

## **The Use of Bitumen in the Stabilization of Lead Contaminated Iraqi Soil**

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### **Abstract**

Iraq is one of the biggest countries in oil production and its processes. Bitumen is a by-product material that remains after the process of crude oil. This study is designed to investigate the role of bitumen in the stabilization of lead contaminated Iraqi soil; as measured by a newly developed “mini” Jet Device and the dispersion ratio method (DR,%).

The results revealed that both bitumen amount and its method of mixing with soil, as a direct or as emulsion mixing, can significantly improve the stability of lead contaminated soil. Hence, only 3% bitumen as emulsion mixing was required to get the optimum soil stabilization, compared with 9% in direct mixing. A 3% bitumen emulsion has reduced both the scouring depth (SD, from 24.5 to 1.6 mm;  $R = -0.90$ ) and the erodibility coefficient ( $K_d$ , from 1.09 to 0.14;  $R = -0.86$ ) as a result of improving soil engineering properties related to soil stabilization. However, the 3% of bitumen emulsion has also improved the soil chemical properties that have an important role in soil aggregation and lead mobility; such as pH, EC and SAR. Therefore, 3% of bitumen emulsion has successfully decreased both the dispersion ratio (DR, % from 7.03 to 4.48;  $R = -0.90$ ) and the lead solubility (Pb from 48.8 to 4.9ppm;  $R = -0.96$ ) in the solutions of lead contaminated soil.

**Keywords:** Bitumen Emulsion, Soil, Stabilization, lead, contamination, Iraq.

### **1.Introduction:**

Baghdad can be considered as one of the most important cities in Iraq. Lead soil contamination becomes as a serious problem in Iraq; due to increasing in the population, wars, human activity and as a result of increasing of car usage; especially after 2003 when about one million used cars entered Iraq from the neighboring countries. The increasing of used cars led to the increase of lead concentration; because the oil ministry in Iraq is still adding the tetra alkyl lead additive as a chemical agent in the benzene production. For

example, lead concentration in Al-Dora region south of Baghdad was very high (more than the minimum range, 2ppm, but less than the maximum, 300ppm, [1]. due to the lead additives in Al-Dora Refinery. Additionally, the presence of Babil car batteries factory in Al-Waziryah (north east of Baghdad) and Al-Noor Batteries factory in Abu-Ghraib (west of Baghdad), which are still using old techniques, have no emission control devices and no waste water treatment plants. All these lead emissions can be deposited and adsorbed by soil. Iraq is one of the biggest countries in both oil production and manufacturing and bitumen is a very available as a by-product of oil industry. Bitumen hence is available and supposed to be of a low cost and can be utilized in the stabilization of soils against erosion. Bitumen is a building material usually used in Iraq under the name “Flank Coat” useful in coating the concrete structures against ground water effect.

The main objective of this study is to evaluate the effect of bitumen, as a stabilizer, on soil engineering and chemical properties that have an impact on the aggregate stability of an artificially lead contaminated soil. The procedure also includes the shaking of 1:2 soil-water mixture to determine both the dispersion ratio (DR, %) and the soluble (i.e unstable or mobile) Pb in soil solution. The DR was measured by the gravimetric method to be related later to the chemistry properties of soil solution (EC, PH, and SAR).

### **1.1. Treatment of Contaminated Soil**

Immobilization technologies were designed to reduce the mobility of contaminants by changing the physical or leaching characteristics of the contaminated matrix. Contaminant mobility is usually decreased by physically restricting its contact with the surrounding waters, or by chemically fixing to make it more stable with respect to dissolution in water. Several methods are available for immobilization of metal contaminants, including those that used chemical materials and/or thermal treatment to physically bind the contaminated soil or sludge. Solidification and stabilization (S/S) treatments are the most commonly used in heavy metals-contaminated sites. [2]

The general approach for S/S treatment processes involves mixing or injecting treatment agents to the contaminated soils. Inorganic binders, such as cement, lime, fly ash, or blast furnace slag, and organic binders such as bitumen are used to form a crystalline, glassy or

polymeric framework around the waste. [3] The S/S is achieved by mixing the contaminated material with appropriate amounts of binder/ stabilizer and water. The mixture sets and cures to form a solidified matrix and contain the waste.

