

On electrokinetic soil radon mitigation: a first theoretical approach

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Abstract

Electrokinetics and its potential significance with regard to soil radon (^{222}Rn) mitigation is investigated. Whereas the use of electrical fields for chemical soil decontamination, also known as electrokinetic remediation (ER), is a consolidated commercial technology, however, its potential use to tackle the radon-soil problem has not yet been explored. One explanation behind is that traditional ER requires the use of electrolytic solutions injected into the soil to form chemical species in an ionic state and then being affected by the electrical potential. Radon is a chemically inert gas unable to form chemical species, and in any case, the continuous injection of electrolytic solutions underneath houses is clearly not an option. Here, it will be demonstrated that the same radioactivity of the soil responsible for the generation of radon might also provide a key for its removal. Utilizing a simplified physical model, it was shown that owing to radioactive background surrounding the pores of the soil through which radon travels toward the surface, they become the preferential centers of ionization, and in fact, for very small pores (through which rocks and specially granite stones absorb and diffuse gases), they are positively polarized.

Keywords: *environmental radon; radon mitigation; electrokinetic remediation*

Electrokinetic remediation (ER) is the technique used to remove chemical ions from soils. This technique, also known as electrokinetic soil processing or electroreclamation, is a consolidated commercial technology, which has been used for soil decontamination such as pesticides, mine tailings, or heavy metals. In ER, a direct power source generates an underground electrical potential, which makes that chemical ionic species migrate beneath the soil surface to the electrodes either by electrophoresis or ionic migration (1).

On the other hand, radon (^{222}Rn), which is generated from the radioactive decay of uranium contained in rocks and soils, enters homes through crevices and cracks and small pores and gaps in the floor, and nowadays represents the most important cause of lung cancer after smoking (2). Several approaches and strategies for radon mitigation have been developed in the past decades, for example, ventilating indoor concentrations, reducing radon entry, or removing radon source (3). Among all the sources to indoor radon level, it is acknowledged, since the early research, that the direct infiltration of soil gas into the house is by far the largest contribution to indoor levels (4).

The objective of this work was to analyze the possibility to use underground electric potentials to tackle the radon-soil problem, which has neither been explored nor

mentioned until now, as far as the author knows. It must be pointed out that although ER has already been discussed with regard to nuclear waste-contaminated soils and to soils contaminated for uranium or ^{137}Cs (5–8), it is based in traditional ER, i.e. requiring the formation of chemical ionic species and in the use of complexing agents to assist the extractability and solubility of the radionuclides.

Although the electrokinetics for radon removal, discussed in this work, keeps resemblance with traditional approach for contaminated soils in the sense that both use external underground electric potentials, there are similarity ends. The electrokinetic removal technique investigated in this work is based on an entirely new hypothesis, which may properly be called the *electrostatic pore hypothesis*, in which pores become preferential centers of ionization owing to the radioactive background. This idea opens the possibility to apply dry-ER, in which the formation of aqueous solutions is not needed in clear contrast with the traditional approach.

Materials and methods

The electrostatic pore theory

To begin with, let us consider a homogeneous soil with certain porosity and, for the sake of generality,

a spherical pore with an effective radius R . Let us fix the origin of coordinates at the center of the pore. The pore is air-filled with certain fraction of atoms of radon, which have been percolated from the surrounding. The radioactivity of the soil, which is responsible for the presence of radon in the soil as a by-product of the disintegrations, is also creating a background of ionizing radiation. The problem that we want to solve is to know what is the resulting distribution of electrons and ions inside the porous.

To do this, we will proceed as follows: first, gas is mainly absorbed in small pores with a diameter of <10 nm and diffused in pores with diameters around 10–100 nm (9), and then it seems allowable to use the Bragg–Gray cavity theory (10), which relates the radiation dose inside the cavity with the dose in the surrounding region. Within the framework of this theory, the gamma rays pass through the pore or cavity without colliding with any atom of gas contained in it, and then the absorbed dose in the cavity is deposited entirely by charged particles crossing it and the deposited energy is directly related with the ionization activity.

