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# 1 **Experimental study on the optimisation of chemical treatment to** 2 **reduce waste rubber aggregates absorption properties**

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## 10 **Abstract**

11 Utilisation of abundant waste rubber generated from scrap tyres in pavement engineering applications could be a  
12 useful route to consume large extent of the waste from the scrap tyres. It may contribute towards maintaining a  
13 greener environment by reducing the associated environmental and social impacts as well as improve the  
14 properties of bituminous mixture used for pavement engineering applications. This paper focuses on the  
15 upgrading of waste rubber aggregates surface properties by identifying and applying optimised chemical  
16 treatment (oxidation and cross-linking). The focus was to increase the life span of bituminous mixtures  
17 containing waste rubber as an aggregate by reducing a phenomenon of migration of light fractions (maltenes)  
18 from bitumen to rubber which occur at elevated temperatures ranging between 160-180°C. The results of various  
19 tests conducted showed the potential of the treatment to reduce the absorption of maltenes into rubber. The  
20 existence of the surface treatment was verified through the Scanning Electron Microscope (SEM) analysis of the  
21 rubber aggregate. The reduction in the absorption properties of rubber due to the existence of the treatment was  
22 verified using the Microwave Spectroscopy technique. The results of both the tests were promising in terms of  
23 indicating the durability of the treatment and the reduction in the absorption properties of rubber. Mechanical  
24 tests such as Indirect Tensile Stiffness Modulus (ITSM) and Repeated Load Axial (RLA) tests were conducted  
25 on the newly devised bituminous mix design with 10% of added rubber (by mass of the sample). The results  
26 were promising in the case of the aged samples with both the stiffness and load bearing capacity being higher  
27 for the aged samples. However, the results need improvement in terms of its applications on the light traffic  
28 areas through the replacement of the ordinary filler with the mineral filler. Also, a higher percentage of waste  
29 rubber should be added to study the suitability of its use in the flexible surface applications such as children's  
30 playgrounds, sports pitches and surfaces, etc.

31 **Keywords:**

32 Waste rubber; scrap tyres; pavement engineering; sustainable environment; chemical treatment; microwave  
33 analysis; bituminous mix; bitumen analysis; mechanical testing.

34 **1. INTRODUCTION**

35 **1.1. Scrap tyres, their environmental impact and applications**

36 The waste rubber from scrap tyres poses an environmental threat if not utilised efficiently. Since 2006  
37 legislation such as End of Life Vehicles (ELV) salvage, waste incineration and EU directives on the waste  
38 landfill management prohibited the disposal of waste tyres arising to the landfill and similar waste disposing  
39 routes, etc. [1, 2]. Also, stockpiling of the scrap tyres is not allowed because of it being a potential source of fire  
40 hazard, cause of environmental damage and other health risks associated with it. It also does add to the disposal  
41 costs of the scrap tyres [3-5]. In Europe 95% of the scrap tyres are recycled [6]. The emphasis on recycling the  
42 scrap tyres encouraged the introduction of new end use market applications as well as alternative recovery  
43 options. Although the market for scrap tyres and its material usage has been established, the amount of tyres that  
44 reach their end-of-life cycle worldwide significantly exceeds the end use market of the scrap tyre material. The  
45 only exception to this is the UK market of scrap tyres as it is meeting 100% of the recovery target through  
46 various routes such as recycling, recovery and reuse [1].

47 One of the potential applications to utilise a significant number of scrap tyres is to use them in rubberised  
48 asphalt pavements surfaces, pavement engineering and flexible rubberised surfaces whereby large quantities of  
49 scrap tyres can be consumed. This could potentially reduce the amount of waste generated and can improve  
50 some of the engineering properties of the pavements such as fatigue resistance, reduced low temperature surface  
51 cracking, improve tensile strength, adhesion, resistance to rutting, elasticity, noise reducing characteristics,  
52 safety in wet conditions, flexibility to reduce injuries in the case of playground surfaces, and many more [4, 5,  
53 7-15].

54 **1.2. Brief problem statement and the literature**

55 Applications of waste rubber aggregates in bituminous mixtures, both in the wet and dry process, resulted in two  
56 main problems identified by researchers and professionals [4, 5, 7-9, 11, 16-19]. At elevated temperatures (140-  
57 170°C) the migration of light fractions (constitute resins, aromatics, and saturates) from bitumen to rubber  
58 occurs. This causes the swelling of rubber between 3-9 times its original size causing it to lose its rigidity and

59 shape. Also, the residual bitumen becomes brittle and hard resulting in the breaking of the bond between the  
60 rubber, bitumen and aggregates. This causes the particles of rubber to become loose and become distributed in  
61 the bitumen in the case of fine particles (wet process) and on the surface of the pavement leaving gaps on the  
62 surface in the case of crumb rubber (dry process). This adversely affects the performance and durability of the  
63 surface.

64 Shakhnazarli [20] has utilised secondary polyethylene, polyamide fiber waste, and crumb rubber to modify  
65 bitumen. Airey et al [7] pointed out the use of modifiers like sulphur, rubbers, thermoplastic polymers, and  
66 thermosetting resins to modify the bituminous mixture to improve mechanical properties of the bituminous mix.  
67 Stabilising agents such as cellulose and mineral fibers have been used to prevent the drain down [21]. Polymers  
68 such as styrene-butadiene-styrene (SBS), styrene-butadiene rubber (SBR), Elvaloy, ethylene vinyl acetate  
69 (EVA), polyethylene, and others have also been used to modify asphalt binders [22, 17]. Frantzis [23] discusses  
70 methods to modify rubber including a devulcanization process, heavy paraffinic distillate solvent extract and a  
71 water activated method. Additives like Poly phosphoric acid and vestenamer were also used for improving the  
72 properties of crumb rubber modified bitumen [24]. Although these treatments, up to an extent, improve the  
73 performance in terms of mechanical behaviour and engineering properties (resistance to deformation, low  
74 temperature cracking, flexibility, stiffness, etc.) of bitumen and bituminous mixtures that ultimately has an  
75 influence on the service life of pavements, the problem of migration of maltenes has not been explicitly  
76 addressed by any of the above.

### 77 **1.3. Waste rubber aggregates treatment**

78 This paper demonstrates the use of suitable chemical additives and their optimised quantities to modify the  
79 rubber surface properties to subsequently reduce the migration of maltenes from bitumen to rubber. This could  
80 potentially result in a strong adhesive properties of rubber which subsequently helps in improving the bonding  
81 between the bitumen and rubber. If successful in reducing the migration of light fraction from bitumen to rubber,  
82 the proposed treatment is also expected to improve the resistance to permanent deformation, reduce low  
83 temperature surface cracking, and improve the flexibility of the material over a long period of time for the  
84 applications such as use in school playgrounds and sports stadiums, shock absorbing surfaces,  
85 walkways/pathways, artificial turf for football stadiums. Additionally, it could have potential to be used on light  
86 traffic road surfaces [4, 5, 7-9, 11, 13, 16-19] (Al-Nageim and Robinson 2007; Rahman et al 2003; Huang et al  
87 2007; Cao 2007; Navarro et al 2005; Lee et al 2008; Rahman et al 2005; Airey et al 2002; Read and Whiteoak

88 2003; Xiao et al 2009; Hernández-Olivares et al 2009). A Microwave spectroscopy technique was used to  
89 analyse the absorption properties of rubber through indirectly studying the properties of bitumen and changes  
90 incurred in the spectrum of bitumen after its interaction with the treated rubber. This technique has been  
91 demonstrated in previous studies by the same author as this article [25, 26] to analyse bitumen properties. The  
92 mechanical properties were tested using the Indirect Tensile Stiffness Modulus (ITSM) test and Repeated Load  
93 Axial (RLA) test.

94 Ateeq et al [25] and Ateeq et al [26] attempted to carry out the chemical treatment, however, the optimum  
95 chemical treatment parameters were not achieved. This study extends the work by identifying the optimum  
96 chemical treatment (percentage of oxidation and cross-linking agents). It also presents the mechanical testing of  
97 the bituminous samples prepared in the laboratory with the treated rubber and its results. Despite the attempt to  
98 identify and explore optimum chemical treatment in the current study, further work is required to improve the  
99 mechanical behaviour from the test results as well as to optimise the chemical treatment in large size samples.  
100 This can either be achieved at the chemical treatment stage (through other quantities of chemical treatment  
101 systems), at the time of manufacturing the laboratory test samples and/or through field trials. More detailed  
102 testing is also required including the use of more established analysis techniques to study the migration of light  
103 fractions from bitumen to rubber.

