

Produced Water Treatment by Adsorption: A Review

*Ataa Ali, **Nawras Jassim, #M.N. Faris

*College of Engineering / Department of Chemical Engineering / Basrah University / Basrah, Iraq

ORCID: <https://orcid.org/0000-0003-0030-1164>

**Basra Engineering Technical College/ Department of Chemical and Petrochemical Techniques
Engineering/Southern Technical University/ Basra/ Iraq

#College of Engineering /Department of Chemical Engineering/ Basrah University/ Basrah/ Iraq

ORCID: <https://orcid.org/0000-0003-3089-7026>

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INTRODUCTION

Oily wastewater currently poses a serious hazard to human life and generates serious ecological and environmental issues. The oil industry and oil production are the main sources of oily wastewater [1]. Treatment of oily wastewater is therefore essential to minimize its impact on the environment and people. Oil recovery from oily wastewater treatment could also have a positive economic impact [2]. Adsorption is a phenomenon that occurs at the interface between a gas or solute and a solid adsorbent, where the gas or solute components are collected on the surface of the solid. The behavior in question is linked to the attractive forces between gaseous and solute molecules, which are often referred to as Van der Waals forces. Adsorption is a physical phenomenon where chemicals adhere to a solid surface without undergoing any chemical reactions. The material that absorbs substances is called the adsorbent, whereas the chemical being absorbed from the liquid or gas phase is referred to as the solute. The adsorptive capacity and kinetics are influenced by the kind of activated carbon, particle size, pore size, and pore size distribution [3]. This chapter provides a brief overview of previous investigations and ongoing research.

PRODUCED WATER AND ITS TREATMENT

Oil is considered a valuable commodity due to its significance as a crucial worldwide energy source and raw material for life. Hence, the implementation of an effective system is crucial for the retrieval of spilled or residual oil. Oil removal and recovery technologies are categorized as physical, mechanical, biological, photochemical recovery, filtering, and the widely employed method is adsorption. These technologies are utilized either independently or simultaneously with one another. Table 1 presents the benefits and constraints of each treatment approach [4].

Table 1: Methods for Treating Oily Wastewater [4].

Methods	Advantages	Disadvantage	Reference
Adsorption	Highly effective in removing oil Straightforward procedure Cost-effective processing	Requires a significant amount of manual labor. Inadequate elimination of small emulsions measuring less than 100 μm .	[5,6]

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Coagulation & flocculation	High efficacy in removing oil	Expensive upfront and ongoing expenses- Highly labor-intensive process that results in the production of secondary pollutants.	[7,8,9]
Electrocoagulation	Superior oil elimination effectiveness Cost-effective operation	High initial cost	[10,11]
Flotation	High oil removal efficiency	Intricate procedure (including the use of coagulants and flocculants) Inadequate elimination of small oil droplets measuring less than 50 μm . Expensive operational expenses	[12,13,14,15]
Coalescences (depth filtration)	Highly effective oil removal efficiency Straightforward procedure Cost-effective initial and ongoing expenses	Oil removal efficiency highly depends on the quality of filter bed media Slow oil removal process	[16,17,18]
Membrane filtration	Superior effectiveness in removing grease	Must require prior processing of wastewater Significant upfront and subsequent costs Early onset of membrane fouling	[19,20,21]
Biological treatment	Highly effective oil removal efficiency Cost-effective operation	Extremely temperature and pH sensitive Requires skilled operator Requires wide space	[22]

CATEGORIES OF OIL AND GREASE

Oil and grease compounds can be classified into two distinct categories, characterized by their origin and chemical composition. The first category of substances originates from minerals, specifically petroleum and its byproducts. It is a combination of hydrocarbons with varying chemical structures. The second category comes from living organisms, either animals or plants, and is primarily made up of triglycerides, which are esters formed from glycerin and fatty acids [24]. The presence of hydrocarbons, including benzene, toluene, ethyl benzene, xylenes (BTEX), and polyaromatic hydrocarbons (PAHs), in oily wastewater is a significant cause for Attention. Petroleum refineries and petrochemical businesses generate significant quantities of pollutants, such as petroleum hydrocarbons, derived from many sources including crude oil storage areas, washing sections, and vessel cleaning sections. If this wastewater is not discharged and treated appropriately, it can have a substantial impact on the environment. Therefore, it is essential to conduct a comprehensive examination and analysis of the properties of oily wastewater prior to initiating any treatment processes [25].