However, the Bragg–Gray cavity theory is insufficient by itself for the purpose of our study. Indeed, Bragg–Gray cavity theory is only concerned with the total energy deposited inside the cavity, which can be reckoned by simple calculation of the ion-pair production per unit of volume inside the cavity, and then it does not take into account how many ions or electrons are being destroyed inside the cavity by, say, recombination process. Although ion-electron recombination could not be important for the calculation of the dose inside the cavity (the deposited energy remains inside the cavity), it plays an important role in our study. In fact, for electrokinetics removal, we are not concerned with the ion-pair production inside the pore but with the total ionization of the pore, i.e. the total number of free ions and electrons, which can be affected by the electric potential. Therefore, it is necessary to account recombination. To start with, we consider the recombination equation for positive ions n_i and electrons n_e (11).

$$\frac{\partial n_{i,e}}{\partial t} = q - \beta n_i n_e \quad (1)$$

where n is the concentration (number of particles per unit volume), the subscripts i and e stand for positive ions and electrons, respectively, t is the time, q is the ionization rate (ion–electron pairs created per unit volume and per unit time), and β is the recombination coefficient. At the steady state, Eq. 1 becomes

$$q = \beta n_i n_e \quad (2)$$

It may be possible to derive an approximated relationship between n_i and n_e by using the diffusion theory. Let us consider Fig. 1, in which a pore with an effective radius R is sketched. In this pore, primary or secondary electrons coming from the surrounding radioactive environment are generating ion–electron pairs inside. Some electrons generated inside the pore will remain inside the pore together with the ions, and other electrons will manage to escape or migrate away from the pore. Contrariwise, because ions are much more heavier, its migration away from the pore can be considered almost zero (in comparison with the electronic migration), and if so, all the ions generated inside the pore are trapped inside the pore. As a result, there will be a buildup of positive charges inside the pore. From the diffusion transport theory, the probability that a certain particle does not escape from a certain region by elastic scattering is estimated as (12)

$$p_n = e^{-B^2 M^2} \quad (3)$$

which is known as the non-leakage probability, where B is the so-called buckling factor, which for a spherical region with radius R is given by

$$B = \frac{\pi}{R} \quad (4)$$

and M^2 is the migrational area, which can be approximated to the diffusion length path by (13)

$$M^2 \approx \lambda^2 \quad (5)$$

where λ is the diffusion length in the gas.

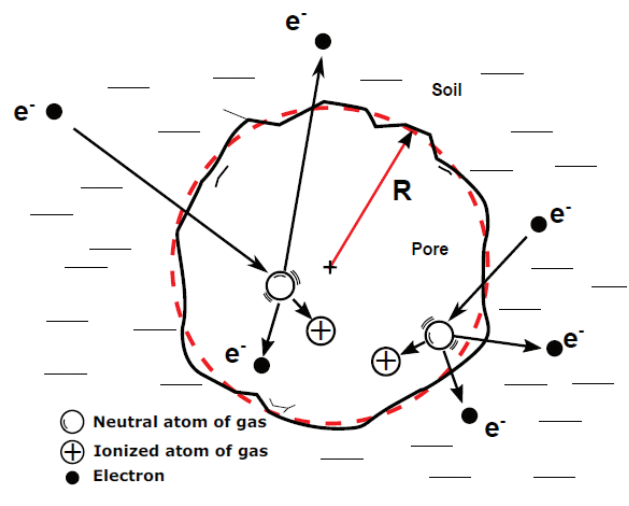


Fig. 1. Physical model for ionization and polarization of the pore. Because the length path of electrons is much higher than ions, then the electronic leakage is also higher, resulting in the build up of positive charge inside the pore.

Therefore, if after a certain number of ionizations inside the pore an amount of ion–electron pairs are created and only remains n_i ions (with a negligible leakage $\lambda_i \approx 0$), then the concentration of electrons inside the porous could be approximately estimated as equal than the number of ions n_i multiplied by the electronic non-leakage probability given by Eq. 3

$$n_e = n_i e^{-B^2 M_e} \quad (6)$$

and taking into account, Eqs.4 and 5 become

$$n_e = n_i e^{-\frac{\pi^2 \lambda_e^2}{R^2}} \quad (7)$$

by inserting Eq. 7 into Eq. 2, one obtains

$$q = \beta n_i^2 e^{-\frac{\pi^2 \lambda_e^2}{R^2}} \quad (8)$$

For small pores within the Bragg–Gray cavity theory, the ionization may be related with the soil-activity as (10)

$$q = \frac{a_s}{w\Lambda} \quad (9)$$

where a_s is the specific soil-activity energy per unit volume, W is the energy required to create an ion pair (≈ 33 eV for air), and Λ is the ratio of the stopping power of the soil and the gas.

