

1        **Optimizing the Mixing Procedure of Warm Asphalt Rubber with**  
2                    **Wax-Based Additives through Mechanism Investigation and**  
3                                    **Performance Characterization**

4    Zhen Leng<sup>a,\*</sup>, Huayang Yu<sup>b,\*\*</sup>, Zeyu Zhang<sup>c</sup>, Zhifei Tan<sup>d</sup>

5    **Abstract**

6    Wax-based additives can be used as flow improvers to enhance the workability of  
7    asphalt rubber (AR). Conventionally, warm asphalt rubber (WAR) is produced by  
8    preparing AR first and then blending it with warm mix asphalt (WMA) additive.  
9    However, directly mixing WMA additive, base asphalt and crumb rubber together  
10   may save more energy since the early incorporation of WMA additive also helps  
11   decrease the production temperature of AR. To assess the feasibility of incorporating  
12   wax-based additives at an earlier stage, this study investigates the influence of the  
13   mixing procedure on WAR prepared by two wax-based additives, i.e., commercial  
14   Sasobit and conventional paraffin wax. Rheological tests on WAR revealed no  
15   significant difference between WARs prepared by different procedures. However, the  
16   direct mixing method led to worse WAR workability compared to the traditional

---

<sup>a</sup> Assistant Professor, Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong, zhen.leng@polyu.edu.hk (\*Corresponding Author)

<sup>b</sup> PhD Candidate, Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong, huayang.yu@connect.polyu.hk (\*\*Co-Corresponding Author)

<sup>c</sup> Research Assistant, Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong

<sup>d</sup> Research Assistant, Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong

17 mixing procedure. Chemical analysis on the liquid phase of WARs (crumb rubber  
18 removed) indicated that independent of the type of wax-based additive, there is less  
19 wax in the liquid phase of WARs when the additive is added earlier, which may be  
20 caused by the absorption of wax by crumb rubber during the interacting process. Thus,  
21 it is not recommended to replace the traditional mixing procedure with the direct  
22 mixing method.

23 **Keywords:** Asphalt rubber; warm mix asphalt; wax; rheological property; workability;  
24 interaction

## 25 **1 Introduction**

26 Asphalt rubber (AR), which is defined as raw bitumen modified by no less than 15%  
27 of crumb rubber modifier (CRM) by total binder weight [1], has gained increasing  
28 interest due to its excellent mechanical performance and tyre-road noise reduction  
29 function [2] and [3]. During its preparation process at elevated temperature, CRM  
30 absorbs the light fractions of base binder and releases polymer chains, such as natural  
31 rubber and styrene-butadiene rubber, resulting in higher percentage of heavy  
32 molecules in asphalt, thus higher viscosity [4], [5], [6] and [7]. Although the high  
33 viscous behavior enhances the rutting resistance of asphalt, it brings the concerns of  
34 worse pumpability, mixability and workability. In general, the production temperature  
35 of AR is 20-30 °C higher than that of base binder, leading to more energy  
36 consumption and higher construction emission [8]. During the past decade, warm-mix  
37 asphalt (WMA) technology has been successfully applied to alleviate the workability

38 concern of AR [9] and [10]. Warm asphalt rubber (WAR) binders with lower  
39 viscosities at mixing and compacting temperatures can be prepared by incorporating  
40 WMA additives into AR binder before mixing it with aggregates. A 15-30 °C  
41 reduction can be achieved by using different WMA additives [8], [10], [11], [12], [13],  
42 [14] and [15]. Among various types of WMA additives, organic additives, in most  
43 cases wax-based additives, have been reported to be effective in improving AR's  
44 workability without compromising its mechanical properties [11], [12], and [13].

45 Attributed to its low melting point and good flowability at elevated temperature, wax  
46 is usually recognized as flow improver of asphalt binder. Various studies have shown  
47 that commercial wax product prepared by Fischer-Tropsch (FT) synthesis process  
48 positively affects not only workability, but also rutting and fatigue resistance of  
49 asphalt [2], [7], [8], [10], [11], [15], and [16]. Meanwhile, despite its potential  
50 negative effect on low-temperature cracking resistance, traditional paraffin wax was  
51 found to be a potential WMA additive for AR binders, since its adverse effect on  
52 low-temperature performance can be compensated by CRM [13]. To prepare WARs  
53 with wax additives, the following two procedures can be adopted: 1) conventional  
54 method: mixing CRM and base binder first and then adding wax additive; 2) direct  
55 mixing method: directly mixing CRM, base binder and wax additives together.  
56 Between these two methods, the direct mixing method may save more energy as the  
57 preparing temperature of AR can also be reduced due to the earlier incorporation of  
58 WMA additive. Once the wax is incorporated, the viscosity of binder decreases,

59 which brings positive effect on homogenous distribution of crumb rubber in base  
60 binder and makes the mixing work easier. However, it is still unclear that whether  
61 these two mixing procedures may lead to different interactions among CRM, raw  
62 binder and WMA additive, thus different final workability and rheological properties  
63 of WAR.

64 The performance of WAR with wax additives prepared by the conventional method  
65 has been well studied, while the research on the direct mixing method is relatively  
66 limited [12] and [17]. Thus, this study aims to evaluate the feasibility and  
67 effectiveness of incorporating wax-based WMA additives at an earlier stage of WAR  
68 production. To achieve this objective, the rheological properties, including penetration,  
69 softening point, viscosity, Superpave rutting parameter, and Superpave fatigue  
70 parameter of WAR binders were characterized and compared. In addition, to reveal  
71 the interaction mechanism, chemical analyses including Differential Scanning  
72 Calorimeter (DSC) test, Gel Permeation Chromatography (GPC) test and wax content  
73 test were also conducted.

## 74 **2 Experimental program**

### 75 *2.1 Preparation of AR and WARs*

76 Asphalt with a penetration grade of 60/70 (Pen 60/70), a common type of asphalt in  
77 Hong Kong, was used as the base binder. Crumb rubber with 40-mesh size was used  
78 and the content was 18% by weight of base binder. Two different types of wax

79 additives were selected and used, namely Sasobit (commercial WMA additive  
80 produced by the Fisher-Tropsch process) and 56<sup>#</sup> paraffin wax (conventional wax),  
81 and their dosages were determined as 3wt% and 1.5wt%, respectively, based on the  
82 manufacture's recommendation and preliminary test results [13]. Both the  
83 conventional and direct mixing methods were applied to produce WARs, leading to in  
84 total four WARs. These WARs are labeled as ARS, ARW, ARSD and ARWD,  
85 representing AR with Sasobit prepared by conventional method, AR with Wax  
86 prepared by conventional method, AR with Sasobit prepared by direct mixing method,  
87 and AR with Wax prepared by direct mixing method, respectively. Table 1 provides  
88 detailed description on the sample IDs and the corresponding mixing conditions of  
89 each test binder.