Table 2: Typical Characteristics of oil and grease bearing wastewater [25].

Sources of oily wastewater	Total amount generated globally	Available forms of oil	Typical concentration of oil and grease, mg/L	Concentration of allied major constituents (in mg/L except pH)	References
Petroleum industry-Produced	39.746*10 ⁹ L/d	Dissolved, Free and suspended form, emulsions	2-565	TSS (1.2-1000), pH (4.3-10), COD =1220, TOC (0-1500), Volatile (BTX)- (0.39-35), Nonvolatile oil and grease (0.275), HCO ⁻³ (77-3990), Cl ⁻ (80-2,00,000), SO ₃ ⁻² (10), SO ₄ ⁻² < (2-1650), VFA (2-4900), Ammonia-N (10-300), phenols (0.009-23)	[26,27]
Petroleum industry-wastewater from refining process	5.342*10 ⁹ L/d	Exist in 4 forms (free, dispersed, emulsified, and dissolved oil)	20-4000	COD (300-600), Phenol (20-200), Benzene (1-100), Lead (0.2-10), Chromium (0.1-100)	[29,30,31]
Bilge water	0.5-50*10 ³ L/ day/ boat	Emulsion	Not Available	Not Available	[32,33]
Edible oil refining	Not Available	Not Available	4000-6000	Not Available	[29]
Leather processing (Tannery effluent)	Not Available	Not Available	200-40,000	Chloride (5000), COD (1500-2500), Settleable substances (10000-2000)	[29,30]
Pharmaceutical industry	Not Available	Not Available	Not Available	COD (5000-15000)	[34]
Metal processing and finishing	Not Available	Not Available	100-20,000	Not Available	[29]
Bio-diesel production process wastewater	Not Available	Emulsion	7000-44,330 mg/L	COD (60,000-545,000), pH (8.5-10.5), BOD (105,000) 300,000, SS (1,500-28,790)	[28]

TREATMENT OF OILY WATER BY ADSORPTION

Adsorption is a prevalent technique employed for the remediation of wastewater. Oil molecules undergo adsorption by adhering to the surface of a solid adsorbent Upon getting into contact. The adsorbent is crucial in the process of adsorption because different adsorbents can engage distinct adsorption mechanisms, resulting in variations in adsorption efficiency. In general, adsorbents can be categorized into two categories: natural adsorbents and synthesis adsorbents: natural adsorbents, which include earth crust and bio-adsorbents, and non-natural adsorbents, which encompass laboratory-synthesized adsorbents and commercial adsorbents. The Figure (1) provides an overview of

the classification of adsorbents [23].

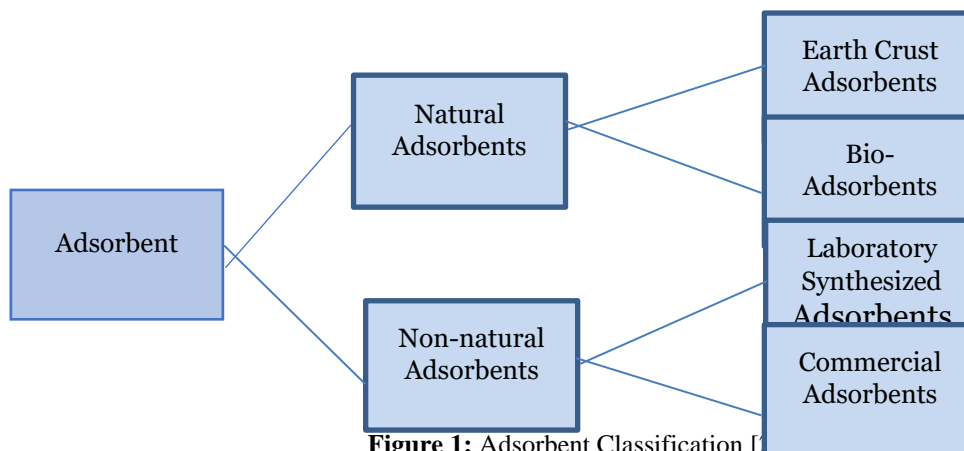


Figure 1: Adsorbent Classification [23]

