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Comparative Analysis on Adsorption of Glipizide from Deionized and Spiked Pharmaceutical Liquid Waste Using Surface Functionalized Activated Carbon from Velvet Tamarind Shell

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ABSTRACT

Glipizide (GLI) is one of the commonly produced drugs in the pharmaceutical industries in our environment. The adverse effect as unused drugs or expired drugs on humans, animals, aquatic life and environment is a serious public health problem. This study involved the development of a surface functionalized Activated Carbon (AC) from velvet tamarind shell. The AC was oxidized with HNO_3 toproduce oxidized activated carbons (OAC) that were surface functionalized using ethylene diamine to producebasic surfaces (BAC) and ethylamine to produce hydrophobic carbonaceous surfaces (HAC).The adsorption of GLIin deionized water and spiked pharmaceutical liquid waste each, on these carbons were investigated using quantities adsorbed/adsorption capacity (qe) and percentage (%) of the drug adsorbed. The adsorption capacity, q_e and %Adsorbed with time, of GLI from the spiked Pharmaceutical Liquid Waste (PLW), of the different carbons follow similar order to that from deionized water. The adsorption of GLI from both deionized water and spiked PLW, using the carbons, show the trend: HAC > OAC > BAC, both for adsorption capacity (qe) and % adsorption .GLI adsorption from spiked Pharmaceutical Liquid Waste (PLW), showed slightly less capacity than that from deionized water but the same trend and the different percentage adsorbed showed significant difference having $P<0.05$.

Keywords: Glipizide, public health and Activated Carbon (AC)

INTRODUCTION

Glipizide belongs to the class of drugs known as sulfonylureas. It lowers blood sugar by causing the release of your body's natural insulin. Common side effects include Nausea, vomiting,loss of appetite, diarrhea, constipation, upset stomach, headache, and weight gain may occur. Its common side effects include Nausea, vomiting, loss of appetite, diarrhea, constipation, upset stomach, headache and weight gain may occur. Activated carbon, also called activated charcoal, is a form of carbon processed to have small, low-volume pores that increase the surface area available for adsorption or chemical reactions [1]. *Activated* is sometimes substituted with *active*.Due to its high degree of microporosity, one gram of activated carbon has a surface area in excess of 3,000 m^2 (32,000 sq ft) as determined by gas adsorption [2]; [3]. In this study, surface functionalized activated carbon were produced from velvet tamarind shell.

MATERIALS AND METHODS

All chemicals used were of analytical grade. Pure sample ofGLI was supplied by Sigma Aldrich. 25 g of clean dry shells were charred 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 nitrogen atmosphere at 500°C for 2 hours. The chars were impregnated in saturated KOH solution in a weight ratio of 1:3.The mixtures were left in the oven (Hobersal Mon X B2-125 furnace, Hobersal, Spain) overnight at 120°C before being transferred to the tube furnace described above. The temperature was raised from room temperature to 550°C in a heating rate of ~8.6°C/min and was kept at 550°C for 1 hour under nitrogen for activation. Activated Carbon (AC) produced was washed thoroughly with deionized water to remove residual alkalinity. To keep the acidic functional groups on the carbon in H-form, AC was washed with 0.1M HCl followed by deionized water until no acidity was detected in wash water. AC of velvet tamarind shell (VTS) was left to dry at 120°C until constant weight was obtained. After cooling in a desiccator and grinding, a size range between two sieves of 1.19 mm and 0.25 mm was selected for characterization.AC surfaces was heated with concentrated HNO_3 (1 g AC: 10 mL acid) at 80°C to almost dryness to produce Oxidized Activated Carbons (OAC), that was washed thoroughly until no acidity was detected in wash water. OAC was dried at 120°C until constant weight was achieved. OAC surface was functionalized to produce Basic Activated Carbons (BAC) as follows; 15 g of dry OAC was allowed to react with 25 % thionyl chloride in toluene (100 mL) under reflux for 6 hours at 70°C. During this stage, surface carboxylic groups were converted to acetyl chloride groups. The carbon was left to dry in the oven at 85°C for 2 hours and the carbon product was allowed to react with 100 mL 0.75 M 1,2-diaminoethane (ethylene diamine) at 90°C under reflux for 24 hours. By the end of the reaction, nitrogen-containing functional groups were immobilized on the carbon surface via amide coupling. For the preparation of hydrophobic activated carbons (HAC), 15 g of dry OAC was allowed to react with 50 % thionyl chloride in toluene under reflux for 2 hours at 70°C. The product was allowed to cool and the solvents were dried using rotary evaporator. After evaporation, the product was immediately mixed with 100 mL of ethylamine and the mixture was kept at 90°C for 2 hours under reflux. By the end of the functionalization steps for both types of surface functionalized carbons (BAC and HAC), the carbons were purified via Soxhlet extraction using 150 mL of acetone for 6 hours followed by washing with deionized water. Further washing using 2M HCl was carried out to remove residual amines from carbon surface. Finally, the carbons were thoroughly washed with deionized water to remove residual acid. The carbons were allowed to dry at 70°C in an oven under vacuum until constant weight was reached. Surface functionalization using EDA produced BAC of VTS. For hydrophobic carbons, surface modification using EA produced HAC of VTS. The Activated Carbons (AC) was characterized by determining their moisture content, ash content, pore volume, density, and surface area. For the preparation of a standard solution of GLI, an initial ***diluent*** 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 same into a 1000ml volumetric flask. Adding 50ml of diluent and kept in an ultrasonic bath until it dissolved completely. Make up to the mark with the mobile phase and mix.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 mixedtogether in equal volumes making a representative sample. For the adsorptionofGlipizide from spiked PLW, samples of the stock solution of Glipizide, was spiked with the filtered PLW to achieve the rangeof initial concentrations as in the study of equilibrium adsorption from deionized water [4]. 0.06 g of the carbons (OACs, HACs, BACs) prepared from VTS, was mixed with25 mL of spiked drug solutions and was left at 25 °C under mechanicalagitation at 30, 60, 90, 120, 150, and 180 minutes, after separated and analyzed.

