



Comparative Study on Inhibitors Comprising Aromatic and Non-Aromatic Solvents towards Flow Assurance of Crude Oil

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ABSTRACT

The petroleum industry is facing a critical issue in transporting crude oil through the pipelines from the seashore where crude oil is being drilled off. The problem arises when crude oil exhibits higher sensitivity to the changes of temperature. This actually causes some alterations occurring in the composition, pour point of the oil and flow of the crude oil itself. Thickening of some components such as wax and asphaltenes causes the deposition to occur in the pipelines due to changes in temperature. Eventually, these depositions cause blockage of the pipelines due to reduction in the diameter of the pipelines and causing disruption in the flow of crude oil. The experiments were carried by mixing different ratio of polymer and solvent such as ethylene-vinyl acetate (EVA40) with 40% vinyl acetate, methylcyclohexane (MCH), toluene and butanol together to form an inhibitor. The response surface methodology (RSM) had been used to identify the best formulation of solvents that could act as inhibitors. The final results show that the most optimum ratio of inhibitor that gives the highest reduction in viscosity of the crude oil is 30% EVA, 30% MCH and finally 40% ratio of solvent which is either toluene or butanol.

Keywords: Asphaltene, crude oil, deposition, inhibitors, response surface methodology, wax

ARTICLE INFO

Article history:

Received: 26 April 2019

Accepted: 22 July 2019

Published: 13 January 2020

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INTRODUCTION

Crude oil is one of the components in petroleum that is very important to be commercially sold over the world and which is known as non-renewable energy. Crude oil is an unrefined product that is obtained from the exploration process and it varies from light to heavy crude oils based on molecular weight (Aldahik et al.,

2017). It is composed of hydrogen and carbons. The major petroleum constituents from crude oil that can be used for almost all types of manufacturing industries such as fuel for transporting, rubber industries, and leather industries (Aldahik et al., 2017; Amghizar et al., 2017; Sharma, 2006).

There are many problems arising during the transportation of crude oil and the main concern or issue is that the solid deposition in the pipelines (Kriz & Andersen, 2005). Solid deposition here is known as waxes and asphaltene components. Due to this phenomenon, the production lines can be clogged and the production of oil can be effected. The highest molecular weight of n-paraffin or known as n-alkane with hydrocarbon bonding is one of the main components in the wax formation in crude oil (Kriz & Andersen, 2005). Asphaltenes are found in the crude oil and they are known as high molecular polyaromates or called as resin too sometimes (Adebiyi & Thoss, 2014; Wilt et al., 1998).

Although wax and asphaltenes act as structural stabilizer during formation of crude oil, at a certain temperature along the transportation can cause these to turn into large clumps which lead to the deposition process. High deposition of wax and asphaltene causes the blockage in the pipeline due to reduction of space or diameter. As a result, the cost of maintenance of the industries increases gradually as the cost to improve the pipeline is too high and need to be maintained frequently. Thus, an effective method is devised to prevent any issues found from the deposition along the pipelines by improving the flow properties of crude oil (Anisuzzaman et al., 2017a; Anisuzzaman et al., 2017b; Tinsley et al., 2009).

There are many methods that have been introduced in previous studies to reduce the deposition of the wax. One of the traditional methods that had been practiced in the new research was by using acrylate polymer, acting as pour point depressant (PPD) which was a solid form polymer (Admiral et al., 2016). Although there are many traditional methods to remove wax precipitation, the most frequent method is the wax inhibitor. An inhibitor is being categorized as the modifier of the crystals, depressants of the pour point and inhibitors of paraffin followed by the flow improvers (Kriz & Andersen, 2005). A wax inhibitor can ensure that the growth of the wax stopped and able to prevent any wax blockage occur in the pipeline (Theyab & Diaz, 2016).

Asphaltenes are known as impure compounds as they consist of thousands different species with different molecular weights but similar behaviors chemically (Wei et al., 2016). As mentioned before, the best treatment is by using chemical inhibitors to reduce the blockage in the pipelines due to the asphaltene deposition. Thus, solvent treatment becomes the best treatment to treat this issue and the most common solvents that can be used as inhibitors are toluene, benzene and xylene which they are aromatic compounds. Asphaltenes have higher solubility in the non-polar solvent such as toluene and they are less soluble in the polar solvents such as glycerine, water, n-heptane and n-pentane compounds (Adebiyi & Thoss, 2014; Zhang et al., 2014). They are also insoluble in normal

alkane solvent. Moreover, asphaltenes always exist in suspended solid form or known as colloidal particles and the structures of the asphaltenes being interconnected by bridges that are formed from some aromatic compounds as sulphur or alkyl (Ghloum et al., 2010).