Taking Eqs. 9 and 8 into account, we obtain

$$n_i = \psi e^{-\frac{\pi^2 \lambda_e^2}{2 R^2}} \quad (10)$$

and

$$n_e = \psi e^{-\frac{\pi^2 \lambda_e^2}{2 R^2}} \quad (11)$$

where

$$\psi = \sqrt{\frac{a_s}{\beta w \Lambda}} \quad (12)$$

It is convenient to define a degree of ionization of the pore by reckoning the total number of ions divided by the total number of particles per unit of volume inside the pore n_o

$$I = \frac{n_i}{n_o} \quad (13)$$

where the number of total particles n_o of the gas is estimated by the relation of ideal gases

$$n_o = \frac{p N_A}{R_g T} \quad (14)$$

where N_A is the Avogadro’s number, p and T are the pressure and temperature inside the pore, respectively, and R_g is the gas constant. On the other hand, the length path of electrons can be estimated from the kinetic theory (14)

$$\lambda_e \approx \frac{\kappa_B T}{p \pi r_i^2} \quad (15)$$

where K_B is the Boltzmann’s constant and r_i is the radius of the atoms of the gas.

Electrokinetics for soil radon removal

In the preceding section, an expression was derived for the degree of ionization of the gas inside a small pore, where recombination and leakage of electrons were both considered. In this section, some rough study is addressed to know how radon can be removed by the application of an electric potential in the ionized pore.

First of all, the concentration of radon inside the pore is a tiny fraction in comparison with the atoms of air as previously stated, and the cross-section of ionization of radon is very small and direct ionization of radon inside the pore can be neglected. This assumption means that the atoms of radon are neutral and then unaffected by the electric field and cannot be directly removed by Lorentz force acting on them, an assumption that is clearly a conservative one.

Therefore, radon removal will proceed indirectly by the momentum exchange from the accelerated ions of air colliding against the neutral atoms of radon, which can be regarded as target atoms at rest. A similar process is actually the driven mechanism in traditional electrokinetic soil remediation, which is called electromigration. However, as easy to see, there are substantial qualitative and quantitative differences. On the one hand, in traditional electromigration for decontamination of soils, the transport of species is mostly due to the momentum transfer between the current of electrons, which are moving toward the anode and then the species are also preferentially migrate toward the anode (electromigration), whereas, in our case, it is opposite, i.e. the atoms of radon are preferential moved by the momentum transfer from the stream of heavy ions of air moving toward the cathode and then the atoms of radon preferentially migrate toward the cathode. On the other hand, mobility in gases is by about four orders of magnitude higher than ion mobility in

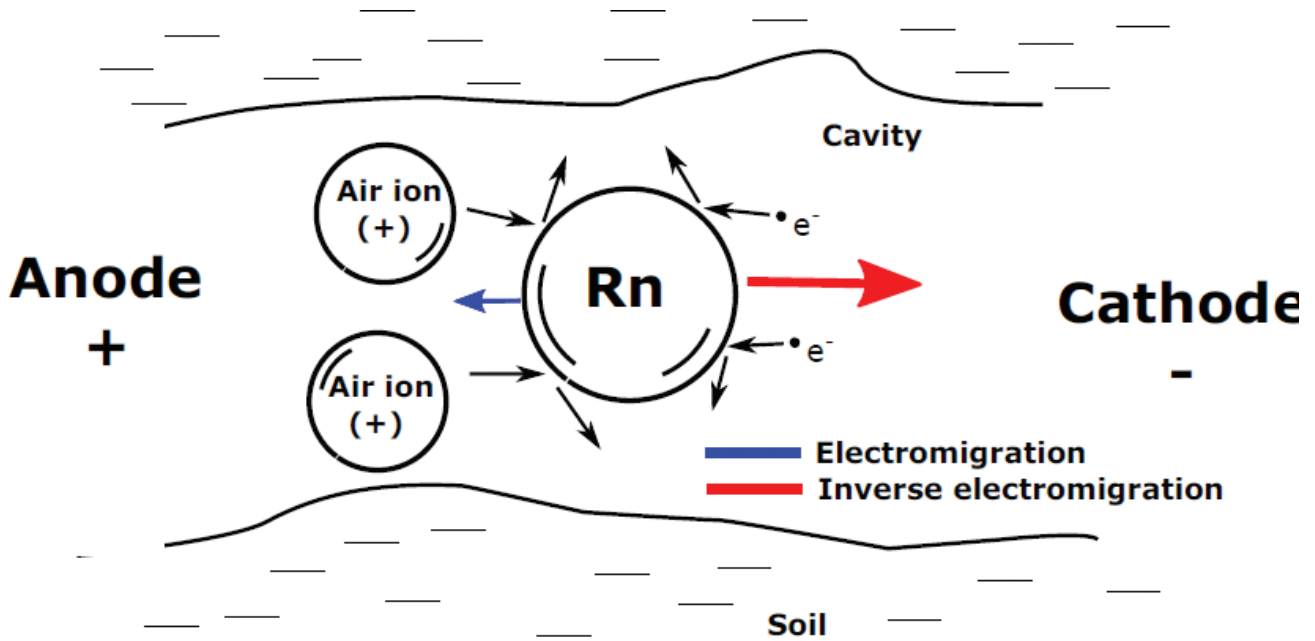


Fig. 2. Radon drift mechanism due to the momentum exchange with the current of ions and electrons under an electric field.

electrolytes (15). In Fig. 2, the drift motion acting on a radon atom is pictorially shown.

