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Percentage adsorption of Chlorpheniramine, Ibuprofen and Glipizide from deionized water and spiked Pharmaceutical Liquid Waste (sPLW)

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ABSTRACT

The undesirable effects of release of some pharmaceutical drugs into the environment have been a serious public health concern. This study was aimed at investigating percentage adsorption of Chlorpheniramine, Ibuprofen and Glipizide from deionized water and spiked Pharmaceutical Liquid Waste using oxidized activated carbons (OAC), hydrophobic activated carbons (HAC) and basic activated carbons (BAC) prepared from mango kernel and avocado pear seeds. Activated carbons (ACs) were prepared from mango kernel seed and avocado pear seed, through KOH activation. The AC was oxidized with HNO_3 to produce OACs that were surface functionalized using ethylene diamine to produce BACs and ethylamine to produce HACs. The adsorption capacity, qe and % adsorbed with time, from the sPLW and deionized water were determined and compared. Drug adsorption depends mainly on solution pH and the adsorbent surface nature, and initial pH 7 was found optimal for the removal of the three drugs. The Chlorpheniramine uptake follows the order: HAC > OAC > BAC for mango kernel seed and avocado pear seed, whereas the adsorption of Ibuprofen and Glipizide follow the order: OAC > HAC > BAC for the two adsorbent materials. The adsorption capacity, qe and % adsorbed with time, from the sPLW of the different carbons follow similar order to that of Chlorpheniramine, Ibuprofen and Glipizide, from deionized water. The percentage removal of drugs as function of sample treatment (OAC, HAC, BAC), drug type (CHP, IBU, GLI), and sample type (Mango kernel seed, Avocado pear seed), showed that there is a significant difference in % removal from main effect of sample treatment at F= 54.633, P value = 0.000. Drug adsorption from spiked Pharmaceutical Liquid Waste (PLW), showed slightly less capacity than that from deionized water but the same trend in the percentage adsorbed by the different carbons as in the deionized water. Such decrease in drug uptake from sPLW is probably because of the competition of dissolved organic substances available in sPLW, with Chlorpheniramine, Ibuprofen and Glipizide molecules, for adsorption sites on the adsorbents. OAC, HAC, BAC prepared from mango kernel and avocado pear seeds displayed good capability for drug removal from spiked pharmaceutical liquid waste. Therefore, surface functionalization of activated carbon using mango kernel and avocado pear seeds showed a promising solution for pharmaceutical removal from deionized water as well as pharmaceutical liquid waste.

Keywords: Chlorpheniramine, Glipizide, deionized water, percentage adsorption, spiked Pharmaceutical Liquid Waste.

INTRODUCTION

Discharge of drug active ingredients into nearby waterways by pharmaceutical companies can lead to environmental pollution. The undesirable effects of release of some pharmaceutical drugs into the environment have been a serious public health concern [1]. Thus, researchers have been making concerted efforts towards development of effective methods for the adsorption of pharmaceutical drugs. Chlorpherinamine is an antihistamine used to treat

the symptoms of allergic diseases [2]. Some undesirable effects of chlorpherinamine intoxication include drowsiness, dizziness, confusion, constipation, anxiety, nausea, blurred vision, restlessness, decreased coordination, dry mouth, shallow breathing, hallucinations, irritability, memory or concentration problems, tinnitus, and difficulty urinating [2]. Chlorpheniramine maleate has been shown to increase oxidative stress and hence cardiotoxic and hepatotoxic in young male Wistar rats [3].Ibuprofen is commonly used to treat pain, fever, and various inflammatory disorders [4]. Reports of ibuprofen toxicology in fish are well-documented [5-9].

Glipizide belongs to the group of medications known as sulfonylureas. Glipizide boosts the natural release of insulin by the pancreas. Thus, it is very useful in the management of high blood sugar [10]. Glipizide is one of the commonly produced drugs in the pharmaceutical industries whose effluent into the environment can lead to environmental pollution. Commonest side effect of Glipizide is hypoglycemia (severe low blood sugar) which can cause loss of consciousness, seizures, or death in some cases [10].

Mango fruit is produced from the tropical tree *Mangifera indica*. It is a common edible fruit present in many countries including Nigeria. However, it originated in Myanmar, Bangladesh, and northeastern India. Mangos are members of the family Anacardiaceae's genus *Mangifera* [11]. Mango fruit contains seeds which are often discarded after eating the fruit or used by industries for various purposes [12]. The avocado tree *Persea americana*, is native to south-central Mexico. Its fruit is known as an avocado. It can also be called avocado pear or an alligator pear and usually has a large berry with a sizable seed. It is self-pollinating plant and usually spread by grafting so that the number and quality of its fruits stay the same [13]. Despite the enormous medical importance of drugs, its discharge into the environment produces undesirable results to man and his environment. Therefore, development of effective method for the adsorption and elimination of pharmaceutical drugs is of utmost importance. Surface modification of activated carbon has been crucial in this quest [14, 15]. Surface functionalization can introduce adsorption forces such as H-bonding, electrostatic interaction, and hydrophobic bonding. Hence, activated carbon surface can be prepared to make use of such adsorption forces via chemical modification. This study was designed to investigate the percentage adsorption of chlorpheniramine, ibuprofen, and glipizide from deionized water and spiked pharmaceutical liquid waste using activated carbon, oxidized activated carbon, and surface functionalized activated carbon prepared from mango kernel seed and avocado pear seed.

MATERIALS AND METHODS

Materials

All chemicals used were of analytical grade. Mango kernel seed and Avocado pear seed were collected from Orji village, in Udi Local Government area, Enugu State, Nigeria. They were athenticated by a Taxonomist in Botany Department of Nnamdi Azikiwe University, Awka.

METHODS

Preparation of Activated carbon (AC)

Clean dry seeds (25g) were charred differently in a carbon steel tube (internal diameter 5.1 cm and length 61 cm) that was heated in a tube furnace (GSL-1100X-110V, MTI Corporation, USA) under a nitrogen atmosphere at 500 oC for 2 hours. In a weight ratio of 1:3, the chars were impregnated with saturated KOH solution. The mixtures were left in the oven (Hobersal Mon X B2-125 furnace, Hobersal, Spain) overnight at 120^{o}C before being transferred to the tube furnace. The temperature was raised from room temperature to 550^{o}C at a heating rate of ~8.6^{o}C/min and was kept at 550^{o}C for 1 hour under nitrogen for activation. The ACs produced are washed thoroughly with deionized water to remove residual alkalinity. To keep the acidic functional groups on the carbon in H-form, ACs were washed with 0.1M

HCl followed by deionized water until no acidity was detected in the wash water. All the ACs of mango kernel and avocado kernel seeds (MKS and AKS) were dried at 120°C until they reached a constant weight. After cooling in a desiccator and grinding, a size range of each between two sieves of 1.19 mm and 0.25 mm was selected for characterization.

