# A new look at rubber-modified asphalt binders

G. R. Morrison, S. A. M. Hesp

Department of Chemistry, Queen's University, Kingston, Canada K7L 3N6

The high-temperature rheological characteristics and the low-temperature fracture properties of asphalt binders containing crumb and devulcanized rubber waste have been investigated. Asphalt binders containing crumb rubber of different mesh sizes, with and without surface modification, and a commercially available binder containing devulcanized rubber, were tested and compared with an unmodified asphalt and three commercially available polymer-modified binders. Interfacial modification of asphalt systems containing crumb rubber was found to give binders that were far superior in their low-temperature performance to commercially available products. The data suggest that a crack-pinning or crack-blunting mechanism is responsible for the increase in toughness found in these systems. A commercially available binder containing devulcanized rubber showed reasonably good high-temperature properties; however, its low-temperature fracture performance was disappointing in that it was not significantly better than that of unmodified asphalt binders.

# 1. Introduction

For the last three decades discarded rubber tyres have found end-use applications in asphalt binders in order to improve upon both the low- and hightemperature performance of the road surface [1]. During hot summer months the rubber adds resilience to the viscous asphalt binder and this results in a pavement with better rutting resistance and durability. In cold climatic conditions, the rubber tyre can lower the stiffness of the asphalt binder, and advocates of this technology often claim that this will result in a reduction of the transverse stress cracking tendency of the pavement [2]. However, actual field trials have so far been inconclusive in their assessment of the performance/cost benefits of these materials [3].

Research on rubber modification of asphalt binders has recently gained significant attention in North America and around the world since the United States Government passed legislation which forces individual States to add waste rubber to their asphalt pavements in order for these States to remain eligible for financial assistance. This legislation will require the States to use as much as 15% crumb rubber on the asphalt binder in 10% of their federally funded highway projects starting in 1995 increasing to 20% of their projects by 1997.

# 2. Background

The development of rubber-modified asphalt binders, as they are now most often used throughout North America and in many other countries, was pioneered by the Roads Department of the City of Phoenix, Arizona. Since the developments in the late 1960s by McDonald and co-workers, a number of variations on the initial technology have been developed, often with mixed results in terms of performance enhancement.

In the so-called "wet process", as developed by McDonald, 14-20 wt % ambiently ground rubber tyre (8-20 mesh) is swollen in the asphalt's oily phase at high temperatures to form a gel-like material after mixing for approximately 45 min at 175-220 °C. To this gel is added some aromatic kerosene fraction to increase its workability. This so-called "asphaltrubber" is mixed with aggregate to form the pavement. The process requires the use of at least 20% more liquid asphalt than is used in a conventional hot-mix pavement. In some cases 40%-60% more asphalt is used, accounting for most of the increase in both cost and performance [4]. The thicker films of asphalt at the aggregate interface result in greater durability. The resulting lower void content in the mix results in slower oxidation of the binder films and improved freeze/thaw resistance of the pavement. These asphalt-rubber binders also have higher softening points which can result in less bleeding and permanent deformation. Moreover, a 90% reduction in traffic noise has been observed on pavements with asphalt-rubber mix overlays as compared to conventional pavements [5]. Because of the substantially higher initial cost of the wet process, the technology has been primarily used for the control of reflective stress cracking. Asphalt-rubber has found applications in stress-absorbing membranes, in stress-absorbing membrane interlayers, as a crack and joint sealer and to a lesser degree as a binder in thin asphalt concrete overlays [6].