Treatment by using chemical solvent can save the cost as well as acts a preventive method for this critical issue. The treatment that has been frequently used is by direct injection process into asphaltene deposition and it is known as a physical-chemical process and chemical inhibitors. The chemical structures of the asphaltene strongly influence the deposition in the crude oil (Ghloum et al., 2010; Rogel, et al., 2001). Thus, it is very important in choosing the proper chemical inhibitors with accurate structural functional groups to treat asphaltene issues in petroleum pipelines so that this issue can be solved easily with less costing (Ridzuan et al., 2016).

Hence, overall purpose of this study is to differentiate the effect of the inhibitors that were formulated from different types of solvents which consisted of aromatic solvent and non-aromatic solvent towards to the solubility of wax and asphaltene. Moreover, response surface methodology (RSM) has been used to identify the best formulation of solvents that can act as inhibitors.

EXPERIMENTAL SECTION

Materials and Chemicals

The main chemical that was used in this experiment is the crude oil that was obtained from Sabah platform, Malaysia. Thus, the type of crude oil is known as Malaysian crude oil where they contain higher fractions of asphaltene compared to the wax fraction (Anisuzzaman et al., 2017a; Anisuzzaman et al., 2017b). Another chemical was the wax inhibitor which was ethylene vinyl acetate (EVA40) with 40% vinyl acetate. EVA is a type of polymer that can increase adhesion process to the surface of wax molecules. The EVA40 had been obtained from the Sigma Aldrich, USA. Another reagent used was the Methylcyclohexane (MCH) which could act as EVA solvent and this was due to EVA properties that would dissolve easily in MCH which had a higher boiling point, 100.4°C (Almeida et al., 2011). The most appropriate inhibitor for asphaltene is toluene but in this experiment, butanol was used together so that the comparison of the aromatic and non-aromatic compounds could be studied. These types of solvents were used to decrease the formation of crystals of asphaltene and increase the adhesion process to the surface of the asphaltene so that flocculation was prevented on the surface of the crude oil. The viscometer was used to obtain the viscosity of the crude oil samples that were being mixed with the different formulation of inhibitors. The viscometer used was the Brookfield Programmable Viscometer DV-III + Rheometer and the standard settings of viscometer such as spindle size and rotational speed was 63 and 100 rpm respectively. Binder oven was used to heat the crude oil pre-night until they reached 90°C before mixing with the inhibitors the next day. Water Bath

was used to decrease the temperature of the sample gradually. All the experiments were carried out at one atmospheric pressure.

Pre-experimental Preparation of Chemicals and Crude Oil

The pre-step for the experiment was that the crude oil was heated in the Binder oven at the temperature about 90°C for an overnight. This step was to melt any deposition of wax crystals that had been formed earlier and the structure of asphaltene being agglomerated in the crude oil. The preliminary step before this experiment was being conducted so that the precipitation of wax and asphaltene at the point of contact between the hot crude oil and cold apparatus could be avoided and to obtain accurate results. Before mixing of the inhibitor together EVA, MCH and toluene or butanol were heated in a water bath to increase the temperature around 50°C to 60°C (Anisuzzaman et al., 2017a; Anisuzzaman et al., 2017b).

Preparation of Inhibitor

The inhibitors were prepared by conducting them on a hot plate at a temperature of about 90°C. Instead of using the oven, using hot plate can help to save time and space for the experiment to be conducted as the limitation of lab equipment in preparing the chemicals. The individual chemicals, EVA, MCH and Toluene were measured separately of its respective volume and weight in accordance with the manipulated percentage composition. The unit for the EVA is grams of mass while MCH is in mL and toluene and butanol are in mL. The total volume of inhibitor used was 0.4g. Thus, for example, if the percentage composition of the samples prepared are 50% EVA, 10% MCH and 40% Toluene, then 0.2 g EVA was measured using a mass balance, 0.04 mL of MCH was measured using a micropipette and 0.16 mL of Toluene was measured using a micropipette. The ratio of the EVA40 used was 30%, 40% and 50%.

The precaution step here was to replace the tube for the micropipette for each new sample that was taken so that contamination can be avoided and accuracy of the results obtained can be improvised. Care was taken to replace the micropipette tube for both chemicals to avoid contamination. The purpose of the reaction to be in high temperature was around 50°C which is to ensure that the EVA pellets are completely melted in the inhibitors and to develop the influence of inhibitors on formed wax crystals (Theyab & Diaz, 2016). After complete melting of EVA, then the crude oil that was placed in the oven overnight was mixed with the EVA and the inhibitor solutions. If the crude oil managed to be heated overnight, then complete dissolved wax crystal was obtained on the next day. Later, the samples obtained were shaken around 30 seconds by magnetic stirrer. This was to ensure the crude oil and the inhibitors completely mixed. Then, the samples are placed again in the oven for 15 minutes to allow the reaction to take place.

