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Determination of optimum limestone filler content in asphalt mastic by dielectric spectroscopy

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ABSTRACT

The rheological and dielectric characteristics of asphalt mastics with different ratios of limestone filler were analyzed and compared with each other for road pavement applications. The dielectric properties of asphalt mastics are studied as a function of frequency (0.1–10⁶ Hz) at different temperatures ((–)40°C–40°C). The glass transition temperature and the relaxation behaviour were determined by Vogel–Fulcher–Tamman and Cole–Cole equations, respectively. The free volume fraction was calculated by the Doolittle equation. The filler ratio dependence of the dielectric properties was attributed to the variation of the void fraction. This result was related to the glass transition temperature and ductility. The sample with 6 wt% limestone filler showed the highest dielectric constant value among all samples for all temperatures due to the excellent adhesion between binder and filler. Consequently, it was determined to be the optimum value to design hot mix asphalt for use in territories with high-temperature differences.

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1. Introduction

Asphalt mastic is a composite material composed of asphalt binder, called bitumen and filler (Wu et al., 2021). The filler is the mineral aggregates with a particle size of less than 075 µm (Wei et al., 2022). Today, asphalt mastics are important in controlling asphalt mixtures' mechanical and rheological properties because most roads and airfields are constructed using asphalt pavements (Apeagyei et al., 2014; Yang et al., 2023). The fillers have been widely used to improve the performance of asphalt mixtures and pavements, including strength, plasticity, porosity, water resistance, and weatherability (Guo et al., 2017; Roman & García-Morales, 2018; Tan & Guo, 2013). Besides, choosing a suitable filler and its optimal content is one of the key factors influencing the performance of mastic (Wasfi Al-Mistarehi et al., 2021). In recent years, limestone, volcanic ash, silt, Portland cement, mineral mud, slaked lime, rock flour, fly ash, and recycled brick dust have been commonly used for asphalt pavement construction (Diab & Enieb, 2018; Guha & Assaf, 2020; Guo et al., 2012; Huang et al., 2007).

Mineral fillers are divided into active and inert fillers, and adding one will have different effects on the mastic (Lesueur et al., 2016). To understand this effect, Das and Singh selected basalt (B) as

an inert filler and hydrated lime (HL) as an active filler, and mastics with different filler percentages were prepared to evaluate its rutting, fracture, and thermal cracking resistance. The results showed that the ageing resistance of asphalt mastic increased with the addition of HL for better rutting performance. In addition, the medium- and low-temperature performance of mastic with basalt increased with the additive of HL (Das & Singh, 2017). Little and Petersen stated that Hydrated lime improved the performance of mastic when the impact of hydrated lime and limestone on the rheology of the asphalt mastic is compared, and they assessed the differences in the chemical composition of mastics which contain hydrated lime and limestone (Little & Petersen, 2005). In another study, the effects of fillers on the properties of the asphalt mixture were investigated by Zulkati. The mixtures are prepared with hydrated lime, granite, and kaolin, selected as fillers. Their optimum asphalt content was determined using the Marshall mix design method, and the results showed that the properties of the filler affect its interaction with asphalt and improve the performance of mixtures (Zulkati et al., 2012).

On the other hand, understanding adhesion and the interaction mechanisms between filler and asphalt binder produces asphalt mixtures with better mechanical properties (Diab & Enieb, 2018). Thus, many studies have reported that three primary mechanisms reinforce the bitumen with fillers: volume filling, particle structuralization, and physical–chemical interactions (Underwood & Kim, 2011). Among these mechanisms, physical–chemical interaction, which occurs at the asphalt–filler interface, includes adsorption and diffusion of asphalt on the surface of the filler (Davis & Castorena, 2015). As a consequence, the thermodynamic and viscoelastic properties, such as penetration, ductility, softening point, and glass transition temperature of asphalt mastic, are significantly affected by asphalt–filler interaction when the fillers are added to asphalt (Clopotel et al., 2012). Therefore, the dielectric characterisation of asphalt mastic will give information about these interactions and their effects on dielectric, thermodynamic, and viscoelastic properties. In addition, this method can help improve characterisation of the performance of asphalt mastic and asphalt mixture (Kara et al., 2021). In addition, determining suitable mineral fillers and their concentration in asphalt mastic plays a significant role in investigating dielectric properties since the addition of different mineral fillers will change the dielectric properties of asphalt mastic. In literature, Zhai et al. investigated the effect of varying particle sizes in mineral aggregates on dielectric properties of bulk asphalt materials, and they stated that particle size is one of the main factors affecting the polarisation of asphalt–particle mixtures and increases the polarisation (Zhai et al., 2022). In another study, Zhai et al. investigated the dielectric properties of styrene–butadiene–styrene (SBS) asphalts in the frequency range from 300 MHz to 3 GHz at room temperature. They compared the predicted results with those obtained using the classical models and explored a particle polarisation mechanism with an electrically independent interface (Zhai et al., 2018). Zhong et al. prepared six samples of asphalt mastics with different filler–asphalt ratios, and dielectric measurement was operated in the temperature range -30 – 60 °C and. The results showed that the relative dielectric constant of asphalt mastic increases with the increase of filler–asphalt ratio (Zhong et al., 2022). Yang et al. reported that asphalt mastic’s dielectric properties depend on temperature and frequency. The filler type affected mastic’s polarisation (Apeagyei et al., 2014). Wang Y. et al. produced asphalt with graphene and carbon fibre to improve its conductivity and explore the influence of graphene on bitumen. Thus, the optimum additive concentration of graphene was determined to obtain high-conductive material (Wang et al., 2022).

A survey of the literature reveals many studies have shown the effect of mineral filler on the mechanical and rheological properties of asphalt mastic. Still, there is no adequate study about the dielectric properties of asphalt mastic. So, this study has been focused on utilising dielectric characterisation to investigate asphalt mastic’s dielectric properties with varying limestone filler ratios. Also, determining the optimal filler ratio has been proposed for better performance and durability in low-temperature climate conditions. Investigating the glass transition of asphalt mastics by dielectric spectroscopy can contribute to understanding the effect of asphalt mastic on the anti-rutting performance of hot mix asphalt mixture.

