8, the average results related to mechanical performance (e.g. master curves of dynamic modulus, fatigue life, and rutting) are shown as a function of type and dosage of polymers. Both average values and dispersion bars, displaying the maximum and minimum result of each mixture, are given in the bar charts.

As far as the dynamic modulus is concerned, based on the master curves plotted in Fig. 6, there is a reasonable degree of correspondence between data collected from the tests and the predicted values of the model. This can be observed from the superimposed curves showing stiffness data points for all the mixtures.

Graphs in the figure indicate that polymers reduce the frequency sensitivity of the mixtures. In fact, due to the presence of polymers, the typical S-shape of the master curves is less accentuated. In particular, mixtures containing polymers show a decrease in the dynamic modulus at high frequencies, implying low temperature in the field, according to the time–temperature superposition principle. This would prove to be a positive characteristic during winter, as at low service temperatures in the field (or high frequencies), cracking phenomena can occur due to the excessive stiffness of the mixture.

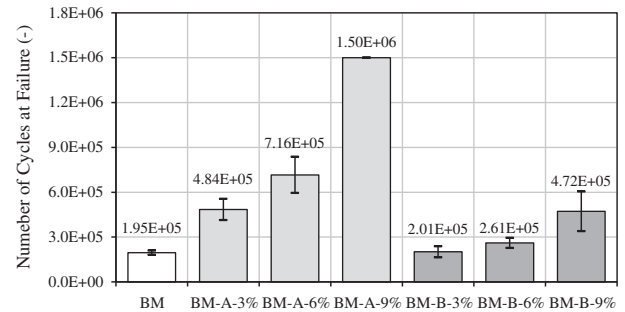
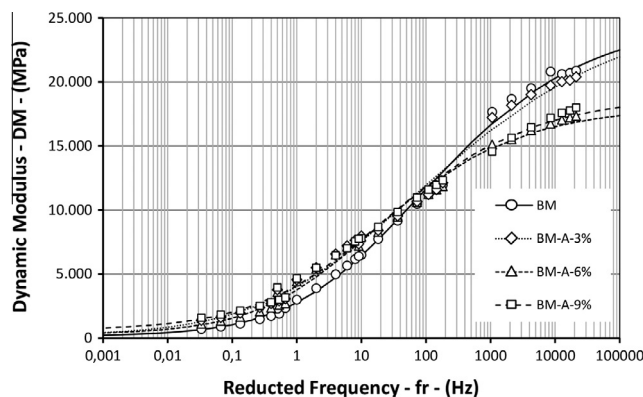


Fig. 7. Fatigue life – load cycles to failure at 20 °C.

However, the polymers investigated show some differences, particularly associated to the dosages.

It is notable that mixtures with lower dosages of polymer A (3%) show the same recorded performances as the reference mixture (BM, with no polymers). On the contrary, mixtures with high dosages of polymer A (6% and 9%) are characterized by a lower stiffness at high frequencies, which remains almost constant. As such, an increase in polymers over 6% does not lead to an increase in performance.

Mixtures containing polymer B demonstrate that the best performance in terms of stiffness is exhibited by the 6% dosage, while the 9% dosage appears to be detrimental to performance.

Results from fatigue tests (Fig. 7) indicate that polymers influence the mixtures' behavior, as demonstrated by the increase in fatigue resistance (represented by the number of load cycles to failure) with higher polymer dosages. However, the values reported show notable differences concerning fatigue life levels. In particular, at a given dosage, mixes with polymer A content exhibit better performances than mixes with polymer B. As such, one can hypothesize that the better fatigue performances are guaranteed by the mixture with 9% of polymer A, which displays a fatigue life greater than 1.5 million cycles, the maximum number of cycles set before halting the tests. As for mixtures with polymer B, results show that both lower (3%) and medium (6%) dosages enable a slight increase in fatigue life when compared to the reference mixture (BM, with no polymers). Furthermore, it is possible to note that the mixture with the maximum content of polymer B (9%) had a fatigue life similar to the mixture with 3% of polymer A.

As far as rutting resistance is concerned, graphs in Fig. 8 shows that polymers greatly improve the mixtures' performance, dependent, obviously, on the type and dosage. Regarding the type, it is possible to note that polymer A exhibits the best performance in terms of RD and WTS, which decrease as the dosage increases.

