An Evaluation of Solubility of Wax and Asphaltene in Crude Oil for Improved Flow Properties Using a Copolymer Solubilized in Organic Solvent with an Aromatic Hydrocarbon

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Abstract-Wax and asphaltene are high molecular weighted compounds that contribute to the stability of crude oil at a dispersed state. Transportation of crude oil along pipelines from the oil rig to the refineries causes fluctuation of temperature which will lead to the coagulation of wax and flocculation of asphaltenes. This paper focuses on the prevention of wax and asphaltene precipitate deposition on the inner surface of the pipelines by using a wax inhibitor and an asphaltene dispersant. The novelty of this prevention method is the combination of three substances; a wax inhibitor dissolved in a wax inhibitor solvent and an asphaltene solvent, namely, ethylene-vinyl acetate (EVA) copolymer dissolved in methylcyclohexane (MCH) and toluene (TOL) to inhibit the precipitation and deposition of wax and asphaltene. The objective of this paper was to optimize the percentage composition of each component in this inhibitor which can maximize the viscosity reduction of crude oil. The optimization was divided into two stages which are the laboratory experimental stage in which the viscosity of crude oil samples containing inhibitor of different component compositions is tested at decreasing temperatures and the data optimization stage using response surface methodology (RSM) to design an optimizing model. The results of experiment proved that the combination of 50% EVA + 25% MCH + 25% TOL gave a maximum viscosity reduction of 67% while the RSM model proved that the combination of 57% EVA + 20.5% MCH + 22.5% TOL gave a maximum viscosity reduction of up to 61%.

Keywords—Asphaltene, ethylene-vinyl acetate, methylcyclohexane, toluene, wax.

I. INTRODUCTION

As the crude oil travels across the pipelines which stretch from under the sea level to the surface, the temperature of crude oil will gradually decrease until it is lower than a certain temperature known as the wax appearance temperature (WAT) or the pour point temperature. When this occurs, the wax will crystallize and asphaltene will flocculate. When a temperature gradient exists between the crude oil and the cold pipeline inner wall surface, it will lead to wax and asphaltene solids deposition on the inner walls of the pipelines [1]. Asphaltene, unlike waxes, will aggregate and flocculate rather than form a full solid or solid-like structure [1].

There are several methods available of which the more common ones are mechanical removal and chemical removal [2]. Another method is to use coiled tubing with jet attachment to remove the deposits in the wellbore. Chemical removal makes use of different types of chemicals to remove the wax and asphaltene precipitation.

Many researches have been done to formulate a wax inhibitor that can provide the best results. Among those are alkyl derivatives of polymeric acrylates, commercial inhibitor containing growth arrestor and nucleator, EVA, polymeric fatty ester, vinyl pyridin, methacrylic acid ester, maleic acid anhydride, alkyl fumarate-vinyl acetate, polyethylenepoly(ethylene-propylene), poly(maleic anhydride amide $co-\alpha$ hydrophobically modified polybetaines, olefin). polyoctadecylmethacrylate, phosphoric ester mixture with sodium aluminate, poly(N,N-diallyl-N-octadecylamine-alt-(maleic acid) [3]-[19]. The studies proved polymeric acrylates and EVA to be the most efficient wax copolymers that can inhibit the formation of wax. However, researches have also proven that the vinyl-acetate weight content of EVA and the concentration of inhibitor also play a vital role in prevention of wax deposition. Research conducted by Machado and Lucas [20] has proven that the efficiency of the EVA is dependent on the type of crude oil and that the optimum VA content and concentration is also dependent on the type of crude oil. Similar studies were done by Ashbaugh et al. [21] and Taraneh et al. [22] which concluded similar results. Some studies showed that a solvent can increase the efficiency of the wax inhibitor. Different types of solvents were experimented for the efficiency such as 1,2-dichloroethane, cyclohexane, xylene, chloroform, ethyl acetate, hexane, butanone, heptane, acetone and petroleum ether [23]-[25]. The results of the studies proved 1,2- dichloroethane, acetone and xylene to be the best wax inhibitor solvent so far.

