

Applications of aluminum oxide and nano aluminum oxide as adsorbents: review

Hawraa Kassem Hami, Ruba Fahmi Abbas*, Emad Mahmoud Eltayef, Neda Ibrahim Mahdi

Department of Chemistry, College of Science, Al-Mustansiriyah University, Baghdad, Iraq (suha_rrr_1983@yahoo.com)

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Abstract

Metal oxides are widely used in adsorption technology as adsorbent surfaces because of their efficiency, low cost and unique physical properties. The aim of this review to clarify the role of aluminium oxide and Nano aluminium oxide in removing some chemicals contain that influence on human health such as dyes, antibiotics, and heavy metals. This paper also includes the affective of some adsorption parameters like pH, contact time, removal percentage and temperature. The Adsorption nature, kinetic adsorption models and isotherm models are also reported here.

Introduction

Pollution is currently one of the most widespread environmental problems that began to emerge in the 20th century because of industrial growth associated with the industrial revolution, all this lead to the emergence of new categories of previously unknown chemicals[1]. Thus, it is necessary to find cheap and effective ways that contribute to decontamination and maintain a clean and healthy environment for future generations.

Over the past years, adsorption has emerged as an efficient technique widely used in removing pollutants from the environment .This technology is simple, cheap and environmentally friendly, one of the most important things for effective adsorption is chosen the right surface[2,3]. For all these reasons, there is an increasing trend of research papers published in adsorption removals in the aqueous phase[4].

Metal oxides are the most commonly used surfaces in the removal of contaminants; they are cheap and widely manufactured, In addition to their high mechanical properties and resistance to thermal decomposition compared to biological surfaces[5]. One of the renowned oxides is Aluminium oxide (Al_2O_3) which is an amphoteric oxide known as alumina and found in many crystalline structures like ' $\alpha-Al_2O_3$, $\gamma-Al_2O_3$, $\theta-Al_2O_3$, $\eta-Al_2O_3$ ' etc. "Fig. 1 and 2". different in physical and chemical properties as well as applications[6]. Aluminium oxide is characterized by its non-toxic, easy used, chemical stability and has many hydroxide groups all these property mad it efficient adsorbent. From 1923 Aluminium oxide had usage as adsorbed to remove Pigments, antibiotics, heavy metals, dissolved organics etc.[7,8].

In the light of the continuous development in the field of nanotechnology and its use in treatment pollution of the environment, interest increasing in the use of metal oxides as nanoparticles as absorbers to remove pollutants from the environment[9], especially Aluminium oxide nanoparticles inasmuch to it inexpensive, high surface area, surface reactivity, well adsorption ability, surface acidity and thermal stability[10,11]. There are many techniques to prepared aluminium oxide nanoparticles powder including ball milling, hydrothermal, sol-gel, co-precipitation, pyrolysis, laser ablation, vapour phase reaction and combustion methods[12].

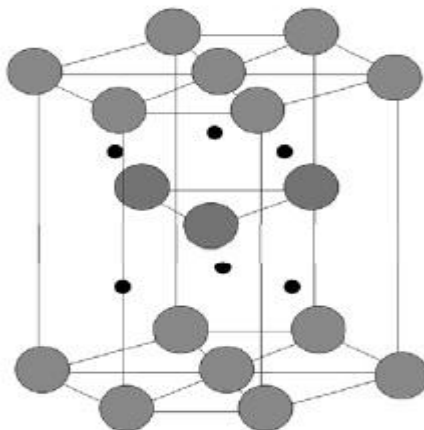


Fig. 1: Structure of α -Al₂O₃

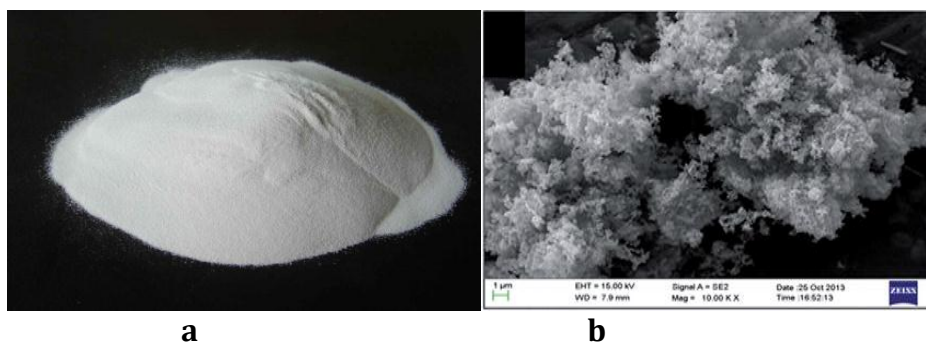


Fig. 2: Characterization of alumina oxide (a)- Al₂O₃ powder (b)- SEM form at enlargement of 10.00 KX.14428 for Al₂O₃nanoparticle[13].