Another method for incorporating recycled rubber crumb into asphalt pavements which has received considerable attention is the so-called "dry process". It was originally developed in Sweden under the trade name Rubit [7] and subsequently registered in the USA under the trade name Plusride. It differs from the wet process in that the rubber crumb is slightly larger  $(1/16-1/4 \text{ in}; \sim 1.58-6.35 \text{ mm})$  and is directly mixed with the aggregate before the asphalt binder is added. The rubber is added at a loading of 3-4 wt % of the aggregate. This process requires a special aggregate gradation in order to avoid any interference of the rubber crumb with the aggregate which can lead to premature stripping. In addition, this process also calls for typically 1.5-3% more liquid asphalt than a conventional hot-mix [8, 9]. The increased asphalt content is needed to achieve a voids content below 3% in order to prevent premature ravelling of the pavement  $\lceil 10 \rceil$ . In cold climates, the Plusride technology has been used quite successfully to reduce the harmful effects of ice formation on roads. Compared to conventional hot-mix pavements, a 25% reduction in stopping distances on ice-covered roads has been recorded for pavements made by the process [11, 12]. Takallou [13] has developed a dry process which uses conventional aggregate gradations by specifying finer crumb rubber grades. The process, which is known under the Generic name, has been used in a number of paving test sections throughout the United States [14].

One serious concern which has hindered the largescale acceptance of asphalt-rubber technology is the high initial cost combined with the uncertainty regarding future benefits. The fact that the cost of rubber-modified hot-mix pavements, in general, is currently anywhere from 60%-150% above the cost of a conventional asphalt pavement, has deterred many municipal and state governments from using this technology [15]. Today, asphalt-rubber is mainly used in low-volume applications. It may very well be more sensible to use less crumb rubber in the asphalt so that initial costs will not be a deterrent to the widespread application of the technology. If modest amounts (4-10 wt %) of fine crumb rubber are applied to an asphalt binder, a pavement may be constructed with normal binder contents and aggregate gradations which would result in only a slight overall increase in cost. This by itself may make the technology more widely acceptable which could eventually mean that much more scrap tyre will find its way into hot-mix asphalt pavements.

Recently, paving trials in Florida [16] and Ontario [17] have used binders which contain only 7%-9% fine crumb rubber (80 mesh) directly blended into the asphalt cement. Initial laboratory test results are quite promising, but it is far too early to draw any conclusions from the performance of these trial pavement sections [18].

This paper describes fundamental rheological and fracture properties of a number of such recently developed asphalt binders containing only modest amounts of recycled rubber and compares them with conventional asphalt-rubber binders. Moreover, a new process is investigated in which the crumb rubber is *in situ* reacted with the asphalt phase in order to improve upon interfacial adhesion and stability. Thus far, the only way in which crumb rubber has been added to asphalt pavements is by simply mixing the rubber and the binder or aggregate prior to paving. Recently, however, new binders have been developed containing both crumb rubber and soluble styrene-butadiene rubbers [19, 20]. In this paper we will show that an actual reaction between a soluble rubber and the crumb rubber can yield binders of even greater toughness.

A commercially available homogeneous asphalt binder containing devulcanized rubber tyre is also investigated for its rheological and fracture performance. This binder contains 10 wt % rubber tyre and the company which produces this product states that its price is only 3%-5% higher than conventional hot-mix asphalt binders with an increase in performance over unmodified products [21].

# 3. Experimental procedure

#### 3.1. Materials

The asphalts used in this study were an 85–100 and a 150–200 penetration grade both from the Bow River area in Alberta, Canada and a 200–300 penetration grade from Venezuela. The Bow River samples were obtained from the Lake Ontario refinery of Petro-Canada in Oakville, Ontario and the 200–300 sample was obtained from the Imperial Oil refinery in Montreal, Quebec.

The wet-ambiently ground scrap passenger car tyre was obtained from Rouse Rubber Industries of Vicksburg, MS. The 10, 20, 30 and 40 mesh cryogenically ground passenger car tyre samples were obtained from Recovery Technologies of Mississauga, Ontario. The 40 mesh ambiently ground tyre sample for the preparation of the wet process asphalt–rubber binder was obtained from Baker Rubber of South Bend, IN. The asphalt sample containing 10% devulcanized scrap tyre, Ecoflex<sup>TM</sup>, was obtained from Bitumar Inc. of Montreal, Quebec.

The liquid polybutadiene (LPBD) was obtained from Ricon Resins of Grand Junction, CO. It contains 80% 1,4-*trans* and 1,4-*cis* units and 20% 1,2 vinyl and it has a number average molecular weight of  $12000 \text{ g mol}^{-1}$ .

