Electrokinetic Remediation of a Contaminated land in Cities

VLADIMIR A. KOROLEV

Lomonosov Moscow St. University, Geology Faculty, Russia (e-mail: korolev@geol.msu.ru)

Abstract: The large territories of modern cities are polluted by the various components, including toxic pollutants. Remediation of urban territories from pollution is one of the important problems of engineering geology and ecology. We develop electrokinetic methods of soil remediation for contaminated land in cities. In the electrokinetic remediation-method, contaminants are removed from soil and soil water by the action of an electric potential applied across electrodes embedded in the contaminated medium. This method is applied to removal from the soil pore solution of charged toxic components, such as ions of heavy metals (Pb, Cd, Zn, Cu, As, Cr, Hg etc.), cyanide complexes, nitrates, chlorine, radionuclides etc. This method can also be applied to remove many organic toxic substances, such as phenol, chlorinated ethane and hydrocarbons, pentachlorophenol and others. We have established that this method removes the petroleum and mineral oils from soils. During the past 10 years some electroremediation laboratory projects have been finished successfully on the Department of Civil and Environmental Engineering of Geological Faculty of MSU.

Résumé: De grandes places des villes modernes sont polluées par les composants divers nuisibles. Le nettoyage des territoires des villes selon les pollutions - un de des importants problèmes de la géologie de génie et l'écologie. Nous élaborons électrique les moyens du nettoyage des territoires pollués dans les villes. Dans cette méthode la pollution bouge dans le sol et l'eau dans ses époques sous l'effet du champ du courant constant électrique. Cette méthode est appliquée pour l'éloignement du sol des tels composants nuisibles comme les métaux lourds (Pb, Cd, Zn, Cu, As, Cr, Hg etc.), les nitrats, le pétrole et les produits pétroliers, les composants radioactifs etc. Au Cours des derniers 10 ans sur la chaire de la géologie de génie et écologique de l'université De Moscou la série de tels projets était accomplie avec succès.

Keywords: contaminated land, ecology, environmental protection, hazardous waste, remediation, pollution

INTRODUCTION

According to the United States Environmental Protection Agency (USEPA 1989), there are over 217,000 sites in the U.S. that urgently require remediation, and the estimated cost is over $187 billion U.S. dollars. The cleaning of the polluted territories of cities from the hazardous waste is one of the major problems of modernity. Unfortunately, the area of the polluted soils in territories of cities, contaminated lands is more and more increased from year to year. Thus the pollution of the urban territories is caused by deliberate release, or spontaneously as a result of various failures during transport, in places of warehousing and processing of the toxic components, as a result of various emissions, dumps, drains etc. In the polluted urban territories, toxic components are contained in various parts of the ecosystem: in air, in plants, in soil, in underground waters etc. Therefore development of effective technologies of cleaning soils that contain toxic pollution has huge practical importance.

Electrokinetic methods represent a novel approach to cleaning soils. These methods of cleaning are based on use of electrokinetic processes, which occur in soils under action of a constant electrical current. The development of electrokinetic methods of soil cleaning has been mainly carried out in the advanced industrial countries, such as Great Britain, USA, Netherlands, Germany, Denmark, Japan, Canada etc. (Eykholt & Daniel 1994, Hansen et al. 1999, Ho, et al. 1999a, 1999b, Lageman 1993, Lageman & Pool 2001, Probstein. & Hicks 1993, Reddy, Adams & Richardson 1999, Reddy et al. 1999, Yanko-Hombach & Korolev 2002.). In Russia, unfortunately, this problem has not received the necessary attention. But during the past 10 years some electroremediation laboratory projects have been finished successfully on the Department of Civil and Environmental Engineering of Geological Faculty of MSU (Korolev 2001a,b, Korolev & Nekrasova 1996, 2000, Nekrasova & Korolev 1997b,c, Korolev, Babakina & Mitojan 2001). The advantage of these methods is caused by high efficiency, by ecological safety and the opportunity to influence soils insitu. The present paper is devoted to the analysis of opportunities of the given methods for the rehabilitation and remediation of a contaminated land in cities.

