THE CONTRIBUTION OF CROSS-LINKED POLYMER MODIFIED BINDERS TO ASPHALT PERFORMANCE

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ABSTRACT

The paper presents the results of a laboratory investigation on the mechanical performance of an asphalt mix modified with cross-linked elastomer modified binders at various polymer contents. The most comprehensive study has been conducted on a typical high modulus bituminous concrete used for structural reinforcement and based on 20/30 unmodified and modified paving grade bitumen. The asphalt has been tested for stiffness modulus, fatigue, permanent deformation and low temperature fracture temperature (TSRST test). Similar measurements, however restricted to permanent deformation and low temperature performance, have been performed with the homologous range of binders based on a 35/50 bitumen grade. In parallel, the binders have been tested for a number of properties likely to be linked to the asphalt behaviour. Those properties included both conventional (such as Penetration, Softening Point, Fraass breaking point) and rheological properties (dynamic viscosity, complex modulus, Bending Beam Rheometer). The final part of the communication discusses the correlation of the observed asphalt properties to these candidate binder performance indicators. This leads to some general considerations on how to select and identify the "best performing binder", considerations which are however only applicable to the specific "family" of cross-linked binders which have been evaluated.

Keywords: Low-Temperature, Mechanical Properties, Performance Testing, Polymer Modified Bitumen, Rheology

1. INTRODUCTION

The development of performance based specifications is still an on-going process for the binder and paving industry. It is of major concern for polymer modified binders but, so far, no satisfactory answer has been given. Many test methods and performance indicators have been proposed and debated but there is still a lack of validation with regard to asphalt characteristics as measured in the laboratory and, even more so, with regard to field performance. The study presented herewith adds some further results and ideas to this debate. It has been devoted to a specific type of elastomer modified binders, i.e. in-situ cross-linked binders. The first objective of the study was to evaluate the incidence of the level of modification (polymer content) on the mechanical performance of a typical asphalt mix. These results were then to be used as a basis for identifying relevant “performance indicators” among a panel of possible ad-hoc binder characteristics.

2. EXPERIMENTAL

2.1 Materials

The selected asphalt mix formulation is a typical continuously graded 0/10 asphalt concrete likely to be used for binder as well as wearing courses (AC-10 as per the EN 13108-1 standard). It has been formulated at a relatively high binder content of 5.7% which is usual for asphalt mixes to be used in high modulus asphalt concrete in France (designated as BBME). The aggregates and sand are a fully broken quartzite with an addition of 4% of limestone filler.

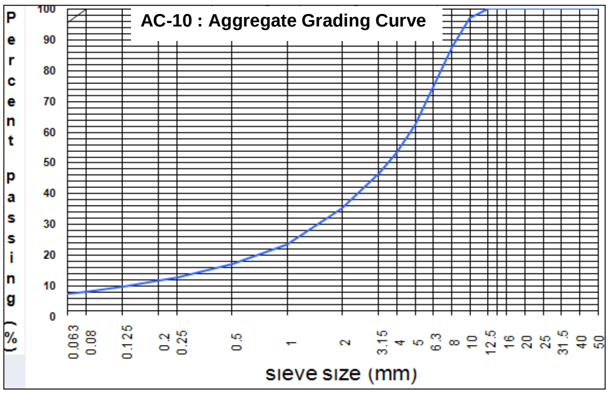


Figure 1 : AC-10 Aggregate grading curve

The investigated binders are obtained through an in-situ cross-linking process of an elastomer polymer. Those binders are well known for their homogeneous polymer distribution, conferring the binder a nearly mono-phasic behaviour with, in particular, an excellent storage stability. We have basically worked with two grades (20/30 and 35/50) of straight-run paving bitumen and three different levels of modification. Maximum amount of polymer was around 5% and two lower levels of modification have been contemplated. These polymer contents will be further referenced to, from lowest to highest, as x%, xx% and xxx%. All these binders correspond to commercially available products. The bitumens used were of the same origin (A) but two additional types (B and C) have also been considered for the 35/50 grade when studying low temperature performance.

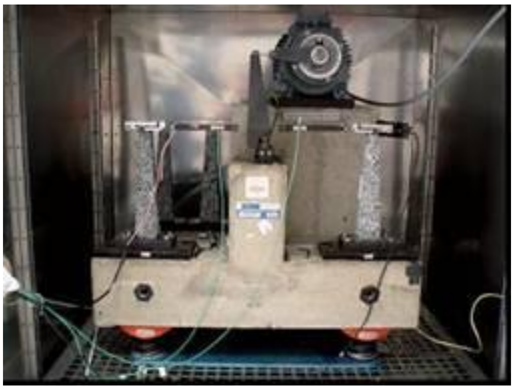
Picture 1 : Polymer network in a cross-linked elastomer modified bitumen

2.1 Measured asphalt properties

The mechanical properties measured on the asphalt concrete are those usually applied in France for performance assessment.

