

Article

Assessment of Dispersed Oil Sorption in Oily Wastewater onto Hydrophobized/Oleophilized Autoclaved Aerated Concrete (AAC) Grains

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Abstract: The discharge of untreated oily wastewater into the environment has serious impacts on human health, living nature, and ecosystems and leads to significant economic losses. Many engineering techniques have been proposed and applied to treat oily wastewater, but limited studies have investigated low-cost and effective techniques using by-products and waste/scrap materials from the construction industry. Materials to treat oily wastewater are needed not only to mitigate environmental pollution but also to promote the reuse and recycling of industrial by-products, especially in developing countries. This study, therefore, examined the sorption capacity of dispersed oil in wastewater (dispersed soybean oil in water; initial oil concentrations, $C_i = 10\text{--}1000$ mg/L; oil droplet size in water <2 μm) onto the hydrophobized/oleophilized autoclaved porous aerated concrete (AAC) grains made from waste scrap in Vietnam by using batch sorption tests in the laboratory. The AAC grains (sizes 0.106–0.25, 0.25–0.85, and 0.85–2.00 mm) were hydrophobized/oleophilized using oleic and stearic acids (coating concentrations of 1.0, 5.0, and 10 g/kg), and two sands (0.18–2.00 and 0.30–2.00 mm) were used as control samples. The results showed that the hydrophobized/oleophilized AAC grains had high sorption capacity for dispersed oil (i.e., high oil removal efficiency) compared to the control sands. Especially, the removal of AAC grains coated with stearic acid was $>80\%$ in high oil concentration solutions ($C_i = 100$ and 1000 mg/L), indicating that the hydrophobized/oleophilized AAC grains have high potential as useful adsorbents to trap dispersed oil in oily wastewater. Moreover, adsorption isotherms were drawn to examine the sorption characteristics of dispersed oil onto AAC grains. For all tested samples, the sorption of dispersed oil increased linearly with increasing equilibrium concentration. The commonly used Langmuir model, on the other hand, did not capture the measured isotherms.

Keywords: dispersed oil; oily wastewater; autoclaved aerated concrete (AAC); sorption; Vietnam



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1. Introduction

The improper discharge and treatment of oily wastewater cause water pollution. In particular, developing countries with rapid urbanization, industrialization, and population growth face severe water pollution from oily wastewater, seriously affecting human health and natural ecosystems [1–3]. For example, Vietnam (one of the growing, developing countries) reports that improper treatment of oily wastewater and oil spills from human activities and industry have caused severe water pollution throughout the country, with the total amount of oil and grease exceeding environmental standards [4–6]; see also Tables A1–A3 in the Appendix A. Therefore, urgent action is required to conserve the water environment and sustainable development [1–3].

The treatment of oily wastewater is required worldwide, and the allowable concentration of fats and oils must be regulated before the treated wastewater is released into

the environment. The Water Pollution Prevention Act (1970) of Japan, for example, limits the maximum permissible levels of treated water discharge to 5 mg/L for mineral oil and 30 mg/L for animal and vegetable oils. In Vietnam, on the other hand, the permissible levels of surface water, groundwater, seawater, and others range between 0.05–1.0 mg/L, and the permissible levels of animal and vegetable oils and grease in domestic wastewater and specific wastewater from industrial and service sectors range between 5–30 mg/L depending on the destination of the discharge (see national standards shown in Table A1 of the Appendix A; note that the oil and wastewater standards in Vietnam are similar to those in Japan [7–18]).

Now, many technologies have been developed and applied to treat oily wastewater [19–22]. Among them, oil/water separation technologies that use hydrophobic and lipophilic membranes, meshes, and grains as filtration and adsorption materials have been intensively developed due to their high treatment performance and cost-effectiveness, e.g., [23]. Since the oil/water separation technology does not require centralized wastewater collection systems or large numbers of water treatment tanks and ponds, it can be incorporated into small-scale decentralized wastewater treatment systems with fixed-bed filtration tanks and floating filtration devices for domestic wastewater in rural areas (small number of households) generated from food processing factories and craft production villages in developing countries including Vietnam [24]. The oil/water separation technique, however, mainly targets treating oil and grease in wastewater existing in the forms such as free and floating oil [25–28]. Limited studies have been conducted to treat the dispersed oil in wastewater (the typical size of oil droplet ranges in the scale of μm to nm), and the adsorption capacity and mechanism of dispersed oil in water onto adsorbent grains have not been fully examined [29–36].

In Vietnam, moreover, construction and demolition waste (CDW) has not been fully reused and recycled and is mostly dumped without any treatment [37–39]. Among CDW, autoclaved aerated concrete (AAC) has a unique pore structure consisting of inner pores (μm to nm scale) and inter-pores (mm to μm scale) [40–45]. It has been reported that crushed AAC grains act as good sorbents for the simultaneous removal of Cd^{2+} and Pb^{2+} in wastewater [46,47]. In addition, the AAC grains, as well as stearic acid coated (i.e., hydrophobized/oleophilized) AAC grains, showed high performance of oil and removal chemical oxygen demand (COD) in the filtration flow system in the laboratory [48]. These suggest that the AAC grains may act as effective adsorbents to treat dispersed oil in wastewater.

This study, therefore, aimed to assess the sorption capacity of dispersed oil onto AAC grains as well as hydrophobized/oleophilized AAC grains coated with oleic and stearic acids using batch sorption tests in the laboratory. For comparing the sorption capacity of AAC grains, commercially-available filtration sands were also used as control samples. Moreover, the applicability of typical adsorption isotherm models (Langmuir, Freundlich, and linear) was examined against the measured data from batch sorption tests.

2. Materials and Methods

2.1. AAC Grains and Sands

AAC scrap was obtained from Viglacera Joint Stock Company, Bac Ninh Province, Vietnam ($21^{\circ}11'50.8''$ N, $106^{\circ}00'42.8''$ E) [49]. These were crushed into 10 mm or fewer grains, and the grains were gently washed with a low-foaming neutral detergent and thoroughly rinsed with distilled water. The AAC grains were then air-dried and sieved into three particle size fractions: 0.106–0.250 mm, 0.250–0.850 mm, and 0.850–2.00 mm [50]. For comparison with test data for AAC grains, two commercially available grains of control sands of particle size fractions: 0.18–2.00 mm and 0.30–2.00 mm (Nippon Genryo Material Co., Ltd., Kanagawa, Japan) meeting Japanese filtration standards [51] were also used in this study.

2.2. Hydrophobic Agents and Coating

Two hydrophobic agents (HA), stearic acid and oleic acid (SA and OA), were chosen to hydrophobized/oleophilized coat the grains tested in this study [48,52–56]. OA (Chemical formula: $\text{CH}_3(\text{CH}_2)_7\text{CH}=\text{CH}(\text{CH}_2)_7\text{COOH}$ molar mass: 282.46 g/mol, density: 0.895 g/cm^3) (Kanto Chemical, Tokyo, Japan) and SA (Chemical formula: $\text{CH}_3(\text{CH}_2)_{16}\text{COOH}$, molar mass: 284.47 g/mol, density: 0.940 g/cm^3) (Fuji Film Wako Pure Chemicals, Tokyo, Japan) were used. Both OA and SA are materials that are harmless to humans and the environment and are inexpensive in Vietnam.

The hydrophobized/oleophilized coating was performed according to [56]. The target HA concentrations were OA, SA = 0, 1, 5, 10 g/kg for AAC and OA, SA = 0, 2, 5 g/kg for sands, following the results of [56].