## 104 **2. MATERIALS AND METHODS**

### 105 **2.1. Materials**

106 The crumb rubber utilised to carry out the treatment was 1-6 mm size aggregates from truck scrap tyres. The  
107 size range of the sample was obtained using a set of sieve by passing through a 6 mm sieve and retaining on 1  
108 mm particle size sieve. It was supplied by J. Allcock & Sons based in Manchester, UK. The sample of waste  
109 rubber is shown in Figure 1 and its physical properties are presented in Table 1.



110

111 Figure 1. Crumb rubber from scrap tyres of the size 1-6 mm used in the study using 1-6 mm sieve set

112

113 Table 1: Physical properties of coarse waste rubber aggregates

Properties	Value
Bulk specific gravity, gm/cm <sup>3</sup>	2.79
Apparent specific gravity, gm/cm <sup>3</sup>	2.82
Water absorption, %	0.4
Dielectric constant (20°C)	8.0

114

115 The bitumen was 100/150 penetration grade (pen) supplied by Nynas UK AB. The typical properties of the  
 116 bitumen used in the study is shown in Table 2.

117 Table 2: Properties of 100/150 pen bitumen used in the study

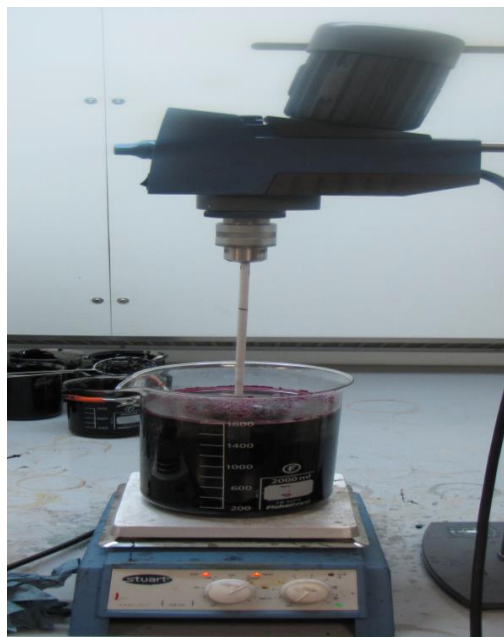
Parameters	Bitumen (100/150 pen) values
Penetration at 25°C	100-150x0.1 mm
Softening point	39-47°C
Penetration index	-1.5 to +0.7
Dynamic viscosity at 60°C	NPD
Fraass breaking point	≤ -12
Dielectric constant (20-25°C)	2.55-3.5

118

119 The chemicals used for the oxidation were potassium dichromate,  $\geq 99.5\%$  supplied by Sigma Aldrich and  
120 potassium permanganate, 99% supplied by BDH Laboratory, England. The cross-linking agent used was  
121 polyethyleneimine supplied by Sigma Aldrich. Petroleum (petrol special) with aromatic basis (~18%, bp. 180-  
122 220°C) was supplied by Sigma Aldrich that was used to replicate the interaction effect of bitumen (because of  
123 their similar properties). This was to analyse the polyethyleneimine coating and its durability in petroleum.

## 124 2.2. Design of the chemical treatment

125 • The modification of waste rubber surface was a two stage process. In the first stage, *oxidation* of the crumb  
126 rubber was carried out. It was followed by the *cross-linking* of the rubber in the second stage. The most  
127 important factor in the current study was to optimise the chemical treatment and to identify the combination  
128 of the oxidation and cross-linking agents and their suitable percentages to obtain the treated rubber to be  
129 used in the preparation of the bituminous mix design. It follows the previous work carried out by Ateeq et al  
130 [25] and Ateeq et al [26]. From the detailed combination of chemical treatments and their analysis the most  
131 suitable and effective treatment identified was as follows: Waste rubber aggregates were treated with the  
132 oxidising solution comprising 0.1 mol/L of potassium dichromate ( $K_2Cr_2O_7$ ), 0.5 mol/L of sulphuric acid  
133 ( $H_2SO_4$ ) and water ( $H_2O$ ). The oxidation was carried out at 60-70°C for 90 minutes to accelerate the  
134 oxidation reaction on the hot plate and thereafter kept at room temperature for 24 hours for further reaction  
135 to take place. The process is shown photographically in Figure 2.



136

137

Figure 2. Oxidation of the waste rubber using potassium dichromate, sulphuric acid and water

- 138 • The oxidised rubber was then cross-linked using 3% of the polyethyleneimine in the rubber-bitumen blend.  
139 The blend was mixed for one hour and further cured for 6 hours at 160-170°C in an oven.

### 140 **2.3. Testing and analysis**

141 Two types of analyses were carried out to confirm the sustainability, durability and effectiveness of the chemical  
142 treatment. They included a surface analysis as well as an absorption analysis of rubber. The surface analysis of  
143 the treated waste rubber was carried out using a Scanning Electron Microscope (SEM). It was important to  
144 obtain evidence of the existence of the cross-linking agent (polyethyleneimine) on the surface of the treated  
145 waste rubber aggregates after the treatment at high temperature (160-170°C). The key factor was the existence  
146 of the thin film coverage on the rubber surface and to examine if the harsh temperature environment still left the  
147 thin film intact during and after the treatment. Since it was difficult to analyse the surface of the treated rubber  
148 out of the rubber-bitumen blend due to the bitumen coating on rubber particles, a suitable and alternative  
149 approach was designed. Petroleum (petrol special) was used to simulate the effect of bitumen. The oxidised  
150 rubber was mixed with petroleum and heated to 160-170°C. For the purpose of this paper, the results of two  
151 samples treated with polyethyleneimine was presented and compared with the untreated rubber sample. Cross-  
152 linking agent at 6% and 9% by weight of the rubber was added in the blend and mixed thoroughly for 1 hour to  
153 produce the uniform coating on the rubber surface. The blend was further mixed for additional 2 hours until all  
154 the petroleum evaporated. Rubber particles from the blend were then analysed under SEM.

155 The analysis of the absorption properties of rubber was carried out using the Microwave Spectroscopy (MS)  
156 analysis technique. The technique has been proven effective, accurate and repeatable in the analysis of the  
157 absorption properties of bitumen when it interacts with rubber [25, 26]. This was achieved through the indirect  
158 analysis of the bitumen taken from the rubber-bitumen blends with and without oxidised and/or cross-linked  
159 rubber. The aim was to monitor the surface resistance of the rubber to bitumen by indirectly measuring the  
160 changes in the bitumen properties extracted from the blend. Based on the theory of the microwave analysis  
161 technique it was assumed that if the treatment was effective the spectra obtained from the microwave analysis of  
162 the bitumen would be similar or close to the pure bitumen curve because of the least change having taken place  
163 in the dielectric properties of bitumen. This is because the two bitumen samples will have almost identical  
164 percentage of light fractions and will exhibit similar properties. Otherwise the spectra would shift away from the  
165 pure bitumen sample result. For results to be accurate and repeatable, the bitumen samples were kept in a  
166 temperature controlled chamber for 24 hours before conducting the analysis. Furthermore, the weight of the



167 bitumen samples to be analysed need to be precisely the same to produce accurate results because of the  
168 instrument sensitivity.

169 The samples were analysed in the frequency range of 2.329-2.347 GHz. This is because of the effective  
170 response of the physical properties of bitumen at this band of frequencies. The results analysed were for the  
171 reflected power also known as reflection coefficient ( $S_{11}$ ) due to the detailed and accurate information available.  
172 The samples were repeated twice to prove the validity of the results.

#### 173 **2.4. Sample preparation for microwave spectroscopy analysis**

174 The aim of the surface treatment of coarse waste rubber was to use it as an aggregate replacement in the  
175 bituminous mix (dry process). However, to study the absorption properties of waste rubber (treated & untreated),  
176 blends of rubber-bitumen were prepared in a similar fashion as in the wet process where the fine rubber is mixed  
177 with the bitumen and heated. This approach was adapted to study and confirm the direct effect of the treatment  
178 on the surface of waste rubber and to analyse the impact in terms of absorption of bitumen into rubber through  
179 its surface. Treated and untreated rubber was mixed at 10% by weight to 100 mL of the bitumen sample at  
180 medium shear strain to bring a solution to a continuous phase. The mixing was carried out for an hour at 160-  
181 170°C. It was then followed by further 6 hours of curing for the maximum absorption of light fractions from  
182 bitumen into rubber. After the blend was ready, bitumen was extracted off the blend into the sample tube (15  
183 mL) to be analysed using the microwave spectroscopy analysis technique.