2. Experimental

2.1. Raw materials

To prepare asphalt mastics, a 50/70 penetration of neat bitumen was used as a binder supplied by Turkish Petroleum Refineries (TÜPRAŞ). The physical properties of the binder are in accordance with the ASTM D5] as seen in Table 1[20]. The limestone as a mineral filler. It was supplied from the limestone quarries of Ömerli, Istanbul, located in the northwestern part of Turkey and passed through the #200 sieve (75 µm). The mineralogical composition of the limestones in this region is 75% of calcite except for the main oxide elements such as SiO₂ of 1,93%, MgO of 1,53%, Al₂O₃ of 0,33%, and Fe₂O₃ of 0.76% (Kayabaşı et al., 2018; Yardim et al., 2019).

2.2. Preparation of asphalt mastics

First, the filler is heated at 150 °C for 24 h to remove the water. Then, a specific amount of filler is slowly added into the softened bitumen and premixed with a mixer for 5 min to obtain a homogeneous dispersion of the filler in bitumen.

According to the literature, the optimum content value of bitumen is 4.8% by mass, while the average content of the filler is 6% to design the Hot Mix Asphalt (HMA) (Bastidas-Martínez et al., 2021). The limestone and 50/70 penetration bitumen were used as filler and a binder, respectively. The mass ratio (by percentages) of filler to asphalt mastic is 4%, 5%, 6%, 7%, and 8%, and the HMA mix proportions adopted are listed in Table 2.

2.3. Rheological characterizations

Rheological characteristics and dielectric properties of samples were determined and compared to each other for road pavement applications.

The specific gravity test, the softening point test, the penetration test, the Fraass breaking point test, the force ductility test, the dynamic viscosity, the elastic recovery test. The specific gravity of samples was determined using the pycnometer method, following the ASTM D70 standard. It was calculated by comparing the weight of mastic to the weight of distilled water at the same volume and temperature of 25°C.

Table 1. Rheological properties of neat bitumen.

Test	Method	Unit	Requirement		Value
			Min	Max	
Specific gravity (25°C)	ASTM D 70	g/cm ³	–	–	1.009
Flashpoint (Cleveland)	ASTM D 92	°C	230	–	345
Penetration (25°C, 100 g, 5 s)	ASTM D 5	0.1 mm	50	70	59.8
Softening point	ASTM D 36	°C	45	55	48.5
Ductility (25°C, 5 cm/min)	ASTM D 113	cm	100	–	> 100
Thin film heating loss (163°C, 5 hr)	ASTM D 1754	%	–	80	11
Penetration percentage aft. heating loss	ASTM D 5	%	54	–	65.9
Ductility after hearing loss	ASTM D 113	cm	50	–	65.4

Table 2. Design of all HMA mixtures used.

Type of mastic	Bitumen (%)	Filler (%)	Bitumen (gr)	Filler (gr)
MA4	4.8	4.0	54.55	45.45
MA5	4.8	5.0	48.98	51.02
MA6	4.8	6.0	44.44	55.56
MA7	4.8	7.0	40.68	59.32
MA8	4.8	8.0	37.50	62.50

To measure the penetration of mastic, a standard needle is pressed into the material vertically with a load of 100 g for 5 s. The penetration depth of 0.1 mm is recorded according to the ASTM D-5 standard.

The ASTM D-92 standard was used to determine the softening point of mastic through the ring-ball method, which measures the temperature at which the material starts to flow.

The viscosity of mastic was measured to determine its flow resistance, following the ASTM D-2170 standard.

The Fraas breaking value test was conducted following the IP 80 standard to determine the low-temperature behaviour of the mastic. The test measured the temperature at which the mastic remained pliable before becoming critically stiff and breaking (up to -30°C).

The elastic recovery of mastic was determined by measuring the returning elastic deformation of bitumen at 25°C using the ductility tool according to the ASTM D-6084 standard.

These processes have been described in (Kara et al., 2021). In addition to characterisations above, RTFOT and BBR test were performed.

2.3.1. Rolling thin film oven test (RTFOT)

The Rolling Thin Film Oven Test (RTFOT) is a commonly used process for simulating the short-term ageing of asphalt binder to evaluate its physical characteristics. The RTFOT test involves exposing the binder to elevated temperatures, and emulating the ageing during manufacturing and placement processes. The results of the RTFOT test can also be used to quantify the amount of volatiles lost throughout the ageing process. In addition, this test is important for selecting asphalt binders because it provides valuable data on their performance characteristics under various conditions (Southern, 2015).

The samples were aged by RTFOT according to the method specified in ASTM-D 2872. First, the samples were cooled at room temperature, and then they were weighed. Secondly, they were subjected to rotational movement in an experimental device at 163°C for 75 min. After this period, the samples were removed from the device and cooled down. Next, the samples are weighed again, and the percentage change in weight is recorded compared to the initial weight. Finally, the penetration (ASTM D-5) and softening point (ASTM D-36) tests are performed on the aged samples to determine the permanent penetration value and the increase in the softening point value.

2.3.2. Bending beam rheometer test

The aim of the test is to determine the flexural creep stiffness of bituminous binders and bitumen at different temperatures. In this test, a beam-shaped specimen is subjected to a constant point load in a three-point bending test, and the deflection is measured as a function of time. The flexural creep stiffness measures the behaviour of bitumen and bituminous binders at low temperatures. The most commonly determined parameters are the flexural creep stiffness at a given temperature or the temperature at which the flexural creep stiffness equals a given value, along with the corresponding m-values. If you require a single result for the flexural creep stiffness and the m-value, the values at a load duration of 60 s are used (Southern, 2015).

Each sample for the BBR was prepared and tested according to the ASTM D6648-08 standard to determine the material's behaviour at low temperatures. The test measures the amount of creep or deflection the material experiences at a specific temperature and under a constant load.