### **1.2. Bitumen Properties**

Bitumen is a by-product that remains after the processing of crude petroleum. Bitumen emulsions are complex fluids, their stability depend on the intermolecular forces, a result of a balance of repulsive and attractive forces. An emulsion is a dispersion of small droplets of one liquid in another one. Emulsions can be formed by any two immiscible liquids, but in mostly one of the phases is water. In multiple emulsions, the disperse phase contains another phase which may not have the same composition as the continuous phase [4]. Bitumen emulsion can be divided into anionic, cationic and nonionic types depending on the charge of their head groups in water; although this may be a pH-dependent. However, the charge on the emulsion head group mostly determines the net charge on the bitumen surfaces [5].

The basic process involved in bitumen stabilization of fine-grained soils is a waterproofing phenomenon. Soil particles or aggregates are coated with bitumen that can stop or minimize the penetration of water which could normally result in a decrease in soil strength. In addition, it can improve soil stability by making the soil resistant to the detrimental effects of water. In soil stabilization with bitumen, two basic mechanisms are active: Waterproofing and adhesion. The aggregate particles adhere to the bitumen and the bitumen becomes a as binder and that increases both the cohesion and the shear strength [6].

## **2. Materials and Methods**

### **2.1. Soil Samples**

In this study, a Silt Clay Loam soil (Lean Clay, Table (1)) were used as a sample to carry out the experiments, acquired from Al-Taji region, North of Baghdad, from depth (0 to 90) cm. The soil samples were tested and analyzed according to the ASTM standards

(ASTM, 2006). Other soil properties, of the concern in this study, are listed in Tables (3) and (4).

**Table (1) The physical and chemical properties of studied soil**

Soil Properties	Value
Sand (%)	13
Silt (%)	57
Clay (%)	30
Specific gravity	2.48
Organic Matter(%)	1.09
CaCO <sub>3</sub> (%)	0.3
CaSO <sub>4</sub> (%)	0.6
PH , (1:2 soil water)	7.4
EC (mS/cm) , (1:2 soil water)	1.03
SAR (1:2 soil water)	7.11
Pb ppm (1:2 soil water)	19.5

## **2.2. Bitumen Material**

The bitumen used in this study is a product under the name “ Prakcoat” made by the Oasis Grease & Lubricants Company (U.A.E) and bought from the local market. Bitumen properties are shown in Table (2). Bitumen features are: cold applied, easy to apply, adhere to (concrete, metal, wood), nonflammable, resist the chloride and sulphate salts attack in soil, and economical. The bitumen addition to soil was carried out in two methods. The first method (named as a Direct Method) was by adding the wanted amount of bitumen directly to the soil and then mixing both materials properly. The second method (named as Emulsion) was by adding the wanted amount of bitumen to the amount of water required to reach the Optimum Moisture Content (O.M.C.) to get a homogeneous emulsion and then mixing it with soil properly.

**Table (2) The properties of bitumen used in this study**

Properties	Values	Test standard
Form	Thick viscose liquid	Visible
Color	Dark brown	
Density (g/cm <sup>3</sup> )	1.03 ±0.02	ASTM D2939
Penetration at 25 C	Null	ASTM D5
Soild Content%	40 ±5	ASTM D2939
Service Temp ©	6 to 47	
Setting time	8 hrs. touch dry 24 hrs. firm set	ASTM D2939

### **3. Experimental Work:**

Most physical and engineering properties were determined at the Hydraulic laboratory of the Environmental Engineering Department, Engineering College, Al-Mustansiriya University according to the ASTM standards, as described by the same author. [7] However, scouring depth (SD), erodibility coefficient (Kd) and critical shear stress ( $\tau_c$ ) of soils were determined by a “mini” Jet Erosion device developed by Al-Madhhachi et al., [8]. Digital Shore-D Durometer to measure the degree of hardness was also used at the Materials Department laboratory, Engineering College, Al-Mustansiriya University.

#### **3.1. Preparing Soil Samples for Chemical Tests:**

- All the soil samples were air-dried, broken into small sizes and sieved through a 4.75 mm sieve according to ASTM standard. The sieving was performed to ensure that the soil was of uniform grade.
- The artificial Pb-contaminated soil samples were prepared by mixing lead nitrate, as the source of lead (Pb), to produce a soil of 4000 mg/kg lead concentration from the natural soil. As a reminder, 300ppm in soil is the maximum acceptable Pb concentration in EU countries [9].

