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Action 		Manuscript Number	Title 
<a href="#">Action Links</a>		APYA-D-21-02809	Kinetic, isot and thermodyna studies on C adsorption cellulose acetate/gra oxide comp nanofibers 

# Applied Physics A

## Kinetic, isotherm and thermodynamic studies on Cr(VI) adsorption using cellulose acetate/graphene oxide composite nanofibers --Manuscript Draft--

<b>Manuscript Number:</b>	APYA-D-21-02809R2
<b>Full Title:</b>	Kinetic, isotherm and thermodynamic studies on Cr(VI) adsorption using cellulose acetate/graphene oxide composite nanofibers
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<b>Order of Authors Secondary Information:</b>	
<b>Funding Information:</b>	
<b>Abstract:</b>	<p>In the present study, cellulose acetate/graphene oxide (CA/GO) composite nanofibers were prepared via electrospinning method to remove Cr(VI) ions from aqueous solution via adsorption process in a batch mode. The impact of adsorption parameters, including contact time, pH, Cr(VI) concentration and temperature was investigated to obtain the optimum conditions for the uptake of maximum Cr(VI) ions from water within a short time. The kinetic data of Cr(VI) adsorption were well fitted by both pseudo-first-order and pseudo-second-order kinetic models, whereas, Redlich-Peterson, Langmuir and Freundlich, isotherm models were used to describe the equilibrium data of Cr(VI) adsorption by the CA/GO nanofibers. The effect of temperature on the adsorption capacity of Cr(VI) ions using the nanofibers indicated that the higher temperatures were favorable for higher adsorption of Cr(VI) ions using the nanofibers. The thermodynamic parameters results indicated the spontaneous and endothermic of Cr(VI) sorption nature using the CA/GO nanofibrous adsorbent. The maximum monolayer adsorption capacity of nanofibers toward Cr(VI) ions sorption was 422.3 mgg<sup>-1</sup> which was comparable with other adsorbents. The reusability of composite nanofibers was carried out for five adsorption-desorption cycles. The obtained results exhibited the high capability of CA/GO nanofibrous adsorbents for Cr(VI) ions sorption from actual wastewater.</p>

**Response to Reviewers:**

Dear editor,  
I would like thank you for your uninterrupted and praiseworthy concern. I am also so grateful for the referee attention on this work, being appreciative to his/her precious suggestions and reminders, which helped us to improve the manuscript. The comment was carefully considered, and the detailed point-by-point response to it is listed below, and also highlighted in the main text.

Comments for APYA-D-21-02809R1:

The authors have carefully revised the whole manuscript and the quality of this work has been greatly improved. However, there is still a minor comment that the authors should take into consideration before publication:

The authors responded that: "As observed in SEM images, the fiber diameter was increased by loading GO into the nanofibers up to 2 wt.% which resulted in increasing fiber diameter. By increasing fiber diameter, the voids between fibers were increased which could be resulted in increasing porosity, specific surface area and adsorption capacity." However, to the extent of our knowledge, the specific surface area will decrease as the diameter of nanofiber increase. [Ref: 10.1016/j.apmate.2021.09.004] In this work, the interruption of GO can reasonably lead to the increase in porosity, specific surface area and adsorption capacity, because of the inherently large surface area and abundant active adsorption sites in the carbon nanomaterials.

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By loading GO into the nanofibers up to 2 wt.%, the increase in the adsorption capacity of Cr(VI) ions by the composite nanofibers could be attributed to the high porosity, large specific area and higher available active sites of GO for Cr(VI) ions sorption.

We hope the responses and corrections made in the revised manuscript meet with the reviewer's considerations.

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# Kinetic, isotherm, and thermodynamic studies on Cr(VI) adsorption using cellulose acetate/graphene oxide composite nanofibers

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## Abstract

In the present study, cellulose acetate/graphene oxide (CA/GO) composite nanofibers were prepared via the electrospinning method to remove Cr(VI) ions from aqueous solution via adsorption process in a batch mode. The impact of adsorption parameters, including contact time, pH, Cr(VI) concentration, and temperature was investigated to obtain the optimum conditions for the uptake of maximum Cr(VI) ions from water within a short time. The kinetic data of Cr(VI) adsorption were well fitted by pseudo-first-order and pseudo-second-order kinetic models, whereas, Redlich-Peterson, Langmuir and Freundlich, isotherm models were used to describe the equilibrium data of Cr(VI) adsorption by the CA/GO nanofibers. The effect of temperature on the adsorption capacity of Cr(VI) ions using the nanofibers indicated that the higher temperatures were favorable for higher

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adsorption of Cr(VI) ions using the nanofibers. The thermodynamic parameters results indicated the spontaneous and endothermic of Cr(VI) sorption nature using the CA/GO nanofibrous adsorbent. The maximum monolayer adsorption capacity of nanofibers toward Cr(VI) ions sorption was  $422.3 \text{ mgg}^{-1}$  which was comparable with other adsorbents. The reusability of composite nanofibers was carried out for five adsorption-desorption cycles. The obtained results exhibited the high capability of CA/GO nanofibrous adsorbents for Cr(VI) ions sorption from actual wastewater.

**Keywords:** Nanofibers; Cellulose acetate; Cr(VI); Graphene oxide; Adsorption.

## 1. Introduction

The discharge of chromium compounds from wastewater of different industries such as pigments, glass, metal finishing, electroplating, electrical equipment, and magnetic tapes into the environment produces high toxicity and endanger animal, plant, and human life [1-7]. The permissible concentration of Cr(VI) for drinking water should be lower than  $50 \text{ }\mu\text{g mL}^{-1}$  based on World Health Organization [2]. The high level of Cr(VI) ions in water is mutagenic and carcinogenic for various organs [2]. Therefore, the Cr(VI) ions in water event its lower level should be removed before its discharge to the environment. Various techniques including precipitation [8-13], adsorption [14-17], advance oxidation [18-20], membrane filtration [21], coagulation [22] and so on were used to remove Cr(VI) ions and other contaminants from water. The adsorption process is a favorable method for decreasing the level of Cr(VI) ions in water due to its low cost and good efficiency [23]. The efficiency of adsorption process is related to the level of available active sites of adsorbent for conjugating with adsorbate ions and their adsorption. Nanofibers prepared by the electrospinning method are good candidates for the adsorption of various contaminants from water due to their high porosity and large specific surface area [24-31]. Cellulose-based nanofibers could be considered as effective adsorbents for the removal of toxic matters from water [32,33]. However, using pure cellulose is limited due to the easier dissolving in water. Cellulose acetate (CA) as ester cellulose exhibited higher mechanical properties in water than pure cellulose. Recently, the electrospun CA nanofibers prepared by the electrospinning method are recommended for different wastewater treatments [34-36]. Furthermore, inorganic nanomaterials such as carbon nanotubes [26], titanium dioxide [37], ferrite nanoparticles [38] were incorporated into the CA nanofibers to increase the strength and adsorption capacity of CA nanofibers. For instance, single-walled carbon nanotubes, ferrite, and titanium oxide nanoparticles into the CA nanofibers were incorporated to remove metal ions and dyes from water [26]. CA/TiO<sub>2</sub> composite nanofibrous adsorbent was synthesized for the removal of heavy metal ions [37]. The potential of CA/chitosan coated by ferrite nanoparticles