Surface modification of activated carbon AC

Oxidized activated carbons (OAC), hydrophobic activated carbons (HAC) and basic activated carbons (BAC) prepared from mango kernel and avocado pear seeds. Activated carbons (ACs) were prepared from mango kernel seed and avocado pear seed, through KOH activation. The AC was oxidized with HNO_3 to produce OACs that were surface functionalized using ethylene diamine to produce BACs and ethylamine to produce HACs.

Preparations of Stock Solutions of Chlorpheniramine, Glipizide, and Ibuprofen:

Chlorpheniramine: A stock solution containing 50mg/L chlorpheniramine in maleate form, was prepared by dissolving 50mg of chlorpheniramine in deionized water in a 1000mL volumetric flask.

Glipizide: An initialdiluent was prepared through a mixture of water, acetonitrile, and methanol (3:1:1) and a mobile phase consisting of acetonitrile: 0.01M potassium di-hydrogen phosphate buffer (pH 3.5) in a ratio of 35:65, which was degassed by sonification. A stock solution containing 50mg/L glipizide was prepared by accurately weighing about 50mg of glipizide and transferring the same into a 1000mL volumetric flask. Adding 50mL of diluent and keeping it in an ultrasonic bath until it dissolved completely. Make it up to the mark with the mobile phase and mix.

Ibuprofen: In a 1000mL volumetric flask, a stock solution containing 50mg/L ibuprofen was prepared by dissolving 50mg of ibuprofen in some 20% methanol to achieve complete solubility with deionized water. Adsorption experiments involving temperature, concentration, pH, and contact time variations will be carried out after serial dilution of standard solutions.

Drug analysis: High performance liquid chromatography (HPLC) equipped with a diode array detector (Agilent technologies, 1260 Infinity Series, USA) was used for the analysis of chlorpheniramine, ibuprofen, and Glipizide, at λmax 260 nm. The drugs were separated using a C18 analytical column and a mobile phase consisting of methanol and 20mm ammonium format buffer (pH 4.8) in a gradient elution mode with a flow rate of 45 µL/min and a column temperature of 40 °C. Calibration standards of the three drugs (1–20 mg/L) were prepared and standard curves were obtained by linear regression of the mean values of peak areas. Retention times for Chlorpheniramine, Ibuprofen, and Glipizide were found to be 2, 6.5, and 7 minutes, respectively. For chlorpheniramine, the linear range was found to be between 1–20 mg/L (R2: 0.9995). For Ibuprofen, the linear range was found to be between 1–20 mg/L (R2: 0.9996). For Glipizide, the linear range was found to be between 1–20 mg/L (R2: 0.9994). The accuracy of the method of analysis shows more than 98.2% recovery for both drugs [16].

Determination of the percentage adsorption of Chlorpheniramine, Ibuprofen and Glipizide, present in the selected spiked Pharmaceutical Liquid Waste (PLW):

Several samples of Pharmaceutical Liquid Waste (PLW) were collected from the effluents of Gauze Pharmaceutical and Juhel Pharmaceutical companies, both in Awka, Anambra state of Nigeria, in a working week day. PLW samples were kept in ice during transport and were filtered using membrane filter (0.45 µm pore size). PLW filtrate samples were mixed together in equal volumes making a representative sample. For the adsorption of Chlorpheniramine, Ibuprofen and Glipizide from spiked PLW, samples of the stock solution of Chlorpheniramine, Ibuprofen and Glipizide, were spiked with the filtered PLW to achieve the range of initial concentrations as in the study of equilibrium adsorption from deionized water mentioned above [16]. 0.06 g each of the carbons (OACs, HACs, BACs) prepared from mango kernel see and avocado pear seed, were mixed

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with25 mL of spiked drug solutions and was left at 25 °C under mechanical agitation at 30, 60, 90, 120, 150, and 180 minutes. After equilibrium was obtained, samples of supernatant were separated and analyzed.

RESULTS AND DISCUSSION

Table 1: Percentage adsorption of CHP from deionized water and sPLW using OAC, HAC, and BAC prepared with mango kernel seed.

time		qe (Deionized)			% Removal (Deionized)			qe (sPLW)		% Removal (sPLW)					
time	sqrt (t)	qe $_{(OAC)}$	qe HAC	qe BAC	$\%_{(OAC)}$	%HAC	%BAC	HAC	BAC	qe(OAC)	qe(HAC	qe(BAC)	%OAC	%HAC	%BAC
30	5.477226	14	18	7	56	72	28	0.30429	0.782461	13.6	17.8	6.8	54.4	71.2	27.2
60	7.745967	16.8	19	10	67.2	76	40	0.407682	0.774597	15.2	18.7	9.6	60.8	74.8	38.4
90	9.486833	17	19.2	11.6	68	76.8	46.4	0.494106	0.81783	16.4	18.95	10.2	65.6	75.8	40.8
120	10.95445	17.2	19.5	11.84	68.8	78	47.36	0.561767	0.925207	16.8	19.45	10.8	67.2	77.8	43.2
150	12.24745	17.45	19.8	11.9	69.8	79.2	47.6	0.618558	1.029197	17.25	19.76	11.5	69	79.04	46
180	13.41641	17.5	17.5	17.5	70	70	70	0.766652	0.766652	17.35	17.35	17.35	69.4	69.4	69.4

