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*Original Research Article*

# Fenton Processes, Adsorption and Nano Filtration in Industrial Wastewater Treatment

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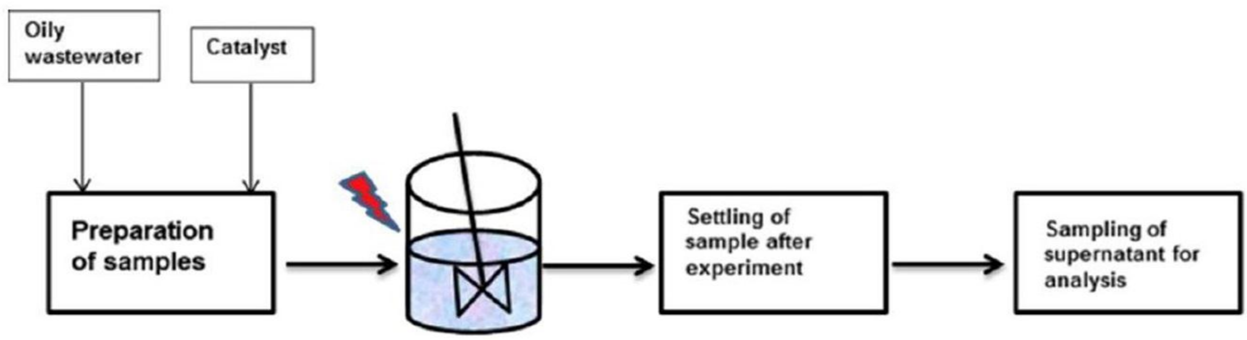
ABSTRACT

This paper reviews Fenton processes, adsorption and nano filtration processes. Wastewater is one of the substances that has been neglected even in recent years in the country. Industrial effluents can have a great variety depending on the type of production and process of industrial units. In other words, for each production line in industrial units, a specific effluent may be produced, the quality and environmental effects of which will depend on the type of material and its content. These effluents are often caused by chemical processes and therefore will create more potential hazards than normal effluents in the environment. Although refineries and petrochemical industries are widely expected to benefit the community, they produce significant volumes of effluents. Effluents produced in refineries are generally considered as hazardous effluents due to their high amounts and high concentrations of oily and petroleum compounds, as well as acids and heavy metals. Protecting the environment against pollution of oil, gas and petrochemical industries has created great concerns from the environmental point of view. Therefore, it is necessary to identify and study the effluents of these units and provide solutions to control their pollution.

GRAPHICAL ABSTRACT

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## Introduction

Today, increasing production of municipal, industrial and agricultural wastewater as a result of increasing population growth and economic development is considered as one of the most important environmental problems in the world. Environmental crises threaten the entire natural system and pollution and destruction of nature and land ecosystems is undeniable. Preserving the environment and natural resources is one of the most important challenges facing humanity in the 21st century, while the need to improve living standards in all countries has not lost its importance [1]. The expansion of urbanization and industrialization, as well as changes in lifestyle and consumption patterns in recent years, especially in developing countries, have led to production of large volumes of effluents in urban, industrial and environmental areas. How to plan and manage wastewater is one of the most important health problems in societies today, and if the environment is not taken into account along with the industrialization of a society, not only will economic development not be achieved, but also many problems will arise in the association with the benefits of an industry. Wastewater management, development and improvement of its operational quality depend on elements such as production control, collection, transmission and disposal [2-4]. When this set acts in the form of a series of coherent and coordinated instructions, it can be effective and useful in promoting the health of the community. In recent years, the interest in measuring pollutants in the effluents of urban and industrial areas has increased significantly because of the destructive effect of these compounds on human health. In addition, the oil and fat in the effluent play an effective role in increasing other pollutants such as COD and BOD as well as the effect on the aquatic ecosystem of the affected areas [5-7]. The possibility of adverse health and environmental impacts caused by pollutants emitted from oil and gas refineries in neighboring areas has been one of the serious concerns of communities. Especially when industries with this type of activity are located in the vicinity of urban communities, residential houses, schools, shopping malls and other public facilities located nearby [8-10].

