EREM 78/2
Journal of Environmental Research, Engineering and Management
Vol. 78 / No. 2 / 2022
pp. 28–37
DOI 10.5755/j01.erem.78.2.29695

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Received 2021/08 | Accepted after revision 2022/01

http://dx.doi.org/10.5755/j01.erem.78.2.29695

# Adsorptive Removal of Ibuprofen, Ketoprofen and Naproxen from Aqueous Solution Using Coconut Shell Biomass

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The use of commercial activated carbon (AC) to remove organic micropollutants from aqueous solution is expensive and unsustainable. In this study, coconut shell activated carbon (CSAC) was synthesized and applied for the removal of ibuprofen, ketoprofen and naproxen from aqueous solutions. The effects of carbonization and acid activation on the CSAC were studied using Fourier-transform infrared spectroscopy, scanning electron microscope, proximate and ultimate analyses. The influence of initial concentration (200–1000 mg/L), contact time (10–200 min), and temperature (30–60°C) was also investigated. The adsorptive capacity of CSAC for various pollutants was found to increase with concentration up to 150 min. Ibuprofen, ketoprofen and naproxen removal obeyed Langmuir ($R^2$ = 0.9978), Temkin ($R^2$ = 0.9551) and Freundlich ($R^2$ = 0.9879) isotherm, respectively. The kinetic data obtained for various pollutants are best described by the pseudo-first-order model with correlation coefficient values in the range 0.96–0.99. The free energy (G) values ranged between 1.0 and 9.0 kJ/mol for all the pollutants investigated. The mechanism of adsorption is physical, endothermic, and non-spontaneous. This study shows that CSAC is an effective alternative adsorbent for sequestering mixture of organic pollutants from aqueous solution.

**Keywords:** synthesis of activated carbon, biomass conversion, sustainable adsorbent, characterization, thermodynamics, isotherms, kinetics, adsorption capacity, percentage removal.

## Introduction

Organic micropollutants including active pharmaceutical compounds such as naproxen, ibuprofen, and ketoprofen constitute serious environmental and health hazards that require immediate attention. The predominance of these drugs in aquatic settings is associated with their clinical and widespread acceptability for the treatment of painful and inflammatory ailments (Zhang et al., 2008). Studies (Acosta et al., 2018, Wang et al., 2018) have shown that organic pollutants including active pharmaceuticals find ways into groundwater via sewage systems of pharmaceutical manufacturing plants, hospitals, private house, households, and landfills. Excessive consumption of naproxen, ibuprofen, and ketoprofen can lead to the formation of hydroxylated and methoxylated derivatives in free and/or glucuronide-conjugated form as observed in human urine and plasma (Landry et al., 2015; Scheurell et al., 2009). Liver and kidney toxicity, gastrointestinal damage, platelet malfunction, and human convulsion have all been reported where these compounds were accidentally consumed in large quantities (Varga et al., 2019).

Several technologies (Landry et al., 2015; Sophia and Lima, 2018; Wang and Wang, 2016; Yang et al., 2017) have been deployed for the abatement of pharmaceuticals and other organic compounds from water sources. Liu et al. investigated the removal of thirty-five pharmaceuticals spatially distributed in twelve China municipal wastewater treatment plants with caffeine as the dominant drug in most of the plants (Liu et al., 2017). Song et al. applied a composite adsorbent for the removal of tetracycline (Song et al., 2018). Ooi et al. investigated the removal of multiple pharmaceuticals from hospital wastewater using biological method (Ooi et al., 2018). The acceptability and effectiveness of various techniques adopted have been hampered due to relatively low efficiency (Ooi et al., 2018). The high polarity and/or chemical persistence of pharmaceuticals make them escape from the wastewater treatment process and only 21–40% removal efficiency has been reported (Zhang et al., 2008). Apart from the aforementioned limitations, conventional techniques are expensive and associated with toxic by-products (El Naga et al., 2019; Ogunleye et al., 2020).