Chemicals Pollution

All living and non-living substances consist of chemicals and every manufactured product we use in our daily lives includes chemicals. These substances can contribute to improving our health and well-being when used duly. In contrast, hazardous chemicals can cause health and environmental damage when used improperly[14].

International organizations such as "WHO, USEPA, ATSDR and EU" analysis of the impact of toxic metal ions and pollutants on human health the increase of concentration of these substances cause serious health damage. Table 1 summarizes some of the toxic elements and their admissible limits in water and their impact on human health according to these organizations.

Table 1: Toxic contaminants in drinking water and their influence on human health

Contaminant	Allowable Limits of concentration mg/l	Possible impact of toxic elements on human health	Ref.
Arsenic	0.01	Skin harm or circulatory problems, may increase the risk of cancer, black foot disease	15
Azo dyes		Causes chromosomal damage and cancer.	16
Cadmium	0.05	Kidney damage, lowering red blood cells, hypertension,	17
Chromium	0.1	Loss Calcium from the bone, staining the yellow teeth (forming a cadmium ring) and damage to the bone marrow, renal failure, lung irritation	18
Copper	1.3	Short-term exposure: digestive distress Long-term exposure: damage in liver or kidney	18
Fluoride	4	Bone diseases and children may lose their teeth	18
Eosin yellow	-	Skin irritation	5
Lead	0.015	Infants and children: delay in physical or mental development, high level cause coma and die Adults: kidney problems, Hypertension, Memory or focus problems	17
Iron	0.3	Lung cancer	19
Nickel	0.02	Cancer of the lung , nose, laryngeal and nickel itch	20
Nitrate	11.3	It poses a risk to infants younger than 6 months and pregnant women	18
Malachite green dye	-	Lung cancer	9
Mercury		Affects the digestive and immune systems, Cancer	18
Molybdenum	10–15	Joint pain, gout-like symptoms, and high uric acid in the blood	18
Phenol	-	Extremely Toxic, mutagenic and absorbed by skin	16
Phosphate	-	Affect the calcium in the body and cause damage to the kidneys	21
Selenium	0.05	Hair loss and nails circulatory problems, cancer	18
Thallium	0.0005	Hair loss; Blood changes, Problems with the kidney, intestine and liver.	16
Zinc	5	Short time: stomach cramps, nausea, vomiting may occur. Long time : anemia, pancreatic damage, and low-density lipoprotein cholesterol levels	22

Continuing the production and use of chemicals worldwide requires a safety system to dispose of these chemicals. In this article, we will talk about some hazardous chemicals that removed by adsorption technology on the surface of Al_2O_3 and Nano Al_2O_3 .

Adsorption parameter

Adsorption process happens either in one step or in a set of steps like Film or external diffusion, pore spreading, surface spreading and adsorption on the surface of the pores. Fig. 3 shows the pathways of adsorption process[23].

