

Investigating the influence of fine RAP on bituminous mixtures at the mastic scale: viscoelastic analyses and micromechanical modelling

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1 **Investigating the influence of fine RAP on bituminous mixtures at the**  
2 **mastic scale: viscoelastic analyses and micromechanical modelling**

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## 15 **Investigating the influence of fine RAP on bituminous mixtures at the** 16 **mastic scale: viscoelastic analyses and micromechanical modelling**

17 This paper studies the potential effects of RAP fine particles on bituminous mixtures by  
18 means of a laboratory investigation performed at the mastic scale. Bituminous mastics,  
19 characterized by a constant filler to bitumen volume ratio, were prepared by combining  
20 virgin filler and virgin bitumen with several dosages of RAP fine particles. The  
21 influence of RAP type was evaluated by employing two RAP products supplied by  
22 different sources. The rheological behaviour of the mastics was investigated by means  
23 of oscillatory tests carried out with a dynamic shear rheometer. The time-temperature  
24 superposition principle and the generalized self-consistent scheme (GSCS)  
25 micromechanical model were used to explore the role played by RAP at the mastic  
26 scale. Results indicate that the adopted experimental and modelling approach allows a  
27 proper assessment of the effects of RAP on the linear viscoelastic properties of mastics  
28 and of the degree of blending occurring between virgin and RAP binder.

29 **Keywords:** Reclaimed Asphalt Pavement (RAP); bituminous mastic; viscoelasticity;  
30 micromechanics; modelling.

### 31 **Introduction**

32 The use of Reclaimed Asphalt Pavement (RAP) in substitution of virgin aggregates is an  
33 absolute priority in the road construction industry. This is due to the savings in environmental  
34 and economic costs that derive from the avoided exploitation of natural resources and from  
35 the reduction of landfill waste (Arm *et al.* 2017). Past experience has proven that RAP can be  
36 successfully employed in all layers of road pavements, including bitumen-bound courses,  
37 cement-stabilized foundations, unbound sub-bases and improved subgrades (Santagata *et al.*  
38 2013, Farina *et al.* 2017). However, various technical concerns have originated from research  
39 studies focused on the performance properties of bituminous mixtures containing RAP, with  
40 the consequent identification of limits to RAP dosage to be adopted in practice. Several  
41 researchers have reported that for RAP contents higher than 25 %, a specific and  
42 comprehensive mix design is required in order to prevent detrimental effects on pavement

43 performance. In the case of RAP percentages comprised between 15 % and 25 %, a reduction  
44 in the grade of virgin binder may be sufficient to account for the stiffening effect induced by  
45 RAP, while at percentages lower than 15 % a change in binder grade is not considered to be  
46 necessary (McDaniel *et al.* 2002, Copeland 2011).

47 In addition to the effects related to RAP content, one key point that must be  
48 thoroughly taken into consideration during the design of bituminous mixtures, is the  
49 prediction of the degree of blending that occurs at the interface between virgin binder and  
50 aged RAP bitumen (McDaniel *et al.* 2001, Shuai *et al.* 2018). In this regard, three different  
51 scenarios can be conjectured to describe the complex system composed of RAP, virgin  
52 bitumen and mineral aggregates. These refer to the so-called “black rock”, “total blending”  
53 and “partial blending” models. According to the “black rock” model, interactions and  
54 interdiffusion processes between virgin and aged bitumen are neglected. RAP particles,  
55 which are composed of mineral aggregates coated by aged binder, behave as lithic aggregates  
56 in the virgin bituminous matrix. In the sector literature it has been reported that this theory  
57 can be properly used to model the real behavior of bituminous mixtures only when relatively  
58 low RAP contents are used (generally lower than 15 %). On the opposite, in the case of “total  
59 blending” it is assumed that the binder from RAP is completely blended with virgin bitumen.  
60 In the intermediate scenario of the “partial blending” model, the binder from RAP is  
61 considered to be only partially blended with virgin bitumen, with a blending degree that  
62 strongly depends upon the specific features of the employed materials and upon mixture  
63 production processes (McDaniel *et al.* 2001, 2002, Kriz *et al.* 2014, Rinaldini *et al.* 2014,  
64 Shuai *et al.* 2018).

65 The crucial issue of understanding the role played by RAP in bituminous mixtures has  
66 been extensively studied in the laboratory at different scales, ranging from the micro-scale  
67 (binder level) to the macro-scale (mixture level) (Lee *et al.* 1999, Kriz *et al.* 2014, Rinaldini

68 *et al.* 2014, Mangiafico *et al.* 2019). However, since it is widely recognized that mixture  
69 properties are mostly governed by the rheology of the mortar/mastic phase (Craus *et al.* 1978,  
70 Underwood and Kim 2011, Hesami *et al.* 2013, Riccardi *et al.* 2018), a significant number of  
71 research works have also been carried out at the meso-scale (mortar/mastic level), focusing  
72 on the effects due to the presence of RAP in the fine fraction of bituminous mixtures.

73 From the results of an experimental study carried out on bituminous mortars  
74 containing RAP and Reclaimed Asphalt Shingle (RAS) materials, Bahia and Swiertz (2011)  
75 found that both RAP and RAS strongly affect the linear viscoelastic properties of virgin  
76 materials, as proven by stiffness measurements performed at low in-service temperatures.  
77 Riccardi *et al.* (2016) focused on the fatigue properties of mortars containing RAP with the  
78 specific purpose of back-calculating the maximum amount of reclaimed material that can be  
79 added to Stone Matrix Asphalt (SMA) mixtures. In another research work performed at the  
80 mastic scale, Gundla and Underwood (2017) used micromechanical models to predict the  
81 degree of blending between RAP binder and newly added bitumen, thereby observing that  
82 blending decreases as RAP dosage increases. Mannan *et al.* (2015) focused their research on  
83 the performance properties of RAP mastics at different RAP dosages, demonstrating that  
84 RAP mastics exhibit an enhanced rutting resistance. However, it was shown that the use of  
85 RAP fines leads to an embrittlement of mastics, with a corresponding increase of their fatigue  
86 and low-temperature cracking potential.

87 The research work presented in this paper aims to increase the knowledge about the  
88 effects of the presence of RAP in bituminous mixtures, focusing on phenomena occurring at  
89 the meso-scale level. Bituminous mastics, characterized by a constant filler to bitumen  
90 volume ratio, were prepared in the laboratory by combining virgin filler and virgin bitumen  
91 with several dosages of RAP fine particles (composed of filler and aged binder). The effect of  
92 RAP type was also evaluated by considering two RAP products supplied by different sources.

93 The rheological behaviour of the mastics was investigated by means of oscillatory tests  
94 carried out with a dynamic shear rheometer. The time-temperature superposition principle  
95 and the generalized self-consistent scheme (GSCS) micromechanical model were used to  
96 quantify the effects of RAP fine particles on the linear viscoelastic properties of mastics and  
97 the degree of blending between virgin and RAP binder, respectively.

## 98 **MATERIALS AND METHODS**

### 99 *Base materials*

100 The base materials used for the preparation of bituminous mastics included one virgin  
101 bitumen, one virgin filler and two types of RAP fine particles.

102 The virgin bitumen (indicated as BIT), which belonged to the 70/100 penetration  
103 grade, was preliminary subjected to rheological and chemical analyses, as detailed in  
104 (Santagata *et al.* 2015). These included tests for the evaluation of Performance Grade (PG)  
105 and Thin Layer Chromatography (TLC) tests carried out to determine the relative amounts of  
106 saturates, aromatics, resins and asphaltenes (SARA analysis). Based on the overall results of  
107 the rheological characterization, the bitumen was classified as a PG 58-22 binder. From  
108 SARA analysis, the composition of the bitumen was found to correspond to 3.0 % saturates,  
109 65.3 % aromatics, 15.9 % resins, and 15.8 % asphaltenes.

