International Journal of GEOMATE, Jan., 2017, Vol. 12, Issue 29, pp. 17-24
Geotec., Const. Mat. & Env., ISSN: 2186-2982(P), 2186-2990(O), Japan

RETENTION STUDIES ON ARSENIC FROM AQUEOUS SOLUTIONS BY LIME TREATED SEMI ARID SOILS

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ABSTRACT: In this study, locally available semi-arid soils (Al-Ghat and Al-Qatif) having different chemical and mineralogical characteristics are considered as barrier materials, and their response to arsenic adsorption at varying initial concentrations, pH conditions, temperature and dilution ratios is studied. Empirical models (Langmuir and Freundlich) are applied to ascertain monolayer or heterogeneous adsorption. Lime is added to these soils in order to enhance their geotechnical properties. Kinetic models are employed to validate the type and nature of arsenic sorption onto these soils (whether pseudo first-order or second-order). Also Elovich and intraparticle diffusion models revealed that along with surface adsorption, chemisorptions and diffusion are the other processes occurring concurrently in the system, but it was not able to identify the dominant phenomenon among these models. It was concluded that both Al- Ghat and Al- Qatif soils when amended with lime can attenuate arsenic, and the experimental results correlate well with selected empirical models.

Key Words: Sorption, Arsenic, Langmuir isotherm, Elovich model, Intraparticle diffusion model.

1. INTRODUCTION

Arsenic is one of the most commonly found metal contaminants in wastes and at waste disposal sites. Arsenic can occur naturally in rocks and soil, water, air, plants and animals, or it can be released into the environment through natural activities such as volcanic action, erosion of rocks, and forest fires. Moreover, anthropogenic contributions of arsenic in the environment can occur through its use as wood preservatives in paints, dyes, metals, drugs, soaps and semi-conductors. High arsenic levels can also be released from certain fertilizers and animal feeding operations. Industry practices such as copper smelting, mining and coal burning also contribute arsenic to our environment. Higher levels of arsenic tend to be found in ground water sources than in surface water sources (i.e., lakes and rivers) of drinking water. The demand on ground water from municipal systems and private drinking water wells may cause water levels to drop and release arsenic from rock formations and exacerbate the arsenic levels in ground water [1-3]. Arsenic exposure can result in carcinogenic and non-carcinogenic responses in humans. Inorganic arsenic compounds are known human carcinogens; USEPA ascertained this designation based on increased lung cancer mortality in multiple human populations exposed primarily through inhalation, increased mortality from multiple internal organ cancers (liver, kidney, lung and bladder), and increased incidence of human cancer in populations consuming drinking water high in inorganic arsenic. The critical effects are hyper pigmentation, keratosis, and possible vascular complications based on human chronic exposure The maximum limit for human consumption of arsenic is limited to 10 ppb by USEPA.

Arsenic speciation can be quite complex insoilsAs^{+3} or As(III) is the dominant form under reducing conditions, while As^{+5} or As(V) is generally the stable form in oxidizing environments. Arsenic compounds adsorb to soils, clays, organic matter and metal oxides/ hydroxides [4]. Arsenic is challenging to treat, and some of the common remediation methods may include containment using barriers, soil washing/flushing, electrokinetics, solidification and stabilization [5]. Solidification and stabilization (S/S) is considered to be the cost effective method for treating high levels of arsenic in soils [6]. Additives and binders used in S/S oxidize As^{+3} andAs^{+5} and form insoluble complexes or become immobilized due to formation of strongly adsorbed species or form co-precipitates with calcium or iron [7]. Calcium has been found to retain arsenic by replacing itself for arsenic in a ratio of 1: 1 mole [8]. However, a sufficient amount of $Ca(OH)_2$ is also required for the hydration to proceed in the system [9-11].

