A two level factorial experimental design for evaluation of viscoelastic properties of bitumens containing a surfactant warm additive

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ABSTRACT

In this paper, a two level factorial experimental design was developed to evaluate the effects of a surfactant warm additive and test temperature on the elastic and viscous properties of bitumen under unaged, short-term aged and long-term aged conditions. These rheological properties were measured by the dynamic shear rheometer (DSR) under fixed strains oscillatory loading at high and intermediate temperatures. The results showed that additive content and test temperature are significant factors influencing prediction of loss modulus and storage modulus of selected bitumen at various ageing conditions, but interaction effect of additive content and temperature is not significant factor on the prediction of loss modulus in unaged and long-term ageing conditions. Using higher amount of this additive decreases both loss and storage modulus in various ageing conditions, while increasing the test temperatures result in lower values for loss and storage modulus.

Keywords: Additives, Ageing, Rheology, Testing, Warm Asphalt Mixture

No. 13
A TWO LEVEL FACTORIAL EXPERIMENTAL DESIGN FOR EVALUATION OF VISCOELASTIC PROPERTIES OF BITUMENS CONTAINING A SURFACTANT WARM ADDITIVE

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ABSTRACT

In this paper, a two level factorial experimental design was developed to evaluate the effects of a surfactant warm additive and test temperature on the elastic and viscous properties of bitumen under unaged, short-term aged and long-term aged conditions. These rheological properties were measured by the dynamic shear rheometer (DSR) under fixed strains oscillatory loading at high and intermediate temperatures. The results showed that additive content and test temperature are significant factors influencing prediction of loss modulus and storage modulus of selected bitumen at various ageing conditions, but interaction effect of additive content and temperature is not significant factor on the prediction of loss modulus in unaged and long-term ageing conditions. Using higher amount of this additive decreases both loss and storage modulus in various ageing conditions, while increasing the test temperatures result in lower values for loss and storage modulus.

Key words: Warm asphalt, Rheology, Additive, Testing, Ageing.

1. INTRODUCTION

Test procedures in engineering and scientific experiments are often guided by subjective consideration of practicality and established laboratory protocols [1]. While these kinds of experiments are viewed economically based on the number of test runs, statistical approaches in design of experiments are used to ensure that experiments are designed by economical criteria [1]. With this in mind, a two level factorial experiment design with limited test samples was used to evaluate the influence of a chemical warm mix additive on viscoelastic properties of a polymer modified bitumen in terms of storage modulus and loss modulus using statistical methods.

In the last two decades, warm mix asphalt additives have been introduced to the asphalt industries to reduce air pollutions and fuel usage in production process and to improve the workability of asphalt mixtures at lower compaction temperatures [2]. These additives are classified into three groups: chemical additives, foamed bitumen technology, and organic additives [2-3]. In this study, a water-free surfactant warm additive was selected that can reduce compaction and mixing temperatures of asphalt mixtures by about 35 °C [4-6]. Studies on the influence of this additive on the bitumen's properties have not been completed and more detailed studies are necessary [7].

Xiao et al. (2012) evaluated the effects of 1.5% of this warm additive on the viscoelastic properties of different bitumen at high temperatures for unaged condition [8]. It was found that this dosage of additive decreases storage modulus of a PG 76-22 bitumen at temperatures ranging from 76 to 82 °C in unaged condition, but it increases the storage modulus of PG 64-22, PG 58-28, and PG 52-28 bitumen at temperature ranges 64 to 70 °C, 58 to 60 °C, and 52 to 58 °C, respectively. In addition, 1.5% of this additive slightly changed the loss modulus of the PG 76-22 bitumen when tested between 76 to 82 °C under unaged condition.

This study quantifies the rheological properties of bitumen blended with different additive contents under three ageing conditions (unaged, short-term ageing, and long-term ageing) via statistical approach. For this purpose, the loss modulus (viscous modulus) and storage modulus (elastic modulus) of unaged and short-term aged bitumens at high temperatures were measured. Additionally, loss modulus and storage modulus were determined for long-term aged bitumen at intermediate temperatures. In the next step, statistical analyses were conducted on the results and corresponding models were developed for appropriate ageing conditions.

2. MATERIALS AND METHODS

2.1 Materials

Polymer modified bitumen was chosen as a hard bitumen and a surfactant additive named Rediset produced by AkzoNobel was used as a chemical warm additive. The properties of the bitumen are shown in Table 1.

