

Rudiments of Electrokinetic Method of Soil Remediation in Civil Engineering Perspective



Science

KEYWORDS : Electrokinetic method, Removal of Sulphates, Chlorides, Extended anode systems, Ionic mobility, Electroosmosis, Electrophoresis, pH maintenance.

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ABSTRACT

Reinforced and prestressed concretes are being used for foundations of many mega structures and some of these underground structures are located in contaminated soil. The contaminants like sulphates and chlorides attack concrete and steel to greater extent and thus many catastrophic structural failures occur. In this paper the theory and philosophy of electrokinetic method of extracting sulphates and chlorides are discussed with their critical issues for effective functioning. Although this method is used widely from 1983 for extracting heavy metals it can also be adopted for removal of detrimental species to concrete and steel in contaminated soil.

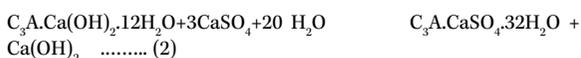
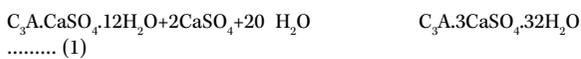
Introduction

Soil pollution has become a very big issue in recent years all over the world [1]. This is due to rapid growth of many industries such as textile industries, metal working industries, electroplating industries, battery industries, paint industries, fertilizer industries, chemical industries and also due to uncontrolled, untreated and treated sewage and waste water to the ground. The organic substances decayed in soil also produce organic acids and microbes [2-11]. Excessive and widely used fertilizers also an important reason for contamination of soil [10, 11]. Mostly clay soil, marine clay soil are contaminated by sulphates and chlorides with many other organic impurities that causes deterioration to concrete [1, 12].

Reinforced concrete sub structures built on contaminated soils or clay, the contaminants such as sulphates (SO_4^{2-}) and chlorides (Cl) along with anaerobic bacteria present in wet soil large amount attacks concrete and reinforcing steel. Sulphates attack concrete and results destruction of concrete by expansion. However, the nature of destruction depends on the cation associated with sulphate anion [13]. The severity of destruction depends on the solubility of various sulphates and pH of their solutions. Usually it is important to consider hydroxide of sulphate salt [14].

Calcium Sulphate Attack

Low reactivity of calcium sulphate results from its low solubility (0.23g/100 g H_2O) and very slight acidity (pH =6.6 at a concentration of 0.01 mol/dm³). This sulphate is neutral. This kind of sulphate has calcium as cation and cement is also having calcium in all its phases and therefore it can form only ettringite from monophases of calcium aluminoferrite hydrates (AFm). The Portlandite [$\text{Ca}(\text{OH})_2$] takes part in the reaction as shown below.

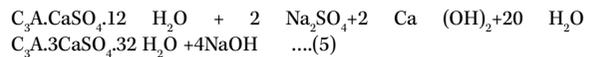
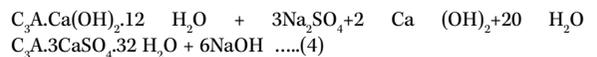


Sodium Sulphate Attack

The solubility of sodium sulphate ($\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$: 44 G/100g H_2O) is greater than that of calcium sulphate. Therefore sodium sulphate attacks on concrete are much higher and severe than calcium sulphate. The pH of sodium sulphate as salt is 7.7 at a concentration of 1 mole/dm³. Gypsum formation from Portlandite [$\text{Ca}(\text{OH})_2$] attack Na_2SO_4 by cation exchange and result in formation of CaSO_4 which will precipitate and crystallize in cement paste microstructure and cause expansion and NaOH is the by-product of the reaction.



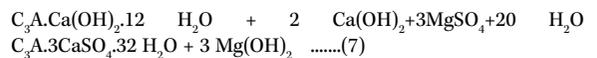
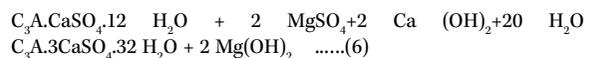
The formation of NaOH increases pH and as a result it reduces the solubility of Portlandite in pore solution and this retards formation of gypsum. When sulphate ion concentration increases locally close to gypsum and monosulphate (AFm) the precipitation of ettringite and expansion will be as shown in following equations.



Therefore numerous microcracks are formed and expansion will be followed and thus failure of concrete by swelling and expansion.

