RECYCLED POLYMERS FOR USE AS BITUMEN MODIFIERS

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ABSTRACT

The volume of waste polymer being produced continues to rise. Disposal is difficult and waste exceeds acceptable levels. The purpose of the research is to examine the possibility of incorporating waste polymer into bitumen as a modifier. The aim was to find recycled polymer modified binders that would be similar to proven modified binders or that would augment the properties of 100 penetration grade bitumen.

A wide range of recycled polymers was tested including polyethylenes, polypropylenes, polyetherpolyurethane, ground rubber and truck tyre rubber. Tests included viscosity, penetration, softening point, ageing and rheology. Stiffness tests on samples of bituminous mixes made using a selection of binders also formed part of the research programme.

Although stability problems were evident with some recycled modified bitumen some cases were found to be successful. The blend with 3% low density polyethlyene substituted for 1% sytrene butadiene styrene had similar properties to that of Polyflex 75 although it had lower stiffness. The most impressive was a combination of low density polyethylene, bitumen and ethyl vinyl acetate.

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INTRODUCTION

Modification of binders with polymers can increase the rutting resistance and decrease temperature susceptibility as well as combating the effects of fatigue in a pavement (Johnston, 1994). Many different polymer modified binders are currently on the market. Polymers currently used include styrene butadiene styrene (SBS) and ethylene vinyl acetate (EVA).

Thousands of tonnes of waste polymers are produced every year (PIA, 1997) but disposal is particularly difficult. New methods are being explored to help relieve this problem. One of these methods is the use of recycled polymers as a bitumen modifier. The aim of the research is to evaluate the performance of recycled modified bitumens and compare their properties with those of a standard bitumen and a polymer modified bitumen. The first section of the paper includes a description of the binders and polymers followed by an examination of the properties of the binders such as penetration, viscosity and softening point. Nine binders were selected for ageing and rheology testing, the results of which are presented, followed by the results of stiffness testing on bituminous mixes made using the three binders showing most potential from the earlier tests. The paper ends with a discussion of the results after which conclusions including suggestions for further work follow.

MATERIALS

The three base bitumens and the recycled polymers are described below.

Bitumen

Three types of bitumen were used as the base binders: a Middle East sourced 200 pen. bitumen and a Venezuelan sourced 200 pen. bitumen. A modified binder, with commercial name Polyflex 75 (P75) was the other base binder. Styrene butadiene styrene (SBS) is the

modifier used in Polyflex. In the tests involving P75, some SBS was removed and replaced with a waste plastic polymer.

Waste Polymers

A wide variety of polymers were used to represent the range of waste plastics currently available. Brief descriptions of the polymers are given below.

Polyethylene belongs to a group of thermoplastic polymers known as polyolefins. The polyethylenes used in this project were reprocessed high density (HDPE) and low density (LDPE) polyethylenes, the properties of which are given in Table 1. Two types of polypropylene were used; homopolymer polypropylene (HPP) and copolymer polypropylene (CPP) (also presented in Table 1).

Polyetherpolyurethane (PEPU), the density of which was 1.09 g/cm^3, was supplied in the form of cut fibres. The rubber used (TTRB) was a sample of reclaimed truck tyres. The sample was derived from ambient grind where the tyre was torn apart at room temperature and then ground. Particle sizes are less than 0.02 mm and its density is 1.1-1.2 g/cm^3. Table 2 contains information provided by the supplier and is an indication of content only.

Rubber flour (GRRB) was another type of rubber examined. Content data for this polymer is not available. It was sourced from the elastic in diapers and swimwear and was supplied in powder form. Ethyl vinyl acetate (EVA) was used in some of the blends in the forms presented. Its basic constituents are polyethylene and a vinyl acetate, a copolymer. 14%, 20% and 33% of vinyl acetate were present in EVA1, EVA2 and EVA3 respectively.

TEST PROCEDURES

Blending of the polymers and bitumen was conducted in a Silverson Blending Machine set at 1300 rpm during addition of polymer but at 700 rpm while blending. LDPE, HDPE and EVA were blended for 45 minutes at 165^0C. CPP and HPP were blended for a similar length of time but at 175^0C. P75 with LDPE was blended for 90 minutes at 175^0C, whereas the PEPU was blended for a similar amount of time but at 200^0C. The TTRB and GRRB mixes were blended for 120 minutes and at a temperature of 200^0C. The so-called gap effect is only likely to have been a problem with the PEPU because of the fibrous nature of this polymer; the size of the fibres were 0.5 mm in thickness by 1-2 mm in length. The particle sizes of the other materials were much smaller e.g. the truck tyre rubber particles were 0.02mm in diameter.

Blends were first tested using empirical tests for viscosity, softening point and penetration. Nine blends were then selected and tested for ageing susceptibility and rheology. A smaller selection of blends was then used in bituminous mixes to evaluate their stiffness properties and compared with conventional mixes. The tests used at each stage are referred to in this section.

The softening point and penetration test methods are presented in IP (1989). The rolling thin film oven test (RTFOT) is described in CEN (1997). Rheology testing was conducted to determine how polymer modified binders behave under certain loading conditions at various temperatures. The equipment used was a Bohlin Dynamic Shear Rheometer II using the 25 mm diameter parallel plate geometry. Each sample was tested using the research option and the SHRP test option provided by the Bohlin Instrument software.

A stress sweep test, where a range of stresses and frequencies was applied to the binder and the resulting strains on the binder was measured, was used to determine the linear viscoelastic region of the binder under consideration. The stresses selected ranged between 50 Pa and 3,240 Pa. The frequencies selected were 0.1 Hz, 1 Hz and 1.5 Hz and the test temperature was 25°C. The sample is subjected to the stress imposed by the oscillating upper plate and the resulting strain is observed. The output graph is one of stress vs. complex modulus (G*), storage modulus (G'), loss modulus (G''), viscosity and phase angle.

A frequency sweep is a test carried out at different frequencies and temperatures. The frequencies ranged from 0.01Hz to 10 Hz and the temperatures from 15°C to 65°C. The sample is tested in the linear viscoelastic region using the strain and stress acquired from the stress sweep.

The SHRP binder test is conducted at a frequency of 1.6 Hz., a temperature of 46°C and a predetermined stress. 46°C is chosen because this is the annual maximum seven-day average temperature for Ireland. The maximum allowable stress is 200 Pa, a relatively low stress level. Some of the binders tested required much higher stresses to attain a strain in their linear viscoelastic region.

Tests Conducted on Bituminous Mixes

The Nottingham Asphalt Tester (NAT) was used to conduct the Indirect Tensile Stiffness Modulus (ITSM) test (BS, 1993) and the Repeated Load Axial (RLA) test (BS, 1990) on bituminous mixes made with a selection of the binders. The ITSM test is used to determine the stiffness modulus of an aggregate mix whereas the RLA test is conducted to

determine the resistance of an aggregate mix to permanent deformation. The RLA test conducted in this work is a variation of the British Standard method. The variation is used in Sweden and is found to have a better correlation with the Wheel Tracking test (Ulmgren, 1996).

The ITSM test is conducted at 20 °C with a 124ms rise time and a Poisson's ratio of 0.35. Four moulds from mixes made with each of the following blends were produced. The test is conducted on perpendicular diameters of each mould. The average is taken to obtain the average stiffness of the mix.

