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RESORCINOL-FORMALDEHYDE CARBON GELS ADSORPTION: A COMMENTARY

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Abstract: This work is aimed at highlighting the recent progress of resorcinol-formaldehyde carbon gels adsorption of water pollutants. The synthesis strategies of the carbon gels were discussed to shed some light on the development of mesoporous matrix of carbon gel via the agglomeration of colloidal particles. The surface area of adsorbent can reach as high as 3000 m^2/g by CO_2 activation, while the surface functionalities are introduced through modification techniques for improving the removal performance. However, most of the recent studies are inclined at batch mode of adsorption with lack of information on the scale-up of the process in continuous mode. Carbon gel is a special class of porous material that can be moulded into desired size, hence a promising adsorbent candidate for monoliths and packings in column adsorption. Therefore, more dedicated works should be established to materialize the applications of carbon gel in column adsorption, particularly at industrial scale.

Keywords**:** Adsorbent; Adsorption; Resorcinol-formaldehyde carbon gel; Water pollutant

Introduction

The issues related to the release of pollutants into the water bodies have become a subject of considerable concern. The presence of water

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contaminants such as dyes, heavy metals and organic compounds brings about damaging effects not only to the aquatic ecosystem but also to human who relies on water resources for routine life.[1] Hence, the polluted effluents must be treated prior entering the streams. Amongst the available methods of water and wastewater treatment, adsorption has appeared to be the mostly applied because it is simple, economical and efficient. Adsorption is a process in which the adsorbate (contaminant) is adhered to the solid adsorbent through dipole-π interaction, π-π interaction, ion exchange, pore filling and complexation.[2]

Adsorption can be operated through batch or continuous mode. For a massive volume of wastewater, the continuous mode is more practical because less space is needed for the process at minimum manufacturing cost. However, the commercially available adsorbents are generally in the powder and granular forms, making them unfit for continuous process. Despite of large specific surface area and rich surface chemistry for satisfactory removal performance in batch mode, the column operation may suffer in escalating pressure drop as a result of hydraulic resistance if adsorbent with tiny particles is employed.[3] Consequently, the column performance decreases at undoubtedly high operating cost.

A potential solution for this problem is a packing-type adsorbent, known as carbon gel. Carbon gel is normally synthesized through polymerization of resorcinol and formaldehyde in the presence of solvent (water) and basic catalyst (sodium carbonate).[4] Also, carbon gel structurally doped with aromatic S or N can be synthesized through co-polycondensation of resorcinol with heterocyclic aldehyde.[5,6] The mesoporous matrix of carbon gel is established via the aggregation of nanometer-sized colloidal particles. The formation of gaps between the nanoparticles develops a hierarchical pore system of mainly mesopores, and some micropores and macropores.[7] Carbon gel is a porous material with large carbon density that can be moulded into desired size and customized for definitive utilizations as catalysts for chemical synthesis, adsorbents for chemical separation and environmental protection, and carbon electrodes for energy storage.[7-9] In the adsorption field, carbon gel is a promising candidate to overcome the resistance of flow and provide a short path for efficient connection between the adsorbate and solid surface in continuous system.[3] Nevertheless, the application of carbon gel as adsorbent for the removal of water pollutants especially in continuous mode is not widely available in literature. This commentary is aimed to highlight the issues revolving around synthesis strategies and adsorption properties of carbon gels, and the limitations and recommendations for future directions.

Synthesis strategies of resorcinol-formaldehyde carbon gels

In general, there are three steps during the preparation process to control the structure of carbon gel. The first step is the solution-sol transition, in which nanoscale sol particles are created automatically or catalyst mediated through hydrolysis and condensation reactions. The second step is the sol-gel transition, in which the sol particles are cross-linked and amass into wet gel with consistent network called hydrogel.[10] The final step is the drying phase that is imperative for the formation of porous gel. There are three drying methods adequate to convert the hydrogel to solid resorcinol-formaldehyde gel. The identity of resorcinol-formaldehyde gel depends on the drying method. Xerogel is a dried dense polymer obtained through drying in a dormant atmosphere or conventional oven.[11,12] In freeze-drying, the solvent within the voids is frozen and evacuated by sublimation to evade the formation of vapour-liquid interface in a polymer called cryogel. The aerogel is formed when the liquid solvent is removed by supercritical drying with CO_2 or organic solvent.[12] Aerogel normally has a high pore volume with wide texture range. Nevertheless, the pore structure giving rise to the surface area of carbon gel depends not only to the drying methods alone but also the interplay between the synthesis conditions and carbonization techniques.[13]

