Diffusion of Radon in Porous Media Saturated with Gels and Emulsions

I. LAKATOS¹, K. BAUER¹, J. LAKATOS-SZABÓ¹, I. CSIGE², J. HAKL² and H.-J. KRETZSCHMAR³

 ¹Research Laboratory for Mining Chemistry, Hung. Acad. Sci. 3515 Miskolc-Egyetemváros, PO Box 2, Hungary. e-mail: lakatos@mta.bkkl.hu
 ²Institute of Nuclear Research, Debrecen, Hungary
 ³DBI Gas and Umwelttechnik GmbH, Freiberg, Germany

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Abstract. The effective diffusion coefficient of radon was determined in polymer/silicate gels and clay suspension used as sealing materials in environmental protection. On the basis of the experimental findings, it was concluded that both materials drastically decrease the convective mass transport in porous media. Simultaneously, the effective diffusion coefficient was reduced significantly. Thus, the radon flux might be decreased by 5 to 6 orders of magnitude in porous systems originally having gas or low water saturation by injection of gel-forming materials or placement of clay suspensions. At high water saturation, however, the diffusion transport of radon can be slightly restricted in consolidated and unconsolidated porous media. The laboratory studies may firmly allow us to conclude that hydrogels and clay suspensions are prospective candidates in an integrated environmental technology to be used for restriction of radon migration in subsurface regions.

Key words: diffusion, radon, gel, emulsion, environmental protection.

Symbols

| c_m | activity concentration of radon in the sample, $Bq m^{-3}$ |
|------------------|--|
| c(x,t) | activity concentration of radon in the pore volume, Bq m ⁻³ |
| j_c | diffusion flux of radon at the surface of model, Bq $m^2 s^{-1}$ |
| t | time, s |
| x | distance from the surface of sample, m |
| A | cross-section of sample, m ² |
| $D_{ m eff}$ | effective diffusion coefficient in the porous media, $m^2 s^{-1}$ |
| D^* | effective diffusion coefficient in gels and in emulsions, m ² s ⁻¹ |
| \mathbf{E}_i | experimental radon exposure, Bq m ⁻³ h |
| E^*_i | theoretical radon exposure, Bq m ⁻³ h |
| H | total thickness of sample, m |
| V | volume of sample, m ³ |
| Φ | porosity |
| η | partition of radon between the sample and the gas phase |
| λ | decay constant of radon, 2.098 10^{-6} s ⁻¹ |
| au | tortuosity |
| | |

1. Introduction

The geochemical radon sources and man-made nuclear waste depositories emanating high intensity radon flux represent continuous and serious hazard for humans. The radon gas $(^{222}$ Rn) originating from 238 U isotope is omnipresent in the natural environment. Its high mobility can be attributed to gas and liquid phase transport of radon itself and the leaching process of precursor nuclides. As a result, the average indoor radon concentration changes between 20 and 180 Bq m⁻³ in different European countries, but in individual homes values higher than 10000 Bg m^{-3} have also been measured, exceeding 50–100 times the recommended action level of 200 Bg m⁻³ set by the UN WHO [1]. Despite predominant (40–90%) contribution of radon to the total radiation exposure in European countries, its role in fatal records is negligible compared to other factors (e.g. smoking). Nevertheless, roughly 2500/year fatal cases are attributed to external and internal radon effect only in the U.K. Consequently, an efficient restriction, or total blocking of radon migration in subsurface regions must form a central issue in environmental protection. Clays, silts, sands, compost and varying mixtures of them are usually proposed as covering or barrier forming materials in porous media to decrease radon flux from disposal sites [2, 3]. According to industrial pilots, different polymer (e.g. polyethylene, polysulfide copolymer, etc.) mats also efficiently block the diffusion of radon transport in the vicinity of uranium deposits [4, 5]. Recently, self-conforming gels are proposed to decrease the flow of radon gas in porous and fractured media. This later technical solution may raise hopes because they can be easily adapted to a great variety of local circumstances and tailored to any integrated environmental technologies without extensive destruction of surface facilities.

