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APPLICATION OF RESPONSE SURFACE METHODOLOGY FOR MIX DESIGN OPTIMIZATION OF NANOCOMPOSITE MODIFIED ASPHALT MIXTURES

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ABSTRACT: Recently the application of polymer nanocomposite has been increasingly recognized as an alternative to polymer modified binders for durable asphalt mixtures. In this study, the optimization of nanosilica and binder content for nanocomposite modified asphalt mixtures has been examined to obtain optimum quantities for higher volumetric properties. Response Surface Methodology (RSM) based on face centered central composite design (FCCCD) was utilized. Interaction effects of independent variables nanosilica and binder content on nanocomposite volumetric properties namely stability, flow, void in total mix (VTM) and voids in mineral aggregate (VMA) were evaluated. The result indicates that the individual effects of nanosilica and binder content are both important; however, the asphalt binder content had more significance on the volumetric properties. Higher Stability was achieved with the mixtures prepared with 2% nanosilica and binder content range of 5 - 6%. Also, the mean error obtained in all the responses from optimization results are all less than 5%, this indicates that predicted values are in agreement with experimental results. Furthermore, it was concluded that asphalt mixture design with high performance properties, optimization using RSM is a very effective approach.

Keywords: Response surface methodology, Nanosilica, Nanocomposite, Marshal mix, Polypropylene

1. INTRODUCTION

Asphaltic pavements during their service life are generally subjected to different external factors which include mechanical loading induced through axle loads and thermal loading induced due to changes in adverse environmental conditions [1]. The applied loads, together with harsh environmental conditions cause a deterioration of pavement which reduces the expected service life of the pavement [2]-[5]. For reducing the effects of such pavement distresses, there in need to use binder materials with improved properties. Based on this, bitumen modification using various materials has been used as suitable way as it transforms the chemical properties of the binder as well as its rheological and physicochemical properties [6]. These changes in binder properties resulted in higher performance and service life extension of asphalt pavements [7].

Previously Polymer materials such as thermoplastic elastomers and plastomers are widely used to improve bitumen properties after yielding some improvements on the modified asphalt binders characteristics [8]-[9]. Common polymers used are polypropylene, polyethylenes, acrylonitrile butadiene styrene, styrene-butadiene-styrene, polyvinyl chloride and polyethylene terephthalate [10]. Recently, nanomaterial additives have extensively gained a great attention by pavement researchers for the preparation of durable asphaltic mixtures with high performance due to their excellent beneficial properties such as large surface area, excellent dispersion ability, strong absorption, excellent stability as well as high chemical purity [1] and [12].

Silica is one of the abundant compounds deposited worldwide which is largely used by industries to produce colloidal silica, silica gels and fumed silica. Nanosilica is used by industries producing medicines, in cement and concrete mixtures and in industries to reinforce elastomers as rheological solutes [13].

Application of statistical modeling and optimization techniques is useful as it is excellent in terms of its ability to deal with various constraints and objectives and in describing the interactions among dependent variables that affect a particular response [14]-[15]. Factorial design of experiments (DOE) through the application of methods such as response surface methodology (RSM) is used to consider several factors simultaneously at different levels, and suggest a suitable predictive model for the relationship between the various factors [16]. RSM methods have been effectively applied in various areas such as concrete technology, material as well as mechanical engineering areas, biomass and clay sciences. Recently, the application of RSM in asphalt mixtures designs is increasingly recognized. [17].

It is clear that incorporating polymers as modifiers for bitumen enhances its performance characteristics. However, polymer modified bitumen is subjected to phase separation caused by poor compatibility of polymers with bitumen, these consequently affects the performance of polymer modified binders. Based on that, there is need on improving the performance of polymer modified binders, in response to the above, this research investigates the application of polyolefenic polymer namely polypropylene (thermoplastic plastomer) due to its availabity as daily waste and addition of nanosilica at lower contents to mitigate the reduction in performance properties of polymer modified binders. The current paper applies response surface methodology (RSM) to optimize and develop models for predicting the volumetric properties of composite nanosilica/polypropylene modified asphalt mixtures.