Figure 1 shows the formation of resorcinol-formaldehyde molecule with the present of basic catalyst. Resorcinol is a trifunctional benzyl compound with two hydroxyl groups in positions 1 and 3, which allows formaldehyde to be added in positions 2, 4 and 6. The condensation reaction takes place when the hydroxymethyl derivatives loses OH^- groups, forming a benzyl-type cation that reacts with other benzene rings, resulting the methylene and ether bonds to form polymeric particles. The primary particles aggregate and cross-link with each other via polymer chains, forming a three-dimensional porous network in a liquid medium.[14]

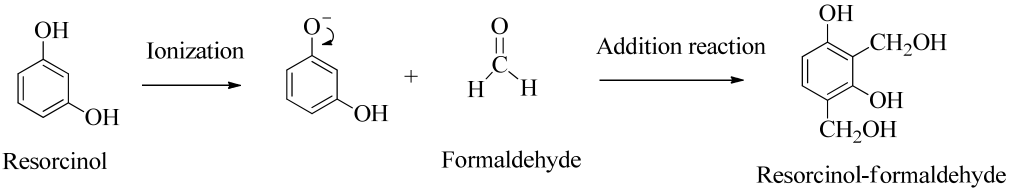

Figure 1. Reaction of resorcinol-formaldehyde.[14]

Generally, the preparation process of carbon gel consists of mixing, gelation, aging, solvent exchange, drying, carbonization and activation.[8,15] The mixing of resorcinol and formaldehyde in a solvent at an appropriate molar ratio in the presence of a basic catalyst such as sodium carbonate (Na_2CO_3) is to initiate the gelation process. The gel is formed due to the crosslinking intensity between the bonds. It is a crucial step that determines the structural characteristics of carbon gel. Gelation is a pH-dependent process, that could be driven by particle nucleation in acidic condition or by addition reaction in basic condition. The solution pH in the sol-gel process is adjusted by controlling the amount of basic catalyst in water.[16] Figure 2 shows the particle growth mechanisms at different pH conditions.

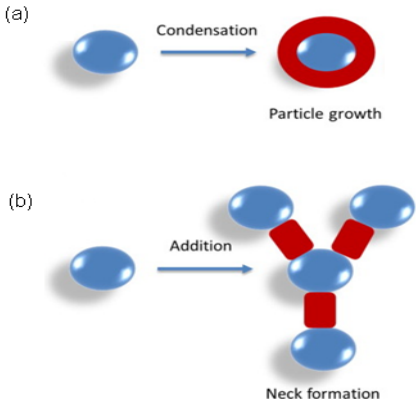

Figure 2. Particle growth mechanisms, (a) condensation reaction at acidic condition, and (b) addition reaction at basic condition.[16]

The resultant hydrogel is then incubated in a controlled environment for a sufficient period of time at 60°C to 80°C for curing to strengthen the bonds through particles aggregation. Next, the material is subjected to solvent exchange for extracting the residual water from the sample interiors. After that, it is carbonized at temperatures between 300°C to 2500°C to eliminate the remaining oxides and hydrogen groups, from which the organic gel (hydrogel) is transformed into carbon gel with highly porous network. This step also enlarges the specific surface area of carbon gel by increasing the number of mesopores and micropores. As the carbonization temperature increases, the carbonization process modifies the porous structure of the gel and creates more pores in the structure by releasing volatile matters. In addition, the increment of carbonization temperature also increases the total surface area and total pore volume of the carbon gel. However, at the temperature of 1000 °C and above the volume of pores in carbon gel decreases due to the collapse of the porous structure.[17] A further oxidation process is introduced thereafter to functionalize the carbon gel with oxygenated surface groups. The physical oxidation in air forms phenolic and carbonyl groups, while chemical oxidation with nitric acid establishes carboxylic group on the carbon surface.[18] The physical oxidation is usually held at a higher temperature than chemical oxidation, but the former does not require a washing post-treatment.[19] Figure 3 illustrates the synthesis of carbon gel.