110 The virgin mineral filler was supplied by an Italian limestone quarry, while the two  
111 RAP materials were obtained from an Iranian and an Italian source. RAP fine particles were  
112 drawn from the RAP materials by passing them through the 0.075 mm sieve. The resulting  
113 fine RAP samples, hereinafter indicated as IR-RAP and IT-RAP, included both uncoated  
114 mineral particles and coated clusters of particles containing a combination of aged bitumen  
115 and fine aggregates. Due to the lack of additional information, the particle size distribution of  
116 virgin filler and RAP filler was assumed to be the same.

117 Ignition tests were conducted on the two different fine RAPs, as per ASTM D6307,  
118 with the twofold objective of retrieving filler particles to be subjected to further  
119 characterization tests, and of assessing their bitumen content. This was found to be equal to  
120 11.65 % and 7.84 % (by weight of filler) for IR-RAP and IT-RAP, respectively.

121 Information regarding the chemical composition of the virgin filler and of the mineral  
122 particles recovered from the two types of fine RAP were obtained by making use of X-Ray  
123 Fluorescence (XRF) and X-Ray Diffraction (XRD) tests.

124 From the results presented in Table 1 it can be noticed that the virgin filler was mainly  
125 composed of calcium oxide, with a lower non-negligible amount of silica. On the opposite,  
126 the Iranian RAP filler (extracted from IR-RAP) was primarily constituted of silica with a  
127 lower percentage of calcium oxide, while the Italian RAP filler (coming from IT-RAP) was  
128 characterized by intermediate amounts of the two oxides, with a prevalence of silica. All the  
129 other components tracked in the test were found in percentages that were always lower than  
130 10 %.

131 *[Table 1 near here]*

132 From the XRD graphs shown in Figure 1 it can be observed that calcite was the  
133 predominant element in the virgin mineral filler, while quartz was the main component of  
134 both the Iranian and Italian recovered fillers (coming from IR-RAP and IT-RAP,  
135 respectively). These results indicate that the virgin mineral filler may be more hydrophobic in  
136 comparison to the two fillers retrieved from RAP, with direct implications on interfacial  
137 interactions which can be established between mineral particles and the surrounding bitumen  
138 in mastics (Pasandín *et al.* 2016). It can be also observed that the outcomes of XRD tests are  
139 in good agreement with those obtained from XRF tests (Table 1), with clear correlations  
140 existing between SiO<sub>2</sub> and quartz, and between CaO and calcite.

141 *[Figure 1 near here]*

142 Specific gravity of the three mineral fillers was determined according to ASTM D854.  
143 Filler drawn from the Iranian fine RAP (IR-RAP) exhibited a specific gravity that was similar  
144 to that of the reference filler, equal to 2.757 and 2.760, respectively. On the other hand, filler  
145 extracted from the Italian fine RAP (IT-RAP) was characterized by a significantly higher  
146 value of specific gravity, equal to 2.840.

#### 147 ***Bituminous mastics***

148 Bituminous mastics were prepared in the laboratory by combining fine RAP, virgin filler and  
149 virgin bitumen. For all mastics, the filler to bitumen volume ratio was fixed at 27 %, which  
150 was considered to be a representative value for typical dense graded bituminous mixtures  
151 with 19 mm nominal maximum aggregate size (Gundla and Underwood 2017).

152 Depending upon the case, filler contained in each mastic was composed of a different  
153 combination of virgin filler and filler contained in fine RAP. In particular, volumetric  
154 concentration of RAP filler ( $f_{RAP}$ ) was varied between 0 % (for mastics containing  
155 exclusively virgin filler) and 100 % (for mastics containing only RAP filler). Additional  
156 concentrations considered in the study were set equal to 30 %, 50 %, and 70 %.

157 Mastics were prepared with the five  $f_{RAP}$  values indicated above and by making use of  
158 the two types of fine RAP (IR-RAP and IT-RAP) referred to in the previous section.  
159 Composition of each mastic to be adopted during laboratory preparation was identified by  
160 taking into account the results of the preliminary tests carried out to determine RAP binder  
161 contents and specific gravity of fillers, and by consequently back-calculating the exact  
162 amounts of each component to be employed in order to achieve the target values of filler to  
163 bitumen volume ratio (equal to 27 %) and of volumetric concentration of RAP filler (variable  
164 between 0 % and 100 %).

165 Table 2 synthesizes relevant information on the composition of the nine mastics  
166 considered in the investigation. Listed parameters include the previously mentioned  
167 volumetric concentration of RAP filler ( $f_{RAP}$ , expressed with respect to total filler in the  
168 mastic), volumetric concentration of RAP binder ( $b_{RAP}$ , expressed with respect to total binder  
169 in the mastic), volumetric percentages of the four components present in the mastic (virgin  
170 bitumen, bitumen from RAP, virgin filler, and filler from RAP), and the filler to bitumen  
171 ratio by weight ( $f/b$ ).

172 *[Table 2 near here]*

173 By analysing the data listed in Table 2 it should be underlined that the mastic with  
174  $f_{RAP}$  equal to 0 %, referred to as the virgin mastic (VM), contains exclusively virgin filler and  
175 virgin binder and therefore is representative of standard bituminous mixtures. The mastics  
176 with  $f_{RAP}$  equal to 30 %, 50 % and 70 % (IR-30, IT-30, IR-50, IT-50, IR-70 and IT-70)  
177 represent the cases of bituminous mixtures with medium, high and very high RAP dosages,  
178 respectively. The mastics with  $f_{RAP}$  equal to 100 % (IR-100 and IT-100) are relevant to the  
179 case of full replacement of virgin filler in the mastic, although these materials still require the  
180 addition of virgin binder to attain the reference 27 % filler to bitumen volume ratio. With  
181 respect to the  $f/b$  ratios, it can be noticed that values obtained for the mastics prepared with  
182 IR-RAP are the same as that calculated for the reference virgin mastic (equal to 1), whereas  
183 the mastics containing IT-RAP were characterized by slightly higher values (comprised  
184 between 1.01 and 1.03). This is due to the different specific gravity of the filler recovered  
185 from IT-RAP with respect to those of both the reference filler and the filler retrieved from IR-  
186 RAP.

187 For the preparation of the bituminous mastics, base materials were mixed by making  
188 use of a low-shear mixer at a temperature of 160 °C. Prior to mixing, the virgin filler was  
189 heated for 6 hours at 160 °C, while in the case of RAP the conditioning temperature and

190 duration were set at 110 °C and 2 hours, respectively (McDaniel *et al.* 2001, Gundla and  
191 Underwood 2017). After manually blending the virgin filler with RAP in proper proportions,  
192 the heated bitumen was added to the compound and mixed with a mechanical stirrer for 60  
193 minutes at a speed of 900 rpm. In order to avoid undesired phase separation between bitumen  
194 and filler, blends were poured into metal moulds to obtain beams of 4 mm thickness, and then  
195 stored at low temperature.