90 Both Sasobit and 56<sup>#</sup> paraffin wax can be completely dissolved in asphalt, while  
91 CRM remains in small particulate form in asphalt after interaction. To investigate the  
92 interaction among different components of AR and WARs, the liquid phase of AR and  
93 WARs were extracted by passing the hot binders through a mesh #200 sieve [5]. Right  
94 after they were prepared, the AR and WARs were dropped onto the sieve which was  
95 placed on top of a custom-designed container. Then the whole extraction system was  
96 placed into an oven at 150 °C for 30 minutes to drain the liquid phase through the  
97 sieve. The extracted liquid phase was stored at 0 °C to prevent further ageing or  
98 reaction. Each extraction process could produce approximately 50g extracted liquid

99 phase from 400g AR or WAR binders. The liquid phases of WARs were labeled as  
100 L-WAR in this paper.

## 101 *2.2 Testing Program*

102 Conventional binder property tests conducted in this study included penetration,  
103 softening point and ductility tests [18], [19], and [20].

104 Viscoelastic properties of the AR and WAR binders as well as their liquid phases were  
105 characterized by the dynamic shear rheometer (DSR) test. The high- and  
106 intermediate-temperature performances were characterized by the Superpave rutting  
107 parameter and fatigue parameter, respectively [21]. 2mm gap was used for all DSR  
108 tests to reduce the influence of CRM particles [5], [22], and [23]. Unaged binders  
109 were used for Superpave rutting parameter measurement (with 25mm-diameter plates)  
110 and Pressure Aging Vessel (PAV) aged binders were used for Superpave fatigue  
111 parameter measurement (with 8mm-diameter plates). Besides, the complex modulus  
112 and phase angle were recorded for rheological analysis. For each test, two replicates  
113 were prepared.

114 The workabilities of AR and WARs were evaluated by three parameters, including  
115 rotational viscosity [24], air void content of Marshall Specimen (SMA10, 4.0%  
116 design air void) corresponding to each binder [25, 26], and number of gyrations of  
117 Superpave Gyratory Compactor (SGC) samples (SMA 10, 7.0% air void) to achieve  
118 the same specimen height [26]. The mixing and compaction temperatures of the

119 samples with AR were 176 °C and 160 °C, respectively, while the samples with  
120 WARs were mixed at 160 °C and compacted at 144 °C. Three replicates were  
121 prepared and tested.

122 The interaction among asphalt, CRM and WMA additives was investigated through  
123 chemical analyses, including Differential Scanning Calorimeter (DSC) test, Gel  
124 Permeation Chromatography (GPC) test, and wax content test [27]. All these tests  
125 were performed on the extracted liquid phases of the test binders.

126 The thermal properties of L-WARs were measured using the Mettler Toledo  
127 instruments DSC3. The melting temperatures ( $T_m$ ) of the binder components were  
128 determined by heating the samples from -20 °C to 150 °C at a rate of 5 °C/min.

129 The molecular weight distribution of L-WARs was evaluated by GPC test. A P230  
130 Elite GPC with three columns (M, NT and NN) was used to separate the constituents  
131 of asphalt binder based on molecular size. Each sample was dissolved into  
132 Tetrahydrofuran (THF) and then filtered through a 0.2µm Polytetrafluoroethylene  
133 (PTFE) syringe filter prior to being placed into the injection module. During the GPC  
134 test, the asphalt-THF solution was drained through columns and allowed to flow at a  
135 rate of 0.5 ml/min, and the temperature of the columns were maintained at 40 °C. The  
136 components' concentration in the eluent was recorded using a differential  
137 refractometer, and the resulting chromatogram was analyzed to obtain the molecular  
138 size distribution.

139 To measure the wax content, a distillation process was applied to extract the wax from

140 asphalt components at 550 °C. The distilled components were dissolved in  
141 ether/ethanol (50/50, V/V) solvent and crystallized at -20 °C. The crystallized waxes  
142 were collected by filtration and their weights were measured.

### 143 **3 Test results**

#### 144 *3.1 Rheological Properties of AR and WARs*

145 Figure 1 describes the rheological properties of the test binders. According to Figure  
146 1a and 1b, the incorporation of CRM decreased the penetration and ductility and  
147 increased the softening point of base asphalt. Regardless of the mixing procedure, the  
148 effects of Sasobit on penetration and softening point were similar to those of CRM,  
149 while paraffin wax provided the opposite modification effects. WMA additives had  
150 insignificant effect on ductility, since the homogeneous structure of asphalt was  
151 destroyed by CRM. Figure 1c shows the Superpave rutting parameters of all test  
152 binders at various temperatures. The failure temperatures of ARS and ARSW were  
153 above 88 °C, while those of AR, ARW and ARWD were between 82 °C and 88 °C  
154 [21]. The Superpave rutting parameters of ARW and ARWD were lower than AR, but  
155 still much higher than that of the base binder. These results indicate that Sasobit is  
156 beneficial to the rutting resistance of AR while paraffin wax has negative effect,  
157 which is consistent with the results of penetration and softening point tests. No  
158 obvious difference can be observed between WARs prepared by different mixing  
159 procedures. Figure 1d shows that all binders with CRM have superior fatigue  
160 resistance than base asphalt. Both Sasobit and paraffin wax negatively affected the



161 fatigue properties of AR. Among the four WARs, ARS and ARSD showed the best  
162 fatigue resistance, with the threshold temperature lower than 16 °C ( $G^* \sin \delta < 5 \text{MPa}$ ).  
163 Besides, the intermediate-temperature fatigue performance of WAR is independent on  
164 the mixing procedure.

### 165 *3.2 Workability Comparison of WARs*

166 As aforementioned, the workabilities of WARs were measured by their rotational  
167 viscosities (Figure 2a), the air void contents of corresponding Marshall Specimen  
168 (Figure 2b) and the number of gyrations of corresponding SGC samples to achieve the  
169 same specimen height (Figure 2c). According to Figure 2a, both Sasobit and paraffin  
170 wax were effective in reducing the viscosities of AR at all three testing temperatures.  
171 For both WMA additives, the mixing procedure had insignificant influence on  
172 rotational viscosity values.

173 Figure 2b shows the air void contents of the prepared Marshall Specimens, which  
174 indicate that only the mixtures with ARS and ARW achieved similar air voids in  
175 comparison to hot AR mixtures when the mixing and compaction temperatures were  
176 16 °C lower. Besides, unlike the rotational viscosity test results, the air void content  
177 results illustrated significant effect of mixing procedure. Under the same preparation  
178 condition, the air void contents of the Marshall Specimens with ARSD and ARWD  
179 are 1.3% and 0.7% higher than those of ARS and ARW, respectively, indicating that  
180 WARs prepared by direct mixing procedure had worse workability. Similar finding  
181 can also be obtained from the number of gyrations of SGC samples (Figure 2c). The

182 mixtures with ARSD and ARWD required more gyration numbers to achieve the same  
183 sample height as those with ARS and ARW. Since the air void contents and number of  
184 gyrations can take into the effect of aggregate-asphalt interaction during mixing on  
185 workability, while rational viscosity cannot, they are believed to be better indicators for  
186 workability of WARs.