The RLA test is conducted at 30°C under a stress of 100 kPa. 1800 pulses are applied to the test specimen and the resulting strain on the sample is observed. Four specimens were tested and the average result is presented.

PROPERTY TEST RESULTS

The objective of this part of the research was to find recycled polymer modified blends, the properties of which would be similar to those of the Polyflex 75 (P75) and 100 pen. bitumen (Murphy, 1999). In the case of comparison with P75 the desired binder would have a similar penetration and softening point with a lower viscosity. Any binder with properties better than that of 100 pen. bitumen would have a higher softening point with similar penetration and viscosity. The properties of the base bitumens and 100 pen. bitumen are presented in Table 3.

Each blend is made up of a 200 pen. base bitumen and a polymer. The results of the

standard tests on the polymer modified bitumens including bitumens with HDPE, LDPE, HPP, CPP, EVA, PEPU, TRRB and GRRB added in different proportions are presented below. V indicates a blend based on Venezuelan bitumen whereas M refers to Middle East bitumen.

Penetration Results

The penetration test results for polyethylene, polypropylene and EVA V blends are presented in Figure 1. The penetration value shows how easily the bitumen can be penetrated where high values suggest a soft bitumen while low values suggest a hard bitumen. The general trend for all blends is decreasing penetration (increasing stiffness) as the proportion of polymer is increased. None of the EVA 3 blends appear to alter the penetration of the original 200 pen. bitumen. CPP hardens the bitumen considerably compared with the other polymers where only 3% of CPP is required to give the same penetration as 4-5% of the others. An addition of about 4% or above of any of the polymers except CPP tends to produce binders with penetrations of 100 dmm. Similar trends are evident for the M blends. The penetration results for EVA combined with LDPE blends are presented in Table 4.

Penetration values for the EVA 3 blends remain relatively high in the range 138 - 159 dmm whereas in the case of the EVA 2 blends the V blend values are around 100 dmm but the M blends are lower at 85 dmm indicating an increase in stiffness. The other blend with a penetration value of 100 dmm is the V2%EVA1+4%LDPE blend. General trends are more difficult to discern for EVA 1.

Penetration tests were also conducted on P75 where LDPE replaced in part the modifier (SBS) in this polymer. In the cases where the SBS content was 1% lower and

increasing quantities of LDPE were added, penetration increased to 125 dmm for 1% of LDPE but fell to 102 dmm in the cases of +2%LDPE and +3%LDPE respectively. Where the SBS content was 2% and LDPE was added, penetration values of 123, 121 and 92 dmm were observed for additions of 2%, 3% and 4% LDPE respectively.

Varying proportions of PEPU were added to Middle East and Venezuelan bitumens. All penetration values are relatively high where most of them are in the 150 to 200 dmm range. The general trend appears to be a reduction in penetration value as the proportion of PEPU is increased. The result for the M6%PEPU+3%GRRB+Oil blend is particularly high at 310 dmm. The addition of oil and ground rubber to this blend aimed to help improve the structure formed within the blend. Unfortunately, the only effect of the oil was to soften the bitumen and reduce its penetration value.

The last series of penetration tests was conducted on truck tyre rubber (TTRB) blends. An addition of 11% TTRB gave a penetration result of 140 dmm and for 16% TTRB the result was 145 dmm. These values are somewhat higher than the 100 dmm required.

Viscosity Results

Viscosity is a very relevant property when evaluating bitumen given that the bitumen is required to be sufficiently workable to coat chippings. Viscosity tests were conducted at different temperatures using a shear rate of $10{,}000s^{-1}$ on all blends and compared with the viscosity results of Polyflex 75 and 100 pen bitumen. The aim is to find a blend that has lower viscosity values than P75 (130 pen) but values similar to those of 100 pen bitumen. Reproducibility and repeatability of the viscosity test are both quite high where in most cases it is possible to reproduce a result to within 4% for most temperatures and to achieve

repeatability to within 2% for all temperatures.

The viscosity results for the polyethylene and polypropylene additions to V are presented in Figure 2. A decrease in viscosity with temperature is apparent as well as an increase in viscosity with increasing levels of polymer. Similar trends are evident for the M series of tests. The viscosities of the Middle East blends generally tend to be higher than the Venezualan blends for HDPE but the trend is reversed in the case of LDPE.

The results for M and V HPP blends are more or less the same. This is particularly evident at the three higher temperatures. The addition of 3% HPP gives the closest set of viscosity results to those of 100 pen bitumen at all temperatures.

In general, HPP and CPP give lower results than those of HDPE and LDPE. The 6% polymer content blends with HDPE raised the viscosities of the base bitumens from 2.1 Poises and 2.15 Poises to 7.8 Poises and 7.35 Poises (at the lowest temperatures) respectively. The viscosities of the 6% CPP blends reached 4 Poises and 3.6 Poises respectively at the same temperatures.

The viscosities of the VEVA1 combinations are lower than in the case of VEVA2 and VEVA3 and that VEVA2 exhibits the highest viscosities of all three. The higher additions of 4% and 6% increase the viscosity for the VEVA2 and VEVA3 more than in the case of VEVA1 where little change is evident with increasing amounts of VEVA1. The 2% additions in the case of VEVA2 and VEVA3 exhibit the closest viscosities to those of a 100 pen bitumen whereas an addition of 4% of VEVA1 is required.

The results of the EVA and LDPE combinations are presented in Table 5. The mixtures of EVA1 and LDPE show increased viscosity results when compared with EVA1 blends. The combinations of EVA2 and LDPE show similar viscosity results to the 6%EVA2 results whereas the EVA3 and LDPE blends are similar to the 6%EVA3 results.

The viscosity results for the P75 blends are shown in Figure 3. Similar trends are apparent at all temperatures. The higher the level of LDPE in the sample the higher the viscosity level achieved. In the case where P75 has a 1% lower quantity of SBS and 1% LDPE is added and in the case where the SBS content is lower by 2% and 2% LDPE is added the viscosities are lower than that of the original P75. The viscosities of all P75 blends are much higher than those of 100 pen bitumen.

The viscosities of PEPU blends are relatively low compared with other blends, the highest being 3.6 Poises. The results for M+6%PEPU+3%GRRB+Oil are particularly low. When comparing M+6%PEPU and M+10% PEPU blends one may observe an increase in viscosity in the case of the 10% addition. When comparing the results with those of 100 pen bitumen it can be observed that a 10% addition of PEPU in the M and V cases achieve similar viscosities to that of the 100 pen bitumen.

In the case of TTRB, a general increase in viscosity is evident as the level of TTRB is increased. At the higher temperatures the viscosities are similar to those of 100 pen bitumen but at lower temperatures the viscosities of the TTRB blends are much higher; 5 Poises for 11% TRRB and 6.4 Poises for 16%TRRB at 135^0C.

Softening Point Results

The softening point results for the polyethylene, polypropylene and EVA V blends are shown in Figure 4. The general trend is increased softening point with increasing amounts of polymer in the binder. The highest increases in softening point were those for CPP. Some of the blends show little change, particularly HPP where the softening point is similar for all blends. 4% of EVA1 gives a softening point value of around 65^0C as does 6% of EVA2. 6% of CPP gives a softening point value of 70^0. Similar relationships exist for the M blends.

