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ORIGINAL ARTICLE

Assessing the effect of fillers on LVE properties of asphalt mastics at intermediate temperatures

Mohit Chaudhary · Nikhil Saboo · Ankit Gupta · Bernhard Hofko · Michael Steineder

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Abstract This study aims at examining the performance of asphalt mastics in the Linear Viscoelastic (LVE) domain incorporating three Indian and three Austrian fillers. The various physical, morphological, and chemical properties of the fillers were analyzed with the help of respective characterization tests. In addition to the particle size distribution curve and fineness modulus, a new parameter called Filler Grain Coefficient (FGC) has been introduced in this study to quantify the distribution of particles in the system. This paper also attempts to find a correlation between various physical parameters. There are two variables i.e., $|G^*|_{LVE}$ and $|G^*|_{ratio}$ in order to quantify the Linear Viscoelastic complex modulus. The effects of both volume and surface area of fillers have been manifested by three ratios denoted as *V*/FM, *V*/FGC, and *V*/RV. To compare the outcome of the study, the Strategic Highway Research Program Linear Viscoelastic strain criteria is also included, which examines the applicability of the criteria to the asphalt mastics. The research incorporates a wide range of fillers with variable properties, as confirmed by the test results. Filler parameters Rigden Voids and Filler Grain Coefficient were found to be strongly correlated with almost all physical properties. On the grounds of variation in LVEM with volume concentration of filler, the highest rank can be attributed to Red Mud and LimeStone, respectively, followed by other fillers with Marble Dust being the lowest. The reinforcing effect of fillers and higher surface area contributed to the exponential increase in $|G^*|_{LVE}$, with an increase in volumetric concentration. The variation of both variables: $|G^*|_{LVE}$ and $|G^*|_{ratio}$, presented *V*/FM as the most worthy parameter, as it illustrated variation at different temperatures. The Linear Viscoelastic limits obtained from the study were relatively conservative compared to those from the SHRP study, this confirms the unsuitability of applying SHRP equations to the asphalt mastics. Moreover, the relationship instigated in the study can be used to reckon the LVE strain limit and to further analyse asphalt mastics.

Keywords Linear viscoelastic · Filler grain coefficient · LVEM · Mastic · SHRP

M. Chaudhary (✉) · N. Saboo · A. Gupta
Department of Civil Engineering, IIT (BHU), Varanasi 221005, India
e-mail: mohitchaudhary.rs.civ18@iitbhu.ac.in

N. Saboo
e-mail: nikhilsaboo.civ@iitbhu.ac.in

A. Gupta
e-mail: ankit.civ@iitbhu.ac.in

B. Hofko · M. Steineder
Institute of Transportation, Vienna University of Technology, 1040 Vienna, Austria
e-mail: bernhard.hofko@tuwien.ac.at

M. Steineder
e-mail: michael.steineder@tuwien.ac.at

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1 Introduction

The particles finer than 75 micrometers, i.e., passing through 0.075 mm or P200 sieve, are termed as fillers or P200 materials [1]. The mineral filler plays a dual role in asphalt mixes: facilitate the contact points between large aggregates by filling interstices between them as well as forms a high consistency matrix with bitumen known as mastic that binds the aggregates together [2–4]. The filler is a primal parameter liable for the behavior of mastics in the asphalt mixtures [5]. The physiochemical nature of fillers, their shape, size, texture, surface area, the elemental composition of mineral fillers, and surface energy are the vital parameters swaying the behavior of mastics and, eventually, the bituminous mixes [6]. The role of asphalt mastics is very prominent in paving mixtures as elucidated by many researchers [7–9]. The mastics provide better resistance to deformation under loading, owing to its higher stiffness [10]. The relative stiffening of asphalt mastics is contingent on properties of both filler and binder as well as the specific interaction between them rather than filler content alone. The inclusion of solid particles into the fluid medium and the specific interaction between bitumen and filler contributes to the modification effect [11]. Kallas and Puzinauskas [12] speculated the role of mineral fillers as active material in physio-chemical interaction instead of considering it as inert material for void filling. The affiliation between filler and binder cannot be simply explained as the adsorption process, the reinforcing effect pointed towards the extensive physio-chemical interaction between bitumen and filler, as shown by Anderson and Goetz [13]. Tunnicliff [14] introduced the concept of gradient stiffening, where he postulated that the interaction between filler and asphalt binder is more prominent at the surface of the filler and reduces outwards. This interaction at the surface is hypothesized to be physiochemical and may vary with the mineralogy of the fillers. Antunes et al. [15], in a recent study, demonstrated that the geometrical characteristics of the fillers are more critical in comparison to the chemical properties while selecting the optimum filler dosage. Clopotel et al. [16] concluded that with the increase in particle surface area, bitumen adsorption increases. It was also found that the chemical composition of asphalt binders and fillers do not affect the properties of the filler-binder system significantly. In general, it is believed that the polar groups of asphalt binders (asphaltenes and resins) are responsible for the adsorption at the surface and describes the interaction between filler and binder.

The characteristics and role of the mastic in an asphalt mix are primarily controlled by the relative amount of filler with respect to the bitumen content of the mix. The ratio of the amount of filler to that of bitumen, which is taken to prepare the mastic, is commonly known as Filler–Bitumen (*F–B*) ratio. It directly influences the viscoelastic properties of the mastics, thereby affecting the performance of the asphalt mix. [17]. The effect of *F–B* ratio on the permanent deformation susceptibility of mastics has been shown in previous studies [18–20].

Many researchers have emphasized on visualizing the asphalt mixture as a mixture comprised of mastic coated aggregates rather than asphalt coated aggregates [21]. The actual binder which holds the aggregate together is not bitumen but the mastic [22]. The rheological exploration of mastic is, therefore, a pressing priority for further assessment of the performance of Asphalt pavements [1, 23–25].

From the literature review, it is evident that the physical and chemical properties of fillers affect the rheological properties of the asphalt mastics. Most of the previous research works have cited physical properties as the most influential factor affecting the asphalt mastic properties. Mixed conclusions are derived from the existing literature on the effect of chemical/mineralogical properties of fillers for the characterization of mechanical aspects of asphalt mastics. An extensive study on the effect of temperature on the LVE characteristics of mastics considering various fillers, *F–B* ratios and covering major intermediate temperature range was the gap which has been bridged by this study. None of the previous studies have attempted to combine the volumetric and filler size variation to assess the stiffening effect of fillers on the mastics. There are two objectives of this study. The first is to understand the effect of various physical properties on the LVE complex modulus denoted as $|G^*|_{\mathrm{LVE}}$ or LVEM of asphalt mastics. Another sub-objective is to identify the most appropriate physical parameters, which can quantify the change in LVEM with the change in filler concentration and temperature. The second objective is to evaluate the relationship between the LVE limit and

LVEM for asphalt mastics. In this study, the effect of six different fillers on the linear viscoelastic properties of asphalt mastics is analyzed at three different temperatures of 10, 20, and 30 °C. Additionally, the effect of physical properties and derived parameters are assessed to understand the interaction of filler-binder composite comprehensively. The LVE complex modulus, and the ratio of $|G^*|_{LVE}$ of mastics with $|G^*|_{LVE}$ of asphalt binder (denoted as $|G^*|_{ratio}$) are used to study the rheological response of the mastics at intermediate temperatures. A relation between linear viscoelastic complex modulus and LVE strain has been established and compared with the standard relationship for asphalt binders.

