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Studies on the evaporation of waxy crude oil

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Abstract
Spilled oil threatens groundwater resources in many parts of the world. Evaporation increases crude oildensity, viscosity and fraction of lower molecular weight substances which reduce its infiltration into the soil and groundwater. In this work, we studied the impact of wax crystallization on the evaporation rate of waxy crude oil.The characterization of this crude oil shows that it has a high wax content. To characterize its tendency towards solidification,the variation of its viscosity as a function of the temperature was measured, which shows that it is very viscous from a temperature of 40 °C, it crystallizes at 27°C. Subsequently,we determined its evaporation rate at room temperature, the change in composition by GC-FID chromatography analysis. The results indicate that wax in crude oil can greatly decrease its evaporation rate.

Keywords*: Waxy crude oil; Evaporation rate; Viscosity; Density; Gaz chromatography analyses*

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Abbreviations
API: American Petroleum Institute; ASTM: American Society for Testing and Materials;Ev: EvaporationSARA: Saturated Aromatic Resin and Asphaltene; DCS: Differential Scanning Calorimetry; SG:Specific Gravity; WAT: Wax Appearance Temperature

I. Introduction

Oil exploitation and pipelines are sometimes the cause of accidental oil spills(Liu and Kujawinski 2015).The spilled crude oil can undergo several physicochemical and biological transformations that are related to its composition, and involve oxidation (degradation in the presence of oxygen, light), leaching (transport by water), dissolution, solidification (melting point), emulsification, bacterial degradation and evaporation(Xu et al. 2018). Once spilled, the volatile fraction of crude oil evaporates, increasing its viscosity and specific gravity(Fine et al. 1997).The increase in viscosity decreases the permeability coefficient and therefore the infiltration of oil into the soil(Overton et al. 2016; Speight et al. 2017) .Evaporation can thus play an important role in the crude oil solidification / stabilization, which reduces its infiltration into the soil and groundwater(Heidarzadeh et al. 2010) .However, crude oil evaporation can be influenced by parameters such as specific gravity, temperature and spillarea(Fingas 2004).Decreasing in the temperature increases waxesprecipitation. The precipitating waxes form a complex, interacting three-dimensional structures which essentially ties up the lighter volatile components (Majhi et al. 2015)(Sousa et al. 2019; Janamatti et al. 2019). The high complexes molecular weight componentsreduce the evaporation rate of thecrude oil(fig 1).Several studies have focused on the crude evaporation based on parameters such as density, wind speed, time and vapor pressurebut according to M. Fingas 2004 the important factors for the evaporation of petroleum compounds are time and temperature(Fingas 2004). In this work, we studied the evaporation rate of waxy crude.

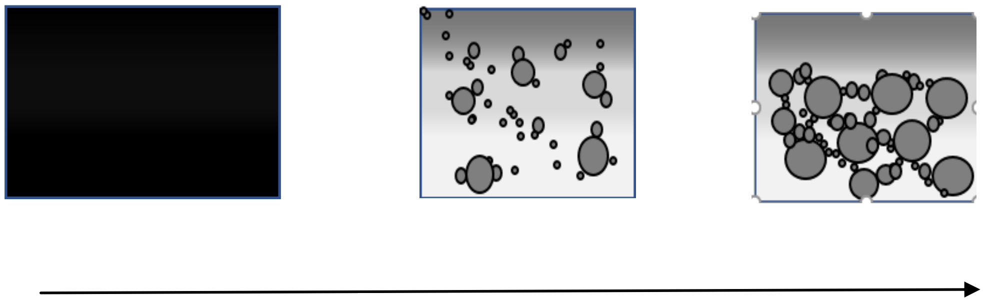

Figure1. *decrease in evaporation rate δ waxes agglomeration*

II. Materials And Methods

2.1. Crude oil characterization

Thestudied crude oil is from Niger. The bulk characteristics of analyzed crude oil include specific gravity, viscosity, pour point, freezing point, wax content, sulfur content, SARA fraction and the Colloidal Instability Index. The standard methods used and the results obtained are presented in the Table I.

2.2.1. Colloidal instability index (CII)

The CII expresses the deposition potential of asphaltenes, which are non-volatiles compounds with very high molecular weight. This parameter is expressed as the sum of Asphaltenes and Saturates per the sum of Resins and Aromatics.