*Literature review*

Alizadeh et al. (2000) conducted a study entitled "adsorption of some heavy metal ions from aqueous solutions using activated carbon and comparing the results of adsorption percentage of activated carbon with other adsorbents" [11]. They concluded that adsorption process by three activated carbon materials, chitosan and agar should be investigated with first-order reaction kinetics in a pH range due to their effect on the buffering solution and not at a constant pH-value [12-15]. In this project, the use of these three materials can be compared with the extracted sample. In 2001, in a paper entitled "synthesis of mercury from effluent using activated carbon obtained from fertilizer wastes", Deeudomwongsa (2019) was able to evaluate the rate of mercury (II) absorption, considering the dependence of the process on the concentration and physicochemical behavior of the adsorbent. With increasing pH-value, the process is exothermic, which is a film adsorption mechanism at low concentrations and with a partial mechanism at high concentrations [16]. Also Mao et al. conducted a study entitled "selective adsorption of chromium (VI) from industrial effluents using low-cost and abundant adsorbents" and concluded that wool with an adsorption capacity of 81% of a sample of 100 ppm chromium was the best type of chromium adsorbent under residence time of 2 hours of stirring the sample [17]. Also, it was possible to compare the absorption rate of the sample with wool in this project. Samimi et al. (2020) examined degradation of a uniform photocatalyst called reactive orange in aqueous solution with

$TiO_2$ photocatalyst and sunlight. The suspension was magnetically stirred in the dark for 35 minutes to equilibrate the dye solution and $TiO_2$, and then the effect of various parameters was investigated [20]. Samimi et al. (2006) investigated the effectiveness of nano filtration membrane in removing nitrate from drinking water under different operating conditions [21-22]. The aim of this study was to investigate the effect of water pressure and ionic composition on reduction of nitrate from drinking water using a nano filtration membrane. In order to investigate the effect of pressure, the rate of nitrate removal at a concentration of 150 mg nitrate per liter of sodium nitrate and calcium nitrate in the pressure range up to 11 times was investigated. To investigate the effect of concentration on nitrate removal from different concentrations of sodium nitrate and potassium nitrate salts, calcium nitrate and magnesium nitrate were used to simulate water. To investigate the effect of nitrate type anion on its removal rate, nitrate removal rate was measured by adding two different salts of sodium fluoride and sodium sulfate to a solution containing sodium nitrate and the effect of the presence of these anions in different concentrations was evaluated and compared. The results showed that with increasing pressure, the amount of nitrate removal increased, which was more pronounced, especially when nitrate was present with a single-capacity cation. In addition, this increase in removal rate was up to 8 times higher and the aftermath was almost constant.

The type of cation with nitrate also had an effect on its removal and the maximum removal of nitrate was observed in the simulated solution with calcium nitrate and the minimum in the simulated solution with potassium nitrate. Regarding the effect of concentration on the rate of nitrate removal, this effect varies depending on the type of cation associated with nitrate. According to the results of this study, when the cation was accompanied by divalent nitrate such as calcium nitrate and magnesium nitrate, as the nitrate concentration increased, its removal rate increased. But in the case of presence of cations with the nitrate such as sodium nitrate and potassium nitrate, the rate of nitrate removal decreased with increasing concentration. The effect of the type and amount of anion associated with nitrate on its removal rate also varies depending on the type of associated anion. The results of this study showed that with increasing the concentration of sulfate in solution, the amount of nitrate removal decreased, while with increasing the concentration of fluoride in solution, the amount of nitrate removal increased (Fig. 1 & 2). In 2007, they removed tertiary butyl ether from the effluent using photocatalytic degradation processes. Photocatalytic process was performed in a catalytic reactor and sodium lamp was used and final samples were analyzed using chromatography [23-25].

*Oxidation*

As the world's population grows rapidly, so does the water resources available to humans. According to the United Nations, about 43% of the world's population has access to adequate water and 22% access to safe water. Increasing the amount of wastewater produced has caused more pollution of surface and groundwater resources.