2 Materials

Six different fillers were used in this study. The selection has been attributed by the properties of the fillers to identify these impacts to LVEM. The general motive was a wide range of filler characteristics like specific gravity (SG), fineness modulus (FM), filler grain coefficient (FGC), Rigden voids (RV) etc. Therefore, a wide range of fillers were selected covering a broad domain of filler characteristics. These fillers included red mud (RM), marble dust (MD), limestone (LS), granite (GR), basalt (BA), and quartz (QZ). Three (RM, MD, LS) out of the six fillers were obtained from India, while the others (GR, BA, QZ) were procured from Austria. RM, also known as bauxite tailings, is a by-product of the Bayer process, method of refining bauxite en route to alumina [26]. Quarrying, processing, and polishing of marble generate tonnes of waste known as MD [27]. The LS dust is extracted from the slurry generated during the cutting and polishing of limestone slabs in the dimension stone industry [28]. Both GR and BA fines are generated from stone crushing operations in respective quarries [29, 30]. The high purity quartz powder had been produced by using iron-free grinding along with the separation process and hence successfully encompassed as a filler in the study.

The fillers obtained from different sources were sieved through 75-micron sieve, dried in an oven, and were stored in separate cans. Asphalt mastics in asphalt mixes has different filler to binder (*F–B*) ratios for different mixes. In typical asphalt mixes the range of filler to binder ratio is broad-ranged. Three filler to binder (*F–B*) ratio (0.5, 1, and 1.5) by weight were used to fabricate asphalt mastics using one viscosity graded (VG) asphalt binder (VG-30) which is most commonly used binder in India. This range of *F–B* ratio covers most common ratios for typical asphalt mixes. Though the fillers were added to the asphalt binder by weight, the concentration of the fillers is described by volume. This is attributed to the difference in specific gravity (SG) of the individual fillers. The volume/volumetric concentration (*V*) refers to the volume of filler with respect to the volume of bitumen in the asphalt mastic. Table 1 presents the description of the fillers with the respective volume concentration at different *F–B* ratio. The basic properties of VG-30 are shown in Table 2.

2.1 Preparation of asphalt mastics

A simple procedure was adopted for the preparation of asphalt mastics in the laboratory. The required amount of filler corresponding to the *F–B* ratio was taken in a pan and weighed accurately on a measuring scale. The pan was then heated in a temperature controlled oven for 1 h at 180 °C followed by the heating of the measured quantity of asphalt binder for 10 min at the same temperature of 180 °C. After mixing the binder and filler in the can, it was agitated continuously with the help of a manually operated mixer for 10 min. The mixing was done in such a way that the homogenous composite prepared resulted in uniform distribution of mineral filler in the mastic. 18 cans of asphalt mastics were prepared and stored for further testing.

Table 1 Volume concentration (%) of different fillers at varying *F–B* ratio

Filler type	Denotation	SG	*F–B* ratio		
			0.5 (%)	1 (%)	1.5 (%)
Red mud	RM	3.218	16	31	47
Marble dust	MD	2.753	18	36	54
Limestone	LS	2.808	18	36	53
Granite	GR	2.741	18	36	55
Basalt	BA	2.895	17	35	52
Quartz	QZ	2.627	19	38	57

Table 2 Basic properties of asphalt binder used in the study

Properties	Values
Penetration, dmm	62
Softening point, °C	48
Viscosity @ 60 °C, Poises	2704
High temperature PG grade	PG 70-22
True fail temperature, °C	74.3

3 Experimental investigation

The experimental investigation carried out in the study included the characterization of fillers and evaluating the LVE properties of asphalt mastics at + 10 °C, + 20 °C, and + 30 °C. The details of these testing protocols are outlined below.

3.1 Characterization of fillers

The fillers greatly influence the mechanical properties of asphalt paving mixtures by influencing the stiffness, oxidation, workability, rutting, moisture damage, and fatigue characteristics [31–33]. They serve different purposes in the bituminous mixture, such as meeting the gradation requirements, stabilizing the mix, increasing the compaction resistivity, stiffening of mix, improving bond strength, etc. [34, 35]. Evidently, the type and amount of filler in the mix are critical. In this study, the characterization of filler was accomplished by various physical, morphological, and chemical tests, as discussed below.

3.1.1 Specific gravity

The specific gravity of the fillers was evaluated using a standard pycnometer method for converting the weight fractions at different *F–B* ratios to volumetric concentration. ASTM D854 [36] was referred for determining the specific gravity of the fillers. The SG values of the fillers are outlined in Table 1.

3.1.2 Particle size analysis

The particle size analysis (PSA) of fine materials with grain size greater than 75 μm is done by allowing the material to pass through a stack of sieves arranged in decreasing order of their sizes. However, for particles smaller than 75 μm, the above-mentioned process cannot be used as the smaller particles tend to stick to the surface of the sieves, being electrically charged. Hydrometer analysis [37] was used for assessing the sieve size distribution of the fillers. The hydrometer measures the specific gravity of the suspension prepared by filler, water, and deflocculating agents at the center of its bulb. Specific gravity depends upon the mass of solids present, which in turn depends upon the particle size. Hence, the particle size curve was obtained with the help of specific gravity.

PSA offers only a qualitative process of understanding the distribution of fine particles in the fillers. The quantification of particle size distribution was done by calculation of Fineness modulus (FM). It represents the relative fineness of the materials. It was calculated by dividing the sum of the percentages of filler coarser than 75, 50, 30, 20, 10, 5, 3, and 1 μm with 100. The finer the composition of filler particles, the smaller the value of FM.

3.1.3 Rigden voids

The intergranular porosity, which is the air-filled space in the bulk material, can be quantified by a parameter popularly known as Rigden voids (RV). BS 812 [38] was used to measure the RV of the fillers used in the study. The test routine involves determining the volume of voids in a dry, compacted bed of fines. The oven-dried filler was compacted in a small mold by a compaction hammer to achieve maximum packing density, which served as an input to obtain the volume of voids. It is an eminent test for the characterization of fillers as the void volume in dry compacted fines is sensitive to change in gradation as well as particle characteristics (shape, size, and texture). As a result, the Rigden voids influences the uniformity and stiffening effect of fillers.