2. MATERIALS AND METHODS

2.1 Materials

The aggregate used in this study for the preparation of asphalt mixture samples is crushed granite coarse aggregate having a maximum nominal size of 19 mm, together with fine aggregate. For obtaining appropriate aggregates interlocking, a dense gradation was selected.

Bitumen binder grade 80/100 penetration was used for the preparation of modified binders blend in this study. Polypropylene polymer in resin form was used and blended with both bitumen and nanosilica to form nanosilica/polypropylene nanocomposite blends.

The nanosilica used in this research has 99.8% SiO_2 content and average particle size of 10-25 nm.

2.2 Mix design and specimen preparation

2.2.1 Preparation of modified bitumen

The composite nanosilica/polypropylene modified binders were prepared by adding 5% polypropylene polymer together with 1%, 2% and 3% nanosilica by weight of bitumen binder. 80/100 pen grade binder was first heated in an oven at a temperature of 150 °C to achieve desirable viscosity for mixing, polypropylene was then added to the required amount of base binder prior to the composite modification at a high shearing rate of 4000 rpm, the mixing continued until polypropylene dissolves completely on the base binder. Different percentages of nanosilica were added gradually and sheared at a high shearing rate of 4000 rpm for 2 hours. Composites mixing was done using a propeller blade laboratory bench top multi mix high shear mixer. During the mixing process, the temperature was maintained at a rate of 150 ± 5 °C throughout.

2.2.2 Mixture design

The Mix design used for the preparation of asphalt mixture samples were based on standard Marshall Mix design method by applying 75 blows on both cylindrical samples sides having dimension approximately 101 mm diameter and height of 64 mm. Marshal stability and flow are obtained according to ASTM D1559 while the bulk specific gravity of compacted mixture was obtained according to standard specification ASTM D2726. Volumetric characteristics of compacted asphalt mixtures were estimated on the basis of bulk specific gravity of asphalt mixture, and consist of Void in Mineral Aggregate (VMA) and Void in Total Mix (VTM).

2.3 Method of analysis

The Central composite design (CCD) is the most common applied design method used with RSM for statistical evaluation of the relationship between independent variables and responses. In this study, the influence of two independent variables binder content (A) from 4% to 6% and nanosilica (B) in the range of 1% to 3% were studied at three levels based on face-centered central composite design (FCCCD). FCCCD is a distinct case of CCD in which α is equal to 1.0, in FCCCD the α forces the axial points to locate on the surface of the cubic rather than on the sphere space as in CCD design which makes FCCCD design a three-level CCD. For this study, Design Expert software version 9.0.2.0 was used to produce statistical analysis and experimental designs. Related literature [18]-[19], as well as preliminary studies, were used to select the independent variables as well as their respective experimental ranges.

An optimal predictor quadratic model, shown in Equation 1, was used to obtain optimal conditions for the responses (binder content and nanosilica).

$$y = \beta_o + \sum_{j=1}^{k} \beta_{jj} x_j^2 + \sum_{i\angle 1}^{k} \beta_{ij} x_i x_j + e \qquad (1)$$

where y is the predicted response; β_0 is the experiment central point fixed response value, β_j and β_{jj} are linear and quadratic coefficients, βij is the interaction coefficients of the factors x_i and x_j, k is the number of factors while e is the model random error.

3. RESULT AND DISCUSSION

3.1 Statistical analysis

A statistical analysis has been done to have a good understating of the model's performance in terms of volumetric properties. After regression analysis has been applied, a fitted quadratic model was developed for prediction of all the responses. Quadratic models were choose based on the highest order polynomials in which the additional terms were significant and are not aliased by the software. The final regression models equations in terms of coded factors after reduction to exclude all the insignificant terms in the models are presented in equations 2 to 5. The positive and negative signs before the terms in the equations show the synergistic and antagonistic effects of the individual variables on the responses.