The Styrene–butadiene (SBR) rubber-modified asphalt, Styrelf<sup>™</sup>, was received from McAsphalt Industries of Scarborough, Ontario. The ethylene vinyl acetate (EVA) modified binder, EB-330, was obtained from Imperial Oil in Sarnia, Ontario and the stabilized polyethylene (PE) binder was prepared according to procedures described in the literature [22].

# 3.2. Procedures

# 3.2.1. Sample preparation

The rubber-modified asphalt binders were prepared by slowly adding a known amount of rubber to the asphalt at  $170 \pm 10$  °C. Small amounts of low molecular weight polybutadiene and sulphur were added to a number of the samples. The mixtures were processed under high shear employing a laboratory mixer (Polytron Mixer, Brinkman Instruments), for a period of 2 h.

# 3.2.2. Dynamic mechanical testing

Samples were heated until liquid at 130-140 °C, poured into the combined melts and solids (CMS) test fixture, and then allowed to cool to room temperature. After cooling, the sample was smoothed flat using a hot metal scraper, and allowed to cool once again. No conditioning of the sample prior to testing was employed.

Testing was done with a Rheometrics Dynamic Analyser RDA II equipped with a liquid nitrogen supply to maintain low-temperature conditions. The CMS fixture consists of a 42 mm diameter cup and a bilevel plate which has an 8 mm diameter serrated surface concentric with and projecting from a 25 mm diameter plate. Data for both the solid and the melt are obtained from the same sample by way of the instrument automatically lowering the height of the bilevel plate at 40 °C. Data for master-curve generation was obtained using programmed frequency-temperature sweeps which control temperature and soak time at a given temperature prior to performing the actual test. Tests were done from -20-60 °C, using eleven frequencies ranging from  $0.1-10 \text{ rad s}^{-1}$  at each temperature. The sample was conditioned at each test temperature for a minimum of 11 min. Curves for tan  $\delta$ , G' and G'' versus temperature were generated using programmed temperature sweeps between -80 and  $10^{\circ}$ C at  $0.1 \text{ rad s}^{-1}$  taking a measurement every 2 °C after a soak time of 1 min. All data are calculated automatically by the RDA II software based on the input strain,  $\gamma$ , and strain angle,  $\theta_{v}$ , and the measured stress,  $\tau$ , and stress angle,  $\theta_{\tau}$ , using Equations 1–5 [23]

$$\delta = \theta_{\tau} - \theta_{\gamma}, \qquad (1)$$

$$G' = \cos(\delta)(\tau/\gamma),$$
 (2)

$$G'' = \sin(\delta)(\tau/\gamma),$$
 (3)

$$|G^*| = (G'^2 + G''^2)^{1/2}$$
(4)

$$\tan(\delta) = G''/G'$$
 (5)

where  $\delta$  is the angle (rad), G' the storage modulus (Pa), G" the loss modulus (Pa),  $|G^*|$  the complex modulus (Pa), and tan  $\delta$  the loss tangent. The raw G\* versus  $\omega$  curves were shifted based on the time-temperature superposition principle (TTS), using the curve at 20 °C as the reference point.

#### *3.2.3.* Low-temperature fracture testing

The reacted rubber-modified binders were poured into silicone rubber moulds and subsequently left in a freezer at -20 °C for a minimum of 18 h before testing. The samples were tested using a three-point bending test based on ASTM method E 399–90 [24]. Testing was done in a temperature-controlled environmental chamber, maintained at -20 °C, using a computer-interfaced Sintech 2/G testing frame. The sample bars measured 25 mm wide by 12.5 mm deep by 175 mm long, and had a 90 ° starter notch 5 mm deep in their centre which was sharpened with a razor blade just prior to testing. The length of the loading span was 100 mm. Brittle fracture studies were completed for all rubber-modified samples and the 85–100 and 150–200 penetration control samples. Measured data included the failure load and the modulus. The fracture toughness was calculated according to Equation 6 [25, 26].

