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OPTIMIZATION OF OPERATING PARAMETERS USING RESPONSE SURFACE METHODOLOGY FOR PARAFFIN-WAX DEPOSITION IN PIPELINE

K. K. Salam^{1*}, A. O. Arinkoola¹, E. O. Oke¹, J. O. Adeleye²

¹Petroleum Engineering Unit, Department of Chemical Engineering, P.M.B. 4000, Ladoke Akintola University of Technology (LAUTECH), Ogbomoso. Nigeria ²Dept. of Petroleum Engineering, University of Mines and Technology, Tarkwa. Ghana *Corresponding Author: kaykaysalam@yahoo.co.uk

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Abstract

The influence of operating parameters on weight of wax deposit in oil-pipeline was investigated in this study. Response surface methodology (RSM) was used to develop polynomial regression model and investigate the effect changes in the level of wax temperature differential, flowrate and residence time on weight of wax deposit using Box Behnken design. The contribution of Flowrate, time,square of significant to the model developed temperature, square of time and interaction between flowrate and time (B, C,

 A^2 , C^2 , BC) are. it was observed that the experimental data fitted better because of the Pred R-Squared of 0.9618 is in reasonable agreement with the Adj R-Squared of 0.9945. The agreement between the predicted and experimental values describe the accuracy of the model developed and can be used to navigate within the design space. The minimum value of wax deposit of 0.0195075 was achieved at Temperature, Flowrate and Residence time values as 53.25°C, 499.54ml/min and 3.01min respectively.

Keyword: RSM; residence time; wax deposit; flow rate; temperature differential; optimization.

1. Introduction

Wax deposition is one of the problems encountered in the Petroleum Industry which can be attributed to decrease in pressure and/or temperature. Decrease in Pressure cause loss of light and components which serves as natural solvents for the waxes and decreased temperature which affect solubility of wax in crude oil ^[1]. Wax deposition leads to increased in viscosity and pipeline roughness, reduction in effective area which all result in increased in pressure drop and in extreme cases it leads to blocking of flow line that either increased overhead cost or abandonment of facilities ^[2].Techniques like thermal treating of pipeline, addition of chemical inhibitor and pigging are commonly used to prevent wax accumulation. Cost associated with remediation could be drastically reduced if accurate means to predict wax precipitation region in available ^[2]. Deposition modeling has become an important method to ensure efficiency in utilizing remediation techniques ^[3]. Different modeling approaches will be reviewed.

Mehrotra and Bhat ^[4] developed a mathematical model used to predict solid deposition from multicomponent paraffin waxy mixture under turbulent flow condition. They made use of a moving boundary problem formulation in which the deposit formation and growth are modeled as a heat transfer process that incorporated effect of shear stress and deposition aging through a cubical cage deformation approach. Effect of shear stress was introduced through viscoplastic model to account for the time-dependent variation in the deposit composition. The model was solved numerically and the result of ate numerical analysis show that deposit thickness increases with lower inlet temperature, lower pipe wall temperature, mixture wax concentration and low Reynolds number. The combined effect of shear stress and aging (deposition time) caused wax deposit content to increase together with a enrichment of heavy alkanes and correspond depletion of higher n-alkanes.

In order to enhance understating of wax deposit, Tabatabaei-Nejaa and Khodapanah used probability model to characterized the plus fraction as a series of pseudo components with continuous molecular weight. Two different single carbon number, C_7 and C_{10+} were chosen and distributed to multicomponent of fine, six, and/or ten using continuous method. Different wax data published for volatile oil, gas condensate, aromatic, paraffin and waxy oil were used to validate the proposed model. The result show that the model show superior accuracy with validated data and two published models used for comparison studies ^[2].

Banki *et al.* ^[5] developed a mathematical model for the prediction of wax deposition in pipeline under laminar flow situation. The model was developed by coupling momentum, energy, species balanced equations and thermodynamic precipitation model. Molecular and thermal diffusion are represented from thermodynamics of irreversible processes. The model was solved using finite volume method. The result of their model show that molecular differs an thermal diffusion affect the flux towards the pipe wall by diffusion, both diffusion gradient are not proportional in the wax phase. The model is too complex to apply and made some assumption like neglecting effect shear dispersion and Brownian diffusion.