### 187 *3.3 Rheological Properties of Extracted Liquid Phase of WARs*

188 The rheological test results have shown limited difference among the WARs prepared  
189 by different mixing procedures, regardless of the type of WMA additives. However,  
190 the effects of different mixing procedures on the performance of the extracted liquid  
191 phases are obvious (Figure 3). Figure 3a compares the failure temperatures of  
192 L-WARs and their corresponding WARs, which were determined as the temperatures  
193 when their rolling thin film oven (RTFO) aged samples have a  $G/\sin\delta$  value of 2.2KPa.  
194 In general, WARs have 8-10 °C higher failure temperatures compared with their  
195 corresponding liquid phases. This is because the CRM particles may act as fillers in  
196 rheological asphalt system, which increase the complex shear modulus and thus  
197 enhance the rutting resistance. For ARS and ARSD, the failure temperature difference  
198 was less than 1 °C. However, the difference between L-ARS and L-ARSD was 2.2 °C.  
199 Similarly, ARW and ARWD had close failure temperatures while L-ARW has 1.9 °C  
200 lower failure temperature compared with L-ARWD. The effect of mixing procedure  
201 on L-WARs seems more significant.

202 Figure 3b, 3c and 3d illustrate the results of phase angle, viscous modulus and elastic

203 modulus. L-ARS has larger viscous modulus, similar elastic modulus and larger phase  
204 angle compared with L-ARSD, while L-ARW has lower modulus and higher phase  
205 angle than L-ARWD. According to the previous studies, Sasobit leads to higher  
206 modulus and lower phase angle, while 56<sup>#</sup> paraffin wax has the opposite effect [11]  
207 and [13]. One possible reason is that there may be more wax additives in the liquid  
208 phase of ARS and ARW, compared with their corresponding ARSD and ARWD. The  
209 existence of CRM may narrow the difference in rheological test results of WARs. But  
210 once the CRM is removed, the distinction caused by mixing procedure becomes more  
211 noticeable.

212 Figure 4 presents the viscosity test results of the extracted liquid phases. It can be  
213 observed that at both 135 and 160 °C, the viscosity values of L-ARS and L-ARW  
214 were less than 2/3 of L-ARSD's and L-ARWD's values, respectively. Both Sasobit  
215 and 56<sup>#</sup> paraffin wax could enhance the flowability of asphalt binder, and the  
216 enhancement effect is more significant with a higher wax content [2], [8], and [13].  
217 Therefore, it is believed that the direct mixing method results in lower wax content in  
218 the liquid phase of WARs.

### 219 *3.4 Chemical Analysis of the Extracted Liquid Phase*

220 Chemical tests were conducted to further verify the difference of wax amount in  
221 liquid phase of WARs. The thermal behaviors of L-WARs are shown in Figure 5. In  
222 the DSC tests, the differences in heat flow between the testing materials and the  
223 reference sample (an empty aluminum pan in this study) were monitored. The peaks

224 in the DSC curves reveal either endothermic behaviors, such as melting and  
225 evaporating, or exothermic behaviors, such as cross-linking and oxidation. Figure 5  
226 shows that the DSC curve of L-AR is relatively smooth within the range between -20  
227 and 140 °C. L-ARS and L-ARSD exhibit two characteristic peaks with maximum  
228 melting temperatures around 100 °C and 110 °C, which is attributed to the mixture of  
229 linear long-chain aliphatic hydrocarbons with those melting temperatures in Sasobit.  
230 L-ARW and L-ARWD show only one characteristic peak at about 57 °C, which is  
231 very close to the melting point of 56<sup>#</sup> paraffin wax. The thermal test results  
232 demonstrate that there are certain amounts of wax additives in all WARs. Besides, it is  
233 noticed that the normalized heat flow of L-WARs prepared by the traditional  
234 procedure is higher, which verifies that there are more wax in L-ARS and L-ARW  
235 than in L-ARSD and L-ARWD. The findings of the thermal analysis support that the  
236 direct mixing procedure results in lower wax content in the liquid phases of WARs,  
237 which is consistent with the findings of the rheological tests results.

238 The wax content results of L-AR and L-WARs, tested according to the European  
239 standard method EN 12606-1, is shown in Figure 6. It is noted that L-AR has lower  
240 paraffin compared with base binder, indicating the wax absorption effect of crumb  
241 rubber. All L-WARs have higher wax content than L-AR and Pen 60/70, which is due  
242 to the incorporation of wax additives. Consistent with the DSC results, L-ARS and  
243 L-ARW contain more wax than their corresponding L-ARSD and L-ARWD.

244 In GPC studies of asphalt, the asphalt binder constituents are generally classified into

245 several groups according to the molecular weight [28], [29], [30] and [31]. In this  
246 study, the GPC chromatogram was divided into three parts according to the  
247 occurrence of peaks. The large molecular size (LMS), medium molecular size (MMS)  
248 and small molecular size (SMS) were defined corresponding to the earliest part, the  
249 middle part and the latest part, respectively. Table 2 presents the molecular weight  
250 distributions of L-AR and L-WARs.

251 Sasobit is a crystalline, long-chain aliphatic polymethylene hydrocarbon with carbon  
252 chain length ranging from C45 to C100 plus, while conventional macrocrystalline  
253 paraffin waxes have carbon chain lengths ranging from C25 to C70. According to  
254 literature, the average molecular weight of Sasobit and 56<sup>#</sup> paraffin wax are  
255 1000-1200 g/mol and 400-500 g/mol, respectively [32] and [33]. Therefore, based on  
256 the molecular weight data in Table 2, Sasobit and 56<sup>#</sup> paraffin wax molecules should  
257 belong to MMS and SMS, respectively. Figure 7 compares the molecular weight  
258 distributions of different WARs. L-ARS was found to have higher percentage of  
259 MMS than L-ARSD, while L-ARW had higher percentage of SMS compared with  
260 L-ARSD, which are consistent with the findings of the DSC tests, i.e., there are more  
261 wax additives in the liquid phases of WARs prepared by the traditional procedure.  
262 Besides, it can be noticed that the LMS percentages of ARSD and ARWD are higher  
263 than those of their corresponding ARS and ARW, possibly due to the more complete  
264 dissolution of CRM polymers.

## 265 **4. Discussion**

### 266 *4.1 Feasibility of Adding WMA Additives at an Earlier Stage*

267 Table 3 provides a summary on the similarity and difference of the two mixing  
268 procedures, based on the test results of this study. It was found that the mixing  
269 procedure did affect the interaction among various components of WAR and thus its  
270 final performance. Compared with the traditional mixing method, the direct mixing  
271 method allows for longer interaction time for wax additive and other components.  
272 Despite the lower interaction temperature, the longer interaction time promoted the  
273 penetration of wax additives into CRM, as evidenced by the wax content test results.  
274 Rheological analysis on WARs showed almost no difference between different mixing  
275 procedures, because the effect of CRM is more dominant. Finally, since the direct  
276 mixing method was found to compromise the workability of AR, it is not  
277 recommended to replace the conventional mixing procedure.