$$K_{Ic} = \frac{P_f S}{BW^{3/2}} \left\{ 3 \left( \frac{a}{W} \right)^{1/2} \left[ 1.99 - \frac{a}{W} \left( 1 - \frac{a}{W} \right) \right] \right\} \times \left( 2.15 - 3.93 \frac{a}{W} + 2.7 \frac{a^2}{W^2} \right) \right\} \times 2 \left( 1 + 2 \frac{a}{W} \right) \left( 1 - \frac{a}{W} \right)^{3/2} \right\}$$
(6)

where  $K_{Ic}$  is the fracture toughness (N m<sup>-3/2</sup>),  $P_f$  the applied failure load (N), S the load span (m), B the specimen depth (m), W the specimen width (m), and a the crack length (m).

### 3.2.4. Environmental scanning electron microscopy

Fracture surfaces were examined in an Electroscan environmental scanning electron microscope (ESEM) in order to learn more about the effect of the rubber on the low-temperature fracture toughness. Notched beams were broken in a three-point bend test as described in the previous section. The fracture surfaces were cut with a sharp knife and subsequently glued on to a 25 mm microscope stub. The specimens were kept frozen with dry ice prior to examination in the ESEM.

# 4. Results and discussion

#### 4.1. Rheological testing

An example of the raw  $G^*$  versus  $\omega$  curves is shown for the 150–200 binder in Fig. 1 along with the master curve resulting form the time-temperature superposition procedure. As expected, increasing the amount of rubber in the binder increases the resistance to deformation, as shown in Fig. 2 for the 80 mesh samples at 4% and 10% loading. At the high-temperature end of



Figure 1 Time-temperature superposition principle demonstrated for a 150-200 penetration grade asphalt binder.



Figure 2 Master curves for a 150-200 grade binder modified with 0, 4 and 10 wt % 80 mesh crumb rubber tyre.

the curves (corresponding to low frequency), the addition of the ground rubber tyre has a pronounced effect on the material's ability to resist deformation, i.e. the addition of rubber increases the complex modulus.

Fig. 3 is included for comparison of the rubbermodified binders with commercially available polymer-modified binders. At temperatures of 50-60 °C the SBR- and polythylene-modified asphalts compare with the more economical 4% 80 mesh GRT, and are surpassed by 10% GRT at 20, 40 and 80 mesh sizes.

Plots of G', G" and tan  $\delta$  versus temperature have been used extensively as a means of determining lowtemperature properties of modified and unmodified asphalt binders. This would appear to be a logical procedure from the point of view that it is better for a material to relieve stresses by viscous flow (i.e. have a low value of G' and thus a high value of tan  $\delta$ ) rather than through crack formation at low temperatures. Along the same lines, attempts have been made to determine the  $T_g$  of asphalt binders using transitions in the tan  $\delta$  versus temperature curve in correlation with the  $T_g$  of added polymers.

Goodrich [27] found that the loss tangent correlated well with the performance of an asphalt mix, and also found a correlation between asphalt binder rheology at 0.1 rad s<sup>-1</sup> and asphalt concrete creep. An oscillation frequency of 0.1 rad s<sup>-1</sup> was used in the present work for determining plots of G', G'' and tan  $\delta$ versus temperature for both this reason as well as for the fact that it is of the same order of magnitude as traffic loading frequencies. However, it should be noted that frequencies that are relevant to processes like thermal shrinkage could be much lower than 0.1 rad s<sup>-1</sup> depending on the cooling rate of the pavement. At present very little is known regarding the exact mechanism involved in thermal stress cracking of asphalt pavements.

Fig. 4 shows a plot of the storage and loss moduli, and loss tangent versus temperature for an unmodified 150-200 asphalt. In this graph there is a maximum in G'' as may be expected at low temperatures where a material's tendency for viscous flow diminishes. However, the maximum in this graph is rather broad



*Figure 3* Master curves for unmodified 85–100, Styrelf, Ecoflex and stabilized polythylene-modified binders.