Huang *et al.* ^[3] developed a mathematical model to predict the increase in both deposit thickness and wax fraction of the deposit using a fundamental analysis of the heat and mass transfer for laminar and Turbulent flow conditions. The kinetic model accurately predicted the wax deposition for different conditions while the independent heat and mass transfer model and solubility model over-predicted and under-predicted the experimental result. The kinetic model accurately predicted the wax deposition for different was deposition for different while the independent heat and mass transfer model and solubility model over predict and under predict and under predict the experimental result. The kinetic model and solubility model over predict and under predict the experimental result. The kinetic model was tested under field condition and there was agreement in result generated. The kinetic model accurately predicted the growth of wax deposit and wax fraction in the deposit with wax precipitation parameter as adjustable parameter.

Mechanistic models have two noticeable shortcomings: Inability to accommodate all assumptions in their models and they are computationally demanding because they require the solution of coupled differential equations which resulted in formulation of simple or simplified model ^[6]. Bank *et al.* did not account for water phase, considered only molecular and diffusion parameters while Osokogwn and Otung didn't account for gel type behavior of wax ^[7]. Accuracy of these models are subject to effect of heat transfer coefficient, mass transfer coefficient, wax aging rates, industrial physical processing the wax and old which makes it difficult to analyzed the accuracy of individual assumption within the simulation ^[6]. The two shortcomings of Mechanistic models can be adequately taken care of simple or simplified model developed from experimental investigations where wax deposit will be describe with empirical or semi empirical term.

With the assistance of transparent experimental set up, Todi and Deo studied deposition phenomena in relation to particle transport at three types of fluxes. The result of the experimental studies show that shear dispersion position in the pipe section, and Brownian diffusion tends cause a uniform dispersion across the channel and particle concentration distribution depended on the competition between the two forces and deposition will occur as long as temperature is below NAT irrespective of the native of flux ^[8].

Edmonds et *al.* ^[6] developed a model for wax deposition treating wax phase as a continuous distribution of n-paraffin components. Shear was introduced into mass transfer model which was coupled with thermodynamic model in order to predict mass of wax deposited, the model was validated using experimental result from flow loop. The result from the model show that using compositional treatment given prediction of wax deposition profile in pipeline appear more physically realistic than simultaneous that are non-compositional, removal of wax accounted for through shearing and compatible with experimental investigation.

Kelechukwu *et al.* ^[9] experimentally studied effect of three variables temperature differential, flow rate and residence time of wax concentration percentage weight in the deposit. Series of experiment were conducted and results were presented which show that all the variables affect wax deposit weight. This research found out that the integration of these conditions could have significant influence of wax deposition. The investigated variables have significant effect on wax % in the deposit but there is no way the quantitative effect can be determined as the experimental result stood. The result of the experiment show that the amount of wax deposited decreased with increasing flow rate, the temperature difference between the waxy

fluid in let temp and subsea condition represented at the pipeline surface is considered important factor in controlling wax deposit. The amounts of wax deposited increase with time attain a maximum value and then gradually tailed off.

Johnson *et al.* ^[1] studied Wax deposition of coated and non-coated surface using a novel cold baffle wax deposition stirred reactor (COBWAD). They achieved this by investigating effect of different surface Treatment, heat flux and flow velocity/wall shear stress dependency, on wax deposition by adjusting flow velocity, temperature of both test surface and test fluid, surface Treatment of text session and duration of the experiment. The result of their investigation show that amount of deposit increased with bare steel and could either increase or decrease for coated surfaces, wax deposit increased with decrease in impeller velocity, influence of flow velocity decreased with increasing exposure to heat as deposit approached steady state and heat flux is affected by coated layers. They concluded that deposition rate has a direct relationship with wall flux, there exist a critical velocity/shear stress for maximum deposition.

