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ANALYTICAL MODEL OF MIGRATION OF RADIONUCLIDES IN POROUS MEDIA

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A mathematical model of transfer of radionuclides in a porous medium has been considered. The analytical solution for this model has been obtained in a generalized form suitable for practical use. A procedure which makes it possible to determine the time of attainment of the maximum concentration by the radionuclides and the coordinates of the zone of influence of the radioactive contamination source has been proposed. The Padé approximation of the scaled error function has been obtained; it can be employed in solving a wide range of problems of heat and mass transfer.

In evaluations of the safety of burial sites of radioactive waste, one often assumes that the effective individual equivalent irradiation dose is absorbed by man only when he drinks contaminated water. In this case it becomes topical to predict the state of ground water in the zone of influence of storages for the entire period of their potential hazard. The concentrations and flows of radionuclides in aqueous media are employed as the safety indicators. The problem of search for the influence zone is in turn reduced to determination of the coordinates of the boundary of the region at which the concentration of the radionuclide in water is lower than the Republic allowable levels (RALs) or is equal to it.

Study of the processes and parameters of mass transfer in water-bearing strata is based on the laws of the theory of migration of underground water, which provides a physicomathematical description of different mechanisms of complex processes of hydrodynamic and physicochemical character.

Migration of chemical components in water-bearing strata occurs within the framework of convective-diffusion processes (with allowance for the mechanism of gravitational differentiation) on which the processes of physicochemical transformations in underground water and those of interaction with enclosing rocks are imposed, and in the general case it is determined by:

(1) the indices of convective (filtration) transfer — filtration rate and active porosity (fracturing);

(2) the intensity of gravitational differentiation of solutions in water-bearing strata;

(3) the parameters of scattering — molecular diffusion and hydrodispersion in the pore (fracture) space;

(4) the indices of heterogeneous processes of physicochemical absorption (release) of a substance (sorption, ion exchange, dissolution, and others);

(5) the intensity of transformation (in particular, destruction) of a substance directly in the liquid phase on long-duration migration.

Numerical modeling is undeniably of primary importance in investigation (not only in prediction) of migration processes, since it replaces physical experiment under the conditions where the latter is either irrational or entirely impossible. In the latter case (if not only in it), the logic of investigations is dictated to an increasing extent by a mathematical apparatus subjectively appealing to a certain group of experts rather than by the character of the actual geological medium and the possible practical applications. As far as the existing limitations of the most widespread modifications of this apparatus are concerned, they follow primarily from the assumption of the stationarity of a random process which is often considered in a two-dimensional formulation, strongly distorting the resulting conclusions.

However, all this has long been known to the researchers themselves, as has been the impracticability of supporting the modeling by reliable initial data.

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In the touched-upon problem of *information support of the models*, the situation concerning investigation of inverse problems is no less disappointing. On the one hand, a stream of publications with constant optimism proposing increasingly more sophisticated procedures for their solution has not become weaker. However, even though these works did add something useful to the initial publications of ten to twenty years ago, they did this only in terms of negative conclusions, having confirmed once again the impossibility of doing away with incorrectness based on complicated formal manipulations with actual (insufficiently complete and reliable) initial data. Either by intuition or in view of the deeper insight into the problem, practical hydrogeologists have proceeded, as previously, from the principle "the simpler (the model and the scheme of solution of the inverse problem), the better," confining themselves, in complication of the model, to the predominantly determined fragmentation of it according to the geological base and the quality of the input information. It goes without saying that the possibilities of the so-called determined modeling (by mutual complementing and mutual correction of the initial data of different character, say, filtration and migration data) are by no means excluded [1].

For on-line analysis of the processes of transfer of radioactive water-soluble compounds one must have analytical solutions which show the character and laws of transfer and distribution of radioactive water-soluble compounds in soils and grounds in addition to numerical solutions.

For this purpose, we have employed in the present work a model the basis for which is the law of conservation of mass for the process of movement of radionuclides in porous media. The model is based on analytical solution of the convective-dispersion equation for contamination in a porous medium with the following assumptions: flow is nonuniform and unidirectional; the zone is modeled as an isotropic homogeneous porous medium; the dispersion is constant throughout the region; the contaminant moves as a soluble substance; the solid and liquid phases are in equilibrium and their concentrations are related by the linear distribution coefficient K_d.