Physical Adsorption

Due to the interatomic connections that connect these atoms to neighboring atoms within the same substance. These surfaces facilitate adsorption by natural attraction forces or van der Waals forces. Under specific pressure and temperature circumstances, this type of adsorption can lead to the formation of several layers of the adsorbent material on its surface. Such adsorption phenomena can occur at low temperatures, resembling the condensation of vapors on liquid surfaces, as the activation energy involved is hypothesized to be below 40 kJ/mol [35].

Chemical Adsorption

Adsorption of this nature takes place on surfaces that are not electrically unsaturated. These surfaces have a tendency to establish chemical interactions with the atoms or molecules that have undergone adsorption. In the first stage of the chemical reaction between the adsorbent surface and the adsorbent material, this particular form of adsorption requires a significant amount of activation energy. Additionally, the temperatures involved are high, exceeding 40 kJ/mol.

This particular adsorption process is very selective and irreversible, and its effectiveness is constrained by the presence of oxygen layers on the coal surface.

Adsorption of hydrogen chloride on the surface of iron [5].

Activated Carbon (AC) As Adsorbent

The effectiveness of activated carbon in adsorbing pollutants from air and water, as well as its utility as an energy storage device and catalytic support, can be attributed to its very large internal specific surface area and well-developed porous structure. Activated carbon is mostly utilized in water treatment, with an estimated annual usage of approximately 350,000 tones, as reported [48]. AC, in its different forms (powder, granular, or brittle), has been utilized in the treatment of both industrial and municipal wastewater due to its ability to adsorb a diverse array of organic and inorganic compounds on non-volatile substances [3].

OILY WASTEWATER ADSORPTION

Carlo et al. (2002) studied Adsorption experiments were conducted on two distinct depleted oils to mitigate their pollution and health risks. Two efficient adsorbents were prepared using an inexpensive recovery material: (a) a combination of calcium and magnesium oxide produced by heating dolomite at 1800°C, and (b) an activated material

created by subjecting this product to chemical treatment with hydrochloric acid. The findings presented and analyzed in this study indicate that the byproducts of dolomite calcination can effectively substitute traditional adsorbent materials in the elimination of organic pollutants, specifically in the case of spent soluble oils that are typically not recyclable. This substitution can lead to a reduction in the operational expenses associated with their treatment [54].

Ehsan et al. (2022) studied three alternative synthesis techniques were used to create magnetic powder and granular activated carbons (MPAC and MGAC) as recoverable adsorbents to extract polycyclic aromatic hydrocarbons (PAHs) from aqueous solutions. The result of the removal percentages ranging from 87.2% to 99.3%, the manufactured magnetic ACs were particularly efficient in removing PAH chemicals from aqueous solution. The maximum PAHs removal efficiency (99.3%) came from the precipitation technique of magnetization using PAC as the base AC, whereas the highest PAHs removal efficiency (98.3%) came from the coprecipitation method of magnetization using GAC as the base AC. According to the study, an increase in the amount of PAH rings would result in more covalent connections forming between the adsorbent and the adsorbate [37].

MORUWOU et al. (2022) Investigators looked at the coconut shell activated carbon's ability to absorb total hydrocarbons from produced water. Activated carbon's ability to adsorb total hydrocarbons was analyzed and optimized using the response surface approach. Adsorption process factors' impacts on this capacity were taken into account. Adsorbent dose of 0.4 to 1.0 g, contact duration of 20 to 60 min, and temperature of 30 to 50°C are the adsorption process factors taken into account, along with a range of values for each. Produced water had a high proportion of pollutants, including hydrocarbons, according to the physicochemical characteristics of the water before it was treated. The highest concentration of THC that could be adsorbed throughout the procedure was shown to be 1,068,451.73 mg/g. This maximum was attained using an adsorbent dose of 0.40 g, a 60-minute contact period, and a 30°C temperature. The significant quantity of THC adsorption demonstrated the effectiveness of coconut shell activated carbon for the treatment of wastewater streams containing hydrocarbons [38].