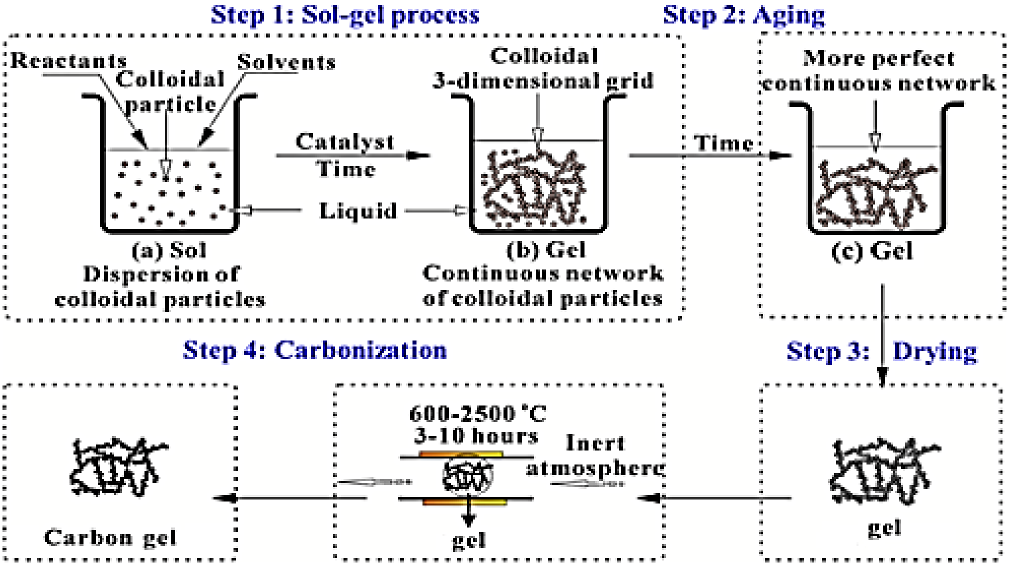

Figure 3. Steps in the production of carbon gel.[20]

The configurations of carbon gel can be classified into pearl-necklace and fibril. Resorcinol formaldehyde carbon gel normally bears a pearl-necklace configuration of poor mechanical character due to the small contact area between particles.[16] In general, the ratio between the catalyst and solvent (water) is controlled to render a strong mechanical structure of carbon gel. At a high water content, the water molecules weaken the bonds between particles, making the hydrogel more susceptible to collapse into tiny particles, particularly at the later stage of synthesis. A strong mechanical structure of carbon gel is therefore imperative for successive cycles of separation and regeneration in continuous mode of adsorption.

Table 1 summarizes the synthesis strategies and adsorptive properties of carbon gels for water pollutants removal. In general, carbon gels are highly mesoporous with surface area in the range of 300 m^2/g to 3000 m^2/g. The structural properties of carbon gels are enhanced through physical activation using CO_2 and steam, and chemical activation using KOH. In addition, the surface properties of carbon gels could also be improved for high adsorption affinity of water pollutants by incorporating titanium tetraisopropoxide and CeO_2, and oxygen functional groups through HNO_3 oxidation. These physicochemical attributes endow them with excellent removal of various dyes and heavy metals, as displayed in Table 1. Carbon gel is a potential adsorbent for water pollutants removal because of its unique textural characteristics that can be tailored by controlling the synthesis strategies. The material can be moulded in the form of monolith and packing to suit column adsorption.[7,9]

Table 1. Synthesis strategies of carbon gels for water pollutants removal[12]

(R: resorcinol, F: formaldehyde, TTP: Titanium tetraisopropoxide, W: solvent, C: catalyst, R/F & R/C in molar ratio, R/W in g/cm^3, r.t: room temperature, *T*: Temperature in °C, *t*: time in h, Yield in %, S_{BET}: BET surface area in m^2/g, R_{Meso}: mesopore content, d_p: average pore diameter in nm; Q_m: maximum adsorption capacity in mg/g)

Reactant	**R/F**	**W**	**R/W**	**C**	**R/C**	**pH**	**Carbonization**			**Activation**			S_{BET}	R$_{Meso}$ (%)	d_p	**Adsorbate**	Q_m
							T	***t***	**Yield**	**Agent**	***T***	***t***					
Xerogels																	
R: Furfural, Hexa-methylene-tetramine	2, 33.3	-		KOH	-	7.0	900	3	-	-			887	28.8	30.9	Methylene Blue	240
																Acid Blue 40	222
																Reactive Black 5	35
						8.1							944	39.7	26.3	Methylene Blue	249
																Acid Blue 40	246
																Reactive Black 5	25
						8.8							898	29.9	23.0	Methylene Blue	238
																Acid Blue 40	223
																Reactive Black 5	20
						9.9							600	5.96	12.7	Methylene Blue	199
																Acid Blue 40	115
																Reactive Black 5	10

