



Uranium leaching followed by bio Sorption using *A. clavatus* of Gattar ore

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Abstract

The main objective of this work focused on choosing the most suitable conditions to leach uranium out from its mineral ores in G. Gattar Area, Northern Eastern Desert, Egypt. A sequence of primary experiments was conducted to select the most effective conditions for U extraction. The results showed that these conditions were usage of sulfuric acid at a concentration of 6molar, pH value of 2, contact time of 60 minutes, temperature of 100° C. *Aspergillus clavatus* fungi were tried as a new bio-sorbent of uranium ions. The appropriate factors to achieve the highest sorption efficiency of U by these fungi were studied and found to be 1 hour contact at pH 4, centrifugation speed of 300 rpm, and temperature of 100° C. The aforementioned indicate that the combination between leaching uranium by sulfuric acid and its bio-sorption by *Aspergillus clavatus* can be considered as a promising technique for removal or, at least, reduction of uranium concentration in uranium-contaminated regions such as G, Gattar, Northern Eastern Desert, Egypt.

Key words: Uranium, *A. clavatus*, leaching, adsorption

Introduction

The growing demands for the fossil energy became highly expensive. The increased awareness about the global carbon dioxide emission (global warming), forced scientists to find out a suitable alternative for the fossil energy (Michhaeldes and Michaelides, 2020). Nuclear energy seems to be a valid alternative to fossil fuels (Tsai and Huang, 2023). It is expected to grow annually by a percentage exceeding 1% (OECD/IAEA, 2010). Most of the uranium, which is the motive power of the nuclear energy stations, exists in the upper crust of soils as U⁴⁺ and U⁶⁺ (Rich et al., 1980). Usually, it is found as Uraninite (UO₂), which reacts with oxygen to form U₃O₈, the mineral pitchblende. Uranium extraction involves complex hydrometallurgical processes such as solid-liquid extraction, solvent extraction, and ion exchange to obtain pure triuranium octaoxide (U₃O₈) from uranium ore (Edwards and Oliver, 2000).

In Egypt, there are many areas of Uranium mining. G. Gattar Area, Northern Eastern Desert, Egypt. (latitudes 27° 07' 00" N to 27° 07' 47" and longitudes 33° 16' 50" E to 33° 18' 0") is an important one of these regions. Uranium leaching is the main process by which the uranium is extracted from the raw ore (Liu et al., 2023). This process is performed using either an alkaline or an acid leaching agent. The selection of leaching agent depends on some conditions among which the economics of the

process and the nature of the ore itself. Depending on the leaching agents used, the uranium production process also involves several precipitation and purification processes. Hence, leaching of these precipitates is required to get rid of these contaminants.

If the leaching solution is sufficiently clean, fractional crystallization can be applied straight to it, as in alkali carbonate leaching of uranium ore. That is rarely the case, particularly in As a result, either a solid/liquid or a liquid-liquid extraction would be required prior to purification. It may be required to use a combination of both processes in some circumstances. Apart from that, the eluate or the solvent extraction strip Solution would be tested. to Uranium precipitation in the form of a preliminary marketable Uranium product known as yellow cake, which would then be subjected to proper refining techniques to produce nuclear fuel material.

To create the preliminary commercial product, many techniques have been created and tested. (Yellow cake) from Uranium ores of various kinds Grinding Uranium ores to generate sized ore suitable for acid or alkaline leaching is the first step in traditional Uranium processing. For Uranium leaching from its ores, sulfuric acid or Na₂CO₃-NaHCO₃ systems are almost exclusively utilized. Several leaching processes have been used depending on the conditions, including a) atmospheric pressure leaching (acid and alkaline); b)

pressure leaching (acid and alkaline). (acid and alkaline); c) acid pugging and curing (acid); d)Heap leaching (acid); e)In situ leaching (mainly alkaline)

Green algal biomass, such as *Ulvalactuca* (Linnaeus) (green marine algae) and *Cladophora glomerata* (Linnaeus) Kützing (freshwater algae), are being studied for their ability to absorb Uranium from Gattar Pilot Planet treated effluent in Egypt's Eastern Desert. The highest biosorption efficacy of Uranium was achieved at 1 hour contact period and pH Furthermore, in their natural condition, each type of algae has a particular surface structure. FTIR was used to characterize the contributing groups, which were variable and contained several pathways depending on the type of Uranium and algae used. It suggests that Uranium biosorption is a single-layer physicochemical process on two masses of green algae (Mansour *et al.*, 2020)

With the advent of nuclear power plants, the research of Uranium extraction and recovery from ores and environmental samples has resulted in the formation of massive amounts of radioactive waste with significant environmental consequences (Kazy *et al.*, 2009). The ability of biomaterials to bind and concentrate heavy metals and radionuclides from aqueous solutions is known as biological adsorption. It is thought to be a cutting-edge method for eliminating pollutants from industrial effluents (Wang, 2002).

