Quest Journals
Journal of Research in Environmental and Earth Sciences
Volume 7 ~ Issue 6 (2021) pp: 01-09
ISSN(Online) :2348-2532
www.questjournals.org

Research Paper

Leaching Study to Decide the Suitable Leaching Device for Uranium Extraction from Gattar Ore, Eastern Desert, Egypt

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ABSTRACT
The importance of Gattar uranium occurrence (Eastern Desert, Egypt) paid attention to further experimental works. The wet chemical processing of ore technological sample was carried out to choose the suitable uranium leaching device regarding the concentration and amount of the chemical leaching agent, the economic percent of the consumed water and the total of contact time to obtain the maximum uranium leaching efficiency.
To achieve the present work, two leaching devices, agitation and vat percolation leaching were used to perform all the different leaching experiments. Uranium concentration of the Gattar collected mineralized sample was about 1293 $mgkg^{-1}$. The obtained optimum leaching conditions of the agitation device were: sulfuric acid 70 g/l concentration as a leaching agent, - 0.125 to + 0.09 mm sample grain size, 14% total percent of acid amount used based on weight of the sample, 161.1% water used based on weight of the sample while it was 80.5% based on the total volume of solution at 32.5% saturation percent and 120 min. contact time, 1/2 solid liquid ratio and 150 rpm at room temperature. According to these conditions, the obtained uranium leaching efficiency attained 92.1%.
On the other hand, in the case of vat percolation leaching under the following optimum condition: sulfuric acid 60 g/l concentration as a leaching agent, 1/3 solid/liquid ratio, - 0.125 to + 0.09 mm grain size, 22% total percent of acid amount used based on weight of the sample, 262.32 % percent of water used based on weight of the sample while it was 87.5% based on the total volume of solution at 27.77% saturation percent and 3 hours contact time at room temperature. The uranium leaching efficiency obtained was 93.5%.
Comparing the obtained results directing the decision to the agitation leaching device as the suitable technique for uranium leaching applying the most economic conditions especially the concentration and amount of the chemical leaching agent, consumed water amount and the taken time.
KEY WORDS: *Uranium - Extraction - leaching technique – Gabal Gattar*

Received 25 May, 2021; Revised: 06 June, 2021; Accepted 08 June, 2021 © The author(s) 2021. Published with open access at www.questjournals.org

I. INTRODUCTION

The area of Gabal Gattar ore is located in the Central Eastern Desert of Egypt (35 km to the west of Hurghada City. The mineralized sample of the study area revealed that the main uranium minerals are represented by uranophane and beta uranophane in association with hematite and magnetite which could be referred to the effect of hydrothermal solutions up on the mineralization **(Salaman et. al., 1990)**.

Khalaf, (1995) indicated the presence of uranophane, beta- uranophane as secondary uranium minerals. She added that fluorite and iron oxides are always associated with the uranium mineralization.

El Hazek et al., (1996) studied the distribution and recovery of uranium and molybdenum from their minerals at Gabal Gattar area. He identified some secondary uranium minerals such as uranophane, beta-uranophane and masuyite. Molybdenite, as a representative mineral for molybdenum ore, has also been identified. A technical flow sheet for the extraction of both uranium and molybdenum was proposed at the end of the leaching study.

This zone was investigated at depth by mapping the surface features and projecting the geometry to the subsurface. Primary uranium mineralization was first time discovered in the tunnel by the geological team of G. Gattar prospect **(El-Feky et al., 2004)**.

Abd El-Naby, (2009) mentioned that secondary uranium mineralization is controlled by shear zones in which the degree of fluid-rock interaction was very high and argillic alteration is abundant. The argillic alteration, represented mainly by kaolinite and illite, played an important role in uranophane precipitation within the altered parts of the Gattar granites. Presence of calcite as void-filling in association with uranophane within

the shear zones of Gattar granites may indicate that uranium was probably transported as uranyl-carbonate complexes. The sorption of U (VI) in the Gattar clay minerals should be low due to the presence of dissolved carbonate ions and to the slightly alkaline conditions under which U (VI) occurs as the weakly sorbed $UO_2(CO_3)_3^{4-}$. U could be removed from solution through dissolution of feldspar, formation of clay minerals and subsequent precipitation of uranophane. Dissolution of the feldspars resulted in a diffusional gradient of Ca and SiO_4 ions away from its surface. Formation of uranophane would likely form near the clay minerals surface, where there was increased probability of successful interaction between U and both Ca and SiO_4. As the uranium loading increases, surface-precipitation of uranophane on clay mineral surfaces can occur.

