Nigerian Journal of Textiles (NJT)
A Publication of Textile Researchers Association of Nigeria (TRAN)
Volume 6 August 2020
http://tran.org.ng

Ceric Induced Grafting of Acrylonitrile onto Kenaf Shive for Crude Oil Spillage Adsorbtion

Salisu, Z. M [1,*], Ishiaku, S.U[2], Abdullahi, D[2], Yakubu, M.K[2], Diya'uddeen B.H [3]
1. National Research Institute for Chemical Technology (NARICT), Basawa, Zaria, Nigeria
2. Department of Polymer and Textile Engineering, Ahmadu Bello University (ABU), Zaria, Nigeria
3. Center for Energy Research and Training (CERT), Ahmadu Bello University (ABU), Zaria, Nigeria
*ruddeen.zms@gmail.com (+2348067990749)

ABSTRACT

A global challenge that has cast the attention of most environmentalist of recent is the crude oil spillage, and the need for extensive study of improving and developing viable natural sorbents in recent years is increasing due to the enormous negative environmental impact of oilspill to the surroundings and the inhabitants. In this paper, crude oil sorbents were developed by varying initiator concentration, monomer ratio and particle sizes from 0.5-2.5wt%, 0.5-2.0wt % and 125-1000µm respectively. The effect studies of the individual and combine factors was carried out using a statistical experimental design matrix using five-level central composite design (CCD). Respond surface methodology (RSM) was used to optimise and develop equations of the aforementioned variables (initiator concentration, monomer ratio and particle sizes). The optimal absorption- 516% as swelling capacity and 267% lower retention - was achieved at initiator concentration, monomer ratio and particle size of 1.25wt %, 1.50wt % and 562µm respectively. Instrumental analysis were carried out on the optimized sorbent such as: DT-TGA, XRD. However, FIRT analysis was carried out on the unmodified (raw kenaf shive) and modified sorbents. Further computations were done for grafting efficiency (GE), homopolymers and density.

Key words: *Sorbent, initiator, crude oil, kenaf shive, Respond surface methodology (RSM)*

Introduction

Crude oil spillage is the release of a liquid petroleum hydrocarbon into the environment due to human activity, and is a form of pollution. The term often refers to marine oil spills, where oil is released into the ocean or coastal waters. Oil spills include releases of crude oil from tankers, offshore platforms, drilling rigs and wells, as well as spills of refined petroleum products (such as gasoline, diesel) and their by-products, and heavier fuels used by large ships such as bunker fuel, or the spill of any oily white substance refuse or waste oil. Spills may take from a month up to years to clean up. Oil also enters the marine environment from natural oil seeps.

Crude oil is one of the major sources of income bestowed under the earth's crust of many countries; Nigeria is one of the most endowed countries with this resource. But because of environmental issues associated with exploration, transportation and refining of the crude oil, this very important revenue earner becomes a menace for most of developing countries mainly due to spillage. [1,2] reported that oil spills cost Nigeria more than 1.89 million barrels annually from more than 10,000 accidental spills which less than 4% and 22.5% as a result of tanker accidents and operational discharge respectively.[3-5]; reports Tokyo oil spill incident—where about 4 million gallons was spilled out—many lives were lost. Apart from the attendant loss in revenue, aquatic organisms also suffer a lot from this oil spills.

Oil spills is considered as one of the most serious disasters that is threatening the marine ecosystem [6,7]. Many techniques have been devised to combat this problem [8]. Historical synopsis of oil spill showing the economic impact from 1967-2018 is listed in Table1.

1.1. Methods of Combating Crude Oil Spillage

In the past, containment and recovery measures have been utilized for oil spill mitigation. The conventional techniques include: containment and mechanical recovery; burning; bioremediation; chemical dispersant and the use of absorbent[10]. The recovery techniques are dependent on various factors such as weather conditions, sea condition, oil type and environmental considerations, which could necessitate the combinations of these measures for clean-up. These techniques include: burning in situ, bioremediation, chemical dispersion and synthetic sorbents in spite of their secondary effect of nondegradability [8]. The most widely accepted by many researchers and industries is the one prepared from polypropylene fibres and is now being considered hazardous [11-13].

Table 1: Oil spill history (www.marine group); [9]