PRINCIPLE OF ELECTROKINETIC REMEDIATION OF CONTAMINATED LAND

The electrokinetic cleaning of soils is based on processes of electroosmosis and electromigration, which arise in a water-saturated soils at imposing a field of a constant electrical current (Acar & Alshawabkeh 1993,1995, Chaparro1999, Dzenitis 1996, Korolev 2001a, 2001b, Shapiro & Probstein 1993, Yeung 1994.). If a toxic components in the water environment (in the water-saturated soils) dissociate on the anions and cations in a field of a constant electrical current the negatively charged components (anions) will be migrate to a positive pole (cathode), and the positive - to negative (anode). As a result of this electrical migration the division of toxic components in soils can be generated (Fig.1) (Korolev 2001a).
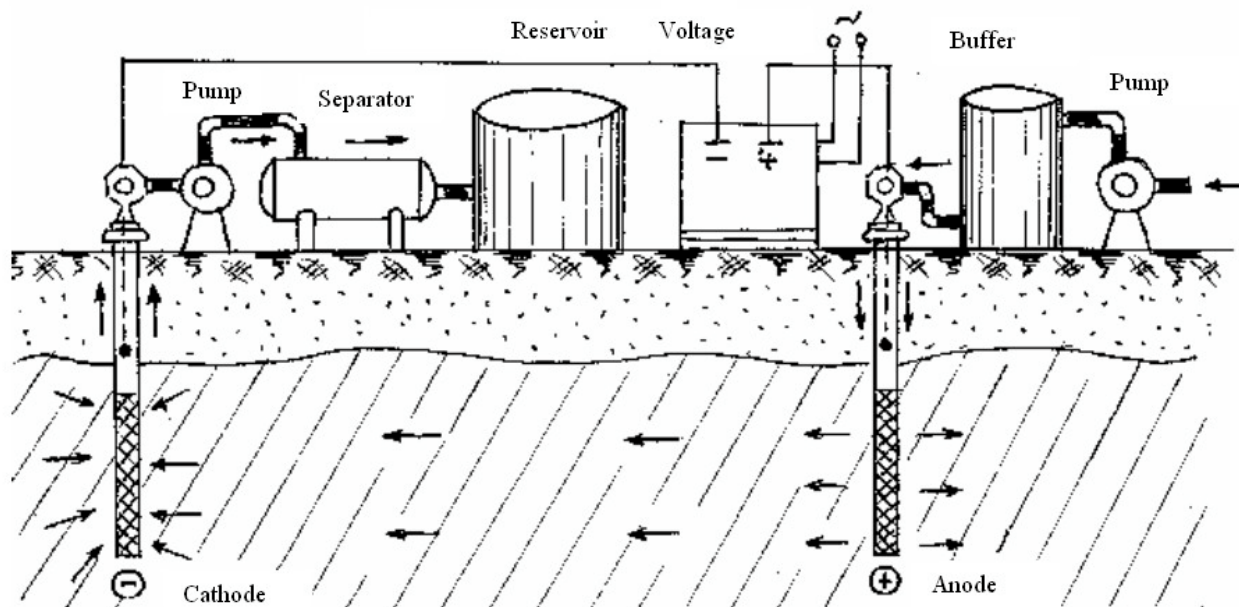
Figure 1. Scheme of electrokinetic remediation of the contaminated land

The electrokinetic cleaning of soils is increasing in many countries of the world. In the different countries many commercial companies are formed and successfully work which realize the various projects on cleaning territories from the many toxic substances: heavy metals, organic connections, petroleum and radionuclides. The countries, in which there are a lot of such companies, are Great Britain, Germany, the Netherlands, USA, Denmark and over (Hansen et al. 1999, Lageman 1993, Lageman & Pool 2001).

METHODS OF LABORATORY RESEARCH

We conducted a laboratory study of electrochemical migration of pollutants in the electrokinetic cells under both flowing and non-flowing conditions (Korolev 2001a).

The non-flowing variant the electrokinetic cell consists of a glass tube of length 10 cm and diameter 20 mm, in which the researched soil was located. The sample was loaded in the tube and compressed by condensing covers from Plexiglas, and they were separated from the soil by rubber rings and paper filters. The platinum electrodes were established on end faces and contacted the soil. For removal of gas and liquid formed during electrochemical cleaning, drainage via an aperture was allowed in the electrokinetic cell. After preparation of the device the difference of a voltage cleaning applied resulted in a current of 10 mA.
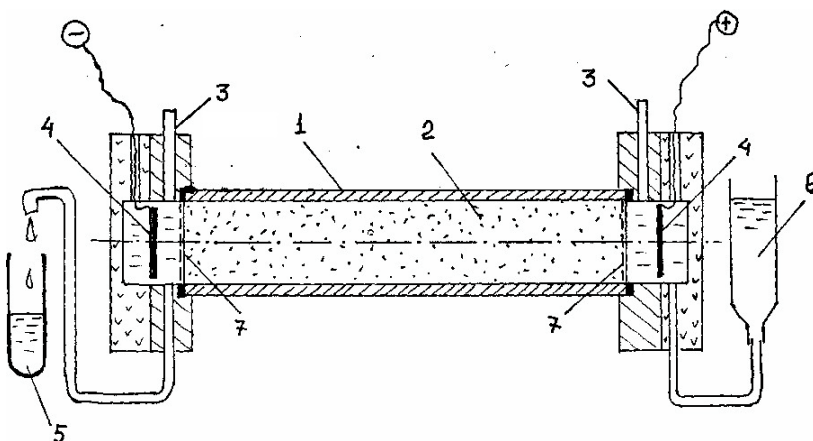
Figure 2. The device for tests in flowing mode: 1- tube, 2-soil, 3 - tube for removal of gases, 4 - platinum electrodes, 5 - capacity for the tax filtrate. 6 - vessel for flushing liquid (water), 7 - paper filters