El-Feky, (2011) recorded different forms of uranium mineralization by autoradiographic studies mainly as massive or disseminated mineralization along tectonic fractures, interstitially in granular minerals, and as cements of breccias. Hydrothermal alteration is found in association with primary (uraninite, pitchblende and coffinite) and secondary uranium mineralization (uranophane and kasolite), sulphides (galena, chalcopyrite, pyrrohtite and pyrite) and iron oxides (Hematite and magnetite).

However, the presence of silica gangue minerals (feldspars and quartz) which are inert for acid attack encouraged the application of acid leaching technique (with sulfuric acid). In addition, uranium present in the study sample is leachable. This is due to the fact that the ore is of oxidized nature (the predominance of uranophane). In general, the acid leaching is more widely used than alkaline leaching because it needs only relatively coarse preparatory grinding and comparatively mild reagent concentration, requires shorter leaching time and usually applied under normal atmospheric temperature and pressure (**Woody et al., (1958).**

Several leaching techniques are available for extracting uranium from its ore such as atmospheric agitation leaching and also, pressure (autoclave) leaching **(Boydell and Viljoen (1982)** and **Girmes (1982).**

The present work was carried out to choose the suitable uranium leaching device to investigate the maximum uranium extraction from the representative technological sample from Gabal Gattar area, using two leaching devices: the first device is the agitation and the second is the vat percolation leaching. The study technological sample was found to assay 1293 mg/kg uranium.

II. MATERIAL AND METHODS

To achieve the main purpose of the present study, a representative technological sample was collected from Gattar (GII) uranium occurrence area. The study sample was properly crushed by jaw crusher and sieves to one grain size. A part of this sample was ground to (200) mesh size for complete chemical analysis involving both major element oxides and some of trace elements with emphasis on uranium as the main objective radioactive element in the present work (Table **(1, 2).** Another part was ground to (-0.125 +0.09 mm) for the technological experiments. The chemical analysis of the study sample was determined in the labs. of the Nuclear Material Authority of Egypt (NMA).

Table (1): Chemical analysis of major oxides of Gattar study sample

Element oxide	Wt.%	Element oxide	Wt.%
SiO_2	70.07	K_2O	4.16
Al_2O_3	12.42	Na_2O	3.95
Fe_2O_3	5.1	Mn O	0.19
Ca O	0.48	TiO_2	0.1 5
P_2O_5	0.13	L.O. I	1.3
MgO	0.35	Total	99.32

Table (2): Trace elements analysis of Gattar study sample

Element	$mgkg^{-1}$	Element	$mgkg^{-1}$
Cu	8.5	Co	437
Mn	24.7	Ni	74
Cd	3.4	U	1293
Pb	0.43		
Zn	30.60		

2.1. Leaching operation

In general, there are two main leaching methods for uranium element dissolution: acid and alkali leaching (agitation, percolation, bug and pressure). In the present work, leaching process was carried out using two devices, acid agitation and acid vat percolation leaching.

2.1.1. Agitation leaching device

Uranium leaching efficiency was studied under the effect of acid concentration and percent of total amount used, total time taken and consumed amount of water. In addition to, solid/liquid ratio, revolution per min. and grain size at room temperature. Each agitation leaching experiment was performed by mixing a proper weight (10 grams) of the study sample with certain volume of sulfuric acid solution at different concentrations. The obtained slurry after agitation for a certain time was filtered and the residue was washed and the solution was completed to a fixed volume where it was chemically analyzed for uranium content.

2. 1. 2. Vat percolation leaching device

Percolation leaching is passing the leaching solution through porous material of Gattar study sample. First, the study sample was crushed, sieved, ground and mixed well to obtain the suitable particles size. A glass column was packed with 10 grams study sample. Different sizes of pebbles of grid or glass wool was put into the bottom of the vat as support. The obtained leached solutions were sampled for uranium analysis to determine the uranium leaching efficiency attained.