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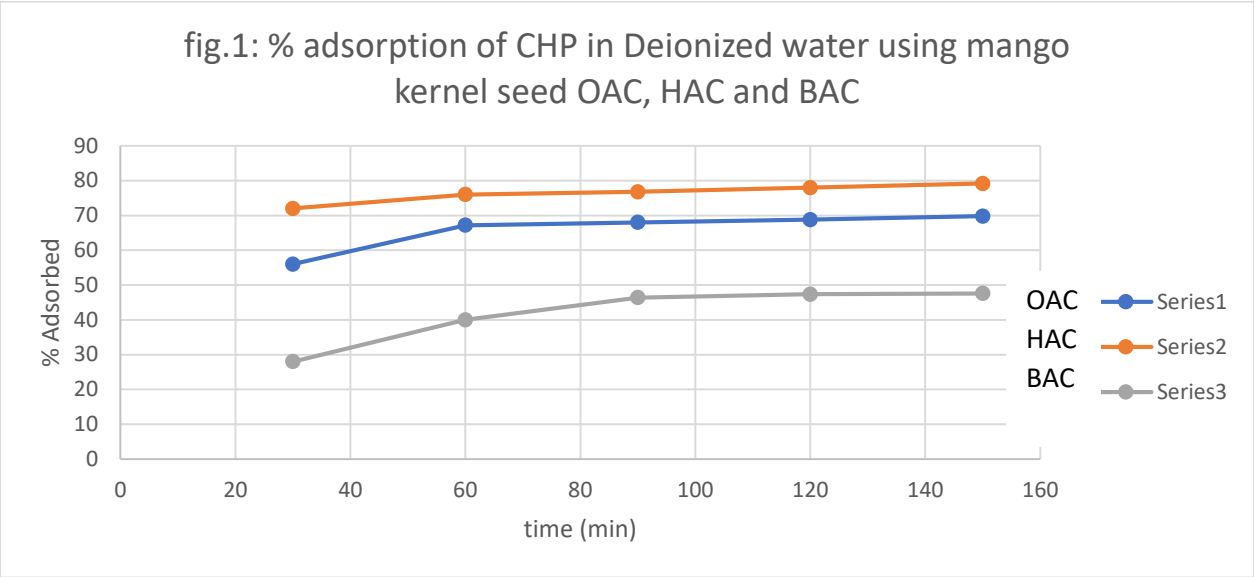
fig.1: % adsorption of CHP in Deionized water using mango kernel seed OAC, HAC and BAC
% Adsorbed
time (min)
OAC
HAC
BAC
Series1
Series2
Series3

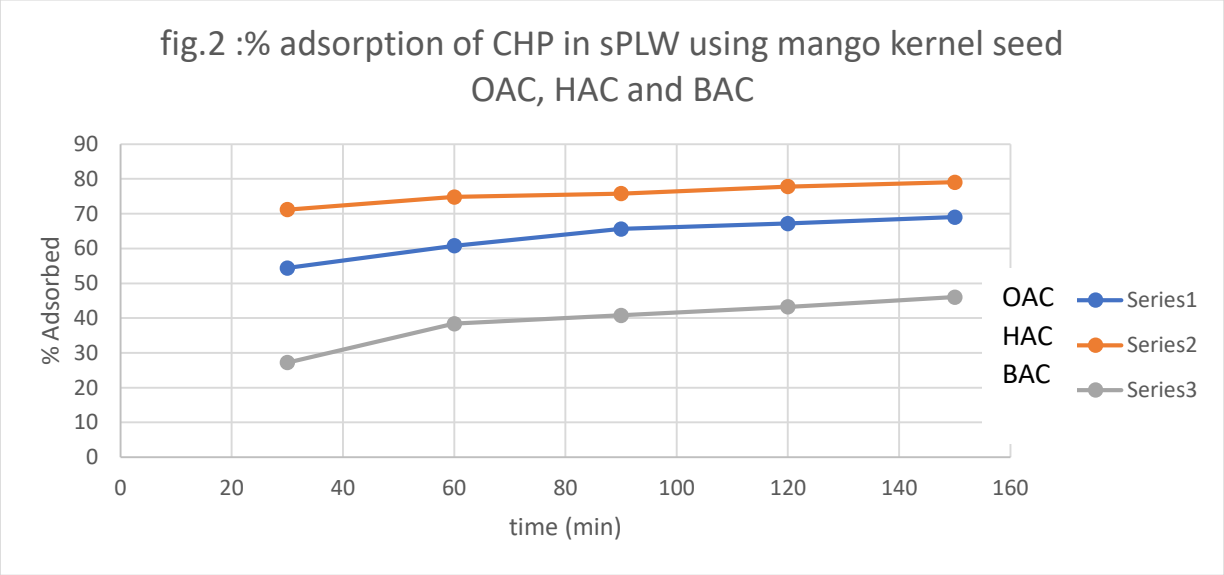
fig.2 :% adsorption of CHP in sPLW using mango kernel seed OAC, HAC and BAC
% Adsorbed
time (min)
OAC
HAC
BAC
Series1
Series2
Series3

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Table 2: Percentage adsorption of IBU from deionized water and sPLW using OAC, HAC, and BAC prepared with mango kernel seed.

Time		qe (Deionized)			% Removal (Deionized)			qe (sPLW)			% Removal (sPLW)		
Time	sqrt (t)	qe $_{(OAC)}$	qe HAC	qe BAC	$\%_{(OAC)}$	%HAC	%BAC	qe(OAC)	qe(HAC	qe(BAC)	%OAC	%HAC	%BAC
30	5.477226	17.5	13.5	7.5	70	54	30	16.9	13.18	6.77	67.6	52.72	27.08
60	7.745967	18	15	10	72	60	40	17.8	14.2	9.6	71.2	56.8	38.4
90	9.486833	19.8	15.5	10.8	79.2	62	43.2	19.45	14.76	10.26	77.8	59.04	41.04
120	10.95445	20.2	16	11.2	80.8	64	44.8	19.87	15.66	10.75	79.48	62.64	43
150	12.24745	20.43	16.4	11.32	81.72	65.6	45.28	20.16	15.94	10.94	80.64	63.76	43.76
180	13.41641	20.5	16.5	11.42	82	66	45.68	21.1	16.03	11.03	84.4	64.12	44.12