Year	Country	Incident	Quantity (tonnes)
1967	United Kingdom	Torrey Canyon ran aground off cornwall spilling	125,352
1970	Sweden	Collision involving Othello in Tralhavet Bay	59,743
1972	Oman	Collision of Brazilian tanker Horta Barbosa with the South Korean tanker Sea Star	114,576
1976	United State of America	Argo Merchant ran aground off Nantucket	24,961
1978	France	Amoco Candiz ran aground near Portsall	218,240
1979	Mexico	Gulf of Mexico	454,667
1979	Trinidad & Tobago	A collision off Tobago between the Atlantic Express and the Aegean Capital	300,080
1983	South Africa	Spanish tanker Castillo de Bellver Fire	245,520
1989	United State of America	Exxon Valdez hit rocks in Prince William Sound	32,736
1990	United State of America	The tanker, American Trader	974
1991	Kuwait	Final phase of Iraqi attack of Kuwait	7,557,935
1992	Indonesia	Nagasaki Spirit collided with container Ocean Blessing in the Malacca Straits spilling some 1993	12,000
1993	Singapore/ Indonesia/ Malaysia	Singapore-registered tanker Maesk Navigator collided With the empty tanker Sanko Honour in the Andaman Sea	272,800
1995	United Arab Emirates	Panamanian-flagged supertanker Seki	15,900
1996	Australia	Iron Baron, ran aground on a reef	500
1998	Nigeria	Ruptured pipeline to one of Mobil's terminals	5,456
1999	France	Registered tanker Erika breaks up in stormy seas	15,000
2000	Brazil	Jeaked from a refinery	31,491
2000	Malaysia	Sunken Chinese cargo ship at Tanjung Poanchorage point at the Sarawak River mouth	5,000
2003	Pakistan	An oil tanker has broken up off Pakistan's Arabian sea port, Karachi	10,000
2004	Canada	Occurred at the Terra Nova offshore oil platform	1,386
2005	United State of America	Murphy oil refinery spill	2,660
2006	Lebanon	Jiyeh power station oil spill	20,000
2007	Norway	Statfjord oil spill	4,000
2008	United State of America	New Orleans oil spill	8,800
2009	Australia	Montara oil spill	4,000
2010	Nigeria	Exxon Mobil oil spill	3,246
2011	Canada	Little Buffalo oil spill	3,800
2012	United State of America	Arthur Kill storage tank oil spill	1,090
2013	United State of America	Magnolia refinery spill	680
2013	Thailand	Rayong oil spill	43
2014	United States	North Dakota pipeline spill	1,10
2014	United States	MV Miss Susan/MV Summer Wind	5,46
2014	Israel	Trans-Israel pipeline	4,300
2015	United States	2015 Yellowstone River oil spill	1,60
2015	Canada	MV Marathassa	2.3
2016	United States	Shell Gulf of Mexico oil spill, Brutus offshore platform	3,16
2016	Canada	Conocophilips Canada Pipeline spill	3,23
2017	Greece	Agia Zoni II	2,500
2017	United States	Delta House oil spill	1,820
2017	United States	Keystone Pipeline	3,82
2018	East China	Sanchi oil tanker collision with CF crystal	1,3800

Nanocellulose aerogel, carbon nanotubes are the recent ones that gave high absorbency capacity (g/g), but these cannot be sustained because of the high cost of raw materials and processing [14-17]. Most recent research discovery shows that natural materials are the best for oil cleaning-up [18]. Moreover, cotton and kapok are the best amongst the green plants. This is because of their higher oil sorption capacity, biodegradability and recyclability which are preeminent materials for oil spill cleanup. Cotton has loose fibers, which presumably limited their application [8, 19]. The problem with cotton necessitated the use of kenaf shive as a possible alternative, it has other advantages over cotton which has to be converted into pads in the preparation sequence consequently reducing the diameter of the capillaries [20]. Kenaf fibres is used in producing bags for agricultural packaging and, by utilizing its shives makes a lot of economic sense since it is normally thrown away [21, 22].

The approaches used in recent years toward super hydrophobicity of cellulosic materials can be classified into two categories, based on the generation of roughness:

(I) Roughness offered by coating cellulosic substrates, which include:
   a. Chemical grafting to modify the surface chemistry and surface morphology of cellulosic fiber/surface simultaneously.
   b. Sol-gel processes to render cellulose fiber/surface with porous outer-layer and to reduce surface energy by post-treatment or by mixing precursors with low surface energy side chains.
   c. Nanoparticle deposition, for example by using metal, metal oxide, mineral and polymers that modify the morphology of the cellulosic fiber/surface, followed by surface energy reduction by post-treatment.
   d. Chemical vapor deposition

(II) Roughness offered by regeneration or fragmentation of cellulosic materials, which among others include:
   a. Electrospinning and elecrospraying
   b. Use of nanoparticles (cellulose nanocrystals and nanofibrillated cellulose)
   c. Use of cellulose composites [23, 24]

Natural sorbents were developed by surface modification for oil sorption. Owing to preeminent properties such as low cost, high efficiency and biodegradable of natural sorbents gained a high exploration. A high number of natural organic oil sorbents were reported, namely: wood chips, sugarcane bagasse, cotton and jute [25-28. Jute plant having many a common properties with kenaf plant deems to be investigated. Jute and kenaf constitute of cellulose, hemicellulose (82-85%) and lignin [29, 30].

Kenaf shive/core crude oil sorbents is aimed with facile/robust because of low cost, durable (strong lumen) and ecofriendly via regenertion technique that is handy and practicable in oil/seawater system. Kenaf shive was selected because it consist (60-70%) of the plant and vibrant in withstanding different climatic changes. Synergizing the abundance of kenaf plant and non- collapsing lumen tipped for new research direction in crude oil sorption. Furthermore, optimizing and instrumental analysis were performed.

2.0 Materials and methods

2.1 Materials and chemical preparation

All chemicals are analytical grades and used how it was received without further purification, except for the monomer (acrylonitrile) which inhibitors were removed by washing it severally with 5% concentration of NaOH. Dried Kenaf stalks were obtained from National Research Institute for Chemical Technology (NARICT), Zaria.