184 The experimentation was carried out in a temperature controlled chamber and the temperature was set at 20°C.  
185 All the samples were kept in the chamber for 24 hours to stabilise the temperature before any analysis tests were  
186 conducted.

#### 187 **2.5. Microwave spectroscopy analysis theory**

188 Microwave analysis monitors the changes in the molecular structure of the material under consideration via  
189 measuring the changes in the permittivity of the material. Any changes in the molecular structure of the material  
190 affects its permittivity. This change in the permittivity of the material can be captured in the form of microwave  
191 spectra when the microwaves interact with the material [27-29]. These response signals could be in the form of a  
192 reflected power, also known as the reflection coefficient or  $S_{11}$  parameter as well as the transmitted power, also  
193 referred to as the transmission coefficient or  $S_{21}$  parameter [30]. Collectively, they are termed as S-Parameters  
194 (scattering parameters) [31]. Permittivity simply measures the response of the material to the applied  
195 microwaves in the form of a change in the complex electric permittivity, termed as  $\epsilon_r$ , and is defined as, [31]:

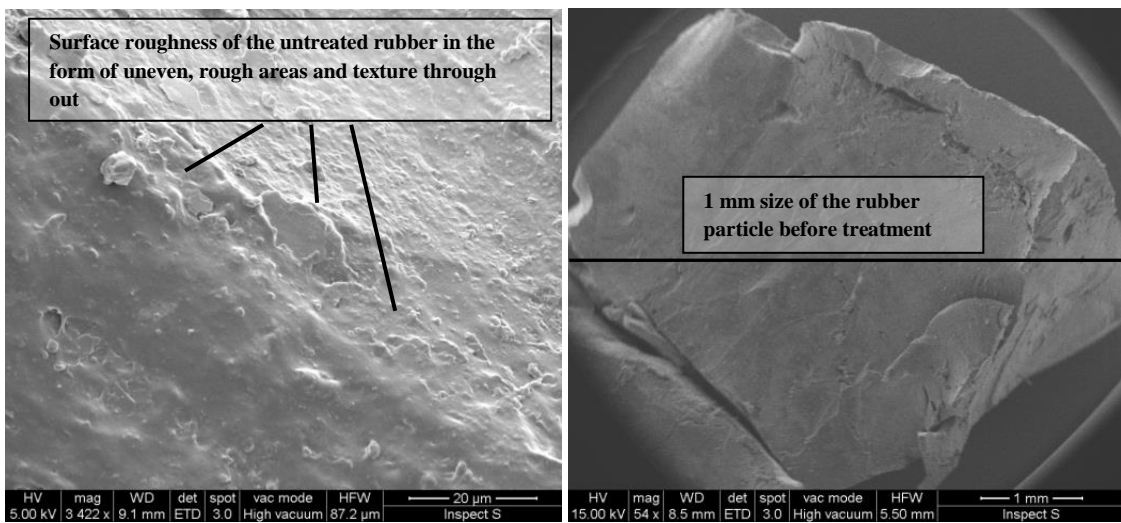
196  $\epsilon_r = \epsilon' + j\epsilon''$  (1)

197 Where  $\epsilon'$  represents the energy stored by the material and  $\epsilon''$  represents any losses of the energy. Measuring the  
 198 influence of the material on the s-parameters at discrete frequencies one can relate it to various material  
 199 characteristics such as material composition, its type, concentration, size/size distribution, etc. in the sample  
 200 under test [31].

201 **3. RESULTS AND DISCUSSION**

202 **3.1. Scanning Electron Microscope (SEM) surface analysis**

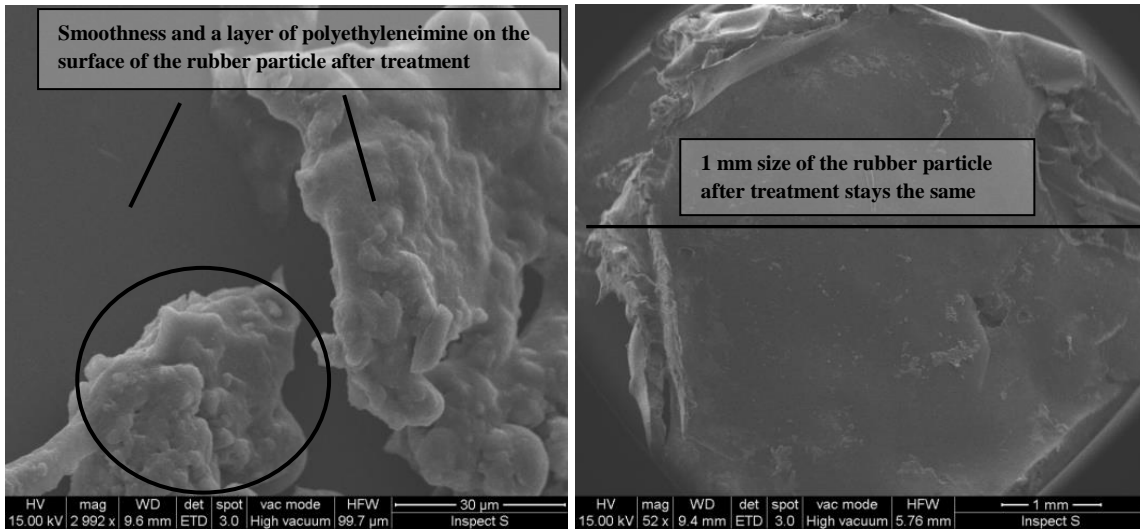
203 The results of the SEM analysis of the surface of the untreated and treated rubber samples is shown in Figure 3,  
 204 4 and 5 for comparison. The same particle size was chosen for a like-to-like comparison both in the case of  
 205 untreated and treated rubber which was about 1 mm. The surface treatment for the SEM analysis was carried out  
 206 as described in section 2.2.



207  
 208 (a)

(b)

209 Figure 3. (a) SEM analysis of the untreated crumb rubber surface at the magnification of 3422x (b) SEM  
 210 analysis showing the particle size of the untreated rubber to be approximately 1 mm



211

212

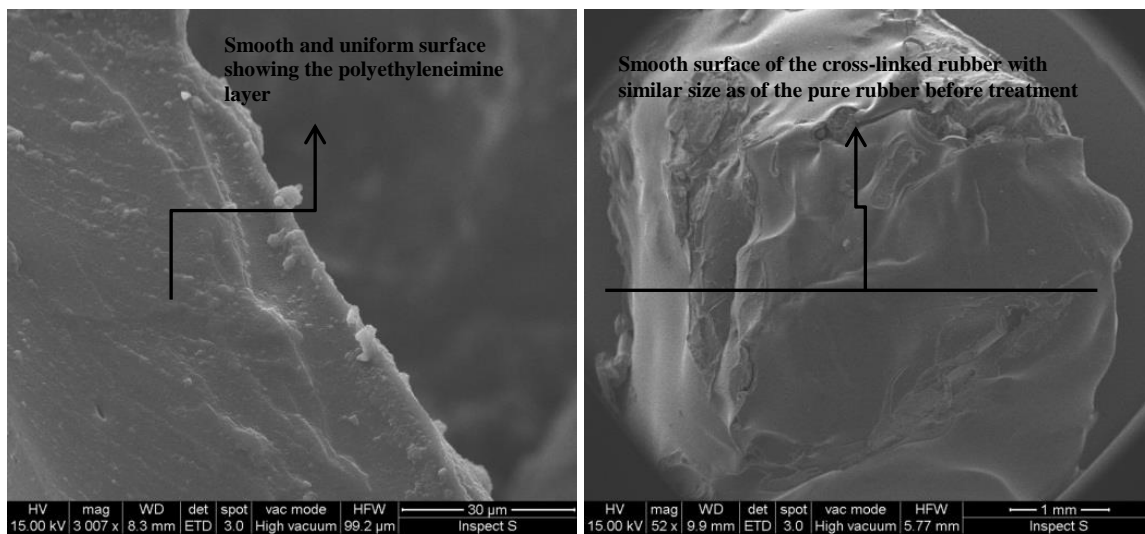
(a)

(b)