Adsorption is a highly preferred method because it is economical, safe, and efficient (Zhang et al., 2008; Aremu et al., 2020). But, commercial ACs are expensive, and for practical purposes and to ensure maximum return on investment, many agricultural residues and industrial wastes have been investigated (El Naga et al., 2019; Baccar et al., 2012; Bello et al., 2019; Khadhri et al., 2019; Okeowo et al., 2020). Some of these residues and industrial wastes constitute environmental problems. In this study, AC was synthesized from coconut shells using a wet activation method and investigated in the laboratory for sequestration of naproxen, ibuprofen, and ketoprofen from aqueous solution.

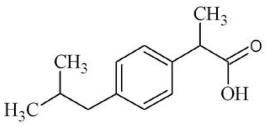
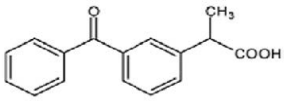
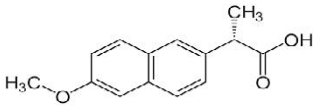
## Materials and methods

The coconut shell (CS) was obtained from local coconut sellers at the Arada market, Ogbomoso, Oyo State, Nigeria. 99.9% purity naproxen, ibuprofen, and ketoprofen purchased from Shasun Chemicals and Drug Ltd, United Kingdom, were used. The physicochemical properties and chemical structure of the drugs are shown in *Table 1*. The reagents used are HCl and distilled water. The equipment used include weighing balance (ZC20602), UV-vis. (UV752D), FTIR, SEM (JSM-7600F), crusher, stopwatch, desiccators, water bath shaker, rotary shaker (HZ 300), muffle furnace (Carbolite ELF 11/68,) and electric oven (Techmel TT-9083, USA).

### Preparation of adsorbate and synthesis of activated carbon

1000 mg/L of the drugs were prepared by dissolving 1.0 g in 1000 mL of distilled water. The other concentrations (200 and 600 mg/L) were prepared by serial dilution. The coconut shell (CS) after being washed with water was dried at 110°C for 24 h to eliminate the moisture. A portion of the dried CS was pulverized, ground to powder, and sieved to a particle size of 1–2 mm. The sample (20 g) was then mixed by

**Table 1.** *Physicochemical property of selected pharmaceutical drugs (Tixier et al., 2003)*

| Trade Name | Chemical Formula | Chemical structure | Molecular Mass (g/mol) | Log Kow* | pKa* |
|---|---|---|---|---|---|
| Ibuprofen | $C_3H_{18}O_2$ |  | 206.29 | 3.97 | 4.91 |
| Ketoprofen | $C_{16}H_{14}O_3$ |  | 254.28 | 3.12 | 4.45 |
| Naproxen | $C_{14}H_{14}O_3$ |  | 230.25 | 3.18 | 4.15 |

** Kow: octanol/water partition coefficient*

impregnation in 200 mL of 0.5 mol/dm$^3$ HCl. The acid-soaked samples were kept for 24 h in a furnace at ambient conditions. Excess HCl was drained and evaporated at 100–105°C for 4 h. The slurry CS was activated at a temperature of 700°C for 90 min in a muffle furnace. The activated char was neutralized using deionized water until the pH of the washing solution was between 6.5 and 7.0 before dried in the oven at 80–90°C for 2 h, and granular CSAC was obtained.

## Characterization study

Fourier transform infrared (FTIR) spectroscope (FTIR-2000, Perkin Elmer) and scanning electron microscope (JSM-7600F) was deployed for investigating the functional group and surface morphology of the AC respectively. The thermogravimetric analyzer (Perkin Elmer TGA7) was used to determine the proximate and ultimate properties.

## pH and point of zero charge ($pH_{pzc}$) determinations

To determine the $pH_{pzc}$ of the CSAC, 0.05 g of CSAC was mixed with a predetermined pH solution containing 100 mL of 0.1 M NaCl. The pH of the mixture was adjusted using the NaOH or HCl. The corked sample holder was agitated in a shaker for 24 h at 250 rpm and the final pH was measured. The difference of pH between the final and the initial (ΔpH) was calculated and plotted against the initial pH from where the $pH_{pzc}$ was determined.