2.3. Dispersed Oil

This study used soybean oil (Wako 1st Grade; Fujifilm Wako Pure Chemicals Corporation, Tokyo, Japan), a domestic oil commonly consumed in Vietnam, and liquid at room temperature [57]. Figure 1a shows an image of dispersed oil (soybean oil) after treatment with ultrasonic oil in water at each initial concentration ($C_i = 0, 10, 50, 100, 1000 \text{ mg/L}$). In this study, dispersed oil was targeted, so soybean oil was dispersed with an ultrasonic device (UP400st, Hielscher, Germany).

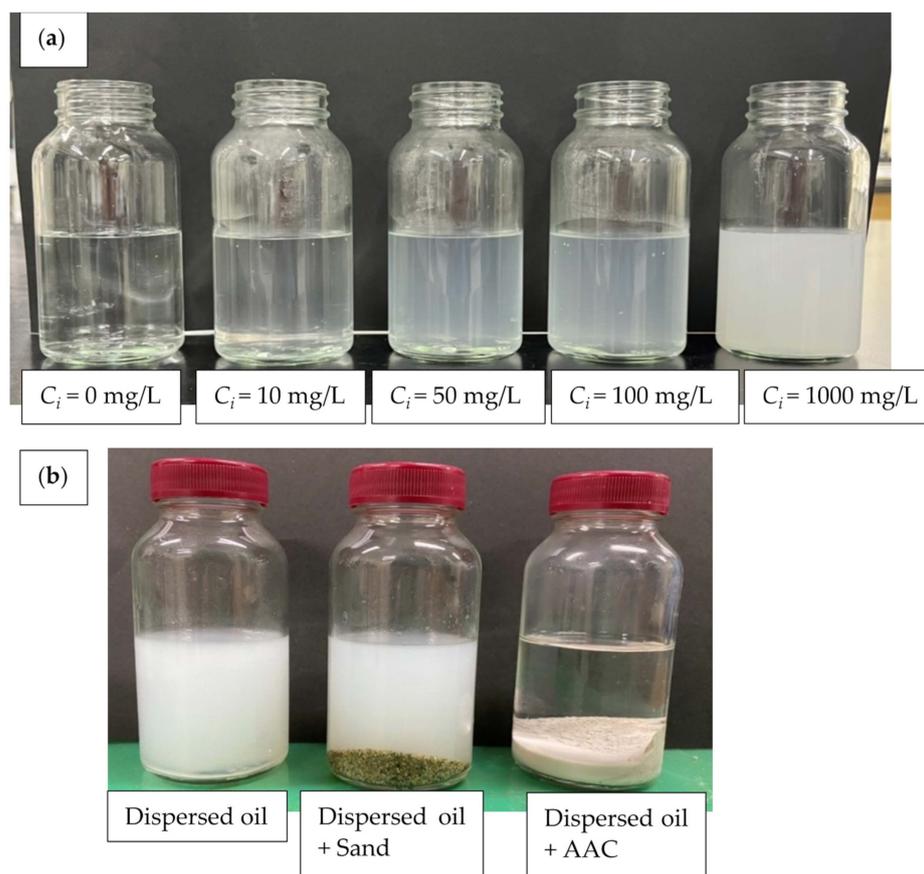


Figure 1. (a) Dispersed oil (soybean oil) after ultrasonic mixing. (b) Before and after batch sorption test of dispersed oil ($C_i = 1000 \text{ mg/L}$).

Figure 2 shows the particle size distributions of dispersed oil in water measured by a laser diffraction nanoparticle size distribution analyzer (SALD-7100, Shimadzu, Kyoto, Japan). For both $C_i = 1000 \text{ mg/L}$ and $C_i = 100 \text{ mg/L}$, dispersed oil had stable particle sizes for 24 h. In addition, the most frequent particle size was about $0.1\text{--}1.0 \mu\text{m}$.

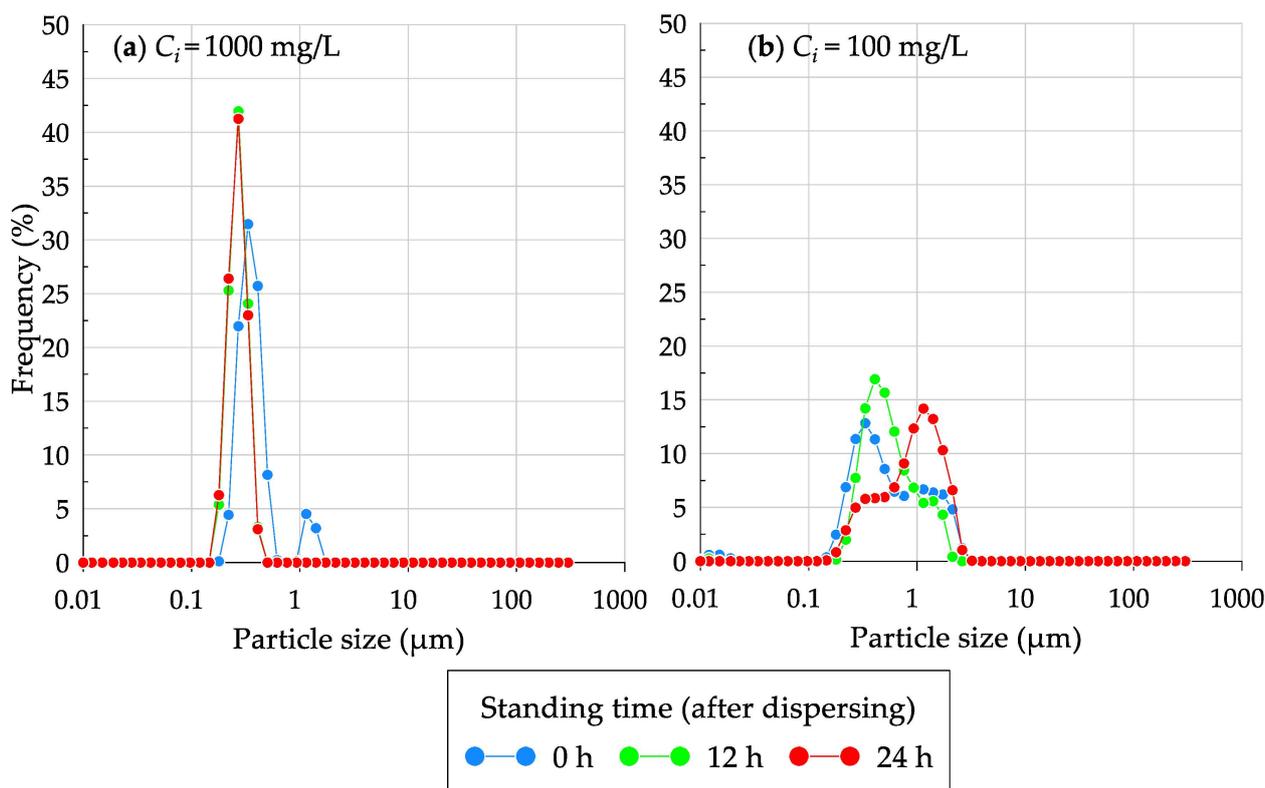


Figure 2. Particle size distributions of the dispersed oil in water.

2.4. Batch Sorption Test

Batch sorption tests were carried out following protocols of the OECD (2000) [58]. The horizontal shaking speed was modified from 100 rpm/min to 20 rpm/min in this study to avoid the peeling of the coated hydrophobic agents (the same as ASTM F726–99 [59]). The liquid–solid ratio (L/S) was set to 10, and the horizontal shaking times were set to 1, 3, 6, 12, 24, and 48 h at 20 °C. Before and after the batch test of tested samples are shown in Figure 1b.