The basic idea of subsurface blocking is that by placing a gel or clay barrier around a heavily contaminated area, the convective mass transport of hazardous materials can be reduced to extremely low levels. Since the convective mass transport and the permeability in porous media can be decreased by 5 to 6 orders of magnitude [6], optimistic expectations were revealed about the application of gel and clay barriers. Sources of misunderstanding should be established in the fact that extensive laboratory and field studies on the determination of radon transport in clays, cements, concrete, polymers and other covering materials have been carried out though, the real contributions of the convective and diffusion factors were often not eliminated [4, 5, 7–11]. These approaches completely ignore the fact that even though the contaminants might be effectively localised by these methods, the diffusive mass transport driven by the difference in concentrations between the isolated area and the surroundings may remain mostly unchanged. Thus, as was definitely shown in the past years, the diffusive mass transport is not only commensurable with the convective one, but sometimes it becomes the major migration factor of species. Therefore, a joint research program was initiated by the co-operating Hungarian and German institutes with the aim of determining the diffusion transport of radon through polymer/silicate gels and concentrated clay suspensions. The measuring and calculation techniques, the materials and models and finally, the effective diffusion coefficients of radon obtained in different media are compared and discussed in the paper.

2. Experimental Conditions

The gel and the clay barrier significantly differ from each other regarding their chemical nature and placement technology. Reasonably, these facts must be considered when constructing the experimental arrangements and the samples [12] to be analysed. Further, it was also necessary to design a mounting device so that the hydrodynamic (permeability) measurements had also to be carried out after each exposure with the aim of determining the possible degradation of sealing materials in the presence of radon. As such deteriorating effects could not be detected, this problem is not detailed here.

2.1. POLYMER/SILICATE GEL

It is well known that chain-like polymers can be cross-linked or gelled by different chemicals having side groups with an opposite electrical charge. In the present case, a cationic polymer (trade name Polyquat 20) containing quaternary ammonium groups is cross-linked by anionic ortho silicates. A sealing phase can be formed from these compounds by the sequential injection of solutions containing the appropriate chemicals into the porous or fractured media or by mixing them at the surface before placement. The rate of gelation is controlled by the reaction rate, hydrodynamic dispersion and diffusion of reacting species. Accordingly, the former technique is applied if deep penetration is needed, while a bulk mixture is used when only shallow penetration is required. Both methods have special advantages and they are not alternatives but rather complementary procedures. Taking these practical aspects into account, the laboratory studies were extended to the following chemical system:

Solution A : 100 g water 100 g Polyquat 20 Solution B : 100 g water 100 g water glass (with 17% SiO₂ content).

Under natural conditions, the blocking effect is accomplished by filling up the pore space of the target subsurface region with a self-conforming mixture of solutions A and B. Both fluids and their mixture at the beginning have a dynamic viscosity of less than 10 mPa s. Namely, the solid structure acts as a carrier medium, while the gel-forming liquid is the saturating phase. After placement of the treating fluids by sequential or bulk phase injection, a gel forms in-situ, which usually has a dynamic viscosity higher than 10 000 mPa s. The samples to be exposed to



Figure 1. Schematic of the core holder applied for the study of polymer/silicate gels (1, 4 -etched track radon monitors; 2 – O-ring; 3 – aluminium cylinder; 5 – aluminium block; 6 – metal sealing ring; 7 – connection fitting; 8,9 – water saturated paper sheet; 10 – epoxy resin; 11 – porous core saturated with gel)

radon must be prepared accordingly. For the serial analysis, artificially consolidated porous media (Meissen filter rock) were used with the following dimensions and properties:

| Diameter : | 30 mm, |
|----------------|---------------------|
| Length : | 20 mm, |
| Porosity : | $\sim 0.5,$ |
| Permeability : | $> 10 \mu { m m}^2$ |

At ambient temperature, a 1:1 mixture of solutions A and B mentioned above has a setting time of about 2 min, and that time was too short to place the partially gelled liquid into a highly permeable porous system. Since the high blocking efficiency makes the control of the setting time impossible, injection of the premixed liquid into the porous media was carried out at 271 K. The setting time at this temperature was sufficiently long, roughly 6 min, for complete saturation of even low permeable porous media.

The porous core was mounted by an impermeable epoxy resin enriched with silica sand. The thickness of resin wall around the porous core was 5 mm. Then the cores were saturated with the gel-forming bulk liquid immediately after mixing the solution A and B and then they were placed into a core holder, as shown in Figure 1. The core holder was designed to fulfil the requirements of both the diffusion and the hydrodynamic measurements.