The ASTM D-113 standard determines the amount of elongation by force ductility. The material's cohesive strength is determined by its ductility at low temperatures. To find the elongation at break, the material is pulled at a speed of 50 mm/min in a water bath at a particular temperature, and the deformation energy is calculated based on the load-extension curve.

2.4. Dielectric measurements

Broadband dielectric spectroscopy is an effective technique for determining complex mixtures like asphalt mixtures, asphalt mastic, etc., where applications and properties depend on the control of

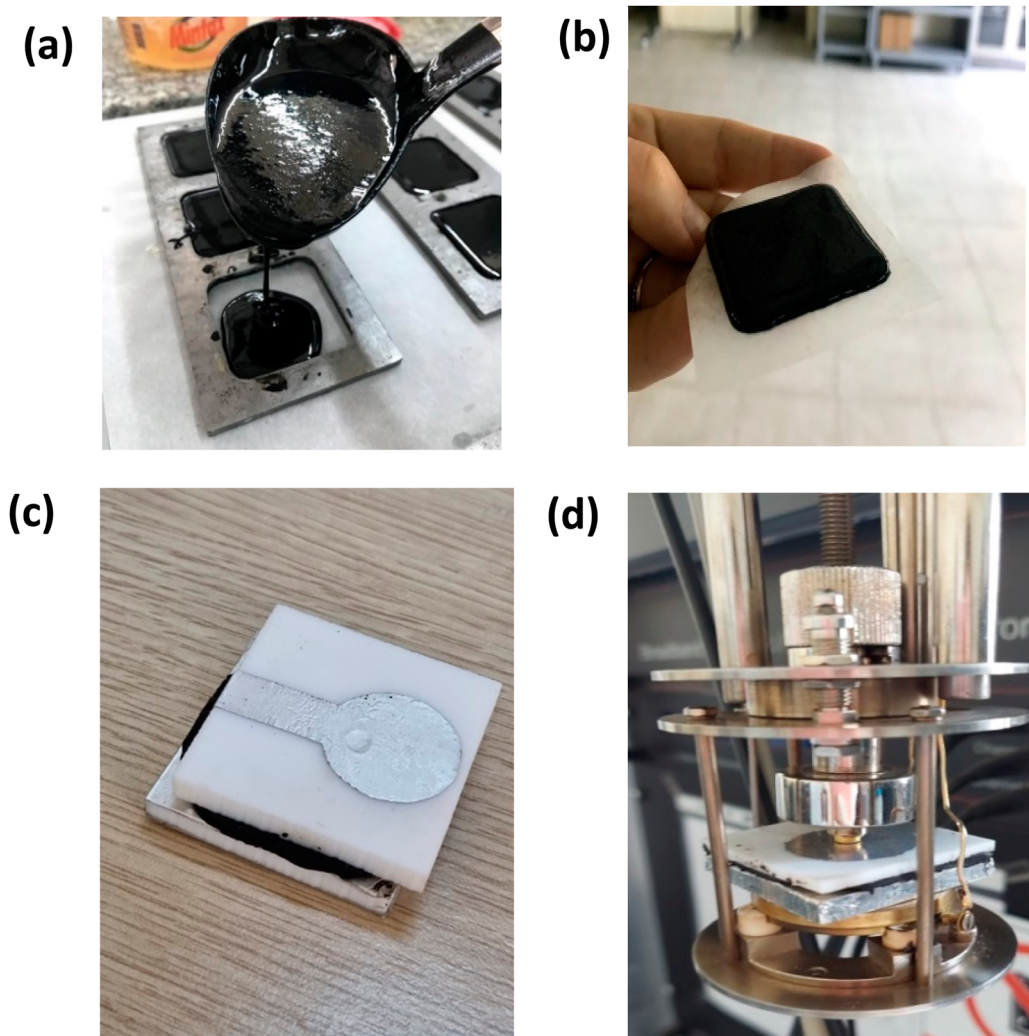


Figure 1. (a) Sample poured into a specially designed sample mold for dielectric measurements. (b) ready-to-measure mastic sample (c) Schematic picture parallel plate capacitor form of bitumen for dielectric measurement. (d) Standart sample cell.

compositions, fillers, and their distributions (Nilagiri Balasubramanian & Ramesh, 2018). For the dielectric measurements, the samples that have a thickness of 2.5 mm were mounted between two Al electrodes with a diameter of 20 mm to obtain a capacitive form, as seen in Figure 1 (c) and (d). The dielectric properties of asphalt mastics are studied as a function of frequency ($0.1\text{--}10^6$ Hz) at different temperatures ($-40^\circ\text{C--}40^\circ\text{C}$.) using Alpha-A high-resolution dielectric, conductivity, and impedance analyzer (Novocontrol Technologies under an alternative voltage of 1.0 V (rms)).

3. Results and discussions

3.1. Rheological results

Investigation of the rheological properties of asphalt mastics is crucial in evaluating issues such as identifying its components, temperature sensitivity, strength, and deterioration under traffic loads. Therefore, the material's rheology should be determined before analyzing the dielectric properties of asphalt mastic depending on parameters such as temperature, frequency, and thickness.