Because there are no toxicity issues, no need for growing media or nutrients, and simple technology for its capture and reuse, dead biomass is preferable to live biomass. (Mathialagan *et al.*, 2009). Polysaccharides and proteins in fungal cell walls include several functional groups (such as carboxyl, OH-, SO₄-, phosphate, and amino groups) that can bind metal ions. The use of freely dispersed biomass for biosorption has numerous disadvantages. (Liu *et al.*, 2003). Increased mechanical strength, density, chemical resistance, ease of separation of cells and effluents (solid and liquid), and high biomass loading/performance are only a few of the benefits of inactivated biomass. (Arica *et al.*, 2001) Agar, agarose, alginate, k-carrageenan, polyacrylamide, polyurethane, cellulose, collagen, chitin, chitosan, polysulfone, and epoxy resins have all been successfully employed for cell entrapment. (Akhtar *et al.*, 2009).

The heavy metal ion biosorption system is improved by trapping microbial cells in polymer supports. Pure resin is commercially available, and it has a consistent pore distribution, a large surface area, is durable, and has a chemically homogeneous structure. (Kocaoba and Arisoy, 2011) .

The main objective of this research is to purify the

soil G. Gattar Area, Northern Eastern Desert, Egypt from contamination with uranium resulting due to its extraction processes and hence making the land suitable for the agricultural use on one side and be safe from the environmental point of view on the other side.

Material and methods

A representative technological sample was collected from G. Gattar Area, Northern Eastern Desert, Egypt. This sample (2kg) was crushed, ground and passed through a 2 mm sieve thereafter was chemically analyzed to determine its contents of the major oxides such SiO₂, TiO₂, Al₂O₃, Fe₂O₃, MnO₂, MgO, CaO, Na₂O, K₂O, P₂O₅, and the trace elements UO₂, Re₂O₃, ThO₂ according to the methods outlined by Shapiro and Bronouk (1962).

Uranium leaching by acid

A portion of the previously prepared technological sample was exposed to dissolution process using H₂SO₄, HCl, H₃PO₄ and H₂CO₃ under different concentrations, pH values, contact times, temperatures, sample size fractions and liquid :solid ratios to select the optimum conditions to obtain the highest solubility efficiency of the uranium in the investigated technological sample.

Uranium sorption by *Aspergillus clavatus*

The *Aspergillus clavatus* fungi were selected as the bio-sorbent of uranium and its efficiency in sorption of uranium was tried under several factors, namely the weight of the *Aspergillus clavatus*, contact time, pH, stirring rate and temperature.

Reagents of the study and chemical analysis

The chemicals and reagents used in this study are of extremely high purity. De-ionized water was used for preparation of all the solutions and standards. Radioactive element Uranium was measured using arsenazo III as described by Marczenko (1976) and titration was conducted as in Mathew *et al.* (2009).

Results and Discussions

Data presented in Table (1) reveal concentrations of the elemental oxides of the major elements while those shown in Table (2) illustrate the elemental oxides of the trace elements found in G Gattar technological sample. It is obvious from Table (2) that concentration of uranium in this sample was about 70 mg kg⁻¹. This concentration seems to be the highest among the trace elements. Therefore, the current study aimed at getting rid or, at least, reducing the concentration of uranium through leaching of this element with a suitable acid at optimum conditions for leaching e.g. type of the leaching agent, particle size fraction of the technological sample, its molar concentration, pH

value of the leaching process ,period of contact between the leaching agent and the technological soil sample under leaching .

Table 1. Major element oxides (wt. %).

Ore	SiO ₂	TiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MnO ₂	MgO	CaO	Na ₂ O	K ₂ O	P ₂ O ₅	L.O.I
wt. %	74.75	00.09	13.47	01.30	00.02	00.20	00.52	03.85	04.55	00.11	00.77

Trace elements

Table 2. Trace element, (mg kg⁻¹).