The experimental arrangement of the diffusion test applied for gels is illustrated in Figure 2. The radon source was placed into an air-tight stainless steel cylinder on a porous plug. At the bottom of the cell 100 ml of water was always present to maintain equilibrium vapour saturation in the system to prevent water loss from the gel. A manifold attached to the radon source served to connect the



Figure 2. Experimental arrangement of the diffusion test applied for gels (1 -stainless steel container; 2 -radon source; 3 -porous plate; 4 -water; 5 -cores (Nos. 2–5) filled up with gel; 6 -reference cores (Nos. 7–11) free of gel; 7 -manifold and christmas tree; 8 -semiconductor based continuous radon monitor).

diffusion cells. The cell Nos. 2–6 were filled up with gel-containing cores, while cell Nos. 7–11 contained only dry porous cores used for reference. The effective diffusion coefficients of radon in gels were determined using a constant inlet radon concentration and different exposure times. To obtain a relatively high and constant initial radon concentration in the whole system, the cylinder was closed for a few weeks prior to measurements. During the exposure period, one pair of cells (a dry and a gel-containing one) were disconnected from the christmas tree after 1–2–3–4–5 days. The inlet and outlet radon exposures in the cells were determined by etched track radon monitors using computerised image analysis for track density measurements. Occasionally, semiconductor-based radon monitors were connected to the inlet and outlet points to directly record the radon concentration.

2.2. MONTAX/CLAY SUSPENSION

The fundamental difference between the gel and clay barriers is that no solid carrier is required in the latter case. The sealing material is usually prepared at the surface from liquid and solid components and then poured into a ditch sited around the contaminated area. The present studies were extended to the following system

Component A : Montanwax emulsion R12 Component B : sand gravel bentonite.

Particle size distribution of the solid component is characterised by the data listed in Table I. Since the pore volume of the clay depends significantly on particle size distribution, the diffusion studies were carried out by sand-enriched clays having porosities of 40 and 50%, respectively. Therefore, the mixing ratio was different in test series 1 and 2 and the blocking phases were made of the following quantities:

| Particle size, μm | Series 1, % | Series 2, % |
|----------------------|----------------|----------------|
| < 160 | 23.9 | 27.4 |
| 160-315 | 26.8 | 20.5 |
| 315-800 | 49.3 | 37.7 |
| > 800 | - | 14.4 |

Table I. Particle size distribution of the solid phase.

Series 1 : 120 g component A 150 g component B Series 2 : 95 g component A 200 g component B

Since the Montanwax/clay dispersion is highly plastic and its solid phase is unconsolidated, the previous mounting device developed for gel studies could not be applied. Therefore, both the sample size and the exposure technique were modified. Actually, samples with different thickness were exposed to radon for a constant exposure time. Therefore, the experimental arrangement and the sample preparation were relatively simple. A series of glass holders having a diameter of 40 mm, and a height of 80 mm, were filled up with the appropriate amount of sealing material. Prior to this etched track radon monitors were placed at the bottom of the glass vials. During each test series the thickness of the sealing material varied between 0.5 and 4.5 cm by steps of 0.5 cm.

The schematics of the experimental arrangement is shown in Figure 3. The same radon source was used as earlier, but the dead volume of the exposure device was much greater and, hence, the equilibrium radon concentration was lower. A closed desiccator containing the samples in glass vials was connected to the radon source via a flexible tubing. Reliable results were obtained by 5–7 days of exposure. Since the clay suspensions were prone to shrinking, special efforts were made to minimise the water loss from the samples. For example, all manipulation on samples and their exposure were carried out in a vapour-saturated environment. For the determination of the inlet and outlet radon exposures, the etched track radon monitors were similar to those used as in case of gels. The radon monitors were protected from the sample and the water by rubber sleeves.