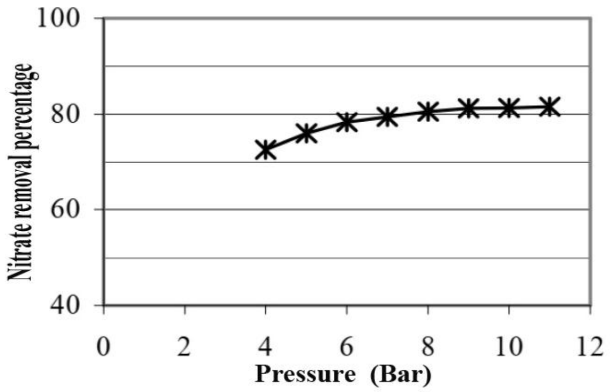


**Fig. 1.** Effect of pressure on nitrate removal with sodium nitrate solution under nitrate concentration of 150 mg/l

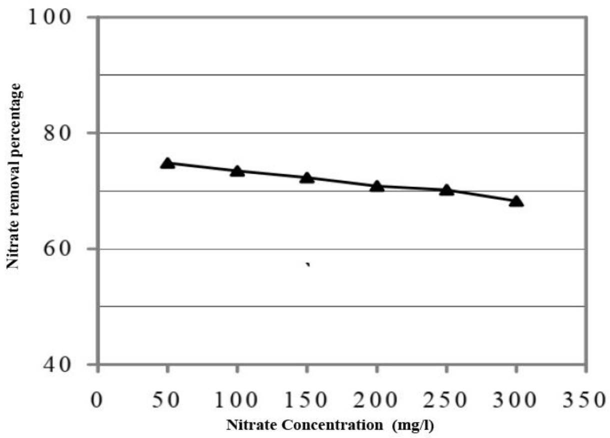


**Fig. *2*.** Effect of concentration on nitrate removal with potassium nitrate solution at operating pressure 8 bar

Since conventional physical and chemical processes alone are not able to remove pollutants from the effluents of some industries, the use of cost-effective complementary processes can solve this problem. Many newly developed treatment methods have joined new research to address health and environmental challenges. Due to the importance of the issue and based on examining the effluents from refineries that contain organic aromatic compounds, they are both dangerous and relatively toxic in nature and biodegradable, which cannot be eliminated by the best conventional technologies available for waste treatment and they may not be economically viable. The need for new and more efficient technologies for treatment of such effluents is becoming more and more important. The importance of the issue is also increasing with the consideration of new guidelines of the environmental organization. Some conventional oxidation methods are in some cases ineffective in bringing the concentration of organic matter to the permissible level. Therefore, the use of new technologies such as advanced oxidation processes can be a good alternative. The mentioned processes have a great potential in eliminating the pollution caused by effluents. Also, these processes have many advantages such as eliminating the environmental pollution along with disinfecting the effluent. Chemical oxidation in wastewater treatment typically involves the use of oxidizing agents such as ozone ($O_3$), hydrogen peroxide ($H_2O_2$), potassium permanganate ($KMnO_4$), chlorine dioxide ($ClO_2$), and chlorine ($Cl_2$). Advanced oxidation process in which the hydroxyl radical ($OH^{\circ}$) is used as a strong oxidant to destroy specific major organic components cannot be oxidized by conventional oxidants such as ozone and chlorine. In advanced oxidation, the process is based on the production of hydroxyl free radicals through various reactions involving the joint action of two oxidants, a catalyst, and an oxidizer [26].

*Fenton reaction*

The Fenton reaction is defined as the reaction between Fe (Π) iron and hydrogen peroxide, in which the hydrogen peroxide is converted to HOH and $OH^-$ according to the following equations (Table 1). The strong HOH radical combines with carbon in organic matter to break the double bond in the benzene ring, eventually breaking the ring and releasing hydrogen molecules. Many organic materials are converted to less harmful substances, carbon dioxide and water. On the other hand, it has the ability to completely destroy aromatic compounds such as benzene, toluene, cresol and oxylene. Double-bonded chemical compounds, such as organic acids, ketones and aldehydes are attacked by these radicals. Hydrogen peroxide alone is used to oxidize substances such as cyanides, sulfides, sulfites, chromiums and heavy metals by batch operation [27]. Amino and nitro compounds are also converted to simple compounds under the influence of this strong radical; but some organic compounds are resistant to radicals, such as acetic acid, acetone and oxalic acid. However, radicals do not react with saturated hydrocarbons (Table 1). Figure 1 shows an example of a Fenton process laboratory pilot [28].

After adding Fenton reagent, an equivalent amount of 200 ml of wastewater sample was exposed to a magnetic stirrer. Also, UV light is produced by a UV grid lamp with a high intensity of 254 nm, produced by UVP Inc. Model (R-52). In this study, physicochemical treatment of oil refinery wastewater has been investigated. This research includes typical physical separation processes related to Fenton and photo-Fenton processes [29].

Exposure was rapid mixing for 10 minutes and then slow mixing for 30 minutes to increase clotting. Also, the turbidimeter of HACH 2100N IS measured the turbidity of the wastewater. The pH-value of wastewater was also measured using a pH meter [30].