Element	Cu	Zn	Co	Ni	U	Pb	Cd	V	Mo	Mn	Sr
mg/kg	34	26	15	8	70	12	6	11	8	26	36

Uranium leaching process was,therefore, studied taken into account the aforementioned factors.

1--Effect of leaching agent type on the efficiency of uranium leaching percentage

The Effect of leaching uranium from G Gattat ore sample with several acids i.e. sulfuric acid, HCl, HNO₃ and H₃PO₄ at a concentration of 6 molar

was conducted to find out which one of these acids is the most efficient in leaching uranium from the studied technological sample. Results showed that all the acids under study could dissolve uranium, however, H₂SO₄ seemed to be the best acid in dissolving uranium from the ore sample under study as shown in Figure. (1)

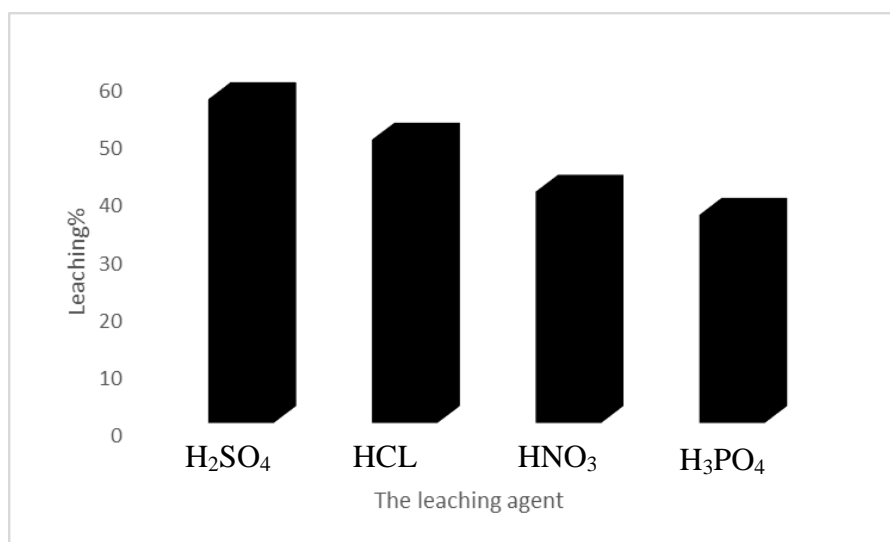


Figure (1): Effect of Uranium leaching agent type on the efficiency of Uranium leaching.

2-Effect of the sample grain size fraction on efficiency of uranium leaching percentage

To study the effect of the sample grain size on uranium removal, eight samples with scales of -2.5, 5-, 10-, 15-, 20-, 30-, 40-, and -50mesh were leached with portions of sulfuric acid 100 mL each with stirring continuously for an hour at room temperature. Results shown in Figure (2) reveal that the best leaching percentage of uranium occurred at the size fraction of -30 mesh .It is expected , however, that dissolution rate of U increases as the particle size fraction decreases due to the increase in

contact surfacethat, accompanies the decrease in particle size per the solid. This is since; smaller particles have a large surface area available for carrying out metal's dissolution (Manaa et al.,2020). This factis not absolute, where the dissolution of uranium observed herein achieved maximum value at a particle size fraction of -30 mesh but at particle size fraction less than that fraction size, dissolution of uranium decreased. This finding agrees with that of Manna et al. (2020) who indicated that dissolution of uranium was not affected by increasing fineness of particles beyond a certain fraction size .