CII=$(Saturates + Asphaltènes)/(Aramatics + Resins)$

If crude oil has a CII value below to 0.7, it is defined as stable, but if CII is higher than 0.9 it is considered as instable, the crude oil tendency towards asphaltenes deposition is so important(Ashoori et al. 2017).

2.2.2. Crude oil evaporation

Evaporation rate was measured as weight loss by using an electronic LCDbalance with an accuracy of 0.01 g. Approximately 15.96g of the crude oil whose composition is determined by GC-FID chromatographic analysis was evaporated at room temperature in the glass beakers of 50 ml. The daily mass loss and the temperature were measured(Fingas 2004). During the experiment the maximum temperatures were recorded between 12.00 p.m and 14.00 p.m and the minimum temperatures between 4.00 a.m and 6.00 a.m (Fig 3). The differences in weight at time zero and the weight at other cumulative times was used to calculate the percentages of evaporation losses by: $\%EV_{evap}=100*(w_o-w)/wo$ (Okamoto et al. 2010)

At the end of the experiment the composition of the residual crude oilwas determined by GC-FID chromatographic analysis.

III. Results And Discussion

3.1. Crude oil properties

The properties of the crude oil that influence its evaporation rate are presented in Table1 and figure1.

Table 1. physicochemical properties of the crude oil

Parameter	Result	Type	Classification criteria	Test method
Density at 20°C **Densité at 50°C** **°API at 15°C**	**0.8565 g/cm^3** **0.8349 g/cm^3** **32.85**	**Light crude oil**	**Light crude oil:** **°API>31.1** **Medium crude oil:** **22.3<°API<31.l** **Heavy crude oil:** **10<°API<22.3** **Extra heavy crude oil** **°API<10**	**ASTM D5002**
Viscosity at (50°)	**13,5 mPa.s**			**ASTM D4402**
Freezing point **Pour point**	**27 °C** **29 °C**			**SY/T 0541** **ASTM D 5853**
Wax content %(m/m)	**24.8**	**High wax content**	**%(m/m) ＞10% high**	

WAT	**64.5 °C**		**crude oil content**	**DSC**
SARA	**Saturates** 52.58% **Aromatics** 14.81% **NSO** 29.88% **Asphaltenes** 2.41%			
CII	**1.23**			
Sulfur content	**0.101**	**Sweet crude oil**	**Crude oil with sulfur content <0.5% is qualified as sweet** **Crude oil with sulfur content 0.5 to 2.0% medium** **Crude oil with sulfur content > 2.0% is qualified as Sour**	

The studied crude oil is light(°API=32.85) and paraffinic with high wax content(24.8%)(García 2000). Waxes are n-paraffins(C12-C35) compounds which deposition and crystallization increase crude oil viscosity when the fluid temperature is lower than the wax appearance temperature (WAT) [14]. With 24.8% of wax content and 27°C freezing temperature, this crude oil crystallization tendency at room temperature is very high. The composition of the crude oil was determined using GC-FID chromatography. Its alkanes fraction is constituted according to the chromatographic analysis (Fig5) of 22.18% of the volatiles alkanes and 43.18% of the semi-volatiles alkanes which can completely evaporate(Kumar and Saravanan 2019). Its sulfur content of 0.101% makes it a sweet crude oil. The sulfur emissions after evaporation is thenbelow the sulfur content limited by legislation(Demirbas et al. 2015). This light crude oil has API gravity of 32.85. The evaporation of crude oil is related to its density. According toM. Fingas 2004 , a light crude oil can lose more than 75% of its volume, a medium crude oil up to 40% of its volume and a heavy crude can lose only 5% of its volume(Fingas 2011). On the other side, the viscosity of this paraffinic crude oil increases a lot when temperature approaches the pour point (Fig2). It reaches its freezing point at 27°C. The transition from the liquid state to the solid state increases the viscosity of the crude oil and consequently the agglomeration of paraffinic waxes, which can reduce its evaporation rate.