2.5. Analysis

The oil concentration of the separated oil in water was measured by an oil content analyzer (OCMA–505–H, Horiba, Kyoto, Japan). Figure 3 compares the oil concentration of the n–hexane extract and the oil concentration measured by the oil content analyzer. A good linear relationship could be seen, so the actual value of the oil concentration was taken as the value obtained by multiplying the output value of the oil concentration meter by the linear regression equation in this study ($y = 0.42x$; $R^2 = 0.99$).

Using this relationship, the equilibrium concentration C_e can be set for the measured oil concentration C from Equation (1) in the case of OA, SA = 0 g/kg:

$$C_e = C/0.42 \tag{1}$$

In the case of OA, SA = 1, 2, 5, 10 g/kg:

$$C_e = (C - C_0)/0.42 \tag{2}$$

where C_0 is the oil concentration of $C_i = 0$ mg/L when OA and SA are 1, 2, 5, and 10 g/kg. Using the measured C_i and C_e , the removal percentage of dispersed oil in water (R , %) was calculated by Equation (3):

$$R = 100 \times (C_i - C_e)/C_i \tag{3}$$

The oil sorption amount at equilibrium (mg/g) was obtained in Equation (4):

$$q_e = (C_i - C_e)V/1000 \times m_s \quad (4)$$

where V is the sorbate volume (=100 mL of dispersed oil in water), and m_s is the sorbent amount (=10 g of AAC grains or sand).

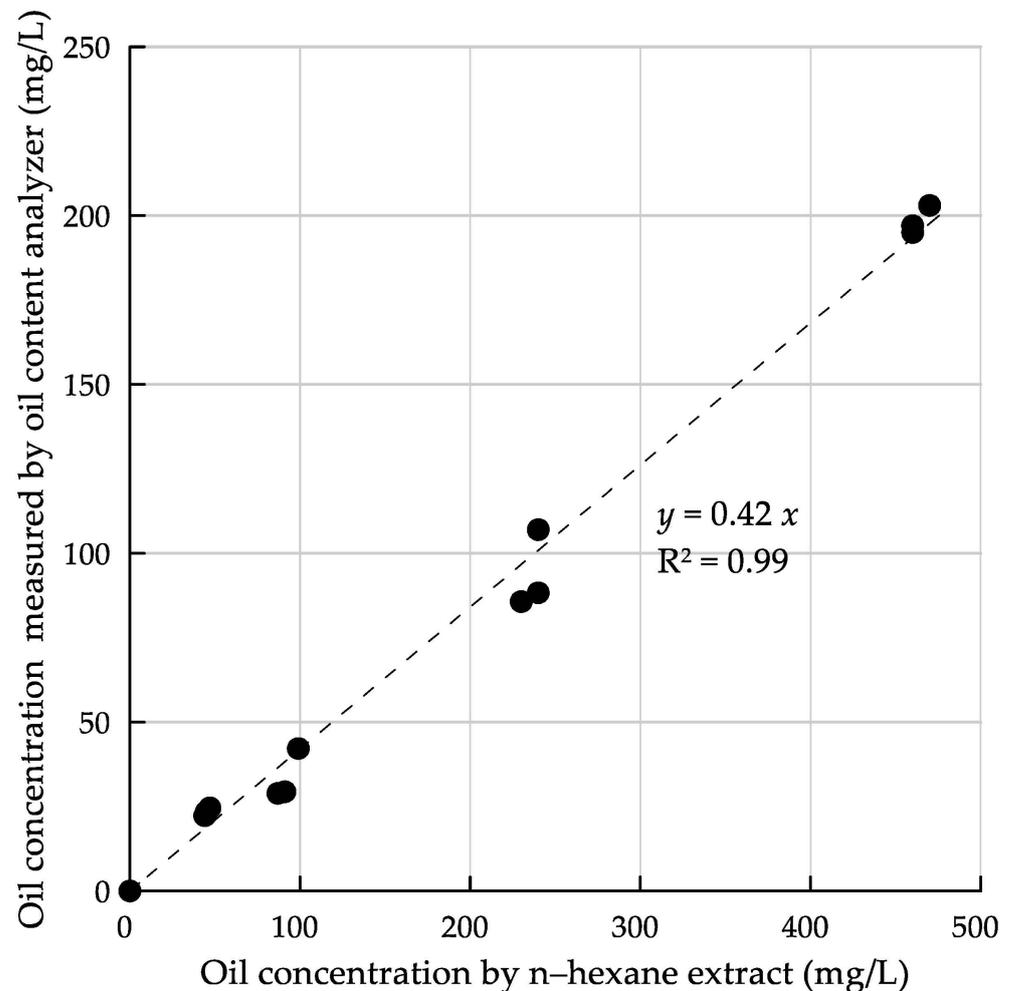


Figure 3. Relationship between oil concentration measured by oil concentration analyzer and normal hexane measurement.

3. Results and Discussions

3.1. Effects of Shaking Time on Removal of Dispersed Oil in Water

Figure 4 shows the effect of shaking time on the removal of dispersed oil, R (%), for tested AAC grains with different sizes at $C_i = 1000$ mg/L. Although some variations in measured R values for all tested samples (non-coated, OA-, and SA-coated) were observed, the R values mostly increased with increased shaking time and became stable at 24 h of shaking. This suggests that the sorption process of dispersed oil onto AAC grains (especially 0.250–0.850 and 0.850–2.00 mm in Figure 4b,c) depends on the contact time to some extent and that 24 h of shaking time would be suitable to examine the sorption capacity of tested AAC grains.