Now, when the ion (ionized atom of air) is under the action of an uniform electric potential, it will be accelerated until it reaches a constant drift velocity according to the well-known formula

$$v_i = K \frac{\Delta V}{L} \tag{16}$$

where K is the ion mobility and Δv is the electric potential and considering a system of electrodes of parallel plates each other at a distance L , as depicted in Fig. 3. The ion mobility may be calculated from the Mason–Schamp equation, (16)

$$K = \frac{3}{16} \left[\frac{2\pi}{\mu \kappa_B T} \right]^{\frac{1}{2}} \frac{Q_i}{n\sigma} \tag{17}$$

where μ is the reduced mass of the ion and the gas molecules, Q_i is the ion charge, n is the gas number density, and σ is the collision cross-section between the ion and the gas molecules. Thus, if a fraction I of the initial amount of gas n_0 is ionized, given by Eq. 13, then the ion mobility can be expressed as function of the mobility in a neutral gas K_0 as

$$K = K_0 \left[\frac{1}{1-I} \right] \tag{18}$$

Combining Eqs. 16 and 18, we have for the ion drift velocity

$$v_i = \frac{K_0 \Delta V}{1-I} \frac{1}{L} \tag{19}$$

For the momentum exchange from the collision between the ion and the atom of radon, we will proceed as follows: on the one hand, the average logarithmic energy decrement ξ_i of the ion after an elastic collision with a given target (in our case an atom of radon) is given by (13)

$$\xi_i = \ln \frac{E_i}{E_f} \tag{20}$$

where

$$\xi_i = 1 + \frac{(\alpha-1)^2}{2\alpha} \ln \frac{\alpha-1}{\alpha+1} \tag{21}$$

where E_i and E_f are the energy of the ion before and after collision, respectively and $\alpha = \frac{m_r}{m_i}$ is the ratio between the mass number of the ion m_i and the radon atom m_r . Thus, the kinetic energy gained by the atom of radon after collision is the kinetic energy lost by the ion atom, i.e. $\Delta E = E_i - E_f$, and thus the velocity of radon as function of the velocity of the incident ion atom v_i yields

$$v_r = v_i \left[\frac{1 - e^{-\xi_i}}{\alpha} \right]^{\frac{1}{2}} \tag{22}$$

and inserting Eq. 19 becomes

$$v_r = \frac{K_0 \Delta V}{(1-I)L} \left[\frac{1 - e^{-\xi_i}}{\alpha} \right]^{\frac{1}{2}} \tag{23}$$