$$VIM = 51 - 16.39A - 1.6B + 1.4A^2 + 0.3B^2 \quad (2)$$

$$VMA = 45.36 - 12.33A - 0.27B - 0.02AB + 1.2A^2 - 0.05B^2 \quad (3)$$

$$Stability = -32.42 + 18.3A + 0.39B - 0.58AB - 1.8A^2 + 0.63B^2 \quad (4)$$

$$Flow = 2.74 - 0.05A - 0.25B + 0.053AB + 0.02A^2 - 0.01B^2 \quad (5)$$

Table 1 presents the statistical analysis summary for different responses analyzed in this investigation. The fitness of the model developed was evaluated based on determination coefficient R^2 and R^2_{adj} values calculated. Determination coefficients R^2 were obtained as 0.9953, 0.9376, 0.9102 and 0.8969 for VTM, VMA, stability and flow respectively. R^2_{adj} for the models were obtained as 0.9920, 0.8930, 0.8460 and 0.8233 for VTM, VMA, stability and flow respectively. For a well-fitted model, the minimum determination coefficient should be greater than 0.80. From Table 1 it can be seen that a high R^2 and R^2_{adj} values of approximately 1.0000 was obtained for all the models, this indicates a reasonable as well as a desirable agreement between actual and predicted model values [20].

In addition, adequate precision (AP) is used to check the adequacy of the developed models, AP measures the model signal to noise ratio through comparisons of the predicted values at the design points to the mean prediction error. For a good model, the AP ratio should be greater than 4 [18], in this study AP ratio of 49.63, 13.69, 10.77 and 11.23 were obtained for responses VTM, VMA, stability, and flow, respectively. Higher AP ratios greater than 4 obtained with the models indicates a sufficient signal and models adequacy to navigate the design space defined by FCCCD to provide parameters for optimum mix design.

Analysis of the variance (ANOVA) shown in Table 1, indicate that the models' F-values of 298.31, 21.03, 14.19 and 12.18 and low p-values of <0.05 are significant for 95% confidence level, also the models and terms are statistically significant for responses VTM, VMA, stability and flow respectively. The p-values of <0.05 for the models also indicate that only a 0.01% chance exists that a model F-value of this level can occur due to noise. ANOVA analysis confirms that both nanosilica and binder content have significant effects on volumetric properties based on the *t-test* at a 5% significance level (P< 0.05) as shown in Table 1.

The lack of fit (LOF) F-test was also analyzed for the evaluation of the model's adequacy. LOF is used to show data variation around the fitted model, LOF errors are generally associated with the selection of the model, for correctly selected models LOF error becomes small and insignificant [21]. For the volumetric responses investigated in this research, the LOF errors obtained as presented in Table 1, show that except the VTM which was found significant, all other responses including VMA, stability, and flow have low and insignificant LOF error. This further confirms that the models selected are highly significant and can be used to describe the correlations that exist between the factors and the responses. Similar findings were reported by [22] and [23].

For an understanding of the developed model's satisfactoriness, plots of predicted versus actual values are analyzed as shown in Fig. 1. As seen all the points for the responses were spread relatively very close to the line of equality, the distribution of the points observed indicates a satisfactory fitting precision of the models and the experimental results are in agreement with each other. In addition, the result verifies that the predicted models developed can adequately navigate the design space defined by FCCCD.

3.2 Volumetric Responses Analysis

Significant interactions among the variables are analyzed and presented using 3D response surface and 2D contour plots as shown in Fig. 2 to 5. These plots provide sufficient information on the interaction effects among mix design parameters on volumetric responses.

The response surface plots for VTM are presented in Fig. 2. It is evident that the individual effects of binder content and nanosilica were both significant. Furthermore, it can be realized that the VTM value decreased by 3.53% from 6.49% to 2.96%, this indicates that the binder content has more influence on the VTM when increasing the binder from 4% to 6%. It can also be seen that the VTM value rises at lower and higher nanosilica content and decreases slightly at intermediate concentrations and adding 2% nanosilica results in lowest VTM, the higher VTM could be attributed to the huge surface area of nanosilica and elastic deformation of polypropylene under compaction effort which can ultimately result in higher VTM within the mixture.