Table 1. Properties of bitumen

Ageing condition	Properties	Value
Original bitumen	Softening point (°C)	69
	Penetration (0.1 mm)	50
	Ductility (cm)	90
	Flash point (°C)	344
	$G^*/\sin\delta$ at 82°C (Pa)	2374
Short-term aged bitumen (RTFO)	$G^*/\sin\delta$ at 82°C (Pa)	3968
Long-term aged bitumen (RTFO+PAV)	$G^*\sin\delta$ at 25°C (MPa)	5.41

2.2 Samples Preparation and Laboratory Tests

To investigate the viscoelastic properties of bitumen incorporating additive in various ageing conditions, the base bitumen was heated and blended with 0, 2, and 4% of additive by weight of bitumen using a low-shear stirrer. These bitumen were short-term aged and then long-term aged artificially via Rolling Thin Film Oven (RTFO) and Pressure Ageing vessel (PAV), respectively. Unaged and short-term-aged samples with 1 mm thickness and long-term-aged samples with 2 mm thickness were tested using the dynamic shear rheometer (DSR). The storage modulus (G') as an indicative of elastic properties of bitumen and loss modulus (G") as an indicative of viscous properties were defined as responses of the experiments.

2.2 Design of Experiments

Test plan was designed based on a two level factorial experimental design for two numerical factors (additive content and test temperature) and one categorical factor (ageing condition) using a central point as middle level of factors. Tables 2, 3, and 4 are the experimental matrixes for unaged, short-term-aged and long-term-aged conditions, respectively. Tests were conducted on the samples containing 0 to 4% additive for test temperatures ranging from 46 to 70°C for unaged and short-term-aged conditions, while bitumen subjected to long-term ageing were tested between 18 to 28°C.

Table 2. Experimental matrix for unaged condition

Sample No.	Temperature (°C)	Additive content (%)	G' (KPa)	G" (KPa)
1	70.00	4.00	1.781	4.924
2	70.00	0.00	2.843	6.238
3	46.00	4.00	26.614	65.319
4	58.00	2.00	8.267	19.33
5	58.00	2.00	7.727	18.118
6	58.00	2.00	7.953	18.476
7	70.00	0.00	2.629	6.121
8	46.00	0.00	33.984	65.283
9	46.00	0.00	36.507	71.098
10	46.00	4.00	27.949	65.808
11	46.00	0.00	33.653	67.424
12	70.00	0.00	2.368	5.782
13	70.00	4.00	1.784	4.673
14	46.00	4.00	25.558	62.86
15	70.00	4.00	1.805	5.07

Table 3. Experimental matrix for short-term ageing condition

Sample No.	Temperature (˚C)	Additive content (%)	G' (KPa)	G" (KPa)
1	46.00	4.00	55.547	121.76
2	46.00	0.00	78.667	139.64
3	58.00	2.00	16.682	34.16
4	46.00	0.00	74.522	132.37
5	58.00	2.00	16.863	36.6
6	70.00	4.00	2.4481	7.6529
7	70.00	0.00	4.638	10.917
8	46.00	4.00	49.072	105.31
9	58.00	2.00	16.063	34.977
10	46.00	0.00	75.527	127.26
11	70.00	4.00	2.2019	7.4131
12	46.00	4.00	51.703	111.45
13	70.00	0.00	4.564	10.566
14	70.00	0.00	5.2645	11.85
15	70.00	4.00	2.648	8.4849

Table 4. Experimental matrix for long-term ageing condition

Sample No.	Temperature (°C)	Additive content (%)	G' (MPa)	G" (MPa)
1	28	4	2.552	2.85
2	28	0	5.142	4.648
3	28	0	4.311	4.034
4	16	0	11.409	8.773
5	22	2	5.794	4.274
6	28	0	5.267	4.82
7	16	0	10.408	8.015
8	16	4	6.958	6.465
9	16	4	5.083	4.721
10	28	0	4.399	4.201
11	22	2	6.116	4.147
12	22	2	5.051	3.648
13	22	2	7.07	5.238
14	16	4	5.099	4.844
15	16	4	4.446	4.121
16	16	0	11.741	9.503
17	28	4	1.784	2.014
18	16	0	10.407	8.272
19	28	4	1.957	2.187
20	28	4	1.588	1.778

2.3 Analysis Method

Analysis of variance (ANOVA) was performed to determine the significant terms for prediction of storage modulus and loss modulus using Design-Expert 6.0.6 software. In the next step, 2 interaction factor models were developed, related graphs are plotted, and main interoperations were extracted.

