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REMOVAL OF THE HYPOGLYCAEMIC DRUG GLICLAZIDE FROM AQUEOUS SOLUTIONS BY CARBON-BASED MAGNETIC MATERIALS

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ABSTRACT

Adsorption of gliclazide from aqueous solutions with carbon-based adsorbents is the aim of this study. For this purpose, magnetic, versatile, and inexpensive magnetic activated carbon (MAC) and magnetic multi-walled carbon nanotube (MMWCNT) adsorbents have been developed. MAC and MMWCNT characterization were analyzed using Fourier-transform infrared spectroscopy (FTIR), X-ray diffraction (XRD) and Thermogravimetric analysis (TGA) techniques. In order to find the optimum conditions in batch adsorption experiments, the effects of parameters such as adsorbent dose, gliclazide solution pH, contact time, temperature and presence of foreign ions were investigated. Parameters were calculated for adsorption isotherms using Langmuir and Freundlich models. The adsorption studies of MAC and MMWCNT materials with gliclazide were in accordance with the nonlinear Langmuir and Freundlich isotherm models for all temperatures (298, 308, 318 K). The correlation coefficients of the Langmuir isotherm model are slightly higher than the Freundlich model. MAC adsorbent has maximum monolayer adsorption capacity at T=298 K ($q_{\max}=101.66$ mg/g, $R^2=0.88$) while MMWCNT has maximum monolayer adsorption capacity at T=308 K ($q_{\max}=71.59$ mg/g, $R^2=0.90$). For MAC and MMWCNT - Gliclazide adsorption systems, pseudo-first and second-order kinetic models were examined and found to be more appropriate to the pseudo-second-order kinetic model. The results showed that the MAC and MMWCNT could be could be promising adsorbents for gliclazide removal.

Keywords: Adsorption, gliclazide, magnetic nanoparticles, multiwalled carbon nanotubes, powdered activated carbon.

1. INTRODUCTION

In recent years, the use of personal care products (PPCPs) such as medical drugs and cosmetics has been increasing rapidly with the improvement in people's living standards (Zhu et al., 2017). Such substances that can be detected in water have been identified as micropollutants found in wastewater streams and groundwater around the pharmaceutical industry. These compounds, which are the cause of many unknown diseases, pose great risks to water and humans (Balasubramani et al., 2020; Alnajjar et al., 2019; Niaei et al., 2020).

The fact that the amount of daily use of such pollutants worldwide is very high is a big problem. Diabetes has become a huge problem globally. According to the 2017 report of the International Diabetes Federation (IDF), diabetes is a worldwide epidemic in which 415 million adults lived with this disease in 2015, and it is estimated that this number will reach 629 million by 2045. (IDF, 2017). Pharmaceuticals used to treat these diseases include substances such as sulfonylureas (gliclazide, glibenclamide, glimepiride) or biguanidine (metformin) (Mrozik and Stefanska, 2014).

Gliclazide is used to treat patients with non-insulin dependent diabetes mellitus (Type II diabetes). It is a second generation sulfonylurea oral hypoglycemic agent (Al Salami et al., 2008; Palmer and Brogden, 1993). Because large amounts of anti-diabetic drugs are used, these pharmaceuticals can accumulate over time and cause environmental problems (Mrozik and Stefanska, 2014). Therefore, with the aim of effectively eliminating this type of contamination, researchers began looking for quick and simple methods of treating contaminated water (Zhu et al., 2017).

Some reported methods for removing micro-pollutants from contaminated water such as adsorption, photocatalytic degradation, catalytic ozonation, reverse osmosis, membrane separation, solvent extraction, ion exchange, electro-fenton process, phytoremediation, etc (Balasubramani, 2020; Niaei and Rostamizadeh, 2020). Most of these methods require high operating costs to remove contaminants (Niaei and Rostamizadeh, 2020). Of the aforementioned methods, adsorption has proven to be the most functional technique for remediation or treatment of a large number of pollutants due to its low cost, accessibility, excellent performance and environmentally friendly nature (Alnajjar et al., 2019; Zhu et al., 2017). The use of various adsorbents for the removal of drug molecules has been extensively studied (Hoseininezhad et al., 2017). In recent years, various materials have been widely

applied to eliminate various contaminants such as activated carbon, biochar, ion exchange resin, carbon nanotubes, chitosan, and organic resin (Zhu et al., 2017).