Osokogwn and Otung ^[7] developed a semi-empirical model for wax precipitation and deposition along oil field installation under various condition of flow. Four various transport phenomena model molecular diffusion is shear dispersion, Brownian diffusion and gravity setting were included in the model developed. The result of the model was validated with three different published works on pressure and temperature profile, wall thickness and wall thickness generated through enthalpy-porosity approach. There was agreement between the output from the model developed when compared with published result. The model slightly under predict wall thickness in Banki *et al.* model which could be as a result of formation of a gel layer between the main fluid stream and the wax surface or could be as a result some assumption in Banki *et al.* model which was implemented in their work ^[5].

Experimental models either empirical of semi empirical are capable of predicting response within the range of experimental condition, therefore the use of assumptions will not arise but interaction of variables involved in the modeling might not be adequately quantified. Response Surface Modeling is the collection of mathematical and statistical technique that are useful for modeling and analysis in application where a response of interest is influenced by several variables ^[10-11]. It has been employed in various engineering problems to account for the influence of both main interaction variables and optimal use of available resources without compromising the quality of targeted objectives ^[12]. Modeling from experimental investigation is proposed by this study and A simple mathematic model which will consider the agreed factors that affects deposition of wax in oil facilities namely flowrate, temperature gradient and time will all be independent variables for a dependen variable wax deposit using experimental result presented by Kelechukwu *et al.* ^[9]. Our interest is to play around the experimental values with the aim of developing a relationship that can simulate the wax deposit.

RSM is a statistical technique for the design of experiments, building models, evaluating the effects of several factors and searching optimum factors for desirable responses. By using this technique, the interactions of possible influencing parameters on treatment efficiency can be evaluated and optimized with a limited number of planned experiments ^[13].

2. Materials and method

The solution of paraffin wax - kerosene system was prepared by dissolving into kerosene solvent (a 3:1 mixture of refined paraffin wax and kerosene oil), as a reconstituted wax of average Malaysian crude oil. A laboratory flow-loop system was designed and constructed to simulate wax deposition of paraffin wax in heat exchanger. The detail of the experimental procedure and results were reported in the work of Kelechukwu *et al.* ^[9].

3. Model Development

Response surface method (RSM) is a collection of mathematical and statistical techniques that are useful for modeling and analysis in applications where a response of interest is influenced by several variables and the aim is to optimize this response with limited number of experimental runs.

It was reported that temperature differential, flow rate and residence time significantly affect wax concentration percentage weight in the deposit ^[9]. After the identification of the important parameters, the experimental range was tabulated in Table 1.

Independent variables	Codod symbol	Level used		
	Coded Symbol	-1	0	1
Temperature differential (°C)	А	28	41.5	55
Flowrate (ml/min)	В	100	300	500
Residence time (min)	С	3	9	15

Table 1 Range of independent variables used for model development

A Box-Behnken statistical design with three factors and three levels was employed to fit second order polynomial model and seventeen experiments were required for this procedure. With the aid of Design-Expert software (version 6.0.3, Stat-Ease Inc., Minneapolis, USA), the data obtained from the design of experiment, as shown in Table 2, were used for the analysis and optimization

Std	Run	Factor A	factor B	Factor C	Std	Run	Factor A	factor B	Factor C
6	1	1	0	-1	10	10	0	1	-1
11	2	0	-1	1	7	11	-1	0	1
2	3	1	-1	0	1	12	-1	-1	0
9	4	0	-1	-1	4	13	1	1	0
8	5	1	0	1	12	14	0	1	1
17	6	0	0	0	3	15	-1	1	0
14	7	0	0	0	16	16	0	0	0
13	8	0	0	0	5	17	-1	0	-1
15	9	0	0	0					