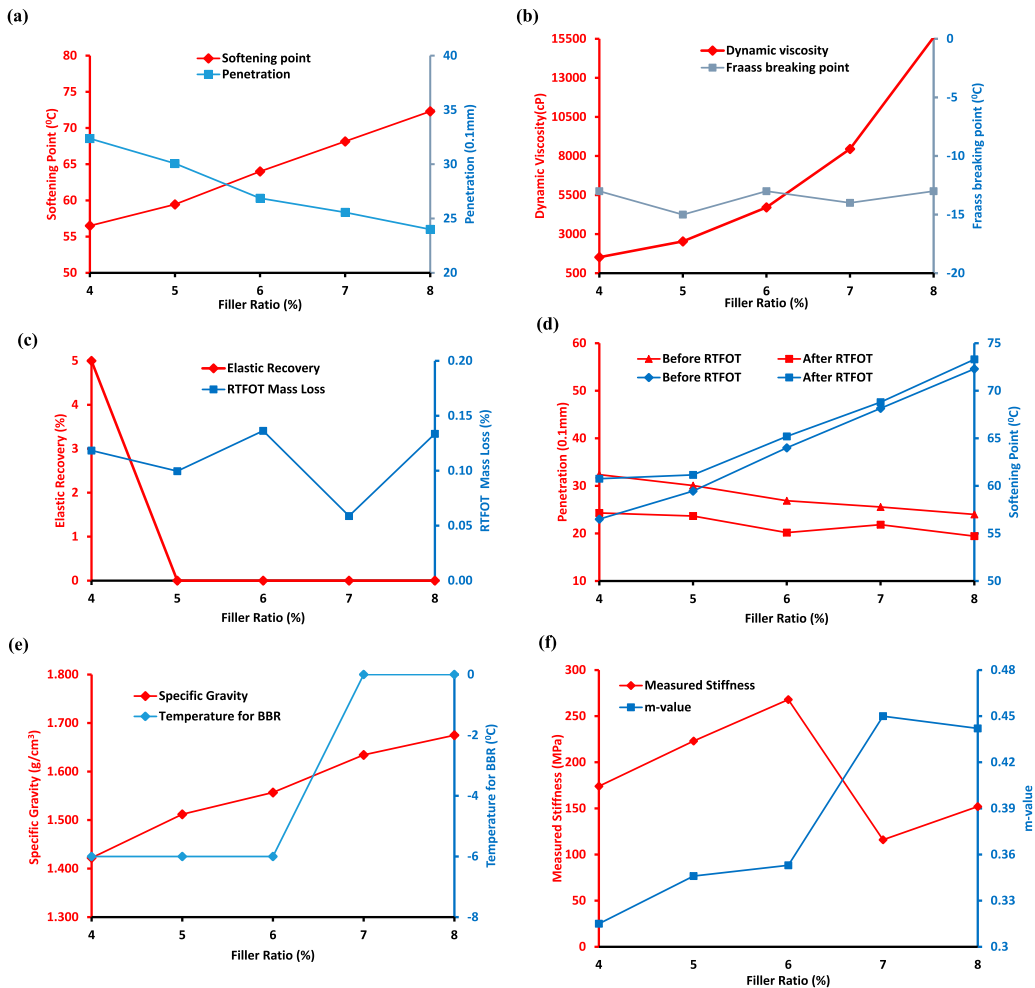


Figure 2. (a) Penetration and softening point with respect to filler ratio (b) Dynamic viscosity and fraass breaking point with respect to filler ratio (c) Elastic recovery and RTFOT mass loss with respect to filler ratio (d) Penetration and softening point before/after RTFOT with respect to filler ratio (e) Specific gravity and temperature for BBR with respect to filler ratio (f) Measured stiffness and m-value for BBR with respect to filler ratio of asphalt mastics.

The rheological properties of neat bitumen are seen in Table 1 (Kara et al., 2021; Ulutas et al., 2023).

The analyses mentioned in section 2.2 were performed to determine the parameters of the rheological properties of five asphalt mastics with different filler ratios. Accordingly, the penetration value of asphalt mastics was much lower than that of neat bitumen.

While the penetration value of neat bitumen is 57.8 (Table 1), this value varies between 24 and 32.4 for asphalt mastics. In addition, it is seen that the penetration value decreases with increasing filler content in asphalt mastic in Figure 2 (a) (blue x-axis).

The softening point value of asphalt mastics is higher when compared to neat bitumen's value. While the softening point of neat bitumen was 49.10°C, this value of asphalt mastics increases as the filler content increases in Figure 2(a) (red x-axis). Rodgers has reported this increase in softening point and exhibits a linear behaviour (Rodgers et al., 2010).

It was found that the dynamic viscosity value of asphalt mastics has higher values when compared to the neat bitumen. While this value was obtained as 450 cP for neat bitumen, it was seen that this value increases with increasing filler content in asphalt mastic in Figure 2(b) (red x-axis). Especially

for the MA7 and MA8 samples, this increase is more pronounced. This behaviour can be explained as agglomeration as the filler contribution increases.

As the filler concentration in the asphalt mastic increased, the softening point and viscosity tended to increase against the decrease in penetration. This result was consistent with the literature (Mukhtar et al., 2021).

When the Fraass Breakpoint value of asphalt mastics is compared to neat bitumen, it is shown that high ductility can be achieved at low temperatures in Figure 2(b) (blue x-axis). While it is -10°C for neat bitumen, this value decreases with adding filler. The results obtained in this study confirm the fracture energy results of mastics at low temperatures (Bardini et al., 2017). It has been stated that adding filler to the asphalt binder reduces the energy-breaking value compared to the neat asphalt binder, and the breaking energy value decreases as the amount of filler increases, depending on the filler type. Also, the results showed that the variation of filler ratio in asphalt mastics has little effect on the Frass Breaking Point value in Figure 2(b).

The percentage of elastic recovery for neat bitumen is zero (Table 1). To reverse this situation, bitumen needs to be modified or fillers added. It is seen that the additive of 4% limestone filler only imparts an elastic recovery capability to the bitumen in Figure 2(c) (red x-axis). As noted in the study by Bao et al., the behaviour of the asphalt mastic under pressure depends on the amount of pressure. The increase of filler in the mastic under low pressure does not affect the amount of elastic recovery much. Still, under high pressure, there is a visible improvement depending on the filler type (Bao et al., 2023).

The results of the RTFOT test, representing the short-term ageing exposure of bitumen, show that the asphalt mastics provide a better performance, and the loss of mass is less compared to neat bitumen. As seen in Figure 2 (c) (blue x-axis), these values are close for asphalt mastics.

The penetration value of neat bitumen is higher than that of asphalt mastics before and after the RTFOT test in Figure 2 (d) (red x-axis). However, the penetration values after RTFOT are lower than before the RTFOT test for asphalt mastics in Figure 2 (d) (red x-axis). The filler additive shows a noticeable improvement by keeping the bitumen particles together in the short term. This behaviour can be explained as evaporation, removing the material's lightweight components from the structure. Penetration values decrease as the filler ratio increases.

