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Relationship Between Rheological Properties of Nano Polymer Modified Asphalt Binder and Permanent Deformation of Asphalt Mixture

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Abstract: Asphalt binder plays an important part in determining many aspects of road performance. However, the rheological properties of asphalt binder are very complex and the parameters depend purely on the viscosity, various loading time and temperature. Therefore, relationship study on asphalt binder rheological properties and asphalt mixture is vital to predict the performance of the mixture. This paper evaluates the relationship between rheological asphalt binder and asphalt mixture performance containing nanopolymer modified binder. Five sets of asphalt binder rheology were tested to determine their viscosity, effect of short term and long-term aging using the dynamic shear rheometer (DSR). The asphalt mixtures performance test was then conducted to evaluate the permanent deformation of the mix. Findings from this study indicate that the rheological properties of asphalt binder acts as indicator for the asphalt mixture performance. The G*/sin δ and viscosity of the asphalt binder significantly agree with the resilient modulus and rut depth results obtained. The dependent (resilient modulus at 40°C and rut depth) and dependent (G*/sin δ and viscosity) variables show that these variables significantly affects each other. An effective prediction models can also be developed according to predicted and measured permanent deformation values.

Keywords: Rheological properties, viscosity, permanent deformation, resilient modulus and nanopolymer

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1. Introduction

Over the years, pavement failures due to permanent deformation or rutting has been a big challenge to the authority to maintain existing road networks. Increase in traffic volume especially heavy vehicles and environmental condition contributes to the deterioration of the pavement. Recently, many studies have been conducted on modification of asphalt binder using different types of polymer modifier. The addition of polymers typically increases the rigidity of the asphalt binder and improves on the temperature susceptibility. Polymer modified asphalt binders also showed improvement to the mechanical properties of hot mix asphalt (HMA) [1]. Several properties of asphalt mix are improved which include fatigue life, temperature susceptibility and resistance to permanent deformation [2[-[4]. All of these attributes improve the overall pavement performance.

Modification of the asphalt binder can increase temperature stiffness, which boosts resistance to rutting, bleeding, and flushing. It can also reduce low temperature stiffness, improve fatigue resistance, improve asphalt age hardening resistance and provide stiffer hot mix layers [5]-[7]. From literature review, few studies were conducted using nano polymer such as nanoclays, carbon nanotubes, nanopolyacrylate, etc. The effectiveness of nanopolymer in altering hierarchical structure of composite materials due to their surface properties has seen a tremendous development in recent years [6]-[9]. Zafari also conducted a study on the potential benefits of nanosilica particles in asphalt mixtures and concluded that introduction of nanosilica to asphalt binder can improve the anti-aging property, rutting performance and rheological properties of asphalt binder [8]. Study by Yao reported that addition of nanoclay and carbon microfiber improves the permanent deformation mixtures performance. In addition, nanosilica modified asphalt binder performed better for rutting and fatigue cracking resistance compared to conventional asphalt binder and carbon microfiber modified asphalt [9].

The study on rheological properties of asphalt binder is an important phenomenon to determine the overall performance of modified asphalt binder. The Superpave tests which are, rolling thin film oven, pressure aging vessel and dynamic shear rheometer are related to performance parameter of the asphalt mixture which is influenced by the asphalt binder. These tests are used in the Superpave performance grade asphalt binder specification to characterize the viscous and elastic behaviour of asphalt binders at high and intermediate service temperatures. The rheological testing such as dynamic shear rheometer (DSR) measures the complex shear modulus (G*) and phase angle (δ) of the binder over a temperature range from 46°C to 76°C. The complex modulus (G*) and the degree of phase angle (δ) is used to determine the relationship between asphalt binder stiffness and the type of deformation: recoverable and non-recoverable. In the Superpave requirements, the G*/sinδ relationship is used as an indicator to determine the rutting of the asphalt binder at high-performance temperature[5-6]. This relationship follows the rationale that an asphalt binder with a high G* value is stiffer, which increases its resistance to deformation. Asphalt binder with low sinδ value is more elastic, hence the ability to recover part of the deformation is increased [12]. It is important to understand this relationship to ensure stable and durable asphalt pavement. Hence, this study focussed on the rheological properties of the nano modified asphalt binder and asphalt mixture performance.