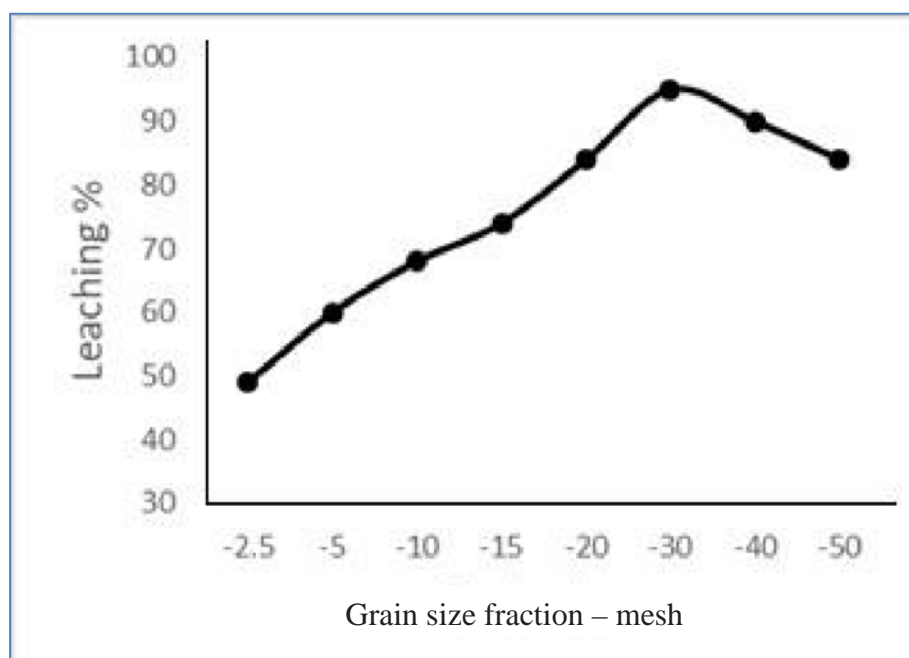


Fig (2): Effect of t grain size fraction on uranium leaching efficiency

3- Effect of the different concentrations of sulfuric acid on efficiency of uranium leaching percentage.

The effects of molarity of the sulfuric acid i.e., 1, 2, 4, 6, 8 and 10 M on the leaching of uranium from the investigated technological sample of G

Gattar ore sample at the room temperature, rain size fraction -30 mesh, pH 3-, and 45-minute contact time are shown in Fig. (3). The best leaching efficiency i.e., 90% was attained upon using sulfuric acid at a concentration of 6 M .

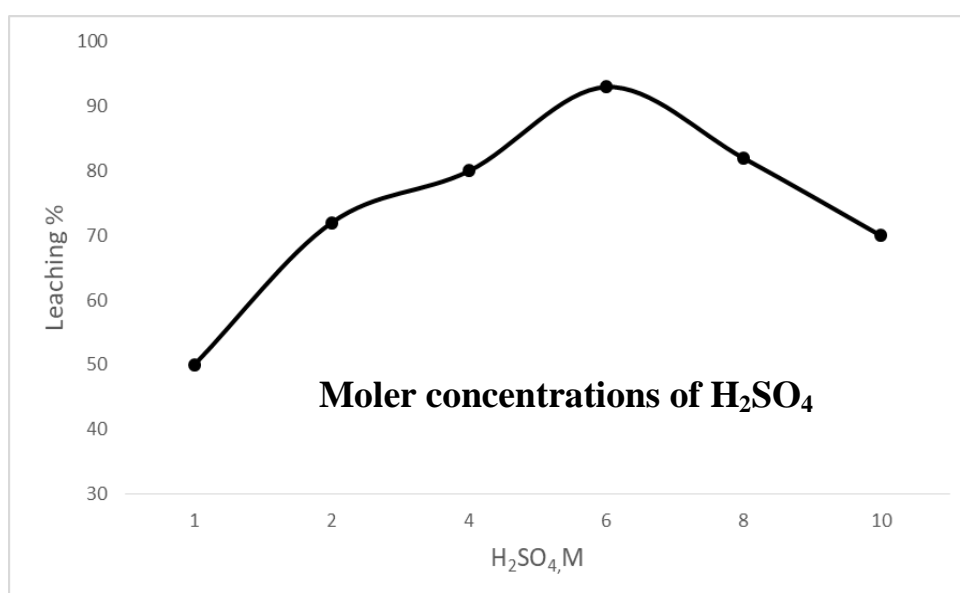


Fig (3): Effect of different concentrations of sulfuric acid on leaching efficiency of uranium

4-Effect of the pH value of the solution on efficiency of uranium leaching percentage

Fig. (4) shows the effects of pH value on the leaching of uranium by sulfuric acid within the pH range of 1: 3.5 to find out what is best one for leaching uranium from the ore at a temperature of 25 Celsius degrees

for 45 minutes and sizeore fraction of 30 –mesh. Results showed that the best pH in this concern was

2 since it resulted in leaching efficiency of uranium estimated by 92% .

