

APPLICATION OF MULTIVARIABLE REGRESSION MODELS FOR PREDICTION OF COMPOSITE NANOSILICA/POLYMER ASPHALT MIXTURE OBC

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ABSTRACT: In this research, the effects of nanosilica particles and polymer on conventional properties of hot mix asphalt have been investigated. The study also investigates the application of various regression models for the prediction of optimum binder content (OBC). The proposed models use values for stability and flow obtained from Marshall test results. The asphalt binder was modified using polyethylene and polypropylene polymers with varying percentages of nanosilica. The fundamental mechanical and physical properties of composite nanosilica/polymer modified binder and aggregate-binder mixtures were estimated through penetration, softening point, rolling thin film oven tests (RTFOT) aging and Marshall test. The results show that application of nanosilica improves the stability, reduces optimum binder content (OBC), increases stiffness as well as strength characteristic of the asphalt mixtures. The regression models analyzed was found to yields good predicted values with a high coefficient of determination R^2 and very low percentage errors of less than 5%.

Keywords: Nanosilica, polyethylene, polypropylene, nanocomposite, optimum binder content, multivariable models

1. INTRODUCTION

The reduction in asphalt mixtures strength and durability as a result of moisture effects or other factors decreases the performance of asphaltic concrete [1]. Previous research shows that several types of pavement failures like cracking, raveling, rutting and potholes may be caused due to moisture damages [2]. The cohesive and adhesive bonding between aggregates and bitumen binder in asphalt mixture determines the performance of hot mixed asphalts (HMA) pavements [3]. The main process of moisture damage for HMA are a loss of bitumen binder film stiffness as well as the failure of cohesion (strength) and adhesion bond between bitumen binder and aggregate in the mixture. Moisture infiltrates the asphalt mixture through surface infiltration (permeability), vapour diffusion and capillary rise [4].

Generally, two main methods are applied for improving adhesion and cohesion properties between aggregate and binder for increasing resistance of asphalt mixtures [5, 6], these are through the use of liquid anti-stripping agents and modifiers [7, 8]. Anti-stripping additives are chemical activators which improve moisture resistance through altering the structure of bitumen binder with an aim to increase the cohesion as well as adhesive bond existing between aggregates and binder by providing a strong coating of binder on

the aggregates [9]. The modifiers most commonly polymers are anticipated to increase the resistance of HMA through adhesion bond improvement between the aggregate surface and bitumen binder. Asphalt modifiers also improve adhesion bond between aggregates and binder through different mechanisms such as promoting binder spread all over the aggregate particles through binder surface tension reduction or enhancing at the same time the chemical behaviors of the bitumen and aggregate surface [10].

Despite the polymers have been immensely popular modifiers, several challenges such as poor stability and moisture damages [11-14] limits there applications and encouraged researchers to find new alternative methods towards improving the performance of polymer modified bitumen. Recent applications of nanomaterial's in conjunction with polymers enhances bituminous binder properties due to excellent properties of nano materials such as small particle size which makes them easily blend and more compatible with bitumen [15, 16]. The most common nanomaterials applied include nanosilica, carbon nanotubes, titanium dioxide, nanoclay, nanolime and nano zinc oxide [17].

Optimum binder content (OBC) is the most important parameter for conducting Marshall test. The Marshall testing procedure for the determination of OBC for asphalt mixtures involves lengthy steps which are time consuming.

However, after conducting the Marshall test only two parameters stability and flow are obtained directly while other parameters such as specific gravity of mixture, theoretical specific gravity, voids in mineral aggregate (VMA), stiffness, air voids (VA) as well as voids filled with bitumen (VFB) for estimating OBC are obtained through extra lengthy calculations. Therefore if a suitable relationship can be developed for prediction of OBC it will be very useful. The rest of the calculations can just be skipped and obtained an approximate OBC. Regression models can be a suitable mean to obtain relationships between marshal parameters for the prediction of OBC values for asphalt mixtures with reasonable accuracy [18].

From previous researches, it is clear that incorporating polymers as modifiers for bitumen enhances its performance characteristics. However, poor improvements were reported with regard to moisture damages. Based on that, there is need on improving the performance of polymer modified binders, in view of that, this research investigates the application of polyolefinic polymers namely polypropylene and polyethylene due to their availability as daily waste and nanosilica as an addition to polypropylene to improve the moisture resistance performance of polymer modified binders. Furthermore, the study investigates the application of various regression models for the prediction of optimum binder content (OBC) for asphalt mixtures.

