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Rheological models for non-newtonian viscosity of modified asphalt binders and mastics

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ABSTRACT

This paper is aimed at providing a comprehensive experimental work on the viscosity behavior of different modified asphalt binders and mastics, as well as exploring mathematical representations of the results in attempts to predict the viscosity and flow behaviors according to Vinogradov-Malkin and Phillips-Deutsch models. The modified asphalt binders were prepared in the unaged and aged states using styrene-butadienestyrene (SBS), ethylene-vinyl acetate (EVA), and crumb rubber (CR); while the hydrated lime (HL) and fly ash (FA) were selected to produce the binder-filler mastics. To widen the study findings, the additives (polymers, rubber, and fillers) were applied at different possible levels of modification. To prepare the aged bituminous materials, the rolling thin-film oven (RTFO) was utilized for the short-term aging, while the pressure aging vessel (PAV) was further used to induce the possible longterm aging of same materials. Binders and binder-filler mastics were tested using a bob and cup geometry. The viscosity behavior was investigated under various effects of testing conditions including ranges of shear rates and temperatures. It was found that the effect of shear rate dominates and therefore the non-Newtonian shear thinning prevails at low temperatures particularly for binders containing high concentrations of additives. Even the base asphalt binder exhibits shear thinning behavior after a certain limit of shear rate. Based on the studied 38 curves, in general, Vinogradov-Malkin and Phillips-Deutsch models were found in a good agreement with the corresponding measurements especially the latter. © 2019 Egyptian Petroleum Research Institute. Production and hosting by Elsevier B.V. This is an open

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1. Introduction

Due to the limited performance of the base asphalt binder against the ever-increasing traffic loads and severe environmental conditions, several additives with different benefits had been used for many years by highway agencies worldwide to meet an adequate performance of the constructed pavements to minimize the expected distresses. Polymeric additives, crumb rubber, and natural or manufactured mineral fillers (particles less than 75- μ m in size) have been widely researched for the asphalt industry to do so. The incorporated additives interact chemically and/or physically with the asphalt binder [1,2]. As the asphalt binder is the cementing phase in the mixture, therefore plays a major contributing factor to the overall performance of the asphalt pavement. Understanding its rheology or flow behavior is a critical step toward constructing a well-performing pavement [3]. The modified asphalt binder and/or mastic (a blend of mineral filler and asphalt binder) can be considered a composite material (or additive(s) containing binder) consisting of different sized additive(s), viscous asphalt binder, and other phases (might be produced from any possible chemical reactions). The modified asphalt binders and mastics can be treated as structured fluids comprised of multi-phases, exhibit different rheological behaviors due to many factors such as the physical and/or chemical interactions between the additive and binder medium. The rheological behavior of an asphalt binder is very much related to its viscosity [4]. Among asphalt binder's rheological properties, the viscosity is of special interest to the industry. Characterizing the mechanisms that stand behind the non-Newtonian phenomenon of fluids can be very diverse, and it is a dilemma to generalize a scheme

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covering all these mechanisms for different multi-component materials such as modified binders and mastics.

There are a diverse range of conditions that could influence the rheological properties of bituminous materials such as temperatures, oxidative aging, and shear rates during the mixing process and compaction [5]. The viscosity or flow of asphalt binders plays a very important part in the production of the asphalt pavements. Modified asphalt binders might show various signs of non-Newtonian rheology. Thus, understanding the non-Newtonian flow of multicomponent fluids such as the polymers, colloid solutions, and dispersions is a cornerstone of their rheology [6-8]. For the modified asphalt binders and mastics, there is a difficulty to understand experimentally their viscosity behaviors and efforts have been devoted to find better models to understand this behavior. The shear viscosity of those materials is still disputable and requires more answers on their behavior because of the nondefinite non-Newtonian behavior of viscosity [9]. The use of mathematical models, capable of describing the behavior of asphalt binder's viscosity under different possible field conditions could help a better understanding of its rheological properties and contribute to finding better methods to benchmark the mixing and compaction temperatures of their mixtures rather than using approximate methods, such as the equiviscous principle [10,11]. The objective of this work is to present and discuss experimental results of the viscosity behavior of different modified binders and mastics under a wide range of conditions. Vinogradov-Malkin and Phillips-Deutsch models are used hereafter in attempts to explore suitable regression models of the acquired experimental results to represent the viscosity behavior of asphalt binders and mastics under various conditions.

