LONG-LASTING OVERLAYS BY USE OF HIGHLY MODIFIED BITUMINOUS MIXTURES

Delphine Simard\textsuperscript{1}, Francois Olard\textsuperscript{2}, Alain Perrot\textsuperscript{1}, Patrick Huon\textsuperscript{1}, Jérôme Dherbecourt\textsuperscript{3}
\textsuperscript{1}EIFFAGE Travaux Publics, Central Laboratory, Corbas, FRANCE
\textsuperscript{2}EIFFAGE Travaux Publics, Research & Development Department, Corbas, FRANCE
\textsuperscript{3}EIFFAGE Travaux Publics Rhône-Alpes Auvergne, Corbas, FRANCE

ABSTRACT

Polymer modified asphalt mixtures have usually been used in wearing courses in order to improve both crack growth resistance and rutting performance where temperatures, vertical stresses and shear strain levels are more severe. Nonetheless, the need for either thinner yet high-performing wearing courses or ever-increasing durability provides the motivation for using higher polymer contents in the wearing course of bituminous pavements. Therefore, instead of the conventional use of 2-3\% styrene-butadiene-styrene (SBS) in polymer modified binders (PMB's), the resort to higher SBS contents in the range 6-7\% allows for a phase inversion in the PMB microstructure: the swollen polymer becomes the continuous phase in which asphaltene nodules are dispersed. This significant change in PMB microstructure brings about significantly higher performances. Besides, some other benefits may be related to the possible layer thickness reduction: less natural materials (aggregate, bitumen) used, less resources required for construction (man-hours, emissions during transport and laying) and, overall, cost saving.

Microstructure, both empirical and rheological characteristics were investigated in laboratory for two different PMB's with very high SBS content. In addition, in-situ testing was carried out: a brief follow-up of the highly trafficked Millau Viaduct surfacing (constructed in France in 2004) where this type of PMB was used, is proposed.

The paper illustrates that the proposed innovative highly modified bituminous mixes may be from now on potentially considered as a relevant solution for sustainable long-life and high-performance overlays, needing only rare surface maintenance.

Keywords: PMB, SBS, high SBS content, high-performance asphalts, perpetual pavements
1. INTRODUCTION

1.1. Technical background to highly polymer modified binders (HPMB’s)

The development of highly polymer modified bitumens (HPMB’s) and the use of additives (such as thermoplastics) is very much linked with the development of new mix designs for bituminous mixtures for thin surfacings, which provide improved practical qualities and durability. Indeed, phase inversion with polymer continuous phase is the main goal of HPMB use. Fluorescence microscopy is the most frequently used technique to assess the state of dispersion of the polymer and bitumen phases (Brion and al) [1]. It is based on the principle that the polymers, swollen by some of the constituents of the bitumen to which they have been added, fluoresce in ultraviolet light. They emit yellow-green light while the bitumen phase remains black (Boudlin and al) [2] (Figure 1).

Thermoplastics triblock copolymers such as Styrene Butadiene Styrene (SBS) have a styrenic end-block (PS) and a rubbery midblock (PB). In bitumen blends, the PS end-blocks form spherical micelles acting as physical cross-links while the PB part is swollen by the maltene portion of asphalt (saturates, aromatics and resins) (Adedeji and al) [3] (Kraus and al)[4]. This physical network can be enhanced by adding a cross-linker (based on sulphur) thus creating a chemical network between the PB parts. Usually, phase inversion occurs when adding around 5% of SBS, but this rate depends on the base bitumen (grade and nature) and whether the binder is cross-linked or not (Planche and al) [5]. Hence, as SBS swollen in the maltene portion of the bitumen, in the past, HPMB’s were mainly developed using high grade base bitumen.

![Figure 1 : Microstructure of bitumen / polymer blends: a) Continuous bitumen phase b) Bitumen with inter-twisted phases c) Continuous polymer phase (AIPCR) [6]](image)

1.2. Technical & environmental challenges

Polymer Modified Binders (PMB’s) started being used extensively about thirty years ago in the quest to improve the mechanical performance of bituminous pavements particularly on the wearing course, such as better resistance to rutting and reduction in reflective cracking. This development came in response to traffic increase, to reduce maintenance periods, which are a major source of costly traffic disturbance. In this new area of sustainable development where materials enabling durability enhancement are being demanded, highly polymer modified binders (HPMB’s) are being developed. Moreover, the ever-increasing production of porous asphalts and open-graded asphalts revolves around an increase in the use of PMB’s.