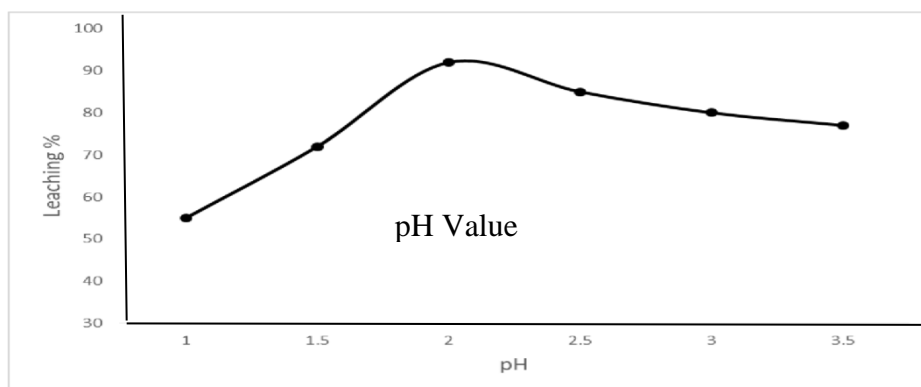


Fig (4): Effect of pH value on leachin efficiency of uranium

5-Effect of contact time on efficiency of uranium leaching percentage

The effects of contact time between the applied sulfuric acid and the ore sample at an ore size fraction of 30- mesh, sulfuric acid concentration of 6 M, pH value of 2, and at room temperature for a period ranging from 15 to 120 minutes are shown in

Fig. (5).. It was found that the best time of contact was 60 minutes, where it caused leaching estimated by 91% for uranium content of the ore sample . The results of Rashad et al. (2020) ,however, indicated that increasing the contact time from 40 to 240 min. increased the leaching efficiency of U up to 67.77% .

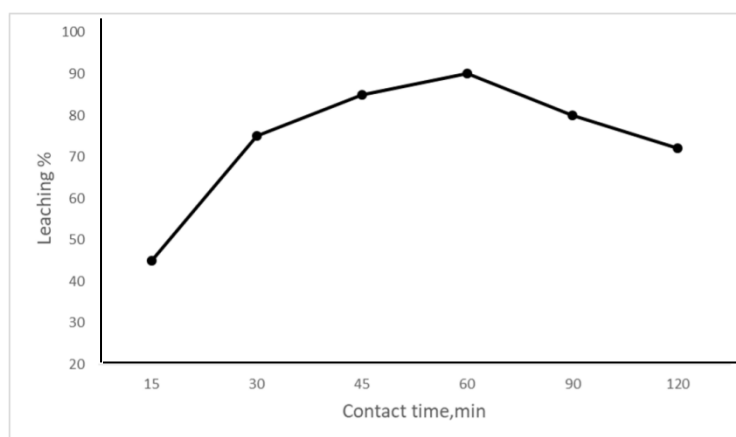


Fig. (5): Effect of contact time on leaching efficiency of uranium

6 - Effect of solid / liquid (S/L) ratio on efficiency of uranium leaching percentage

Table (6) shows the effects of the solid/liquid ratio on the leaching efficiency of uranium under the conditions of contact time 60 minutes, sulfuric acid concentration 6 M, room temperature (25 °C), a particle size fraction of - 30 mesh and S/L ratios

varied between 1/1 to 1/4. It was found that the best solid-to-liquid ratio is 1/1, where 91% of uranium in the investigated sample was leached. Rashad et al. (2020) indicated ,however ,that the best leaching efficiency of U, 58% , was attained at S/L ratio of 1/4.