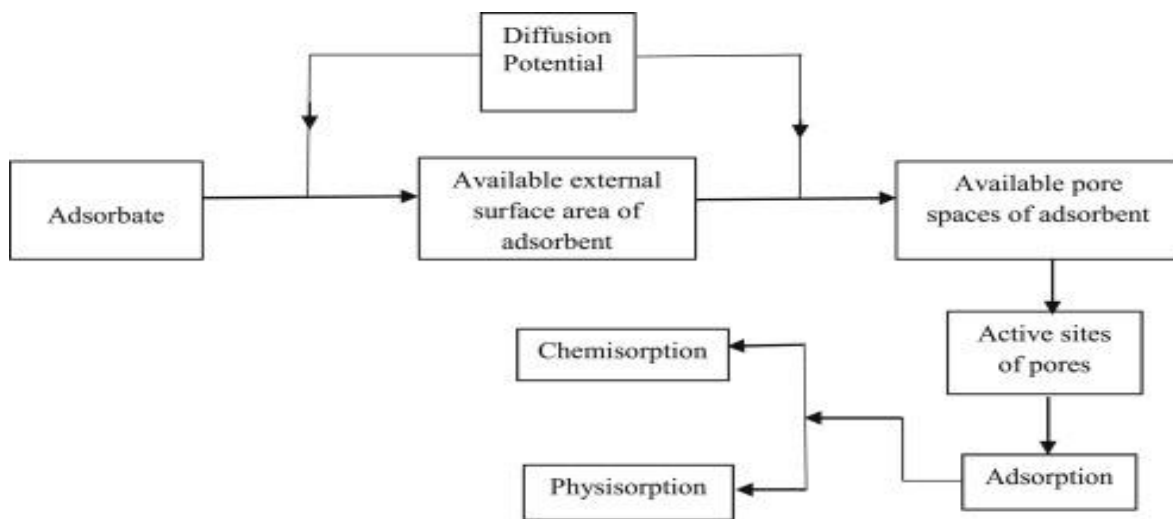


Fig. 3: Pathways of adsorption process Knowing the optimum conditions will help increase the efficiency of the adsorption process and choose the right surface for best results.

There are several parameters that impact in the adsorption process which are studied to reach optimal conditions that give the highest removal percentage. In this review we take removal percentage, pH, temperature, time, and isotherm.

1. Removal percentage: It is known that the higher the percentage of removing pollutants from the environment, the more efficient the adsorption process. Therefore, knowing the percentage of removing pollutants contributes in choosing the appropriate conditions that give the highest efficiency [4].
2. pH of solution is vital factor that impact in the adsorption efficiency. It enhances adsorption removal of cationic or basic adsorbate but reduces that of anionic or acidic adsorbate[13].
3. Temperature: An increase in the adsorption capacity with an increase in temperature is recognized as an endothermic and a low absorption capacity with an increase in the temperature of the solution is called exothermic. So Temperature reinforces adsorption removal of water pollutant by increasing activity of the surface and kinetic energy of the adsorbate but may damage adsorbent physical structure[10].
4. Contact time: as shown in Fig. 4 the reinforces adsorption removal rate of adsorption pollutant by less its mass transfer resistance[10].
5. Isotherm modelling: It helps to know the adsorption mechanism, clarify surface properties and also helps in designing an effective adsorption system[16].