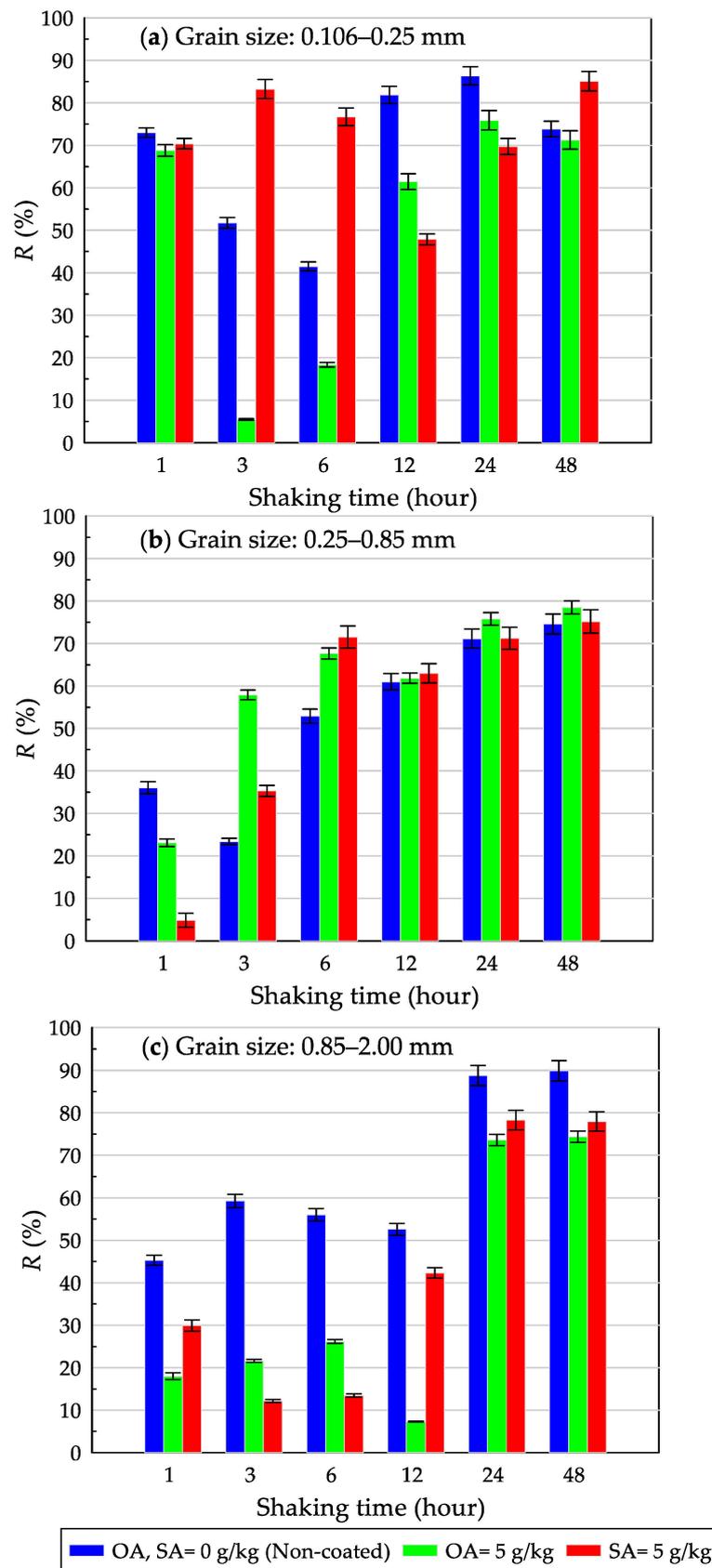


Figure 4. Measured removal percentage of dispersed oil in water (R , %) values for AAC grains with different shaking times at $C_i = 1000$ mg/L: (a) 0.106–0.25 mm, (b) 0.25–0.85 mm, (c) 0.85–2.00 mm. Error bars indicate standard deviations of measured data.

3.2. Effect of Initial Dispersed Oil Concentration on Removal Percent of Dispersed Oil

Measured R values for all tested AAC grains with different coating percent (OA/SA = 0 (non-coated), 1, 5, and 10%) at $C_i = 10, 50, 100,$ and 1000 mg/kg are shown in Figure 5. Overall, the grain size of AAC did not affect the measured R values for tested uncoated, OA-coated, and SA-coated samples in this study. For low C_i solutions such as 10 and 50 mg/L, AAC grains coated with OA = 1 g/kg (Figure 5b) and SA = 1 g/kg (Figure 5e) removed dispersed oil well, and measured R values became >80% irrespective of grain size. For high C_i solutions such as 100 and 1000 mg/L, on the other hand, high SA-coated AAC grains (10 g/kg in Figure 5g) gave high R values of >80%. It is interesting that non-coated AAC grains also removed the high-concentration dispersed oil solution ($C_i = 1000$ mg/L) well, and the R values became > 80% (Figure 5a).

Based on the measured R values in this study, the coating of hydrophobic/oleophilic agents yields both positive and negative effects on AAC grains (adsorbents) from the viewpoint of removal of dispersed oil in water. As shown by Matsuno and Kawamoto, the coating of AAC grains with hydrophobic/oleophilic agents reduces the specific surface area of AAC grains and affects the hydrophobicity/oleophilicity of AAC grain surface in water [56]. Moreover, the affinity between dispersed oil and the hydrophobized/oleophilized AAC grain surface depended on the initial dispersed oil concentration, resulting in suitable coating conditions that gave high R values. This strongly suggests that suitable coating should be examined against the concentration of the target oily wastewater.

Figure 6, for reference, shows the measured R values of control sands with different sizes at $C_i = 10, 50, 100,$ and 1000 mg/L. The measured R values were lower than those from AAC grains (Figure 5) in the whole range of C_i . Especially, the coating of hydrophobic/oleophilic agents for sands did not contribute to the removal of dispersed oil at the low concentration of $C_i = 10$ mg/L (i.e., R values became <5%).

3.3. Application of Adsorption Isotherm Models to Characterize the Dispersed Oil Sorption onto AAC Grains

In order to understand the sorption properties of dispersed oil in water onto AAC grains, adsorption isotherm models, the Langmuir model [60], the Freundlich model [61], and a simple linear model commonly used to characterize the adsorption process and mechanism of dissolved ions and metals onto adsorbents were applied. The Langmuir model described the relationship between C_e and q_e :

$$C_e/q_e = 1/(bq_{\max}) + C_e/q_{\max} \quad (5)$$

where q_{\max} (mg/g) is the maximum adsorption capacity, and b (L/mg) is the Langmuir constant related to binding strength. The Freundlich model is described as follows:

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

where K_F (mg/g) is the Freundlich constant and $1/n$ is the adsorption intensity factor or surface heterogeneity. A simple linear adsorption model is described:

$$q_e = K_d C_e \quad (7)$$

where K_d is the linear adsorption coefficient (L/mg).

The fitted parameters for adsorption isotherms models are summarized in Table 1, and the measured relationship between C_e and q_e for non-coated AAC grains, non-coated Sands, AAC grains coated at OA/SA = 5 g/kg, and Sands coated at OA/SA = 5 g/kg are exemplified in Figure 7.

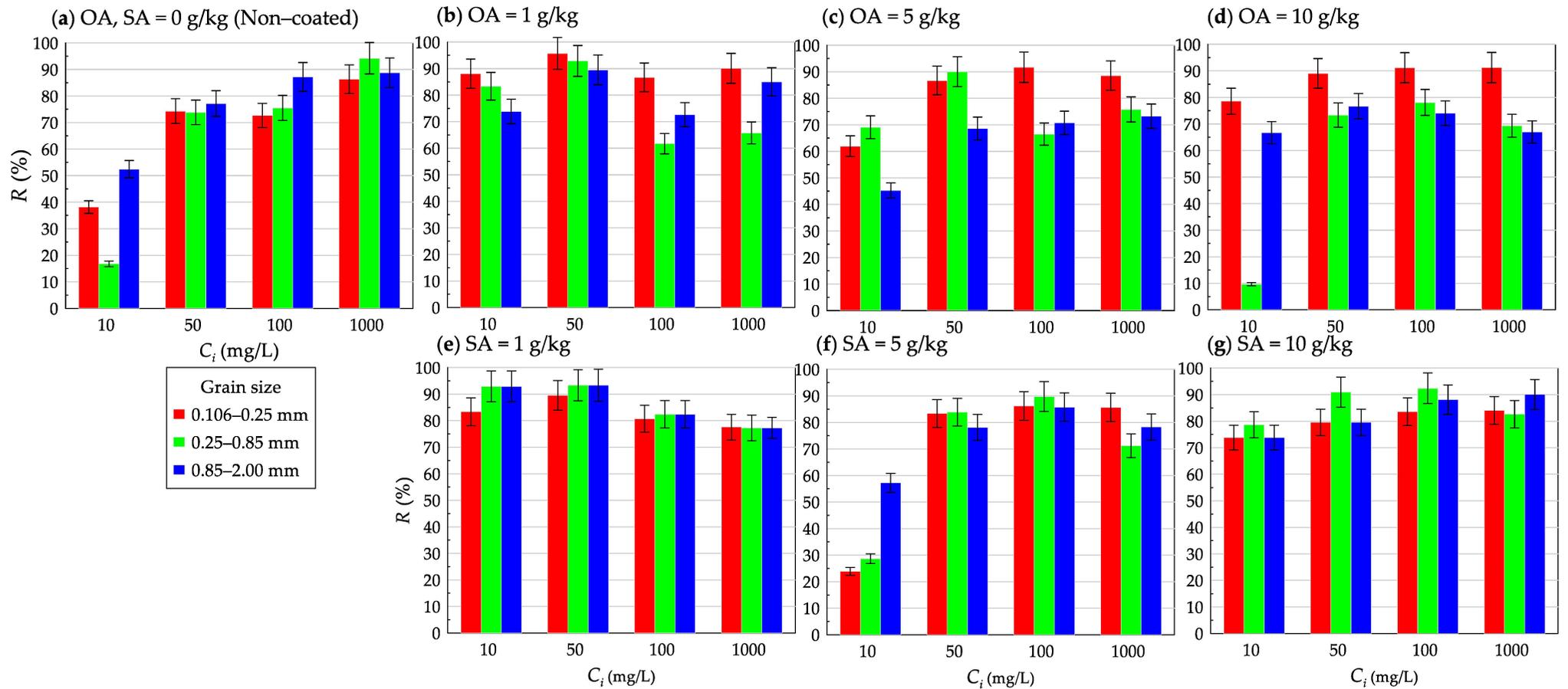


Figure 5. Measured removal percentage of dispersed oil in water (R , %) for AAC grains with different sizes and different coating conditions at $C_i = 10, 50, 100,$ and 1000 mg/kg. Error bars indicate standard deviations of measured data.