Flocculation of aspahltene is another worrying issue in the deposition of solids on pipelines. Several studies had been done to formulate an asphaltene inhibitor for the crude oil flow assurance. This is because the wax inhibitor does not work to inhibit asphaltene, but as asphaltene is dispersed by

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hydrocarbons, the inhibition of wax coagulation would increase the amount of hydrocarbon to form stable micelles. Studies by Bouts et al. [26], Buriro and Shuker [27], Palermo et al. [28] experimented various range of dispersants such as oil-soluble polymeric dispersant, dodecylbenzenesulfonic acid, cardanol and some commercially available dispersant. The results mainly showed that an aromatic compound is an efficient asphaltene dispersant.

This study aims to prove the efficiency of a pour-point depressant made up of a wax inhibitor, EVA dissolved in MCH and TOL to inhibit the precipitation and deposition of wax and asphaltene. The novelty of this paper is the calibration of weightage composition percentage of EVA, MCH and TOL needed in the inhibitor to give a maximum viscosity and WAT reduction which directly relates to the success of wax and asphaltene inhibition. Besides that, RSM software is used to optimize the experimentally obtained results analytically to present the highest possible efficiency in reduction of crude oil viscosity.

II. MATERIALS & METHODS

A. Materials

The crude oil to be tested in this paper is Malaysian crude oil, specifically taken from Sabah platform. Malaysian crude oil is known to have minimal amount of wax but high content of asphaltene. EVA copolymer is used as the wax crystal modifier or wax inhibitor which adheres to the active surface of wax molecules, thus, controlling the size of wax crystals from increasing in size or depositing by the pipeline walls. EVA18 is used in this experiment with 18 wt.% vinyl acetate. This is because, according to past researches, EVA with lower VA content works best for crude oil with higher asphaltene content. MCH is used as a solvent for the EVA in this experiment. The melting point of EVA18 is at 84 °C. Therefore, an efficient organic solvent with boiling point higher than 84 °C is required to be able to dissolve the copolymer as the reaction has to take place at a high temperature. TOL is used as the asphaltene dispersant which adheres to the free active surface of the asphaltene molecules forming stable micelles which otherwise will form micelles with each other, forming flocculates. All these reagents were supplied by Merck Sdn. Bhd, Selangor, Malaysia.

B. Viscosity Measurement

Viscometer is used to evaluate the viscosity of the crude oil samples. The viscometer used is the Brookfield Programmable Viscometer DV-III + Rheometer. The standard settings of the equipment are shown in Table I.

TABLE I							
STANDARD SETTINGS OF VISCOMETER							
Spindle Size	7						
Rotational Speed	100 rpm						
Units	cgs						
Readings	Viscosity (cP) Torque (%)						

The viscometer is left to warm up for about 10 minutes after

startup, and then, auto-zeroed without the spindle and at a leveled position. Once, the auto-zeroing completed, the spindle was fixed with care while holding and pushing the thread head upwards in order to avoid disturbance of the calibration (Brookfield Engineering Laboratories Inc.).

C. Experimental Procedure

Prior to the experiment, the crude oil is heated in an oven at 90 °C overnight to remove any primitively formed wax crystals and asphaltene agglomerates. The apparatus is heated up to avoid wax and asphaltene formation upon contact with the crude oil. EVA, MCH and TOL are also heated in a water bath to increase the efficiency of the inhibitor.

For the preparation of the inhibitor, the reaction is conducted at a temperature of about 90 °C. Each component is measured separately in accordance to the manipulated percentage composition and mixed together at 90 °C to form a 10000 ppm of 0.01 mL inhibitor. When the inhibitor has completely melted, 10 mL of crude oil is poured into the inhibitor and mixed evenly before it is heated up to allow the reaction to occur. The samples are shaken again before testing.

Each set of samples is tested of its viscosity from 40 °C to 5 °C at every 5 °C intervals. Firstly, the control experiment is conducted which is the blank crude oil at the absence of inhibitor. The Brookfield Programmable Viscometer DV-III + Rheometer was used with spindle 7 and a rotational speed of 100 rpm.

The procedure is repeated for each of the other samples that contain inhibitor. A total of 39 samples were tested, one of which is the control sample of blank crude oil and the rest at varying percentage composition of EVA from 10% to 90%, MCH from 5% to 80% and TOL from 5% to 80%. The results are graphically analyzed to find the viscosity reduction and the WAT temperature.