$Fe^{+2} + H_2O_2 \rightarrow Fe^{+3} + OH^{-} + OH^{\circ}$ (R1)

$Fe^{+3} + OH- \rightarrow Fe^{+2} + OH^{\circ}$ (R2)

$RS + 2\ OH^{\circ} \rightarrow H_2O + RSO$ (R3)

$RSO + 2\ OH^{\circ} \rightarrow H_2O + RSO_2$ (R4)

Below you can see how the advanced homogenization process progresses without the use of light (Table 2). In the final reaction of the combination of $H_2O_2$ with iron (II), iron is known as the catalyst (Fenton reactor) [31].

Advanced homogeneous oxidation process in the presence of light:

$R_1\ 2O_3 + H_2O \rightarrow OH^{º} + O_2 + HO^{2-}$

$R_2\ O_3 + H_2O_2 \rightarrow OH^{º} + O_2 + HO_2^{º}$

$R_3\ Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^{º} + OH$

Fenton's response can be a step towards controlling pollution from effluents and then providing other effective solutions to other parameters. The liquid part of the effluent is ultimately water that will enter groundwater and surface water contaminated by industrial activities, and sometimes physical and chemical treatment is required to treat it, and sometimes in sensitive cases, many biological methods must be used.

*Advantages of the Fenton reaction*

There are advantages with the Fenton reaction. The reaction (oxidation) takes place in less time, the chemicals used (iron salt) are cheap and expensive, and the chemicals used (hydrogen peroxide and iron salts) are non-toxic.

Further, we do not have restrictions on the transfer of mass with the chemicals used. Also, equipment and energy are easily provided. Another advantage is the degradation agent (hydroxyl radical) formed rapidly as well as operating conditions that are easy to control.

In addition, it is suitable for wastewater treatment with various organic compounds. And, the reaction takes place under ambient temperature and pressure

*Application of the Fenton reaction*

Fenton's powerful reaction can be used in the following cases:

- Industrial effluent treatment containing aromatic amino compounds;
- Industrial effluent treatment containing different colors;
- Industrial wastewater treatment containing pesticides;
- Industrial effluent treatment containing surfactant;
- Industrial effluent treatment containing explosives;
- Wastewater treatment from the textile industry;
- Wastewater treatment from fuel terminal plant;
- Wastewater treatment from a refinery; and,
- Preparation of toxic and non-biodegradable effluents for biological treatment.

*Adsorption*

In the adsorption operation, a component of the gas or liquid phase is transferred to the solid surface.