To evaluate the surface acidity and basicity of CSAC, $NaHCO_3$ and HCl were used for the neutralization of acidic and basic groups, respectively, and concentration of the acidic and basic groups was determined by Boehm titration (Bello et al., 2017).

## Adsorption study

Batch adsorption experiments were performed for the investigation of the adsorption capacity and percentage removal. The residual pollutant concentrations were determined using a standard curve (linear Lambert curve) established between absorbance and concentration (plots not included). The concentrations of aqueous solutions prior to and after adsorption were quantified with UV-Vis. spectrophotometer (UV – 752, USA) at a maximum wavelength of 317, 264, and 193 nm for naproxen, ibuprofen, and ketoprofen, respectively. The percentage of pollutants removed was estimated using equation 1 while the adsorption capacity was estimated using equation 2.

$$\%\, removal = \frac{(C_o - C_e)}{C_o} \times 100 \quad (1)$$

$$q_t = \frac{(C_o - C_e)V}{M} \times 100 \quad (2)$$

Where: $C_o$ – Initial and equilibrium pollutant concentrations (mg/L)
$C_e$ – Equilibrium pollutant concentrations respectively (mg/L)
M – Adsorbent dosage (g/L)
V – Solution volume (mL)

### Adsorption isotherm

Isotherms describe the interaction of adsorbates with adsorbents. The amount of drugs attached to the surface of CSAC was determined as a function of concentration at 303 K. The linearized form of the Langmuir, Freundlich, and Temkin isotherms evaluated based on the correlation coefficient ($R^2$) is portrayed in *Table 2*.

**Table 2.** *Linearized isotherm models and corresponding plot variables*

| Isotherm | Linearization | plot |
|---|---|---|
| Langmuir | $\frac{C_e}{q_e} = \frac{1}{k_l q_m} + c_e \frac{1}{q_m}$ | $\frac{C_e}{q_e}$ Vs $c_e$ |
| Freundlich | $log\, q_e = \frac{1}{n}\, log\, c_e + log\, k_f$ | $log\, q_e$ Vs $log\, c_e$ |
| Temkin | $q_e = B\, ln\, A + B\, ln\, c_e$ | $q_e$ Vs $ln\, c_e$ |

### Adsorption kinetics

To determine the sorption design parameter and predict the transport dynamics for the rate of adsorption of these drugs on CSAC, kinetic data were fitted to pseudo-first-order and pseudo-second-order models, since empirical models such as logarithmic, power, exponential, and hyperbolic are often difficult to correlate with the sorption mechanism (Ogunleye et al., 2020). The linearized form of these models is represented by equations 3 and 4, respectively. The selection of suitable kinetic parameters was based on the Average Absolute Relative Percentage Error (AARPE) and $R^2$ criteria.

(1) Pseudo-first-order

$$\log(Q_e - Q_t) = \log Q_e - \left(\frac{k_1}{2.303}\right) t \quad (3)$$

(2) Pseudo-second-order

$$\frac{t}{Q_t} = \frac{1}{k_2 Q_e^{\,2}} + \left(\frac{1}{Q_e}\right) t \quad (4)$$

### Adsorption thermodynamics

Equation (5) was used to calculate the free energy change (ΔG). The apparent changes in entropy (ΔS) and enthalpy (ΔH) were estimated from the Van't Hoff's graph by plotting $ln\, K_c$ vs $\frac{1}{T}$. The slope $(\frac{-\Delta H}{R})$ and intercept $(\frac{\Delta S}{R})$ were used to estimate standard ΔS and ΔH according to equation (6).

$$\Delta G = -RT\ ln\, K_c \quad (5)$$

$$ln K_c = \frac{-\Delta H}{RT} + \frac{\Delta S}{R} \quad (6)$$

Where: $K_c$ - Apparent equilibrium constant;
R - Gas constant (8.314 J/mol K);
T - Absolute temperatures (K).