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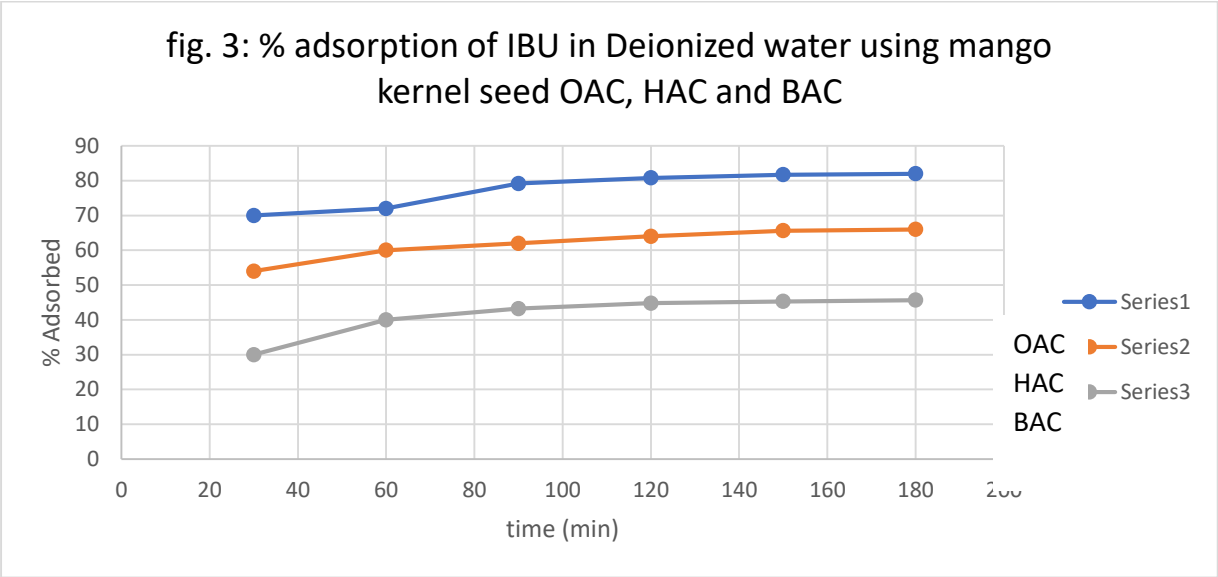
fig. 3: % adsorption of IBU in Deionized water using mango kernel seed OAC, HAC and BAC
% Adsorbed
time (min)
Series1
Series2
Series3
OAC
HAC
BAC

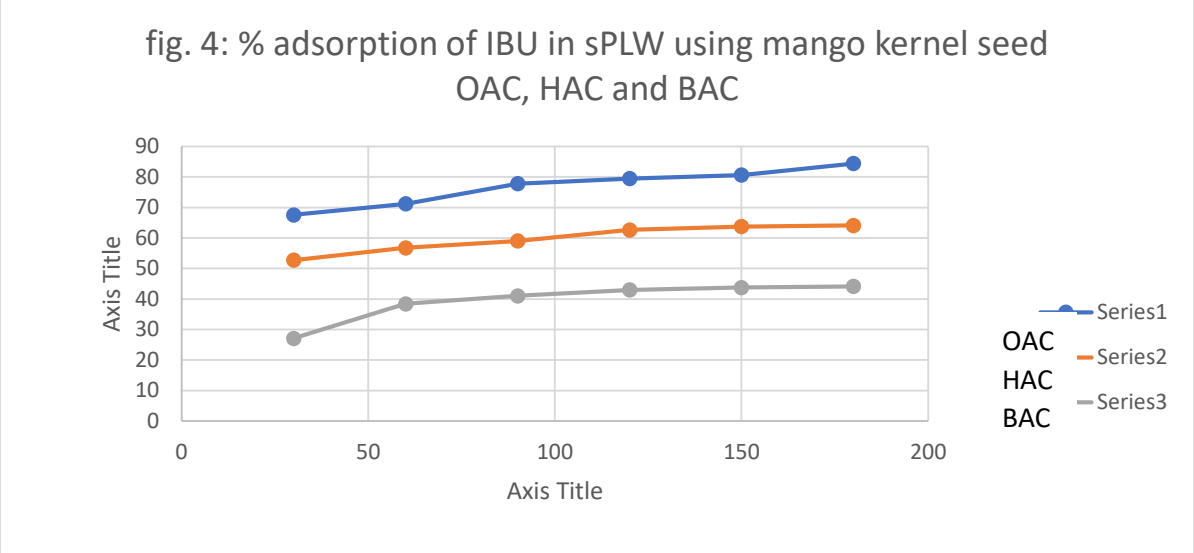
fig. 4: % adsorption of IBU in sPLW using mango kernel seed OAC, HAC and BAC
Axis Title
Axis Title
Series1
Series2
Series3
OAC
HAC
BAC

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Table 3: Percentage adsorption of GLI from deionized water and sPLW using OAC, HAC, and BAC prepared with mango kernel seed.

Time		qe (Deionized)			% Removal (Deionized)			qe (sPLW)			% Removal (sPLW)		
Time	sqrt (t)	qe $_{(OAC)}$	qe HAC	qe BAC	$\%_{(OAC)}$	%HAC	%BAC	qe(OAC)	qe(HAC	qe(BAC)	%OAC	%HAC	%BAC
30	5.477226	21	11.4	6.4	84	45.6	25.6	20.12	10.45	5.4	80.48	41.8	21.6
60	7.745967	22.5	12.5	9.3	90	50	37.2	21.2	11.08	8.12	84.8	44.32	32.48
90	9.486833	23.6	12.8	9.85	94.4	51.2	39.4	21.8	12.24	8.78	87.2	48.96	35.12
120	10.95445	24.4	13	10.2	97.6	52	40.8	23.6	12.76	9.37	94.4	51.04	37.48
150	12.24745	24.6	13.35	10.25	98.4	53.4	41	24.23	13.08	9.66	96.92	52.32	38.64
180	13.41641	24.85	13.5	10.42	99.4	54	41.68	24.5	13.24	9.84	98	52.96	39.36

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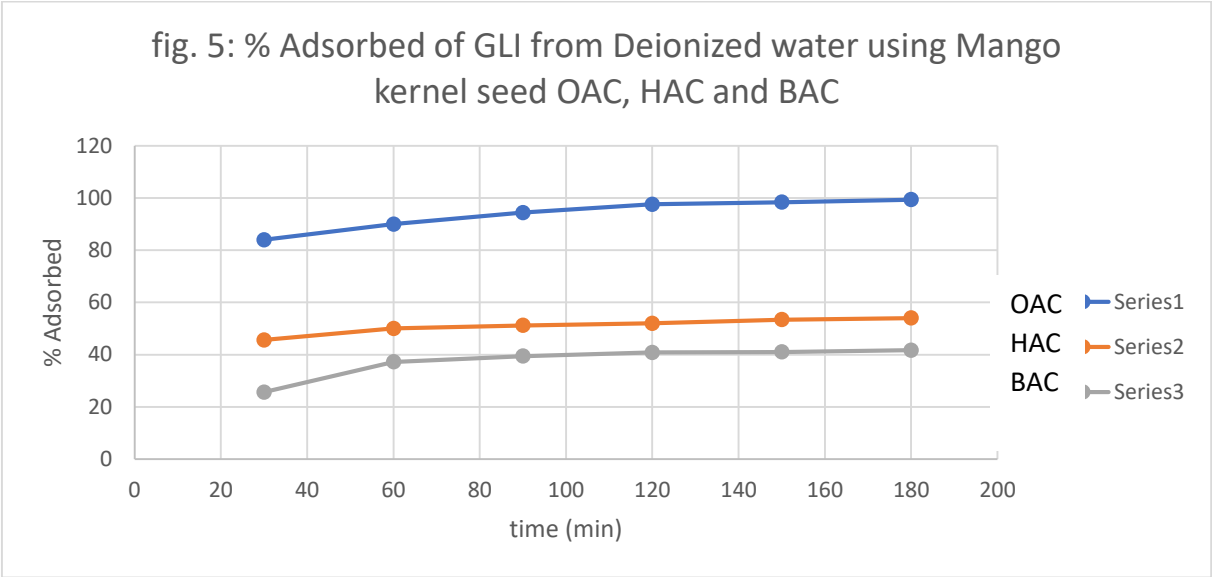
fig. 5: % Adsorbed of GLI from Deionized water using Mango kernel seed OAC, HAC and BAC
% Adsorbed
time (min)
OAC
HAC
BAC
Series1
Series2
Series3