2. Theoretical approach

Under viscosity testing for a wide range of shear rates, the fluid can reveal the general shape of the viscosity curve (Fig. 1). The curve indicates a constant viscosity at low and very high shear rates. These regions are referred to as first and second Newtonian regions, respectively. The viscosity of the first region is called zero-shear viscosity, η_0 , while that at the second region is referred to as the infinite shear viscosity, η . Basically, no single model is recommended by researchers to capture all the signs of non-Newtonian viscosity, and hence multiple models vary in their complexity have been explored to examine their ability to capture the viscosity characteristics of modified asphalt binders [9].

With reference to the shear-thinning behavior, Vinogradov-Malkin and Phillips-Deutsch models have been selected herein to discuss the viscosity results of asphalt binders and mastics in order to explore the applicability of such models for different modified binders and mastics. Although these models are less known for



Fig. 1. Typical viscosity behavior of shear-thinning fluid [6].

asphalt binders, further exploration for simple and more appropriate models is necessary. These models include a shear rate dependency of the viscosity and have been proved effective for many fluids. Vinogradov and Malkin model allows the prediction of the viscosity behavior of many fluids over a wide range of shear rates. This model has been formulated taking into account the fluid viscosity ranging from zero to ∞ [12].

$$\eta = \frac{\eta_0 - \eta_\infty}{1 + a\dot{\gamma}^p + b\dot{\gamma}^{2p}} + \eta_\infty \tag{1}$$

where η is the apparent viscosity, η_0 is the limiting viscosity at a very low shear rate $\dot{\gamma}$, η_{∞} is the limiting viscosity at a very high shear rate and, a, b, and p are constants.

Phillips and Deutsch [13] have formulated a model containing coefficients (C_1 , C_2 , and C_3) to describe the flow behavior of a fluid that has two Newtonian regions at very low and high shear rates and in between, the fluid exhibits shear thinning behavior. The model describes the shear stress τ as follows:

$$\tau = C_1 \left[\frac{1 + C_2 \dot{\gamma}^2}{1 + C_3 \dot{\gamma}^2} \right] \dot{\gamma} \tag{2}$$

3. Methodology

3.1. Materials and preparations

Different additives namely styrene-butadienestyrene (SBS) copolymer, ethylene-vinyl acetate (EVA) copolymer, and crumb rubber (CR), representing the generic types that mostly used in the field, were used as the modifying agents. The utilized polymer products SBS (from Dynasol company) and EVA (from Noble Polymers company) are categorized as elastomeric and plastomeric additives, respectively. The SBS imparts elastic characteristics to binder from the cross-linking of the elemental polymers into its three-dimensional networks while the EVA is characterized by a random structure of ethylene and vinyl acetate [14]. According to the information provided by the suppliers, 30% total styrene in the SBS and the EVA has 22% vinyl acetate. The rubber was prepared using the grinding process from waste tires with a maximum size of 600-µm (30 mesh). In addition, hydrated lime (HL) and fly ash (FA) mineral fillers passing through No.200 sieve were used to prepare asphalt-filler mastics. The HL and FA are characterized by basic and acidic nature respectively [15]. Each polymer type or rubber additive was blended separately at percentages of 3% and 5% with a PG 58-28 base asphalt binder. The base binder PG 58-28 used in the present study is supplied from a refinery in Michigan, USA. The measured Superpave properties of the base binder are presented in Table 1.