3.1.4 Methylene blue value (MBV) test

MBV estimation is a chemical test that was done to find the suitability of filler for the production of asphalt mixes. It is a French Test method recommended by the International Slurry Seal Association (ISSA) [39]. The spot test specified in EN 933-9 [40] quantifies the amount of harmful clays of the Smectite

(Montmorillonite) group, organic matter, and iron hydroxides present in fine aggregates.

3.1.5 X-ray diffraction (XRD) analysis

XRD analysis was used to characterize the crystallographic structure of fillers. It is based on the constructive interference of X-rays. The atoms of a crystal scatter the x-rays falling on them, which produces an interference so that the information about the crystal structure is obtained from the diffraction pattern. The constructive interference is produced when conditions satisfy Bragg's Law ($n\lambda = 2d \sin \theta$). This law relates the wavelength of electromagnetic radiation to the diffraction angle and the lattice spacing in a crystalline sample [41]. The X-ray Diffractometer known as Rigaku benchtop XRD device with Cu-Kα radiation and 2θ scanning ranging between 5° and 70° (2θ) with 1.5406 Å wavelengths was used to perform the XRD analysis. The analysis was done to determine the prevalent minerals in the composition of the filler. The XRD scans were performed with a scan rate of 5° per minute.

3.2 Linear viscoelastic properties of asphalt mastics

The viscoelastic behavior of asphalt binders can be assessed with the help of Dynamic Mechanical Analysis (DMA) by using a Dynamic Shear Rheometer (DSR). The complex shear modulus (G^*) and the phase angle (δ) are the primal viscoelastic parameters, where G^* is the total resistance to deformation on the application of shear loading to the asphalt binder and δ is the time lag or phase lag between the applied shear stress and strain responses during a test [42, 43].

Traditionally, an amplitude sweep test is conducted using a DSR to obtain the LVE range of an asphalt binder. The LVE strain is defined as the strain level at which the measured complex shear modulus reduces to 95% of its initial value [44]. A plethora of research has been done to evaluate the linear viscoelastic characteristics of asphalt binders [45–50]. Previous researches also showed the limits of linearity of bituminous binders under different loading conditions [51–53].

In the Strategic Highway Research Program (SHRP), the shear stress and strain LVE limits were found to be functions of complex modulus defined by the following equations [54]:

$$\gamma_{\mathrm{LVE}} = \frac{12}{|G^*|^{0.29}} \quad (1)$$

$$\tau_{\mathrm{LVE}} = 0.12 \times |G^*|^{0.71} \quad (2)$$

where γ_{LVE} is the shear strain (%),τ_{LVE} is the shear stress (kPa), and $|G^*|$ is the complex modulus (kPa).

The LVE limits of the asphalt mastics were determined following the similar protocol used for asphalt binders. Anton Paar MCR 102 Dynamic Shear Rheometer (DSR) was employed to determine the LVE range of asphalt mastics obtained by the Amplitude sweep test at a frequency of 10 rad/s for all LVE tests carried out in this study. The sample fabrication involves setting the mastic sample in a silicon mold, followed by sandwiching between the 8 mm diameter parallel plates keeping the testing gap to a value of 2 mm. The trimming of the excess sample was done by the hot trimming tool, and then the sample was allowed to equilibrate with the temperature for 10 min before starting the test. The test was conducted at intermediate temperatures, viz. 10 °C, 20 °C, and 30 °C on three replicates. Therefore, a total of 162 samples were tested to obtain the results.

4 Results and analysis

4.1 Proposed approach

The description of LVE modulus is quantified in two ways: One is the value of $|G^*|_{\mathrm{LVE}}$ obtained from strain sweep test, and the other is the value of $|G^*|_{\mathrm{ratio}}$. Though fillers are defined as materials passing 75-micron sieve, their size may vary considerably [55]. No physical property, as described previously, can quantify the effect of both concentration and size of grains. It is hypothesized in this study that physical parameters when combined with volumetric concentration, can reveal the possible interaction between the asphalt binder and the filler, which can be used for explaining the variation in linear viscoelastic properties with the change in filler concentration and temperature. Literature review in this domain proves that there is a definite change in mastics stiffness and rheological behavior with an increase in volume

concentration. However, it is believed that in addition to the amount of filler, there is a physiochemical interaction, depending on the type of filler, which affects its mechanical behavior. This study also conjectures that this physiochemical interaction is confounded in the physical property of the fillers. It is believed that the surface area distribution of the fillers can give more accurate information about the stiffening behavior of the asphalt mastics. The identification of this physical parameter is made using a hit and trial process. Three physical parameters including, fineness modulus (FM), filler grain coefficient (FGC), and Rigden voids (RV) can be used to describe the distribution of surface area of fillers in the composite. To combine the effect of volume and surface area, *V*/FM, *V*/FGC, and *V*/RV are used for the identification of the most appropriate combination. These parameters will be used to study the variation of $|G^*|_{\mathrm{LVE}}$ and $|G^*|_{\mathrm{ratio}}$ with change in the values of the parameters at different temperatures.

4.2 Physical properties of mastics

Table 3 presents the results obtained for the various physical properties of fillers. All the results presented in the study are an average of three tests done on each sample combination. The variability in the data has been manifested by the standard deviation (SD) values corresponding to Specific gravity, Rigden voids, and MBV test. Additionally, it is ascertained that the test values are perceived to be well below 10% coefficient of variability. The six fillers used demonstrated a wide range for each property. Fillers displayed a broad range of specific gravity with RM having the highest value attributed to the presence of Hematite, whereas QZ has the lowest specific gravity. It can be easily depicted from Fig. 1 that RM has the finest particles among all the fillers. The distribution of particles in RM can be attributed as well graded because of the presence of a wide range of sizes of particles. MD has a very less amount of small sized particles, i.e., < 10 μm. QZ has uniformly distributed particles having almost all sizes. It is very fine in nature, in fact, one of the finest among all the fillers. The lowest curve corresponding to BA shows that the BA is the coarsest amongst all fillers. The particle size distribution in GR and LS is almost similar and in proximity to each other.

