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# Development of crumb rubber reinforced bituminous binder under laboratory conditions

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Bituminous binders are widely used in the construction of flexible pavements. However, in some applications, the performance of conventional binders is not considered to be satisfactory. Reinforcing these binders with selected polymers prevents premature failure of a pavement by improving the properties of the binder. Another source of reinforcement comes from crumb (ground) rubber produced from waste tyres. After they have been worn-out during their limited service life, millions of used tyres are discarded every year and are hauled to a dump. The fatigue resistance at temperatures below normal service temperatures (25°C), one of the key engineering properties of crumb rubber reinforced binders, has been found to be lower than that of neat binders. This paper is concerned with the development of a rubber reinforced binder. It was shown that the binder has the potential to be used as an all-weather wearing course in flexible roads, whilst at the same time recycling a considerable amount of waste rubber. 

## 1. Introduction

Bituminous binders are a class of materials which have been widely used in the construction of flexible pavements for a very long time. However, it is desirable to improve the performance of such binders to meet modern construction demands. Recently, it has been demonstrated that various types of additives or chemical modification can lead to improvements [1]. Combining bituminous binders with polymers enhances the elastic behaviour and hence improves the engineering properties of the binder, particularly the resistance to rutting (ability to rebound) at high temperatures whilst suppressing fatigue cracking at normal service temperatures [2, 3]. Rutting is the permanent (irreversible) deformation (in the form of ruts or corrugation) caused by horizontal or lateral displacements of the binder and mineral grains in a pavement under a shear stress, produced by a wheel load at a high pavement service temperature [4]. In New Mexico a small company called Tewa recently claimed to have produced a bituminous binder with improved deformation properties compared to conventional binders [5]. The binder is reinforced with recycled plastic from waste materials in everyday use. The use of crumb rubber (crumb-sized particles) from scrap tyres in pavement applications also provides an economic and alternative source of elastomeric materials. Today, the use of crumb rubber is also coupled with environmental concerns. For example, an average of 38 million tyres, a massive 400,000 tonnes, are scraped in the United Kingdom every year and disposal has proved very difficult because these materials do not disintegrate easily. Since April 2002 a new European Union directive is in force. It aims to ensure that motor manufacturers assume responsibility for dismantling and recycling their cars when they reach the end of their performance life [6]. The new directive is also expected to ban the admission of whole tyres to landfill by 2006 [7]. In the United States of America, is estimated that 3 billion tyres are currently found in landfills and stockpiles across the country and this is growing by approximately 200 million annually. Scrap tyres can cause health hazards. For example, colonies of aggressive, potential disease carrying mosquitoes can establish themselves: waste tyres are a favourite site for certain species to lay their eggs [8]. Scrap tyres can also cause fire hazards and are part of the solid waste management problem. Novel ways of utilisation of waste tyres include methods such as retreading and burning as fuel in cement kilns [9]. One solution is pyrolysis, that is the burning of whole tyres to generate electricity or, the cooking of tyres to produce gas, oil, carbon and steel. Burning tyres is not the ideal environmental solution, as it does not make any use of the polymeric components in the tyres and it requires a major infrastructure investment. Another way of reusing tyres is to process them into crumb-sized particles for use in the manufacture of carpet underlay, children's play areas and sports surfaces.

Crumb rubber is also used in cementitious materials [10–13]. Research since the 1960s involving the use of

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crumb rubber in the manufacture of mixes (bituminous binders + aggregates) in road applications in the United States of America is well documented [14, 15]. The production of crumb rubber reinforced binders in this country continues to increase rapidly. On average, the total amount of mix containing crumb rubber for pavement applications exceeds 1 million tonnes annually.

In Europe, crumb rubber reinforced binders and mixes for pavement applications have been developed and used extensively over the past 30 years or so. These binders are also used as the wearing course (uppermost layer) in roads to reduce traffic noise [16] and improve durability.

However, crumb rubber is used only as an elastomeric filler in binders. This is because a small percentage of the three dimensional network of the vulcanised rubber [17] can be dissolved in the bituminous binder. Most of the rubber remains intact, as a somewhat loosened rubber filler swollen by the binder's oils [2].