- Stiffness measurement at 15°C and 0.02s loading time under a direct tension mode on cylindrical specimens (DT-CY, EN 12697-26, Annex E). The test sample (Φ = 80mm, H = 200mm) is cored out of a rolling wheel compacted slab.
- Permanent deformation at 60°C with the French (LCPC) rutting tester according to the EN 12697-22. The retained performance indicator is the rut depth, expressed as a percentage of slab thickness (10cm) after 30 000 load cycles.
- Fatigue at 10°C-25Hz on trapezoidal specimen, in a 2-point bending, controlled strain, loading mode according to EN 12697-24. The retained performance indicator is the ε_6 value, i.e. the strain level (µstrain) which leads to conventional failure after 10^6 loading cycles.
- Low temperature fracture test (TSRST – Thermal Stress Restrained Specimen Test). In this procedure, a cylindrical sample (Φ = 57mm, H = 250mm) is maintained at a constant length whereas temperature is dropped

at a constant rate of 10°C per hour. This leads to a constantly increasing stress in the sample, until failure occurs. The retained performance indicator is the fracture temperature. Test results are given as the average of at least three samples, a test result being rejected if it differs by more than 2.5 °C from one of the others.



Picture 2 :Trapezoïdal 2-point bending fatigue test

Picture 3 : Sample used fro TSRST test

2.2 Measured binder properties

The binders have been tested for conventional properties such as Penetration (EN 1426), Softening Point (EN 1426), Fraass brittleness temperature (EN 12593) and Dynamic viscosity in the range 120°C–180°C (EN 13302).

Visco-elastic behaviour has been assessed through the measurement of complex stiffness modulus (norm |G*| and phase angle δ) over a wide range of temperatures and frequencies with an AR-2000 Dynamic Shear Rheometer (EN 14770). A particular emphasis has been placed on low temperature by determining the performance indicators usually derived from measurements with the Bending Beam Rheometer (EN 14771), i.e. the temperature corresponding to a stiffness modulus (S) of 300 MPa at 60s loading time ($T_{S=300\ MPa}$) and the temperature corresponding to an m value of 0.3 ($T_{m=0.3}$) at again 60 s loading time. The m-value is the slope of the tangent to the log S versus log(loading time) curve and reflects the ability of the binder to relax stress under an applied load. These indicators have been determined on original binders, RTFOT-163°C aged binders (EN 12607-1) and RTFOT + PAV (EN 14769) aged binders.

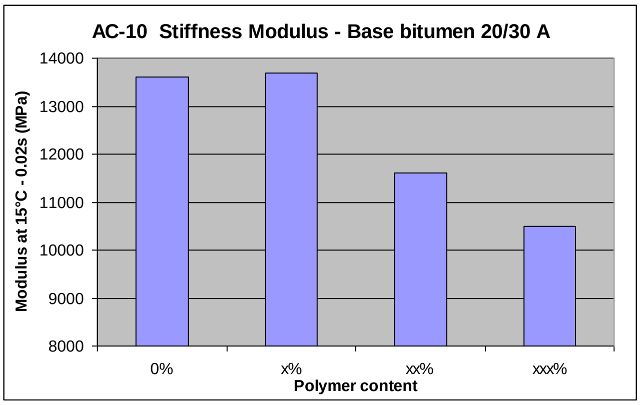
Picture 4 : AR-2000 rheometer in operation

2. PERFORMANCE OF CROSS-LINKED BINDERS IN HIGH MODULUS ASPHALT CONCRETE

In France, the most stringent performance requirements for a 0/10 high modulus asphalt concrete (BBME), which correspond to a “Class 3” material, are as follows :

Stiffness modulus at 15°C-0.02s	≥	11 000 MPa
ε_6 value (fatigue) at 10°C-25 Hz	≥	100 µstrain
Rut depth after 30 000 cycles at 60°C	≤	5 %

As it can be seen on Figure 2 the stiffness modulus is not affected at low polymer content (x%) but does then constantly decrease with increasing polymer content. The fatigue performance is very significantly improved (well above the specified minimum value), even at the lowest level of modification. A bit surprisingly, the ε_6 value stays however the same whatever the polymer content (Figure 3).