RESULTS

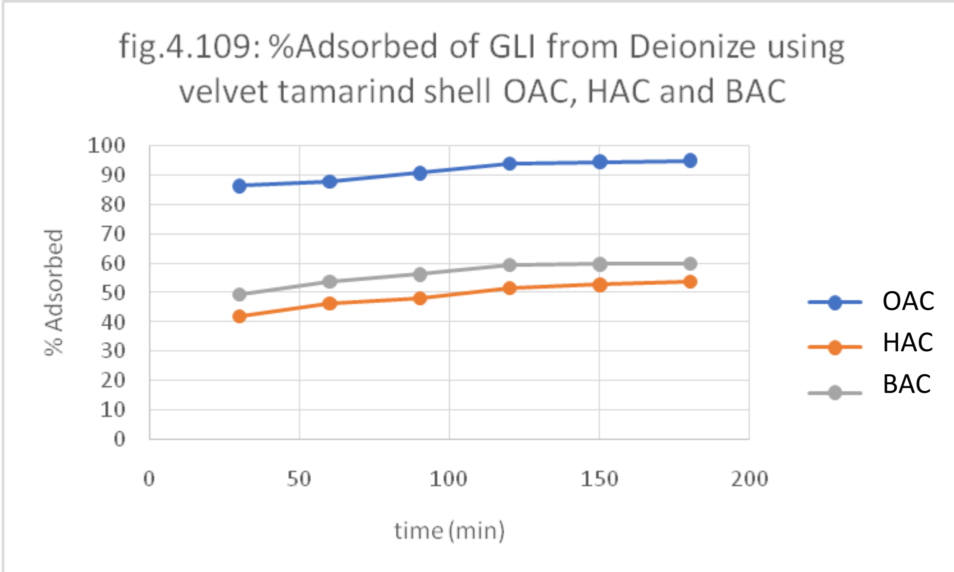
Table 1: The physical characteristics of Mango Kernel Seed Activated Carbon

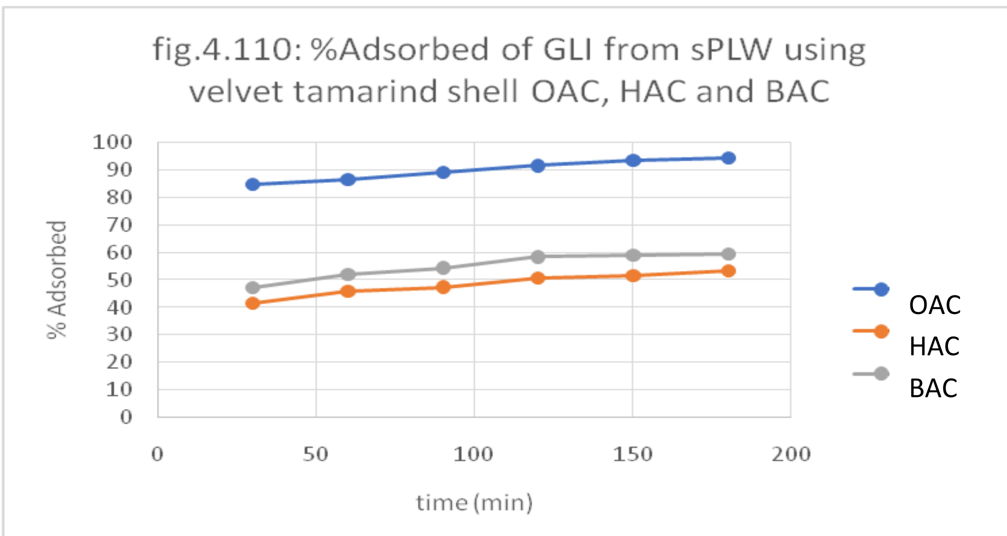
Parameter	**AC(VTS)**
Moisture content %	11.94
Ash content %	8.30
Pore volume cm^3	1.11
Density gcm^{-3}	0.29
Surface area m^2g^{-1}	570