The softening point values of neat bitumen before and after RTFOT are lower than the asphalt mastics in Figure 2 (d) (blue x-axis). The softening point values obtained after RTFOT are higher than those received before RTFOT. The softening point values for asphalt mastics increase as the filler ratio increases.

The increase in the filler ratio causes the bitumen molecules to be retained in the asphalt mastic, reducing the free binder volume content, and an increase in the specific gravity of the mastic occurs with the effect of the increasing particle volume (Mukhtar et al., 2021). The specific gravity of asphalt mastics is higher than neat bitumen. Also, the specific gravity value increases with increasing filler ratio in the asphalt mastics, as seen in Figure 2 (e) (red x-axis).

The results of BBR applied to examine the cracking resistance and fatigue performance of bituminous materials at low temperatures are given in Figure 2 (d and e). The purpose of the BBR test (Measured Stiffness) is to determine the temperatures that provide $S \leq 300 \text{ MPa}$ and $(m\text{-value}) m \geq 0.300$. All asphalt mastics fulfilled BBR conditions, but desired values were obtained at different temperatures. While the neat bitumen fulfilled the desired condition at -12°C , the desired conditions for asphalt mastics with 4%, 5%, and 6% filler ratios were fulfilled at -6°C . Apart from these samples, asphalt mastics with 7% and 8% filler ratios fulfilled the desired condition at 0°C . These observations are agreement the results obtained by Qiu, H. et al., who applied the BBR test to observe the effect of the filler content on the low-temperature performance and stated that the performance deteriorated with the increase in the filler-bitumen ratio significantly when the percentage increased from 1.2–1.4 (Qiu et al., 2013).

It can be seen in Figure 2 (f) that the creep stiffness (S) increases up to 6% filler ratio while S decreases at the filler ratio 7%. Meanwhile, the m -value increases with increasing filler ratio.

The high m -value and low creep stiffness for the asphalt mastics show good resistance at low temperatures (Roberto et al., 2020). According to this, MA7 and MA8 have good low-temperature cracking resistance but, MA4, MA5, and MA6 worsens due to the poor adhesion between binder and filler (Chen et al., 2022; Clopotel & Bahia, 2013).

3.2. Dielectric results

3.2.1. Dependence of dielectric constant on frequency, temperature, and filler ratio

The complex dielectric function represents the polarisation and conduction of dielectric materials in an alternating electric field. The complex dielectric function is as follows (Harun et al., 2009):

$$\kappa^* = \kappa' - i\kappa'' \quad (1)$$

Where,

κ' : the dielectric constant, a measurement of how much electrical energy is stored in the dielectric sample.

κ'' : dielectric loss, a measurement of the electrical energy loss in the dielectric sample.

The dielectric constant can be calculated using the below relation (Abdullah et al., 2011)

$$\kappa' = \frac{C^*d}{\varepsilon_0^*A} \quad (2)$$

where d is the thickness of the sample, A is the surface area of the electrodes, ε_0 is the electrical permittivity of the vacuum, and C is the capacitance of the sample.

The chemical composition of the bitumen consists of a long C–H chain and other organic components such as Sulphur (S), Oxygen (O), Nitrogen (N), and Hydrogen (H). Limestone, used as a filler, contains high CaO content and numerous voids (Muniandy et al., 2013).

Figure 3 shows that the dielectric constant decreases with increasing frequency for all asphalt mastics. The long chain can respond to the change in the electric field at low frequencies. However, the polarisation ability of the chain will decrease as the frequency increases. Thus, the dielectric constant decreases as the frequency increases. Other side branches and smaller polarisable groups will contribute to polarisation at high frequencies. The dielectric constant at the high-frequency region is lower than at the low-frequency region. Moreover, more polarisable groups in the medium contribute to the polarisation as the temperature increases (Zhong et al., 2022).

The relationship between filler content ratio and dielectric constant is observed in Figure 4.

In a recent article, the neat bitumen's dielectric constant is reported as 5.5 at room temperature (Kara et al., 2021). So, the dielectric constant increases with the increasing filler ratio; asphalt mastic with different filler ratios is higher than neat bitumen because more polarisable groups in the medium are influential (Xie et al., 2019).

When the dielectric constants of asphalt mastics are investigated, the dielectric constant increases up to MA6 and then decreases with increasing filler ratio. It is detected that the MA6 has the highest dielectric constant, and dielectric constant values for other samples are very close to each other. This behaviour can be expressed through the variation of free volume fraction in the medium (Değer & Ulutas, 2003; Tuichai et al., 2021; Xie et al., 2019; Yahsi et al., 2019; Yang et al., 2023).

Considering the MA7 and MA8 samples, the rigidity of asphalt mastic increases due to the agglomeration of asphalt mastics when the filler ratio increases, as stated in the dynamic viscosity test results. So, the polarizability of samples decreases. Thus, the dielectric constant decreases with the increasing filler ratio (Bozoglu et al., 2019).