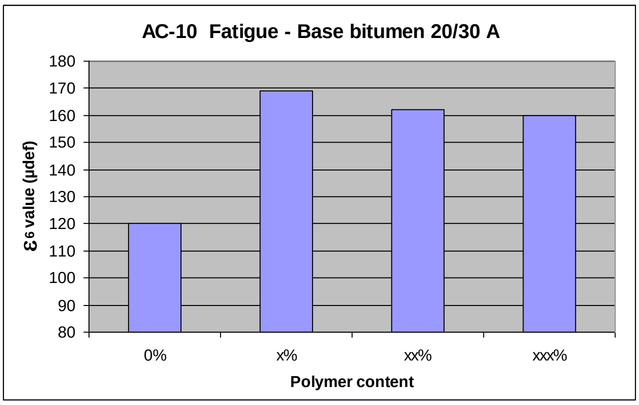


Figure 2 : AC-10-20/30 Stiffness Modulus

Figure 3 : AC-10-20/30 Fatigue

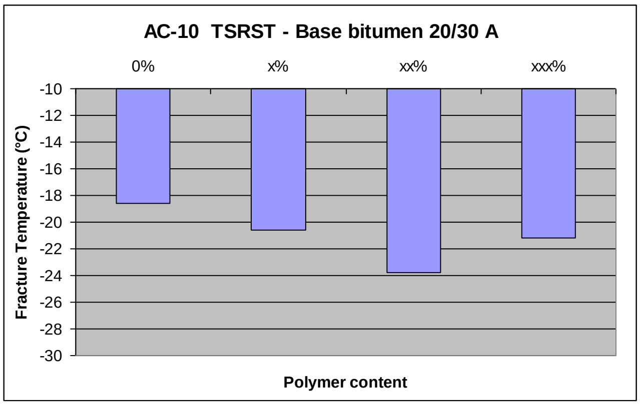
Structural pavement design is based on stiffness modulus and fatigue data. From this point of view, one would certainly privilege the lowest polymer content since it is sufficient to significantly boost the ε_6 without impairing the high stiffness of the mix. Going to higher polymer content would rather be counter-productive since it leads to a drop in stiffness and thus structural ability. This disadvantage must however be put in perspective since it will probably be more than compensated by the above mentioned gain for the ε_6 value. Indeed, in pavement design, it is generally admitted that the tensile strain (ε) resulting from the application of a traffic load and the stiffness modulus (E) of the corresponding pavement layer are related by following equation :

$$\varepsilon.\ E^{1/2} = \text{cte}$$

A drop in stiffness from 13 600 MPa to 10 500 MPa (extreme case in Figure 2) would thus entail an increase in tensile strain of about 14% whereas the gain in the ε_6 value amounts to at least 25%.

To benefit from the other potential advantages traditionally expected from polymer modification, it is thus possible to increase the amount of polymer while still enhancing the structural reinforcement ability. Those other advantages are for instance an improvement of low temperature characteristics (fears connected to the use of “hard” bitumen) as well as an improvement of resistance to rutting in the case of extreme traffic and climate conditions.

In that respect, the TSRST data show an improved behaviour for all modified mixes. This improvement, which stays quite limited at the lowest polymer content, goes through an optimum for the “medium level” of modification. Resistance to permanent deformation, which is already satisfying the specification for the unmodified bitumen, is impacted by the polymer in the expected way (continuous decrease of rut depth with increasing amounts of polymer).



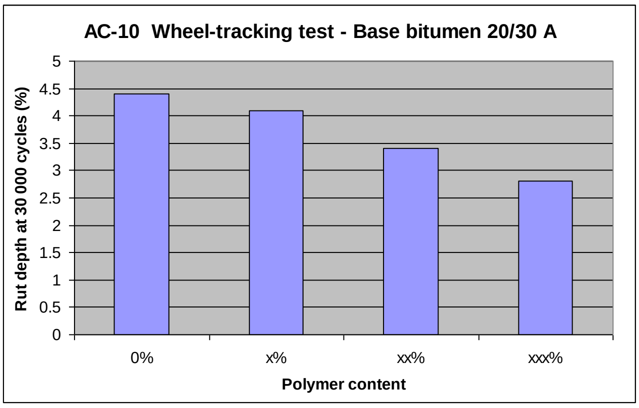
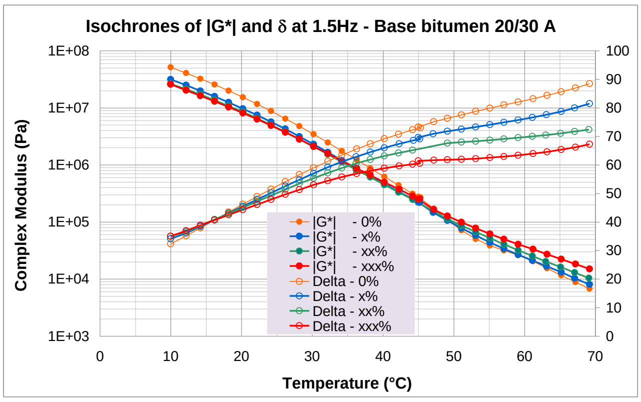


Figure 4 : AC-10-20/30 Fracture temperature

Figure 5 : AC-10-20/30 Wheel-tracking test

The behaviour in the wheel-tracking test is to be compared to the evolution of the stiffness modulus of the binder (|G*|) and of its visco-elastic character (as measured by the phase angle). At high service temperatures, as it can be seen on Figure 6, |G*| increases with increasing polymer content whereas δ continuously decreases with a more and more pronounced “levelling-off” effect. It is also important to notice (Figure 7) that these evolutions get even more pronounced when the frequency of loading decreases. The improved performance seen in the wheel-tracking test (which works at a frequency of 1 Hz) should therefore be further enhanced under slow motion or stationary traffic conditions.



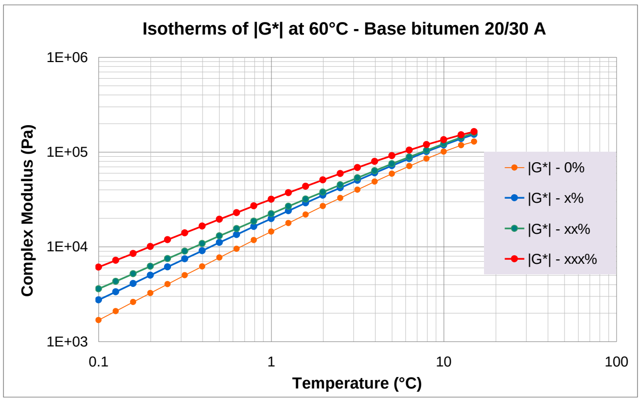


Figure 6 : Complex Modulus - Isochrones at 1.5 Hz

Figure 7 : Complex Modulus – Isotherms at 60°C

At this stage, it is however to be reminded that, especially in association with hard bitumen, an increase in polymer content will also lead to higher and higher viscosities. This is of high concern from a practical point of view since a high binder viscosity will require high coating temperatures and make the laying and compacting more and more difficult. A too high viscosity may thus rapidly impair the benefits expected from the modified binder (especially in the case of works performed at night or under adverse climatic conditions).