Each mineral filler was blended with the asphalt binder at mass fractions (i.e. by %wt. of asphalt binder) of 70% and 115% within the recommended range of dust-to-binder ratio (0.60–1.60) in the Superpave design. The specific gravity of asphalt binder γ_b was

Table 1			
Measured properties	of base	asphalt bind	ler.

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	Binder state	Measured property	Result
	Unaged binder	Viscosity at 135 °C (mPa.s) (≤3000)	430
		G*/sin δ at 58 °C (kPa) (≥1)	1.85
	RTFO-aged binder	G*/sin δ at 58 °C (kPa) (≥2.2)	4.13
	PAV-aged binder	G*.sin δ at 19 °C (kPa) (≤5000)	2400.80

RTFO: rolling-thin film oven); PAV: pressure aging vessel; G*: complex shear modulus; δ : phase angle

assumed as 1.02 GCC while the specific gravities of the mineral fillers γ_f were calculated using the helium pycnometer test (ASTM D5550). The calculated volume fraction V_f (Eq. (3)) [16] corresponding to a prespecified mass fraction M_f of each mineral filler is presented in Table 2.

$$V_f = \frac{\gamma_b M_f}{\gamma_f + \gamma_b M_f} \tag{3}$$

All the modified binders and mastics were blended using a laboratory higher shear mixer. Supplemental hand mixing was performed prior to the mechanical mixing to initially agitate the additives to help disperse the particles, especially for the highly modified mastics. Before mixing, the base asphalt binder and each prespecified amount of additive were preheated to 180 °C and after reaching the target temperature, the mechanical mixing was performed continuously for 2 h at 2000 rpm. A portion of each prepared sample of base binder and modified asphalt binder was subjected to the short-term aging (at 163 °C for 85 min) using the rolling thin film oven (RTFO), ASTM D2872. The pressure aging vessel (PAV) procedure (ASTM D6521) also was used to induce the long-term aging of the RTFO-aged binder (at 100 °C for 20 h). Sedimentation and storage problems in the binders were minimized by testing right after the preparation.

3.2. Measurements and method

To study the viscosity behavior of the modified asphalt binders and mastics, the bob-and-cup (concentric cylinder) geometry of an MCR 302 DSR (Anton Paar) was employed. In this geometry, a prespecified volume of the test sample is poured into a cup (outer cylinder) and the inner cylinder (bob) is inserted concentrically (coaxially) during the test. In this measuring system, the bob rotates and the cup with a narrow gap apart from the bob is fixed. The bob-and-cup measuring technique has been well-described previously by others (e.g. [17]). AASHTO T 316 is the commonly used protocol for evaluating the viscosity of asphalt binders. However, for the modified asphalt binders, in particular, the equiviscous principle is not fulfilling the practical requirements as the viscosity is highly shear rate dependent. In the plant mixing process or in laboratory mixers, the asphalt binders exhibit turbulent mixing with extremely high shear rates [5]. In this study, the RheoCompass software (ver. 1.17) was used to calculate the resulting shear stress corresponding to shear rates ranging from 5 to 1000 s⁻¹ (~5 s⁻¹ interval). Hence the flow and viscosity data over the prespecified range were recorded automatically. A matrix of the prepared materials and testing temperatures are provided in Table 3. The ranges of testing temperatures 105 °C to 165 °C for binders and 135 °C to 165 °C for mastics were selected to suit the conditions. It is worth noting that these temperatures (105 °C and 135 °C) were selected as the lowest temperatures that can be used to run the test, i.e. if lower testing temperatures are used the material will be too sticky and this would exceed the maxi-

Table 2

Densities and fractions of mineral fillers.

Filler type	Density (γ_f) (gcc)	Volume fraction (V_f) corresponding to mass fraction (M_f)	
		Mastic 1 (M _f =70%)	Mastic 2 (M _f =115%)
HL	2.2915	23.76	33.86
FA	2.6087	21.49	31.02

Table 3

Prepared materials and testing temperatures.