213

214

Figure 4. (a) SEM analysis of the 6% polyethyleneimine treated crumb rubber surface at the magnification of approximately 3000x (b) SEM analysis showing the particle size of the treated rubber to be approximately 1 mm



215

216

(a)

(b)

217

218

Figure 5. (a) SEM analysis of the 9% polyethyleneimine treated crumb rubber surface at the magnification of approximately 3000x (b) SEM analysis showing the particle size of the treated rubber to be approximately 1 mm

219

220

221

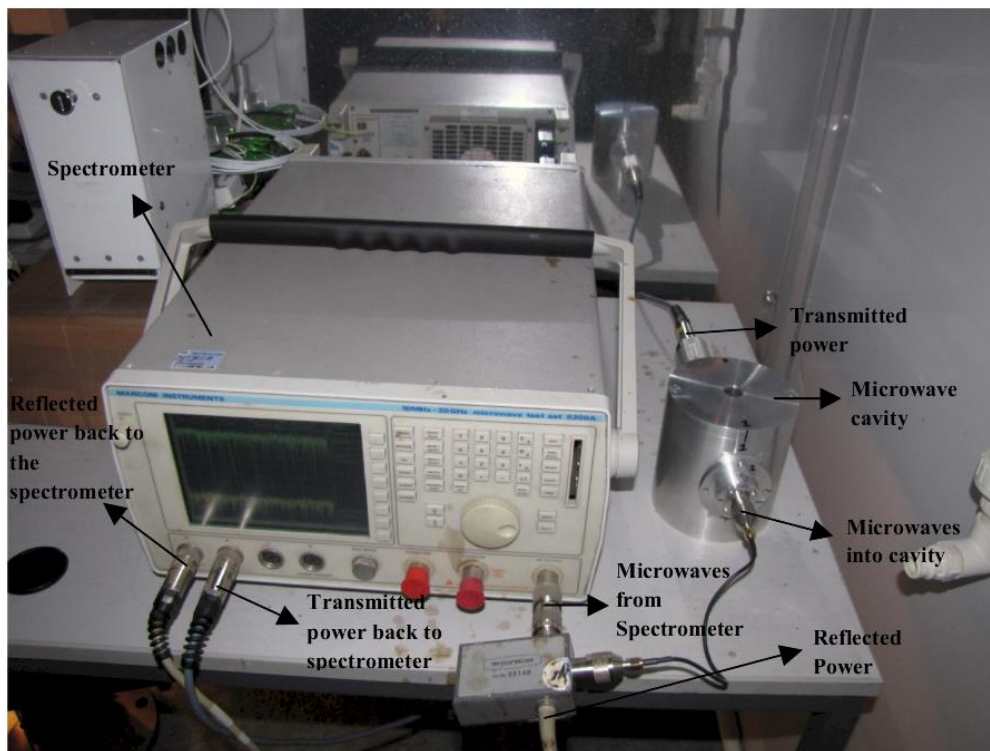
The comparison of the two rubber samples in Figure 3 and 4 in terms of both their surface properties (appearance, texture, smoothness) and change in the size shows promising results and demonstrate the effectiveness of the treatment. Looking at the surface of the untreated rubber in Figure 3 (a) shows the surface

222 being rough at approximately 3000x magnification. Comparing it with the treated rubber surface in Figure 4 (a)  
223 shows the smooth surface labelled, Figure 4(a), covered by the film of polyethyleneimine even after going  
224 through the harsh temperature conditions that resembles to the actual treatment and its interaction with bitumen.  
225 It is noticed in Figure 4(a) that some parts of the treated rubber has an excess of polyethyleneimine, an example  
226 of which is indicated and encircled in the Figure. This might be mitigated in the case of bitumen with further  
227 mixing to spread the layer uniformly. Figure 3(b) and 4(b) shows that size comparison before and after  
228 treatment of the similar size rubber sample. It can be observed that the particle sizes remain similar before and  
229 after treatment, hence demonstrating the least (which would be confirmed with the microwave analysis in  
230 section 3.2) as well as the durability and effectiveness of the treatment to reduce the absorption of light fractions  
231 into the rubber. Additional result of the treated rubber sample with higher percentage of polyethyleneimine is  
232 presented in Figure 5 to verify the existence and effectiveness of the coating. A better uniformity in the coating  
233 was achieved as shown in Figure 5(a) at approximately 3000x magnification compared to Figure 4(a). When  
234 comparing the treated surface, Figure 5(a), with the untreated rubber in Figure 3(a) it is noticed that the treated  
235 rubber surface has smoother appearance in comparison to the untreated rubber surface that shows rough surface  
236 and texture as in Figure 3(a). In terms of the change in the size of the sample after treatment, it can noticed in  
237 Figure 5(b) that the sample size of 1 mm doesn't change in comparison to Figure 3(b), untreated rubber. This  
238 indicates the effectiveness of the cross-linking treatment to reduce the swelling of rubber particles.

### 239 3.2. Microwave analysis

240 The microwave cylindrical resonant cavity was utilised for the microwave analyses of the sample. It is a  
241 bespoke cavity resonator designed at the Radio Frequency and Microwave (RFM) group, Liverpool John  
242 Moores University to carry out the analysis of various materials such as carbon, silver products, polymers, etc.  
243 The technique is non-invasive and non-destructive. The resonant cavity is classed as a closed box short circuited  
244 at both ends. Both the electric and magnetic energy is stored within the cavity. These fields together form a  
245 standing wave at certain frequencies causing the resonance to occur inside the cavity. Because of this  
246 phenomena various modes may exist in the cavity, each of them with its own resonant frequency and a quality  
247 factor (Q) associated with it. For the cylindrical cavity, the fundamental modes are  $TE_{111}$  and  $TM_{010}$ . The TE  
248 (Transverse Electric) mode has a magnetic component in the direction of propagation whereas the TM  
249 (Transverse magnetic) mode has an electric component in the direction of propagation of wave. In this way, the  
250 power dissipated in the metallic walls of the cavity due to the applied electromagnetic wave (in the microwave

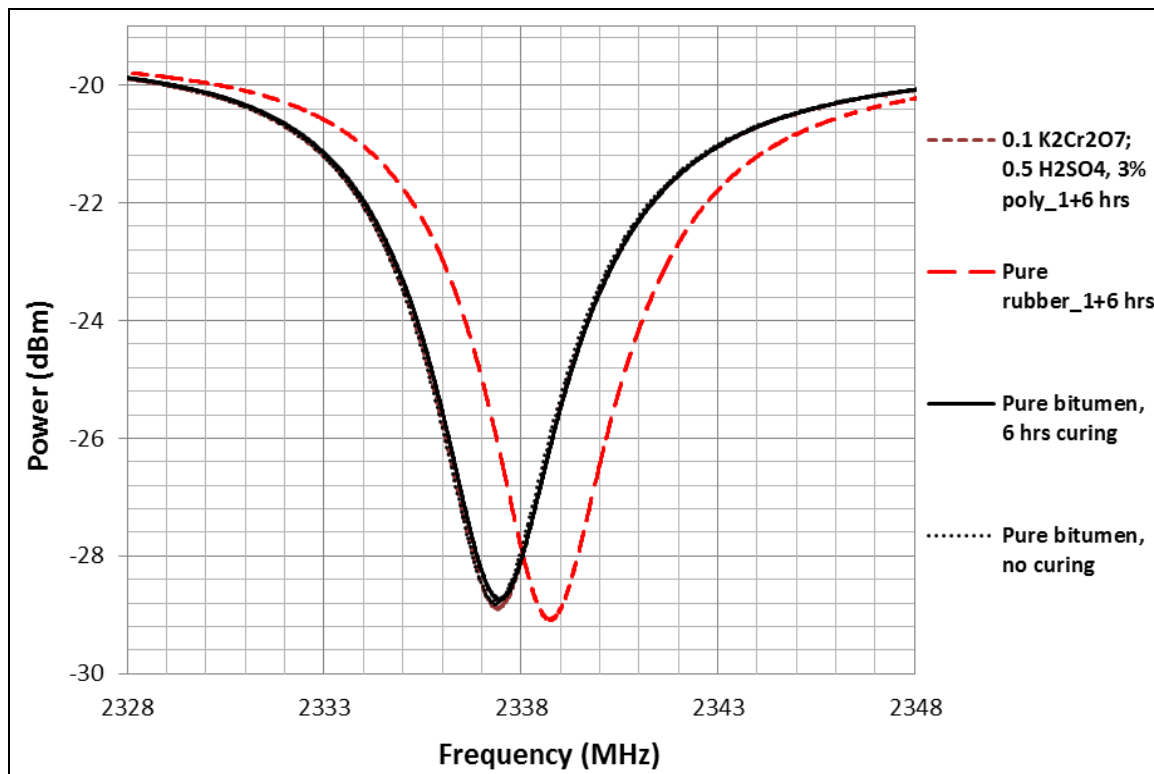
251 frequency region) can be used to study the material under test based on its interaction with the material [32, 33].  
 252 The setup of the microwave spectroscopy system is shown in Figure 6.  
 253 Due to the response of the microwaves to the bitumen, the reflected power,  $S_{11}$ , described in the section 2.5 was  
 254 considered for the analysis before and after the treatment. The signal represents the change in the dielectric  
 255 properties of the bitumen samples based on its absorption into rubber, and any changes incurred as a result of  
 256 the reduction in the light fractions percentage. After the range of experiments conducted it was found that the  
 257 reflected power in the frequency range of 2.329-2.347 GHz was best suited to indicate the absorption of waste  
 258 rubber for each sample and the effectiveness of the treatment through the resonant peaks obtained.