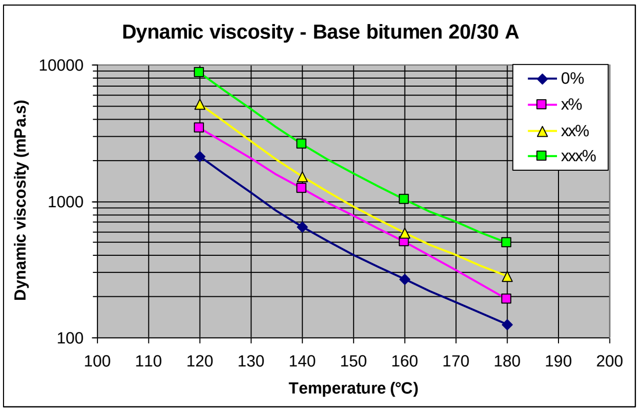


Figure 8 : Dynamic viscosity

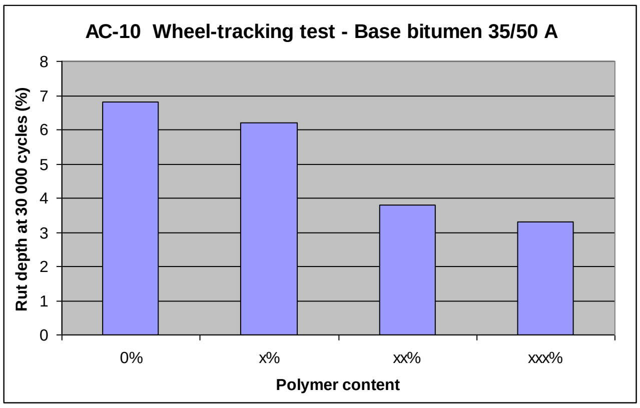
For high modulus asphalt concrete of the type investigated here, one may thus conclude that a relatively low amount of polymer seems sufficient to significantly enhance fatigue performance without diminishing the stiffness. In comparison to the use of a pure bitumen of the same penetration range, one would then get a maximum benefit in terms of pavement design life, with however only a marginal to small gain in fracture temperature and resistance to rutting. The associated increase in viscosity stays acceptable, which allows to keep manufacturing and placing conditions close to those applied when using a pure bitumen.

Higher amounts of polymer may however be justified to answer particularly severe requirements in terms of rutting and low temperature behaviour. The TSRST results do however suggest the existence of an optimum. Since, in terms of rutting, the overall performance of such mixes is already quite high, it does not seem relevant to go for too high polymer contents, all the more that the resulting surge in viscosity may rapidly compromise a satisfactory laying and compacting of the mix.

3. PERFORMANCE OF CROSS-LINKED BINDERS IN WEARING COURSES

When stepping over from the relatively thick pavement layers used for structural reinforcement to dedicated wearing courses (of which the thickness does generally not exceed 5cm), the requirements in terms of low temperature performance and resistance to rutting become more stringent. Softer base bitumen (typically 35/50 or 50/70) are then used for polymer modification. In our investigations, we have thus replaced the 20/30 base bitumen by a 35/50 bitumen of the same origin, while keeping the same levels for the polymer content. Only TSRST and wheel-tracking experiments have been conducted.

As expected, the polymer modification improves the resistance to rutting, the impact becoming really significant as from a “medium” amount (xx%) of polymer (Figure 9). This time, the impact of polymer modification on the low temperature fracture temperature is less pronounced than with the 35/50 base bitumen, since the maximum gain (again observed at xx% polymer) only reaches 3°C (Figure 10).



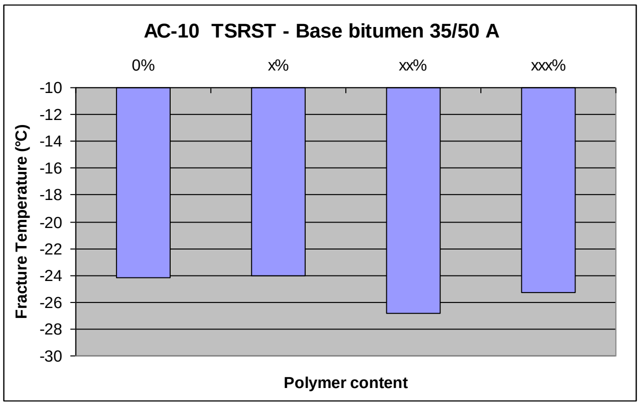


Figure 9 : AC-10-35/50 Wheel-tracking test

Figure 10 : AC-10-35/50 Fracture temperature

It is especially to be noticed that the use of a softer base bitumen leads to a global shift of all fracture temperatures by more than 5°C. As observed many times, the penetration value of the base bitumen appears once more as a dominant factor with regard to low temperature performance.

The importance of base bitumen has also triggered the question of the possible incidence of bitumen origin. Also this was not part of the initial scope of our studies, we have nevertheless been able to compare three 35/50 bitumen of different origins, either as such (unmodified) or modified with xx% of polymer. The thermal stress experiments do indeed show significant differences in-between the pure bitumen. A good surprise is however the fact that these differences in behaviour are markedly less pronounced for the corresponding cross-linked modified binders.