Magnesium Sulphate Attack

Magnesium Sulphate solution is acidic (pH = 5.7 at a concentration of 1 mole/dm³). The solubility is 30.8 g/100 g H_2O . The impact of this magnesium sulphate on concrete is a double chemical attack because of Mg^{2+} and SO_4^{2-} ions. This sulphate is more dangerous for concrete due to the following reactions.

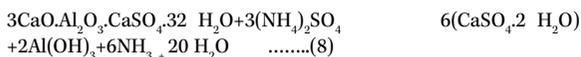


The solubility of $\text{Mg}(\text{OH})_2$ is lower than MgSO_4 magnesium hydroxide gel is precipitated in all reactions of MgSO_4 with $\text{Ca}(\text{OH})_2$ and other phases. $\text{Mg}(\text{OH})_2$ gel and then brucite is formed larger pores of cement paste. Although Mg^{2+} leaves solution by lowering the pH value, the brucite cover retards the attack. Acceleration of calcium ions dissociation in solution and formation of gypsum continues. Therefore, C-S-H gel phase converted to M-S-H phase which will not have any binding properties.

Ammonium Sulphate

Ammonium Sulphate [$(\text{NH}_4)_2\text{SO}_4$] solution is highly soluble salt and its solubility is 75.4 g/100 g H_2O and ammonium sulphate solution is acidic, the pH is 4.62 at a concentration of mole/dm³. The reaction of this salt with calcium phases will be similar to the reaction of sulphuric acid. Ammonium Sulphate attack on concrete causes overlapping dissolution process like acid attack and expansion like salt attack. Ammonium Sulphate solution

acts very rapidly with Calcium Hydroxide. Gypsum precipitates and ammonia gas liberated. The by-product is ammonium hydroxide which decomposes into ammonia in gaseous form. Ammonium Sulphate solution attacks AFm phase and Ca (OH)₂. It results in formation of ettringite and expansion of hardened cement paste. In all reactions between (NH₄)₂SO₄ and calcium phases, ammonium cations NH₄⁺ leave solution. Due to low pH, calcium phases dissociated and gypsum forms.



Finally the decalcification and ultimate decomposition of all cement paste phases will follow. The ultimate product of sulphate attack is gypsum [15].

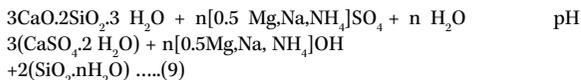


Table 1 Properties of Sodium and Magnesium Sulphate Salts

Salt	Formula	Density	Molecular Volume	Crystallisation pressure(N/mm ²)					
				C/C _s =2		C/C _s =10		C/C _s =50	
				0°C	50°C	0°C	50°C	0°C	50°C
Mirabilite	Na ₂ SO ₄ · 10H ₂ O	1.46	322.19	7.2	8.3	23.4	27.7	39.7	47.3
Thenardite	Na ₂ SO ₄	2.69	142.04	29.2	34.5	97.0	115.0	165.0	196.5
Epsomite	MgSO ₄ · 7H ₂ O	1.68	246.40	10.5	12.5	35.0	41.5	59.5	70.8
Hexahydrate	MgSO ₄ · 6H ₂ O	1.75	228.00	11.8	14.1	39.5	46.9	67.1	30.0
Kieserite	MgSO ₄ · H ₂ O	2.45	56.55	27.2	32.4	91.0	107.9	154.3	184.0

Similarly the chlorides present in soil plays a major role for corrosion of embedded steel in concrete. Free chlorides present in contaminated soil moves into concrete through the pores, voids, cracks, fissures, capillaries and other surface defects. Due to pore connectivity the free chloride travel into concrete and reaches the steel reinforcements surface. Once the accumulated chloride level is increased to threshold limit, the passive film formed on steel surface due to high alkalinity of concrete will be broken and anodic pit will be formed and corrosion cell will act as anode and the rest of the surface area near by anodic pit will act as cathode and the concrete will act as electrolyte. The steel gets corroded as shown below.



This leads to accumulation of rust products around steel bar results radial pressure and subsequently cracking of concrete.

