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Proceedings of the 6th International Conference on Fundamental and Applied Sciences

ICFAS 2020

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The 6th International Conference on Fundamental and Applied Sciences (ICFAS 2020), 13–15 July 2021

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Preface

The Fundamental and Applied Sciences Department, Universiti Teknologi PETRONAS, is honoured to host the Virtual Event of 6th International Conference on Fundamental and Applied Sciences (ICFAS 2020) from 13 to 15 July 2021. It is our pleasure to welcome delegates from Malaysia, Indonesia, Kazakhstan, Nigeria, Vietnam, Pakistan, Ghana, New Zealand and Brunei to share and exchange ideas on the latest advancement in mathematics, chemistry and physics with the theme of "Innovative Science towards Sustainability and Industrial Revolution 4.0". This conference has certainly provided a platform for sharing and discussion on recent advances in sustainable chemistry, nano and sustainable technology, and mathematical and analytical methods in science and technology. The conference topics include green materials, molecular modelling, catalysis, nanodevices and nanosystems, smart materials applications, solar cell technology, computational mathematics, data analysis and visualization, but not limited to numerical analysis. It is our hope that the contents of this book will benefit researchers, postgraduate students and industrial practitioners in the areas of mathematics, physics and chemistry as most of the topics are in line with IR 4.0. On behalf of the sponsors and the Conference Committees, we would like to gratefully acknowledge all the keynote and invited speakers, presenters and participants in contributing to the success of ICFAS 2020. Special appreciation and thank you to all the reviewers for providing their expertise in improving the quality of the papers, without which this book will not be possible. Finally, thank you to Springer for publishing our conference in Springer Proceedings in Complexity.

Seri Iskandar, Malaysia

Samsul Ariffin Abdul Karim
Mohd Fadhlullah Abd Shukur
Chong Fai Kait
Hassan Soleimani
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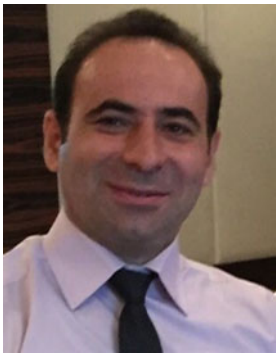
Chong Fai Kait is Associate Professor at Universiti Teknologi PETRONAS. She graduated with B.Sc. (Hons) in Industrial Chemistry from Universiti Putra Malaysia in 1993. In 1998, she received her Ph.D. from University of Dundee, Scotland, in the area of heterogeneous catalysis for naphtha reforming process. She has more than 20 years of academic and research experience, and her main research interests are in photocatalysis and its integration with ionic liquids or other techniques for sustainable environmental applications.



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UiO-66 and ZIF-8 Metal-organic Frameworks for Acenaphthene Adsorption

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UiO-66 and ZIF-8 Metal-organic Frameworks for Acenaphthene Adsorption

Zakariyya Uba Zango, Anita Ramli, Khairulazhar Jumbri, and Muslim Abdurrahman

Abstract Comparative adsorption of acenaphthene onto UiO-66 and ZIF-8 metal-organic frameworks (MOFs) were investigated. Response surface methodology (RSM) was employed for the process optimizations according to the central composite design (CCD) with 5 inputs variables. The adsorption efficiency achieved were 99.7 and 60.7% for the UiO-66 and ZIF-8, respectively according to the RSM optimization conditions. The model was significantly described according to statistical analysis of variance (ANOVA). The adsorption process was well fitted by pseudo-second order kinetic model with R^2 values of 0.9947 and 0.97780 for the UiO-66 and ZIF-8, respectively.

Keywords Acenaphthene · Adsorption · Metal-organic frameworks · Response surface methodology