Carbon-based materials are used for different pharmaceutical adsorption studies such as tetracycline, sulfamethoxazole, ciprofloxacin, metformin, aspirin, acetaminophen and caffeine (Zhu et al., 2017). Carbon-based materials such as MWCNT have a very wide surface area and superior pore volume for adsorption systems (Çavuşoğlu et al., 2021). At the same time, carbon nanotubes have perfect conductivity or semiconductivity, high thermal stability, chemical stability and tensile strength (Hoseininezhad et al., 2017).

Nanomaterials with magnetic properties have found use in many applications such as magnetic separation (Zhu et al., 2017). These magnetically modified materials are highly effective adsorbents, because such components can make easy separation of materials from one another and provide ease of processing (Çavuşoğlu et al., 2021).

Some researchers have studied the removal of various drugs using multi-walled carbon nanotubes (MWCNTs). Lio et al. performed adsorption of resorcinol on intact MWCNTs and HNO₃ treated MWCNTs (Liao et al., 2008). Zhao et al. examined the adsorption mechanism of sulfonamides and non-antibiotic pharmaceuticals using MWCNTs (Zhao et al., 2016). Oleszczuk et al. performed removal of oxytetracycline and carbamazepine by MWCNTs (Oleszczuk et al., 2009). There are few studies on the removal of gliclazide in the literature. Mrozik and Stefanska investigated the adsorption and biodegradation of antidiabetic drugs such as gliclazide, glibenclamide, glimepiride, metformin in soil (Mrozik and Stefanska, 2014). Radi et al. removed the hypoglycemic drug gliclazide using electrochemical oxidation method (Radi et al., 1999). There are also many adsorption studies using magnetic materials as adsorbent. Çavuşoğlu et al. prepared magnetic carbon composites for removal of antidiabetic agent metformin hydrochloride (Çavuşoğlu et al., 2021). Zhou et al. used magnetic dendritic materials for the adsorption of dyes and drugs (Zhou et al., 2010). Bayazit prepared magnetic MWCNTs for methyl orange removal from aqueous solutions (Bayazit, 2014). Huo et al. prepared core/shell nanoadsorbent based on Fe₃O₄ magnetic nanoparticles for adsorption of anionic dyes (Huo et al., 2018). Fan et al. performed adsorption of heavy metal ions by magnetic chitosan beads (Fan et al., 2018). Baghdadi et al. synthesized magnetic activated carbon for adsorption of carbamazepine from urban wastewater (Baghdadi et al., 2016).

In this work, magnetically enhanced MAC and MMWCNT adsorbents were used to remove gliclazide contaminants from aqueous solutions. Various parameters affecting the adsorption

process were investigated. For this purpose, the effects of parameters such as adsorbent dose, pH of gliclazide solution, contact time, temperature and presence of foreign ions (NaCl) were examined. As a result, isotherm models were created by using adsorbate solutions at different temperatures and concentrations, and the kinetics of the adsorption process were examined with the data obtained at different times.

2. EXPERIMENTAL SECTION

2.1. Chemicals

Gliclazide (purity>99%) was supplied by Ali Raif Pharmaceutical Industry Co. (Turkey). The physical properties of gliclazide are presented in Table 1. Activated carbon (AC) was purchased from Merck Co. MWCNT (purity>98% , length: 1–2 μm , outer diameter: 10–20 nm) was purchased from Shenzhen NanoTech. Port. Co. (China). $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$, $\text{Fe}(\text{SO}_4) \cdot 7\text{H}_2\text{O}$, HCl (36.5%), NaOH, NaCl, CH_3OH and NH_4OH solution (25%) were received from Merck Co. High purity water from a Millipore Direct-Q3 water treatment system was used in the experiments.