1 for the adsorption of phenol and metal ions has been investigated [38]. Electrospun CA/hydroxyapatite  
2 nanocomposite nanofibers were synthesized for the adsorption of Fe(III) and Pb(II) from water [39]. Graphene  
3 oxide (GO), due to its large scale production, low cost, high mechanical properties, and hydrophilic nature, is  
4 often blended with the polymeric solution to obtain the composite nanofibers with improved mechanical strength  
5 and high removal efficiency [40-43]. The aim of this study is to incorporate the GO into the CA nanofibers for  
6 increasing the mechanical strength and adsorption capability of nanofibers toward Cr(VI) ions sorption.  
7

8 The present work studies the capability of CA/GO composite nanofibers for the adsorption of Cr(VI) ions in a  
9 batch system under different adsorption variables such as contact time, adsorbent dosage, pH, Cr(VI)  
10 concentration, and temperature. The kinetic and equilibrium data of Cr(VI) adsorption by the composite  
11 nanofibers were analyzed by the pseudo-first-order and pseudo-second-order kinetic and Redlich-Peterson,  
12 Langmuir, Freundlich, isotherm models, respectively. The thermodynamic parameters were also investigated  
13 based on the impact of temperature on the Cr(VI) ions sorption using the CA/GO composite nanofibers.  
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## 26 **2. Experimental**

### 27 **2.1. Material and Methods**

28 Cellulose acetate (Mw:30 kDa, CA), and trifluoroacetic acid (TFA) supplied from Sigma-Aldrich (Germany) were  
29 used for the preparation of CA solutions. Cr(VI) ions solution was prepared by dissolving a predetermined amount  
30 of potassium dichromate (Sigma-Aldrich, Germany) in distilled water. The morphology and mean fiber diameter  
31 were determined by using a scanning electron microscope (JSM-6380, JEOL, Germany, SEM) and an image  
32 analyzer, respectively. The gold coating was implemented on nanofibrous samples using a sputter coater (Polaron,  
33 SC7620). The as-prepared nanocomposites were dispersed in water and cast onto a copper grid to study the size  
34 and morphology of the synthesized materials by TEM (Transmission Electron Microscope) using a Philips –  
35 CM300 - 150 kV microscope.  
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### 48 **2.2. Preparation of CA/GO composite nanofibers**

49 The modified Brodie method was used to synthesize graphite oxide nanosheets from graphite powder, potassium  
50 permanganate, sulfuric acid and orthophosphoric acid as described previously [44]. CA solution (10 wt.%) was  
51 prepared by dissolving 1 g CA in 10 mL TFA under stirring for 4 h. Various GO content (0, 0.5, 1, 1.5, and 2  
52 wt.%) was added to the CA solution and sonicated for 30 min to prepare CA/GO solutions. The prepared solutions  
53 were transferred into a 5 mL plastic syringes attached to the syringe pump. The electrospinning condition for  
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1 fabrication of CA and CA/GO nanofibers was an applied voltage of 20 kV, the feeding rate of 0.3 mLh<sup>-1</sup>, the  
 2 distance of 12 cm, the temperature of 25%, and the humidity of 40%.

### 6 2.3. Adsorption process in a batch system

7 Adsorption experiments were implemented by immersing 20 mg nanofibers (CA and CA/GO composite  
 8 nanofibers) into the 250 mL flasks containing 100 mL of 50 mgL<sup>-1</sup> Cr(VI) at 200 rpm and 25 °C. The effect of  
 9 GO content (0-3 wt.%), pH (2-8), contact time (5-60 min), initial concentration of Cr(VI) (10-1000 mgL<sup>-1</sup>), and  
 10 temperature (25-45°C) on the Cr(VI) sorption using nanofibers was investigated. The adsorption capacity (q mgg<sup>-1</sup>)  
 11 was evaluated based on the initial (C<sub>0</sub>, mgL<sup>-1</sup>) and equilibrium (C<sub>e</sub>, mgL<sup>-1</sup>) concentrations of adsorbate, solution  
 12 volume (V, L) and nanofibrous adsorbent weight (g) as follows:

$$13 q_e = \frac{(C_0 - C_e)V}{M} \quad (\text{Eq.1})$$

14 The known pseudo-first-order (Eq. 2), pseudo-second-order (Eq. 3) kinetic models and Freundlich (Eq. 4),  
 15 Langmuir (Eq. 5), and Redlich-Peterson (Eq. 6) isotherm models were utilized for describing the kinetic and  
 16 equilibrium data of Cr(VI) ions sorption using nanofibers [45]. The adsorption nature of Cr(VI) ions by the  
 17 composite nanofibers was further investigated by evaluating the thermodynamic parameters such as Gibbs free  
 18 energy change (ΔG°), enthalpy change (ΔH°), and entropy change (ΔS°). The reusability of nanofibers for Cr(VI)  
 19 sorption was implemented for five cycles by their immersion in 1 M HCl for 2 h. The final concentration of Cr(VI)  
 20 ions is measured by an inductivity-coupled plasma atomic emission spectrophotometer (ICP-AES, Thermo Jarrel  
 21 Ash, Model Trace Scan).

$$22 \text{ Pseudo-first-order } q_t = q_e(1 - \exp(-k_1 t)) \quad (\text{Eq. 2})$$

$$23 \text{ Pseudo-second-order } q_t = \frac{k_2 q_e^2 t}{1 + k_2 q_e t} \quad (\text{Eq. 3})$$

$$24 \text{ Freundlich } q_e = k_F C_e^{1/n} \quad (\text{Eq. 4})$$

$$25 \text{ Langmuir } q_e = q_m \frac{b C_e}{1 + b C_e} \quad (\text{Eq. 5})$$

$$26 \text{ Redlich-Peterson } q_e = \frac{P C_e}{1 + \alpha C_e^\beta} \quad (\text{Eq. 6})$$