Figure2 shows the relationship of crude oil viscosity at different temperatures

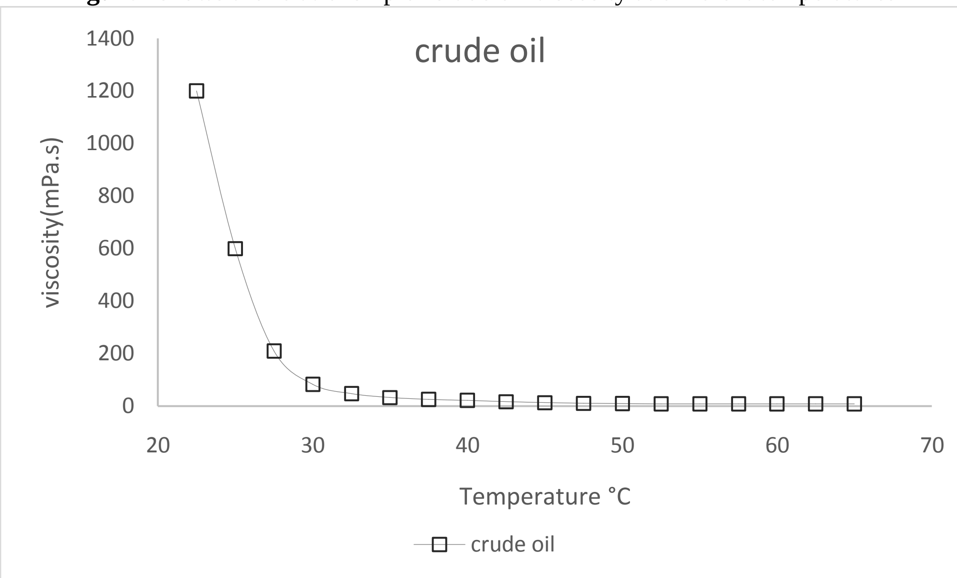

Figure2. *viscosity-temperatureprofil*

A large increment inviscosity wasobserved when the temperature drops from 40°C. The significant increase in the viscosity is linked to its paraffin content and wax precipitation at temperatures below WAT(Zhao et al. 2019). The crude oil viscosity is so controlled by the temperature (Ridzuan et al. 2016). The saturates content of 52.58% also promotes the deposition of asphaltenes which are non-volatilescrude oil components(Alcazar-Vara and Buenrostro-Gonzalez 2011). The calculated Colloidal Stability Index of 1.23 is greater than 0.9, the tendency of this crude oil to form colloids is very high. The formation of colloidal which are high molecular weight compounds can also reduce its evaporation rate(Ashoori et al. 2017).

3.2. Evaporation rate

The daily mass loss is represented in Figure 2 and the temperature variation in figure 3