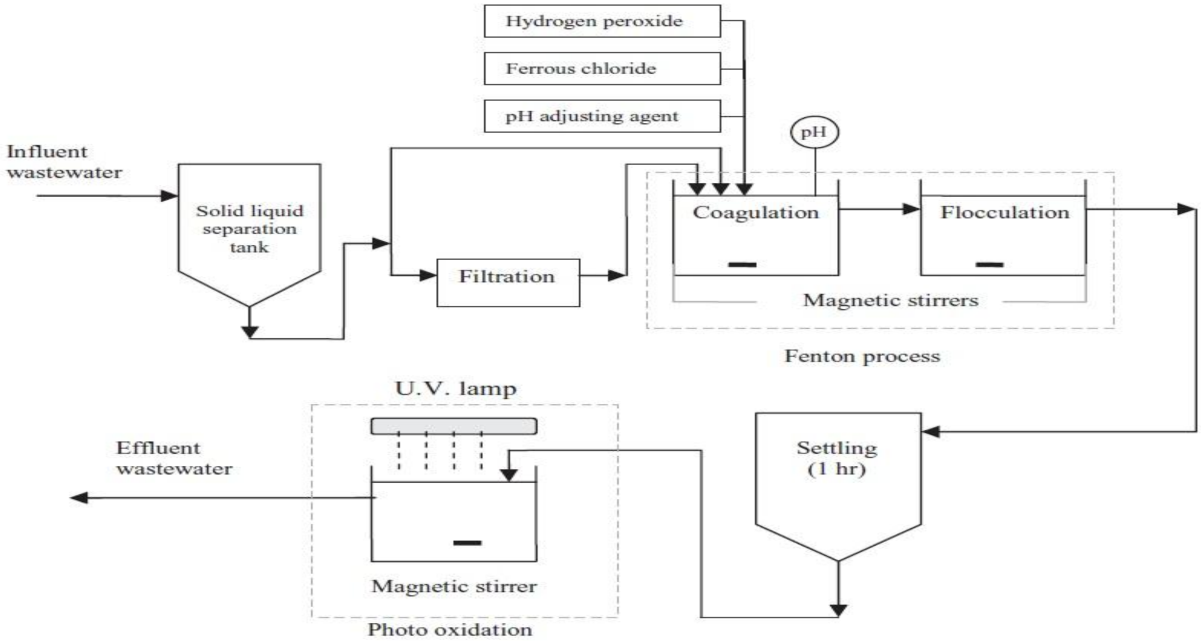


**Fig. 3.** Sample Fenton process laboratory pilot

This process is applied in bleaching sugar syrup and refining industrial or edible oils, and removing contaminants from the air or mixtures of other gases. The term adsorption is used to describe the fact that concentration of adsorbed molecules at the solid contact surface is greater than the gas or solution phase.

Adsorption on a solid surface is due to the gravitational force of atoms or molecules on that solid surface. In practice, adsorption of various forces, both physical and chemical, is effective and its amount depends on the nature of the adsorbed material and the adsorbent body. In the case of gaseous separations from the adsorption process, in dehumidification of dry air and other gases, decontamination and separation of impurities from industrial gases such as carbon dioxide, recovery of valuable solvents from their dilute mixture with air or other gases, and separation of mixed gases from mixed mixtures take place. Liquid separation processes include dehumidification of gasoline, decolorization of petroleum products and aqueous solutions of sugar, desalination of water, and separation of aromatic and paraffinic hydrocarbons, each of which has a wide range of applications in the industry.

The similarity of these processes is that the mixture in contact with another insoluble phase (such as solid adsorbent) and the uneven distribution of raw materials between the adsorbed phase on the solid surface and the fluid mass cause separation. The phenomenon of surface adsorption can be described as a high affinity of the adsorbed component towards the solid body. Therefore, the adsorption phenomenon is divided into two general categories: Physical surface absorption and chemical adsorption.

Physical adsorption is due to the presence of van der Waals forces, similar to what happens when gas liquefies between molecules. The forces involved in this type of absorption are permanent dipole, inductive dipole and quadrupole.

There is no electron transfer in this type of adsorption, the nature and strength of the adsorption depends on the physical properties visible in the adsorbent and not on the type of adsorbent. This type of adsorption is fast because it has an activation energy close to zero and the heat from the adsorption is very low. In chemical surface adsorption, electrons are transferred between a particle and a surface, and the forces involved are electron interactions, exchange, and electron sharing.

In chemical adsorption, the activation energy is high and the adsorption rate is low. Due to the formation of chemical bond, the heat of adsorption is high and nature of adsorption depends on the solid and adsorbent properties. The solid which adsorption occurs on its surface is called the adsorbent or substrate, and the adsorbed liquid is called the adsorbent. Adsorption occurs on the solid-liquid interface. Absorbent solids are usually consumed in the form of granules (spherical particles with a diameter of several millimeters) and their size varies from 12 mm to 50 micrometers.

Many solids have the property of being able to absorb some of the gas or solute dissolved in the solvent. The amount of a gaseous substance that is physically adsorbed on the surface of a solid changes with pressure. The idol equation allows us to measure the volume of gas required to form a single layer of liquid on a solid surface, $V_m$. The idol equation is as follows (Equation 1):

$$\frac{Y}{V(1-Y)} = \frac{1}{V_m} + \frac{(B-1)Y}{V_m} \quad (1)$$

Y is the relative pressure ($P/P_0$), V is the volume of adsorbed gas ($m^3$), P is the gas pressure and $P_0$ is the saturation pressure at the temperature of the adsorbent storage chamber.

$V_m$ is the volume of gas required to form a layer on the adsorbent at system temperature and B is a constant value. Therefore, the curve Y/V(1-Y) in terms of Y must be a line with slope ((B-1)) / $V_m$ and width from the origin $1/V_m$ to get the value of $V_m$ from the values obtained from the Figure so two equations will be solved together. The linearity of Y / V (1-Y) in terms of Y is usually seen at relative pressures ($P / P_0$), in the range of 0.05 and 0.3, although in some cases it has been seen outside this range.