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1 Introduction

Pollution caused by the presence of organic contaminants in environmental waters has been a disturbing phenomenon affecting the peaceful existence of living organisms. Acenaphthene, a member of Polycyclic aromatic carbons (PAHs) heavily found in crude oil and coal [1, 2]. It consisted of naphthalene connected by the ethylene at position 1 and 8 (Fig. 1). It also occurs in small quantity as a result of biomass decay [3]. It has found variety of applications in industrial processing as a precursor and intermediate material for the production of products such as dyes production, plastics, insecticides, fungicides and other manufacturing processes [4, 5]. It is an important component in coal tar for road and highway constructions and as preservative in wood industries [6]. It is among the most prominent organic pollutant ubiquitously found in environmental waters [7]. It is resulted from oil spillage, effluents discharged from petroleum and petrochemical industries, coal conversion process, urban air due to automobile exhaust and cigarette smoke, as well as municipal wastewater due to the soil surface and street asphalt runoff [8, 9]. It is relatively insoluble in water (0.4 mg/100 mL), thus easily transported into the water bodies. Findings have shown the presence of acenaphthene in various ground and surface waters. It is recalcitrant to atmospheric degradation, thus undergoes bioaccumulation in the soil and sediments and subsequently consumed by the plants and aquatic organisms [10]. It was known to cause significant impact to both plants and animals due to its toxic and hazardous effects such as eye and skin irritations, lungs cancer and other respiratory complications [11, 12]. It was listed among the priority PAHs organic pollutant according to USEPA. Thus, essentially required to be eliminated from environmental waters.

Adsorption process have been recognized as an alternative wastewater remediation technology. It is a low-cost and environmental-friendly technique for variety of pollutants removal from the water. The availability of various natural and synthetic adsorbents materials for both organic and inorganic pollutants removal has been capitalized as the way forward compared to other water treatment technologies such as coagulation, reverse osmosis and enzymatic degradation processes [13, 14]. Thus, various adsorbents from natural and sources such as biomass [15], clays, soil and sand particles [16], etc., have been well reported for the acenaphthene adsorption from environmental waters. Despite showing lots of promise in water treatment, most of these adsorbents are not suitable for PAHs removal from water due to the low specific

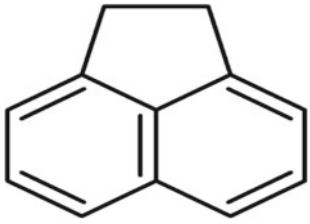

Fig. 1 Chemical structure of acenaphthene

surface area and pore volumes of the adsorbents [17]. The use of synthetic materials such as activated carbons [18], biochar [19], carbon nanotubes, graphene [20], and silica have also been reported. However, AC is not suitable for organic pollutant removal at low concentrations.

Advancement in materials sciences have paved the way for the invention of new classes of materials with versatile applications. Metal-organic frameworks (MOFs) are among the most advanced materials that attracted the attention of researchers from scientific and engineering fields. They are comprised of coordination sphere made from interaction of metal-ion with organic moieties, forming extensive crystalline frameworks [21, 22]. They acquired unique properties such as exceptionally high porosities, mechanical and thermal strength [23, 24]. Thus, they have found tremendous applications in various fields such as carbon dioxide capture, energy storage, sensors, drug delivery, catalysis and wastewater remediations [25, 26]. They offered promising applications as super adsorbents for both organic and inorganic pollutants removal from water [27–29], due to their ultrahigh porosities and good stabilities.

This work is thus aimed at adsorption of acenaphthene from aqueous medium using UiO-66 and ZIF-8 MOFs. They were chosen due to their high BET surface area and pore volumes as well as their stabilities in both aqueous and organic phases. Response surface methodology (RSM) was used for the experimental design using central composite design (CCD). The kinetics of the adsorption process was evaluated, and the reusability of the MOFs was studied.

2 Materials and Methods

The materials purchased in this work were analytically graded and thus were used as received. Acenaphthene standard (99% purity), ZIF-8 (99% purity) zirconium tetrachloride (99.99% purity), terephthalic acid (97% purity) obtained from Sigma Aldrich (USA), and supplied by Avantis Laboratory, Malaysia.

2.1 Synthesis and Characterizations of the MOFs

UiO-66 was synthesized and characterized according to the procedure reported in our previous work [30]. While ZIF-8 was commercially purchased.

2.2 Preparation of the Stock Solution

The acenaphthene stock solution was prepared in acetone by using 10 mg of the standard in 100 ml volumetric flask to make a solution of 100 mg/L. Working solution was prepared from the stock solution in double deionized water using serial dilutions.