## Results and discussion

### Characterization of coconut shell adsorbents

The proximate and ultimate analyses result for CS and CSAC are shown in *Table 3*. It was observed that there was a significant enhancement of proximate properties of CSAC when compared with the CS. Many reported agricultural residues processed for AC have also shown similar trends (Bello et al., 2019; Andas et al., 2017). The ultimate analysis shows high organic content (74.3% C). It can, therefore, be inferred that by activating the CS, the carbon content of CSAC increased significantly. The active carbon of adsorbent determines its adsorption capacity (Dada et al., 2012). It can also be deduced that the adsorption capacity of CSAC improved after the acid activation. The SEM micrographs (*Fig. 1*) revealed poor development

of surface pores of the non-activated sample and well-developed pores and cavities in the activated sample. The formation of rough cavities and pores in CSAC can be attributed to the disintegration of lignocellulosic material and given off of volatile matter at high temperatures (Bello et al., 2019). The FTIR spectra (*Fig. 2*) indicates that some peaks were shifted; some disappeared while some new peaks were also detected. These observations can be attributed to the activation process. *Table 4* shows the FTIR data summary of CS and CSAC.

**Table 3.** *Surface characteristics, proximate contents, and elemental analyses of CSAC*

| Properties | CS | CSAC |
|---|---|---|
| Proximate composition (%) | | |
| Moisture content | 7.4 | 5.3 |
| Volatile matter | 72.7 | 17.5 |
| Fixed Carbon | 16.8 | 71.2 |
| Ash | 3.1 | 6.0 |
| Elemental composition (%) | | |
| Carbon | 51.5 | 74.3 |
| Hydrogen | 5.7 | 4.8 |
| Nitrogen | 0.6 | 0.5 |
| Others | 42.2 | 20.4 |

**Fig. 1.** *SEM images showing (a) char and (b) CSAC*

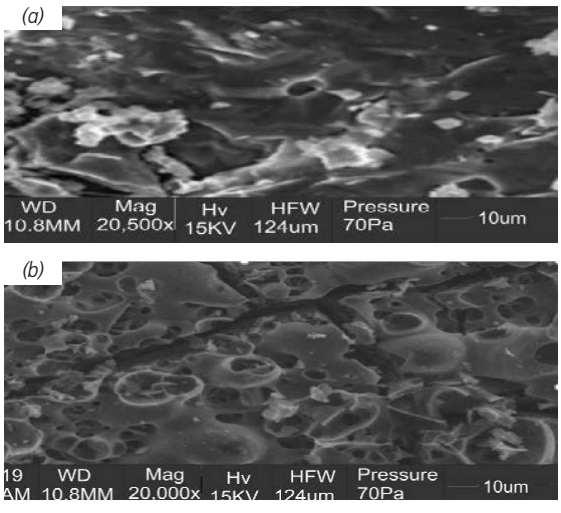
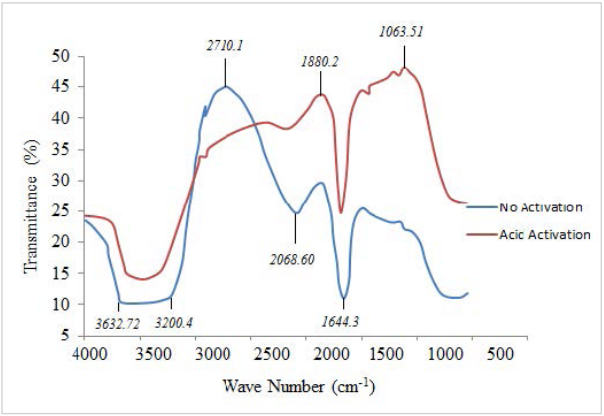