### 278 *4.2 Appropriate Method to Measure Workability of Asphalt Rubber*

279 In this study, the rotational viscosity test results were found to contradict to the results  
280 of the air void content and number of gyrations measurement. To measure the  
281 rotational viscosity of asphalt binder, the commonly used spindle is the number 27<sup>#</sup>  
282 and the volume of asphalt sample is 10.5 ml. After interaction with base binder, CRM  
283 particles swell to three to five times of their original volumes by absorbing the light  
284 fractions of asphalt [4] and [34]. As a result, the test AR sample cannot be treated as a  
285 simple Newton fluid anymore, because of the solid rubber particles inside. Besides,

286 the space for liquid asphalt among the chamber wall, spindle wall and CRM particles  
287 is very limited (Figure 8). During the testing, the CRM particles may produce  
288 resistance to the rotation of the spindle, demanding additional torque to maintain the  
289 constant rotational speed. As a result, the viscosity difference of liquid phases might  
290 be masked by the particle effect.

291 In addition, the size effect of insoluble CRM particles may lead to different speeds  
292 between liquid asphalt phase and the CRM particles when pumping and mixing the  
293 binders with aggregates. The size of CRM is larger than part of the fine aggregates  
294 and fillers even before swelling, and the density difference makes CRM unable to  
295 maintain the same moving speed with asphalt and aggregate. Therefore, the relative  
296 movement among CRM, liquid asphalt and aggregate is very complicated. Therefore,  
297 the rotational viscosity test is not recommended to be conducted directly on AR to  
298 evaluate its workability. Instead, it is worth to further investigate whether the  
299 viscosity value of the liquid phase of AR can better describe its workability.

## 300 **5. Findings and Recommendations**

301 In this study, a series of rheological tests and chemical analyses were conducted on  
302 AR and wax-additive-based WAR binders to characterize the effects of different  
303 mixing procedures. Based on the outcome of this study, the following findings and  
304 recommendations have been obtained:

- 305 1. Sasobit enhances the high-temperature performance of AR binder while 56<sup>#</sup>  
306 paraffin wax has the opposite effect. Both additives negatively affect the  
307 intermediate-temperature fatigue performance.
- 308 2. For WAR binders with either Sasobit or 56<sup>#</sup> paraffin wax, the effect of the  
309 mixing procedure on their mechanical performance is insignificant.
- 310 3. The direct mixing method leads to poorer workability of WAR compared with  
311 the traditional method. But the rotational viscosity test cannot effectively  
312 detect such difference.
- 313 4. The mixing procedure affects the interaction among the components of WARs.  
314 The wax content of the liquid phase of the WARs prepared by the direct  
315 mixing method is lower than that prepared by the conventional method.
- 316 5. It is not recommended to replace the traditional mixing method with the direct  
317 mixing method because of the compromised workability of WAR.
- 318 6. Further study on a more appropriate workability index for AR binders is  
319 recommended.

## 320 **Acknowledgements**

321 The authors sincerely acknowledge the funding support from the Hong Kong  
322 Research Grants Council (Project Number: 539113). Trademark or manufacturers'  
323 names appear in this paper only because they are considered essential to the object of  
324 this paper.



325 **References**

- 326 [1] ASTM Standard D6114, 2009. Standard Specification for Asphalt-Rubber Binder.  
327 American Society for Testing and Materials. West Conshohocken, PA, USA.
- 328 [2] Shu, X., & Huang, B. (2014). Recycling of waste tire rubber in asphalt and  
329 Portland cement concrete: an overview. *Construction and Building Materials*, 67,  
330 217-224.
- 331 [3] Liu, Y., Han, S., Zhang, Z., & Xu, O. (2012). Design and evaluation of  
332 gap-graded asphalt rubber mixtures. *Materials & Design*, 35, 873-877.
- 333 [4] Gawel, I., Stepkowski, R., & Czechowski, F. (2006). Molecular interactions  
334 between rubber and asphalt. *Industrial & Engineering Chemistry Research*, 45(9),  
335 3044-3049.
- 336 [5] Ghavibazoo, A., Abdelrahman, M., & Ragab, M. (2016). Changes in composition  
337 and molecular structure of asphalt in mixing with crumb rubber modifier. *Road  
338 Materials and Pavement Design*, 1-14.
- 339 [6] Yu, H., Leng, Z., & Gao, Z. (2016). Thermal analysis on the component  
340 interaction of asphalt binders modified with crumb rubber and warm mix  
341 additives. *Construction and Building Materials*, 125, 168-174.
- 342 [7] Moreno, F., Sol, M., Martín, J., Pérez, M., & Rubio, M. C. (2013). The effect of  
343 crumb rubber modifier on the resistance of asphalt mixes to plastic  
344 deformation. *Materials & Design*, 47, 274-280.
- 345 [8] Oliveira, J. R., Silva, H. M., Abreu, L. P., & Fernandes, S. R. (2013). Use of a

- 346 warm mix asphalt additive to reduce the production temperatures and to improve  
347 the performance of asphalt rubber mixtures. *Journal of Cleaner Production*, 41,  
348 15-22.
- 349 [9] Rodríguez-Alloza, A. M., Gallego, J., Pérez, I., Bonati, A., & Giuliani, F. (2014).  
350 High and low temperature properties of crumb rubber modified binders  
351 containing warm mix asphalt additives. *Construction and Building Materials*, 53,  
352 460-466.
- 353 [10] Yu, X., Wang, Y., & Luo, Y. (2012). Effects of types and content of warm-mix  
354 additives on CRMA. *Journal of Materials in Civil Engineering*, 25(7), 939-945.
- 355 [11] Xiao, F., Punith, V. S., & Amirkhanian, S. N. (2012). Effects of non-foaming  
356 WMA additives on asphalt binders at high performance temperatures. *Fuel*, 94,  
357 144-155.
- 358 [12] Kim, H. H., & Lee, S. J. (2015). Effect of crumb rubber on viscosity of  
359 rubberized asphalt binders containing wax additives. *Construction and Building*  
360 *Materials*, 95, 65-73.
- 361 [13] Yu, H., Leng, Z., Xiao, F., & Gao, Z. (2016). Rheological and chemical  
362 characteristics of rubberized binders with non-foaming warm mix additives.  
363 *Construction and Building Materials*, 111, 671-678.
- 364 [14] Yu, H., Leng, Z., Zhou, Z., Shih, K., Xiao, F., & Gao, Z. (2017). Optimization of  
365 preparation procedure of liquid warm mix additive modified asphalt rubber.  
366 *Journal of Cleaner Production*, 141, 336-345.