R:F	-	-		-		6.5	800	6	-	-			617	100	31.3	Ciprofloxacin	112
R:F	0.5	Water	35	Na_2CO_3	1000	-	900	2	-	-			630	78.5	28-40	Methylene Blue	212
										CO_2	900	4	1015	75.3	29-40		438
												6	1365	71.5	28-43		532
												8	2180	65.0	29-41		714
R:F	0.5	Water	0.2	Na_2CO_3	500	-	750	2	-	-			78	-	-	-	-
R:F	0.5	Water	0.1	Na_2CO_3	100	-	-			-			489	79.8	2.84	-	
					200								522	94.0	4.32		
					500								310	96.6	15.7		
					600								217	96.3	25.6		
					700								175	98.3	29.9		
					800								127	98.4	31.3		
			0.05		200								376	95.7	3.94		
			0.1										454	96.6	5.20		
			0.15										465	95.7	7.92		
R:F	0.5	Water	-	Malonic acid	1	-	850	3	-	-			1201	26.3	1.91	Cu^{2+}	260
										HNO_3	50	10	1150	39.7	1.98		299
R:F	0.5	Water	0.5	Na_2CO_3	50	-	1000	4	-	-			190	35.5	2.40	-	
										CO_2	1000	0.6	460	34.6	2.40		

												1.05	230	37.5	2.40		
					100					-			640	64.3	3.80		
										CO_2	1000	0.65	1400	47.5	3.30		
												1.2	1900	44.0	2.40		
					200					-			700	84.7	9.30		
										CO_2	1000	0.85	1900	76.5	9.30		
												2	2421	76.1	7.10		
					1000					-			630	83.3	44.0		
										CO_2	1000	0.75	1700	76.2	69.0		
												2	2965	85.0	44.0		
R:F	0.5	Water	0.3	KOH	150	-	600	-	-	-			684	73.9	-	Ni^{2+}	0.415
																Cu^{2+}	0.585
																Zn^{2+}	0.428
										Steam	850	-	1870	66.9		Ni^{2+}	0.508
																Cu^{2+}	0.352
																Zn^{2+}	0.232
										KOH	600	-	276	23.1		Ni^{2+}	0.453
																Cu^{2+}	0.574
																Zn^{2+}	0.578

					400					-			741	86.1		Ni^{2+}	0.044
																Cu^{2+}	0.209
																Zn^{2+}	0.084
										KOH	600	-	556	81.4		Ni^{2+}	0.553
																Cu^{2+}	0.601
																Zn^{2+}	0.537
R:F	0.5	Water	0.25	Na_2CO_3	100	4	-			-			189	0	3.80	-	
					150	5							248	0	3.10		
					200	6							277	0	2.50		
					250	7							202	0	2.80		
R:F	0.5	-		Na_2CO_3	-	-	800	12	-	-			606	83.8	9.79	Pb^{2+}	64.2
R:F, Melamine	0.5, 5.2				100								531	64.4	3.41		74.5
					150								573	82.6	8.02		83.8
					200								530	58.0	3.75		71.7
	0.5, 10.4				150								560	71.9	4.21		69.9
	0.5, 6.9												609	78.5	6.38		73.2
R:F	0.5	Water	0.3	NaOH, HNO_3	-	3	750	2	46.6	-			484	8.80	9.0	Methylene Blue	102
						5			49.6				498	52.7	8.0		108
						6			51.8				513	19.8	9.0		123

						6.5			66				622	42.7	5.9		128
						7.2			54.2				412	15.7	6.4		91
						6.5	600	2	72				308	94.9	8.0		90
						6.5	700	2	68.6				449	86.2	9.7		93
						6.5	800	2	60				433	6.40	9.0		113
						6.5	750	2	56				541	12.0	6.2		-
R:F	0.5	Water	0.3	Na_2CO_3	1000	6	750	3	-	-			444	-	3.61	Pb^{2+}	17.0
																Cu^{2+}	6.64
																Co^{2+}	5.46
																Ni^{2+}	4.16
R:F	0.5	Water	0.5	Na_2CO_3	100	-	600	3	42.8	-			476	23.7	1.86	Reactive Orange 16	0
										HNO_3	60	5	301	0	1.33		0
					200				49.6	-			608	46.3	2.36		2.73
										HNO_3	60	5	567	44.9	2.33		0
					1000				42.6	-			639	69.8	6.89		6.80
										HNO_3	60	5	712	75.1	7.68		6.52
					2000				53.7	-			631	59.2	3.56		6.46
										HNO_3	60	5	586	59.3	3.57		5.84
R:F	0.5	Water	0.5	Na_2CO_3	1000	-	600	3	42.6	-			639	69.8	6.89	Congo Red	2.25