The degree of cleaning of a soil was also studied in flowing mode (fig. 2). In this variant the tube containing a researched soil was fixed from both ends by electrode chambers filled with water or solutions, to which the platinum electrodes which are not contiguous closely to a soil were attached. But, as well as in the first variant, for prevention of washing out of a soil in electrode zones the rubber rings and paper filters were used. The capillary from the tank with water fell to the lateral chamber with the positively charged electrode (fig. 2). Water from the tank acted in the

electrode chamber at downturn of a level, thus capacity with water was fixed so as to prevent occurrence of a pressure head gradient in a capillary. From the electrode chamber with the negatively charged electrode, in which acted following from a soil filtrate, the solution got in capacity-store, with which help the charge of a liquid was supervised. Thus, in a flowing mode on a measure electroosmotic of cleaning water from the lateral chamber acted in a soil and gradually washed out it. To avoid non-uniform drainage, current was maintained at 1-4 mA and field intensity no more than 20 V/cm.

Upon termination of experiment the anode, cathode and intermediate parts of a sample were analysed for the remaining pollutant and moisture content. Quantity of heavy metals in tests we determined by a X-ray dispersed method, quantity of nitrates and phenol - by a photo-colorimetric method, quantity of petroleum and oil - by a thermogravimetric method, and quantity of radioactive substances - spectrometer method. Clay soils of different mineral composition and structure were investigated.

ELECTROCHEMICAL REMOVAL OF HEAVY METALS

The heavy metals are widespread among others pollutants in territories of many cities. In a field of a constant electrical current heavy metals moved in the clay soils in the several forms. Numerous works as domestic (Korolev 2001a, Korolev, Babakina and Mitojan 2001, Korolev, Nekrasova and Mitojan 1998, Nekrasova and Korolev 1997a), and foreign authors (Chinthamreddy 1999, Chinthamreddy and Reddy 1999, Hamed, Acar and Gale 1991, Reddy and Chinthamreddy 1999, Reddy, Chinthamreddy and Al-Hamdan 2001, Reddy. And Parupudi 1997, Reddy et al. 1997, 2002, Reddy and Shirani 1997, Reddy, Xu and Chinthamreddy 2001) are devoted to the analysis of electrokinetic cleaning of soils from heavy metals.

The results of our laboratory researches of electrochemical migration of ions cadmium (Cd^{2+}), lead (Pb^{2+}) and zinc (Zn^{2+}) in the monoionic forms of loam (tab. 1) have shown, that the features of interaction of heavy metals with clay minerals influence their electrochemical migration in a soil. Under action of the constant electrical current the concentration of heavy and transitive metal ions is reduced by 50-90%. It testifies to the large efficiency of this cleaning method.

Table 1. Estimation of the electrochemical redistribution of metal ions in the loams

Quantity of ions, %:	Mg-loam	Zn- loam	Pb- loam	Cd- loam
Removed from the soil with the filtrate	20.5	-	41.2	42.3
Precipitated on the electrode	0.003	0.001	4.5	1.6
Redistributed in the pore solution	10.5	81.8	34.3	24.1
Redistributed in the exchange complex	69.0	12.8	20.0	32.0
In total:	100	94.6	100	100

The remaining quantity of heavy metals ions in the soil was redistributed along the length of the sample. The forms of a presence of heavy metals in the soil depend on the kind of metal and environmental factors. Change of environment pH and chemical properties of ions exert the large influence on electrochemical migration of heavy metal ions. During action of an electrical field in an anodic zone the pH is reduced, and in cathode zone, raised.
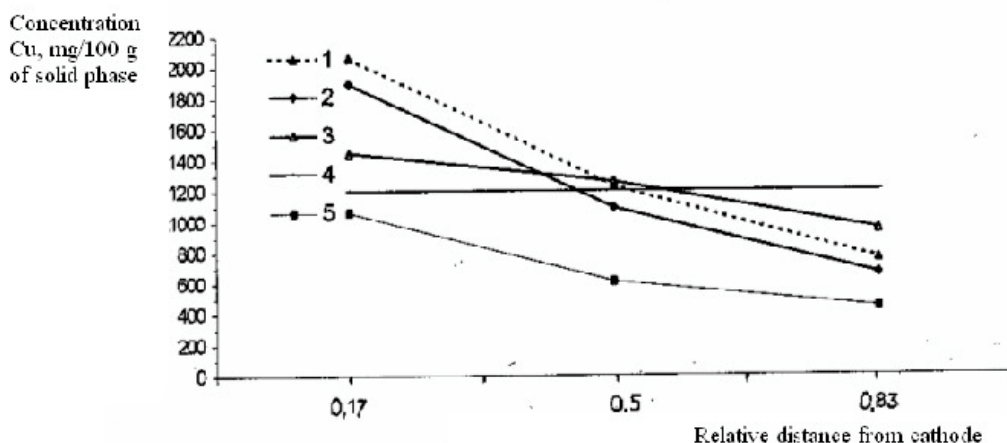

Figure 3. The change of the cupper ion content in the clay soils under the influence of a constant electrical current field: 1 – moraine clay sand, 2 - moraine loam, 3 - kaolinite clay, 4 - initial concentration of copper; 5 – illite clay

The comparative data on cleaning the soils of different mineral and lithological composition contaminated with copper are submitted in a fig. 3. From this figure it is visible, that in kaolinite clay, clay sand and loam the change of the pollutant content in cathode, intermediate and anodic zones is almost identical, and the change of the contents of copper ions in the illite clay occurs strongly.