- 367 [15]Rubio, M. C., Martínez, G., Baena, L., & Moreno, F. (2012). Warm mix asphalt:  
368 an overview. *Journal of Cleaner Production*, 24, 76-84.
- 369 [16]Jamshidi, A., Hamzah, M. O., Kurumisawa, K., Nawa, T., & Samali, B. (2016).  
370 Evaluation of sustainable technologies that upgrade the binder performance grade  
371 in asphalt pavement construction. *Materials & Design*, 95, 9-20.
- 372 [17]Liu, J., Saboundjian, S., Li, P., Connor, B., & Brunette, B. (2011). Laboratory  
373 evaluation of sasobit-modified warm-mix asphalt for Alaskan conditions. *Journal*  
374 *of Materials in Civil Engineering*, 23(11), 1498-1505.
- 375 [18]ASTM Standard D5, 2013. Standard Test Method for Penetration of Bituminous  
376 Materials. West Conshohocken, PA, USA.
- 377 [19]ASTM Standard D36, 2006. Standard Test Method for Softening Point of  
378 Bitumen (Ring-and-Ball Apparatus). West Conshohocken, PA, USA.
- 379 [20]ASTM Standard D113, 2007. Standard Test Method for Standard Test Method for  
380 Ductility of Bituminous Materials. American Society for Testing and Materials.  
381 West Conshohocken, PA, USA.
- 382 [21]AASHTO Standard M320, 2010. Standard Specification for Performance-Graded  
383 Asphalt Binder. Washington, DC, USA.
- 384 [22]Bahia, H. U., & Davies, R. (1994). Effect of crumb rubber modifiers (CRM) on  
385 performance related properties of asphalt binders. *Asphalt paving technology*, 63,  
386 414-434.
- 387 [23]Mo, L., Shu, D., Li, X., Huurman, M., & Wu, S. (2012). Experimental

- 388 investigation of bituminous plug expansion joint materials containing high  
389 content of crumb rubber powder and granules. *Materials & Design*, 37, 137-143.
- 390 [24]AASHTO Standard T316, 2013. Standard Method of Test for Viscosity  
391 Determination of Asphalt Binder Using Rotational Viscometer. Washington, DC,  
392 USA.
- 393 [25]AASHTO Standard M312, 2015. Standard Method of Test for Preparing and  
394 Determining the Density of Asphalt Mixture Specimens by Means of the  
395 Superpave Gyrotory Compactor. Washington, DC, USA.
- 396 [26]AASHTO Standard T166, 2013. Standard Method of Test for Bulk Specific  
397 Gravity (Gmb) of Compacted Hot Mix Asphalt (HMA) Using Saturated  
398 Surface-Dry Specimens. Washington, DC, USA.
- 399 [27]European Standard EN 12606-1. Bitumen and bituminous binders -determination  
400 of the paraffin wax content – Part 1: method by distillation.
- 401 [28]Zhao, S., Bowers, B., Huang, B., & Shu, X. (2013). Characterizing rheological  
402 properties of binder and blending efficiency of asphalt paving mixtures  
403 containing RAS through GPC. *Journal of Materials in Civil Engineering*, 26(5),  
404 941-946.
- 405 [29]Yang, S. H., & Lee, L. C. (2016). Characterizing the chemical and rheological  
406 properties of severely aged reclaimed asphalt pavement materials with high  
407 recycling rate. *Construction and Building Materials*, 111, 139-146.
- 408 [30]Kim, S., Lee, S. H., Kwon, O., Han, J. Y., Kim, Y. S., & Kim, K. W. (2016).

- 409 Estimation of service-life reduction of asphalt pavement due to short-term ageing  
410 measured by GPC from asphalt mixture. *Road Materials and Pavement*  
411 *Design*, 17(1), 153-167.
- 412 [31]Zhao, S., Huang, B., Shu, X., & Woods, M. E. (2016). Quantitative evaluation of  
413 blending and diffusion in high RAP and RAS mixtures. *Materials & Design*, 89,  
414 1161-1170.
- 415 [32]Qin, Q., Farrar, M. J., Pauli, A. T., & Adams, J. J. (2014). Morphology, thermal  
416 analysis and rheology of Sasobit modified warm mix asphalt binders. *Fuel*, 115,  
417 416-425.
- 418 [33]Jamshidi, A., Hamzah, M. O., & You, Z. (2013). Performance of warm mix  
419 asphalt containing Sasobit®: State-of-the-art. *Construction and Building*  
420 *Materials*, 38, 530-553.
- 421 [34]Dong, D., Huang, X., Li, X., & Zhang, L. (2012). Swelling process of rubber in  
422 asphalt and its effect on the structure and properties of rubber and asphalt.  
423 *Construction and Building Materials*, 29, 316-322.
- 424

## 425 **List of Figures and Tables**

### 426 *Figures:*

427 Figure 1. Rheological Properties of WARs: (a) Penetration; (b) Softening point and Ductility;  
428 (c) Superpave rutting parameter; and (d) Superpave fatigue parameter

429 Figure 2. Workability Evaluation of WARs: (a) Rotational Viscosity; (b) Air voids of Marshall  
430 Specimens; (c) Number of Gyration of SGC Specimens

431 Figure 3. Rheological Properties of L-WARs: (a) Failure temperature; (b) Phase Angle; (c)  
432 Viscous Modulus; and (d) Elastic Modulus

433 Figure 4. Rotational Viscosity of L-WARs

434 Figure 5. Thermal Behavior of DSC L-WARs (heat flow was normalized by the sample  
435 weight)

436 Figure 6. Wax content test results

437 Figure 7. GPC test result: (a) Typical curve of L-ARS and L-ARSD; (b) Molecular size  
438 distribution of L-ARS and L-ARSD; (c) Typical curve of L-ARW and L-ARWD; and (d)  
439 Molecular size distribution of L-ARW and L-ARWD