Reference [12] studied, lime stabilization of the fly ash and found it was not completely effective. Through three cycles of sequential leaching, lime amendment effectively immobilizes cadmium. Considerable reductions in concentration are also observed for arsenic and selenium, although it is difficult to isolate an optimal value from the data collected. Lime amendment appears to increase chromium leachability. There is a threshold level of lime amendment, beneath which may result in an increase in pH and leachability of oxyanions such as arsenic, chromium, and selenium. Also the high correlations detected between trace element mobility and dissolved organic carbon (DOC) it suggests that As, Cu and Pb formed soluble organic complexes [13]. Sodium carbonate and bicarbonate ions extracted arsenic most efficiently; Na_2CO_3 leached maximum 118.12 µg/l of arsenic, and $NaHCO_3$, 94.56 µg/l of arsenic from the Ganges delta sediments after six days of incubation. The arsenic concentrations extracted in the batch experiments correlated very well with the bicarbonate concentrations [14].Extraction with $CaCl_2$, EDTA and CH_3COOH, revealed that the leaching curves for As differed from those of the other heavy metals because it occurs mainly as arsenate in oxidized samples. The effect of pH on the surface complexation model based on the sorption at surface sites in Fe–Al oxides, clay and organic matter do not fully apply to this trace element. Arsenic showed a high leaching rate at basic pH because the surfaces are negatively charged above the point of zero charge (when the sorption of anions is less favorable). This finding was made for all the samples examined, especially the montmorillonitic soils, which showed As extraction yields near to 100% at pH over 7. The leaching pattern of As at acid pH depended on the samples examined and the soil phases solubilized in each case. Further the geotechnical behavior of selected soil is dependent on the nature of fluid as well [15]. For instance, a distinctive pattern in the mineral samples with soluble contamination was detected with respect to the samples from the accidental spill of pyritic sludge. The increase in the As leaching rate at acid pH in the latter samples is attributed to the solubilization of the sludge itself and related minerals (e.g.,As-jarosite), rather than to the variation of the surface charge as a function of pH[16-23]. The above study suggests lime amendment to be used with caution.

In this study, the retention of As in two semi-arid soils, originating from Al-Ghat and Al-Qatif, is investigated to evaluate their suitability as barrier materials. Further, the effects of amending these soils with lime on enhanced arsenic retention are explored. The amount of lime was standardized at 6% by dry weight of soil. This percentage was determined based on initial lime consumption and optimum lime content requirements [24-25]. Several series of batch equilibrium tests are conducted to systematically investigate the effects of initial concentration, solution pH, temperature, and time dependent kinetics on arsenic retention in the soils with and without lime amendment.

2. MATERIALS AND METHODS

Soil samples collected from Al-Ghat and Al-Qatif are selected for testing in this study. Al-Qatif is a historic coastal oasis region located on the western shore of the Persian Gulf in the Eastern Province of Saudi Arabia (26° 56′ 0″ N, 50° 1′ 0″ E). Al-Ghat is a town located 270 km to the Northwest of Riyadh at latitude 26° 32' 42" N and longitude 43° 45' 42" E. The physicochemical properties of soils were determined and the USCS (Unified Soil Classification System) classified both soils as CH (clay with high plasticity) with specific gravity of 2.84 and 2.71, respectively. It was found that Al-Ghat soil to be a kaolinitic soil and Al-Qatif soil to be a montmorillonitic soil. Analytical grade calcium hydroxide, has been used as lime and its dosage was fixed at 6% based on initial lime consumption and lime leachability criteria [26]. Analytical grade Arsenic Trioxide, was used as source chemical for arsenic.

2.1 Batch Equilibrium Test Procedure

Batch adsorption experiments were conducted by shaking mechanically a series of bottles containing the soil sample and heavy metal ion solutions maintained at different pH values. Soil sample of 5g was mixed with 100 ml of the solution maintained at a particular pH in 500 ml polyethylene bottles to obtain the soil slurry. In all the tests, the liquid to solid dilution ratio was maintained at 20. This slurry was agitated with a mechanical shaker at room temperature (25±2°C) for 2 h until the pH was stabilized. The pH of the slurry was adjusted to the desired value in the range of 2 to 10 with 0.1M HNO_3 and NaOH [27-29].

Predetermined amounts of source chemical were added to the bottles to result in the desired arsenic concentrations (10, 15, 20, 25 and 30 mg/l), and the bottles were further agitated until equilibrium concentrations were attained. The slurry was filtered using Whatman 42 ashless filter paper and the residual concentration of arsenic in the filtered solution was measured using atomic absorption spectrophotometry (AAS). The amount of arsenic adsorbed by the clay fraction was taken as the difference between the initial and residual concentrations of the arsenic in the solution. In order to determine the removal by hydroxide precipitation at various pH values, a set of blank tests were also conducted using solutions maintained at different pH values without soil.

Tests were conducted in triplicates and the average concentrations are calculated. In order to investigate the effect of temperature on adsorption of arsenic, a series of tests was conducted in controlled constant water bath conditions following the same adsorption testing procedure [30].