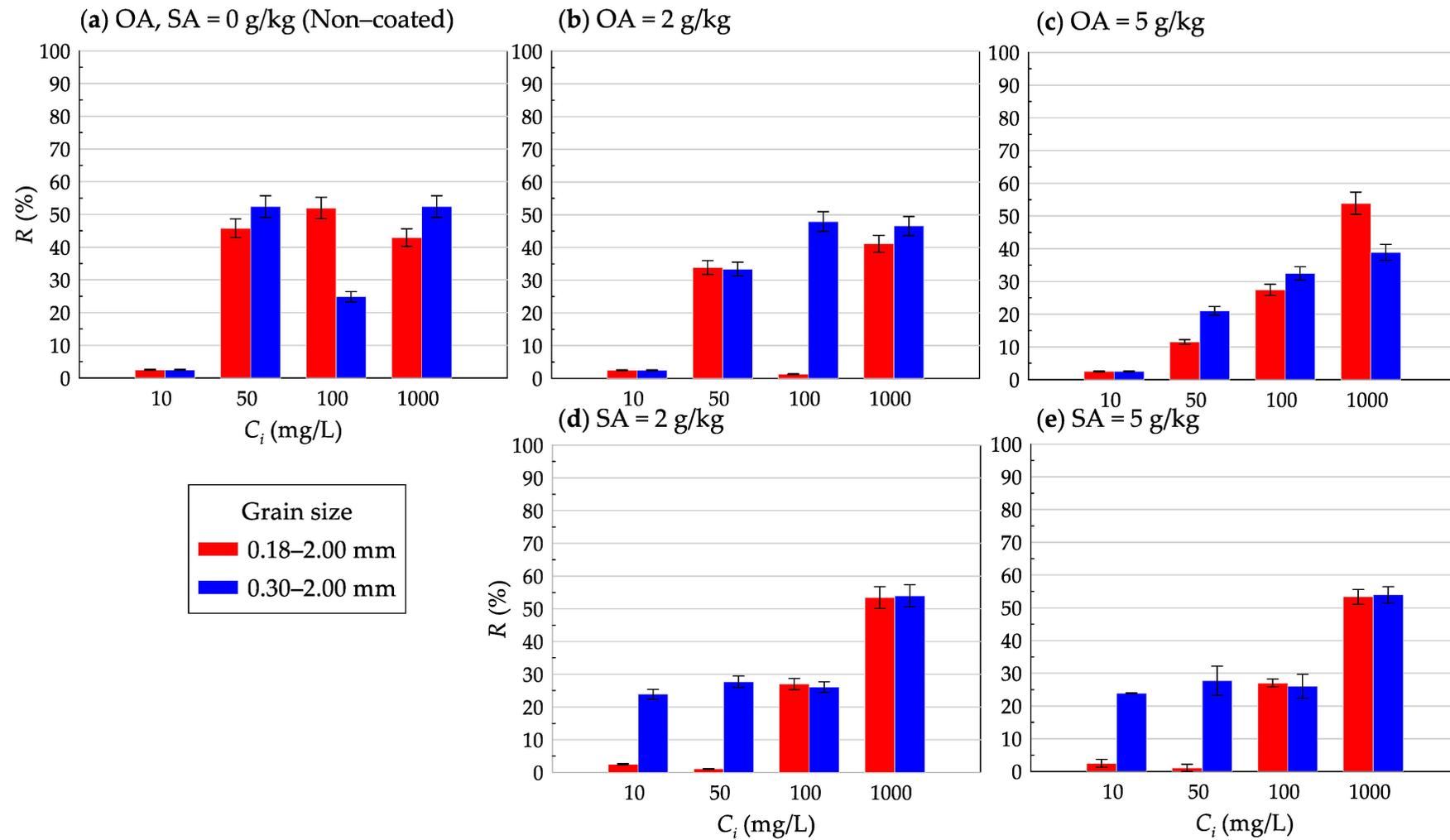


Figure 6. Measured removal percentage of dispersed oil in water (R , %) for sands with different sizes and different coating conditions at $C_i = 10, 50, 100,$ and 1000 mg/kg. Error bars indicate standard deviations of measured data.