Experimental Procedures

The viscosity of the samples was taken from the temperature range from 5°C and 20°C. The control sample in this experiment was blank crude oil that was free from inhibitors. The viscosity of the blank crude oil was measured at earlier stage. The procedure was repeated for each of the other samples that contain the different formulation of inhibitors with different ratio of the composition of solvents and polymers.

Optimization of Experimental Design of Aromatic Inhibitor by using Response Surface Methodology (RSM)

RSM was used to optimize the ratio of each chemical and to test the optimized ratio of formulation inhibitor on the wax and asphaltene solubility. Table 1 shows the design summary of the model for the formulation of inhibitors consists of toluene.

Table 1
The summary of the design model

File Version				Design Expert 7.0					
Study type				Mixture					
Design type				D-optimal					
Design model				Quadratic					
Subtype				Split-plot					
Runs				36					
Factor	Name	Units	Type	Minimum	Maximum	Coded Values			
A	EVA	%	Mixture	20	60	0.00			
B	MCH	%	Mixture	20	60	0.00			
C	Toluene	%	Mixture	10	40	0.00			
Response	Name	Units	Observed value	Analysis	Minimum	Maximum	Std. Dev.	Ratio	Model
R1	Viscosity	cP	36	Polynomial	72	755	181.99	10.49	Special cubic

Based on the Table 1, the study type that had been used was the mixture as there were three different types of chemicals to form a specific inhibitor. The total ratio of chemicals used was 100%. From the Table 1, it shows that that the total runs or samples that we used were 36 samples at different temperature which was around 5°C to 20°C. The parameter that acted as a response was the viscosity of crude oil.

RESULTS AND DISCUSSION

Characterization of Crude Oil

Fourier transform infrared spectroscopy (FTIR) analysis was used as an analytical method to determine the presence of asphaltene content in crude oils qualitatively. The results FTIR analysis of blank crude oil is shown in the Figure 1.

The results of spectrum obtained from FTIR analysis was evaluated by comparing with the literature review that had done in analyzing the asphaltene contents in crude oil using the FTIR (Table 2) (Wilt et al., 1998). Based on the results, there are some similarities on the characteristic of spectrum retrieved from FTIR spectroscopy analysis between the blank crude oil used for the experiment and blank crude oil used in the literature review. The spectrum of FTIR analysis in Figure 1 had been compared with the result of evaluation of the crude oil obtained from existing research sources and confirmed that there was presence of asphaltenes in the Sabah blank crude oil used in this study (Wilt et al., 1998). The peak at 1607.15 cm^{-1} in the Figure 1 shows that the results of FTIR analysis corresponded to the aromatic C=C stretching vibrations. Besides, the absorbance at 1456.07 cm^{-1} shown predominantly is due to its CH_2 bending modes and a part of CH_3 bending modes. Methyl bending vibrations are the main reason for the peak at 1377 cm^{-1} to be occurred in this analysis. The ester linkages present in the asphaltene molecule as indicated by the peak