### 196 ***Testing***

197 The base bitumen and the nine bituminous mastics considered in the investigation were  
198 subjected to rheological characterization in the linear viscoelastic domain. Temperature-  
199 Frequency Sweep (TFS) tests were carried out by using a dynamic shear rheometer from  
200 Anton Paar Inc. (Physica MCR 302), according to AASHTO T 315. Frequency sweeps were  
201 performed at several temperatures, comprised between 4 °C and 82 °C, with 6 °C increments  
202 between consecutive steps. Thermal conditioning of the specimens was guaranteed by  
203 imposing a conditioning time of 12 minutes before each temperature step. Two log-decades  
204 of angular frequency were covered during testing, from 1 rad/s to 100 rad/s. The parallel-  
205 plate geometry with 8-mm diameter (PP08) and 2-mm gap between plates was used at  
206 temperatures comprised between 4 °C and 34 °C, while 25-mm parallel plates (PP25) with  
207 1-mm gap were used between 34 °C and 82 °C. Shear strain values were varied depending  
208 upon temperature and frequency conditions, thus allowing the rheological properties of the  
209 materials to be always assessed in their linear viscoelastic region. At least two replicates were  
210 performed for each test and average results were used in the following analyses.

## 211 RESULTS AND DISCUSSION

### 212 *Raw data analysis*

213 Black and Cole-Cole plots were constructed from raw data of TFS tests in order to perform  
214 an overall assessment of the linear viscoelastic properties of the mastics and to verify the  
215 applicability of the time-temperature superposition principle. As illustrated in Figure 3a  
216 (Black) and 3b (Cole-Cole), these diagrams provide a graphical representation of the norm of  
217 the complex modulus  $|G^*|$  versus the phase angle  $\delta$  and of the loss modulus  $G''$  versus the  
218 storage modulus  $G'$ , respectively. By comparing the results obtained for the virgin mastic  
219 (VM) with those collected for the mastics prepared with 100 % RAP filler (IR-100 and  
220 IT-100), it can be noticed that all materials exhibited continuous single smooth curves. This  
221 outcome indicates that all the considered materials showed a rheologically simple response,  
222 thus supporting the validity of the time-temperature superposition principle. As expected, it  
223 was also observed that the presence of fine RAP affected the rheological behaviour of the  
224 mastics to a non-negligible extent.

225 When considering the results plotted in the Black space, the curve associated to base  
226 bitumen (BIT) exhibited the typical gradual transition from a glassy to a viscous state. A  
227 similar trend was found for both VM and IT-100, that showed an increase in stiffness and a  
228 decrease of the phase angle when approaching the viscous asymptote. In the case of mastics  
229 containing IR-RAP, it is worth noting that, besides displaying a higher stiffness at high  
230 temperatures (and/or low frequencies), the considered mastic did not show a liquid-like  
231 response. This is proven by the phase angle that did not present a monotonic increase at  
232 decreasing values of the norm of the complex modulus, thus giving rise to a hump in the  
233 Black curves. This behaviour can also be found in TFS tests performed on bituminous  
234 mixtures, in which the binder phase is no longer able to lubricate the lithic skeleton particles  
235 at high temperatures (Ramirez Cardona *et al.* 2015).

236 From an inspection of the raw data plotted in the Cole-Cole plane, it can be seen that  
237 in the range of low temperatures (and/or high frequencies) the presence of IT-RAP led to an  
238 increase in both the storage and the loss moduli. On the other hand, mastics containing IR-  
239 RAP showed lower values of the loss modulus, thus indicating that such a fine RAP mainly  
240 affected the degree of elasticity exhibited by the material, that shifted towards a more elastic  
241 response.

242 *[Figure 2 near here]*

243 For a straightforward comparison of raw data in the same testing conditions, norm and  
244 phase angle values of the complex modulus, normalized with respect to those of the virgin  
245 mastic, are reported in Table 3 and Table 4, respectively. All results refer to an angular  
246 frequency of 10 rad/s and temperatures of 4 °C, 22 °C, 58 °C and 82 °C.

247 *[Table 3 near here]*

248 *[Table 4 near here]*

249 As expected, by increasing fine RAP dosage, an increase in  $|G^*|$  and a decrease in  $\delta$   
250 were generally observed. When considering the ratios of the norm of the complex modulus,  
251 these values were found to be significantly higher at the higher temperatures. On the contrary,  
252 in the case of the phase angle, changes with respect to the reference virgin mastic were more  
253 evident at low temperatures and progressively decreased at increasing temperatures, reaching  
254 a unit value at the highest investigated temperature (the only exception being IR-100 with a  
255 value of 0.98). It is interesting to observe that such trends associated with fine RAP content  
256 are coherent with those reported in previous research studies focused on the effects of  
257 bitumen ageing (Dalmazzo *et al.* 2019). This seems to suggest that the presence of aged  
258 binder coatings around RAP filler particles can have a strong impact on the overall linear  
259 viscoelastic response of mastics.

260 By taking into consideration the effects of the type of fine RAP, it can be clearly  
 261 noticed that changes were always found to be higher in the case of the IR-RAP, probably as a  
 262 consequence of the higher volumetric content of aged bitumen and/or a more severe ageing  
 263 state.

### 264 *Master curves*

265 By making use of the time-temperature superposition principle, values of the norm and phase  
 266 angle of the complex modulus were modelled in the form of master curves. Raw data  
 267 collected from frequency sweep tests carried out at different temperatures were shifted along  
 268 the frequency axis, thus defining continuous rheological functions at a reference temperature  
 269 (set equal to 22 °C). Master curves were constructed using an optimization process by means  
 270 of which experimental data were fitted to the Christensen and Anderson analytical functions  
 271 (Christensen and Anderson 1992). These are presented in Equations 1 and 2, for the norm and  
 272 the phase angle of the complex modulus, respectively:

$$273 \quad |G^*|_{LVE}(\omega_r) = G_g \left[ 1 + \left( \frac{\omega_c}{\omega_r} \right)^{\frac{\text{Log } 2}{R}} \right]^{-\frac{R}{\text{Log } 2}} \quad (1)$$

$$274 \quad \delta_{LVE}(\omega_r) = \frac{90 \text{ m}}{\left[ 1 + \left( \frac{\omega_r}{\omega_c} \right)^{\frac{\text{Log } 2}{R}} \right]} \quad (2)$$

275 where  $G_g$  is the glassy modulus,  $R$  is the rheological index,  $\omega_c$  is the cross over  
 276 frequency, and  $\omega_r$  is the reduced angular frequency defined by Equation 3.

$$277 \quad \omega_R = \omega \cdot a_T \quad (3)$$

278 where  $\omega$  is the physical angular frequency and  $a_T$  is the shift factor between a generic  
 279 test temperature  $T$  and the reference temperature  $T_R$ . Shift factor values at each test  
 280 temperature were directly derived from the optimization process.

281 Master curves of the norm and phase angle of the complex modulus are presented in  
 282 Figure 3 for the base bitumen, the virgin mastic ( $f_{RAP}$  equal to 0 %) and the two mastics with

283  $f_{\text{RAP}}$  equal to 100 %. For the sake of clarity, results obtained for mastics with intermediate  
284 values of  $f_{\text{RAP}}$  were omitted from the representation since a progressive variation in results  
285 was found when increasing the volumetric concentration of RAP filler from 0 % to 100 %.  
286 As expected, it can be seen that the presence of the virgin filler caused a general increase in  
287 stiffness and elasticity over the entire range of reduced angular frequencies. This is clearly  
288 observed when comparing the curves corresponding to VM and BIT. Although to a lesser  
289 extent, the incorporation of fine RAP particles also produced an increase in stiffness and  
290 elasticity, consistently with the outcomes drawn from raw data analysis presented in the  
291 previous section.