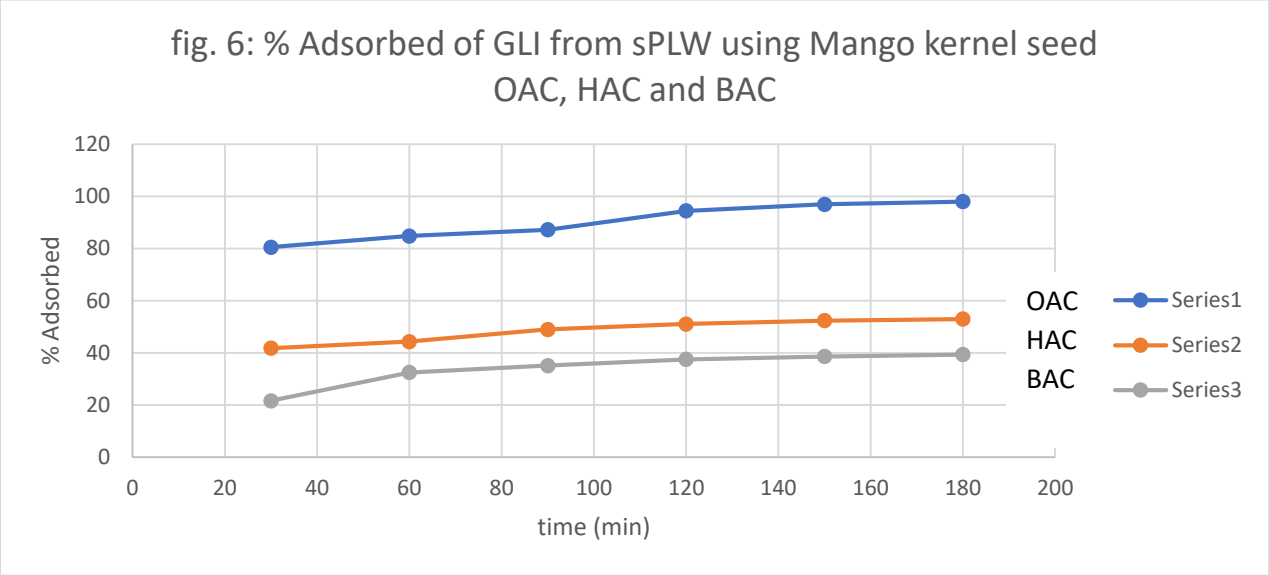
fig. 6: % Adsorbed of GLI from sPLW using Mango kernel seed OAC, HAC and BAC
% Adsorbed
time (min)
OAC
HAC
BAC
Series1
Series2
Series3

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Table 4: Percentage adsorption of CHP from deionized water and sPLW using OAC, HAC, and BAC prepared with avocado pear seed.

Time		qe (Deionized)			% Removal (Deionized)			qe (sPLW)			% Removal (sPLW)		
Time	sqrt (t)	qe $_{(OAC)}$	qe HAC	qe BAC	$\%_{(OAC)}$	%HAC	%BAC	qe(OAC)	qe(HAC	qe(BAC)	%OAC	%HAC	%BAC
30	5.477226	17.28	21.5	8.4	69.12	86	33.6	17.12	21.23	7.2	68.48	84.92	28.8
60	7.745967	18.55	22.45	10.6	74.2	89.8	42.4	18.24	22.2	9.2	72.96	88.8	36.8
90	9.486833	19.75	22.74	11.75	79	90.96	47	18.88	19.24	9.46	75.52	76.96	37.84
120	10.95445	19.9	23.5	13.23	79.6	94	52.92	19.34	19.5	11.68	77.36	78	46.72
150	12.24745	20.1	23.66	13.4	80.4	94.64	53.6	20.04	19.87	11.8	80.16	79.48	47.2
180	13.41641	20.28	23.97	13.45	81.12	95.88	53.8	20.2	20	12.04	80.8	80	48.16

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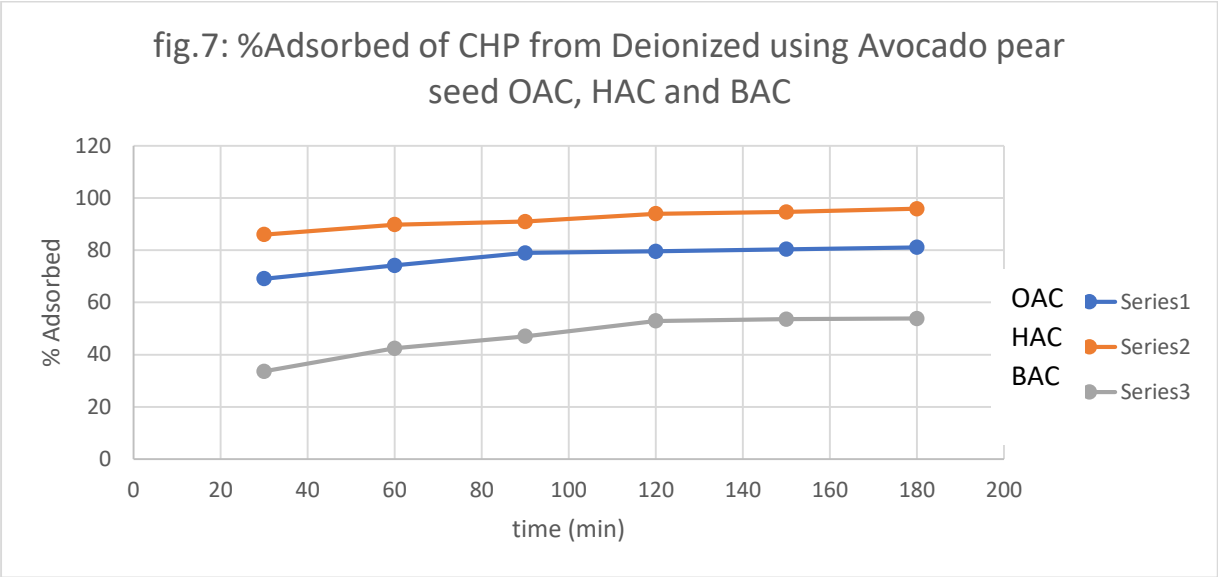
fig.7: %Adsorbed of CHP from Deionized using Avocado pear seed OAC, HAC and BAC
% Adsorbed
time (min)
OAC
HAC
BAC
Series1
Series2
Series3