440 Figure 8. Diagram of Rotational Viscosity Measurement

### 441 *Tables:*

442 Table 1 Description of Prepared Binders

443 Table 2 Molecular Weight Distributions of L-AR and L-WARs

444 Table 3 Comparisons between Two Mixing Procedures

445

446

447

448

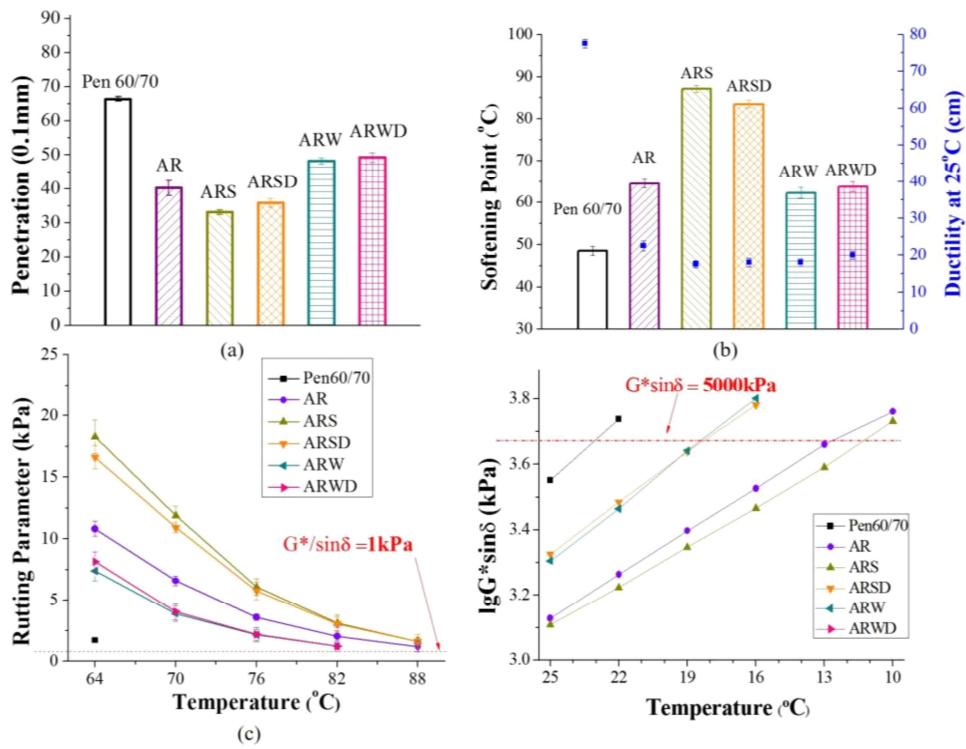
449

450

451

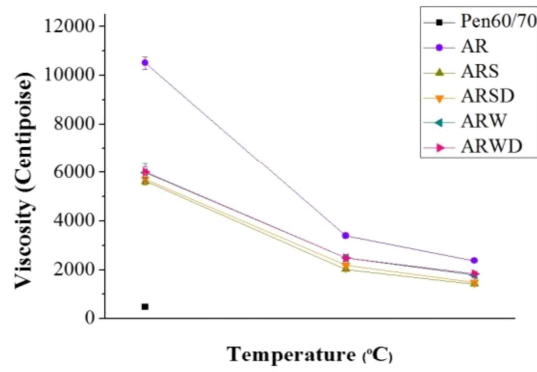
452

453

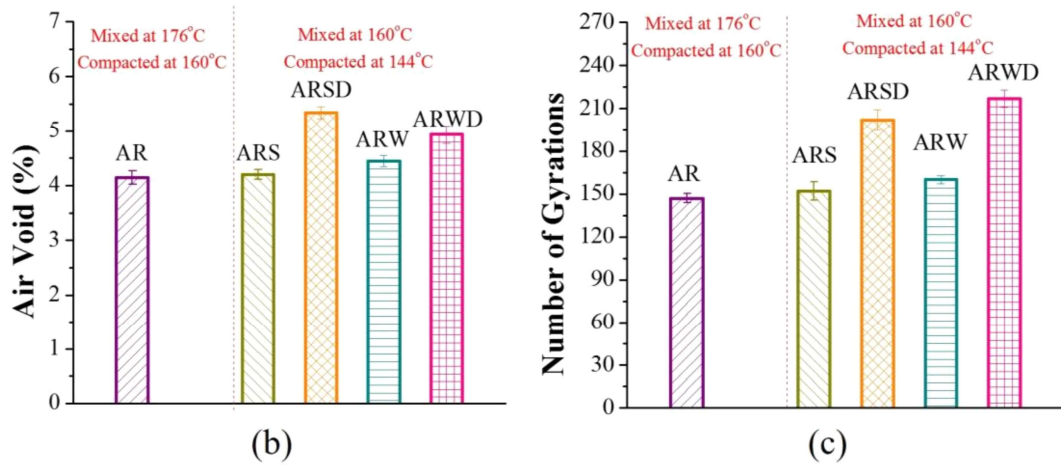


454

455 **Figure 1.** Rheological Properties of WARs: (a) Penetration; (b) Softening point and Ductility;  
 456 (c) Superpave rutting parameter; and (d) Superpave fatigue parameter



(a)



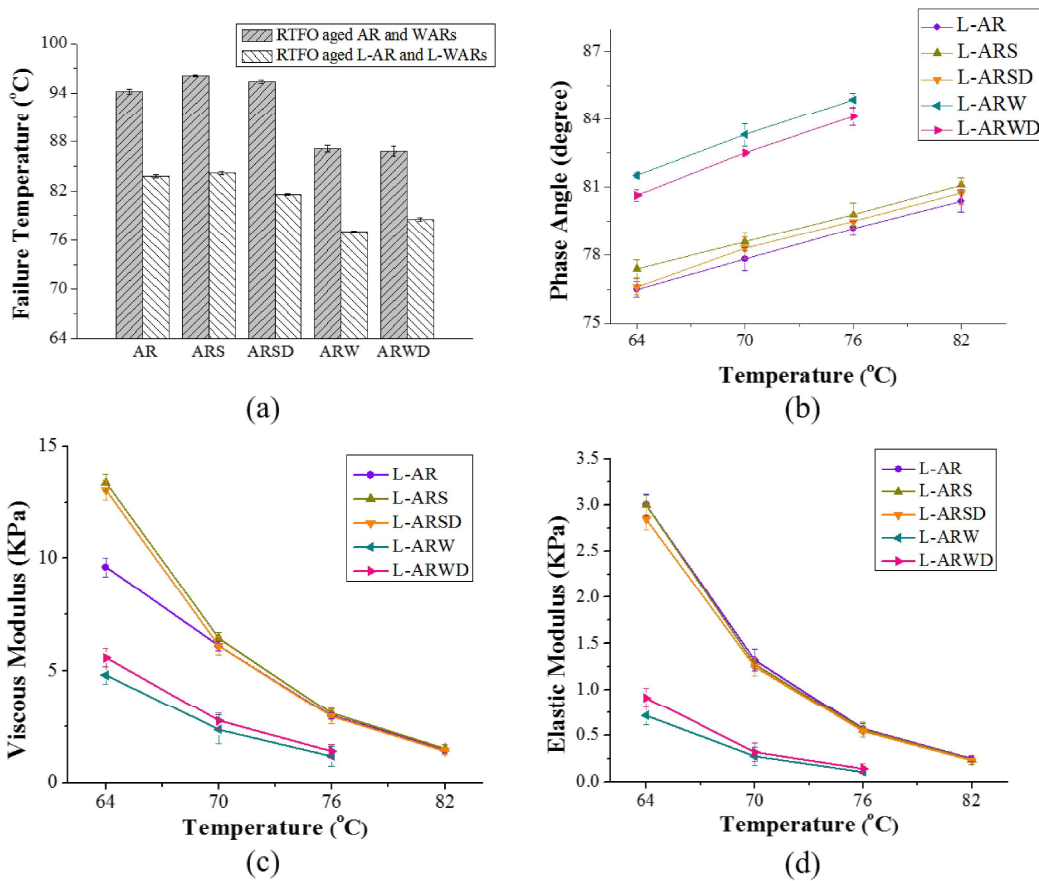
(b)

(c)

457

458 **Figure 2.** Workability Evaluation of WARs: (a) Rotational Viscosity; (b) Air voids of  
 459 Marshall Specimens; (c) Number of Gyration of SGC Specimens





460

461 **Figure 3.** Rheological Properties of L-WARs: (a) Failure temperature; (b) Phase Angle; (c)