3. RESULTS AND DISCUSSIONS

3.1 Effects of Warm Additive Content on Storage Modulus for Unaged Condition

Table 5 shows the analysis of variance (ANOVA) for factorial model of storage modulus for unaged condition. As can be seen, additive content (A), test temperature (B), and interaction between temperature and additive content (AB) are statistically significant terms in prediction of elastic part of bitumen properties for corresponding ageing condition. A “Prob>F” value less than 0.05 is indicative of a statistically significant factors or their interactions. From Table 5, F-value of additive content is higher than its interaction with test temperature. This means that the effect of additive content is higher than its interaction with temperature on the storage modulus of bitumen.
Eq. (1) shows a mathematical equation for the storage modulus as a function of additive content and test temperature. The F-value of 1055.74 in Table 5 shows high significance of the model in prediction of the elastic characteristics of the bitumen for unaged condition. The predicted and actual values of storage modulus for unaged condition are shown in Fig. 1.
Interaction effects between additive content and test temperature are illustrated in Fig. 2. From this figure, additive content decreases the storage modulus of the selected bitumen at unaged condition. This result is consistent with the results obtained by Xiao et al. (2012b) for a PG 76-22 under unaged condition. They showed that although 1.5% additive content decreases the storage modulus of a PG 76 bitumen at temperature from 76 to 82°C, this additive increases the storage modulus of PG 64, PG 58, and PG 52 at temperatures ranging from 64 to 70°C, 58 to 60°C, and 52 to 58°C, respectively.

Table 5. Analysis of variance for factorial model of storage modulus in unaged condition

Source	Sum of Squares	DF	Mean Square	F-Value	Prob > F
Model	2535.52	3	845.17	1055.74	< 0.0001*
Test temperature	2438.32	1	2438.32	3045.80	< 0.0001*
Additive content	58.49	1	58.49	73.06	< 0.0001*
Test temperature×Additive content	38.71	1	38.71	48.36	< 0.0001*

Note: * Significant at 95% confidence interval

$$G'=96.24 - 1.34\times(\text{Test temperature}) - 5.44\times(\text{Additive content}) + 0.07\ (\text{Testing tmperature} \times \text{Additive content}) \quad (1)$$

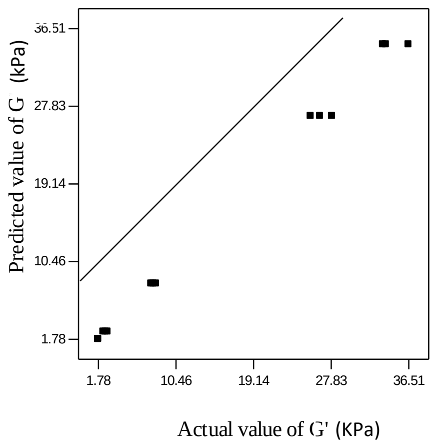

Fig. 1. Predicted and actual storage modulus in unaged condition

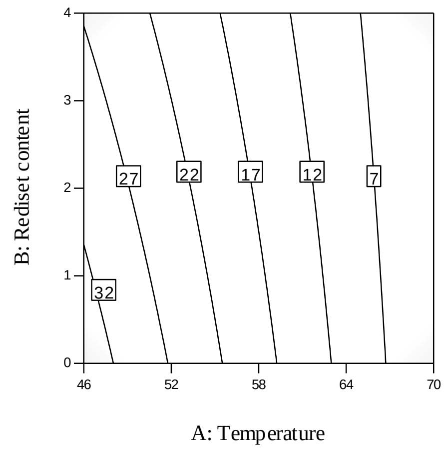

Fig. 2. Storage modulus in unaged condition

3.2 Effects of Additive Content on Loss Modulus for Unaged Condition

The ANOVA results for the loss modulus in unaged condition shown in Table 6 shows that A (test temperature) and B (additive content) exhibit statistically significant effects on the measured loss modulus, while their interactions, AB does not exhibit significant effect. The proposed model of loss modulus for unaged conditions is expressed by Eq. (2). This model was developed without using insignificant interaction term of additive content and test temperature (AB). The predicted and actual values of loss modulus for unaged condition are shown in Fig. 3. Interaction effects between additive content and test temperature are illustrated in Fig. 4. From this figure, increase of additive content from 0 to 4% slightly decreases the loss modulus, while increase of temperature from 46 to 70°C decreases the loss modulus by more than 50 KPa.