D. RSM Modeling

RSM was applied using a commercial statistical package, Design Expert, version 10.0.3.1. The type of design used is the D-optimal design with split plot and coordinate exchange. The design has 39 runs and no blocks. There are three factors in the design which is the percentage of EVA, percentage MCH and percentage of TOL. Eight responses are observed, the viscosity at each temperature, namely, 40 °C, 35 °C, 30 °C, 25 °C, 20 °C, 15 °C, 10 °C and 5 °C. A second order polynomial equation was used to express the responses as a function of the independent variables. The constraint of the experiment is set as $0 \le a + B + C \le 100$ as the total amount of composition of each chemical has to make up 100% of the inhibitor.

III. RESULTS & DISCUSSION

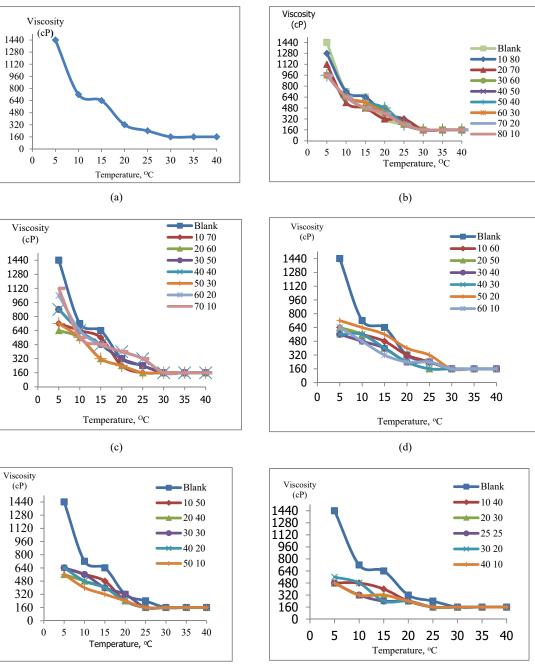
A. Viscosity Reduction Analysis

The results were separated into 10 sets according to the percentage of EVA. In each set of results, the data are graphically analyzed with varying MCH and TOL (Fig. 1).

The experimental data proved that as EVA is increased in amount in the crude oil, the viscosity of crude oil will eventually decrease until a certain point. After that point, the presence of EVA contributes to the thickening of the crude oil which then increases the viscosity. This can be observed in the graph above. Figs. 1 (a)-(f) show the positive effect of EVA which managed to reduce the viscosity at 5 $^{\circ}$ C from 1440 cP up to 480 cP. However, Figs. 1 (g)-(j) show that the viscosity

starts to increase again. This is because EVA is a solid copolymer. Although it is used as a wax inhibitor, excess of this plastic will increase the presence of solids or viscous substance in the crude oil, causing an increase in viscosity. Table II shows the summary of the experimental results.

(f)





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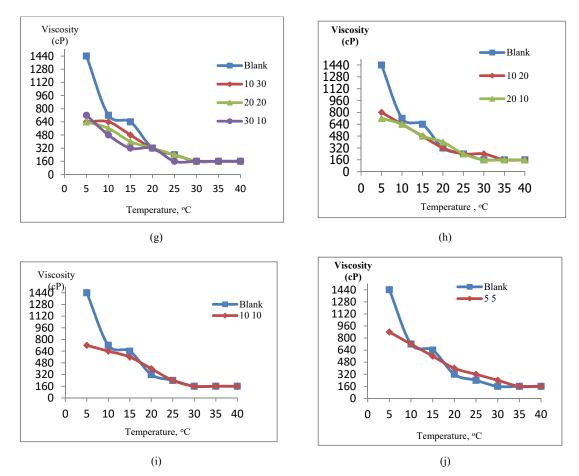


Fig. 1 Viscosity of crude oil with EVA against temperature (a) blank crude oil (b) crude oil with 10% EVA (c) crude oil with 20% EVA (d) crude oil with 30% EVA (e) crude oil with 40% EVA (f) crude oil with 50% EVA (g) crude oil with 60% EVA (h) crude oil with 70% EVA (i) crude oil with 80% EVA (j) crude oil with 90% EVA

SUMMA	SUMMARY OF VISCOSITY REDUCTION ANALYSIS OF SAMPLES						
EVA (%)	Viscosity reduction (%)	Optimum amount of MCH and TOL (% MCH: % TOL)					
10	11 - 33	70:20 80:10					
20	22 - 56	20:60					
30	50 - 61	30 : 40 40 : 30					
40	56 - 61	50:10					
50	61 - 67	25:25					
60	50 - 56	20:20					
70	44 - 50	20:10					
80	50	-					
90	39	-					