Table 1: Physical properties of gliclazide

Name	Chemical structure	Molecular formula	M_w (g/mol)	λ_{max} (nm)
Gliclazide		$\text{C}_{15}\text{H}_{21}\text{N}_3\text{O}_3\text{S}$	323,4	227

2.2. Synthesis of Carbon Based Magnetic Materials

Two different carbon-based materials were used in this study, namely AC and MWCNT. To eliminate amorphous carbon remnants, AC and MWCNT were processed in a ash furnace at 350°C for 30 minutes. Subsequently, the materials were oxidized with concentrated HNO_3 solution for 100 minutes under ultrasonication. After the ultrasonication process was completed, AC and MWCNT particles were washed with distilled water until the solution pH reached 7. As a final treatment, the neutralized carbons were dried in a vacuum oven at 60°C.

Magnetic activated carbon (MAC) and magnetic MWCNT (MMWCNT) materials were magnetized using the co-precipitation technique. In this method, carbon materials are magnetized with Fe (II) and Fe (III) ions. The ratio of "Carbon material: Fe (II) + Fe (III)" was

applied as "1: 4". Fe (II) and Fe (III) ions were dissolved in distilled water and mixed separately. Mixtures prepared by adding carbon material were kept under ultrasonic bath. The 8 M NH_4OH solution was slowly added dropwise to the mixtures, reaching the pH of the solution to 11-12. The resulting precipitates were washed at least 5-6 times using distilled water followed by ethanol and dried in a vacuum oven at 60°C .

2.3. Characterization of Synthesized Magnetic Materials

In the first stage of characterization, FTIR analyzes were used to determine the structural properties of magnetic materials. Analyses were carried out using the Bruker Alpha brand device and the KBr method. XRD analyses were performed at room temperature using Rigaku D/Max-2200 diffractometer brand device ($\text{Cu K}\alpha$ radiation, $\lambda = 0.15418 \text{ nm}$). The intensities of the samples were measured at a scanning rate of $2^\circ/\text{min}$ between 20° and 60° , with a range of 2θ . TGA analyzes were performed to determine the thermal properties. Analyses were carried out by heating the samples from room temperature to 1000°C at a rate of $10^\circ\text{C}/\text{min}$ with 5-10 mg of materials and Hitachi STA-7200 simultaneous thermogravimetric analyzer device under nitrogen (200 mL/min).

2.4. Batch Gliclazide Adsorption Tests

Adsorption experiments were carried out as a batch process using 100 mL flasks and 10 mL adsorbate solution. These flasks were kept in a temperature-controlled orbital shaker for certain equilibration times under constant shaking at 120 rpm. The adsorbate stock solution was prepared by dissolving gliclazide in distilled water with some methanol at a concentration of 10 ppm. In order to determine the optimal dose of adsorbents with the first parameter effect, different amounts of adsorbents (0.1, 0.25, 0.5, 0.75 and 1 g/L) and gliclazide stock solution were left to shake. Based on preliminary experiments, the parameters were determined as reaction time 120 minutes, temperature 298 K, solution normal pH value (5.86). After determining the optimal dose, the times to equilibrium of the adsorption for both adsorbents were determined by experiments performed at certain time intervals (1, 5, 10, 15, 30, 45, 60, 90, 120, 180 and 240 minutes). Pseudo-first-order (PFO) and pseudo-second-order (PSO) kinetic models were modeled using experimental data from kinetic studies. The effect of the initial gliclazide concentration (2-10 mg/L) was determined at normal solution pH using the optimal adsorbent dose and equilibration time. These adsorption tests were carried out at three different temperatures (298, 308 and 318 K). Model parameters were determined by using the data of experimental sets and Langmuir and Freundlich isotherm models. Finally, the effects of

different solution pH (3, 7, 9 and 11) and presence of foreign ion (NaCl: 0.005, 0.01, 0.05 and 0.1 mol/L) on the adsorption capacity were investigated using the optimal adsorbent dose, equilibrium time and 10 mg/L gliclazide solution. The pH values of the stock solution were adjusted using 0.1 M HCl and 0.1 M NaOH solutions.