- Different percentages of bitumen (0%, 1%, 2%, 3%, 6% and 9%) by soil weight were added to the soil and mix by hand until the mixture seems to be homogeneous and then left to air dry. As explained in Section (2.2), two mixing methods were used to prepare soil samples; as Direct Mixing and as Emulsion.
- 25 g of soil for each bitumen percent placed in a conical flask and 50 ml of distilled water was added (1:2 soil water ratio) to the soil sample and shaken by hand for 1 minute then left for 2 hours to settle down. The soil solution remaining after sedimentation was subjected to the tests of the dispersed solids (DR %) and then filtered to measure Pb, PH, EC, and SAR of soil solutions.

### **3.2. Soil Solution Testing Methods**

All tests on soil sample solutions were carried out at the sanitary laboratory of the Environment Department, Engineering College, Al-Mustansiriya University.

#### **3.2.1. Dispersion Ratio**

The term "dispersed ratio" refers to matter suspended in water or in soil solution, and is related to the percentage of clay (<0.002mm) in soil sample. Dispersed solids includes both total suspended solids; the portion of total solids retained by a filter, and total dissolved solids; the portion that passes through a filter [10].

Dispersed solids can be measured by evaporating a soil-water mixture in a weighed dish, and then drying the residue in an oven at 103 to 105° C. The increase in weight of the dish represents the dispersed solids and hence soil erodibility. The dispersion ratio in this study was measured according to Middleton principles and by a method modified by Mutter [7, 11] In this study, 25g of lead contaminated soil were mixed in 250ml conical flask with 50ml water to get a 1:2 soil water mixture. The mixture was shaken by hand for 1min. and then left for 2 hours [12]. After 2hours the soil particles greater than the clay (>0.002mm) may settle down. A 10ml of the suspension was transferred to a weighed beaker and put in the oven to determine the dispersed soil materials in suspension. DR (%) was calculated by the following equation:

$$\text{DR, (\%)} = (\text{Clay dispersed in Soil solution} / \text{Clay content in soil}) * 100$$

### 3.2.2 Chemistry of Soil Solution

The soil solution chemical properties, namely pH, EC and SAR, were measured in the filtered 1:2 soil solution prepared in the previous suction. pH was measured by a pH-meter, EC by an Electrical Bridge, soluble Na, Ca and Mg (in meq/l) and Pb (as ppm) were measured at Ib Al-Haitam Education College, University of Baghdad. The sodium adsorption ratio was calculated by the following formula [13]:

$$\text{SAR} = \frac{\text{Na}^{+1}}{\frac{\sqrt{\text{Ca}^{+2} + \text{Mg}^{+2}}}{2}}$$

; where sodium, calcium, and magnesium are in meq/liter.

Sodium adsorption ratio (SAR) is a measure of the suitability of water for use in irrigation to prevent soils from the sodium hazard. In general, the higher the sodium adsorption ratio, the less suitable the water is for irrigation. It is also an index of the soil sodicity and can be determined from the chemical analysis of water extracted from the soil.

## 4. Results And Discussion

Due to results obtained in this work and the findings of another related detailed study published by the same author [14], Bitumen has markedly improved soil both engineering and chemical properties related to soil stabilization. It should be noted, however, that the 9% direct mixing of bitumen has given better results than the 3% as emulsion. But in terms of feasibility and costs analysis, the 3% emulsion has achieved the optimum results.

### 4.1 Bitumen Direct Mixing

Table (3) shows the bitumen direct mixing impact on both engineering and chemical properties of lead contaminated soil. A 9% of bitumen was required to get the best soil properties against erosion and lead movement. According to the correlation coefficient (R), bitumen has a significant impact on shear stress ( $\tau_c$ , pa), Atterberg limits and the degree of hardness (DH); followed in a lesser extent by the optimum moisture content. Hence the Scouring Depth (SD) was reduced from 24.5 to 0.31mm and the erodibility coefficient (Kd) from 1.09 to 0.0. On chemical properties, bitumen seems to have a positive impact on soil properties related to soil stability; by reducing the dispersion ratio (from 7.03 to 2.9%)

and soluble Pb in soil solution (from 48.8 to 9.8ppm) due to the reduction in both SAR (from 6.3 to 4.2) and EC (from 3.5 to 1.7) which is accompanied with greater pH values (7.18 to 7.45) in soil solution.