2. Materials and Method

2.1 Aggregate Properties

In this study, granite aggregates were supplied by Blacktop Quarry, Rawang. The aggregates were prepared through washing, oven drying and sieving processes. The aggregate tests conducted for this study are Los Angeles Abrasion Value, Aggregate Impact Value, Value and Flakiness Index Value. Table 1 presents the aggregate properties result. The aggregate properties conformed to Public Works Department of Malaysia's (PWD Malaysia) Specification for Road Works requirement which indicates good quality thus acceptable for use in road works.

Table1 - Aggregate properties.

Aggregate Test	Specification	Result
LA Abrasion	<45%	25.35%
Aggregate Impact Value	<45%	21.75%
Flakiness	<20%	3.1%
Elongation	<20%	16.6%

2.2 Asphalt Binder Modification

In this study, a base binder of PEN80-100 was modified with nanopolyacrylate (NP). The NP was supplied by Nan Pao Resin Chemical Co. Taiwan which acts as modifier and is commonly known as acrylics which belongs to a group of polymers commonly used. The NP consist of 39% to 40% polyacrylate resin with an average diameter of 50nm.

Initially, 500g of base asphalt binder was heated to 110˚C and poured into a 500 ml container. The asphalt binder was further heated in the oven at 150˚C until it liquefies. Next, the NP was added slowly into the liquid asphalt binder and sheared using a mechanical stirrer attached to a high shear mixer until the temperature reach 160°C. At this stage, the mixing cycle speed is increased to 500 rpm for 30 minutes. The mixing temperature is then maintained at 160 ± 5°C throughout the mixing progress. The NP modified asphalt binder was then evaluated for further rheological properties tests. From the previous study, the optimum NP polymer used to modify the asphalt binder is 6% [13].

2.3 Marshall Mix Design

The volumetric properties and the optimum binder content (OBC) for control mix and nanopolymer modified asphalt binder mixes were prepared based on the Marshall mix design method in accordance with ASTM D1559. Asphalt wearing course of AC14 aggregate gradation was selected as shown in Fig. 1 selected according to Public Works Department of Malaysia's (PWD Malaysia) Specification for Road Works [14]. A total of 15 samples were prepared with asphalt binder content ranging from 4.0% to 6.0% for the control mix (0% NP) and modified asphalt binder mixes with nanopolyacrylate (NP) contents of 2% (NP2), 4% (NP4) and 6% (NP6) by weight of asphalt binder. All the AC14 Marshall samples were compacted at 75 blows/face. The volumetric properties analysis was determined by obtaining the bulk specific gravity, theoretical maximum density, voids in total mix (VTM), voids in mineral aggregate (VMA) and voids filled with asphalt (VFA). Individual plots for VTM, VFA, density, stability and flow against asphalt binder content were analysed to determine the optimum binder content (OBC) of the asphaltic mix. The OBC value is then used to prepare samples for the resilient modulus and rutting resistance test.

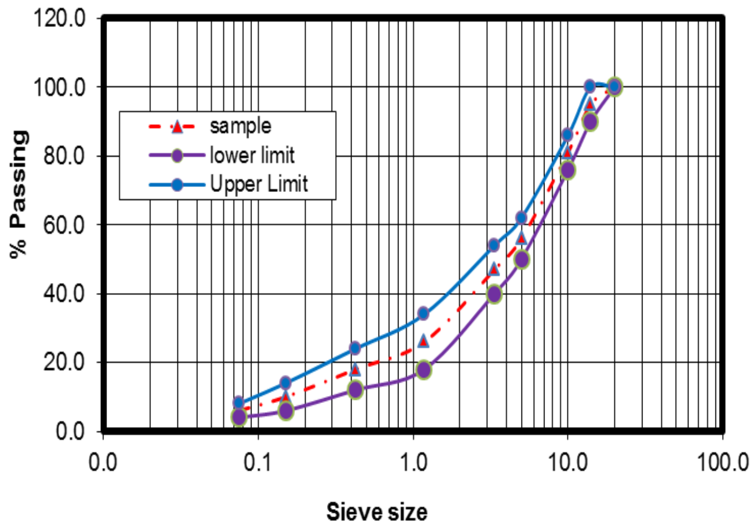

Fig. 1 - Gradation Limit for AC14.