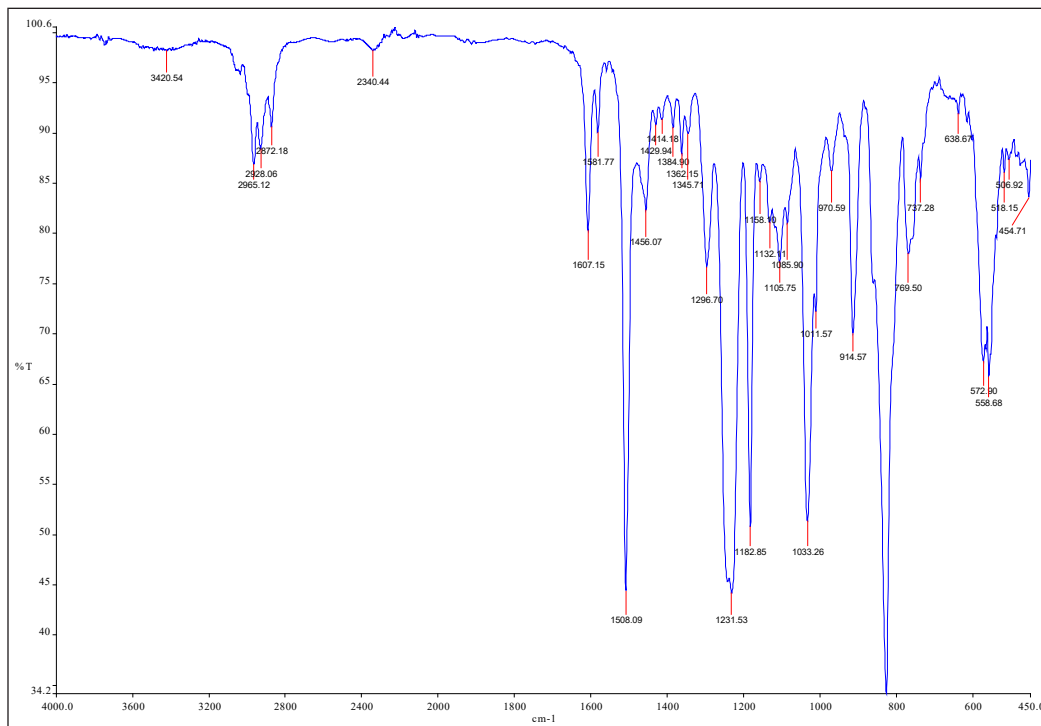


Figure 1. Illustration of blank crude oil FTIR analysis

at 1033.26 cm^{-1} . The three absorbances observed between 870 and 722 cm^{-1} can be the indicator to the aromatic CH out-of-plane bending vibrations.

Table 2
The peaks values of two different sources

Peaks showing presence of asphaltenes, cm^{-1} (Wilt et al., 1998)	Peaks in the FTIR results of Sabah blank crude oil, cm^{-1}	Peak assignment
1602	1607.15	C=O
1032	1033.26	C-O-C
812	819	C-H
752	769	C-H

The spectrum of FTIR analysis also proved that the blank crude oil contained wax as one of the components. Wax paraffin is known as hydrocarbons with the C-H and C-C bonding. Once wax paraffin become saturated hydrocarbons, then their C-H stretching peaks in FTIR will be around 1377 to 1461 cm^{-1} . Thus based on the blank crude oil used in the experiment, the paraffin wax is proven to be saturated (Khan et al., 2017).

Viscosity of Blank Crude Oil

The viscosity of the control sample of the experiment was the blank crude oil as shown as in Figure 2. These samples viscosities were measured at different temperatures in range around 5°C to 20°C .

From the Figure 2, the results show that the viscosity of the crude oil decreases as the temperature of the crude oil increases.

The reduction of the viscosity using aromatic solvent (toluene) is shown in Figure 3.