Table 6. Analysis of variance for factorial model of loss modulus in unaged condition

Source	Sum of Squares	DF	Mean Square	F-Value	Prob > F
Model	11119.19	3	3706.40	1593.51	< 0.0001*
Test temperature	11101.11	1	11101.11	4772.77	< 0.0001*
Additive content	14.72	1	14.72	6.33	0.0306*
Test temperature×Additive content	3.35	1	3.35	1.44	0.2575**

Note: *, Significant at 95% confidence interval; **, Not significant at 95% confidence interval

$$G''=183.99 - 2.53\times(\text{Test temperature}) - 0.55\times(\text{Additive content}) \quad (2)$$

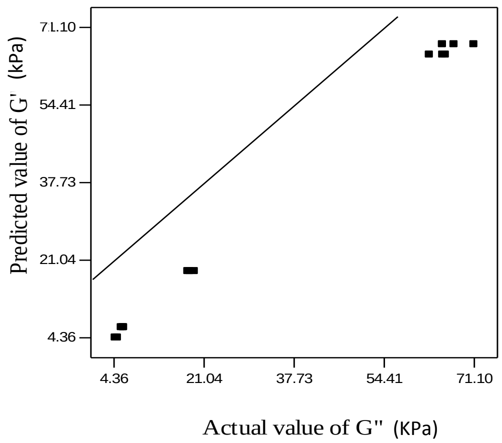

Fig. 3. Predicted and actual loss modulus in unaged condition

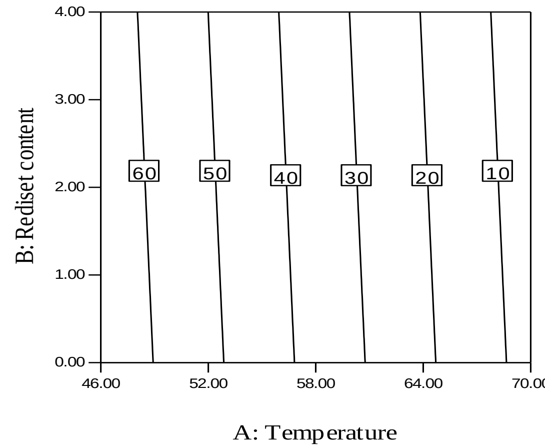

Fig. 4. Loss modulus in unaged condition

3.3 Effects of Additive Content on Storage Modulus for Short-Term Ageing Condition

The ANOVA results for the storage modulus in short-term aged condition, given in Table 7, indicate that A (test temperature), B (additive content), and the interaction of test temperature and additive content (AB) exhibit significant effects on the prediction of storage modulus. From this table, the F-value of test temperature is greater than additive content and interaction of test temperature and Additive content. This implies that the effect of test temperature is higher than other factors on the storage modulus of bitumen for corresponding ageing condition. Fig. 5 shows the predicted and actual value of storage modulus. The closeness of the predicted and actual values to the 1:1 line is illustrated in this figure. Eq. (3) shows a mathematical equation for prediction of storage modulus in short-term ageing condition. This model indicates that higher test temperature and higher additive content decrease storage modulus of bitumen, whereas the interaction effect of test temperature and additive content has inverse effect on storage modulus. This interpretation can be inferred from Fig. 6.

Table 7. Analysis of variance for factorial model of storage modulus in short-term ageing condition

Source	**Sum of Squares**	**DF**	**Mean Square**	**F-Value**	**Prob > F**
Model	11879.35	3	3959.78	1264.85	< 0.0001*
Test temperature	10997.30	1	10997.30	3512.81	< 0.0001*
Additive content	527.52	1	527.52	168.50	< 0.0001*
Test temperature×Additive content	354.53	1	354.53	113.25	< 0.0001*

Note: * Significant at 95% confidence interval

$$G'=213.12 - 2.98\times(\text{Test temperature}) - 16.45\times(\text{Additive content}) + 0.23\times(\text{Test temperature} \times \text{Additive content}) \quad (3)$$