2.3 Synthesis of the MOFs

Batch adsorption experiment was designed by the RSM software (Design Expert 11) using full factorial CCD comprises of 5 input variables: contact time (min), dosage (mg), concentration (mg/L), pH and temperature (°C) with 5 center points. The adsorption was conducted using 30 mL of the acenaphthene solution in an incubator shaker (Incubator ES 20/60, bioSan) at 200 rpm. The sample was analyzed using UV-visible spectrophotometer (GENESYS 30) analysis at 220 nm. The responses were determined as the percentage adsorption efficiency achieved by the UiO-66 and ZIF-8 MOFs accordingly to the formula:

$$\%R = \frac{C_0 - C_e}{C_0} \times 100 \tag{1}$$

And the amount of the acenaphthene adsorbed onto the MOFs at certain time (qt) and equilibrium (qe) were determine from the formula:

$$q_t = \frac{(C_0 - C_t)V}{w} \tag{2}$$

$$q_e = \frac{(C_0 - C_e)V}{w} \tag{3}$$

where C_0, C_t and C_e are the initial, time and equilibrium concentrations (mg/L), respectively, w is the weight of adsorbent (g), and V is the volume of the solution (L).

3 Results and Discussion

3.1 RSM Optimizations for Acenaphthene Adsorption onto UiO-66 and ZIF-8

The synthesis of UiO-66 was achieved using solvothermal technique described in our previous studies [30], while ZIF-8 was commercially purchased. The BET surface

area of the MOFs were 1421 m^2/g and 1299 m^2/g, while the pore volumes were 0.91 and 0.60 m^3/g for the UiO-66 and ZIF-8, respectively.

The adsorption studies for the removal acenaphthene were thus conducted using both UiO-66 and ZIF-8. The experimental conditions were determined by the CCD consisting of 5 parameters input variables with 47 number of experimental runs as described in Table 1. The adsorption efficiency of the two MOFs were determined according to each set of the experimental conditions given. The adsorption efficiency of UiO-66 was found to be much higher than that of ZIF-8, which was attributed the higher surface area and pore volume of the MOF [31, 32]. Thus, the highest removal efficiency achieved was 99.7% and 60.7% according to the described multi-variate conditions given in Fig. 2a,b for both UiO-66 and ZIF-8, respectively.

The corresponding 2-dimension (2D) and 3-dimension (3D) graphs for the RSM optimizations of the adsorption process. According to the Fig. 2, the adsorption efficiency was shown to increase with increase in the dosage of the MOFs due to the increase in the available number of adsorption sites on the surface of the MOFs. UiO-66 has shown to efficiently adsorbed larger organic molecules such as dyes and pharmaceuticals from aqueous solutions, attributed to its potential breathing capacity [33], high porosity [34], hydrophobicity and extensive stabilities in both organic and aqueous medium [35]. Similarly, the adsorption efficiency was increase with the increasing the contact time for the adsorption due to the increase in the interaction of the acenaphthene with the surface of the MOFs.

The model fittings were described by the analysis of variance (ANOVA) as presented in Table 2 according to the Fischer test (F-test) and p-values (probability > F) with 95% confidence level for the input variables. The overall model p-values were significant (< 0.0500), with F-value of 33.44 and 126.34 for the UiO-66 and ZIF-8, respectively. The significant terms for the adsorption and parameters combination were A, B, C, D, AD, A^2 and A, B, C, D, AB, BD, A^2 for the UiO-66 and ZIF-8 respectively. The significance of the model was also described by the reasonable agreements between the adjusted R^2 (0.9338 and 0.9820 for UiO-66 and ZIF-8, respectively) and predicted R^2 (0.8733 and 0.9638 for UiO-66 and ZIF-8, respectively) values with less than 20% differences. The lack of fit test for the replicated design points of the residual errors against the pure errors for the model were 0.1915 and 0.5840. The adequate precision of the model was 17.3082 and 32.6708 for the UiO-66 and ZIF-8, respectively, implying that the model can be used to navigate the design space for the adsorption studies. The scatter plots (Fig. 3) have shown significance of the model in terms of good agreement between the experimental and predicted adsorption efficiencies. Thus, RSM model was significant for the design optimizations of acenaphthene adsorption onto UiO-66 and ZIF-8.