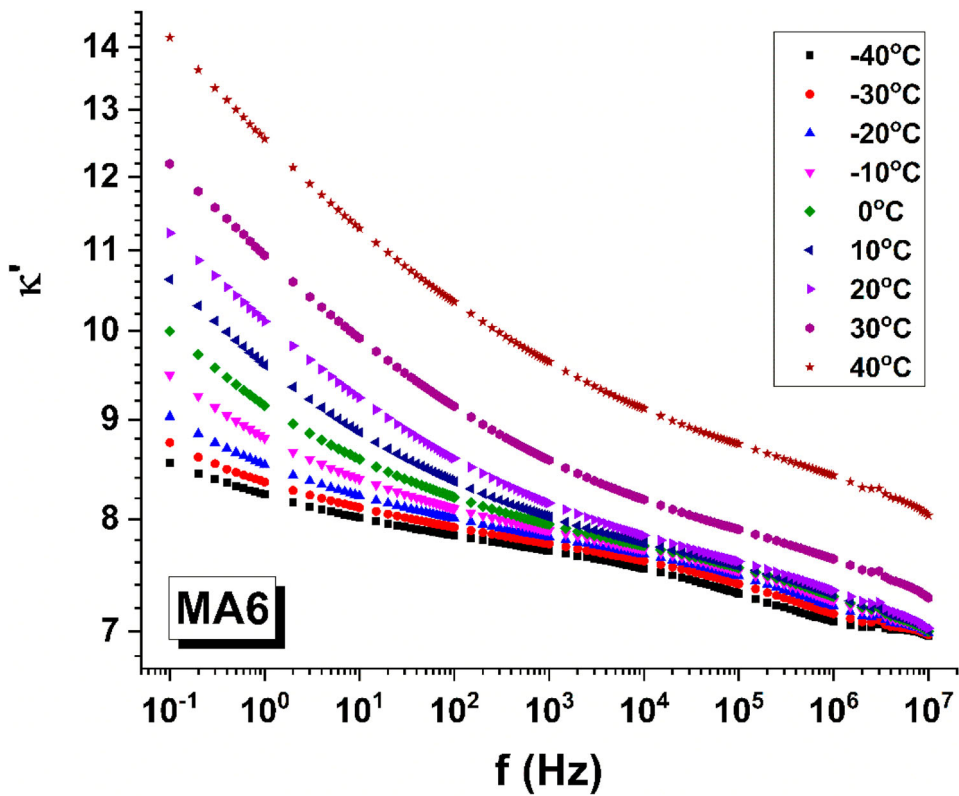


Figure 3. κ' as a function of frequency at different temperatures of MA6 (representative sample)

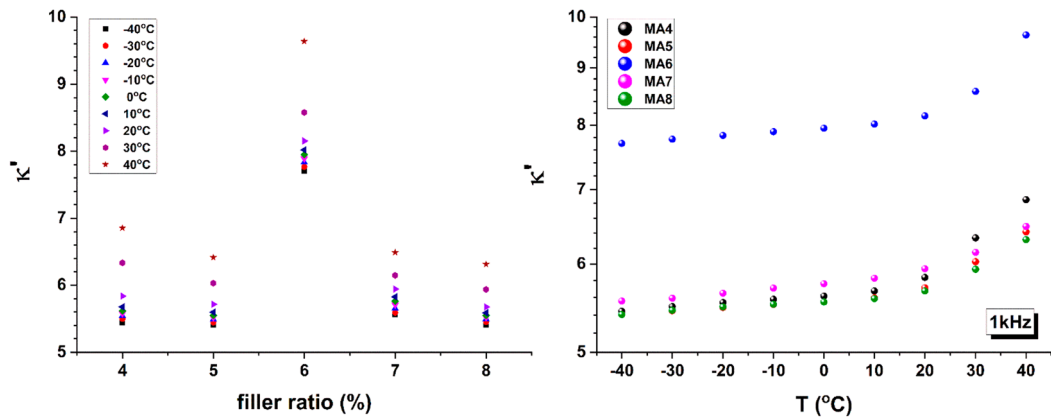


Figure 4. Temperature dependence of dielectric constant of asphalt mastics at 1 kHz.

3.3. Dynamic glass-transition temperature studies

3.3.1. Electrical modulus

The complex electric modulus (M^*), which helps understand the relaxation process in structure, is used for distinguishing the influences of electrode polarisation during the conductivity process at low

frequencies, and it can be expressed using the relationship below (Wang, 2022):

$$M^* = \frac{1}{K^*} = \frac{1}{K'^2 - iK''^2} = \frac{K'}{K'^2 - K''^2} + i \frac{K''}{K'^2 - K''^2}, \quad (3)$$

Here, M' and M'' are the real and imaginary parts of M^* , respectively, that can be written as:

$$M' = \frac{K'}{K'^2 + K''^2} \quad (4)$$

$$M'' = \frac{K''}{K'^2 + K''^2} \quad (5)$$

Also, analysis of the imaginary electric modulus of bitumen and modified bitumen can be done by fitting the following Cole–Cole equation:

$$M''(\omega) = M_\infty \left[\frac{\sin(\beta\pi/2)}{(\omega\tau_0)^{-\beta} + 2\cos(\beta\pi/2) + (\omega\tau_0)^\beta} \right], \quad (6)$$

In Eq.6, τ and β represent the relaxation time and symmetrical broadening for the relaxation function. Also, β is recognised as the shape of the parameter that follows the conditions ($0 \leq \beta \leq 1$) (Hodge et al., 2005).

From Figure 5 (a-e), a shoulder that is attributed to the relaxation of a long C–H chain is observed for all samples at temperatures above 0 °C, and the shoulder shifts towards higher frequencies with increasing temperature. The polarisable entities are thermally activated as temperature rises. This causes fast polarisation, which has a short relaxation time (τ). As a result, the resonance frequency will shift to higher frequencies if the temperature increases.

Inset graphs show that the experimental data are well fitted to the Vogel–Fulcher–Tamman model (VFT), which describes the α -relaxation including the segmental motion of the main chain (Kirtay et al., 2022)

$$\omega_{\max\kappa''}(T) = \omega_\infty e^{\left(\frac{-DT_0}{T-T_0}\right)} \quad (7)$$

Here, the obtained values of Vogel temperature (T_0) and ductility (D), which correspond to the change of viscosity for all samples from VFT's model, are presented in Table 3.

Using the results of the temperature sweep of κ'' at 100 kHz, all samples were analyzed to characterise the glass transition temperature (T_g) in more detail in Figure 6(a-e). For all samples, two relaxation regions are observed. The first peak is the primary α -relaxation process associated with T_g . The shoulder with higher intensity corresponds to the normal mode relaxation process. The T_g values of samples are very close to each other except MA8. T_g of MA8 is determined as -15°C. So, it can be said that the MA8 is unsuitable for use in road pavements at low temperatures due to the cracking easily. In our recent article, it was found that the T_g of neat bitumen is -20 °C (Kara et al., 2021). T_g decreases with increasing filler ratio when the neat bitumen is compared to the asphalt mastics. This behaviour can be attributed to the physico-chemical interaction between bitumen and filler. Because the number of voids on the structure will change the dielectric properties of samples (Clopotel et al., 2012). Consequently, changes in viscosity with increasing filler ratio affect the molecular dynamic of structure and T_g .