Table 1 Analysis of ANOVA for responses

Source	F-value	P-value	R^2	Adj R^2	AP
VTM					
Model	298.31	<0.0001	0.9953	0.9920	49.628
A	1146.7	<0.0001			
B	55.21	0.0001			
AB	1.22	0.305			
A^2	208.77	<0.0001			
B^2	7.55	0.0286			
Lack of fit	22.59	0.0057			
VMA					
Model	21.03	0.0004	0.9376	0.8930	13.69
A	9.85E-03	0.9237			
B	26.13	0.0014			
AB	0.02	0.8912			
A^2	69.46	<0.0001			
B^2	0.1	0.7588			
Lack of fit	3.07	0.1534			
Stability					
Model	14.19	0.0015	0.9102	0.8460	10.77
A	11.45	0.0117			
B	0.04	0.8479			
AB	7.86	0.0264			
A^2	51.55	0.0002			
B^2	6.47	0.0384			
Lack of fit	5.89	0.0599			
Flow					
Model	12.18	0.0024	0.8969	0.8233	11.23
A	56.71	0.0001			
B	2.24	0.1784			
AB	1.76	0.2263			
A^2	0.15	0.7075			
B^2	0.12	0.741			
Lack of fit	0.29	0.8285			

Fig. 3 presents the variation of VMA versus binder contents and nanosilica. It can be depicted that there are less changes in VMA values with increase in nanosilica as compared to binder content and mixture prepared with 5% binder content shows a considerable lower VMA values.

Moreover, the greatest VMA values are achieved in the mixtures prepared with highest binder content and minimum nanosilica content. This behavior can be attributed due to the properties of nanosilica such as the huge surface area of interaction and strong surface free energy which makes it fill voids in the mixture at lower concentrations.