292 *[Figure 3 near here]*

293 Curves of the shift factors plotted as a function of temperature are displayed in Figure  
294 4, where the results of the mastics containing IR-RAP and IT-RAP ( $f_{\text{RAP}}$  equal to 100 %) are  
295 compared to those of the virgin mastic and of the base bitumen. From an inspection of the  
296 plot, it can be seen that at temperatures lower than the reference one, changes were non-  
297 negligible only when comparing bitumen and mastics, while a complete overlapping between  
298 mastic curves was found. On the contrary, the time-temperature dependency can be  
299 discriminated at higher temperatures, where the difference between the generic test  
300 temperature and the reference temperature is greater. From a first comparison between base  
301 bitumen and virgin mastic, it was highlighted that the presence of filler particles in the  
302 bituminous matrix caused a reduction in the high-temperature shift factors. The same trend  
303 was observed when analysing the effects of fin RAP in mastics, with a reduction of the  
304 high-temperature shift factors at the higher RAP dosage for both mastics containing IR-RAP  
305 and IT-RAP.

306 *[Figure 4 near here]*

### 307 ***Thermal susceptibility***

308 In order to gain valuable insights into the effects of fine RAP on the rheological behaviour of  
309 mastics, the thermal susceptibility parameter  $\vartheta$  was evaluated as indicated in Equation 4:

$$310 \quad \vartheta = \frac{\text{Log}[|G^*|(4)/|G^*|(82)]}{\text{Log}[a(4)/a(82)]} \quad (4)$$

311 where  $\text{Log}[a(4)/a(82)]$  is the difference between the logarithms of the shift factors  
312 at 4 °C and 82 °C, , and  $\text{Log}[|G^*|(4)/|G^*|(82)]$  is the difference between the logarithms of  
313  $|G^*|$  at 4 °C and 82 °C (at the reference angular frequency of 10 rad/s).

314 The main outputs of the analysis are summarised in Figure 5, where the stiffness  
315 parameter  $\text{Log}[|G^*|(4)/|G^*|(82)]$ , the time-temperature dependency parameter  
316  $\text{Log}[a(4)/a(82)]$ , and the thermal susceptibility parameter  $\vartheta$  are presented as a function of  
317 both volumetric concentration of RAP filler ( $f_{\text{RAP}}$ ) and volumetric concentration of RAP  
318 binder ( $b_{\text{RAP}}$ ). Such a double representation allows the influence of the different phases  
319 present in the mastics to be graphically assessed and compared.

320  $\text{Log}[a(4)/a(82)]$  was calculated to analyse the time-temperature dependency,  
321 regardless of the value of  $T_{\text{R}}$  arbitrarily selected in master curve construction. This parameter,  
322 that in the  $T\text{-Log}[a(T)/a(T_{\text{ref}})]$  plot of Figure 4 quantifies the y-axis extension of the  
323 curves in the constant range of investigated temperatures (from 4 °C to 82 °C), is a measure  
324 of the stretching of master curves in the frequency domain. Hence, higher values of  
325  $\text{Log}[a(4)/a(82)]$  denote higher extensions of master curves in the axis of reduced angular  
326 frequencies.

327 The evolution of  $\text{Log}[a(4)/a(82)]$  as a function of  $f_{\text{RAP}}$  and  $b_{\text{RAP}}$  are presented in  
328 Figure 5a and 5b, respectively. Although a well-defined relationship between time-  
329 temperature dependency and mastic volumetrics cannot be clearly identified in both  
330 representations, it is evident that an increase in RAP content led to a greater widening of the  
331 range of reduced frequencies. Moreover, by comparing  $\text{Log}[a(4)/a(82)]$  values of neat

332 bitumen and virgin mastic (both represented in the graph at 0 % RAP content, corresponding  
333 to 0 % values of both  $f_{RAP}$  and  $b_{RAP}$ ), the effects induced by the presence of mineral particles  
334 in the bituminous matrix can be identified.

335 Coherently with the above described analysis focused on the stretching of the range of  
336 covered frequencies obtained by means of the time-temperature superposition principle,  
337  $Log[|G^*(4)/|G^*(82)]$  was calculated to quantify the extension of the corresponding range  
338 of stiffness values. The parameter  $Log[|G^*(4)/|G^*(82)]$  is presented as a function of  $f_{RAP}$   
339 and  $b_{RAP}$  in Figure 5c and 5d, respectively. It is interesting to observe that when plotting the  
340 stiffness parameter as a function of the volumetric concentration of RAP filler, two distinct  
341 trends can be identified for IR-RAP and IT-RAP. By considering the stiffness parameter as a  
342 function of the volumetric concentration of RAP binder, the differences in the two curves are  
343 definitely reduced.

344 *[Figure 5 near here]*

345 Plots of  $\vartheta$  as a function of  $f_{RAP}$  (Figure 5e) and  $b_{RAP}$  (Figure 5f) reveal information on  
346 the effects caused by the presence of fine RAP on the thermal susceptibility of mastics.  
347 Although in both representations two distinct curves can be identified, it is worth noting that  
348 when plotting thermal susceptibility as a function of the volumetric concentration of RAP  
349 binder, a reduction in the divergence between the two curves was obtained, coherently with  
350 observations made for the stiffness parameter. These results support the idea that the amount  
351 of RAP binder can have a greater impact on the rheology of mastics in comparison to the  
352 amount of RAP filler. Focusing on the residual divergence found between IR-RAP and IT-  
353 RAP curves in the  $b_{RAP}$ -  $\vartheta$  plot, it can be postulated that part of such a discrepancy can derive  
354 from the different RAP filler concentrations associated to constant values of  $b_{RAP}$ . Moreover,  
355 influencing factors can also include the ageing degree of the two RAP binders and the  
356 physicochemical characteristics of the mineral particles composing the different RAP types,

357 that in turn are not the same of those of the virgin mastic. These specific aspects will be  
358 certainly explored in future research studies.

### 359 ***Micromechanical modelling***

360 Further insights into the role played by RAP in bitumen-filler blends were gained by means  
361 of micromechanical modelling. When considering the stiffening effect caused by the presence  
362 of mineral filler in a bituminous matrix, three main reinforcement contributions can be  
363 distinguished: (i) volume-filling reinforcement, related to the presence of rigid inclusions in a  
364 softer matrix; (ii) physiochemical reinforcement, generated by interfacial effects between  
365 filler and bitumen; and (iii) particle-interaction reinforcement, generated by interconnected  
366 contacts between rigid particles which form a networked structure. This last contribution is  
367 assumed to be relevant only at relatively high filler concentrations, well above the reference  
368 value of 27 % considered in this study.

369         Following the approach developed for traditional bituminous mastics by Buttlar et al.  
370 (1999), the stiffening occurring beyond volume filling was quantitatively assessed in all  
371 bitumen-filler systems considered in the investigation. The volumetric concentration of  
372 spherical inclusions theoretically needed to reach the stiffening level experimentally  
373 measured in the laboratory ( $c_{cal}$ ) was at first calculated by modelling the reinforcement  
374 contribution given by volume filling only. Consequently, by subtracting from  $c_{cal}$  the real  
375 concentration of filler particles ( $c$ ), the volumetric concentration of hard inclusions not  
376 associated to aggregate mineral particles ( $V_{imm}$ ) was estimated for the virgin mastic and for  
377 all mastics containing fine RAP.

378         In Figure 6, a schematic representation of the modelled system composed of spherical  
379 inclusions floating in a softer continuous matrix is reported for a virgin mastic (Figure 6a)  
380 and for a mastic containing fine RAP (Figure 6b), both characterized by the same volumetric

381 concentration of mineral filler particles. In neat mastics,  $V_{imm}$  can be envisaged as the  
 382 volumetric concentration of the hard shells of binder around filler particles that are generated  
 383 by physiochemical phenomena, when the effects of particle-interaction reinforcement are  
 384 negligible. In the case of mastics containing fine RAP, it is believed that  $V_{imm}$  is also  
 385 significantly affected by the presence of coatings of aged bitumen around RAP filler  
 386 particles. This is illustrated in Figure 6b, in which a generic system composed of both virgin  
 387 and RAP filler particles is considered.