Figure 4 Temperature sweep of G', G" and tan  $\delta$  for a 150–200 grade asphalt binder.

and it is therefore difficult to find an accurate glass transition from this measurement. With respect to the tan  $\delta$  curves, for the 150–200 asphalt there was no descernible transition, which follows from the fact that asphalt is a mixture of many components of varying structure and molecular weight. Moreover, a dissolved polymer will have the same effect on  $T_g$ , given by the rule of mixtures, regardless of its molecular weight. And if the polymer is insoluble, a two-phase system will occur in which case special toughening mechanisms may play an important role in the low-temperature fracture performance of the binders (i.e. enhanced shear yielding mechanisms due to stress concentration effects and crack-pinning or crackblunting mechanisms). Instead of relying on ill-defined glass transition measurements for predicting low-temperature performance, this work describes a test at low temperatures which actually fractures the material and measures the force required for fracturing the binder.

#### 4.2. Low-temperature fracture testing

Low-temperature fracture data for ground rubbermodified asphalt binders (with 90% confidence limits)



Figure 5 Effect of the interfacial grafting reaction on the fracture toughness of a rubber-modified 150–200 grade asphalt binder.  $(\Box, \Delta)$  ungrafted 40 mesh samples; ( $\diamond$ ) surface-grafted wet-ambiently ground, 40 mesh sample; ( $\bigcirc$ ) surface-grafted cryogenically ground, 40 mesh rubber-modified sample.

are given in Fig. 5 and in Table I. Various quantities of wet-ambiently and cryogenically ground rubber tyre in the 80 and 40 mesh size range were added to a Bow River 150–200 penetration grade asphalt. The samples were also reacted with minor amounts of liquid polybutadiene and 1 wt % sulphur. In addition, Table II gives the results for a series of differently sized ground rubber tyre samples tested at 7 wt % rubber, with and without surface reactive processing with 2 wt % liquid polybutadiene and 1 wt % sulphur.

Fig. 5 shows that the effect of the interfacial grafting reaction is most pronounced for the wet-ambiently ground sample in this 150–200 grade binder. Currently the reason for this difference is not clear. However, it is important to note that for paving applications it would be better to use the wet-ambient over the cryogenically ground material.

To distinguish between the effect of the dissolved from the grafted polybutadiene on the toughness, control samples of the 150–200 grade asphalt containing 2, 4 and 6 wt % liquid polybutadiene were also tested. The results for the effect of dissolved polybutadiene on the fracture toughness of 150–200 grade asphalt are given in Table III.

From these results, it can be concluded that for each 1% liquid polybutadiene added to the 150-200 Bow River asphalt, the fracture toughness increases by approximately 4.2 kN m<sup>-3/2</sup>. With this information, it is now possible to show the effect which the interfacial reaction between the polybutadiene and the rubber particles has on the fracture toughness of the binders. Fig. 6 gives the added benefit to the fracture toughness imparted by the crumb rubber, the liquid polybutadiene, and the interfacial reaction between the polybutadiene and the rubber particles, for 10, 20, 30 and 40 mesh cryogenically, and 80 mesh wetambiently ground samples. These results show that the interfacial modification is beneficial in all samples. However, the effect is largest for the coarse crumb rubber-modified binders. The unreacted 10 and

2588

TABLE I Fracture toughness of 80 mesh wet-ambiently ground rubber tyre-modified 150-200 Bow River asphalt

Ground rubber (wt %)	LPBD (wt %)	Modulus (GPa)	$K_{\rm Ic}$ (kN m <sup>-3/2</sup> )
0	0	$0.76 \pm 0.09$	64.9 ± 7.1
4	0	$0.79 \pm 0.05$	$77.3 \pm 5.2$
7	0	$0.68\pm0.07$	$96.0 \pm 4.1$
10	0	$0.53 \pm 0.04$	$120.1 \pm 3.2$
4	1	$0.87 \pm 0.04$	95.6 ± 4.1
7	1	$0.94 \pm 0.09$	$101.2 \pm 7.0$
10	1	$0.94 \pm 0.06$	$123.6 \pm 10.4$
4	2	$0.85 \pm 0.08$	$100.6 \pm 5.7$
7	2	$0.65 \pm 0.05$	$119.3 \pm 9.6$
10	2	$0.62 \pm 0.04$	$137.3 \pm 6.4$