**Fig. 2.** *FTIR spectra of (a) non-activated and (b) acid activated carbon from coconut shell*




## The effect of pH

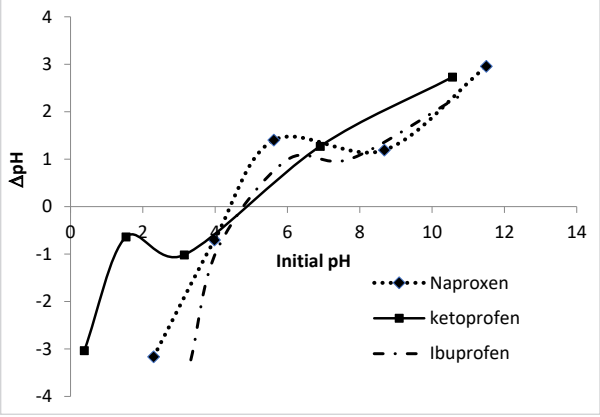
The solution pH was previously observed to influence adsorption of dyes and pharmaceuticals (Yang et al., 2017; Bello et al., 2019). The optimum pH for the removal of the three drugs was determined experimentally using the pH point of zero value (*Fig. 3*). The values of $pH_{pzc}$ (pH where net CSAC surface charge equals zero) of 4.22, 4.57 and 4.89 obtained for naproxen, ibuprofen and ketoprofen solutions were highly influenced by all the functional groups that present on the CSAC surface. According to Bello et al. (2019), a net positive charge on carbon surface is obtained when the pH of the solution is below the $pH_{pzc}$ and a negative charge is obtained when it is higher. *Fig. 3* shows the graph of ΔpH against initial pH ($pH_o$) for a different drug solution. Cationic adsorption is favored when $pH_{pzc}$ is lesser than pH while adsorption of anions is enhanced at pH less than $pH_{pzc}$ (Bello et al., 2017). The total acidic and basic groups determined through the Boehm titration for CSAC using the naproxen, ibuprofen and ketoprofen solutions are 0.127, 0.110, 0.104 and 0.057, 0.072, 0.091 mol/g, respectively. The lower values of the basic groups when compared with the acidic groups for all the drug solutions indicated that the adsorbent surface is predominantly acidic (Bello et al., 2017; Bello et al., 2019).

The adsorption of the three drugs on CSAC increased at pH higher than their $pH_{pzc}$ values. The % removal of various pollutants was optimum at pH of 4 (89.28, 80.5 and 75.38% for naproxen, ibuprofen and

**Table 4.** *FTIR spectra of char and activated coconut shell*

| Carbonized coconut shell | | Activated Coconut shell | | Remark |
|---|---|---|---|---|
| Wavenumber ($cm^{-1}$) | Functional group | Wavenumber ($cm^{-1}$) | Functional group | |
| 3632.72 | O-H | 3471.24 | O-H | Shifted downward after activation |
| 3208.44 | O-H | – | – | Disappeared when activated |
| 2068.6 | C≡C | 2087.6 | C≡C | Shifted upward when activated |
| 1644.33 | C=C | 1637.99 | C=C | Shifted downward when activated |
| – | – | 1083.91 | C-N | Appeared after activation |

**Fig. 3.** *Graph of point zero charge of acid activated CSAC*

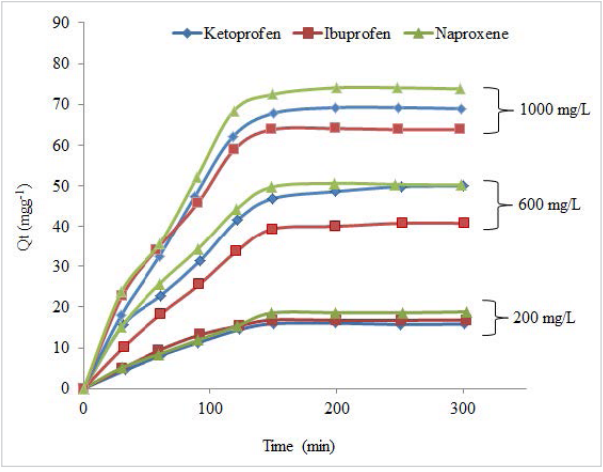



ketoprofen solutions, respectively). Above the pH of 4, there was a reduction of the percentage of the drug removed perhaps due to increasing repulsive electrostatic force (Khazri et al., 2017). The $pK_a$ value of ibuprofen (4.91), ketoprofen (4.45), and naproxen (4.15) is constant and unique for each molecule of the drugs regardless of concentration and determines the ease in which the solution will donate protons to the surface of the adsorbent. The lower the value of $pK_a$, the stronger is the acid. Hence, the greater is its ability to donate protons (Khazri et al., 2017).