2.2 Source of crude oil

The crude oil sample used for the sorption test was obtained from Petroleum Research Laboratory, Warri, Delta state, Nigeria. The raw crude oil was kept at room temperature and the adsorption test was carried out at 40^0C after the grafting of the acrylonitrile onto the kenaf shive. Table 1 shows the significant properties of the oil that perhaps affect the oil sorption.

2.3 Experimental procedures

The obtained dried kenaf stalks were subjected to chemical retting, 1% w/v NaOH for 2hrs in order to extract its shive from the two components (shive and bast fibres). The product was washed with distilled water until neutrality was achieved and further drying took place for 72hrs at room temperature. The extracted shive was ground into different particle sizes as prescribed by the DoE software result sheet.

Synthesis of the grafted sorbent was done by soaking requisite quantity (1.00g) of kenaf shive in 5mls of distilled water for 24hrs. The mixture was transferred to reaction kettle and 1.00ml of

2% acetic acid, 10.00ml of 0.4M of nitric acid and 0.5ml of the weight percent of the requisite quantities of initiator as well as monomer concentration as in Table 2 were added, however, the monomer was added after purging nitrogen gas for 5min. The reaction continued for 3hrs at 60^0C. The sample was then washed, filtered and oven dried at 40^0C.

Three neck flask was quarterly filled with about 25ml of acetone for homopolymers removal. The initial weight of the thimble was taken after which the thimble plus grafted shive was noted. The latter was inserted into the extractor for the extraction process. This was done at 60^0C for 24hrs in which the homopolymers weight were calculated as in Table 3. The experimental processes were repeated for twenty samples using requisites regressors as in Table 3.

Table 2- Specifications of crude oil samples

Sample	Viscosity (m^2/s)	Speed (m/s^2)	Torgue (Nm)	Temp. (^{0}C)	Density (g/cm^3)
Crude oil	1.33	30.00	0.10	24.5	0.8965
	0.67	60.00	0.00	24.5	

Table 3: FTIR result for modified kenaf shive.

WAVE NO. (CM^{-1})	VIBRATION	STRUCTURE
2922	δs (CH)	–CH3–H
3291	δs (OH)	–OH
2113	δ s(CH2)	–CH2
1736	δ s(CO)	–C=O
1640	δs(CC)	–C=C
1226	Vs(CO)	–C–O–
1021	Vs(CN)	–C≡N

δ s- Asymmetrical stretch, δs- symmetrical stretch, Vs- symmetrical vibration

The extracted grafted sorbents were tested for crude oil sorption using requisite quantity of the sorbent (0.10g) into watch glass containing 5mls of the oil. This was done at 40^0C for 5mins to achieve proper sorption.

2.4 Analytical test

Infrared spectra of the sorbent in KBr pellets was analysed and scanned from 4000 – 400 cm^{-1} using Shimadzu FTIR-8400S. The test was carried out on the raw (unmodified) and modified optimized unextracted sorbent that bears the highest oil sorption to confirm the modifications by taking the advantage of the unique vibration/stretching property for each functional group. The sorbent crystallinity was determine using Shimadzu XRD 6000 (Tokyo, Japan) with CuKαradiation (λ= 1.542 Å) operated at 30 kV and 30 mA. Surface area was determined using Brunauer, Emmette and Teller(BET) technique by (Quantachrome Instruments, Model Nova1000e series, USA), however, the heat properties was not set aside but determine using DTA-TGA60 Shimadzu, Japan.

2.5 Experimental design, data analysis and process optimisation

There are several variable which potentially affects the grafting efficiency and absorption. Response surface method (RSM) would be of tremendous important in the analysis of such multivariate system. The objective of using RSM is to get a quick insight in the interaction amongst the investigated variables, facilitate the optimisation conditions and locate optimal response in the region of interest [30, 31]. For these purpose, a reduction empirical model describing the process was developed for predicting and determining future responses in such system. The generated results of the responses from the experimental runs would be obtained by employing Eq. 1 through fitting with a second-polynomial equation was used to predict the studied variable factors as independent variable and interaction between them:

$$Y = b_0 + \sum_{i-1}^{k} b\ X\ + \sum_{i-1}^{k} b\ X^2 i + \sum_{i-1}^{k} 1 \sum_{i-1}^{k} b\ + \varepsilon \quad \ldots. (1)$$

where, Y is the predicted dependent variable, b_0 is constant coefficient, *bi*, *bii* are regression coefficient, *i* and *j* are index numbers, k is number of patterns, X'_is are independent variables and ε is the random error. The analysis of variance (ANOVA) was used to assess the significance and adequacy of the model. The fitness of the

polynomial model was express by coefficient of determination, R^2, R_{adj}^2 and R_{pred}^2. The main indicators that were used to show the significance of the model were Fisher Variation Ratio (F-value), probability value (Prob > F) with 95% confidence level and adequate precision. The final model for each response was obtained after elimination of insignificance term ($p > 0.05$) base on F-test and 3D plots were presented. In addition, the optimum values of the independent variables were identified and further development of the absorbent was carried out at this condition to confirm the regression models.