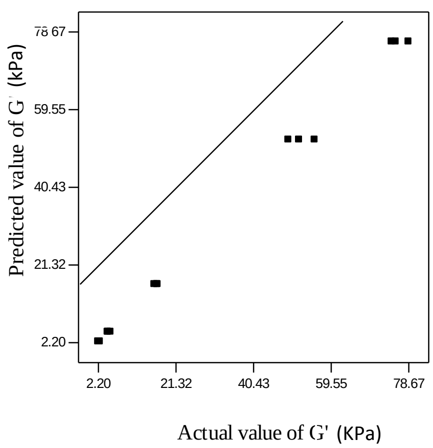

Fig. 5. Predicted and actual storage modulus in short-term ageing condition

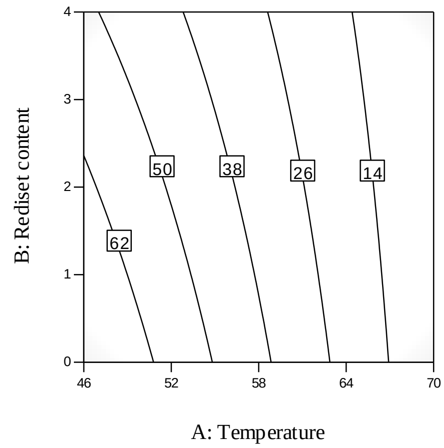

Fig. 6. Storage modulus in short-term ageing condition

3.4 Effects of Additive Content on Loss Modulus for Short-Term Ageing Condition

Table 8 shows the ANOVA results for factorial model of loss modulus in short-term ageing condition. "Prob>F" value less than 0.05 indicates that test temperature (A), additive content (B), and interaction effect of test temperature and additive content (AB) are significant factors in prediction of loss moduls. Eq. (4) exhibits the factorial model for prediction of loss modulus for a corresponding ageing condition. According to this model, test temperature and additive content decrease the loss modulus, but their interaction increases the loss modulus.

Table 8. Analysis of variance for factorial model of loss modulus in short-term ageing condition

Source	**Sum of Squares**	**DF**	**Mean Square**	**F-Value**	**Prob > F**
Model	39267.14	3	13089.05	594.39	< 0.0001*
Test temperature	38636.09	1	38636.09	1754.53	< 0.0001*
Additive content	414.56	1	414.56	18.83	0.0015*
Test temperature×Additive content	216.48	1	216.48	9.83	0.0106*

Note: * Significant at 95% confidence interval

$$G'' = 366.88 - 5.08\times(\text{Test temperature}) - 13.20\times(\text{Additive content}) + 0.17\times(\text{Test temperature} \times \text{Additive content}) \quad (4)$$

The F-value of 594.39 in Table 8 shows high significance of the model in prediction of the loss modulus of the bitumen for short-term ageing condition. The predicted and actual values of loss modulus in short-term ageing condition are shown in Fig. 7. Interaction effects between additive content and test temperature on loss modulus are illustrated in Fig. 8 for short-term ageing condition. Lower loss modulus is related to higher test temperature and higher additive content, while higher loss modulus is related to lower additive content and lower test temperature.