### 3. Results and Discussion

#### 3.1. Morphology of GO and CA/GO composite nanofibers

SEM images from GO and GO-loaded CA nanofibers with various concentrations (0-2.5 wt.%) are presented in Figure 1. As shown in the figure, the sheet structure with nano-scale thickness was obtained for synthesized GO. For pure CA, the homogenous nanofibers with free of beads with an average diameter of  $210\pm 70$  nm were achieved. By loading 0.5 wt.% GO into the nanofibers, the fiber diameter was gradually increased due to its physical loading and increase in the solution viscosity. By increasing GO concentration up to 2 wt.%, the fiber thickness was increased and GO nanosheets were successfully incorporated into the nanofibers without aggregation in the nanofibers surface. By incorporating 2.5 wt.% GO, the fiber diameter was significantly increased and some GO have been aggregated on the surface of the nanofibers. The uniform dispersion of GO into the CA solution resulted in production of uniform nanofibers for CA/GO solutions containing 0.5, 1, and 2 wt.% with an average diameter of  $260\pm 110$ ,  $320\pm 130$ , and  $430\pm 140$  nm, respectively. Whereas, the beaded-fibers with an average diameter of  $530\pm 150$  nm were obtained for CA/GO 2.5 wt.% composite nanofibers.

**Insert figure 1 here.**

TEM images of the nanocomposites nanofibers are presented in Figure 2. The images shown in the figure demonstrate the wire morphology of the nanocomposites. However, GO nanosheets exhibited a bulk structure (Figure 2a). Figure 2 b-f shows that compositing CA with GO increases the thickness size of the composite. According to the images, it is found that the thickness size of the structures is increased by increasing the GO weight content ratio in the nanocomposite mixture. According to the TEM images and the scale bar sizes, the thickness size of the rod-like structure samples is 50, 70, 120, 200, 300 nm, respectively, for CA, CA/GO 0.5 wt.%, CA/GO 1 wt.%, CA/GO 2 wt.%, and CA/GO 2.5 wt.% composite nanofibers.

**Insert figure 2 here.**

#### 3.2. Effect of GO concentration on Cr(VI) ions adsorption

The effect of GO content in the CA/GO nanofibers on the Cr(VI) adsorption capacity under 0.2 g/L adsorbent dosage, pH:2, Cr(VI) concentration of 50 mg/L, 25 °C, and GO content ranging from 0-3 wt.% is presented in Figure 3. As shown, by increasing GO concentration up to 2 wt.%, the adsorption capacity of Cr(VI) ions by the composite nanofibers was increased and further increase in GO content resulted in decreasing the adsorption capacity due to aggregation of GO on the nanofibers matrix and decreasing the available active sites for Cr(VI) ions sorption as confirmed by SEM images. **By loading GO into the nanofibers up to 2 wt.%, the increase in the adsorption capacity of Cr(VI) ions by the composite nanofibers could be attributed to the high porosity, large**

specific area and higher available active sites of GO for Cr(VI) ions sorption. Moreover, the high amounts of hydroxyl groups of GO and the redox reaction of -OH groups with Cr(VI) ions led to the removal of Cr(VI) ions from water by loading GO and enhancement of GO content up to 2 wt.% [46]. Furthermore, hydroxyl groups of GO could be oxidized into the carboxylic acid by reducing Cr (VI) ions to Cr (III) ions. After that, Cr (III) ions could be easily adsorbed on the nanofibers surface. Moreover, the carboxylic groups of CA could bond with Cr (VI) ions, via the surface complexation mechanisms. Therefore, the main mechanisms to remove Cr(VI) ions, are absorbance through the nanofibers porosity, ionic exchange, surface complexation and reduction. The CA/GO 2wt.% composite nanofibrous adsorbent is selected for further experiments. The schematic of Cr(VI) ions sorption mechanism using CA/GO composite nanofibers is illustrated in scheme 1.

**Insert figure 3 here.**

**Insert scheme 1 here.**

### **3.3. Effect of pH on the Cr(VI) ions sorption**

The effect of pH on the Cr(VI) ions sorption using CA/GO 2 wt.% composite nanofibers under 0.2 g/L adsorbent dosage, Cr(VI) concentration of 50 mg/L, 25 °C, and different pH values ranging from 2-8 wt.% is presented in Figure 4. As shown in the image, increasing pH value resulted in decreasing chromium ions adsorption capacity of composite nanofibers and the maximum adsorption capacity was found at pH=3. The positive charge of CA and GO led to adsorb Cr(VI) ions ( $\text{HCrO}_4^-$  and  $\text{Cr}_2\text{O}_7^{2-}$ ) at pH 3. By increasing pH, the positive charge of the adsorbent was decreased, which resulted in decreasing the adsorption capacity. The significant decrease in Cr(VI) adsorption after pH=7 could be attributed to the competition between chromium ions and hydroxyl groups, which blocked the large numbers of nanofibrous adsorbent active sites and significantly reduced its adsorption capacity. The obtained results indicated that the electrostatic interaction plays a significant role in the adsorption of Cr(V) ions by the composite nanofibers. Therefore, pH=3 is selected for further experiments.

**Insert figure 4 here.**

### **3.4. Effect of contact time on the Cr(VI) ions sorption and kinetic models**

The impact of contact time on Cr(VI) ions sorption using CA/GO 2 wt.% composite nanofibers under 0.2 g/L adsorbent dosage, pH=3, Cr(VI) concentration of 50 mg/L, 25 °C and contact time in the range of 5-60 min is presented in Figure 5. It is found that the fast adsorption of Cr(VI) ions using CA/GO composite nanofibers occurred for 15 min and reached to equilibrium after only 30 min. The fast adsorption during 15 min could be attributed to the high surface area, high porosity and high amounts of functional groups for Cr(VI) ions sorption. The lower adsorption capacities of Cr(VI) ions by the adsorbent were due to the diffusion of Cr(VI) ions to the

inner pores of nanofibers, and interaction of some functional groups of GO doped-CA nanofibers. Therefore, 30 min is selected as an equilibrium time for Cr(VI) ions sorption. The experimental Cr(VI) sorption data were fitted by pseudo-first-order, and pseudo-second-order kinetic models which results are presented in Table 1. The data revealed that the pseudo-second order kinetic model described the kinetic data of Cr(VI) ions sorption using the synthesized composite nanofibrous adsorbent. This behavior indicated that the chemisorption is a predominant mechanism for the Cr(VI) ions sorption by the CA/GO nanofibers.