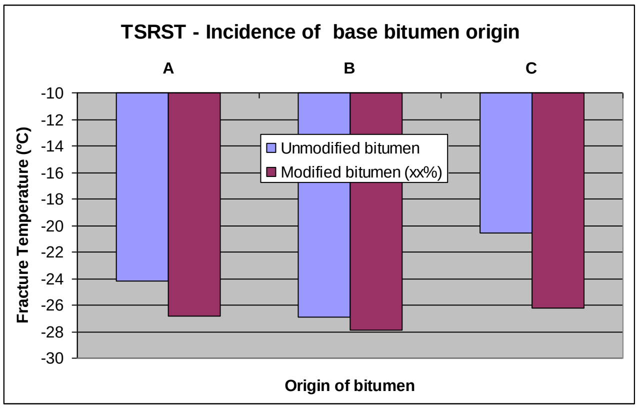


Figure 11 : AC-10-35/50 Incidence of bitumen origin on TSRST results

Overall, it may be concluded that, when increasing the penetration of the base bitumen, we observe similar behaviours and evolutions than with the 20/30 bitumen. Considering the higher initial penetration, it will probably be necessary to use somewhat higher amounts of polymer to satisfy high requirements in terms of resistance to rutting. This will be more easily achievable than in association with harder bitumen due to the lower overall viscosity levels. But, also here, it is not justified to go for unnecessarily high polymer contents. Above an optimum amount (in our case, again around xx%), the potential benefits are either uncertain (low temperature performance) or only marginal (rutting performance) and are further likely to get impaired by the negative impact of an excessive increase in viscosity.

4. CORRELATION TO BINDER PROPERTIES

4.1 Stiffness modulus

For the asphalt mixes based on the 20/30 base bitumen, we observe a good correlation between the stiffness moduli measured on the mixes at 15°C-0.02s and the |G*| value measured on the corresponding binders at 15°C-10Hz. This is well in line with existing experience since numerous researchers have shown the good correlation between the visco-elastic behaviour of binder and mix [1]. This implies also that at higher temperatures, the more elastic behaviour which is conferred to the binder by the polymer modification (see Figure 6 and 7) will also reflect in the mix behaviour and should lead to less permanent deformation.

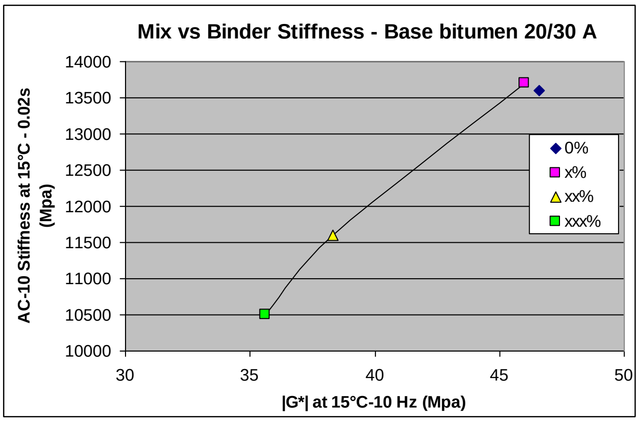


Figure 12 : AC-10-20/30 Stiffness of asphalt mix vs stiffness of binder

4.2 Fatigue

So far, for many reasons, there is no consensus on what may be an adequate binder performance indicator for the fatigue behaviour of asphalt mixes. In the limited frame of our study, and considering that the ε_6 values measured for the various polymer modified binders based on the 20/30 bitumen are nearly identical (see Figure 4), the search for meaningful correlations had of course no chance of being successful and has therefore not been attempted.

4.3 Resistance against permanent deformation

In our wheel-tracking tests we have observed a continuous decrease of the rut depth after 30 000 loading cycles with increasing polymer contents (Figures 5 & 9). As expected, a similar trend is followed by indicators usually associated with the high temperature performance of the corresponding bituminous binders such as, for instance, the softening point or rheological indicators based on |G*| and δ. Figures 13 and 14 illustrate some possible correlations.

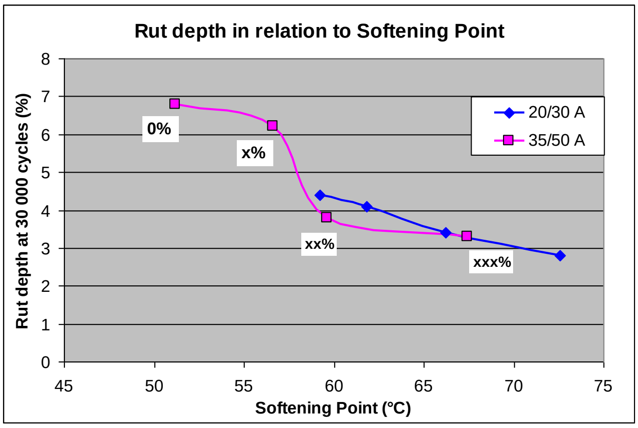


Figure 13 : AC-10 Rut depth vs. Softening Point

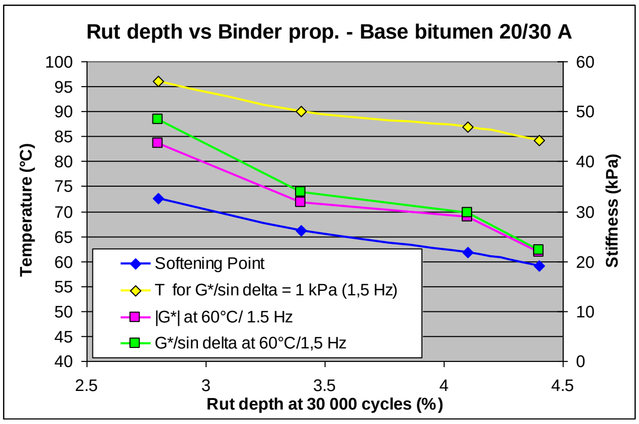


Figure 14 : AC-10-20/30 Rut depth vs. binder prop.