Material	Percentage of additives (by mass of base binder)	Case of material	Testing temperatures
Base binder	-	Unaged	105 °C and 165 °C
		RIFO-aged	
SBS-modified	3%	I Inaged	
binder	5/0	RTFO-aged	
		PAV-aged	
	5%	Unaged	
EVA-modified	3%	Unaged	
binder		RTFO-aged	
		PAV-aged	
	5%	Unaged	
CR-modified	3%	Unaged	
binder		RIFO-aged	
		PAV-aged	
	5%	Unaged	
HL-mastic	70%	Unaged	135 °C and 165 °C
	115%		
FA-mastic	70%		
	115%		

mum torque of the rheometer. The proprietary software is capable of fitting the acquired data to Vinogradov-Malkin and Phillips-Deutsch models, and the goodness of fitting could be examined with the aid of the provided coefficient of determination denoted by R^2 to determine how close the data are to the fitted regression model. The higher the coefficient of determination, the better the selected model as it fits the data well.

4. Analysis of results

Fig. 2 presents the viscosity-shear rate plots for the unaged binders (at 105 °C and 165 °C) and mastics (at 135 °C and 165 °C). As seen, the shear-thinning prevails at low testing temperatures for both the binders and mastics. The addition of polymers generally allows for substantial differences in the viscosity of the asphalt binder. In the case of CR, the increase was not that high compared to polymer-modified binders. For asphalt mastics, the increase in the mass/volume fraction of the mineral filler from low or moderate $(M_f=70\%)$ to concentrate $(M_f=115\%)$ also changes the viscosity significantly. Previous work by Diab [10] showed that the viscosity of polymer-modified binders and HL-mastic is highly shear rate dependent, also depending on the concentration of the additive. Clearly, the increase in the temperature results in an obvious decrease in the viscosity. The viscosity of base asphalt binder kept constant which appeared as an upper Newtonian plateau within a specified range of shear rates (up to 200 s⁻¹), i.e. can be considered a Newtonian fluid within this range of shear rates, then the shear thinning started. The shear-thinning behavior of base binder has been proven experimentally by a number of studies (e.g. [7,18]). Although, apparently the base asphalt binder seems homogeneous, in fact, it is not, or in other words, a multi-component matter consisting of several components-SARA (saturates, aromatics, resins, and asphaltenes) [19]. After a certain limit of shear rate, the base binder shows shear-thinning as well, but to a lesser extent compared to the modified binders or mastics. When shearing begins, the viscosity of the asphalt binder is high but it drops with increased rate of shearing. This conclusion is shared with others (e.g. [7]). One possible mechanism, in the shearing process of polymer fluids, randomly orientated long molecules tend to align which reduces the apparent viscosity of the whole fluid with

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Fig. 2. Viscosity-shear rate of unaged binders at: (a) 105 °C, (b) 165 °C, and of mastics at: (c) 135 °C, and (d) 165 °C.

increased shear rates [20]. The rubber-modified binder exhibited lower viscosity compared to the polymer-modified binders because of the strong cross linking of polymers. Not only the polymer content has a pronounced effect on the viscosity of modified binders, also the type of the polymer additive largely contributes to the measured viscosity; however mixed results were attained with this regard. In other words the chemistry between polymers and binder, especially after age-hardening complicates the interpretation of the viscosity of polymer-modified binder, thereof chemical studies are required to provide insight in this regard. In

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Fig. 3. Viscosity-shear rate curves of RTFO-aged binders at: (a) 105 °C and (b) 165 °C.