										HNO_3	60	5	712	75.1	7.68		10.6
					2000				53.7	-			631	59.2	3.56		2.51
										HNO_3	60	5	586	59.3	3.57		4.36
Aerogels																	
R:F	0.5	Water	1.5	Na_2CO_3	500	-	1000	1	-	-			420	-	6	-	
			0.068										630		139		
			0.016										717		563		
	0.5		0.061		50								618		14		
					300								616		51		
					800								710		193		
R: Furfural, Hexa-methylene-tetramine	0.5, 30	Ethanol	339	acetic acid	-	5.1	900	6	-	CO_2	1000	1	803	71.3	0.65	Benzene	242
																Hydroquinone	32.4
																Catechol	27.5
																Resorcinol	21.5
R:F	0.5	Water	0.2	Na_2CO_3	500	-	750	2	-	-			570	-	-	Cd^{2+}	11.5
																Cu^{2+}	8
R:F	0.5	Water	0.5	Na_2CO_3	5	6	-			-			846	19.0	-	-	
R:F, TTP	0.5, 1												223	27.7			

R:F	0.5						900	1	38				1990	17.1			
R:F, TTP	0.5, 1								50				718	32.5			
R:F	0.5								38	TTP	r.t	24	301	7.04			
R:F	0.5	Water	0.3	KOH	50	-	600	-	-	-			623	72.3	-	Ni^{2+}	0.048
																Cu^{2+}	0.494
																Zn^{2+}	0.332
										KOH	600	-	830	37.2		Ni^{2+}	0.256
																Cu^{2+}	0.427
																Zn^{2+}	0.25
Cryogels																	
R:F	0.5	Water	0.2	Na_2CO_3	100	-	1000	4	-				500				
R:F	0.5	Water	20	Na_2CO_3	100	-	800	-	-	CeO_2	r.t	0.25	614	-	14	As^{3+}	4.34

Carbon gels adsorption

Adsorption has been accepted as a preferable method of pollutants treatment. Its performance relies on the physicochemical characteristics of adsorbent, chemical properties of adsorbate, and operating conditions such as concentration, contact time, temperature, pH, ionic strength and the presence of foreign elements. The adsorption of water pollutants is synergistically driven by porous texture and surface functionalities of adsorbent. At low concentration, the interactions may not have sufficient driving force to achieve equilibrium. Similarly, the equilibrium may be slow if the transport process is rate-determining at high concentration. The concentration of solute on the surface of solid continues to increase until the solute in the solution remains in equilibrium with that at the surface.

Table 2 summarizes the operating conditions of carbon gels adsorption in batch mode. In general, the concentration of adsorbate provides the driving force to overcome the resistance of mass transfer at adsorbent phase, from which the adsorption increases with increasing concentration.[21-22] Likewise, a longer time would be needed to achieve the equilibrium as the concentration increases.[21-22] The post-treatment using HNO_3 increases the mesoporosity of carbon gel at the expense of slight decrease in surface area.[18,21,23] Xiao and co-workers[21] reported an increase of Cu(II) removal by HNO_3-oxidized carbon gel as a result of complexation and ion-exchange interactions with surface acidic oxygen groups. For anionic dyes removal by HNO_3-modified carbon gel, Tang & Zaini[22] reported an increase of Congo Red removal, while Zhi & Zaini[23] reported a decrease of Reactive Orange removal. A variation in the removal pattern could be attributed by the interplay between the accumulation of protons (surface charge) and pore restriction for the transport of dyes molecules.

The restricted movement of adsorbate molecules into the adsorbent pore channels could also be associated with a longer contact time to reach the equilibrium. Tang & Zaini[22] reported a 4200 min of equilibrium for 10 mg/L of Congo Red by HNO_3-oxidized carbon gel, while Zhi & Zaini[23] showed a 9000 min of equilibrium for 10 mg/L of Reactive Orange by un-modified carbon gel. In a related work, the restriction for adsorption as a result of the aggregation of colloidal particles within the mesoporous matrix of carbon gel is also evident from the CO_2 activation of carbon gel. Despite the fact that its surface area elevates from 630 m^2/g to 2180 m^2/g upon activation, the removal of Methylene Blue remains unchanged at about 4.4 mg/g. Nevertheless, the time taken to attain the equilibrium (Methylene Blue concentration: 3.7 mg/L, dosage: 1 g/L) has slightly improved from more than 500 min to 410 min.[24]

Table 2. Adsorption properties of carbon gels.