## The effect of contact time and initial concentration

*Fig. 4* shows the adsorptive capacity of CSAC for the various drugs at various initial concentrations (200–1000 mg/L), contact time (30–200 min), and at pH of 4.0. Generally, at the onset, the uptake of various drugs by the CSAC increases with time due to the presence of many empty sites on the surface of the adsorbent. After a period of time, there was a reduction in adsorption because the pore surface site was difficult to fill as a result of electrostatic hindrance until the equilibrium is reached (Bello et al., 2019). At the equilibrium, the drug uptake was at maximum with the onset of the plateau which began approximately after 150 min for all the drugs. According to Zhang et al. (2008), the pores on the adsorbent at the equilibrium would have become saturated with molecules of various pollutants. It is clear from *Fig. 4* that CSAC had the highest adsorption capacity for naproxen (75.1 mg/g) and lowest for ibuprofen (64.8 mg/g).

**Fig. 4.** *Adsorptive capacity of CSAC for the removal of ibuprofen, ketoprofen and naproxen in (a) 200 mg/L, (b) 600 mg/L and (c)1000 mg/L initial pollutant concentrations (dosage = 10 g/L, pH = 6.5, T = 303 K)*




The highest adsorption capacities (ibuprofen: 63.78 mg/g, ketoprofen: 68.92 mg/g, naproxen: 73.78 mg/g) were obtained when 1000 mg/L initial concentration of various drugs was used. Also, the lowest

**Table 5.** *Comparison of percentage removal (%) of selected pharmaceuticals from an aqueous solution by CSAC and some reported activated carbons*

| Adsorbent | Activated coconut shell | | | References |
|---|---|---|---|---|
| | Ibuprofen | Ketoprofen | Naproxen | |
| Activated sludge | 64 | – | 48 | Carballa et al. (2004) |
| Activated sludge | 82.5 | 51.5 | 85.1 | Radjenovic et al. (2007) |
| Olive-waste cake | 70.1 | 88.4 | 90.5 | Baccar et al. (2012) |
| Coconut shell AC | – | – | 79.1 | Bo et al. (2015) |
| Rice hull | 95.4 | – | – | Mukoto et al. (2015) |
| Medicinal based AC | 89.5 | – | – | Omer et al. (2019) |
| Bituminous oil-based AC | 51.0 | – | – | Mellah and Harik (2019) |
| Coconut shell AC | 84.5 | 80.0 | 88.2 | This study |

adsorption capacities (ibuprofen: 16.83 mg/g, ketoprofen: 15.86 mg/g, naproxen: 18.83 mg/g) were obtained when 200 mg/L initial concentration of various pollutants was used. This result is consistent with previous studies (Bello et al., 2017; Kumar and Kumar, 2014; Rafatullah et al., 2010). *Table 5* shows how the CSAC's removal efficiency compared with some known adsorbents.

## Isotherm studies

Adsorption isotherm studies are important for designing the adsorption system (Aremu et al., 2020; Okeowo et al., 2020). Isotherms provide information about the degree the various molecules of the pollutants interact with surfaces contacted (Ogunleye et al., 2020). In this study, the amount of ibuprofen, ketoprofen, and naproxen attached to the surface of CSAC was determined graphically at different initial concentrations and at 303 K. *Table 6* presents the parameter estimated from various isotherm models for various pollutants. It was observed that ibuprofen, ketoprofen and naproxen obey Langmuir ($R^2$ = 0.9978), Temkin ($R^2$ = 0.9551) and Freundlich ($R^2$ = 0.9879) isotherms, respectively.