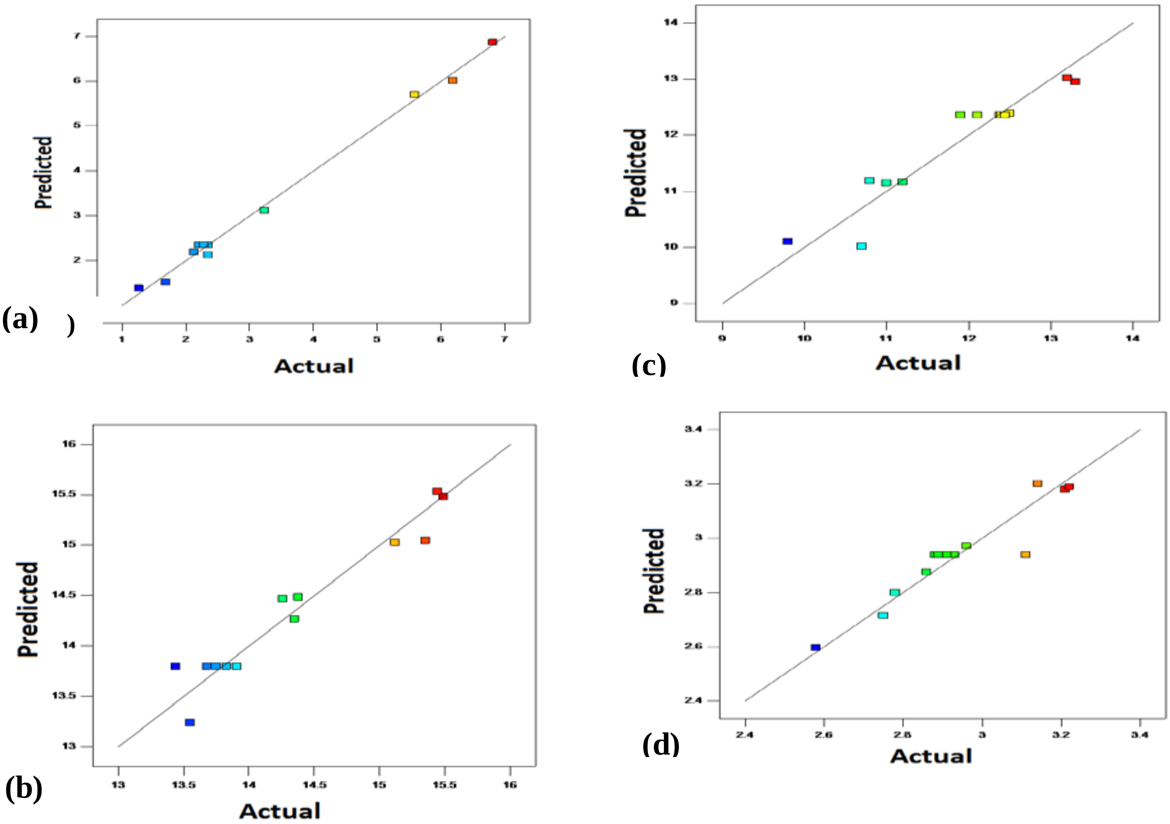

Fig.1 Predicted versus actual plots (a) VTM (b) VMA (c) Stability (d) Flow

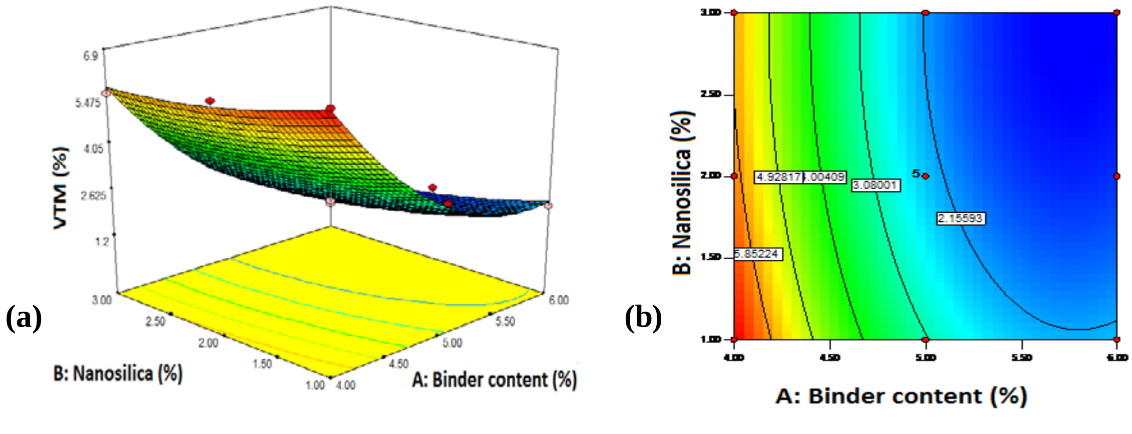

Fig.2 Effect of binder content and nanosilica content on VTM (a) 3D plot and (b) 2D contour plot

Fig. 4 shows the variation of stability at different concentrations of nanosilica and binder content. The result indicates that increasing nanosilica from 1 – 2% shows higher influence on the stability of the asphalt mixtures while the increase in binder content only shows a negligible effect on the mixtures. This behaviour can be attributed to the high energy and surface activity of nanosilica particles. In Fig. 4b elliptical contour lines were obtained meaning there is a perfect interaction between variables [10] and [24], the elliptical shape contours also show that there is an area of optimum performance within 1.5 – 2.5% nanosilica and 4 – 5.5% binder content.

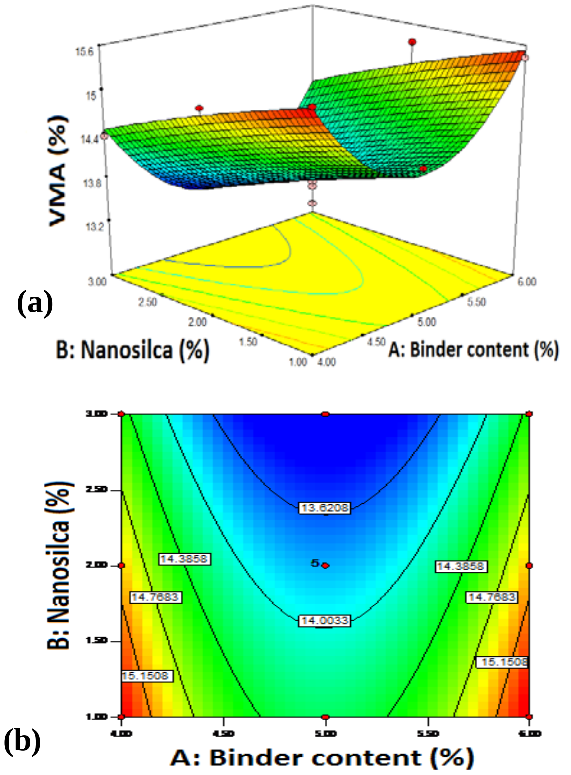

Fig.3 Effect of binder content and nanosilica content on VMA (a) 3D plot and (b) 2D contour plot