**Table (3) Bitumen percentages in relation to engineering and chemical soil properties  
(As Direct Mixing)**

Engineering Property	Bitumen,% (Direct Mixing)				Corr.,R*
	0	3	6	9	
SD, mm	24.5	3.35	2.25	.38	-0.84
Kd, cm <sup>3</sup> /N.s	1.09	0.17	0.17	0.0	-0.85
$\tau_c$ , pa	0.05	4.13	5.09	7.74	0.97
DH	76.5	92.3	93.35	97.75	0.90
M.D.D, g/cm <sup>3</sup>	1.77	1.80	1.80	1.75	-0.29
O.M.C, %	18.0	18.3	18.6	18.5	0.88
L.L,%	39	39	38	38	-0.89
P.L, %	25	25.7	26	27	0.98
P.I, %	14	13.3	12	11	-0.99
<b>Chemical property (1:2 soil:water)</b>					
DR,%	7.03	4.07	3.83	2.93	-0.91
Pb,ppm	48.8	12.2	9.8	9.8	-0.81
pH	7.18	7.4	7.43	7.48	0.91
EC,mS/cm	3.50	2.9	2.6	1.7	-0.91
SAR	6.3	5.2	4.9	4.2	-0.98

\*R must equal 0.90 or 0.98 to be significant at 0.05 or 0.01 level, respectively.

#### **4.2 Bitumen Emulsion**

Table (4) reveals that the addition of only 3% bitumen as an emulsion has improved the engineering properties related to soil stabilization and the Pb solubility and movement reduction. However, shear stress ( $\tau_c$ , pa), maximum dry density (M.D.D), degree of hardness (DH), the Atterberge limits and optimum moisture content have markedly affected by the bitumen. Hence, both the scouring depth (SD) and the erodibility coefficient (Kd) were reduced (from 24.5 to 1.6 and 1.09 to 0.14, respectively).



**Table (4) Bitumen percentage in relation to engineering and chemical soil properties  
(As Emulsion)**

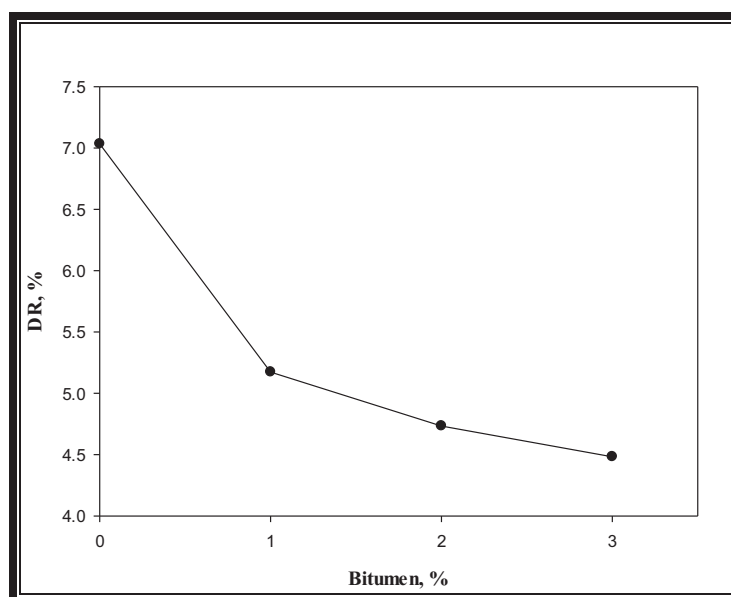
Engineering Property	Bitumen,% (as emulsion)				Corr.,R*
	0	1	2	3	
SD, mm	24.5	14	2	1.6	-0.90
Kd, cm <sup>3</sup> /N.s	1.09	0.31	0.25	0.14	-0.86
$\tau_c$ , pa	0.045	2.6	4.5	6.1	0.99
DH	76.5	82.5	85	92	0.98
M.D.D, g/cm <sup>3</sup>	1.77	1.78	1.79	1.80	0.99
O.M.C, %	18	18.1	18.15	18.3	0.98
L.L,%	39	38.8	38.8	38.5	-0.93
P.L, %	25	25.25	25.5	25.7	0.99
P.I, %	14	13.4	12.6	12	-0.99
<b>Chemical property (1:2 soil :water)</b>					
DR, %	7.03	5.17	4.73	4.48	-0.90
Pb,ppm	48.8	40.8	24.4	4.9	-0.96
pH	7.18	7.19	7.20	7.21	0.85
EC, mS/cm	3.50	2.55	2.37	2.34	-0.86
SAR	6.3	6.2	5.4	5.01	-0.96