Adsorbent	**Surface area (m^2/g)**	**Mesoporosity (%)**	**Dosage (g/L)**	**Adsorbate**	**Concentration (mg/L)**	**Equilibrium time (min)**	**Solution pH**	**Equilibrium adsorption (mg/g)**	**Rate constant (g/mg.min)**	**Thermodynamics**
HNO_3-oxidized carbon gel[22]	711	75.1	1.5	Congo Red	5	1800	5.4	3.19	3.0×10^{-3}	-
					10	4200		4.73	6.32×10^{-3}	
Carbon gel[21]	1201	26.3	0.5	Cu(II)	50	20	5.0	53.6	5.1×10^{-3}	Endothermic
					200	60		246	1.2×10^{-3}	
HNO_3-oxidized carbon gel[21]	1150	39.7			50	20		56.1	5.2×10^{-3}	
					200	60		262	1.0×10^{-3}	
N-doped carbon gel[26]	573	82.6	1	Pb(II)	50	180	4.5	37.6	3.55×10^{-3}	Endothermic
HNO_3-modified carbon gel[18]	418	76.8	2.5	Methylene Blue	10	3600	Natural	4.41	-	-
Carbon gel[27]	444	-	2	Pb(II)	25	220	3.0	9.20	-	-
				Ni(II)		195		4.12		
Expanded graphite carbon gel[27]	547	-		Pb(II)		250		11.0		
				Ni(II)		200		5.03		
Carbon gel[12,23]	631	59.2	1.5	Reactive Orange	10	9000	Natural	6.88	6.67×10^{-5}	-

HNO_3-oxidized carbon gel[12,23]	586	59.3				4470		2.64	1.33×10^{-4}	
Carbon gel[24]	630	78.5	1	Methylene Blue	3.7	>500	6.5	4.99	7.0×10^{-4}	-
CO_2-activated carbon gel[24]	2180	65				410		4.34	3.0×10^{-3}	

The pseudo-second-order kinetics model has been commonly used to describe the carbon gels adsorption rate of water pollutants. The model equation is expressed as, $q_t = q_e^2k_2t/(1+q_ek_2t)$, where q_t is the adsorption capacity at time, t, q_e is the adsorption at equilibrium, k_2 is the pseudo-second-order rate constant, and t is the contact time. The model indicates that the transport of adsorbate molecules is driven by the external diffusion at low concentration due to strong adsorbent-phase mass transfer resistance, and chemical adsorption through the formation of imaginary bonds due to electrostatic and dispersive forces interactions at high concentration.[12,24] In general, a high concentration subsides the solid-phase mass transfer resistance for more adsorbate-adsorbent interactions and adsorption. Yet, the vacant active sites progressively decrease with time, hence decreasing the rate of adsorption. The rate constant generally decreases with increasing concentration because of the intensified collision of dye molecules.[21] On the other hand, a greater rate constant with increasing concentration is attributed to a stronger driving force to overcome mass transfer resistance.[22] According to Arrhenius theory, a weak activation energy endows a high rate constant that is associated with a rapid adsorption due to less repulsion of molecules and mutual interactions between the adsorbate and adsorbent surface characteristics.[22,24]

To date, the continuous mode of adsorption using carbon gel for water pollutants removal is still limited in much of published literature. In a related work, Wu and co-workers[25] studied the adsorptive properties of carbon gel (583 m^2/g, 87.4% mesoporosity) and CO_2-activated carbon gel (852 m^2/g, 81.5% mesoporosity) for the removal of Reactive Red 2 using bench scale continuous adsorption. The carbon gel adsorbent with an average particle size of 0.285 mm is introduced inside a column with dimensions of 30 cm length and 1.1 cm internal diameter. Despite a promising adsorption of about 700 mg/g of Reactive Red 2, the column operation may be compromised at large scale due to the use of very slow flow rates (0.6 mL/min – 1.2 mL/min) and small particle size of adsorbent, which are not representative to the real applications. Furthermore, the minute size of adsorbent in column may trigger the resistance to flow which giving rise to high pressure drop. Therefore, systematic studies to evaluate the effectiveness of carbon gel in the form of monoliths or packings in continuous mode of adsorption for industrial scale applications should be established as the directions for future research.