Fig. 5 depicts the effects of variation in both nanosilica and binder content on the asphalt mixtures. It can be realized that almost a linear relationship was obtained between nanosilica and binder content. The result illustrates that increase in the amount of binder content has higher influence on the flow; additionally the increase in nanosilica content resulted in reduction of flow values in the asphalt mixtures, this indicates that sufficient flow value can be obtained at binder content of 4.5 – 5.5% and nanosilica ranges of 1.5 – 2.5%.

3.3 Optimization of design variables

In this study, a numerical optimization method was applied to optimize design variables and evaluates the accuracy of the developed models. To minimize materials nanosilica was set in minimum to reduce the quantity, binder content was set in range (4-6%), stability was set to maximum and flow was set minimum to achieve high performance and VTM was set in range (3-5%) to obtain a durable dense mixture.

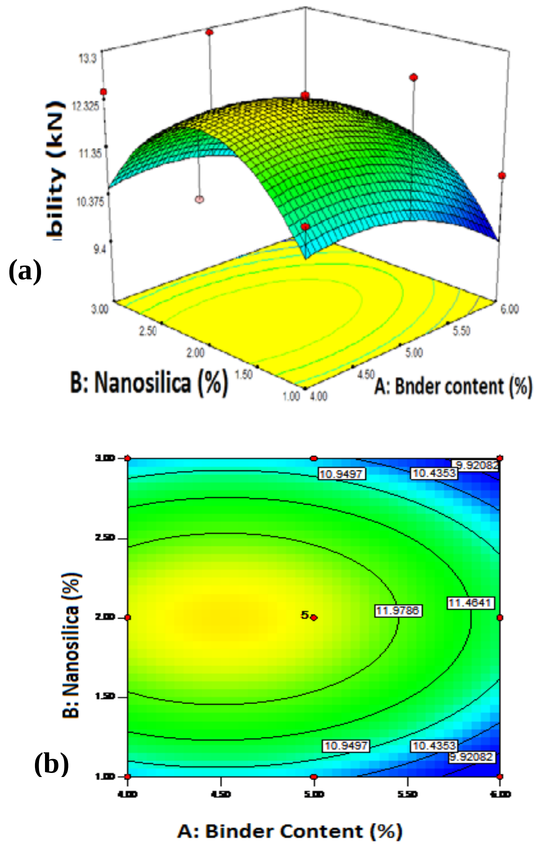

Fig.4 Effect of binder content and nano silica content on stability (a) 3D plot and (b) 2D contour plot

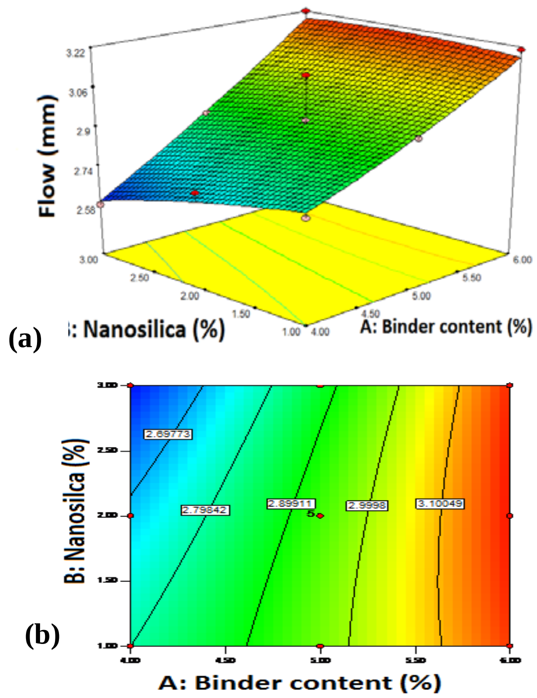

Fig.5 Effect of binder content and nano silica content on Flow (a) 3D plot and (b) 2D contour plot