3.0. Results and discussions

3.1. The crude oil sample was characterised using Rheometer instrument.

3.2. Experimental designs and ANOVA analysis

The obtained results from the absorbent experiments and predicted values by the developed model for the studied dependents variables are presented in Table 3.

From the interaction results of the variables, two multivariate models were derived to describe the DOC reduction and the accompanying sorbents development (eqn. (2) and (3)).

$$G = +17.5092 + 0.662519(A) - 0.04072(B) + 0.393868(C) - 0.00041(A*B) - 0.24583(A*C) - 0.00179(B*C) - 0.01776(A)2 + 3.13E - 05(B)2 + 1.670909(C)2 \ldots.(2)$$

$$O\ s = +337.23974 - 5.06312(A) + 0.71128(B) - 175.61128(C) - 9.95810E - 003(A*B) + 3.91333(A*C) - 0.15183(B*C) + 0.13552(A)2 - 1.61318E - 004(B)2 + 56.77273(C)2\ g/g \ldots(3)$$

The synergistic and antagonistic effects are shown by the positive and negative signs in the equations respectively. Based on these models, grafting efficiency and absorption can be predicted as a function of particle sizes, monomer ratio and initiator concentration.

The models were found statistically significant ($p<0.05$) and therefore included in the models, analysis of variance (ANOVA) was performed (Table 3). Based on the results, it is seen that the p-values for both responses were both less than 0.05 (<0.0001), this indicates that is significance and both could be used for response prediction. Regression coefficient R^2, adjusted R^2 and predicted R^2 were used to evaluate the quality of the developed equations. The adjusted R^2 values that display the total variation of the response were 0.7092 and 0.5780 for grafting efficiency and adsorption, respectively. The R^2 values close to 1.000 is desirable as it shows the acceptable adjustment of the suggested model with experimental data. The absorption and grafting efficiency of the regression coefficient (R^2 = 0.8469 and 0.7779) and predicted (R^2= 0.7197 and 0.6032) indicated that the models are highly reliable in terms of repetition of the experiments and also adequate in the actual relationship between the responses and the variables. Furthermore, the adj-R^2 and pre-$R^2$are in good agreement where the difference between them was less than 0.2 [32,33]. However, the adequate precision that is signal to noise ratios were 8.0 and 9.0 respectively, which are greater than 4.0 (desirable value). This indicates adequate signals that shows the models could be used to navigate the design space. The lack of fit shows the variation around the fitted model. In the adsorption is 0.1222 implies that the lack of fit is significant relative to the pure error. The insignificant lack of fit was good and it showed that this model was good to predict the amount of absorption within the studied range of variables. On the contrary, the lack of fit was significant (*F*-value was 0.0001) for the graft efficiency, the model could still be used for design space navigation defined by the CCD, due to good agreement between the adjusted and predicted R^2 values [34]. To ascertain the adequacy of the models in exploring the response surface arising from the variability observed in the data owing random errors, residual were evaluated in addition to regression coefficient.

3.3 Analytical tests

3.3.1 Fourier Transmitted Infrared (FTIR) spectroscopy

The FTIR spectrum of the starch in Figure 3, gives strong absorption bands at 3324 cm^{-1}, an –OH broad and ingle intermolecular polymeric band the C–H stretching at 2922 cm^{-1}, and C=O stretching in the 1673cm^{-1} band, characteristic of the parent unmodified material contained in the kenaf shive (UG). For the optimized sorbent (GAN), the peaks at 2922, 3291, 2113, 1736, 1640, 1226, 1020 cm^{-1} that were assigned to –CH3–H, –OH, –CH2, –C=O, –C=C, –C–O– and –C≡N respectively as shown in Table 3.

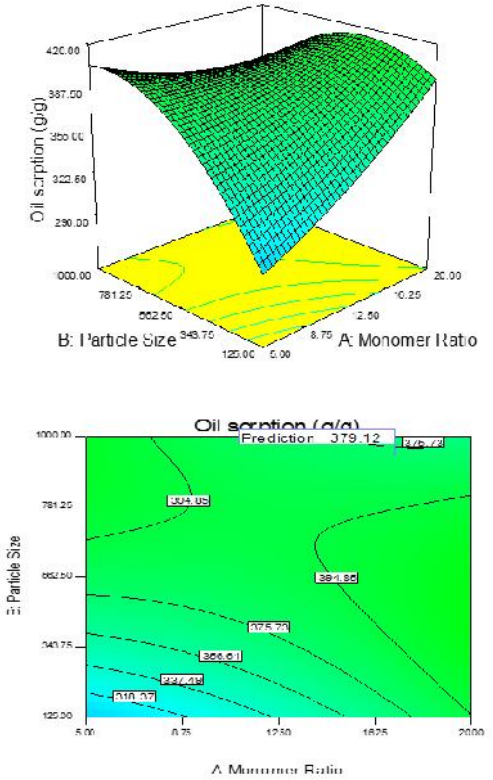

Figure 1- Response surface (a) and contour plots (b) for oil sorption as function of particle sizes and monomer ratio.