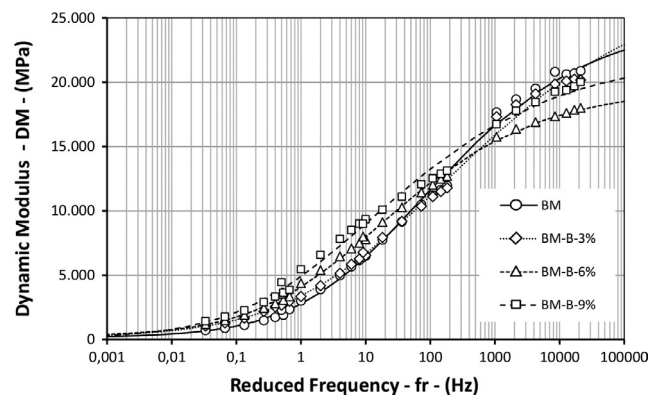


Fig. 6. Master curves of the dynamic modulus.

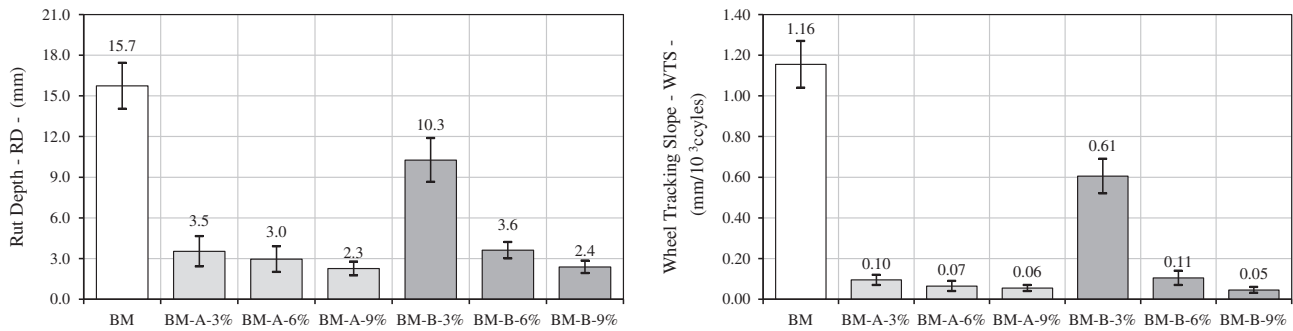


Fig. 8. Rut Depth (RD) and Wheel Tracking Slope (WTS) at 60 °C.

With regards to polymer B content, the corresponding mixtures show the same rutting behavior as mixtures containing polymer A at medium and maximum dosages (6% and 9%). Additionally, low dosages of polymer B (3%) exhibit the worst performance; even though these are better than those obtained by testing the reference mixture (BM, with no polymers).

4. Summary and conclusions

The comparative laboratory investigation described in this paper focused on the effects of commercially available polymers as an additive for bituminous mixtures used as binder courses. The analysis was divided into two main phases: the first one was dedicated to the comparison of mixtures to which polymers were added with standard and modified bitumen-bound mixtures; the second one aimed at evaluating the effects of different types and dosages of polymers on the corresponding mixtures by using simulative mechanical tests.

Based on the results reported in this paper, the following conclusions can be drawn:

- the addition of polymers does not significantly affect constructability parameters (k and C_1) or the volumetric characteristics (air voids, VMA and VFB) of the corresponding mixtures;
- polymers grant the same benefits as modified bitumen in terms of dynamic modulus; benefits increase when a high dosage is used;
- in terms of frequency sensitivity, polymers noticeably affect the dynamic modulus of the corresponding mixtures. Specifically, the master curves of mixtures containing polymers, especially those with medium and high dosages (e.g. 6% and 9%) show a reduction in stiffness at high frequencies (low temperatures); whereas, at low frequencies (high temperatures), their behavior is quite similar to the reference mixture (without polymers). Therefore, in winter, a reduction in the propensity of a mixture containing polymers for thermal cracking can reasonably be expected;
- fatigue life is generally improved by the presence of polymers, thus a high dosage (9%) of polymer A (EVA) grants the best performance;
- the addition of polymers clearly benefits a mixture's rutting resistance, with the exception of the mixture with 3% polymer B (LDPE) in which a slight reduction of RD and WTS can be observed compared to the reference mixture (without polymers). Generally, in summer, a reduction in rut deformation proneness in mixtures containing polymers is to be expected.