TABLE II Fracture toughness of 10, 20, 30 and 40 mesh cryogenically ground rubber tyre-modified 150–200 Bow River asphalt binders

Ground rubber (wt %)	Mesh size	LPBD (wt %)	Modulus (GPa)	$K_{\rm Ic}$ (kN m <sup>-3/2</sup> )
0	_	0	0.76 ± 0.09	64.9 ± 7.1
7	10	0	$0.95 \pm 0.12$	$66.8 \pm 4.8$
7	10	2	$0.88 \pm 0.11$	$93.6 \pm 4.9$
7	20	0	$0.88 \pm 0.08$	$67.2 \pm 2.5$
7	20	2	$1.01\pm0.06$	$105.3 \pm 5.7$
7	30	0	$0.84 \pm 0.07$	$81.0 \pm 4.3$
7	30	2	$1.05\pm0.08$	$107.9 \pm 4.1$
7	40	0	$0.85\pm0.09$	$80.3 \pm 2.1$
7	40	2	$0.88\pm0.08$	104.4 ± 3.4

TABLE III Fracture toughness of liquid polybutadiene-modified150-200 Bow River asphalt binders

Polybutadiene (wt %)	Modulus (GPa)	$K_{1c}$ (kN m <sup>-3/2</sup> )
0	$0.76 \pm 0.09$	64.9 ± 7.1
2	$0.88 \pm 0.05$	$71.3 \pm 3.8$
4	$1.01 \pm 0.21$	84.9 ± 5.1
6	$0.95\pm0.11$	$90.4 \pm 6.2$

20 mesh crumb rubber-modified binders do not show any significant improvement in fracture toughness over a 150–200 Bow River control binder. However, when the interface is strengthened, the added toughness is vastly improved. The added gain in fracture toughness for the interfacially modified versus the unmodified 10 mesh sample was nine-fold. For the reactively processed 20 mesh sample, the toughness increase due to surface modification of the crumb rubber was as much as 15 times higher than for the unreacted sample. For the 30 mesh sample, the added benefit in fracture toughness due to the interfacial modification was 200%, for the 40 mesh sample it was 100% and for the 80 mesh sample it was still a respectable 50%.

As a comparison, a 20 mesh ambiently ground rubber tyre sample was mixed according to the wet process (20 wt % rubber mixed for 45 min at 190 °C). The fracture toughness for this material was determined to be 164.0  $\pm$  4.4 kN m<sup>-3/2</sup> which is higher than all other samples. However, some of the reacted 10% crumb



*Figure 6* Effect of the interfacial grafting reaction on fracture toughness of 150-200 grade binders modified with 7 wt % rubber. Black: rubber contribution; white: LPBD contribution; grey: contribution of the grafted interface.



Figure 7 Environmental electron micrograph of a fracture surface for an interfacially modified crumb rubber-asphalt system.

rubber samples come quite close to this level of toughness.

Figs 7 and 8 give environmental scanning electron micrographs of a surface-modified system and an unmodified system, respectively. From Fig. 7 it can be seen that the surface-modified system shows good interfacial adhesion between the asphalt and rubber phase. However, in Fig. 8 the unmodified system shows that sharp cracks surround the rubber particle, indicating a weak interface. These images combined with the fracture toughness data indicate that a crackpinning or crack-blunting mechanism is responsible for the major toughening effect in these systems. Substantial plastic flow dissipates fracture energy into heat when the particles are pulled out of the crack surface upon fracture.

An asphalt binder containing devulcanized rubber tyre was tested for its low-temperature fracture toughness. This asphalt has an 85-100 penetration grade and is therefore compared with a Bow River 85-100penetration grade control sample. The devulcanized sample was also reacted with 1 wt % sulphur at  $170 \,^{\circ}$ C for a period of 2 h in order to increase the molecular



Figure 8 Environmental electron micrograph of a fracture surface for an unmodified crumb rubber-asphalt system.