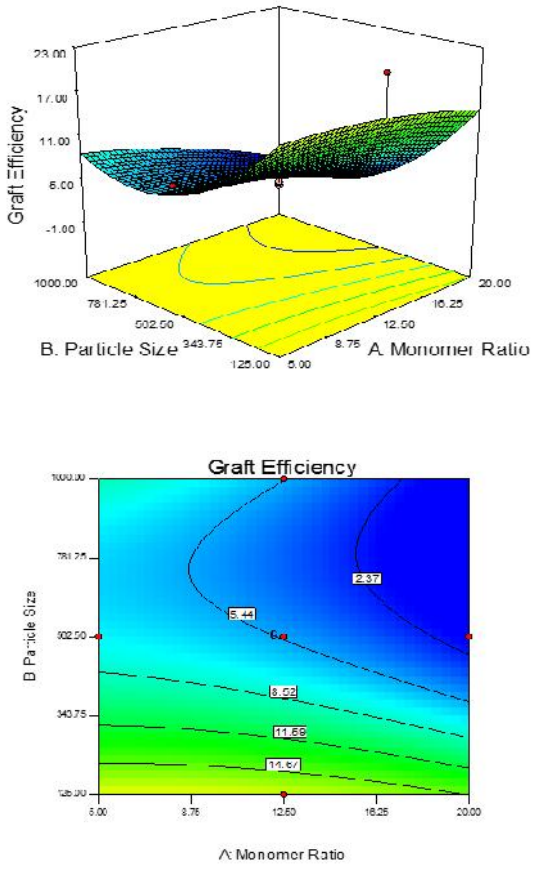

Figure2- Response surface (a) and contour plots (b) for grafting efficiency as function of particle sizes and monomer ratio.

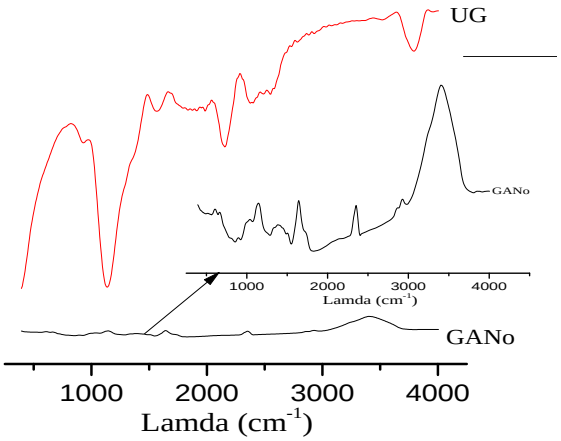

Figure 3- FTIR spectra of the unmodified modified kenaf shive sorbent

3.3.2 DT-TGA test

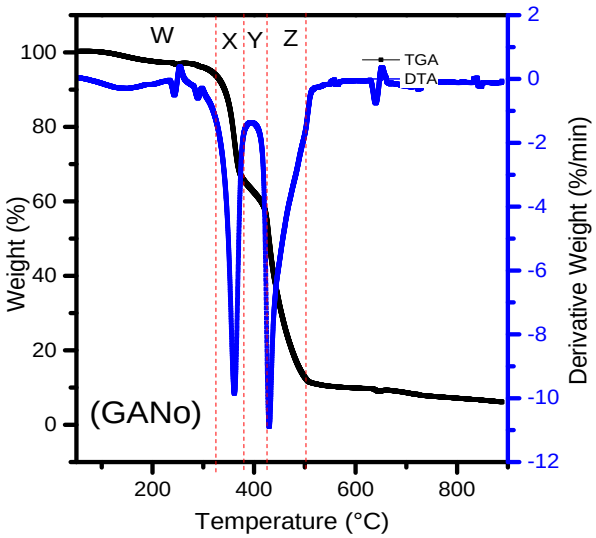

Figure 4: DT-TGA plots showing the effect of temperature on the optimized modified silane sorbent

DTA transition was shown in Figure 4 which is an indication of the water evaporation, crystallization, pyrolysis and decompositions at about 110^{o}C, part W, X, Y and Z respectively.

On the other hand, TGA plot shows that at ambient temperature there is little indication of weight loss but not obvious this indicates the slight hydrophobicity of the sorbent and this is proved from the DTA plot [30]. The major weight loss were shown at part label X, Y and Z where there were, respectively, weight loss of 32, 6 and 58%. Afterwards, there was continuous degradation living about 8% residue. This is an indication of its degree of organic property.

3.3.4 X-ray difraction test

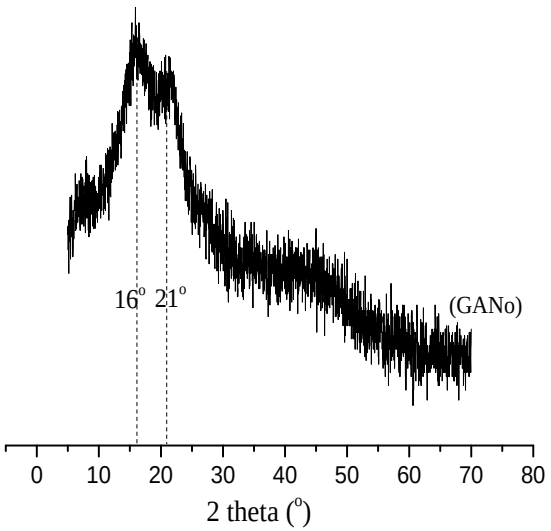

Figure 5: X-ray difractogram for the optimized silane/kenaf shive sorbent

The plot X-ray difractogram (Figure 5) is a plot which indicates the crystallinity properties of the optimized sorbent. It is obvious that there are two plateaus which attributes to the crystalline region at 16^{o} and 21^{o}. Generally speaking, the graph shows the sample is amorphous. This is of advantage in the reaction actualization as it comes prone to the reactants, however, gives it more ability for adsorption.