Table 2 Combination sequence of the three factors

The regression parameters of the developed model and graphical interpretation for each response with statistical significance were calculated using Design-Expert software. The main and interactive relationship between the experimental variables and response were evaluated by generating response surface and contour plots. The response of the experiment is represented in the form of equation (1):

$$Y = \beta_0 + \beta_1 A + \beta_2 B + \beta_3 C + \beta_{11} A^2 + \beta_{22} B^2 + \beta_{33} C^2 + \beta_{12} A * B + \beta_{13} A * C + \beta_{23} B * C$$
(1)

where *Y* is the predicted response (wax deposit) used as a dependent variable; *k* the number of independent variables (factors), *x*i (*i* = 1, 2) the input predictors or controlling variables (factors); β_0 as the constant coefficient, and β_i , β_{ij} and β_{ii} the coefficients of main, interaction and quadratic term, respectively. The coefficient parameters were estimated using a multiple linear regression analysis employing the Design-Expert software. The software was also used to find the 3-D surface and 2-D contour plots of the response models.

4. Result and discussions

4. 1 Development of Regression Model Equation for the Wax deposited/Model Fitting

Figure 1(i-iii) shows individual behavior of the three factors used for the prediction of wax deposit in the pipeline. Figure 1(i) show that at constant flow rate of 300ml/min and time of 9mins; the relationship between the wax deposit and temperature was a concave relationship. There was an increase in wax deposit from 0.115635 to 0.33922 when the temperature increased from 28° C to 41.5° C. Further increase in temperature caused the wax deposit to decrease which led to wax deposit of 0.111685% at 55° C.

Figure 1(ii) show that wax deposition has an inverse relationship with flow rate, increase in flow rate from 100 to 500ml/min led to decrease in wax deposit from 0.427177 to 0.25084 at constant temperature of 41.5oC and time of 9minutes. Shown in Figure 1(iii) was the relationship between wax deposit and time at constant temperature of 41.5oC and flowrate of 300ml/min. There was an increase in deposit from 0.238422 to 0.33922 when the time

varied from 3mins to 9mins and after 9mins, the wax deposit value dropped to 0.257897 at the end of 15mins of the experimental setup.

The effect of flowrate at low and high values (100 & 500ml/min) with increase in temperature from 28-55°C at constant time. It was observed that at the same temperature the wax deposit decreased from 0.208932 to 0.03 when flow rate was increased from 100 to 500ml/min, and increasing value of temperature, the flow rates increased at both low and high values till a maximum value of wax deposit of 0.40606 at 100ml/min and 0.247305 at 500ml/min and at the end of the experimental setup the value was 0.194302 at 100ml/min and 0.028648 at 500ml/min. The percentage increment of wax deposit at 28oC was 85.6% with reference to 100ml/min at a temperature of 41.5°C, we have 39.9% and 85.3% reduction wax deposit when flowrate was increased from 100ml/min to 500ml/min of 55°C.



Figure 1 One Factor interaction of the variables on was deposited

Figure 2(ii) show the simultaneous increase in the values of time and temperature on wax deposit at constant flow rates of 300ml/min. It was observed that increase in time of reaction from 3 to 15mins led to an increase in deposition rate from 0.0164975 to 0.0326525 at 28°C, at a temperature of 41.5°C, the values increased from 0.22528 to 0.24637 when the time was increased from 3 to 15mins. Beyond a temperature of 41.5°C, the graph dropped at a final wax deposit of 0.0092275 and 0.0320225 at time 3mins and 15mins respectively.

Figure 2(iii) show the combine effects of time and flowrate on wax deposit at a temperature of 41.5°C. It was observed that an increase in flowrate from 100 to 500ml/min led to a decrease in wax deposit values at low and high time. The values of wax deposit was increased from 0.28811 at 3mins to 0.39571 at 15mins at a constant flowrate of 100ml/min and linearly

decreased with increase in flowrate to a value of 0.188315 at 3mins and 0.13125 at 15mins at a flowrate of 500ml/min. It was noticed that at high flowrate, a high value of wax was experienced at low time of 3mins and lower value of wax deposit at a high time of 15mins.



Figure 2 Two Factor interactions of variables on wax deposited



Figure 3 Surface plot of interaction between flowrate and temperature on wax deposited.



Figure 4 Surface plot of interaction between time and temperature on wax deposited.



Figure 5 Surface plot of interaction between time and flowrate on wax deposited.