**Insert figure 5 here.**

**Table 1.** Kinetic parameters of Cr(VI) ions sorption using synthesized nanofibers.

Experimental		Pseudo-first-order			Pseudo-second-order			
$q_t$ (mg g <sup>-1</sup> )	$q_e$ (mg g <sup>-1</sup> )	$k_1$ (min <sup>-1</sup> )	$R^2$	RMSE	$q_e$ (mg g <sup>-1</sup> )	$k_2$ (g.mg <sup>-1</sup> .min <sup>-1</sup> )	$R^2$	RMSE
212.4	185.23	0.05428	0.965	5.241	224.5	0.00781	0.990	0.491

### 3.5. Effect of initial concentration on the Cr(VI) ions sorption and isotherm models

The impact of initial concentration on the Cr(VI) ions sorption using CA and CA/GO 2 wt.% composite nanofibers under 0.2 g/L adsorbent dosage, pH=3, contact time 30 min, 25 °C, and different initial concentrations ranging from 10-1000 mg/L is presented in Figure 6. Figure 6 shows that increasing the Cr(VI) concentration increases the adsorption capacity of nanofibers toward Cr(VI) ions sorption up to an initial concentration of 500 mg/L Cr(VI) due to increasing the driving force. After that, the adsorption capacity did not change by further enhancement of Cr(VI) ions concentration due to the saturation of active sites of nanofibrous adsorbent.

The equilibrium data were analyzed by Freundlich, Langmuir, and Redlich-Peterson isotherm models. The results are presented in Table 2. The Redlich-Peterson isotherm model described well the equilibrium data of Cr(VI) ions sorption compared to the Langmuir and Freundlich isotherm models by comparing  $R^2$  and RMSE values. The “n” value of Redlich-Peterson model was close to 1, which indicated that the monolayer sorption of Cr(VI) using synthesized nanofibers was predominant. The maximum experimental and monolayer sorption capacities of composite nanofibers for Cr(VI) sorption were about 410.20 and 422.3 mg.g<sup>-1</sup>, respectively, at 45 °C. The increasing the adsorption capacity by increasing temperature indicated that the higher temperature is favorable for the adsorption of more Cr(VI) ions from water. The units of T,  $K_F$ ,  $q_{max}$ ,  $K_L$ , P, and  $\alpha$  are °C, mg.g<sup>-1</sup>, mg.g<sup>-1</sup>, L.mg<sup>-1</sup>, L.mg<sup>-1</sup>, and L.mg<sup>-1</sup>, respectively.

**Table 2.** Isotherm parameters of Cr(VI) ions sorption using synthesized nanofibers at different temperatures.

T	Freundlich isotherm					Langmuir isotherm				Redlich-Peterson isotherm				
	q <sub>exp</sub>	K <sub>F</sub>	n	R <sup>2</sup>	RMSE	q <sub>max</sub>	K <sub>L</sub>	R <sup>2</sup>	RMSE	P	α	n	R <sup>2</sup>	RMSE
25	367.3	131.4	4.258	0.955	4.578	395.6	0.0457	0.982	2.774	22.11	0.524	0.915	0.993	0.482
35	390.2	142.5	4.444	0.962	4.014	408.8	0.0518	0.985	2.451	23.54	0.652	0.925	0.992	0.512
45	410.2	153.2	4.782	0.959	4.235	422.3	0.0548	0.988	2.154	25.31	0.845	0.948	0.994	0.459

Insert figure 6 here.

### 3.6. Thermodynamic parameters for Cr(VI) ions sorption

Thermodynamic parameters are evaluated based on the thermodynamic equations and Van't Hoff plot of  $\ln K_C$  versus  $1/T$  as follows [47]:

$$k_C = \lim_{C_{el} \rightarrow 0} \frac{C_{es}}{C_{el}} \quad (\text{Eq. 7})$$

$$\Delta G^0 = -RT \ln k_C \quad (\text{Eq. 8})$$

$$\ln k_C = \frac{\Delta S^0}{R} - \frac{\Delta H^0}{RT} \quad (\text{Eq. 9})$$

where R is the gas constant (8.314 J mol<sup>-1</sup> K<sup>-1</sup>), T is an absolute temperature (K) and k<sub>C</sub> is the adsorption equilibrium constant. C<sub>es</sub> and C<sub>el</sub> are the values of solid-phase concentration and liquid phase concentration at equilibrium in mg L<sup>-1</sup>. The results of Gibbs free energy change ( $\Delta G^0$ ), enthalpy change ( $\Delta H^0$ ), and entropy change ( $\Delta S^0$ ) of the adsorption process are listed in Table 3. The increasing the negative value of  $\Delta G^0$  (KJ/mol) by increase in temperature indicated the spontaneous nature of Cr(VI) ions sorption using nanofibers. The positive  $\Delta H^0$  (KJ/mol) values indicated the endothermic nature of the Cr(VI) ions sorption. The positive  $\Delta S^0$  value confirmed the enhancement of disorder in the solid-solution interface during the Cr(VI) ions sorption.

**Table 3.** Thermodynamic parameters of Cr(VI) ions sorption using synthesized nanofibers.

	K <sub>C</sub>			ΔH°	ΔS°	ΔG°		
	25°C	35°C	45°C			25°C	35°C	45°C
	4.12	13.42	25.31	100.899	0.3435	- 3.507	-6.649	-8.543

### 3.7. Reusability of nanofibers

The synthesized nanofibers regenerated successfully using 1 M HCl and used for five sorption/desorption cycles under 0.2 g/L adsorbent dosage, pH: 3, contact time 30 min, 25 °C and initial concentrations 50 mg/L, which results are presented in Figure 7. The data indicated that the adsorption capacity did not significantly change after five cycles. The obtained results indicated the high capability of synthesized nanofiber for the adsorption of Cr(VI) ions from real wastewater in the future.

**Insert figure 7 here.**

The maximum adsorption capacity of Cr(VI) ions using synthesized nanofibers was compared with other nanofibrous adsorbents in the literature, which results are presented in Table 4. As shown, the adsorption capacity of CA/GO composite nanofibers for Cr(VI) ions sorption was comparable and moderately higher than those of many corresponding nanofibrous adsorbents in the literature.