The softening points of the Polyflex and LDPE blends are shown in Table 6. Two values of softening point are given for some blends because the two tests did not produce results within 1% of each other as required by the standard. Those blends having softening points around 70^0C include most of them except P75-2%SBS+2%LDPE which has a value of 46.4^0C and P75-2%SBS+3%LDPE (57^0C) and P75-2%SBS+4%LDPE (58 - 59.4^0C). The softening points of PEPU blends are all quite low in the 33 - 45^0C range as are those of 11%TTRB and 16%TTRB where the values are 46.6^0C and 46^0C respectively.

SELECTION OF BINDERS FOR MORE DETAILED INVESTIGATION

There are a number of blends, at first glance, that could be considered for further testing. Of the polyethylene samples only the 3% blends of LDPE were chosen. Their properties are not that similar to P75 or 100 pen. bitumen to any great degree. They were chosen more to investigate the difference between the two base bitumens. The reason more polyethylenes were not chosen is the degree to which they separated from the base bitumens. The higher percentage blends have similar viscosities to the P75 but do not have softening points of 70+°C. An addition of 4% LDPE to the Middle East bitumen was required to give the most promising results although this blend has a higher viscosity but the softening point is

less at 44.2°C and the penetration is higher at 125 dmm.

None of the polypropylenes were deemed suitable for further work for reasons similar to that of polyethylene. Although high softening points were evident they proved difficult polymers to work with and separated too easily to make them feasible alternatives. Of the EVA blends, none of the EVA3 blends were deemed suitable for further work because of the minimal impacts they made in comparison with other blends. The lowest penetration value measured was 138dmm and the highest softening point was 52.2°C. The M4%EVA1+2%LDPE blend was chosen because its penetration is similar to 100 pen. bitumen. Although its softening point is a little low at 57.6 °C it has an acceptable viscosity.

The EVA2 blends chosen were the M4%EVA2+2%LDPE blend and the V2%EVA2+4%LDPE and 4%EVA2+2%LDPE blends. The former has a comparable penetration to 100 pen. and a higher softening point and viscosity; all desirable properties. The V blends are quite similar to each other and were chosen because of their raised softening points and viscosities while retaining a penetration close to 100dmm.

The P75-1%SBS+3%LDPE blend was chosen from the P75 substitution blends because it has a comparable penetration to P75 and higher softening point and viscosity. The P75-1%SBS+2%LDPE blend is the only other blend that could be considered in comparison to the P75. It has comparable penetration and viscosity but, unfortunately, it does not have the required softening point.

The M10%PEPU blend has a comparable softening point and viscosity to the 100 pen. but its pen. value is high. It was considered for the next stage of the project with a view to

testing it in an aggregate mix. Fibres are used to keep binder drainage to a minimum in open graded mixes. The 11%TTRB blend has a higher viscosity than a 100 pen. bitumen but is softer in terms of penetration and its softening point is only minimally increased. It was considered because of its elastic nature.

AGEING TESTS

The objective of this part of the research was to investigate the response of the selected blends to ageing. The results of the rolling thin film ageing are given in Table 7. The binders all pass the required specification limits (NRA, 1998) where the requirements are a minimum retained penetration of 45%, a maximum softening point increase of 12°C, a maximum softening point decrease of 5°C and a maximum change in mass of 1%.

Of the binders tested, the minimum retained penetration was 50%, a result yielded by M3%LDPE and 4%EVA1+2%LDPE blends (Table 7). The maximum softening point increase was provided by the V3%LDPE blend and the maximum softening point decrease of 4^0C was found for the P75-1%SBS+3%LDPE blend. The blend with the largest loss in mass of 0.41% mass was the 11%TTRB blend.

RHEOLOGICAL TESTS

Each blend was tested using the SHRP rutting resistance test (SHRP, 1994) and the rheology research option on the dynamic shear rheometer. Unaged and aged samples were tested and compared. The results are presented below.

SHRP Rutting Resistance Test Results

Table 8 shows the limiting values for the rutting parameter ($G^*/\sin\delta$) permitted by the

SHRP (1994) specifications. This parameter is an indication of the rutting resistance of the binder. Table 9 shows the test results for the modified binders and their 200 pen. bitumen bases, and the ratios of the modified binder result to an unmodified binder result. This ratio is a guideline (not used for specification purposes) given in the protocol for modified bitumens (AASHTO, 1994). The comparison of the results is a means of determining how the binder performs relative to an unmodified binder. According to the protocol the ratio of the rutting parameter results ($G^*/\sin\delta$) of modified to unmodified binder should be greater than 2.5.

From the data in the table, it can be seen that each binder has passed the limiting conditions. However, if the guideline for modified binders is considered some of them do not meet the recommendations for rutting parameter (resistance). The M and V 3%LDPE blends do not meet the values suggested in the protocol, although the V blend is more resistant to rutting having higher $G^*/\sin\delta$ values and higher ratios for both the unaged and aged samples. The 10%PEPU blend also does not meet the protocol requirements as is also the case with the TTRB blend.

All other blends passed the guideline criterion with the V4%EVA2+2%LDPE aged blend giving the highest ratio of 4.9. While the rutting resistance ($G^*/\sin\delta$) values increased on ageing their ratios did not. In the case of the M blends the ratios of the unaged and aged samples remain much the same suggesting similar ageing to that of 200 pen. bitumen. In the case of the V blends the ratios either decreased or remained the same with only the 4%EVA2+2%LDPE blend ratio increasing.

The Polyflex 75, a proven binder, has a relatively low ratio of 2.79 which decreased on ageing indicating poorer ageing quality than that of 200 pen. bitumen. The rutting

resistance of the P75-1%SBS+3%LDPE blend is better in both the unaged and aged state than that of the P75.

Shear Rheometer Rheology Research Option

Three samples of both the unaged and rolling thin film (RTFOT) aged binders were tested. Two samples for each test are deemed sufficient if the acceptability criteria of the proposed Highways Agency Clause 928 method (1997) are met (i.e. complex modulus must not differ by more than 15% and phase angle must not differ by more than 3^0). If the acceptability criteria were not met then the average of the three results is required.

Data from the stress and frequency sweeps are presented by plotting graphs referred to as black diagrams. Black diagrams are constructed by plotting the log complex modulus vs. phase angle at all frequencies. Each curve on a black diagram represents one of the six temperatures at which the test was conducted (15-65°C in 10°C increments). Generally, the curve for an unmodified binder shows a phase angle increase as the stiffness modulus decreases (i.e. the material becomes more fluid-like). However, a polymer-modifier imparts a degree of elasticity as the stiffness modulus decreases (or temperature increases) - phase angle reduces.

The black diagram for P75-1%SBS+3%LDPE, in Figure 5, shows an increase in stiffness and tendency towards elastic behaviour. The maximum phase angle is approximately 65° demonstrating its improved elasticity in comparison with the 200 pen. bitumen. At the higher temperatures it has a stiffness between $1x10^2$ Pa and $1x10^4$ Pa. The curve exhibits its reduced temperature susceptibility compared with other binders, suggesting potential as a binder.

Comparison with the black diagram of P75 reveals that the modified binder has a decreased elasticity at higher temperatures but an increased elasticity at lower temperatures. The presence of LDPE causes a loss in elasticity at higher temperatures because it behaves in a plastic manner.