3.2 Contact Time and Kinetics Study

The uptake of acenaphthene by the UiO-66 and ZIF-8 MOFs were studied. Adsorption experiment was studied using the optimized RSM data. Figure 4 highlighted the

Table 1 RSM optimizations for acenaphthene adsorption onto UiO-66 and ZIF-8 MOFs

Name	Units	Low	High	− Alpha	+ Alpha
A: Time	min	5	25	0	35
B: Dosage	mg	3	5	2	6
C: Concentration	mg/L	1	3	0	4
D: pH		2	6	0	8
E: Temperature	°C	25	35	20	40

Std	Run	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Response 1	Response 2
		Time	Dosage	Concentration	pH	Temperature	UiO-66 (%R)	ZIF-8 (%R)
		min	mg	mg/L		°C		
1	1	5	3	1	2	25	67.3	30.2
3	2	5	5	1	2	25	77.1	33.5
22	3	25	3	3	2	35	84.2	50.2
24	4	25	5	3	2	35	97.5	53.6
14	5	25	3	3	6	25	96.1	51.2
16	6	25	5	3	6	25	98.4	55.2
36	7	15	6	2	4	30	87.4	45.7
45	8	15	4	2	4	30	85.2	44.3
35	9	15	2	2	4	30	72.2	42.1
5	10	5	3	3	2	25	58.5	29.6
17	11	5	3	1	2	35	63.2	31.5
20	12	25	5	1	2	35	98.5	57.4
42	13	15	4	2	4	40	79.3	44.4
7	14	5	5	3	2	25	58.6	33.8
46	15	15	4	2	4	30	79.1	43.6
31	16	5	5	3	6	35	56.3	32.4
33	17	5	4	2	4	30	54.2	32.1
26	18	25	3	1	6	35	95.5	56.3
28	19	25	5	1	6	35	97.1	57.2
32	20	25	5	3	6	35	92.4	55.4
43	21	15	4	2	4	30	77.3	43.7
39	22	15	4	2	10	30	76.2	44.2
18	23	25	3	1	2	35	94.3	56.3
25	24	5	3	1	6	35	55.7	33.5
34	25	35	4	2	4	30	99.7	60.7
2	26	25	3	1	2	25	94.1	56.4
41	27	15	4	2	4	20	76.5	44.5

(continued)

Table 1 (continued)

Std	Run	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Response 1	Response 2
		Time	Dosage	Concentration	pH	Temperature	UiO-66 (%R)	ZIF-8 (%R)
		min	mg	mg/L		°C		
10	28	25	3	1	6	25	98.5	56.7
27	29	5	5	1	6	35	57.2	34.8
4	30	25	5	1	2	25	99.1	57.5
8	31	25	5	3	2	25	97.8	55.4
11	32	5	5	1	6	25	59.5	35.1
29	33	5	3	3	6	35	56.4	32.5
15	34	5	5	3	6	25	58.3	33.3
47	35	15	4	2	4	30	77.7	44.2
30	36	25	3	3	6	35	92.6	54.6
21	37	5	3	3	2	35	56.4	32.3
23	38	5	5	3	2	35	58.4	33.6
38	39	15	4	4	4	30	75.5	43.6
44	40	15	4	2	4	30	77.3	45.2
6	41	25	3	3	2	25	91.5	53.7
12	42	25	5	1	6	25	98.5	55.2
9	43	5	3	1	6	25	55.4	33.5
40	44	15	4	2	8	30	77.3	46.3
19	45	5	5	1	2	35	56.4	35.2
13	46	5	3	3	6	25	55.4	33.4
37	47	15	4	5	4	30	75.2	43.2

effect of contact time for the adsorption process. Sudden adsorption of the acenaphthene was observed at the onset of the experiment, attributed to the higher BET surface area of the MOFs [36]. The equilibrium of the adsorption was attained within 30 min with the MOFs achieving equilibrium adsorption capacities of 23.814 mg/g and 14.627 mg/g for UiO-66 and ZIF-8, respectively. The adsorption efficiency achieved at equilibrium was 99.23% and 60.95% for UiO-66 and ZIF-8, respectively, confirming the superior adsorption capacity of UiO-66 and its higher porosities as previously reported in our studies [37].

To understand the mechanism and the rate controlling step for the process, the adsorption data for the UiO-66 and ZIF-8 effect of contact time was kinetics models was treated by the kinetics models of pseudo-first order (Eq. 4), pseudo-second order (Eq. 5), intra-particle diffusion (Eq. 6) and Elovich (Eq. 7) models linearly expressed as.