The principal cause of bituminous binder ageing in service is commonly known to be the oxidation by the oxygen from the air of certain molecules resulting in the formation of highly polar and strongly interacting oxygen containing functional groups. The chemistry of asphalt oxidation reactions is based on a dual sequential hydrocarbon oxidation mechanism (Petersen) [7] leading to the formation of both ketones and sulfoxides which are the major oxidation products. In the latter stages of oxidation in highly oxidized asphalts, dicarboxylic anhydrides and carboxylic acids are being formed in a smaller quantity. However, during road service life, (i) standard binder characterizations showed that plain asphalt is much more affected by ageing than the PMB and (ii) cross-linked PMB features a significantly lower oxidation degree as measured by Fourier Transformed InfraRed Spectroscopy IRTF (Dressen and al) [8]. This can be explained by a very homogenous polymer distribution in the binder matrix (Mouillet and al) [9].

Traffic and weather conditions affect binder ageing. Nevertheless, the great chemical stability of the SBS polymer is responsible for its reliable long-term performances and great durability (Gallet and al.) [10]. Indeed, Gel Permeation Chromatography GPC linked to IRTF studied over 20 years shows that only the really thin surface (~15mm) is affected whereas the inferior layer remains intact giving great durability to the PMB over years [10].

In an environmental context, what is mainly being sought is the reduction of the emissions of polycyclic aromatic hydrocarbons (PAHs) and semi-volatile organic compounds (SVOCs) during the manufacturing stage. As mentioned previously in the technical background, the SBS swollen in the maltene part of asphalt leads to a two-phase composed PMB, a first one infiltrated by bitumen maltenes and a second one containing all the bitumen components which have not been absorbed by polymer. As a result, the greater the SBS content, the more emissions can be limited due to volatile compounds from binder trapping by polymer (Gaudefroy and al) [11].

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2. EXPERIMENTAL LABORATORY STUDY

Three highly polymer modified bitumens (HPMB’s) with 6% of SBS triblock copolymer and using 3 different base bitumens (hard, medium and soft) were studied at EIFFAGE Travaux Publics central laboratory in Corbas (France). All of them are cross-linked. Their respective compositions are proprietary. These three HPMB's are referred to as PMB.A, PMB.B and PMB.C.

2.1. Empirical tests

Properties of HPMB’s (Table 1) were obtained from empirical tests and in accordance with European norms: penetration grade (NF EN 1426) [12], ring and ball softening point (NF EN 1427) [13], elastic recovery (NF EN 13398) [14], Brookfield viscosity (MOPL 102) [15], storage stability (NF EN 13399) [16] and Fraass breaking point (NF EN 12593) [17].

Table 1: Empirical tests on HPMBs: PMB.A hard bitumen, PMB.B medium bitumen and PMB.C soft bitumen

<table>
<thead>
<tr>
<th>Properties</th>
<th>PMB.A</th>
<th>PMB.B</th>
<th>PMB.C</th>
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<tbody>
<tr>
<td>Penetration (dmm)</td>
<td>30</td>
<td>39</td>
<td>62</td>
</tr>
<tr>
<td>Ring &amp; ball softening point (°)</td>
<td>88</td>
<td>86</td>
<td>92.5</td>
</tr>
<tr>
<td>Elastic recovery (%)</td>
<td>88</td>
<td>90</td>
<td>97</td>
</tr>
<tr>
<td>Viscosity at 160°C (Po)</td>
<td>5.8</td>
<td>4.5</td>
<td>5.51</td>
</tr>
<tr>
<td>Storage stability test (%)</td>
<td>91</td>
<td>100</td>
<td>99</td>
</tr>
<tr>
<td>Fraass breaking point (°C)</td>
<td>-13</td>
<td>-13</td>
<td>-20</td>
</tr>
</tbody>
</table>

Adding 6% of SBS to a hard bitumen grade is more than usual; penetration, R&B softening point and elastic recovery are acceptable; however viscosity is high and storage stability is within the limits. When working with softer bitumen with an even higher level of polymer, viscosity is more acceptable and storage stability remains unaffected. Indeed, when the maltene content increases (= when the grade of bitumen became softer) polymer is more swollen. This leads to better properties such as outstanding elastic recovery and Fraass breaking point with very soft bitumen.

2.2. Microscopy on binders

Fluorescence measurements of HPMB's were carried out at EIFFAGE Travaux Publics laboratory using a Zeiss Axioskop microscope (Figure 2 - a). The microscope was equipped with a specific filter set (Figure 2 – b) composed of a blue violet excitation filter (395-440nm), a beam splitter (FT 460nm) and an emission filter (LP 470nm). The light source is based on mercury and emits in all wavelengths from 350 to 650nm. The principle of fluorescence is based on electronic transitions between an excited singular state and a fundamental state; according to the Jablonski diagram (Figure 2 – c).