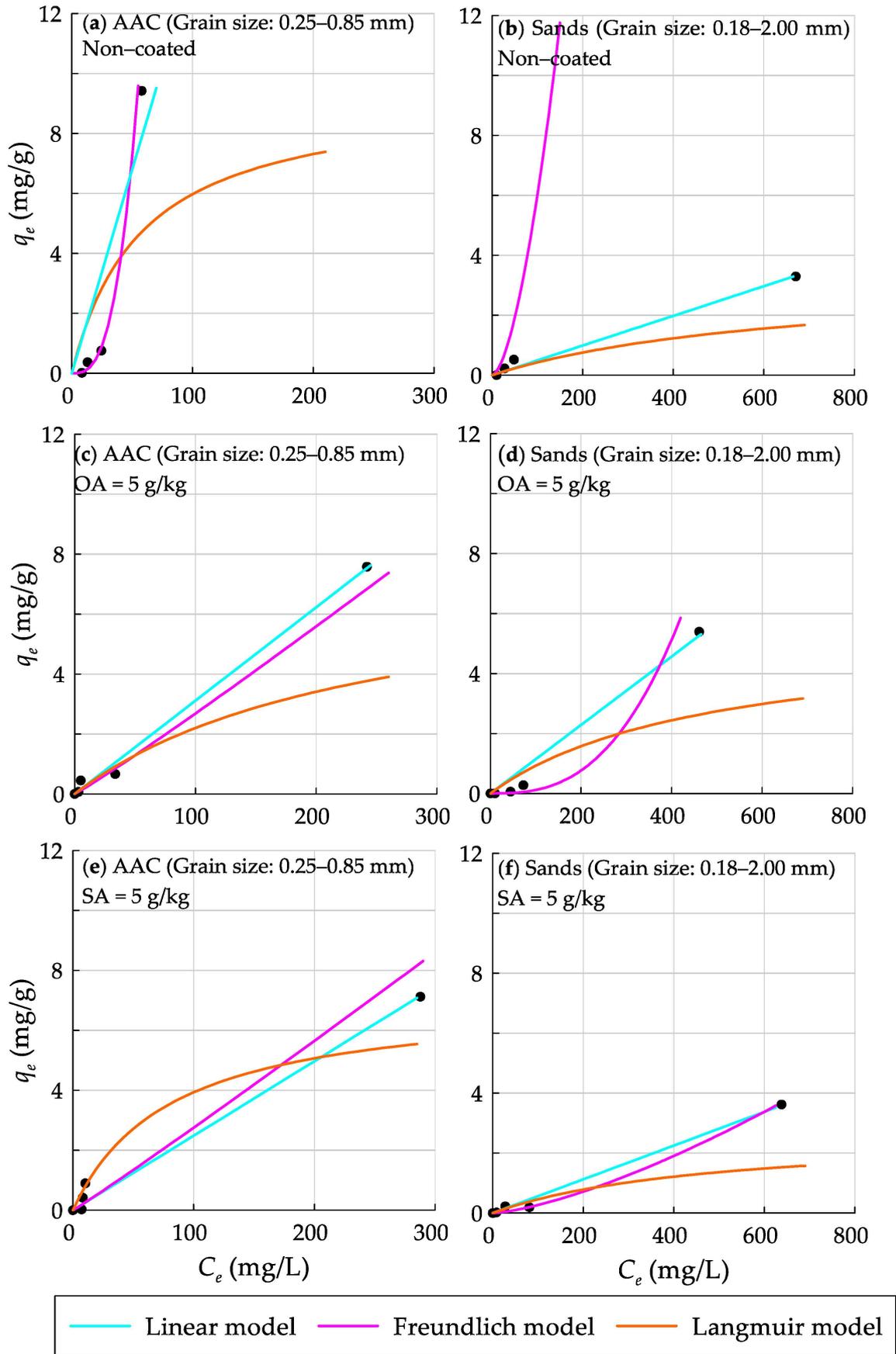


Figure 7. Relationship between q_e and C_e for AAC grains and Sands.

Table 1. Measured and reported parameters for adsorption isotherm models.

Sample	Particle Size (mm)	Coating (g/kg)	Langmuir			Freundlich			Linear		Target Oil	Ref.
			q_{max} (mg/g)	b (L/mg)	R^2	$1/n$	K_F (mg/g)	R^2	K_d (L/mg)	R^2		
AAC	0.106–0.25	0	8.6	7.3×10^{-3}	0.97	1.7	3.0×10^{-3}	0.99	6.1×10^{-2}	0.99	Soybean oil *	This Study
	0.25–0.85		9.4	1.7×10^{-2}	0.86	3.0	6.3×10^{-5}	0.99	1.4×10^{-1}	0.94		
	0.85–2.00		8.9	8.9×10^{-3}	0.99	1.5	8.6×10^{-3}	0.99	7.8×10^{-2}	0.99		
	0.106–0.25	OA = 1	9.0	1.0×10^{-2}	0.98	0.92	1.2×10^{-1}	0.99	9.1×10^{-2}	0.99		
	0.25–0.85		6.6	2.9×10^{-3}	0.96	0.70	8.9×10^{-2}	0.98	1.9×10^{-2}	0.99		
	0.85–2.00		8.5	6.7×10^{-3}	0.85	1.0	4.0×10^{-2}	0.99	5.6×10^{-2}	0.99		
	0.106–0.25	OA = 5	8.9	8.7×10^{-3}	0.81	1.3	2.6×10^{-2}	0.99	7.7×10^{-2}	0.99		
	0.25–0.85		7.6	4.1×10^{-3}	0.93	1.1	2.1×10^{-2}	0.99	3.1×10^{-2}	0.99		
	0.85–2.00		7.3	3.7×10^{-3}	0.94	1.4	3.6×10^{-3}	0.99	2.7×10^{-2}	0.99		
	0.106–0.25	OA = 10	9.1	8.5×10^{-3}	0.82	3.0	8.3×10^{-6}	0.99	1.0×10^{-1}	0.99		
	0.25–0.85		6.9	3.0×10^{-3}	0.96	2.1	4.2×10^{-5}	0.99	2.3×10^{-2}	0.99		
	0.85–2.00		6.7	2.8×10^{-3}	0.96	1.6	7.8×10^{-4}	0.99	2.0×10^{-2}	0.99		
	0.106–0.25	SA = 1	7.8	4.5×10^{-3}	0.92	0.89	6.9×10^{-2}	0.99	3.5×10^{-2}	0.99		
	0.25–0.85		7.7	4.4×10^{-3}	0.93	0.73	1.4×10^{-1}	0.99	3.4×10^{-2}	0.99		
	0.85–2.00		6.3	6.2×10^{-2}	0.39	0.60	1.8×10^{-1}	0.99	1.7×10^{-2}	0.99		
	0.106–0.25	SA = 5	8.6	6.9×10^{-3}	0.86	1.6	4.5×10^{-3}	0.99	6.0×10^{-2}	0.99		
	0.25–0.85		7.1	1.2×10^{-2}	0.75	1.0	2.3×10^{-2}	0.99	2.5×10^{-2}	0.99		
	0.85–2.00		7.8	4.6×10^{-3}	0.74	1.2	2.0×10^{-2}	0.99	3.6×10^{-2}	0.99		
0.106–0.25	SA = 10	8.4	6.3×10^{-3}	0.87	1.2	2.8×10^{-2}	0.99	5.3×10^{-2}	0.99			
0.25–0.85		8.3	1.3×10^{-2}	0.73	0.95	7.6×10^{-2}	0.99	4.8×10^{-2}	0.99			
0.85–2.00		9.0	1.0×10^{-2}	0.78	1.3	2.3×10^{-2}	0.99	9.0×10^{-2}	0.99			
Sands	0.18–2.00	0	3.3	1.5×10^{-3}	0.99	1.5	2.0×10^{-2}	0.99	4.9×10^{-3}	0.99	Soybean oil *	This Study
	0.30–2.00		5.2	2.1×10^{-3}	0.97	1.7	5.6×10^{-3}	0.99	1.1×10^{-2}	0.99		
	0.18–2.00	OA = 2	4.1	1.7×10^{-3}	0.96	1.8	1.8×10^{-5}	0.99	6.8×10^{-3}	0.98		
	0.30–2.00	OA = 5	4.7	1.8×10^{-3}	0.98	2.0	3.3×10^{-5}	0.99	8.7×10^{-3}	0.99		
	0.18–2.00	OA = 2	5.4	2.1×10^{-3}	0.96	2.8	3.7×10^{-7}	0.99	1.1×10^{-2}	0.99		
	0.30–2.00	OA = 5	3.9	1.6×10^{-3}	0.98	2.2	5.9×10^{-6}	0.99	6.3×10^{-3}	0.99		

Table 1. Cont.

Sample	Particle Size (mm)	Coating (g/kg)	Langmuir			Freundlich			Linear		Target Oil	Ref.
			q_{\max} (mg/g)	b (L/mg)	R^2	$1/n$	K_F (mg/g)	R^2	K_d (L/mg)	R^2		
	0.18–2.00	OA = 2	5.3	2.1×10^{-3}	0.96	2.1	1.2×10^{-5}	0.99	1.1×10^{-2}	0.99		
	0.30–2.00	OA = 5	5.4	2.2×10^{-3}	0.96	1.4	6.7×10^{-4}	0.99	1.1×10^{-2}	0.99		
	0.18–2.00	SA = 2	3.6	1.5×10^{-3}	0.98	1.4	4.0×10^{-4}	0.99	5.6×10^{-3}	0.99		
	0.30–2.00	SA = 5	4.0	1.7×10^{-3}	0.99	1.3	1.4×10^{-3}	0.98	6.7×10^{-3}	0.99		
Graphite powders	<0.03	Activated carbons	25	3.9×10^{-3}	0.9	0.82	0.16	0.77	–	–	Diesel oil *	Huang et al. (2018) [62]
Activated carbons	0.5–2.0	0	16–86	7.0×10^{-3} -1.0×10^{-2}	0.94–0.99	0.18–0.5	0.4–5.0	0.93–0.99	–	–	Vegetable oil	Gong et al. (2007) [34]
Wakame	–	Biochar /Ni	8.65–116.5	0.113–0.481	0.964–0.999	0.03–0.1	3.45–7.21	0.806–0.965	–	–	Diesel oil	Jing et al. (2022) [63]

* Dispersed or Emulsified oil.