Table 2 presents the optimum binder content (OBC) result and the volumetric properties for AC14 mixtures in this study. The optimum binder content for AC14 mixtures. The OBC for AC14 control mix is 5.2% and AC14-NP2, AC14-NP4 and AC14-NP6 is 5.3%, 5.5% and 5.6% respectively. The stability of the mixtures also increase as more NP is added to AC14 mix ranging from 1120 kg to 1298 kg. The flow value results also showed decreasing trend with increase in stability of the AC14 mixtures. All the AC14 mixtures conform to JKR specifications and hence were further evaluated for performance tests.

Table 2 - Volumetric properties for AC14 mixtures.

Mixture type	AC-Control	AC14-NP2	AC14-NP4	AC14-NP6	Specification (JKR/SPJ/ 2008-S4)
Polymer content (%)	0	2	4	6	-
OBC (%)	5.2	5.3	5.5	5.6	-
Stability (kg)	1120	1170	1270	1298	>815
Flow (mm)	3.56	3.46	3.35	3.28	2.0-4.0
Stiffness (kg/mm)	314	341	382	392	>203
VTM (%)	3.6	3.8	4.2	4.4	3.0-5.0
VFB (%)	77	74.5	74	73.5	70-80

2.4 Rheological Asphalt Binder Tests

The rheological properties of asphalt binder are conducted using the viscosity, rolling thin film oven (RTFO), pressure aging value (PAV) and dynamic shear rheometer (DSR) test. These tests are important to evaluate the overall performance of the modified asphalt binder.

The RTFO test is developed according to AASHTO T240 and ASTM D2872. This test provides an aged asphalt binder for further testing by the dynamic shear rheometer (DSR) and is used to simulate the short-term aged asphalt binder at elevated temperatures to simulate aging during manufacturing and construction of HMA pavement. The asphalt binder samples were poured in glass containers and placed in RTFO carriage with the opening of glass containers facing the jet air in the chamber. The ageing process continues for 85 minutes at 163ºC with the carriage rotating speed of 15 rpm.

The standard DSR test is based on AASHTO T315 and this technique is used for the determination of fundamental characteristics of modified asphalt binders. This test is applied in the Superpave performance grade asphalt binder specification to characterize the viscous and elastic behaviour of asphalt binders at high and intermediate service temperatures. The DSR also known as dynamic shear rheometer or oscillatory shear rheometer is used in the plastic industry for many years due to its capability of quantifying both elastic and viscous properties [15]. The DSR evaluates the complex shear modulus (G^*) and phase angle (δ) of asphalt binders at the desired temperature and frequency of loading. The complex modulus (G^*) can be considered as the total resistance of the sample deformation when repeatedly sheared and the phase angle (δ), is the lag between the applied shear stress and the resulting shear strain. The results of G^* and δ are generally used as indicators of HMA rutting and fatigue cracking [16]. Rutting and fatigue cracking are the greatest concern in the early and later stage of a pavement life.

The PAV is used to simulate the asphalt binder aging that occurs during 5 to10 years of in service HMA pavement. There are few advantages of pressure aging of the asphalt binder such as: limited loss of volatiles, the oxidative process is accelerated without resorting to high temperatures, an adequate amount of asphalt binder can be aged at one time for further testing, and the test is practical for routine laboratory testing. The PAV is used to age RTFO residue which provides simulated long term aged of the asphalt binder in the HMA pavement.

2.5 Resilient Modulus Test

Resilient Modulus is an important parameter that is used in the mechanistic pavement design as an input to the multilayer layer elastic analysis or finite element models to compute the pavement response under traffic loading. Indirect tensile test for resilient modulus of bituminous mixtures is performed in accordance with ASTM D4123. Resilient modulus of asphalt mixtures, measured in the indirect tensile mode, is the most popular form of stress-strain measurement used to evaluate the elastic properties. The indirect tensile resilient modulus test is carried out using the Universal Testing Machine (UTM-5P) as shown in Fig. 2. The test parameters are shown in Table 3.

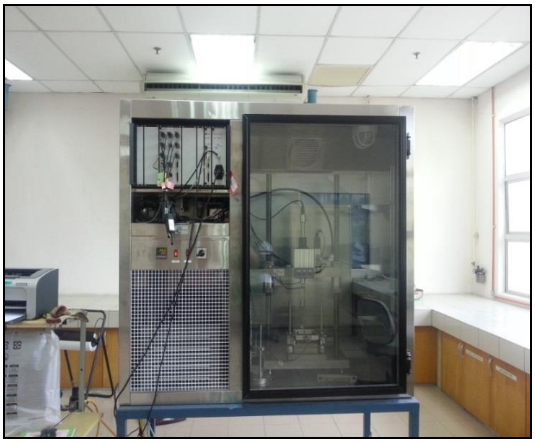

Fig. 2 - Resilient modulus test device.