Monica et al. (2022) studied the effective synthesis of an activated carbon-Fe₃O₄ composite. In this research, magnetic activated carbon was created synthetically, and its effectiveness as a gasoline adsorbent was examined. The adsorption capacity of AC-M for gasoline adsorption was 1252.22 mg/g for adsorption period of 4 min and mass of adsorbent 0.09 g and followed the adsorption kinetic pseudo-second-order model from Ho and McKay with adsorption rate constant 0.0012 g/mg/min [39].

Rajak et al., 2018 The article described the chemical activation technique used to create charcoal from sawdust using multiple chemical agents, including phosphoric acid, zinc chloride, and ferrous sulfate heptahydrate. The ability of several charcoal samples to adsorb oil from an oil-in-water emulsion was studied. Investigations have been done into the effects of various factors, such as adsorbent dose, contact duration, and temperature, on the adsorption of oil from oil-in-water emulsions by synthetic charcoals. Although it also relies on the starting oil content in the emulsion, under ideal conditions, oil separation efficiency is greater than 98%. The adsorption of oil from an oil-in-water emulsion has also been examined kinetically and isothermally [40].

Eman et al. (2021) Adsorption studies (continuous fixed-bed column) were performed using researched activated carbon, zeolite, silica gel, and mixed adsorbents (activated carbon and silica gel) to remove oil and chemical oxygen demand (COD) from generated water. The findings show that the Thomas kinetic model was appropriate for the adsorption of COD and oil on all different types of adsorbents. Additionally, a drop-in flow rate and concentration led to an increase in the elimination of contaminants. With a flow rate of 1.25 mL/min, an adsorbent dose of 0.5 g, an oil concentration of 40 ppm, and a bed height of 2 cm, mixed adsorbents achieved the highest removal of oil and COD (83.62% and 78.81%), followed by powdered activated carbon (72.98% and 69.5%), silica gel (67.8% and 64.74%), granular activated carbon (64.87% and 60) [41].

Eman et al. (2021) examined the treatment of produced water (PW) using powdered activated carbon (PAC), clinoptilolite natural zeolite (CNZ), and synthetic zeolite type X (XSZ), which results in the adsorption of chemical oxygen demand (COD), oil, and turbidity. Adsorption was carried out in a batch adsorption setup. To determine the ideal working parameters, the effects of adsorbent dose, duration, pH, oil concentration, and temperature were researched. Maximum oil removal efficiencies (99.57, 95.87, and 99.84 percent), COD, and total petroleum hydrocarbon (TPH) were identified at a PAC adsorbent dosage of 0.25 g/100 ml. Furthermore, the maximum turbidity removal efficiency (99.97%) was attained when zeolite X was applied at a concentration of 0.25 g/100 ml. It is comparable to the 99.65% you would receive from PAC. The results shown that adsorption on PAC is the most

effective technique for eliminating organic pollutants from PW when compared to zeolites [42].

Yuanyuan et al. (2020) studied the primary mechanisms for effectively removing emulsified oils (EOs) in produced water are electrostatic adsorption and hydrophobicity. In this study, sepiolite (Sep) is subjected to modification using two types of organic cation surfactants: stearyl trimethyl ammonium bromide (STAB) and dimethyl dioctadecyl ammonium bromide (DDAB). The highest adsorption capacities were 957.0 mg/g for STAB-Sep and 1031.5 mg/g for DDAB-Sep, indicating that the modified Sep with double chains had a strong attraction to EOs. Both the Langmuir and Freundlich isotherms are applicable for elucidating the adsorption phenomenon. The thermodynamic characteristics provided evidence that the adsorption process was a spontaneous and endothermic physical phenomenon. This study establishes a theoretical basis for the development of an organoclay adsorbent that is both highly efficient and cost-effective [49].