2.2. Analytical procedures

A representative part of the collected technological sample of Gattar area was subjected to complete chemical analysis for major elements oxides using wet chemical methods of **Shapiro and Barnnock (1962).** To follow the uranium leaching efficiency, the collected leach liquors of the working sample were subjected to uranium analysis by using the oxidemetric titration method against ammonium metavanadate (**Davies and Gray, 1964**).

III. RESULTS AND DISCUSSION

In the present work, a comparative study between uranium agitation and percolation leaching devices was performed. For this purpose, uranium leaching efficiency was investigated under most of the effective factors especially the concentration and used amount of acid, taken time, the percent of water amount used, the solid/liquid ratio, the revolution per minute and the particles size.

3.1. Agitation leaching device.

3.1.1. Uranium distribution along different particle size and its saturation percent

Uranium content was analyzed in various particle size to suggest the optimum size range for the following experiments. From **Table (3)** it is clear that the high percent (50.24%) found in the range (- 0.125 and + 0.09) which represent 37.78 wt.% and the saturation percent attained 32.5%.

Table (3): Uranium distribution along various particle size and saturation %

Ore size, mm	Weight %	Uranium conc., mg/kg	Uranium content, (%)	Saturation %
- 0.5 to + 0.25	29.1	296.77	22.95	29.2
- 0.25 to + 0.125	32.56	346.44	26.79	30.7
- 0.125 to + 0.09	37.78	649.70	50.24	*32.5
Total	99.46	1293.2	99.98	

3.1.2. Effect of leaching agent type

Uranium leaching efficiency was studied using different types of acid and alkali to determine the best leaching agent can be used. This factor was studied under the following conditions: 100 g/l chemical agent concentration, 30% weight of acid based on weight of the study sample, - 0.5 mm particle size**,** 100 rpm.**,** 1 hour contact time, (1/3) solid/liquid ratio and 31.5% saturation percent at room temperature.

The obtained results **(Table 4)** showed that, uranium leaching efficient by sulfuric acid solution attained 47.98% which is a little low than that of HCl solution (49.3 %) while the other leaching agents examined gave lower efficiencies. Sulphuric acid is the most common acidic reagent used in uranium leaching because of its availability and low cost. In addition, sulphoric acid may be generated autogenously by treating uranium ores that contain sulphide minerals by air or oxygen under pressure or by bacterial action.

Table (4): Effect of the leaching agent type upon uranium leaching efficiency %

Leaching agent type	Uranium leaching efficiency (%)
H_2SO_4	47.98
HCl	49.30
HNO_3	46.36
Citric acid	40.36
$Na_2CO_3/NaHCO_3$(1:1)	26.49
Na OH /Na_2CO_3(1:1)	13.24

3.1.3. Effect of grain size

Effect of grain size of the study sample upon uranium leaching efficiency was studied using different sizes ranging between **(**- 0.5 + 0.25 mm) to (- 0.125 + 0.09 mm) to determine the optimum grain size can be used. This factor was studied under the following conditions: (100 g/l) H_2SO_4 concentration, (30%) amount (w/w) of acid, (1/3) S/L ratio, (100) rpm and 1 hour agitation time at room temperature. The obtained results (Table 5) indicated that, the maximum leaching efficiency attained (64.58%) at (- 0.125 + 0.09 mm) grain size while the other sizes gave 56.4% and 47.98% respectively. So, the optimum grain size was - 0.125 to + 0.09 mm.

Table (5): Effect of grain size upon uranium leaching efficiency

Grain size, mm	Uranium leaching efficiency%
- 0.5 to + 0.25	47.98
- 0.25 to + 0.125	56.4
- 0.125 to + 0.09	64.58

3.1.4. Effect of acid concentration, its amount and the used water

Effect of acid concentration and its amount upon uranium leaching efficiency were studied between (20 and 100 g/l) and between (6 and 30%) amount per sample. under the following conditions: (-0.125 +0.09) mm grain size, (1/3) S/L ratio, (100) rpm and 1 hour stirring time at room temperature. On the other side, the percent of water used ranged between (88.1 and 84.8%) depending on the total volume of solution and between (265.2 to 251.9%) based on weight of the sample. Results of uranium leaching efficiency (Table 6) indicated that the optimum acid concentration is (60 g/l) and 18% of used amount based on the weight of sample which gives (62.93%) uranium leaching efficiency. Also**,** 86% of water the used based on the total volume of the used solution or 258.1% based on the sample.