Sieve size distribution curve is a qualitative method of identifying the distribution of particle size in a filler system. As described previously, FM can be used to quantify the grain size distribution curve. A lower value of FM indicates finer particle sizes in the filler. D_{60} and D_{10} are also used to quantify the distribution of particles in any soil system. D_{60} is defined as the particle diameter at which 60% of the filler mass is finer than this size, whereas D_{10} is the particle diameter at which 10% of the filler mass is finer than this size. In the filler, a low value of D_{60} indicates the presence of finer grain sizes. Moreover, larger is the difference between D_{60} and D_{10}, wider will be the range of particle sizes. Therefore, a new derived parameter known as Filler Grain Coefficient (FGC) has been introduced in this study to quantify the distribution of particles in the filler system.

$$\mathrm{FGC} = \frac{D_{60} - D_{10}}{D_{60}} \tag{3}$$

A higher value of FGC represents a finer and well graded distribution of particles. The high values of FGC pointed towards the well graded nature of RM and QZ, which is complemented by the particle size

Table 3 Physical and chemical properties of different fillers

Filler	SG	SD, SG	RV (%)	SD, RV	MBV (g/kg)	SD, MBV	FM	D_{60} (μm)	D_{10} (μm)	FGC
RM	3.22	0.016	45.70	1.570	2.5	0.191	4.32	41.00	1.11	0.97
MD	2.75	0.041	12.10	0.586	3	0.144	6.06	50.00	10.00	0.80
LS	2.81	0.015	22.40	1.010	1.5	0.072	5.81	70.00	8.40	0.88
GR	2.74	0.065	27.60	1.890	2.5	0.144	5.70	60.00	7.20	0.88
BA	2.90	0.075	6.33	0.433	3	0.072	6.13	72.00	10.50	0.85
QZ	2.63	0.057	27.8	1.374	1	0.072	5.12	67.00	3.30	0.95

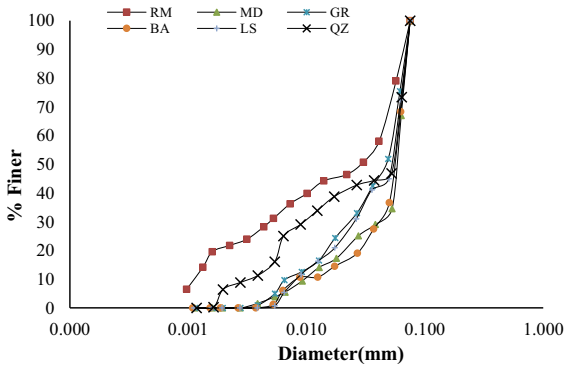


Fig. 1 Sieve size analysis of fillers used in the study

distribution curve too. The FM values also show the results in accordance with the results obtained from Fig. 2. The Highest FM value corresponds to BA, whereas RM has the lowest FM, followed by QZ. Hence it can be concluded from these FM values that Basalt is the coarsest filler, and Red Mud is the finest filler. ISSA [39] specifies that the MBV of fillers should not be more than 10 g/kg. Higher MBV value represents a high amount of harmful clay/smectite, which is undesirable for any asphalt mix. From Table 3, it is clear that all the MBV values are under permissible limits. The lower values indicate that the fillers used in this study do not contain any harmful clays.

Table 4 presents the correlations obtained between various physical parameters. The correlations were established by using a linear fit between the variables. In general, appreciable correlations were observed between FM, FGC, and RV. Previous researches have shown mixed correlations between FM and RV. Faheem and Bahia [56] have observed an appreciable correlation between FM and RV, whereas other researchers reported a poor correlation between the same [57, 58]. The aforementioned correlation matrix portrays a negative correlation of RV with FM, which is also confirmed by Wang et al. [59]. This can be attributed to the corresponding specific surface area (SS) of the fillers. It is known that there is an increase in SS with an increase in the fine particle content. In addition to that, RV are proportional to the SS of the fillers [15]. A negative correlation has been observed in the study. As shown in Table 4, all measured properties strongly correlate with RV and the derived parameter FGC. This indicates that for the chosen set of diverse fillers, FGC and RV can be used as an indicator for characterizing the physical behavior of fillers. MBV, being a chemical test, was not included in the correlation table of the physical properties.

4.3 X-ray diffraction results

The X-ray diffractograms of the studied fillers have been shown in Fig. 2. Silica in the form of Quartz has been found in the composition of almost all the fillers in varying quantities. Calcite is another compound that is found in many of the aforementioned fillers. The occurrence of plagioclase feldspar minerals has been observed in MD and GR. BA had a predominant amount of Anorthite and Pyroxenes. The LS mainly contains Quartz and Calcite, with only a small amount of Aragonite. Microcline is found in a reasonable amount in GR. The predominant minerals in the various fillers have been presented in Table 5. It is difficult, if not impossible, to quantify the mineralogical composition in the fillers. The XRD data reveals the presence of silica and calcite in varying amounts in the different fillers used in the study.

From Table 5, it is evident that most of the minerals, except BA, contains silicate group. To quantify the XRD data concerning the presence of silicate, the total intensity was calculated from the diffractograms. The calculated values, in thousands, were found to be 33, 12, 27, 94, and 242 for RM, MD, LS, GR, and QZ, respectively. An attempt was made to correlate the intensity counts with various physical properties. It was found that the counts, excluding the data of RM, correlated well with the values of FM (R^2 = 0.96), SG (R^2 = 0.87), D_{60} (R^2 = 0.97), and FGC (R^2 = 0.77). The presence of RM results in a poor correlation between the physical properties and silicate intensity. This is attributed to the complexity associated with the characterization of Bauxite residue in comparison to other fillers. Cheng et al. [60] stated that the use of XRD for quantification of mineralogical characteristics of Diatomite produces complicated results. A similar nodus has been noted in this study due to the presence of analogous minerals in RM.

4.4 Linear viscoelastic properties of asphalt mastics

The variation of $|G^*|_{LVE}$ with volume concentration (V) at different temperatures are presented in Fig. 3.