The black diagram of 4%EVA2+2%LDPE is shown in Figure 6 and is similar to that of P75. At 55°C and 65°C a dramatic increase in elasticity occurs coupled with a decrease in stiffness as frequency decreases. This behaviour may have to do with the combination of polymer or the level of EVA2 in the blend. Aged samples follow the patterns described for other polymer modified binders. The aged M blend curve follows the curve of its unaged counter blend more than the V one but at higher temperatures the V blend curve retains its shape. There is some scatter evident in Figure 6 but it is difficult to interpret the reasons for it.

An increase in stiffness and tendency towards more elastic behaviour was found for the V2%EVA2+4%LDPE blend and the RTFOT aged sample shows increased elastic and stiffness behaviour at all temperatures. The diagram for M4%EVA1+2%LDPE is shown in Figure 7. While the elasticity of P75 and P75-1%SBS+3%LDPE blends (Figure 5) tend to decrease or level off at higher temperatures, the elasticity of M4%EVA1+2%LDPE increases at the same termperatures. It also maintains its high stiffness at these temperatures. This part of the black diagram represents high temperatures and long loading times, thus indicative of standing traffic and fast moving traffic (high temperatures). If a binder exhibits high elasticity and high stiffness in these conditions, it would be expected to be more rut resistant.

In the case of the 11%TTRB blend, increased elastic behaviour is evident and does not

appear to vary with temperature. Its stiffness is increased at higher temperatures (longer loading times) but not as much as that of the P75 or EVA2 blends. Of all the blends, this is the one that seems least affected by ageing. The aged binder curve follows the unaged curve almost exactly. It has shifted slightly to the right showing increased stiffness.

BITUMINOUS MIXES

A selection of blends were used in the manufacture of bituminous mixes, on which stiffness tests were conducted. The Marshall mix design method was used to design a standard mix for the comparison of the binders. The grading of aggregate used was to ensure a voids content of 14%. The optimum binder content was calculated to be 4.3% and this value was used for all mixes.

The binders used in the mixes are as follows:

- 50 pen.,
- P75,
- P75 - 1% SBS + 3% LDPE,
- 11 TTRB
- 4% EVA1 + 2% LDPE (Middle East source).

Four samples of each were tested and the numbers 1 to 4 at the end of each blend designation represents the mould number. The ITSM results are presented in Table 10.

The 50 pen. mix is the stiffest at 4746 MPa whereas the P75 mix is less stiff (2687 MPa). This is due in part to the presence of SBS, an elastomer, and thus makes the blend

more elastic and less stiff. The fact that the P75 is stiffer than P75–1%SBS+3%LDPE is an unexpected result. In the rheology tests, the latter was slightly stiffer but here the P75 mix, on average, is 1000 MPa stiffer.

The 11%TTRB blend, which indicates lower stiffness than the P75-1%SBS+3%LDPE blend in rheology tests, appears to be stiffer than the latter in the ITSM tests. The Middle East blend of 4%EVA1+2%LDPE is the stiffest of the recycled polymer modified binder mixes. EVA is a plastomer and this could account for the additional stiffness observed. The extra stiffness provided by EVA over SBS mixes has also been found in other research (Clifford, 1996).

Repeated Load Axial Test Results

The repeated load axial tests revealed some interesting results, presented in Figure 8. The P75, M4%EVA1+2%LDPE and P75–1%SBS+3%LDPE blends all behaved in a similar manner. Of the three blends, the P75–1%SBS+3%LDPE mix was the most susceptible to stresses imposed upon it and exhibited the highest strain. The M4%EVA1+2%LDPE showed less damage and P75 exhibited the least damage.

Although the 50 pen. mix exhibited high stiffness values, the strain results shown in Figure 8 are relatively high. 50 pen bitumen is quite plastic and thus its response to the load is not as good as the polymer modified binder mixes. Likewise, the 11%TTRB deforms significantly. At the beginning of the test its response was like other polymer modified binder mixes. However, after 20 pulses deformation increases steadily until its maximum deformation is almost twice that of the other polymer modified binder mixes and about 1500 μstrain lower than that of 200 pen. bitumen at the end of the test. Although the 11%TTRB

blend appeared to be elastic by nature, the polymer itself was not absorbed into the bitumen. Thus, it would have acted as a separate entity in the mix, increasing the structural aspect and increasing stiffness.

DISCUSSION

All polymers showed similar trends when added to bitumen. Increased viscosity and softening point and decreased penetration were evident. Some polymers were more effective than others. EVA3 was the least effective additive with 6% polymer content only decreasing the penetration of the base bitumen to 138 dmm and increasing the softening point to 52.2°C.

The polypropylenes were the most difficult to blend. The 11%TTRB blend showed a slight improvement in the properties of the base bitumen. It should be noted that an addition of 5% extra rubber produced the same fundamental property results as the 11% blend although the 16% blend appeared more elastic. The addition of PEPU did not influence the properties of bitumen significantly. The Polyflex substitution blends demonstrated the effect SBS has on the fundamental properties of a binder. Lowering the SBS content by 1% caused a decrease in softening point of 20°C and an increase in penetration of 21dmm.

It was observed from analysis of the black diagrams that stiffness increased at lower temperatures (short loading times) and elasticity decreased at high temperatures (long loading times), but to varying degrees. The Polyflex 75 substituted binder, M4%EVA2+2%LDPE blend and the 11%TTRB showed the largest increase in elastic behaviour with the 11%TTRB blend retaining the highest degree of elasticity at lower temperatures.

The 3%LDPE binder exhibits little difference from the 200 pen. bitumen in both

cases. The 10%PEPU binder shows an increased elasticity and stiffness compared with 200 pen. bitumen especially at the lower temperatures but exhibits lower stiffness at higher temperatures. The V2%EVA2+4%LDPE binder loses its elasticity at high temperatures while the M and V 4%EVA2+2%LDPE binders appear to gain elasticity.

The samples chosen for the aggregate mix stage were the P75-1%SBS+3%LDPE blend, the 11%TTRB blend and the M4%EVA1+2%LDPE blend. The first one was chosen because it behaved like P75, a binder known to work in practice. The second one was chosen because of the nature of its elastic behaviour. It was the blend least susceptible to temperature. This blend did not meet the SHRP rutting resistance guideline but because of the nature of its black diagram it was chosen. The last one was chosen because of its retention of increased stiffness and elastic nature at high temperatures and longer loading times. These properties are desirable in order to combat rutting.

Overall, the M4%EVA1+2%LDPE blend is the most impressive. It has a high stiffness and performed well in the repeated load axial test, which means it has a high rutting resistance. This behaviour is confirmed by the black diagram; the binder showed high stiffness and high elasticity values across the full temperature range.

The P75–1%SBS+3%LDPE performed well in the RLA test but its lack of stiffness demonstrated in the ITSM test is not encouraging. Although its stiffness is relatively low, it is worth noting that it exhibited the same stiffness as that of a core taken from a two-year old 100 pen. 20mm roadbase.

Although the P75, P75-1%SBS+3%LDPE and the M4%EVA1+2%LDPE blends had

varying stiffnesses they performed very similarly in the repeated load axial test. Another interesting point is that the performance of the mixes in the repeated load axial test appears to reproduce the performance of the binders in the rheology tests. Those that exhibited high elasticity and stiffness in the black diagrams were the ones that performed best in the repeated load axial test. This suggests that the black diagram can be considered as a performance indicator for rutting resistance.

The research conducted was limited to an examination of the rutting performance of the materials tested. Clearly other parameters such as thermal cracking and fatigue properties would require evaluation before recommendation of the recycled polymer bitumens for common usage.