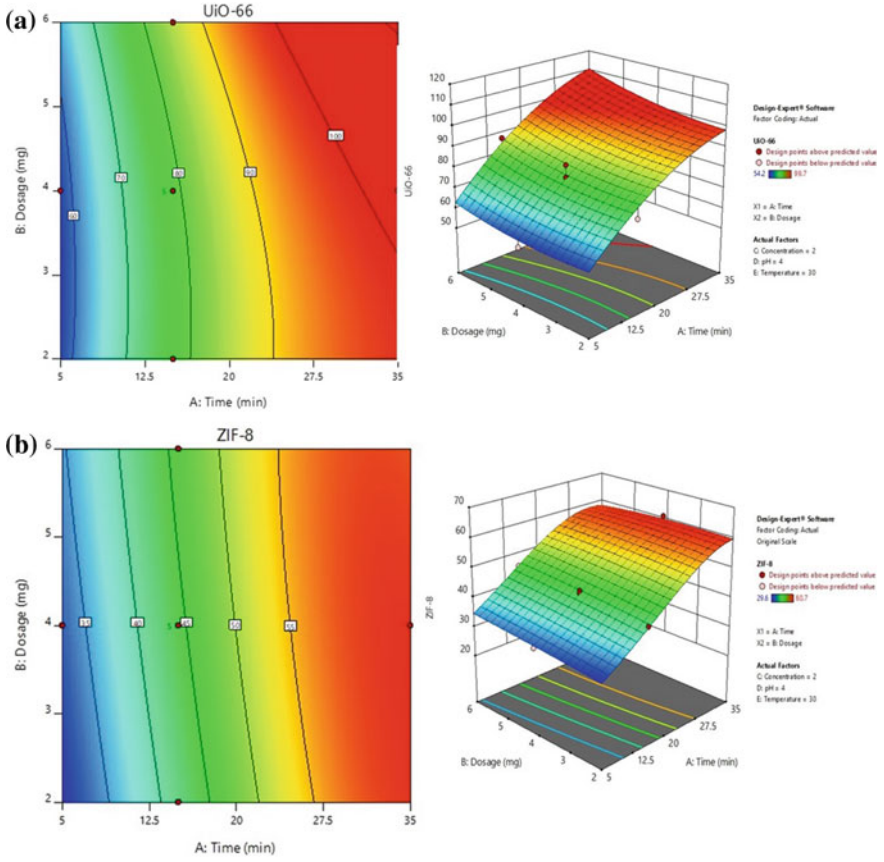

Fig. 2 2D and 3D RSM plots for the adsorption of acenaphthene onto **a** UiO-66 and **b** ZIF-8 MOFs under the condition: contact time of 35 min; dosage of 4 mg; concentrations of 2 mg/L, pH of 4 and temperature of 30 °C

$$\ln(q_e - q_t) = \ln q_e - k_1 t \tag{4}$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \tag{5}$$

$$q_t = K_P t^{\frac{1}{2}} + C \tag{6}$$

$$q_t = \frac{1}{\beta} In() + \frac{1}{\beta} Int \tag{7}$$

where q_e (mg/g) and q_t (mg/g) represent the amount of the acenaphthene adsorbed by the MOFs at equilibrium and t (mins). k_1 (1/min) k_2 (q/mg/min) represented the