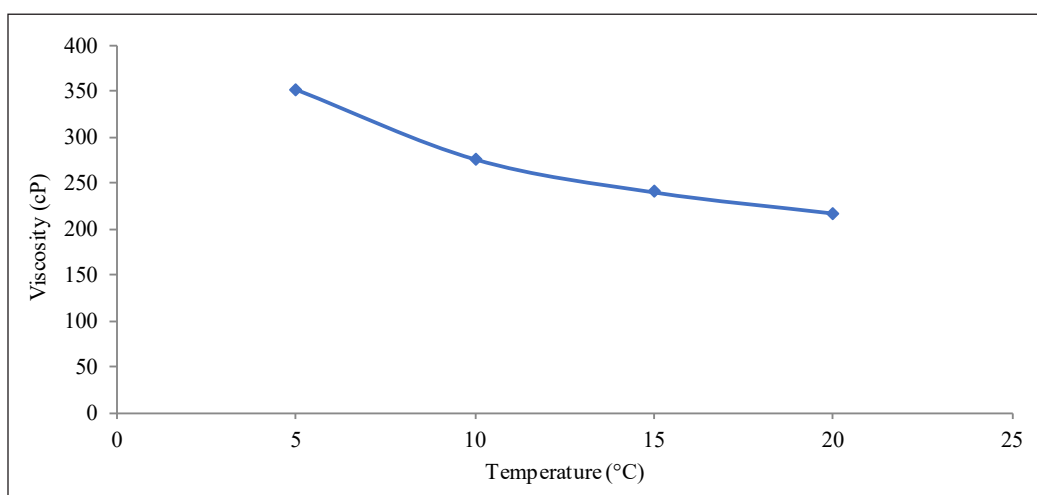


Figure 2. Viscosity of the blank crude oil against temperature

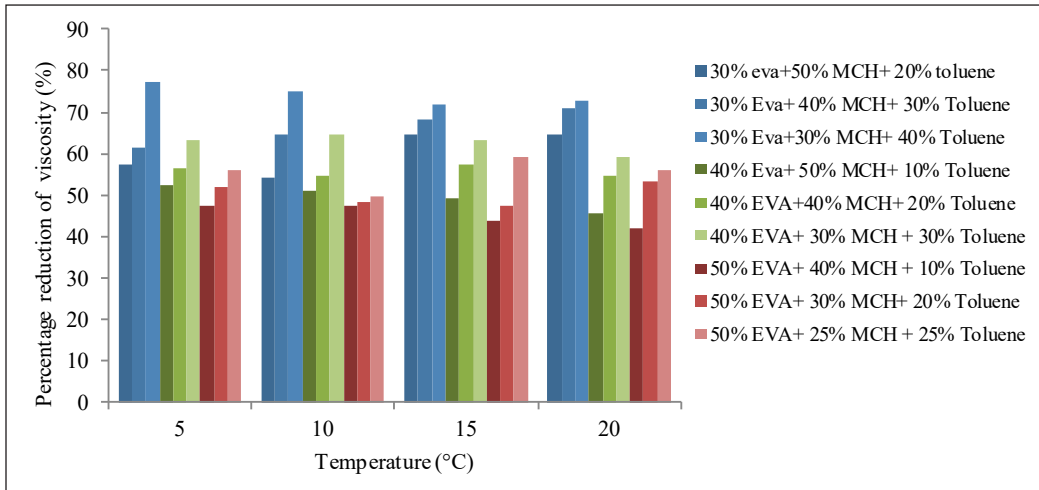


Figure 3. Percentage reduction in viscosity against temperature using aromatic solvent

The results show that higher the percentage of EVA in the solvent used higher the viscosity of the crude oil. In addition to that, it also shows that at higher temperature, the viscosity was too low. Increasing the amount of EVA eventually caused increase in viscosity of crude oil (Anisuzzaman et al., 2017a; Anisuzzaman et al., 2017b; Quan et al., 2016). Thus, an optimum amount of EVA is required to act as an inhibitor for wax. This is because the solubility of asphaltene in the presence of toluene is actually a balance between the π - π stacking of aromatic ring systems from the toluene and the repulsion which comes in steric form that comes from the alkane chains, which tends to increase the solubility. Thus, it helps to delay the deposition or aggregation of asphaltene in the crude oil (Zarkar et al., 2015).

The reduction of the viscosity using non-aromatic (butanol) is illustrated in Figure 4.

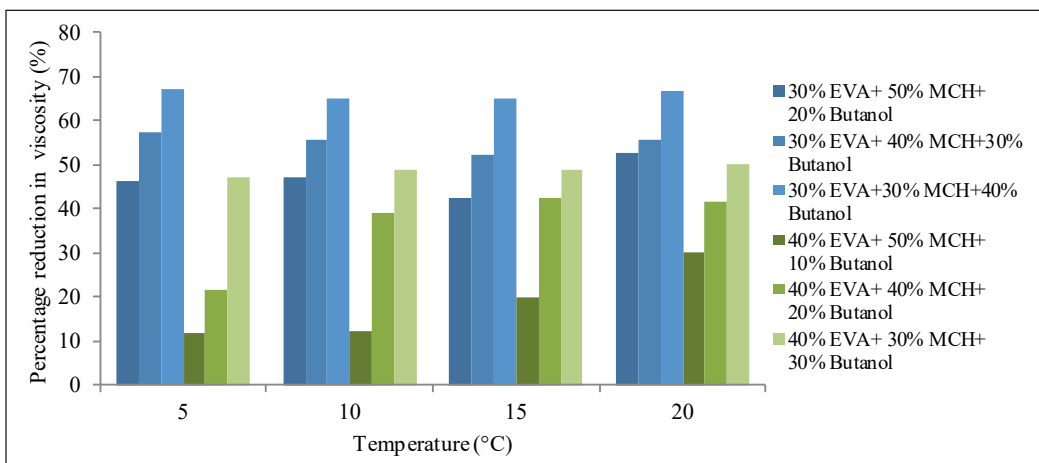


Figure 4. Percentage reduction in viscosity against temperature using non-aromatic solvent

A clear observation was obtained where the viscosity of the crude oil becomes greater than the blank crude oil when the fixed percentage ratio of EVA is the highest which is at 50%. In this situation, there is a significant change where the crude oil was assumed to become thicker and hard to flow with higher percentage of EVA and butanol. As it reaches a saturation point where the level of EVA and butanol in crude oils is maximum, the crude oil becomes thicker and more viscous because there is higher agglomeration of EVA and butanol. Thus, butanol which is the solvent that could not coagulate the EVA sufficiently and it tended to spread over the surface of coagulant causing deposition to occur (Matsumoto et al., 1974). Hence, the usage of non-aromatic compound, butanol is less efficient to aid the wax and asphaltene solubility in the crude oil and to improve the flow assurance of crude oil in pipelines.