From Table 1, VTSAC surface area was found to be 570 $m^2 g^{-1}$ which indicates VTSAC to be a very good adsorbent for efficient adsorption of pollutants from pharmaceutical liquid waste. The other physical properties like density, pore volume, moisture content, and ash content may affect the adsorption capacity.

Table 2: Percentage adsorption of GLI from deionized water and sPLW using OAC, HAC, and BAC prepared with velvet tamarind shell.

Time		**qe (Deionized)**			**% Removal (Deionized)**			**qe (Effluent)**			**% Remova**	
Time	sqrt (t)	$qe_{(OAC)}$	qe HAC	qe BAC	$\%_{(OAC)}$	%HAC	%BAC	qe(OAC)	qe(HAC	qe(BAC)	%OAC	%H
30	5.477226	21.6	10.5	12.4	86.4	42	49.6	21.24	10.38	11.8	84.96	4
60	7.745967	22	11.6	13.5	88	46.4	54	21.64	11.48	13	86.56	4
90	9.486833	22.7	12	14.1	90.8	48	56.4	22.3	11.86	13.6	89.2	4
120	10.95445	23.5	12.9	14.9	94	51.6	59.6	22.9	12.68	14.6	91.6	5
150	12.24745	23.64	13.2	14.96	94.56	52.8	59.84	23.4	12.94	14.78	93.6	5
180	13.41641	23.78	13.45	15	95.12	53.8	60	23.58	13.36	14.86	94.32	5





A plot of the percentage adsorption with time of Glipizide on the carbons of Velvet tamarind shell, from spiked PLW at 25 °C are shown in figure above. The adsorption data show a gradual increase in the percentage adsorbed with time. This is in agreement with that obtained from the drug in deionized water as can be seen in table above. The adsorption capacity, q_e and % Adsorbed with time, from the spiked PLW of the different carbons follow similar order to Glipizide, from deionized water. For carbons of velvet tamarind shell on Glipizide, from both deionized water and spiked PLW, the trends are: OAC >HAC > BAC, OAC > HAC > BAC, OAC > HAC > BAC respectively. The little deviation here could be as a result of high porosity of velvet tamarind shell activated carbon. BAC show the lowest uptake GLI. This could be because, at the pH of the deionized and spiked PLW when the adsorption was done, both the drugs and BACs surfaces remain positively charged leading possibly to electrostatic repulsion and less GLI adsorptions. However, there were less uptake from spiked PLW than from deionized water. Such decrease in drug uptake from spiked PLW is probably because of the competition of dissolved organic substances, available in spiked PLW, with Glipizide molecules for adsorption sites on the adsorbents.

CONCLUSION

Activated carbon prepared using VTS possesses high surface area. Oxidized activated carbon possesses acidic surface properties with much low surface area than activated carbon. Surface functionalization of OAC to produce basic and hydrophobic activated carbons (BAC and HAC) were largely successful with a further decrease in their surface area. GLIinteract differently with the carbons under investigation depending on the nature of carbon surface. Adsorption forces involved in GLIadsorption include hydrophobic bonding, electrostatic interaction, H-bonding and van der Waals forces. Despite possessing a very low surface area (by nitrogen adsorption), HAC shows the best performance in GLIremoval in terms adsorption capacity and percentage adsorption

The current study showed that functionalized surfaces can play betterrole in the removal of Glipizide, from aqueous solution than activated carbon with high surface area. From literature, Activated Carbon (unoxidized) possesses high surface area and well-developed porous structure with dominating van der Waals forces. However, it shows the slowest process of drug uptake compared with the other carbons. OAC and other functionalized activated carbons show good capability of drug removalfrom spiked PLW. Surface functionalization of activated carbon shows a promising solution for pharmaceuticals removal from deionized water as well as pharmaceutical liquid waste.

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