**Table 6.** *Parameter of isotherm models for ibuprofen, ketoprofen and naproxen pollutants*

| Isotherm | Parameter | Ibuprofen | Ketoprofen | Naproxen |
|---|---|---|---|---|
| | $Q_{max}$ (mg/g) | 89.29 | 92.59 | 111.111 |
| Langmuir | $K_L$ (L/mg) | 0.0077 | 0.0062 | 0.0065 |
| | $R_L$ | 0.1153 | 0.1393 | 0.1332 |
| | $R^2$ | 0.9978 | 0.7045 | 0.8569 |
| | $K_F$ (mg/g) | 2.4757 | 2.7221 | 2.6321 |
| Freundlich | N | 1.7501 | 1.8467 | 1.6923 |
| | $R^2$ | 0.9691 | 0.9407 | 0.9879 |
| | B | 12.715 | 10.573 | 22.004 |
| Temkin | $B_T$ | 198.124 | 238.262 | 114.486 |
| | A | 0.8029 | 0.8693 | 0.6208 |
| | $R^2$ | 0.9001 | 0.9551 | 0.9115 |

## Adsorption kinetics

The dynamics of solute uptake by adsorption can be influenced by many factors including movement of solutes through the protective film around the particle, solute diffusion to the surface, movement from the surface to the internal sites and solute trapping by adsorption, precipitation or ion-exchange (Bello et al., 2019). In this study, the design parameters and the transport model for adsorption of ibuprofen, ketoprofen and naproxen on CSAC were determined. The value of $R^2$ obtained ranged 0.851–0.965 compared with 0.960–0.999 obtained for the pseudo-first-order model. The calculated kinetic parameters, corresponding $R^2$, and AARPE are presented in *Table 7*. Based on these criteria, the pseudo-first-order model best described the experimental data for adsorption of various drugs onto CSAC.

**Table 7.** *Ibuprofen, ketoprofen and naproxen kinetic parameters at 303 K*

| Pollutants | Co (mg/L) | $Q_{e, exp.}$ (mg/g) | Pseudo-first order | | | | Pseudo-second order | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | $Q_{e, cal}$ (mg/g) | $K_1$ ($min^{-1}$) | $R^2$ | AAPRE (%) | $Q_{e, cal}$ (mg/g) | $K_2$ ($min^{-1}$) x$10^{-3}$ | $R^2$ | AAPRE (%) |
| Naproxen | 200 | 17.6 | 23.9 | 0.05 | 0.995 | 0.362 | 42.74 | 0.61 | 0.926 | 1.43 |
| | 600 | 45.5 | 58.4 | 0.05 | 0.999 | 0.282 | 109.89 | 0.26 | 0.909 | 1.41 |
| | 1000 | 74.5 | 82.9 | 0.03 | 0.982 | 0.112 | 158.73 | 0.16 | 0.917 | 1.13 |
| Ketoprofen | 200 | 17.2 | 23.9 | 0.04 | 0.960 | 0.39 | 50.7 | 0.52 | 0.925 | 1.956 |
| | 600 | 40.0 | 57.1 | 0.05 | 0.985 | 0.43 | 111.1 | 0.28 | 0.851 | 1.783 |
| | 1000 | 70.0 | 123.0 | 0.06 | 0.993 | 0.36 | 238.1 | 0.14 | 0.873 | 2.408 |
| Ibuprofen | 200 | 16.8 | 21.23 | 0.05 | 0.985 | 0.26 | 33.0 | 0.86 | 0.965 | 0.96 |
| | 600 | 46.7 | 45.22 | 0.04 | 0.973 | 0.03 | 119.0 | 0.15 | 0.925 | 1.55 |
| | 1000 | 64.8 | 90.60 | 0.04 | 0.969 | 0.39 | 322.6 | 0.12 | 0.859 | 3.98 |

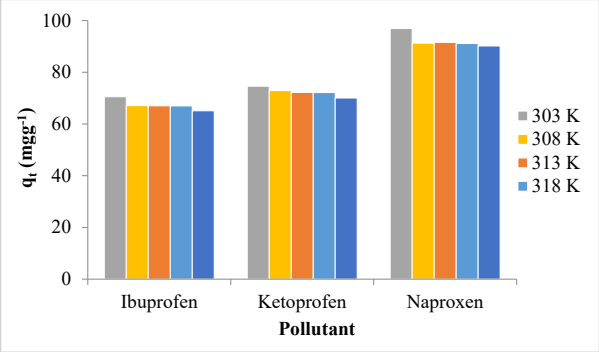
**Table 8.** *Estimated parameters for thermodynamic study of ibuprofen, ketoprofen and naproxen*