The effects of solids to liquid ratio on adsorption results was assessed by preparing the samples with solid to liquid (S/L) ratios of 1:5, 1:10, 1:20, 1:50, and 1:100 and shaking them for 24 hours. Then, 100 mgL^{-1} equivalent arsenic mass was added to all samples and again shaken for 24 hours. The samples were then removed, filtered and the filtered liquid was analyzed for arsenic concentration. Another series of experiments was conducted to investigate pH effects by employing the similar procedure except the pH of the contaminant solution was first adjusted to the required level and then the adsorbent was added while maintaining a constant S/L ratio of 1:20 throughout the procedure.

2.2 Adsorption Kinetics Test Procedure

A series of kinetic adsorption tests was conducted to study the time dependent arsenic adsorption and help determine the time necessary to achieve the maximum removal capacity. For this test series, batch experiments were conducted for different predetermined time intervals in different polyethylene bottles prepared and maintained under similar conditions. At the end of each fixed time interval, the soil slurry was filtered and the concentration of arsenic was determined as above. This procedure was repeated till the concentration of arsenic in the filtrate remained unchanged with time. The experimental results were validated by applying them to isotherm and kinetic models.

3 RESULTS AND DISCUSIONS

The results obtained from batch equilibrium and kinetic adsorption experiments are assessed to study the adsorption behavior of arsenic in the tested two soils.

3.1 Effects of Initial Concentration, Dilution Ratio and pH on Arsenic Adsorption

Results in Fig.1 show that sorption coefficient increases with initial concentration, with the increase in concentration the competition from other ions reduced resulting in an efficient sorption of arsenic. A decrease in soil amount decreases the number of active sites that are available for sorption and the amount of sorption decreases.

This is evident from Fig.2 in that as dilution ratio increases, more amount of metal ion become available for sorption. Only stable metal ions get sorbed leading to an effective and permanent sorption. Heike [31] also reported similar results in that an increase in the solids results in an increase in retention. This is attributed to an increase in the available surface area and thus an increase in active sites. However, the increase in contaminant retention is not proportional to the amount of the solids; which is attributed to the decrease in the mass transfer gradient as the remaining concentration of contaminant diminishes [32].

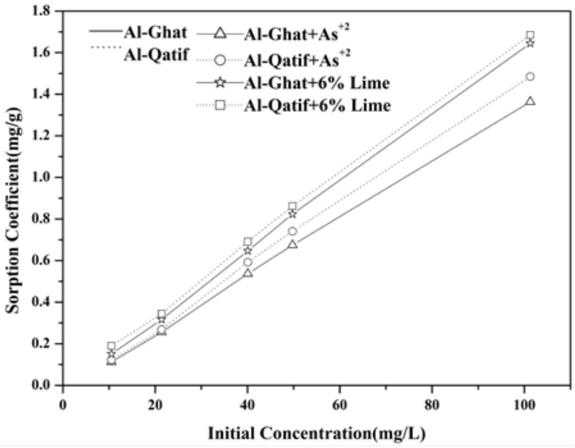

Fig. 1 Sorption of As in soils with and without lime amendment under different initial concentration

The pH plays a major role in sorption of arsenic, and the results showed a gradual increase in the amount of sorbed arsenic with the increase in pH. This increase in sorption can be explained by the changes that occur to the sorbate (soils and their oxide surfaces) and the formation of hydroxides of metal complexes under different pH conditions. The pH of the solution affects the protonation of the functional group on the adsorbent surface of soils as well as metal complexation. At low pH values, adsorbed protons that are exchanged on surface can form proton bonds between surface and metal complexes.

The sorbed protons, also, generate positive charges at the surface repelling or attracting positively or negatively charged metal complexes respectively. It was found that the presence of calcium in lime played a major role in sorption as it increased the pH of the solution there by precipitating the As ions.

It was found that the effective sorption took place at a pH range of 4.5 to 8.5 which can be attributed to the formation of As-Ca complexes at this pH range [33, 34].