Table 6 shows the swelling capacities of the modified shive in which the highest is 637% and the lowest is 205% while Table 5, the control i.e. unmodified shive is having 525 as the highest and 23% as the lowest. However, this is not the yardstick for the best between the two because we have to consider percentage retention that is the determining factor for the oil recovery. In a nutshell, the grafted shive is preeminent because of its merits like ease recovery, low retention and high swelling percentage than the ungrafted ones.

Table 4- Design matrix for crude oil sorbent

Run no	Experimental design			Density (g/cm^3)	Homopolymer (g)	Results			
						Experimental		Predicted	
	Monomer ratio (%)-A	Particle size (µm)-B	Initiator conc. (%)-C			Grafting efficiency	% swelling	Grafting efficiency	% swelling
1	5	1000.00	0.50	0.075	0.021	8.78	637.20	8.013	675.74
2	5	125.00	2.50	0.068	0.049	24.18	326.30	23.32	312.75
3	12.5	562.50	1.50	0.088	0.036	4.47	474.30	5.36	383.75
4	20	125.00	0.50	0.106	0.027	14.63	342.90	16.07	307.71
5	5	125.00	0.50	0.140	0.004	11.70	305.00	15.41	322.16
6	20	125.00	2.50	0.120	0.026	15.18	453.90	16.61	415.70
7	5	1000.00	2.50	0.058	0.054	13.57	365.10	12.79	400.63
8	12.5	562.50	1.50	0.072	0.045	4.67	365.10	5.36	383.75
9	12.5	562.50	0.50	0.078	0.012	11.52	516.70	5.71	482.30
10	12.5	562.50	1.50	0.071	0.058	4.67	365.10	5.36	383.75
11	12.5	562.50	2.50	0.102	0.010	4.99	365.70	8.36	398.74
12	20	1000.00	2.50	0.074	0.001	3.69	389.70	0.70	372.88
13	12.5	1000.00	1.50	0.085	0.019	2.29	501.70	5.53	430.56
14	12.5	562.50	1.50	0.083	0.042	4.57	365.10	5.36	383.75
15	12.5	125.00	1.50	0.079	0.034	22.86	205.40	17.18	275.18
16	20	562.50	1.50	0.074	0.022	2.72	304.50	1.51	380.82
17	12.5	562.50	1.50	0.083	0.042	4.17	365.10	5.36	383.75
18	20	1000.00	0.50	0.075	0.032	1.72	516.70	3.30	530.59
19	12.5	562.50	1.50	0.083	0.042	4.37	365.10	5.36	383.75
20	5	562.50	1.50	0.073	0.044	8.45	479.60	7.22	401.92

Table 5- Analysis of variance (ANOVA) for the grafting efficiency and absorption process

Source	Sum of squares	Degrees of freedom	Mean square	*F*-ratio	*P*-value (Prob >*F*)
Grafting Efficiency					
Model	**442.41**	**3**	**147.47**	***6.11***	**0.0057**
Residual	**386.25**	**16**	**24.14**	**-**	**-**
Lack of fit	**386.04**	**11**	**35.09**	**822.52**	**0.0001**
Pure error	**0.21**	**5**	**0.043**	**-**	**-**
Cor total	**828.66**	**19**	**-**	**-**	**-**
Absorption					
Model	**1.297E5**	**6**	**21609.49**	**5.38**	**0.0054**
Residual	**52241.38**	**13**	**4018.57**	**-**	**-**
Lack of fit	**42304.18**	**8**	**5288.02**	**2.66**	**0.1480**
Pure error	**9937.20**	**5**	**1987.44**	**-**	**-**
Cor total	**1.819E5**	**19**	**-**	**-**	**-**

Table 6- Control (Unmodified kenaf shive)

Particle Size (µm)	**Mass of Sorbent before sorption (g)**	**Mass of Sorbent after sorption (g)**	**Mass of Sorbent after squeezing (g)**	**Oil sorption (g)**	**% Swelling**	**% Retention**
1000	0.1	0.625	0.506	0.119	525	406
562	0.1	0.580	0.312	0.268	480	212
250	0.1	0.548	0.397	0.151	448	297
125	0.1	0.123	0.079	0.044	23	-21

4. Conclusion

A crude oil sorbents was achieved from kenaf shive by chemical modification using effluence of monomer ratio, particle size and initiator concentration based on the statistically designed experimental matrix. The effect of the process-specific variable were investigated using 6-level central composite design (CCD) and response surface methodology (RSM) used for the process optimisation. FTIR result also ascertained the modification achievement, where the expected peaks were glarely seen. This succeeded sorbent was tested for oil recovery, the result sound if compared with the control (ungrafted). The former shows a low retention and high swelling while the latter has high retention which is an indication for poor recovery. Amongst the modified ones, the sorbent with 1000um, 5% , 0.50 % w/v of particle sizes, monomer ratio and initiator concentration, respectively, gives the higher oil sorptionof(516%, swelling capacity and lower retention of 267%). This perhaps is due to lower homopolymers. However, apart from it having preeminent properties of average density which makes it unsinkable-rather to flow in the oceans-it is also hydrophobic based on the grafting temperature and particle size that is nearly to its natural length of the fibre [35-37].