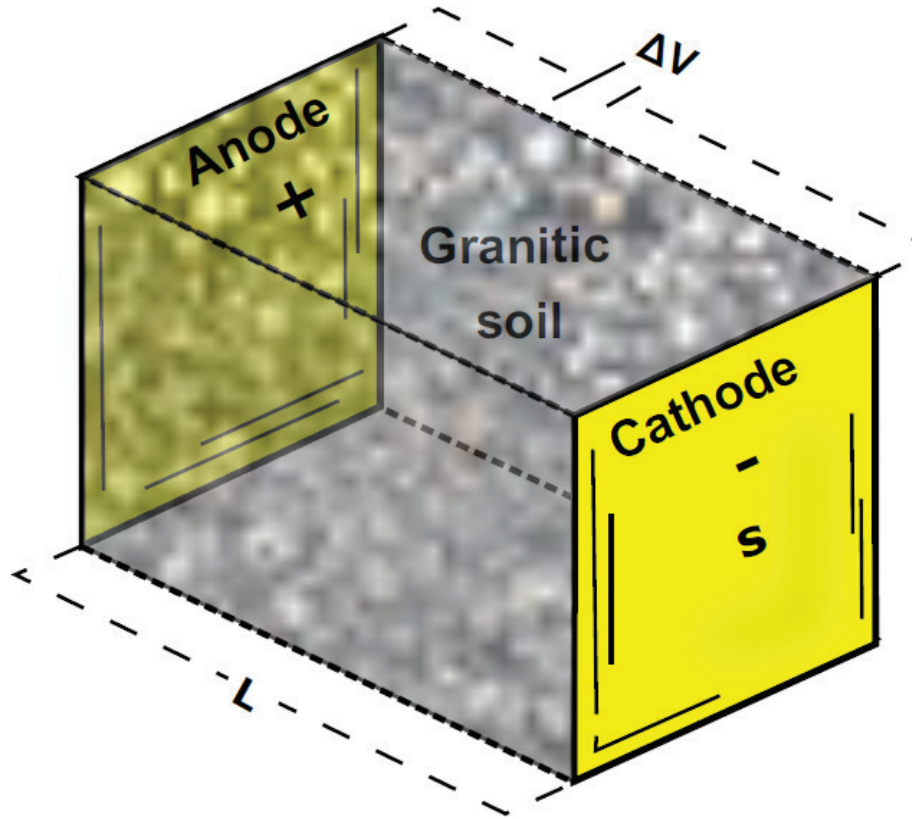


Fig. 3. Soil-parcel electrokinetic model.

The flux of ions j_i across any plane parallel to the electrode-plates is given by

$$j_i = v_i n_i \tag{24}$$

or considering Eq. 19 becomes

$$j_i = \frac{K_o \Delta V n_i}{(1-I)L} \tag{25}$$

On the other hand, the radon flux J_r is related with the ionic flux according to the following equation, derived in the Appendix:

$$j_r = \frac{2j_i}{3\alpha(1-I)} \left[\frac{v_r}{v_i} \right] \left[\frac{n_r}{n_o} \right] \tag{26}$$

Inserting Eqs. 13, 23, and 25 into Eq. 26, we have

$$j_r = \frac{2K_o \Delta V n_r I}{3L(1-I)^2} \left[\frac{1 - e^{-\xi_i}}{\alpha^3} \right]^{\frac{1}{2}} \tag{27}$$

If the specific generation of radon in the soil per unit of time and volume is a_r , then the amount of radon generated in the parcel of soil limited by the electrodes with a cross-section area s , distance between them L , and volume

sL is sLa_r . Therefore, by a balance of mass in steady state, the radon flux must satisfy

$$\begin{aligned} sj_r &= sLa_r; \\ j_r &= La_r, \end{aligned} \tag{28}$$

In addition, the concentration of radon in the soil must be equal or lower than the equilibrium concentration attained by its radioactive decay; therefore, an upper limit is obtained by

$$a_r \approx n_r \frac{\ln(2)}{t_{1/2}} \tag{29}$$

where n_r is the concentration of radon and $t_{1/2} \approx 3.3 \times 10^5$ s is the half-life of radioactive decay for radon (^{222}Rn). Inserting Eq. 27 into Eqs. 28 and 29, we have

$$\Delta V = \frac{3\ln(2)L^2}{2K_o t_{1/2}} \left[\frac{\alpha^3}{1 - e^{-\xi_i}} \right]^{\frac{1}{2}} \frac{(1-I)^2}{I} \tag{30}$$

Results

In order to obtain some idea of the degree of ionization and the require electric potential, we assume some typical values of the parameters: a γ -soil activity for a soil with ^{40}K , ^{226}Ra , and ^{232}Th with specific activities (17) 500, 35,

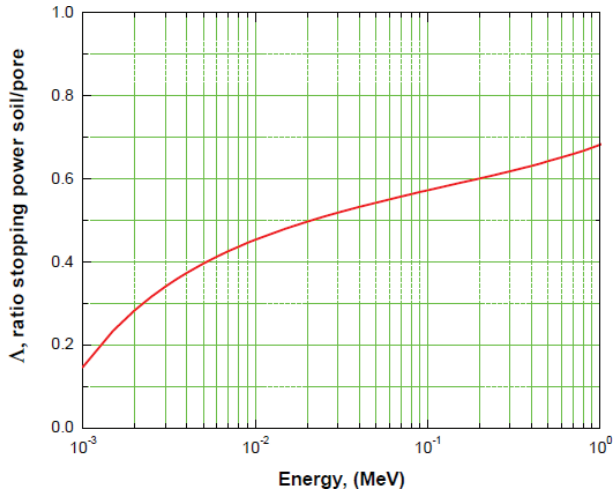


Fig. 4. The soil-pore stopping power ratio, Λ .