Khaled et al. (2011) investigated the process of extracting oil from oil-water emulsions by the adsorption method using bentonite, powdered activated carbon (PAC), and deposited carbon (DC). The results provided proof of the adsorbents' capacity to adsorb oil, and indicated that the adsorptive characteristics of the three adsorbents (bentonite, PAC, and DC) were influenced by various circumstances. The Freundlich isotherm accurately matches the removal of oil by bentonite, PAC, and DC, with DC and bentonite having greater adsorptive capabilities than PAC [50].

M. Sharafimasoooleh et al. (2011) Modified Nano clay was created by substituting CTAB with inorganic/metal ions/cations in montmorillonite structure. The adsorption capacity of the sorbent materials was tested using crude oil, kerosene, petrol, and toluene. Results showed a higher adsorption capacity than unmodified clay, with the CHN study verifying these findings [51].

Albatrni et al. (2019) examined the application of four artificial resins to eliminate emulsified oil from generated water. The experimental study assessed crucial parameters like adsorbent dosage, contact time, beginning oil content, and pH for Optipore L493, Amberlite IRA 958, Amberlite XAD 7, and Lewatit AF 5. AF 5, XAD 7, and L493 were able to obtain oil removal rates above 98%. IRA 958 observed removal rates that were significantly low, measuring less than 25%. The isotherm data were analyzed and modelled using the Langmuir, Freundlich, Toth, Flory Huggins, and Dubinin-Radushkevich models. The findings indicate that the process of adsorption onto XAD 7 and L 493 involves many layers forming on a surface that is not uniform. This phenomenon can be well described by the Freundlich model for XAD 7 and the Toth model for L 493 [52].

M. Fathy et al. (2018) studied a unique technique is employed to generate amorphous carbon thin film (ACTF) from oil palm leaves. The ACTF invention is composed of thin films that resemble graphene sheets with a coiled surface. The study examined the adsorption efficiency of both batch and fixed bed adsorption systems. The study involved conducting batch tests to investigate the effects of contact time, initial concentration of condensate oil ($C_o = 100\text{--}2500\text{ mg/l}$), and temperature. The results showed that the adsorption capacity and removal efficiency improved over time, reaching a maximum of 132.77 mg condensate per gramme of adsorbent and 66.38% respectively, after 6 hours at a temperature of 308 K. The thermodynamic adsorption studies were carried out at temperatures of 288, 308, and 318 K, indicating that the adsorption process is exothermic in nature [53].

M. El-Sayed et al. (2016) Studied an amorphous carbon thin film (ACTF) was synthesized by hydrolyzing wood sawdust and removing the lignin to obtain cellulose. The cellulose was then reacted with cobalt silicate nanoparticles as a catalyst, with the addition of strong sulfuric acid at 23°C . The efficacy of ACTF in adsorbing oil from synthetic generated water was assessed by employing the Thomas and Yoon-Nelson models. The performance research is explained using the notion of breakthrough curves under certain operating parameters, including column bed heights of 3.8, 5, and 11 mm, and flow rates of 0.5, 1, and 1.5 mL/min. The study revealed that the oil absorption process is more advantageous for taller bed heights. The maximum bed capacity of 700 mg oil per gramme of activated carbon textile fabric (ACTF) was attained at a bed height of 5 mm and a flow rate of 0.5 mL/min [54].

FIXED BED ADSORPTION STUDIED

Ali et al. (2022) concerned with employing an activated carbon fixed-bed column run in batch recirculation mode to remove chemical oxygen demand (COD) from petroleum refinery effluent obtained from Iraq's Al-Diwaniyah petroleum refinery facility. The fixed bed column that was employed in this study was divided into three compartments: an upper, middle, and lower compartment. The center adsorption chamber is fed from the bottom

compartment, while the effluent is collected in the upper compartment. The effects of several operational factors, including packing level, pH, and duration on the effectiveness of COD removal were examined by using response surface methodology (RSM). The best circumstances led to a COD removal efficiency of 96.70%: an activated carbon packing level of 80%, a pH of 5.7, and an estimated adsorption duration of 73 min. The findings showed that time had only a little impact on COD removal, whereas the amount of activated carbon packed in had the biggest impact. The current study shows that the activated carbon adsorption system is a useful technique for eliminating wastewater from the condom Odom Al-Diwaniyah petroleum refinery [43].