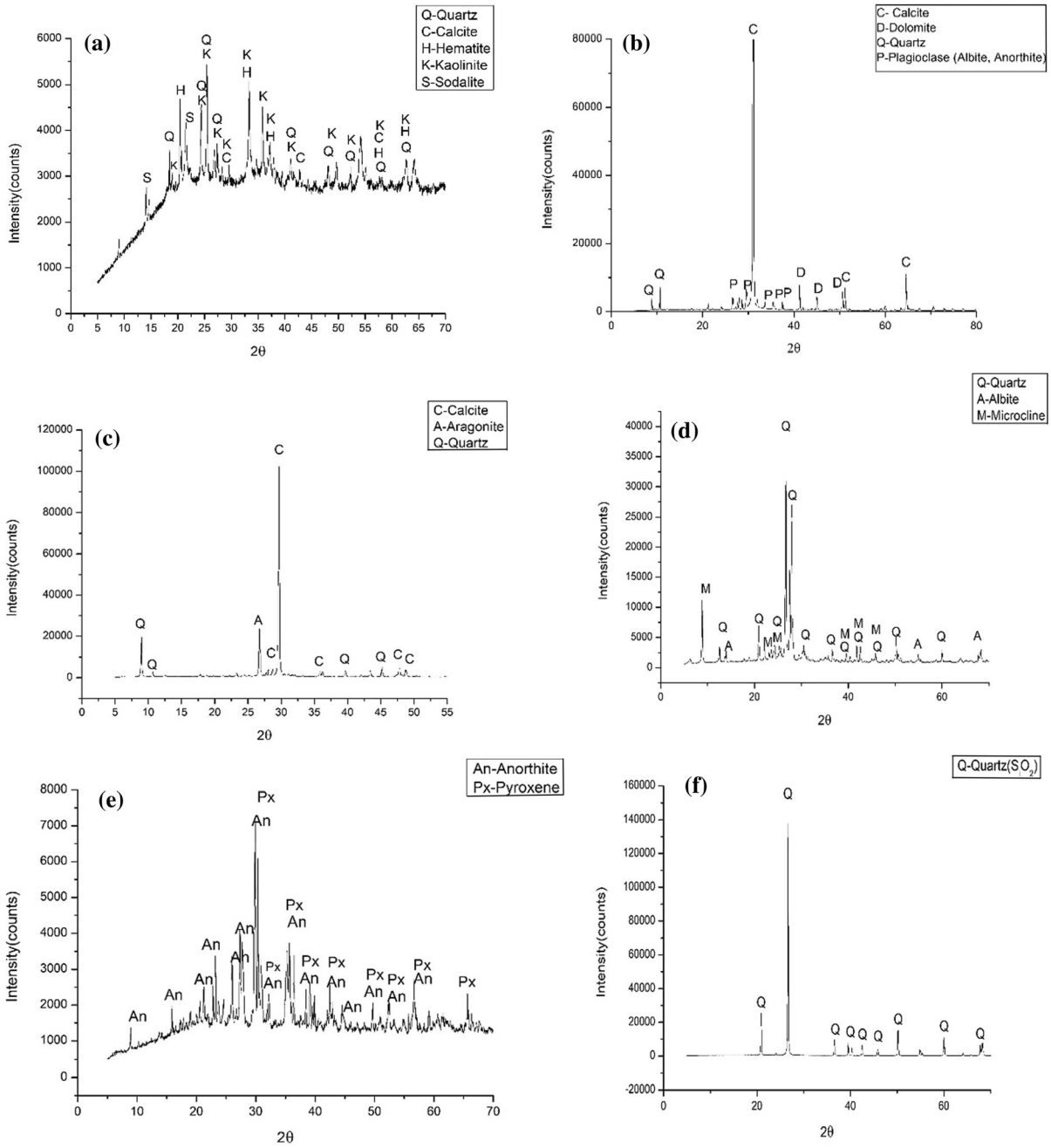


Fig. 2 X-ray diffractograms of **a** RM **b** MD **c** LS **d** GR **e** BA and **f** QZ

The individual curves for all the fillers indicate an exponential increase in LVEM with the increase in *V*. This is in agreement to the conceptual model described by Faheem and Bahia [56, 61] in which there is a transition from diluted to a concentrated regime, with an increase in the concentration of the filler volume in the asphalt mastic. The initial stiffening rate at the lower end of the curve is low and increases gradually with the increase in the concentration of the fillers. As is obvious, with an increase in temperature, the LVEM of the modulus decreases. However, the rate of change in LVEM with an

Table 4 Correlations between different physical properties of fillers

Physical properties	FM	SG	D_{60}	D_{10}	FGC	RV
FM	1.00					
SG	− 0.58	1.00				
D_{60}	0.59	− 0.59	1.00			
D_{10}	0.98	− 0.41	0.48	1.00		
FGC	− 0.91	0.39	0.81	− 0.94	1.00	
RV	− 0.94	0.51	− 0.89	− 0.92	− 0.87	1.00

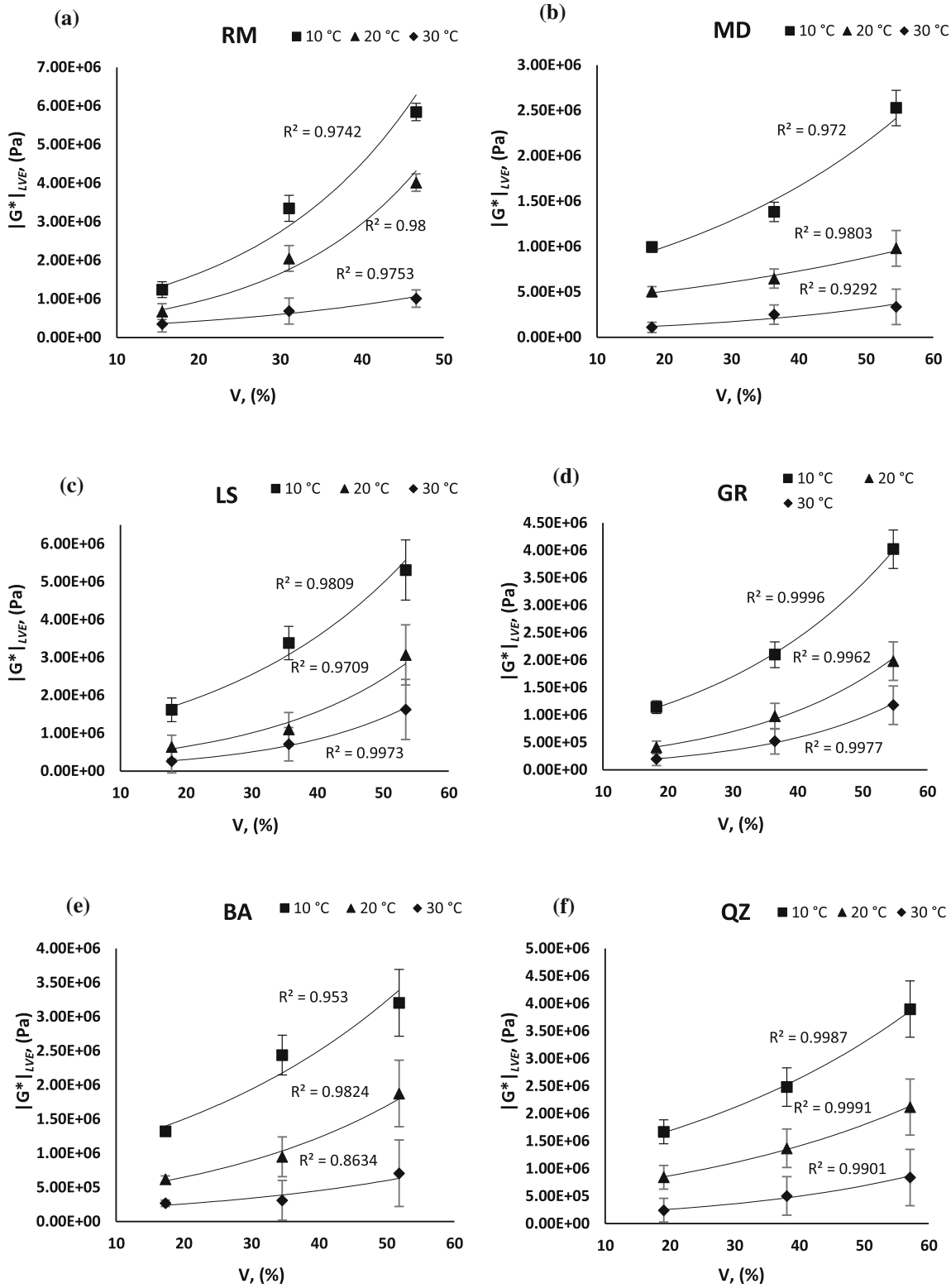
Table 5 Predominant minerals in different fillers