3.1.5. Effect of solid/liquid ratio

In this factor, the highest uranium leaching efficiency (Table 7) was that at solid/liquid ratio 1/5 giving (66.2%) while from the economic point of view the solid/liquid ratio of 1/2 which give (62.6%) leaching efficiency at 12 % as weight of acid per weight of the sample, 80.5% of water used per solution and consuming a little amount of acid and water at (60 g/l acid concentration, (- 0.125 + 0.09 mm) grain size, 100 rpm and 1 hour stirring time at room temperature). On the other hand, weight of water percent per weight of the sample became 161.1% comparing to that obtained from the previous factor (258.1%).

Table (6): Effect of acid concentration, its amount and water used upon uranium leaching efficiency

Acid leaching solution			ore Grain size, (mm)	Solid /liquid ratio	Used water			U. leaching efficiency (%)	Round per (minute)	Stirring time (hour)
Type	Conc, g/l	% of used amount based on weight of ore	-0.125 to +0.09	1/3	saturation percent %	Percent of used water%			100	1
					32.5	Based on weight of the ore	Based on the total volume of solution			
H_2SO_4	20	6				264.0	88.1	45.04		
	40	12				261.0	87.1	47.68		
	*60	*18				258.1	86.0	*62.93		
	80	24				254.5	85.0	63.22		
	100	30				251.3	84.3	64.58		

Table (7): Effect of solid/liquid ratio upon uranium leaching efficiency

Acid leaching solution			Solid/ liquid ratio	Grain size, (mm)	Water used			Uranium leaching efficiency%
					Saturation percent	Percent of water used		
Type	Conc., g/l	% of used amount based on weight of ore		(-0.125 to +0.09)	32.5	Based on weight of the ore	Based on the total volume of solution	
H_2SO_4	60 g/l	6.0	1/1			64.2	64.2	51.7
		*12.0	*1/2			161.1	80.5	*62.6
		18.0	1/3			258.1	86.1	63.5
		24.0	1/4			354.5	88.6	64.9
		30.0	1/5			451.9	90.4	66.2

3.1.6. Effect of stirring time

This factor was studied under the following conditions: 60 g/l acid concentration, 1/2 solid /liquid ratio, (- 0.125 + 0.09 mm) grain size, 100 rpm and 1 hour stirring time at room temperature and 161.1% of water based on the sample. Uranium leaching efficiency obtained after 2 hours stirring time (69.3%) while that after 4 hours attained 73.9%. For the economic aspect it was decided to continue this study with 2 hours contact time.

Table (8): Effect of agitation time upon uranium leaching efficiency

Acid leaching solution			Solid/ liquid ratio	Agitation time, Hour	Grain size, (mm)	Water used			Uranium leaching efficiency %
Type	Conc., g/l	% of used amount based on weight of ore				Saturation Percent	Percent of water used		
					-0.125 +0.09	32.5	Based on weight of the sample	Based on the total volume of solution	
*H_2SO_4	60g/l	12	1/2						
				1			161.1	80.5	62.6
				*2					69.3
				3					72.5
				4					73.9

3.1.7. Effect of agitation speed (rpm)

Investigation of uranium leaching efficiency through a range of agitation speed (50, 100, 150, 200 rpm) (Table 9), it was concluded that 150 rpm is the optimum speed giving 75.2% at 60g/l acid concentration, 12 % of used amount based on weight of sample, 2 hour stirring time, 1/2 solid/ liquid ratio and 161.1% of the used pregnant water based on weight of sample while 80.5% based on solution.

Table (9): Effect of agitation speed (rpm) upon uranium leaching efficiency

Acid leaching solution			Solid/ liquid ratio	Stirring time, Hour	agitation speed (rpm)	ore Grain size, (mm)	Used water			U. leaching efficiency %
type	Conc., g/l	% of used amount based on weight of ore					saturation percent%	Percent of used water%		
						-0.125 to +0.09	32.5	Based on weight of the ore	Based on the total volume of solution	
H_2SO_4	60g/l	12	1/2	2.0				161.1	80.5	
					50					47.9
					100					69.3
					*150					*75.2
					200					76.16

3.1.8. Effect of excess amount of acid

Estimation of the amount of the used acid percent based on weight of sample (12%) led to the ability of raising its amount percent and consequently the acid concentration as follow: 15%= 75 g/l, 14%= 70 g/l, 12%= 60 g/l, 10%= 49 g/l, 6%= 30 g/l (Table 10). According to achieve the optimum uranium leaching efficiency using the lowest amount of acid and water, it was decided to use the 15% acid amount (75 g/l acid concentration), where it reached to 94.94% uranium leaching efficiency.