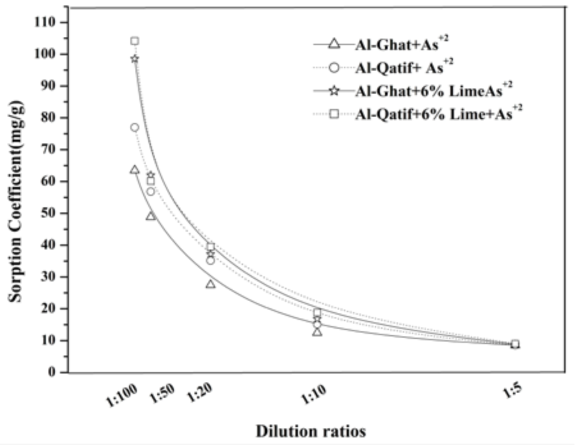

Fig. 2 Sorption of As in soils with and without lime amendment under different solids to liquid ratios

3.2 Effect of Temperature on Arsenic Adsorption

Temperature is known to affect the complexation of metal ions. The influence of the temperature on retention of arsenic was investigated in the range from 25° to 55°C. The selected temperature range encompasses the typical temperature expected during the course of the year in Saudi Arabia. The average temperature of 25°C corresponds to winter, while 55°C is the maximum possible temperature during the summer. All the tests were conducted at neutral pH value (6.8 to 7.2). The effect of temperature under different pH conditions was beyond the scope of this study. For a given initial concentration, the amount of arsenic retained increased considerably with increase in temperature. This effect was more pronounced at higher initial concentration values. The increase in retention levels for arsenic could be attributed to conditions favoring enthalpy and entropy changes in both Al-Ghat and Al-Qatif soils. The increased retention capacity at elevated temperatures is primarily due to entropy increase arising due to increased disorder of water molecules and respective cations in the soil slurry. The results obtained indicate that for both Al-Ghat and Al-Qatif, the increase in temperature aids in higher arsenic removal capacity.

3.3 Sorption Isotherms

Batch equilibrium sorption data was modeled using two common sorption isotherm models, namely Langmuir model and Freundlich model. The isotherm parameters were calculated and are summarized in Table 1. The values of monolayer sorption capacity calculated using Langmuir isotherm are in the similar range for all the soil lime mixtures. For Freundlich isotherm, the R^2 is close to one, but its adsorption values are unrealistic. Hence, Freundlich isotherm does not hold good for these samples. The Langmuir isotherm may be expressed in terms of equilibrium parameter R_L which is a dimensionless constant referred as separation factor. R_L value indicates the sorption nature to be unfavorable if $R_L > 1$; linear if $R_L = 1$; favorable if $0 < R_L < 1$; and irreversible if $R_L = 0$. A separation factor of less than one is obtained for all the samples, which indicates that Langmuir isotherm favors this sorption.

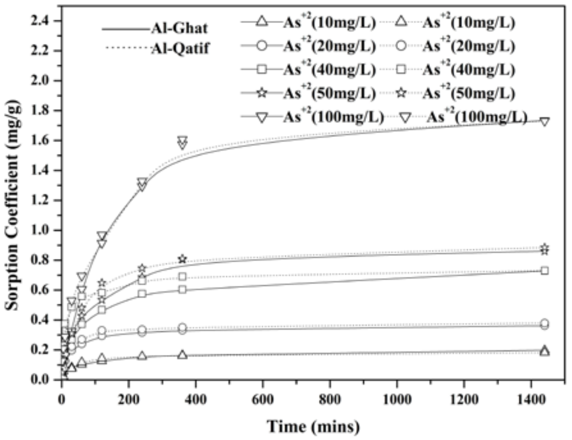

Fig. 3 Time dependent adsorption of As on soils without lime amendment

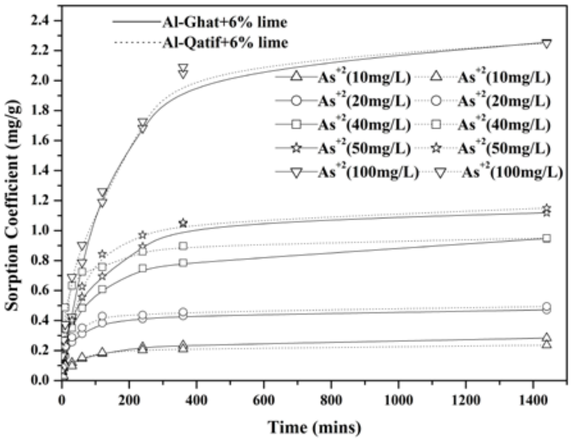

Fig.4 Time dependent adsorption of As soils with lime amendment

3.4 Adsorption Kinetics of Arsenic in Soils

It can be observed from Fig. 3 and Fig.4 that significant sorption occurs immediately and it increases with time with maximum sorption levels in 30 minutes, and thereafter it remains constant till 1440 minutes (24hrs). Sorption increased with time as well as with initial arsenic concentration. It can be clearly seen that with the addition of lime the amount of As adsorbed is higher than without lime addition and remains same for over 24 hours period.