462 Viscous Modulus; and (d) Elastic Modulus

463

464

465

466

467

468

469

470

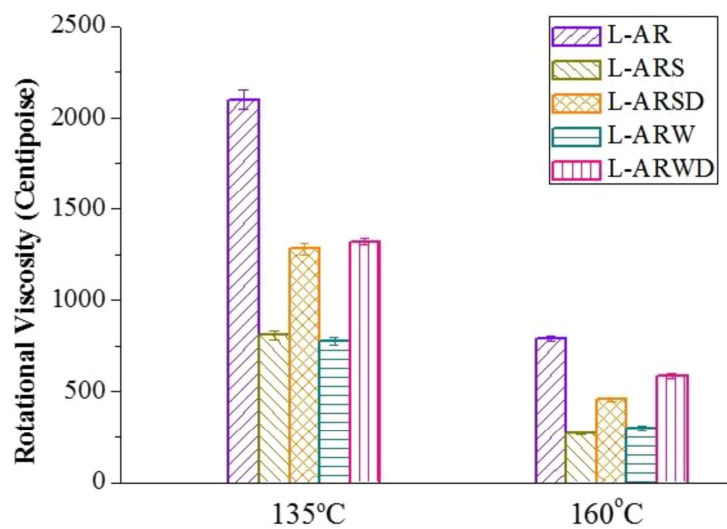


Figure 4. Rotational Viscosity of L-WARs

471

472

473

474

475

476

477

478

479

480

481

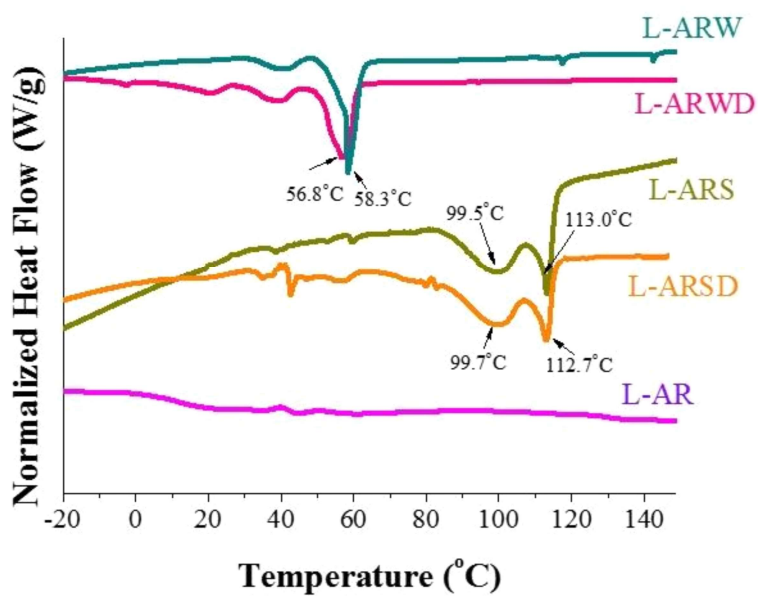
482

483

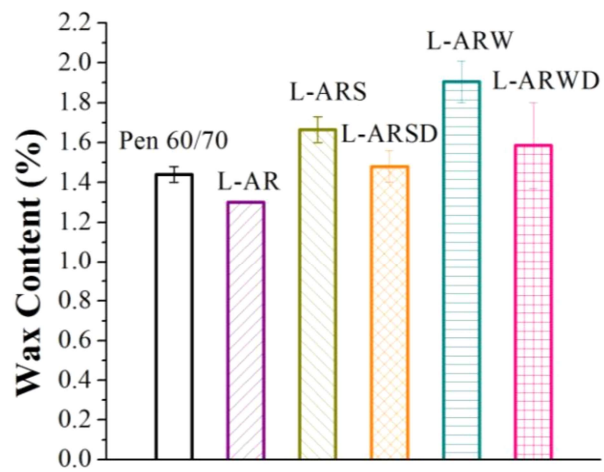
484

485

486



487  
488 **Figure 5.** DSC results of L-WARs (heat flow was normalized by the sample weight)  
489  
490  
491  
492  
493  
494  
495  
496  
497  
498  
499  
500  
501  
502  
503  
504  
505  
506  
507  
508  
509  
510  
511  
512  
513  
514



**Figure 6.** Wax content test results

515

516

517

518

519

520

521

522

523

524

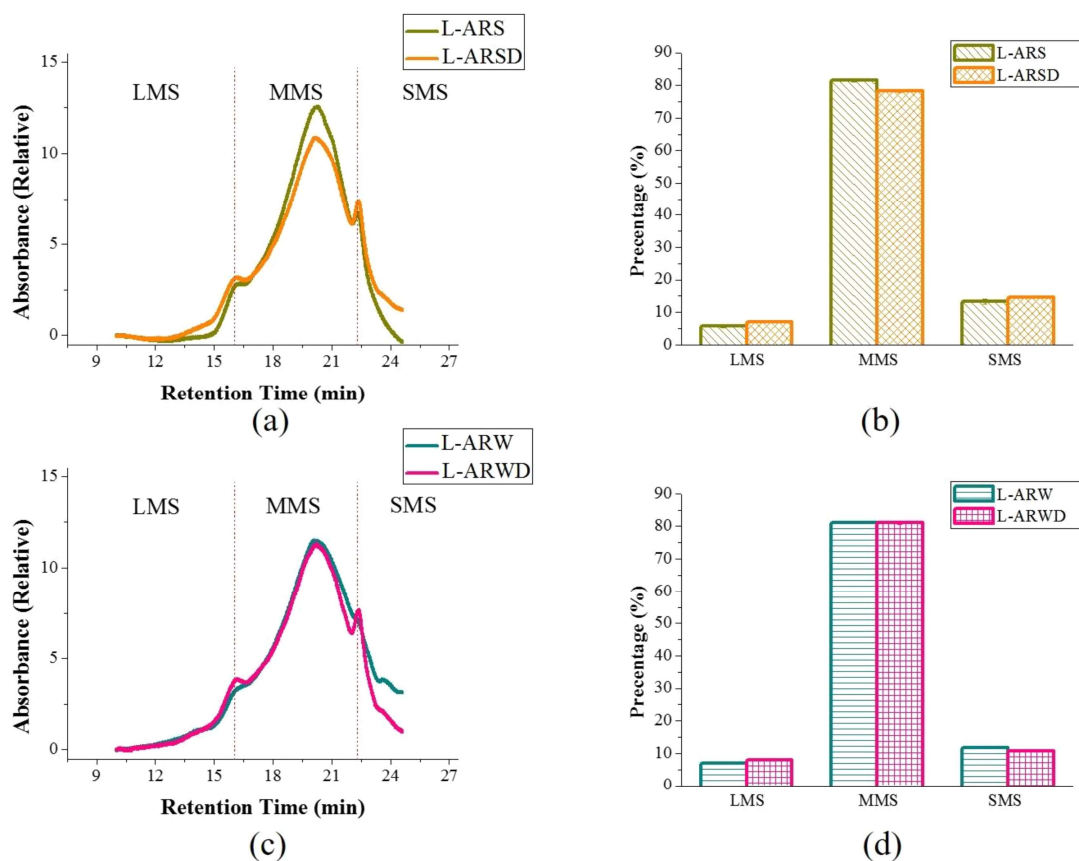
525

526

527

528

529



530

531 **Figure 7.** GPC test result: (a) Typical curve of L-ARS and L-ARSD; (b) Molecular size