Figure 3 described the behavior of A and B and constant C. It was shown that wax deposit decreased with increase flowrate and gave an highest value at 230ml/min while an increase in temperature increases the value of wax deposit till 41.5°C after which there will be reduction in the value. Similar trend was observed when the interaction between the factors was considered.

Surface interaction of A and C at constant B was shown in Figure 4. The figure show that an increase in C leads to a corresponding increase in the wax deposit to a value of 0.292091

at 41.5°C before the value of wax deposit decrease to a value of 0.0894608 at 55°C. The trend is similar to the behavior of interaction coefficient.

Figure 5 show the interaction of B and C at constant A. The surface plot show that the highest value of 0.359634 wax deposited was experienced at 246ml/min and 9mins; also the deviation from the two points will either increase or decrease the wax deposit. If the flow rate and time was increased beyond the above values, there will be reduction in wax deposit and in decrease beyond the value; there would be an increase in wax deposit value in the pipe.

4.2 Interaction of variables on wax deposition

The complete design matrix for 17 experimental run, including the coded factors of variables together with the results obtained for wax deposit was used in determination of all constants in equation 1 above. The quadratic model was selected by the software for the response which was the mass of wax deposited. Multiple regression analysis was used to correlate the responses of the three variables studied using a second order polynomial as shown in Eq. (2).

$$Y = 0.34 - 1.975E - 003 * A - 0.088 * B + 9.737E - 003 * C - 0.23 * A^{2} - 2.1E - 004 * B^{2}$$

-0.091 * C² + 5.34E - 003 * A * B + 1.66E - 003 * A * C - 0.038 * B * C (2)

Here, in Eq. (2) A, B, and C represents the coded values for temperature, flow rate and time. Positive sign in front of these three terms represent synergistic effect, while negative sign represents antagonistic effect. The coefficients with one factor of temperature, flowrate and time and represent the effect of that particular factor for the wax deposit weight. Figure 6 show the influence of all the coefficient of equation 2 of weight of wax deposit. The behavior of equation (2) was plotted in Figure 6 which show that C, AB and AC have a positive influence on the prediction while BC, C^2 , B^2 , A^2 , B and A all have negative impact on the prediction of mass of wax deposited.



Figure 6: Influence of all the independent variables on the model developed



4.3 Model Validation

The coefficients with two factors and others with second order terms show the interaction between the two factors and quadratic effect, respectively. The accuracy of the model developed can be understood by the value of R^2 , adjusted R^2 and standard deviation. R^2 indicates the ratio between sum of the squares (SSR) with total sum of the square (SST) and it describes up to what extent perfectly the model estimated experimental data points. Based on R^2 , it was observed that the experimental data fitted better because of the "Pred R-Squared" of 0.9618 is in reasonable agreement with the "Adj R-Squared" of 0.9945. The accuracy of the predicted and experimental was demonstrated in Figure 4 which show the agreement between the predicted and experimental values. This model can be used to navigate the design space of the experiment.

Determination of CV value is essential as it indicates the ratio between standard error of estimate with the mean value of the observed response as percentage. It measures the reproducibility of the model. If the value The "Adeq Precision" measures the signal to noise ratio with a minimum value of 4 as desirable value. 46.075 indicates an adequate signal. The standard deviation of 0.011, mean of 0.19, and standard error of 10^{-3} .

4.4 ANOVA and statistical significance of the model

The competence and significance of the model was justified by analysis of variance (ANOVA). The ANOVA for the quadratic model for wax deposit weights in the pipeline listed in Table 3. The Model F-value of 325.05 implies the model is significant. There is only a 0.01% chance that a "Model F-Value" be large and it could occur due to noise. Values of "Prob > F" less than 0.0500 indicate model terms are significant. In this case B, C, A², C², BC are significant model terms. Values greater than 0.1000 indicate the model terms are not significant.