388 *[Figure 6 near here]*

389 The stiffening effect generated by volume-filling reinforcement was modelled  
 390 according to the generalized self-consistent scheme (GSCS) model (Buttlar *et al.* 1999). The  
 391 corresponding formulation is reported in Equations 5 to 11:

$$392 \quad A \left( \frac{|G^*|_m}{|G^*|_b} \right)^2 + 2B \left( \frac{|G^*|_m}{|G^*|_b} \right) + C = 0 \quad (5)$$

$$393 \quad A = 8 \left( \frac{G_i}{|G^*|_b} - 1 \right) (4 - 5\nu_b) \eta_1 c^{\frac{10}{3}} - 2 \left[ 63 \left( \frac{G_i}{|G^*|_b} - 1 \right) \eta_2 + 2\eta_1 \eta_3 \right] c^{\frac{7}{3}} + 252 \left( \frac{G_i}{|G^*|_b} - 1 \right) \eta_2 c^{\frac{5}{3}} - 50 \left( \frac{G_i}{|G^*|_b} - 1 \right) (7 - 12\nu_b + 8\nu_b^2) \eta_2 c + 4(7 - 10\nu_b) \eta_2 \eta_3 \quad (6)$$

$$395 \quad B = -4 \left( \frac{G_i}{|G^*|_b} - 1 \right) (1 - 5\nu_b) \eta_1 c^{\frac{10}{3}} + 4 \left[ 63 \left( \frac{G_i}{|G^*|_b} - 1 \right) \eta_2 + 2\eta_1 \eta_3 \right] c^{\frac{7}{3}} - 504 \left[ \left( \frac{G_i}{|G^*|_b} - 1 \right) \eta_2 \right] c^{\frac{5}{3}} + 150 \left( \frac{G_i}{|G^*|_b} - 1 \right) (3 - \nu_b) \nu_b \eta_2 c + 3(15\nu_b - 7) \eta_2 \eta_3 \quad (7)$$

$$397 \quad C = 4 \left( \frac{G_i}{|G^*|_b} - 1 \right) (5\nu_b - 7) \eta_1 c^{\frac{10}{3}} - 2 \left[ 63 \left( \frac{G_i}{|G^*|_b} - 1 \right) \eta_2 + 2\eta_1 \eta_3 \right] c^{\frac{7}{3}} + 252 \left( \frac{G_i}{|G^*|_b} - 1 \right) \eta_2 c^{\frac{5}{3}} + 25 \left( \frac{G_i}{|G^*|_b} - 1 \right) (\nu_b^2 - 7) \eta_2 c - (7 + 5\nu_b) \eta_2 \eta_3 \quad (8)$$

$$399 \quad \eta_1 = \left( \frac{G_i}{|G^*|_b} - 1 \right) (49 - 50\nu_i \nu_b) + 35 \frac{G_i}{|G^*|_b} (\nu_i - 2\nu_b) + 35 (2\nu_i - \nu_b) \quad (9)$$

$$400 \quad \eta_2 = 5\nu_i \left( \frac{G_i}{|G^*|_b} - 8 \right) + 7 \left( \frac{G_i}{|G^*|_b} + 4 \right) \quad (10)$$

$$401 \quad \eta_3 = \frac{G_i}{|G^*|_b} (8 - 10\nu_b) + (7 - 5\nu_b) \quad (11)$$

402 where  $|G^*|_m$  and  $|G^*|_b$  are the norm of the complex modulus of mastic and bitumen  
 403 (collected from direct measurements);  $G_i$  is the shear modulus of inclusions (set equal to 24  
 404 GPa);  $\nu_b$  and  $\nu_i$  are the Poisson ratios of bitumen and inclusions (set equal to 0.40 and 0.15,  
 405 respectively).

406 By back-calculating the values of  $c$  that lead to a convergence between predicted and  
407 measured  $|G^*|_m$  data by means of Equations 5 to 11, the effective volumetric concentration of  
408 filler ( $c_{cal}$ ) was computed. With the purpose of taking into account the role of temperature in  
409 stiffening effects, the optimization process was performed separately at each test temperature  
410 by considering only  $|G^*|$  values collected at the reference angular frequency of 10 rad/s.  
411  $V_{imm}$  was then calculated as the difference between the effective volume concentration,  
412 which takes into consideration the spherical inclusions composed of both filler particles and  
413 influenced binder layers, and the actual filler concentration (Equation 12).

$$414 \quad V_{imm} = c_{cal} - c \quad (12)$$

415 The outcomes of micromechanical modelling are summarized in Figure 7a, where  
416  $V_{imm}$  is plotted as a function of the volumetric concentration of RAP binder ( $b_{RAP}$ ). For the  
417 sake of clarity, only results obtained at 4 °C, 10 °C, 22 °C and 58 °C are displayed in the  
418 graph.

419 *[Figure 7 near here]*

420 For values of  $b_{RAP}$  equal to 0 % (which correspond to the case of the virgin mastic), it  
421 can be seen that the stiffening effect associated with interfacial phenomena taking place  
422 between filler and bitumen was of the same order of magnitude of the actual filling  
423 contribution. This was proven by the  $V_{imm}$  values, comprised between 0.14 and 0.20, that are  
424 comparable to the volumetric concentration of filler, equal to 0.27. Moreover, it is interesting  
425 to observe that the value of  $V_{imm}$  depends on test temperature, showing higher values at  
426 increasing temperatures.

427 Regardless of the considered temperature, mastics containing fine RAP showed  
428 superior values of the “fictitious” immobilized binder, coherently with the outcomes of the  
429 overall rheological characterization. By analysing the relation between  $V_{imm}$  and  $b_{RAP}$ , linear  
430 trends were highlighted at each test temperature regardless of RAP origin, with  $V_{imm}$  values

431 approaching those found for the virgin mastic at  $b_{RAP}$  values tending to zero. From a general  
432 overview of the results, it can be stated that the layer of binder immobilized by virgin filler is  
433 significantly smaller than the layer activated by RAP mineral particles. While in the case of  
434 virgin mastics it is believed that the dimension of spherical inclusions is mainly governed by  
435 bitumen-filler interfacial phenomena, in the case of mastics containing fine RAP, these  
436 effects are also mixed with interactions triggered by the aged binder coatings of RAP filler  
437 particles. Moreover, it can be hypothesised that the increased dimensions of the RAP  
438 spherical inclusions can also promote the onset of local particle-to-particle interaction  
439 reinforcement effects.

440         When focusing on the influence of temperature on  $V_{imm}$ , it can be noticed that in line  
441 with the trend highlighted for the virgin mastic (displayed in the plot at  $b_{RAP}$  equal to 0 %), at  
442 increasing temperatures the sensitivity of  $V_{imm}$  on the amount of RAP binder dramatically  
443 increased. This is proven by the  $V_{imm}$  values that, at the maximum value of  $b_{RAP}$ , were  
444 doubled passing from 4 °C to 58 °C. Such a finding can be partially explained by considering  
445 the difference in stiffness between the neat bituminous matrix and the immobilized binder  
446 constituting the rigid shells which cover filler particles. At low temperatures this difference is  
447 expected to be limited since all bituminous materials asymptotically tend to a glassy modulus.  
448 On the other hand, at high temperatures the altered bitumen, generated by interfacial  
449 phenomena and/or ageing, seemed to be less sensitive to temperature variations with respect  
450 to the bitumen composing the matrix phase. This result is also consistent with the reduction in  
451 thermal susceptibility found at increasing  $b_{RAP}$  values (Figure 5f). Based on these  
452 observations, it can be concluded that at low temperatures the effects produced by fine RAP  
453 can be adequately described by the “total blending” model, as revealed by the slight  
454 differences in  $V_{imm}$  values found between virgin mastic and blends containing RAP. On the

455 opposite, by increasing temperature, the response of the mastics containing RAP definitely  
456 deviates from that predicted with a “total blending” approach.