Thus, the high degree of the clay soils cleaning from the various heavy metals is achieved in anodic parts of samples, that allows us to apply this method for the cleaning of contaminated land.

ELECTROCHEMICAL REMOVAL OF PHENOL

A large number of papers are devoted to a problem of the electrokinetic cleaning of soils contaminated with phenol, spirits and others toxic organic substances (Haus et al. 2001, Korolev, Babakina and Lazareva 2001, Li, Cheung and Reddy 2000, Saichek 2002). Efficiency of phenol removal was investigated in the various clay soils (clay sands and loams), in which the known phenol quantity, exceeded the limiting allowable concentration for soils by tens or hundreds of times.

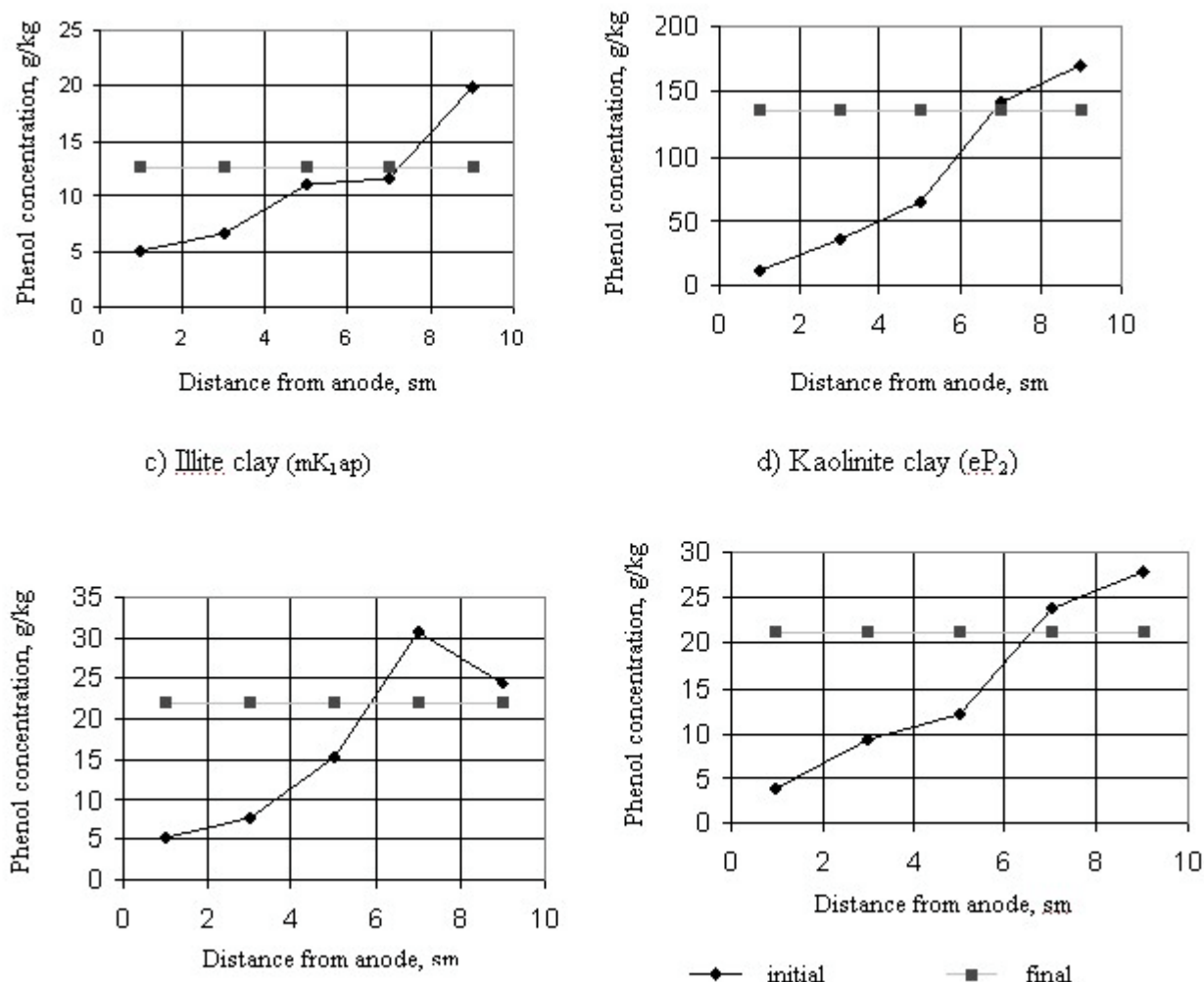On fig. 4 the diagrams of the phenol redistribution on the length of samples clay soils are shown.

c) Illite clay (mK₁ap)

d) Kaolinite clay (eP₂)

Figure 4. The diagrams of the phenol redistribution in the clay samples of various mineral compositions (a-d) in the result of the electrokinetic cleaning

From this figure it follows that the phenol moves from the anode to the cathode in a field of a constant electrical current. Thus the anodic zone is cleared from the phenol to the greatest degree.