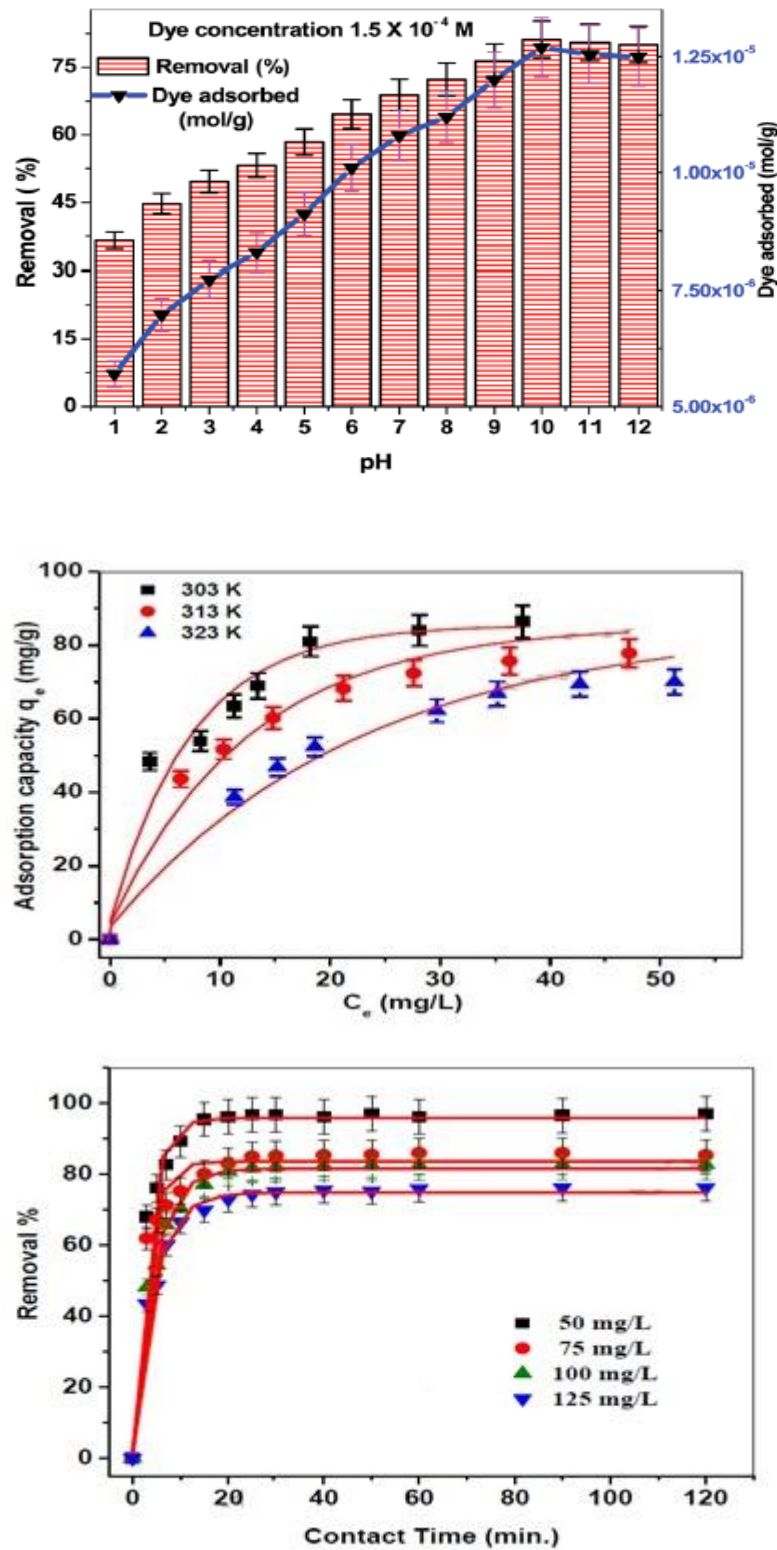


Fig. 4: Sample of adsorption parameter that effect of adsorption in Al_2O_3 and Nano Al_2O_3 surface[10, 13].

Applications

1-Dyes

Dyes are dangerous and carcinogenic organic pollutants that affect living organisms. Dyes are used in many industries like food colouring, cosmetics, paper and textile industries and they are characterized by being light-stable and non-biodegradable as well as causing water

discoloration. There were many techniques to depose dyes . Adsorption technology is most efficiency among them[24,25].

The dye adsorption process involves two mechanics (adsorption and ion exchange) and impacted by numerous parameters like dye/adsorbent reaction, Surface area of adsorbents, pH, contact time, particle size and temperature. The adsorption feature has become a cheap use of sorbents, which reduces the cost of the step[26].

Many Azo dyes like "Methylene Blue, Orange G, Acid Orange-7 and Eriochrom Black T" had removed by Al_2O_3 and Nano Al_2O_3 surface, Fig. 5. Table 2 listed the removal of some dyes in these two surfaces.

Table 2: Adsorption of different dyes in Al_2O_3 and Nano Al_2O_3 surface

Dyes	Adsorbed	Removal %	Contact Time (min.)	Isotherm	Temp.	Adsorption nature	pH	Ref.
Methylene Blue	Nano Al_2O_3	81.2	25	Langmuir	Endothermic	Physical	10	13
Orange G	Nano Al_2O_3	-	30	Langmuir	-	Physical	-	10
Malachite green	Al_2O_3	-	60	Langmuir	-	Physical	7	3
Eosin yellow	Nano Al_2O_3	99.3	120	Langmuir	Endothermic	Chemical	4	27
Acid Orange-7	Nano Al_2O_3	100	60	-	Endothermic	-	2	28
Eriochrom Black T	Al_2O_3	88	30	Langmuir	Endothermic	Chemical	7	29

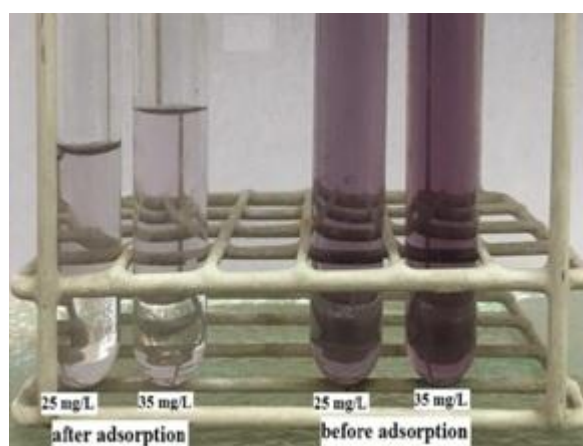


Fig.5: photographs of Eriochrom Black T before and after adsorption[29]

2-Antibiotics

Antibiotics are deemed as an environmental and health challenges due to their genotoxic and mutagenic influences and continuance in natural ecosystem[30].