After the flasks were kept at the specified time and temperature, the adsorbents were separated with the help of magnets. Adsorbates (gliclazide) concentrations remaining from adsorption were analyzed using UV–Vis spectrophotometer (Jasco V-730) at 227 nm, the maximum absorbance wavelength of gliclazide molecule.

3. RESULT AND DISCUSSIONS

3.1. Characterization Results of Magnetic Materials

Characterizations of Magnetic Materials

The structural properties of MAC and MMWCNT, which are the synthesized adsorbents, were characterized by using FTIR, XRD and TGA methods.

FTIR spectra of MAC (1a) and MMWCNT (1b) obtained before and after gliclazide adsorption are shown in Figure 1. FTIR spectra were measured in the range of 400-4000 cm^{-1} . When the spectrum is examined, it is seen that there are similar peaks for both adsorbent before and after adsorption. Fe-O vibrations were seen at 584 cm^{-1} and 447 cm^{-1} in MAC, and at 576 cm^{-1} and 443 cm^{-1} in MMWCNT. These peaks indicate that a band of approximately 583 and 456 cm^{-1} is observed in the tetrahedral site ($\text{Fe}_{\text{tetra}} \leftrightarrow \text{O}$) and octahedral site ($\text{Fe}_{\text{octa}} \leftrightarrow \text{O}$), which is attributed to the intrinsic stress vibrations of the metal (Lei et al., 2017). Carbon materials have similar results for MAC and MMWCNT nanoparticles, such as the peak at 3441-3446 cm^{-1} is indicative of O-H stretching. At the same time, a broad characteristic band of about 3460 cm^{-1} indicates the stretching modes of H_2O molecules or -OH groups on the surface of magnetite particles (Lei et al., 2017). O-H bending peaks are seen at 1380 cm^{-1} for MAC and 1378 cm^{-1} for MMWCNT (Çavuşoğlu et al., 2021). On the other hand, peaks are seen at 1055 and 1043 cm^{-1} , which peaks at around 1100 cm^{-1} corresponding to the bond stretching vibration of C–O (Lei et al., 2017). The peaks observed at 1637-1640 cm^{-1} can be related with C=C vibrations (Liu et al., 2020). C-H peaks occur at around 2903 cm^{-1} for MAC and 2922 cm^{-1} for MMWCNT. These peaks indicate the bond stretching vibration of C-H (Liu et al., 2020). As can be seen, FTIR results are compatible with the literature.

Figure 1: FTIR spectrum before and after gliclazide adsorption of MAC (a) and MMWCNT (b)

XRD patterns for AC, MAC (2a), MMWCNT (2b) and Fe_3O_4 nanoparticles are shown in the Figure 2. All adsorbent materials were compared with those of bare- Fe_3O_4 . In the XRD model of MAC and MMWCNT, six characteristic peaks of the magnetite spinel structure (Fe_3O_4) at 30.16° , 35.62° , 43.30° , 53.72° , 57.14° , and 62.82° were observed to match up to crystal planes 220, 311, 400, 422, 511 and 440, respectively. (Danalıoğlu et al., 2017; Habibi, 2014).

MMWCNT nanoparticles present specific peaks at 25.91° (Shanmugharaj et al., 2007). Therefore, XRD results of MAC and MMWCNT nanoparticles are compatible with the literature.