3. Results and Discussion

3.1. CALCULATION OF THE EFFECTIVE DIFFUSION COEFFICIENT

The spatial and time-dependent distribution of radon concentration in porous media and in the detection volume can be described by the differential equations of



Figure 3. The experimental arrangement of diffusion test applied for Montax/clay suspensions; (1 - stainless steel container; 2 - radon source; 3 - porous plate; 4 - water; 5 - exposition chamber; 6 - porous diaphragm; 7 - glass vials; 8 - etched track radon monitors; 9 - clay suspension).

diffusion (Fick I and II) corrected with the spontaneous decay of radon along the transport path:

$$\frac{\partial c(x,t)}{\partial t} = D_{\text{eff}} \frac{\partial^2 c(x,t)}{\partial x^2} - \lambda c(x,t).$$
(1)

The interstitial diffusion flux at the outlet surface of the samples is

$$j_c = -D_{\text{eff}} \frac{\partial c(x,t)}{\partial x} |_{x=H}.$$
(2)

Thus, the change of the radon activity concentration in the detection volume is given by the following differential equation:

$$\frac{\mathrm{d}c_m(t)}{\mathrm{d}t} = \frac{j_c \Phi A}{V} - \lambda c_m(t). \tag{3}$$

The initial conditions are c(x, 0) = 0 and $c_m(0) = 0$, and the boundary conditions are $c(0, t) = \eta c_{in}$ and $c(H, t) = \eta c_m(t)$, respectively. The tortuosity of the carrier porous media, determined by electric measurements, was less than 1.1, therefore it was neglected. Since all parameters, except the effective diffusion coefficient, are known in Equations (1)–(3), the diffusion mass transport can be modelled on the basis of the change of radon concentration as a function of time or thickness of sample. The most probable diffusion coefficient is then obtained by fitting the arbitrary chosen 'theoretical' values to the experimentally measured data.

About 20 MBqm⁻³ and 0.6 MBqm⁻³ inlet radon activity concentration were used during the gel and clay suspension studies. Earlier it was found that, in some

cases, emanation of radon from radium may contribute significantly to the radon concentration present in the detection space. In our case, however, the daughter nuclides of radon had no measurable effect on the accuracy of determination.

As a first step of the modelling procedure, the radon exposures were determined using different arbitrarily chosen 'theoretical' effective diffusion coefficients:

$$E_i^*(D_{\rm eff}) = \int_0^{t_i} c_m(t) \,\mathrm{d}t.$$
(4)

The upper limit of integration was different during the analyses of the gelcontaining samples, while it was constant when the Montanwax/clay suspensions were tested. Having determined the radon exposure, the total relative squared error was then calculated:

$$S = \sum_{i=1}^{n} \left(\frac{E_i - E_i^*(D_{\text{eff}})}{E_i} \right)^2.$$
 (5)

The adjusted 'theoretical' diffusion constant belonging to the minimum of this error function was accepted as the most real value of the effective diffusion coefficient in gel-containing samples and clay suspensions.

3.2. DETERMINATION OF THE EFFECTIVE DIFFUSION COEFFICIENT IN POROUS MEDIA SATURATED WITH GELS

In the initial experimental program three series of measurements were carried out using nearly identical experimental conditions. The porosity of the carrier porous media in the first two series was about 50%, while in the third series it was only 40%. The pore space of all cores were saturated with the same gel. For series 1, the measured outlet radon exposures of samples as a function of exposure time is shown jointly with the results of the mathematical prediction in Figure 4. The modelling may allow us to conclude that the probable effective diffusion coefficient of radon in that case is 3.5×10^{-6} cm² s⁻¹. As is evident in Table II, the average value of the effective diffusion coefficients, obtained at constant radon concentration exposure for the same gel and porous cores is 3.3×10^{-6} cm² s⁻¹. Since the correlation coefficient in the present cases is over 0.9 and the relative standard deviation is negligible, we may firmly state that on the one hand the method is reliable, while on the other, the most realistic effective diffusion coefficient of radon is between the values mentioned.

The effective diffusion coefficient found for series 3 is smaller than those for series 1 and 2. That fact might be explained partly by the lower porosity of the carrier phase and partly by the greater relative standard deviation of the measurement. The later circumstance is attributed to the higher equilibrium activity concentration of radon in the exposure device and, hence, to the shorter exposure times applied.

| Series | Radon activity conc., MBq/m ³ | Correlation coefficient | Effective diffusion coefficient, cm ² /s | S |
|--------|--|-------------------------|---|------|
| 1 | 11.6 | 0.987 | 3.5×10^{-6} | 0.73 |
| 2 | 10.2 | 0.910 | 3.3×10^{-6} | 0.70 |
| 3 | 27.0 | 0.880 | 3.0×10^{-6} | 2.50 |

Table II. Experimental results of the diffusion tests in gels.