CONCLUSIONS

A range of waste polymers were tested to evaluate their potential for use as modifiers for bitumen. Although some were found not to be useful others showed potential. The conclusions are as follows:

1. Polypropylenes were not useful in improving the properties of bitumen and displayed practical difficulties during mixing and testing suggesting poor cohesion with bitumen.
2. The addition of 11% of truck tyre rubber showed a slight improvement in the base properties of bitumen.
3. Polyetherpolyurethane, like polypropylene, did not show much potential for improvement of bitumen.
4. An addition of 2% of low density polyethylene to a bitumen with 4% of ethyl vinyl acetate showed the most promise in terms of improved stiffness.
5. Substitution of styrene butadiene styrene with low density polyethylene exhibited good

results in the repeated load axial test but indicated a lack of stiffness in the indirect tensile stiffness modulus test.

There is considerable potential for further research in this area particularly to examine more fully those blends showing most promise but also to conduct a similar series of tests on other combinations of recycled polymers with bitumen and polymer modified bitumen.

ACKNOWLEDGEMENT

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Table 2. Contents of TTRB

Material	Content (% by weight)
Natural rubber	25-50
SBR/Butadiene content	5-30
Carbon black	25-30
Processing agents (Oil, zinc oxide, sulphur, etc.)	20-25
Benzene Extraction	3.5-9
Acetone extraction	10 (max.)
Ash content	4-6

Table 1. Properties of LDPE, HDPE, HPP and CPP

Property	LDPE	HDPE	Homopolymer Polypropylene (HPP)	Copolymer Polypropylene (CPP)
Density (g/cm^3)	0.953	0.952	0.922	0.91
Melting point (average °C)	115.1	129.1	164.2	163.8
Melt flow index (g/600s)	36.07 (@190^{0}C)	31.4 (@190^{0}C)	19.23 (@230^{0}C)	3.65 (@230^{0}C)

Table 3. Classification of base bitumens

Property	Middle East Bitumen	Venezuelan Bitumen	Polyflex 75	100 pen. bitumen
Softening point (°C)	39.6	37.4	72	45.2
Penetration (dmm.)	185	186	100	108
Viscosity (Poise) @				
135°C	2.1	2.15	7.8	3.05
150°C	1.2	1.2	4.85	1.65
180°C	0.4	0.4	2.05	0.6
195°C	0.3	0.3	1.4	0.4

Table 4. Penetration of EVA and LDPE blends

Blend Type	Penetration (dmm)
M 2% EVA1 + 4% LDPE	138
V 2% EVA1 + 4% LDPE	102
M 4% EVA1 + 2% LDPE	95
V 4% EVA1 + 2% LDPE	119
M 2% EVA2 + 4% LDPE	85
V 2% EVA2 + 4% LDPE	103
M 4% EVA2 + 2% LDPE	86
V 4% EVA2 + 2% LDPE	102
M 2% EVA3 + 4% LDPE	154
V 2% EVA3 + 4% LDPE	138
M 4% EVA3 + 2% LDPE	159
V 4% EVA3 + 2% LDPE	151

Table 5. Viscosity results of EVA and LDPE blends

Viscosity (Poises)	2%EVA1+ 4%LDPE M/V	4%EVA1 +2% LDPE M/V	2%EVA2+ 4%LDPE M/V	4%EVA2 +2% LDPE M/V	2%EVA3+ 4%LDPE M/V	4%EVA3+ 2%LDPE M/V
@135^{0}C	5 / 5.2	4.4 / 3.9	6.7 / 6	6.6 / 6.05	5.9 / 5.5	5.8 / 5.4
@150^{0}C	2.9 / 3	2.55 / 2.25	3.9 / 3.4	4 / 3.55	3.4 / 3.1	3.2 / 3.1
@180^{0}C	1.2 / 1.15	1 / 0.9	1.6 / 1.35	1.6 / 1.45	1.4 / 1.2	1.3 / 1.2
@195^{0}C	0.85 / 0.85	0.7 / 0.65	1.05 / 0.9	1.1 / 0.95	1 / 0.9	0.9 / 0.8

Table 6. Softening points of P75 and LDPE blends

Blend	Softening Point (^{0}C)
P75 - 1% SBS +1% LDPE	56 / 69.6
P75 - 1% SBS +2% LDPE	64.6 / 66.2
P75 - 1% SBS +3% LDPE	73.2 / 74.8
P75 - 2% SBS +2% LDPE	46.4
P75 - 2% SBS +3% LDPE	57
P75 - 2% SBS +4% LDPE	58 / 59.4
P75 + 2% LDPE	73 / 75.6

Table 7. Rolling thin film oven test results

	Percen. change in mass	Softening Point (°C) (Original)	Softening Point (°C) (RTFOT)	Increase in Softening Point (°C)	Pen. (dmm) (Orig)	Pen. (dmm) (RTFOT)	Percent. of Retained Penetration
Middle East Blends							
200 Pen.	-0.05 -0.07	39.6	46.2	6.6	188	100	53.19
3% LDPE	-0.16 -0.33	44.2	49.2	5	156	78	50.00
4% E1 + 2% LDPE	-0.01 0.00	53.5	60.6	7.1	110	55	50.00
4% E2 + 2% LDPE	-0.01 0.00	58.2	60.3	2.1	86	56	65.12
Venezuelan Blends							
200 Pen.	-0.36 -0.35	37.4	44	6.6	184	112	60.87
3% LDPE	-0.39 -0.37	41.9	52.2	10.3	135	73	54.07
2% EVA2 + 4% LDPE	-0.36 -0.36	56.2	58.00	1.8	103	57	55.34
4% EVA2 + 2% LDPE	-0.35 -0.37	56.8	60.8	4	102	55	53.92
Others							
P75	-0.01 -0.01	72	70.6	-1.4	100	68	68.00
P75 - 1%SBS + 3% LDPE	0.00 -0.01	74	70	-4	102	62	60.78
11% TTRB	-0.38 -0.41	46	50.5	4.5	145	114	78.62
10% PEPU	-0.17 -0.15	44.6	50.1	5.5	142	86	60.56

Table 8. Limiting values for binders tested using SHRP (1994)

Distress Mechanism	Property	Test Procedure and binder preconditioning	Limiting Value
Rutting	G*/sinδ	Dynamic Shear Rheometer	
	Dynamic Shear at maximum pavement temperature (46°C for Ireland)	Original (untreated) binder	1kPa (min.) @10rad/s
		After RTFOT treatment	2.20 kPa (min.) @ 10 rad/s

Table 9. SHRP rheology test results

	Unaged binder ($G^*/\sin\delta$) kPa	Ratio (Modified/ 200 pen.)	RTFOT aged binder ($G^*/\sin\delta$) kPa	Ratio (Modified/ 200 pen.)
Middle East Blends				
100 pen.	7.3	-	16.9	-
200 pen.	4.3	1	8.5	1
3%LDPE	7.5	1.7	12.3	1.5
4%EVA1 + 2%LDPE	19.9	4.6	35.4	4.2
4%EVA2 + 2%LDPE	17.9	4.2	40.4	4.8
10%PEPU	5.4	1.3	11.8	1.4
Venezuelan				
200 pen.	4.1	1	7.85	1
3%LDPE	8.5	2.1	16.3	2.1
2%EVA2+4%LDPE	18.0	4.3	28.4	3.6
4%EVA2+2%LDPE	18.3	4.5	38.6	4.9
11%TTRB	10.0	2.4	12.3	1.6
Others				
Polyflex 75	12.0	2.79	21.4	2.51
Polyflex75-1%SBS+3%LDPE	16.2	3.77	24.8	2.92