	Hematite	Aragonite	Anorthite	Calcite	Pyroxene	Albite	Kaolinite	Quartz	Dolomite	Plagioclase
RM	✔			✔			✔	✔		
MD				✔				✔	✔	✔
LS		✔		✔				✔		
GR						✔		✔		
BA			✔		✔					
QZ								✔		

increase in *V* is filler dependent. As the fillers dosages were taken corresponding to the *F–B* ratio, different volume concentrations are yielded depending on the SG of fillers. Hence, a definite comparison of the ranking of asphalt mastics cannot be defined. Figure 4 illustrates the comparison between LVEM of mastics at different temperatures. Due to the difference in the rate of increase of LVEM with the increase in *V* for different variables, the ranking changes at different volume concentrations. In general, it was observed that asphalt mastics prepared with MD had the lowest value of LVEM, irrespective of temperature for the major range of *V*. Mastics incorporating RM and LS are ranked highest with GR, BA, and QZ having mixed performance.

Figure 5 presents the variation of $|G^*|_{\text{LVE}}$ with the volumetric concentration (*V*) and the three different assumed parameters, while Fig. 6 depicts the similar variation for $|G^*|_{\text{ratio}}$. The variation of LVEM indicates that *V*/FM appears to the best parameter for describing the variation at different temperatures. In comparison to the volumetric concentration, better correlation (exponential) is obtained between LVEM and *V*/FM. *V*/FGC and *V*/RV were unable to explain the variation in LVEM with coefficient of correlation less than 0.4 for different cases (and hence the values are not shown). For a given *F–B* ratio, *V* is an indicator of the SG of the material. Higher is the SG of the material, lower will be the volume occupied. On the other hand, FM describes the surface area characteristics of the filler. A higher value of FM indicates a coarser distribution of materials and thus smaller surface area. Therefore, a high value of *V*/FM would indicate a higher concentration of finer filler distribution. With an increase in *V*/FM, $|G^*|_{\text{LVE}}$ increases, which is attributed to the increase in filler concentration and higher surface area of the fillers, making the stiffening effect more pronounced. This explains that there is an increase in the gradient of stiffening with the increase in surface area. As is expected, with increase in temperature, $|G^*|_{\text{LVE}}$ decreases. It is also observed that with the increase in temperature, the rate of increase in LVEM increases. However, the rate of increase is not high enough to change the ranking of $|G^*|_{\text{LVE}}$ with the change in temperature within the limits of *F–B* ratio used in this study.

A very interesting observation could be comprehended with the use of $|G^*|_{\text{ratio}}$ for quantifying the effect of filler and temperature on the rheological property of the asphalt mastics. In general, the correlation of $|G^*|_{\text{ratio}}$ and *V*/FM was found to be the highest in comparison to other physical parameters. Unlike the variation of LVEM, $|G^*|_{\text{ratio}}$ ratio gave a


(a)
RM
10 °C 20 °C 30 °C
R² = 0.9742
R² = 0.98
R² = 0.9753
|G*|LVE, (Pa)
V, (%)
(b)
MD
10 °C 20 °C 30 °C
R² = 0.972
R² = 0.9803
R² = 0.9292
|G*|LVE, (Pa)
V, (%)
(c)
LS
10 °C 20 °C 30 °C
R² = 0.9809
R² = 0.9709
R² = 0.9973
|G*|LVE, (Pa)
V, (%)
(d)
GR
10 °C 20 °C 30 °C
R² = 0.9996
R² = 0.9962
R² = 0.9977
|G*|LVE, (Pa)
V, (%)
(e)
BA
10 °C 20 °C 30 °C
R² = 0.953
R² = 0.9824
R² = 0.8634
|G*|LVE, (Pa)
V, (%)
(f)
QZ
10 °C 20 °C 30 °C
R² = 0.9987
R² = 0.9991
R² = 0.9901
|G*|LVE, (Pa)
V, (%)

◀**Fig. 3** Variation of LVEM (at 1.59 Hz) with increase in volume concentration at different temperatures for **a** RM **b** MD **c** LS **d** GR **e** BA and **f** QZ

variable ranking of the mastics with the change in temperature. For lower values of *V*/FM, the ranking of mastics with the change in temperature is as expected: with increase in temperature, the $|G^*|_{\text{ratio}}$ decreases. However, with an increase in *V*/FM, the ranking changes. After a *V*/FM of approximately 6.5, the $|G^*|_{\text{ratio}}$ of mastics at 10 °C is lower than at 30 °C. This is attributed to the rate of change of $|G^*|_{\text{ratio}}$ with increase in *V*/FM at different temperatures. At higher temperatures, the molecular movement in the asphalt binder is more rapid. This allows more adsorption of the binder in the surface of the filler leading to rapid stiffening with the increase in the concentration of filler dispersion. Though previous literatures have indicated that the increase in filler concentration increases the stiffness of the binder, its variation with change in temperature has not been reported previously. Thus this analysis is an important contribution by this study.