Since test conditions applied in binder testing (even in so-called “rheological” tests) are markedly different from the stress and strain conditions prevailing in mechanical asphalt mix testing (and even more so from those encountered in an actual pavement), such correlations can of course not be generalised and need to be re-established for every new asphalt mix formulation and type of binder. In addition, they are often questioned, especially in the case of polymer modified binders, due to the poor reproducibility of binder tests (as an effect, for instance, of the thermal history undergone by the test sample) or to specific test artefacts (highly modified soft bitumen may, for instance, lead to artificially high values of the softening point). In the present case, the correlations shown in Figure 13 and 14 may however be given some extra credit due to the fact that all modified binders have been made from bitumen of the same origin and that modification occurred via a cross-linking process which confers the binder a nearly mono-phasic structure. From these considerations, we may even conjecture (Figure 13) that the rut depths measured for the asphalt mix based on 35/50 bitumen at x% and xx% of polymer could be respectively over- and under-estimated.

4.4 Low temperature behaviour

The low temperature fracture temperatures from the TSRST tests have been confronted to “usual” binder performance indicators such as the Fraass brittleness temperature and the characteristics measured with the Bending Beam Rheometer.

The Fraass values follow quite well (with a difference of about 10°C) the evolution with polymer content as seen by the TSRST test. For the binders based on the 20/30 bitumen, the Fraass results span a maximum range of 5°C, which is similar to the range covered by the TSRST fracture temperatures but small in comparison to the (poor) reproducibility of the Fraass test (6°C for pure bitumen). Differences in Fraass values are more pronounced for the 35/50 based binders, which is what one would have expected but somehow in contradiction with the TSRST results which are much less differentiated. It is further surprising to notice that the Fraass test, contrary to the TSRST test, did not prove to be sensitive to the different types of 35/50 unmodified bitumen (Figures 15 & 16).

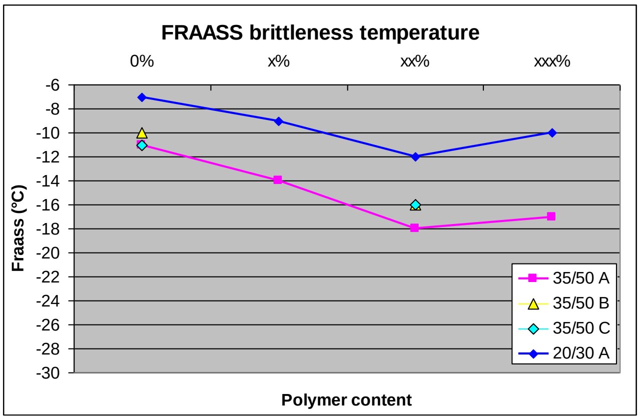


Figure 15 : Evolution of Fraass brittleness temperature

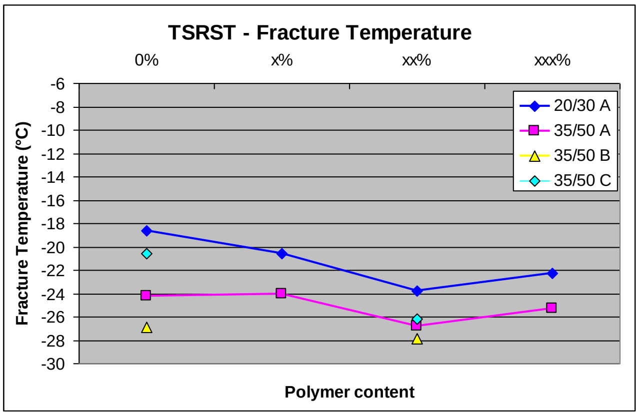


Figure 16 : Evolution of TSRST Fracture Temp.

BBR characteristics after RTFOT ageing are generally quite close to those measured on the un-aged binder. They are more substantially affected by RTFOT+PAV ageing, which is the state of binder which has been considered when establishing the SUPERPAVE criteria. When searching for possible correlations with the TSRST results, we have considered both the BBR results obtained on the un-aged binder and those obtained on the RTFOT+PAV aged binder. The TSRST test samples being made from asphalt mixes prepared in the laboratory, we may estimate that the “degree of ageing” of the binder is somewhere in-between the un-aged and RTFOT aged condition.