The degree of electrokinetic cleaning of the various clay soils from phenol and the separation efficiency is shown in table 2. It can reach size 92-95%.

Table 2. The separation efficiency of clay soils from phenol

Clay Soils	Degree of Remediation (%) in:		Phenol accumulation in cathodic zone, %
	Anodic zone	Middle zone	
Montmorillonite	92	74	25
Clay	86	56	32

Kaolinite Clay	66	55	11
Polymineral Clay	59	48	39
Polymineral Loam	55	32	4
Illite Clay			

Thus, the efficiency of cleaning clay soils from the phenol grows with increasing dispersibility. The peak efficiency of the soil electrochemical cleaning from the phenol occurs in samples with the free swelling humidity.

ELECTROCHEMICAL REMOVAL OF PETROLEUM

In 1995 we have established, that the drops of petroleum in the dispersive soils can be removed from the soil in a field of a constant electrical current. After that we have conducted numerous studies on factors influencing on the electrokinetic cleaning of soils from petroleum pollution (Korolev 2001a,b, Korolev et al. 2001). We have established, that the drops of petroleum move to the cathode together with a electroosmotic water flow and are involved in movement at the expense of viscous friction.

The results of experiments have shown increase of moisture content from the anode to the cathode along an axis of a soil sample after realization of the electrochemical remediation. Hydrocarbon remaining in the sample was redistributed from the anode to the cathode along the axis of a sample. Thus, the concentration of the hydrocarbon is reducing except near the cathode.

The researches have shown increasing of pH during the electrochemical soil remediation from petroleum pollution along the axis of the sample from anode to the cathode.

In the whole amount removal petroleum made from 30 up to 50 % from initial oil content. In some cases the amount of the remaining petroleum in the sample at the anodic part exceeded 4 times amount the pollutant remaining at the cathode part. The degree of sample remediation from petroleum pollution reached up to 70 – 80 % in the cathode part in some cases.

The structure of hydrocarbons affects their electrochemical migration in clay soils. The high-molecular fraction on some data reduces its electrochemical mobility. And the low-molecular hydrocarbon compounds are removed from the sample with greater efficiency.
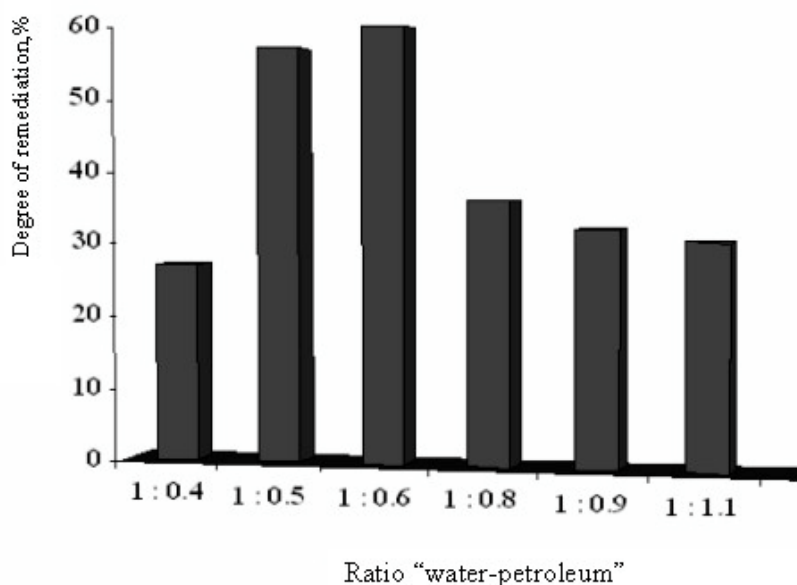

Figure 5. Influence of a ratio "water - petroleum" on a degree of the electrokinetic cleaning of the grey forest soil from the petroleum

The intensity of hydrocarbon electrokinetic migration increases with the specific fraction of petroleum in water, but at the ratio of volumes "water - petroleum" close to 1:0.8 and more the migration intensity begins to reduce. The maximum quantity of petroleum (60 %) was removed from a sample at the ratio "water - petroleum" of 1:0.6 (see fig. 5).