The part of structures i.e., foundations like pile foundation, raft or mat foundation and isolated footings are of reinforced concrete and thus affected significantly by sulphates and chlorides. Therefore, soil pollution has become more and more serious in recent years. In response to a growing need to address soil contamination many remediation technologies are developed among all these electrochemical method or electrokinetic method is best suited for removal of SO₄²⁻ and Cl⁻. Already this method is being adopted for removal of heavy metals from contaminated soil all over the world but not being adopted for removal of SO₄²⁻ and Cl⁻. Therefore, there is a great need for developing effective economical remediation method. Electrokinetic method is most suited and easy removal of SO₄²⁻ and Cl⁻. This can be explored for protecting sub structures, buried oil and other pipelines in Civil Engineering projects either in-situ or ex-situ.

Mechanism of sulphate attack expansion of concrete is not clearly understood and it is still a subject of numerous discussions and hypothesis. The two most discussed concepts in recent decades say that the expansion occurs due to the additional volume generated by ettringite. The relationship between the volume expansion and the stress developed is not well documented. Ettringite formation seems to be a necessary condition for expansion. It is reported [16] that the expansion is caused by the crystallization pressure, due to the formation of ettringite from supersaturated solution in small pores. The theory of crystallization pressure has been explained in a detailed manner by recent observation [15]. Table 1 shows the properties of sodium and magnesium sulphates. Therefore, sulphates disintegrate cracks and disrupting by swelling and volume increase and concrete reduces its cross section area and leads to poor load bearing capacity.

Electrokinetic Method

Electrokinetic method of soil remediation technique involves appreciation of low voltage direct current (DC) is in the order of 1 volt per cm distance between anode and cathode or current is applied in the order of mA/cm² of electrode area. The soil is a medium and the water acts as electrolyte. This low level DC results in physio-chemical and hydrological changes in the soil mass resulting in species transport in the porous soil medium, voids, water, organic, decomposed organic matters, mild organic acids and microbes.

When the DC current is applied across the electrodes in electrokinetic bath, the water present in the soil undergoes water splitting reaction i.e., oxidation of water at the anode generating acidic environment near anode and reduction of water at the cathode creating an alkaline front at the cathode as per equations 12 and 13. The Fig.1 shows the principle of electrochemical method of treating soil for removal of SO₄²⁻ and Cl⁻ at anode.

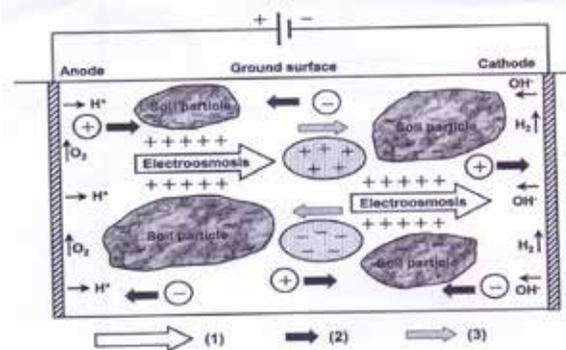


Fig.1 Principle of Electrokinetic Method

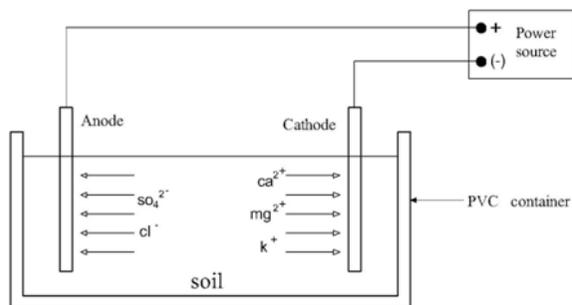


Fig.2 Mechanism involved for removal of sulphates and chlorides

The Fig.2 shows the principle mechanisms involved in electrokinetic method are electromigration (movements of charges species in a solution caused by an applied electric field),electroosmosis (movement of liquid relative to a stationary charged surface caused by an electric field) and electrophoresis (movement of charged particles e.g. colloids, clay particles and organic particles relative to stationary liquid caused by an electric field).Several external variables also contribute to this transport mechanism such as total mass flux, soil mineralogy, pore fluid composition and conductivity, electrochemical properties of the species in the pore fluid and porosity of the soil medium, relative humidity and pH of the soil medium. The ionic mobility of some selected cations and anions are shown in Table 2 [21, 22].