Concluding remark and future outlook

Resorcinol-formaldehyde carbon gel is a promising class of adsorbent for water pollutants removal. The inherent mesoporous matrix of carbon gel endows the hierarchical pathways to accommodate adsorbate molecules for effective adsorption process. Also, the adsorptive properties can be improved through activation and surface modification strategies. However, the earlier works are mainly emphasized on batch mode of adsorption without proper insights into continuous mode of adsorption. In addition, the adsorption performance of carbon gels in some studies is not satisfactory due to very long contact time necessary to achieve equilibrium. The continuous adsorption is a non-equilibrium mass transfer process that is dynamic in nature until the bed saturation limit is attained. The adsorption performance via column mode relies upon influent concentration and flow rate, and adsorbent bed height. At industrial scale, the application of column adsorption is preferred to treat large volume of wastewater. Yet, the column operability and performance may be compromised due the fact that most commercial adsorbents are in the form of small particles, leading to long diffusion path, high resistance to flow, high pressure drop and high operating costs. Carbon gel is a potential adsorbent candidate for column adsorption because it could be moulded as monoliths and packings to overcome the aforementioned hurdles. It is inspired by the promising physicochemical attributes of carbon gel with high surface area rich surface chemistry that have been proven to offer considerable removal performance of water pollutants like Methylene Blue, Congo Red, Methyl Orange, Reactive Black 5, Acid Blue 40, residual antibiotics and toxic heavy metals in batch mode.

Nevertheless, dedicated studies for carbon gels adsorption in dynamic mode are still scarce in literature. The adsorption properties from batch operation such as maximum capacity, rate constant and equilibrium time should therefore be established and integrated for systematic evaluation of column operation at pilot scale. Also, the mass production of carbon gel and the associated manufacturing cost should also be taken into account to ensure the sustainability of the process in a long run.

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References

1. Zaini, M. A. A.; Cher, T. Y.; Zakaria, M.; Kamaruddin, M. J.; Setapar, S. H. M.; Yunus, M. A. C. Palm oil mil effluent sludge ash as adsorbent for methylene blue dye removal. *Desalin. Water Treat.* **2014**, *52*(*19-21*), 3654-3662.
2. Ming-Twang, S.; Lin-Zhi, L.; Zaini, M. A. A.; Zhi-Yong, Q.; Pei-Yee, A. Y. Activated carbon for dyes adsorption in aqueous solution. In

Advances in Environmental Research, J.A. Daniels, Ed.; Nova Science Publishers: New York, 2015, *36*; pp 217-234.

3. Masuda, T.; Ogino, I.; Mukai, S. R. Immobilization of magnesium ammonium phosphate crystals within microchannels for efficient ammonia removal. *Water Sci. Technol.* **2013**, *67(2)*, 359-365.

4. Rojas-Cervantes, M. L. Some strategies to lower the production cost of carbon gels. *J. Mater. Sci.* **2014**, *50(3)*, 1017-1040.

5. Kicinski, W.; Norek, M.; Dziura, A. ; Polanski, M. Copolycondensation of heterocyclic aldehydes: A general approach to sulfur and nitrogen dually-doped carbon gels. *Micropor. Mesopor. Mat.* **2016**, *225*, 198-209.

6. Strachowski, P.; Fronczak, M.; Olechno, E.; Kowalik, M.; Kicinski, W.; Kaszuwarac, W.; Bystrzejewski, M. Magnetic organic xerogels: efficient adsorbents for the removal of heavy metal ions from aqueous solutions. *New J. Chem.* **2018**, *42*, 7073-7082.

7. Mukai S. R. Controlling the morphology of carbon gels. *Bol. Grupo Espanol Carbon.* **2012**, *26*, 8-11.

8. Al-Muhtaseb, S. A.; Ritter, J. A. Preparation and properties of resorcinol-formaldehyde organic and carbon gels. *Adv. Mater.* **2003**, *15(2)*, 101-114.

9. Tsuchiya, T.; Mori, T.; Iwamura, S.; Ogino, I.; Mukai, S. R. Binder free synthesis of high-surface-area carbon electrodes via CO_2 activation of resorcinol–formaldehyde carbon xerogel disks: Analysis of activation process. *Carbon* **2014**, *76*, 240-249.