Optimization Experimental Design of Aromatic Inhibitor by using RSM

ANOVA analysis was used to determine the regression model equations for this mixture design of these combinations of three chemicals (Bono et al., 2008; Bono et al., 2014). For the ANOVA analysis, the model used for the mix-design model is Scheffe Mix Model as this model is typically used to handle any natural constraints that occur in the mixture design. However, the mix order that we decided to use here was the special cubic as there would be around three different components blend together to form an inhibitor. The equation for the actual responses for the viscosity of the crude oil with a constant of 100 rpm is expressed as shown in equation 1.

$$\begin{aligned} \text{Viscosity (cP)} &= +2.94605A + 1.25404B - 1.09010C - 0.021691AB - 0.02986AC \\ &= +2.94605A + 1.25404B - 1.09010C - 0.021691AB - 0.02986AC \end{aligned} \quad (1)$$

wherein the equation above each term represented as:

A is the ratio of percentage of EVA, B is the ratio of percentage of MCH and C represent the ratio of the toluene.

Statistical Analysis of the Design Model of Aromatic Solvent

Statistical analysis of experiments depends on the ANOVA analysis. The ANOVA analysis can be done by studying the values of probability (p), coefficient of determination (R^2), and adjusted R^2 . The analysis was tabulated as shown in Table 3 based on ANOVA variance analysis.

Table 3 shows that the 'Model F' and value of p are 4.90 and 0.0014 respectively. These values clearly show that the model is significant. Besides, the model terms are significant as the values of "Prob > F" obtained is less than 0.05. From the analysis, predicted R^2 is

actually in a reasonable agreement with the adjusted R^2 . As “Adequate precision” measures the signal to noise ratio, any ratio greater than 4 is desirable. Signal to noise ratio is the interference occurs due any electrical strength as computer signal. Based on the analysis, the obtained ratio is 7.082 which show that it is an adequate signal. R^2 in this model is 0.5037 which indicates that there is about 50% effect that is given to the input variable by the variation output variable.

Table 3
Statistical analysis of variance (ANOVA) for the viscosity using aromatic solvent

Source	Sum of squares	DF	Mean square	F-value	P-value Prob > F	
Model	19004.59	6	3167.43	4.90	0.0014	Significant
Linear mixture	18859.14	2	9429.57	14.60	<0.0001	
AB	3.65	1	3.65	5.659x10 ³	0.9405	
AC	0.70	1	0.70	1.09x10 ³	0.9739	
BC	3.33	1	3.33	5.15x10 ³	0.9432	
ABC	1.06	1	1.06	1.638x10 ³	0.9680	
Residual	18729.05	29	645.83			
Cor Total	37733.64	35				
Std. Deviation = 25.41		Mean = 114.31		Adequate precision = 7.082		
$R^2 = 0.5037$		Adj. $R^2 = 0.4010$				

Interactions between each Component

Each interaction between the three main components was studied. Figure 5 shows the interactions between each component in three-dimensional group based on ANOVA.

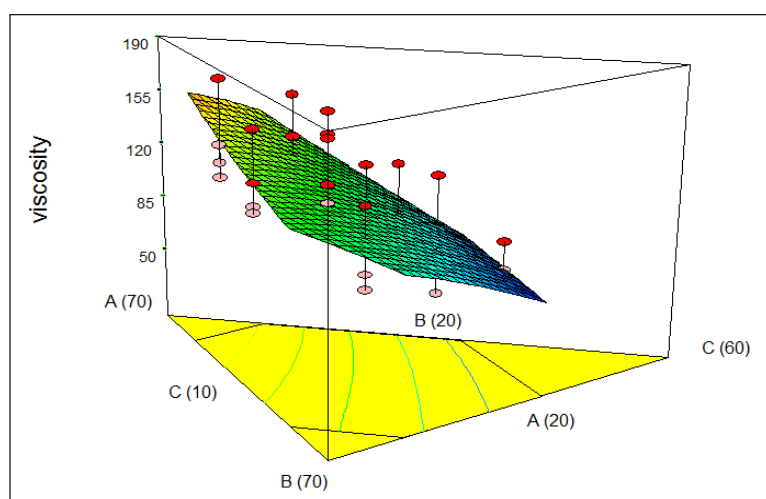


Figure 5. 3D illustration on the interaction between each component using aromatic solvent

Based on Figure 5, each component had been labelled respectively in alphabetical orders such as A represents the percent ratio of EVA, B represents the percent ratio of MCH and last but not least C was the term to describe the toluene ratio in forming an inhibitor. The most effective interaction was between AC where at this region, the lowest viscosity was obtained. This observation clearly proves that the EVA is the wax inhibitor while the toluene acts as asphaltene inhibitor (Wei et al., 2016).