TABLE IV Fracture toughness of devulcanized rubber tyremodified asphalt binders

Asphalt	Additive	Modulus (GPa)	$K_{\rm Ic}$ (kN m <sup>-3/2</sup> )
85-100	_	1.35 + 0.11	44.1 + 3.9
Ecoflex <sup>a</sup> Ecoflex	10 wt % rubber 10 wt % rubber +	$1.14 \pm 0.17$	$55.8 \pm 6.1$
	1 wt % sulphur	$1.15\pm0.12$	$66.4 \pm 4.6$

<sup>a</sup> Bitumar Inc., Montreal, Quebec.

TABLE V Fracture toughness of other polymer modified asphalt binders

Binder	Additive	Modulus (GPa)	$K_{\rm Ic}$ (kN m <sup>-3/2</sup> )
85-100	_	1.35 + 0.11	44.1 + 3.9
Stvrelf <sup>a</sup>	4 wt % SBR	$0.71 \pm 0.03$	86.1 + 8.8
EB-330 <sup>b</sup> 150–200	5.5 wt % EVA 4 wt % PE +	$1.46 \pm 0.09$	$91.37 \pm 5.4$
	1 wt % LPBD	$1.47 \pm 0.21$	$84.3\pm3.3$

<sup>a</sup> McAsphalt Industries Ltd., Scarborough, Ontario.

<sup>b</sup> Imperial Oil Ltd, Sarnia, Ontario.

weight of the devulcanized rubber. As is shown in Table IV, the fracture toughness of the devulcanized sample is only marginally better than that of the 85–100 penetration grade control sample. Reacting the devulcanized rubber with sulphur increases the fracture toughness by about 18% but the performance is still far below that of the (vulcanized) crumb rubber-modified binders. However, devulcanized rubber may be an ideal material to be used in the interfacial reaction between crumb rubber and asphalt. The low price of this material makes it a much more attractive reactive agent than the (expensive) liquid polybutadiene used in this work.

The results for the commercially available SBR, EVA and polythylene-modified asphalts are summarized in Table V.

#### 5. Conclusions

The high-temperature rheological properties of a 150–200 penetration grade asphalt can be greatly

enhanced through the addition of minor amounts of ground rubber tyre. The degree of deformation resistance was found to increase with increased rubber loading and smaller particle size. It was also found that these rubber-modified asphalt binders perform as well as, or better than, commercially available polymer-modified asphalts.

A commercially available binder containing 10 wt % devulcanized rubber which had undergone an *in situ* sulphur revulcanization reaction, performed equally well at high temperatures as the 10% 80 mesh rubber-modified sample.

Plain 10 or 20 mesh crumb rubbers do not significantly improve the low-temperature fracture toughness of a 150–200 penetration grade Bow River binder at additive contents as high as 10 wt %. However, the gain in fracture toughness due to interfacial modification in such systems can be very large.

Plain 30, 40 and 80 mesh crumb rubber tyre samples do increase the fracture toughness of the binder to some extent. However, in these samples, an interfacial *in situ* grafting reaction also gives substantial improvements over unmodified mixtures. The data suggest that a crack pinning or crack-blunting mechanism is responsible for the large increase in fracture toughness that are found in these systems.

Whether the results obtained will translate into improved low-temperature thermal stress cracking resistance has to be investigated further with studies on asphalt–aggregate mixes.

The commercially available asphalt binder containing devulcanized rubber tyre showed only a marginal improvement in fracture toughness over an unmodified 85–100 control sample. However, increasing the molecular weight of the rubber through a sulphur revulcanization reaction did increase the fracture toughness of this binder by an additional 18%.

#### Acknowledgements

Joseph Hui is thanked for his assistance with the preparation of some of the asphalt binders containing crumb rubber tyre. Financial support from the Ontario Ministry of the Environment (Project ET367TT), the Ontario Centre for Materials Research, the Advisory Research Committee at Queen's University and the National Science and Engineering Research Council of Canada is gratefully acknowledged.