References

[1] Diya'uddeen, B.H.; Mohammed, I.A.; Ahmed, A.S.; Jibril, B.Y.; 2008, "Production of Activated Carbon from Corncobs and its Utilization in Crude Oil Spillage Clean Up". Agricultural Engineering International: the CIGR Ejournal. Manuscript EE 08 004. Vol.X.

[2] Adelana, S.O; Adeosun, T.A; Adesina, A.O; Ojuroye, M.O; 2011 "Environmental pollution and remediation: challenges and management of oil Spillage in the Nigerian coastal areas" American Journal of Science and Industrial Research.

[3] Liu, H., Geng, B., Chen, Y., Wang, H., 2017. Review on the aerogel-type oil sorbentsderived from nanocellulose. ACS *Sutainable Chem. Eng.* 5, 49-66

[4] NOAA, 2017b. Spill Containment Methods. Office of Response and Restoration[Online] Available at: https://response.restoration.noaa.gov/oil-and-chemicalspills/oil-spills/spill-containment-methods.html.

[5] Salisu, Z.; Umaru, IS.; Danladi, A.; Yakubu, MK.; Diya'uddeen, BH. 2019a. Optimisation of crude oil adsorbent developed from a modified styrene kenaf shive.*J. Scientific Research.* Unpublished paper.

[6] Son, T. Nguyen; Jingduo, Feng; Nhat, T. Le Ai; T. T. Le; Nguyen, Hoang; Vincent, B.

C. Tan; Hai, M. Duong; 2013, "Cellulose Aerogel from Paper Waste for Crude Oil Spill Cleaning". *Industrial and Engineering Research*,dx.doi.org/10.1021/ie4032567

[7] Viet, H. P and James, H. D; 2014, "Superhydrophobic Silanized Melamine Sponges as High Efficiency Oil Absorbent Materials". American Journal of Science and Industrial Research, dx.doi.org/10.1021/am503503m| ACS Appl. Mater. Interfaces2014, 6, 14181−14188

[8] Singh, V.; Sudheer, Jinka; Kater, Hake; Siva, Parameswaran; Ronald, J. Kendall; Seshadri, Ramkumar; 2014, "Novel Natural Sorbent for Oil Spill Cleanup". Industrial and Engineering Research, dx.doi.org/10.1021/ie5019436| Ind. Eng. Chem. Res. 2014, 53, 11954−11961

[9] Bland, B. 2018. "Rescuers battle toxic oil blaze off China coast". Financial Times.

[10] IOGP, 2016. Controlled In-situ Burning of Spilled Oil, Good Practice Guidelines forIncident Management and Emergency Response Personnel. IOGP Report 523.

[11] Bayat, A.; Aghamiri, S. F.; Moheb, A.; Vakili-Nezhaad, G. R.; 2005, "Oil spill cleanup from sea water by sorbent materials.Chem. Eng. Technol. 2005, 28, 1525.

[12] Yuan, X and Mike Chung, T. C.; 2012, "Novel Solution to Oil Spill Recovery: Using Thermodegradable Polyolefin Oil Superabsorbent Polymer (Oil−SAP)" *American Journal of Science and Industrial Research*, dx.doi.org/10.1021/ef300388h| Energy Fuels2012, 26, 4896−4902.

[13] Azubuike, C.C., Chikere, C.B., Okpokwasili, G.C., 2016. Bioremediationtechniqueseclassification based on site of application: principles, advantages,limitations and prospects. World J. Microbiol. Biotechnol. 32, 180.

[14] Gui, X. C.; Wei, J. Q.; Wang, K. L.; Cao, A. Y.; Zhu, H. W.; Jia, Y.; Shu, Q. K.; Wu, D. H.; 2010, "Carbon Nanotube Sponges.Adv. Mater.(Weinheim, Ger.)" 2010 22, 617.

[15] Korhonen, J. T.; Kettunen, M.; Ras, R. H. A.; Ikkala, O.; 2011 "Hydrophobic Nanocellulose Aerogels as Floating, Sustainable, Reusable, and Recyclable Oil Absorbents". ACS Appl. Mater. Interfaces 2011, 3, 1813.

[16] Al-Majed, A. A.; Adebayo, A. R.; Hossain, M. E.; 2012, "A sustainable approach to controlling oil spills". J. Environ. Manage. 2012, 113, 213.

[17] Wahi, R.; Chuah, L. A.; Choong, T. S. Y.; Ngaini, Z.; Nourouzi, M. M.; 2013, "Oil removal from aqueous state by natural fibrous sorbent: An Overview". Sep. Purif. Technol. 2013, 113, 51.