ELECTROCHEMICAL REMOVAL OF RADIONUKLIDS

The dispersive soils, infected by the radioactive substances, also can be cleaned with the help of the electrokinetic method. As a substances simulating radionuclids Cs-137 and Sr-90, we used the salt of their stable isotopes having the similar ion radiuses, and also physical and physical-chemical characteristics in solutions of the appropriate salts

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cesium CsCl and strontium $Sr(NO_3)_2$. In the other part of experiments which are spent together with the firm "Radon", as a radioactive pollutants we used directly radioactive elements Cs-137 and Sr-90.

It was established, that the electrokinetic cleaning from radionuclids is possible also in artificial sandy screens, fixed by the aluminosilicate gel and created as the protective barriers on the way of the liquid radionuclide migration. In the result of the electrokinetic processing of such soils the general redistribution of the strontium contents (Sr-38) is formed on length of the sand-gel sample from the concentration 1.4 mg/g in the anodic sector up to the concentration 5.6 mg/g in the cathodic sector. In the same soil the concentration of cesium (Cs-133) varies within the limits of 2.0-4.8 mg/g and is formed in that a direction at imposing a field of the constant electrical current (at the initial concentration of elements in the sand-gel samples 3.5 mg/g).

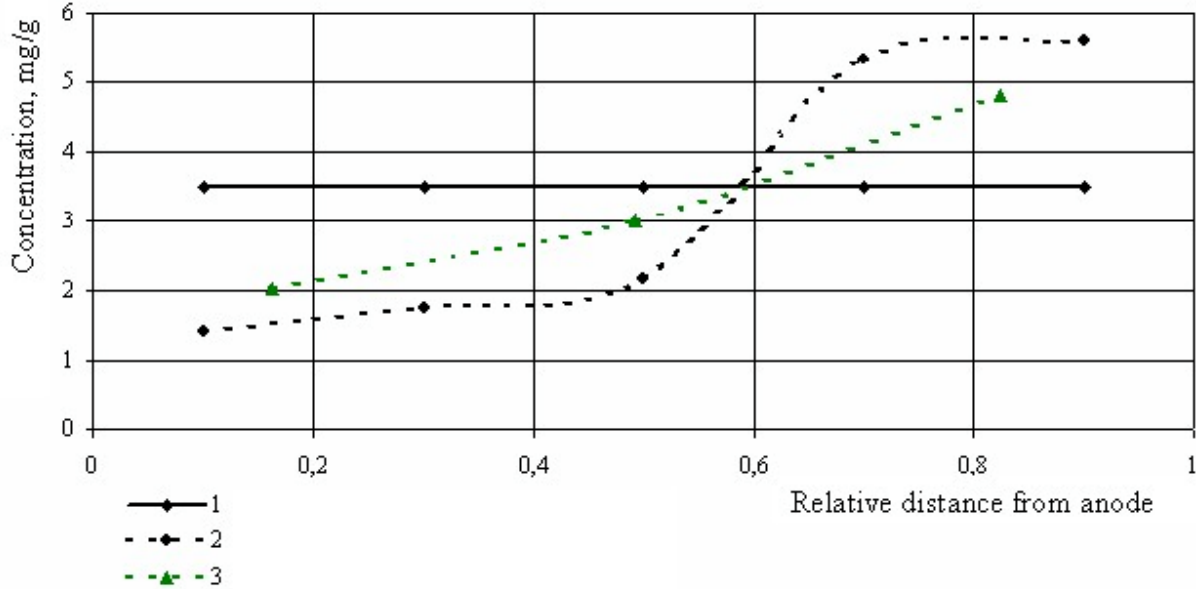On the fig.6 the experimental results on cleaning of soils from radionuclids are shown.

Figure 6. The change of strontium (Sr) and cesium (Cs) concentration on the length of a sand-gel sample in result of the electrokinetic cleaning: 1 – initial Sr and Cs concentration; 2 –Sr concentration after cleaning; 3 –Cs concentration after cleaning

Conducted by us together with the firm "Radon" the laboratory and half-industrial stand experiments have shown rather high efficiency of the electrokinetic extraction of radioactive Cs-137 from the clay soils. In this case NH_4NO_3 was used as a buffer solution in the flushing mode. The degree of radionuclid extraction for 2 months was about 25 %; with increase of duration of electroprocessing the degree of extraction can be raised up to 50 % and more.

MATHEMATICAL MODELING OF THE ELECTROKINETIC REMEDIATION

The modeling of electrokinetic redistribution of cations-pollutants in the soils can be carried out by the different ways. First, this process can be described on the basis of thermodynamics of irreversible processes theory. In a basis of this approach the analysis of system of three linear equations for streams of a moisture (J_w), an electricity (J_i), and cations-pollutants (J_c), arising under the influence of a gradient of potential of a moisture (∇P), electric potential ($\nabla \varphi$) and a gradient of concentration (∇C), accordingly lays:

$$\begin{cases} J_w = K_w(\nabla P) + K_{eo}(\nabla \varphi) + K_{13}(\nabla C) \\ J_i = K_{pp}(\nabla P) + \chi(\nabla \varphi) + K_{23}(\nabla C) \\ J_c = K_{31}(\nabla P) + K_{32}(\nabla P) + D(\nabla C) \end{cases} \qquad (1)$$

here K_{ij} – is the phenomenological coefficients which have the following sense: K_w - coefficient of a filtration; K_{eo} - coefficient of electroosmosis; K_{pp} - coefficient of flow conductivity; χ - electroconductivity; D - coefficient of effective diffusion. These thermodynamic ratios are observed at small size of gradients. Such approaches to modeling electrokinetic transport of cations in clay soils was applied in a number of works (Korolev 2001a; Hansen et al. 1999, 2001). However, application of such method demands knowledge of many phenomenological coefficients which cannot always easily can be determined experimentally.