2. MATERIALS AND METHODS

2.1 Materials

A bitumen binder grade 80/100 penetration was used for modification with polymers and nanosilica. The asphalt mixtures used for this research are prepared with composite nanosilica/polymer modified binders. Polymers linear low density polyethylene (LLDPE) in pellet form and polypropylene polymer in resin form were used in this research. The nanosilica used in this research for the preparation of nanocomposites has 99.8% SiO₂ content and average particle size of 10-25 nm.

A crushed granite coarse aggregate having a maximum nominal size of 19 mm, together with fine aggregate are selected for the preparation of asphalt mixture samples.

2.2 Experimental methods

2.2.1 Preparation of composite modified binders

The composite nanosilica/polymer modified binders were prepared by modifying the optimum

concentration of 6% polyethylene and polypropylene polymers with the addition of 1%, 2%, 3% and 4% nanosilica by weight of bitumen binder. Bitumen binder was first subjected to heating inside the oven at a high temperature of 150°C for 1 hour. For the composites mixing a propeller blade bench top Multimix high shear mixer was used at a high shear rate of 4000 rpm for 2 hours. During the mixing process, the temperature was maintained at a rate of 150 ± 5°C throughout.

2.2.2 Marshall Mix design

The Mix design used for the preparation of asphalt mixture samples were based on standard Marshall Mix design method (ASTM D 2967) by applying 75 blows on both cylindrical samples sides having dimension approximately 102 mm diameter with a mean height of 64 mm. After preparation, the samples are immediately placed on a flat and smooth surface at room temperature prior to flow and stability testing (ASTM D1559) as well as bulk specific gravity testing (ASTM D2726).

2.2.3 Conventional test

Conventional penetration tests at a temperature of 25°C (ASTM D5 13) as well as softening point temperature test (ASTM D36 12) for consistency were conducted on the modified binders. Results found from penetration together with softening point temperature tests were used to obtain penetration index (PI) [19] which is estimated using equation 1.

$$PI = \frac{1952 - 500 \log P_{25} - 20 T_{R\&B}}{50 \log P_{25} - T_{R\&B} - 120} \quad (1)$$

where $PI_{TR\&B}$ is the penetration index, P_{25} is the binder penetration tested at 250 °C, and $T_{R\&B}$ refers to samples softening point temperature.

3. RESULT AND DISCUSSION

3.1 Conventional tests

Influence of nanosilica concentration on conventional properties of polymer modified binder before and after aging is shown in Table. 1. It can be seen that while penetration values for the binders are decreasing, softening point temperature is increasing with the addition of nanosilica. However, after RTFOT aging of the binders, the penetration values decreases and the softening point temperatures increases, this indicates that the binders are resistant to oxidative aging due to the

formation of a strong chemical bond between bitumen and polymers. Penetration index (PI) is often estimated for quantification of the relative temperature susceptibility of the binders. The greater the PI value of a binder, the less the temperature susceptibility of the binder material [20]. It is obvious from Table 4 that PI values increase with the addition of more nanosilica content; hence this indicates binders susceptibility to temperature decreases and also the softening point increment observed after RTFOT aging of the binders, indicates that nanosilica reduces the effects of aging on the binders modified.

3.2 Marshall volumetric properties

Marshall stability, as well as flow values for the mixture sample tested, are summarized in Table 2, all the values presented are an average of at least three mixture samples tested in order to obtain higher reproducibility of the results. Marshall stability is one of the most important properties considered in the design of asphalt pavement wearing course. Stability is the measure of asphalt mixture ability to resist rutting under condition of traffic loading. It can be seen that Marshall stabilities for all the samples increases with nanosilica content, this indicates that addition of nanosilica provides an improvement in stiffness of the binders. However, the mixture having 3% nanosilica in both cases has higher stability values compared to other mixtures samples. This indicates that the mixture is harder and more resistant than other mixtures. Also, it is observed that nanosilica addition reduces the optimum binder content (OBC) for the mixtures. This can be attributed due to the surface nature of nanosilica

particles.

Marshall quotient (MQ) is estimated to ascertain the resistance of the asphalt mixtures to shear stresses and rutting deformation of the modified mixtures. As it is can be seen the MQ increases with nanosilica content and 3% nanosilica mixtures has the highest values of MQ, this shows that nanosilica stiffen the mixtures and inhibit high flow which results in greater MQ values.