Table 2 ANOVA for the adsorption of acenaphthene onto UiO-66 and ZIF-8 MOFs

	UiO-66						ZIF-8					
Source	SS	df	MS	F-value	p-value		SS	df	MS	F-value	p-value	
Model	0.0004	20	0.0000	33.44	< 0.0001	Significant	0.0014	20	0.0001	126.34	< 0.0001	Significant
A-Time	0.0004	1	0.0004	636.44	< 0.0001		0.0013	1	0.0013	2455.38	< 0.0001	
B-Dosage	3.434E−06	1	3.434E−06	5.77	0.0238		0.0000	1	0.0000	22.62	< 0.0001	
C-Concentration	3.811E−06	1	3.811E−06	6.40	0.0178		5.770E−06	1	5.770E−06	10.76	0.0029	
D-pH	2.554E−06	1	2.554E−06	4.29	0.0484		3.105E−06	1	3.105E−06	5.79	0.0235	
E-Temperature	1.980E−06	1	1.980E−06	3.33	0.0797		2.672E−07	1	2.672E−07	0.4985	0.4864	
AB	8.016E−09	1	8.016E−09	0.0135	0.9085		3.309E−06	1	3.309E−06	6.17	0.0197	
AC	8.144E−07	1	8.144E−07	1.37	0.2528		1.668E−07	1	1.668E−07	0.3112	0.5817	
AD	4.568E−06	1	4.568E−06	7.67	0.0102		2.068E−06	1	2.068E−06	3.86	0.0603	
AE	7.053E−07	1	7.053E−07	1.18	0.2865		4.333E−07	1	4.333E−07	0.8083	0.3769	
BC	4.427E−08	1	4.427E−08	0.0743	0.7873		2.146E−08	1	2.146E−08	0.0400	0.8430	
BD	2.858E−08	1	2.858E−08	0.0480	0.8283		3.603E−06	1	3.603E−06	6.72	0.0154	
BE	5.467E−07	1	5.467E−07	0.9181	0.3468		2.582E−07	1	2.582E−07	0.4816	0.4938	
CD	1.962E−06	1	1.962E−06	3.30	0.0810		9.847E−08	1	9.847E−08	0.1837	0.6718	
CE	4.490E−07	1	4.490E−07	0.7540	0.3932		2.190E−07	1	2.190E−07	0.4085	0.5283	
DE	7.972E−07	1	7.972E−07	1.34	0.2578		4.070E−07	1	4.070E−07	0.7592	0.3915	
A^2	0.0000	1	0.0000	33.65	< 0.0001		0.0000	1	0.0000	86.03	< 0.0001	
B^2	2.631E−08	1	2.631E−08	0.0442	0.8352		8.056E−08	1	8.056E−08	0.1503	0.7014	
C^2	1.937E−07	1	1.937E−07	0.3253	0.5734		4.969E−07	1	4.969E−07	0.9270	0.3445	
D^2	2.434E−07	1	2.434E−07	0.4088	0.5282		3.000E−07	1	3.000E−07	0.5595	0.4612	
E^2	9.380E−09	1	9.380E−09	0.0158	0.9011		1.998E−08	1	1.998E−08	0.0373	0.8484	

(continued)

Table 2 (continued)

	UiO-66						ZIF-8					
Source	SS	df	MS	F-value	p-value		SS	df	MS	F-value	p-value	
Residual	0.0000	26	5.955E−07				0.0000	26	5.361E−07			
Lack of fit	0.0000	22	6.564E−07	2.52	0.1915	Not significant	0.0000	22	6.146E−07	5.90	0.0484	Not significant
Pure error	1.043E−06	4	2.607E−07				4.170E−07	4	1.043E−07			
Cor total	0.0004	46					0.0014	46				

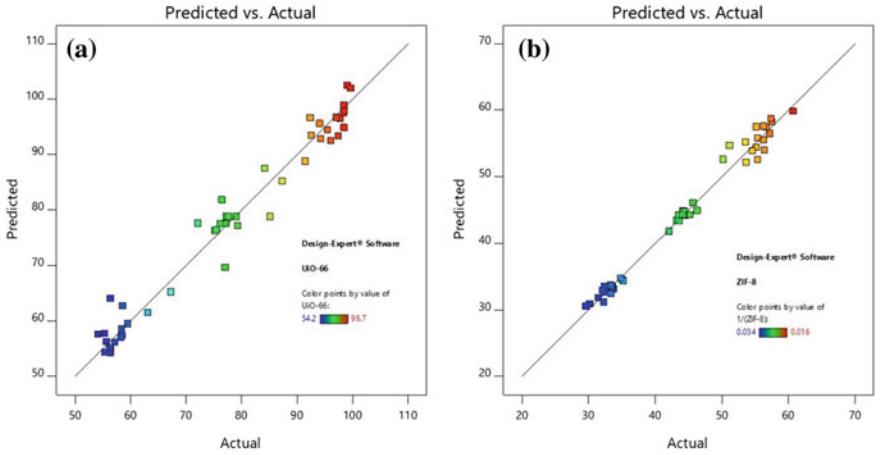

Fig. 3 Scatter plots for the RSM adsorption of acenaphthene onto UiO-66 and ZIF-8 MOFs

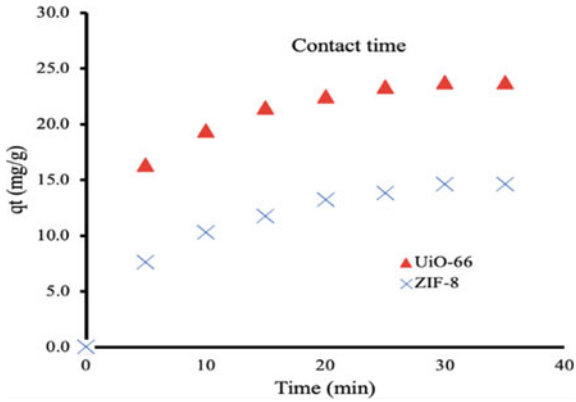

Fig. 4 Effect of contact time for the adsorption of acenaphthene onto UiO-66 and ZIF-8 MOFs

pseudo-first order and pseudo-second order rate constants, respectively. Kp represented the intraparticle rate constant and C is constant. The α (mg/gmin) and β (g/mg) were the initial desorption rate and desorption constant for the Elovich model, respectively.