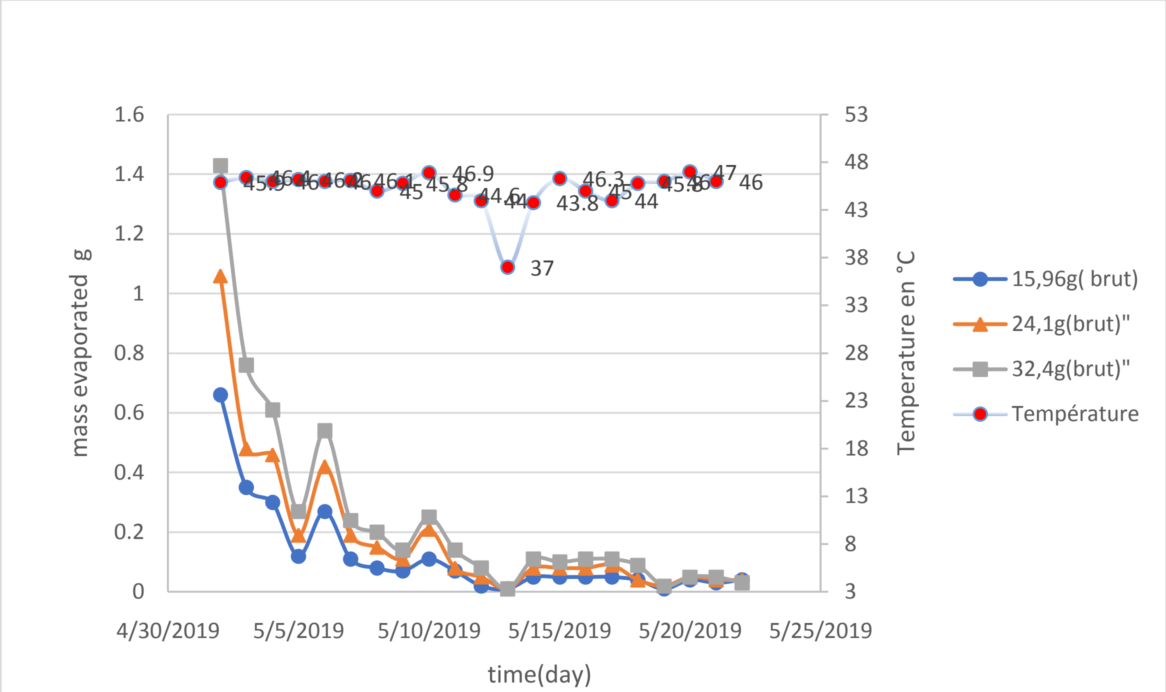

Figure 3. *mass loss per day and the temperature*

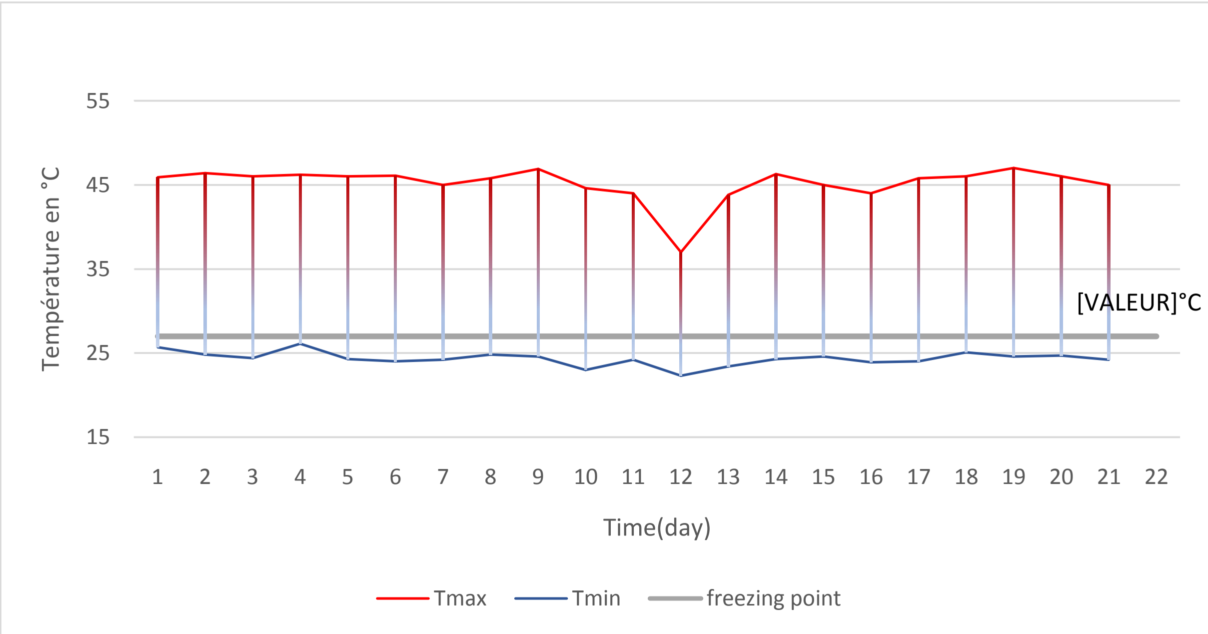

Figure 4. *temperature variation*

The evaporated mass per day increases with temperature. For the same temperature, the evaporated mass decreases with time this is due to the depletion of petroleum volatile components(Bufarasan et al. 2002). The maximum temperature of 47°C recorded (fig4) during the experiment is less than the crude oil WAT (64.5 °C). The minimum temperatures recorded are less than the freezing temperature of this crude oil (27°C). In Figure 2 the viscosity of this crude oil increases greatly when the temperature drops from 40°C. At the temperatures recorded during the experiment, the viscosity of this crude oil is then very high. The amount of crude oil evaporates per day became almost zero for samples without acetone after 23 days of evaporation. To determine its evaporation rate, we presented the cumulative mass percent loss versus time in the figure 6.