Table 2 Ionic Mobility of selected anions and cations in free electrolytes at 25°C

Element	Ionic Mobility ($10^{-8} \text{ m}^2/\text{Vs}$)	Element	Ionic Mobility ($10^{-8} \text{ m}^2/\text{Vs}$)
H ⁺	36.23	Na ⁺	5.19
Al ³⁺	6.32	Pb ²⁺	7.36
Ca ²⁺	6.17	Zn ²⁺	5.47
Cd ²⁺	7.36	OH ⁻	20.64
Cr ²⁺	6.94	Cl ⁻	7.19
Cu ²⁺	5.56	NO ₃ ⁻	7.40
Fe ³⁺	5.60	SO ₄ ²⁻	8.29
K ⁺	7.62	CO ₃ ²⁻	7.46

It can be seen from Table 2 that the ionic mobility of H⁺ and OH⁻ ions are significantly higher than other ions. It is reported [23] that the electroosmotic mobility in soil is usually in the order of $5.0 \times 10^{-9} \text{ m}^2/\text{Vs}$. This order is lower than the ionic mobility of heavy metals in soil. The electrophoretic mobility in soil varies between 10^{-10} and $10^{-9} \text{ m}^2/\text{Vs}$ [23].This order is also lower than electroosmotic mobility and the ionic mobility. Therefore, electrophoresis becomes significant in electrokinetic soil remediation. But the colloidal particles transport by electrophoresis is of little importance in compacted soil.

Critical Analysis of the Method

In this electrokinetic method as stated earlier, acidic and alkaline environments are created at anode and cathode respectively. Higher alkalinity causes precipitation of salts nearby cathode. Similarly the pH is not equal in soil body. Therefore to control pH and zeta potential and also to increase mobility of ions many studies are carried out [24-27].Ion exchange membrane was introduced near electrode to improve the electrokinetic activity [24] to control hydroxyl advancements at cathode area, cation exchange membrane was introduced by Li et.al. [25].Soil membrane were introduced to reduce or eliminate the drawback of energy consumption and to ensure proper distribution of H⁺ ions and their mobility [28] .As a further development, extend-

ed anode systems that is providing larger anode area and also to reduce the distance between anode and cathode, Reddy and Chinthamreddy [29] suggested a new technique. Many bench scale models and pilot plant scale field electrochemical extraction of heavy metals from different kinds of soils including sand, silt, kaolinites, illites, bentonites, natural soils and various artificial soil mixtures were studied by various researchers [30-42] and results of these studies reports, that soil type does not pose any significant limitations on the technology but transport rates of the contaminant and efficiencies depend heavily on soil type. Saturated soil provides favourable condition for transport of contaminants by electroosmotic process and ionic migration. It is also reported that [43,44] electrochemical method provides effective way to treat heterogeneous soils with varying hydraulic conductivity.

Electromigration primarily depends on applied electric field and as a result it helps electroosmosis advection arête and thus more uniform clean up of heterogeneous soil deposit. The energy consumption for different soil depends on electrical conductivity of each soil. It is also reported [45-47] that contaminants in the form of colloidal can be removed by the combined effect of electroosmotic advection and electrophoresis. It is also reported that type of contaminant does not pose a significant limitation on the electrokinetic technology provided that the contaminants does not exist in a sorbed phase on soil particles. It is possible to remove high concentration of ions but this high concentration of ions increases electrical conductivity of the soil and the efficiency of electroosmotic fluid flow is reduced, therefore in such a case, the applied electric field shall be reduced [48].The decomposition of water into H⁺ and OH⁻ ions transported in soil mass and as a result the pH of soil is reduced. This low pH of soil impedes electroosmosis and efficiency is reduced. Therefore it is essential to maintain pH in sufficiently low so that it can make the metal contaminants in a mobile phase.

Maintaining pH in soil is an important aspect for successful contaminant removal. Many researches [49-57] suggested using various chemicals including acetic acid, NaOH, use of membrane, buffer solutions, and use of some special electrodes and finally circulating anolyte and catholyte. The method and chemicals to be used will vary according to the contaminated to be removed.

Conclusions

- Electrokinetic method of soil remediation is easy, efficient and eco friendly for removal of sulphates and chloride from sub soil.
- Extended anode system with large anode area will reduce the acidic front at anode and will reduce the energy consumption.
- Various chemicals, methods are discussed to bring the pH to effective functioning.
- Transport properties by electroosmosis, ionic migration and electrophoresis are discussed with their mechanisms to improve.

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