Table 10. Indirect tensile stiffness modulus test results

Binder	Stiffness Ø1 (MPa)	Stiffness Ø2 (MPa)	Average Stiffness (MPa)	Mix Stiffness (MPa)
50Pen1	5169	5060	5114.5	
50Pen2	4304	4116	4210	
50Pen3	5571	5600	5585.5	
50Pen4	4033	4114	4073.5	4746
P751	2542	2435	2488.5	
P752	2622	2639	2630.5	
P753	2808	2647	2727.5	
P754	3011	2788	2899.5	2687
P75-1%SBS+3%LDPE1	1940	1894	1917	
P75-1%SBS+3%LDPE2	1548	1523	1535.5	
P75-1%SBS+3%LDPE3	1753	1707	1730	
P75-1%SBS+3%LDPE4	1447	1468	1457.5	1660
11%TTRB1	2130	2141	2135.5	
11%TTRB2	2069	2065	2067	
11%TTRB3	2058	1993	2025.5	
11%TTRB4	1943	1754	1848.5	2019
M4%EVA1+2%LDPE1		3296	3296	
M4%EVA1+2%LDPE2	2860	2811	2835.5	
M4%EVA1+2%LDPE3	3339	3043	3191	
M4%EVA1+2%LDPE4	3544	3216	3380	3176

FIGURE CAPTIONS

Figure 1 Penetration values of polyethylene, polypropylene and EVA V blends

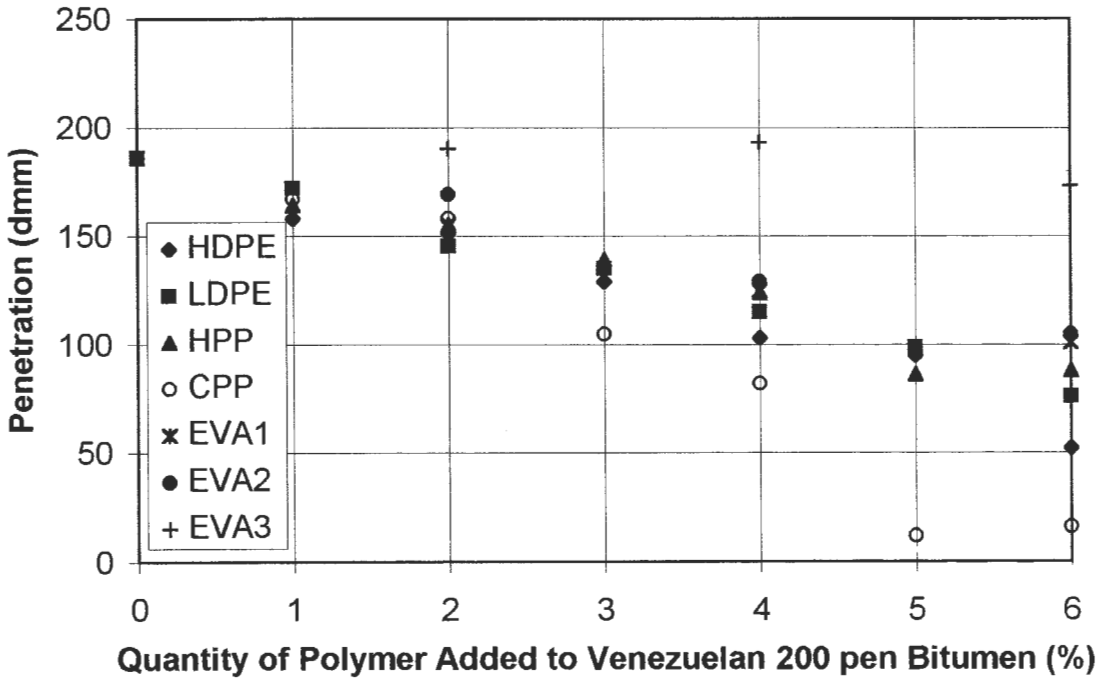

Figure 2 Viscosity results of polyethylene and polypropylene V blends

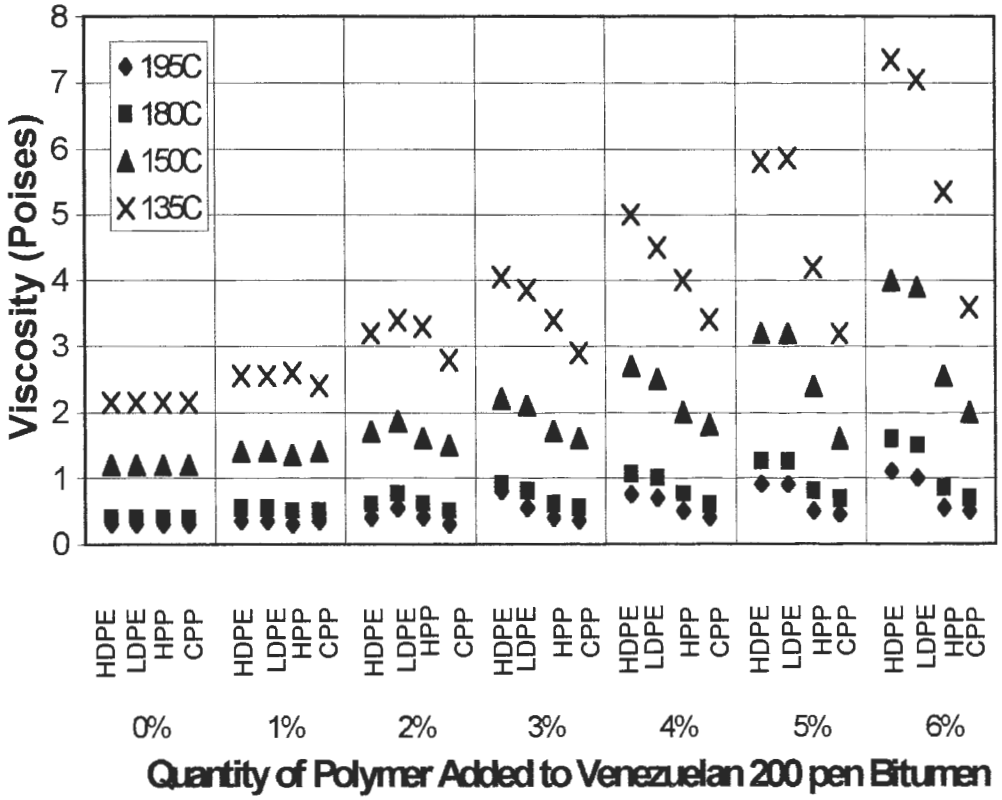

Figure 3 Viscosity results of Polyflex and LDPE blends

(Note: Negative percentages refer to reduction in SBS and positive percentages refer to addition of LDPE)