It can be seen that the Langmuir model did not capture the measured relationship between C_e , q_e , and q_{\max} values were given (shown in Figure 7), indicating that the sorption of dispersed oil onto AAC grains did not follow the basic adsorption concept of dissolved ions (i.e., a saturation of adsorption site) and surface chemical reactions (e.g., formation of surface complexation). Both the Freundlich and linear models, on the other hand, captured the C_e and q_e relationships well. However, the Freundlich model did not capture the measured relationships of SA-coated AAC grains with 5 g/kg in this study (e.g., Figure 7e). Thus, the simple linear model performed the best among the models (R^2 values became mostly 0.99), and the fitted K_d values became a good indicator to characterize the sorption ability of dispersed oil in water onto both non-coated and coated AAC grains with hydrophobic/oleophilic agents. In addition, it is interesting that non-coated AAC grains 0.25–0.85 mm gave the highest value of 0.136 among fitted K_d values (Table 1). This may support choosing a suitable grain size for treating oily wastewater because we did not observe a clear difference in measured R values among three grain size samples (Figure 5). For reference, the measured relationship between q_e to C_e for sands is shown in Figure 7b,d,f. It can be seen that only the linear model captured the relationships well.

Some previous studies examined the applicability of oil sorption onto adsorbents based on adsorption isotherm models (Langmuir and Freundlich) and reported the adsorption parameters [34,62,63] that are given in Table 1. Those studies showed good applicability of Langmuir and Freundlich models and determined the parameters such as q_{\max} and K_F . In this study, on the other hand, those models that assume monolayer and/or multilayer adsorption of dissolved ions onto the surface of the adsorbent were not applicable to measured data. This strongly implies the adsorption mechanisms of dispersed oil onto porous AAC grains were more complicated, probably due to the accessibility of dispersed oil (<2 μm) to micro-scale pores of AAC grains and the affinity of dispersed oil to cementitious AAC grain surface. Moreover, the inapplicability of adsorption isotherm models (Langmuir and Freundlich models in this study might be attributed to the emulsification of dispersed oil in water, and the oil emulsion made the adsorption mechanisms more complicated.

4. Conclusions

The study examined the applicability of hydrophobized/oleophilized AAC grains to treat dispersed oil in water. Based on the tested results from batch experiments, low-coated AAC grains with OA and SA (1 g/kg) removed from dispersed oil well (approximately $R > 80\%$) for low oil concentration solutions, and high SA-coated AAC grains (10 g/kg) gave high removal efficiency (approximately $R > 80\%$) for high oil concentration solutions. Typical adsorption isotherm models for dissolved ions, such as Langmuir, Freundlich, and a linear model, were adapted to measured data. It was observed that the sorbed dispersed oil increased linearly with increasing equilibrium concentration for all tested samples, while the Langmuir and Freundlich models did not capture the measured isotherms. This implies that the sorption mechanism of dispersed oil onto porous AAC grains was supposed to be complicated due to the accessibility and affinity of dispersed oil (<2 μm) to micropores of cementitious AAC grain surface.

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Abbreviations

The following abbreviations are used in this manuscript:

AAC	Autoclaved aerated concrete
C_e	Equilibrium concentration of dispersed oil in water
C_i	Initial concentration of dispersed oil in water
HA	Hydrophobic agents
OA	Oleic acid
q_e	Oil sorption amount at equilibrium (mg/g)
R	Removal percentage (%) of dispersed oil in water
SA	Stearic acid

Appendix A

Table A1 summarizes National standards of emission limits for oil and grease, and Table A2 shows some reported values of oil and grease concentrations in water and wastewater in Vietnam. Among surface waters, the Ha Thanh River has a very high measured value of 250 mg/L. The source of pollution in this area is not shown. For coastal water, the maximum was 1.3 mg/L, and all values were below 1.0 mg/L. Some domestic wastewater showed high values of up to 140 mg/L. Regarding industrial wastewater, the landfill leachate showed a very high value of 3.79×10^4 mg/L, causing environmental problems [64–72].

Table A3 shows reported values of oil pollution loads and oil spills in Vietnam. Oil leaks from crude oil exploitation. A total of 9.2×10^4 tons is estimated to flow into the coastal and marine environment. This is the high environmental impact of the oil spill, with oil pollution loads approaching 7.72×10^5 tons/year, which significantly impacts the environment and the economy. Total financial losses caused by a significant oil spill in 2001 were estimated at 250 billion VND (17 million USD), while costs for cleaning up polluted waters and beaches reached 60 billion VND (4 million USD). The oil pollution load of the river has a value of 3.45×10^4 kg/day in the southern Ho Chi Minh City area. The northern Cau River basin and Cau Bay basin had values of 66 tons/day and 6.34 kg/day. This is less than that in the southern part of the country. The source of these contaminants appears to be domestic wastewater [4,64,65,73–75].

Table A4 shows various information on wastewater in Vietnam, such as BOD and COD [24,64,72,73,76–78]. Since no concentration and river flow data were available for BOD and COD, it is difficult to say for sure, but it can be said that they are causing significant pollution loads. Concentrations of BOD and COD after sewage treatment were also measured at the sewage treatment plant in Hanoi in this study, with BOD = 4.0 mg/L and COD = 8.0 mg/L. When n-hexane extract (mineral oils) and the concentration of n-hexane extract (animal and vegetable oils) were also measured, no oil concentration was detected. Further, according to Do Nam Thang, the percentage of the waste is higher in domestic wastewater than in industrial wastewater [77], and it can be said that there are issues with their treatment.

Table A1. National technical regulations of oil and grease concentrations in Vietnam.

Act Name	Parameter	Environmental Standards (mg/L)				Analysis Method
Environmental standards: Surface water, groundwater, seawater, and others						
QCVN 08–MT:2015/BTNMT [7]	Total oil and grease	Domestic use		Irrigation		TCVN 7875: 2008 ⁵ [8] SMEWW 5520B: 2012 [9]
		A1 ¹ 0.3	A2 ¹ 0.5	B1 ¹ 1.0	B2 ¹ 1.0	
QCVN 10–MT:2015/BTNMT [10]	Total mineral oil and grease	Area of aquaculture and conservation 0.5		Beach and water sport area 0.5	Other areas 0.5	TCVN 7875: 2008 ⁵ [8] SMEWW 5520B: 2012 [9] SMEWW 5520C: 2012 [11]
QCVN 38:2011/BTNMT [12]	Total mineral oil and grease	0.05				
Emission limits: Domestic and industrial wastewater						
QCVN 14:2008/BTNMT [13]	Animal and vegetable oil and grease	A ² 10		B ² 20		EPA Method 1664: 2010 [14]
QCVN 40:2011/BTNMT [15]	Total mineral oil and grease	A ² 5		B ² 10		TCVN 7875: 2008 ⁵ [8] EPA Method 1664: 2010 [14]
Emission limits: Specific wastewater from industries and service sectors						
QCVN 14:2008/BTNMT [13]	Animal and vegetable oil and grease	A ³ 10		B ³ 20		EPA Method 1664: 2010 ⁵ [14]
QCVN 29:2010/BTNMT [16]	Mineral oil (Total hydrocarbon)	A ⁴ 5	Warehouse 15	B ⁴ Car wash 18	Stations 30	TCVN 7875: 2008 ⁵ [8]
QCVN 52:2013/BTNMT [17]	Total mineral oil	A ³ 5		B ³ 10		TCVN 7875: 2008 ⁵ [8] SMEWW 5520C: 2012 [11]
QCVN 11–MT:2015/BTNMT [18]	Total animal and vegetable oil	A ³ 10		B ³ 20		TCVN 7875: 2008 ⁵ [8] SMEWW 5520C: 2012 [11]

¹ A1: Good for domestic uses and other purposes (A2, B1, and B2); A2: Good for domestic uses with prior suitable treatment, protection of aquatic habitats, and other purposes (B1 and B2); B1: Good for irrigation and agricultural uses; B2: Others (water transport and low priority of water treatment). ² A: Effluents reach water sources used for domestic purposes; B: Effluents reach water sources used for other purposes. ³ A: Effluents reach water sources used for drinking purposes; B: Effluents reach water sources used for other purposes. ⁴ A: Effluents reach water sources used for aquaculture and aquatic conservation; B: Effluents reach water sources used for other purposes. ⁵ Analysis methods with equivalent or higher accuracy than those of the listed standards can be accepted. N/D: Not detected.