Flow is an ability of asphalt mixture to adjust to gradual settlements and movements without cracking. Flow is used to determine the reversible behavior of the wearing course under traffic loads. The flow values of the composite modified mixtures are measured and also presented in Table 2. Form the flow values it can be seen that lower flow values were obtained for the composite modified mixtures in comparison with the control mixture, thus indicating that modified binder helps in enhancing the strength of bituminous mixture due to formation of strong adhesion bond between the binder and aggregates.

Air voids (AV) is the amount of air presence in compacted asphalt mixtures. AV shows the percentage volume between the aggregate particles coated by asphalt binder in compacted bituminous mixture. It can be seen that the composite mixtures have sufficient flow values.

3.3 Prediction of OBC using stability and flow values

Statistical multivariable regression analysis is a modeling technique used for analyzing multiple variables in order to find the correlation between a dependent and one or sets of independent variables.

Table 1 Conventional properties of modified binders before and after RTFOT aging

Properties	Modified binders							
	PE1%	PE2%	PE3%	PE4%	PP1%	PP2%	PP3%	PP4%
	NS	NS	NS	NS	NS	NS	NS	NS
Unaged								
Penetration (100g, 0.1 mm, 5 s, 25°C)	64	59	55	51	59	56	48	50
Softening point (°C)	58.4	60.8	64.3	65.8	57.3	64.5	62.9	63.4
Penetration index (PI)	1.35	1.62	1.91	2.17	1.63	2.21	2.42	2.11
RTFOT Aged								
Mass loss (%)	0.15	0.11	0.06	0.1	0.09	0.05	0.07	0.13
Penetration (100g, 0.1 mm, 5 s, 25°C)	48	44	41	37	44	33	31	32
Softening point (°C)	64.2	66.4	69.4	71.6	66.6	73.9	75.8	73.5
Softening point increment (°C)	5.8	5.6	5.1	5.8	9.3	9.4	12.9	10.1
Penetration index (PI)	1.72	1.89	2.22	2.33	1.92	2.43	2.55	2.28

Table 2 Marshal volumetric properties

Mixture type	Stability (KN)	Flow (mm)	VA (%)	VMA (%)	VFB (%)	OBC (%)	MQ KN/mm
PE Composite							
Control PE	9.100	2.96	2.9	15.2	81	5.44	2.38
PE1%NS	11.00	2.92	3.8	15.4	75	5.09	2.39
PE2%NS	13.00	2.83	3.4	15.1	75	4.99	2.37
PE3%NS	14.71	2.77	3.2	14.2	71	4.80	2.38
PE4%NS	13.10	2.81	3.5	14.0	77	4.81	2.40
PP Composite							
Control PP	11.01	2.97	3.4	14.2	77	5.19	2.35
PP1%NS	12.97	2.90	3.6	15.1	75	4.90	2.34
PP2%NS	14.40	2.81	3.2	14.4	77	4.94	2.37
PP3%NS	16.80	2.71	3.5	13.6	78	4.68	2.38
PP4%NS	13.02	2.84	3.1	14.2	78	4.88	2.63

Table 3 ANOVA result for multivariable regression models

Models	R ²	F-value	Standard error (%)
PE Mixture			
Linear	0.88	7.64	0.138
Exponential	0.89	8.34	0.026
Power	0.92	11.19	0.023
PP Mixture			
Linear	0.94	18.65	0.059
Exponential	0.94	19.52	0.012
Power	0.95	19.61	0.012

In this study, multivariable regression models are developed for prediction of OBC directly from average stability (S) and flow (F) values from Marshal test. Linear, exponential and double power multivariable regression models have been developed.

For yielding good prediction accuracy, a multi variable regressions were used which are highly useful for fitting experimental data with more than one independent variable [21]. The LOGEST function from Microsoft Excel has been applied to estimate best fits straight lines for the data using the method of least squares, the general form for the exponential equations used is shown in equation 6.

$$Y = ab^S * c^F \quad (6)$$

Where Y is optimum binder content (OBC), S is stability, F is flow, a, b and c are constants.

Also for multivariable power regression model, the experimental stability and flow values are used. Initially, the natural logarithms for the data have been analyzed, after which 'LINEST' function in Microsoft Excel is applied to estimate the best fits straight lines for the data using the method of least squares [22].

$$Y = aS^bF^c \quad (7)$$

Where Y is the optimum binder content (OBC), S is the stability, F is the flow and a, b, c are the constant dependent on the material.