**\*R must equal 0.90 or 0.98 to be significant at 0.05 or 0.01 level, respectively.**

Chemically and because the use of 3% bitumen emulsion has yielded the optimum soil stabilization, a detailed coverage focusing on the impact of the 3% bitumen emulsion on the soil chemical properties are discussed in Figures (1 to 5).

Figure (1) shows the relationship between the dispersion ratio of the lead contaminated soil and bitumen percentage (0%, 3%, 6%, and 9%). The figure indicates that whenever the bitumen percentage increased in soil, the dispersion ratio (DR, %) in soil solution decreased (from 7.03 to 4.48). In soil stabilizing with bitumen, two basic

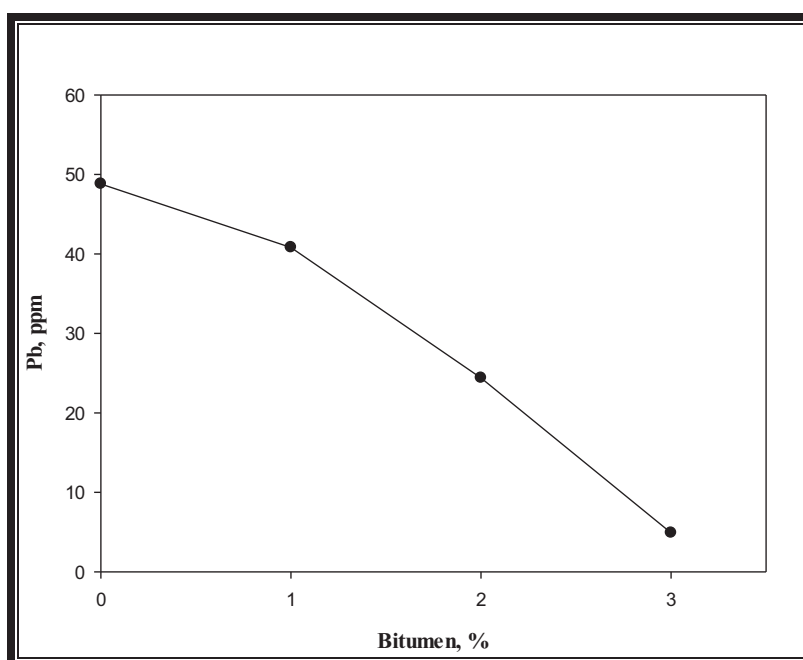
processes are active: water proofing and adhesion. Soil particles are coated with bitumen that stops or reduces the penetration of water, which could result in decreasing soil strength. The soil aggregates adhere to the bitumen and it behaves as a binder and thus the cohesion forces and shear strength increase [6]. When a contact happened between the bitumen emulsion and soil aggregates, adsorption of free emulsifier and soil particles may occur and the pH rises. This rise in pH leads to the flocculation of soil particles. Coalescence of bitumen droplets also takes place and the trapped water diffuses out. Droplets in contact with clay minerals spread on the surface eventually displace the water film on the aggregate surface [15, 16].