| Parameter | Time (min) | Ibuprofen | | Ketoprofen | | Naproxen | |
|---|---|---|---|---|---|---|---|
| | | 303 K | 333 K | 303 K | 333 K | 303 K | 333 K |
| $K_c$ | 60 | 0.05 | 0.07 | 0.05 | 0.07 | 0.06 | 0.09 |
| | 120 | 0.19 | 0.22 | 0.19 | 0.22 | 0.13 | 0.21 |
| | 180 | 0.25 | 0.29 | 0.25 | 0.27 | 0.49 | 0.58 |
| ΔG (KJ/mol) | 60 | 7.99 | 7.22 | 7.48 | 6.59 | 7.12 | 6.69 |
| | 120 | 4.54 | 4.27 | 4.21 | 3.78 | 5.07 | 4.38 |
| | 180 | 4.00 | 3.60 | 3.46 | 3.08 | 1.76 | 1.52 |
| ΔH (KJ/mol K) | 60 | – | 8.07 | – | 9.40 | – | 4.09 |
| | 120 | – | 5.91 | – | 4.76 | – | 12.02 |
| | 180 | – | 4.41 | – | 4.06 | – | 10.78 |
| ΔS (J/mol K) | 60 | – | 0.53 | – | 6.57 | – | 7.79 |
| | 120 | – | 5.21 | – | 1.80 | – | 22.79 |
| | 180 | – | 1.36 | – | 2.05 | – | 12.39 |

## Thermodynamics studies

As shown in *Fig. 5*, the adsorption capacity of CSAC decreases with temperature for the three drugs under the investigation. This observation can be ascribed to the decreasing mobility of molecules of various pollutants as temperature increases from 303–333 K (Bello et al., 2019). *Table 8* shows the thermodynamic parameters estimated for ibuprofen, ketoprofen, and

**Fig. 5.** *Effect of temperature on adsorption capacity of CSAC for removal of ibuprofen, ketoprofen and naproxen (initial concentration = 1000 g/L, adsorbent dosage = 10 g/L, contact time = 150 min, pH = 6)*




naproxen data when plotted according to Equation 6. The decreasing values of $\Delta G$ with temperature indicate decreasing adsorption of pollutants on CSAC. This may be attributed to the effect of increased degree of freedom which promotes desorption rather than adsorption at such a higher temperature. The positive value of $\Delta G$, however, indicates that the process is non-spontaneous without any external influence (Olakunle et al., 2017). Since values of $\Delta G$ obtained are in the range of 1.0–8.0 KJ/mol for all the pollutants, the adsorption process can be regarded as physical (Başar, 2006). The positive values of $\Delta H$ at all temperatures showed that the adsorption process is endothermic and the positive $\Delta S$ values indicate increasing randomness at the solid-solution interface during the adsorption process (Qasemi et al., 2018).

## Conclusion

The potential of a cheap and environmentally benign activated carbon produced from CS was investigated in this study for sustainable removal of pharmaceuticals from aqueous solution. The adsorptive capacity of the developed adsorbent improved with increasing concentrations of the pharmaceuticals and decreased with increasing temperature. The adsorption of ibuprofen, ketoprofen, and naproxen onto the activated carbon was confirmed to obey Langmuir, Temkin, and Freundlich isotherms, respectively. The kinetic data are best described by the pseudo-first-order kinetic model. The values of the enthalpy, entropy, and Gibbs free energy changes suggested endothermic, non-spontaneous, and physical adsorption of various drugs onto the surface of the developed activated carbon. CSAC had proven to be a suitable alternative adsorbent in lieu of expensive commercial activated carbon for the abatement of multiple micro-organic pollutants from wastewater. Due to its high affinity for naproxen, it is highly recommended for sequestration of naproxen drug from aqueous solution.

### Acknowledgments

The authors acknowledge the Departments of Chemical Engineering Ladoke Akintola University of Technology (LAUTECH) and African Development Bank (ADB) Laboratory at the African University of Science and Technology (AUST), Abuja, for the usage of their laboratories.

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