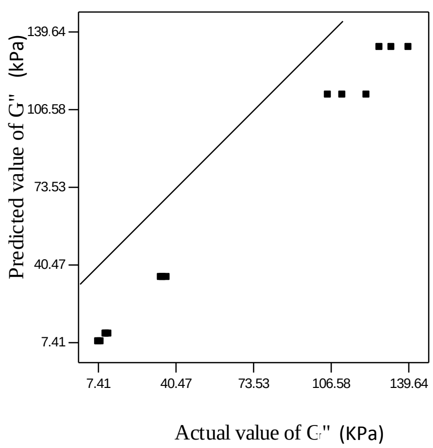

Fig. 7. Predicted and actual loss modulus in short-term ageing condition

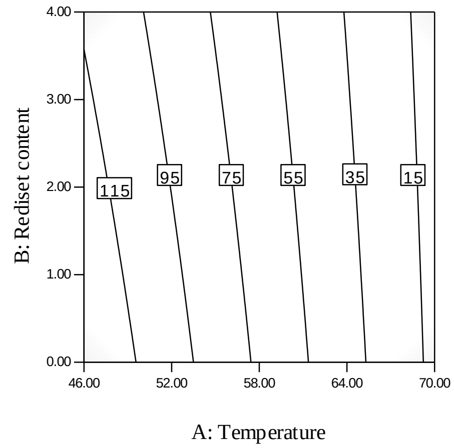

Fig. 8. Loss modulus in short-term ageing condition

3.5 Effects of Additive Content on Storage Modulus for Long-Term Ageing Condition

Table 9 shows the analysis of variance for factorial model of storage modulus in long-term ageing condition. As can be seen, additive content (A), test temperature (B), and interaction between temperature and additive content (AB) are statistically significant factors in prediction of elastic part of bitumen properties at corresponding ageing condition. From Table 9, F-value of additive content is higher than its interaction with temperature. This means that the effect of additive content is higher than its interaction with temperature on the storage modulus of bitumen.
Eq. (5) shows a mathematical equation for the storage modulus as a function of additive content and test temperature. The F-value of 103.17 in Table 9 shows high significance of the model in prediction of the elastic characteristics of the bitumen for long-term ageing condition. Predicted and actual values of the storage modulus for long-term ageing condition are shown in Fig. 9. The interaction effects between additive content and test temperature are illustrated in Fig.10. From this figure, lower additive contents and lower test temperatures show higher elastic modulus for bitumen.

Table 9. Analysis of variance for factorial model of storage modulus in long-term ageing condition

Source	**Sum of Squares**	**DF**	**Mean Square**	**F-Value**	**Prob > F**
Model	171.28	3	57.09	103.17	< 0.0001*
Test temperature	92.89	1	92.89	167.85	< 0.0001*
Additive content	70.63	1	70.63	127.64	< 0.0001*
Test temperature×Additive content	7.76	1	7.76	14.02	0.0020*

Note: * Significant at 95% confidence interval

$$G' = 19.27 - 0.52\times(\text{Test temperature}) - 2.33\times(\text{Additive content}) + 0.06\times(\text{Test temperature}\times\text{Additive content}) \quad (5)$$

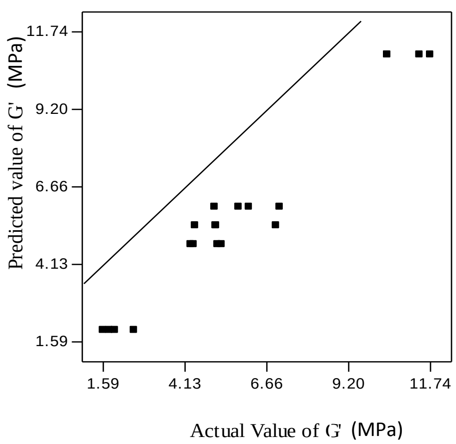

Fig. 9. Predicted and actual storage modulus in long-term ageing condition

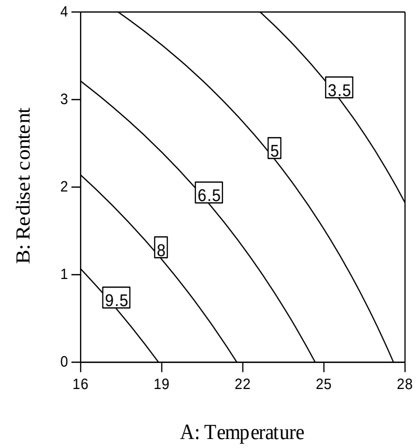

Fig. 10. Storage modulus in long-term ageing condition

3.6 Effects of Additive Content on Loss Modulus for Long-Term Ageing Condition

The proposed equation for loss modulus in long-term ageing conditions is expressed by Eq. (6). This equation is developed using significant factors illustrated in Table 10. The ANOVA results show that test temperature (A) and additive content (B) are significant factors for prediction of loss modulus. The predicted and actual loss modulus values for long-term ageing condition are plotted in Fig. 11. In Fig. 12, contour plots of loss modulus versus additive content and test temperature are illustrated. When additive content and test temperature decrease, the loss modulus increases for long-term ageing condition.