Figure 2: XRD of raw Fe₃O₄, AC and MAC (a) raw Fe₃O₄, MWCNTP and MMWCNT (b)

220 TGA and DTA graphics of MAC (a) and MMWCNT (b) are shown in Figure 3. As can be seen,
 221 MAC and MMWCNT show similar thermal behavior. The average magnetite content in MAC
 222 adsorbent is 66.7%, while it is 72.1% in MMWCNT. As seen in Figure 3b, the decomposition
 223 temperatures of MAC and MMWCNT determined are 868°C and 911°C, respectively.
 224 According to the final thermal oxidative decomposition temperatures, it was seen that
 225 MMWCNT had higher thermal resistance than MAC adsorbent. The reason for this is the
 226 amount of carbon in their content.

228
 229
 230 **Figure 3:** TGA (a) and DTA (b) curves of MAC and MMWCNT

3.2. Gliclazide Adsorption Studies

In adsorption studies, gliclazide concentrations were measured at 227 nm by UV–Vis spectrophotometer. Figure 4 (a) shows the effect of the adsorbent dose in the range of 0.1–1 g /L on the adsorption capacity of gliclazide.

$$qe = \frac{(Co - Ce).V}{m} \quad (1)$$

The adsorption capacity of the adsorbents (q_e , mg/g) were calculated using Eq. (1). Where Co (mg/L) is the initial concentration of gliclazide, C_e (mg/L) is the equilibrium concentration of gliclazide and V (L) is the volume of the solution. m (g) is the amount of the adsorbent.

At the same time, adsorption efficiencies are calculated by Eq. (2) and its graph is given in Figure 4 (b).

$$\% Eff. = \frac{(Co - Ce)}{Co} \times 100 \quad (2)$$

The effect of adsorbent dose on adsorption capacity (a) and percent removal of gliclazide (b) are shown in Figure 4. The q_e values for MAC ranged from 82.3 mg/g to 12.02 mg/g, while the MMWCNT ranged from 42.59 mg/g to 9.81 mg/g. When the results were examined, as the amount of adsorbent increased, the adsorption capacity decreased. This result is due to the reduction of the total surface area per active adsorption site and/or due to the agglomeration of the nano-adsorbent.

The adsorption efficiency for MAC reaches 97.83% while for MMWCNT it is 75.63%. With a small increase in adsorbent dose, more adsorption surface area regions are available for the drug to be adsorbed. Further increase of the adsorbent does not change the result much because of the absence of adsorbate. In subsequent experimental studies, the adsorbent dose was chosen as 1 mg. The results are in agreement with the literature.

Figure 4: Effect of the adsorbent amount on adsorption capacity (a) and adsorption efficiency (b). Operating conditions: pH = 5.86, T = 298 K, t= 120 min and Co=10 mg/L.

Adsorption kinetics of gliclazide on MAC and MMWCNT adsorbents, nonlinear PFO and PSO kinetic model parameters were calculated using Eq. (3) (Lagergren, 1898) and Eq. (4) (Ho and McKay, 1999), respectively. Where, q_t (mg/g) is the adsorption capacity at t time, k_1 (min^{-1}) is PFO rate constant, k_2 (g/mg min) is PSO rate constant and t (min) is the contact time.

$$q_t = q_e \times (1 - e^{-k_1 x t}) \quad (3)$$

$$q_t = \frac{t_2 q_e^2 t}{1 + k_2 q_e t} \quad (4)$$

Kinetic curves for MAC and MMWCNT generated using experimental data and kinetic models are shown in Figure 5 and model parameters in Table 2. In order to determine the equilibration times for both adsorbents, experiments were carried out at different time intervals using kinetic models. Equilibrium times of MAC and MMWCNT were determined as 120 and 180 minutes, respectively.

Table 2: Pseudo first order and pseudo second order kinetic model parameters for MAC and MMWCNT.