Figure 4. Measured and the calculated radon exposures as a function of exposure time using polymer/silicate gels as saturating media in a consolidated porous system (Series 1).

The diffusion coefficient of radon in a pure gel phase can be calculated using the analogy of diffusion in porous media [13, 14]:

$$D^* = \frac{D_{\text{eff}}\tau^2}{\Phi}.$$
(6)

Supposing that the tortuosity in highly permeable porous cores is close to unity, the following effective diffusion coefficients of radon are predicted in gel:

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Series 1 : 7.0 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}
Series 2 : 6.6 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}
Series 3 : 7.5 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}
Average : 7.0 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}.
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In a recently published paper [15], it was pointed out that the diffusion mass transport of chromium ions in polymer/silicate gels can be described correctly using the considerations applied earlier for consolidated porous media [14]. According to this approach, the gels consist of a flexible and dual network of cross-linked polymer and polymerised silicate, and their pore space is filled up with hydrating and bulk phase water. Consequently, the 'porosity' of gel can be determined if the solid content of the gel is known. Following this concept and taking the 25% solid content of gel into account, $D^* = 8.0 \ 10^{-6} \ cm^2 \ s^{-1}$ is found which is very close to $D = 10^{-5} \ cm^2 \ s^{-1}$ characteristic for radon in pure water. The difference between the values is reasonable because the 'tortuosity' of gel is unknown and neglected. Despite that uncertainty, we may conclude that the diffusion coefficients of radon in water and in polymer/silicate gels are similar, and the slight difference can be explained satisfactorily by the porosity and the tortuosity of gel. In this respect, the experimental results are converging with those reported in literature for diffusion of cations in aqueous solutions and hydrated gels [16, 17].

3.3. DETERMINATION OF THE EFFECTIVE DIFFUSION COEFFICIENT IN NONCONSOLIDATED CLAY SYSTEMS SATURATED WITH EMULSION

As was mentioned, the diffusion coefficient of radon in clay suspensions was determined using different sample thicknesses but a constant inlet activity concentration and exposure time. The measured and calculated data are listed in Table III. The effective diffusion coefficients were obtained in a similar way to those made earlier by fitting the theoretical curve to the experimental points. The precision of the model here also proves that the accuracy of the elaborated method is satisfactory (Figure 5).

| Series | Exposure time, h | Inlet radon expo- sure, MBq/m ³ h | Porosity | Effective diffusion coefficient, cm ² /s |
|--------|---------------------|---|----------|---|
| 1 | 168 | 113 | 0.5 | 8×10^{-6} |
| 2 | 114 | 147 | 0.4 | 4×10^{-6} |

Table III. Experimental results of the diffusion test in Montax/clay suspension

According to the experimental data, the effective diffusion coefficient of radon in clay suspensions with a porosity of 40–50% is in the range of $4-8 \times 10^{-6}$ cm² s⁻¹. Taking these values and the corresponding porosities as a basis, the following diffusion coefficients were obtained for the filling liquid (emulsion):

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Series 1 : 1.6 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}
Series 2 : 1.0 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}
Average : 1.3 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}.
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Figure 5. Measured and the calculated radon exposures as a function of exposure time using an emulsion as a saturating media in a nonconsolidated porous clay system (Series 1).

Although the latter value is equal to the diffusion coefficient of radon in pure water, the average exceeds it by 30%. If $D^* = 1.3 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$ is accepted as the most realistic diffusion coefficient of radon in emulsion, then we have to suppose that the characteristic diffusion coefficient of radon in an organic phase is greater than in aqueous media. The high solubility of radon in polymers and plastics makes this statement reasonable [3, 4].