TABLE II

From the summary of each analysis as presented in Table II, the addition of EVA will increase the viscosity reduction as compared to that of the blank crude oil. However, 60% and above EVA addition will cause an adverse effect and increase the viscosity of crude oil. As discussed at each case earlier, an excess of EVA will cause presence of solids in the crude oil due to the nature of the copolymer as a solid plastic. This affects the viscosity reading by increasing it. On the other hand, a lower amount of EVA is insufficient to adhere to all the wax molecules to avoid the crystallization growth of wax.

The experimental analysis proved that adding inhibitor containing EVA, MCH and TOL will reduce the viscosity spike at lower temperatures. However, it does not affect viscosity at temperature higher than 25 °C because the wax and asphaltene does not form precipitates at higher temperatures; thus, the inhibitor is dormant at that temperature. The analysis also proves that the MCH does not affect the efficiency of the inhibitor much as it is only intended to dissolve the EVA. However, the MCH has to be sufficient to be able to dissolve all the EVA. Insufficient amount will cause inefficiency of the EVA to act as the wax inhibitor because EVA in solid form cannot react with the wax molecules. Analysis of the data also showed that increasing the amount of TOL can decrease the viscosity. This is because TOL acts as the asphaltene solvent, thus, will dissipate the asphaltene flocculates which improved the flow properties of crude oil. Therefore, based on the experimental data above, the optimum percentage of EVA needed to be used is 50% which gives the optimum viscosity reduction of up to 67% with the combination of 25% MCH and 25% TOL. This can be compared to the research by Lashkarbolooki et al. [25]

which proved that a combination of 50% EVA + 25% Acetone + 25% P- xylene added at 800 ppm gave a 53% viscosity reduction.

B. WAT Reduction Analysis

The WAT of the blank crude oil is graphically determined by analyzing of the viscosity graph of blank crude oil. The viscosity graph of blank crude oil shows a sudden spike at temperature lower than 20 °C. This shows that 20 °C, which is the final point at which the viscosity is still low, is the WAT as the drastic increase in viscosity after that shows that the wax crystals and asphaltene precipitations have started to form.

Using the WAT of blank crude oil, the viscosity at that point, 320 cP is set as a benchmark for other set of samples. It is assumed that viscosity higher than 320 cP will contain wax crystals and asphaltene precipitation. Using this assumption, the viscosity graph of each sample was analyzed to predict the temperature at which the viscosity starts to increase higher than 320 cP. That point of temperature was taken as the new WAT of the samples containing inhibitor.

Analysis of the WAT of each sample shows that with lower percentage of EVA content, the WAT will increase to be higher than the crude oil WAT. This means that the wax and asphaltene precipitates form even at a higher temperature of about 25 °C. This phenomenon could be due to the interference of the MCH and TOL solvents on the wax crystallization mechanism. Even when excessive amount of EVA is used, the WAT increases above the WAT of blank crude oil. This could be due to the excess EVA that solidifies in the crude oil due to insufficient solvent to dissolve it. Solid EVA within the crude oil would also contribute to the increase of viscosity of crude oil, which gives an impression that wax and asphaltene precipitates have formed, when it is only the copolymer that is giving a negative effect on the viscosity of the crude oil.

C. Optimization of % Composition of EVA, MCH and TOL Using RSM

At response 1 and 2 which were the viscosity at 40 °C and 35 °C, there were no optimizations or models designed as the viscosity of all samples at this temperature were constant. At response 3 and 4 which were the viscosity at 30 °C and 25 °C, the optimization model had an R^2 value of 1.0 as the values were almost constant. At responses 5 to 8 which were the viscosity at 20 °C to 5 °C, the optimization is more significant with the p-value of below 0.05 for most of the cases. Figs. 2-5 show the optimization of different % composition of EVA, MCH, and TOL using RSM.

Fig. 2 shows that maximum viscosity reduction at 20 °C is when EVA and MCH is increased (Fig. 2 (a)), EVA and TOL is increased (Fig. 2 (b)) and when TOL is increased and MCH is decreased (Fig. 2 (c)).