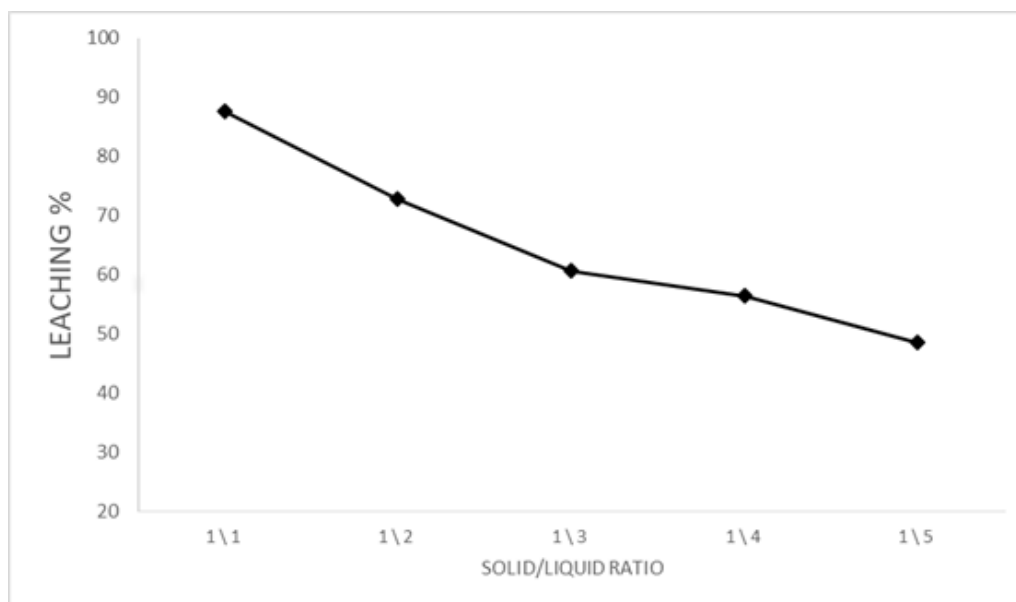


Fig (6): Effect of S/L ratio on efficiency of uranium leaching percentage

7-Effect of temperature on efficiency of uranium leaching percentage

The effects of temperature on the leaching efficiency of uranium have been studied through several experiments carried out at temperatures of 25, 35, 45, 55, 70, 85 and 100 ° C under constant conditions

of sulfuric acid concentration at 6 M, pH value of 2 and contact time of 60 min., particle sample size fraction of 30- mesh and ratio of solid to liquid of 1/1 .The results showed that the best temperature was 100 Celsius, where the dissolution percentage of uranium reached 93%, as shown in Fig. (7).

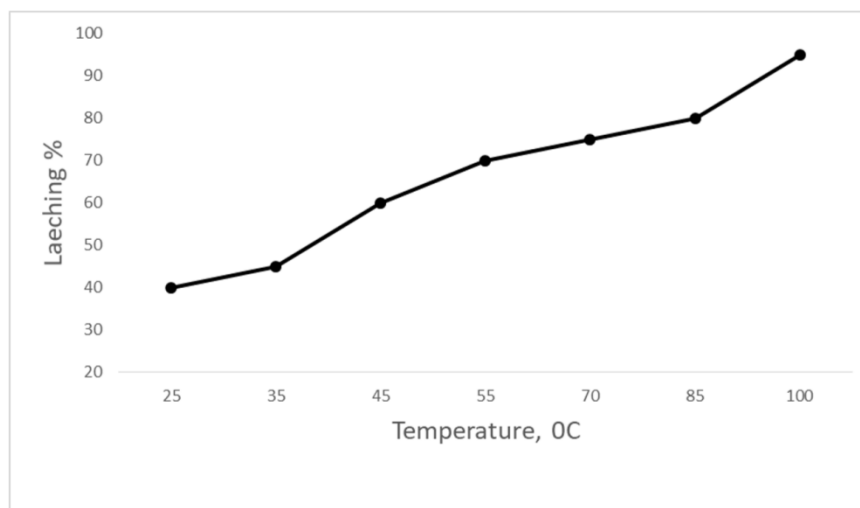


Fig (7): Effect of temperature on efficiency of uranium leaching percentage

Summary of the total optimum conditions for uranium leaching process:

- The best acid is H₂ SO₄,
- the best particle size fraction is 30-mesh,
- The best acid concentration is 6M,
- The best pH is 2

- The best contact time is 60 minutes
- The best ratio of solid to liquid is 1/1
- The best temperature is 100 C⁰

Uranium bio-sorption by *Aspergillus clavatus*

A sample weighing 1/2 kg of *Aspergillus clavatus* was prepared and tested for selecting the best conditions for uranium sorption through previous experiments. Uranium filtration was done until we got a 2-liter solution, and then the best conditions for uranium sorption by *Aspergillus clavatus* were tried as shown hereafter.

1-Effect of weight of the fungi

A quantity of fungi differing in weight from 0.1 to 0.7 g was placed in a 100 mL portions of the uranium filtrate to find out the highest percentage of sorbed uranium under room temperature conditions, contact time of 120 minutes, pH 3, and stirring speed of 200 rpm. The results revealed that the 0.5 gram weight of the *Aspergillus clavatus* was the fungi weight that could sorb the highest percentage of uranium i.e.85%, as shown in Fig(8).