259  
 260 Figure 6. Non-invasive real-time cylindrical microwave cavity setup for the bitumen analysis

261 The microwave analysis results of four samples are presented in Figure 7. The samples analysed include:

- 262
- Bitumen sample, un-cured.
  - 263 • Bitumen sample, cured for 6 hours to compare with the 6 hours cured bitumen extracted from the blend
  - 264 with treated rubber.
  - 265 • Bitumen extracted from the blend containing pure/untreated rubber.
  - 266 • Bitumen extracted from the blend containing the 10% (by weight) of the treated rubber oxidised with
  - 267 0.1 mol/L of potassium dichromate ( $K_2Cr_2O_7$ ), 0.5 mol/L of sulphuric acid ( $H_2SO_4$ ) and water ( $H_2O$ )
  - 268 and cross-linked with 3% of polyethyleneimine.

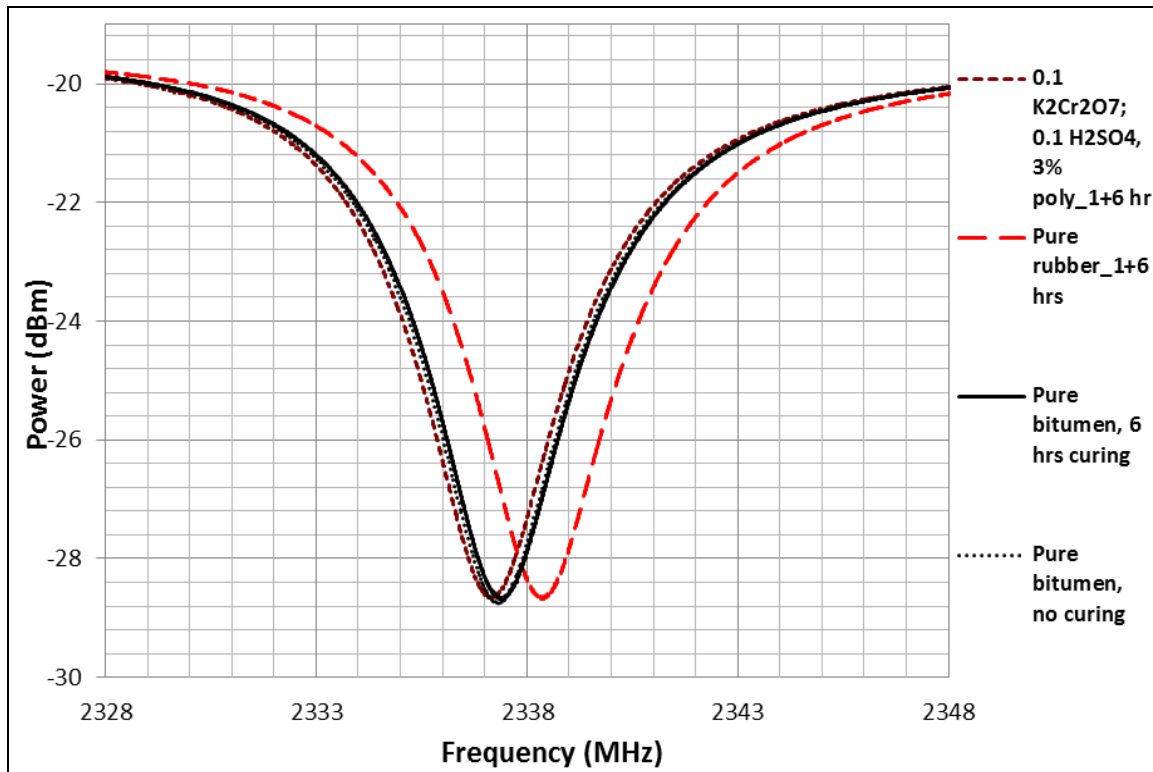


269

270 Figure 7: Effects of 1 hour mixing plus 6 hours curing using 3% polyethyleneimine treatment in the rubber-  
 271 bitumen blend (10:100 ratio) containing rubber oxidised with 0.1 mol/L K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub>, 0.5 mol/L of H<sub>2</sub>SO<sub>4</sub>, and  
 272 water

273 The resonant peak of the bitumen (cured and uncured) sample were detected at around 2.3374 GHz and 2.3373  
 274 GHz respectively. When these peaks were compared with the bitumen sample extracted from the blend  
 275 containing the untreated rubber mixed for 1 hour and further cured for 6 hours, a shift to the right was observed  
 276 with a resonant peak detected at the frequency of 2.3387 GHz. The shift was approximately 1.34 and 1.4441  
 277 MHz from the cured and uncured bitumen sample. This amount of shift (in MHz) is significant keeping in  
 278 consideration the sensitivity of the instrument. This signifies that the bitumen has changed its properties due to  
 279 its light fractions absorbed by the rubber. When the curve of the bitumen retrieved from the treated rubber  
 280 bitumen blend was analysed the peak frequency was observed at 2.3373 which is an overlap on the uncured  
 281 bitumen sample curve. Thus, the bitumen was exhibiting the dielectric properties similar to the pure bitumen  
 282 and could be linked with the loss of light fraction in the bitumen. On this occasion, the results indicate minimal  
 283 absorption of light fractions into the treated rubber.

284 To indicate the effectiveness of the chemical treatment, microwave analysis of a sample oxidised with 0.1 mol/L  
 285 K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub>, 0.1 mol/L of H<sub>2</sub>SO<sub>4</sub>, and water followed by 3% polyethyleneimine is also presented in Figure 8.



286

287 Figure 8: Effect of 1 hour mixing plus 6 hours curing and 3% polyethyleneimine on rubber-bitumen blend

288 (10:100) containing rubber oxidised with 0.1 mol/L K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub>, 0.1 mol/L of H<sub>2</sub>SO<sub>4</sub>, and water

289 The four samples presented in Figure 8 are:

- 290 • Pure bitumen sample, un-cured
- 291 • Bitumen sample, cured for 6 hours to compare it with the bitumen sample extracted from the cured  
292 blend with treated rubber.
- 293 • Bitumen sample extracted from the blend with pure/untreated rubber.
- 294 • Bitumen sample extracted from the blend containing 10% (by weight) of the treated rubber, oxidised  
295 with 0.1 mol/L K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub>, 0.1 mol/L of H<sub>2</sub>SO<sub>4</sub>, and water and cross-linked with 3% of the  
296 polyethyleneimine.