Optimized Results for the Aromatic Compound

The objective of this study is to optimize the ratio of chemicals used and the temperature by using the RSM. Thus, the optimum value for each component is tabulated in Table 4.

Table 4
Optimized result for aromatic solvent

Type of components	Goal to achieve	Optimized value
EVA (%)	In range	30.00
MCH (%)	In range	30.48
Toluene (%)	In range	39.51
Viscosity of crude oil (cP)	Minimize	69.98
Desirability		1.00

Table 4 shows the optimized viscosity of the crude oil obtained is 69.98. In conclusion, addition of the inhibitors can cause reduction in viscosity of the crude oil and caused the flow assurance of crude oil in the pipelines becomes more effective. As the desirability is 1, the optimized ratio for the formulation of inhibitor might give a great effect on the viscosity of the crude oil.

Optimization Experimental Design of Non-Aromatic Inhibitor by using RSM

As mentioned before in aromatic compound design model, the type of model used here was Scheffe Mix model as this model helped to readjust back the natural constraints compared to Slack Model. The mix model used here is special cubic where there are three components used in this design to form the best inhibitors. The equation for the actual responses for the viscosity of the crude oil with a constant of 100 rpm was defined as shown in equation 2.

$$\begin{aligned} \text{Viscosity (cP)} = & 25.71679A + 8.92852B - 60.42534C - 0.449041AB + 1.77294AC \\ & + 1.75675BC - 0.072701ABC \end{aligned} \quad (2)$$

wherein the equation above each term represented as:

A is the ratio of percentage of EVA, B is the ratio of percentage of MCH and C represent the ratio of the toluene.

Statistical Analysis of the Design Model of Non-Aromatic Inhibitor

Table 5 shows results obtained for the inhibitors that consist of non-aromatic compound.

Table 5
Statistical analysis of variance (ANOVA) for the viscosity using non-aromatic solvent

Source	Sum of squares	DF	Mean square	F-value	P-value Prob > F	
Model	9.92×10^5	6	1.665×10^5	25.0	0.0038	Significant
Linear mixture	7.489×10^5	2	3.774×10^5	56.21	<0.0001	
AB	21454.37	1	21454.37	3.22	0.0831	
AC	1038.81	1	1038.81	0.16	0.6958	
BC	2205.68	1	2205.68	0.33	0.5694	
ABC	13904.29	1	13904.29	2.09	0.1592	
Residual	1.932×10^5	29	6661.06	5.54		
Cor Total	1.192×10^6	35				
Std. Deviation = 81.62		Mean = 254.67		Adequate precision = 14.664		
$R^2 = 0.8380$			Adj. $R^2 = 0.8045$			

From the Table 5, the P-value is lower than 0.05, proving that the model is significant. It can be seen that the larger the standard deviation, the accuracy of the results from the mean of the result is low.

Interactions between Components

Interaction between each component in this mixture had been explained with the Figure 6.

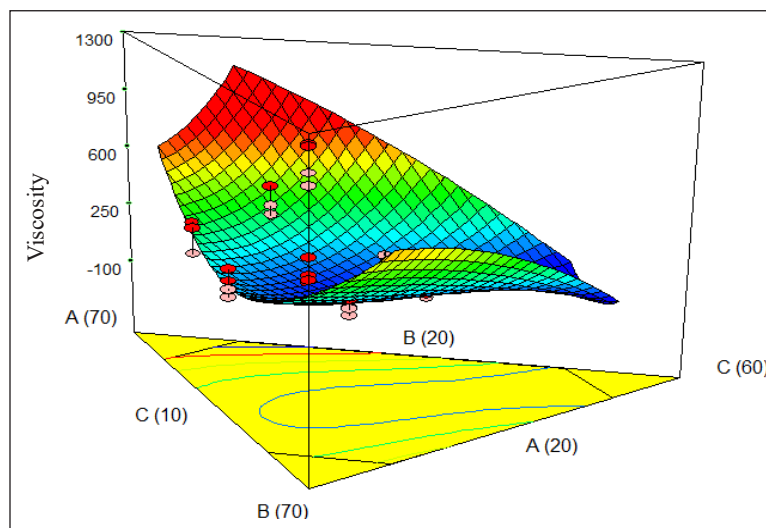


Figure 6. 3D illustration on the interaction between each component using non-aromatic solvent

As shown in Figure 6, the interaction between A which is the EVA and C, butanol (AC) did give significant effect on the viscosity of crude oil. This is due to the higher point of A (EVA) on the plane, the higher the viscosity. However, the higher the point of C (butanol), the viscosity decreases.