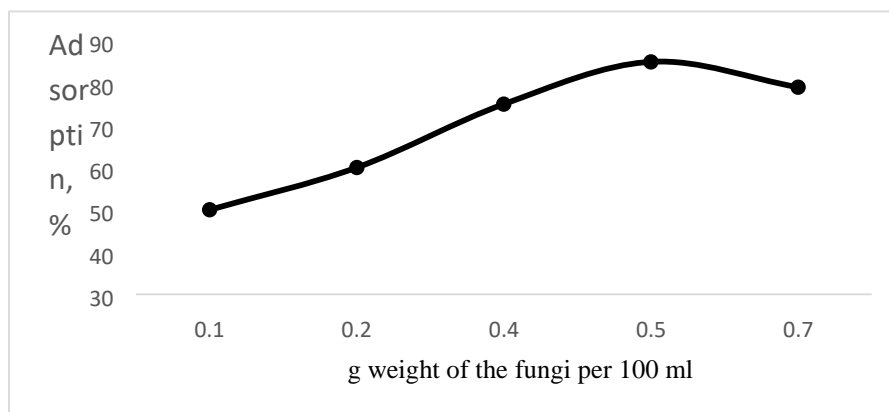


Fig. (8): Effect of the weight of the fungi

2-Effect of time of contact

Weights of 0.5 g portions of the fungi were placed in 100 mL of the uranium solution and left for periods ranging from 30 to 180 minutes to find out the best period for uranium sorption from the solution. The

pH was 2 and the stirring speed was 500 rpm. It was found that the contact time was 60 minutes that caused the highest sorption i.e.90% as shown in Fig.(9)



Fig. (9): Effect of time of contact

3- Effect of the solution pH

Solutions of different pH values ranging from 1-6 were prepared to find out the best pH value for sorption of uranium from the solution. The amount of fungus was half a gram, the contact time was 60

minutes, the stirring speed was 250 rpm, and the temperature was 50 Celsius degrees. Results showed that the best pH value was 4, where sorption of uranium achieved 90%, as shown in Fig.(10)

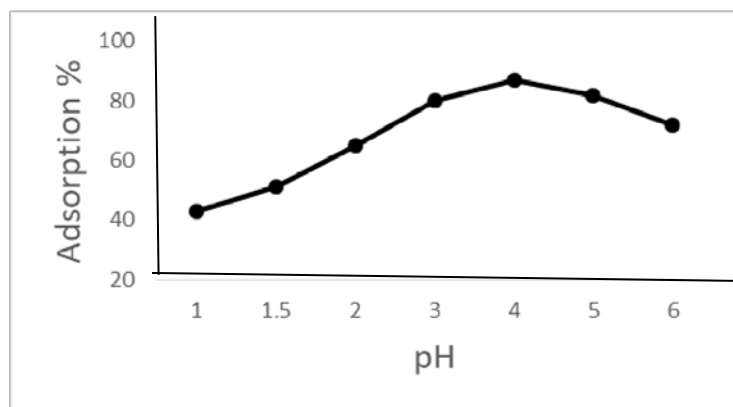


Fig (10): Effect of the solution pH

4-Effect of the speed of flipping

Several solutions were prepared, the volume of each one was 100 mL, in each of which half a gram of the fungi was placed. The pH was 4, the contact time was 60 minutes, and the temperature was 60 Celsius

degrees. The speed of flipping in these solutions was either 100, 200, 300, 400 or 500 rpm. Results indicated that the speed 300 rpm was the flipping speed which caused the highest sorption by the fungi i.e., 91% as shown in Fig. (11)

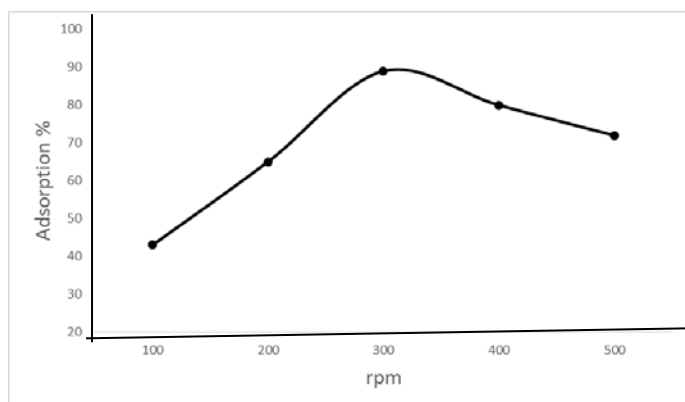


Fig (11): Effect of the speed of stirring

5-Effect of temperature

Several solutions were prepared, and a weight of half a gram of fungi was placed in each of them. The contact time was 60 minutes, the pH was 4, and the stirring speed was 300. They were placed under different temperatures 25, 40, 55, 75, 90, and 100

Celsius degrees to find out which of these temperatures is the best for uranium sorption. The experiments showed that the best temperature is 100 Celsius degrees, where the sorption percentage was 93 %, as shown in Fig. (12).