297 It can be noticed from Figure 8 that the resonant frequencies of pure bitumen sample (cured and un-cured) are  
298 detected at around 2.33742 & 2.33732 GHz. In comparison, the curve of the bitumen sample retrieved from the  
299 pure/untreated rubber-bitumen blend shifted to the right showing the peak at approximately 2.33834 GHz, a  
300 difference of 0.92 and 1.02 MHz from the cured and un-cured sample respectively. It is a considerable shift  
301 when compared to the peak value of the bitumen sample retrieved from the blend with modified rubber. It has a  
302 peak at 2.33713 GHz, representing a shift of approximately 0.29 & 0.19 MHz to the left, a value closer to the

303 cured and un-cured sample of the pure bitumen. Similar to the result in Figure 7, the peak value of the modified  
 304 bitumen in Figure 8 indicates the dielectric properties of the modified bitumen sample close to the pure bitumen  
 305 showing a reduction in the migration of light fractions into rubber. However, it is slightly towards the left in  
 306 comparison to modified bitumen in Figure 7 indicating a slight change in the dielectric properties of the  
 307 modified bitumen. Hence, the combination of treatment in Figure 7 was more effective in comparison to the  
 308 treatment in Figure 8, i.e. 0.1 mol/L K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub>, 0.1 mol/L of H<sub>2</sub>SO<sub>4</sub>, and water followed by 3%  
 309 polyethyleneimine in comparison with the later 0.1 mol/L K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub>, 0.5 mol/L of H<sub>2</sub>SO<sub>4</sub>, and water followed  
 310 by 3% of polyethyleneimine. This shows that a higher molar concentration of H<sub>2</sub>SO<sub>4</sub> was more effective.

### 311 3.3. Mechanical testing and results

#### 312 3.3.1. Mix design

313 The mix design was selected based on the target application. The target applications included surfaces such as a  
 314 light traffic areas, sports pitches, golf courses, walkways/pathways, etc. The available recipe of the mix designs  
 315 in the British and European Standard for this kind of application includes a 6 mm medium graded surface course  
 316 (MGSC) mix and a dense graded surface course mix. Since, the purpose was to modify the mix design and  
 317 accommodate the waste rubber aggregates (along with their swelling properties) the MGSC mix design was  
 318 selected for carrying out the mix designs and mechanical testing. A typical MGSC mix gradation target limit  
 319 according to the PD6691 [34] is shown in Table 3.

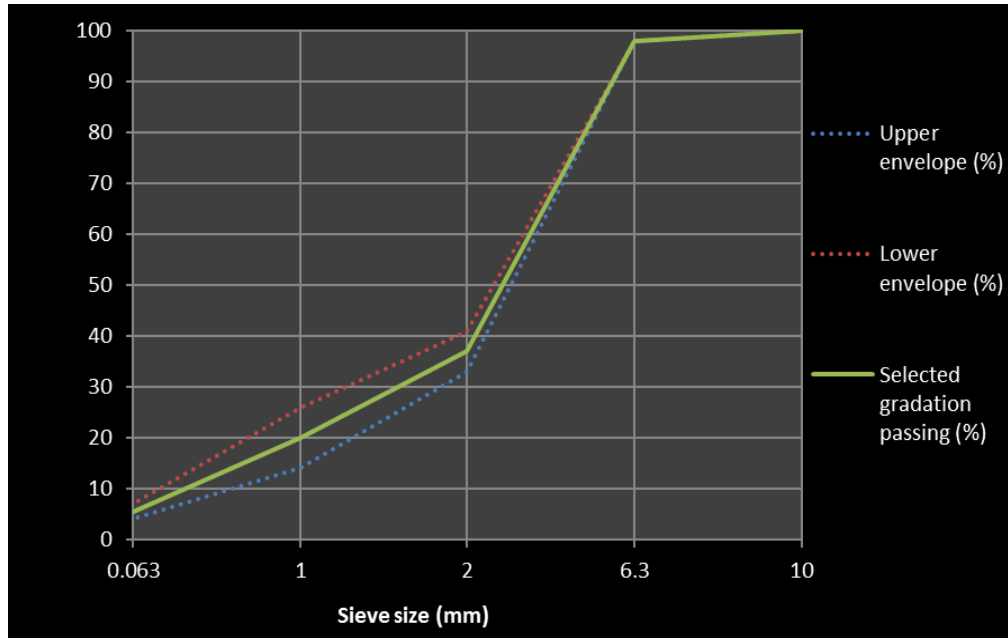
320 Table 3. The target limit for the composition of Medium Graded Surface Course (MGSC) mix [34]

<b>Mixture description</b>	<b>AC 6 med surface</b>
<b>Test sieve aperture size</b>	<b>% by mass passing</b>
<b>10</b>	100
<b>6.3</b>	98
<b>4</b>	-
<b>2</b>	33-41
<b>1</b>	14-26
<b>0.250</b>	-
<b>0.063</b>	4-7

321



322 For the purpose of the laboratory based mix design of the samples, the mean value of the percentage of the  
323 respective size of the aggregates was selected for the new mix designs. The mix gradation chart for the target  
324 limit and composition is shown in Figure 9.



325

326 Figure 9: MGSC mix gradation chart for the target limit and composition

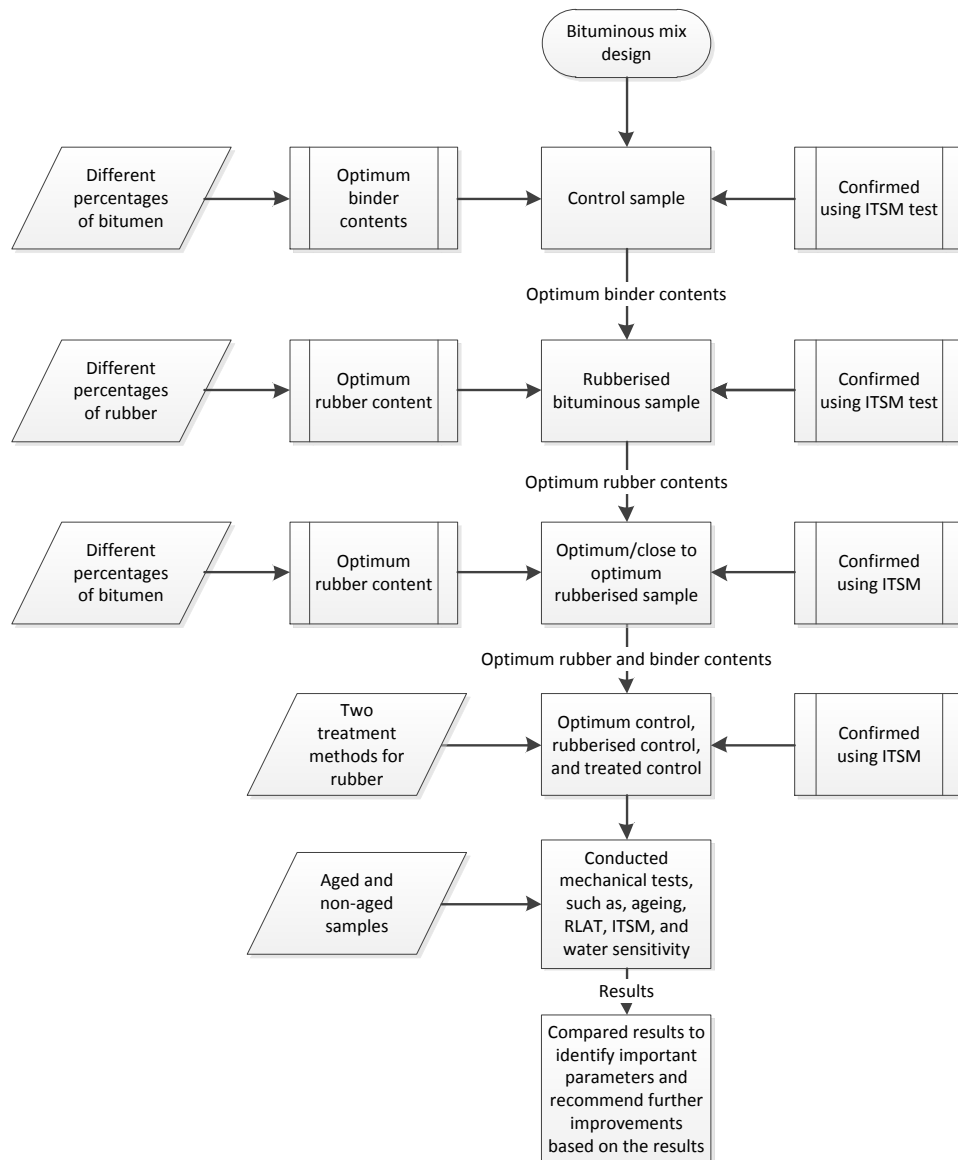
### 327 3.3.2. Optimisation of the mix design and samples prepared

328 The mix design of the control and rubberised samples in terms of the amount of bitumen and rubber was  
329 optimised using the flow chart of Figure 10. The samples prepared were as follows:

- 330 • Control sample
- 331 • Sample with untreated rubber
- 332 • Sample with treated rubber using method 1 (TP1)
- 333 • Sample with treated rubber using method 2 (TP2)

334 Typically, the difference between the TP1 and TP2 treatment method was that in TP1 the rubber was first  
335 oxidised and then cross-linked in the rubber-bitumen blend mixed for 1 hour and cured for further 6 hours as  
336 stated in section 2.4. However, to examine the impact of a variation in the treatment method, in TP2 the oxidised  
337 rubber was first cross-linked using the polyethyleneimine and cured for 3 hours in the oven at 170-180°C. The  
338 rubber obtained was then used in the preparation of rubberised bituminous mix samples.