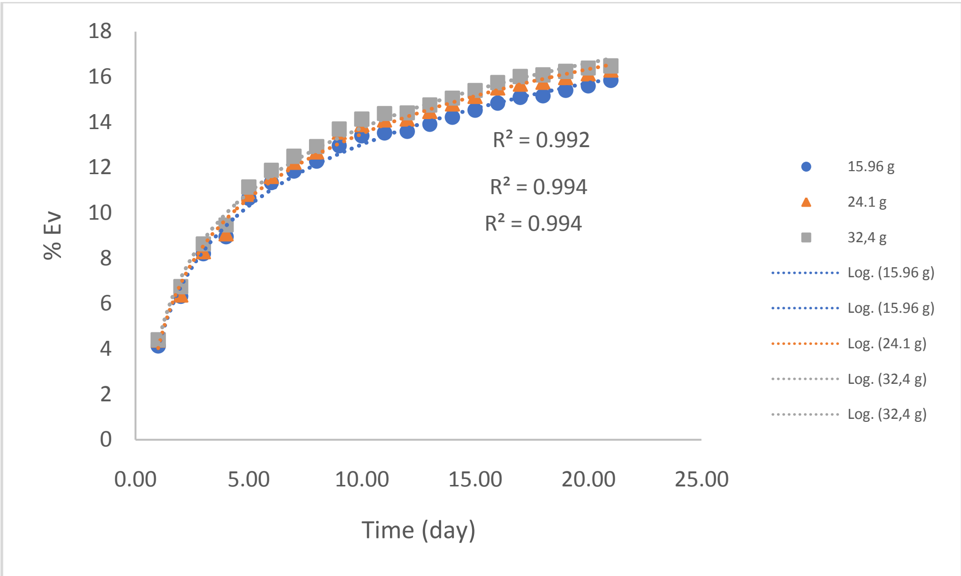

Figure 5. *mass percent evaporated*

The maximum evaporative loss is determined after 23 days when the mass loss is very minimal.The mass percent lost is 25.68% despite the high API gravity of this light crude oil.

This suggest that evaporation rate at room temperature can be limited by wax agglomeration because light crude can lose more than 75 % of its mass.The figure 6 shows that the crude oil evaporates at logarithmic rate (M. F. Fingas 1997).

3.3. Compositional changes induced by evaporation

The n-alkanes distribution (nC1-nC32) before and after evaporation was analyzed using a Gas Chromatograph coupled with a Flame Ionization Detector (GC-FID) and presented in Figures 7. The crude oil consists of a large quantity of n-alkanes with a number of carbon atoms greater than 15 which correspond to paraffinic wax molecules(fig7a)(García 2000).The chromatogram of the unevaporated crude oil (fig7b) shows the loss of n-alkanes in the C1 to C4 range, the decrease from 22.2% to 1.7% of the volatile alkanes(C5-C9) andapersistence of semi-volatile alkanes(C10-C15) that can evaporate at room temperature(Tianhong Zhou and Kau-Fui V. Wong 1996; Kumar and Saravanan 2019).