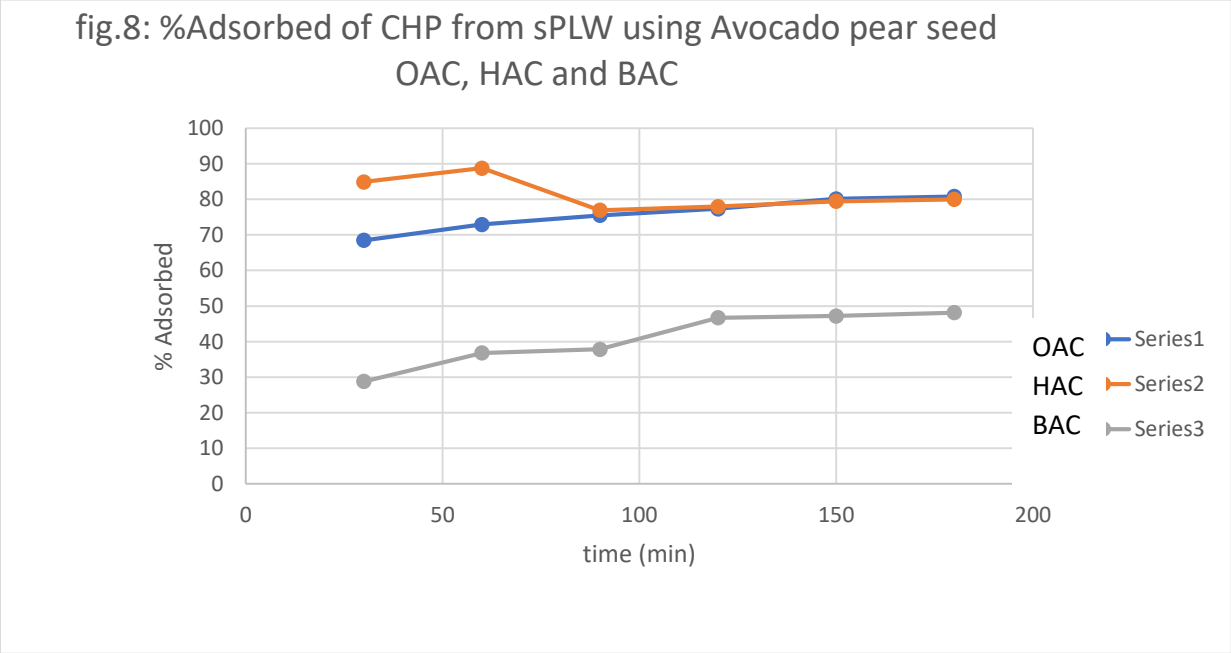
fig.8: %Adsorbed of CHP from sPLW using Avocado pear seed OAC, HAC and BAC
% Adsorbed
time (min)
OAC
HAC
BAC
Series1
Series2
Series3

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Table 5: Percentage adsorption of IBU from deionized water and sPLW using OAC, HAC, and BAC prepared with avocado pear seed.

Time		qe (Deionized)			% Removal (Deionized)			qe (sPLW)			% Removal (sPLW)		
Time	sqrt (t)	qe $_{(OAC)}$	qe HAC	qe BAC	$\%_{(OAC)}$	%HAC	%BAC	qe(OAC)	qe(HAC)	qe(BAC)	%OAC	%HAC	%BAC
30	5.477226	15.4	13.45	6.5	61.6	53.8	26	14.8	13.2	5.6	59.2	52.8	22.4
60	7.745967	16.3	14.66	11.7	65.2	58.64	46.8	15.6	13.85	10.54	62.4	55.4	42.16
90	9.486833	16.8	15.4	11	67.2	61.6	44	16	15.06	10.4	64	60.24	41.6
120	10.95445	17.2	15.95	11.26	68.8	63.8	45.04	16.7	15.7	10.8	66.8	62.8	43.2
150	12.24745	17.25	16.04	11.56	69	64.16	46.24	16.94	15.95	11.26	67.76	63.8	45.04
180	13.41641	17.37	16.21	11.98	69.48	64.84	47.92	17.15	16.45	11.54	68.6	65.8	46.16

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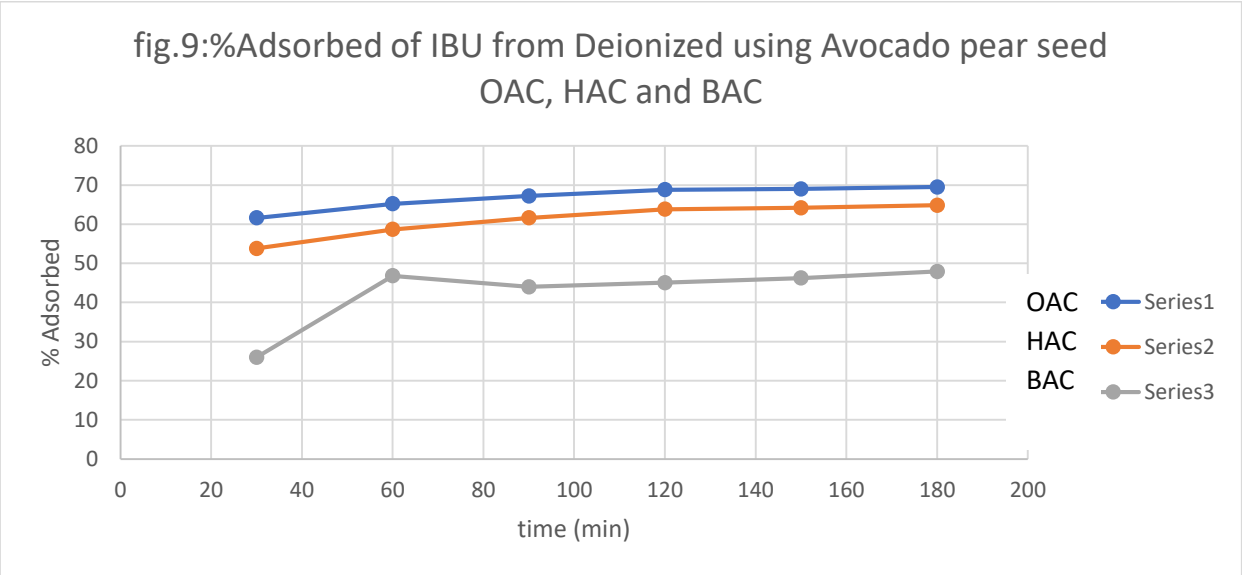
fig.9:%Adsorbed of IBU from Deionized using Avocado pear seed
OAC, HAC and BAC
% Adsorbed
time (min)
OAC
HAC
BAC
Series1
Series2
Series3