Fig. 3 shows that maximum viscosity reduction at 15 °C is when EVA and MCH is increased (Fig. 3 (a)), EVA is increased with little regards to TOL (Fig. 3 (b)) and when TOL is decreased and MCH is increased (Fig. 3 (c)).

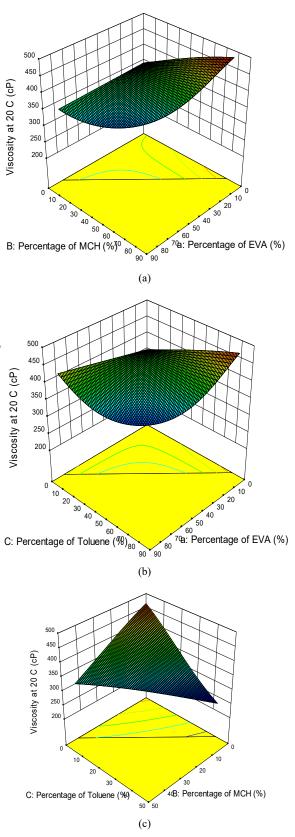


Fig. 2 Optimization at 20 $^{\rm o}{\rm C}$ (a) EVA & MCH (b) EVA & TOL (c) MCH & TOL

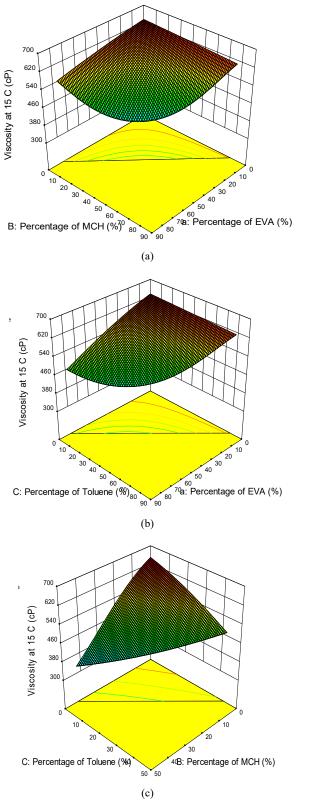
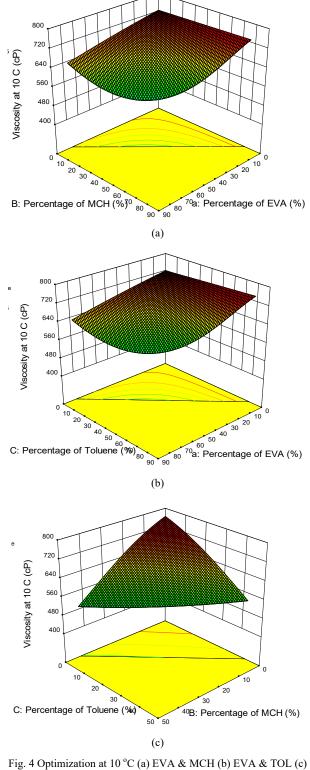


Fig. 3 Optimization at 15 $^{\rm o}{\rm C}$ (a) EVA & MCH (b) EVA & TOL (c) MCH & TOL



MCH & TOL

Fig. 4 shows that maximum viscosity reduction at 10 $^{\circ}$ C is when EVA and MCH are increased (Fig. 4 (a)), EVA and TOL is increased (Fig. 4 (b)) and when TOL and MCH is

decreased (Fig. 4 (c)).

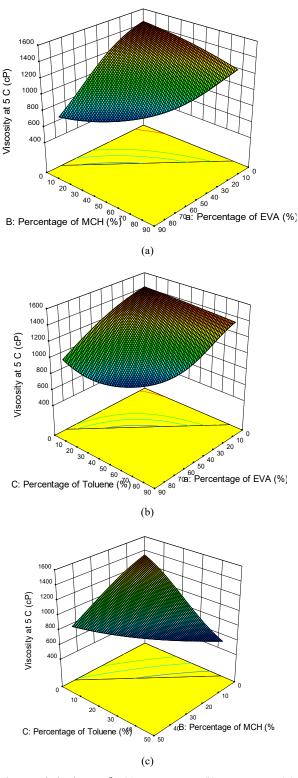


Fig. 5 Optimization at 5 °C (a) EVA & MCH (b) EVA & TOL (C) MCH & TOL

Fig. 5 shows that maximum viscosity reduction at 5°C is

when EVA increased and MCH is decreased (Fig. 5 (a)), EVA and TOL is increased (Fig. 5 (b)) and when TOL is increased and MCH is decreased (Fig. 5 (c)).