To prevent fatigue cracking a softer (for example, 100 or 200 penetration grade [18]) bituminous binder may be used at the expense of possibly severe rutting on hot summer days. Use of a harder binder (for example, 50 penetration grade) may prevent rutting in summer but will result in poor fatigue at lower temperatures. Adding crumb rubber as an elastomer to this hard binder may, if applied properly, improve the fatigue performance. This requires that all the rubber is finely dispersed, but it cannot occur unless the three dimensional rubber network is broken. This process is known as the "devulcanization of the rubber" and a summary of the numerous methods used [19] and the mechanisms involved [20] may be found in the literature. The devulcanized rubber becomes soft and can be reprocessed and reshaped into new rubber articles when blended with virgin rubbers in quantities up to 40%. Devulcanized rubber can also be incorporated into bituminous binders, as the short literature review below demonstrates. Following the review, the materials used to include the rubber treatment methods, the experimental procedure and the results and discussion are presented. Finally, the main conclusions and plans for further work are made.

## 2. Background

- Chemically devulcanized waste rubber was used to reinforce binders [21]. Devulcanization of the rubber was carried out by refluxing the rubber crumbs in an organic solvent at high temperature (over 200°C) in the presence of a chemical devulcanizing agent. The results indicated that the devulcanized rubber reinforced binders investigated had the potential to give improved low temperature (between −10 and −20°C) performance.
- Pretreated crumb rubber was incorporated into binders for the preparation of mixes [22]. The rubber pretreatment product was a heavy paraffinic distillate solvent extract commonly used in the manufacture of tyre rubber (Hydrolene 90 extender oil). The results indicated that mixes made with the pretreated rubber binder had some potential for reducing thermal cracking (−18°C).
- A study was carried out to develop crumb rubber reinforced binders with improved engineering properties by the devulcanization of the rubber through use of heat, shear or a combination of both [2]. The results showed that the treated rubber improved the properties of mixes.
- A reinforced binder with improved low temperature (−20°C) fracture properties was obtained by devulcanizing the crumb rubber [23]. The rubber was mixed with crude oil in a pot at a temperature between 280 to 300°C. Oil was slowly distilled off the mixture until all the volatile low molecular weight hydrocarbon fractions of the crude oil were removed. The pot residue from the distillation, containing the devulcanized rubber, was mixed with a bituminous binder to give a homogeneous composition. Low temperature testing results showed an increase in fracture toughness and fracture energy [24] of the binders reinforced with the devulcanized crumb rubber. It was therefore concluded that binders with higher fracture toughness performed better in a pavement, which is under stress from traffic induced loads and thermal contraction during summer and winter months.
- A simple method for treating crumb rubber for use in binders has been developed [25]. According to this method, a slurry of crumb rubber was formed by adding water. The slurry was heated to a temperature of 85–90°C for a period of at least 5 minutes to release the excess oils and chemicals from the rubber particles into the water. The slurry was next dried to produce a fine mesh rubber product. The results showed an improvement in the rheological properties of the reinforced binders investigated.
- Another method of treating crumb rubber for use with bituminous binders has been developed [26]. The method provides a procedure for partially devulcanizating waste rubber. The crumb rubber was mixed with an aromatic oil under the application of heat (at a temperature of up to 250°C) and shear. The treated product comprised 65% of rubber and 35% of oil. Sulphur was added in an amount of 1% of the amount of treated rubber. The resulting cross-linked product was mixed with binders. It was found that the elastic properties of the reinforced binders were improved.

In the present work, crumb rubber from waste tyres was treated under laboratory conditions using two industrial methods recently reported in the literature [25, 26]. The treated product was incorporated into a 50 penetration grade binder at a concentration of up to 30% by weight of the neat binder. The fatigue characteristics of the reinforced binders at 10°C were investigated and compared using a method recently reported in the literature [27]. The aim was to produce a binder suitable for use as an all-weather wearing course in flexible roads and to meet environmental concerns.

## 3. Materials

The materials selected were a hard (50 penetration grade) bituminous binder from Kuwait and crumb rubber (produced by an ambient grinding process) from truck tyres (natural rubber) of 300 $\mu$m maximum particle size. For the preparation of control specimens (the unreinforced material), the binder (approximately 750 g in weight) was heated in a container to 180°C. Sufficient amount of the material (approximately 100 g) was then collected and stored in a fridge for use as specimens (each specimen was approximately 0.125 g). For the preparation of untreated reinforced specimens, the untreated (as received) particles were added to the binder in the container at a concentration of 10% by weight of the neat binder and mixed at 180°C using a Silverson high shear mixer for 1/2 hour at a rate of 2000 rpm (revolutions per minute). The same amount of the material was then collected and stored in a fridge. The same procedure was followed for the preparation of the treated reinforced materials for use as specimens after the following rubber treatment methods.