Table 3 - Indirect tensile resilient modulus test parameter.

Test Condition	Parameter
Temperature (ºC)	25 and 40
Load Pulse (ms)	100
period (ms)	1000, 2000, 3000
Applied Load	Induce 10% of the indirect tensile strength

2.6 Rutting Test

Rutting or permanent deformation of the laboratory designed asphalt mixtures are determined through Asphalt Pavement Analyzer (APA) test conducted according to AASHTO TP 63. The APA wheel tracking device operates under a pair of wheels by applying moving loads above two rubber hose to the specimens to simulate the rutting performance. The test measures the depth and number of wheel passes until failure. Each moving steel wheel of APA machine is 8 inches (203.6 mm) in diameter and 1.85 inches (47 mm) wide. For APA testing, eight cylindrical samples were compacted using Superpave Gyratory Compactor as depicted in Table 4. The desired density of HMA mixture was obtained by adjusting the weight of the mixture. Prior to testing, the specimens were conditioned in the APA chamber for two hours at 60°C with full confinement in the test mould. The pressure of rubber hose and wheel load was respectively set up at 690 kPa (100psi). The rut measurements were obtained at 0, 25, 4000 and 8000 loading cycles respectively. Fig. 3 and Fig. 4 display the condition of the specimens before and after the test.

Table 4 - Design matrix for APA rutting test.

Asphalt Mixture Type	Nanopolymer Content (%)	Rutting Specimen
AC14-Control	0	2
AC14-NP2	2	2
AC14-NP4	4	2
AC14-NP6	6	2

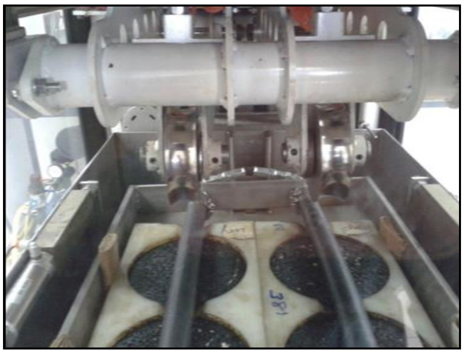

Fig. 3 - Specimen before the APA test.

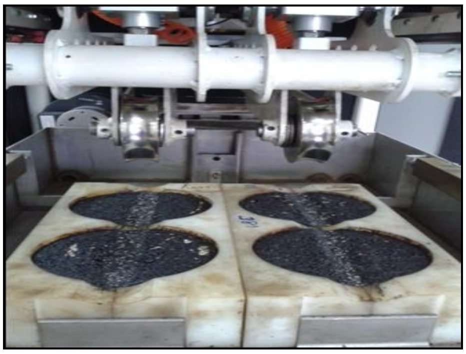

Fig. 4 - Specimen after the APA test.

2.7 Regression Model of Permanent Deformation of Nanopolymer Modified Asphalt Mixture

In this study, the effect of nanopolymer modified asphalt binder on rutting and resilient modulus performance were established through regression models. The relationship between rheological properties of the nanaopolymer modified asphalt binder and asphalt mixture performance are evaluated based on the results obtained. Simple linear regression is used to analyse relationships and is characterized by straight line, or by generalizations of straight lines. Apart from knowing the strength and direction of relationship, regression analysis can also be used to make prediction. Prediction is made possible from knowledge on the regression model. In simple linear regression, one dependent variable is regressed with one independent variable. A straight line relating two quantities Y and X can be described by the Eq. (2).

$$y = \beta_0 + \beta_1 x_1 + e_1 \quad (2)$$

where $_1$ = 1,2, 3…n, y = dependent or response variable, x = Independent or explanatory variable (the engineering properties value of modified asphalt binders based on selected tests), βo = the intercept, the value of y when $x = 0$, β_1 = slope of the rate of change in y for any one unit change in x and e_1 = residual (y_1-y).