For description of the processes of migration of radionuclides in a porous medium we selected the convective-diffusion model written for both the liquid and solid phases of the skeleton of the rock:

$$\frac{\partial (nC)}{\partial t} = \frac{\partial^2 (nDC)}{\partial^2 z} - \frac{\partial (nVC)}{\partial z} - \lambda nC - \alpha \, [C - F(N)] \,, \tag{1}$$

$$\frac{\partial (\rho N)}{\partial t} = \frac{\partial^2 (D_m \rho N)}{\partial^2 z} - \lambda \rho N - \alpha \, [C - F(N)] \,, \tag{2}$$

$$N = K_d C \,. \tag{3}$$

Combining Eqs. (1) and (2) and employing the isotherm of sorption according to Henry (3), on condition that the lumped parameters are constant we finally obtain

$$nR \frac{\partial C}{\partial t} = nD_{ef} \frac{\partial^2 C}{\partial^2 z} - VC - \lambda nRC \,, \tag{4}$$

where $R = 1 + \rho K_d / n$ is the retrogression factor which determines the slowing-down of the velocity of propagation of contamination due to the sorption processes and $D_{ef} = D + \rho K_d D_{mol} / n\psi$ is the effective coefficient of diffusion.

From the experimental data for the coefficients of dispersion of mixtures moving in a porous medium we have established the linear dependence of D on the velocity of the flow and the size of porous-medium particles [2, 5].

As a result of these experiments it was shown that the process of mixing of liquids is quite satisfactorily described by the diffusion equation with a convective term; the dispersion coefficient for both gases and liquids is written in the form $D = nD_{fr.v}/\psi + \gamma V/n$.

To solve Eq. (4) we introduce into consideration the similarity numbers of mass transfer in migration of radionuclides in a porous medium according to [2]: analog of the diffusion Péclet number $u = zV/nD_{ef}$, analog of the mass-exchange relaxation Fourier number $\tau = tV^2/Rn^2D_{ef}$, analog of the rate constant of the reaction of first order $\beta = \lambda n^2 R D_{ef}/V^2$, and dimensionless specific activity in the liquid phase $S = C/C_0$.

Within the framework of the assumptions made, the equation of transport of the contaminant was reduced to dimensionless form:

$$\frac{\partial S}{\partial \tau} + \frac{\partial S}{\partial u} = \frac{\partial^2 S}{\partial u^2} - \beta S \qquad (5)$$

with the initial conditions $S(0, \tau) = \exp(-\beta\tau)$.

The analytical solution has been obtained with the use of the Laplace transformation [2] and it has the form

$$S = \frac{1}{2} \exp(-\beta\tau) \left[\operatorname{erfc}\left(\frac{u - \tau}{2\sqrt{\tau}} \right) + \operatorname{erfc}\left(\frac{u + \tau}{2\sqrt{\tau}} \right) \exp(u) \right] . \qquad (6)$$

where $\operatorname{erfc}(Y) = 1 - \operatorname{erf}(Y)$ is the residual error function.

Expression (6) yields satisfactory results for $Y < 3$. However in evaluating the zones of influence of burial sites of radioactive waste and propagation of radioactive contaminants, the values of Y are more than 3 as a rule.

We introduce into consideration the scaled residual error function $\operatorname{erfcx}(Y)$, which is defined as $\operatorname{erfcx}(Y) = \exp(Y^2) \operatorname{erfc}(Y)$. With this function, after simple transformations, expression (6) will take the form

$$S = \frac{1}{2} \exp(-\beta\tau) \exp\left[-\left(\frac{u - \tau}{2\sqrt{\tau}} \right)^2 \right] \left[\operatorname{erfcx}\left(\frac{u - \tau}{2\sqrt{\tau}} \right) + \operatorname{erfcx}\left(\frac{u + \tau}{2\sqrt{\tau}} \right) \right] . \qquad (7)$$

Based on the theory of analytical approximations [3], for calculation of $\operatorname{erfcx}(Y)$ for $Y > 3$ we have obtained in this work the Padé approximation of sixth order, represented in the form

$$\operatorname{erfcx}(Y) = (\mathrm{F1}/\mathrm{F2} + \mathrm{F3}/\mathrm{F4})/(2Y\sqrt{\pi}) ,$$

where

$$\mathrm{F1} = 128Y^{14} + 6656Y^{12} + 127776Y^{10} + 1140480Y^8 + 4891320Y^6 + 9344160Y^4 + 6267870Y^2 + 645120 ;$$

$$\mathrm{F2} = 128Y^{14} + 6720Y^{12} + 131040Y^{10} + 1201200Y^8 + 5405400Y^6 + 11351340Y^4 + 9459450Y^2 + 2027025 ;$$

$$\mathrm{F3} = 128Y^{14} + 5760Y^{12} + 93280Y^{10} + 676800Y^8 + 2224152Y^6 + 2917320Y^4 + 1019970Y^2 ;$$

$$\mathrm{F4} = 128Y^{14} + 5824Y^{12} + 96096Y^{10} + 720720Y^8 + 2522520Y^6 + 3783780Y^4 + 1891890Y^2 + 135135 .$$