Polar structured binders get absorbed on mineral fillers. As the filler ratio increases, the absorption in the structure increases, leading to an increase in free volume. However, as the binder absorption increases, it reduces the mobility of the binder, which in turn increases T_g . The 8% ratio is accepted as a maximum filler value as stated in literature (Guo et al., 2012; Liu et al., 2020).

To support this evaluation based on ductility, the change of free volume can be investigated by Eq.9 related to the VFT equation.

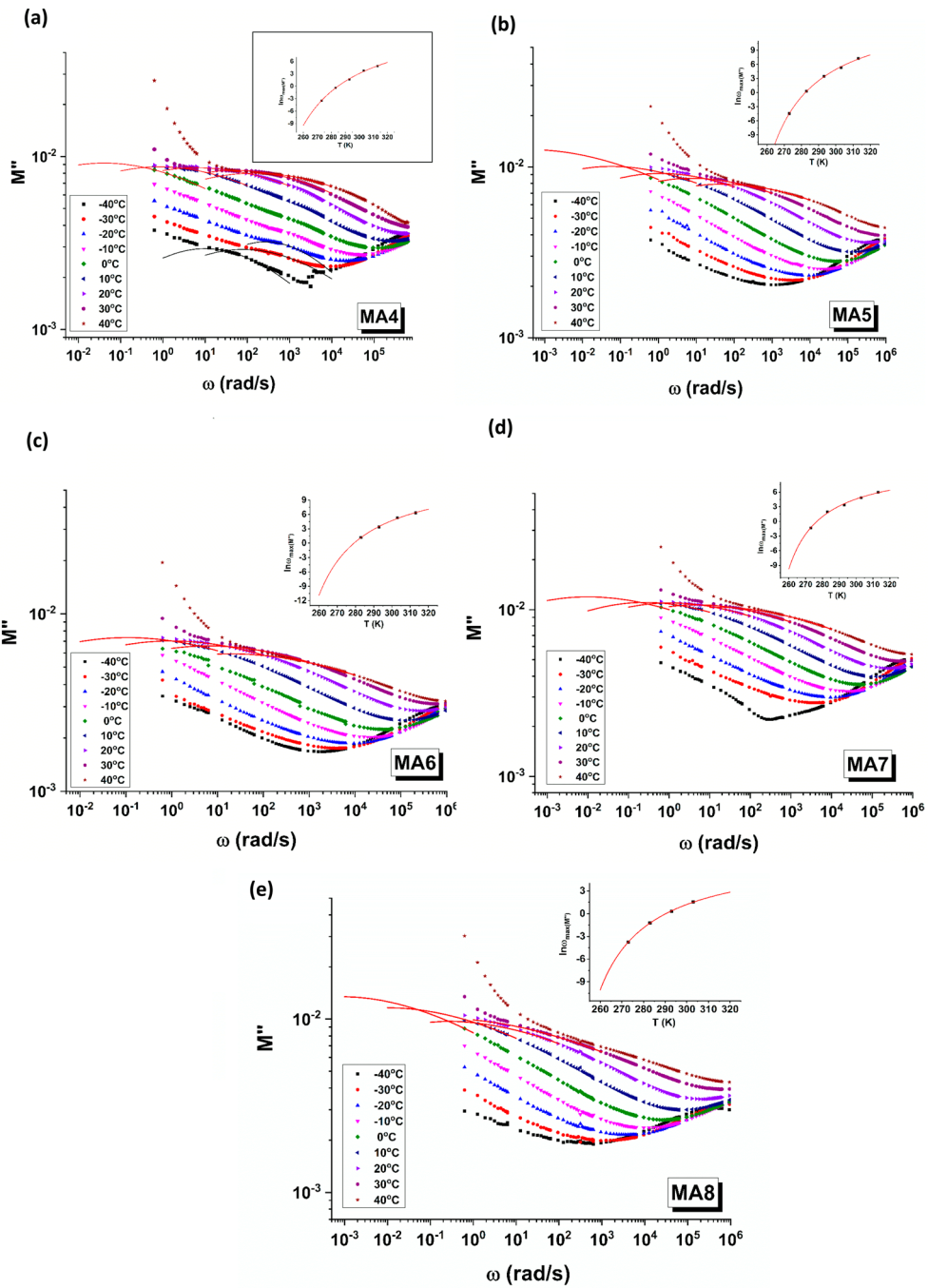


Figure 5. M'' as a function of angular frequency fitting by Cole-Cole equation at different temperatures (a) MA4 (b) MA5 (c) MA6 (d) MA7 (e) MA8. Fitting curves are represented as lines. Inset graphs: $\ln \omega_{M''} - T$ fitting by VFT equation at different temperatures for all samples.

Considering the Doolittle equation, the relaxation time for α - the relaxation process depends on the free volume (Ortiz-Serna et al., 2011):

$$f_{max} = f_o \exp \left(\frac{B}{\phi} \right) \quad (9)$$

Table 3. Ductility, Vogel temperature, Glass transition temperature, and free volume fraction values for all samples.

Samples	D	T ₀ (K)	T _g (K) (at 100kHz)	ϕ_B
MA4	0,51	206,04	240	0,323
MA5	0,79	197,42	253	0,356
MA6	1,08	190,55	238	0,230
MA7	0,74	203,87	233	0,193
MA8	0,8	208,78	258	0,294