457 In order to further discriminate between the stiffening provided by the RAP aged  
458 binder and other reinforcement effects, the “black rock” hypothesis was also considered.  
459 While according to the “total blending” approach the binder deriving from RAP is treated in  
460 modelling as the binder constituting the matrix phase, in the “black rock” approach the  
461 volume of RAP binder is considered as part of the filler volume concentration. The  
462 corresponding definition of immobilized binder in the “black rock” hypothesis is given in  
463 Equation 13:

$$464 \quad V_{imm} = c_{cal} - (c + b_{RAP}) \quad (13)$$

465 As shown in Figure 7b, by totally excluding RAP binder from the volume of  
466 immobilized binder, further effects related to physiochemical phenomena and other  
467 contributions deriving from contact reinforcement can be highlighted. At high temperatures,  
468  $V_{imm}$  still increased for increasing  $b_{RAP}$  values, thus indicating that in mastics containing  
469 RAP, stiffening effects are not exclusively related to the presence of the hard aged bitumen  
470 coming from RAP. While at intermediate temperatures  $V_{imm}$  was almost constant, at low  
471 temperatures a decrease in  $V_{imm}$  was observed. This last result suggests that the presence of  
472 the aged RAP binder can hinder interfacial effects occurring between the bitumen of the  
473 matrix phase and filler particles, thus confirming a deviation from the “black rock”  
474 hypothesis at low temperatures.

475 In the assumption that mastics containing RAP will exhibit intermediate behaviours  
476 between those depicted in the extreme ideal cases of “total blending” and “black rock”,  
477 results displayed in Figure 7a and Figure 7b represent the range of immobilized binder  
478 volumes that can actually form in any mastic as a function of RAP dosage.

479 **Conclusions**

480 Based on the results presented in this paper, it can be concluded that fine RAP can have a  
481 significant impact on the overall rheological behaviour of mastics. This outcome strongly  
482 suggests that the use of fine RAP in substitution of virgin filler requires a thorough  
483 characterization of base materials in order to avoid detrimental effects on pavement  
484 performance. In such a context, the micromechanical approach adopted in the present study  
485 has proven to be an effective tool to assess the role played by RAP at the mastic scale.

486 Mastics containing RAP always showed higher stiffness and elasticity, which progressively  
487 increased at increasing RAP dosages to an extent that was dependent on RAP type. It was  
488 observed that RAP caused an overall reduction in the thermal susceptibility of mastics, that  
489 was mainly related to the volumetric concentration of RAP binder. However, it is worth  
490 noting that the type of RAP also affected the thermal dependency of mastics, probably as a  
491 consequence of the key role played by the ageing degree of different RAP binders and/or by  
492 the physicochemical characteristics of RAP mineral particles.

493 From the micromechanical modelling it was highlighted that RAP particles generate higher  
494 volume concentrations of fictitious immobilized bitumen, used to quantitatively assess the  
495 stiffening effects in mastics beyond volume filling. While the influence of RAP source was  
496 quite limited, the volume of bitumen behaving as a hard coating around mineral inclusions  
497 was found to be strongly dependent upon temperature, thus indicating the need to perform an  
498 extensive viscoelastic characterization of the composite materials.

499 Further studies are needed in order to support the findings of this research work, possible by  
500 extending the investigation to the macro-scale with an appropriate characterization of the  
501 viscoelastic and damage properties of bituminous mixtures containing different types and  
502 quantities of fine RAP. Modelling can also be improved by adopting a multi-scale approach,

503 with the final goal of fully understanding the implications of RAP use in bituminous  
504 mixtures.

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581         behaviour of asphalt concrete. *International Journal of Pavement Engineering*, 12 (4),  
582         357–370.

583 Table 1. Results of XRF tests carried out on fillers.

Components	Percentages		
	Virgin mineral filler	Iranian RAP filler	Italian RAP filler
SiO <sub>2</sub>	10.651	51.518	35.073
CaO	44.644	15.855	27.095
Al <sub>2</sub> O <sub>3</sub>	1.304	7.359	4.849
Na <sub>2</sub> O	0.048	2.016	0.928
MgO	3.712	3.253	5.087
K <sub>2</sub> O	0.471	2.888	1.169
Fe <sub>2</sub> O <sub>3</sub>	1.006	4.784	4.051
Other components	0.324	2.127	1.848

584

585 Table 2. Composition of mastics.

Fine RAP type	Mastic ID	Volumetric concentration of RAP filler, ( $f_{RAP}$ )	Volumetric concentration of RAP binder, ( $b_{RAP}$ )	Volumetric percentages in bituminous mastic				Filler to bitumen ratio by weight, ( $f/b$ )
				Virgin binder	Binder from RAP	Virgin Filler	Filler from RAP	
-	VM	0	0.0	73.0	0.0	27.0	0.0	1.00
IR- RAP	IR-30	30	4.0	70.1	2.9	18.9	8.1	1.00
	IR-50	50	6.6	68.2	4.8	13.5	13.5	1.00
	IR-70	70	9.2	66.3	6.7	8.1	18.9	1.00
	IR-100	100	13.2	63.4	9.6	0.0	27.0	1.00
IT- RAP	IT-30	30	2.6	71.1	1.9	18.9	8.1	1.01
	IT-50	50	4.4	69.8	3.2	13.5	13.5	1.02
	IT-70	70	6.1	68.5	4.5	8.1	18.9	1.02
	IT-100	100	8.8	66.6	6.4	0.0	27.0	1.03

587 Table 3.  $|G^*|$  ratios for mastics containing IR-RAP (variable volumetric concentration of  
588 RAP filler).

$ G^* $ ratios	IR-30	IR-50	IR-70	IR-100	IT-30	IT-50	IT-70	IT-100
4	1.04	1.11	1.17	1.12	1.13	1.14	1.15	1.30
22	1.25	1.40	1.60	1.70	1.22	1.27	1.35	1.58
58	1.34	1.48	1.86	2.17	1.15	1.27	1.52	1.74
82	1.19	1.31	1.61	1.80	1.15	1.25	1.44	1.63

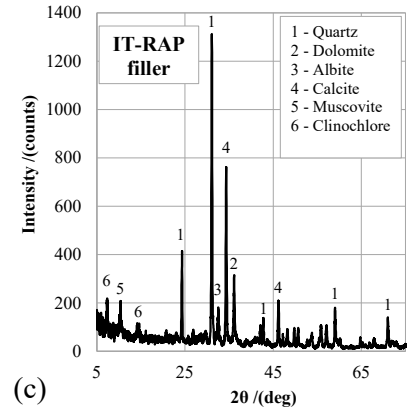
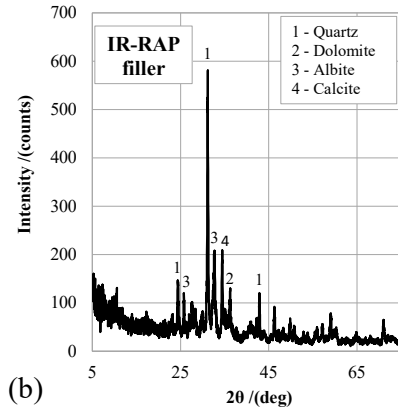
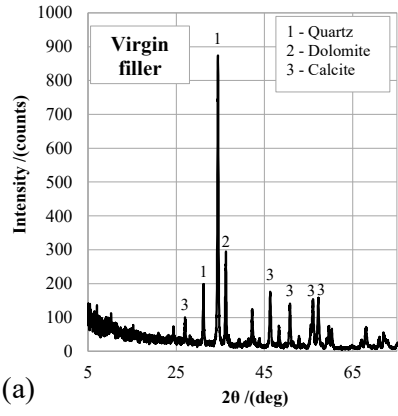
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590 Table 4.  $\delta$  ratios for mastics containing IR-RAP (variable volumetric concentration of RAP  
591 filler).