The widespread use of antibiotics in the treatment of human and animal infections has led to the throw of these antibiotics in the form of wastes that cause soil and water contamination, So it is of vital to remove the antibiotic Waste from source like house, hospitals and pharmaceuticals factories before release them to ecological. One of these antibiotics is the tetracycline family which widely used in the treatment of many infections ‘The cheapness and availability of these antibiotics as well as their therapeutic effect has led to their widespread

use in developing countries. The majorities of these antibiotics are not absorbed by the body and released into the environment causing environmental pollution[31,32].

Previous studies have shown a strong relationship between tetracycline derivatives and the surface of aluminium oxide that made aluminium oxide surface is one of the best surfaces used in the removal of these antibiotics from the environment, as shown in table 3. The adsorption of tetracycline group on the surface of Al_2O_3 is chemical adsorption and ligand-promoted dissolution is obtained during the adsorption process[33]. The adsorption of tetracycline hydrochloride, chlortetracycline hydrochloride and oxytetracycline hydrochloride show that the deformation energy is much less than their non-bonding energy and the adsorption happen by non-bond reaction[34]. Other research reveals existence strong transformation reaction rate correlates by the formation of surface complex between tetracycline and Al_2O_3 [35], also the nature of adsorption for this Antibiotics is physical.

Table 3:Adsorption of tetracycline derivatives in Al_2O_3 and Nano Al_2O_3 surface

Antibiotics	Adsorbed	Removal %	Contact Time (min.)	Isotherm	Temp.	Adsorption nature	pH	Ref.
Chlorotetracycline	Al_2O_3	22	180	-	-	-	5	35
Chlortetracycline hydrochloride	Al_2O_3	66.21	120	Freundlich	Endothermic	Physical	5	34
Doxycycline	Al_2O_3	81.32	90	-	-	Physical	10	36
Oxytetracycline	Al_2O_3	15.8	180	-	-	-	5	35
Oxytetracycline hydrochloride	Al_2O_3	69.48	120	Freundlich	Endothermic	Physical	5	34
Tetracycline	Al_2O_3	18.8	180	-	-	-	5	35
Tetracycline hydrochloride	Al_2O_3	95	1440	-	-	Physical	5.3	34
Oetracycline hydrochloride	Al_2O_3	72.62	120	Freundlich	Endothermic	Physical	7	34

3-Heavy metals

One of the serious environmental problems is the pollution of Heavy metal even when it is present in low concentrations significantly and non-degradability affects in the environment around us and accumulate in the food chain and causes many risks on human's health, These metals discharged into water and soil in the form of liquid waste from various industrial processes causing water and soil pollution. Also it cannot bio degradable and tends to piling up in living organisms[37-38].

World Health Organization "WHO" had list arsenic, cadmium, mercury and lead in its list of ten chemicals of a major public health concern in 2018 [39]. Also cadmium, mercury, and arsenic are deem as the most toxic (Big Three) group of heavy metals with the utmost potential risk to humans and ecological [4]. Adsorption is one of the best methods used to remove heavy metals from waste water because of sorbent regeneration, chemical and biological sludge reduction, high efficiency, and possibility to recover metals. Also the adsorption process is appropriate even when metal ions are present low concentration up to 1 mg/L [23].

Metal oxide like Al_2O_3 and Nano Al_2O_3 is efficiency in separation of menace metal ion from aqueous solutions and that can see in table 2.

It is observed from 'Table 2' is that the contact time is reduced for same heavy metals when we used nano aluminum oxide surface instead of aluminum oxide, the contact time of adsorption Cd decrease by "25%, 16.6% for Pb, 33.3% for Ni and 55% for Zn" when we used nano Al_2O_3 , also the % removal was found to be in order:

For Al_2O_3 :

Ar (II)>Zn(II)>Ar(V)> Cd(II)>Pb(II)> Cu(II)> Ni(II)

For Nano Al_2O_3 :

Cu(II)> Fe(III)> Cr (VI)>Pb(II)>Th(II) ~Se(VI)>Se (IV)>Cd(II)> Mo(II) ~ Zn(II)> Ni(II).