The values of $\operatorname{erfcx}(Y)$ computed according to this approximation have an error of less than 10^{-11}.

We have employed approximation by rational functions of the form of [4] for $Y < 3$.

For a prescribed distance from the contamination source the time of attainment of the maximum concentration is determined from the condition $\frac{\partial S}{\partial \tau} = 0$. Employing (6), we obtain

$$-\beta S + \frac{1}{2\sqrt{\pi}} \frac{u}{\tau^{3/2}} \exp(-\beta\tau) \exp\left[-\frac{(u - \tau)^2}{4\tau} \right] = 0 . \qquad (8)$$

Let us consider the ratio $\frac{\tau}{u} = \frac{t}{t_{\mathrm{p.s}}}$, where $t_{\mathrm{p.s}} = Rt_{\mathrm{p}}$ is the calculated time of the concentration front approaching the point z, $t_{\mathrm{p}} = nz/V$. We can write the relation $t_{\mathrm{p.s}} = \beta\tau/\lambda$ for $t_{\mathrm{p.s}}$ with the use of the dimensionless parameters.

We analyze the solution obtained for the condition where $t = t_{\mathrm{p.s}}$ for $u = \tau$. We rewrite Eq. (7) for this case in the form

$$S = \frac{1}{2} \exp(-\beta\tau) \left[1 + \operatorname{erfcx}(\sqrt{\tau}) \right] . \qquad (9)$$

For a prescribed distance we can determine the time of approach of the radionuclide-contamination front $t_{\text{p.s}}$ and compute the concentration of the radionuclides in water S from (9). At $\tau > 100$, (9) with an error of less than 1% takes the form

$$S = \frac{1}{2} \exp(-\beta\tau) \left(1 + \frac{1}{\sqrt{\tau\pi}}\right). \tag{10}$$

Let us consider the case where $\tau > u$. Then Eq. (7) takes the form

$$S = \frac{1}{2} \exp(-\beta\tau) \left\{2 - \exp\left[-\left(\frac{u-\tau}{2\sqrt{\tau}}\right)^2\right]\left[\operatorname{erfcx}\left(-\frac{u-\tau}{2\sqrt{\tau}}\right) - \operatorname{erfcx}\left(\frac{u+\tau}{2\sqrt{\tau}}\right)\right]\right\}. \tag{11}$$

The solutions obtained for the prescribed parameters enable us to evaluate the zone of influence of the radioactive contamination source, which is determined by the distance from the source where the maximum concentration in the aqueous solution corresponds to the RALs for a given radionuclide.

Thus, the problem is reduced to solution of the transcendental equation (8) (the values of S are computed from (11) for τ with a fixed β and different u). The program of computations has been realized in the Microsoft Excel environment. We have used the MATLAB mathematical system for solution in the graphic variant and for numerous calculations.

It is well known that among the radionuclides of Chernobyl origin the greatest migration capacity is exhibited by ^{90}Sr with a decay constant of $\lambda = 0.02415$.

According to RALs-99, the concentration in water for this radionuclide is 0.37 Bq/liter. Let us consider the cases where the initial concentrations take on values of 3.7 Bq/liter, 37 Bq/liter, 370 Bq/liter, and 3700 Bq/liter. The maximum dimensionless concentrations $S_{\max}$ will respectively be equal to 0.1, 0.01, 0.001, and 0.0001. Based on the solutions obtained we have calculated the basic characteristics (presented in Table 1) of migration of ^{90}Sr.