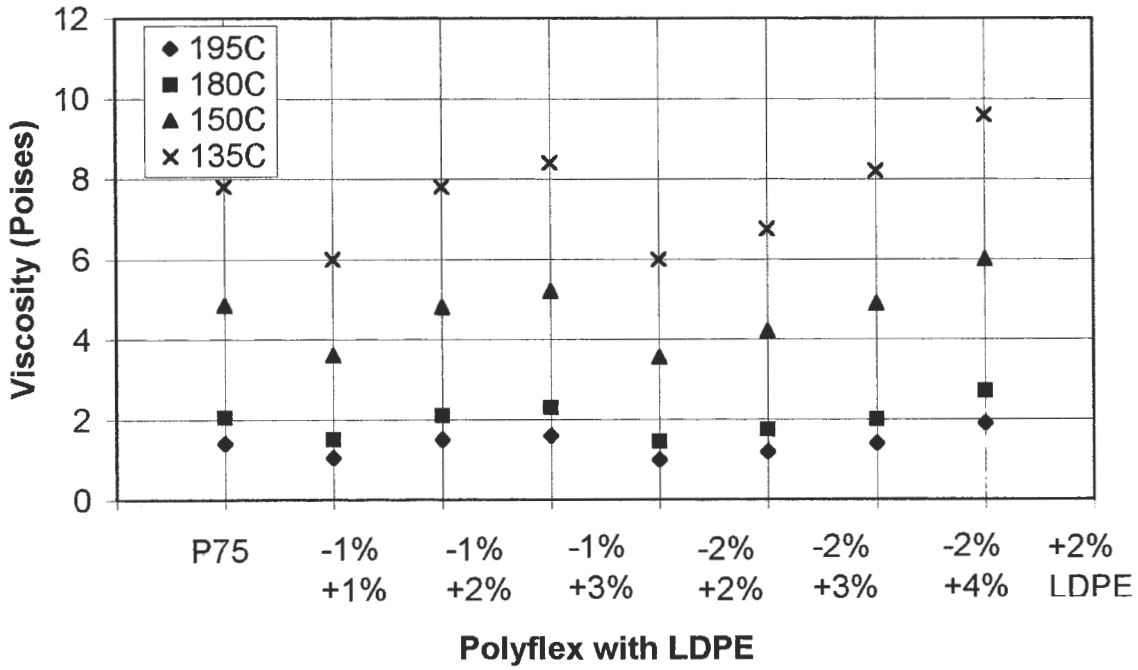

Figure 4 Softening point results of polyethylene, polypropylene and EVA V blends

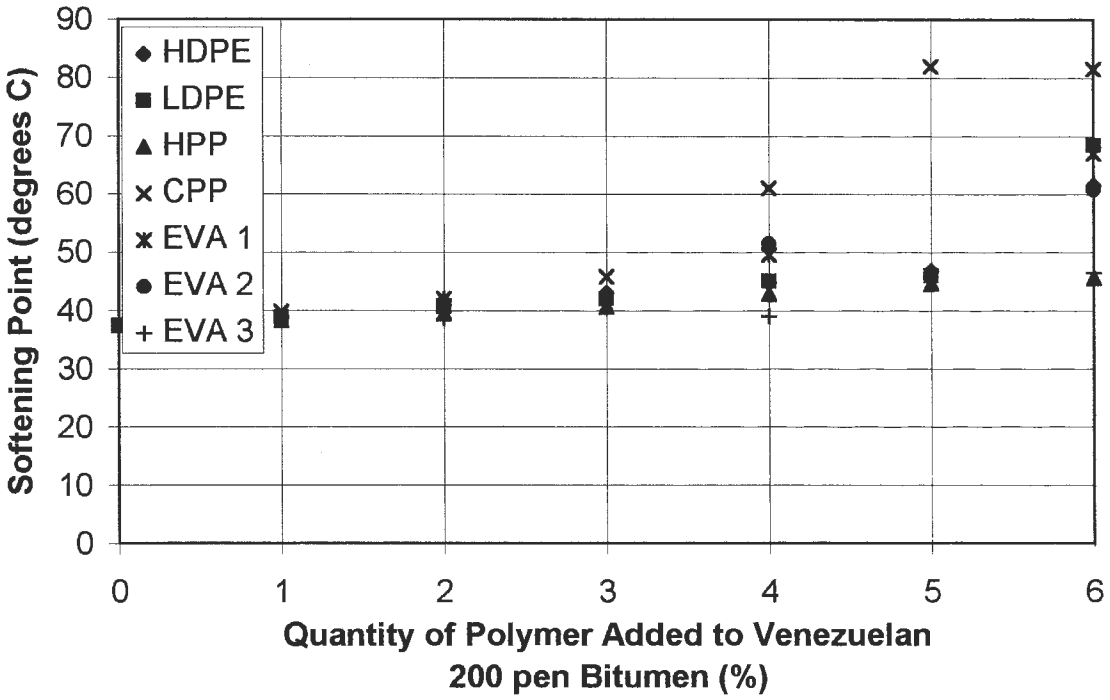

Figure 5 Black diagrams for P75 and P75-1%SBS+3%LDPE