Figure 5 depicted the plots of the kinetic models studied. The adsorption data was found to be precisely suited by the pseudo-second order model in comparison to other models evaluated. It is R^2 value was 0.9947 and 0.97780 for the UiO-66 and ZIF-8, respectively. Also, its calculated adsorption capacities were 24.80 and 15.87 mg/g for the UiO-66 and ZIF-8, respectively, very closer to the obtained experimental values of 23.814 and 14.627 mg/g for UiO-66 and ZIF-8, respectively, indicating the chemisorption is the rate limiting step of the process and the availability of the adsorption sites on the surface of the MOFs. Other reports have also described

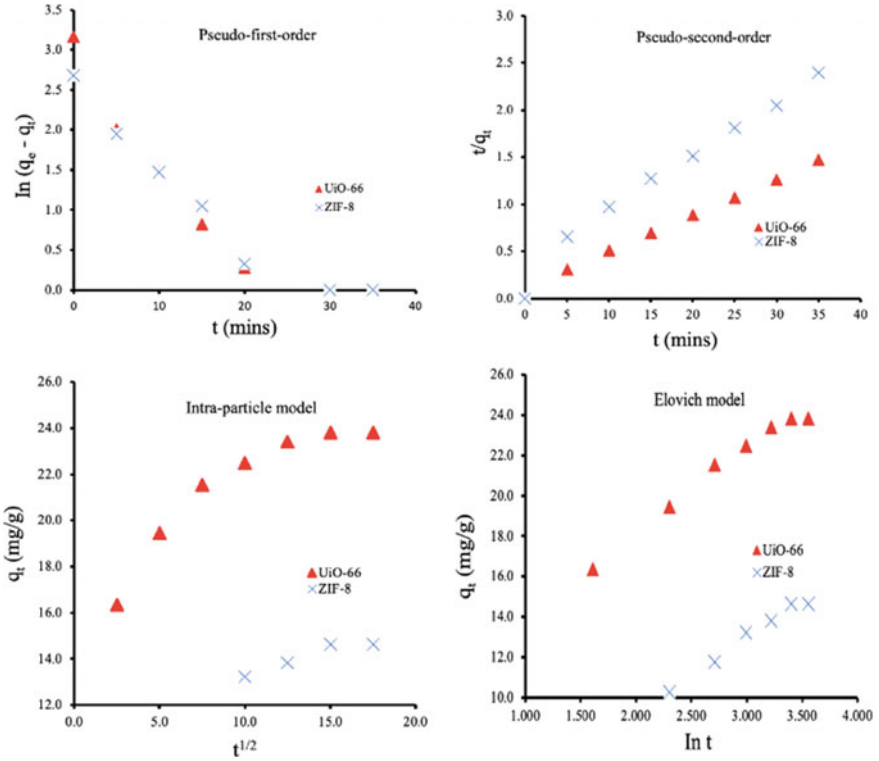

Fig. 5 Adsorption kinetics plots for acenaphthene adsorption onto UiO-66 and ZIF-8 MOFs

the adsorption of organic pollutants onto UiO-66 [38, 39] and ZIF-8 [40, 41] as pseudo-second order process.

4 Conclusion

RSM was used for the experimental design of acenaphthene adsorption onto UiO-66 and ZIF-8 MOFs. Five factor parameters were employed, and best performance of the MOFs was achieved for UiO-66 with adsorption efficiency of 99.7 due to its higher porosity in comparison to ZIF-8 which recorded adsorption efficiency of 60.7%, according to the experimental condition with contact time of contact time of 35 min, dosage of 4 mg, concentrations of 2 mg/L, pH of 4 and temperature of 30 °C. The overall model p-values were significant (< 0.0500) for both UiO-66 and ZIF-8 MOFs. The lack of fit test for the model was insignificant, which was desirable for both MOFs and the adequate precision was 17.3082 and 32.6708 for the UiO-66 and ZIF-8, respectively. Good agreement between the RSM and experimental findings.

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