**Fig. (1) Relationship between dispersion ratio of soil solution and bitumen percentage, as emulsion (R= -0 .90).**

Figure (2) focuses on the reduction of soluble Pb in artificially contaminated soil solution (from 48.8 to 4.9ppm), due to the impact of soil properties which have clearly improved by the addition of bitumen. As a reminder and according to Table (1), the Pb concentration in the original natural soil sample is 19.52ppm which means that bitumen has even stabilized part of lead existing in original sample. Depending on the Pb cationic strength, soil type and bitumen characteristics, Pb may be strongly adsorbed on the colloidal surfaces of soil leading to the reduction of Pb concentration in soil solution. Pb

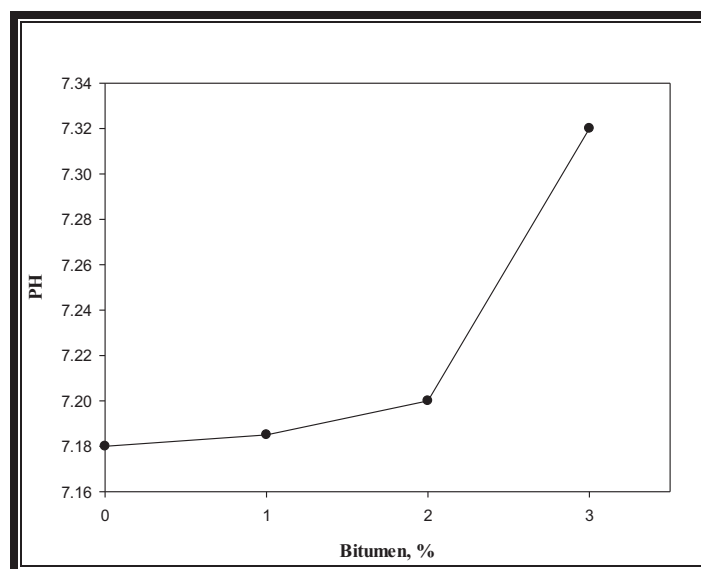
precipitation may also occur in the soil and water systems as Pb carbonates [17]. As reviewed by [18], lead in sulphide solutions may be reduced to precipitate as Pb sulphides. This occurs usually by the action of sulphate-reducing bacteria or through an encounter with organic matter, which may be bitumen in this case. In alkaline conditions and in the presence of carbonate, sulphides and silicates; as in the case of the current studied soil Table (1), many metals including Pb may precipitate and be less in soil solution.



**Fig. (2) Relation between Pb conc. In soil solution and the bitumen percentage (R= - 0.96).**

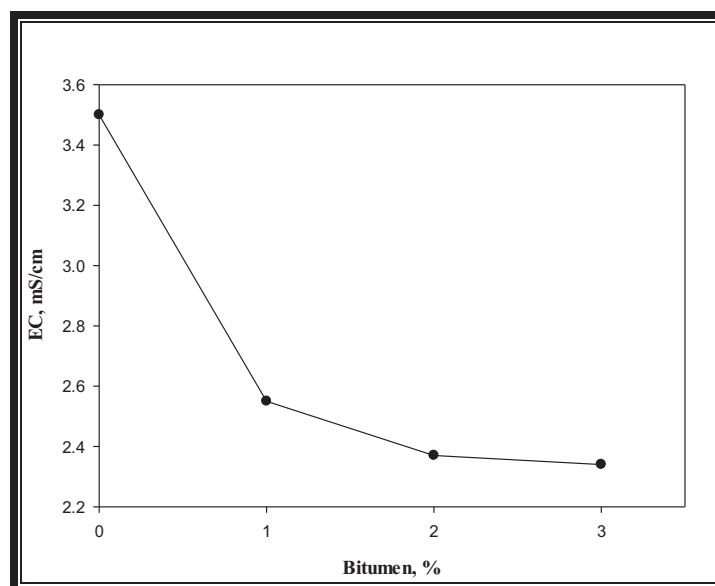
Figure (3) shows the relationship between the pH of soil solution, which is contaminated with lead, and bitumen percentages (0%, 3%, 6%, and 9%). Bitumen has increased pH from 7.18 to 7.32. Depending on the type of emulsifier used, the bitumen particles can either get positive or negative charge. Bitumen particles getting negative charge are classified as anionic bitumen emulsion and the ones having positive charge are known as cationic bitumen emulsion [19]. From the figure it could be noticed that the pH increase with increasing bitumen percentage and that may be because bitumen has acted as

positively charged cation and may actually neutralize the acid in the cationic emulsion causing the pH to rise [20].