Table 4: Adsorption of different heavy metals in Al_2O_3 and Nano Al_2O_3 surface

Heavy metals	Adsorbed	Removal %	Contact Time (min.)	Isotherm	Temp.	Adsorption nature	pH	Ref
Arsenite	Al_2O_3	-	60	Langmuir	-	-	6	40
Arsenic(III)	Al_2O_3	99	60	Langmuir	-	-	6	41
Arsenate(V)	Al_2O_3	95	60	Langmuir	-	-	6	41
Cadmium (II)	Al_2O_3	94.25	120	Langmuir	Exothermic	Chemical	5	2
Cadmium (II)	Nano Al_2O_3	87	30	Freundlich	-	-	5	42
Copper(II)	Al_2O_3	85.42	60	Langmuir	-	Physical	6	43
Copper(II)	Nano Al_2O_3	100	360	-	-	-	7.5	44
Chrome(VI)	Nano Al_2O_3	99	60		Exothermic	Physical	3	45
Iron (III)	Nano Al_2O_3	99.99	50	Langmuir	Exothermic	Physical	4	46
Led (II)	Al_2O_3	92.10	120	Langmuir	Endothermic	Chemical	5	2
Led (II)	Nano Al_2O_3	97	20	Freundlich	-	-	5	42
Nickel(II)	Nano Al_2O_3	7.75	240	Langmuir		-	5	45
Nickel	Al_2O_3	84.8	720	-	-	-	7.5	38
Nickel-Zinc	Al_2O_3	92.6	360	-	-	-	6	38
Mercury (II)	Nano Al_2O_3	-	1440	Langmuir	-	-	6	7
Molybdenum	Nano Al_2O_3	80	30	Langmuir		-	5	47
Selenite(VI)	Nano Al_2O_3	95	2160	Langmuir	-	-	6.5	5
Selenate(IV)	Nano Al_2O_3	90	2160	Langmuir	-	-	6.4	5
Thallium(IV)	Nano Al_2O_3	95	70	Langmuir	Endohermic	-	4	48
Zinc(II)	Al_2O_3	98.9	180	-	-	-	7.5	38
Zinc(II)	Nano Al_2O_3	80	10	Freundlich	-	-	6.5	49

3- Other compounds

There are many hazardous compounds that the surface of aluminium oxide used to remove them efficiently such as phenol, carboxylic acids in vegetable oils etc.[50-51]. Materials such as fluorine, phosphate and phosphorus are widely used in our daily life and the increasing of them poses a health hazard and causes environmental pollution[52-53], these materials can also remove by adsorption technique in Al_2O_3 and Nano Al_2O_3 surface, table 5.

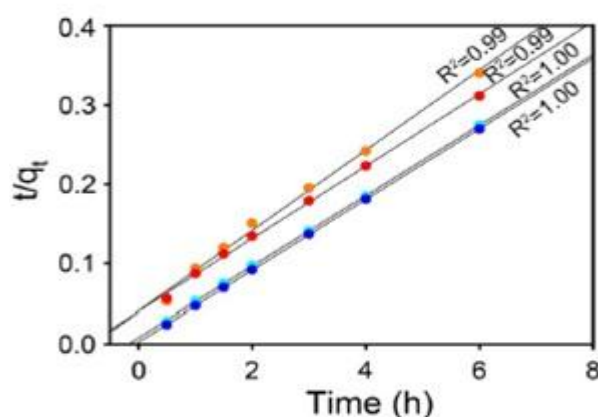
Table 5: Adsorption of some other compounds in Al₂O₃ and nano Al₂O₃ surface