The sorption versus time data was used to test different kinetic models such as pseudo first order, pseudo second order, Elovich, and intraparticle diffusion. The results obtained are summarized in Table 2 and 3. It was observed from regression analysis that pseudo second order kinetic model was better than the first order kinetic model as its regression coefficient was closer to one. Also for a heterogeneous material like soil, monolayer sorption alone does not occur. Instead, a number of processes such as chemi-sorption, ion exchange and precipitation occur simultaneously.

Pseudo second order being a better model reinforces this assertion and this conclusion is also in general agreement with previous studies. Elovich kinetic model tries to model the processes of desorption and chemisorption. It can be seen from the regression analysis, as given in Table 3, that the regression coefficient is closer to 1 which indicates that chemisorption is also possibly occurring, where the ά and β values represent sorption and desorption. Further, sorption increases with concentration, while desorption decreases with concentration [35-36].

At a higher concentration, sorption takes place predominantly on sorption sites whereas at lower concentration desorption is dominant as competing ions predominate the sorption sites. Similarly, intraparticle diffusion model tries to predict the phenomenon of diffusion in the sorption process. If the results follow a linear fit passing through the origin, then it implies that only diffusion acts as the predominant phenomenon. It can be observed from Table 3 that, the regression value is closer to 1, but the regression line does not coincide with the origin which shows that along with sorption, diffusion is also active, but it is not the dominant process in sorption.

4. CONCLUSIONS

The retention of metal ions by soils is a complex process involving complex mechanisms, and is controlled by different variables that can interact. In this study, two field soils (Al-Ghat and Al-Qatif soils) having variable chemical composition were selected and amended with 6% lime to enhance their retention capacity of arsenic. The results obtained indicate that both initial concentration and rise in temperature favor the retention characteristics for both soils with lime quite significantly. Surface adsorption is known to be dominant at lower pH levels, while precipitation favored at higher pH levels.

The presence of calcium in lime played a major role and formed stable precipitates of calcium arsenate at highly oxidizing conditions and moderate pH conditions of 4.5 to 8.5. At elevated temperatures the hydration of calcium occurs efficiently which contributed to efficient sorption of arsenic.

The addition of lime in soil to retain arsenic creates a solidified monolith where the mobility is diffusion controlled. This reduces the leaching of arsenic significantly and this soil lime mixture can be considered as a suitable barrier material (e.g. soil liner for a landfill).

Relatively higher retention of arsenic was recorded, in case of Al-Qatif (montmorillonitic soil) with lime than Al-Ghat (kaolinitic soil) with lime due to difference in chemical composition, entropy and enthalpy changes. This is also consistent with the published studies that montmorillonitic soils retain arsenic better than kaolinitic soil.

The following specific conclusions can be drawn from this study:

1. Al- Ghat soil and Al- Qatif soil amended with 6% lime were found to be good sorbents for arsenic. Al-Qatif is a better adsorbent than Al-Ghat soil.
2. Sorption is pH dependent and the maximum sorption occurs over a pH range of 4.5-8.5 for arsenic in both the soils. Retention of arsenic increased with increase in initial concentration, dilution ratio, and pH.
3. Langmuir monolayer sorption capacity is shown to decrease with concentration, whereas Elovich’s model sorption is found to increase with concentration and desorption to decrease with concentration.
4. The adsorption kinetic experiments show for both soils that the maximum sorption occurs within an hour and thereafter reaches equilibrium.

Arsenic retention in soils us attributed to several processes such as sorption, chemisorptions, ion exchange, precipitation and diffusion occurring simultaneously, with no one process is being dominant.

5. ACKNOWLEDGEMENTS

This project was funded by the National Plan for Science, Technology and Innovation (MAARIFAH), King Abdul Aziz City for Science and Technology, Kingdom of Saudi Arabia, Award Number (12ENV2583-02).