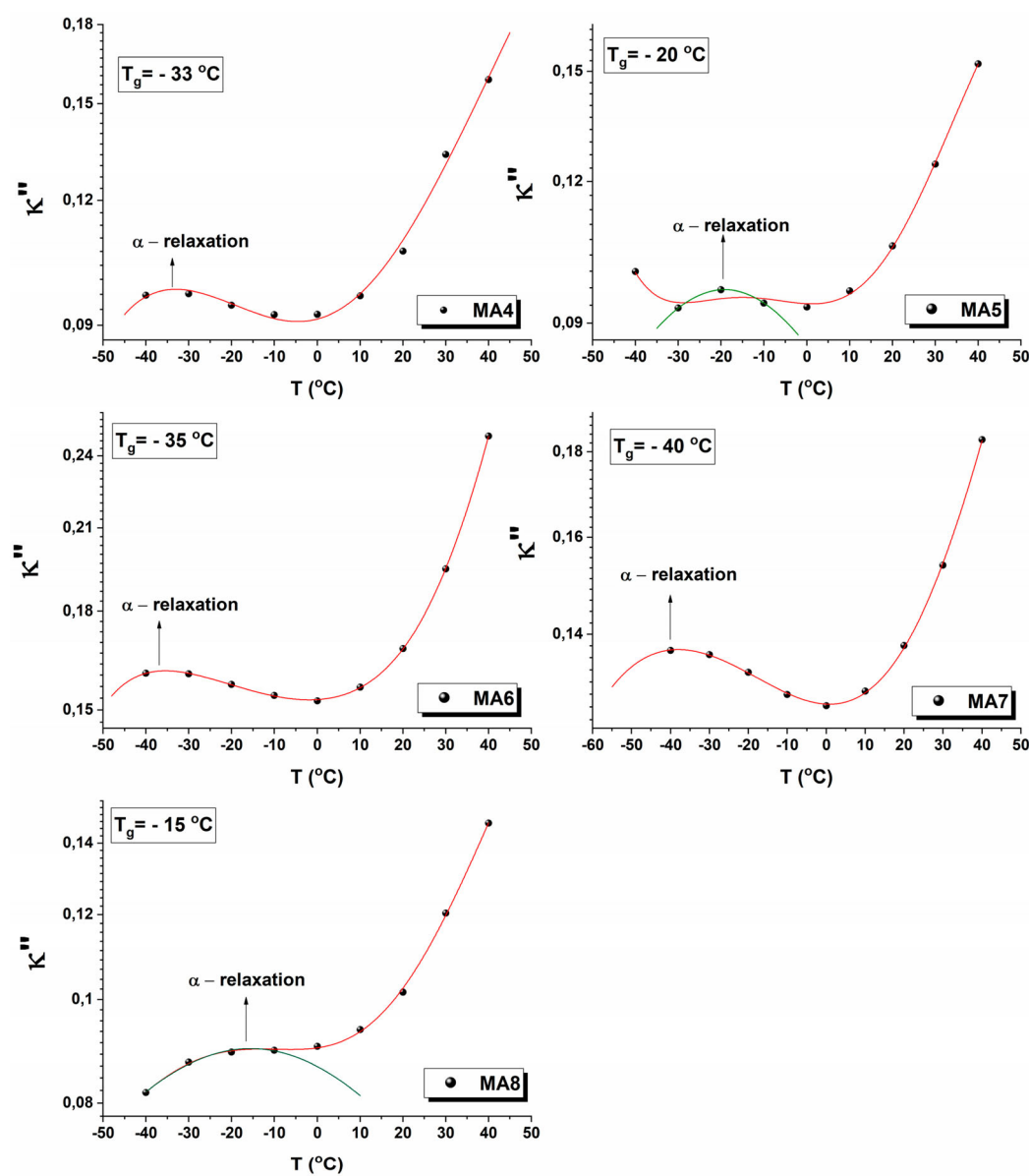


Figure 6. Temperature dependent κ'' for (a) MA4 (b) MA5 (c) MA6 (d) MA7 (e) MA8 at 100 kHz.

ϕ : the fraction-free volume

$B = \sim 1$ for the volume occupied by the segment that participates in α - the relaxation process

When combining Equations (8) and (9), the free volume value obtained by the formula given in Equation (10):

$$\left(\frac{B}{\phi}\right) = \frac{(T - T_0)}{(D.T_0)} \quad (10)$$

In Table 3, the effect of the filler ratio on filler volume fraction and glass transition temperature was given. These results show that ductility reached a maximum for MA6 because the interaction between bitumen and limestone has more adhesion for this sample in comparison to all samples. These results are compatible with the results of low T_g and T_0 for MA6.

Ductility and T_g are related to free volume fraction for polymeric and asphalt samples based on Eq. (9) and (10). For this reason, MA6 is one of the samples with a low free volume fraction. When the parameters, T_g , ductility, and free volume fraction are considered together, MA6 is detected as the most suitable sample for road pavement application because the low free volume fraction of MA6 with high ductility and low T_g , the road pavements containing this sample will be resistant to temperature variations. Thus, these longevity road pavements, including MA6, will prevent cracks in territories with high-temperature differences. According to the results, the most suitable filler ratio can be selected for road pavement performance based on sensitivity to changes in temperature and volume fraction.

4. Conclusions

In this study, the rheological and dielectric properties of asphalt mastic with various filler ratios of limestone were investigated and compared with neat bitumen. The results are presented below.

The asphalt mastic has a lower penetration, while the softening point is higher than neat bitumen's. The dynamic viscosity increases with added filler, particularly in asphalt mastic, with 7% filler (MA7) and 8% (MA8) because the viscosity becomes more brittle due to agglomeration. Compared to neat bitumen, the Fraass Breakpoint of the asphalt mastic displays greater durability at lower temperatures. However, the BBR test revealed that the low-temperature performance of asphalt deteriorated significantly with an increase in the filler-bitumen ratio.

The dielectric properties of the asphalt mastic were found to be influenced by the amount of filler used. The saturation point was discovered at a limestone content of 6%. Toward higher percentages of this content, the free volume of the sample increases due to agglomeration, which influences rigidity, a situation of the structure that controls the charges' polarisation ability. As the filler ratio increases over 6%, the dielectric constant decreases due to the increased rigidity. Increased rigidity causes the glass transition temperature to be increased while the ductility to be decreased. It was seen that the characteristics of dielectric constants, glass transition temperature, and free volume fractions for MA6 are in accordance with each other. According to these results, in designing hot mix asphalt, asphalt mastic with 6% filler ratio is the most suitable sample for road pavements because of its high efficiency and longevity under low-temperature weather conditions.

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