Table 4 Comparison of adsorption capacity of Cr(VI) sorption using CA/GO nanofibers with other nanofibers in the literature [24, 25, 38, 48-52].

Adsorbent	Adsorbate	$q_{\max}$ (mgg <sup>-1</sup> )	Ref.
Chitosan/graphene oxide fibers	Cr(VI)	310.4	[24]
Chitosan-g-PNVCL/ZIF-8 nanofibers	Cr(VI)	495.6	[25]
CA/chitosan/Ferrite nanofibers	Cr(VI)	193.2	[38]
Chitosan/MWCNTs/Fe <sub>3</sub> O <sub>4</sub> nanofiber	Cr(VI)	360.1	[48]
Polypyrrole-polyaniline	Cr(VI)	227.0	[49]
Polyacrylonitrile/NH <sub>2</sub> nanofibers	Cr(VI)	156.0	[50]
Stacked chitosan nanofibers	Cr(VI)	131.58	[51]
Sodium alginate-polyaniline nanofibers	Cr(VI)	73.34	[52]
CA/GO nanofibers	Cr(VI)	422.3	This study

#### 4. Conclusion

In this work, different GO concentrations (0-2.5 wt.%) were successfully loaded into the CA nanofibers to produce CA/GO composite nanofibers for the adsorption of Cr(VI) ions from aquatic systems. The mean fiber diameter of CA/GO nanofibers containing 0, 0.5, 1, 2, and 2.5 wt.% GO was about 210±70 nm, 260±110, 320±130, 430±140 nm, and 530 ±150 nm, respectively. The maximum experimental and monolayer adsorption capacities of CA/GO composite nanofibers for Cr(VI) sorption were about 410. 20 and 422.3 mg.g<sup>-1</sup>, respectively, under 2 wt.% GO,

0.2 g/L adsorbent dosage, pH: 3, contact time 30 min, and 45 °C. The spontaneous and endothermic nature of Cr(VI) sorption using the CA/GO nanofibrous adsorbent were obtained by evaluating thermodynamic parameters. Nanofibers were successfully regenerated for five adsorption-desorption cycles without significant change in the adsorption capacity of composite nanofibers for Cr(VI) ions sorption.

### Conflict of interest

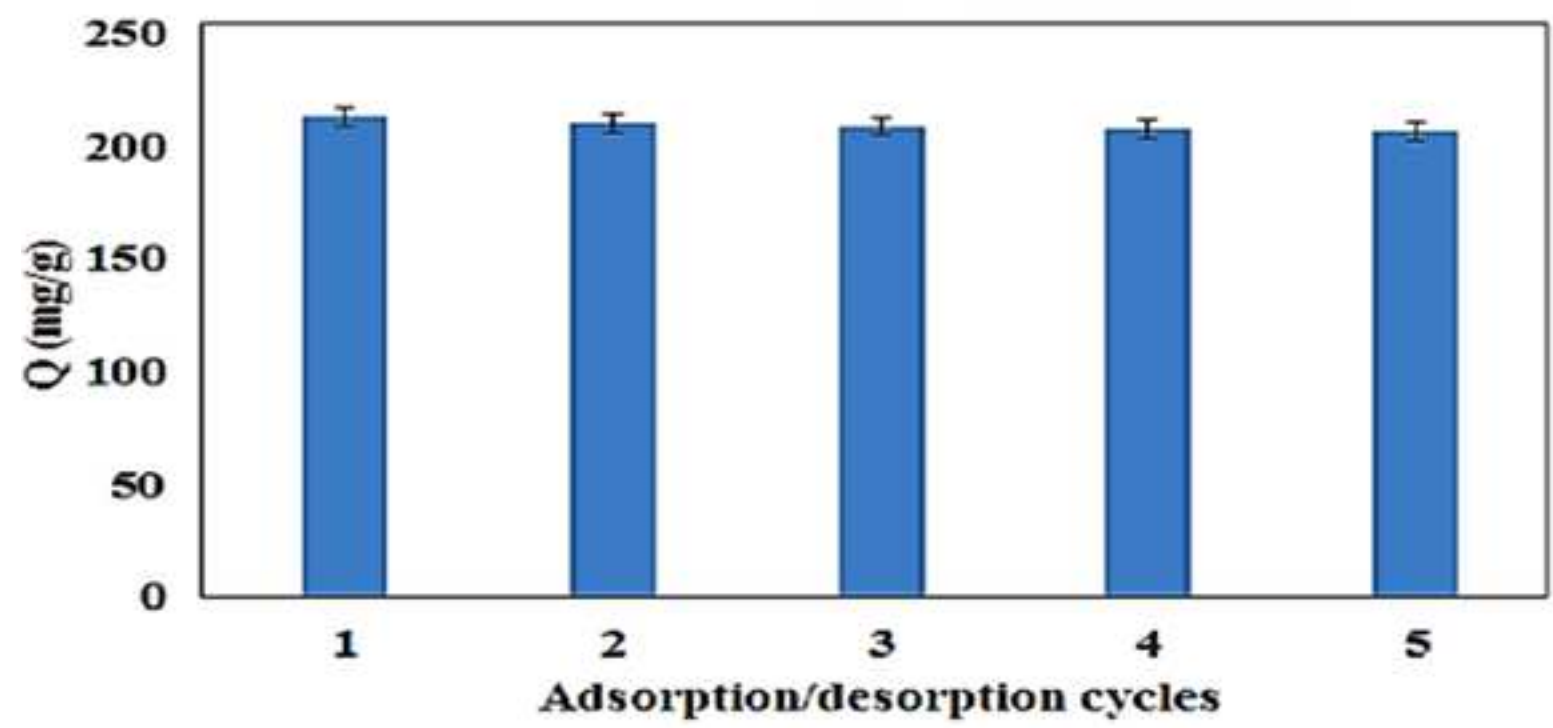
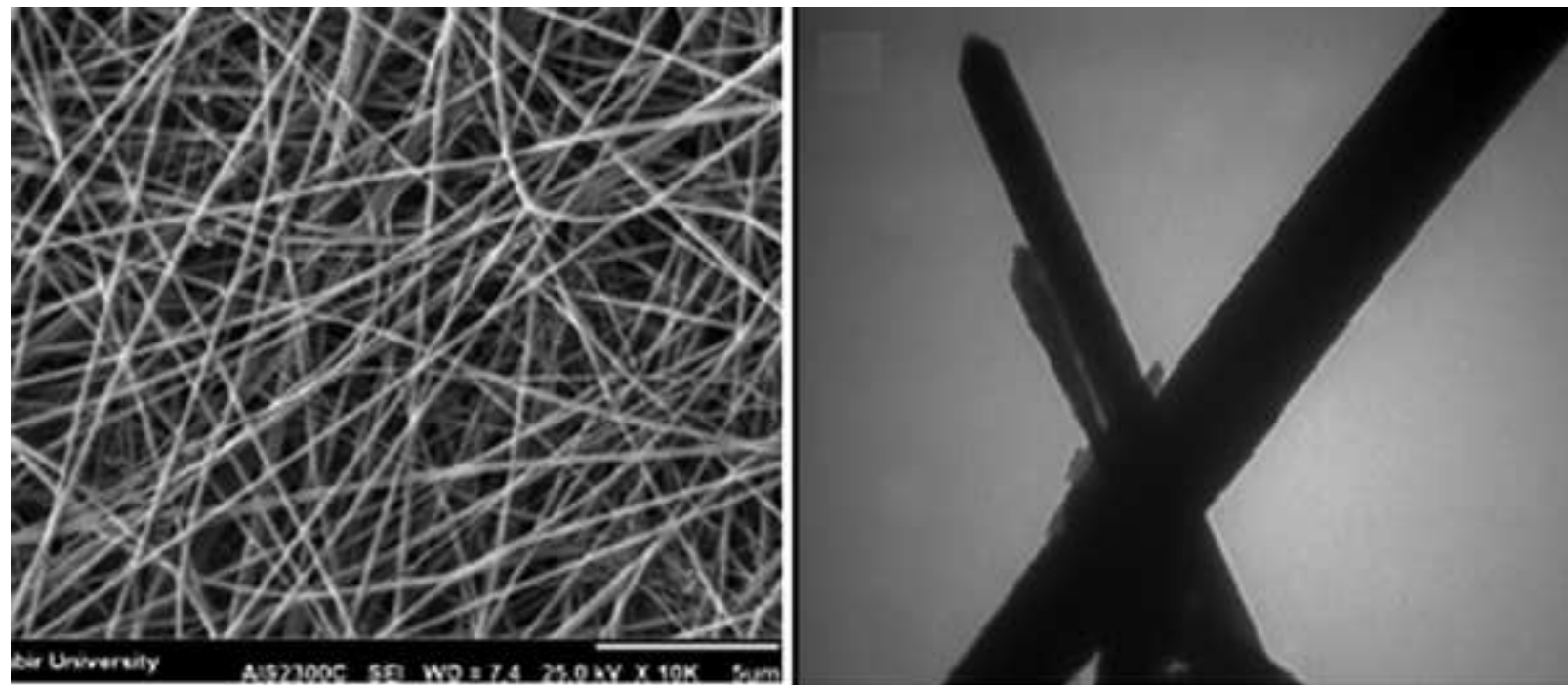
The authors declare that they have no competing interests or personal relationships that could have appeared to influence the work reported in this paper.