The transition from $u_{\max}$ to the coordinate prescribing the zone of influence of the radioactive source is determined by the expression

$$z_{\max} = \frac{u_{\max} n D_{\text{ef}}}{V}, \tag{12}$$

while conversion to the time of attainment of the maximum levels is written as

$$t_{\max} = \frac{\tau_{\max}\beta}{\lambda}. \tag{13}$$

The dimensionless concentration for which we determine $z_{\max}$ can be found from the formula

$$S_{\text{m.al.c}} = \frac{N}{K_{\text{d}} C_{\text{m.al.c}}}. \tag{14}$$

As an example of the employment of the results obtained in Table 1, we give calculations for specific cases. The initial data for the radionuclide ^{137}Cs are as follows: velocity of the moisture flow $V = 2.45$ m/g, effective coefficient of diffusion $D_{\text{ef}} = 0.41$ m^2/g, $R = 520$, active porosity $n = 0.35$, $\lambda = 0.023$, $\beta = 0.1$, $C_0 = 1000$ Bq/liter, $C_{\text{m.al.c}} = 10$ Bq/liter, $S_{\text{m.al.c}} = 0.01$, $u_{\max} = 40.12$, and $\tau_{\max} = 40.65$; the data for the radionuclide ^{90}Sr are $V = 2.45$ m/g, $D_{\text{ef}} = 0.41$ m^2/g, $R = 5$, $n = 0.35$, $\lambda = 0.02415$, $\beta = 0.001$, $C = 37$ Bq/liter, $C_{\text{m.al.c}} = 0.37$ Bq/liter, $S_{\text{m.al.c}} = 0.01$, $u_{\max} = 4398.27$, and $\tau_{\max} = 4560.97$. Employing (12) and (13), we obtain $z_{\max} = 2.35$ m and $\tau_{\max} = 176.7$ g for ^{137}Cs and $z_{\max} = 257.6$ m and $\tau_{\max} = 188.9$ for ^{90}Sr.

Thus, if the initial concentration of ^{90}Sr is thirty times lower than that of ^{137}Cs, the zone of its influence is two orders of magnitude larger. The smaller the generalized parameter β, all other parameters being the same, the larger the size of the influence zone.

TABLE 1. Coordinates and Time of Attainment of the Maximum Allowable Concentrations for Different Values of the Parameter β for ^{90}Sr

β	$S_{\text{m.al.c}}$	$t_{\text{p.s}}$, year	$\tau_{\text{p.s}}$	$t_{\max}$, year	$\tau_{\max}$	$u_{\text{p.s}}$	$u_{\max}$
0.1	0.1	71.78	17.3349	75.70	18.2816	17.33	17.41
0.05	0.1	70.43	34.1626	81.82	39.5191	34.01	35.37
0.01	0.1	68.43	165.258	90.26	217.978	165.23	194.06
0.005	0.1	67.92	328.054	92.05	444.602	328.01	403.06
0.001	0.1	67.23	1623.6	94.12	2273	1623.34	2146.39
0.0005	0.1	67.06	3239	94.53	4565.8	3238.6	4369.57
0.0001	0.1	66.84	16141.9	95.03	22949.7	16138.69	22430.52
0.1	0.01	165.51	39.9707	165.51	40.65	39.97	40.12
0.05	0.01	164.54	79.4728	169.46	81.8492	79.46	79.66
0.01	0.01	163.17	394.056	182.95	441.824	394.0	414.57
0.005	0.01	162.84	786.517	185.71	896.979	786.39	846.81
0.001	0.01	162.39	3921.72	188.86	4560.97	3921.0	4398.27
0.0005	0.01	162.28	7838.12	189.49	9152.37	7836.75	8896.04
0.0001	0.01	162.13	39154.4	190.24	45943	39148.7	45247.96
0.1	0.001	260.25	62.8504	260.35	63.50	62.83	63.0
0.05	0.001	259.40	125.29	260.80	125.966	125.27	126.2
0.01	0.001	258.30	623.795	276.04	666.637	623.69	638.78
0.005	0.001	258.03	1246.28	279.64	1350.66	1246.09	1296.27
0.001	0.001	257.67	6222.73	283.71	6851.6	6221.73	6665.13
0.0005	0.001	257.58	12441.1	284.52	13742.3	12439.31	13444.95
0.0001	0.001	257.47	62179	285.47	68941	62168.68	68121.77
0.1	0.0001	355.17	85.9016	360.31	87.0149	85.76	85.9
0.05	0.0001	354.48	171.214	362.38	175.03	171.19	171.53
0.01	0.0001	353.53	853.775	369.34	891.956	853.63	864.80
0.005	0.0001	353.30	1706.44	373.72	1805.07	1706.15	1748.50
0.001	0.0001	352.99	8524.71	378.63	9143.91	8523.29	8939.4
0.0005	0.0001	352.92	17046	379.60	18334.7	17043.01	18005.04
0.0001	0.0001	352.81	85203.6	380.63	91922.1	85190.0	91000.0