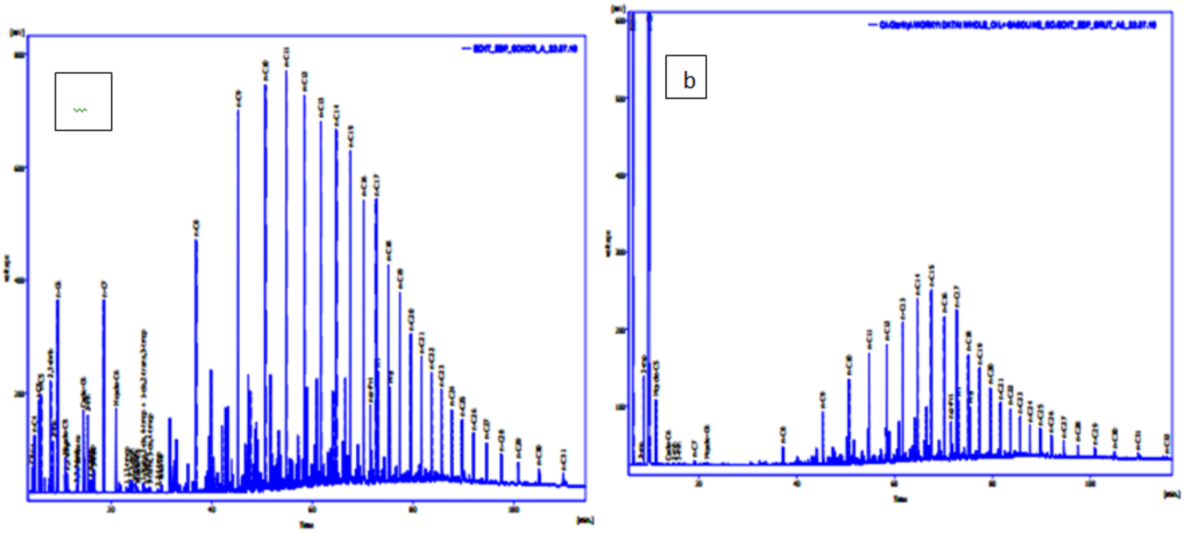

Figure 6. *Crude oil chromatograms before(a) and after(b) evaporation according to the GC-FID method*

Table 2. Percentage of volatiles and semi-volatiles n-alkanes

		% Before Evap	%AfterEvap
% volatils n-alkanes	n-C5	2.2	0
	n-C6	3.8	0
	n-C7	3.8	0.1
	n-C8	4.9	0.3
	n-C9	7.4	1.3
	TOTAL	**22.2**	**1.7**
% semi-volatils n-alkanes	n-C10	7.69	2.2
	n-C11	8.13	2.5
	n-C12	7.47	2.7
	n-C13	7.14	3.2
	n-C14	6.59	3.7
	n-C15	6.26	3.8
	TOTAL	**43.28**	**18.1**

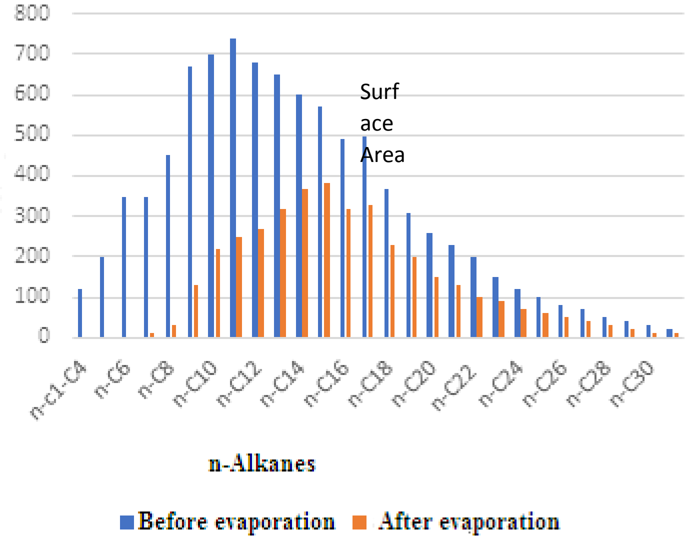

Figure 7. *Variation of n-alkanes before and after evaporation*

3.4. Effect of evaporation on specific gravity (SG)
The relationship between specific gravity and time is represented in figure8.