The final multivariable regression models equations for both PE and PP mixtures obtained from linear, exponential and double power models are presented in equation 8 through 13.

PE Composite Mixtures

Linear: $OBC = -0.119S + 0.134F + 6.038$ (8)

Exponent: $OBC = 5.63 * S^{0.977} * 1.054^F$ (9)

Power: $OBC = 8.387 * S^{-0.271} * F^{0.139}$ (10)

PP Composite Mixtures

Linear: $OBC = 0.041S + 2.418F - 2.5549$ (11)

Exponent: $OBC = 1.128 * 1.008^S * 1.616^F$ (12)

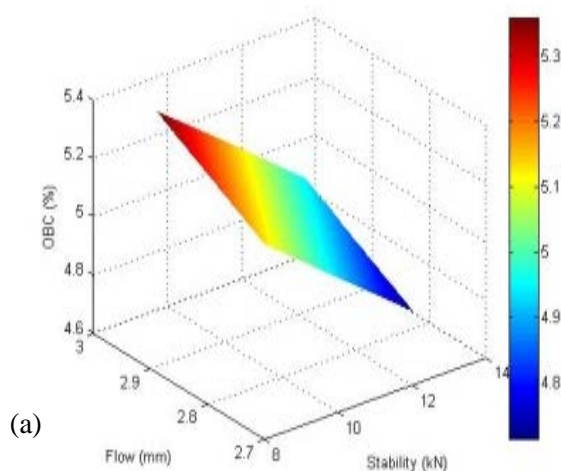
Power: $OBC = 0.813 * S^{0.125} * F^{1.404}$ (13)

where OBC is optimum binder content, S is stability values and F is flow obtained from Marshall test.

The ANOVA statistical analyses for the developed relationships of the mixtures are presented in Table 3. For PE composite modified mixtures, it can be seen that high coefficient of determination R^2 values in the range 0.89 – 0.91 have been found for all the regression models developed and double power regression model has the highest R^2 value of 0.92, while in the case of linear and exponential regression models, approximately the same R^2 of 0.89 have been obtained. However, highest statistical F-value of 11.19 has been obtained in power model while linear and exponential model shows the least F-values of less than 10. The results indicate that the power regression model has been found to lead with higher F-value and coefficient of determination R^2 value among all the developed regression models.

For PP composite modified mixture, it was also observed that high R^2 values have been found for all the regression models developed and also double power regression model has the highest R^2 of 0.95. Similarly, in the case of linear and exponential regression models, the R^2 value of 0.94 has been obtained. However, highest statistical F-value of 19.61 has been obtained with linear regression model while linear and a power regression model shows lower F-values. The results indicate that all regression models developed are found to be adequate for the analysis.

The developed regression analysis was plotted on 3D surface plots using Matlab program and presented in Fig. 2 and Fig. 3 for PE mixture and PP mixture respectively. In general it can be seen from both figures that the stability values for the mixtures increases with corresponding increase in OBC values up to around 5.5% OBC.



between predicted and actual values obtained from

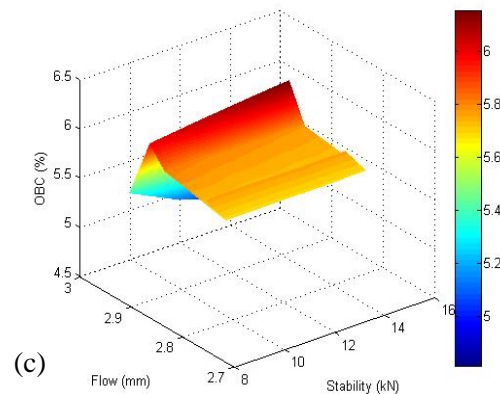
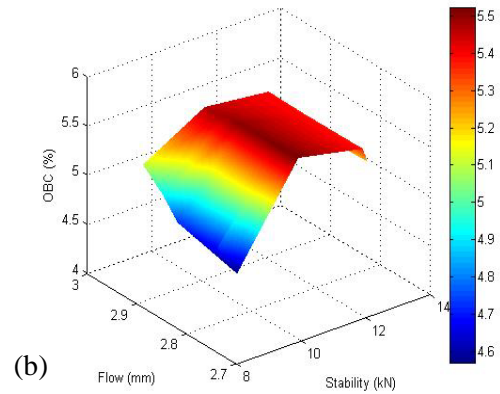