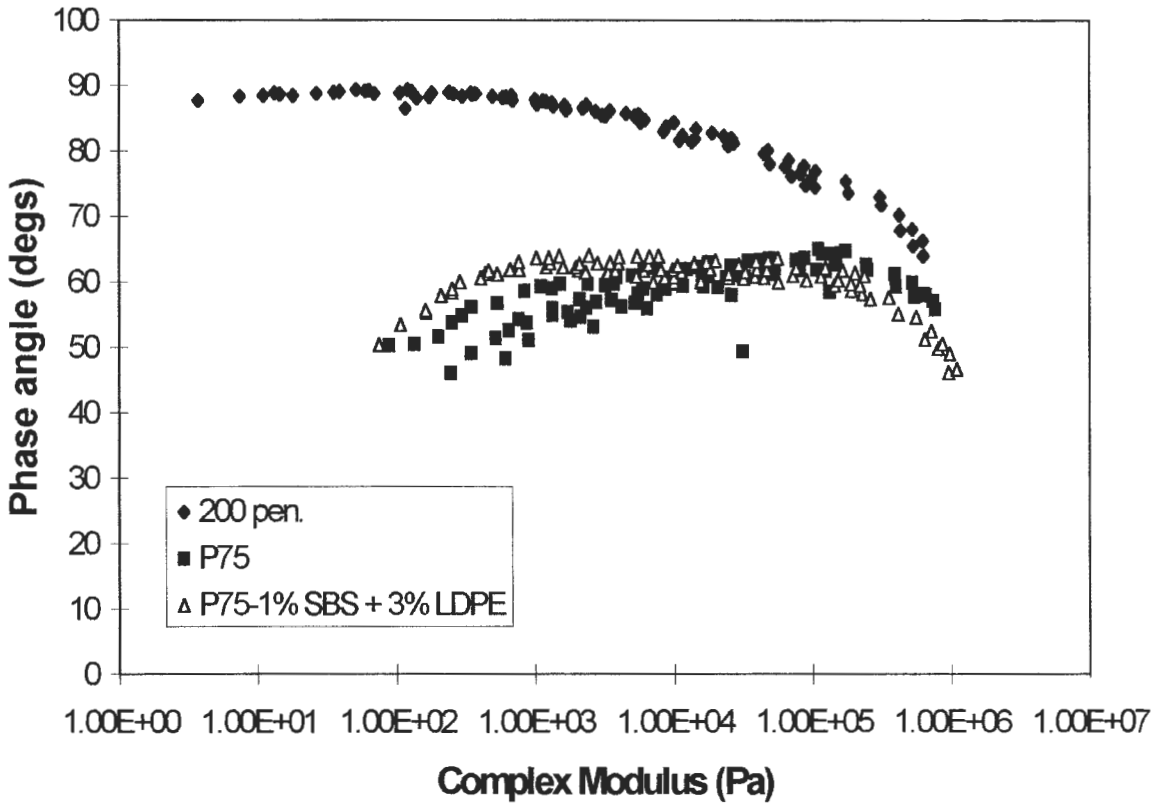

Figure 6 Black diagram for 4%EVA2+2%LDPE blends

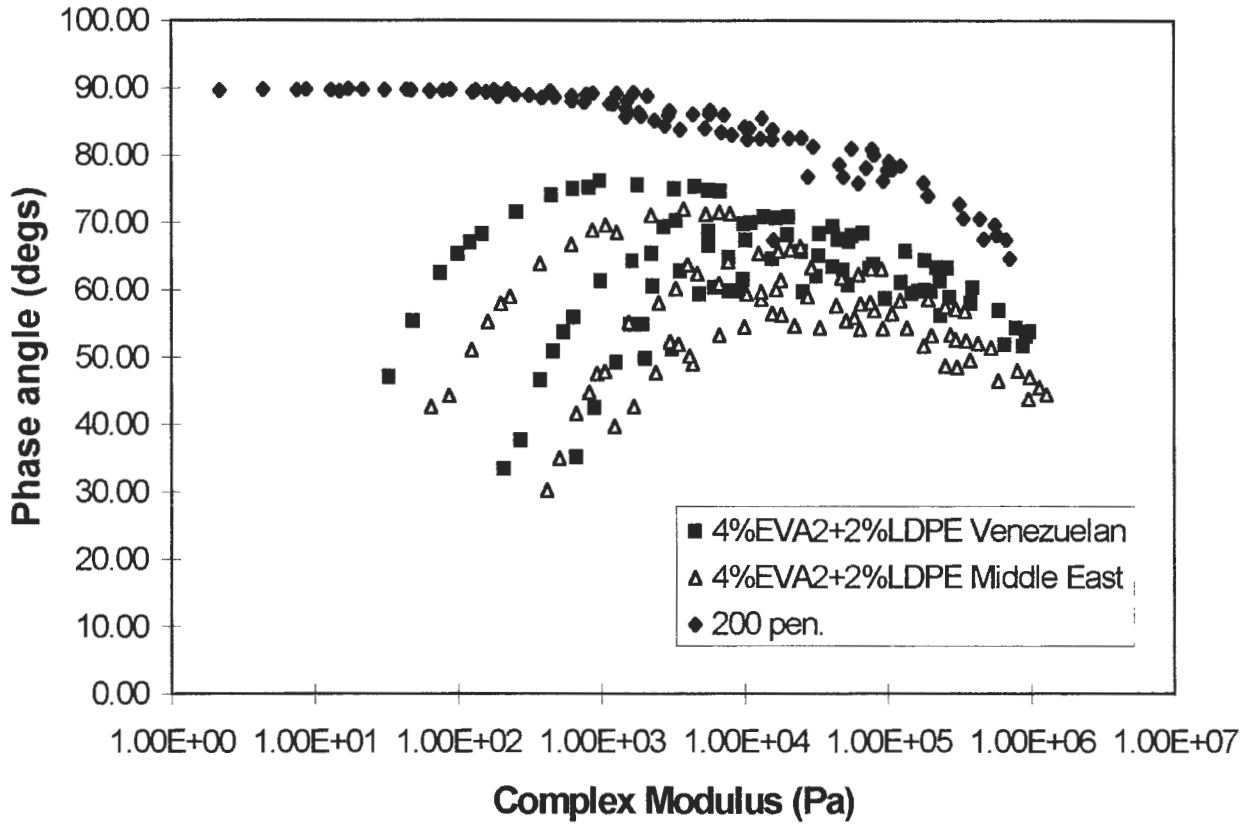

Figure 7 Black diagram for M 4%EVA1+2%LDPE

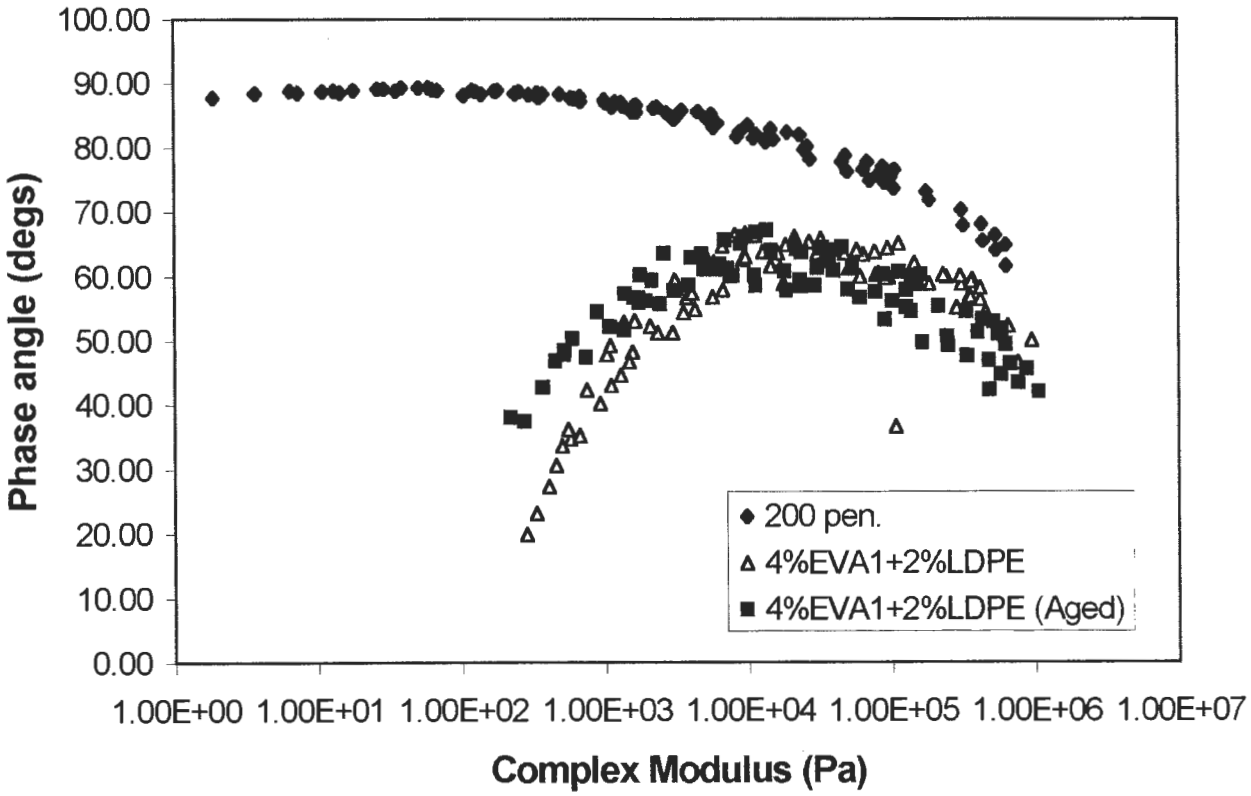

Figure 8 Repeated load axial test results