Gliclazide Adsorption	Pseudo first order kinetic model			Pseudo second order kinetic model		
	q_e (mg/g)	k_1 (min ⁻¹)	R^2	q_e (mg/g)	k_2 (g/mg.min)	R^2
MAC	63.66	0.066	0.89	71.45	0.001	0.93
MMWCNT	17.19	0.060	0.80	18.95	0.005	0.81

The parameters of the kinetic models showed that the adsorption process for both adsorbents was compatible with the PFO and PSO kinetic models ($R^2 > 0.80$). When the results were compared with the experimental data and the correlation coefficients were examined, it was seen that the PSO kinetic model was more compatible with a small difference. The experimental data of the adsorption capacity and the results of the model parameters were found to be compatible (MAC: $q_t = 71.45$ mg/g, $q_e = 77.35$ mg/g; MWCNT: $q_t = 18.95$ mg/g, $q_e = 17.64$ mg/g).

Figure 5: Non-linear PFO and PSO kinetic model plots of gliclazide adsorption for (a) MAC and (b) MMWCNT. Operating conditions: pH = 5.86, T = 298 K, m = 0.001 g and Co = 10 mg/L.

To determine the mechanisms and equilibrium conditions of the adsorption systems, the Langmuir Eq.(5) (Langmuir, 1918) and Freundlich Eq. (6) (Freundlich, 1909) isotherms were plotted and the parameters were calculated. Where, K_L (L/mg) is Langmuir isotherm constant,

q_m (mg/g) is the maximum adsorption capacity, K_F is Freundlich isotherm constant (L/g), and n is the heterogeneity factor.

$$q_e = \frac{K_L q_m C_e}{1 + K_L C_e} \quad (5)$$

$$q_e = K_f C_e^{1/n} \quad (6)$$

The nonlinear Langmuir and Freundlich isotherm plots of MAC and MMWCNT are shown in Figure 5 (a) and (b), respectively, and the isotherm model parameters are shown in Table 3. Although temperature is not a very influential parameter, the optimum temperature for MAC and MMWCNT is 298 K and 308 K, respectively. When the regression coefficients were compared, higher values ($R^2 = 0.76-0.96$) were obtained for the Langmuir isotherm for both adsorbents. For this reason, it is seen that the adsorption system of gliclazide ions on carbon-based adsorbents is more compatible with the Langmuir isotherm. The maximum adsorption capacities (q_m) for MAC and MMWCNT ranged from 101.66-109.20 mg/g and 28.29-71.59 mg/g, respectively. The maximum adsorption capacity for MAC was 109.20 mg/g at 298 K, while the maximum adsorption capacity for MMWCNT was 71.59 mg/g at 318 K. Although the temperature did not make a significant difference on the adsorption capacity, better results were obtained at lower temperatures. It is known that the adsorption process is positive when the n parameter takes values between 1 and 10. The n values for both adsorbents vary between 1 and 2, and it can be said that gliclazide adsorption brings to a successful conclusion.

Table 3: Langmuir and Freundlich isotherm parameters for MAC and MMWCNT.

Gliclazide Adsorption		Langmuir isotherm			Freundlich isotherm		
	T	q_m (mg/g)	K_L (L/mg)	R^2	K_F (L/mg)	n	R^2
MAC	298 K	109.20	0.55	0.88	37.71	1.84	0.82
	308 K	101.66	0.68	0.92	39.09	1.91	0.88
	318 K	105.58	0.49	0.76	33.91	1.80	0.73
MMWCNT	298 K	48.45	0.07	0.96	3.59	1.29	0.94
	308 K	71.59	0.03	0.90	2.45	1.14	0.88
	318 K	28.29	0.14	0.93	4.20	1.65	0.84

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320 **Figure 5:** Langmuir and Freundlich adsorption isotherms for (a) MAC and (b) MMWCNT at
 321 different temperatures (298, 308, 318 K). Operating conditions: pH=5.86, t=120 min (MAC);
 322 180 min (MMWCNT), m= 0.001 g and Co=2,4,6,8,10 mg/L.