### 3.1. Water activated method

- A quantity of distilled water was added to the particles to form a slurry [25]. The rubber/water ratio was 1:2. The slurry was heated to 90°C and heating continued for a further 20 minutes at this temperature. The addition of water and the elevated temperature caused the particles to expand which enabled excess oils, chemicals and some residual metals in the rubber particles to be released. This was observed by the change in the colour of the water. The slurry was next dried in an oven and the particles were ready for mixing with the binder. Mixing was carried out as in the case of the untreated rubber and at a concentration of 10% by weight of the neat binder.

### 3.2. Devulcanization method

- A quantity of rubber was treated at 180°C with an aromatic oil, "Strukdex 270", which was a similar oil to the one reported in the literature ("Suntex 790") [26]. For the processing of the crumb rubber in the laboratory, a modified method to that used in the industry [26] was followed: 310 g of rubber was mixed with 500 g of oil at 180°C using a domestic food mixer. Mixing of the rubber with the oil using a high shear mixer [26] was found to be impossible. The rubber was slowly added to the oil because it was difficult to mix. When all the rubber was added mixing at this temperature continued for a further 3 hours. The temperature of the mixture rose to about 250°C, as reported in the literature [26]. The mixture was then split into two equal parts of 405 g each. This was done because of the limitation in the sizes of the equipment and mixing facilities available in the laboratory. To each part, a further 310 g of rubber was added until the mixture contained in total 930 g of rubber and 500 g of oil. Thus, the final composition of the product was 65% of dissolved rubber and 35% of aromatic oil. Lastly, 4.65 g of sulphur was added to each part and mixed for 5 minutes using the domestic mixer, giving a total of 9.3 g of sulphur or 1% of the amount of treated rubber in the oil. The mixture was further mixed at 180°C for 1 hour. The final product was then stored at room temperature. Mixing with the binder was carried out as in the case of the untreated material and at concentrations of 5% (giving a rubber content of 3%), 10% (rubber content 6%) and 30% (18% rubber) by weight of the neat binder.

## 4. Experimental procedure

In an effort to develop a bituminous binder reinforced with crumb rubber from waste tyres with engineering properties at least equal to those of the neat binder, the fatigue properties of the binders at 10°C were investigated and compared. Preliminary creep/recovery tests (a measure of rutting resistance) at higher service temperatures [2] using a Bohlin dynamic shear rheometer (DSR II) have shown no significant differences between the (untreated and treated rubber) reinforced binders (up to 10% reinforcement) and the neat binder. It was therefore assumed that rutting performance of the reinforced binders was similar to that of the neat binder. The same rheometer (connected to a microcomputer) was used and the test procedure followed was based on a method reported in the literature to measure and assess the fatigue behaviour of bituminous binders [27]. According to the test method, subjecting a specimen of the binder to repetitive shear oscillation at low enough temperatures (for example, 10°C) in a shear rheometer generated a fatigue failure. Loss of strength or, in this case, decrease in complex shear modulus (a measure of stiffness) with time was obtained as a result of repeated application of strains or stresses at levels below those required to produce immediate failure. The fatigue life of binders could thus be obtained and compared. Such fatigue tests provided experimental evidence showing the impact of binders on mix fatigue behaviour without the need to prepare mix samples [27]. It should be noted that fatigue behaviour strongly depends on pavement structure.

Similar fatigue tests were therefore carried out for this study at a frequency of 10 Hz and at a testing temperature of 10°C (these are typical laboratory fatigue testing conditions of mixes using beams as specimens). Plate-plate geometry (4 mm radius) was chosen (the specimens were placed between the plates) with a 2 mm experimental gap to eliminate machine compliance errors [27]. Tests were performed by applying continuous oscillatory shear loading under either constant strain (0.015) or constant stress ($3.5 \times 10^5$ Pa) conditions. Note that the rheometer can only apply oscillatory shear loading. One of the reasons the rheometer was selected is the lack of a convenient fatigue test for bituminous binders. Data produced in this study showed that this type of loading was able to generate fatigue failure. The time needed for a specimen to fail was taken as the fatigue life. This was observed directly by the rapid decrease in the values of the complex shear modulus from plots of the complex shear modulus against time in logarithmic form. The plots were displayed on the monitor and stored by the microcomputer. The data produced was used to compare the fatigue life of the binders and assess the influence of the rubber treatments.