Thus, any particular formula might provide good or poor descriptions of how y relates to x. In a simple word, y is said to be known as a linear function of x where y can be used as regression model based on estimated prediction value of the dependent variables. In regression analysis, coefficient of determination (R^2) is useful because it shows the proportion of the variance for one variable which can be predicted from other variable. The predicted value will allow researchers to further determine of predictions value based on their model or graph. The value of R^2 is often represents in a percentage value of data that is the closest to the best fit line.

3. Results and Discussion

3.1 Rheological Asphalt Binder and Asphalt Mixture Performance Test Results

Table 5 shows the effect of nanopolyacrylate (NP) on the viscosity of the modified asphalt binder. As observed, the viscosity of NP modified asphalt binder increases gradually as the percentage of NP polymer in the asphalt binder increase. The viscosity of control asphalt binder tested at 135ºC is 0.4 Pa.s and highest value is at NP6 (0.65 Pa.s).

The RTFO aged asphalt binder test result shows that the NP modified asphalt binder can improve the workability (viscosity) and rutting resistance (G*/sinδ) of mixtures. Investigation of the effects of nanoparticles on the rheological characteristics of NP modified asphalt binder showed that the addition of this polymer improved the rheological properties of bituminous binders after short-term aging (STA) treatments [11]. There is an increase in G*/sin δ for the NP modified asphalt binder at NP2, NP4 and NP6 compared to control asphalt binder which is helpful in increasing the failure temperature, complex modulus, and elastic modulus values and in improving rutting resistance of the RTFO binder. Asphalt binder NP4 showed highest value of G*/sin δ indicating its higher viscous behaviour. The phase angle of the binders generally decreased with an increase in nano content and RTFO aging procedure [17].

For long term aging, the G*sin δ values for the nanopolymer modified asphalt binder decrease with the addition of NP. However, these values increase slightly at NP6. Similar finding from a study by Xiao indicated that nano particles can be beneficial in enhancing the rheological properties of bituminous binders after long-term aging treatments [17].

Table 5 - Rheological properties of unmodified and nanopolymer modified asphalt binder.

Asphalt Binder Sample	Viscosity, n tested 135ºC (Pa.s)	STA G*/sinδ tested at 64ºC (Pa)	G* sinδ tested at 25ºC (kPa)
Control	0.40	4849	2498
NP2	0.50	5223	2247
NP4	0.60	5343	2187
NP6	0.65	5220	2387

Table 6 summarizes the resilient modulus and permanent deformation results of the control and nanopolymer modified asphalt mixtures. All the HMA mix samples were prepared according to Marshall Mix design to obtain optimum asphalt binder content. The resilient modulus (M_R) and APA rut depth test were conducted on both control and NP modified asphalt mixtures. In general, the results show that the resilient modulus values increase for modified AC14-NP mixes compared to the AC14-Control mixes. The resilient modulus values for control mix is 542 MPa and 614 MPa, 716 MPa and 761 MPa respectively for AC14-NP2, AC14-NP4 and AC14-NP6 asphalt mixtures. It is observed that, the resilient modulus values increase as the NP content increase.

Table 6 - Resilient modulus and permanent deformation of asphalt mixtures.

Sample	M_R (0.1s) @ 40ºC (MPa)	Rut Depth @ 64ºC (mm)
AC14-Control	542	7.1
AC14-NP2	614	4.9
AC14-NP4	716	4.5
AC14-NP6	761	4.7

In addition, the APA rut depth of the nanopolymer modified asphalt mixtures also significantly decrease. The rut depth of control mix is 7.1mm and decrease to 4.9mm, 4.5mm and 4.7mm for AC14-NP2, AC14-NP4 and AC14-NP6 asphalt mixtures. The AC14-NP4 mix is the least susceptible to permanent deformation with rut depth of 4.5mm compared to AC14-NP2 and AC14-NP6 respectively.

3.2 Relationship Between Asphalt Mixture Performance Tests and G*/Sinδ of Asphalt Binder

Rutting is one of the most common pavement permanent deformation due to repetitive traffic load which accumulate small deformations of pavement material appearing as longitudinal depression in the wheel paths of the roadways [18]. APA rut depth test is conducted in order to evaluate rutting resistant of asphalt mixtures. According to Strategic Highway Research Program (SHRP), the minimum G*/sin δ value is 2.2 kPa. Higher G*/sin δ value indicates higher stiffness of the asphalt binder and is more resistant to rutting.