Second, the modeling of the cations-pollutant flow can be carried out on the basis of the decision of the differential equations of diffusion and convection as:

$$\partial C/\partial t = D\ (\partial^2 C/\partial x^2) - v_e(\partial C/\partial x), \tag{2}$$

here D - is the effective diffusion coefficient; v_e - the effective velocity of cations movement in an electrical field . This equation is supplemented with the different ratios which allow taking into account such processes, as adsorption, electromigration, dissolution etc.

In our work the mathematical description of electrokinetic migration of cations-pollutants on a basis of the binomial linear equations is offered.

The electrokinetic removal of pollutants (such as heavy metals, radionuclids, organic toxically components etc.) from soils is carried out under influence of several processes: the cations movement together with electroosmotic filtrate, the ions movement as a result of electromigration and electrolysis of a pore solution, the movement of the charged pollutants particles under action the electrophoresis etc. Diffusion is a factor interfering electrokinetic migration of ions, directed against a gradient of ions concentration. We have applied to mathematical modeling of this complex process model, which takes into account only the electroosmotic and electromigration movement of cations with velocity (v_{ek}), and also the diffusion process of ions by velocity (v_D), interfering the electrokinetic migration. In this case we have:

$$v = v_D - v_{ek}, \tag{3}$$

hear v - is the linear effective velocity cations movement.

As is well known, this process is described by the differential equation of a kind:`

$$\partial C/\partial t = D\ (\partial^2 C/\partial x^2) - v_e(\partial C/\partial x), \tag{4}$$

here D - is the effective diffusion coefficient; v_e - the effective velocity of cations movement in an electrical field . The value v_e can be expressed through the effective cations mobility U:

$$v_e = E(U/F), \tag{5}$$

here   - is the intensity of an electrical field; F - the Faraday constant.

The value of linear cations velocity (v) is defined by the equation in a binomial form:

$$v = v_D - v_{ek} = \text{D}\bullet(\partial C/\partial x) - \text{K}\bullet(\partial\varphi/\partial x), \tag{6}$$

here D - is the diffusion effective coefficient; K - the electrokinetic coefficient of the cations migration; v_D and v_{ek} − is the linear velocity of diffusion and electrokinetic migration, accordingly.

Let's consider the experimental circuit of electrokinetic migration, which is shown in Fig.7. Here diagrams of distribution of cations concentration at the various moments of time t_i are shown.
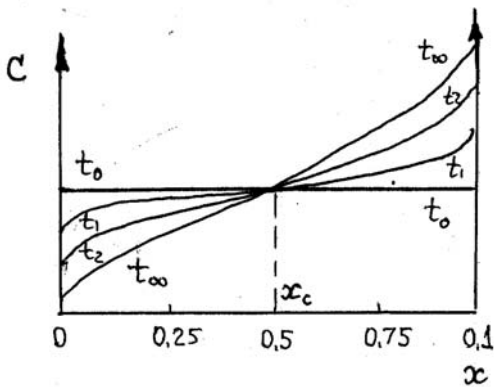

Figure 7. The redistribution of the cations concentration (C) on the length of a soil sample (x) in the various moments of time t_i

Here are designated: C - is the concentration of the cation-pollutant (mg/g of solid phase of soil), $C = C_i/C_o$, where $_i$ - is the concentration at any moment of time t_i; C_o - the initial concentration in sample (on time t_o); x - the relative distance from the anode ($x = x_i/L$, where L - the length of a sample, or distance from the anode up to the cathode; x_c – the middle of sample); t - the time of action of the enclosed electrical voltage φ.

The experiments show, that as a result of electrokinetic migration the cations distributed on the length of a sample so, that their concentration is increased from the anode to the cathode: $C = f(x)$, (Fig. 7). This distribution is formed not at once, but for the some interval of time Δt, i.e. $C = f(x,t)$. A series of diagrams distribution of a cations concentration may be received for the different intervals of time: t_o, t_1, $t_2...t_\infty$ etc. Finally at the moment of time of the experiments ending ($t_i = t_\infty$) the structure of the maximal cations redistribution on the length of a sample is formed.