Other	Adsorbed	Removal %	Contact Time (min.)	Isotherm	Temp.	pH	Ref.
Andiroba oil	Al ₂ O ₃	40.29	30	Langmuir	-	-	51
Carbondots	Al ₂ O ₃	50	1440	Freundlich	-	3	54
Ferrocyanide	Al ₂ O ₃	100	1980			7	55
Fluoride	Al ₂ O ₃	99	180	-	-	6.5	56
Fluoride	Nano Al ₂ O ₃	94	60	Freundlich	Endothermic	4	52
Nitrate	Nano Al ₂ O ₃		60	Langmuir	Endothermic	4.4	57
Palm oil	Al ₂ O ₃	21.7	30	Langmuir	-	-	51
Phenols	Al ₂ O ₃	14.7	1440	Freundlich	-	4.5	50
Phosphorus	Nano Al ₂ O ₃	38	180	Langmuir	Endothermic	4	53
Phosphate	Al ₂ O ₃	86.3	-	Langmuir	-	5	58
Sodium dodecyl sulfate	Al ₂ O ₃	90	180	-	-	4	59
Sodium tetradecyl sulfat	Al ₂ O ₃	80	180	-	-	4	59

Kinetic studies

Research in the adsorption kinetics helps clarify the mechanism that controls absorption Processing. Adsorption kinetics is important as it provides value insight into the interaction paths and mechanism reactions[36].

In this review the studies of kinetic adsorption of dye, Antibiotics and heavy metals in Al₂O₃ and Nano Al₂O₃ surface show agreement with the pseudo second-order except the adsorption of Cd (II) in Nano Al₂O₃ surface was fit to pseudo first -order[45]. Adsorption mechanism of "Pd(II), Cd(II)[2], Cu(II)[43], Fe(III)"[46] were complex and involve two or more steps such as surface adsorption and intraparticle diffusion, Fig. 6. Nano Al₂O₃ surface had contact time less than Al₂O₃ surface.

**Fig. 6:** kinetic plots of "Ni" and "Zn" adsorption on γ -Al₂O₃; pseudo second-order[36].

Conclusions

There are many researchers studied the use of Al₂O₃ and nano Al₂O₃ surface for removal some chemical compounds such as dyes, antibiotics and heavy metals through the discussing of some adsorption factor such as time, isotherm, removal percentage, isotherm, temperature and pH.

The results showed:

- 1- That most of these compounds prefer the pH in the range 4-7.
- 2- Adsorption isotherms of Freundlich and Langmuir isotherms were used for studies and it was observed that most of them preferred Langmuir isotherms; also in general the result gave good agreement with endothermic and physical adsorption for most compounds.
- 3- Nano Al_2O_3 surface gave best removal percentage and less contact time in comparison with Al_2O_3 surface.
- 4- Adsorption kinetics for the adsorption processes follows the pseudo-second-order kinetic mode except "Cd (II)" in nano Al_2O_3 surface was fit to pseudo first-order.

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Hawraa Kassem Hami, Ruba Fahmi Abbas*, Emad Mahmoud Eltayef and Neda Ibrahim Mahdi
E-mail: rubaf1983@uomustansiriyah.edu.iq

Ruba Fahmi Abbas* (Department of Chemistry, College of Science, Al-Mustansirriyah University, Baghdad, Iraq)

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تطبيقات أكسيد الألومنيوم وأوكسيد الألومنيوم النانوي كمتز مراجعة

حوراء قاسم حامي، ربا فهمي عباس*، عماد محمود الطيف، نداء ابراهيم مهدي
قسم الكيمياء، كلية العلوم، الجامعة المستنصرية، بغداد، العراق (suha_rrr_1983@yahoo.com)

معلومات البحث:	الخلاصة:
تأريخ الاستلام: 2020/01/24 تأريخ القبول: 2020/04/05	تستخدم أكاسيد المعادن على نطاق واسع في تكنولوجيا الامتزاز كونها سطوح مازة بسبب كفاءتها، وانخفاض تكلفتها وخصائصها الفيزيائية الفريدة. تهدف هذه المقالة إلى توضيح دور أوكسيد الألومنيوم وأوكسيد الألومنيوم النانوي في إزالة بعض المواد الكيميائية التي تؤثر على صحة الإنسان مثل الأصباغ، والمضادات الحيوية والمعادن الثقيلة. وشملت أيضا تأثير بعض معاملات الامتزاز مثل درجة الحموضة ووقت الاتصال ونسبة الإزالة ودرجة الحرارة، وطبيعة الامتزاز، ونماذج الامتزاز الحركية ونماذج الأيزوثيرم.
الكلمات المفتاحية:	
أوكسيد الالمنيوم، نانو اوكسيد الالمنيوم، الممتز، اصباغ، مضادات حيوية	