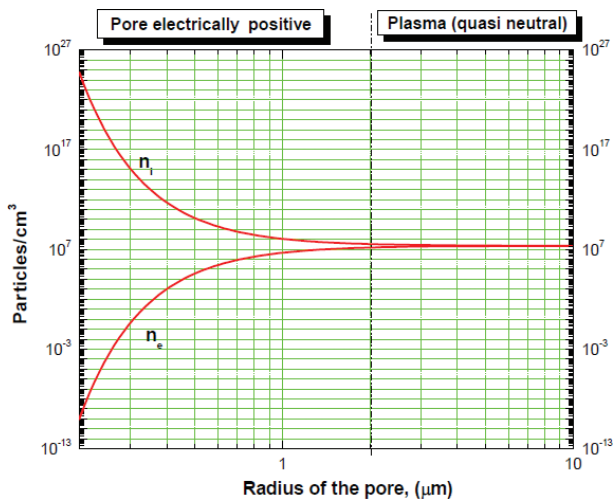


Fig. 5. The charge distribution inside the pore.

and 50 Bq/kg, respectively, which for a soil with an average density of 1.5 g/cm³ yields an activity around 2.3 MeV/cm³ s; an average energy lost per pair of ions formed in the gas (10), $w = 33$ eV; a recombination coefficient (18), $\beta = 10^{-9}$ cm³/s; an ionic mobility $K_o = 1.29 \times 10^{-2}$ m²/Vs; $\alpha = 7.68$. For the calculation of the ratio stopping power soil-pore, Monte Carlo computational simulations were performed using the Monte Carlo N-Particle Transport code, which is a general purpose Monte Carlo radiation transport code that tracks nearly all particles at nearly all energies developed at Los Alamos National Laboratory. The calculations were performed for several cavities air-filled with diameters from 0.1 to 10 μ m. The cavity was immersed into an infinite medium with a material composition from a typical granitic soil in weight fraction as follows: SiO₂: 45.26; TiO₂: 0.04; Al₂O₃: 39.9; Fe₂O₃: 0.41; MgO: 0.06; CaO: 0.10; Na₂O: 0.02; K₂O: 0.45; H₂O: 13.76. The resulting curve for the stopping power ratio is

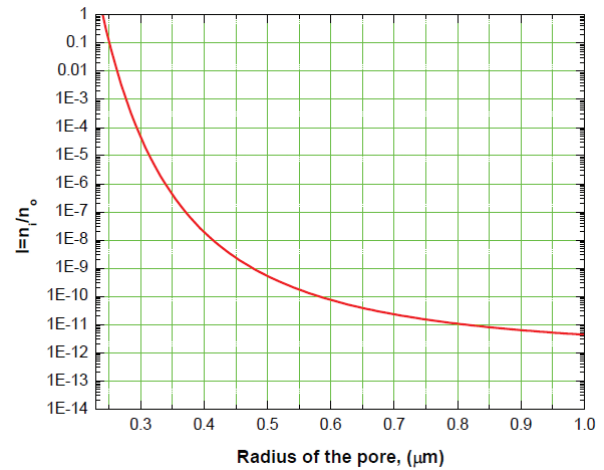


Fig. 6. Ionization degree I as a function of the radius of the pore.

shown in Fig. 4, where, for a first estimate, an average representative value around $\Lambda = 0.5$ may be assumed. Finally, assuming a temperature inside the pore around $T = 320$ K with a pressure $p = 10^5$ Pa and an ionic radius for air molecules $r_i = 2 \times 10^{-10}$ m, we obtain Fig. 5 for the ionic and electronic concentration as function of the radius of the pore. From this figure, it is easy to see that the pore tends to a plasmatic condition (quasi neutral gas) for radius higher than 2 μ m or thereabouts, and with a concentration around 10⁶/cm³. For radius less than 2 μ m, the pore starts to become positively charged owing to the increased leakage of electrons. Fig. 6 shows the ionization degree as function of the radius of the pore. It is seen that full ionization is attained for porosities with diameters around or less than 0.2 μ m or thereabouts. Finally, Fig. 7 shows the electric potential required for radon removal as a function of the diameter of the pore. It is seen that electrokinetics working with practical and realizable potentials with less than 100 V, as used in traditional ER, is able to remove radon from soils with porosities with pore-diameters ≤ 600 nm. This distribution of sizes of the pore is encouraging if one takes into account that, actually, the gas diffusion region in rocks and specially in granite stones (19), where gases are mainly absorbed in micropores (<10 nm) and diffused in small pores (10–100 nm) (9).