Three optimal solutions for the mix design were suggested by design expert software but the optimal value was selected based on the highest desirability of 1.0. An additional experiment was conducted based on the optimal predicted mix design factors of 1.0% nanosilica and 5% binder content. The percentage error (%) between experimental and optimal predicted values was calculated using equation 6.

$$Error = \frac{Experimental - Model}{Experimental} \times 100 \qquad (6)$$

Optimization result suggests that 1% nanosilica and 5% binder content as optimal values to achieve design requirements. The percentage error difference is presented in Table 2, in all the responses analyzed the percent error difference was found to be less than 5%, this indicates that the predicted values for the developed models are in good agreement with the experimental values.

Table 2 Verification of experiment at optimum conditions

Parameter	Predicted	Observe	Error (%)
VTM	3.10	3.21	3.43
Stability	12.95	13.35	2.99
Flow	2.97	3.11	4.5

4. CONCLUSION

The research investigates the application of RSM technique to evaluate the effect of nanosilica and binder content variation on volumetric properties of composite modified asphalt mixtures. Based on the analysis in this research the following conclusions can be drawn:

1. Results from the statistical analysis show that the predicted values obtained are consistent with the experimental results.
2. A high coefficient of determination (R^2) was obtained for all the responses analyzed, this indicates a high degree of accuracy with the developed regression quadratic models and the experimental data are in real agreement with the predictive models.
3. Both the individual effects of nanosilica and binder content are significant; however, the quantity of binder used shows higher influence on the volumetric properties.
4. Higher stability was achieved with the mixtures prepared with 2% nanosilica and binder content range of 5 - 6%.
5. Based on the optimization results a mean error of less than 5% were achieved for all the responses, this indicates that for asphalt mixture design with high performance properties, optimization using RSM is very effective approach.

For further research, performance test such as beam fatique and wheel tracking test can be carried out at higher temperatures and loading conditions.

5. AUTHOR'S CONTRIBUTIONS

The experiments were conducted by Nura Bala as part of his PhD research project, under the supervision of Assoc Prof. Dr M. Napiah. The draft of the manuscript was prepared by Nura Bala and Assoc Prof Dr I. Kamaruddin and revised by Assoc Prof. M. Napiah.

6. ETHICS

This article is original work and not under consideration for publication in any other journal. There is no conflict interest on this paper.

7. REFERENCES

[1] Y. Zaika and L. Djakfar, "Gradation band of some types material for reservoir base of porous pavement," International Journal of Geomate, vol. 11, 2016, pp. 2486-2492.

[2] Y. Zhang, R. Luo, and R. L. Lytton, "Characterizing permanent deformation and fracture of asphalt mixtures by using compressive dynamic modulus tests," Journal of Materials in Civil Engineering, vol. 24, 2011, pp. 898-906.

[3] X. Lu and U. Isacsson, "Effect of ageing on bitumen chemistry and rheology," Construction and Building Materials, vol. 16, 2002, pp. 15-22.

[4] F. Durrieu, F. Farcas, and V. Mouillet, "The influence of UV aging of a styrene/butadiene/styrene modified bitumen: comparison between laboratory and on site aging," Fuel, vol. 86, 2007, pp. 1446-1451.

[5] H. Di Benedetto, C. De La Roche, H. Baaj, A. Pronk, and R. Lundström, "Fatigue of bituminous mixtures," Materials and structures, vol. 37, 2004, pp. 202-216.

[6] L. R. Logitharan, K. Somasundaraswaran, and J. M. Ramanujam, "Rehabilitation works in Queensland using foamed bituminous stabilisation," International Journal of Geomate, vol. 8, 2015, pp. 1308-1315.

[7] C. Miller, D. Little, A. Bhasin, N. Gardner, and B. Herbert, "Surface energy characteristics and impact of natural minerals on aggregate-bitumen bond strengths and asphalt mixture durability," Transportation Research Record: Journal of the

Transportation Research Board, 2012, pp. 45-55.