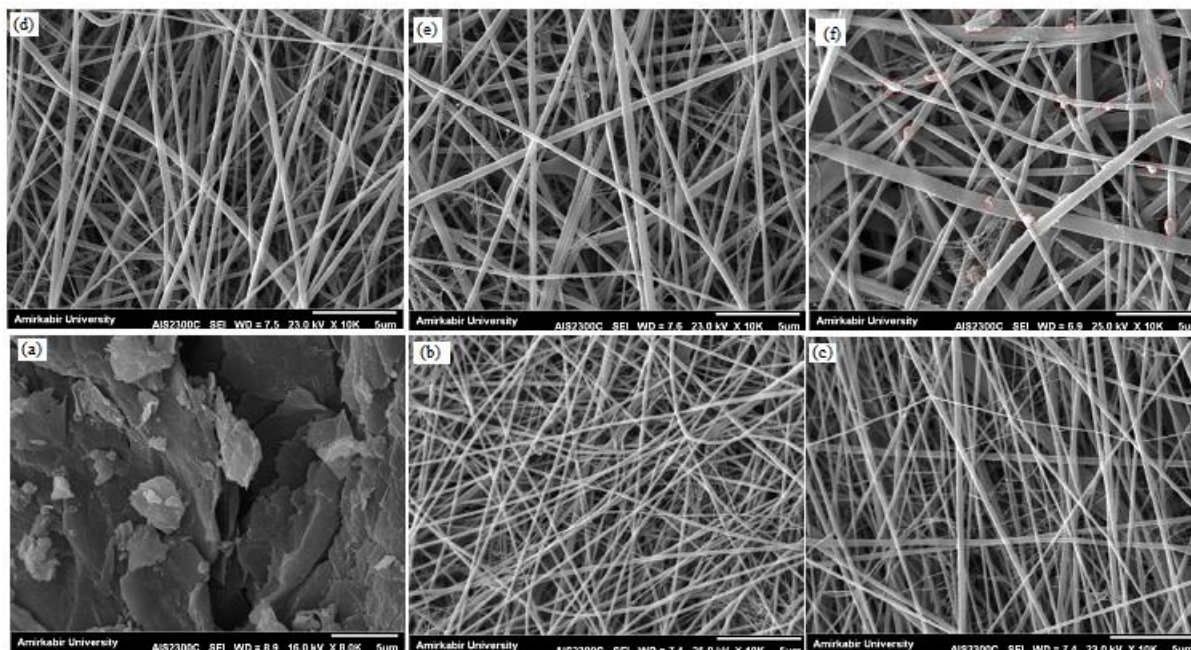
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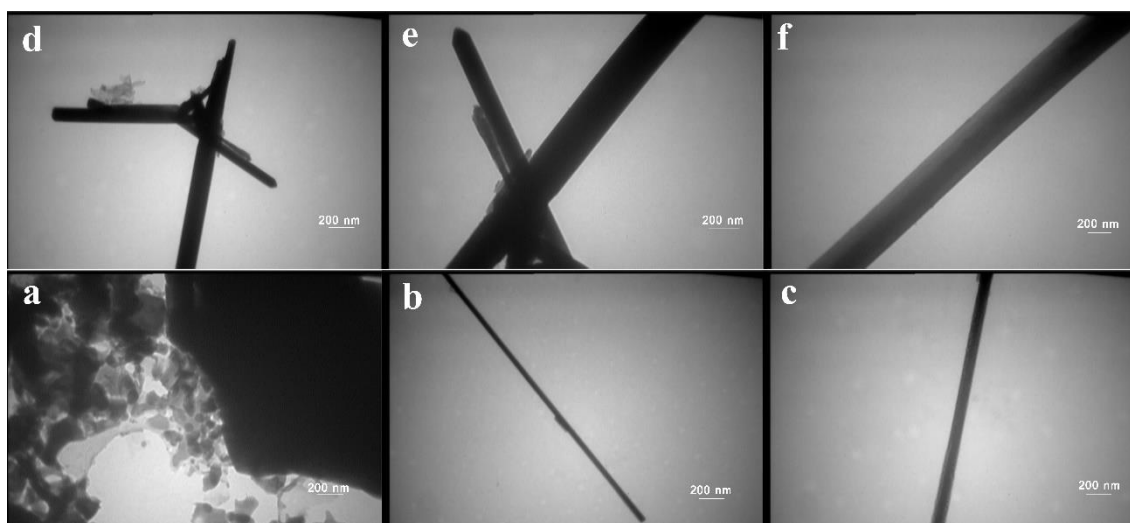
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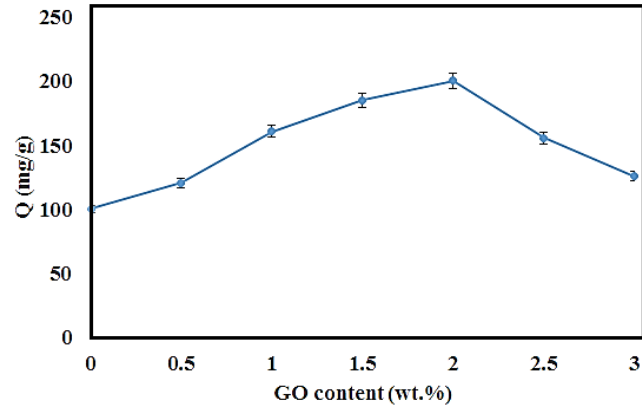