## 5. Results and discussion

Table I summarises the fatigue results for all the binders investigated in terms of the complex modulus and the corresponding fatigue lives. Figs 1 to 4 show typical fatigue results in logarithmic form.

### 5.1. Controlled strain tests

Figs 1 and 2 show typical fatigue data obtained as plots of the complex shear modulus against time in logarithmic form. In Fig. 1, reinforcing the binder with 10% (by weight of the neat binder) untreated rubber leads

TABLE I Summary of complex shear modulus (a measure of stiffness) against fatigue life (10°C, 10 HZ) for all binders under controlled strain (0.015) and controlled stress ($3.5 \times 10^5$ Pa) conditions

| | Controlled strain (0.015) | |
|---|---|---|
| Material | Modulus (Pa) | Fatigue life (sec) |
| Neat (unreinforced) binder | $4 \times 10^6$ | 52900 |
| Neat (unreinforced) binder | $1 \times 10^7$ | 40128 |
| Untreated 10% rubber | $1 \times 10^7$ | 26476 |
| Water treated 10% rubber | $1 \times 10^7$ | 39255 |
| Devulc 5% | $1 \times 10^7$ | 45800 |
| Devulc 10% | $1 \times 10^7$ | 35565 |
| Devulc 30% | $4 \times 10^6$ | 43300 |
| | Controlled stress ($3.5 \times 10^5$ Pa) | |
| | Modulus (GPa) | Fatigue life (sec) |
| Neat (unreinforced) binder | $1 \times 10^7$ | 43840 |
| Untreated 10% rubber | $1 \times 10^7$ | 6045 |
| Water treated 10% rubber | $1 \times 10^7$ | 10400 |
| Devulc 5% | $1 \times 10^7$ | 32200 |
| Devulc 10% | $1 \times 10^7$ | 26597 |

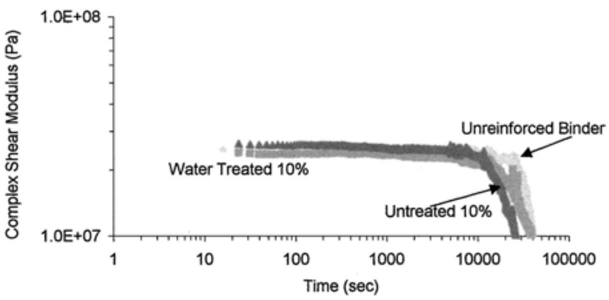


*Figure 1* Typical fatigue behaviour at constant strain of 0.015, 10 Hz and 10°C: bituminous binder reinforced with 10% (by weight of the neat binder) untreated and treated in water crumb rubber (300 $\mu$m maximum size) from truck tyres.

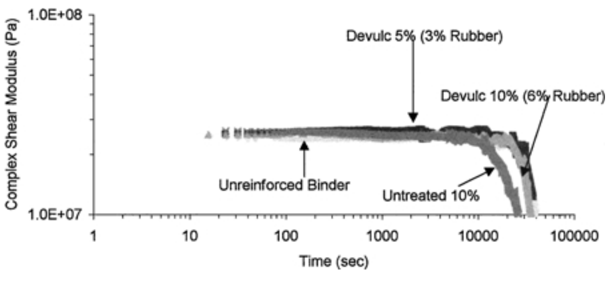


*Figure 2* Typical fatigue behaviour at constant strain of 0.015, 10 Hz and 10°C: bituminous binder reinforced with up to 10% (by weight of the neat binder) devulcanized product and 10% untreated crumb rubber.

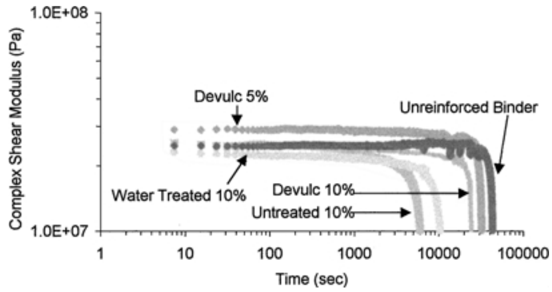


*Figure 3* Typical fatigue behaviour at constant stress of $3.5 \times 10^5$ Pa, 10 Hz and 10°C: bituminous binder reinforced with up to 10% (by weight of the neat binder) devulcanized product, 10% water treated and 10% untreated crumb rubber.