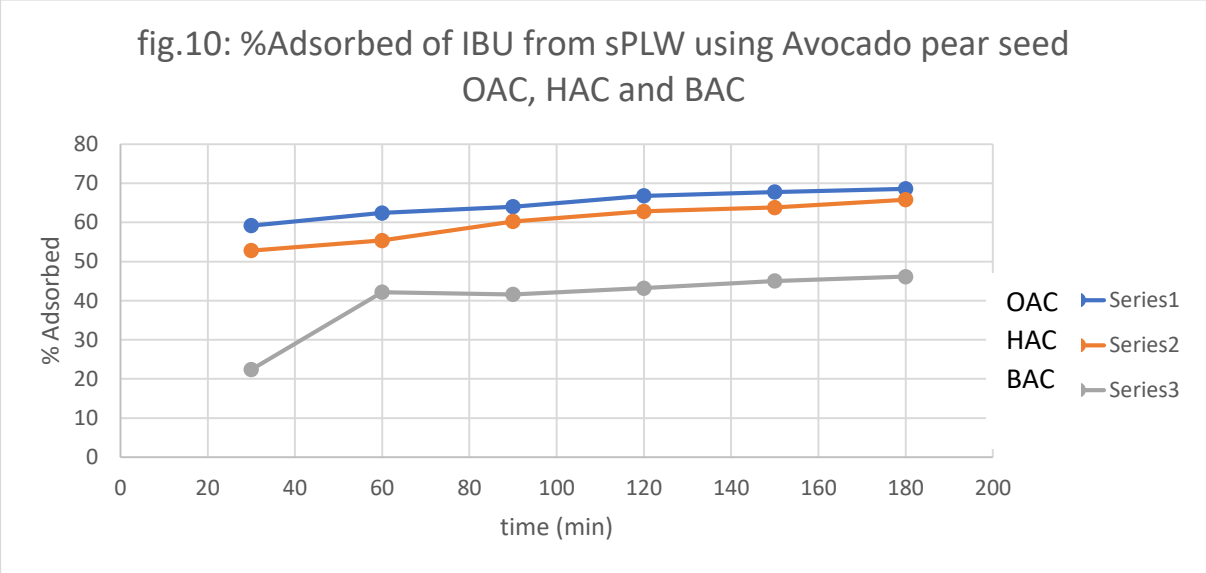
fig.10: %Adsorbed of IBU from sPLW using Avocado pear seed
OAC, HAC and BAC
% Adsorbed
time (min)
OAC
HAC
BAC
Series1
Series2
Series3

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Table 6: Percentage adsorption of GLI from deionized water and sPLW using OAC, HAC, and BAC prepared with avocado pear seed.

Time		qe (Deionized)			% Removal (Deionized)			qe (sPLW)			% Removal (sPLW)		
Time	sqrt (t)	qe $_{(OAC)}$	qe HAC	qe BAC	$\%_{(OAC)}$	%HAC	%BAC	qe(OAC)	qe(HAC	qe(BAC)	%OAC	%HAC	%BAC
30	5.477226	19.5	9.62	5.6	78	38.48	22.4	19.24	9.57	5.02	76.96	38.28	20.08
60	7.745967	20.4	10.5	8.45	81.6	42	33.8	19.87	10.28	7.42	79.48	41.12	29.68
90	9.486833	20.93	10.8	8.67	83.72	43.2	34.68	20.6	10.62	7.45	82.4	42.48	29.8
120	10.95445	21.6	11.02	9.5	86.4	44.08	38	20.96	10.94	8.46	83.84	43.76	33.84
150	12.24745	21.68	11.14	9.55	86.72	44.56	38.2	21.46	11	9.2	85.84	44	36.8
180	13.41641	21.7	11.2	9.57	86.8	44.8	38.28	21.5	11.14	9.4	86	44.56	37.6