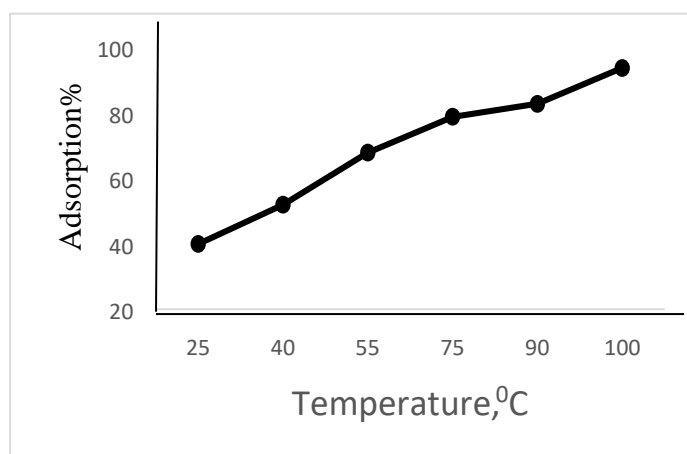


Fig (12): Effect of temperature on adsorption

As a final summary of the aforementioned experiments, it can be deduced that the most optimum.

conditions for sorption of uranium by *Aspergillus clavatus* fungi are:

- a half g portion of the fungi
- 60 minutes contact time.
- pH value of 4,
- 300 rpm stirring speed.
- 100 Celsius degrees.

Conclusion

Leaching uranium ore by sulfuric acid at a molar concentration of 6 M and optimum conditions of liquid – solid ratio ,contact time and temperature seems to be a suitable technique of leaching uranium from its ores .However, bio-sorption of the leached uranium by *Aspergillus clavatus* fungi would be a good complementary technique for bio-remediation of the ores from uranium .Hence it is recommended to use a combined technique comprised of leaching and bio-sorption to reduce processing costs of uranium decontamination and ,at the same time, keep the soil safe from the environmental point of view.

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اذالة التلوث باليورانيوم من منطقة جتار بالصحراء الشرقية بالغسيل بحامض واسترداد اليورانيوم المغسول بالامتصاص الحيوى بفطر كلافاتس

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1- هيئة المواد النووية، صندوق بريد 530 المعادى، القاهرة، مصر

2- قسم الاراضى والمياه، كلية الزراعة، جامعة بنها، مصر

تقع منطقة الدراسة فى جتار شمال غرب مدينة الغردقه فى الصحراء الشرقيه بين خط عرض 27 درجة شرقا و خط طول 33 درجة شمالا . فى هذه المنطقه يتم التخلص من النفايات الاشعاعية المتبقية بعد عمل عجيبة اليورانيوم و بالتبعية فان اراضي هذه المنطقه اصبحت عرضة للتلوث باليورانيوم و ذلك فان البحث الحالي يمثل محاولة استهدفت بصفة رئيسية معالجة التربه المستقبلة لهذه النفايات من الملوثات الاشعاعيه الناتجه من وجود اليورانيوم وحتى تكون كفاءه الخالص اعلي ما يمكن تم المفاضلة بين مجموعه من الاحماض اثبتت التجارب أن أفضلها فى غسيل اليورانيوم هو حامض الكبريتيك بتركيز 6 مولر عند رقم حموضة و نسبة من العينة الي محلول الاستخلاص مقدارها و فى ظل زمن تلامس بين العينة و الحامض مقداره دقيقة . هذا

ثم تم امتصاص اليورانيوم باستخدام فطر الكلافاتس المعالج باستخدام هيدروكسيد الصوديوم حيث انه صديق للبيئه و الحصول عليه سهل و رخيص الثمن بعد اختياره من عدة فطريات حيث تبين انه الافضل و الاعلى كفاءه فى نسبة الامتصاص مع دراسة العديد من المعاملات كتاثير كمية الماده الماصه و تاثير زمن التلامس و تاثير الاس الهيدروجينى و تاثير سرعه الاذابه و تاثير درجه الحراره و كذلك تاثير نوع الحمض فى اعاده و استرجاع اليورانيوم من فطر الكلافاتس