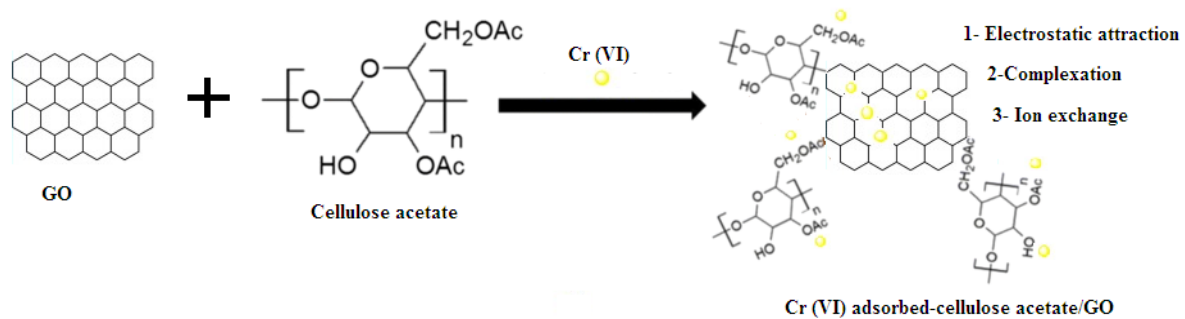
**Figure 1.** SEM images from (a) GO, (b) CA, (c) CA/GO 0.5 wt.%, (d) CA/GO 1 wt.%, (e) CA/GO 2 wt.%, and (f) CA/GO 2.5 wt.% composite nanofibers.



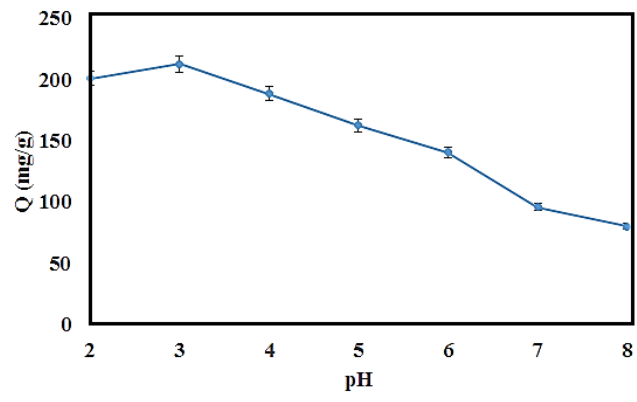
**Figure 2.** TEM images of a) a) GO, (b) CA, (c) CA/GO 0.5 wt.%, (d) CA/GO 1 wt.%, (e) CA/GO 2 wt.%, and (f) CA/GO 2.5 wt.% composite nanofibers.



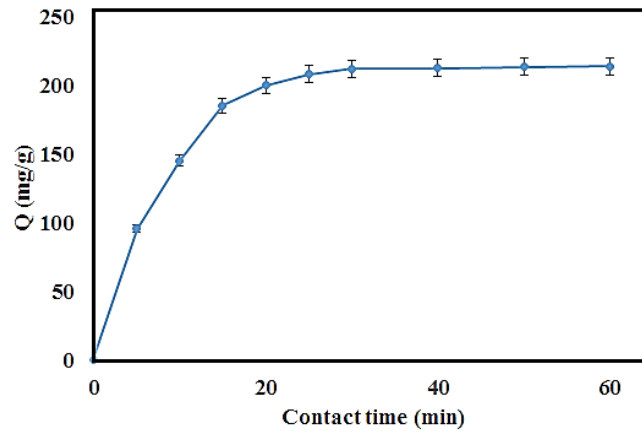
**Figure 3.** Effect of GO content on the adsorption capacity of CA/GO composite nanofibers to Cr(VI) ions.



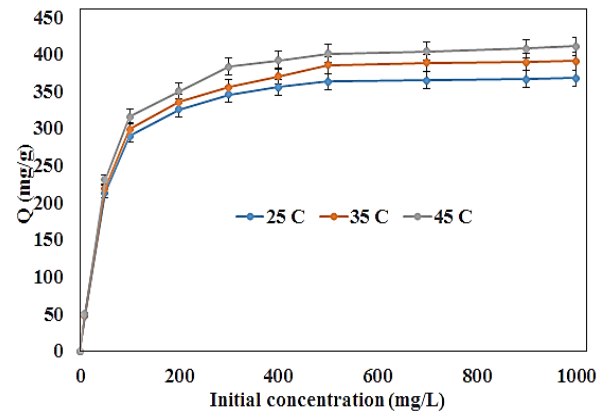
**Scheme 1.** Adsorption mechanisms of Cr(VI) ions using CA/GO composite nanofibers.