532

distribution of L-ARS and L-ARSD; (c) Typical curve of L-ARW and L-ARWD; and (d)

533

Molecular size distribution of L-ARW and L-ARWD

534

535

536

537

538

539

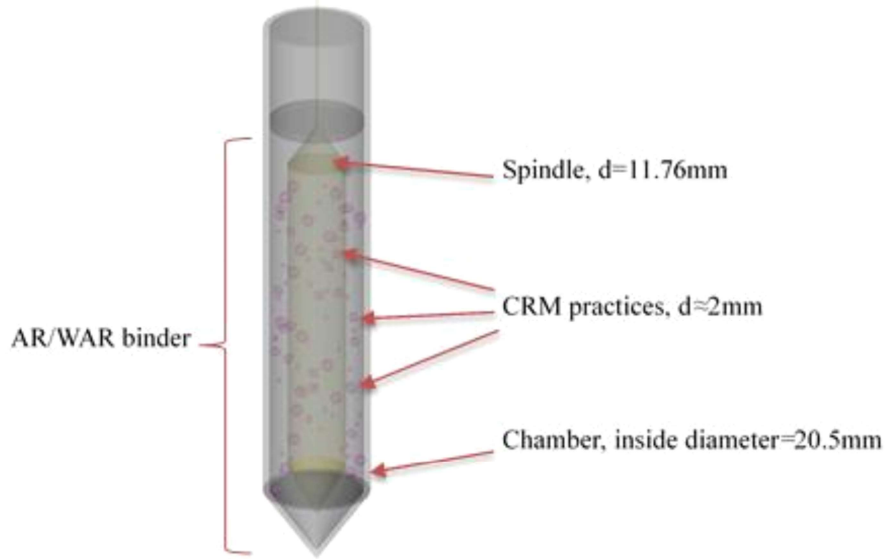
540

541

542

543

544



545

546

547

**Figure 8.** Diagram of Rotational Viscosity Measurement

548

549

550

551

552

553

554

555

556

557

558

559

560

561

562

563

**Table 1** Description of Prepared Binders

Sample ID	Description
Pen 60/70	Base binder, obtained from Anderson Co., Ltd, Hong Kong
AR	Blending 18% of 40-mesh crumb rubber by the total weight of AR with base asphalt at 176 °C and 4000 rpm /min for one hour using a high shear mixer
ARS	Adding 3% of Sasobit into AR binder and high shear mixing for 10 minutes at 160 °C right after the mixing process of AR
ARW	Adding 1.5% of 56 <sup>#</sup> paraffin wax into AR binder and high shear mixing for 10 minutes at 160 °C right after the mixing process of AR
ARSD	Directly high shear mixing Sasobit, crumb rubber and base binder together (same mass ratio as ARS) at 160 °C for one hour
ARWD	Directly high shear mixing 56 <sup>#</sup> paraffin wax, crumb rubber and base binder together (same mass ratio as ARW) at 160 °C for one hour

564

565

566

567

568

569

570

571

572

573

574

575

576

577

578

579

580

581

582

583

584

585

586

587

588

589

590

591

592

**Table 2** Molecular Weight Distributions of L-AR and L-WARs

L-AR						
Peak No.	Retention Time (min)	Area%	Mn	Mw	Mz	Mw/Mn
1 (LMS)	16.19	8.10	1.11E+04	2.15E+04	9.83E+04	1.94
2 (MMS)	20.16	81.11	1.37E+03	1.81E+03	2.52E+03	1.33
3 (SMS)	22.14	10.79	4.26E+02	4.63E+02	4.92E+02	1.09
ALL		100.00	1.17E+03	3.26E+03	5.36E+04	2.78
L-ARS						
1 (LMS)	16.52	6.46	1.09E+04	1.65E+04	3.62 E+04	1.52
2 (MMS)	20.14	82.07	1.38 E+03	1.84E+03	2.60 E+03	1.34
3 (SMS)	22.10	11.48	4.32 E+02	4.73E+02	5.03 E+02	1.09
ALL		100	1.15 E+03	2.63E+03	1.62 E+03	2.28
L-ARSD						
1 (LMS)	16.20	7.05	1.17E+04	1.81E+04	4.08 E+04	1.54
2 (MMS)	20.22	78.26	1.4E+03	1.91E+03	2.67 E+03	1.35
3 (SMS)	22.37	14.69	4.13E+02	4.63E+02	5.04 E+02	1.1
ALL		100.00	1.13E+03	3.1E+03	2.2 E+03	2.7
L-ARW						
1 (LMS)	16.19	7	1.01 E+04	1.71E+04	1.15 E+04	1.70
2 (MMS)	20.16	81.21	1.38 E+03	1.84E+03	2.56 E+03	1.33
3 (SMS)	22.14	11.89	4.30 E+02	4.68E+02	4.97 E+02	1.09
ALL		100	1.15 E+03	2.77E+03	5.21 E+03	2.40
L-ARWD						
1 (LMS)	16.22	8.1	1.11 E+04	2.15E+04	9.83 E+04	1.94
2 (MMS)	20.12	81.11	1.37 E+03	1.81E+03	2.52 E+03	1.33
3 (SMS)	22.28	10.79	4.26 E+02	4.63E+02	4.92 E+02	1.09
ALL		100	1.17 E+03	3.26E+03	5.36 E+03	2.78

593 \*Area%= the percentage of molecules within specific weight range

594 Mn= number-average molecular weight (g/mol, daltons)

595 Mw=weight-average molecular weight (g/mol)

596 Mz=z-average molecular weight (g/mol)

597 Mw/Mn=polydispersity index-relative spread in molecular weights

598

599

600

601

602

603

604

605

606



607

**Table 3** Comparisons between Two Mixing Procedures

	Similarity	Difference
Sample preparation	Binders are prepared by same material and equipment.	The direct mixing procedure is more convenient and energy-saving.
Interaction condition	Two mixing procedures provide similar interaction time for CRM and base asphalt.	Direct mixing procedure enables more complete interaction condition for wax additive, but lower temperature for the interaction of CRM and base binder
Performance	Samples prepared by two mixing procedures have similar rutting and fatigue resistance, as well as very close rotational viscosities.	Direct mixing procedure leads to poorer workability when preparing mixture specimens.
Component interaction	In both cases, three components interact at a certain level, providing satisfactory mechanical performance and relatively good workability.	The longer interaction time results in less wax additives in liquid phase of WARs. The wax additives may be degraded or absorbed by CRM.

608

609

610