[8] N. Bala and I. Kamaruddin, "Physical and storage stability properties of linear low density polyethylene at optimum content," in Engineering Challenges for Sustainable Future: Proceedings of the 3rd International Conference on Civil, Offshore and Environmental Engineering (ICCOEE 2016, Malaysia, 15-17 Aug 2016), 2016, p. 395.

[9] N. I. M. Yusoff, D. Mounier, G. Marc-Stéphane, M. R. Hainin, G. D. Airey, and H. Di Benedetto, "Modelling the rheological properties of bituminous binders using the 2s2p1d model," Construction and Building Materials, vol. 38, 2013, pp. 395-406.

[10] V. Punith and A. Veeraragavan, "Behavior of reclaimed polyethylene modified asphalt cement for paving purposes," Journal of Materials in Civil Engineering, vol. 23, 2010, pp. 833-845.

[11] N. Bala, I. Kamaruddin, M. Napiah, and N. Danlami, "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.

[12] R. Li, F. Xiao, S. Amirkhanian, Z. You, and J. Huang, "Developments of nano materials and technologies on asphalt materials–A review," Construction and Building Materials, vol. 143, 2017, pp. 633-648.

[13] N. I. M. Yusoff, A. A. S. Breem, H. N. Alattug, A. Hamim, and J. Ahmad, "The effects of moisture susceptibility and ageing conditions on nano-silica/polymer-modified asphalt mixtures," Construction and Building Materials, vol. 72, 2014, pp. 139-147.

[14] S. Kikuchi, N. Kronprasert, and S. M. Easa, "Aggregate blending using fuzzy optimization," Journal of Construction Engineering and Management, vol. 138, 2012, pp. 1411-1420.

[15] M. Soltani, T. B. Moghaddam, M. R. Karim, S. Shamshirband, and C. Sudheer, "Stiffness performance of polyethylene terephthalate modified asphalt mixtures estimation using support vector machine-firefly algorithm," Measurement, vol. 63, 2015, pp. 232-239.

[16] T. B. Moghaddam, M. Soltani, and M. R. Karim, "Stiffness modulus of Polyethylene Terephthalate modified asphalt mixture: A statistical analysis of the laboratory testing results," Materials & Design, vol. 68, 2015, pp. 88-96.

[17] L. Chávez-Valencia, A. Manzano-Ramírez, E. Alonso-Guzmán, and M. Contreras-García, "Modelling of the performance of asphalt pavement using response surface methodology—the kinetics of the aging," Building and environment, vol. 42, 2007, pp. 933-939.

[18] M. O. Hamzah, B. Golchin, and C. T. Tye, "Determination of the optimum binder content of warm mix asphalt incorporating Rediset using response surface method," Construction and Building Materials, vol. 47, 2013, pp. 1328-1336.

[19] A. I. Nassar, N. Thom, and T. Parry, "Optimizing the mix design of cold bitumen emulsion mixtures using response surface methodology," Construction and Building Materials, vol. 104, 2016, pp. 216-229.

[20] D. C. Montgomery, Design and analysis of experiments: John Wiley & Sons, 2008.

[21] R. H. Myers, D. C. Montgomery, and C. M. Anderson-Cook, Response surface methodology: process and product optimization using designed experiments: John Wiley & Sons, 2016.

[22] M. Hamzah, S. Omranian, B. Golchin, and M. Hainin, "Evaluation of effects of extended short-term aging on the rheological properties of asphalt binders at intermediate temperatures using respond surface method," Jurnal Teknologi, vol. 73, 2015, pp. 133-139.

[23] T. B. Moghaddam, M. Soltani, M. R. Karim, and H. Baaj, "Optimization of asphalt and modifier contents for polyethylene terephthalate modified asphalt mixtures using response surface methodology," Measurement, vol. 74, 2015, pp. 159-169.

[24] Q. Li, L. Cai, Y. Fu, H. Wang, and Y. Zou, "Fracture properties and response surface methodology model of alkali-slag concrete under freeze–thaw cycles," Construction & Building Mat., vol. 93, 2015, pp. 620-626.