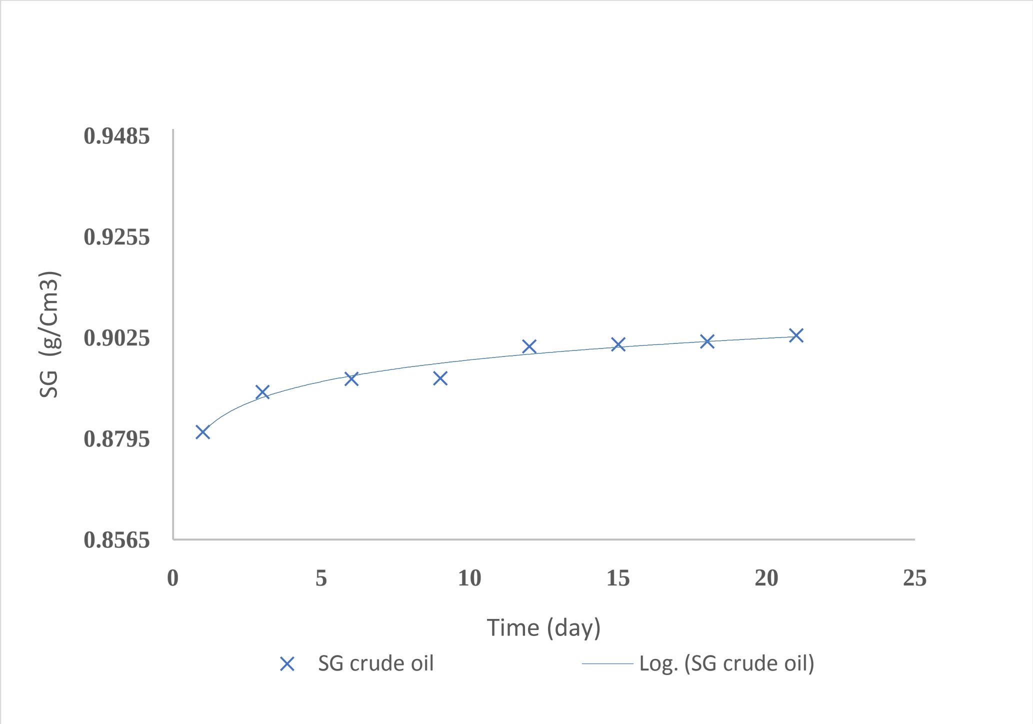

Figure8*. S.G of crude oil after evaporation*

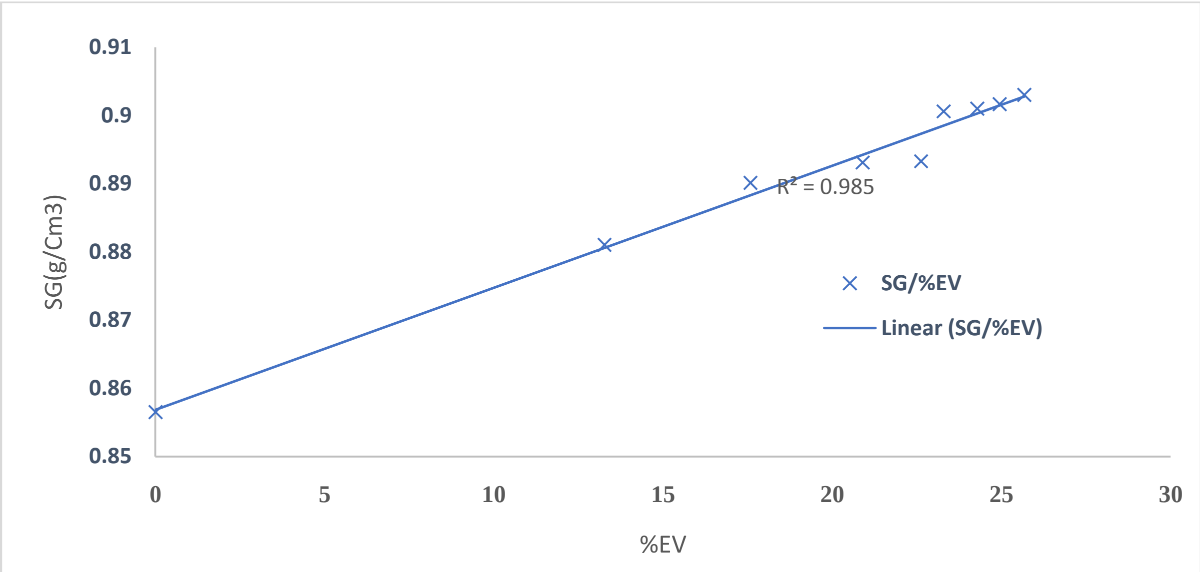

Figure9. *SG variation of theresidualcrude oil as a function of %Evaporated*

The changing in composition caused by evaporation leads in the changes of physical properties of the crude oil(Bufarasan et al. 2002). When time increases, light components evaporate and the specific gravity also increases(Barker and Bufarsan 2001).The effect of increase in specific gravity due to increase in the percentage evaporated of crude oil is represented in figure11. As can be seen, the specific gravity increases linearly with the increase in percentage evaporated.

IV. Conclusion

The study of the physicochemical properties of this crude oil shows that it is a light crude oil which has a very high wax content. The high wax content promotes the increase of its viscosity and its solidification at room temperature which slowed down its evaporation rate. The tendency towards solidification greatly reduces the evaporation rate of semi-volatiles components, especially the semi-volatile alkanes.

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