Table 10. Analysis of variance for factorial model of loss modulus in long-term ageing condition

Source	**Sum of Squares**	**DF**	**Mean Square**	**F-Value**	**Prob > F**
Model	85.45	3	28.48	64.01	< 0.0001*
Test temperature	49.64	1	49.64	111.57	< 0.0001*
Additive content	33.89	1	33.89	76.17	< 0.0001*
Test temperature×Additive content	1.92	1	1.92	4.31	0.0555**

Note: * Significant at 95% confidence interval; **, Not significant at 95% confidence interval

$$G''=12.99 - 0.29\times(\text{Test temperature}) - 0.73\times(\text{Additive content}) \quad (6)$$

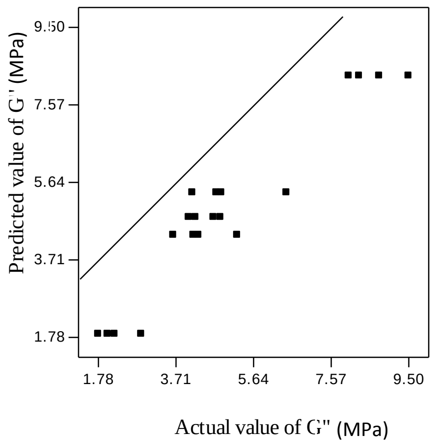

Fig. 11. Predicted and actual loss modulus in long-term ageing condition

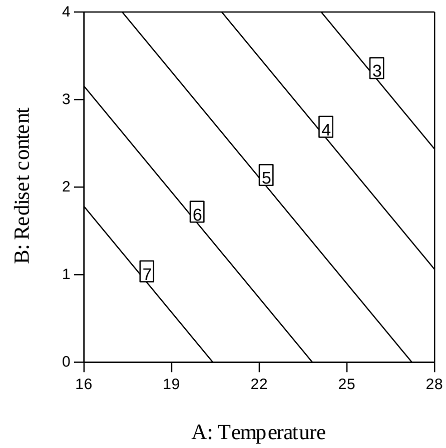

Fig. 12. Loss modulus in long-term ageing condition

4. CONCLUSIONS

This study described the effects of a surfactant warm additive content and test temperature on the elastic and viscous properties of the selected bitumen. The results and statistical analysis showed that additive content is a significant factor in prediction of elastic and viscous modulus of this bitumen in various ageing condition. This warm additive decreases both viscous and elastic modulus values in intermediate temperatures. This may imply that this additive as a warm mix additive improves the fatigue resistance of the selected bitumen at intermediate temperatures. Also, this additive decreases loss modulus and storage modulus of both unaged and short-term aged binder. It implies that this additive may have a negative effect on the rutting properties of the selected bitumen at high temperatures. Interaction effect of additive and temperature increases loss and storage modulus in short-term ageing conditions. This implies that the different effects of this additive at several high temperatures.

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REFERENCES

[1] Robert. Mason, Richard F. Gunset, and James L. Hess, 2003. Statistical design and analysis of experiments with application to engineering and science. Second edition, John Wiley & Sons Inc., Hoboken, New Jersey.

[2] Capitão, S., Picado-Santos, L. and Martinho, F. 2012. Pavement engineering materials: Review on the use of warm-mix asphalt. Construction and Building Materials, 36, 1016-1024.

[3] Rubio, M. C., Martínez, G., Baena, L. and Moreno, F. 2012. Warm mix asphalt: an overview. Journal of Cleaner Production, 24, 76-84.

[4] Arega, Z., Bhasin, A., Motamed, A. and Turner, F. 2011. Influence of Warm-Mix Additives and Reduced Ageing on the Rheology of Bitumens with Different Natural Wax Contents. Journal of Materials in Civil Engineering, 23, 1453-1459.

[5] Banerjee, A., de Fortier Smit, A. and Prozzi, J. A. 2012. The effect of long-term ageing on the rheology of warm mix bitumens. Fuel, 97, 603-611

[6] Hamzah, M. O., Golchin, B., Jamshidi, A. and Chailleux, E. 2014. Evaluation of Rediset for use in warm-mix asphalt: a review of the literatures. International Journal of Pavement Engineering, 1-23.

[7] Xiao, F., Amirkhanian, S. N. and Zhang, R. 2012. Influence of Short-Term Ageing on Rheological Characteristics of Non-Foaming WMA Bitumens. Journal of Performance of Constructed Facilities, 26, 145-152.

[8] Xiao, F., Punith, V. and Amirkhanian, S. N. 2012. Effects of non-foaming WMA additives on bitumens at high performance temperatures. Fuel, 94, 144-155.