339 All the bituminous samples (control, untreated rubberised and treated rubberised) were prepared and compacted  
 340 according to the British Standards [35, 36]. The samples prepared were tested using the Indirect Tensile  
 341 Stiffness Modulus (ITSM), for the stiffness behaviour, and Repeated Load Axial Test (RLAT), to evaluate the  
 342 resistance to permanent deformation. Ageing tests were also conducted on both the control and modified  
 343 bituminous samples to examine the effect of ageing to simulate the theoretical service life from 6 hours to 10  
 344 years. Some of the samples prepared are presented in Figure 11.



345

346

Figure 10: Schematic representation of the overall strategy to optimise the mix design

347



348

Figure 11: Subset of the bituminous mix samples prepared (control and rubberised)

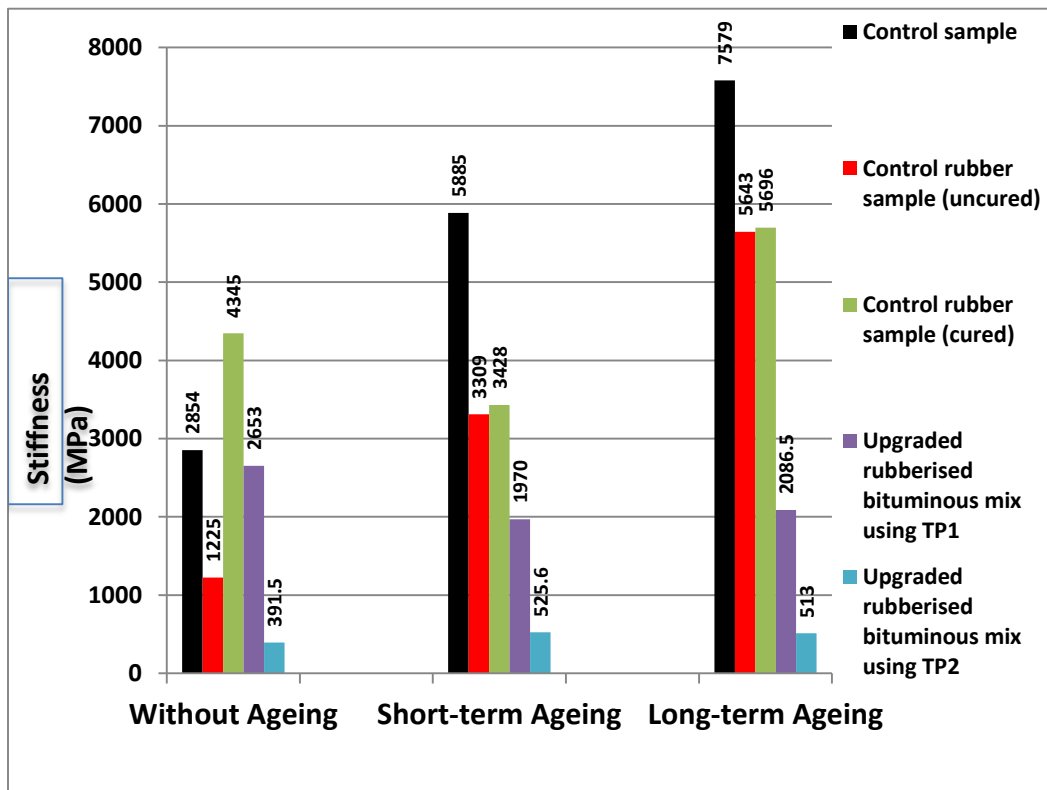
349 **3.3.3. Indirect Tensile Stiffness Modulus (ITSM) test**

350 The test conditions set for the ITSM test were the horizontal strain/deformation of 0.005% of the specimen

351 diameter, the rise time of  $124 \pm 4$  ms (equivalent to the frequency of 1.33 Hz), the specimen diameter of 100

352 mm, the specimen thickness between 30-70 mm, and the test temperature of 20°C. The stiffness results and

353 comparison of various samples are presented in Figure 12.



354

355 Figure 12: Stiffness Modulus of the control and rubberised bituminous samples (aged and unaged)

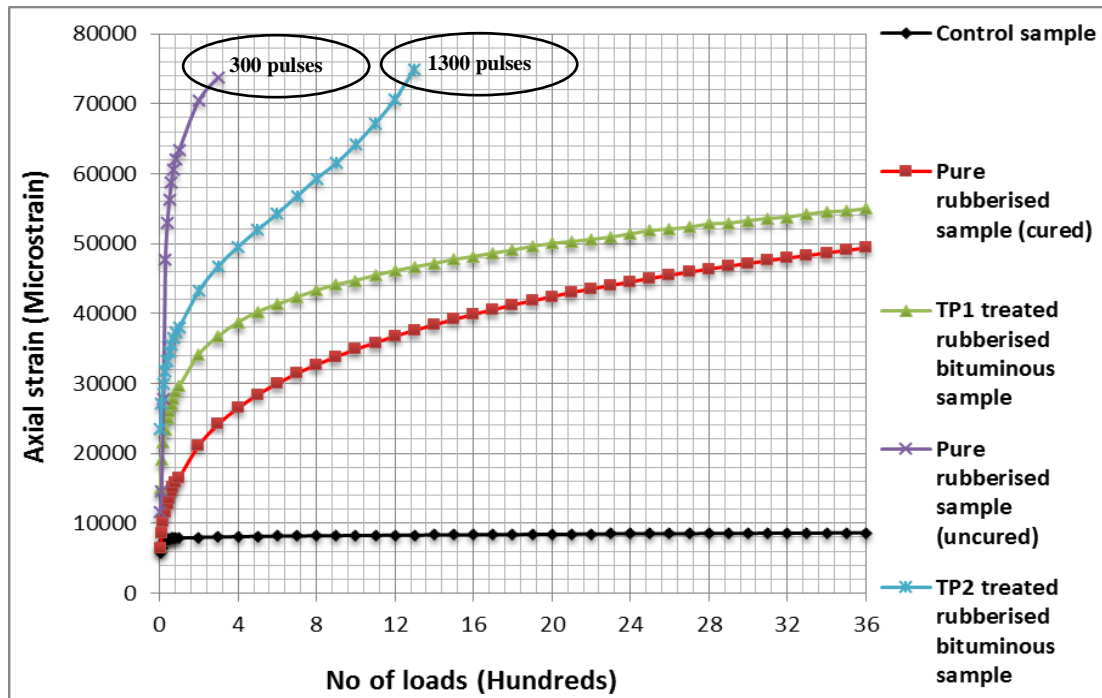
356 The results show that in the case of unaged samples the highest value of the stiffness was obtained for the  
357 untreated rubberised bituminous sample cured for 6 hours. The value of the stiffness of the TP1 treated  
358 rubberised bituminous mix has a value of 2653 MPa, comparable to the control sample of 2854 MPa. The  
359 stiffness values for the TP2 treated rubberised bituminous mix and the uncured control rubber sample has the  
360 lowest values. In comparison to the unaged samples, the short-term aged samples showed variation in the  
361 stiffness values of the samples. The highest value was obtained for the control sample followed by the cured  
362 untreated rubberised bituminous sample that was slightly higher than the uncured untreated rubberised sample  
363 (3309 MPa). The value of the TP1 treated rubberised bituminous sample dropped to 1970 MPa compared to its  
364 unaged version. However, it was still significantly higher than the TP2 treated rubberised bituminous sample.  
365 The last comparison was between the long term aged samples. Significantly higher values were obtained for the  
366 control and untreated rubberised bituminous samples (cured and uncured). However, the stiffness value of the  
367 TP1 treated rubberised bituminous sample increased (2086.5 MPa) with ageing showing improvement in its  
368 stiffness. In contrast the TP2 treated rubberised bituminous mix stiffness reduced further to 513 MPa. On  
369 repetition similar results were obtained. The results show that the stiffness value needs improvement in the case  
370 of lightly trafficked areas applications. However, the TP1 treated mix can be utilised for playground and  
371 footpath applications after changes in the mix design (such as the type of filler and percentage of waste rubber)  
372 to improve its performance. This will be verified in the proposed following study.