Fig. 5 shows the relationship between M_R asphalt mixtures at 40°C with the G*/Sinδ of the NP modified asphalt binders. There is a strong relationship, R^2=0.8 between the M_R value and the G*/Sinδ of nanopolymer modified asphalt binders. The trend observed that the M_R value increase as G*/Sinδ values increase which indicates better resistant to rutting. Results have proven that; it is possible to predict rutting from the asphalt binder stiffness properties.

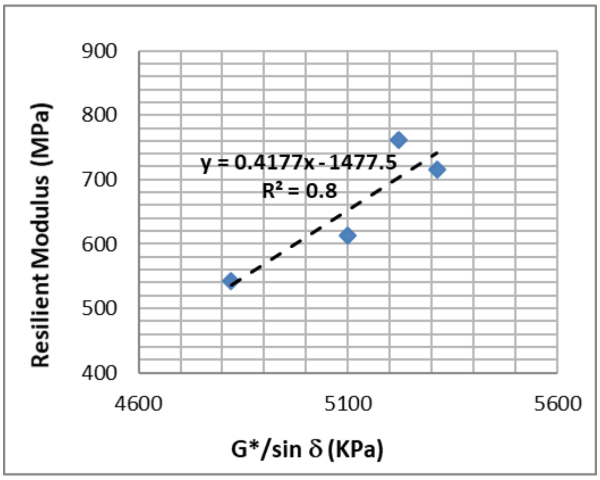

Fig. 5 - Relationship between M_R at 40°C and G*/sinδ.

In addition, Fig. 6 shows the relationship between the APA rut depth of the asphalt mixtures at 40°C and G*/Sinδ from the DSR test on the nanopolymer modified asphalt binder. Results showed a strong relationship of R^2=0.9 between the rut depth and G*/Sinδ of nanopolyacrylate modified asphalt binder. The trend also observed that the rut depth decrease as G*/Sinδ values increase. This indicates that the nanopolymer improves the viscoelastic properties of the asphalt binder and is resistant to permanent deformation. The trend indicates very strong relationships predictions of the rutting characteristic of mixtures based on the G*/Sinδ binder properties.

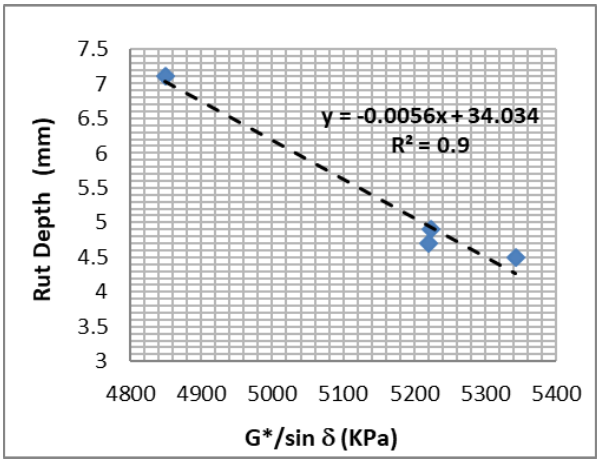

Fig. 6 - Relationship between rut depth and G*/sinδ.

3.3 Relationship Between Asphalt Mixture Performance Tests and Viscosity of Asphalt Binders

Viscosity properties was used to represent the pumpability of asphalt binder. Resilient modulus test and APA rut depth test are conducted to evaluate the effect of viscosity to the asphalt mixture performance. Fig. 7 shows the relationships between M_R of asphalt mixtures with the viscosity of the modified asphalt binder. There is a strong relationship, R^2 = 0.9 between the M_R and the viscosity of asphalt binders for nanopolyacrylate modified asphalt binder. The trend also observed that the M_R value at 40ºC value increase with the increase of viscosity. This trend is obvious as the increase in the viscosity leads to higher stiffness in the asphalt binders, and thus higher stiffness in the mixtures due to the addition of nano polymer and this finding agree with Tayfur et al. [18].

Additionally, Fig. 8 shows the relationship between the rut depth values from APA test with viscosity of the asphalt binder. It shows a strong relationship between the rut depth values and the viscosity of asphalt binder mixtures (R^2 = 0.8). The trend shows that the rut depth decreases as the viscosity increase which indicates that the stiffness of the binders due to higher viscosity improved the rutting resistance of the mixtures, hence better resistant to permanent deformation of pavement.