Table 1 Langmuir and Freundlich Isotherm Parameters for Arsenic Sorption in Soils with and Without Lime Amendment

Parameters		Langmuir isotherm				Freundlich Isotherm		
Sorbent	Initial Conc (mg/L)	Monolayer Adsorption Capacity (mg/g)	K	R^2	Separation factor R_L	K_f	n adsorption intensity	R^2
Al- Ghat Soil + As^{+3}	10	1.483	4.50 X10^{-2}	0.95	0.6897	0.1852	0.0053	0.98
	20	1.462			0.5269		0.0018	
	30	1.472			0.4251		0.0009	
	40	1.586			0.3576		0.0006	
Al- Ghat Soil + 6% Lime + As^{+3}	10	2.011	1.89 X10^{-1}	0.9512	0.3454	0.9160	0.3251	1
	20	1.874			0.2089		0.2399	
	30	1.452			0.1492		0.1980	
	40	1.758			0.1164		0.1742	
Al- Qatif + As^{+3}	10	3.785	0.126	0.904	0.4429	0.8400	0.2699	0.588
	20	3.610			0.2849		0.1897	
	30	1.571			0.2095		0.1548	
	40	1.633			0.1629		0.1375	
Al- Qatif + 6% Lime + As^{+3}	10	3.060	1.157	0.96	0.0795	0.5120	0.5287	0.95
	20	1.510			0.0425		0.5687	
	30	1.812			0.0234		0.5297	
	40	2.310			0.0216		0.5346	

Table 2 Pseudo first order and second order model parameters

Parameters		Pseudo first order		Pseudo second order	
Sorbent	Initial Concentration (mg/L)	K_1 per min	R^2	K_2(g/mg/min)	R^2
Al- Ghat Soil + As^{+3}	10	0.0062	0.9800	4.26 x10^{-8}	1.02529
	20	0.0058	0.9600	9.056 x10^{-2}	1.00
	30	0.0185	0.9885	4.82 x10^{-8}	1
	40	0.0024	0.9871	1.839295	0.9999
Al- Ghat Soil + 6% Lime + As^{+3}	10	0.0119	0.97276	2.91 x10^{-8}	1
	20	0.00374	0.99063	1.88 x10^{-8}	1
	30	0.00407	0.9916	1.92 x10^{-9}	1
	40	0.00359	0.99532	5.76 x10^{-7}	1
Al- Qatif + As^{+3}	10	0.00221	0.9416	5.43 x10^{-10}	1
	20	0.00126	0.99644	1.33972	1
	30	0.00051	0.78949	3.05 x10^{-9}	1
	40	0.000955	0.98272	1.91682	0.99999
Al- Qatif+ 6% Lime + As^{+3}	10	0.00304	0.96947	5.76 x10^{-9}	1
	20	0.00499	0.99175	0	1
	30	0.00363	0.99085	6.16 x10^{-9}	1
	40	0.00316	0.98837	4.77073	1

Table 3 Kinetic model parameters for Arsenic Sorption in Soils with and without lime amendment

Parameters		Elovich			Intra Particle Diffusion		
Sorbent	Initial Concentration(mg/L)	β (g/mg)	α (mg/g/min)	R^2	K_{dif} (mg/g/min$^{-0.5}$)	C (mg/g)	R^2
Al- Ghat Soil + As^{+3}	10	49.4559	0.0928	0.9912	0.0032	0.07337	0.8330
	20	25.6410	0.4626	0.9928	0.0050	0.2683	0.8049
	30	22.7842	3.2682	0.9762	0.0044	0.2621	0.9613
	40	19.688	4.4788	0.9538	0.0148	0.2316	0.9833
Al- Ghat Soil + 6%Lime + As^{+3}	10	48.0769	0.0627	0.9653	0.0035	0.0755	0.9768
	20	35.8551	1.56204	0.8737	0.0053	0.1627	0.9859
	30	26.7952	37.9274	0.9400	0.0061	0.2709	0.9363
	40	24.9812	638.9750	0.8603	0.0075	0.4206	0.9129
Al- Qatif + As^{+3}	10	74.6825	34.1722	0.9805	0.0036	0.1240	0.8727
	20	59.2417	882.4575	0.9297	0.0031	0.2137	0.9391
	30	36.8052	1337.9876	0.9725	0.0047	0.3468	0.9084
	40	28.2167	920.4580	0.9163	0.0069	0.4202	0.9714
Al- Qatif + 6% Lime + As^{+3}	10	236.9668	12.8765	0.9206	0.0041	0.1711	0.8854
	20	65.4450	166.3907	0.9714	0.0158	0.2827	0.9714
	30	53.3617	369.5565	0.9368	0.0199	0.4453	0.9678
	40	42.4448	1925.9	0.9218	0.0054	0.6091	0.9602

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International Journal of GEOMATE, Jan., 2017, Vol. 12, Issue 29, pp. 17-24.

MS No. 5155j received on June 03, 2015 and reviewed under GEOMATE publication policies.

Pertinent discussion including authors’ closure, if any, will be published in Dec. 2017 if the discussion is received by June 2017.

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