From the ANOVA analysis, the equation for optimization of the percentage composition of the inhibitor is

$$\begin{split} & \text{Viscosity at } 20 \ ^{\circ}\text{C} = -0.40547 + (0.028596 \times \text{A}) + \\ & (0.017732)(\text{B}) + (7.67329 \times 10^{-3})(\text{C}) - (1.20439 \times 10^{-3})(\text{AB}) - (1.16560 \times 10^{-3})(\text{AC}) + (3.05883 \times 10^{-4})(\text{BC}) \\ & (1) \end{split}$$

Viscosity at 15 °C = $1.79176 + (4.13023 \times 10^{-3})(A) + (4.13253 \times 10^{-3})(B) + (1.65278 \times 10^{-3})(C) - (1.35407 \times 10^{-3})(AB) - (6.99663 \times 10^{-4})(AC) - (2.92014 \times 10^{-4})(BC)$ (2)

Viscosity at 10 °C = $1.38629 + (0.012024)(A) + (8.43718 \times 10^{-3})(B) + (5.56466 \times 10^{-3})(C) - (1.26399 \times 10^{-3})(AB) - (8.83482 \times 10^{-4})(AC) - (1.91861 \times 10^{-4})(BC) (3)$

Viscosity at 5 °C = $2.63906 - (0.014781)(A) - (7.91329 \times 10^{-3})(B) - (6.10910 \times 10^{-5})(C) - (9.23388 \times 10^{-4})(AB) - (1.35198 \times 10^{-3})(AC) - (4.82119 \times 10^{-4})(BC)$ (4)

where, A is % of EVA; B is % of MCH and C is % of TOL

The R^2 value of (1)-(3) is at 0.84, 0.83, and 0.60 respectively, which means that the fitting of the line which is on data is not very accurate. The adjusted R^2 value is 0.80, 0.79, and 0.49 respectively, which shows that the added terms further decrease the precision of the fitting. However, the R^2 value of equation 4 is at 0.91 which means that the fitting of the line which is on data is accurate. The adjusted R^2 value is 0.89 which shows that the added terms further decrease the precision of the fitting but is still within accepted accuracy. This could be because of the larger range of values for the response which makes the optimization less accurate.

The p-values in these model equations are all low, below 0.05 for this response which makes the model significant. The significance of the model at this point is important to evaluate the model as the temperature lies below the WAT for the blank crude oil without inhibitor. Thus, the priority to optimize the effect of the inhibitor at this point is greater.

D. RSM Point Prediction

From the ANOVA analysis, it can be concluded that the RSM modelling was significant and the point prediction of optimum concentration percentage is 57% EVA, 20.5% MCH, and 22.5% TOL which can give a maximum reduction of up to 61%. Table III represents the comparison of the viscosity reduction results.

TABLE II Comparison of the Viscosity Reduction										
Temperature (°C)	40	35	30	25	20	15	10	5		
Viscosity of blank crude oil (cP)	160	160	160	240	320	640	720	1440		
Viscosity of crude oil with 57% EVA + 20.5% MCH + 22.5% TOL (cP)	160	160	165	229	265	386	497	558		

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IV. CONCLUSION

In conclusion, the objective of this paper was achieved in which it was proven that the combination of EVA, MCH, and TOL can make up a wax and asphaltene inhibitor which can reduce the viscosity of crude oil. The percentage composition was determined experimentally to be optimum at 50% EVA, 25% MCH, and 25% TOL. The optimization of the data obtained experimentally proved the optimum percentage composition to be at 57% EVA, 20.5% MCH and 22.5% TOL. The values determined experimentally and through RSM are similar, which further strengthens the value obtained. The viscosity reduction, through experimental procedure, was up to a maximum of 67%, while the maximum viscosity reduction of the crude oil by RSM optimization was up to 61%. The WAT was reduced up to 10 °C from 20 °C of the blank crude oil to 10 °C of the crude oil containing inhibitor at the optimum composition which is 50% EVA + 25% MCH + 25% TOL. Therefore, the inhibitor containing wax inhibitor, EVA, wax inhibitor solvent, MCH and asphaltene solvent, TOL have been proven to be an effective flow properties improver for Sabah crude oil.

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