TABLE 2. Values of the Approximation Coefficients for Different β for $u_{\max}$ and $\tau_{\max}$

β	$b \cdot 10^{-4}$	$c \cdot 10^{-4}$	d	e	Standard deviation, 10^{-6}
0.1	2.0478257	44.324	–0.9632652	–0.498	1.429
0.05	–5.8704338	–115.525	–1.0532004	–0.6025	1.592
0.01	2.5802604	70.541	–0.9185112	–0.2086	1.610
0.005	2.0068692	54.65	–0.933288	–0.1602	1.558
0.001	2.3986865	47.266	–0.9542312	–0.07294	1.429
0.0005	0.76861726	21.2	–0.9711187	–0.0616	0.3077
0.0001	0.70923031	15.114	–0.9831404	–0.02786	0.2287
β	b_1	c_1	d_1	e_1	
0.1	–2.3208692	13.203	–0.9537831	–0.378	1.017
0.05	–49.011	–655.897	–1.1901479	–0.477	13.17
0.01	–13.864	–182.576	–1.0466606	–0.15065	11.61
0.005	0.41636123	11.977	–0.9754291	–0.02925	1.150
0.001	0.1722904	4.9095629	–0.990787	–0.011222	0.1543
0.0005	0.11467824	3.2865591	–0.9939253	–0.00754	0.04130
0.0001	0.36737994	5.7677462	–0.9957515	$6.83 \cdot 10^{-4}$	0.1023

By employing the data of Table 1, we can solve, for different radionuclides, the problem of search for the influence zone and of determination of the coordinates of the boundary of the region at which the concentration of the radionuclide is lower than or equal to RAL-99.

The analysis of the results in Table 1 has shown that the products $u_{\max}\beta$ and $\tau_{\max}\beta$ are approximated by the expression

$$u_{\max}\beta = bx^3 + cx^2 + dx + e\,, \tag{15}$$

$$\tau_{\max}\beta = b_1x^3 + c_1x^2 + d_1x + e_1\,, \tag{16}$$

where $x = \ln(S_{\text{m.al.c}})$, and the values of the coefficients are presented in Table 2.

By employing (15) and (16) for prescribed $S_{\text{m.al.c}}$ at two values of β, we can obtain the required solution for the selected parameter β using interpolation.

Thus, the analytical solution of the equation of convective diffusion of radionuclides with allowance for the kinetics of sorption and the exact solutions presented enable one to relate in explicit form the hydrodispersion, the velocity of a liquid flow, and the characteristics of transfer and mass exchange of radioactive water-soluble compounds and to determine the zone of influence of the radioactive contamination source and the time of potential hazard of this zone; also, they can be useful in developing experimental methods of determination of the characteristics of transfer and mass exchange of radioactive water-soluble compounds.

NOTATION

n, active porosity of the skeleton of the rock, m^3/m^3; C, specific activity of the radionuclide in the liquid phase, Bq/liter; D, coefficient of dispersion in the z direction, m^2/year; V, velocity of the flow, m/year; λ, decay constant of the radionuclide, 1/year; α, coefficient of mass transfer to the liquid, 1/sec; $F(N)$, sorption isotherm; N, activity of the radionuclide in the solid phase, Bq/kg; D_{m}, coefficient of molecular diffusion in the skeleton of the rock, m^2/year; $D_{\text{fr.v}}$, coefficient of molecular diffusion in the free volume, m^2/year; ψ, crookedness coefficient characterizing the inhomogeneity of a porous medium; ρ, density of the skeleton of the rock, kg/cm^3; D_{ef}, effective diffusion coefficient, m^2/year; z, coordinate in the direction of motion of the liquid; t, time; u, dimensionless coordinate; τ, dimensionless time; S, dimensionless specific activity; γ, hydrodispersion coefficient, m; K_{d}, coefficient of distribution of a water-soluble compound; R, retrogression factor; β, dimensionless constant of decay of the radionuclide; b, c, d, e, b_1, c_1, d_1, and e_1, approximation coefficients; V and x, parameters of the functions. Subscripts: ef, effective; m, molecular; 0, initial; fr.v, free volume; p, piston-type; p.s, piston-type with allowance for sorption; max, maximum; m.al.c, maximum allowable concentration.

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