$\delta$ ratios	IR-30	IR-50	IR-70	IR-100	IT-30	IT-50	IT-70	IT-100
4	0.92	0.89	0.88	0.84	0.95	0.94	0.93	0.91
22	0.96	0.94	0.93	0.90	0.97	0.96	0.95	0.94
58	0.99	0.99	0.99	0.98	1.00	1.00	0.99	0.99
82	1.00	1.00	1.00	0.98	1.00	1.01	1.01	1.00

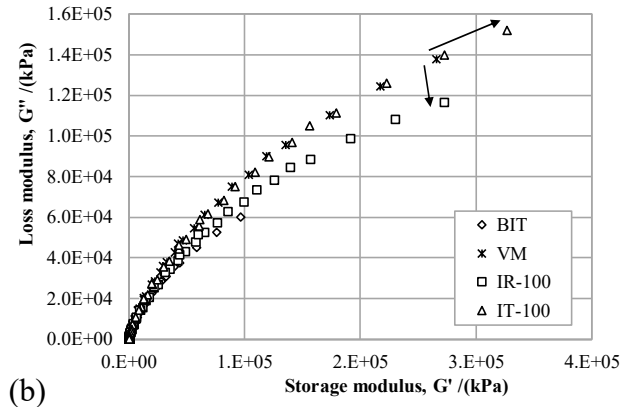
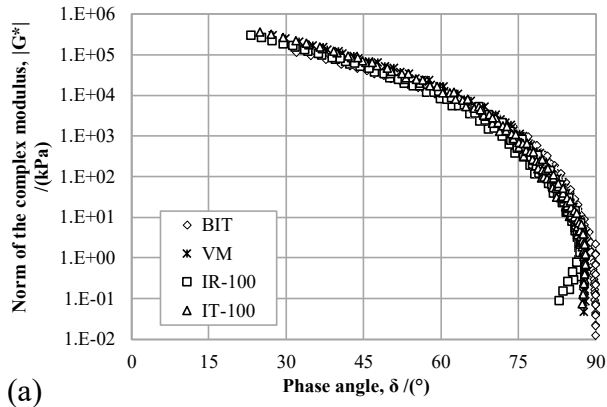
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[Figure 1]



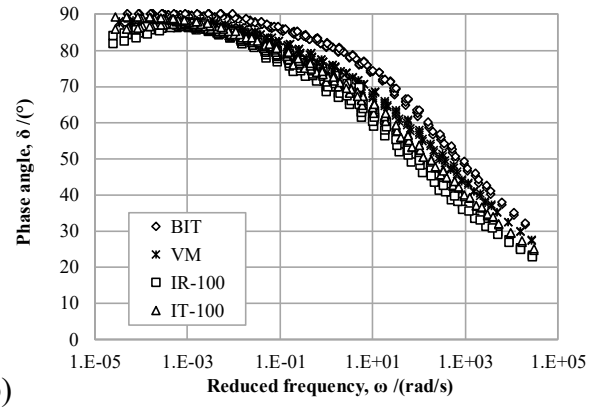
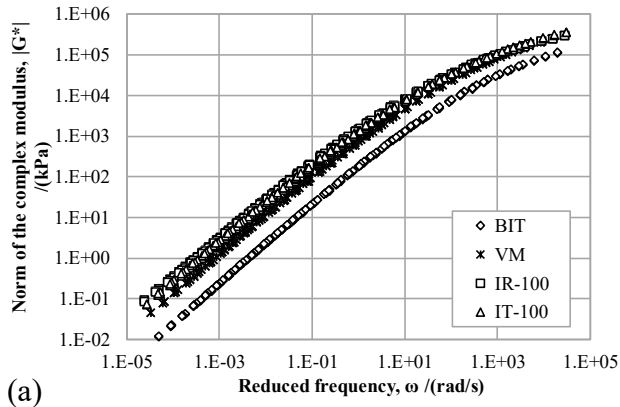
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(a)

(b)

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[Figure 2]



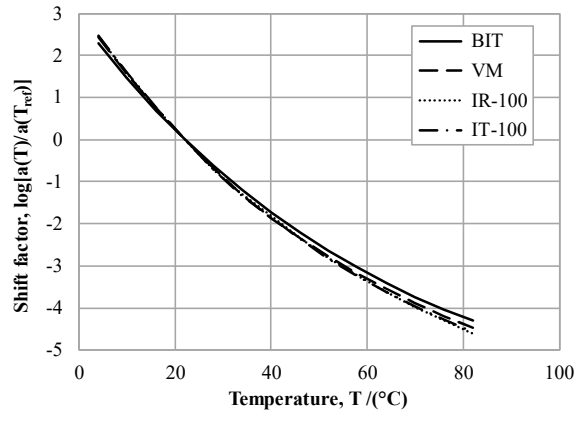
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(a)

(b)

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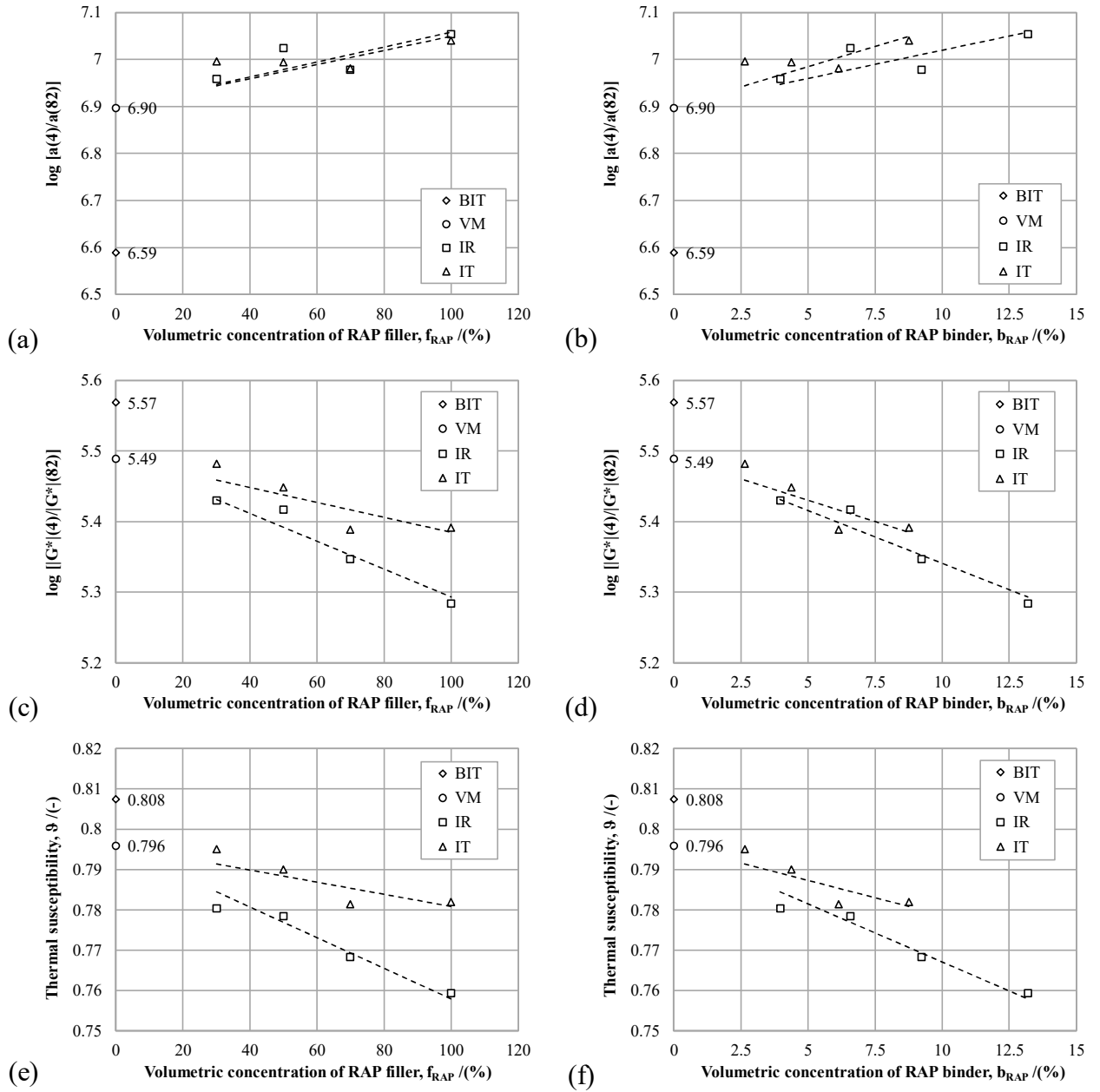
[Figure 3]