Fig. 2 Response surface plot (3D) for optimum binder content (OBC), stability and flow for PP mixtures (a) Linear (b) Exponential (c) Double power

From Figure 3a for the linear model, it can be seen that OBC increases with a decrease in stability and increase in flow, while for PP mixture presented in Figure 3a, OBC increases with increase in both stability and flow, however, the flow has more effect on the OBC. It can also be seen from Figure 2b that lower OBC were achieved with an increase in both stability and flow values while from Figure 2c, Figure 3b and Figure 3c OBC decreases with increase in stability and decrease in flow.

In addition, the degree of determination and fitness of the developed regression models were verified graphically by plotting the predicted OBC against the actual OBC as shown in Figure 4a and Fig. 4b for PE mixture and PP mixture respectively. In general, it can be seen that all the observed points are relatively close to line of equality. This indicates that the models have highest degree of fitness. Furthermore, it was observed that the double power models has the highest degree of fitness compared to linear and exponential models as verified by its highest R^2 values, and therefore will provide the highest prediction accuracy.

In order to quantify the level of accuracy of each predicted model, the percentage error Marshall Test, has been calculated and the %

errors versus actual OBC for the asphalt mixtures are estimated and presented in Fig. 5 It can be observed that all the regression models developed have very low percentage error values of less than 5%, which indicates higher accuracy of the predicted regression models developed. It can also be noted that PP models shows the least values of standard error compare to PE models, this indicates that the models have higher precision accuracy. In addition, it is evident that double power model will provides much accurate results for both mixtures due to minimum percentage error values obtained with the model.

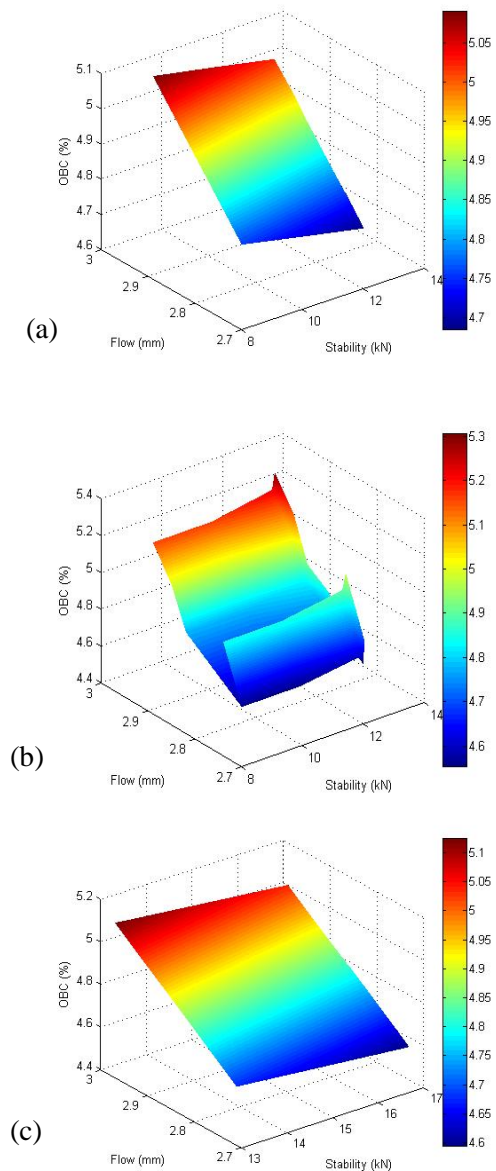


Fig. 3. Response surface plot (3D) for optimum binder content (OBC), stability and flow for PP mixtures (a) Linear (b) Exponential (c) Double power