Using the idol method, the adsorbed monolayer volume of the gas on the surface is obtained and usually nitrogen gas, whose characteristics are well known, is used. Of course, it should be noted that other known gases can also be used. Knowing the molecular cross section of nitrogen, we can calculate the surface area of the adsorbent and the values of surface area of the adsorbent are usually proportional to their capacity. Figure (2) shows the schematic used in this dissertation.

*Anthracite*

Stones and fossils left over from plant and animal remains are called bioliths or biolites. Biolites are divided into combustible or non-combustible groups. Combustible biolites are called caste biolites or flammable rocks. Coal is actually a type of combustible biolite.

Biolites include two groups of plant and animal remains. Plant organs eventually convert to peat, bitumen and coal; and animal organs to hydrocarbons, oil, gas, and oil shale. Lignite coal is also known as soft brown coal. This type of coal is formed in the post-peat stage. In principle, lignites have a low degree of charcoal and some residues of woody material, leaves and bark of plants can still be found in them. The color of lignite coal is black to brown to black, porous and its density is about 1.5.

The porous texture of lignite is similar to that of dry charcoal. Some of them have loose and fibrous texture and some of them have dense and soil-like texture. In this type of coal, the average amount of carbon is 75-80%, the average amount of oxygen is about 10-15% and the average amount of hydrogen is about 5-7%, and their calorific value is low and about 6000 kcal. The moisture content of lignites is also higher than that of bituminous coals, reaching about 35-75%, which is why they produce a lot of smoke when burning. Their volatiles are relatively high and sometimes reach about 40-55%. Quasi-bituminous coal is the boundary between lignite and bituminous coal and for this reason they are called quasi-bituminous coal.

These coals burn easily and have a bright flame with smoke and produce a lot of gas. The texture of these coals has a lot of voids that are usually filled with jelly material in their texture. The color of these coals varies between brown and dark brown. These types of coals have a good layering appearance. The amount of wood materials is very low and their carbon content reaches about 80%. Their volatile content is about 53-53%, which are crushed when exposed to air. Bituminous coals are composed of decomposition of algae and microscopic animal organic matter that contains about 85-80% carbon and about 5% oxygen and hydrogen. These coals usually have a dense, hard and striped texture and consist of alternating glossy and matte layers.

Bituminous coals are divided into two groups in terms of volatiles. The first group contains 30-45% of volatiles and is mostly used as fuel in power plants. The second group has less volatiles of about 20-30% and is mostly used to prepare coke. Bituminous coals release significant amounts of flammable gas when burned. Bituminous coal is the most common type of fossil fuel in nature and a widely used one. When reverse osmosis membranes grew at relatively low pressures and flow rates. The separation mechanism in nanofiltration includes the effect of molecule size (for non-ionic materials), differences in permeability and solubility of feed components, and electrical interaction (Donan phenomenon) between the membrane surface and the ions in the feed.

From the beginning, the water treatment industry has been one of the most important areas of nanofiltration. Among its wide applications, we can mention its use in the softening process. As they are still sometimes referred to as "hardeners".

However, studies show that these membranes also function well in removing natural organic matter and dyes from water. The combined ability to remove organic and inorganic materials along with the removal of hardness in one step makes it possible to replace one membrane unit with several parts of the water treatment process. This has led water treatment companies to do more research in this field and show interest in investing in this sector, followed by new applications such as disinfection by removing viruses, pesticides and other micro-pollutants, and then removal of arsenic.

Today, due to increasing population, declining drinking water resources, strict regulations and increasing operating costs such as energy, it seems necessary to use an optimal process in water treatment. Among the latest water treatment methods, we can mention membrane methods. The advantages of these methods include energy savings, simple technology and ease of use, environmental compatibility, and production of good quality product, high flexibility in system design and the possibility of combining with other separation processes.

In addition to water treatment, nanofiltration has other applications in different fields, including municipal and industrial wastewater treatment, food industry, chemical industry, petrochemical, paper industry, textile industry, metal and acid recovery industries, removal of harmful and excess materials and pharmaceutical industries.