Figure 2: a) Microscope equipped with three growth views (x10, x20, x50) b) Schematic diagram of a fluorescence filter cube c) Jablonski diagram with S0 fundamental state; S1 excited state and 3 fluorescence.

With the fluorescence microscopy technique and using this specific filter set, the asphalt rich phase appears dark, while the polymer-rich phase appears light. The three blends made in the laboratory were studied using the microscope and the differences in the results depending on the bitumen grade are presented here below in Figure 3.
Figure 3: Microstructure of bitumen / polymer blends

At a constant polymer rate of 6%, when the maltene content increases, the microstructure changes. With hard bitumen the continuous phase seems to be bitumen (case a). Indeed, there is a black continuous phase and the high level of polymer appears very shiny. When increasing the oil content and working with softer bitumen, the inter-twisted phase can be seen (case b); yellow and black parts are slightly merged. However, with very soft bitumen and high oil content, the continuous phase clearly becomes the polymer (case c) as black spots are defined among a large yellow continuous phase. This microstructure evolution highlights the role played by the maltene in the swollen polymer.

2.3. Rheological tests on binders

Complex modulus tests were performed at EIFFAGE Travaux Publics laboratory using a Dynamic Shear Rheometer (DSR Physica 501 Anton Paar) over a frequency range from 1 to 100Hz and a temperature range from -30°C to 70°C. Thus, complex modulus and master curves were obtained on modified bitumen and are presented below (Figure 4).

Figure 4: Rheological results a) Black diagram; b) Master curves at 15°C

The complex modulus of a PMB is mainly influenced by the modulus of the polymer in the high-temperature and low frequency domain; as used in this case, the thermoplastic SBS has a phase angle near 0°. These three HPMB’s display a similar level of SBS, but different asymptotic elastic behaviour illustrating the influence of the maltene content on the SBS. All of the three curves are quite similar however, with a softer base binder, the phase angle is lower. This can be explained by the fact that in softer bitumen, polymers are more swollen and thus give better elasticity to the binder.

3. DEVELOPMENT AND LARGE SCALE ROAD WORKS USING HPMB’S

Polymer modified asphalt mixtures are usually used in wearing courses in order to improve both crack growth resistance and rutting performance where temperatures, vertical stresses and shear strain levels are more severe. Nonetheless, the need for either thinner yet high-performing wearing courses or ever-increasing durability provides the motivation for using higher polymer content in bituminous pavements.

The previous laboratory results presented in section 2 were found to be very encouraging and led to many large scale road works using HPMB’s. The two following sub-sections 3-1 and 3-2 present two case studies:

- section 3.1 presents the case study of the Millau viaduct surfacing with a soft bitumen modified with more than 7% of cross-linked SBS (proprietary composition);
- section 3.2 deals with the case study of a high volume traffic bus lane in Lyon city center for which a medium pen grade bitumen was modified with more than 5% of cross-linked SBS (proprietary composition).

3.1. Design of a specific bituminous surfacing for orthotropic steel bridge decks: the Millau Viaduct

Both the geometry of the structure and the very high flexibility of metallic plates make the strains and stresses very severe in steel bridge surfacings (Figure 5). In particular, the repeated loading makes the fatigue phenomena an important parameter for the design of such bituminous wearing courses. In addition, these specific surfacings must also have durability over the expected temperature range: it must be resistant to thermal cracking at low temperature and to rutting at high temperature. The technical studies led in parallel to the construction of the Millau Viaduct (France) –the
The highest bridge in the world—have provided in particular the opportunity for new developments in appropriate laboratory testing equipment and in original highly polymer modified surfacing [18][19][20]. Indeed, EIFFAGE Travaux Publics have pulled out the stops and led a comprehensive research program including a large laboratory testing campaign.

**Figure 5:** a) Example of deformed bridge deck (250x) with transversal stress (Huurman et al. 2003) [21]; b) Bituminous mix on orthotropic plate (Méhue 1981) [22].

These studies led to an HPMB called Orthoprène® being applied successfully on the bridge in December 2004 (Figure 6).

**Figure 6:** Laying BBSG 0/10 Surfacing with Arvieu aggregate and the HPMB, Orthoprène®

Microstructure tests (as explained previously in section 2.2.) were hence performed on Orthoprène® (Figure 7 – a) as well as on the coated aggregate (Figure 7 – b). Photographs were also taken in 2 dimensions for the aggregate (explaining the blurring in the photographs).

**Figure 7:** Microstructure observations a) Soft bitumen modified with more than 7% cross-linked SBS (Orthoprène®) b) Coated Arvieu aggregate with this HPMB.