Optimized Results for the Non-Aromatic Compound

After analyzing the ANOVA of the design model, the optimized data for each component is shown in Table 6.

Table 6
Optimized result for non-aromatic solvent

Type of components	Goal to achieve	Optimized value
EVA (%)	In range	30.00
MCH (%)	In range	30.00
Butanol (%)	In range	40.00
Viscosity of crude oil (cP)	Minimize	77.00
Desirability		0.993

Based on the Table 6, desirability of the optimized model result is just 0.993 which means that there is lacking of “quality” in the component process data thus this optimized ratio for this formulation of inhibitors might be less efficient in giving great reduction in viscosity. The optimized viscosity obtained for this formulation of inhibitors using butanol is 77 cP.

Comparison between Aromatic Compound and Non-Aromatic Solvent

Comparison between these two solvents was done to identify the effect of two different solvents as inhibitors toward solubility of wax and asphaltene in the crude oil. The comparison was done based on percentage of reduction of viscosity for the optimised ratio of both inhibitors. Figure 7 shows the comparison in percentage reduction for this specified ratio of chemicals:

Figure 7 shows that the toluene which is aromatic solvent is the best inhibitor compared to non-aromatic solvent, butanol. This is due to the percentage reduction of viscosity of crude oil at 10°C with formulation of inhibitor is the highest which is 77% compared to butanol which was just 65%. Thus, it can be proven that aromatic solvent plays the effective role as the best inhibitor in improving the solubility of wax and asphaltene. Indirectly, it helps to increase flow assurance of crude oil in pipelines compared to non-aromatic solvent.

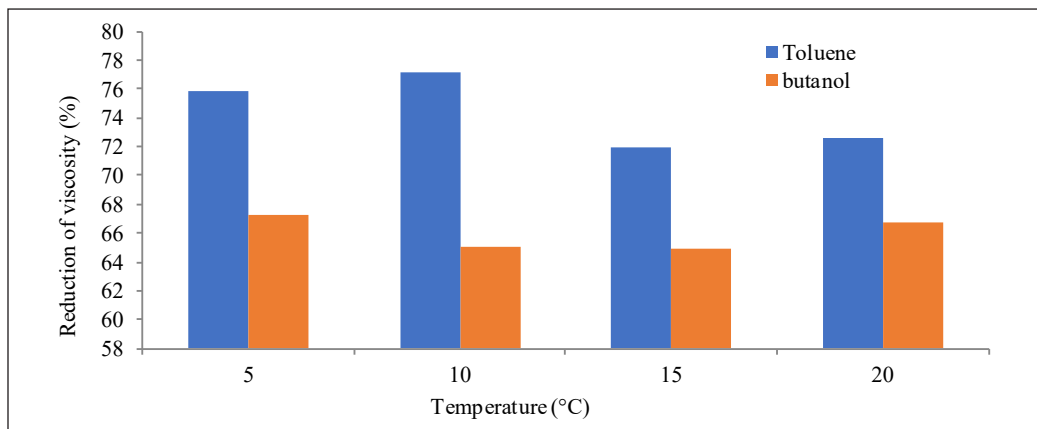


Figure 7. Percentage reduction of viscosity against temperature

CONCLUSION

In conclusion, both inhibitors consisting toluene and butanol showed effect on reduction in viscosity of the crude oil. Thus, it indicates that the aromatic solvent and non-aromatic solvent could act as inhibitors for the deposition of wax and asphaltene and could increase the solubility of wax and asphaltene in the crude oil. However, the level of certainty is that all non-aromatic solvent can reduce the viscosity of the crude oil but still at a very low level as this study was done with a specific type of solvent. From the optimization, the best inhibitor for reduction of viscosity can be formulated in the ratio of 30% EVA, 30% MCH and finally 40% of the solvent either toluene or butanol. The results show that the formulation of inhibitor using butanol as solvent gives higher percentage reduction in viscosity than formulation of inhibitor using toluene.

ACKNOWLEDGEMENTS

The authors would like to express their greatest appreciation to the Centre of Research and Innovation (PPI), Universiti Malaysia Sabah (UMS) for the support to this study (Grant No. SBK0346-2017).

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