[18] Laitinen, O., Ojala, J., Sirvi€ o, J.A., Henrikki, L., 2017a. Sustainable stabilization of oil inwater emulsions by cellulose nanocrystals synthesized from deep eutectic solvents. *Cellulose* 24, 1679-1689.

[19] Khan, E., Virojnagud, W., Ratpukdi, T., 2004. Use of biomass sorbents for oil removalfrom gas station runoff. Chemosphere 57, 681e689.

[20] Kizil, S., Sonmez, H.B., 2017. Oleophilic and hydrophobic gels made from glycerolpropoxylate: efficient and reusable sorbents for oil spill clean-up. *J. Environ. Manag.* 196, 330-339.

[21] Kalliola, S., et al., 2016. The stability of green nanoparticles in increased pH andsalinity for applications in oil spill-treatment. Colloid. Surface. Physicochem. Eng. Aspect. 493, 99-107.

[22] Laitinen, O., Suopajarvi, T.,Osterberg, M., Liimatainen, H., 2017b. Hydrophobic,superabsorbing aerogels from choline chloride-based deep eutectic solventpretreated and silylated cellulose nanofibrils for selective oil removal. *ACS Appl. Mater.* Interfaces 9, 25029-25037.

[23] Junlong, S; Rojas,O.J; 2013 "Approaching super-hydrophobicityfromcellulosic materials: AReview".Nordic Pulp & Paper Research Journal Vol 28 no. 2/2013, pp 216

[24] Bhairavi D.; Mika S.; Simo K. 2018.A review of bio-based materials for oil spill treatment. *Journal of Water Research* 135 (2018) 262-277

[25] Kollarigowda, R.H., Abraham, S., Montemagno, C.D., 2017. Antifouling cellulosehybrid biomembrane for effective oil/water separation. ACS Appl. Mater. Interfaces 9, 29812-29819.

[26] Wang, A., et al., 2017. The enhanced stability and biodegradation of dispersed crude oil droplets by Xanthan Gum as an additive of chemical dispersant. *Mar. Pollut.* Bull. 118, 275e280.

[27] Wu, Z.-Y., et al., 2017. Carbon nanofiber aerogels for emergent cleanup of oil spillageand chemical leakage under harsh conditions. *Sci. Rep.* 4, 4079

[28] Arfaoui, MA.; Dolez, PI.; Dube, M.; David, E. 2017. Development and characterization of hydrophobic treatment for jute fibres based on zinc oxide nanoparticles and a fatty acid. Appl. Surface Sci.397, 19-29.

[29] Na, L.; Xiaoli, W., Shitao, P.; Huaqin, Z.; Lei, L. 2018. Study of the kinetics and equilibrium of the adsorption of oils onto hydrophobic jute fiber modified via the sol-gel method. *International journal of environmental research and public health.*

[30] Salisu, Z.; Umaru, IS.; Danladi, A.; Yakubu, MK.; Diya'uddeen, BH. 2019b.Recovery of crude oil from aqueous medium by optimised styrene/kenaf shive graft-based sorbent via regeneration method: study of the equilibrium, kinetics and activation energy. *World Journal of Innovative Research (WJIR)*. Unpublished paper.

[31] Diya'uddeen, BH, Abdul Aziz, AR, Wan Mohd Ashri Wan Daud, 2011 "Oxidative mineralisation of petroleum refinery effluent using Fenton-like process". Institute of Chemical Engineers,ChemE, Elsevier doi: 10.1016/j.cherd.2011.06.010

[32] Diya'uddeen, BH, Shima Rahim Pouran, Abdul Aziz AR, Daud, WMAW, 2015 "Fenton oxidative treatment of petroleum refinery wastewater: process optimization and sluge characterisation". Royal Society of Chemistry Advance 2015, 5, 68159-681668.

[33] Akhabue, C.E., Osarumwense, J.O., Ekhator, O.H., Oyekunle, D.T., Oyedoh, E.A. 2018. Optimizing Ethanol Concentration from Orange Peel Wastes using Response Surface Methodology. *Nigerian Research Journal of Engineering and Environmental Sciences* 3(1) 2018 pp. 198-206. www. Rjees.com

[34] Osakue, I.Y.; Amenaghawon, N.A.; 2018. Response surface methodology for optimizing alkaline hydrogen peroxide pretreatment of corn stover. *Nigerian Research Journal of Engineering and Environmental Sciences* 3(1) 2018 pp. 118-124.

[35] Taylor, S. Charles, March 2004, "Kenaf – New Crop Fact Sheet", Kenaf International Ltd., McAllen TX, http://www.hort.purdue.edu/newcrop/CropFactSheets/kenaf.html.

[36] Mitul, Zaveri Dilip, 2004. "Absorbency characteristics of kenaf core particles". M. Sc. Thesis, Department of Textle Apparel Technology and Management, Raleigh, College of Textiles North Carolina State University.

[37] Gürdag, G, Sarmad, S 2013. "Cellulose grafting copolymers: synthesis, properties and application". Polysaccharide based graft copolymer, DOI 10.1007/978-3-642-36566-9_2, Springer-Verlag Berlin Heidelberg.