Table (10): Effect of excess amount of acid percent upon uranium leaching efficiency

Acid leaching solution			S/L ratio	Stir., Time, hour	rpm	Ore grain size, mm	Used water			Additive amount of acid (% of used amount based on weight of ore)		Leaching Efficiency %
Type	Conc., g/l	of used amount based on weight of ore, %					SP%	Used water %		(%)	Conc., g/l	
							32.5	Based on weight of the ore	Based on the total volume of solution	15	75	94.94
										14	70	92.1
										12	60	75.2
										10	49	70.2
*H_2SO_4	*60	*12	*1/2	*2	*150	0.125 to +0.09		161.1	80.5	6.0	30	61.2

Summing up the agitation leaching device study, the obtained optimum conditions for uranium leaching efficiency are:

- Type of leaching agent: sulfuric acid.
- Total percent of acid amount used based on weight of the ore: 14%.
- Percent of water used based on weight of the sample: 161.1% while it is 80.5% based on the total volume of solution.
- Total agitation time: 2 hours.
- Grain size: - 0125 + 0.09 mm.
- Solid/liquid ratio: 1/2.
- Uranium leaching efficiency: 92.1%.

3.2. Vat percolation leaching device

3.2.1. Effect of sample permeability

The nature and the particles size of the sample must suit this technique where the flow rate of the penetrating leaching (sprinkling) solution through a porous material such as the working sample is the first parameter we should determine. This factor was examined through a range of flow rate from 0.25 to 1.25 ml/min. in the prepared glass column. The obtained results (Table 11) indicated that the highest and optimum flow rate is (0.75 ml/min.) at which the leaching solution cannot be flooded.

Table (11): Effect of permeability on the flow rate

Particle size, Mm	Flow rate of solution (ml/min.)	Flooding stage
-0.125 to + 0.09	0.25	No flooding
	0.5	No flooding
	0.75	No flooding
	1.0	Flooding
	1.25	Flooding

3.2.2. Effect of acid concentration

The packed study sample in the glass column was subjected to sulfuric acid leaching using different concentrations (20, 40, 60, 80 and 100 g/l). The other fixed factors were the grain size - 0.125 + 0.09 mm, (1/3) solid/liquid ratio, heights flow rate of solution (0.75 ml/min.), rate of out let solution was about 0.2 ml/min. at room temperature. The obtained results (Table 12) showed that, uranium leaching efficiency at 20g/l acid concentration was 61.5 and 6.1% acid percent based on the sample while at 100 g/l it reached to 84.6% and 30% acid percent based on the sample. Economically, the suitable acid concentration was 60 g/l where 18.0% weight of acid consumption, acid / sample giving 78.85% uranium leaching efficiency. On the other hand, the percent of the pregnant water used based on sample and solution were 262.5% and 87.5% respectively, also the saturation percent attained about (27.77%).

Table (12): Effect of acid concentration and its amount upon uranium leaching efficiency

Acid leaching solution			Solid/ liquid ratio	Water used			Uranium leaching efficiency, based on solution (%)	Grain size, mm.
				Saturation %	Water used %			
Type	Conc., g/l	% of used amount based on weight of sample	1/3	27.7	Based on weight of the sample	Based on the total volume of solution		-0.125 +0.09
H_2SO_4								
	20	6			269.0	89.70	61.5	
	40	12			265.8	88.6	70.16	
	60	18			262.5	87.5	78.85	
	80	24			259.3	86.4	81.0	
	100	30.			256.1	85.3	84.6	

3.2.3. Effect of sold/liquid ratio

The effect of solid/liquid ratio upon uranium leaching efficiency using vat percolation leaching device was studied from 1:1 to 1/5. This factor was performed under the following experimental fixed conditions: 60 g/l acid concentration, (- 0.125 to + 0.09 mm) grain size, (0.75 ml/min.) rate of in let solution and 3 hours total leaching time at room temperature. The obtained data (Table 13) showed that, (1/3) solid/liquid ratio is

considered the optimum ratio at which the uranium leaching efficiency attained (78.85%). It is important to mention here that although the S/L ratio of 1/5 gave the highest uranium leaching efficiency (84.6%), the amount and the volume of acid (5 times) is not economic.