Table A2. Reported values of oil and grease content in water and wastewater in Vietnam.

Location	Province/ Prefecture or City	Year	Oil and Grease (mg/L)	Remarks	Reference
Surface water					
Cau River	Thai Nguyen/Thai Nguyen	2004, 2005	0.02–0.16	6 monitoring points	MONRE, WB and DANIDA (2006) [64]
Dong Nai River	Hoa An/Cao Bang	2000–2006	0.025–0.029		
Saigon River	Binh Phuoc/Binh Duong, etc.	2000–2006	0.025–0.12		
Saigon–Nha Be River	Thu Dau Mot/Binh Duong, etc.	N/D	0.0–0.3	5 monitoring points	IGES (2007) [65]
Ha Thanh River	Quy Nhon/Binh Dinh	2016	27–250	Ha Thanh Bridge, Canal, Channel	Binh Dinh PPC (2016) [66]
Ditch	Quy Nhon/Binh Dinh	2019	15	An upstream ditch of Bau Sen Lake	Nguyen et al. (2020) [67]
Coastal water					
Red and Mekong River		1996–2001	>0.05–2.4		MONRE (2003) [4]
Coastal zone (1)	Rach Gia/Kien Giang:	1997–2015	0.12–0.79	Rach Gia monitoring station	Le and Pham (2017) [68]
Coastal zone (2)	Nha Trang/Khanh Hoa	2013–2017	0.35–0.55	Nha Trang monitoring station	Pham (2018) [69]
Coastal zone (3)	Khanh Hoa/Trung binh, etc.	2010–2018	0.007–0.65	4 monitoring stations	Pham (2019) [70]
Coastal zones	Hue/Thua Thien Hue, etc.	2011–2015	0.012–1.3	Total of 16 monitoring stations	MONRE (2015) [71]
Domestic wastewater					
In and Out of WWTP	Quy Nhon/Binh Dinh	2016	0.8–160	Nearby Dam, Nhon Binh, Bau Luc	Binh Dinh PPC (2016) [66]
In and Out of WTF	Hai Ba Trung/Hanoi	2016	2.0–14.6	WTF for condominium in Hanoi	JICA (2019) [72]
Industrial wastewater					
Landfill leachate	Tuy Phuoc/Binh Dinh	2016	3.79×10^4	Long My waste landfill	Binh Dinh PPC (2016) [66]
Wastewater from construction site	Quy Nhon/Binh Dinh	2016	200	Cleaning and washing of equipment and materials at a bridge construction site	

Table A3. Reported values of oil pollution loads and oil spills in Vietnam.

Location	Year	Type and Sources	Oil Pollution Loads/Oil Spills	Descriptions and Other Remarks	Reference
East sea	1995–2002	Oil spills	Approx. 7.72×10^5 tons/year	Oil leaks from crude oil exploitation. A total of 9.2×10^4 tons is estimated to flow into the coastal and marine environment. Total financial losses caused by a major oil spill in 2001 were estimated at 250 billion VND (17 million USD), while costs for cleaning up polluted waters and beaches reached 60 billion VND (4 million USD).	ADB (2006) [73]; MONRE, WB and DANIDA (2003) [4]
Dong Nai River basin	2004	Domestic wastewater: HCMC 5 provinces	3.45×10^4 kg/day $90\text{--}4.1 \times 10^3$ kg/day		MONRE, WB and DANIDA (2006) [64]
Cau River basin	2005	Domestic wastewater from 6 provinces	66 tons/day		
Cau Bay River basin	2013	48 enterprises at Long Bien District, Hanoi	6.34 kg/day		JICA (2013) [74]
HCMC	2000	Industrial wastewater		Major industrial contributors are oil refining, chemical, and food processing industries (20–30% of the total industrial wastewater discharges into river systems in Vietnam from HCMC)	ADB (2000) [75]

Table A4. Information on other pollution loads in Vietnam.

Location	Year	Type and Sources	Oil Pollution Loads/Oil Spills	Descriptions and Other Remarks	Reference
Craft village	2002	Tra Co cassava processing village Bao Loc silk village	(BOD ₅ kg/day, COD kg/day) 218–379, 145–544	Pollution loads of SS, TN, TP, CN andNH ⁴⁺	MONRE, WB and DANIDA (2006) [64]
Whole country	2003	(Volume of wastewater) Total	(m ³ /year) 896 × 10 ⁶		ADB, GEF, UNEP (2006) [73]; WWF (2018) [76]
Whole country: Industrial zones	2005	(Load of pollutants) HCMC, Dong Nai, Binh Duong, BR–VT	(BOD ₅ kg/day, COD kg/day) 2.24 × 10 ² –1.28 × 10 ⁴ , 1.06 × 10 ³ –4.68 × 10 ⁴	Pollution loads of TSS, TN, TP, and wastewater volume	MONRE, WB and DANIDA (2006) [64]
Hanoi	2005	(Wastewater discharge from industrial sectors) Chemical factories, Textile and dyeing, Foodstuff companies, Mechanical factories	(m ³ /day) 3.73 × 10 ³ –2.65 × 10 ⁴		MONRE, WB and DANIDA (2006) [64]
Food processing villages	2005	(Wastewater discharge) Phu Do rice noodle, Vu Hai rice noodle, Ninh Hong rice noodle, Tan Do wine, etc.	(BOD ₅ tons/year, COD tons/year) 10–53, 15–1.3 × 10 ⁴		MONRE (2008) [24]
Red River Delta	2006	(Wastewater discharge) Food processing, husbandry, slaughtering Weaving, dyeing, leather processing, etc.	(COD tons/year) ~1.5 × 10 ⁴ ~1.5 × 10 ³		MONRE (2008) [24]
Nhue–Day River	2006	(Proportion of wastewater discharge) Domestic, Industrial, Farming and husbandry, etc.	4–56%		MONRE, WB and DANIDA (2006) [64]
Hanoi, Hai Phong HCMC	2009	(Wastewater discharge) Domestic wastewater, Industrial wastewater, etc.	(BOD ₅ tons/day) 0.3–214		Nguyen et al. (2020) [67]
Hanoi, Hai Phong Da Nang, HCMC	2010	Share of pollution load on the sum of domestic and industrial wastewater. Domestic wastewater and Industrial wastewater	15–85%	1	Do and Nguyen (2014) [77]
Hanoi, Hai Phong Da Nang, HCMC	2010 2019	Wastewater discharge into canals from agricultural activities Domestic wastewater discharge rate and amount	(BOD ₅ tons/day, COD tons/day) 17–193, 26–305	Pollution loads of SS, TSS are also given.	VEA (2012) [78]
Whole country	2019	Domestic wastewater discharge rate and amount. Upper: Urban area Lower: Rural area	150 L/capita/day (4.66 × 10 ⁶ m ³ /day) 80 L/capital/day (4.85 × 10 ⁶ m ³ /day)	Blackwater 93–94%, Graywater 6–7% (Hanoi, Hai Phong)	JICA (2019) [72]

¹ 90% of domestic wastewater not treated. The capacities of wastewater treatment plants in big cities only meet about 30% of the demand.

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