3.4. EVALUATION OF THE EXPERIMENTAL RESULTS

The absolute and effective diffusion coefficients of radon in different media are listed in Table IV. Comparing the data the following conclusions may be drawn:

- (1) The diffusion mass transport of radon in porous and fractured systems might be reduced by 5 to 6 orders of magnitude using self-conforming hydrogels and clay suspensions if the pore space is originally filled up by gaseous substances (e.g. air [18]).
- (2) The diffusion coefficient of radon is 30% less in polymer/silicate gels than in pure water [19]. The difference can be satisfactorily explained by the solid content of the gel and taking the porosity and tortuosity of gel into account.
- (3) In porous media saturated with polymer/silicate gels, the effective diffusion coefficient of radon is 60–70% less than in the aqueous phase. Considering that fact, it might be said that the diffusion radon transport is slightly (max. by

| Media | Diffusion coefficient $cm^2 s^{-1}$ |
|--|-------------------------------------|
| Air [18] | $\approx 1.0 \times 10^{-1}$ |
| Water [19] | $\approx 1.0 \times 10^{-5}$ |
| Polymer/silicate gel | $\approx 7.0 \times 10^{-6}$ |
| Porous media saturated by polymer/silicate gel | $\approx 3.3 \times 10^{-6}$ |
| Montanwax emulsion | $\approx 1.3 \times 10^{-5}$ |
| Clay suspension | $\approx 6.0 \times 10^{-6}$ |
| | |

Table IV. Diffusion coefficient of radon in different media

30%) reduced by gels in those systems where the original saturating media is water.

- (4) In emulsions used for the preparation of clay suspensions, the calculated effective diffusion coefficient is 30% greater than the value characteristic in water. The phenomena can probably be traced back to the higher diffusion rate of radon in organic phases.
- (5) The diffusion transport of radon in concentrated clay suspensions is similar to porous media filled up with gels. The effective diffusion coefficient of radon in such media is dependent on solid content, particle size distribution, porosity, tortuosity and the composition of the liquid phase used to prepare the suspensions. Converging results have been reported by many authors [7, 8, 10]. The experimental results clearly show that the hydrophobized sealing material is practically impermeable against water. However, the presence of the organic phase in suspension may increase slightly the diffusion mass transport of radon.

Finally, the study of radon diffusion revealed that the gas phase migration of radon in natural porous media can be drastically reduced by the injection of gelforming materials and the placement of clay suspensions in the target area. Hence, their application in different environmental technologies aiming at the restriction of radon migration from geochemical and man-made sources, might be very effective. In natural water saturated surface and subsurface systems, however, the positive effect derives mostly from blocking the convective mass transport, meanwhile the reduction of the diffusion mass transport remains within moderate limits. Despite that unfavourable fact, a beneficial property of both the polymer/silicate gels and clay system is that the quenching process is almost perfect in the blocking phase: on average less than 1% of the inlet radon concentration could be detected at the outlet surface of the sample having merely 3–4 cm thickness. Similar observations were made by Archibald and Hackwood [4] analysing the diffusion transport of radon through polysulfide copolymers and polyethylene terephtalate mats. On the other hand, it should be underlined that in this respect the daughter nuclides of radon

may concentrate in the sealing materials until they completely decay, resulting in stable lead isotopes.

4. Conclusions

- (1) Two experimental techniques were developed for the determination of the effective diffusion coefficients of radon in polymer/silicate gels and concentrated clay suspensions.
- (2) The effective diffusion coefficient was $3.3 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$ in gel-containing porous media that corresponds to $7.0 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$ in the bulk gel phase. That value is 30% less than the value characteristic in pure aqueous solutions. Since the diffusion coefficient of radon in the gas phase is about $10^{-1} \text{ cm}^2 \text{ s}^{-1}$, the reduction of radon flux by 5–6 orders of magnitude makes that sealing technology very attractive.
- (3) Similarly, the concentrated clay suspensions may also restrict both the hydrodynamic and the diffusive mass transport in porous media. The samples proved practically impermeable against convective flow, while the effective diffusion coefficient was found to be 1.3×10^{-5} cm² s⁻¹ for the emulsion and 6.0×10^{-6} cm² s⁻¹ for clay suspensions.

The laboratory studies firmly allow us to conclude that the polymer/silicate hydrogels and the clay-containing suspensions, as sealing agents are prospective candidates for practical application in porous and fractured media with the aim of restricting the radon migration. Further, it is definitely proved that an integrated technology based on their use offers an efficient solution for such environmental problems which jeopardise human health through direct inhalation of radon.

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