The microstructure of Orthoprène®, consisting of soft bitumen modified with more than 7% cross-linked SBS, highlights a polymer continuous phase. Indeed, black spots representing the asphaltene, are surrounded by a homogeneous yellow phase representing the SBS swollen in the maltene portion of the bitumen. This particular microstructure gives outstanding properties (resistance to rutting and fatigue tests, etc.) to the asphalt concrete. Furthermore, photographs taken of the coated aggregate show that the modified binder is homogeneously spread on the surface of the aggregate.

The vertical displacement field of the Millau Viaduct structure under truck lead is illustrated in Figure 8. According to Pouget et al. [23][24], maximum strain magnitudes may reach $500 \times 10^{-6}$ m/m, obviously depending upon the temperature and loading rate.
Furthermore, the high fatigue resistance of this highly SBS modified surfacing is illustrated in Figure 9. The two-point bending fatigue test was carried out in accordance with the NFEN12657-24 standard at 10°C-25Hz. For strain amplitudes below 350.10^{-6}m/m, there is no failure observed up to 10 million cycles (a plateau is observed), which indicates that 350.10^{-6}m/m is the so called "endurance limit", an outstanding value for such bituminous material.

Figure 9: Two-point bending fatigue test results obtained for the Millau Viaduct surfacing.

3.2. Development of a new HPMB for Lyon city centre – France 2011

Making roads in a city centre such as Lyon is challenging as the night traffic cannot be stopped; buses and trolleys are still running until one o’clock in the morning. Therefore, in order to achieve a sustainable wearing course (less oxidation of the really thin surface and thus less maintenance periods) that is resistant to high-traffic roads such as the bus lanes in Lyon city centre, a medium pen grade bitumen modified with 5% of cross-linked SBS was developed. In this street, 2000m² (or 280t) of a BBSG 0/10mm with granitic aggregates (Lafarge La Patte at St Laurent de Chamousset France) were laid with a thickness of 5cm. This asphalt concrete was prepared with 5.1% of the innovative HPMB described previously.

After planing the previous surface course, asphalt concrete was laid on the paving slab just after spraying on a traditional tack coat. The asphalt concrete (Figure 10 – a) was laid using traditional compaction equipment. The asphalt concrete displayed good behaviour during laying and compaction. Surface texture of the asphalt concrete complied with our tests.

Thus, this experiment was successfully completed and traffic flow (car, buses and trolleys) resumed just after the first lane was finished in order not to cause disruption.

After one month of traffic use, the asphalt concrete behaviour is reasonable (Figure 10-b) but a much more accurate follow-up of this work site is planned over the next few years. Indeed, resistance to rutting will be studied and observations made over months and years to confirm the great durability of this new HPMB put to the test by very damaging traffic use, particularly by very slow vehicles such as buses.
As mentioned above, the HPMB (Figure 11 – a) used for this experiment was studied using microscopy. With regard to the aggregates, photographs were also taken in 2 dimensions (Figure 11 – b).

The microstructure of the binder used for the experiment in Lyon city centre using medium pen grade bitumen modified with 5% of cross-linked SBS, highlights a polymer continuous phase. Indeed, black spots representing the asphaltenes are surrounding by a homogeneous yellow phase representing the SBS swollen in the maltene portion of the bitumen. This particularly microstructure gives outstanding properties to the asphalt concrete. Furthermore, photographs taken of the aggregate show that the modified binder is homogeneously spread on the surface of the aggregate.

4. CONCLUSIONS & PERSPECTIVES

Nowadays, the need for either thinner yet high-performing wearing courses or ever-increasing durability provides the motivation for using higher polymer content in the wearing course of bituminous pavements. Moreover, it has been proven that working with HPMB helps to reduce environmental impact and also has longer term durability than normal PMB with less oxidation and less ageing of the binder. As only the really thin surface of the road is the most affected by ageing, using HPMB is very beneficial.

HPMB's developed at Eiffage Travaux Publics (Biprene® and orthoprene®) give interesting results in two particularly outstanding cases studies: the Millau Viaduct completed in 2004 and Lyon city centre in 2011. In the first case, studies were made with soft bitumen with more than 7% of polymer in order to have good fatigue resistance. New HPMB developed in 2011 with hard bitumen and more than 5% of polymer, gives both rigidity and good fatigue test resistance in the same binder; this is very encouraging. In both cases, the inversion phase is obtained and highlights a polymer continuous phase. Moreover, as hard bitumens are cheaper than soft bitumens, HPMB in hard bitumen is not only technically outstanding but also economically advantageous.

To meet new demands, HPMB are still being improved in Eiffage Travaux Publics laboratories, in order to achieve better performing and longer lasting binders.

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