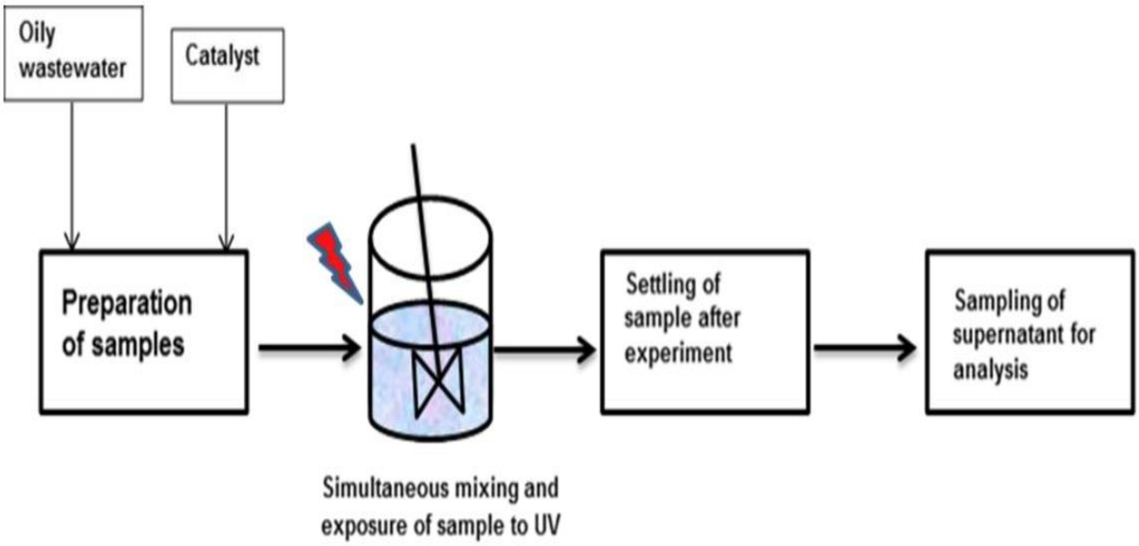


**Fig. 4.** Schematic of the adsorption process

The first and most important application of nanofilters was in water treatment and initially these membranes were known as water hardening membranes. Nanofilters are used to remove polyvalent ions such as calcium and magnesium, pesticides, organic matter, viruses, bacteria, and monovalent ions such as nitrate, chloride and sodium. Today, research focuses mainly on understanding the mechanism by which different compounds pass through membranes and explore new applications, including the removal of monovalent ions. Nanofiltration membranes are located between reverse osmosis and ultrafiltration, in other words, the separation in these membranes is based on the screening mechanism, and because these membranes are pregnant, the reactions between the membrane and the solution also play an important role in this separation. These reactions depend on the properties of the solution to be filtered and the structure of the membrane. The basis of nanofiltration devices is similar to reverse osmosis.

The difference between the two devices is the pressure required and the size of the penetration areas of the two shells. So, the pressure used in NF devices is less and its pores are bigger. As a result, more salt will pass through it.

## Conclusion

Consumption of water contaminated with oil derivatives poses a serious threat to the health of the environment, humans and other living organisms.

Due to the fact that oil spills and leaks into water are inevitable in most cases, and also the adverse effects of water contaminated with oil derivatives on humans and the environment, several methods for water treatment and separation have been devised. Gravity separation, filters, reverse osmosis, biological processes, dissolved air buoyancy, membrane bioreactors, adsorption with activated carbon, chemical coagulation, electrical buoyancy, etc. are some of the solutions that have been used for water treatment so far. Most common processes are not introduced as an efficient treatment method to separate water from oil due to limited and low efficiency, high sludge generation, the need to add chemicals, high cost, the need for pre-treatment and problem management. The wastewater treatment process consists of three main stages, but no definite boundaries can be conceived for these steps and they merge. The first stage involves the physical processes that take place on the effluent. The second stage is the biological stage and the third stage is the chemical stage. Both chemical and biological steps are completed using physical properties such as density difference and adsorption. There is also a fourth stage to complete the treatment, which includes a series of disinfection processes. Sometimes, by combining different techniques, a highly polluted effluent can be treated in such a way that its final discharge is ion-free water or at least suitable for drinking water.

But this is usually acceptable as long as its pollution indices are set to a permissible level that is harmful to the environment or suitable for irrigating farms and purify. The effluent, depending on its origin, may be domestic, industrial, agricultural, or a combination.

On the other hand, the effluent in terms of physical, chemical, biological properties as well as pollution power includes four types of quality: poor, medium, strong and very strong. The health importance of effluent depends on the presence of chemical agents and living pathogens and organic matter that not only cause various diseases but make the environment stink and look bad.

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## Declaration of Competing Interest

The authors declared that they have no conflicts of interest to this work.

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## Disclosure Statement

The authors reported no potential conflict of interest.

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