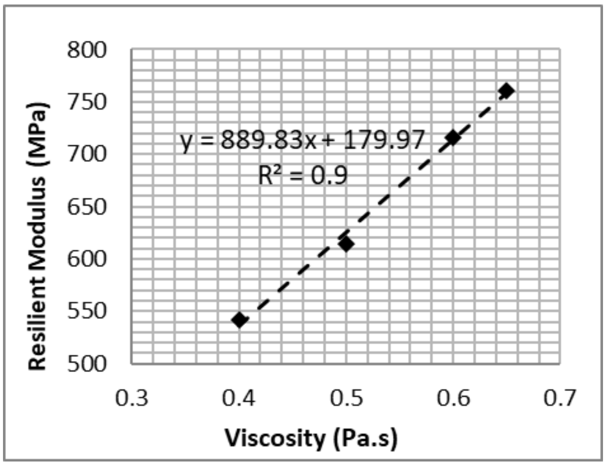

Fig. 7 - Relationship between resilient modulus and viscosity of nanopolyacrylate modified asphalt binder.

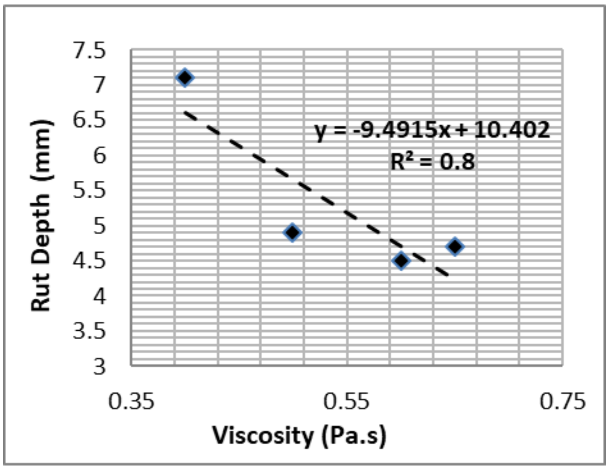

Fig. 8 - Relationship between rut depth and viscosity.

3.4 Predictive Model for Permanent Deformation

Table 7 shows the coefficient of determination values and equations of the selected variables. The regression analysis between the rheological properties of asphalt binder and engineering properties of nanopolyacrylate modified asphalt binder presents reasonable coefficient of determination values. It is found that there is strong relationship between the dependent and independent variables. The results indicate that resilient modulus and rut depth values significantly affects the properties of nanopolyacrylate modified asphalt binder mixtures.

The R^2 values obtained for the developed regression models between dependent and independent variables where the resilient modulus, at 40ºC, and rut depth showed a strong correlation with viscosity and G*/sin δ of the asphalt binder. Therefore, an effective prediction models can be measured based on the equation with higher R^2 values obtained from the regression analysis as depicted in Table 7. These findings are reasonable and the relationship between the predicted and measured permanent deformation values from regression analysis can be used to develop models effectively. It is revealed that the relationship of selected dependent and independent variables can affect the pavement performance [19].

Table 7 - Relationship and predictive equation model for permanent deformation.

Dependent Variables	**Independent variables**	**AC14-NP**	
Y	X	Equation	R^2
M_R (40°C)	Viscosity	y = 889.83x + 179.97	0.9
	Rutting, G*/ sin δ	Y = 0.4177x - 1477.5	0.8
Rut Depth	Viscosity	y = -9.4915x + 10.402	0.8
	Rutting, G*/ sin δ	y = -0.0056x + 34.034	0.9

4. Conclusion

Experimental results from this study shows that the use of nanopolyacrylate polymer as additives can improve the pavement performance through their ability to improve the properties of the asphalt binder and also asphalt mixture performance. The nanopolyacrylate asphalt mixtures demonstrate better resistance to permanent deformation (rutting) than the control mix. The study on rheological properties of asphalt binder is effective as an indicator to determine the asphalt mixture permanent deformation. The relationship of selected independent and dependent variables can affect the pavement performance. The dependent (resilient modulus at 40°C and APA rut depth) and independent (viscosity and G*/sin δ of the asphalt binder) variables significantly affects each other. Therefore, an effective prediction models can be developed according to predicted and measured permanent deformation values.

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