#### References

 C. H. MCDONALD, in "Highway Research Record 146" (Highways Research Board), (1966) pp. 1–16.

- M. R. PIGGOTT, W. NG, J. D. GEORGE and R. T. WOOD-HAMS, in "Proceedings of the Association of Asphalt Paving Technologists", Vol. 46 (1977) p. 495.
- 3. L. FLYNN, Roads Bridges, 12 (1992) 42.
- 4. W. MOORE, "Construction Equipment," February (Cahners, 1991).
- 5. T. KUENNEN, Roads Bridges January (1992).
- J. E. HUFFMAN, ASTM Special Technical Publication 724, (American Society for Testing and Materials, Philadelphia PA, 1980) p. 3–12.
- 7. A. BJORKLUND, in "Proceedings of the VII World Road Congress", Vienna (1979).
- H. B. TAKALLOU and R. G. HICKS, Transportation Research Record 1171 (TRB, National Research Council, Washington, DC, 1988) pp. 113–20.
- 9. J. L. MCQUILLEN, J. F. MEMBER, H. B. TAKALLOU, R. G. HICKS and D. ESCH, J. Transp. Eng. 114 (1988) 259.
- F. P. NARUSCH, "Alaska Experience with Rubberized Asphalt Concrete Pavements, 1979–1982", Alaska Department of Transportation and Public Facilities, Div. of Design and Construction, Central Region, Juneau, Alaska.
- 11. D. C. ESCH, Report no. FHWA-AK-RD-85-07 (FHWA, 1985).
- 12. D. ESCH, Transportation Research Record 860 (TRB, National Research Council, Washington, DC, (1992) pp. 5–13.
- H. B. TAKALLOU, "Evaluation of Mix ingredients on the Performance of Rubber Modified Asphalt Mixtures", Oregon State University, Oregon, 12 June 1988.
- M. HEITZMAN, in Transportation Research Board 71st Annual Meeting, 12–16 January 1992, Washington DC, Preprint 920549.
- 15. P. TARRICONE, Civil Eng. April (1993) 46.
- G. C. PAGE, B. E. RUTH and R. C. WEST, in Transportation Research Board 71st Annual Meeting, 12–16 January 1992, Washington, DC, Preprint 920452.
- P. E. JOSEPH and G. KENNEPOHL, "Trial Section with Polymer-Modified Asphalts on Highway 400", Report PAV-91-03, Ministry of Transportion of Ontario, Downsview, Ontario, June 1991.
- P. E. JOSEPH and J. H. DICKSON and G. KENNEPOHL, in "Proceedings of the Canadian Technical Asphalt Association" (1992), pp. 243–264.
- 19. F. R. DAVIS, US Pat. 4485201, 27 November 1984.
- 20. D. W. CAUSYN and K. THYS, World Pat. Appl. 92/21820.
- 21. ECOFLEX, "Paving the Way to a Better World", Company literature, Bitumar Inc., Montreal, Quebec (1993).
- 22. S. A. M. HESP, Z. LIANG and R. T. WOODHAMS, US Pat. Appl. 92-863734 and WO Pat. Appl. 93-07219.
- 23. Rheometrics Dynamic Analyser RDA II, Owners Manual (Rheometrics Inc., 1990) pp. 8-19 to 8-21.
- "Plane-Strain Fracture Toughness of Metallic Materials", ASTM Method E-399-90, (American Society for Testing and Materials, Philadelphia, PA, 1984).
- D. BROEK, "Elementary Engineering Fracture Mechanics", 3rd revised Edn (Martinus Nijhoff, The Hague, 1982) p. 171.
- 26. A. J. KINLOCH and R. J. YOUNG, "Fracture Behaviour of Polymers" (Elsevier Applied Science, New York, 1983) p. 93.
- J. L. GOODRICH, in "Proceedings of the Association of Asphalt Paving Technologists", Vol. 57 (1988) p. 116

Received 3 March and accepted 25 November 1994