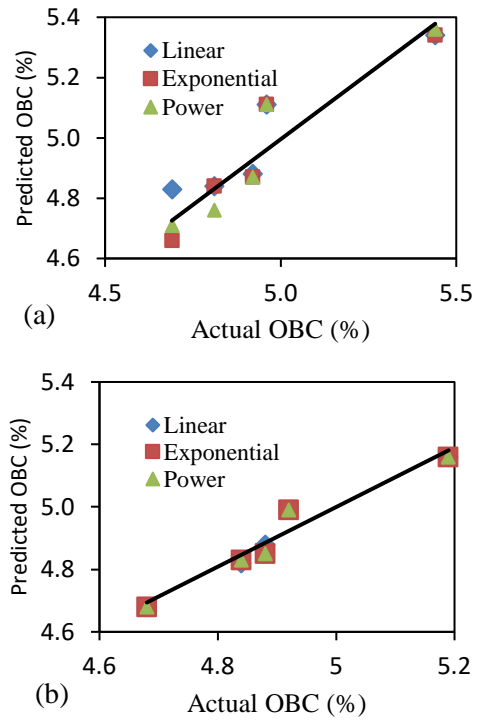


Fig. 4 Extent of agreement between predicted and actual results (a) PE mixture (b) PP mixtures

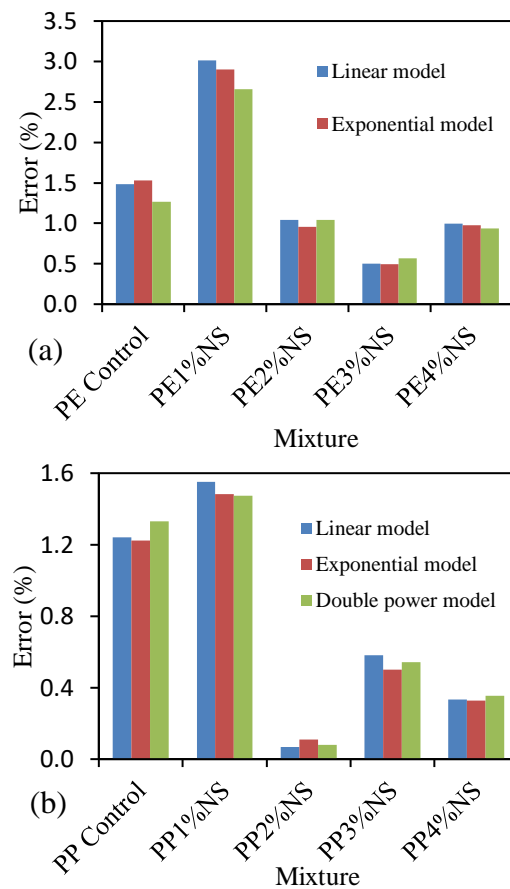


Fig. 5 Percent error for all the regression models (a) PE mixture (b) PP mixture

4. CONCLUSION

In this investigation, the effects of nanosilica and polymer on hot mix asphalt were evaluated and also the reliability of regression models for prediction of optimum binder content has been investigated. Based on the outcomes of the investigation, the following conclusions have been drawn:

1. Conventional penetration and softening point temperature test results have shown that nanosilica content enhances the stiffness of the polymer modified binders. Also, the penetration index for the composite modified binders increases with the addition of nanosilica content. This indicates that nanosilica promotes the decrease in the brittleness and temperature sensitivity reduction of the modified binders.
2. Based on Marshall Stability test, the Marshall stability values increases and optimum binder content decreases with increase in nanosilica content. This indicates an enhancement in binder's properties.
3. The experimental OBC and predicted OBC from models were found to be in good agreement with each other.
4. The predictive regression models analysis shows that in all the models the percentage error has not exceeded 5%, and lowest error of 1.2% have been achieved with double power model for both mixtures this concludes that double power model offers high level of accuracy for the prediction of OBC.