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The effect of solution pH (a) and foreign ion (b) on adsorption capacity is shown in figure 6. pH is one of the most important process variables that directly affects the adsorbate uptake by the adsorbent because it can affect the extent of adsorbate ionization and the surface properties of the adsorbent (Iftekhhar et al., 2018). As seen in Figure 6 (a), the acidity of the environment is a significant factor affecting the adsorption of gliclazide against both adsorbents. The removal of gliclazide from aqueous solution decreased steadily with increasing pH. As a result of the experiments carried out at different pHs, it can be said that the MAC and MMWCNT surfaces are negatively charged with gliclazide adsorption and give better results at low pH values. Therefore, the pH value of the drug solution (pH=5.86) was found to be suitable for adsorption studies.

In order to estimate the adsorption capacities of adsorbents in real environment, different ion effects that exist with the pollutant are examined. In this study, different concentrations of NaCl (0.001-0.1 mol/L) were used to see the effect of different ions. As seen in Figure 6 (b), the presence of different concentrations of NaCl ion did not significantly affect the gliclazide adsorption efficiency on MAC and MMWCNT. The q_e values for MAC ranged from 59.82 mg/g to 62.59 mg/g, while the MMWCNT ranged from 16.42 mg/g to 19.45 mg/g.

Figure 6: Effects of the solution pH (a) and presence of foreign ions (b) on adsorption capacity. Operating conditions: $t=120$ min (MAC); 180 min (MMWCNT), $m= 0.001$ g and $T=298$ K.

4. CONCLUSION

MAC and MMWCNT nanoparticles used as adsorbent were magnetized using co-precipitation method and characterized using FTIR, XRD and TGA techniques. For the removal of gliclazide from aqueous solutions, the adsorption method was chosen and various parameters affecting the adsorption system were investigated. The highest adsorption capacities were obtained for both adsorbent at 1 mg. Equilibrium times for MAC and MMWCNT were determined as 120 and 180 minutes, respectively, as a result of kinetic studies. When the kinetic model parameters of PFO and PSO were examined, gliclazide adsorption reaction kinetics gave results more compatible with PSO. According to the data of kinetic studies, MMWCNT ($k_2=0.005$ g/mg min) adsorbent was found to be a faster adsorbent than MAC ($k_2=0.001$ g/mg.min). Adsorption experiments were carried out at three several temperatures, 298 , 308 and 318 K, to define the mechanism of the hypoglycemic drug-gliclazide adsorption system. Langmuir and Freundlich equilibrium isotherm models were applied to these experimental data. The Langmuir model was more compatible with the adsorption system ($R^2 \geq 0.76$). The maximum adsorption capacities ($q_{\max}$) for MAC and MMWCNT were calculated as 109.20 mg/g ($T=298$ K) and 71.59 mg/g ($T=308$ K), respectively. When the effects of different pH values were examined,

it was observed that the adsorption capacity increased at low pH values. In other words, the pH value (pH=5.86) of the drug solution was found suitable for adsorption studies. The presence of different concentrations of foreign ions, NaCl, did not significantly affect the gliclazide adsorption efficiency on MAC and MMWCNT. As a result of the examination of all experiment results, MAC and MMWCNT showed that there are alternative adsorbents that can be used for the removal of gliclazide contaminants from aqueous solutions. But in addition, it can be said that MAC adsorbent has higher adsorption capacity than MMWCNT and is more economical in terms of cost in removing such a pollutant such as gliclazide.

387 **DECLARATIONS**

388

389 **Ethics approval and consent to participate**

390 Not applicable

391 **Consent for publication**

392 Not applicable

393 **Availability of data and materials**

394 The datasets used and/or analysed during the current study are available from the corresponding
395 author on reasonable request.

396 **Competing interests**

397 Not applicable

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399 Not applicable

400 **Authors' contributions**

401 Ferda Civan Çavuşoğlu: Conceptualization, Methodology, Validation, Writing-Review &
402 Editing.

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