Conclusions

Electrokinetic and its potential significance with regard to soil radon mitigation was investigated. Utilizing a physical model, it was shown that because of the surrounding radioactivity, soil-pores, through which radon travels toward the surface, are preferential centers of ionization, and in fact, small pores become positively charged owing to the much higher migrational areas of electrons in comparison with heavy ions, which are

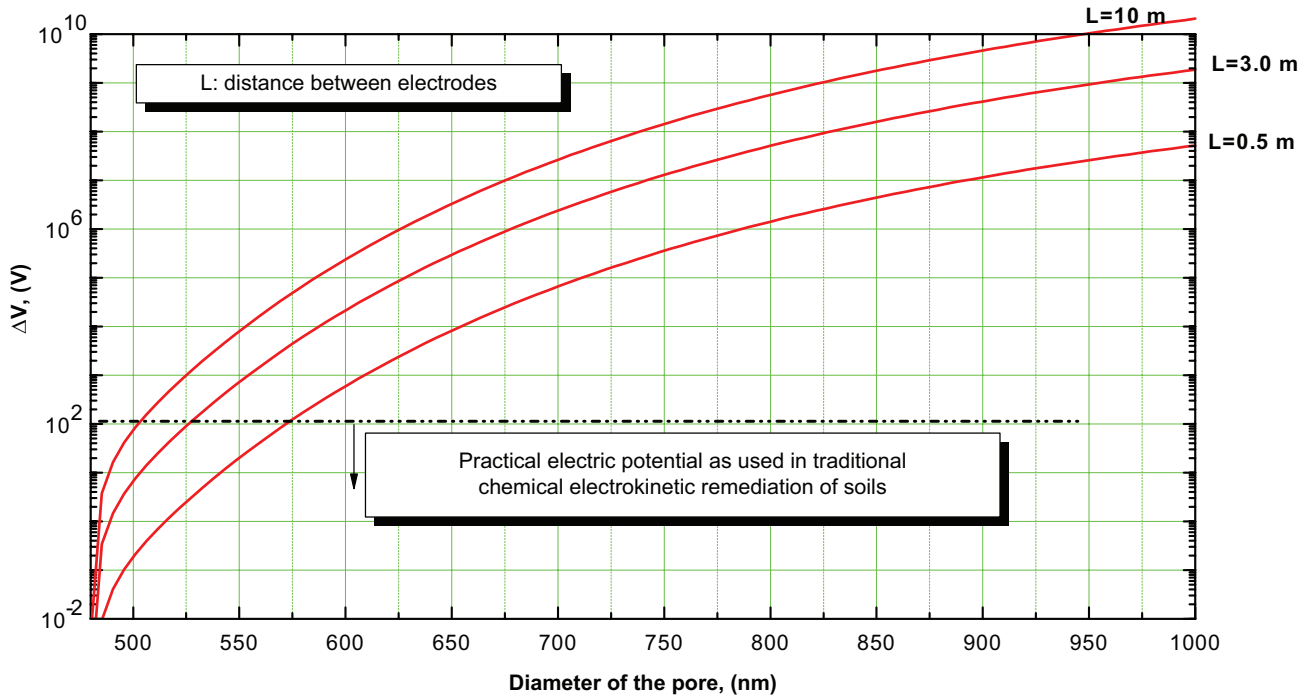


Fig. 7. The electric potential required for radon removal as a function of the diameter of the pore and for several distance between electrode-plates.

trapped inside the pore whereas electrons escape. The electrical activity inside the pores opens the possibility for radon removal by applying electric potentials. It was found that electric potentials with less than 100 V, as used in traditional ER, are able to remove radon from soils with porosities with pore-diameters 600 nm or less. This distribution of sizes of the pore is very encouraging because it constitutes, actually, the so-called gas diffusion region in rocks and specially in granite stones, which feature specially high levels or radon, where it is known that gas is mainly absorbed in micropores (<10 nm) and diffused in small pores (10–100 nm).

Finally, it should be kept in mind that the present report must be taken with caution. Substantial uncertainties were present at every step of the analysis as well as idealizations and simplifying assumptions that are inherent in theoretical treatment. Therefore, the reported results are not intended to typify quantities. This should not be misconstrued as an attempt to produce a definitive mechanistic analysis. Nonetheless, we will provide important guidance for future experimental research, which is required in order to proof the electrostatic pore hypothesis.

Appendix

Derivation of radon flux

Let us consider a single, small volume with cross-section area s , length l and volume sl with a concentration of radon and neutral atoms of air $n_r, n_o(1 - I)$. Those atoms can be

regarded as being at rest if compared with the ions, which are being accelerated by the electrical field. The volume is being bombarded by the directional flux of ions j_i , induced by an external electric field, and then some of the atoms of radon are being energized by the momentum exchange from the collision with the ions. It is desired to know the induced flux of radon j_r as a result of the ionic flux j_i .