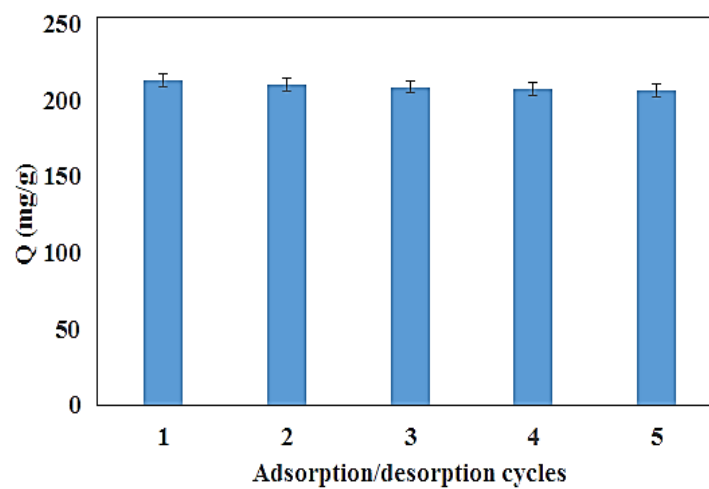
**Figure 4.** Effect of pH on the adsorption capacity of CA/GO composite nanofibers to Cr(VI) ions.



**Figure 5.** Effect of contact time on the adsorption capacity of CA/GO composite nanofibers to Cr(VI) ions.



**Figure 6.** Effect of initial concentration on the adsorption capacity of CA/GO composite nanofibers to Cr(VI) ions at different temperatures.



**Figure 7.** Five adsorption–desorption cycles of Cr(VI) ions sorption by the CA/GO composite nanofibers.

- Cellulose acetate/graphene oxide nanocomposite was used to adsorb Cr(VI) ions from aqueous solution.
- Redlich-Peterson, Langmuir and Freundlich models were used to study equilibrium data of Cr(VI) adsorption.
- CA/GO nanocomposite maximum adsorption capacity was  $0.4 \text{ g.g}^{-1}$  for 2 wt% of GO in the composite mixture.

Manuscript for publication in 'Applied Physics A'

Dear editor

Enclosed please find the original article entitled:

**Kinetic, isotherm and thermodynamic studies on Cr(VI) adsorption using cellulose acetate/graphene oxide composite nanofibers**

In the present study, cellulose acetate/graphene oxide (CA/GO) composite nanofibers were prepared via electrospinning method to remove Cr(VI) ions from aqueous solution via adsorption process in a batch mode. The impact of adsorption parameters, including contact time, pH, Cr(VI) concentration and temperature was investigated to obtain the optimum conditions for the uptake of maximum Cr(VI) ions from water within a short time. Hereby, I confirm that this manuscript is the original work and it has been neither published nor submitted simultaneously elsewhere. We believe that our recent findings may be interesting to the journal. Thank you for receiving our manuscript and considering it for review. We appreciate your time and look forward to your response. We wish you, reviewers and all people from publishing services to be healthy and optimistic. We hope that difficult time for everyone because of covid-19 will be very short, and we will soon return to normal life.

We will be grateful if you would kindly consider this paper for publication in 'Applied Physics A'

I hope this will meet with your considerations.

Sincerely yours.

Indah Raya

Departement of Chemistry

Faculty Mathematics and Natural Science

Hasanuddin University

Makassar

South Sulawesi

90245 Indonesia

## Author's Response To Reviewer Comments

Close

Dear editor,

I would like thank you for your uninterrupted and praiseworthy concern. I am also so grateful for the referee attention on this work, being appreciative to his/her precious suggestions and reminders, which helped us to improve the manuscript. The comment was carefully considered, and the detailed point-by-point response to it is listed below, and also highlighted in the main text.

Comments for APYA-D-21-02809R1:

The authors have carefully revised the whole manuscript and the quality of this work has been greatly improved. However, there is still a minor comment that the authors should take into consideration before publication:

The authors responded that: "As observed in SEM images, the fiber diameter was increased by loading GO into the nanofibers up to 2 wt.% which resulted in increasing fiber diameter. By increasing fiber diameter, the voids between fibers were increased which could be resulted in increasing porosity, specific surface area and adsorption capacity." However, to the extent of our knowledge, the specific surface area will decrease as the diameter of nanofiber increase. [Ref: 10.1016/j.apmate.2021.09.004] In this work, the interruption of GO can reasonably lead to the increase in porosity, specific surface area and adsorption capacity, because of the inherently large surface area and abundant active adsorption sites in the carbon nanomaterials.

As observed in SEM images, the fiber diameter was increased by loading GO into the nanofibers up to 2 wt.% which resulted in increasing fiber diameter. By increasing fiber diameter, the voids between fibers were increased which could be resulted in increasing porosity, specific surface area and adsorption capacity." is removed from manuscript and the following text is modified as follows:

By loading GO into the nanofibers up to 2 wt.%, the increase in the adsorption capacity of Cr(VI) ions by the composite nanofibers could be attributed to the high porosity, large specific area and higher available active sites of GO for Cr(VI) ions sorption.

We hope the responses and corrections made in the revised manuscript meet with the reviewer's considerations.

Sincerely yours  
Indah Raya  
Hasanuddin University  
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Close

**Date:** 10 Jan 2022  
**To:** "Indah Raya" r.indah2021@gmail.com  
**From:** "Alexandra Papavlu" alexandrapalla@yahoo.co.uk  
**Subject:** APYA: Your manuscript entitled Kinetic, isotherm and thermodynamic studies on Cr(VI) adsorption using cellulose acetate/graphene oxide composite nanofibers

Ref.:  
Ms. No. APYA-D-21-02809R2  
Kinetic, isotherm and thermodynamic studies on Cr(VI) adsorption using cellulose acetate/graphene oxide composite nanofibers  
Applied Physics A

Dear Dr. Raya,

I am pleased to tell you that your work has now been accepted for publication in Applied Physics A.

Thank you for submitting your work to this journal.

With kind regards

Alexandra Papavlu, PhD  
Member of the Board of Editors  
Applied Physics A

\*\*\*\*\*

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