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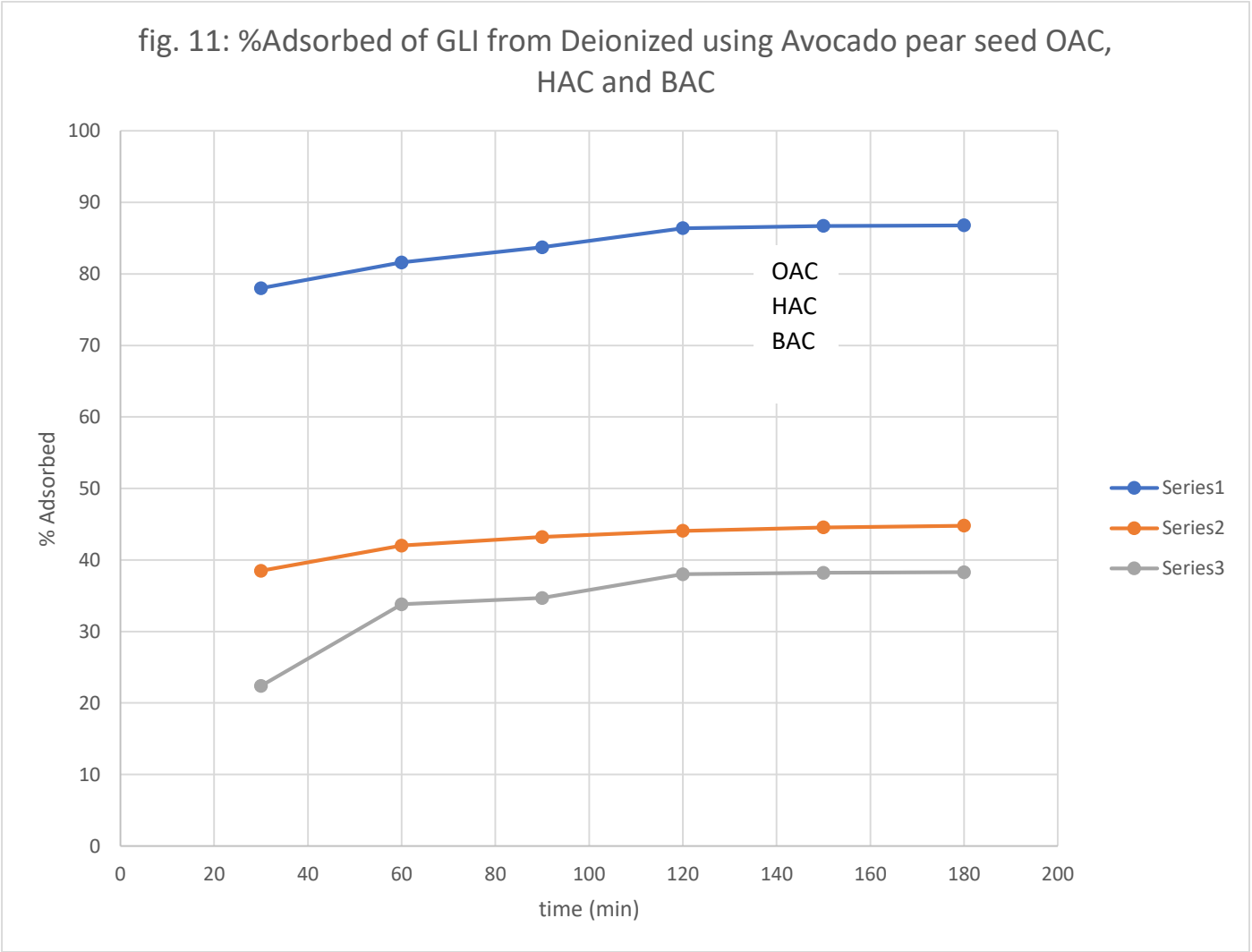
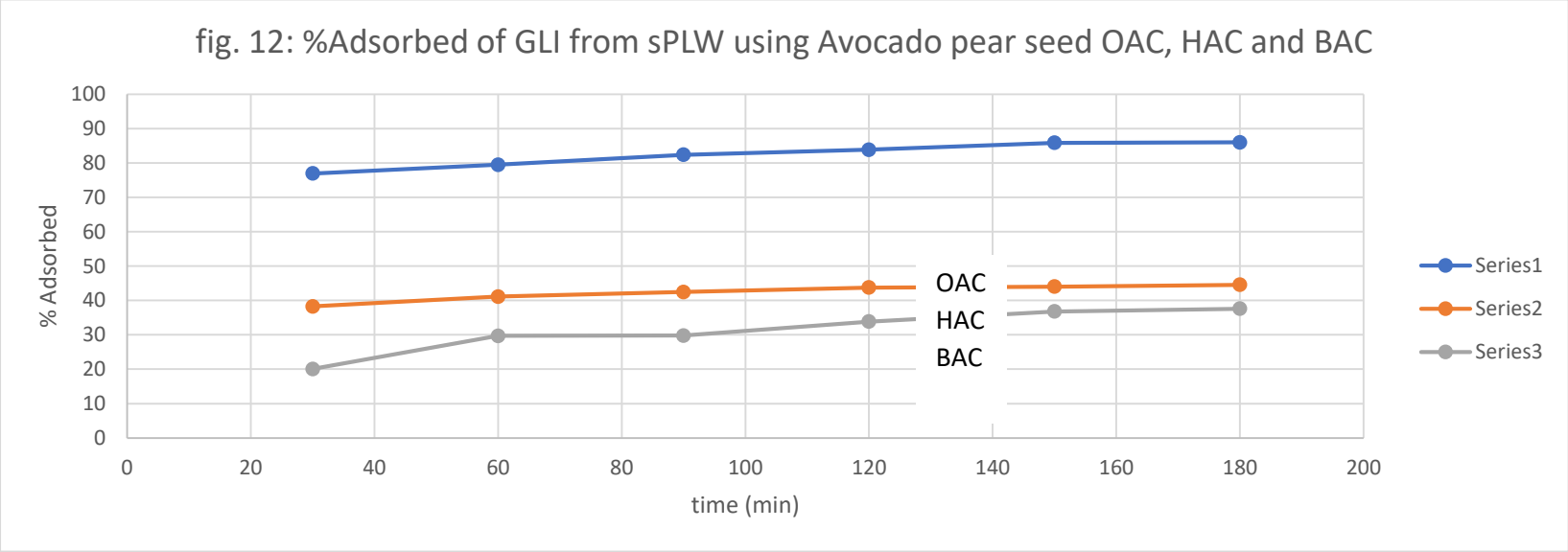


fig. 11: %Adsorbed of GLI from Deionized using Avocado pear seed OAC, HAC and BAC
% Adsorbed
time (min)
OAC
HAC
BAC
Series1
Series2
Series3
0
10
20
30
40
50
60
70
80
90
100
0
20
40
60
80
100
120
140
160
180
200



A plot of the percentage adsorption with time of chlorpheniramine, ibuprofen, and glipizide on the carbons of mango kernel seed and avocado pear seed, from spiked PLW at 25 °C is shown in the figures 1-12 above. The adsorption data showed a gradual increase in the percentage adsorbed with time. This is in agreement with that obtained from the drugs in deionized water, as can be seen in the Tables 1-6. The adsorption capacity, qe, and % Adsorbed with time, from the spiked PLW of the different carbons, follow a similar order to that of chlorpheniramine, ibuprofen, and glipizide, from deionized water. For carbons of mango kernel seed on chlorpheniramine, Ibuprofen and Glipizide, from both deionized water and spiked PLW, the trends are: HAC > OAC > BAC, OAC > HAC > BAC, OAC > HAC > BAC, respectively. This could be due to the expected domination of the hydrophobic interactions between immobilized ethyl chains of HAC and the hydrophobic parts of the chlorpheniramine molecule. For carbons of avocado pear seed on chlorpheniramine, Ibuprofen and Glipizide, from both deionized water and spiked PLW, the trends are: HAC > OAC > BAC, OAC > HAC > BAC, OAC > HAC > BAC, respectively. As previously discussed in mango kernel seed carbons, the reason for the trend is same.

CONCLUSION

OAC, HAC, BAC prepared from mango kernel and avocado pear seeds displayed good capability for drug removal from spiked pharmaceutical liquid waste. Therefore, surface functionalization of activated carbon using mango kernel and avocado pear seeds showed a promising solution for pharmaceutical removal from deionized water as well as pharmaceutical liquid waste.

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