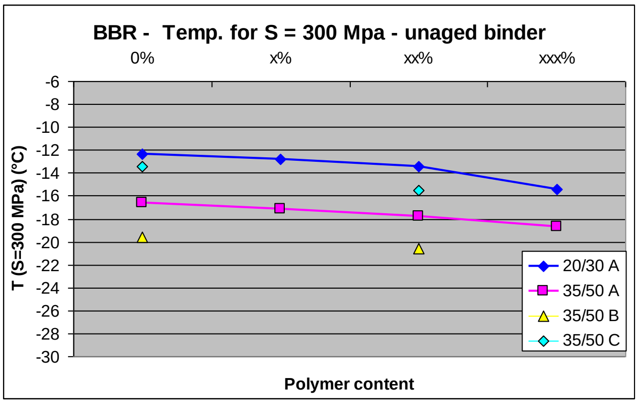


Figure 17 : $T_{S=300\ MPa}$ – unaged binders

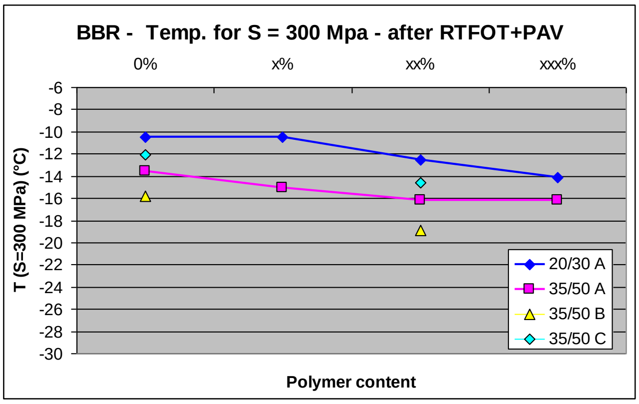


Figure 18 : $T_{S=300\ MPa}$ – RTFOT+PAV aged binders

We observe that, be it before or after ageing, $T_{S=300\ MPa}$ reflects quite accurately the ranking of the various pure bitumen (0% polymer) seen by the TSRST test. It does however keep this ranking for the modified binders (xx% polymer), which is not the case for the TSRST test. With regard to the impact of polymer modification, the predictive power of $T_{S=300\ MPa}$ proves to be rather disappointing since it is even less discriminating than the TSRST fracture temperature and does not evidence any optimum value (slow, but constant, improvement with increasing polymer content). The same conclusions can be made concerning the $T_{m=0.3}$ indicator (Figures 17 to 20).

The fact that the BBR test essentially measures stiffness and not, a failure behaviour as in the TSRST test, could account for these observations.

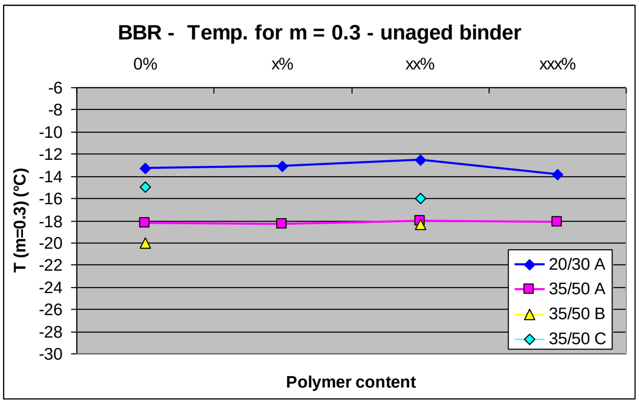


Figure 19 : $T_{m=0.3}$ – unaged binders

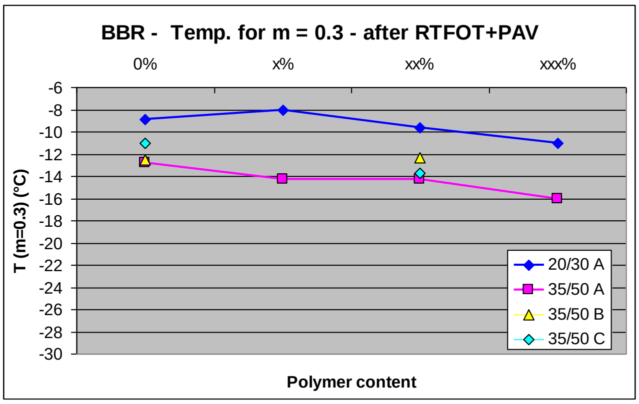


Figure 20 : $T_{m=0.3}$ – RTFOT+PAV aged binders

4.5 Binder selection criteria

From the above attempted relationships between binder and asphalt mix performance, we may try to derive a number of practical conclusions with regard to the choice of adequate selection criteria for cross-linked polymer modified binders.

We have seen that the evolutions of Fraass brittleness temperature are rather well in line with those of the fracture temperatures as seen by the TSRST tests. The variations in relation to polymer content stay however of the same order of magnitude as the (poor) reproducibility value of the test, which may lead to obvious practical problems for product quality control purposes. The BBR test is known to have a much better reproducibility but we have also seen that in our case, the BBR indicators (S or m value) are not of great help to differentiate the different levels of modification. In particular, they do not indicate any optimum or threshold value as suggested by the TSRST results. On the other hand, the BBR indicators may nevertheless be seen as “level indicators”, since they do effectively differentiate the polymer modified binders based on 20/30 bitumen from those made with a 35/50 base bitumen.