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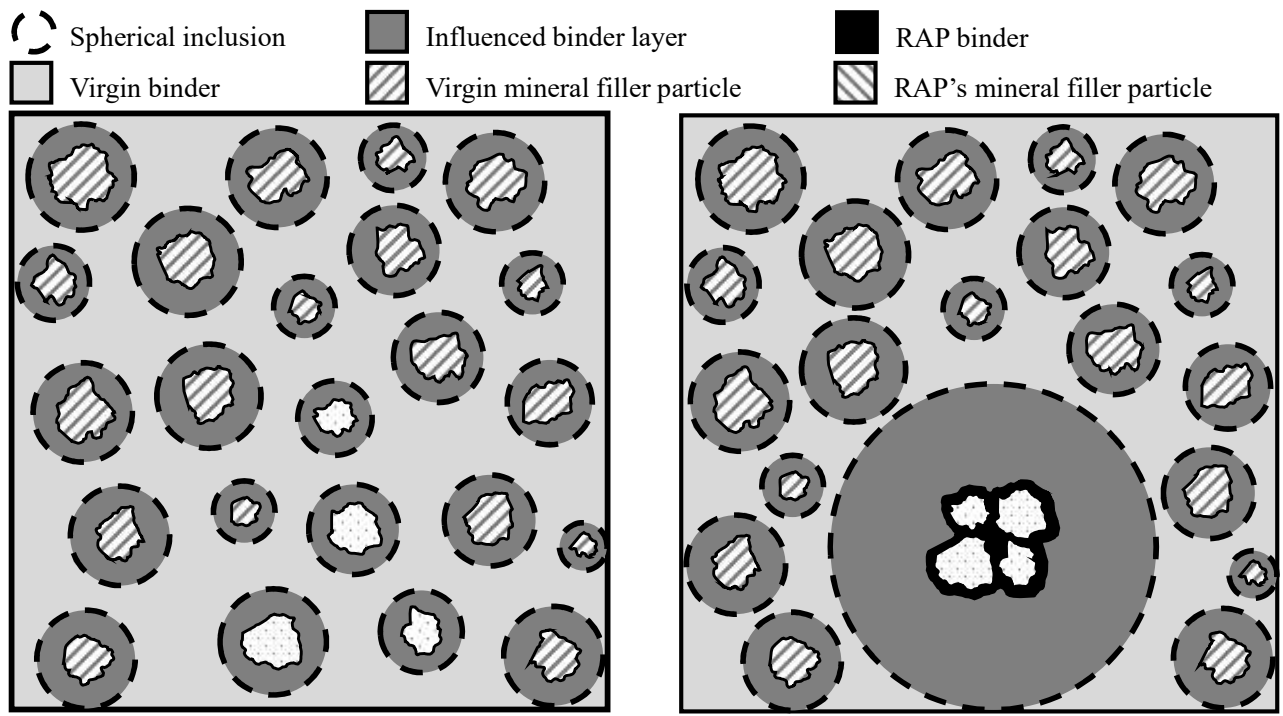
[Figure 4]



601

602

[Figure 5]



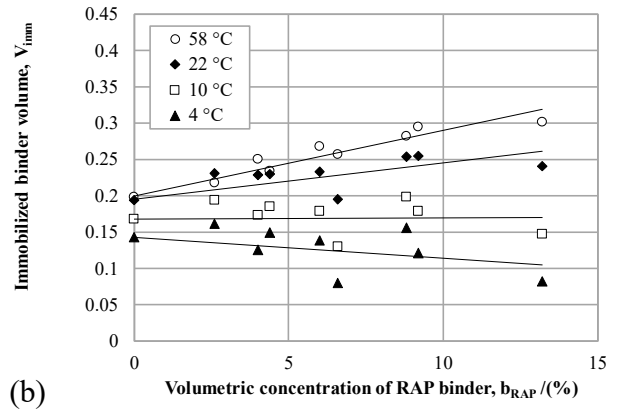
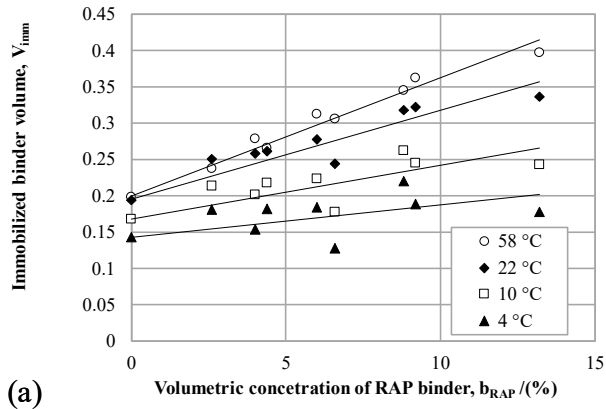
(a) Mineral filler + virgin bitumen

(b) Mineral filler + RAP + virgin bitumen

603

604

[Figure 6]



605

606

(a)

(b)

[Figure 7]

607 *Figure Captions:*

608 Figure 1. Results of XRD tests carried out on fillers: (a) virgin filler, (b) IR-RAP filler and (c)  
609 IT-RAP filler.

610 Figure 2. (a) Black and (b) Cole-Cole diagrams of mastics.

611 Figure 3. Master curves of the norm (a) and the phase angle (b) of the complex modulus.

612 Figure 4. Shift factors of mastics ( $T_{\text{ref.}} = 22 \text{ }^{\circ}\text{C}$ ).

613 Figure 5. (a, b)  $\text{Log}[a(4)/a(82)]$ , (c, d)  $\text{Log}(|G^*|_4/|G^*|_{82})$ , and (e, f)  $\vartheta$  as a function of  
614 RAP volumetric concentration of RAP filler and RAP binder.

615 Figure 6. Representation of particulate composite structure for generic mastic (a) and mastic  
616 containing RAP particles (b).

617 Figure 7.  $V_{imm}$  as a function of volumetric concentration of RAP binder according to (a)  
618 “total blending” and (b) “black rock” hypotheses.