The actual observable linear velocity of the cations flow transfer (v) from the anode to cathode for an interval of time $\Delta t = t_2 - t_1$ through average section of a soil sample (i.e. on distance $x_c = 0.5$) is equal:

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$$v = J_{wc} / C = \frac{\left(\int\limits_{0}^{0,5} C\,dx\right)_{t_2} - \left(\int\limits_{0}^{0,5} C\,dx\right)_{t_1}}{t_2 - t_1},$$ (7)

hear J_{wc} – average volumetric density of an electrokinetic stream of cations-pollutants in the time $\Delta t = t_2\text{-}t_1$ through unit of area S; $J_{wc} = m_c / \Delta t \bullet S$; m_c - mass of cations; C - concentration of cations, $C = m_c/m_s$; m_s - mass of a solid soil phase. Integrand expressions in this equation are easily determined by graphic integration or a numerical way by results of experimental definitions of redistribution cations concentration on the length of a sample at the various moments of time t.
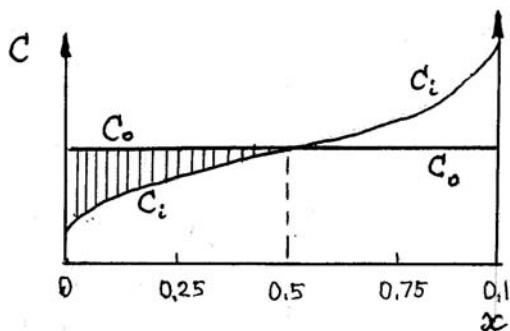

Figure 8. Redistribution of the cations concentration (*C*) on the length of a soil sample in the moment of time t_i

For average section of a sample it is possible to determine quantity of cations, past through this section in the unit of time. On Fig. 8 shaded area on the diagram shows quantity of cations, redistributed through this section in time $\Delta t = t_2\text{-}t_1$.

Then, for the different moments of time (t_0, t_1, t_2 etc.) it is possible to make system of the linear equations (8):

$$\begin{cases} v_1 = D \cdot \text{grad} C_1 \text{ - } K \cdot \text{grad} \varphi_1 \\ v_2 = D \cdot \text{grad} C_2 \text{ - } K \cdot \text{grad} \varphi_2 \\ \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \\ v_i = D \cdot \text{grad} C_i \text{ - } K \cdot \text{grad} \varphi_i \end{cases}$$ (8)

The value of coefficients D and K can be found by the solution of the redefined system of the equations (8) (by the Simpson method). Values of velocities v_1, v_2,...v_i find by the solution of the equation (4).

In the stationary condition, when v = 0, and also $v_D = v_{ek}$ from the equation (4) follows:

$$(\partial C/\partial \varphi) = K/D = \delta_{ek},$$ (9)

here δ_{ek} - is the electrogradient concentration coefficient.

Thus, the knowing electrogradient concentration coefficient is possible to calculate one of coefficients D or K on known to one of them. The offered approach allows simulating the electrokinetic removal of pollutants from soils.

Processing of the skilled data can be carried out in conditions of normalization of the cations concentration values. In this case their concentration (C, mg/g) is replaced with relative dimensionless concentration C ' (Fig. 9).
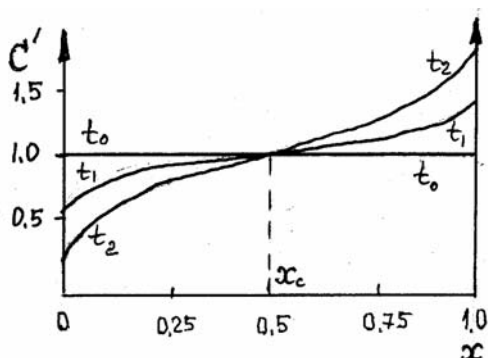

Figure 9. Distribution of the relative cations concentration (*C'*) on the length of a soil sample in the various moments of time t_i

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Here are designated: C' - is the relative concentration of the cations-pollutant; $C' = C_i/C_o$, where C_i - is the concentration at any moment of time t_i; C_o - the initial concentration; x - the relative distance from the anode ($x = x_i/L$, where L - the length of a sample, or distance from the anode up to the cathode; x_c – the middle of sample); t - the time of action of the enclosed electrical voltage φ.

CONCLUSION

Thus, the research reported proves the high efficiency of the electrokinetic cleaning method. It can be applied for the remediation of contaminated lands from the various, organic and inorganic pollutants: heavy metals, nitrates, phenol, petroleum and oils, and also radionuclids.

The practical and the ecological-geological meaning of the executed work is that these data can be used for the development of an industrial method of the electrochemical cleaning of polluted soils. It has the large importance for the decision of many ecological problems connected to the rehabilitation of urban territories, polluted by the heavy metals, nitrates, phenol, petroleum or oils and radionuclids. The further development and perfection of this method can put it on the first place among the ways of the soil cleaning from the pollution in situ, and, hence - for the improvement of the geological environment condition and ecosystems as a whole.

Corresponding author: Russia, 119899, Moscow, Vorobjevy Gory, MSU, Geological Faculty, Dept. of Engineering and Ecological Geology

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