#### 373 **3.3.4. Repeated Load Axial (RLA) test**

374 To study the resistance to permanent deformation of the newly developed rubberised bituminous mix, an RLA  
375 test was used. Again, the comparison was made among the control sample (no rubber), the control rubberised  
376 sample and treated rubberised sample (both aged and unaged state). The results are presented in Figure 13 and  
377 14 for unaged and aged samples respectively.

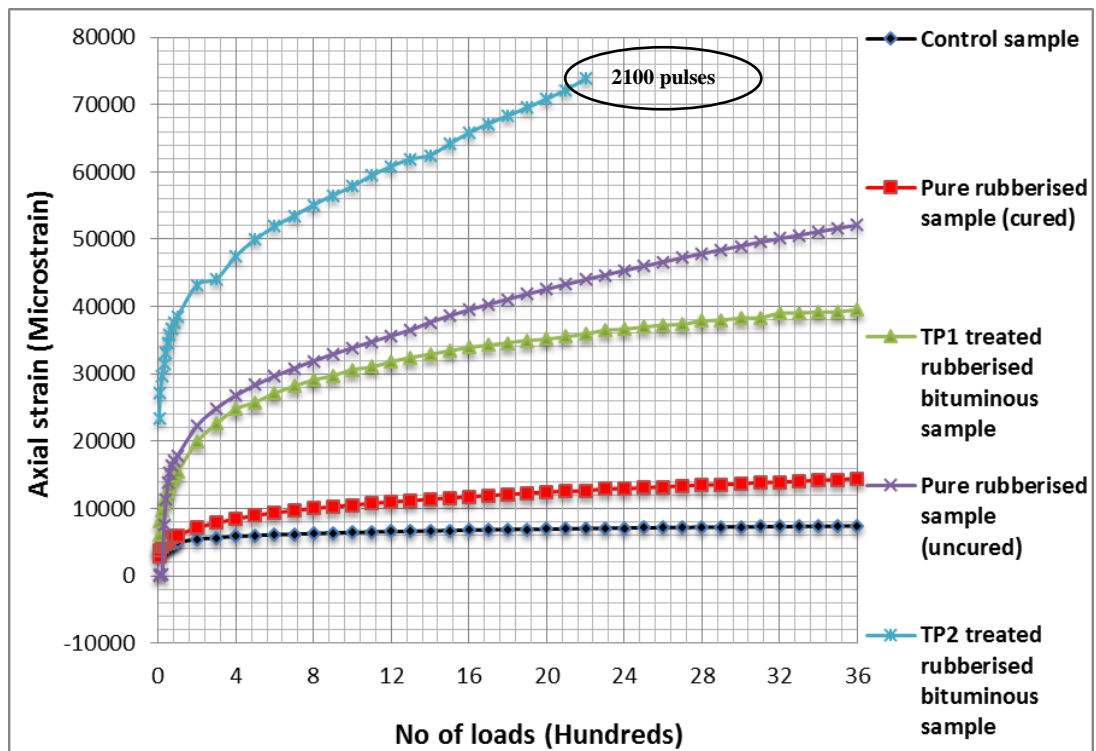
378 By comparing Figure 13 and 14 it can be concluded that the deformation of all the samples decreased after  
379 ageing compared to the unaged samples. Improvement was observed in the results of the untreated rubberised  
380 sample (cured) whereby the axial strain reduced from 50,000 micro-strain to 14500 micro-strain for 3600 pulses.  
381 Also, significant improvement was observed in the RLA value of the TP1 treated rubberised bituminous sample  
382 with the axial strain reduced from 55,000 micro-strain to almost 40,000 micro-strain for 3,600 pulses. In  
383 contrast, for both the untreated rubberised bituminous sample (uncured) and TP2 treated rubberised bituminous  
384 sample, the failure occurred at 300 and 1300 pulses respectively, earlier than the completion of the load pulses

385 (Figure 13). Improvement was observed though in the case of uncured rubberised bituminous sample with an  
 386 axial strain value of 52,000 micro-strain (Figure 14) compared to the failure that occurred at the value of 73,858  
 387 micro-strain following 2100 pulses (Figure 13).



388

389 Figure 13: Repeated load axial test results of the unaged control and rubberised samples



390

391 Figure 14: Repeated load axial test results of the aged control and rubberised samples

392 The results show that the TP1 treated rubberised need improvement in terms of the deformation properties for its  
393 application in the light traffic areas but may be suitable for the applications such as playgrounds, footpaths, etc.  
394 after minor amendments in the design. This could be achieved through the use of an alternative filler or by the  
395 inclusion of fibres into the mix and retested to see the improvement. It is therefore recommended that the future  
396 work should utilise fibres in the mix or replace the filler used with the mineral filler such as hydrated limestone.  
397 Literature [37-40] suggested that by replacing the ordinary filler from the aggregate source with the mineral  
398 filler such as white cement, ordinary Portland cement, hydrated limestone dust, ceramic waste dust, coal fly ash,  
399 etc. and adding fibres substantially improves the mechanical properties of the material which might be suitable  
400 for light traffic areas, etc. It would be interesting to have further laboratory trials and tests using these fillers  
401 with the TP1 treated waste rubber to monitor and analyse the impact of such addition.

#### 402 **4. CONCLUSIONS AND RECOMMENDATIONS**

403 Tests were conducted to monitor and analyse two aspects of the current study. This included:

- 404 • The quality of the waste rubber surface treatment to reduce its absorption properties. The tests  
405 conducted were the SEM surface analysis of the rubber and Microwave spectroscopy analysis of the  
406 bitumen.
- 407 • Mechanical properties of the newly designed bituminous mixes by utilising the treated and untreated  
408 rubber and comparing it with the control samples through Stiffness modulus test and Repeated load  
409 axial test.

410 The following were the conclusions from the analysis and mechanical test results.

- 411 • The SEM analysis in Figure 4 & 5 of the surface treatment showed the existence of the film on the  
412 surface of the rubber particle. This indicates the durability of the thin film on the surface under harsh  
413 temperature conditions of mixing.
- 414 • The microwave spectroscopy analysis of various samples of bitumen such as the pure bitumen, bitumen  
415 out of the untreated rubber bitumen blend and bitumen from the treated rubber bitumen blend  
416 successfully showed the variation in the bitumen properties when the rubber was mixed with the  
417 bitumen. The microwave spectrum obtained for the bitumen from treated rubber bitumen blend showed

418 the properties close to the pure bitumen demonstrating the effectiveness of the treatment. This provided  
419 evidence of the reduction in the absorption properties of the rubber. Whereas, the bitumen from the  
420 untreated rubber bitumen blend had a shifted spectrum from the pure bitumen case showing the change  
421 in its properties due to the absorption of light fraction from bitumen into rubber.

422 • The mechanical test results indicates that the mix design need improvement in the following/future  
423 study to make it applicable for a range of applications. The results show that the material is only  
424 suitable for applications such as indoor and small play areas and possibly walkways and pathways.

425 • As mentioned in the last section, the mechanical behaviour of the mix can be studied in the subsequent  
426 studies to monitor any improvements by replacing the ordinary filler with mineral filler or by the  
427 addition of various types of fibres.

428 • Future work is required on the use of higher percentage of rubber than 10% to make the material more  
429 suitable for sensitive applications such as children’s playgrounds, sports track and pitches, etc. that  
430 require higher flexibility and softness of the surface with shock absorbing properties.

431 • Thorough mechanical testing is required to ensure the durability of the material with additional tests  
432 such as Head Injury Criterion (HIC), etc. to make it suitable for additional industrial applications such  
433 as children’s playground, sports pitches, tennis courts and turfs.

434 • Field trials along with the modifications in the mix design are also recommended for the newly devised  
435 TP1 treated rubberised mix to monitor it over a certain period of time and in a range if weather  
436 conditions.

437 • The results show that the treatment technique has the potential to reduce the absorption properties of  
438 the waste rubber and improve the bituminous mix mechanical properties with further modifications in  
439 the design. However, further studies with additional chemical treatments and testing is recommended to  
440 verify the claim. It would also be interesting to verify the reduction in absorption by analysing the  
441 bitumen samples through a suitable existing techniques such as FT-IR.

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