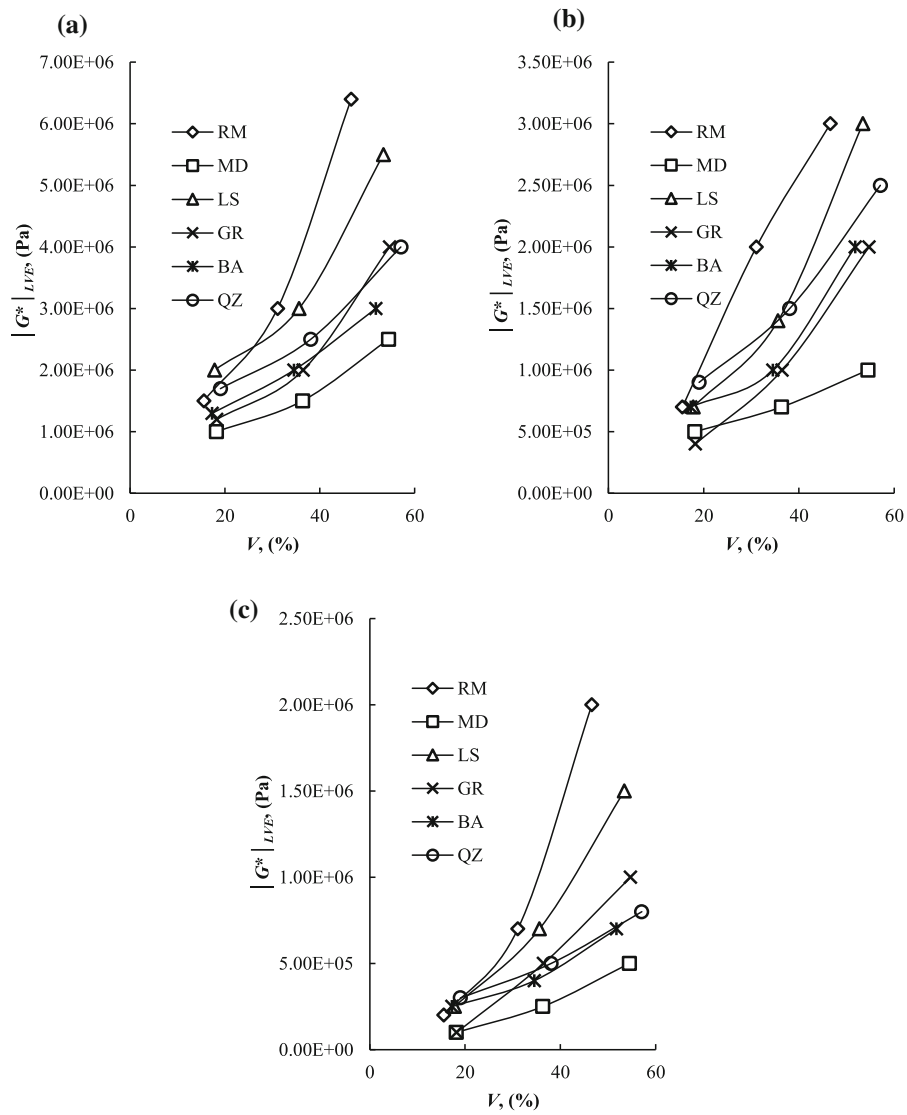


Fig. 4 Effect of filler type on change in LVEM (at 1.59 Hz) at **a** 10 °C **b** 20 °C and **c** 30 °C

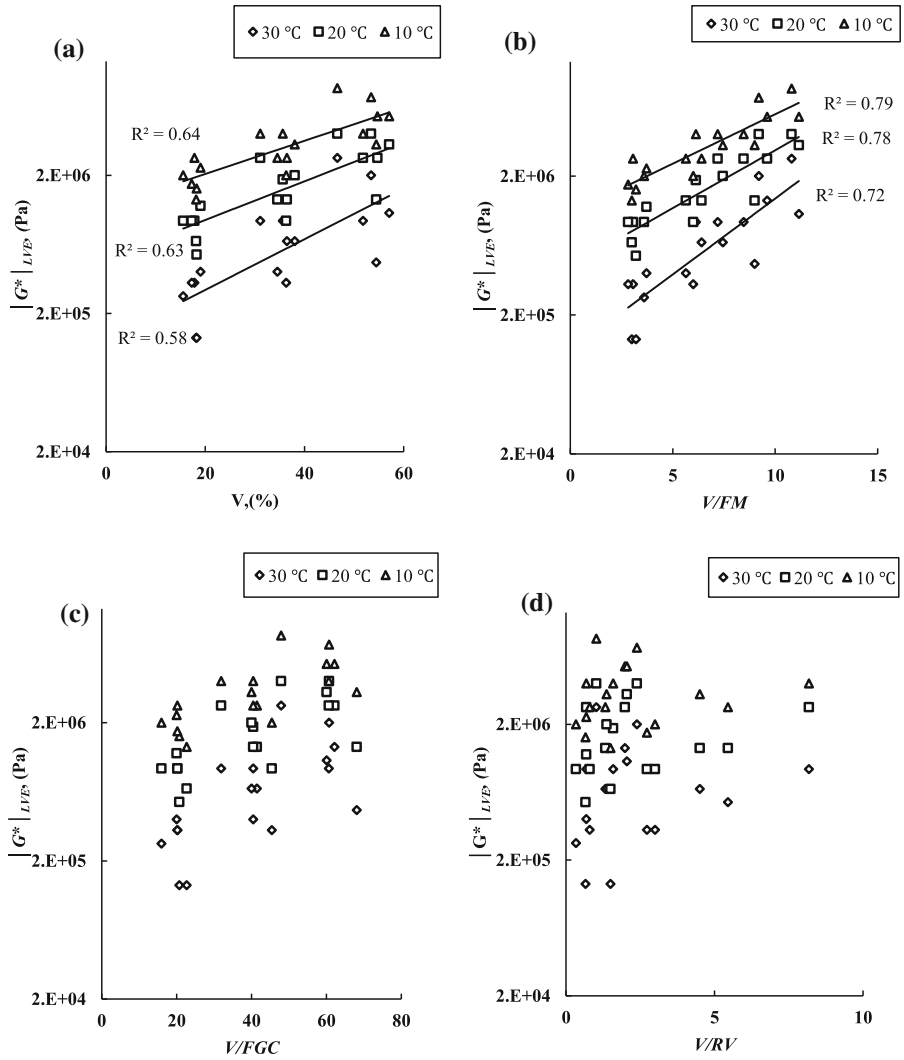


Fig. 5 Correlation of LVEM with **a** *V* **b** *V*/FM **c** *V*/FGC and **d** *V*/RV

4.5 Relationship between LVEM and LVE strain limits

Linear viscoelastic study of any viscoelastic material requires conducting dynamic mechanical experimentation below the LVE strain/stress limit. For asphalt binders, the SHRP relationship as given in Eq. (1) and (2) are adopted for calculating the LVE stress/strain limits. For the study of asphalt mastics, no such relationship has been provided in any literature. Figure 7 presents the relationship between LVEM and LVE strain for the asphalt mastic combinations used in this study. The relationship has also been compared with the existing SHRP protocol for asphalt binders. It can be seen that the LVE strain limit is on the conservative side in comparison to binder values. Additionally, it is found that the relationship between LVEM and LVE strain limit follows an exponential model in contrast to the binder's power law criteria. The relationship illustrated in Fig. 7 is independent of the type of filler and temperature and can be used for evaluating the LVE strain limit in DMA experimentation and analysis for asphalt mastics. However, the results provided are for a single base binder and cannot be generalized. Additional studies are required to support the findings from this work.