10. Du, A. ; Zhou, B. ; Zhang, Z.; Shen, J. A special material or a new state of matter: A review and reconsideration of the aerogel. *Materials* **2013**, *6(3)*, 941-968

11. Czakkel, O.; Marthi, K.; Geissler, E.; László, K. Influence of drying on the morphology of resorcinol–formaldehyde-based carbon gels. *Micropor. Mesopor. Mater.* **2005**, *86(1-3)*, 124-133.

12. Zhi, L. L. ; Hui, T. S.; Zaini, M. A. A. Carbon gel-based adsorbents for anionic dyes removal. In *Activated Carbon Prepared From Various Precursors*, H.S. Min, Ed.; Ideal International E – Publication Pvt. Ltd, Indore, 2017 ; pp. 62-84.

13. Zhang, R.; Li, W.; Liang, X.; Wu, G.; Lu, Y.; Zhan, L.; Lu, C.; Ling, L. Effect of hydrophobic group in polymer matrix on porosity of organic and carbon aerogels from sol–gel polymerization of phenolic resole and methylolated melamine. *Micropor. Mesopor. Mater.* **2003**, *62(1-2),*17-27.

14. Rey-Raap, N.; Arenillas, A.; Menéndez, J. A. Carbon gels and their applications: A review of patents. *Submicron Porous Mater.* **2017**, 25-52.

15. Pekala, R. W. Organic aerogels from the polycondensation of resorcinol with formaldehyde. *J. Mater. Sci.* **1989**, *24(9),* 3221-3227.

16. Alshrah, M.; Tran, M.; Gong, P.; Naguib, H. E.; Park, C. B. Development of high-porosity resorcinol formaldehyde aerogels with enhanced mechanical properties through improved particle necking under CO_2 supercritical conditions. *J. Colloid Interface Sci.* **2017**, *485*, 65-74.

17. Mirzaeian, M.; Hall, P. J. The control of porosity at nano scale in resorcinol formaldehyde carbon aerogels. *J. Mater. Sci.* **2009**, *44*(*10*), 2705-2713.

18. Zaini, M. A. A.; Yoshida, S.; Mori, T. ; Mukai, S.R. Preliminary evaluation of resorcinol-formaldehyde carbon gels for water pollutants removal. *Acta Chim. Slovaca.* **2017**, *10(1)*, 54-60.

19. Elkhatat, A. M.; Al-Muhtaseb, S. A. Advances in tailoring resorcinol-formaldehyde organic and carbon gels. *Adv. Mater*. **2011**, *23*, 2887-2903.

20. Zuo, L.; Zhang, Y.; Zhang, L.; Miao, Y.; Fan, W.; Liu, T. Polymer/carbon-based hybrid aerogels: preparation, properties and applications. *Materials* **2015**, *8(10)*, 6806-6848.

21. Xiao, Z.; Peng, F.; Li, X.; Zhang, R.; He, W.; Zhou, T. Surface modification of malonic acid-catalyzed carbon xerogels and their high performance for adsorption of Cu (II) ions. *Surf. Interface Anal.* **2013**, *45*, 1869-1877.

22. Tang, S. H.; Zaini, M. A. A. Congo red removal by HNO_3-modified resorcinol-formaldehyde carbon gels. *Chem. Eng. Trans*. **2017**, *56*, 835-840.

23. Zhi, L. L.; Zaini, M. A. A. Equilibrium and kinetic adsorption studies of reactive orange onto resorcinol-formaldehyde carbon gels. *Chem. Eng. Trans*. **2017**, *56*, 811-816.

24. Paez, C. A.; Contreras, M. S.; Léonard, A.; Blacher, S.; Olivera-Fuentes, C. G.; Pirard, J.; Job, N. Effect of CO_2 activation of carbon xerogels on the adsorption of methylene blue. *Adsorption* **2012**, *18*, 199-211.

25. Wu, X.; Wu, D.; Fu, R.; Zeng, W. Preparation of carbon aerogels with different pore structures and their fixed bed adsorption properties for dye removal. *Dyes Pigments* **2012**, *95(3)*, 689-694.

26. Yang, B.; Yu, C.; Yu, Q.; Zhang, X.; Li, Z. ; Lei, L. N-doped carbon xerogels as adsorbents for the removal of heavy metal ions from aqueous solution. *RSC Adv.* **2015**, *5*, 7182-7191.

27. Osinska, M. Removal of lead(II), copper(II), cobalt(II) and nickel(II) ions from aqueous solutions using carbon gels. *J. Sol-Gel Sci. Technol.* **2017**, *81*, 678-692.