Table (13): Effect of solid/liquid ratio upon uranium leaching efficiency

Acid leaching solution			**Solid/ Liquid ratio**	**Water used**			**Uranium leaching efficiency, based on solution (%)**
				Saturation, (%)	**Used water%**		
Type	Conc., g/l	Used amount based on weight of sample, %		27.7	Based on weight of the ore	Based on the total volume of solution	
H_2SO_4	60	6	1/1		69.0	69.0	39.73
		12	1/2		165.6	83.1	55.63
		18	1/3		262.5	87.5	78.85
		24	1/4		359.2	89.8	83.2
		30	1/5		456.1	91.2	84.6

3.2.4. Effect of excess amount of acid

In order to obtain the maximum uranium leaching efficiency using percolation leaching device, excess amount of the acid used was added to the same amount of water used in the leaching solution. Five different amounts acid were investigated under the following conditions: 0 g/l acid concentration, 1/3 solid/liquid ratio, 22% weight of acid per weight of sample, 262.5% of water used based on sample while 87.5% based on leaching solution, 3 hours contact time, 0.75 mil/min. flow rate of in let solution while 0.2 ml/min. of out let solution and - 0.125 to + 0.09 mm grain size. Data obtained (Table 14) revealed that uranium leaching efficiency reached to 93.5% at 22% wt. of acid / wet. of sample

Table (14): Effect of excess amount of acid upon uranium leaching efficiency through vat percolation device

Acid leaching solution			**Water used**			**Excess amount of acid (%)**	**Uranium leaching efficiency %**
Type	Conc., g/l	% of used amount based on weight of sample	SP%	Percent of used water %		wt. of acid/ wt. of sample %	
			27.7	Based on weight of the ore	Based on the total volume of solution	22	93.5
						20	89.30
						18	78.85
						15	66.90
						10	59.30
						6	52.20
						15	66.90
*H_2SO_4	*60	*18.0					
				262.5	87.5		

From the above study, uranium leaching efficiency by vat percolation leaching device, it is concluded that the obtained optimum conditions are:

- Type of leaching agent: sulfuric acid solution.

-Acid concentration: 60 g/l.
- Grain size: - 0.125 mm to + 0.09 mm.
- Acid solution percent: (22%) weight of acid per weight of sample.
- 1/3 solid/liquid ratio.
- Saturation percent: 27.77%.
- Water amount used: 87.5% based on the solution while 262.32% based on sample.
- Contact time: 3 hours.
- Uranium leaching efficiency: 93.5%.

IV. CONCLUSION

The present study dealt with the suitable leaching device for uranium extraction from Gabal Gattar project, Eastern Desert, Egypt. A technological sample, was subjected to two leaching devices: the agitation and the vat percolation leaching. This aim was achieved by applying some different leaching technological parameters through both devices. The obtained optimum leaching conditions of the agitation device were: sulfuric acid **60 g/l** concentration and **14%** total acid amount used based on weight of the sample as a leaching agent**, - 0.125 to + 0.09** mm. sample grain size, water used based on weight of the sample **161.1%** while it was **80.5%** based on the total volume of solution at **32.5%** saturation and **120** min. contact time, **1/2** solid liquid ratio and **150** rpm at room temperature. According to these conditions, uranium leaching efficiency attained **94.94%.**

On the other hand, in the case of vat percolation leaching under the following optimum condition: sulfuric acid **60 g/l** concentration as a leaching agent, 1/3 solid/liquid ratio, **- 0.125 to + 0.09** mm. grain size, **22%** total acid amount used based on weight of the sample, percent of water used based on weight of the sample **262.32 %** while was **87.5%** based on the total volume of solution at **27.77%** saturation percent and **3 hours** contact time at room temperature. The uranium leaching efficiency obtained was **93.5%.**

Comparing the obtained results directing the decision to the agitation leaching device as the suitable technique for uranium leaching applying the most economic conditions especially to the consumed water amount, acid volume and saturation percent where they are very important to the project established in the middle of desert.

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