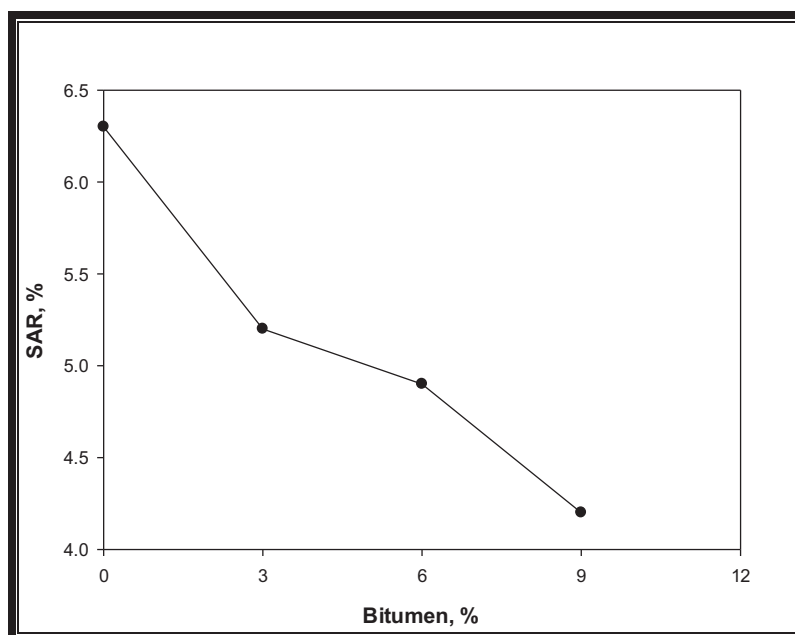
**Fig. (3) Relationship between soil solution PH and bitumen percentage (R= .99).**

Figure (4) shows the relationship between electrical conductivity of soil solution, which is contaminated with lead ions and bitumen percentages (0%, 3%, 6%, and 9%). From the figure it could be noticed that electrical conductivity decreased (from 3.5 to 2.34) with increasing bitumen percentage. According to [21], Soil pH is negatively related with soil electrical conductivity. Low soil pH value should have high soluble salt content and therefore high electrical conductivity. In high pH values, the cation exchange capacity of soil increased and hence more cations are adsorbed on soil particles and this may reduce the soluble cations in soil solution and hence EC [18].



**Fig. (4) Relationship between electrical conductivity and bitumen percentage (R= - 0.89).**

Figure (5) shows the relationship between sodium adsorption ratio of soil solution that is contaminated with lead ions and bitumen percentages (0%, 3%, 6%, and 9%). From the figure it could be noticed that the SAR decreased (from 6.3 to 5.0) with increasing bitumen percentage. The SAR depends largely on the  $\text{Na}^+$  concentration in soil solution. These results are in agreement with [22] who studied the role of  $\text{Na}^+$  in enhancing the clay dispersion in Sodic soils. When sodium ions are adsorbed by soil particles as exchangeable cations, soil becomes sodic and the soil structure is degraded by means of clay swelling and dispersion. In the current study, the lead that existing in the contaminated soil (4000 ppm) may act like Ca and Mg as a divalent cation and can reduce the dispersive action of Na in soil solution [13]. In addition, bitumen may also behave as a positively charged cation [20] and this may enhance the flocculation and aggregation of soil particles.



**Fig. (5) Relationship between Sodium adsorption ratio and bitumen percentage (R= - 0.96).**

### **5. Conclusion:**

Bitumen appears to be as a good Soil stabilizer. Bitumen can significantly improve both the engineering and the chemical properties of soils and that make it more stable and less erodible. In this study, the method of bitumen mixing with soil is also appeared to be of a prime importance. In the direct mixing method, 9% of bitumen was required to completely improve soil properties against erosion. While in emulsion mixing method, only 3% bitumen was successful in minimizing the soil erodibility; by improving shear stress, degree of hardness, maximum dry density, optimum moisture content, Atterberg limits and hence reducing both the scouring depth and erodibility coefficient of lead contaminated clayey soil. In addition, the chemical impact of bitumen has also played a major part in improving soil stability. Therefore, the dispersion ratio, which is considered as a simple soil aggregate stability test, decreased with higher additions of bitumen emulsion. The bitumen has increased pH, but reduced both EC and SAR, in soil solution and this has markedly improved the soil stability and reduced the mobility of Pb in soil solution.

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