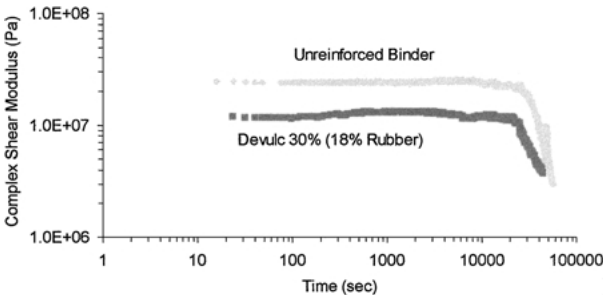


*Figure 4* Typical fatigue behaviour at constant strain of 0.015, 10 Hz and 10°C: bituminous binder reinforced with up to 30% (by weight of the neat binder) devulcanized product (18% crumb rubber content).

to lower fatigue life compared with that of the unreinforced (neat) binder. However, if the rubber is treated with water before mixing, the fatigue life of the reinforced binder becomes similar to that of the neat binder. Fig. 2 demonstrates that rubber devulcanization leads to a development of a binder with fatigue performance equivalent to that of the neat binder.

### 5.2. Controlled stress tests

Fig. 3 shows typical data obtained for the various binders as plots of the complex shear modulus against time in logarithmic form. It can be seen that fatigue performance was significantly improved, with respect to the neat binder, and in the following order: devulcanized 5% > devulcanized 10% > water treated > untreated binder. Clearly these results demonstrate that some form of rubber treatment can lead to improvement in fatigue lives and that the devulcanized binders performed better. However, the binders were subjected to severe stress conditions, as explained below, and therefore it is not possible to give any possible reasons for the order obtained.

### 5.3. Preliminary tests with higher rubber content

In constant stress tests, the applied (very high level of) stress remained constant as the stiffness (complex shear modulus) continuously decreased. The specimens were therefore subjected to severe stress conditions during testing. In a similar way, a constant strain applied to the specimens resulted in the stress being continuously decreased with decreasing stiffness, a less severe condition [27]. This difference in the nature of the two types of fatigue tests may explain the difference between the data in Figs 1–3. The stress level in the constant stress tests could have been reduced, but this would have resulted in longer fatigue times. For the purpose of this work, it was not possible to occupy the rheometer for longer periods. The controlled strain condition was therefore thought to be a more realistic and appropriate type of fatigue testing for the rapid evaluation of the binders.

Fig. 4 shows data obtained under controlled strain conditions for the binder reinforced with 30% by weight of the neat binder (18% devulcanized rubber) as a plot of the complex modulus against time in logarithmic form. It can be seen that similar fatigue life with that of the neat binder was achieved but at lower complex modulus values, which is better for improved resistance to low temperature (below 0°C) cracking. However, this may have an effect on rutting performance and further work is envisaged. Nevertheless, the reinforced binder developed has the ability to resist fatigue cracking at 10°C as well as the unreinforced binder, whilst at the same time a considerably amount of waste can be recycled.

## 6. Conclusion

Polymers are currently used to enhance the elastic behaviour of bituminous binders for use in flexible pavements. Reinforcing the binders with crumb rubber from waste tyres offers an economic and alternative source of elastomeric materials coupled with environmental benefits. However, the resulting binders have been found to have lower fatigue resistance compared with that of the unreinforced (neat) binders at service temperatures below 25°C. Selecting a suitable rubber treatment method can lead to a significant increase in fatigue resistance of the reinforced binders. Two such methods used in the industry have been selected here for the purpose of developing a rubber reinforced binder under laboratory conditions whose performance at least equals that of the neat binder. It should be noted these industrial methods are still in experimental stage and the associated cost implications of the additional treatments is not yet known. One of the methods was modified to enable the development of a binder reinforced with up to 18% crumb rubber by weight of the neat binder. A large amount of waste from tyres can thus be disposed of without significantly changing the performance of the binder. The binder developed has the potential to be used as an all-weather wearing course in flexible roads at service temperatures at least as low as 10°C. This is the first time such findings are reported in the literature. Further work at lower temperatures is envisaged.

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