Since (as shown under § 3) the penetration of the base bitumen turned out to be the dominant factor with regard to low temperature performance, it would be the simplest and most practical criterion for binder selection or specification purposes. In the case of the cross-linked polymer modified binders studied here, the penetration of the modified binders is only slightly different from the penetration of the base bitumen used and it is furthermore not significantly impacted by polymer content (see Figure 21). At least in first instance, the penetration of the polymer modified binder could thus be a sufficiently reliable indicator for the penetration of the base bitumen and hence for low temperature performance.

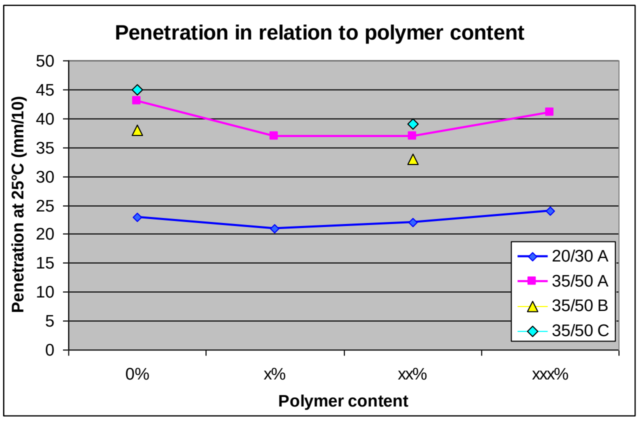


Figure 21 : Penetration of cross-linked elastomer binders in relation to polymer content

If penetration could thus be a first indicator for low temperature behaviour, there remains a need for an indicator of the degree of modification which, as we have shown, conditions more particularly the behaviour at elevated service temperatures. In France, tenders often refer to the Plasticity Range, which is the difference between Softening Point and Fraass brittleness temperature. This leads to a general strive, as well from the side of the specifying bodies as from the side of the producers, for higher and higher Plasticity Range values. The corresponding products are then more and more modified, in part also so as to provision a sufficient safety margin against the poor reproducibility of the Fraass test. However, we have seen that a too heavy modification is not necessarily a guarantee for a better performance and that, in addition, the later may even be impaired by more difficult mixing, laying and compacting conditions.

Replacing Plasticity Range by an alternative indicator not related to Fraass is in our opinion not only desirable but also well possible. Polymer modification impacts both thermal and loading time susceptibility of bituminous binders. It also changes significantly the balance between the viscous and elastic components of rheological behaviour. All these characteristics are easily evidenced by measuring the complex modulus ($|G^*|$, δ) at different temperatures-frequencies and performance indicators based on the evolution of $|G^*|$ with temperature and on the values of δ as compared to pure bitumen have already been proposed in the past [2]. One may rightly object that rheology measurements are difficult to master. The conditions for obtaining reproducible results (one has to strictly follow well identified test protocols) are however better and better known [3]. In that respect, it may also be mentioned that, due to their quasi mono-phasic structure, these difficulties seem to be more easily dealt with in the case of cross-linked binders than in the case of “physical” blends [4]. The fact that a cross-linked structure does in our opinion lead to more reliable and reproducible test results should also apply to less “sophisticated” indicators of polymer modification such as the Softening Point or, better, a Penetration Index based on Softening Point and Penetration.
All these conjectures do of course only apply to the specific family of cross-linked elastomeric binders which have been investigated here and do certainly need to be further validated by additional investigations (in particular on softer binders).

5. CONCLUSION

Our investigations on the mechanical performance of cross-linked elastomer modified asphalt mixes have confirmed all the interest of this kind of modified binders. In particular, we could evidence a significant contribution to the resistance against fatigue and, as it is well known, to the resistance against permanent deformation. The polymer also brings a positive contribution to low temperature performance as seen by the TSRST test. Fatigue and low temperature performance are however not constantly improving with increasing polymer content. Our results suggest some optimum or threshold value above which performance tends to stay level or even to decrease slightly. Resistance against permanent deformation should also tend towards an asymptote with increasing polymer content. These findings need of course to be further validated and the study extended to additional (softer) binders and mixes.

But it may already be concluded that the optimisation of an asphalt mix performance does not systematically call for the highest level of binder modification. In the case of a high modulus asphalt concrete, we have seen that a relatively low polymer content was already sufficient to significantly increase the structural reinforcement capability without impairing the other characteristics. High polymer contents may of course be necessary to cope with particularly stringent requirements at both low and high service temperatures. But this content should not exceed a certain limit which corresponds to either the optimum achievable mix performance (low temperature and probably also fatigue behaviour) or to a limiting viscosity above which proper laying and compacting could be compromised. We have also noticed that the thermal fracture temperature seems to be predominantly controlled by penetration. Our results would thus confirm an often encountered mix design policy in which one starts from a “not too hard” base bitumen and ensures high temperature performance through a high enough polymer content.

It is our belief that Plasticity Range based on Softening Point and Fraass brittleness temperature is not the best selection criterion for polymer modified binders. As a matter of fact, the question of performance related binder specifications stays an open question to which our study can only propose some “lines of thought”. Furthermore, our studies have been focused on cross-linked elastomer modified binders and our findings can of course only be applied to this “family” of binders. But they would imply that, having ascertained that a binder effectively belongs to this family (which could be done via “fingerprints” such as microstructure picture, elastoric recovery and storage stability), one could use relatively simple performance indicators such as the penetration for low temperature behaviour. For high service temperatures, those would be $|G^*|$ and δ and their susceptibility to temperature and frequency, whereas Softening Point and Penetration Index could be sufficient for production control purposes.

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