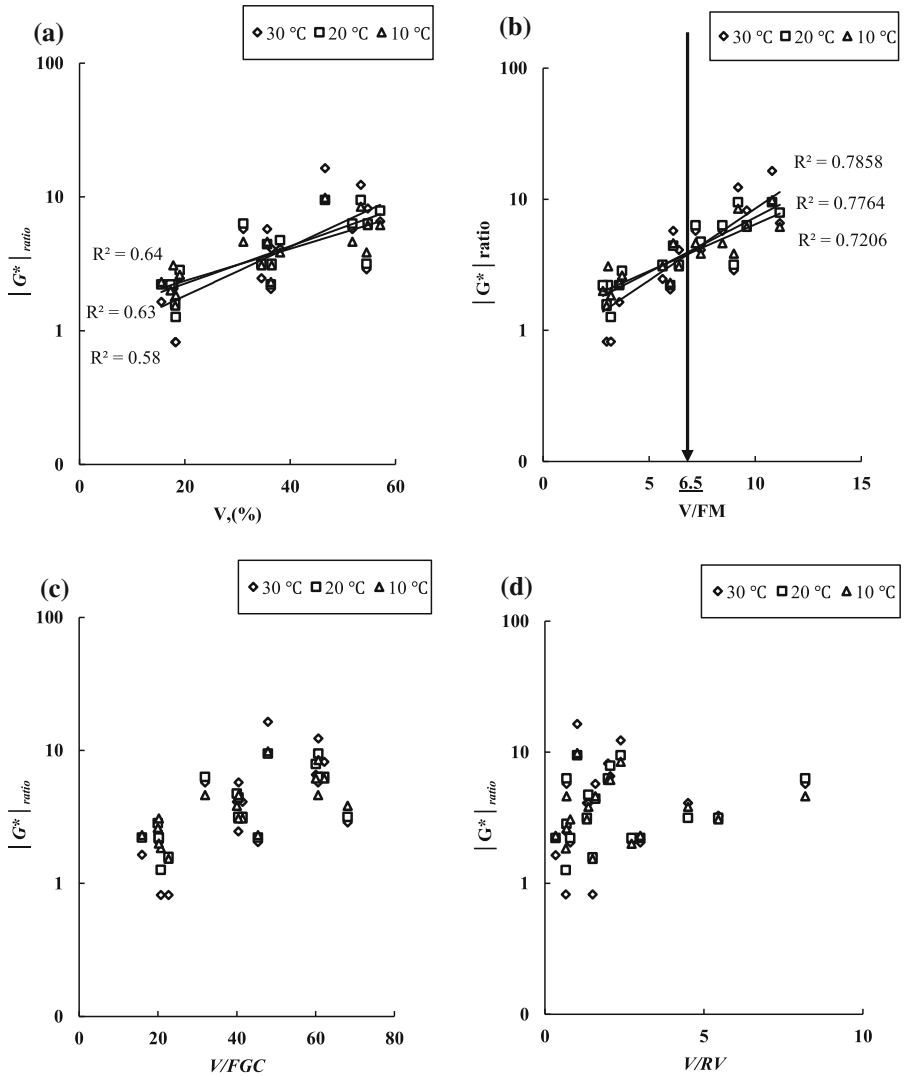


Fig. 6 Correlation of $|G^*|_{ratio}$ with **a** *V* **b** *V*/FM **c** *V*/FGC and **d** *V*/RV

5 Conclusion

In order to assess the filler-binder interaction, this study is pivoted towards the investigation of the physical, chemical, and rheological properties of asphalt mastics by the incorporation of Indian and Austrian fillers. Based on the laboratory investigation and analysis, the following conclusions can be deduced from this study:

- Filler grain coefficient (FGC), which has been introduced as a new parameter for grain size analysis, was found to be reasonable in quantifying the distribution of particle size in the fillers. Hydrometer analysis displayed red mud (RM) as the finest filler, whereas basalt (BA) was found to be the coarsest amongst all. The values of fineness modulus also complemented this observation.
- Correlations established between various physical properties of the fillers indicated that Rigden voids (RV) and FGC are the most suitable candidates, which can be used as an indicator for characterizing the physical behavior of fillers.
- X-ray diffractograms revealed that silicate and calcite are the most common minerals found in most of the fillers. The intensity counts for silicates correlated well with the physical properties of all the fillers except red mud. The XRD data w.r.t

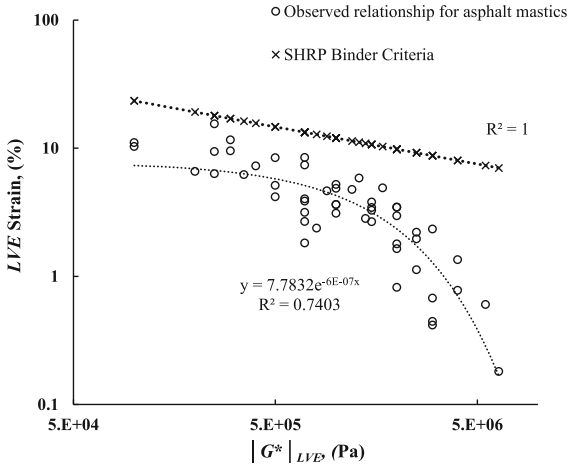


Fig. 7 Relationship between LVEM and LVE strain for asphalt mastics

presence of silicates reveals a good correlation of intensity counts with various physical properties. The presence of heterogeneous mineral groups poses a challenge in the quantification of XRD data for bauxite residue.

- No definite comparison of LVEM ranking could be established with a variation of fillers. Due to the difference in the rate of increase of LVEM with the increase in *V* for different variables, the ranking changes at different concentrations. In general, it was observed that asphalt mastics prepared with MD had the lowest value of LVEM, irrespective of temperature for the major range of *V*. Mastics incorporating RM and LS are ranked highest with GR, BA, and QZ having mixed performance.
- Use of *V*/FM as a parameter to study the variation of LVEM gave the best results in comparison to other parameters, including *V*, *V*/FGC, and *V*/RV. The variation of LVEM evidence *V*/FM as the premier parameter for describing the variation at different temperatures. *V*/FM incorporates the effects of changes in volume as well as the distribution of surface area with the filler system. An increase in filler concentration and surface area leads to a higher stiffening of asphalt mastics, thus increasing the LVEM.
- The variation of $|G^*|_{\mathrm{LVE}}$ and $|G^*|_{\mathrm{ratio}}$ with *V*/FM at different temperatures were not alike. After a *V*/FM of approximately 6.5, the $|G^*|_{\mathrm{ratio}}$ of mastics at 10 °C was found to be lower than at 30 °C. At higher temperatures, the molecular movement in the asphalt binder is more rapid. This allows more adsorption of the binder in the surface of the filler leading to rapid stiffening with the increase in the concentration of filler dispersion.
- The mathematical relationship peculiarly developed for asphalt mastics has been compared with the SHRP protocol for binders. The exponential model followed by mastics shows incompatibility with SHRP criteria for asphalt binders justifying power law.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

Code availability Not applicable.

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