(Daffalla et al., 2022) investigated the potential of rice husk-derived activated carbon as a medium for removing phenol from an aqueous solution in a fixed-bed adsorption column. We looked at the impacts of concentration on the influent phenol concentration (100-2000 mg/L), flow rate (5-10 mL/min), and bed depth (8.5-15.3 cm). It was discovered that the capacity of bed adsorption increased in direct proportion to the rise in influent concentration and bed depth. However, when the input flow rate increased, the bed adsorption capacity also increased. After three regeneration cycles, it was discovered that the regeneration of an activated carbon column employing 0.1 M sodium hydroxide was successful with a 75% regeneration efficiency. Data on adsorption were found to be consistent with numerous well-known models (such as Yoon-Nelson and Adams-Bohart, as well as bed depth service time models) [44].

(Karunarathne & Amarasinghe, 2013) The potential of sugarcane bagasse-derived activated carbon for the removal of aqueous phenol in a fixed column was investigated. Under 600 °C, without any air, sugarcane bagasse was thermally activated. Breakthrough curves were produced by altering the height of the activated carbon bed in fixed bed studies. In light of this, the ideal breakthrough curves (IBC) were constructed, and for each scenario, the bed capacity (BC), length of the unused bed (LUB), time needed for full bed depletion at infinite fast adsorption TS, and breakthrough timings Tb were computed [45].

Himanshu Patel 2020 Utilizing batch and column investigations, an investigation was conducted into the adsorption of heavy metals, including Pb, Cu, Cr, Zn, Ni, and Cd, onto activated charcoal made from neem leaf powder (AC-NLP). Variations in flow rate, beginning concentration, and bed height were used in the column research to interpret breakthrough curves and parameters. The results show that the best circumstances were lower flow rates (5 mL/min), lower initial concentrations (5 mg/L), and higher bed heights (20 cm). Through the evaluation of isotherm models, comparisons between batch and column studies revealed that column study is favored over batch treatment. The maximum Thomas adsorption capacity for Pb, Cu, Cd, Zn, Ni, and Cr, respectively, was 205.6, 185.8, 154.5, 133.3, 120.6, and 110.9 mg/g. Metal ionic characteristics help to explain this removal pattern. For the adsorption and desorption of AC-NLP, a variety of adsorbing agents, including acids and bases, were used [46].

Mojtaba et al. (2017) studied the fixed bed adsorption method of removing tetracycline from wastewater, which uses synthetic mesoporous carbon as a potential adsorbent. Particles were implanted in a Pyrex glass tube that was made for a lab in order to evaluate the adsorptive capacity of the adsorbent. The enhanced adsorption capacity was caused by a decrease in bed height and flow velocity and an increase in starting concentration. The combination of 4 cm bed depth, 4 mL/min, and 50 mg/L influent concentration yielded the greatest bed capacity of 76.97 mg/g. Under every experimental circumstance, the breakthrough curve's initial portion completely matched the Adams-Bohart model [47].

M. Fathy et al. (2018) examined the performance of fixed bed adsorption using the breakthrough curves approach. It focuses on two parameters: column bed heights (5, 10, and 15 mm) and flow rates (2.2, 5, and 8.4 ml/min). The Thomas and Yoon-Nelson models were utilized to determine various parameters of a fixed bed, such as the adsorption capacity and the time required for 50% breakthrough. The findings showed that the optimal parameters for the ACTF column were a feed flowrate of 2.2 ml/min, a bed height of 5 mm, and an initial oil condensate concentration of 1000 mg/l. The observed breakthrough curves exhibited a satisfactory correspondence with the calculated breakthrough profiles derived from the Thomas model [53].

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