	Sum of square	DF	Mean square	F-Value	Prob> F	
Model	0.33	9	0.037	325.05	< 0.0001	significant
А	3.12E-05	1	3.12E-05	0.28	0.6148	
В	0.062	1	0.062	552.55	< 0.0001	
С	7.59E-04	1	7.59E-04	6.74	0.0356	
A2	0.21	1	0.21	1903.37	< 0.0001	
B2	1.86E-07	1	1.86E-07	1.65E-03	0.9687	
C2	0.035	1	0.035	310.21	< 0.0001	
AB	1.14E-04	1	1.14E-04	1.01	0.3476	
AC	1.10E-05	1	1.10E-05	0.098	0.7634	
BC	5.86E-03	1	5.86E-03	52.05	0.0002	
Residual	7.88E-04	7	1.13E-04			

Table 3 Statistical analysis of the model.

4.5 Optimization studies

The numerical optimization was performed to minimize weight of wax deposit in the pipe for selected ranges of variables temperature differential, flow rate and residence time as 28 - 55° C, 100 – 500ml/min and 3 – 15min respectively. By applying the desirability function method in RSM, forty solutions were obtained for the optimum covering criteria with desirability value close to 1. In this case, first solution was selected as good desirability for minimum weight of wax deposit with desirability equal to 1. At this point, weight of wax deposit equal to 0.0195075 at Temperature, Flow rate and Residence time estimated as 53.25° C, 499.54ml/min and 3.01min respectively.

5. Conclusion

The modeling of effect of temperature, flow rate and time on wax deposit was successfully done with minimum number of experimental runs using RSM. The behavior of each of the

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independent variable was observed on dependent variables show that wax deposit weight can only be high at specified conditions of independent variables. The Model F-value implies that the model is significant. The contribution of Flowrate, time, square of temperature, square of time and interaction between flowrate and time (B, C, A², C², BC) are significant to the model developed. The agreement between the predicted and experimental values describe the accuracy of the model developed and can be used to navigate within the design space.

Conflict of Interest

The authors declare no conflict on interest.

References

- [1] Johnsen, S.G., Hanetho, S.M., Tetlie, P., Johansen, S.T., Einarsrud, M.A., Kaus, I. and Semon, C.R. Proceeding of International Conference on Heat Exchanger fouling and cleaning, June 05 10, Crete Island, Greece, 251 258, 2011.
- [2] Tabatabei-Nejaa, S. A. and Khodapanah, E.: (2009) Iranian Journal of Chemical Engineering, 6(1), 50-65.
- [3] Huang, Z., Lee, H.S., Senra, M. and Fogler, H.S.: (2011) American Institute of Chemical Engineers, 57(11), 2955 2964,
- [4] Mehrotra, A. K. and Bhat, N. V.: (2010) Energy & Fuels, 24, 2240 2248.
- [5] Banki, R. Hoteit, H. and Firoozabadi, A.: (2008) International journal of Heat and mass transfer, 51, 3387 3398.
- [6] Edmonds, B., Moorwood, T., Szczepanski, R. and Zhang, X.: (2008) Energy & Fuel, 22, 229-741.
- [7] Osokogwn, U. and Otung, D. A.: (2012) International Journal of engineering sciences and Research Technology, 3, 73-95.
- [8] Todi, S., and Deo, M.: (2006) Offshore Technology Conference (OTC) 18368 Paper prepared for presentation at the 2006 OTC held in Houston, Texas, USA, May 2006.
- [9] Kelechukwu, F.M., Hikmat, S., Aa-Salam, S. and Yassin, A. M.: (2010) International Journal of Physical Sciences, 5(15), 2351-2362.
- [10] Sabet, J. K., Ghotbi, C. and Dorkoosh, F.:(2012), Journal of Nanomaterials, Article ID 340379, 1-15.
- [11] Badwaik, L. S., Prasad, K. and Deka, S. C.:(2012) International Food Research Journal, 19(1), 341-346.
- [12] Biglari, M., Langstaff, A. and Elkamel, A.:(2010) Petroleum Science and Technology, 28, 1788–1798.
- [13] Lee, J., Ye, L., Landen, W.O. and Eitenmillcr, R.R.: (2000) J. Food Compos. Anal., 13, 45-57.