Fig. 4. Viscosity-shear rate curves of PAV-aged binders at: (a) 105 °C and (b) 165 °C.

the case of mastics, same behavior was registered. It appears that filler type also affects the viscosity of binder-filler system. The HL and FA have different chemical natures, basic and acidic, respectively. In other words, the type, nature, and content of the mineral filler have a pronounced effect on the viscosity. Asphalt binders inevitably oxidize during production processes and service life of the pavements and limited results, if any, are available in the literature regarding their viscosity behavior. The shown results (Figs. 3 and 4) confirm the changes in the viscosity due to the effect of oxidative aging. The increase of the oxidative aging allows for the depletion of low fraction components in the asphalt binder and increases the viscosity [21]. From Figs. 3 and 4, it can be seen that the effect of shear-thinning of polymers and rubber also is most pronounced at low temperatures where shear rates are

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Fig. 5. Sample measurements of unaged binders and mastics and corresponding regression curves according to Vinogradov-Malkin model (a) and Phillips-Deutsch model (b).

greatest. Curve fitting using good modeling functions is used to describe the complete behavior of the viscosity for further prediction of the material's behavior under a wide range of conditions. In this study, the goal is not to discuss all the commonly used functions proposed for the approximation of the non-Newtonian shear-thinning viscosity of asphalt binders and mastics, but to explore the possibility of applying new functions namely Vinogradov-Malkin and Phillips-Deutsch models. Examples of the fitted curves based on the selected models are presented in Fig. 5a and b. Vinogradov-Malkin model was fitted to the measured data using RheoCompass 1.17 software, which also provided the correlation coefficient to examine the goodness of fitting of all38 curves. For the adopted models, in general, a good prediction of the experimental data was proven in terms of the acceptable coefficient of determination (Fig. 6). The goodness of fitting was examined with the aid of the values of R² compared to each other, i.e., the higher the better, also the model that is better representing the viscosity for more materials is the most preferable. In this sense, the fitting results of both models indicate that both models can accurately represent the viscosity behavior of binders and mastics over a wide range of conditions. However, in terms of the highest coefficient of determination and number of materials that are best represented, the Phillips-Deutsch model provided higher values for a wider range of tested materials compared to Vinogradov-Malkin model.

5. Summary and conclusions

The strategy of incorporating additives in bituminous mixtures is known as an important direction to increase the quality of pavements. These additives come in contact with the asphalt binder creating complex blends significantly controlling the performance of the asphalt mixtures during construction and service life. The viscosity of the blend, i.e. modified asphalt binder or mastic, is a critical property in the science of asphalt rheology. Knowledge the viscosity characteristics of modified binders and mastics is important because it controls the flow behavior as well as the determination of the mixing and compaction temperatures (or rather the workability) of their asphalt mixtures. The current study was devoted to address the effects of temperature, shear rate, and aging on the viscosity behavior of different asphalt binders and mastics. In addition, representing the experimental data using suitable models was examined through using Vinogradov-Malkin and Phillips-Deutsch models. The modified binders and mastics are expected to exhibit more non-Newtonian behavior compared to the base binder. Therefore, cannot be described by a small range of conditions. A wide range of shear rates was applied along with temperature variations. The modified binders of this study included polymer-modified binders prepared with styrene-buta dienestyrene (SBS) and ethylene-vinyl acetate (EVA) and crumb rubber (CR)-modified binder. To prepare mastics, two mineral fillers namely hydrated lime (HL) and fly ash (FA) were used. The mentioned additives were blended with the base asphalt binder (PG 58-28) with different concentrations. The asphalt binders were produced as unaged and using the laboratory short- and long-term aging ovens. The bob-and-cup geometry of a dynamic shear rheometer was employed to run the measurements. Based on the study findings, the modified asphalt binders and mastics are characterized by a distinctive shear-thinning behavior. The critical shear rate of the base binder at which a transition from Newtonian to non-Newtonian flow occurs was smaller than that for the modified binders and mastics. The regression analysis according to Vinogradov-Malkin allowed obtaining a satisfactory mathematical representation for the studied materials. However, Phillips-Deutsch model better represented most of the studied materials with a higher coefficient of determination.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Fig. 6. Coefficient of determination of the fitted models: Vinogradov-Malkin model (a: unaged binders; b: mastics; c: RTFO-aged binders; d: PAV-aged binders) and Phillips-Deutsch model (e: unaged binders; f: mastics; g: RTFO-aged binders; h: PAV-aged binders).

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