References

- [1] Bahia HU, Hanson DI, Zeng M. Characterization of modified asphalt binder in Superpave mix design. NCHRP Report 459. Washington, DC, USA: National Academy Press; 2001.
- [2] Fernandes MRS, Forte MMC, Leite LFM. Rheological evaluation of polymer-modified asphalt binders. *Mater Res* 2008;11:381–6.
- [3] Lu X, Isacsson U. Modification of road bitumens with thermoplastic polymers. *Polym Test* 2000;20:77–86.
- [4] Ameri M, Mansourian A, Sheikhmotevali AH. Laboratory evaluation of ethylene vinyl acetate modified bitumens and mixtures based upon performance related parameters. *Constr Build Mater* 2013;40:438–47.
- [5] Yildirim Y. Polymer modified asphalt binders. *Constr Build Mater* 2007;21:66–72.
- [6] Airey GD. Fundamental binder and practical mixture evaluation of polymer modified bituminous materials. *Int J Pavement Eng* 2007;5:137–51.
- [7] Hassan HF, Al-Oraini S, Taha R. Evaluation of open grade friction course mixtures containing cellulose fibers and styrene butadiene rubber polymer. *J Mater Civil Eng* 2005;17:416–22.
- [8] Sha AM, Zhou QH, Yang Q. Material composition design method for high modulus asphalt concrete. *J Chang'an Univ* 2009;3:1–5.
- [9] Zou XL, Sha AM, Jiang W, Huang XY. Characteristic analysis of dynamic modulus for high modulus asphalt concrete. *Appl Mech Mater* 2014;505–506:15–8.
- [10] Tayfur S, Ozen H, Akson A. Investigation of rutting performance of asphalt mixtures containing polymer modifiers. *Constr Build Mater* 2005;21:328–37.
- [11] Zhou QH, Sha AM. Permanent deformation of high modulus asphalt concrete pavements. *J Chang'an Univ* 2013;33:13–6.
- [12] Zhi S, Gun WW, Hui LX, Bo T. Evaluation of fatigue crack behavior in asphalt concrete pavements with different polymer modifiers. *Constr Build Mater* 2012;27:117–25.
- [13] EN 12591 Bitumen and bituminous binders – SPECIFICATIONS for paving grade bitumens. European Committee for Standardization; 2009.
- [14] The Asphalt Handbook. MS-4. 7th ed., Lexington, KY, USA: Asphalt Institute; 2007.
- [15] Crispino M, Mariani E, Toraldo E. Assessment of fiber-reinforced bituminous mixtures' compaction temperatures through mastics viscosity tests. *Constr Build Mater* 2013;38:1031–9.
- [16] Toraldo E, Brovelli C, Mariani E. Laboratory investigation into the effects of working temperatures on wax-based warm mix asphalt. *Constr Build Mater* 2013;44:774–80.
- [17] The Superpave mix design system manual of specifications, test methods, and practices. Strategic Highway Research Program SHRP A-379. Washington, DC, USA: National Research Council; 1994.
- [18] Bassani M, Santagata E. Construction: roofing, waterproofing and bituminous materials. In Proc. 3rd international conference on bituminous mixtures and pavements. Thessaloniki, Greece; 2002.
- [19] Cominsky R, Leahy RB, Harrigan ET. Level one mix design: material selection, compaction, and conditioning. Strategic Highway Research Program SHRP-A-408 Report. Washington, DC, USA: National Research Council; 1994.
- [20] EN 12697-26 Bituminous mixtures – test methods for hot mix asphalt – Part 26: Stiffness. European Committee for Standardization; 2004.
- [21] EN 12697-33 Bituminous mixtures – test methods for hot mix asphalt – Part 33: Specimen prepared by roller compactor. European Committee for Standardization; 2007.
- [22] EN 12697-22 Bituminous mixtures – test methods for hot mix asphalt – Part 22: Wheel tracking. European Committee for Standardization; 2007.
- [23] EN 12697-24 Bituminous mixtures – test methods for hot mix asphalt – Part 24: resistance to fatigue. European Committee for Standardization; 2005.
- [24] AASHTO TP-62. Standard method of test for determining dynamic modulus of Hot-Mix Asphalt (HMA). AASHTO – American Association of State Highway and Transportation Officials; 2005.