First, the number of atoms of radon per unit of volume and time \dot{n}_r^* , which are being energized by the collision with the flux of ions, is given by

$$\dot{n}_r^* = \sigma_r n_r j_i \tag{31}$$

where σ_r is the microscopic cross-section of collision between ions and radon targets, n_r is the number of atoms of radon at rest, and J_i is the flux per unit of area of ions. Now, if the length of the volume is equal to the length path of the energized atoms of radon $l = \lambda_r$, then the time of reaction is given by $\tau = \frac{\lambda_r}{v_i}$ where v_i is the velocity of the ions discussed previously. Therefore, the number of radon atoms energized per unit of volume will be

$$n_r^* = \frac{\sigma_r n_r \lambda_r j_i}{v_i} \tag{32}$$

The length path λ_r can be expressed in terms of the concentration of the target atoms, i.e. the atoms not ionized of air n , and the microscopic cross-section as (13), $\lambda_r = \frac{1}{n\sigma_r}$

being the microscopic cross-section of collision between the energized radon and the neutral atoms. Therefore, Eq. 32 becomes

$$n_r^* = \frac{\sigma_i n_r j_i}{\sigma_r n v_i} \quad (33)$$

On the one hand, it is allowable to assume that the microscopic cross-section $\sigma_i \approx \sigma_r$, and on the other hand, after collision, the radon atom is scattered with an average cosine of the scattering angle with regard to the initial direction of the ion (13)

$$\bar{\mu}_o = \frac{2}{3\alpha} \quad (34)$$

and then the average component of the velocity of radon in the direction transverse to the plates is

$$\begin{aligned} \bar{v}_r &= v_r \bar{\mu}_o; \\ \bar{v}_r &= \frac{2v_r}{3\alpha} \end{aligned} \quad (35)$$

and the regulating drift flux $j_r = n_r^* \bar{v}_r$, and thus, we have

$$j_r = \frac{2j_i}{3\alpha} \left[\frac{v_r}{v_i} \right] \left[\frac{n_r}{n} \right] \quad (36)$$

which is physically consistent with what would be expected, namely, if the radon velocity is null, the flux will also be null, or even if the radon velocity is high but the concentration of radon tends to 0, the flux will also tend to 0. Taking into account that the number of atoms of air no ionized as function of the initial number of atoms is given by $no(1 - I)$, then Eq. 36 becomes

$$j_r = \frac{2j_i}{3\alpha(1-I)} \left[\frac{v_r}{v_i} \right] \left[\frac{n_r}{n_o} \right] \quad (37)$$

Nomenclature

a_s = specific activity of the soil in energy per unit of volume and time (J/m³s)
 $a_{r_}$ specific generation of radon in the soil in particles per unit of volume and time (1/m³s)
 B = buckling factor (1/m)
 e = electronic
 E = energy of the accelerated ion (J)
 i = ionic
 I = dimensionless ionization factor defined by Eq. 13
 J = flux per unit of area (1/m²s)
 K = ionic mobility
 L = distance between electrodes
 m = atomic mass (kg)
 M = migrational area (m²)

n = concentration per unit of volume (1/m³)
 N_A = Avogadro's number
 p = pressure (Pa)
 p_n = dimensionless nonleakage probability
 q = ionization rate per unit volume and time (1/m³s)
 Q = charge per unit of volume (C/m³)
 r_i = radius of the ion (m)
 R = radius of the pore (m)
 R_g = gas constant (J/Kmol)
 t = time (s)
 $t_{1/2}$ = half-life of radon ²²²Rn (s)
 T = temperature (K)
 w = energy required to create ion-pair w (J)

Greek symbols

α = dimensionless ion-radon mass ratio
 β = recombination coefficient (m³/s)
 Δv = electric potential (V)
 k_B = Boltzmann's constant (J/K)
 λ = length path (m)
 Λ = dimensionless ratio stopping power soil-pore
 ξ = dimensionless average logarithmic energy decrement
 ρ = density (kg/m³)
 σ = microscopic cross-section (m²)
 v = velocity (m/s)

Subscripts

a = air
 e = electron
 f = final
 i = ionization, ion or initial value
 p = pore
 r = radon
 s = soil

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Author contribution

The author confirms sole responsibility for the study conception, analysis and interpretation of results, and manuscript preparation.

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