5. REFERENCES

- [1] Baldi-Sevilla A., Aguiar-Moya J. P., Vargas-Nordbeck A. and Loria-Salazar A., "Effect of aggregate-bitumen compatibility on moisture susceptibility of asphalt mixtures," *Road Materials and Pavement Design*, pp. 1-11, 2017.
- [2] Kakar M. R, Hamzah M. O. and Valentin J., "A review on moisture damages of hot and warm mix asphalt and related investigations," *Journal of Cleaner Production*, vol. 99, pp. 39-58, 2015.
- [3] Aboufoul M. and Garcia A, "Factors affecting hydraulic conductivity of asphalt mixture," *Materials and Structures*, vol. 50, p. 116, 2017.
- [4] Caro S., Masad E., Bhasin A. and Little D. N., "Moisture susceptibility of asphalt mixtures, Part I: mechanisms," *International Journal of Pavement Engineering*, vol. 9, pp. 81-98, 2008.
- [5] Kok B. V. and Yilmaz M., "The effects of using lime and styrene-butadiene-styrene on moisture sensitivity resistance of hot mix asphalt," *Construction and building materials*, vol. 23, pp. 1999-2006, 2009.
- [6] Behiry E. M., "Laboratory evaluation of resistance to moisture damage in asphalt mixtures," *Ain Shams Engineering Journal*, vol. 4, pp. 351-363, 2013.
- [7] Nejad F. M., Azarhoosh M, Hamed G. H. and Azarhoosh M., "Influence of using nonmaterial to reduce the moisture susceptibility of hot mix asphalt," *Construction and Building Materials*, vol. 31, pp. 384-388, 2012.
- [8] Arabani M. and Hamed G. H., "Using the surface free energy method to evaluate the effects of polymeric aggregate treatment on moisture damage in hot-mix asphalt," *Journal of Materials in Civil Engineering*, vol. 23, pp. 802-811, 2010.
- [9] Hesami S., Roshani H., Hamed G. H. and Azarhoosh A., "Evaluate the mechanism of the effect of hydrated lime on moisture damage of warm mix asphalt," *Construction and Building Materials*, vol. 47, pp. 935-941, 2013.
- [10] Abuawad I. M., Al-Qadi I. L. and Trepanier J. S., "Mitigation of moisture damage in asphalt concrete: Testing techniques and additives/modifiers effectiveness," *Construction and Building Materials*, vol. 84, pp. 437-443, 2015.
- [11] Ouyang C., Wang S., Zhang Y., "Improving the aging resistance of styrene-butadiene-styrene tri-block copolymer modified asphalt by addition of antioxidants," *Polymer degradation and stability*, vol. 91, pp. 795-804, 2006.
- [12] Zhang Y., Zhao S., Xie L., and Yao S., "Improving the aging resistance of styrene-butadiene-styrene tri-block copolymer and application in polymer-modified asphalt," *Journal of applied polymer science*, vol. 116, pp. 754-761, 2010.
- [13] Zhu J., Birgisson B., and Kringos N., "Polymer modification of bitumen: Advances and challenges," *European Polymer Journal*, vol. 54, pp. 18-38, 2014.
- [14] Xiao Y., Van De Ven M., Molenaar A., Su Z. and Zandvoort F., "Characteristics of two-component epoxy modified bitumen," *Materials and structures*, vol. 44, pp. 611-622, 2011.
- [15] Bala N., Kamaruddin I., Napih M., and Danlami N., "Rheological and rutting evaluation of composite nanosilica/polyethylene modified bitumen," *Proceedings of the 7th International Conference on Key Engineering Materials (ICKEM 2017) held between 11th to 13th*

- March 2017, Penang Malaysia. IOP Conference Series: Materials Science and Engineering vol. 201, 2017.
- [16] Arabani M., Tahami S. A. and Hamed G. H., "Performance evaluation of dry process crumb rubber-modified asphalt mixtures with nanomaterial," *Road Materials and Pavement Design*, pp. 1-18, 2017.
- [17] Fang C., Yu R., Liu S. and Li Y., "Nanomaterials applied in asphalt modification: a review," *Journal of Materials Science & Technology*, vol. 29, pp. 589-594, 2013.
- [18] Handle F., Füssl J., Neudl S., Grossegger D., Eberhardsteiner L., Hofko B., "The bitumen microstructure: a fluorescent approach," *Materials and Structures*, vol. 49, pp. 167-180, 2016.
- [19] Read J. and Whiteoak D., *The shell bitumen handbook*: Thomas Telford, 2003.
- [20] Samsudin M. S., Arshad A. K., Ahmad J., and Masri K. A., "Microstructure of nanosilica modified binder by atomic force microscopy," *Jurnal Teknologi*, vol. 78, pp. 33-44, 2016.
- [21] Underwood B., Baek C. and Kim Y., "Simplified viscoelastic continuum damage model as platform for asphalt concrete fatigue analysis," *Transportation Research Record: Journal of the Transportation Research Board*, pp. 36-45, 2012.
- [22] Papworth F., Corbett D. and Barnes R., "In-situ concrete strength assessment based on Ultrasonic Pulse Velocity (UPV), rebound, cores and the SonReb method," in *Concrete Repair, Rehabilitation and Retrofitting IV: Proceedings of the 4th International Conference on Concrete Repair, Rehabilitation and Retrofitting (ICRRR-4)*, 5-7 October 2015, Leipzig, Germany, 2015, p.257.

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